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A MULTI-LAYER MODEL FOR ESTIMATING SULFUR DIOXIDE DEPOSITION TO A DECIDUOUS OAK FOREST CANOPY

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Abstract—A multi-layer 'K-theory' model for gaseous deposition to a deciduous forest is described and discussed. The model incorporates realistic physiological concepts to improve upon earlier multi-layer models found in the literature. Computations of deposition fluxes and velocities are most sensitive to variations in parameters relating to stomatal resistance. The model computations of deposition velocities are less sensitive to aerodynamic parameters since the canopy resistance of a deciduous forest is generally much greater than the aerodynamic resistance. Model computations of SO_2 flux and deposition velocities are tested against deposition measurements made over a deciduous forest; these computations are not significantly different from measured values. Estimates of deposition fluxes, computed with multi-layer model, improve upon those values estimated with a "big-leaf" model.

Key word index: Dry deposition, canopy–atmosphere interactions, K-theory model, micrometeorology.

NOMENCLATURE

$a(z)$	leaf area density at height z
b_{rs}	curvature coefficient of the stomatal resistance–PAR response curve
C_{am}	bulk canopy drag coefficient
$C_m(z)$	effective canopy drag coefficient at height z
d	zero plane displacement height
$df(z)$	leaf area of an incremental layer
D_s	molecular diffusivity of SO_2
$F(z)$	flux of SO_2 at a height z
F_{SO_2}	SO_2 flux between the canopy and the atmosphere
h	height of the canopy
k	von Karman's constant
$K_m(z)$	eddy exchange coefficient for momentum transfer
$K_s(z)$	eddy exchange coefficient for SO_2 transfer
l	characteristic length of leaves
L	Monin–Obuhkov scale length
LAI	canopy leaf area index
PAR	photosynthetically active radiation
r_{sm}	minimum stomatal resistance under optimal conditions
R_a	aerodynamic resistance
R_b	boundary layer resistance of a leaf to mass exchange
$R_b$soil	boundary layer resistance of the soil
R_{cut}	cuticle resistance to mass exchange
R_g	global solar radiation
R_l	total leaf resistance to mass exchange
R_{meso}	mesophyll resistance to mass exchange
R_s	stomatal resistance to mass exchange
R_{soil}	soil resistance to mass exchange
Re	Reynold's number
$s(z)$	SO_2 concentration at height z
Sc	Schmidt number
Sh	Sherwood number
T_a	air temperature
$u(z)$	wind speed at height z
u_m	mean wind speed inside the canopy
$u_*(z)$	friction velocity at height z inside the canopy
u_*	friction velocity measured above the canopy.
V_d	deposition velocity
z_i	height above the ground where a logarithmic wind profile is assumed.
z_0	roughness length
α	wind speed attenuation coefficient
ρ	density of air
τ	momentum transfer
ψ_m	diabatic stability function for momentum transfer
ψ_s	diabatic stability function for SO_2 transfer.

INTRODUCTION

Pollutant gases and particulates are deposited to landscapes via wet and dry pathways. Monitoring of the wet component of pollutant deposition has been continuing for many years and an extensive network of monitoring stations has been established (see Wisniewski and Kinsman, 1982). More recently, a need to monitor the dry component of pollutant deposition has been identified (e.g. Hicks *et al.*, 1982). Establishment of a dry deposition monitoring network has been delayed, relative to the establishment of the wet deposition network, since it is difficult to make continuous measurements of dry deposition to landscapes with present technology and limited resources. As an alternative, it has been proposed to estimate pollutant fluxes to terrestrial surfaces with an inferential method (Hicks *et al.*, 1982, 1985, 1987). This method involves monitoring pollutant concentrations, specific meteorological variables and surface conditions and using this information to estimate dry deposition fluxes with a numerical model.

Scientists at the Atmospheric Turbulence and Diffusion Division (ATDD) are developing and testing a hierarchy of dry deposition models as part of a dry deposition monitoring program (see Meyers and Baldocchi, 1988). The first model uses the 'big-leaf' approach (Hicks *et al.*, 1985, 1987), where deposition velocities are computed as the reciprocal of the sum of the aerodynamic, quasi-laminar and canopy resistances. An extension of the first model is a hybrid, 'big-leaf'-multilayer model (Baldocchi *et al.*, 1987). This second model treats the stomatal resistance component of the canopy with a multi-layer submodel and the aerodynamic and quasi-laminar boundary layer resistances with a 'big-leaf' submodel. More recently, we have developed two multi-layer, gaseous deposition models, which improve upon the 'big-leaf' approach. One model is based on higher-order closure principles and is reported by Meyers (1987). The other model is based on 'K-theory'. In this paper, the latter multilayer model is described. The model's sensitivity to variations in input parameters is also examined and discussed. The third objective of this paper is to examine the utility of a 'K-theory' model in predicting the deposition of SO_2 to a deciduous forest canopy. Such an examination is needed because there are some theoretical limitations associated with 'K-theory' models (Corrsin, 1974; Finnigan and Raupach, 1987); these models are only valid if the length scale of the turbulence is less than that associated with the curvature of the vertical profile of the entity of interest.

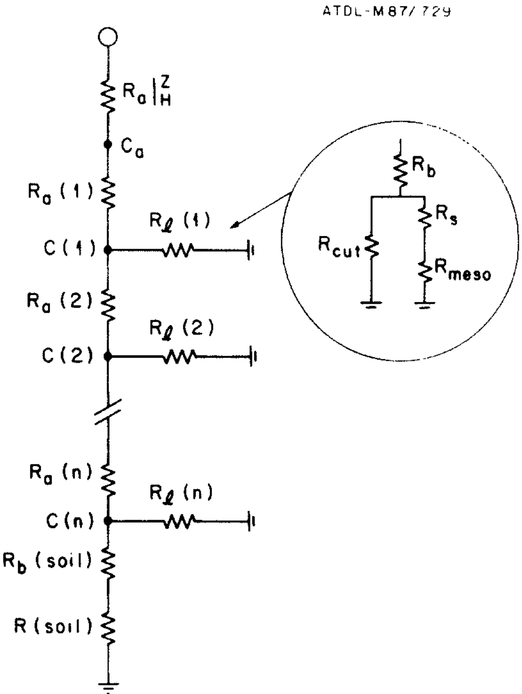

Fig. 1. Resistance network for the multi-layer resistance analog gaseous deposition model. R_a, R_l, R_b, R_s, R_{meso}, R_{cut} and R_{soil} are the aerodynamic, leaf, diffusive boundary layer, stomatal, mesophyll, cuticular and soil resistances, respectively.

THEORY

Deposition velocities and fluxes are computed using a multi-layer, 'K-theory' model, based on concepts originally proposed by Shreffler (1976, 1978), Belot *et al.* (1976) and Murphy *et al.* (1977) for gaseous deposition within plant canopies. The resistance network used to compute deposition velocities is described in Fig. 1. The primary resistances to pollutant uptake are the aerodynamic resistances above the canopy and within the vegetation (R_a), the leaf and soil boundary layer resistances (R_b and $R_b$soil), the soil resistance (R_{soil}) and the leaf resistances (R_l) due to the stomata (R_s), mesophyll (R_{meso}), and cuticle (R_{cut}).

This model is one-dimensional and is applicable over extended, homogeneous canopies in relatively uniform terrain. The model assumes that the interior of the leaf and the soil are perfect sinks for the pollutant. This model is primarily applicable for pollutant transfer to the canopy, as is the case for SO_2, O_3 and HNO_3. However, with information on soil efflux, this model can be modified and used to estimate fluxes of NO_x, N_2O, CH_4, and other trace gases with sources from the soil.

