# A PRELIMINARY MULTIPLE RESISTANCE ROUTINE FOR DERIVING DRY DEPOSITION VELOCITIES FROM MEASURED QUANTITIES

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**Abstract.** Because there is no simple device capable of measuring the dry deposition rates of small particles and trace gases directly, much current activity is focused on the use of an inferential technique. In this method, measurements of atmospheric concentration ($C$) of selected chemical species are coupled with evaluations of appropriate deposition velocity ($V_d$) to yield estimates of dry deposition rate from their product. Difficulties arise concerning the ability to measure $C$, and especially regarding the poor knowledge of $V_d$ for many species. A multiple resistance routine for deriving deposition velocities is presented here. Current knowledge of biological processes is incorporated into a first-generation 'big leaf' model; formulations of resistances appropriate for describing individual leaves are combined to simulate the canopy as a whole. The canopy resistance is combined with estimates of aerodynamic and boundary-layer resistances to approximate the total resistance to transfer, from which deposition velocity is then computed. Special emphasis is given to the influence of the diurnal cycle, to the way in which the various transfer resistances can be inferred from routine data, and to the role of canopy factors (e.g., leaf area index, wetness, temperature response, and sunshade fractions).

## 1. Introduction

At present, there are few experimental programs designed to produce dry deposition flux estimates. The difficulties that arise are well known, and will not be repeated here. Instead, the reader is referred to recent reviews on this subject (e.g., Wesely and Hicks, 1977; McMahon and Denison, 1979; Sehmel, 1980; Hosker and Lindberg, 1982; Hicks, 1984; Hosker, 1986; Voldner *et al.*, 1986), which discuss the factors that influence dry deposition and provide the overall scientific justification for the methods developed here.

'Dry deposition' includes both gas and particle transfer to surfaces exposed to the atmosphere. Dry deposition is a consequence of the same atmospheric exchange mechanisms responsible for the surface fluxes of heat, moisture, and momentum, but is also strongly influenced by a range of surface properties (physical, chemical, and biological). As in the case of the more familiar meteorological fluxes, surface exchange rates of trace gases and small particles can indeed be measured by micrometeorological methods, provided sufficiently sensitive and responsive sensors are available. In general, these micrometeorological methods (usually based on covariances or gradients) are sufficiently complicated that routine application in a monitoring network is not yet practical. Consequently, most dry deposition monitoring programs are designed to allow

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dry deposition rates $-F$* to be computed from field observations of the atmospheric concentrations ($C$) of the chemical species of interest, and sufficient supporting data to permit an estimate of the flux. The factor linking dry deposition to atmospheric concentrations is the deposition velocity, $V_d = -F/C$. (A table of notation is appended.)

The lack of detailed information concerning $V_d$ has been a limitation in all deposition monitoring programs conducted so far. For a small number of chemical species, $V_d$ can now be measured in intensive experimental programs, or calculated by applying an appropriate model driven by field measurements of especially selected controlling (or symptomatic) variables. The latter method is essentially inferential, and gains credibility only when referenced against some more direct, independent measurement technique. This approach is referred to as the 'inferential method', in order to distinguish it from an earlier 'concentration monitoring' philosophy which lacked the $V_d$ component.

This report summarizes a trial operational model developed for use in applying the inferential method to estimate fluxes of $SO_2$, $O_3$, $HNO_3$, and submicron particles. The present model is intended to be a practical simplification of more detailed simulations of the overall atmosphere/surface exchange process. The model is limited in its applicability to only a few well-understood plant species, and only to conditions in which pollutant concentrations are not enough to influence photosynthesis directly. In a companion paper (Matt *et al.*, 1987), results of tests of the model are presented.

## 2. Theoretical Background

A major goal of research programs conducted over the last several years has been to improve understanding of the processes controlling the deposition velocity. Figure 1 illustrates the complexity of the model required to accommodate the natural range of pollutant species and receptor surfaces. It also shows how the processes controlling the atmosphere/surface exchange of these materials can be ordered, quantified, and combined in a logical manner.

In agricultural meteorology, the concept is sometimes referred to as a 'big leaf' approach, in which no attempt is made to represent the detailed structure of the surface; rather, the critical factors controlling surface-atmosphere exchange are characterized in terms of the overall process. The present extension to the case of pollutant exchange is accomplished by introducing a resistance component to account for the chemical and biological processes that control pollutant adsorption and capture at natural surfaces.

For trace gases, the model has three major resistance components: (1) an aerodynamic resistance ($R_a$) that is wholly determined by atmospheric properties (predominantly turbulent exchange), (2) a quasi-laminar boundary layer resistance ($R_b$) that accounts for the fact that pollutant transfer in the vicinity of receptor surfaces is affected by the molecular diffusivity, and (3) a surface or canopy resistance ($R_c$) that combines the consequences of all uptake processes involving individual elements of the surface into a single number that is characteristic of the pollutant in question and the surface at the site under consideration.

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* The negative sign is arranged to conform with micrometeorological convention, in which positive turbulent exchange is away from the surface.

