THE GLOBAL EXPOSURE OF FORESTS TO AIR POLLUTANTS

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Abstract: The tall, aerodynamically rough surfaces of forests provide for the efficient exchange of heat and
momentum between terrestrial surfaces and the atmosphere. The same properties of forests also provide for
large potential rates of deposition of pollutant gases, aerosols and cloud droplets. For some reactive pollutant
gases, including SO_2, HNO_3 and NH_3, rates of deposition may be large and substantially larger than onto
shorter vegetation and is the cause of the so called "filtering effect" of forest canopies. Pollutant inputs to
moorland and forest have been compared using measured ambient concentrations from an unpolluted site in
southern Scotland and a more polluted site in south eastern Germany. The inputs of S and N to forest at the
Scottish site exceed moorland by 16% and 31% respectively with inputs of 7.3 kg S ha^{-1} y and 10.6 kg N ha^{-1}
y^{-1}. At the continental site inputs to the forest were 43% and 48% larger than over moorland for S and N
deposition with totals of 53.6 kg S ha^{-1} y^{-1} and 69.5 kg N ha^{-1} y^{-1} respectively.

The inputs of acidity to global forests show that in 1985 most of the areas receiving > 1 kg H$^+$ ha^{-1} y^{-1} as S
are in the temperate latitudes, with 8% of total global forest exceeding this threshold. By 2050, 17% of global
forest will be receiving > 1 kg H^{-1} ha^{-1} as S and most of the increase is in tropical and sub-tropical countries.

Forests throughout the world are also exposed to elevated concentrations of ozone. Taking 60 ppb O_3 as a
concentration likely to be phytotoxic to sensitive forest species, a global model has been used to simulate the
global exposure of forests to potentially phytotoxic O_3 concentrations for the years 1860, 1950, 1970, 1990
and 2100. The model shows no exposure to concentrations in excess of 60 ppb in 1860, and of the 6% of
global forest exposed to concentrations > 60 ppb in 1950, 75% were in temperate latitudes and 25% in the
tropics. By 1990 24% of global forest is exposed to O_3 concentrates > 60 ppb, and this increases to almost
50% of global forest by 2100. While the uncertainty in the future pollution climate of global forest is
considerable, the likely impact of O_3 and acid deposition is even more difficult to assess because of
interactions between these pollutants and substantial changes in ambient CO_2 concentration, N deposition and
climate over the same period, but the effects are unlikely to be beneficial overall.

Keywords: acid deposition, ozone, forests, nitrogen deposition, pollution climate.

1. Introduction

The major regional pollutants associated with acidic deposition (oxidised sulphur and
nitrogen compounds and reduced nitrogen, and the photochemical oxidant (O_3)
represent a threat to a wide range of ecosystems including agricultural crops, wetlands
and heathland vegetation. However, much of the interest in pollutant effects during the
last three decades has been focussed on forests. Examples include the role of air
pollutants in the decline in health of Ponderosa Pine in Southern California (Miller et

al., 1963), the dieback of high elevation forests in the Appalachian mountains of Eastern North America (Eager and Adams, 1992) and the 'new type' of forest decline in Germany and other areas of central Europe (Schultze and Freer-Smith, 1991). More recently, observations of increased rates of growth of European forests have focussed on the role of elevated CO_2 concentrations and the deposition of fixed nitrogen (Cannell *et al.*1997; Spiecker *et al.*, 1996; Thornley *et al.*, 1991), both oxidised (NO_x) and reduced (NH_x).

Why should forests become such a clear focus of interest in these pollution problems? Is it simply a consequence of their abundance as land cover or size relative to other vegetation in the non-agricultural landscape and their importance for amenity, or, are there factors, which predispose forests to the influence, positive or negative of air pollutants?

In this paper, the properties of forests as sinks for the major gaseous and particulate pollutants are described and contrasted with shorter vegetation. In this way one of the major characteristics of forests, their aerodynamic interaction with the atmosphere, is highlighted as an important contributor to their sensitivity to the presence of reactive air pollutants. The current and projected trends in emissions of acidifying pollutant and the presence of photochemical oxidants are then used to quantify the spatial and temporal trends in pollutant exposure globally.

2. Forests and the Atmosphere

The exchange of mass, sensible heat and momentum between vegetation and the atmosphere is effected by turbulent transfer, molecular processes only becoming important very close (<< 1mm) to terrestrial surfaces. The turbulence generated within the surface layers of the atmosphere occurs as a consequence of frictional drag at the earth's surface and forests, among all natural land cover classes, generate the greatest frictional drag at the surface. Forests are therefore aerodynamically rough surfaces with rates of turbulent exchange between the atmosphere and forests larger by an order of magnitude or more than those over grassland.

The rates of exchange of mass, energy and momentum between forests and the atmosphere are discussed in detail by Jarvis *et al.* (1975) and Thom (1975). The extent to which the larger rates of turbulent transfer above forests lead to larger inputs of pollutants to forests than other vegetation is determined by physical and chemical processes at the foliar surfaces, and within the viscous boundary layer close to foliage. Thus the physical and chemical properties of the pollutants interact strongly with the properties of forests to determine rates of exchange.

3. The Pollutants

The pollutants occur as gases, SO_2, NO, NO_2, HNO_3, NH_3, O_3, as aerosol particles mainly in the size range

0.1 to 1.0 μm (diameter)	SO_4^{2-}, NO_3^-, NH_4^+, H^+
0.1 to 0.5 μm (diameter)	heavy metals (e.g. Pb, Cd, Zn, Cu)
1.0 to 3.0 μm (diameter)	base cations Ca^{2+}, Mg^{2+}

and as cloud droplets in the range 3 μm to 20 μm in diameter, containing the major ions (SO_4^{2-}, NO_3^- NH_4^+, H^+, Cl^-, Na^+, Ca^{2+}, Mg^{2+}).

Generalising the properties of forests necessarily conceals the variability between forests of different species composition and climates. The alternative, of describing the aerodynamic properties of a range of tree species and canopy structures, is outside the scope of this paper. Furthermore, the extent of published literature to provide the basis for such an exercise is limited to relatively few species.

TABLE I

Potential effects of forests on deposition fluxes

		Moorland	Forest	% Increase Forest/Moor
Canopy height	h (m)	0.15	10	-
Zero plane displacement	d (m)	0.1	7	-
Roughness length	z_o (m)	0.01	1.0	-
Friction velocity	u_* (m s^{-1})	0.32	0.82	156%
Momentum flux	τ (N m^{-2})	131	840	541%
Maximum deposition velocity for SO_2	V_{max} SO_2 (mm s^{-1})	18.6	35.1	89%
Maximum deposition velocity for NO_2	V_{max} NO_2 (mm s^{-1})	20.0	43.5	118%
Maximum deposition velocity for NH_3	V_{max} NH_3 (mm s^{-1})	21.4	55.5	160%

In describing and contrasting the micrometeorology of temperate and tropical forests Shuttleworth (1989) showed broad similarities in the rates of exchange of water and energy of boreal conifer forest and tropical rainforests. The forests have had to adapt to common problems of limited water supply and very large potential rates of water loss despite the very different architecture of the forest canopies and the climatologies of these regions. The same processes which create the potential for large rates of exchange of momentum, water and sensible heat also influence the deposition of pollutants.

4. Potential deposition rates of pollutants on forests

The extent to which these generalisations apply to the deposition of pollutants on forests varies with the physical and chemical forms of the pollutant. It is therefore necessary

initially to quantify first the potential deposition rates onto forests as a consequence of their aerodynamic properties and contrast these for different canopies and atmospheric conditions. This background may then be used to work through each of the major pollutant categories to show the extent to which the 'scavenging effect' of forests occurs in the field.

To demonstrate the potential rates of deposition of pollutants onto forest relative to short vegetation conventional micrometeorological approaches may be used to calculate momentum and gas fluxes in typical conditions. For simplicity, taking an extensive forest of uniform height in a flat landscape, the wind velocity profile within the constant flux layer may be used to estimate the friction or eddy velocity (u_*) and the momentum flux ($\tau = \rho\, u_*^2$,where ρ is air density) from an assumed wind velocity at a reference height above the surface (Monteith and Unsworth, 1990).

Using this simple approach to contrast the turbulent exchange over a 10 m forest and a moorland (height 0.15 m) for a constant windspeed of 3 ms^{-1} at a reference height of 10 m above the effective surface (d), the eddy velocity over the trees is increased by nearly a factor of 3 and the momentum flux is increased by a factor of 6.4 (Table 1). It is easy to see from this simple example the magnitude of the effects of enhanced turbulence over the trees.