The transfer of SO_2 inside a canopy is described in terms of the balance between the flux divergence and the sink strength of the foliage:

$$dF(z)/dz = \text{sink} = -s(z)/[\Delta z\, R_l(z)], \qquad (1)$$

where $F(z)$ is the flux of SO_2 at height z, $s(z)$ is the concentration of SO_2, Δz is the thickness of the layer, and $R_l(z)$ is comprised of the leaf boundary layer resistance and the leaf resistance to SO_2 transfer. R_l is expressed on a unit leaf area basis. Near the soil surface, a constant flux layer is assumed, such that $dF(z)/dz$ equals zero and turbulent and the molecular diffusive fluxes of SO_2 into the soil are equal. The canopy deposition flux (F_{SO_2}) is obtained by integrating Equation (1) from the ground surface to the top of the canopy.

Fluxes of SO_2 inside the canopy $[F(z)]$ are computed as

$$F(z) = -K_s(z)ds(z)/dz, \qquad (2a)$$

where $K_s(z)$ is the eddy exchange coefficient for SO_2 at height z and $ds(z)/dz$ is the vertical gradient of SO_2. The flux of SO_2 into the soil is defined as

$$F(\text{soil}) = -s(z_i)/[R_b(\text{soil}) + R_{\text{soil}}], \qquad (2b)$$

where R_b(soil) is the boundary layer resistance of the soil, R_{soil} is the soil resistance to SO_2 uptake and $s(z_i)$ is the SO_2 concentration at the first level above the soil.

Eddy exchange coefficients inside the canopy are computed using momentum transfer theory (Thom, 1975). The momentum transfer $[\tau(z)]$ inside the canopy is expressed as

$$\tau(z) = -\rho K_m(z)du(z)/dz \qquad (3a)$$

$$\tau(z) = -\rho \int_0^z C_m(z)u(z)^2 a(z)dz, \qquad (3b)$$

where $K_m(z)$ is the eddy exchange coefficient for momentum transfer, ρ is air density, $C_m(z)$ is the effective drag coefficient, $u(z)$ is the horizontal wind speed and $a(z)$ is the leaf area density.

Using Equations (3a) and (3b), $K_m(z)$ is computed as

$$K_m(z) = \frac{\int_0^z C_m(z)a(z)u(z)^2\,\mathrm{d}z}{\mathrm{d}u(z)/\mathrm{d}z}. \quad (4)$$

$K_s(z)$ is assumed to equal 0.8 $K_m(z)$ due to differences in the eddy exchange coefficients for momentum and mass (see Halldin and Lindroth, 1986). Below crown closure (about 14 m), the foliage is sparse and the wind speed profile is complex; reversals in the wind speed profile occur (Baldocchi and Meyers, 1988) and yield negative K_m values [Equation (4)], which are unrealistic. Consequently, $K_s(z)$ is assumed to be constant below this level and equal to K_s at 14 m. The eddy exchange coefficients are related to the interstitial aerodynamic resistances, shown in Fig. 1, as

$$R_a(z\!:\!2, z\!:\!1) = \int_{z:1}^{z:2} \mathrm{d}z/K_m(z) \quad (5a)$$

$$\tau(z) = -\rho[u(z\!:\!2) - u(z\!:\!1)]/R_a(z\!:\!2, z\!:\!1), \quad (5b)$$

where the indices 1 and 2 refer to adjacent levels.

Within-canopy wind speeds, used to evaluate Equation (4), are computed from the relationship of Cionco (1972):

$$u(z) = u_h \exp[-\alpha(1 - z/h)], \quad (6)$$

where α is the wind speed attenuation coefficient and u_h is the wind speed at the top of the canopy. u_h is computed by extrapolating the logarithmic wind profile to canopy height:

$$u_h = u(z_r)\left[\frac{\ln(h-d) - \ln(z_0) - \psi_m[(h-d)/L]}{\ln(z_r-d) - \ln(z_0) - \psi_m[(z_r-d)/L]}\right], \quad (7)$$

where d is the zero plane displacement, ψ_m is the diabatic stability function, L is the Monin–Obukhov scale length, $u(z_r)$ is the wind speed at a reference height, z_r, above the canopy, and z_0 is the roughness length. The relationship for ψ_m is given in Kanemasu *et al.* (1979).

The effective drag coefficient of leaves in a canopy layer is different from the drag coefficient of an isolated leaf (Landsberg and Thom, 1971; Landsberg and Powell, 1973; Thom, 1975). This difference is due to the multiple sheltering by leaves and leaf orientation to the flow, which affects the contribution of bluff-body and skin-friction influences on momentum transfer. The effective drag coefficient $[C_m(z)]$ is assumed to be constant with height (Thom, 1975) and is expressed as

$$C_m(z) = C_{am}/\{\mathrm{LAI}[u_m/u(z_r)]^2\}, \quad (8)$$

where C_{am} is the bulk canopy drag coefficient, LAI is the canopy leaf area index, and u_m is the mean wind speed within the canopy. Eddy exchange coefficients, computed with a constant $C_m(z)$, have the feature of an exponential decrease with depth into the canopy (Monteith, 1973).

The mean within-canopy wind speed (u_m) is computed as

$$u_m = (1/h)\int_0^h u(z)\,\mathrm{d}z. \quad (9)$$

Equation (9) is evaluated using $u(z)$ values derived from a fourth-order polynomial fit to the normalized wind speed profile measured in a deciduous forest (Baldocchi and Meyers, 1988). A polynomial relationship is used to estimate u_m because a strong secondary wind speed maximum is observed below crown closure. This technique thus leads to a more realistic estimate of u_m and improves upon the estimates of u_m based on an exponential model for $u(z)$ (e.g. Cionco, 1972).

The leaf boundary layer resistance is the resistance to molecular diffusion through the leaf boundary layer. It is evaluated on a unit leaf area $[\mathrm{d}f(z)]$ basis as

$$R_b(z) = l/[\mathrm{d}f(z)D_s\mathrm{Sh}(z)], \quad (10)$$

where l is the characteristic length of leaves, D_s is the molecular diffusivity of the entity and Sh is the Sherwood number. Relationships for the Sherwood number, derived for flat plates, are used for leaves. These relationships are

$$\mathrm{Sh}(z) = 0.66\,\mathrm{Re}^{0.5}\,\mathrm{Sc}^{0.33} \quad (11a)$$

for laminar flow and

$$\mathrm{Sh}(z) = 0.03\,\mathrm{Re}^{0.8}\,\mathrm{Sc}^{0.33} \quad (11b)$$

for turbulent flow (Grace and Wilson, 1976). Re is the Reynolds number and Sc is the Schmidt number. The critical Re for the transition from laminar to turbulent flow ranges between 8000 and 25,000.

The air flow over a leaf is a mixed regime of turbulent and laminar flow, due to leaf flutter, leaf hairs and transition distances over leaves. Grace and Wilson (1976) found that measured values of Sh were greater than those computed with Equation (11) by a factor exceeding two. Computations of R_b are thus made by multiplying Sh by a factor of two.

The boundary layer resistance at the soil surface [R_b soil] is computed using the technique of Schuepp (1977):

$$R_b\ \mathrm{soil} = [\mathrm{Sc} - \ln(d_0/z_i)]/[ku_*(z_i)], \quad (12)$$

where d_0 is the height where molecular transfer becomes equal in magnitude to turbulent transfer ($d_0 = \nu/ku_*$), k is von Karman's constant ($k = 0.4$), $u_*(z_i)$ is friction velocity at z_i, ν is kinematic viscosity and z_i is the height above the ground where a logarithmic wind profile is assumed. Equation (12) is evaluated by assuming that z_i is equal to the height of the first layer or grid point above the soil surface. Friction velocity of the lowest layer was evaluated as ($kz_i\,\partial u/\partial z$).

