Deposition of Atmospheric Pollutants on Forests [and Discussion]

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Deposition of atmospheric pollutants on forests

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Forests have long been considered as efficient sinks for atmospheric pollutants. The potential for large rates of deposition is provided by the turbulent structure of air above and within forest canopies. Large rates of deposition of pollutant gases, however, are only found for the very reactive gases HNO_3, HCl and NH_3. In contrast, the pollutants SO_2 and O_3 are deposited on forests and short vegetation at similar rates under the control of stomatal resistance.

Deposition of sub-micrometre aerosol particles on forests appears to be inefficient but at high elevations in the United Kingdom (up to 500 m) these aerosols are frequently activated into cloud droplets in the size range 5–10 μm (radius). These droplets are efficiently captured by forest canopies and this deposition pathway may make a large contribution to annual inputs at high elevation sites.

The effects of afforestation on inputs of pollutants to catchments are illustrated by model calculations for inputs of sulphur and nitrogen to Kielder forest in northern England. Inputs of sulphur and nitrogen to this area as moorland are estimated at 17.5 kg ha^{-1} (1 hectare = 10^4 m^2) and 12.4 kg ha^{-1} annually, respectively. Afforestation of the moorland increases sulphur and nitrogen inputs by 30 % and 90 %, respectively.

INTRODUCTION

During the past decade the focus of interest in the environmental effects of atmospheric pollutants has changed. In the early 1970s there was concern over the role of sulphur emissions in freshwater acidification (Drablos & Tollan 1980). Many aspects of freshwater acidification are now reasonably well understood, and the causal links between emissions of pollutants and freshwater acidification have become established. Throughout the late 1970s and 1980s there have been widespread declines in the health of silver fir (*Abies alba* Mill.), Norway spruce (*Picea abies* (L.) Itarst.), and beech (*Fagus sylvatica* L.) in Europe (Krause *et al.* 1986), and of red spruce (*Picea rubens*) in eastern North America (Johnson 1987). Although the declines have been attributed by some to the effects of pollutants acting directly or indirectly, no causal link between individual pollutants and this widespread forest decline has been established in the field.

Reports of forest decline in Europe show that at many sites symptoms are more prevalent at higher altitudes (Krause *et al.* 1986; Ammer *et al.* 1988). Many of the papers describing the problem refer in a non-specific way to 'efficient interception' of air pollutants by forests or 'filtering' by trees, so that an impression is generated that forests are particularly efficient at absorbing or scavenging pollutants from the atmosphere (Mayer & Ulrich 1974; Miller & Miller 1980). Such generalizations take no account of marked differences between the deposition processes for particles and gases to leaf surfaces, or of the great range of chemical reactivity with leaf surfaces for individual pollutants.

In this paper we examine the processes of deposition onto forest canopies, considering particles, cloud droplets and the major gaseous pollutants. In particular, the properties of forests as sinks for the major atmospheric pollutants are contrasted with those of shorter vegetation. In this way, the influence of afforestation on the atmospheric inputs may be quantified. A specific case, of Kielder Forest in northern Britian, is taken as an example to show the influence of afforestation on pollutant deposition. The strengths and weaknesses of current understanding of deposition onto forests are also identified. For several of the pollutant gases, and for cloud-water deposition, there are no direct field measurements, largely because of practical difficulties.

Measurement methods

Micrometeorological techniques

Micrometeorological techniques have been widely used to study fluxes of SO_2 (Garland 1977), HNO_3 (Huebert 1983), HCl (Dollard *et al.* 1987), NH_3 (Duyzer *et al.* 1987) and cloud water (Dollard *et al.* 1983). The flux-gradient technique has generally been used because pollutant sensors with adequate response times for eddy correlation methods have become generally available only recently. These practical restrictions, favouring flux-gradient methods, have led to an emphasis on short vegetation (cereals, grass and moorland vegetation), because of the uncertainty in flux-gradient relations over very tall crops (Thom 1975).

The main virtues of micrometeorological flux measurements are that they integrate over extensive areas of vegetation and, more importantly, the measurements provide the means to identify the relative importance of atmospheric and surface components of the deposition process.

The flux density (F_s) above extensive uniform surfaces may be written:

$$F_s = K_s\,\delta\chi/\delta z, \tag{1}$$

where K_s is the eddy diffusivity for the gas or particle and $\delta\chi/\delta z$ is the vertical concentration gradient. The eddy diffusivities, K_s (for gases) and K_m (from momentum), are essentially independent of the chemical nature of the gas or particle, and are determined by the aerodynamic roughness of the underlying vegetation, the wind speed, and to a lesser extent on the temperature structure of the turbulent boundary layer (Thom 1975).

The vertical gradient in concentration ($\delta\chi/\delta z$) has been shown to provide satisfactory estimates of the fluxes above short vegetation, whereas over forests flux-gradient relations break down close to the canopy surface and therefore introduce important practical restrictions to the technique.

Eddy correlation methods rely on detection of turbulent fluctuations in the vertical component of turbulence (w') and in fluctuations in the concentrations of the pollutant gas (χ'). The flux is given by

$$F_s = \overline{w'\chi'}. \tag{2}$$

Eddy correlation measurements of sulphur fluxes onto forests by this method have been reported by Galbally *et al.* (1979), Fowler & Cape (1983) and more recently by McMillan *et al.* (1987). The techniques are shown to be well suited to aerodynamically rough surfaces such as forests. Results from these studies are used later to estimate annual sulphur dioxide deposition on forests.

Enclosure methods

A mass budget in a 'closed' or 'open' chamber (figure 1) provides an estimate of the loss of material to the vegetation or soil or both within the box (Unsworth & Mansfield 1980; Galbally & Roy 1981). Chambers ('cuvettes') may enclose single leaves or branches, or in the case of short crops and soil they may enclose up to 3 m^2 ground area of vegetation. However, for complex or large canopies of vegetation, there are considerable problems in scaling the measurements to provide fluxes to the entire canopy. The most important progress using cuvette studies has been in studying the role of biological processes in the deposition pathway (Hällgren *et al.* 1982), and in measuring fluxes that are too small to be measured by micrometeorological methods (Johannson 1987).

