

Trees and the deposition of atmospheric pollutants

13. DRY DEPOSITION OF AIRBORNE POLLUTANTS ON FORESTS

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During the last decade, determined efforts have been made to elucidate (i) the factors influencing the removal and fate of atmospheric pollutants, and (ii) their effects on terrestrial and aquatic ecosystems. For the former, methods have ranged from field experiments using SO₂ labelled with the radioisotope ³⁵S (Garland *et al.*, 1973) to laboratory measurements using a mass balance technique (Hill & Chamberlain, 1976), with most information being obtained from micrometeorological methods developed originally for studies of heat and momentum exchange between vegetation and the atmosphere.

Gaseous or particulate material captured directly by the earth's surface from the atmosphere, regardless of the state of the surface or the mechanism operating, is referred to as dry deposition in contrast to wet deposition, the transfer of substances from the atmosphere in aqueous solution (rain) or suspension. All gases and particles smaller than about 10 μm are transported to surfaces by turbulent transfer processes, and, as most atmospheric particles are in this size category, dry deposition tends to be interchangeable with turbulent deposition. In contrast, the relatively small fraction, by mass, of large particles (those with diameters ≥10 μm) and appreciable terminal velocities are deposited under the influence of gravity.

I. The removal of atmospheric pollutants

1.1 Gases

The rates of dry deposition of gases are determined by 2 largely independent processes, (i) turbulent diffusion which transports gas molecules from the free atmosphere to a surface (in reality, surfaces have viscous boundary layers of air through which gases must be transported by molecular diffusion), and (ii) the affinity (chemical or physical) of the absorbing surface for the gas in question.

Crop canopies have at least 3 different sites at which gas molecules may be absorbed, (i) plant cuticles, (ii) the lining of sub-stomatal cavities and (iii) the soil surface. Each of these has a different affinity for different gases, each varying independently of the other. Surface water, whether rain or dew, acts as an additional 'sink' and further complicates the picture.

Of a wide range of methods available for measuring deposition rates, micrometeorological techniques have helped most in defining mechanisms. They average over a large area of surface, are non-destructive and enable fluxes to be obtained over short periods (~20 minutes) during which surface and atmospheric conditions are generally reasonably constant. Although they are confined to the atmosphere, micrometeorological measurements enable the influence of atmospheric transfer on rates of deposition to be separated from that of surface affinity. For this, it is convenient to use a resistance analogue described in detail by Thom (1975) and subsequently used for interpreting flux measurements made of pollutant gases by, among others, Garland (1978) and Fowler (1978).

The vertical flux of pollutant gas, F_p, to suitable surfaces (extensive level areas of uniform vegetation) is proportional to ∂X/∂z, the vertical concentration gradient of pollutant gas in the turbulent boundary layer over these surfaces:

$$F_p = -K_p \frac{\partial X}{\partial z} \quad (1)$$

where K_p, the constant of proportionality, is the eddy diffusivity for the gas in question.

Integrating (1) with respect to height between Z₁ and Z₂:

$$F_p = X_p(Z_2) - X_p(Z_1) \quad (2)$$

$$\int_{Z_1}^{Z_2} \frac{\partial X}{K_p}$$

—a relationship analogous to Ohm's law. Assuming that the surface concentration is zero, and that the lower limit of the integral is the surface, then equation (2) may be simplified:

$$r_t(Z_2) = \frac{X(Z_2)}{F_p}$$

The flux and hence total resistance, r_t , may be obtained from measurements of vertical gradients, above suitable surfaces, of (i) wind-speed, (ii) air temperature and (iii) concentrations of pollutant gas.

The total resistance to pollutant gas deposition is the sum of atmospheric, r_a and r_b , and surface r_c components (Figure 32). Atmospheric terms r_a and r_b may be estimated directly from wind speed and air temperature profiles, and thus the surface components, r_{c1} , r_{c2} and r_{c3} , may be estimated as a single residual term, r_c . If measurements of $r(a + b)$ equal r_t , then the surface is behaving as a perfect sink and is absorbing the gas as quickly as the eddy diffusion processes can

relate measurements being made of fluxes of SO_2 at ITE's experimental forest site, kindly made available by the Forestry Commission, in Ochil Forest. The main sinks for SO_2 in a forest canopy will be measured, and, from them, the different parts of the resistance network (Figure 32) will be evaluated, enabling the prediction of rates of deposition on forests to be made in a wide range of surface and atmospheric conditions.