The leaf resistance (R_l) is comprised of serial resistances exerted by the stomata, boundary layer and leaf mesophyll, which are, in turn, parallel with the cuticular and leaf boundary layer resistances. The leaf resistance (R_l) is computed using the following equations:

$$R_{ss}(z) = R_s(z) + R_{meso} \quad (13a)$$

and

$$1/R_l(z) = 1/[R_{ss}(z) + R_b(z)] + 2/[R_{cut} + R_b(z)], \quad (13b)$$

where R_s, R_{meso}, R_b and R_{cut} are the stomatal, mesophyll, leaf boundary layer and cuticle resistances, respectively. All resistances are expressed on a unit leaf area basis. The coefficients in Equation (13b) arise because leaves of many of the species in a temperate deciduous forest (e.g. *Quercus* species) are hypostomatous (stomata are only on one side of the leaf), whereas the cuticular and boundary layer resistances are effective on both sides of leaves.

Stomatal resistance (R_s) is a complex function of environmental and physiological variables, including photosynthetically active radiation (PAR), air temperature (T_a), humidity, and plant water status (Jarvis, 1976). The stomatal resistance, at discrete layers in the canopy, is computed by coupling a canopy radiative transfer model (Norman, 1979) with the multiplicative leaf stomatal resistance model of Jarvis (1976). This complexity is required because R_s responds non-linearly to light, which varies with height in the canopy and on the sunlit and shaded leaf fractions. Computations of PAR profiles and sunlit and shaded leaf area are based on the assumption that the spatial leaf distribution is random and that the leaf inclination distribution is spherical. Details on the stomatal resistance submodel are provided in Baldocchi *et al.* (1987) and Baldocchi and Hutchison (1986).

COMPUTATIONAL PROCEDURES

Fluxes and concentration profiles are computed with an iterative numerical technique. First, Equation (1) is evaluated with an assumed initial SO_2 profile. The Adams–Bashforth method (Conte and de Boer, 1972), is then used to solve Equation (1) and compute a vertical profile of $F(z)$. Next, vertical profiles of concentration gradients $[ds(z)/dz]$ are computed from Equation (2a). The concentration gradients are then integrated with respect to z to obtain a new concentration profile. In order to attain numerical stability, a digital recursive filter is applied to the new estimates of SO_2 concentration.

Boundary conditions are reset with each iteration. The SO_2 concentration at the top of the canopy is redefined as

$$s(h) = s(z_r) + (F_{SO_2} k u_*)[\ln(z_r/h) - \psi_s], \quad (14)$$

where F_{SO_2} is the integrated canopy SO_2 flux, u_* is the friction velocity measured above the canopy, ψ_s is the diabatic stability function for SO_2 and $s(z_r)$ is the SO_2 concentration at a reference level above the canopy. F_{soil} is re-evaluated in terms of the new SO_2 concentration near the soil [Equation (2b)]. These steps are repeated until desired convergence, which typically takes 40–70 iterations.

Deposition velocities are computed as

$$V_d = F_{SO_2}/s(z_r). \quad (15)$$

The parameter coefficients used to compute SO_2 fluxes and deposition velocities are presented in Table 1. Sensitivity tests are computed assuming that the ambient SO_2 concentration is 20 ppb. The time of the simulations is assumed to be day 180 at 1200 h. The cuticle resistance used in the computations is based on the assumption that the foliage is dry. The meteoro-

Table 1. Parameters and values used to compute SO_2 deposition velocities to a deciduous oak forest

Parameter	Unit	Value	Reference
Canopy height (h)	m	22.5	Hutchison *et al.* (1986)
Leaf area index (LAI)	—	4.9	Hutchison *et al.* (1986)
Leaf length (l)	m	0.1	Measured
Zero plane displacement (d)	m	19(0.84 h)	Baldocchi and Meyers (1988)
Roughness length (z_0)	m	2.2 (0.1 h)	Dolman (1986)
Stability (z/L)		−0.001	
SO_2 diffusivity (D_s)	$mm^2\,s^{-1}$	12	
Bulk canopy drag coefficient (C_{am})	—	0.016	Verma *et al.* (1986)
Wind extinction coefficient (α)	—	6.95	Baldocchi and Meyers (1988)
Leaf reflectivity (ρ, PAR)	—	0.11	Baldocchi *et al.* (1985)
Leaf transmissivity (τ, PAR)	—	0.16	Baldocchi *et al.* (1985)
Soil reflectivity (ρ_s, PAR)	—	0.033	Baldocchi *et al.* (1985)
Minimum stomatal resistance (r_{sm})	$s\,m^{-1}$	150	Baldocchi and Hutchison (1986)
Stomatal resistance curvature coefficient (b_{rs})	$W\,m^{-2}$	40	Meyers and Baldocchi (1988)
Cuticle resistance (R_{cut})	$s\,m^{-1}$	3000	Spedding (1969)
Soil resistance (R_{soil})	$s\,m^{-1}$	300	McMahon and Denison (1979)
Mesophyll resistance (R_{meso})	$s\,m^{-1}$	10	Hosker and Lindberg (1982)

logical input variables needed for the model computations include wind speed (u), friction velocity (u_*), Monin–Obukhov scale length (L), air temperature (T_a), and solar radiation (R_g). Structural inputs include leaf area index and the vertical distribution of leaf area. The canopy was assumed to be well watered and recent experiments show that there is no vapor pressure deficit effect on R_s of oak leaves, which agrees with the data of Appleby and Davies (1983). Diffuse and direct PAR are computed from measurements of R_g using the model of Weiss and Norman (1985).

Computations of SO_2 fluxes and component resistances are performed by dividing the canopy into 70 layers; numerical tests show that no significant improvement in the estimate of deposition fluxes and component resistances is obtained by dividing the canopy into more layers.

MATERIALS AND METHODS

The structure of the deciduous forest canopy is described in Hutchison *et al.* (1986). A continuous function of the vertical profile of leaf area density [$a(z)$] is determined using the Beta distribution (Massman, 1982) from actual measurements of leaf area index.

The multi-layer SO_2 deposition model is tested against experimental data obtained over a fully-leafed, well-watered, deciduous forest near Oak Ridge, TN (see Matt *et al.*, 1987). Architectural characteristics of the canopy are described in Hutchison *et al.* (1986). Measurements of SO_2 flux were made in July 1985 with the eddy correlation technique. SO_2 fluxes were determined as the mean covariance between fluctuations in vertical wind velocity and SO_2 concentration. Deposition velocities were computed as the ratio of SO_2 flux and concentration. The instrumentation used to measure SO_2 fluxes was positioned about 9 m above the zero plane displacement. The instruments included a three-dimensional sonic anemometer and a fast-response Meloy sulfur sensor. Many modifications were made on the SO_2 flame photometric sensor in order to make it applicable for eddy correlation measurements (see Matt *et al.*, 1987). The response time was decreased to less than 0.7 s by shortening the internal plumbing, precisely regulating the hydrogen fuel with a mass flow controller, regulating the sample flow with a critical orifice and installing a faster burner block assembly. Nonlinearities at low levels of SO_2 were minimized by doping the hydrogen fuel with SF_6 to effectively raise the baseline concentration of S being detected by the sensor. Mean ambient SO_2 concentrations, as needed for the deposition velocity computations, were measured independently with a Monitor Labs u.v. pulse fluorescence SO_2 monitor. Data used for the comparison were chosen from periods when the SO_2 concentration exceeded 3 ppb and the time rate of change in SO_2 concentration was less than 20% per 1/2 h. The meteorological conditions encountered during the experiment were moderate; temperatures ranged between 25 and 30°C, solar radiation ranged between 300 and 800 W m^{-2}, wind speeds above the canopy range between 1 and 4 m s^{-1} and SO_2 concentrations ranged between 3 and 20 ppb [see Meyers and Baldocchi (1988) for specific conditions].