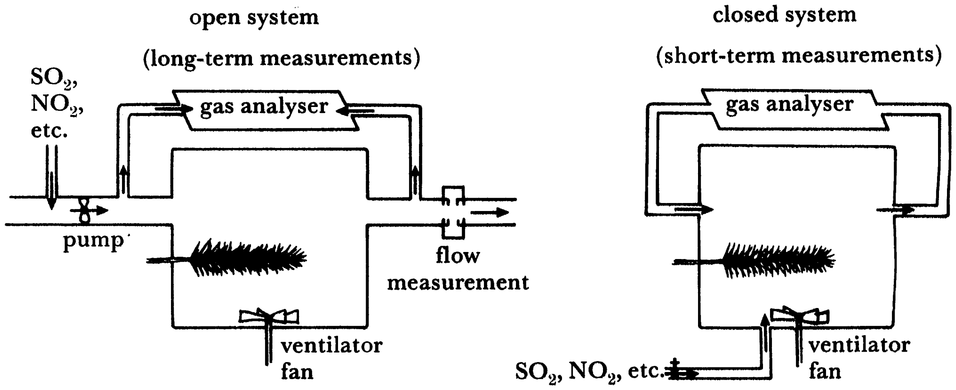

FIGURE 1. Schematic representation of simple open and closed 'box' systems for gas exchange.

A combination of practical and theoretical problems with techniques, summarized in table 1, has therefore steered measurements of dry deposition away from forests, so that for several pollutants there are few (if any) field measurements, and we have to rely on information from studies using shorter vegetation, and from cuvette measurements.

TABLE 1. COMPARISON OF MICROMETEOROLOGICAL AND CHAMBER METHODS FOR FLUX MEASUREMENT

micrometeorological	chamber
Provide average fluxes over extensive canopies (10^3–10^5 m^2 ground area).	Scale: from single leaf to whole plants, 0.1–5 m^2 ground area.
Restricted to uniform, horizontal and extensive canopies.	Difficult to estimate bulk canopy rates of exchange from chamber measurements.
Restricted to a few gases by instrument sensitivity and response time and measurement problems, e.g. advection and storage introduce errors in the measurement of small fluxes.	By modifying chamber conditions, very small fluxes may be measured.
Influence of the natural environment on fluxes may generally be studied in comprehensive micrometeorological experiments.	Chamber effects always modify results and may be difficult to evaluate.
Extensive arrays of sensors and data logging equipment are required to allow detailed interpretation.	Responses of the flux to imposed changes in chamber atmosphere, or vegetation, provide mechanistic studies of the deposition pathway.

Gases

The gases considered below are SO_2, NO_2, NO, O_3, HNO_3, HCl and NH_3. Although this is not an exhaustive list, most of the major pollutant gases are included. Rates of uptake of these gases by forests are determined by turbulent diffusion above and within the canopy, molecular diffusion close to the individual components of the vegetation and within stomata, and by rates of chemical reaction between individual gases and leaf surfaces, or rates of solution in intercellular fluids within the vegetation (illustrated in figure 2). For the range of gases under consideration, control over rates of deposition varies from cases where deposition is determined entirely by rates of atmospheric transfer, to cases with deposition controlled almost entirely by stomatal conductance, and hence by biological processes. The former cases, for exceptionally reactive gases, show no dependence of deposition on surface processes, whereas the latter, for gases that do not react with leaf surfaces, are insensitive to rates of turbulent diffusion, wind speed, etc.

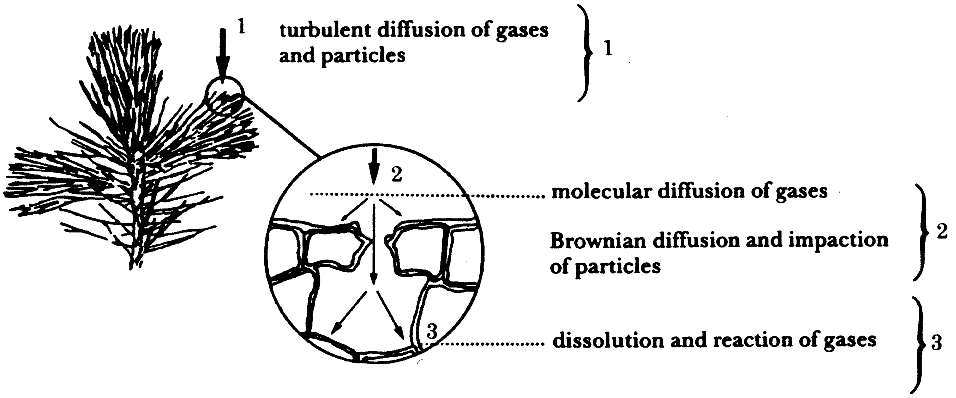

FIGURE 2. Transport processes of gases and small particles to vegetation.

Resistance analogy

Measurements of deposition rates of gases have commonly been interpreted by treating the gas transfer process as analogous to the flow of electrical current through a network of resistances (Monteith 1973).

In a constant flux layer, integration of equation 1 with respect to height between height z_1 and z_2 yields

$$F_s = \{\chi_s(z_2) - \chi_s(z_1)\} \int_{z_1}^{z_2} K_s^{-1}\,dz. \tag{3}$$

The lower limit of the integral is an imaginary absorbing surface within the vegetation canopy. In practice, the sites of absorption of trace gases within the canopy are more complex, but despite the complications, this approach provides a very useful tool to contrast the different gases and their properties.

The total resistance to transfer, r_s, where $r_s = \int_{z_1}^{z_2} K_s^{-1}\,dz$, comprises atmospheric and surface terms, and the reciprocal of r_s is identical to the deposition velocity $v_g(z)$ (Chamberlain 1960), which is widely used to parametrize deposition rates of reactive gases. Deposition velocity may also be regarded as a flux normalized for concentration ($v_g(z)$ = flux/concentration (z)).

The total resistance, r_s, may be subdivided into atmospheric and surface components. The atmospheric components r_a and r_b are given by $r_a = u/u_*^2$ and $r_b = B^{-1}u_*^{-1}$, where u is the wind velocity at a reference height above the canopy, u_* is the friction velocity and B is the sub-layer Stanton number. Whereas r_a can be readily determined from a knowledge of wind profiles over the forest canopy, r_b has seldom been measured directly, and is often estimated from empirical relations making allowances for diffusivities of gases and particles. Thom (1972) developed a theoretical basis for r_b in plant canopies, demonstrating that r_b is necessary for describing mass transfer for two reasons: (i) the absence in mass transfer of bluff body forces, which transfer momentum to the canopy more effectively than mass; (ii) differences in the heights of apparent sinks for momentum and mass within the canopy. Thom (1972) suggested that for many leafy canopies r_b should be almost independent of aerodynamic roughness, but this is not the case for two extremes: (i) canopies with many fibrous elements (such as pine needles), or (ii) surfaces where the roughness elements are rigid and impermeable to the wind. For the former case, Stewart & Thom (1973) found that r_b for evaporation from a pine forest was about half the value estimated from empirical formulae valid for shorter, leafy canopies. On the other hand, values of r_b measured by Chamberlain (1975) for transfer of vapours and particles to rough surfaces were 1–2 orders of magnitude larger than those generally assumed for vegetation. It seems likely that r_b for forests can take a wide range of values depending on the density of the foliage, leaf form, tree spacing and surface topography. It is important to know more about r_b because of the controlling role that $(r_a + r_b)$ may have in the transfer of highly reactive gases and in particle and droplet deposition.