From a compilation of published deposition rates, Garland (1978) showed that canopy resistance to SO_2 deposition increased with vegetation height. Canopy resistance to water vapour transfer also increases with vegetation height, an adaptation towards stricter control of water loss in tall vegetation. It seems probable, therefore, that stomata, the major source of water vapour in vegetation, are important sites of SO_2 uptake in tall vegetation,

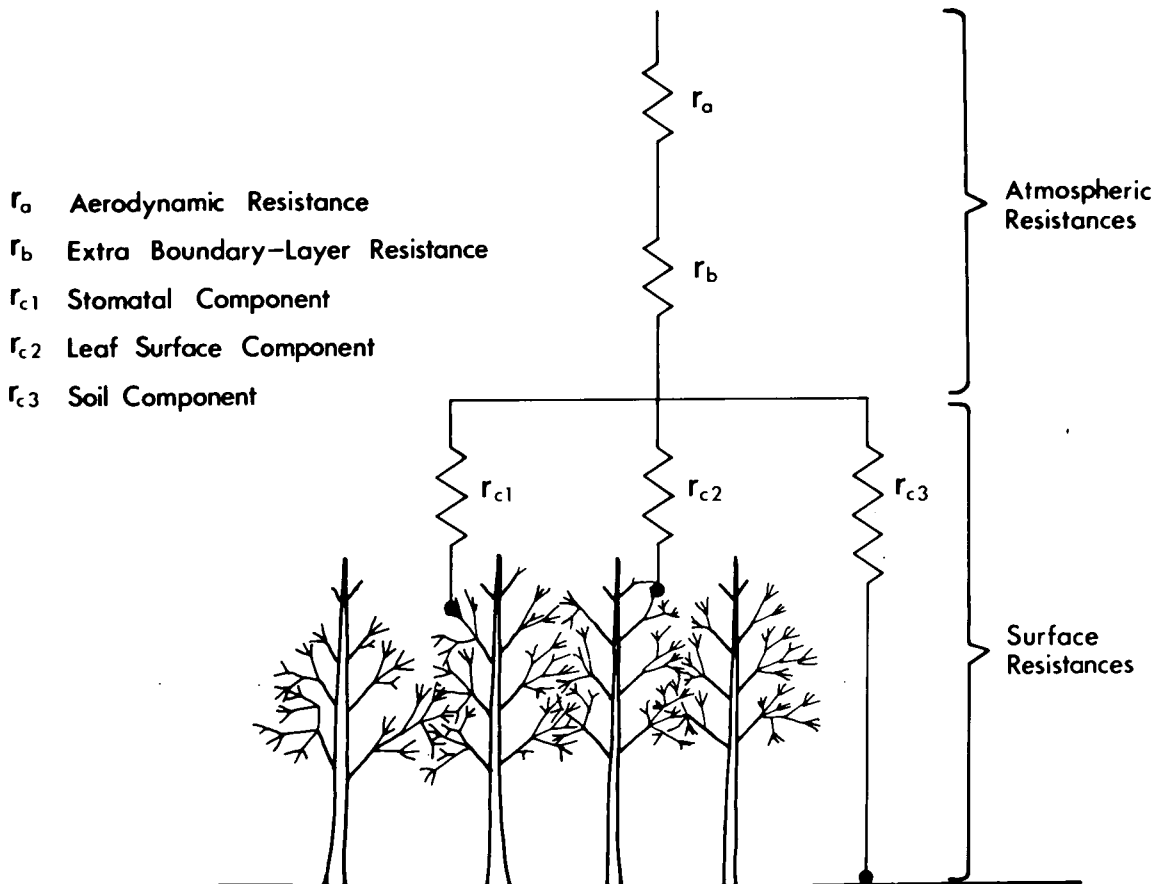


Fig. 32 Resistances to pollutant gas deposition in forest canopies.

supply it. However, when a residual term exists (r_c), the surface or canopy resistance, we have a measure of the affinity of the absorbing surface (all sinks acting in parallel) for the gas in question.

This background provides a framework to inter-

just as they are in short vegetation (Fowler, 1978). Aerodynamic resistance for mass transfer to and from vegetation decreases with increasing vegetation height, so that canopy resistance to SO_2 transfer increases as a proportion of total resistance as vegetation height increases.

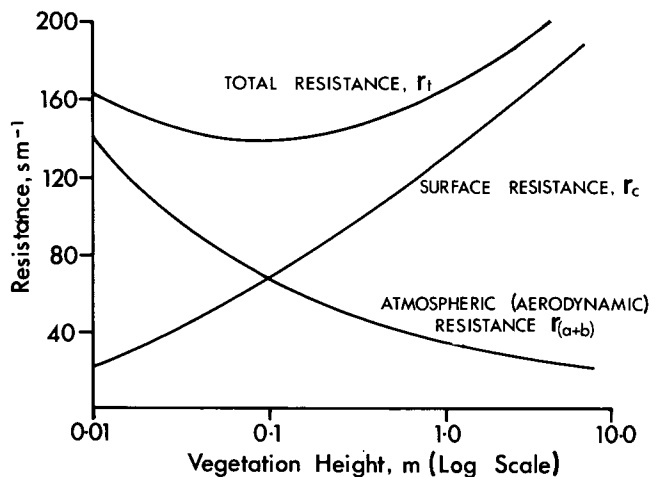


Fig. 33 Relation between vegetation height and mean surface (r_c) and atmospheric ($r_a + r_b$) resistances to SO_2 deposition.