RESULTS

(a) *Model computations*

The vertical distribution of leaf area density [$a(z)$] and the computed profile of SO_2 flux density, normalized by $F_{SO_2}[F(z)/(\Delta z\, F_{SO_2})]$, are presented in Figs 2a and 2b. Both the leaf area and the SO_2 uptake are concentrated in the upper 20% of the canopy. At 18 m, over 95% of the total canopy SO_2 deposition flux and about 70% of the leaf area is observed. It can thus be inferred from these data that any poor assumption

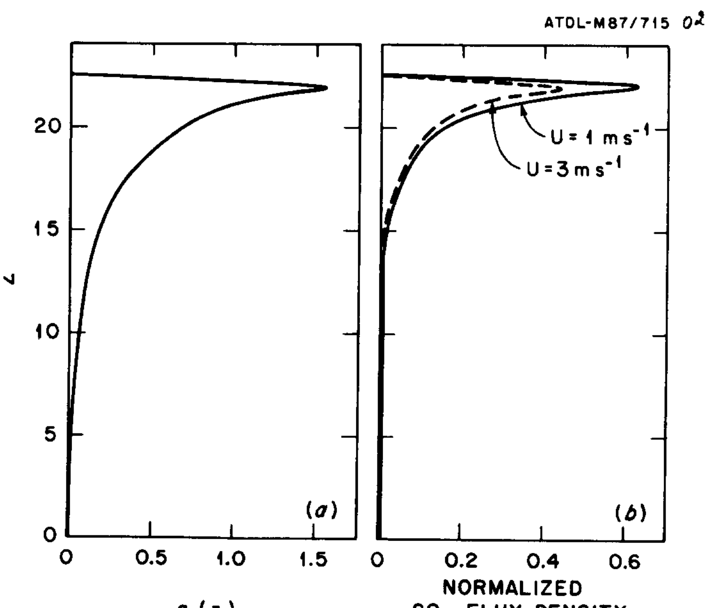

Fig. 2. (a) Vertical profile of leaf area density [$a(z)$]; and (b) the computed vertical profile of SO_2 flux density [$F(z)/\Delta z F_{SO_2}$] for wind speed regimes of 1 and 3 m s^{-1}. These computations are based on the standard conditions presented in Table 1 for a deciduous forest.

regarding turbulence exchange processes in the lower 18 m of the deciduous forest canopy will be relatively inconsequential since little SO_2 exchange occurs in the lower region.

Vertical profiles of SO_2 concentration ($[SO_2]$) are computed for different wind speed regimes (Fig. 3). These computed profiles show that the drawdown in $[SO_2]$, inside a deciduous forest, is generally substantial and increases with decreasing wind speed. The shapes of these profiles and the magnitude of the associated drawdowns resemble the theoretical $[SO_2]$ profiles computed by the 'K-theory' model of Murphy *et al.* (1977) in a coniferous forest canopy. On the other hand, smaller theoretical SO_2 drawdowns are computed in a forest canopy with a higher-order closure model (Meyers, 1987).

Utilization of the computed $[SO_2]$ profile in computing canopy SO_2 deposition, however, does not have a great impact on the estimation of SO_2 uptake in a deciduous forest (Fig. 4). A difference of less than 5 % in the estimation of SO_2 flux typically occurs when comparing SO_2 flux estimates, based on simulated profiles, against those made by assuming a constant $[SO_2]$ profile, equal to the SO_2 concentration measured above the canopy. This small impact is primarily an artifact of canopy structure; although the leaf area is concentrated above 0.8 h, where it provides a strong biological sink, the drawdown in $[SO_2]$ in this region is generally small because turbulent mixing is also great. However, under different circumstances the determination of the concentration profile is crucial in estimating the deposition flux correctly (see Bache, 1986a).

Estimates of deposition velocity are sensitive to environmental and physiological conditions and to the parameters used to compute the component resis-

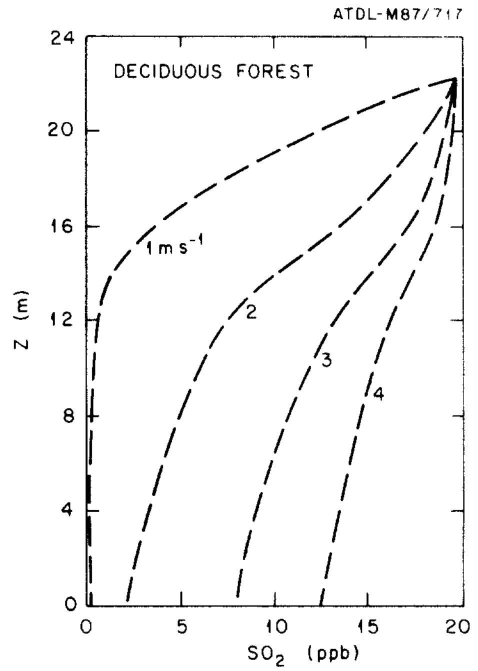

Fig. 3. The computed vertical profile of SO_2 concentration at different wind speed regimes. These computations are based on the standard conditions presented in Table 1 for a deciduous forest.

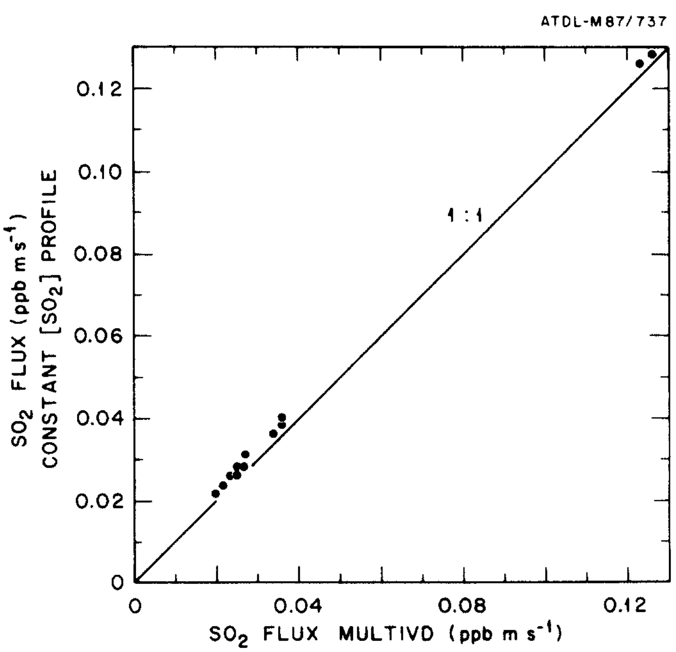

Fig. 4. A comparison of computed SO_2 fluxes with the multi-layer model by computing the $[SO_2]$ profile and by assuming a constant profile of $[SO_2]$ inside the canopy. These data are based on environmental conditions measured in the study of Matt *et al.* (1987).

tances. The sensitivity of V_d to environmental conditions and modeling parameters is examined below. Figure 5 shows the response of computed SO_2 deposition velocities (V_d) to varying PAR and wind speed (u) regimes under moderate temperatures ($T_a = 25\,°C$) and well-watered conditions. SO_2 deposition velocities range from about 0.1 to $0.9\ cm\,s^{-1}$ and increase nonlinearly with increasing PAR and u. The range of V_d values in Fig. 5 correspond with values of SO_2 V_d measured over natural, vegetated surfaces (see Fowler, 1978; Hicks, 1984; Matt *et al.*, 1987).