When the atmospheric resistances have been subtracted from the total resistance, the residual term is known as the surface or canopy resistance r_c, which is determined by the parallel transfer pathways to leaf surfaces, through stomata, and to underlying layers of vegetation or litter after further transport through the canopy space.

RESULTS

It is now appropriate to consider each of the major gases in turn, showing for forests the relative importance of aerodynamic and surface resistances. It is convenient to group the gases by their reactivity.

HNO_3, HCl

Among the gases under consideration, nitric acid vapour is the most reactive, and is readily absorbed by leaf and stem surfaces. All field and laboratory measurements at temperatures above 0 °C show that the canopy or surface resistance for HNO_3 is zero, and that rates of deposition are determined entirely by atmospheric transfer (Huebert 1983; Johannson *et al.* 1982; Johansson & Granat 1986; Dollard *et al.* 1987). For experimental reasons, most of the field measurements have been made over short grass. It is therefore possible to predict rates of deposition in the field from estimates of r_a and r_b; for a typical forest 10 m high, with a wind speed of 5 m s^{-1}, 2 m above the canopy, the deposition velocity would be about 10 cm s^{-1}. At such rates of deposition, with an average HNO_3 concentration in air of 0.5 parts in 10^9 by volume (Derwent 1986), this provides an annual input of nitrogen about 8 kg ha^{-1} from nitric acid vapour. Such very large deposition rates deplete the mixing layer rapidly at night below the nocturnal inversion; for example a shallow inversion, up to 100 m, would be depleted in about 20 minutes. The dry deposition of nitric acid at night would therefore be restricted at sites isolated from the bulk of the mixed layer by a shallow nocturnal inversion to an hour or so after sunset. At windy sites, for example on hills, the nocturnal inversion may develop only infrequently for short periods and at such sites the dry deposition process would continue most of the time. Local topographic features and climate would therefore be expected to strongly influence the magnitude of nitric acid inputs.

As surface resistance is negligible for HNO_3 transfer, rates of deposition are limited only by atmospheric transport and are very sensitive to the aerodynamic roughness of the vegetation.

Rates of HCl deposition on vegetation measured in field conditions by Dollard *et al.* (1987) show no evidence of a surface resistance. The deposition velocity for HCl may therefore be assumed to be identical to that for HNO_3 and be determined by the aerodynamic and boundary layer resistances r_a and r_b. Forests are therefore much more efficient sinks for gaseous HNO_3 and HCl than agricultural crops because of their height and hence their aerodynamic roughness. The effects of vegetation height and windspeed on rates of deposition of HNO_3 and HCl are shown in table 2.

TABLE 2. HNO_3 AND HCl DEPOSITION ONTO VEGETATION

	short grass (h 0.1 m)	moorland (h 0.4 m)	cereal (h 1.0 m)	forest (h 10.0 m)
deposition velocity, v_g (mm s^{-1}) at wind speed of 1 m s^{-1}	5.2	7.6	14.3	40.0
deposition velocity, v_g (mm s^{-1}) at wind speed of 4 m s^{-1}	23.5	33.3	50.0	100.0

surface resistance (r_c) assumed = 0, thus $v_g = (r_a + r_b)^{-1}$.

NH_3

The presence of gaseous ammonia in the atmosphere has been recognized for many years (Healy *et al.* 1970). Within the past decade, however, the concentrations of NH_3 in the air and its removal at the ground in The Netherlands have been regarded as a significant factor in widespread change in the flora of acidic heathland (Heil *et al.* 1987). Concentrations of gaseous ammonia in air range from 0.1 parts in 10^9 to 5 parts in 10^9 by volume for rural air, and recent results of deposition studies over vegetation show very small surface resistances (Duyzer *et al.* 1987). For moorland the surface behaved as a perfect sink whenever the vegetation was wet but in dry conditions a small surface resistance, constituting about 25 % of the total transfer resistance, was observed. An average deposition velocity of 15 mm s^{-1} was reported for this surface. The only published field measurements of ammonia deposition on a forest canopy show, as for moorland, that rates of uptake are controlled primarily by atmospheric transfer (Duyzer *et al.* 1987), with deposition velocities over young Corsican pine (*Pinus nigra* var. *maritima* (Ait.) Melville) trees (2.5 m high) of 20–30 mm s^{-1}.

SO_2, O_3

For gases absorbed through stomata, where there is rapid dissolution or reaction within the sub-stomatal cavity but limited uptake by external surfaces of the plant, the maximum rates of uptake are determined mainly by the bulk stomatal resistance of the canopy (table 3). Forest canopy resistances associated with stomatal diffusion are seldom less than 150 s m^{-1}, comparable to, or rather larger than, for short vegetation; this gives an upper limit to the deposition velocity of about 9 mm s^{-1} (depending on diffusivity) for these gases. For forests, in the absence of uptake by leaf surfaces, the rates of deposition of SO_2, and O_3 are therefore unlikely to exceed rates of deposition onto short vegetation.

Considering first SO_2, for which dry deposition is the best understood of the pollutant gases, rates of deposition to a range of vegetation types have been measured (see Garland 1977; Fowler & Cape 1984). In general, these have shown that for dry canopies the rates of uptake by leaf surfaces are small for agricultural crops, with maximum deposition velocities of around 2 mm s^{-1}. With stomatal uptake dominating leaf surface uptake, a pronounced diurnal cycle is observed, with daily maxima of around 10 mm s^{-1} and night minima of 2 mm s^{-1}. For forests there are fewer measurements, but those available (see Hicks *et al.* 1983; Fowler & Cape 1984) show much smaller deposition velocities, with uptake by external surfaces being very small, of the order of 1 mm s^{-1}, and the daytime maxima reaching only 6 mm s^{-1}. For the measurements cited, the surface resistances were almost always in excess of 90% of the total resistance, and rates of deposition were therefore insensitive to wind speed. An internal resistance to SO_2 uptake was postulated to explain deposition measurements to Scots pine (*Pinus sylvestris* L.) (Fowler & Cape 1983) because measured rates of dry deposition were much smaller than would have been expected on the basis of known stomatal conductances (found by simultaneous measurements of transpiration). An alternative concept to explain these data would involve simultaneous emission of H_2S from surface or internal sources and dry deposition of SO_2. When surfaces are wet and NH_3 is present the canopy resistance for SO_2 may disappear.