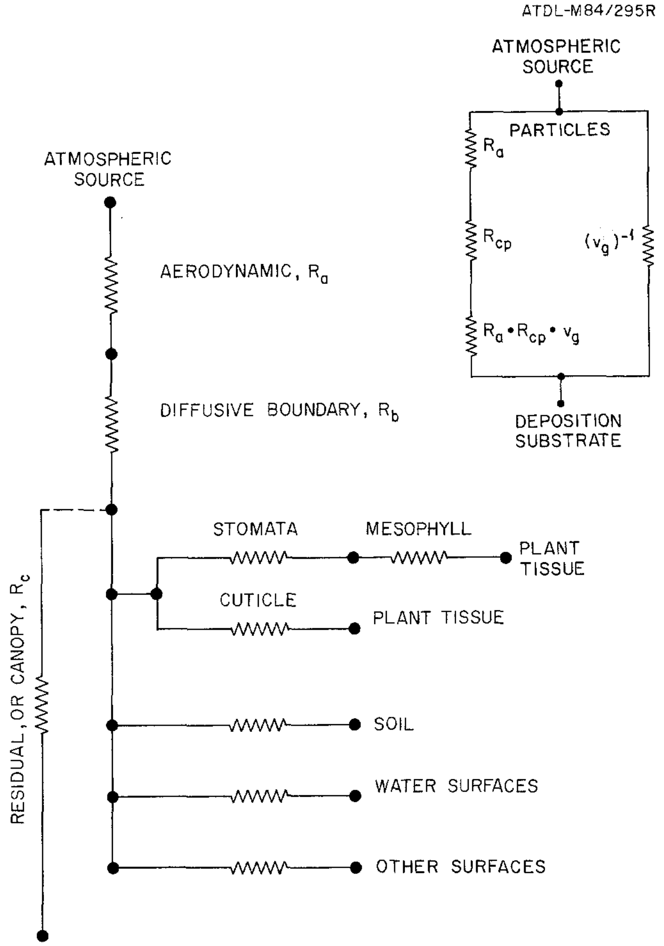

Fig. 1. The resistance model which forms the basis for data evaluation, showing the three major resistance components ($R_a$, $R_b$, and $R_c$) in series, and illustrating the way in which individual resistances to transfer to different receptor surfaces combine into an effective canopy residual resistance, $R_c$.

## 3. Mathematical Derivation

Consider the flux $F$ of material with airborne concentration $C$ and with local concentration gradient $\mathrm{d}C/\mathrm{d}z$ at height $z$ above the zero plane associated with a simple but natural surface (i.e., $z = Z - d$, where $Z$ is height above ground level and $d$ is the zero plane displacement). Following the usual methods of micrometeorology, the flux can be expressed in terms of the local vertical gradient of $C$ as

$$F = -(ku_* z)(\mathrm{d}C/\mathrm{d}z)/\phi_c(z/L), \tag{1}$$

where $k$ is the von Kármán constant (0.4), $u_*$ is the friction velocity, and $\phi_c(z/L)$ is the stability-dependent dimensionless concentration gradient. Following convention, stability is quantified in terms of $z$ and the Monin–Obukhov length scale $L$. Rearrangement and integration over height leads to the familiar form

$$C - C_0 = -(F/ku_*)\,[\ln(z/z_{0c}) - \Psi_c(z/L)]\,, \tag{2}$$

where $\Psi_c(z/L)$ is an integral form of the departure from neutral of the dimensionless concentration gradient. The roughness length, $z_{0c}$, is a constant of integration corresponding to the constraint that $C = C_0$ at the roughness height $z_{0c}$.

The electrical analog illustrated in Figure 1 provides a useful conceptual model. An aerodynamic resistance, $R_a$, can be written as

$$R_a = (1/ku_*)\,[\ln(z/z_0) - \Psi_c(z/L)]\,, \tag{3}$$

where $z_0$ is the familiar roughness length associated with momentum transfer. A second atmospheric resistance component can be written in terms of $z_0$ and $z_{0c}$:

$$R_b = (1/ku_*)\ln(z_0/z_{0c})\,. \tag{4}$$

This near-surface resistance is associated with transfer through the quasi-laminar layer in contact with the surface; it quantifies the way in which pollutant transfer differs from momentum transfer in the immediate vicinity of the surface.

Modeling studies and wind-tunnel investigations confirm that $R_b$ is strongly influenced by the diffusivity of the material being transferred. Effects associated with molecular or Brownian diffusivity lie outside the scope of the micrometeorological

TABLE I

Molecular (for gases) and Brownian (for particles) diffusivities (D; $\mathrm{cm}^2\,\mathrm{s}^{-1}$) for a range of pollutants, and the deduced values of Schmidt numbers (Sc). The viscosity of air is taken to be $0.15\,\mathrm{cm}^2\,\mathrm{s}^{-1}$ (for STP).

| | *D* | Sc |
|---|---|---|
| Gaseous species | | |
| $H_2$ | 0.67 | 0.22 |
| $H_2O$ | 0.22 | 0.68 |
| $O_2$ | 0.17 | 0.88 |
| $CO_2$ | 0.14 | 1.07 |
| $NO_2$ | 0.14 | 1.07 |
| $O_3$ | 0.14 | 1.07 |
| $HNO_3$ | 0.12 | 1.25 |
| $SO_2$ | 0.12 | 1.25 |
| Particles (unit density) | | |
| 0.001 μm radius | $1.28 \times 10^{-2}$ | $1.17 \times 10^{1}$ |
| 0.01 | $1.35 \times 10^{-4}$ | $1.11 \times 10^{3}$ |
| 0.1 | $2.21 \times 10^{-6}$ | $6.79 \times 10^{4}$ |
| 1 | $1.27 \times 10^{-7}$ | $1.18 \times 10^{6}$ |
| 10 | $1.38 \times 10^{-8}$ | $1.09 \times 10^{7}$ |

treatments leading to Equations (2), (3), and (4). However, specialized surface transfer models are available to deal with the problem (e.g., Brutsaert, 1975, 1979). These models predict a functional dependence of $R_b$ on the Schmidt number, Sc, such that in general

$$R_b = c(\mathrm{Sc})^p , \tag{5}$$

where $c$ is a constant and the exponent, $p$, is often taken to be $\frac{2}{3}$ but is suspected to vary according to the circumstance. For most trace gases, the uncertainty associated with the exponent is not critical, but for very slowly diffusing quantities such as aerosol particles, the uncertainties become large. This remains a subject for research. Table I gives values of Sc for a range of pollutants.

Many models have also been developed to describe how $R_b$ varies with surface roughness. Figure 2 (after Garratt and Hicks, 1973) shows results obtained using two such models, and demonstrates the way in which field observations disagree. As the surface becomes rougher, models tend to predict a monotonically increasing form for the dimensionless resistance $ku_* R_b$, as in Equations (3) and (4). However, field studies of sensible heat and water vapor transfer over vegetation yield a substantially different