The stomatal resistance of the canopy (R_{sc}) influences the estimate of the SO_2 deposition velocity to varying degrees (Fig. 6), and is mediated by the degree of turbulent mixing. For example, under strong wind speed regimes ($u = 4\ m\,s^{-1}$), aerodynamic resistance is much smaller than the canopy stomatal resistance. Thus, V_d is strongly inhibited with increasing R_{sc}. On the other hand, under low wind speed regimes ($u = 0.5\ m\,s^{-1}$), the aerodynamic and stomatal resistances are relatively similar, which leads to R_{sc} having a smaller influence on V_d.

The response of leaf stomatal resistance (R_s) to PAR can be expressed, after Turner and Begg (1973), as

$$R_s(z) = r_{sm} + b_{rs} r_{sm}/\mathrm{PAR}(z), \tag{16}$$

where r_{sm} is the minimum stomatal resistance under optimal conditions and b_{rs} is a curvature coefficient,

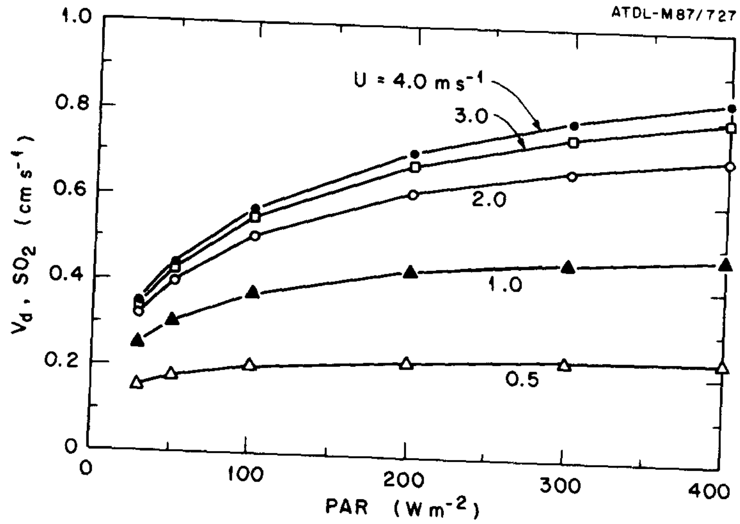

Fig. 5. Sensitivity of the multi-layer, resistance analog deposition model to photosynthetically active radiation (PAR) and wind speed (u). These computations are based on the standard conditions presented in Table 1 for a deciduous forest.

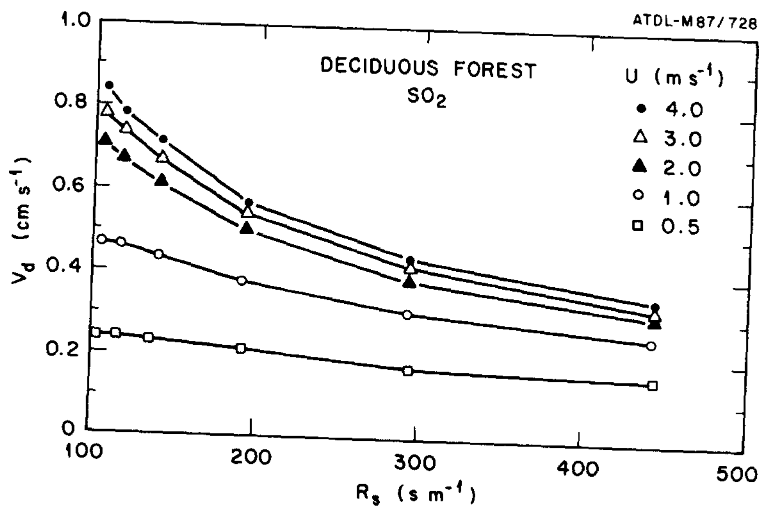

Fig. 6. Sensitivity of the multi-layer, deposition model to variations in canopy stomatal resistance under different wind speed regimes. These computations are based on the standard conditions presented in Table 1 for a deciduous forest.

defined as the PAR level at twice the minimum stomatal resistance. Values of the minimum stomatal resistance range widely in the literature (Korner *et al.*, 1979). For example, minimum stomatal resistances for *Quercus* species typically range between 100 and 200 s m^{-1} (Hinckley *et al.*, 1978; Baldocchi and Hutchison, 1986). Elias (1979), however, reports more extreme values, exceeding 600 s m^{-1}.

SO_2 deposition velocities are very sensitive to values of the minimal stomatal resistance (r_{sm}) (Fig. 7). V_d values increase by about 100–200% with increasing PAR (Fig. 7a) and by about 50–200% with increasing wind speed conditions (Fig. 7b), as r_{sm} decreases from 200 to 50 s m^{-1}. The maximum V_d value decreases from 1.6 to 0.6 cm s^{-1} as r_{sm} increases from 50 to 200 s m^{-1}. Deposition velocities are most sensitive to changes in PAR and u when r_{sm} is small and are least sensitive when r_{sm} is large.

The PAR curvature coefficient for stomatal resistance (b_{rs}) strongly influences the canopy stomatal resistance, and thus V_d. Figure 8 shows that V_d increases by about 25–60% as b_{rs} decreases from 40 to 10 W m^{-2}. Deposition velocities are most sensitive to b_{rs} under low PAR regimes since the relationship between leaf stomatal resistance and PAR exhibits great curvature under such conditions.

The aerodynamic component of V_d is influenced by the parameterization of such factors as canopy drag coefficient [C_{am}, Equation (8)] and the wind speed attenuation coefficient [α, Equation (6)]. Sensitivity