The increased momentum flux for the forest results directly from the increased frictional drag of the canopy, quantified as the drag coefficient (C_m) and given by

$$C_m = [\frac{u_*}{u_{(z)}}]^2 \quad \text{where } u_{(z)} \text{ is the windspeed at height z.}$$

Thus C_m may be calculated directly from a knowledge of u_* and the wind velocity profile used to produce Table I. The drag coefficient is therefore related directly to the roughness length (z_o) within the wind profile equation.

$$u_{(Z)} = \frac{u_*}{k} \ln\, (\frac{z-d}{z_o})$$

In which k is von Karman's constant and d the zero plane displacement (Monteith and Unsworth, 1990).

The direct link between aerodynamic roughness of the canopy and the turbulent transfer of momentum is clear from these simple relationships. The transfer of momentum however, differs from that of trace gases or particles in that momentum transfer occurs both as frictional drag and as form drag, whereas the transfer of gases and particles is restricted to the equivalent of frictional drag, since the gases rely on molecular diffusion at the surface. A convenient method to quantify the reduced rate of transfer of gases relative to momentum is provided within the resistance analogy of mass and energy transfer between the surface and the atmosphere (Thom, 1975). In the analogy the total resistance to transfer of momentum (r_{am}), is provided by ($u_{(Z)}$ / u_*^2) (Fig. 1). For gases and particles the additional transfer resistance through a viscous sub-layer (r_b) completes the transfer pathway from a reference point in the free atmosphere to the external surfaces of vegetation where uptake of the gas or capture of the particle

may occur (Fig. 1). The additional resistance (r_b) for gases and particles may be regarded as a different level within the plant canopy which represents the effective sink for the gas, and since gas transfer is less efficient than momentum, the height of the effective sink for gases will be deeper within the canopy than the sink for momentum. The maximum rates of transfer for the pollutant gases to canopies of vegetation are therefore somewhat smaller than those of momentum, and differ between gases as a consequence of the different molecular diffusivities of the different gases. The individual values for the maximum rate of deposition (v_{max}) for SO_2, NO_2 and NH_3 to a 10m forest and 15cm moorland in Table I differ as a consequence of differences in r_b.

The deposition velocity V_d is simply the reciprocal of the total resistance in the deposition pathway (r_t) i.e. $r_t = V_d^{-1}$. In the case of V_{max}, the maximum rate of deposition is simply the condition in which all molecules transported to the surface are absorbed (or react). In practice, the gas may not react readily at the surface and the extent to which a gas is not removed from the atmosphere by contact with the surface may be expressed as a residual, or surface resistance. The maximum rates of deposition onto a forest, in these conditions, vary between 89% and 160% larger than their maximum rates of deposition onto moorland under the same atmospheric conditions. The largest absolute value of V_{max} and largest forest-moorland difference are for NH_3 the pollutant in this group with the largest molecular diffusivity.
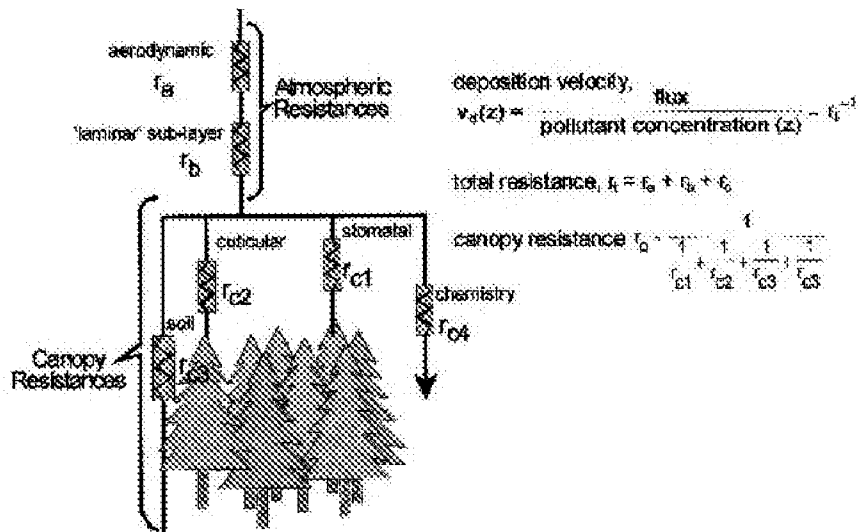

Fig. 1 A simple resistance analogy to simulate the partitioning of pollutant fluxes between the principle sinks in a forest canopy

The bulk of the published deposition velocity data for these gases show the presence of a surface resistance, although the magnitude varies according with surface and atmospheric conditions. The potential enhancement of pollutant deposition by forests is

only realised for circumstances in which atmospheric transfer (i.e. $r_a + r_b$ in Figure 1) significantly influences the magnitude of r_t (and hence V_d).

The overall deposition process is subject to controls by rates of turbulent diffusion above and within the vegetation and to chemical reaction with a range of surfaces, including epicuticular wax, leaf surface water, senescent vegetation and soil. While rates of turbulent transfer and aerodynamic resistances can be estimated directly from a knowledge of roughness characteristics of the surface and wind velocity profiles, the canopy resistance cannot in general be quantified from first principles or theory and requires empirical values from field or laboratory measurement.

5. Measurements of deposition fluxes and quantifying canopy resistance

The measurement of wet deposition fluxes in the field is straightforward and networks of wet deposition samplers provide the basis for wet deposition maps of Europe and North America and parts of Asia. The measurements are more difficult in complex terrain and especially in mountainous regions. However, even in these regions extensive wet deposition measurements have been made (Dore et al. 1990; Mohnen, 1992).

By contrast, the measurement of dry deposition to vegetation requires instrumentation to determine either the co-variance in short term variability in concentration χ^1 and the vertical component of turbulence w^1 to provide the flux (F_s) as

$$F_s = \rho w^1 \chi^1$$

Alternatively, fluxes may be measured by aerodynamic flux gradient or Bowen ratio methods. The detailed methodology lies outside the scope of the paper and is described by Woodward and Sheehy (1983) and Fowler and Duyzer (1989) while the micrometeorological background for the work is provided by Monteith and Unsworth (1990).

These measurement methods have provided a reasonable understanding of the main sinks for the pollutant gases in forest canopies (Erisman and Draaijers, 1995; Fowler et al., 1989; Duyzer et al., 1992) and in shorter vegetation (Erisman & Wyers, 1993; Flechard and Fowler, 1998).

5.1 SO₂ DEPOSITION

The primary control of SO_2 deposition occurs at the surface, with canopy resistances generally exceeding aerodynamic resistance for the majority of the time. Typical aerodynamic resistances (rates) being between 10 s m^{-1} and 20 s m^{-1} for conifer forests (Jarvis et al, 1975) and 30 s m^{-1} to 50 s m^{-1} for cereals (Monteith and Unsworth, 1990), whereas the canopy resistances for SO_2 deposition range from 10 s m^{-1} to 500 s m^{-1} (Erisman and Draaijers, 1995; Fowler and Unsworth, 1979) with median values in excess of 100 s m^{-1}. The large range of canopy resistances arises as a consequence of a combination of different sinks for SO_2 within vegetation. For entirely dry canopies the epicuticular waxes on foliage represent a relatively weak sink for SO_2 with canopy resistances of 500 s m^{-1}. The stomata represent efficient sites of SO_2 uptake, limited only by the diffusive resistance to transport to the apoplast fluids within sub-stomatal cavities. Making allowance for the relative molecular diffusivities of SO_2 and H_2O, canopy conductances for H_2O may be used to calibrate the magnitude of stomatal uptake and the canopy resistance with which it is associated. Thus the minimum values

are typically 100 s m^{-1} for the stomatal component of the canopy resistance to SO_2 uptake by cereals and grasses. For forests, which are characterised by larger canopy resistances for evapotranspiration, the work of Jarvis et al (1975) can be used to estimate the equivalent values of r_{c1} for SO_2 deposition onto forests, of 150 s m^{-1} to 200 s m^{-1} for conifers in the absence of water stress. In practice, the two components of canopy resistance, for stomatal uptake and for uptake onto the external surfaces (r_{c2}) (Fig. 1) operate in parallel, and thus provide the total canopy resistance (r_c) as

$$rc^{-1} = r_{c1}^{-1} + r_{c2}^{-1}$$

This is the simplest representation of the processes within the canopy in which many additional sinks exist (e.g. soil, dead or senescent plant material). Furthermore, the canopy structure is far from uniform and may be better described following sub-division of the canopy into a series of horizontal layers. The more recent work on SO_2 from long-term monitoring of deposition fluxes shows that the overall canopy resistance is strongly linked to the chemistry in surface layers of water on the vegetation (Erisman et al, 1998a; Flechard and Fowler, 1998). The measurements show that the canopy resistance for SO_2 is regulated by the supply of NH_3, which neutralises the acidity formed by SO_2 oxidation to SO_4^{2-} (Flechard and Fowler, 1999). Thus the local chemical climate of the atmosphere regulates the composition of surface water on vegetation and thus the canopy resistance for SO_2. Sufficient data have been accumulated within the recent long-term flux measurement studies to parameterise deposition models in which the interaction of pollutants in regulating deposition rates is simulated.