Table 3. SO_2 transfer to a Scots pine canopy showing the presence of an internal resistance to SO_2 uptake within the leaves

Greenwich Mean Time	deposition velocity $(v_g)/(\text{mm s}^{-1})$	bulk canopy resistance for H_2O (s m^{-1})	bulk stomatal resistance for SO_2 (s m^{-1})	‘internal’ resistance (s m^{-1})
12h30–13h30	6.9	78	151	0
13h30–14h30	3.2	56	356	253
14h30–15h30	4.7	52	232	128
15h30–16h30	1.8	100	714	514
16h30–17h30	1.5	170	909	560

Ozone is a very similar gas to SO_2 in its transport to vegetation in that rather small rates of uptake occur at external surfaces. Canopy resistances for ozone are closely coupled to stomatal resistances and may be readily predicted from a knowledge of water vapour exchange by the canopy. This is surprising for a reactive but relatively insoluble gas such as O_3. The sinks for O_3 within the plant are assumed to be through reaction with solutes in the intercellular fluids or with membranes accessible from the sub-stomatal cavity. Measurements of ozone deposition to forest canopies are few, whereas measurements in cuvette systems are plentiful (Johansson *et al.* 1982). Table 4 shows typical resistances and deposition velocities of SO_2 and O_3 to forest canopies based on literature values.

Because of the importance of stomatal resistance in controlling canopy resistance for O_3 and SO_2 deposition, it is possible to extend mechanistic models of water vapour and CO_2 exchange of forest canopies to allow estimates of deposition velocities. Figure 3 shows some preliminary calculations made by J. C. Wilshaw (personal communication), who modified a model 'MAESTRO' (Grace *et al.* 1988) to include surface deposition as well as stomatal uptake. 'MAESTRO' determines stomatal resistance by first calculating the light distribution in the foliage (the structure of which can be specified in detail) and assuming physiological relations between light and stomatal resistance. The figure shows the strong diurnal cycle in deposition velocity as a consequence of stomatal responses. Further work is necessary to test modelled deposition against field measurements.

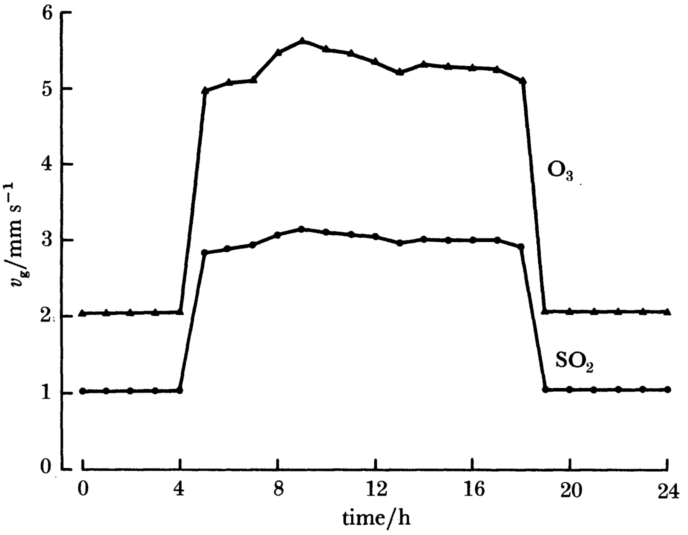

FIGURE 3. SO_2 and SO_3 deposition on to a red spruce (*Picea rubens* Sarg.) canopy 15 m in height simulated by the MAESTRO model (Grace *et al.* 1988).

TABLE 4. SO_2 AND O_3

(Forest 20 m in height with wind speed of 3 m s^{-1}, 2 m above the canopy, and $r_a + r_b$ 10 s m^{-1} for all gases above r_{c1} stomatal resistance, r_{c2} leaf surface resistance, r_i internal resistance.)

		r_{c1} (s m^{-1})	r_{c2} (s m^{-1})	r_i (s m^{-1})	r_{total} (s m^{-1})	v_g (mm s^{-1})
day and summer	SO_2	150	500	200	176	5.7
(i.e. r_{c1} min)	O_3	150	500	0	125	8.0
night and winter	SO_2	∞	1000	—	1000	1.0
	O_3	∞	500	—	500	2.0

NO_2, NO

The rates of exchange of NO_2 and NO with vegetation appear to be the most complex of all pollutant gases studied to date. Physiological processes within plants may produce NO_2 or NO or both, so that in circumstances where the atmospheric concentrations of the gas are less than 50 parts in 10^{12} by volume, a net flux away from the canopy may be expected.

Uptake of NO_2 by trees has been measured by cuvette techniques (Johansson *et al.* 1982; Grennfelt *et al.* 1983; Johansson 1987) but no measurements by micrometeorological methods over forests have been reported. The cuvette studies show maximum deposition velocities for NO_2 at concentrations of 25 parts in 10^9 by volume (p.p.b.v.) of about 1.5 mm s^{-1} expressed on a leaf-area basis and equivalent to about 5 mm s^{-1} for a Scots pine canopy (Grennfelt *et al.* 1983). More recently, the work of Johansson (1987) has shown the deposition velocity for NO_2 on Scots pine to depend strongly on NO_2 concentration. At NO_2 concentrations of 40 p.p.b.v. the deposition velocity was proportional to stomatal conductance for NO_2 up to 2 mm s^{-1} (on a projected needle-area basis) with no significant intercept, so that uptake was solely stomatal. At NO_2 concentrations of 4 p.p.b.v. there was very little uptake of NO_2, with a maximum value of v_g approaching 1 mm s^{-1}. At 1.2 parts in 10^9 there was no net uptake and no dependence of the flux on stomatal conductance. Such findings have important implications for NO_2 uptake in rural areas where concentrations of NO_2 in air are generally less than 5 p.p.b.v. Recent measurements over moorland vegetation showed deposition velocities for NO_2 of only 1–2 mm s^{-1} (table 5) at NO_2 concentrations of around 1 p.p.b.v. and surface resistances around 90% of the total resistance. The site of absorption of NO_2 by vegetation is also in doubt. As shown by Lee & Schwartz (1981), the reaction of NO_2 (aq) with water to give NO_3^- and NO_2^- ions is very slow at concentrations typical of rural areas, typically 5×10^{-13} mol dm^{-3} s^{-1} at 10 p.p.b.v. NO_2 and 25 °C. This implies an equilibrium between $NO_2(g)$ and $NO_2(aq)$, and no dependence of uptake by water on gas phase transport.