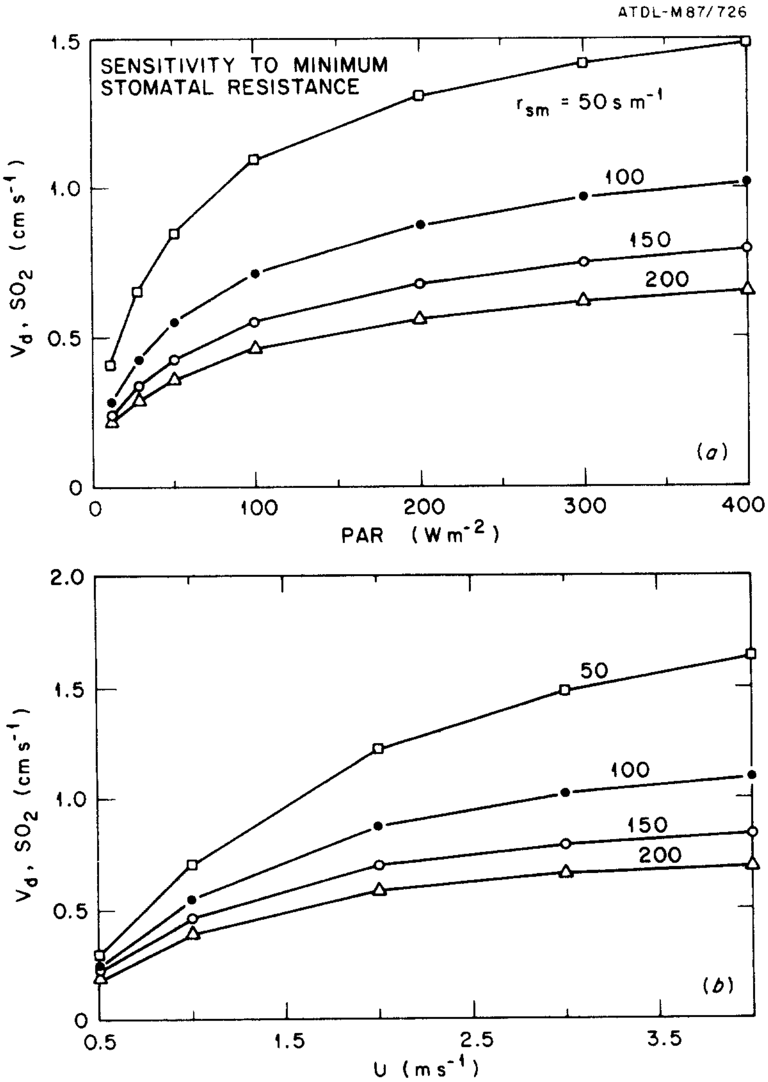

Fig. 7. (a) Sensitivity of SO_2 deposition velocities to variations in the minimum stomatal resistance (r_{sm}) and photosynthetically active radiation (PAR). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and wind speed equals 3 m s^{-1} at 30 m. (b) Sensitivity of SO_2 deposition velocities to variations in the minimum stomatal resistance (r_{sm}) and wind speed (u). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and PAR equals 400 W m^{-2}.

tests suggest that V_d is generally not as influenced by variations in C_{am} (Fig. 9), as it is by variations in the stomatal resistance parameters. For example, V_d increases by less than 15% with a tenfold increase in C_{am} (Fig. 9a), over a wide range of PAR values. V_d, however, is more sensitive to C_{am} under low wind speed conditions (Fig. 9b); V_d increases by over 100% as C_{am} increases from 0.016 to 0.2 at u equal 0.5 m s^{-1}.

Computations of V_d are also relatively insensitive to the wind speed attenuation coefficient (Fig. 10). V_d increases by less than 0.16 cm s^{-1} as the wind speed attenuation coefficient decreases from 6.95 to 3, a range of values characteristic of many forest canopies (Cionco, 1978), with increasing PAR and wind speed. This suggests that errors in the parameterization of the within-canopy wind speed profile do not strongly influence the estimate of gaseous deposition to a deciduous forest.

Leaf length affects the leaf diffusive boundary layer resistance, as shown in Equation (10). As with the aerodynamic parameters discussed above, considerable variations in leaf length have only a minor influence on the SO_2 V_d for a deciduous forest (Fig. 11). A leaf length variation of 0.05 m about a mean leaf length of 0.10 m has less than a 15% influence on V_d.

Deposition velocities are relatively insensitive to values of the cuticle resistance (R_{cut}) exceeding 3000 s m^{-1} (Fig. 12). However, V_d becomes increasingly sensitive to R_{cut} as it approaches zero, a condition which can occur when the canopy is wet and the moisture is unsaturated with SO_2. For example, V_d increases by 40–100% as R_{cut} decreases from 1500 to 500 s m^{-1}. Under these surface conditions the magnitude of the cuticle resistance approaches that of the stomatal and aerodynamic resistances and thus is another major pathway for deposition.

(b) *Model test*

A comparison between measured SO_2 fluxes and those computed with the multi-layer model is presented in Fig. 13. The associated statistics are presented in Table 2. These data show that there is no significant difference between measured and calculated SO_2 fluxes on the 10% level of significance. The computed and measured fluxes are also highly correlated ($r = 0.95$; $r^2 = 0.90$, where r is the correlation coefficient and r^2 is the coefficient of determination), suggesting that the model computations account for much of the variance in SO_2 uptake caused by the influencing environmental and physiological factors.

The comparison between measured and computed SO_2 deposition velocities (Fig. 14; Table 2) also shows that there is no significant difference between measured and computed values on the 10% level of significance. The correlation coefficient between measured and computed V_d values, however, is lower than that observed for SO_2 fluxes ($r = 0.62$; $r^2 = 0.38$). This lower correlation coefficient results from several factors. First, deposition velocities are measured and calculated with less accuracy than deposition fluxes, since an additional source of error is introduced via the measurement of the absolute SO_2 concentration. Second, SO_2 sensors are inherently noisy instruments and, thus, cause substantial run-to-run variability. Therefore, many experimental runs are needed to construct a meaningful estimate of the deposition flux (see Wesely and Hart, 1985).

Based on the current level of technology, we can reasonably conclude that deposition velocities, derived from long-term averages, are valid. On the other hand, we cannot make any such conclusions regarding short-term estimates of deposition velocity, as derived from this model.

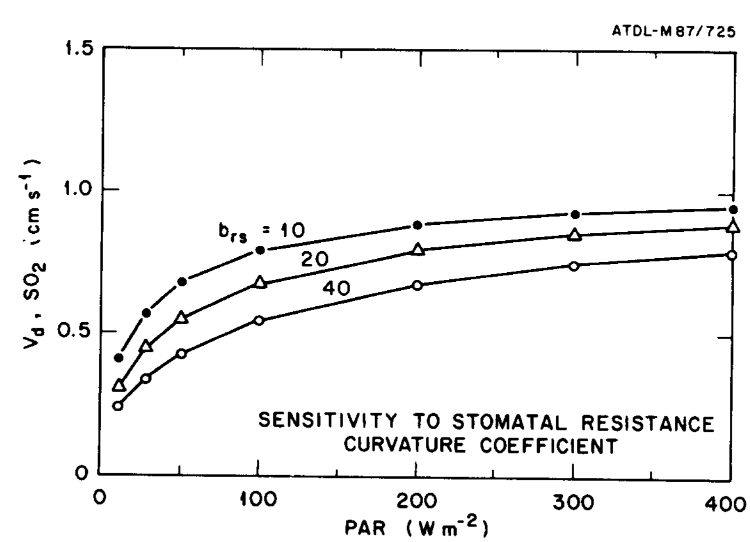

Fig. 8. Sensitivity of SO_2 deposition velocities to variations in the stomatal resistance curvature coefficient (b_{rs}) and PAR. These computations are based on the standard conditions presented in Table 1 for a deciduous forest and u equal to 3 m s^{-1} at 30 m.