5. 2 NO₂ DEPOSITION

The uptake of NO_2 by a variety of tree species has been shown in laboratory conditions to be almost exclusively by stomata (Hanson and Lindberg, 1991). For a grassland canopy, Hargreaves et al. (1992) showed in field conditions that NO_2 uptake is limited to stomata. Such findings greatly simplify the deposition of NO_2 to vegetation, and reduce the differences between forests and other surfaces, since for the aerodynamically rough surfaces ($z_O > 1$ m) bulk stomatal resistances are generally larger than for short vegetation ($z_O < 0.2$ m). A complication arises in the measurement of NO_2 uptake by forests in particular since soil emissions of NO, which does not deposit at significant rates, are readily oxidised within the canopy to NO_2 by O_3 (Duyzer et al., 1995). The NO_2 may then be absorbed within the canopy by stomata, and such an internal cycle for the soil-plant represents no net uptake by the canopy of vegetation (Fowler et al., 1998a).

5.3 NH₃ DEPOSITION

The vegetation-atmosphere exchange of NH_3 is complicated by the presence of NH_4^+ within apoplast fluids of the leaf mesophyll. Thus with open stomata the net flow of NH_3 between the sub-stomatal cavity and the atmosphere is regulated by apoplast pH and $[NH_4^+]$ and the ambient concentration of NH_3 within the plant canopy (Sutton et al., 1995). The process, illustrated schematically in Fig. 2 has been simulated in a simple canopy compensation point model by Sutton et al. (1993c).
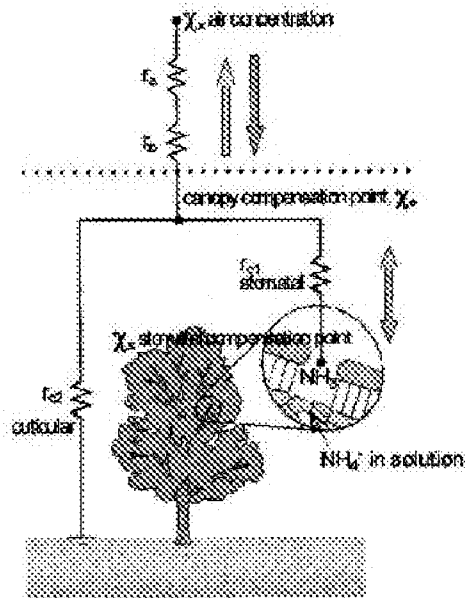
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Fig. 2 A schematic representation of the canopy compensation point model to simulate emission and deposition fluxes of NH_3

Field measurements of NH_3 exchange are therefore influenced by reactions with the leaf surface chemical processes within liquid films on foliage which also SO_2 deposition and by the physiological controls of NH_4 in the apoplast. In practice, the canopies with the greatest potential for emission fluxes of NH_3 are the heavily fertilised agricultural canopies and especially grassland which in intensive production may receive in excess of 300 kg N ha^{-1} annually in fertiliser applications. Forests in contrast, generally receive much smaller quantities of nitrogen, except in the vicinity of large agricultural sources of NH_3, such as livestock farms. In long-term flux measurements of NH_3 over semi-natural (moorland) vegetation Flechard and Fowler (1998) showed a median canopy resistance of 37 s m^{-1}, providing deposition velocities typically in the range 10 mm s^{-1} to 20 mm s^{-1}, and net emission fluxes of NH_3 were observed only 6% of the time during 13 months of continuous measurement. Similarly, Sutton et al. (1993a, b) showed large rates of NH_3 deposition onto acidic moorland vegetation.

5.4 CONTRASTING POLLUTANT DEPOSITION ON FORESTS WITH MOORLAND
While far from comprehensive in land surface cover, these extensive measurements provide the necessary data to parameterise a simple model with canopy resistances for a forest and moorland (or other semi-natural short vegetation canopy). This allows the effect of forest on pollution fluxes to be quantified, given the same ambient pollution climate and an identical meteorological and similar surface conditions.

TABLE II a

Effect of forests on pollutant fluxes: a remote relatively unpolluted site (S. Scotland)

Pollutant Concentrations (μg m^{-3})				Dry Deposition of Gases		
SO$_2$	NO$_2$	NH$_3$		SO$_2$	NO$_2$	NH$_3$
2.4	6.0	0.9		kg S ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}
Canopy resistance R_c (s m^{-1})			Moorland	1.9	0.6	2.7
			Forest	2.1	0.6	4.2
SO$_2$	NO$_2$	NH$_3$				
150	1000	37				
				Dry Deposition of Aerosols		
Wet deposition fluxes (kg ha^{-1} a$^{-1)}$						
SO$_4^{-S}$	NO$_3^{-N}$	NH$_4^{-N}$		SO$_4^{-S}$	NO$_3^{-N}$	NH$_4^{-N}$
4.1	1.9	2.4		kg S ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}
			Moorland	0.3	0.2	0.3
			Forest	1.0	0.6	1.0
					0.6	1.0

TABLE IIba

Effect of forests on pollutant fluxes: a polluted site in Central Europe

Pollutant Concentrations (μg m^{-3})				Dry Deposition of Gases		
SO$_2$	NO$_2$	NH$_3$		SO$_2$	NO$_2$	NH$_3$
13.8	19.8	7.3		kg S ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}
(5 ppb)	(10 ppb)	(10ppb)				
Canopy resistance R_c (s m^{-1})			Moorland	25.3	1.8	22.6
			Forest	35.9	1.9	34.5
SO$_2$	NO$_2$	NH$_3$				
32	1000	37				
Wet deposition fluxes (kg ha^{-1} a^{-1})				Dry Deposition of Aerosols		
SO$_4^{-S}$	NO$_3^{-N}$	NH$_4^{-N}$		SO$_4^{-S}$	NO$_3^{-N}$	NH$_4^{-N}$
9.3	5.2	11.3		kg S ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}	kg N ha^{-1} a^{-1}
			Moorland	3.0	4.1	1.8
			Forest	8.4	11.7	4.9
					0.6	1.0

The concentration of the pollutants SO_2, NO_2 and NH_3, the aerosol concentrations of SO_4^{2-}, NO_3^-, NH_4^+, the canopy resistance values for each and the wet deposition fluxes were all obtained from direct measurements at Auchencorth Moss (Fowler *et al.*, 1996,1998b) and summarised in Table IIa and b. Assuming a constant wind speed of 5 ms^{-1} at a height of 10 m (above d + z_0), a thermally neutral atmosphere and roughness lengths (z_0) of 0.01 and 1.0 m respectively for moorland and forest the annual deposition fluxes were calculated as shown in Table IIa.

The annual input fluxes of sulphur and nitrogen of 6.3 kg. S and 8.1 kg. N (Table III) to moorland and forest are small but are typical of the upland areas in the north and west of the UK (RGAR, 1997). The effect of increasing the aerodynamic roughness of the surface may be seen in the comparison for this relatively unpolluted site between moorland and forest. The change in aerodynamic roughness increases the sulphur and nitrogen deposition by 16% and 31% respectively, the larger increase in N deposition being largely a consequence of NH_3 deposition.

The influence of the pollution climate on both the total input and the importance of the aerodynamic effects of the forest are quantified by running the same procedure to calculate annual inputs for data from a more polluted location. In this case the pollutant data were provided by measurements from Melpitz, a pollution flux monitoring station close to the city of Leipzig (Spindler *et al.*, 1996). The canopy resistances used were identical to those in the previous example to allow the interaction of pollutant concentration and forest moorland to be compared without the complication of different canopy resistances. The input data and output fluxes are summarised in Table IIb with a summary of the annual S and N inputs for both sites and canopies in Table III. The annual inputs of sulphur at the polluted site range from 37 kg S ha^{-1} for m moorland to 54 kg S ha^{-1} for forest, while nitrogen inputs range from 47 kg N ha^{-1} for moorland to 70 kg N ha^{-1} for the forest. These values are all large for the polluted site and the effect of the forest in enhancing the deposition is typically + 50%.