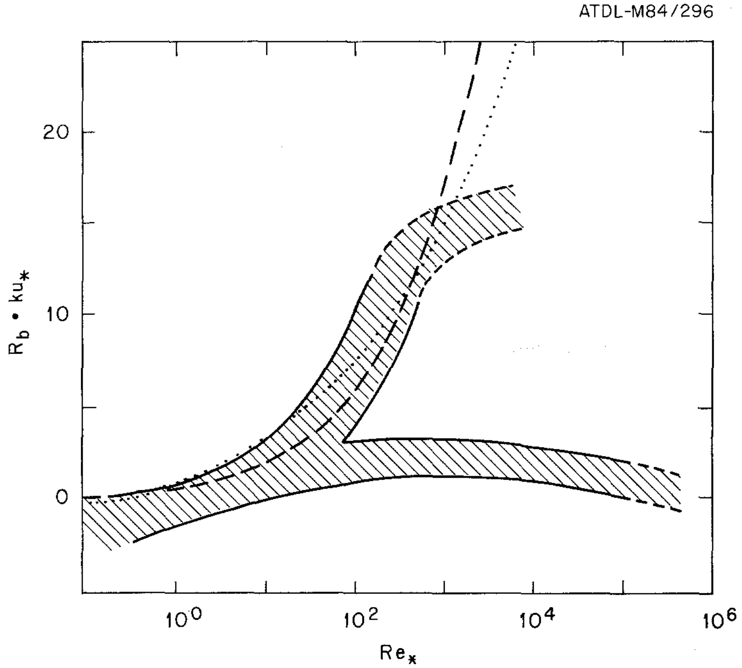

Fig. 2. The variation with surface roughness Reynolds number of the boundary-layer resistance indicator $ku_* R_b$, as determined for the case of heat transfer. Shaded areas represent field data (see Garratt and Hicks, 1973) with the upper branch being associated with bluff roughness elements and the lower branch representing vegetation and fibrous roughness elements. The dashed and dotted curves represent formulae derived by Owen and Thompson (1963) and Brutsaert (1975), respectively.

result; $ku_* R_b$ attains a limiting value of about 2. Thus, for the present purposes the quasi-laminar boundary layer resistance $R_b$ is assumed to be given by

$$R_b = (2/ku_*)(\mathrm{Sc}/\mathrm{Pr})^p , \tag{6}$$

where the Prandtl number for air, Pr ($\simeq 0.72$), enters to account for the fact that the basic observations are primarily of heat transfer (see Wesely and Hicks, 1977).

A surface or canopy resistance $R_c$, is introduced to extend the resistance network analogy into the final receptor, corresponding to the assumption that $C$ tends to zero at the point of removal. $R_c$ is defined as the ratio $-C_0/F$. The total resistance to transfer from the atmosphere is then $R = R_a + R_b + R_c$. Comparison with the definition of the deposition velocity ($V_d = -F/C$) leads to the obvious relationship that

$$V_d = 1/(R_a + R_b + R_c) . \tag{7}$$

It is instructive to consider the meaning of $R_c$ in two ways. On the one hand, $R_c$ is the net resistance corresponding to the entire surface around an observation point (or upwind of it, as dictated by fetch/height considerations). On the other hand, it is the result of integration of all of the individual resistances corresponding to the separate elements that comprise the surface. For clarity of this presentation, we will use lower-case symbols to represent the resistances associated with individual parts of the surface, and upper case symbols to represent the corresponding net resistance, as viewed from a height sufficiently far above the surface that effects of individual elements cannot be distinguished. By analogy with momentum transfer theory and experiment, this height is typically ten times the roughness length above the zero plane (if not higher, as has been suggested by some analyses; see Garratt, 1978).

There are many pathways (Figure 1) by which trace gases and particles can be deposited. For chemically reactive trace gases like $HNO_3$, capture at most natural and man-made surfaces is likely to be highly efficient, and hence an assumption $R_c = 0$ will often be adequate (see Huebert and Robert, 1985). Several water-soluble trace gases have been shown to follow pathways similar to water vapor (especially $SO_2$, but also less soluble $O_3$ and $NO_2$; see Chamberlain, 1980); hence, stomatal, mesophyll and cuticular resistances should be considered. For particles, studies of deposition to surrogate surfaces indicate a need to consider characteristics such as stickiness and microscale roughness, effects of which cannot yet be parameterized except in a relatively crude fashion. In many experimental studies, the complexity associated with $R_c$ is sidestepped by the simple expedient of evaluating $R_c$ directly as the residual from the measured total resistance to transfer $R = -C/F$, and the independent estimates of the other resistance components $R_a$ and $R_b$.

It should be emphasized that the present focus is on chemical species with no surface source; the flux is assumed to be unidirectional, towards the surface. Thus, chemical species with surface sources (e.g., $NH_3$) are outside the present scope. Questions about specific receptors are also beyond consideration here; they will require the use of a more detailed canopy model. A considerably more complicated canopy model, based on a multiple-layer subcanopy simulation of transpiration, has been presented elsewhere

(Baldocchi *et al.*, 1987). A multiple-layer subcanopy turbulence exchange model has also been developed as a contribution to this overall effort (Meyers, 1987). The outputs of such complex models provide many of the details on the biological components of the 'big-leaf' model represented in Figure 1.

## 4. Computation of $R_a$ from Field Data

Aerodynamic resistance is controlled by atmospheric turbulence and, hence, is influenced by both mechanical mixing and buoyancy. Standard micrometeorological approaches are complicated, and require accurate specification of atmospheric stability, an especially demanding task. Direct measurement of the turbulence appears to be a better approach, and has been adopted as a preferred method in the prototype systems described here. In concept, the standard deviation of wind direction, $\sigma_\theta$, contains information on stability and surface roughness, combined in a manner of special relevance to the problem now at hand.

To a close approximation, $\sigma_\theta$ can be written in terms of the standard deviation of the crosswind velocity component $\sigma_v$ as

$$\begin{aligned} \sigma_\theta &\simeq \sigma_v/u\,, \\ &= [\sigma_v)/u_*]\,[u_*/u]\,, \\ &= k[\sigma_v/u_*]\,[\ln(z/z_0) - \Psi_m(z/L)]\,. \end{aligned} \tag{8}$$

In stable and neutral conditions, it can be assumed as a first-order approximation that the integrated stability correction term for momentum $\Psi_m(z/L)$ is the same as that for all of the passive materials being transported, $\Psi_c(z/L)$. The difference between these two terms in unstable conditions seems to be of little practical importance, since the aerodynamic resistance is then typically low. Equations (3) and (8) can then be combined to give

$$R_a \cong [\sigma_v/u_*]^2/[u\sigma_\theta^2]\,. \tag{9}$$

In near-neutral and stable stratification, the ratio $\sigma_v/u_*$ is about two, but increases rapidly after the onset of instability to asymptotically approach about three (see Hicks, 1981). For the present purposes, Equation (9) is simplified to

$$R_a \cong 4/[u\sigma_\theta^2]\,; \quad [\text{neutral and stable}]\,, \tag{10a}$$

$$R_a \cong 9/[u\sigma_\theta^2]\,; \quad [\text{unstable}]\,. \tag{10b}$$

## 5. Computation of $R_b$ from Field Data

Computation of $R_b$ requires knowledge of the friction velocity $u_*$. Having determined $R_a$, an internally-consistent value for $u_*$ can be derived from the familiar neutral approximation $R_a \cong uu_*^{-2}$. Given $u_*$, Equation (6) can then be used without further manipulation to determine $R_b$.