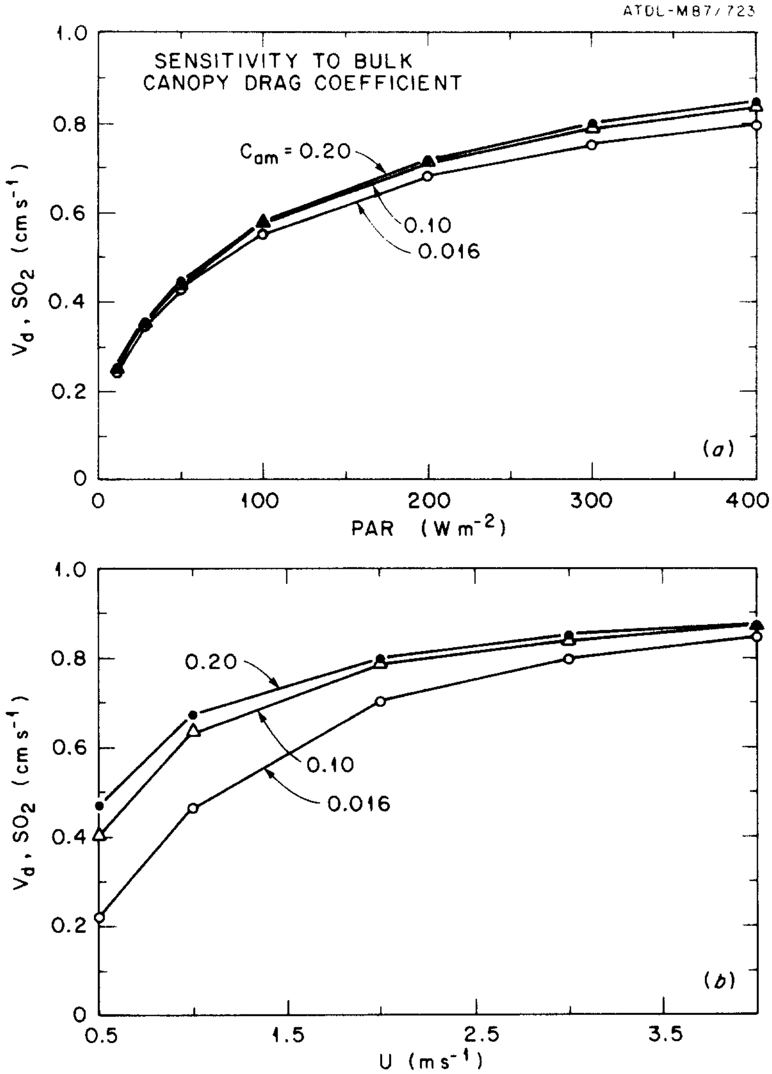

Fig. 9. (a) Sensitivity of SO_2 deposition velocities to variations in the bulk canopy drag coefficient (C_{um}) and photosynthetically active radiation (PAR). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and wind speed equals 3 m s^{-1} at 30 m. (b) Sensitivity of SO_2 deposition velocities to variations in the bulk canopy drag coefficient (C_{um}) and wind speed (u). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and PAR equals 400 W m^{-2}.

DISCUSSION

A hierarchy test of various deposition models by Meyers and Baldocchi (1988) shows that the multi-layer, 'K-theory', gaseous deposition model, presented here, significantly improves upon the hybrid multi-layer, 'big-leaf' deposition model presented earlier by the author and other colleagues (Baldocchi *et al.*, 1987). This improvement in model performance is attributed to a more realistic and improved treatment of the canopy architecture, the canopy microclimate, the SO_2 sink-strength, its sink-location and the leaf and soil boundary layer resistances, as compared with the 'big-leaf' approach. Bache (1986a) also shows that a multi-layer deposition model improves upon a 'big-leaf' model as the surface resistance becomes progressively small, but when this resistance is relatively greater than the boundary layer resistances. The major limitation in utilizing a multi-layer model is the need for additional model parameters and profile measurements of canopy structure, which are often difficult to obtain.

These computations of SO_2 fluxes are based on 'K-theory' (Monteith, 1973). The use of 'K-theory' to simulate within-canopy turbulent transfer has been criticized because within-canopy turbulent transfer is an intermittent process, predominantly driven by large eddies (Denmead and Bradley, 1985). Consequently, it

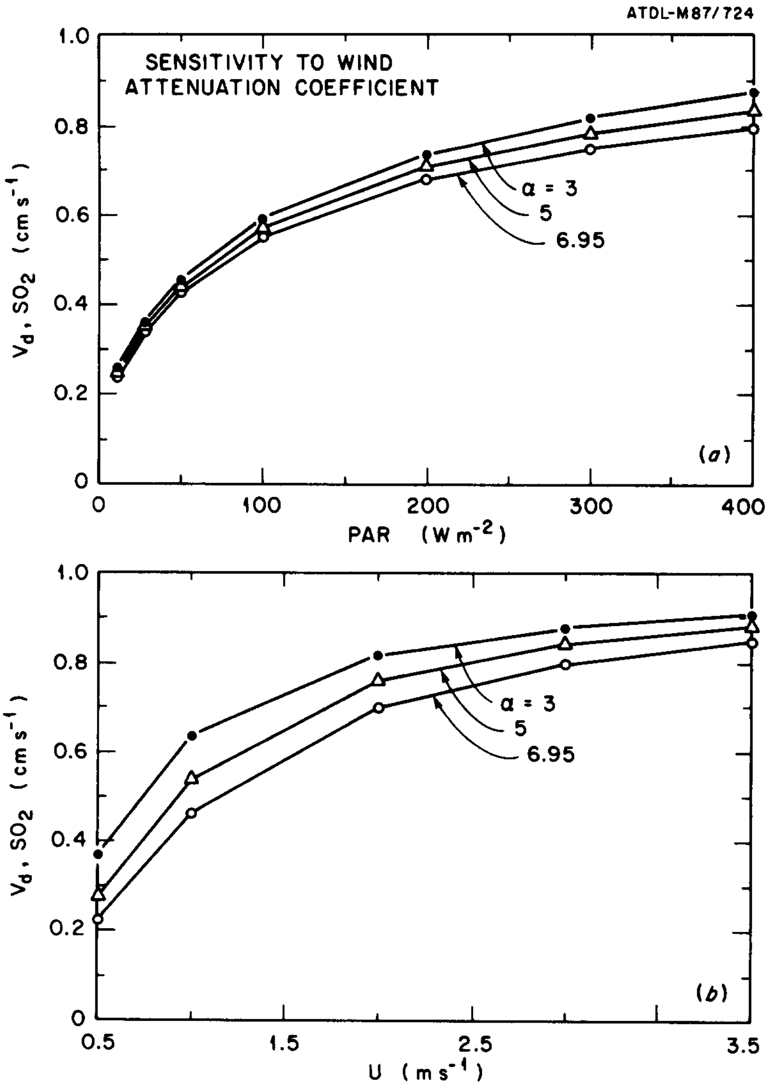

Fig. 10. (a) Sensitivity of SO_2 deposition velocities to variations in the wind speed attenuation coefficient (α) and photosynthetically active radiation (PAR). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and wind speed equals $3\,m\,s^{-1}$ at 30 m. (b) Sensitivity of SO_2 deposition velocities to variations in the wind speed attenuation coefficient (α) and wind speed (u). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and PAR equals $400\,W\,m^{-2}$.

has been suggested that turbulent transfer cannot be described well by mean profiles of eddy exchange coefficients (K) and the entity of interest (Legg and Monteith, 1975; Finnigan, 1985; Finnigan and Raupach, 1987).

On the other hand, 'K-theory' models make reasonable estimates of turbulent exchange if the turbulent length scales are smaller than the distance associated with changes in the vertical gradient of the entity (Corrsin, 1974). Recently, Bache (1986b) showed that 'K-theory' models simulate wind speed profiles well in the upper portion of a canopy, but do not simulate the secondary wind speed maximum deep inside the canopy; this deficiency is considered to be relatively inconsequential since little transfer occurs deep inside the canopy.

Examination of the [SO_2] profiles (Fig. 3) show that the curvature of the concentration gradients is rather weak in the upper portion of the canopy, where most of the SO_2 exchange occurs. This evidence, combined with the work of Bache (1986b) concerning wind speed profiles, suggests that 'K-theory' models may be valid for estimating SO_2 exchange in tall vegetation because the length scales of the turbulence are probably smaller than the distances associated with changes in the concentration and wind speed gradients.