TABLE III

Effect of forests on pollutant fluxes: annual deposition fluxes of S and N at contrasting sites

	Unpolluted area (S. Scotland)		Polluted area (central Europe)	
	S	**N**	**S**	**N**
	kg S ha^{-1} a^{-1}	kg S ha^{-1} a^{-1}	kg S ha^{-1} a^{-1}	kg S ha^{-1} a^{-1}
Moorland	6.3	8.1	37.6	46.8
Forest	7.3	10.6	53.6	69.5
% increase Forests/Moor	16%	31%	43%	48%

Total Wet + Dry Deposition

The magnitude of the effect of forests in enhancing pollutant deposition is therefore very variable, but increases rapidly for the soluble reactive pollutants SO_2 and NH_3 (and HNO_3) with the ambient concentration of these gases. These calculations contrast two lowland locations, one in the windy, relatively unpolluted environment of southern

Scotland and one in the more continental and polluted climate of central Europe. The exercise shows the magnitude of the forest effect on these gases and aerosols and demonstrates that, given adequate monitoring data for the pollutant gases and meteorological variables, the calculation of both total inputs and effects of different land cover is straightforward. Very few sites in rural areas of Europe provide the necessary data for these flux measurements.

6. Aerosols and cloud droplets

The processes which lead to the enhancement in deposition by forest relative to other terrestrial surfaces combine the general aerodynamic properties of the forest with surface processes which regulate the rate of uptake. The aerosols and cloud droplets containing pollutants are present over a broad size range from a few nm in diameter to 20 μm in diameter. Over this size range the control over deposition rates changes from processes regulating the atmospheric transfer for the larger particles to processes controlling the capture processes at the surface for the smaller particles (Beswick et al. 1991).

The large particle sizes (> 10 μm), and especially large cloud droplets, have significant rates of gravitational settling. At 20 μm droplet radius, unit density spheres sediment at about 10 mm s^{-1}, whereas for all particles smaller than 5 μm the gravitational settling velocity is less than 1 mm s^{-1} and can generally be neglected. The rates of turbulent diffusion above forests provide upper limits for deposition rates in the range 30 mm s^{-1} to 50 mm s^{-1} for the gases (Table I) and also particle deposition. The extent to which these large rates apply to the particles is determined by the transport of particles through the viscous sub-layer of air close to surfaces and to the capture efficiency of the surfaces.

The conventional particle deposition theory, largely based on wind tunnel studies, shows efficient deposition of particles and droplets larger than 10 μm diameter onto vegetation (Chamberlain, 1975). For particles smaller than about 0.1 μm diameter, transport through the viscous sub-layer by Brownian diffusion provides significant deposition rates and for particles larger than 5 μm diameter interception and impaction become increasingly efficient. However, for particles in the size range 0.1 μm diameter to 5 μm diameter, the wind tunnel measurements show very inefficient transport and capture of particles on vegetation. The relationship between particle deposition and particle size from the work of Chamberlain (1975) and others is illustrated in Figure III. Also illustrated in Figure III are results of several recent field experiments summarised by Gallagher et al. (1997). This analysis shows much larger rates of deposition of sub-micron aerosols then earlier work and for which there is no underlying theoretical framework. However, the extent of the direct experimental support for larger rates of sub-micron aerosol deposition on forest is now sufficient to be confident that for these surfaces at least, aerosol deposition rates are substantially larger than the 0.1 mm s^{-1} to 1.0 mm s^{-1} indicated by wind tunnel studies. The potential mechanisms available to provide a mechanistic basis for the observations include electrophoresis and other phoretic mechanisms (Goldsmith et al., 1963). Somehow, the small particles are transported through the viscous sub-layer at rates which exceed rates of Brownian diffusion and for particle sizes which have insufficient inertia for efficient impaction.

However, to date the no unifying theory has been advanced to explain the observed rates of deposition of particles in the size range 0.1 μm diameter to 1.0 μm diameter. For this exercise, and based on the data provided in Figure III, a deposition velocity of 5 mm s^{-1} has been assumed for particles in the size range 0.1 μm diameter to 2.0 μm diameter (i.e. including all the aerosols SO_4^{2-}, NO_3^- and NH_4^+).
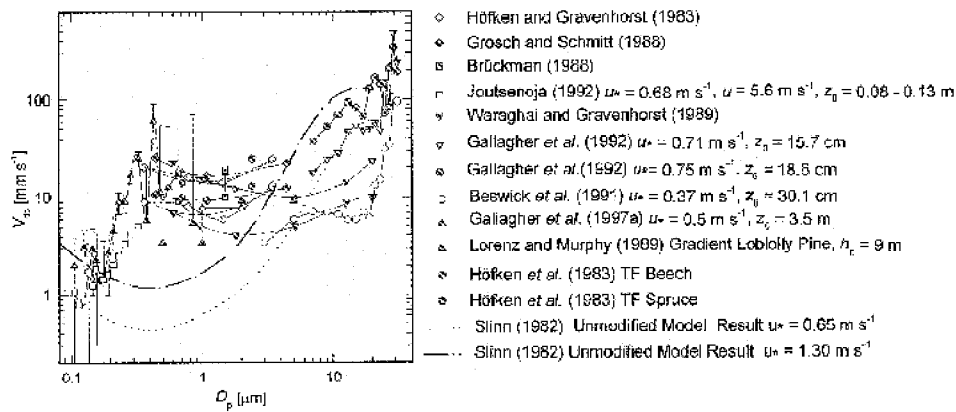

Fig. 3   The relationship between particle diameter and deposition velocity from field measurements compared with the model of Slinn (1982.) (Adapted from Gallagher et al. 1997).

To examine the effect of different canopy heights on the long-term average inputs of aerosols and cloud droplets the inventory of ^{210}Pb in soil has been very valuable (Fowler et al., 1998d). The method relies on the deposition of radioactive ^{210}Pb containing aerosols and cloud droplets onto vegetation, and the capture and retention of the ^{210}Pb by organic matter in soils. The inventory of [210]Pb (I_s) then undergoes radioactive decay with a half life (t $^1/_2$) of approximately 22 years and average lifetime (λ = ln2/t $^{1/2}$), so that the long-term annual deposition flux (F_D) is provided by

$$F_D = \frac{I_s}{\lambda}$$

The data in Figure IV from Fowler et al. (1998d) shows the increase in the ^{210}Pb inventory beneath a forest canopy relative to those beneath a nearby grass canopy in the Scottish Borders. The increase in the inventory occurs largely as a consequence of a greater cloud droplet capture by the trees than by grassland. Measurements of cloud

frequency and windspeed at the site allow independent estimates of cloud droplet deposition to be made for the site and these are in good agreement with the observed increase (~ 40%) in the ^{210}Pb inventory at the site (Fowler et al., 1998d).
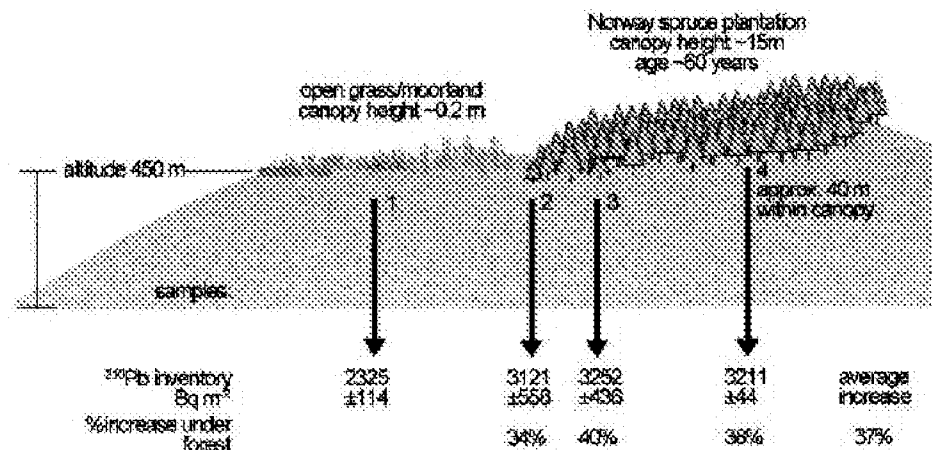

Fig. 4. The soil inventories of ^{210}Pb under moorland and conifer forest, showing the effect of enhanced capture of cloud droplets and aerosols by the forest canopy (from Fowler et al 1998d)

Overall, the 'filtering effect' of forest canopies for pollutants, relative to shorter vegetation, is therefore supported by current understanding of the deposition processes. It is clear however, that the degree of enhancement is highly variable, depending on chemical interaction at foliar surfaces for the gases and particle size for particle deposition, as well as the meteorological and morphological characteristics of individual sites which regulate turbulent exchange between vegetation and the atmosphere.

7. Current and future global patterns of acidic deposition

The period of the 1970s and 1980s provided a series of research programmes investigating effects of acid deposition on forests (e.g. Heij et al., 1991; Eagar and Adams, 1992). The health of forests became an important issue in Europe and North America in particular; during this period air pollutants were believed to be responsible, at least in part, for areas of pronounced decline. No single causal mechanism was found to be responsible for problems of forest decline in the high elevation red spruce forests of the Appalachians of Eastern North America (Johnson, 1987), the fir and spruce decline in Germany and parts of France, in spruce and fir forests of the Czech Republic and Poland and in pine in the South East United States and California.