Table 5. NO_2 deposition onto acidic moorland (*Eriophorum* sp. and *Carex* sp.)

(Vegetation height 10 cm. Fetch, between 100 and 500 m. Conditions 16h00–20h00 Greenwich Mean Time: net radiation 200–0 W m^{-2}; air temperature 13–11 °C, reference height for U, NO_2 etc. is 1 m.)

run	NO_2 p.p.b.v.	U^* (m s^{-1})	U (m s^{-1})	$r_a + r_b$ (s m^{-1})	v_g (mm s^{-1})	r_s (s m^{-1})	$r_s/r_t \times$ 100 (%)
1	0.64	0.23	2.74	62.3	1.6	563	90.1
2	0.54	0.23	2.71	66.4	2.2	388	85.6
3	0.53	0.28	2.98	46.0	2.4	370	88.9
4	0.38	0.39	4.10	33.0	3.6	245	88.4
5	0.37	0.36	3.80	36.0	2.9	308	89.8
6	0.39	0.35	3.50	38.0	2.7	332	89.7
7	0.37	0.17	1.80	78.0	1.8	477	85.9

The potential source or sink represented by NO_2 in the aqueous phase within intercellular fluid may be estimated and its influence on measured fluxes determined. Taking 1 kg water per square metre (ground area) as the water in equilibrium with NO_2 at an ambient NO_2 concentration of 10 parts in 10^9, the storage term amounts to 10^{-9} g m^{-2} ground area. A change in ambient NO_2 concentration from 10 to 1 p.p.b.v. over 1 minute would result in a flux of 5×10^{-11} g m^{-2} s^{-1}, which would be negligible if deposition velocity exceeded 0.1 mm s^{-1}.

The deposition velocity by an aqueous route is therefore determined by the irreversible reaction of NO_2 (aq) within the leaf with substances other than water, and is likely to be controlled by leaf biochemistry, or the presence in solution of other pollutants such as SO_2, or both.

However, Lendzian & Kerstiens (1988) have reported much higher permeability of leaf cuticles to NO_2 than to other gases, including O_3, CO_2 and SO_2. NO_2 was absorbed by isolated cuticles giving an increase of about 10% in cuticle mass at saturation. Most of the absorbed NO_2 was fixed in the cutin matrix of the cuticle, presumably through nitration reactions, but some NO_2 was absorbed reversibly. Assuming a partition coefficient between cuticle and air of 4000 (K. J. Lendzian, personal communication) a cuticle mass of 0.25 mg cm^{-2} and a leaf area index of 4, the amount of reversibly absorbed NO_2 per square metre ground area would be *ca.* 2 g at an NO_2 concentration of 10 parts in 10^9 in air. Although no information on the kinetics of NO_2 absorption by cuticles is available, if uptake were in equilibrium with a change in concentration of 10 parts in 10^9 min^{-1}, from 0 to 10 parts in 10^9 NO_2, the reversible absorption rate would be equivalent to a deposition velocity of *ca.* 3×10^{-3} m s^{-1}. It is therefore possible that measurements of dry deposition of NO_2 could be confounded by reversible absorption and storage of NO_2 by leaf cuticles, and that in field conditions relatively little NO_2 is taken into solution inside leaves.

Nitric oxide fluxes over vegetation in the field have been shown to be generally away from the surface, with fluxes of the order of 5–20 ng m^{-2} s^{-1}, the source of the nitric oxide being primarily denitrification processes in soil under anaerobic conditions (Johansson & Galbally 1984). A forest canopy may reduce the net loss of N to the atmosphere by absorbing a proportion of the NO emission that has been oxidized by ambient ozone to NO_2 within the forest canopy, but for the reasons outlined above this is unlikely to be an important process.

Particles, cloud and rain

(*a*) *Particles*

The interception of aerosol particles, and cloud and rain droplets, by forest canopies has long been considered to be an important mechanism, generating larger inputs to forests than to shorter vegetation (White & Turner 1970; Mayer & Ulrich 1974). However, calculations of the efficiency of collection of particles by vegetation indicate a strong dependence on particle size (figure 4). On theoretical principles sub-micron aerosols, the size range in which most of

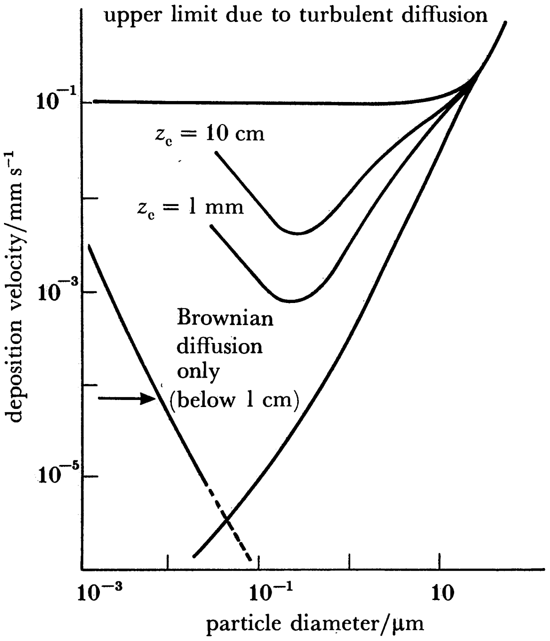

FIGURE 4. Predicted velocities 1 m above vegetation for unit density particles in size range 10^{-3}–10^{2} μm.

the sulphate and nitrate particulate material in the atmosphere is found, are deposited very inefficiently, whereas droplets of cloud water and fine rain are captured rather efficiently by vegetation. The particle size distributions for the sulphate and nitrate aerosol and for typical orographic cloud found in upland Britain are shown in figure 5*a*, *b*.