The changes with vegetation height of aerodynamic and canopy resistance to SO_2 transfer are such that overall resistance to transfer remains approximately constant over a large height range (Figure 33). Extrapolation in this figure to the height range appropriate for forests is fairly speculative, but, if correct, it shows that surface conditions effectively control rates of SO_2 deposition. Stomatal movements and chemical and physical reactions between leaf surfaces and SO_2 are therefore likely to be the major factors influencing forest deposition rates. In the presence of surface moisture, surface resistances would be small and the large rates of deposition would be set by atmospheric resistances, provided that the pH of surface water film exceeds 4.0. Because the solubility of SO_2 declines with increasing acidity, surface resistance would eventually reappear as an important factor if surface moisture films were to become more acid than pH 4, as a result of the absorption and oxidation of SO_2 molecules. The point at which surface resistance becomes dominant lies in the range pH 3.0 to 3.5.

1.2 Particles

Turbulent diffusion processes are responsible for the transport of particles from the free atmosphere to the viscous boundary layer close to surfaces, but transport of particles through this sub-layer (distances of the order of 1mm) and subsequent capture differ considerably from that of gases. The large particles (diameter $> 1\mu\text{m}$) are transported through the sub-layer by their own inertia and by gravitational forces, whereas particles appreciably smaller than $0.1\mu\text{m}$ are transported by Brownian diffusion. In between, in the size range of 0.1 to $1\mu\text{m}$, particles do not have an effective 'vehicle' for transfer through this boundary layer and consequently their deposition rates are minimal (Chamberlain, 1975). Most sulphur-containing

particles are in the size range $0.1 - 1.0\mu\text{m}$ and, inevitably, deposition velocities are small. This low velocity, linked with the generally small ambient concentrations of sulphur particles, 2 to $8\mu\text{g S m}^{-3}$, makes the measurement of their fluxes in a forest very difficult, although there is now a prospect of success using an eddy correlation technique.

2. Effects of atmospheric pollutants

Most primary air pollutants SO_2 , O_3 , NO_x and HF are reactive gases. It would not be surprising if they and/or their secondary and tertiary reaction products were to react with the protective layers of conifer needles. Using an arbitrary system of assessment, it was found that the structure of surface configurations of epicuticular waxes was lost sooner by needles of Scots pine (*Pinus sylvestris*) growing at a polluted site in south Yorkshire than at a clean air site in the Lammermuir hills of south-east Scotland (Figure 34), care having been taken to ensure that both stands of trees were grown from seeds of the same provenance. The differences in wax structure of needles, which survive for 3 years, were greatest in the period 3 to 9 months after needle expansion.

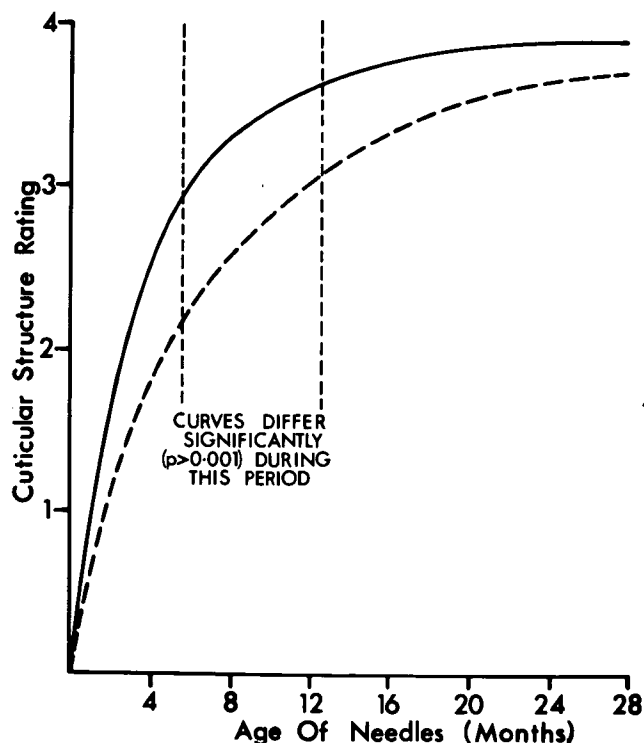


Fig. 34 Changes in the structure of epicuticular waxes of ageing needles of Scots pine grown at sites with either sulphur polluted (---) or clean (—) ambient air. CSR = CUTICLE STRUCTURE RATING, 0 and 4 being the least, and most, eroded, respectively.

References

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