However, acid deposition and sulphur in particular was shown to be the cause of marked reductions in frost hardiness of red spruce (DeHayes, 1992; Cape et al., 1991; Sheppard et al., 1991). Acid deposition was also shown to be the major contributor to

acidification of soils and along with the direct of SO_2 was the cause of widespread decline in high elevation conifer stands in the Czech Republic and Poland.

The control measures introduced in Europe and North America were in part influenced by these findings. Peak sulphur emissions in Europe during the late 1970s were followed by a monotonic decline from approximately 29 Tg Sy^{-1} in 1980 to 15 Tg S in 1997, a 48% reduction. In North America the change has been somewhat smaller from 16 TG S y^{-1} to 12 Tg y^{-1} over the same period (Table IV). Sulphur has been the clear focus for political action because it was perceived as the major contributor to acidic deposition and the major cause of terrestrial effects on freshwaters and forests. The marked reductions in S emissions represent the major results of regional pollution control, and also a considerable financial investment in abatement technology throughout Europe and North America. Elsewhere in the world, and especially in the rapidly developing economies of south and east Asia as well as Africa, South and Central America, emissions of all pollutants, including S have been increasing rapidly. In S.E. Asia in particular the increase in sulphur emissions throughout the last decade has been very rapid and already this region of the world emits more sulphur dioxide into the atmosphere than Europe or North America (Galloway and Rodhe, 1991).

The sensitivity of soils and ecosystems to acidification has now been mapped at global scales by the Stockholm Environment Institute (Foell et al. 1995). This analysis identifies large areas within the tropics and sub-tropics in which soils are very sensitive to acidification (Rodhe and Herrera, 1988).

TABLE IV

Emissions of sulphur in Europe and N America in 1980 and 1997

	1980	1997	
Europe	29 Tg	15 Tg	-48%
N. America	16 Tg	12 Tg	-25%

The global inventory of sulphur emissions for 1985 and the projected emissions for the year 2050 have been used to model the global deposition of sulphur, using the Moguntia model (Crutzen et al., 1997) based on the emission projections IS92a (IPCC 1995). The results of the model are shown in Figure V, which reveal large areas of Asia, Africa and Central America with increased sulphur deposition. To identify the consequence of the sulphur deposition scenarios for forests throughout the world, land cover from Matthews (1983) has been used to identify the areas of forest in two broad categories, temperate/sub polar and tropical/sub-tropical (Figure VI). The modelled sulphur deposition may then be used to calculate the areas of forest receiving atmospheric sulphur input in excess of 1keq ha^{-1} annually for 1985 and 2050 (Figure VII). Making the assumption that at an annual sulphur deposition in excess of 1 keq S ha^{-1} the forest is at risk on acid sensitive soils, the areas of potential forest damage may be calculated. The value of 1 keq S ha^{-1} represents a much lower sensitivity than the most sensitive soil (0.25 keq S ha^{-1}) applied by Fowler et al (1995) and applies a mid-point of the sensitivity scale applied (0.25 to 2.5 keq S ha^{-1}). The exercise, despite its simplifying assumptions, reveals an increase in the area of exceedance by a factor of

2.14 for all forest over the 65 year period (Table Va). If, however, the threshold for damage is taken as 2 kg S ha^{-1} then overall the risk to forests increases by a factor of 7.2, with a 5 fold increase in temperate forests and an increase from zero to 2.3 x 10^6 km^2 of tropical forest at risk (Table Vb).
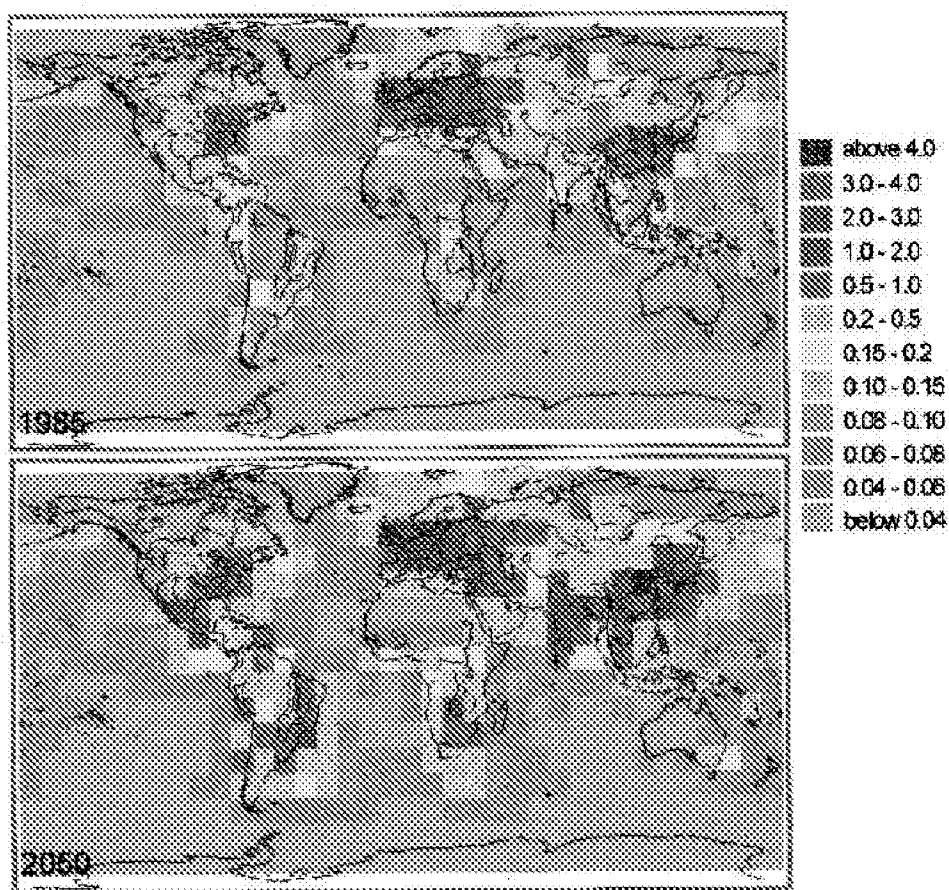

Fig. 5 Total deposition of sulphur at 10^0 x10^0 (keq H$^+$ -S ha^{-1} yr^{-1}) from the MOGUNTIA model based on the IPCC IS92a scenario (data provided by the Stockholm Environment Institute).
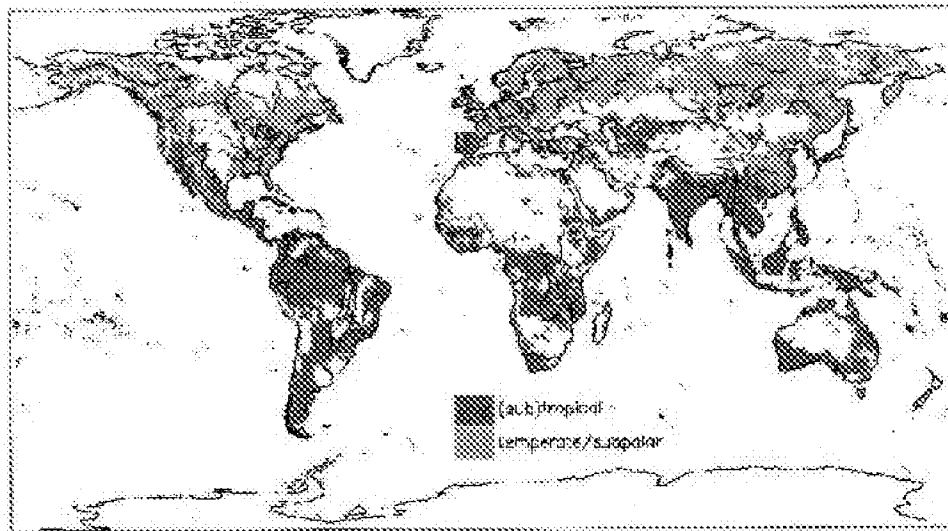
Fig. 6 The global distribution of temperate and tropical (+sub-tropical) forest cover derived from the work of Mathews (1983).