A refinement is possible if an estimate of atmospheric stability is available. If net radiation, $R_n$, is positive, and if $\sigma_\theta$ exceeds some cardinal value $A$, then conditions can be assumed to be unstable. Inspection of Equation (8) indicates that $A$ must be site-specific because of the functional dependence of $\sigma_\theta$ upon $z_0$ and $d$. The value $A \cong 0.17$ (corresponding to $\sigma_\theta \cong 10°$) is used in the initial phases of this program, and will be refined as further experience is obtained.

## 6. Computation of $R_c$ from Field Data

Agricultural and forest meteorologists have focused on the question of what determines $R_c$, with traditional emphasis on the fluxes of water vapor, heat, and $CO_2$, although recently concern has been extended to gaseous pollutants (e.g., Unsworth, 1980). As starting points, we refer to summaries by Jarvis (1976) and Jarvis *et al.* (1981).

Special caution must be exercised with respect to nomenclature and terminology. Many specialists quantify resistances associated with biological pathways (via stomata and leaf cuticle, in particular) in terms of a biological resistance to transfer expressed as the effective resistances per unit area of foliar surface, whereas in the context of the present big-leaf model, we are mainly interested in the integrated consequences of these resistances expressed per unit horizontal area of the earth's surface. One link between the two is the leaf area index (LAI), and its equivalents that refer to other components of the biomass. However, extension of detailed knowledge of the behavior of individual canopy element surfaces to a complete canopy is far from trivial. For some properties (e.g., heat and water vapor), synthesis of complete canopy behavior can be accomplished

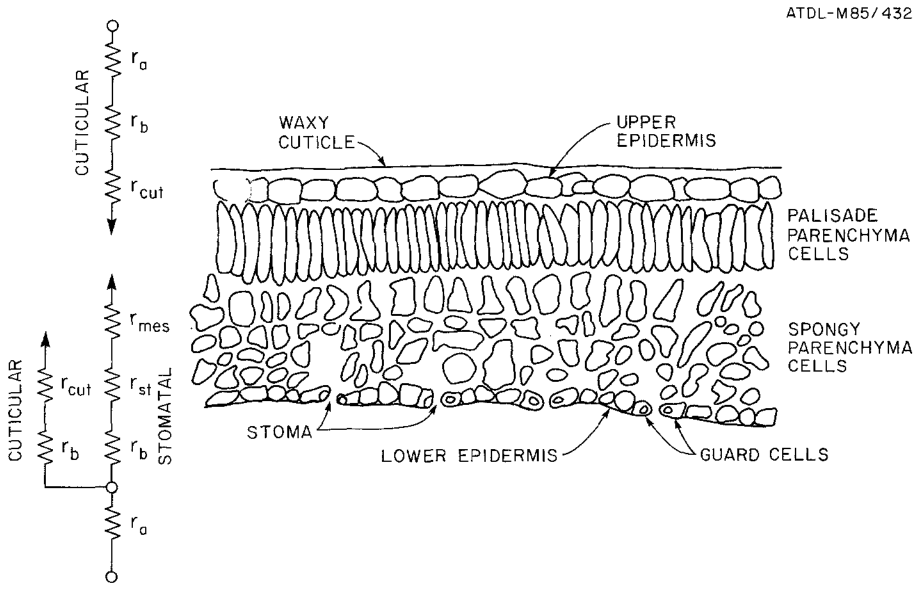

Fig. 3. A schematic illustration of the pathways and processes for transfer between air and plant tissue (following O'Dell *et al.*, 1977).

with some confidence. For other quantities (especially particles), resolution of the problem appears quite distant. We proceed, therefore, with awareness that the methods described here are best suited to trace gas transfer.

Lower case symbols ($r$) will be used to identify resistances expressed in terms of a unit area of foliar surface (i.e., biological resistances). Figure 3 gives details of the interactions among the various resistances, for a single leaf.

### 6.1. STOMATAL RESISTANCE, $r_{st}$

Transfer through stomata is by diffusion; thus, an inverse dependence of stomatal resistance on molecular diffusivity of trace gases is normally accepted (e.g., see Jarvis, 1971). In general, stomatal resistance depends on the incident photosynthetically active radiation ($I_p$):

$$r_{st} = r'_{st}[1 + (b'/I_p)]/(f_e f_w f_T f_s)\,, \tag{11}$$

where $r'_{st}$ is some minimum value which varies with the plant species (Table II), and $b'$ is an empirical constant, also species-dependent (Burrows and Milthorpe, 1976).

TABLE II

Results of experimental studies, as summarized by Baldocchi *et al.* (1987), of the relationship between stomatal resistance and photosynthetic radiation intensity, $I_p$. A relationship of the general form of Equation (11) is assumed.

| Species | $r'_{st}$ ($\mathrm{s\,m^{-1}}$) | $b'$ ($\mathrm{W\,m^{-2}}$) |
|---|---|---|
| Soybean | 65 | 10 |
| Oak | 145 | 22 |
| Maize | 100 | 66 |
| Spruce | 232 | 25 |

The correction factors $f_e$, $f_w$, and $f_T$ in Equation (11) were introduced by Jarvis (1976) to correct for effects of humidity, water stress, and temperature (respectively), and all range between 0 and 1.0. The correction factor $f_s$ modifies the stomatal resistance to account for the differences in molecular diffusivity between the trace gas in question and water vapor.

The literature survey and analyses summarized by Jarvis (1976) suggest a linear reduction of stomatal conductance with increasing atmospheric vapor pressure deficit $D_e$; $D_e = e_s(T) - e$, where $e_s(T)$ is the saturated water vapor pressure at air temperature $T$, and $e$ is the actual atmospheric vapor pressure. To account for this reduction, a factor

$$f_e = 1 - b_e D_e \tag{12}$$

is defined, where $b_e$ is a constant indicative of the response of a given plant species.