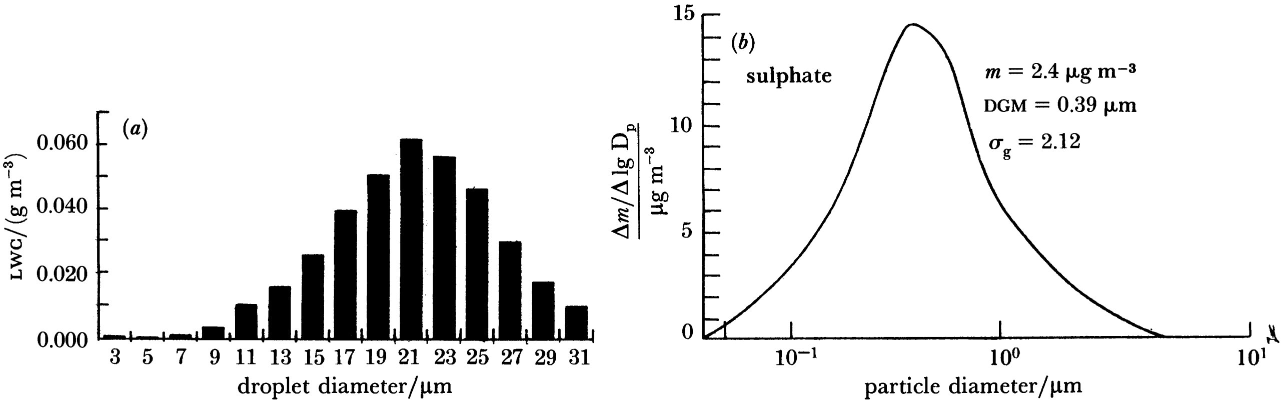

FIGURE 5. (*a*) Drop size distribution for cloud at an upland site in northern Britain (after Fowler *et al.* 1988). (*b*) Typical size distribution for aerosol sulphate (after Whitby 1977). Abbreviations: LWC, liquid water content; DGM, geometric mean droplet diameter; *m*, particle concentration in air.

Theoretical calculations and wind-tunnel measurements of deposition velocities of sub-micrometre aerosols to leaves differ by about a factor of four, with measurements the larger. Reasons for the discrepancy are not known. Belot *et al.* (1976) used results of wind-tunnel measurements to estimate deposition velocities to a pine forest 15 m high, and predicted that v_g for 0.5 μm diameter particles increased from about 0.2 mm s^{-1} at a windspeed of 1 m s^{-1} (canopy top) to about 1 mm s^{-1} when the windspeed was 5 m s^{-1}.

Deposition of particles with diameters between 1 μm and 5 μm is critically influenced by the small-scale roughness of surfaces. In identical conditions, the deposition velocity may be an order of magnitude larger to hairy leaves than to smooth leaves (Little 1977). This is probably because impaction on micro-roughness elements is an important deposition mechanism in this size range. Because of the variation imposed by surface micro-structure, it is unlikely that precise models of particle deposition in this size range to forests can be developed from theoretical principles, and there is a need for more field measurements of particle fluxes in the sub-micrometre and low micrometre size ranges, using methods that can distinguish particle sizes.

(*b*) *Cloud*

In comparison with particles 0.1–5 μm in diameter, cloud droplets in the size range 5–40 μm radius are collected rather efficiently by foliage via impaction and sedimentation. Consequently the nucleation during cloud or fog formation of sulphate and nitrate aerosols, which are ubiquitous in the industrialized countries of the world, transforms relatively non-depositing particles into readily deposited droplets. Recent field measurements over moorland, summarized in table 6 (Gallagher *et al.* 1988) have shown that the average turbulent deposition velocities of cloud droplets with diameters 5–10 μm are very similar to that for momentum, in agreement with the predictions of Chamberlain (1975) and Shuttleworth (1977).

Table 6 shows that particles larger than about 10 μm appeared to be deposited less

TABLE 6. DEPOSITION VELOCITIES CALCULATED (FROM GALLAGHER *ET AL.* 1988) AS A FUNCTION OF DROPLET SIZE

radius µm	v_t (mm s^{-1})	v_d (mm s^{-1})	v_t/v_m
2.5	5	5	0.09
3.5	38	40	0.71
4.5	27	29	0.50
5.5	42	46	0.79
6.5	54	58	1.01
7.5	45	51	0.84
8.5	62	71	1.16
9.5	68	79	1.27
10.5	76	89	1.42
11.5	69	84	1.29
12.5	47	66	0.88
13.5	42	64	0.79
14.5	23	49	0.43
15.5	22	51	0.41

v_t, turbulent deposition velocity; $v_d = v_t$+sedimentation velocity. $v_m = r_a^{-1}$.

effectively than momentum absorption by turbulent transfer. This may be an experimental artefact, but it may also be because the relaxation time of these drops does not allow them to be transported in high frequency eddies. If the latter is a significant feature, it implies that deposition velocities of larger cloud drops will be influenced by the turbulent spectrum generated by aerodynamically rough surfaces.

Results such as those in table 6 have important implications for forests that are frequently enveloped in cloud associated with wind speeds of 5–10 m s^{-1}, which is the case for much of the land in western Britain at altitudes in excess of 400 m. They indicate that rates of cloud-water deposition to forests 10–15 m high, with cloud liquid-water contents of 0.2 g m^{-3}, are about 0.15 mm h^{-1} of water or about 10% of typical rainfall rates. For land above 600 m, which is in cloud for typically 2000 h per year in western Britain, cloud-water deposition may increase precipitation by about 12% and may add up to 40% of additional solutes to the wet deposition, because the concentrations of the major ions in cloud water exceed those in rain water by a factor of between 2 and 3 (Fowler *et al.* 1988). Differences between average cloud-water and rain-water composition are shown in figure 6 for a site in northern England. Cloud-water deposition is much less important over shorter vegetation (Unsworth & Crossley 1987).

The effect of afforestation, is therefore, to increase the inputs of major nutrients and pollutants from the atmosphere for all ions contained in cloud water. The major difference between low and high elevation land is that the activation of relatively non-depositing small sub-micrometre aerosols into cloud droplets over upland areas converts the particles into a readily deposited form, and land use in the uplands simply determines the sink strength. These arguments are illustrated diagrammatically in figure 7.

Within cloud on hilltops, as illustrated in figure 7, there may be variability in the cloud-water deposition and in solute concentrations on foliage. Beginning at the condensation level at the upwind edge, the processes of nucleation, condensation and coalescence lead to a spectrum of drop sizes that shifts to larger diameters as air is lifted up the hillside. Because impaction efficiency increases sharply with drop size, there are likely to be regions near the condensation level, both at the upwind and downwind edges, where cloud-water deposition

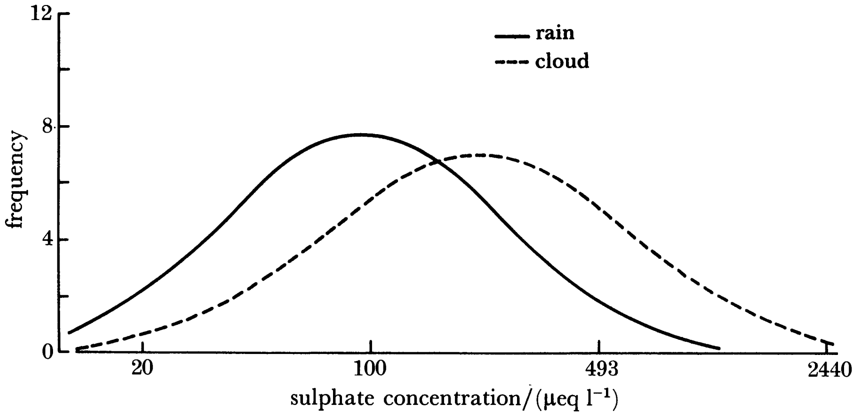

FIGURE. 6. Concentrations of sulphate in rain and orographic cloud collected at the same site in northern Britain (180 samples in each distribution).