TABLE V a

Assuming forest at risk is that receiving > 1 keq H^+ ha^{-1} as S

	Potential area of exceedance		
	all forests	tropical + subtropical	temperate + subpolar
1985	$2.77 \times 10^6 \text{ km}^2$	$0.02 \times 10^6 \text{ km}^2$	$2.75 \times 10^6 \text{ km}^2$
%	8	0.2	15
2050	$5.9 \times 10^6 \text{ km}^2$	$2.3 \times 10^6 \text{ km}^2$	$3.6 \times 10^6 \text{ km}^2$
%	17	15	20
% change 1985 to 2050	114	9200	32

TABLE V b

Assuming forest at risk is that receiving > 2 keq $H^+ ha^{-1}$ as S

	Potential area of exceedance		
	all forests	tropical + subtropical	temperate + subpolar
1985	$0.28 \times 10^6 km^2$	0	$0.28 \times 10^6 km^2$
%	0.8	0	1.5
2050	$5.9 \times 10^6 km^2$	$2.3 \times 10^6 km^2$	$3.6 \times 10^6 km^2$
%	6.0	4.4	7.3
% change 1985 to 2050	624	-	376

The majority of the increase is however in tropical and sub-tropical countries in which the increase in areas of potential exceedance increases by almost two orders of magnitude. By contrast, the temperate and sub-polar forests change by a modest +32% relative to 1985. There are important uncertainties in this simplistic exercise; however, it is clear that the majority of forest acidification effects will be in developing regions of the world, in the tropics and sub-tropics in particular. There is also a clear requirement for improved understanding of processes, and monitoring of pollution climates and their effects on forests in these regions.

8. Eutrophication, nitrogen deposition and forests

The focus on sulphur emissions and deposition and its role in acidification throughout the 1970s and 1980s distracted attention from the other major contributors to acidification, namely NO_y and NH_x. However, in the Scandinavian research of the 1970s it was recognised that the four dominant ions of anthropogenic origin in wet deposition were SO_4^{2-}, NO_3^-, NH_4^+ and H^+. The NO_3^- and NH_4^+ ions are both potentially acidifying depending on their biological and chemical processing within soil and vegetation (Sutton et al., 1993c). Throughout the last four decades, the deposition of nitrogen compounds has contributed a substantial fraction of the acidifying deposition. Furthermore, as sulphur emissions have declined in Europe and North America, gradually the contribution of nitrogen to deposited acidity has increased. In many parts of Europe and North America the atmospheric inputs of NO_y and NH_3 already contribute the dominant fraction of acidifying deposition.

The control measures developed to limit sulphur dioxide emissions from large combustion plant have not been accompanied by similarly effective controls on NO_x
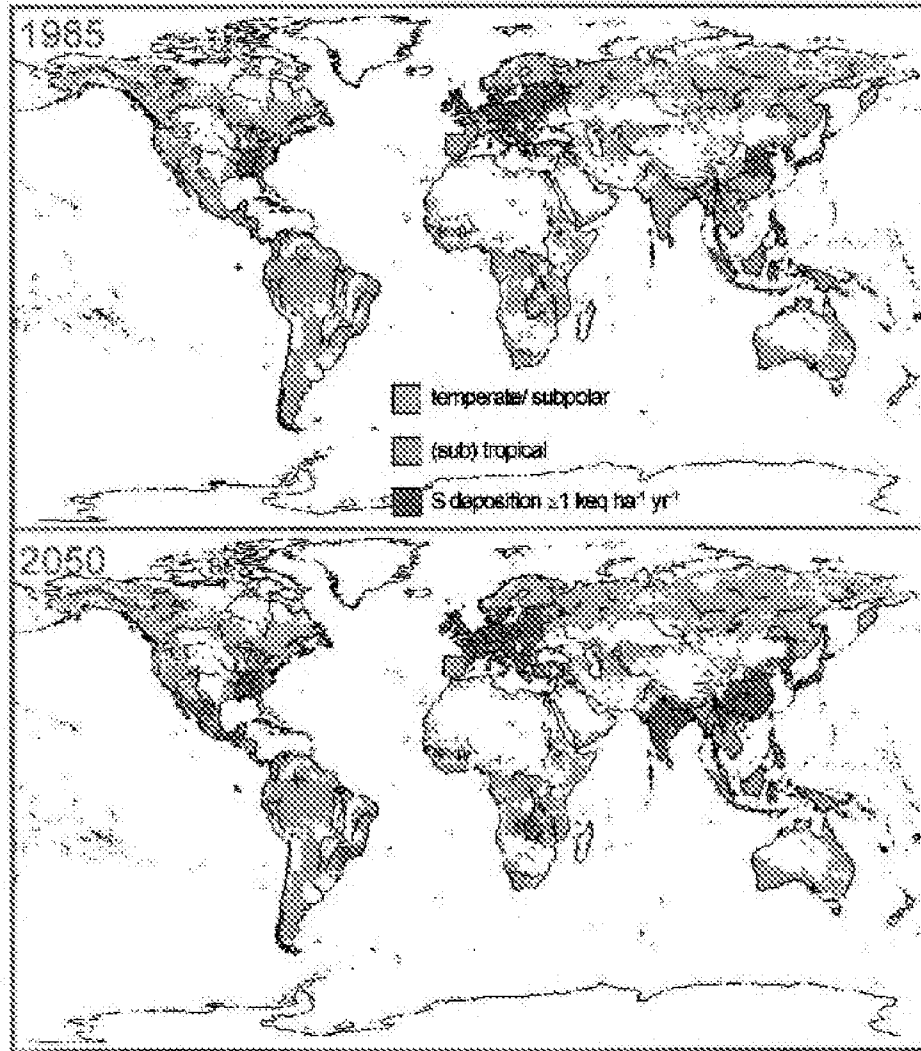
Fig. 7 The global distribution of forest cover on which total sulphur deposition exceeds 1 keq ha^{-1} yr^{-1}, for 1985 and 2050

    emissions, and there are currently very few effective controls on emissions of ammonia (NH$_3$ other than reductions in livestock numbers or intensity of production (Lekkerkirk, 1998). The greatest concern over NH$_3$ emissions has been in the Netherlands, where NH$_3$ deposition provides most of the acidifying input to terrestrial surfaces throughout the country and to forests in particular (Erisman and Draaijers, 1995). The introduction of control measures has not resulted in the expected reductions in NH$_3$ concentrations or deposition (Erisman et al., 1998b). Current understanding of the effects of different control techniques on the net exchange of NH$_3$ between soil-vegetation and the atmosphere has therefore to be significantly improved before

effective controls on NH_3 emissions can be implemented. It seems likely therefore that the deposition of nitrogen compounds will increasingly dominate the acidifying atmospheric inputs to forests throughout Europe and North America over the next decade.

Atmospheric inputs of fixed nitrogen to forests have also attracted recent interest as a potential cause of increased forest growth over a wide geographical scale in Europe (Speicker et al., 1996). Other potential causes of the increased forest growth include CO_2 fertilisation as a consequence of the increase in atmospheric CO_2 concentration which between 1960 and 1998 changed from 320 ppm to 365 ppm (Neftel et al., 1985). The current interest in nitrogen deposited onto forests therefore includes both of the major regional and global environmental problems, acidification and global warming.

Critical loads for nitrogen inputs to forests have been developed from mass balance and empirical approaches (Hornung et al., 1995, Kuylenstierna et al, 1999). While there is still considerable uncertainty in the values selected, both approaches include values in the range of 15 to 20 kg N ha^{-1} y^{-1} for the most sensitive forest ecosystems.

In considering the current interest in deposition of fixed nitrogen onto forests, it is useful to draw a distinction between the regional increases in deposition that have taken place over the last 100 years and local deposition of nitrogen close to point sources in which inputs may exceed 50 kg N ha^{-1} annually. The regional increases from 2 to 5 kg N ha^{-1} to 10 to 20 kg N ha^{-1} y^{-1} have occurred over substantial areas of the industrialised regions of Europe and the USA. Taking the UK as an example, the annual deposition of fixed nitrogen as NO_y and NH_x, is approximately 385 kt N (Fowler et al., 1998e), which averages the deposition to terrestrial surfaces in the UK at about 16 kg N ha^{-1} with a range from 5 kg N ha^{-1} to 40 kg N ha^{-1} (Goulding et al., 1998).

The terrestrial effects of this scale of increase in nitrogen input include increased emissions of the radiatively active N_2O to the atmosphere and also enhanced emissions of NO (Skiba et al., 1994). However, the magnitude of these increased emissions is small relative to the potential for terrestrial sequestration of CO_2 from the atmosphere by increased forest growth as a consequence of increased N deposition.