Plant water stress can be quantified in thermodynamic terms using the leaf water potential, $W_p$. Stomatal resistance is relatively independent of $W_p$, until $W_p$ drops below a threshold value. Below the threshold value, stomatal resistance increases rapidly. The influence of $W_p$ can be incorporated using a 'discontinuous switch' model (Fisher *et al.*, 1981):

$$\begin{aligned} f_w &= 1\,, && \text{if} \quad W_p > \text{threshold}\,, \\ f_w &= aW_p + b\,, && \text{if} \quad W_p < \text{threshold}\,, \end{aligned} \tag{13}$$

where $a$ and $b$ are constants.

Absolute temperature effects can be corrected with a further modifying factor

$$f_T = [(T - T_e)/(T_0 - T_e)]\,[(T_h - T)/(T_h - T_0)]^{B_t}\,,$$

where

$$B_t = (T_h - T_0)/(T_0 - T_e)\,. \tag{14}$$

Here, $T_h$ and $T_e$ are the species-dependent higher and lower temperature extremes at which stomata no longer open (typically 40 °C and 5 °C, respectively, but see Table III), and $T_0$ is the temperature at which stomatal exchange is optimized (typically 25 °C).

TABLE III

Characteristic temperatures (°C) associated with the stomatal resistance of different species, as specified in Equation (14)

| Species | $T_0$ | $T_h$ | $T_e$ |
|---|---|---|---|
| Maize (ref. 1) | 25 | 45 | 5 |
| Spruce (ref. 2) | 9 | 35 | −5 |
| Oak (ref. 3) | 25 | 45 | 10 |

*References*:
(1) Rodriguez and Davies (1982).
(2) Jarvis (1976).
(3) Aubuchon *et al.* (1978).

Because transfer through stomata is by molecular diffusion, the case of trace gases can be addressed by a simple correction introduced as the factor $f_s$ in Equation (11):

$$f_s = D/D_0\,, \tag{15}$$

where $D_0$ is the molecular diffusivity corresponding to the initial specification of $r'_{st}$ (usually water vapor), and $D$ is the molecular diffusivity of the trace gas in question.

### 6.2. ADDING THE MESOPHYLL RESISTANCE $r_{\mathrm{mes}}$

Once cpollutant or other gases enter substomatal cavities, they must be either transferred to plant cells, or destroyed by chemical reactions within the substomatal cavity or interstices among mesophyll cells. In either case, the practical consequence is that a mesophyllic resistance $r_{\mathrm{mes}}$ must be combined with the diffusive stomatal resistance $r_{st}$ in order to describe transfer to the eventual sink (see O'Dell *et al.*, 1977). In classical agrometeorology, mesophyll resistance arises as an important issue in the case of the transfer of $CO_2$, since $CO_2$ is relatively insoluble in water. In the present context, the arguments of Liss and Slater (1974) regarding the surface conductivity associated with partially soluble chemical species appear relevant; $r_{\mathrm{mes}}$ should be a function of the Henry's law constant, $H$. This constant varies strongly with chemical species: at 15 °C, $H$ is 2.19 for $O_3$ and 0.0078 for $SO_2$ (see Peters, 1984). At this time, however, there is little experimental evidence that would permit discussion of the functional relationship of $r_{\mathrm{mes}}$ on $H$. Moreover, there is no basis yet for identifying the independent roles of solubility, chemical reactions in the cavity, and reactions occurring on contact with (wet) cellular surfaces in the mesophyll.

In general, $r_{st}$ and $r_{\mathrm{mes}}$ are additive. The overall resistance to transfer through stomatal openings is then

$$r_s = r_{st} + r_{\mathrm{mes}} \,. \tag{16}$$

### 6.3. CUTICULAR RESISTANCE, $r_{\mathrm{cut}}$

Deposition directly to leaf cuticle is likely to be of importance similar to uptake via stomata for many trace gases, especially fat-soluble hydrocarbons depositing to foliage with waxy leaves. There is little known about how cuticular resistance varies with the chemical characteristics of the material being transferred or with the cuticular characteristic of leaves. At this time, $r_{\mathrm{cut}}$ is introduced for completeness of the analytical model, not because it is a well-known factor.

### 6.4. EXTENSION TO ENTIRE CANOPY

As a first attempt to extend the elemental surface resistances to an entire plant canopy, many workers scale according to leaf area index (LAI):

$$R_{\mathrm{foliage}} = r_{\mathrm{tot}}/\mathrm{LAI} \,, \tag{17}$$

where

$$r_{\mathrm{tot}} = 1/[1/r_{\mathrm{cut}} + 1/r_s] \,.$$

For many canopies (e.g., crops), such simple scaling is often adequate; however, forests present problems that are subjects of current research. Stomatal resistance is a strong function of radiation, and is, therefore, affected by shading by other foliage. It is clear that a layer-by-layer computation of net overall resistance is especially appropriate for a canopy with large LAI.

Baldocchi *et al.* (1987) have presented a more detailed model of canopy stomatal resistance, intended specifically to address questions of relevance here. As expected, shading within the canopy causes results from the more complicated model to differ from the simple LAI-based approximations developed here; scaling according to leaf area index alone leads to increasing errors as the LAI increases. As a first-order correction for this effect, the leaf area index can be partitioned into sunlit ($L_s$) and shaded ($L_{sh}$) portions by assuming a spherical leaf angle distribution:

$$\begin{aligned} L_s &= [1 - \exp(K\,\mathrm{LAI})]/K\,, \\ L_{sh} &= \mathrm{LAI} - L_s\,, \end{aligned} \tag{18}$$