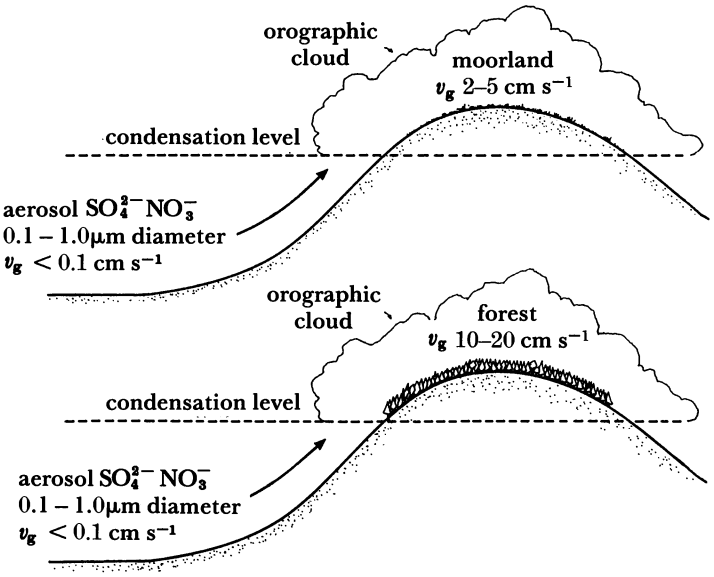

FIGURE 7. Effect of orographic cloud and land use at hill sites on the deposition rate of particulate SO_4^{2-} and NO_3^- to vegetation.

changes rapidly. In addition, entrainment of unsaturated air and penetration of solar radiation through the developing cloud can create conditions in which evaporation from leaf surfaces, occurring simultaneously with deposition, can considerably increase the solute concentrations of water on leaf surfaces (Unsworth 1984; Milne *et al.* 1989). The effect is likely to be even more common at the top of a cloud layer that does not fully cover a hill or mountain, and may lead to damaging chemical concentrations on foliage.

(*c*) *Rain*

For rain droplets, falling under gravity, inputs are the same for moorland or forest, although subsequent evaporation from a wetted forest canopy is more rapid than for moorland, and the pattern of distribution of water to the soil is very different.

Application

One of the most important and contentious aspects of forest–pollution interactions is the extent to which forests modify chemical and hydrological inputs to catchments. The preceding arguments show that for the very reactive pollutant gases, HNO_3 and HCl, forests increase the inputs as a consequence of enhanced turbulence over the aerodynamically rougher surfaces of forest canopies relative to 'smoother' grass or moorland surfaces. For similar reasons, the inputs of cloud water are also enhanced. For NH_3, all available field measurements over 'natural' (unfertilized land) provide deposition velocities similar to those for HNO_3 and HCl. For the gases SO_2, and O_3 the rates of dry deposition to forests do not differ greatly from those to shorter vegetation, as these gases are absorbed through stomata and there is little or no net uptake on external surfaces of leaves.

These arguments are illustrated for an upland region in the Scottish Borders and northern England over which large scale afforestation has taken place. This area, Kielder Forest, was largely open moorland at the turn of the century and now has 60000 ha of conifer plantation forests with an average height of 15 m.

Table 7. The effects of afforestation on inputs of major ions from the atmosphere to Kielder in northern Britain

(An area of moorland of 6×10^4 ha has been afforested with conifers that are now 15 m tall. The budget below shows inputs to the area for moorland and for trees; all other conditions remain the same.)

atmospheric conditions

concentration in air: annual averages (parts in 10^9 by volume)

HNO_3	HCl	SO_2	NH_3†	NO_2†
0.3	0.1	2.0	3.0	5.0

concentrations (for 1985 (RGAR 1987) and cloud (Fowler *et al.* 1988)

	Cl^-	SO_4^{2-}	NO_3^-	H^+	Na^+	NH_4^+	Mg^{2+}	Ca^{2+}
rain	64	46	15	19	58	21	11	10 μeq l^{-1}
cloud water	166	93	42	74	139	50	26	24 μeq l^{-1}

forest/(g m^{-2} per year)

	wet deposition	dry deposition	cloud-water deposition‡	total
SO_4^-	1.31	0.31 (SO_2)	0.65	2.27
NO_3^- (N)	0.35	0.42 (NO_2, HNO_3)	0.09	0.86
NH_4^+ (N)	0.45	0.93 (NH_3)	0.10	1.48
Cl^-	4.98	0.20	0.86	6.04
Na^+	2.81	—	0.46	3.27
Mg^{2+}	0.33	—	0.04	0.37
Ca^{2+}	0.62	—	0.07	0.69
moorland/(g m^{-2} per year)				
SO_4^{2-}	1.31	0.31	0.13	1.75
NO_3^- (N)	0.35	0.22 (NO_2, HNO_3)	0.02	0.59
NH_4^+ (N)	0.45	0.18 (NH_3)	0.02	0.65
Cl^-	4.98	0.04	0.17	5.19
Na^+	2.81	—	0.09	2.90
Mg^{2+}	0.33	—	0.01	0.34
Ca^{2+}	0.62	—	0.01	0.63

† Data from Dr D. H. F. Atkins, Atomic Energy Research Establishment, Harwell.
‡ In cloud for 1000 hours per year (11 %), annual average wind speed (10 m) 4 m s^{-1}.

The details of estimated inputs of major ions in precipitation and cloud water, and of the gases SO_2, NO_2, NH_3, and HNO_3 are provided in table 7. The presence of 15 m trees is estimated to increase annual inputs of sulphur and nitrogen from the atmosphere by 30 % and 90 %, respectively (summarized in figure 8). The additional pollutant deposition, either as cloud water or as HNO_3 or NH_3 gases by dry deposition, does therefore justify in part the view that forests are better sinks for pollutants than other vegetation. The additional inputs of Cl^-, Na^+, Ca^{2+} and Mg^{2+} to forests are much smaller than those of nitrogen.