The average increase in regional deposition of nitrogen conceals 'hot spots' which result from proximity to large sources. The dry deposition rate of NH_3 is substantially larger than that for SO_2 and this, along with the ground level sources of NH_3 exaggerates the spatial variability in N deposition relative to that in S (Fowler et al. 1998c). These hot spots include the areas close to intensive livestock production units (NH_3), hill tops on which cloud droplet deposition may contribute substantial inputs and urban areas with large NO_2 concentrations. At such sites, the local N input from the atmosphere may exceed 50 kg N ha^{-1} (Fowler et al., 1998c). The dispersion of NH_3 from ground level sources and the typical size of livestock units leads to the individual areas of high deposition close to each farm being small. Taking as an example a poultry farm surrounded by woodland, the woodland between 15m and 50m downward of the source experienced a mean NH_3 concentration of 29 μgm^{-3} and received approximately 42 kg NH_3^- N ha^{-1} annually from the source. At a distance of 150m from the source the ambient NH_3 concentration was 4 μg NH_3 m^{-3} and the dry deposition was 8 kg NH_3^- N ha^{-1} annually. Thus the broad regional maps smooth the variability due to landscape features and conceal the presence of hot spots of deposition close to local sources.

9. Photochemical Oxidants

Among the pollutants recognised as potential causes of forest decline in Europe and North America, O_3 was identified as a probable contributor (McLaughlin and Kohut, 1992). From field studies however, in most of the areas in which clear evidence of die-back of forests has been observed, there are several possible causes. In the case of high elevation conifers, these are subjected to prolonged exposure to cloud water containing large concentrations of the major ions SO_4^{2-} NO_3^- NH_4^+ and H^+ (Mohnen, 1992). At some forest locations, the bulk of the sulphur and nitrogen is deposited as cloud droplets, with concentrations of the major ions between 5 and 7 times those in precipitation at the same site (Crossley et al., 1992). The same high elevation locations experience prolonged exposure to elevated ozone largely because at these altitudes the development of a stable nocturnal boundary layer is suppressed by the larger wind speeds (PORG, 1997). However, laboratory and open-top chamber studies in which ozone has been introduced in controlled experiments show clear effects on the physiology and growth of many of the common tree species of temperate forests (Skarby et al., 1998; Chappelka and Samuelson, 1998). The threshold concentration above which ozone may influence physiological processes in trees is not well defined. In practice, a cut-off at 40 ppb has been widely adopted in assessing the potential for ozone damage to both field crops and trees (Karenlampi and Skarby, 1996) even though there may well be important effects at smaller concentrations. The use of the arbitrary cut-off of 40 ppb has also been helpful in developing 'dose'-effect relationships in which biomass yield, especially of cereal crops, has been linearly related to the accumulated exposure to concentrations in excess of 40ppb, the AOT_{40} concept (Fuhrer et al., 1997). However, the threshold is arbitrary and, in practice, there may well be physiologically important effects at concentrations significantly smaller than 40 ppb.

The threshold of 40 ppb is however very close to ambient background concentrations at many exposed locations in mid-northern latitudes during the Spring and Summer months. To contrast the global distribution of ozone concentrations which represent a threat to forest productivity and health, it is helpful to work at higher concentrations. At 60 ppb, surface ozone concentrations are both significantly elevated above the zonal background concentration and are also sufficiently large to represent a potential threat to the productivity of many forests.

Networks of ozone monitoring stations have been established in many of the countries of Europe as well as in the USA, Canada and Japan. These direct measurements are restricted mainly to the last decade and are therefore not adequate to provide a chronology of O_3 concentration over the last century, during which the concentrations are believed to have increased and are of course limited to a small area of the global forests. An alternative method of assessing the global exposure of forests to potentially changing O_3 concentrations is to simulate the photochemical production of ozone within a global model containing the emission inventories for the ozone precursors, the fluid dynamics which transport the pollutants and the processes that remove soluble, and reactive species through wet and dry deposition. Such models have made great progress in recent years in simulating the concentrations and their seasonal and changes (Collins et al., 1997, Wang et al, 1998).

The model used for this paper is a 3-D chemistry-transport model (CTM) run off line from archived meteorological data generated by the UKMO unified model at climate

resolution (3.75° longitude x 2.5° latitude with 19 vertical levels). The archived meteorological information provides winds, pressures, temperatures, humidity, tropopause height, cloud and precipitation and boundary layer and surface properties. The model (STOCHEM) described by Collins et al. (1997) used a Lagrangian method in which the atmosphere comprises 50,000 air parcels in which the chemical processing of pollutants is simulated. The model uses 70 chemical species and 174 reactions. It has been used to simulate the atmospheric transport, and chemistry of the pollutant emissions using 5 scenarios for 1860, 1950, 1970, 1990 and 2100 each for a period of 15 months from which the first 3 months were discarded (Stevenson et al., 1998). Thus, the effect of changes in anthropogenic and biomass burning emissions over 240 year period on the global distribution of tropospheric ozone are simulated.
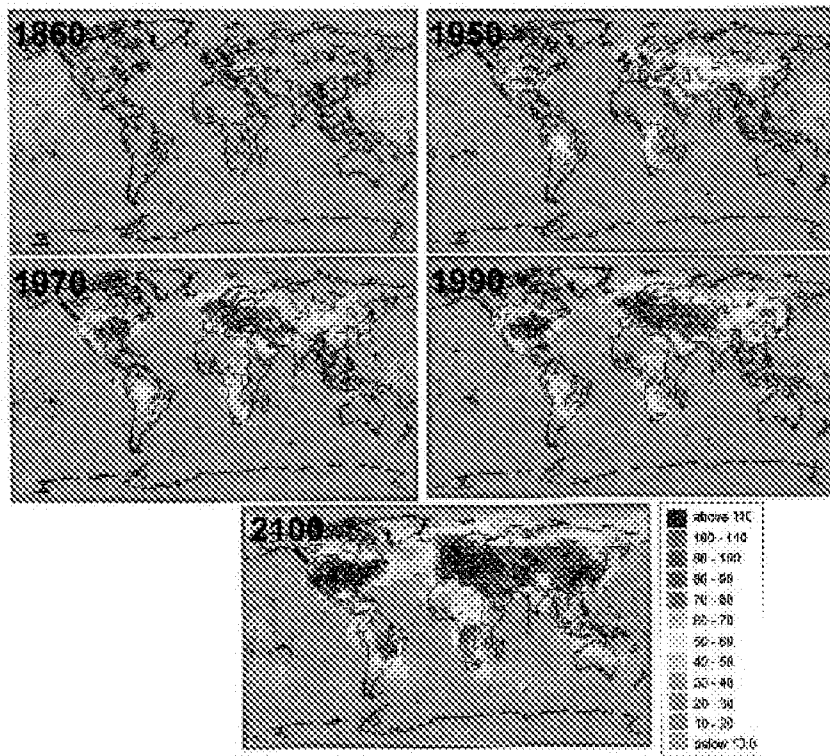

Fig. 8   July average global surface ozone concentrations at 5km x 5km from the STOCHEM model (Stevenson et al 1998).

The results in map form are shown in Figure VIII. The spatial resolution is sufficient to show the regional patterns and the 1860 map shows no areas in which July average surface O_3 concentration exceeds 50 ppb and peak concentrations of approximately 25 ppb at a latitude of 40°N. By 1950, substantial areas of Europe, the Middle East and the southern States of the USA experience peak daily O_3 concentrations in excess of 60 ppb with small areas reaching average concentrations of 70 ppb. The areas of experiencing elevated O_3 concentration expand rapidly between 1950 and 1990 and average

concentrations in the most polluted areas reach 80 ppb. The simulation for the year 2100 is based on emissions according to the IS92a scenario (IPCC 1992) using meteorology derived from 2 x CO_2 climate.

The land cover maps (Figure IX) have been used to quantify the areas of global forest exposed to peak surface concentrations in excess of 60ppb. The use of 60ppb as the threshold to quantify the aereal and temporal changes in the exposure of phytotoxic ozone overcomes some of the problems of uncertainty in the analysis. However, uncertainty in O_3 exposure due to the vertical gradient in O_3 concentration in the lowest 10m to 20m of the boundary layer (Grunhage et al. 1999), and the uncertainties in precursor emission and the overall chemical regulation of boundary layer O_3 concentration, argue for a cautious approach in quantifying the likely distribution of phytotoxic O_3 concentration. Given this approach, the global extent of O_3 exceedance is largely restricted to the temperate latitudes in 1950 and extends to 1.7 x 10^6 km^2 or 9% of the temperate and sub-polar forest (Table VI). For 1970, 6.3 x 10^6 km^2 or 18% of global forest is exposed to potentially phytotoxic O_3 concentrations, of which 2/3 is in the temperate and sub-polar latitudes. By 1990 the aereal extent of the forest exposed to O_3 concentrations in excess of 60ppb has grown to 8.3 x 10^6 km^2, almost a quarter of Fig. 9 Global areas of forest cover where the July peak surface ozone concentration exceeds 60 ppb from the model output of the STOCHEM model (Stevenson et al.1998).