where $K$ is the extinction coefficient ($0.5/\cos\alpha$; $\alpha$ is the zenith angle). To obtain the radiation intensities ($I_p$) to be used in Equation (11), the measured incoming global radiation is separated into beam and diffuse components ($I_{\mathrm{beam}}$ and $I_{p_{\mathrm{diff}}}$) of photosynthetically active radiation (PAR) using the technique of Weiss and Norman (1985). To obtain the PAR on a sunlit leaf, the incoming beam radiation is multiplied by the extinction coefficient ($K$). Then, the corresponding diffuse contribution is computed as $0.5I_{p_{\mathrm{diff}}}$, and the total PAR to sunlit foliage ($I_{p_{\mathrm{sun}}}$) is obtained by simple addition. For shaded foliage, only the diffuse component is used. The net uptake resistance associated with the foliage per unit ground area, $R_{\mathrm{foliage}}$, is obtained by combining the weighted parallel stomatal resistances of the sunlit ($r_{\mathrm{sun}}$) and shaded ($r_{sh}$) leaf area, and the weighted cuticular resistance. Then

$$R_{\mathrm{foliage}} = 1/[1/R_s + \mathrm{LAI}/r_{\mathrm{cut}}]\,,$$

where

$$R_s = 1/[L_s/r_s(KI_{p_{\mathrm{sun}}}) + L_{sh}/r_s(0.5I_{p_{\mathrm{shade}}})]\,. \tag{19}$$

In the initial forms of this model, values of soil resistance have been deduced from papers describing the results of uptake tests of gaseous pollutants to sample building materials, stone, sand, and soil exposed in wind tunnels (see the tabulations presented by Hicks, 1984) and from results of a limited number of field studies (e.g., Turner *et al.*, 1973). It is acknowledged that substantially different values are likely to be appropriate for the complicated mixture of decaying leaves and other biological matter at the floor of a forest, for example, but as yet no relevant data base is available to investigate the problem. Work in this area is continuing. At the time of this writing, estimates of soil resistance are combined with the overall foliar resistance $R_{\mathrm{foliage}}$ as if these components were in parallel, yielding an approximation for the net effective canopy resistance

$$R_c = 1/[1/R_{\mathrm{foliage}} + 1/R_{\mathrm{soil}}]\,. \tag{20}$$

### 6.6. WETTED FOLIAGE

The matter of surface wetness is receiving special attention. All of the relations given above were derived for a dry but transpiring, fully-leafed canopy. Figure 4 shows

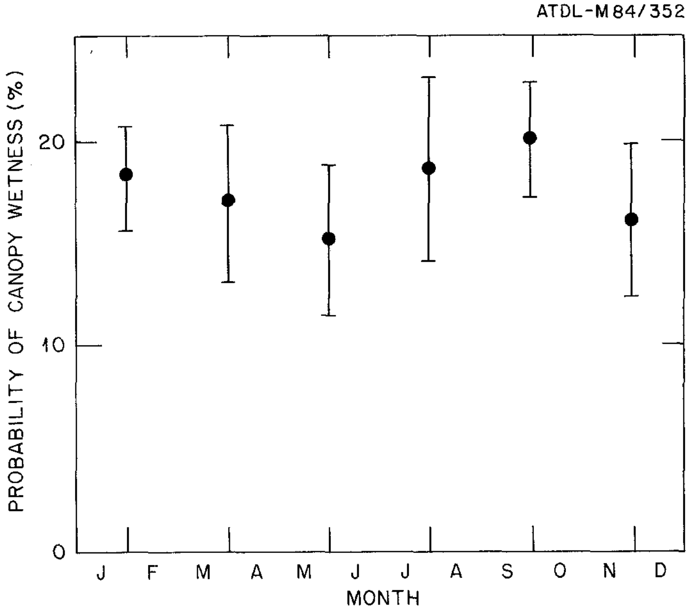

Fig. 4. The probability of surface wetness (due to all causes, including dewfall and precipitation) at the forest meteorology research site adjacent to the Walker Branch Watershed, Oak Ridge, Tennessee, over the period from July 1981 to June 1984.

observed probabilities that a forest canopy is wet, as a function of time of year, including effects of both dew formation and precipitation. The data were generated using an array of wetness sensors (Davis and Hughes, 1970), distributed above and within an oak-hickory forest near Oak Ridge, Tennessee. There is no statistically-significant annual cycle evident in the diagram. Overall, it appears that the canopy is wet 15 to 20% of the time. These data cannot be taken to be representative of all canopies, locations, and times, but nevertheless they indicate that canopy wetness may be a significant boundary condition affecting dry deposition.

Although the consequences of canopy wetness are not yet well documented, mounting evidence appears to be supporting expectations (e.g., see Fowler, 1985). As guiding considerations, the following observations are presently emphasized:

– Ozone is essentially insoluble in water. Hence, $O_3$ canopy resistance is assumed to be large when surface wetness is indicated.

– Sulfur dioxide is only weakly soluble in water having $pH < 3.5$. Early versions of this program assumed that the canopy resistance for $SO_2$ was small whenever surface wetness was detected. Subsequent field tests indicated that this assumption is wrong when the surface is wetted by rain (presumably in equilibrium with atmospheric $SO_2$ concentrations at the time of falling and hence a poor sink). Hence methods for distinguishing between the two major causes of surface wetness (precipitation and condensation) are now being explored.

## 7. Particle Deposition: a Special Case

The fact that particles can fall through the turbulent eddies that transfer trace gases raises questions about how particle transfer can be accommodated within the multiple-resistance framework developed for trace gases. Consideration of the turbulent transport of particles sedimenting with a fall speed $V_g$ leads to a modification of the conventional resistance model (see Hicks *et al.*, 1985):

$$V_p = V_g + 1/(R_a + R_{cp} + R_a R_{cp} V_g)\,, \tag{21}$$

where $R_{cp}$ is a net canopy resistance to particle deposition (incorporating the equivalent to the relevant quasi-laminar term, $R_b$). Equation (21) differs from the usual form by inclusion of a term involving gravitational settling within the resistance expansion. This term is not of great importance unless $V_g$ is relatively large. In initial applications of the model described here, the particle focus has been on submicron sulfate, for which $V_g$ is too small to cause concern in the context of Equation (21).

It is well known that particle size has a strong influence on the deposition velocity. Belot *et al.* (1976), Bache (1979), Davidson *et al.* (1982), Slinn (1982), and de la Mara and Friedlander (1982) present examples of models designed to extend laboratory data on deposition to surrogate surfaces to real-world plant canopies. These models vary widely in their assumptions and in their predictions. However, all predict deposition velocities substantially lower than observed in experiments involving deposition of naturally occurring radionuclides to plant surfaces.