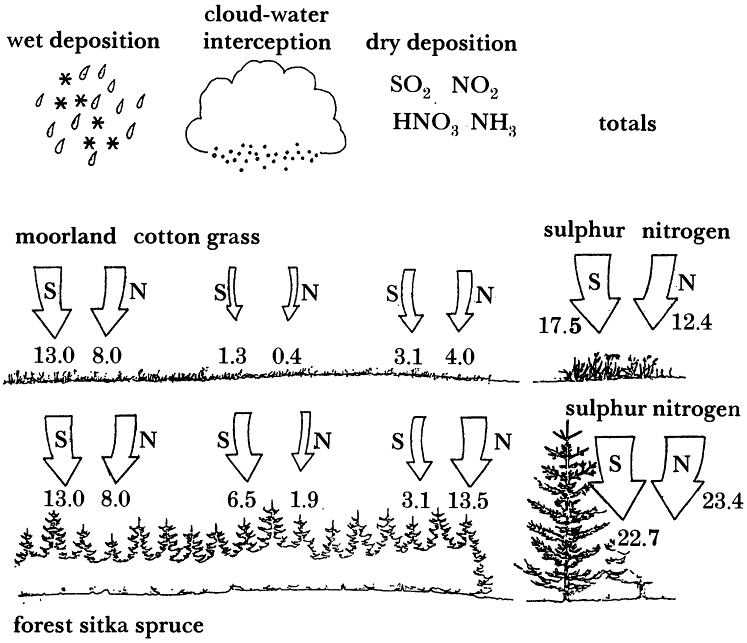

FIGURE 8. Atmospheric inputs at Kielder (6×10^4 ha, 300 m above sea level, 1500 mm rain). Units in kilograms per hectare per year (for details of calculation and assumptions see table 7).

However, the magnitude of the increase in deposition is very site and element specific. Low elevation sites would show little effect of afforestation on sulphur inputs (the effect on sulphur inputs at Kielder results from cloud water deposition), whereas the nitrogen inputs as NH_3 and HNO_3 may be considerably larger.

CONCLUSION

The rates of deposition of pollutant gases, aerosol particles and cloud droplets on forests are shown to differ from shorter vegetation for many of the chemical species. The gases HNO_3, HCl and NH_3 and cloud-water droplets show rates of deposition that are determined by rates of turbulent transfer and yield deposition velocities in the range 2–15 cm s^{-1}. The gases SO_2 and O_3, which are absorbed through stomata, are deposited at much smaller rates, comparable with the deposition rates to short vegetation.

The net effect of afforestation is to increase inputs of nitrogen and acidity, and at sites experiencing cloud water also to increase sulphur and all other elements contained in the cloud water. At moderate elevations (300–500 m) afforestation in western Britain increases the inputs of sulphur and nitrogen by one third and a factor of two, respectively, and is one of the causes of acidification of freshwater draining from afforested catchments.

The modelling of pollutant deposition on forests has reached a stage where valuable estimates of the inputs from the atmosphere may now be made for the nutrients N and S. These estimates provide inputs to catchment chemistry modelling studies and to experiments designed to assess effects of specific pollutants on trees and soils.

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Discussion

H. MAYER (*Lehrstuhl für Bioclimatologie und Angewandte Meteorologie, Universität München, F.R.G.*). Is it possible that forests could themselves be regarded as a source of ozone as a result of terpene emissions?

D. FOWLER. The emission of volatile organic compounds into the atmosphere by forests is regarded by many as a contribution to the organic compounds that take part in the production of photochemical oxidants, including ozone in the troposphere. In that sense therefore forests do contribute indirectly to the production of tropospheric ozone.

J. M. Crowther (*Department of Physics and Applied Physics, University of Strathclyde, U.K.*). In Dr Fowler's paper he has estimated the deposition of sulphur and nitrogen as annual averages. The effects on vegetation are likely to be most severe at high concentrations in rainfall or cloud droplets. Does he know of any work on the effects of high concentration events on forests, specifically linking physiological damage to such events?

D. Fowler. There are several published reports of physiological damage to young trees that have been subjected to applications of simulated acid mist containing H^+, SO_4^{2-}, NO_3^- and NH_4^+ with pHs in the range 2.5–3.5. However, I do not know of any observations of forest damage in the field following a natural episode.

P. G. Jarvis (*Department of Forestry and Natural Resources, University of Edinburgh, U.K.*). In the sulphur and nitrogen budget for Kielder the big increase in nitrogen input appears in the column 'dry deposition'. Would Dr Fowler please explain which of the various gases and particles that he has just described contribute most to this flux?

D. Fowler. The large increase in dry deposition of nitrogen for forests when compared with moorland is caused by the increase of HNO_3 and NH_3 deposition to forests. Rates of NO_2 deposition onto forest and moorland are small and similar. The assumptions underlying the nitrogen and sulphur budget for Kielder are detailed in table 7.

B. Gardiner (*Forestry Commission, Northern Research Station, Roslin, U.K.*). Dr Fowler mentioned that pollutant concentrations in cloud water are found to be largest near the cloud base. He then suggested that this might explain the observation in some European forests that the worst damage occurs at a specific height. Is it really possible to define an average cloud base for a certain location?

D. Fowler. Certainly, the cloud base at a particular site depends on meteorological and surface conditions, but for many upland sites in Britain and on the continent there is a typical altitude for cloud base for the common wind directions, as for example in Cumbria in spring and autumn where it is commonly close to 600 m.

W. J. Shuttleworth (*NERC Institute of Hydrology, Wallingford, U K*). 1. Some years ago, when working on droplet deposition theory (Shuttleworth 1977) I noticed a situation, which might well occur in practice for forests, with droplets efficiently impacting the canopy and the water subsequently re-evaporating. It seems this would be a mechanism giving rise to increased concentrations on forest foliage. Is there evidence that this occurs in practice?

2. Is nitrate deposition a good thing or a bad thing?

D. Fowler. In answer to your first question there is evidence from modelling and field measurements to show increases in the acidity of droplets as they evaporate on leaf surfaces from work by my colleagues at the Institute of Terrestrial Ecology in Midlothian.

On the second question the nitrate may represent a source of mineral nutrition or a damaging pollutant, depending on the plant species, the concentration in solution applied to the foliage, the quantity deposited and the timing of application.

P. G. Jarvis. In relation to the last question, I should like to make the following comment. My colleagues and I have recently measured growth of a pole stage stand of Sitka spruce in relation to interception of photosynthetically active radiation (PAR) (light) over four years in a ± nitrogen, ± phosphorus and ± thinning experiment. In the treatments to which nitrogen was added, there was a 34% increase in the conversion efficiency of PAR, with respect to the other treatments. I believe that the addition of nitrogen at 20–30 kg ha^{-1} per year is having a substantial beneficial effect on the growth of young plantations.