TABLE VI

Assuming forest at risk is that exposed to surface ozone concentrations >60 ppb

	Potential area of exceedance		
	all forests	tropical + subtropical	temperate + subpolar
1860		no exceedance of 60 ppb	
1950 %	2.1 x 10^6 km^2 6.3	0.5 x 10^6 km^2 2.9	1.7 x 10^6 km^2 9.2
1970 %	6.3 x 10^6 km^2 18.5	1.9 x 10^6 km^2 12.1	4.4 x 10^6 km^2 24.1
1990 %	8.3 x 10^6 km^2 24.4	3.0 x 10^6 km^2 19.1	5.3 x 10^6 km^2 29.1
2100 %	17.0 x 10^6 km^2 49.8	6.0 x 10^6 km^2 37.9	11.0 x 10^6 km^2 60.2

The global forest with 60% in the temperate and sub-polar region. During the period 1950 to 1990 as well as the overall increase in area of forest exposed (by a factor of 4), the relative increase in area is almost an order of magnitude over the 40 years while the increase in the temperate zone is only (!) a factor of 3. The modelled forest exposure for 2100 is necessarily dependent on global emissions for this year and is somewhat
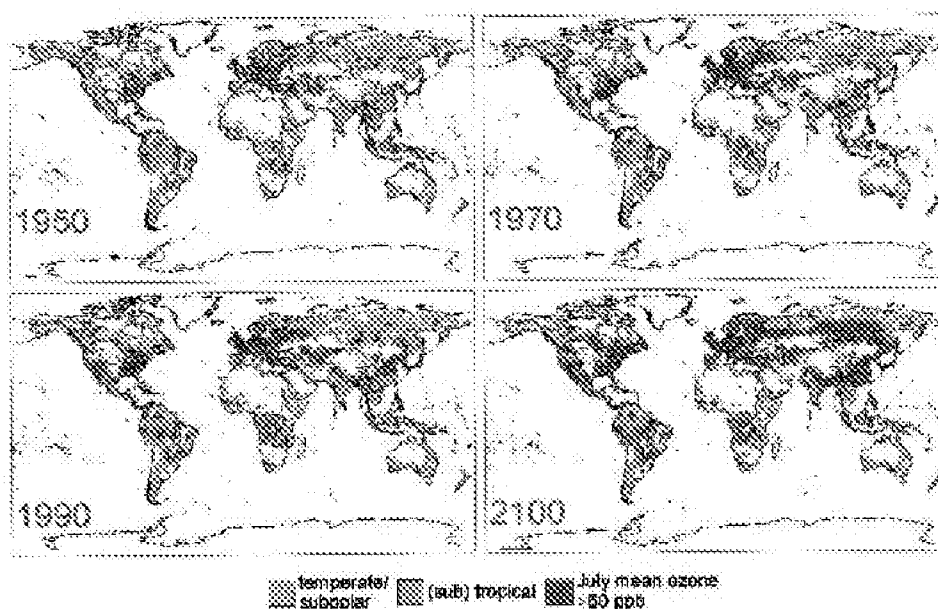
Fig. 9 Global areas of forest cover where the July peak surface ozone concentration
exceeds 60 ppb from the model output of the STOCHEM model (Stevenson *et al.*1998).

speculative. However, the very large area (17 x 10^6 km^2), representing 50% of global
forest exposed to potentially phytotoxic O$_3$ concentrations, identifies clearly the
potential magnitude of the problem. While projected emissions and climate a century in
advance introduce considerable uncertainty in the actual exposure to O$_3$, it seems
probable from the simulation that photochemical oxidants will remain a major global
pollutant threat to forest productivity globally.

Many variables interact to determine the actual exposure to potentially damaging
concentrations. Considering first the damage mechanism, which is much more likely to
be due to the absorbed dose of O$_3$ exceeding a threshold for toxicity than simply
exposure to large ambient concentrations (Fowler *et al.*, 1995). Such a threshold would
almost certainly vary between species and with different environmental conditions. The
cumulative exposure over a growing season in excess of the threshold may then be used
to quantify the likely scale of dry matter lost. A very speculative exercise could be
attempted for the global exceedance to 60ppb O$_3$. However, in practice the elevated CO$_2$
concentrations for the year 2100 will influence the photosynthetic process and net
carbon exchange such that the water-use efficiency may have changed considerably.
Such effects would decrease canopy conductance and both evapotranspiration and the
ozone flux would be appreciably smaller. Regional modification of evapotranspiration,
and therefore leaf temperatures, as a consequence of a larger fraction of net radiation
partitioned into sensible heat loss as a consequence of smaller canopy stomatal

conductances, would also feed-back to further decrease the canopy stomatal conductance. These strong feedbacks would be expected to reduce the canopy ozone fluxes, and thus lead to smaller exceedance of fluxes in excess of a threshold for dry matter loss. Therefore, even given accurate forecasts of the ozone concentrations at dates sufficiently far into the future, the climate would probably have changed significantly, and any attempt to quantify the response of forest productivity becomes extremely speculative.

A further, and probable consequence of elevated CO_2 in a warmer climate on boundary layer ozone concentration would be a reduction in the rate of removal of ozone at the surface by dry deposition due to reduced canopy conductance, and this would increase boundary layer O_3 concentrations.

TABLE VII

% Change in global areas of potential ozone effects

% Change	All forests	Tropical + subtropical	Temperate + subpolar
1950 – 1970	195	318	162
1970 – 1990	32	59	21
1990 – 2100	104	98	107
1950 – 2100	694	1214	553

10. Conclusions

The underlying processes contributing to pollutant deposition on forests are reasonably well understood for the pollutant gases SO_2, NO_2, NH_3, HNO_3 and O_3. While uncertainties remain on the chemical interaction of pollutants on foliar surfaces and especially within water films on vegetation, rates of deposition of the pollutants on specified forest can be quantified given ambient concentrations of the pollutants and knowledge of the key environmental variables (wind velocity, air temperature, solar radiation and the presence of surface water on foliage). The average rates of deposition are appreciably larger for the gases SO_2, NO_2 and NH_3 onto forests than onto shorter vegetation. This leads to inputs of fixed atmospheric nitrogen into forests in the UK exceeding those for most other land classes by between 30% and 50% depending on the pollution climate of the site. Rates of particle deposition onto forest also exceed those for other land classes, although important uncertainties exist in current understanding of the physical processes which create these differences. In particular, the deposition rates of sub-micron aerosols onto forests appear from recent field studies to exceed those suggested earlier from wind tunnel studies by an order of magnitude. Since rates of turbulent transfer and the associated aerodynamic resistance are reasonably well known,

the mechanism(s) leading to efficient transfer through the viscous sub-layer require explanation.

Cloud droplet capture by forest remains a very efficient deposition process for high elevation forest, and in polluted regions this mechanism may lead to very large inputs. The systematic study of chemical composition of orographic cloud composition around the globe may identify important regional pollutants and considerably aid understanding of hydrochemical cycling processes in remote regions.

The current temporal trends of acidifying pollutant emissions show a marked contrast between the total emissions, which are increasing globally, and emissions in Europe and North America, which are declining. These trends will, over the next decade, appreciably reduce the areas of forest in Europe and N. America currently receiving inputs of acidity in excess of 2 keq H^+ ha^{-1} annually. However, increasing emissions in rapidly developing countries, and especially in South East Asia, will greatly increase the area of forest in exceedance of 2 keq H^+ ha^{-1} annually. By 2050 more than half of the global forest subject to acidifying inputs in excess of 2 keq H^+ ha^{-1} annually will be in tropical and sub-tropical countries.

The photochemical oxidants, and O_3 in particular, have been shown to be major pollutants for forests globally, and to have increased rapidly throughout the last half century. Model forecasts of the global exposure of forest to O_3 indicate continued increases in exceedance of thresholds for physiological effects, and probably also dry matter decreases. The timescale of the projections is sufficiently long that important interactions between the effects of climate, CO_2 and O_3 on the responses of forests to O_3 are likely. The scale of the interactions is also likely to be of the same order as the direct effect of the O_3, so that significant positive or negative feedback is likely. Within the same general theme, the steady increase in deposition of nitrogen compounds (as oxidised and reduced compounds NO_2, NH_3, NO_3^-, HNO_3, NH_4^+) with time is also likely to interact with climate change and enhanced CO_2. Thus, the net effect on forest composition, productivity and sensitivity to environmental stresses becomes very difficult to predict. There is little doubt that given the scope for environmental changes in the chemical and physical climates to which forests will be exposed over the next century that current models could simulate dramatic changes to the composition of the afforested landscape. It is much more likely that the actual effects which occur will be unforeseen, and that they may be either positive or negative, and only extensive monitoring and research programmes in this field will provide the opportunity to identify such effects sufficiently early to introduce adequate control measures to prevent important social and economic consequences.

Acknowledgements

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