The state of knowledge regarding particle deposition to vegetative canopies is presently so poor that no 'best' model can yet be identified. As an interim measure, therefore, a purely empirical relationship has been adopted. Wesely *et al.* (1983) present data showing an approximately linear relationship between $V_d$ for small particles and the product $\sigma_\theta$u, for a relatively limited set of conditions:

$$V_d = 0.003\sigma_\theta u\,. \tag{22}$$

It is not yet clear how Equation (22) can best be integrated with the formalism developed earlier for trace gases. However, it is obvious that a dependence on Schmidt number Sc is required. Wesely *et al.* (1983) emphasize the uncertainties involved in any assumption regarding the role of Sc. For testing purposes, Equation (22) is being used as a first-cut attempt to yield results appropriate to the particulate species associated with the entire spectrum of accumulation-size-range aerosols. Experimental programs concurrent with this modeling effort are presently addressing this problem.

## 8. Discussion

The program presently used to compute deposition velocities from routinely collected meteorological data is listed elsewhere (see Hicks *et al.*, 1985). This program takes as input the quantities identified in the main text of this document, with supporting data provided by site operators from weekly visual observations.

Data recorded at individual sites are first examined in order to detect (and reject) errors, and to interpolate through short periods of missing data. On-site checks and data tests are performed to ensure that sensors and recording apparatus are working correctly. After completion of such tests, values of the separate resistances $R_a$, $R_b$, and $R_c$ are computed.

The key parameters required as input for the interpretive scheme are as follows:

(a) For $R_a$, only meteorological data are required. The optimal data are wind speed and stability information (such as net radiation and the magnitude of $\sigma_\theta$, the standard deviation of wind direction). At high-quality meteorological sites, more precise evaluations of $R_a$ could be obtained by direct measurement of eddy fluxes, or by application of flux-gradient relations to gradient data. It should be remembered, however, that gradient data cannot be easily interpreted except over smooth surfaces that are horizontal and uniform. Such sites are not the norm.

(b) For $R_b$, pollutant characteristics are also important. The Schmidt number is used to modify meteorological evaluations of the quasi-laminar layer transfer resistance based on estimates of the friction velocity derived from wind speed data and knowledge of surface roughness.

(c) For $R_c$, knowledge is required of a number of biological factors. Stomatal resistance can be estimated from measurements of photosynthetically active radiation (PAR), air temperature, and humidity, coupled with observations of vegetative species and leaf area index. The value of $R_c$ must be modified according to canopy wetness. Effects of water stress and phenology remain to be addressed.

Instrumentation suitable for evaluating each of these resistances and for tracking the performance characteristics of the associated chemical devices has been set up at a small network of test locations. At each location, measurements of key variables are performed every ten seconds, and average and statistical quantities are calculated every 15 min; the variables are $U$, $\sigma_\theta$, $T$, $RH$, $W_n$, and $I$. Precipitation data are recorded to detect occasions of severe water stress, and to determine periods when canopies are wet from precipitation, as opposed to dew. Weekly site-operator observations of local surface and canopy conditions are also required, to document drought, changes in the status of vegetation, the presence of snow, etc. These various inputs are analyzed by computer programs, which first search for data inconsistencies or omissions and, for satisfactory data, then estimate the appropriate transfer resistances.

All of the procedures used in this analysis are under continuing review, involving both the improvement of relevant theories and simulations and direct comparison with more detailed and sophisticated experimental observations made at specially-selected sites. The model presented here is viewed as a slowly-improving tool to interpret meteorological and surface data, and is as yet not such that general application can be recommended.

## 9. Summary and Conclusions

Knowledge of deposition velocities and their controlling factors is not yet adequate to develop a system for evaluating deposition velocities from routine monitoring data with great confidence. However, sufficient knowledge is present for first-generation interpretive routines to be structured. The 'big leaf' model approach, as described here, combines gross resistances that correspond to those of a single leaf, equivalent to the canopy as a whole. Preliminary computer codes have been written, and are presently being tested in a trial network. This small network is inferring weekly dry deposition from weekly-averaged atmospheric concentration data. At the time of this writing, the emphasis is on $SO_2$ and $HNO_3$ vapor, with a secondary interest in $O_3$ and aerosol particles.

Several limitations of the inferential method described here should be emphasized. First, the technique relies heavily on the accurate measurement of air concentrations at remote locations, for which standardized techniques are not yet well-developed. Second, the body of knowledge on which any interpretive scheme for deposition can be based is quite limited. Most information on gas transfer either deals with average uptake in laboratory conditions (chambers, wind-tunnels, etc.), or is derived from short-term micrometeorological measurements at carefully selected field sites. Third, the method only addresses the case of atmospheric pollutants that are unidirectionally transferred to the surface. Fourth, knowledge is lacking on effects associated with surface heterogeneity and terrain complexity.

Ongoing research programs are addressing these (and other) problems. Until their work is successfully concluded, none of the methods addressed here can be advocated with assurance.

The above comments imply constraints on the applicability of answers derived using the methods outlined here. The results will be useful only for locations that have spatially homogeneous vegetation, and where terrain effects are not dominating. Values derived will be appropriate for only a limited area surrounding observing site. The 'point measurements' on which the techniques are based imply an 'area of applicability' centered on each measurement location, but possibly no larger than 1 km diameter. Answers obtained will be best founded for $SO_2$ and $O_3$, with nitric acid vapor possibly closely following. Other species cannot yet be addressed with more than minimal confidence, even in the best of circumstances.

The measurement program considered here is intended to provide estimates of dry deposition, with quantified uncertainty, over time scales compatible with existing wet-deposition sampling protocols. The goal is to compute weekly averages of dry deposition. To this end, weekly averages of air concentrations have proved adequate, yet it is necessary to consider deposition velocity with much finer time resolution. The main reason lies in the way the various contributing resistances interact, each with the possibility of changing greatly from hour to hour. For this reason, the deposition velocity routines are intended to provide 1-hr time resolution, from which longer period information is then derived by integration. The errors involved in this approach have been investigated by Meyers and Yuen (1987).

It is intended that the dry deposition data should have similar statistical validity to the wet deposition data presently being collected. As a 'rule-of-thumb', the error associated with the average annual wet deposition to any site is likely to be in the range

10 to 50%, varying widely with location. As knowledge of wet-deposition statistics improves, required accuracies for dry-deposition measurement can be simultaneously refined. In some areas time trends might be sufficiently well documented by fewer dry-deposition stations than wet. However, documentation of important spatial patterns will require more dry stations in source regions than in remote areas.

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## Appendix. Table of Notation

| | | |
|---|---|---|
| $b'$ | Constant in expression for $r'_{st}$ | ($\mathrm{W\,m^{-2}}$) |
| $b_e$ | Constant in expression for $f_e$ | |
| $b_w$ | Constant in expression for $f_w$ | |
| $B_t$ | Exponent in temperature expression for $r_{st}$ | |
| $c$ | Constant | |
| $C$ | Concentration in air | ($\mathrm{g\,m^{-3}}$) |
| $C_0$ | Concentration in air in contact with the surface | ($\mathrm{g\,m^{-3}}$) |
| $d$ | Displacement height | (m) |
| $D$ | Diffusivity | ($\mathrm{m^2\,s^{-1}}$) |
| $D_e$ | Vapor pressure deficit | (mb) |
| $D_0$ | Molecular diffusivity of baseline species | ($\mathrm{m^2\,s^{-1}}$) |
| $e$ | Vapor pressure | (mb) |
| $e_s(T)$ | Saturated vapor pressure at temperature $T$ | (mb) |
| $f_e$ | Parameterization expression for effect | |
| $f_s$ | Parameterization expression for effect | |
| $f_T$ | Parameterization expression for effect | |
| $f_w$ | Parameterization expression for effect | |
| $F$ | Vertical flux (deposition, if negative) | ($\mathrm{g\,m^{-2}\,s^{-1}}$) |
| $H$ | Henry's law constant | |
| $I$ | Total insolation | ($\mathrm{W\,m^{-2}}$) |
| $I_p$ | Photosynthetically active radiation | ($\mathrm{W\,m^{-2}}$) |
| $I_{p_{\mathrm{beam}}}$ | Direct component of PAR | ($\mathrm{W\,m^{-2}}$) |
| $I_{p_{\mathrm{diff}}}$ | Diffuse component of PAR | ($\mathrm{W\,m^{-2}}$) |
| $I_{p_{\mathrm{shade}}}$ | PAR to a shaded surface | ($\mathrm{W\,m^{-2}}$) |
| $I_{p_{\mathrm{sun}}}$ | PAR to a sunlit surface | ($\mathrm{W\,m^{-2}}$) |
| $k$ | von Kármán's constant (0.4) | ($\mathrm{W\,m^{-2}}$) |

| | | |
|---|---|---|
| $K$ | Extinction coefficient | |
| $L$ | Monin–Obukhov length scale | (m) |
| LAI | Leaf Area Index | |
| $L_s$ | Sunlit proporties of total leaf area | |
| $L_{sh}$ | Shaded proportion of total leaf area | |
| $p$ | Constant: exponent | |
| Pr | Prandtl number | |
| $r_a$ | Leaf-scale aerodynamic resistance | |
| $r_b$ | Leaf-scale boundary-layer resistance | |
| $r_{\mathrm{cut}}$ | Cuticular resistance | $(\mathrm{s\,m^{-1}})$ |
| $r_{\mathrm{mes}}$ | Mesophyll resistance | $(\mathrm{s\,m^{-1}})$ |
| $r_s$ | Total resistance to exchange through stomata | $(\mathrm{s\,m^{-1}})$ |
| $r_{\mathrm{sh}}$ | Shaded stomatal resistance | $(\mathrm{s\,m^{-1}})$ |
| $r_{st}$ | Stomatal resistance | $(\mathrm{s\,m^{-1}})$ |
| $r_{\mathrm{sun}}$ | Sunlit stomatal resistance | $(\mathrm{s\,m^{-1}})$ |
| $r_{\mathrm{tot}}$ | Total leaf resistance | $(\mathrm{s\,m^{-1}})$ |
| $r'_{st}$ | Minimal stomatal resistance | $(\mathrm{s\,m^{-1}})$ |
| $R_a$ | Aerodynamic resistance | $(\mathrm{s\,m^{-1}})$ |
| $R_b$ | Quasi-laminar boundary resistance | $(\mathrm{s\,m^{-1}})$ |
| $R_c$ | Canopy, or surface resistance | $(\mathrm{s\,m^{-1}})$ |
| $R_{cp}$ | Canopy resistance – particles | $(\mathrm{s\,m^{-1}})$ |
| $\mathrm{Re}_*$ | Roughness Reynolds number | |
| $R_{\mathrm{foliage}}$ | Total foliar resistance of a canopy | $(\mathrm{s\,m^{-1}})$ |
| $RH$ | Relative humidity | |
| $R_{\mathrm{soil}}$ | Soil resistance | $(\mathrm{s\,m^{-1}})$ |
| Sc | Schmidt number | |
| $T$ | Temperature | (°C) |
| $T_e$ | Lower temperature boundary for stomatal exchange | (°C) |
| $T_h$ | Upper temperature boundary for stomatal exchange | (°C) |
| $T_0$ | Optimal temperature level for stomatal exchange | (°C) |
| $u$ | Wind speed | $(\mathrm{m\,s^{-1}})$ |
| $u_*$ | Friction velocity | $(\mathrm{m\,s^{-1}})$ |
| $V_d$ | Deposition velocity | $(\mathrm{m\,s^{-1}})$ |
| $V_g$ | Gravitational settling speed (particles) | $(\mathrm{m\,s^{-1}})$ |
| $W_n$ | Surface wetness | |
| $W_p$ | Leaf water potential | (mb) |
| $z$ | Height above the zero plane | (m) |
| $z_0$ | Roughness length for momentum | (m) |
| $z_{0c}$ | Roughness length for concentration | (m) |
| $Z$ | Height above the surface | (m) |
| $\alpha$ | Zenith angle | (deg) |
| $\phi_c$ | Dimensionless gradient of concentration | |
| $\sigma_\theta$ | Standard deviation of wind direction | (radians) |

| | | |
|---|---|---|
| $\sigma_v$ | Standard deviation of lateral velocity | ($\mathrm{m\ s^{-1}}$) |
| $\Psi_c$ | Integrated departure from neutral of the dimensionless concentration gradient | |
| $\Psi_m$ | Integrated wind profile correction | |

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