The limitations associated with the use of 'K-theory' to compute deposition fluxes are also less severe if the

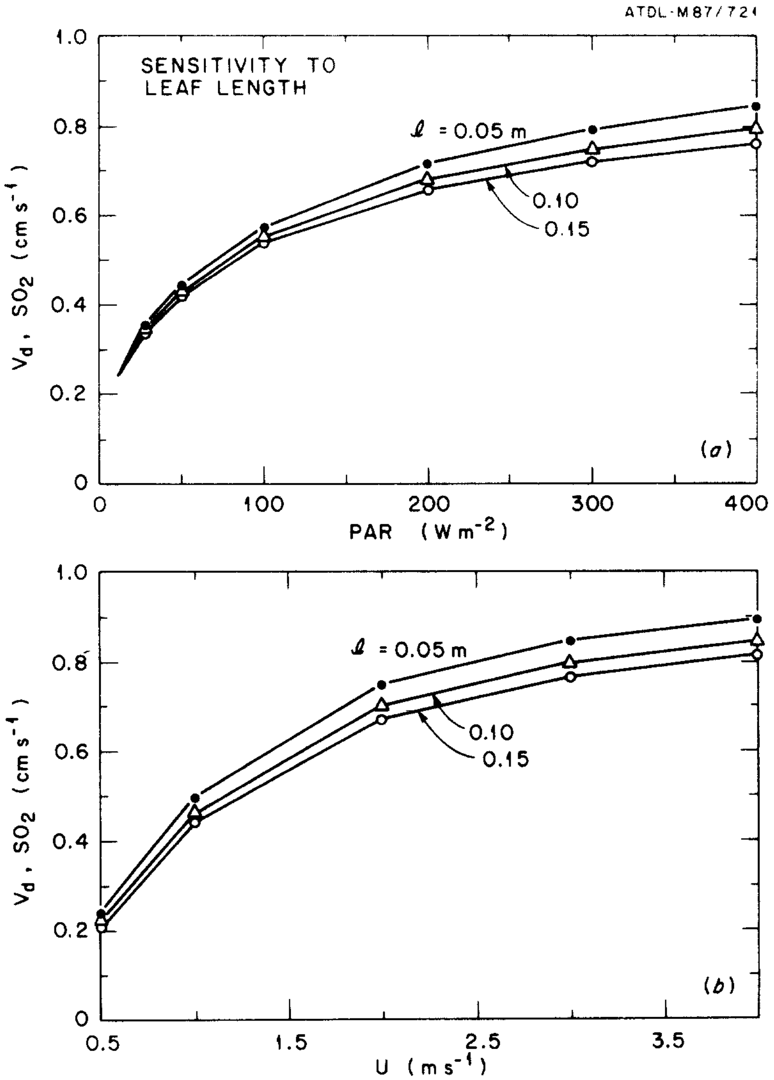

Fig. 11. (a) Sensitivity of SO_2 deposition velocities to variations in the leaf length (l) and photosynthetically active radiation (PAR). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and wind speed equals 3 $m\ s^{-1}$ at 30 m. (b) Sensitivity of SO_2 deposition velocities to variations in the leaf length (l) and wind speed (u). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and PAR equals 400 $W\ m^{-2}$.

canopy aerodynamic resistance is much smaller than the canopy stomatal or surface resistance. This condition generally holds for the case of turbulent transfer above and within forest canopies since the ratio between aerodynamic and canopy stomatal resistances is about 1 : 5 (Jarvis, 1981; Verma *et al.*, 1986). Other supporting evidence on using 'K-theory' to compute SO_2 deposition in a deciduous forest is provided by the strong sensitivity of SO_2 V_d to canopy stomatal resistance and the relative insensitivity to the aerodynamic and diffusive boundary layer resistances.

One may, therefore, conclude that the ability to model gaseous deposition in tall, aerodynamically rough canopies is highly dependent on the quality and accuracy of the parameterization of the canopy stomatal resistance. These conditions do not hold with shorter agricultural crops or for cases when the canopy stomatal or surface resistance is not the controlling factor (see Bache, 1986a), as is the case with HNO_3 deposition.

The resistance-analog or 'K-theory' approach has been used in many dry deposition models (Waggoner, 1975; Shreffler, 1976; Belot *et al.*, 1976; Murphy *et al.*, 1977; Unsworth, 1980; Wiman and Argen, 1985). The dry deposition models available in the literature, however, do not use a detailed stomatal resistance submodel or canopy radiative-transfer submodel to estimate the surface resistance. At best, these models only consider the effect of light on stomatal resistance through the use of empirical relationships for light-

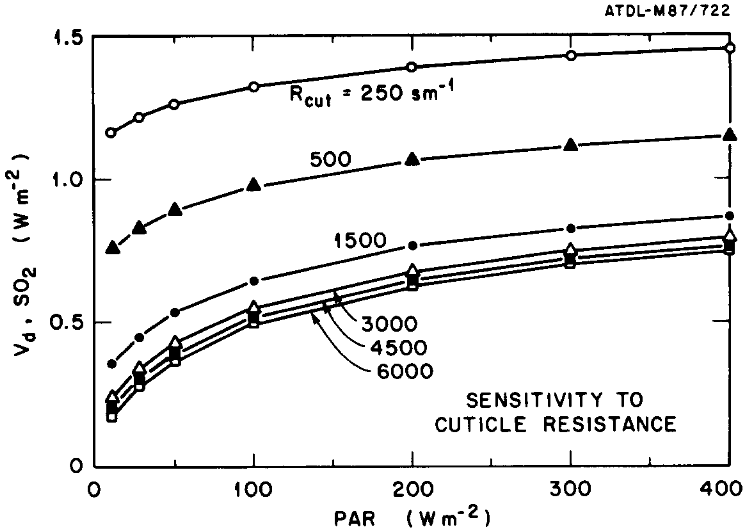

Fig. 12. Sensitivity of SO_2 deposition velocities to variations in the cuticle resistance (R_{cut}) and photosynthetically active radiation (PAR). These computations are based on the standard conditions presented in Table 1 for a deciduous forest and wind speed equals 3 m s^{-1} at 30 m.

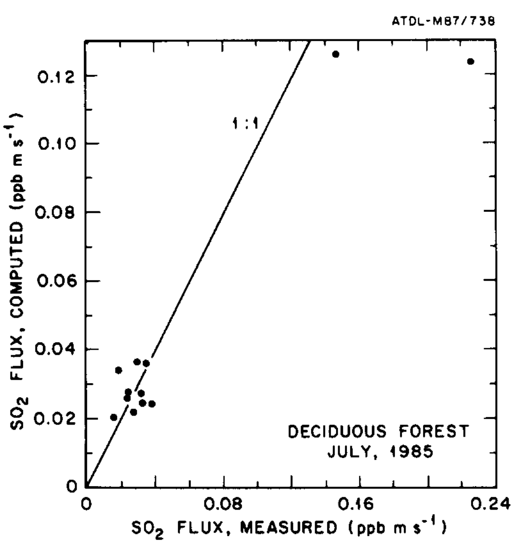

Fig. 13. Comparison between SO_2 fluxes measured and computed with the multi-layer, resistance-analog deposition model (MULTIVD). These data are from a well-watered, fully-leafed deciduous forest, near Oak Ridge, TN. The data were obtained during July 1985.

attenuation in plant canopies (Waggoner, 1975; Murphy *et al.*, 1977). The model described here improves upon these earlier dry deposition models through the use of a more detailed theory to describe the sink term, due to stomatal resistance (e.g. Norman, 1979, 1982; Baldocchi and Hutchison, 1986).

The estimation of canopy stomatal resistance (R_{sc}) in a deciduous forest has been tested by Baldocchi and Hutchison (1986), using different canopy-radiative transfer models. They found that R_{sc}, computed with a spherical Poisson canopy radiative transfer model (e.g. Norman, 1979), underestimates canopy stomatal conductance by about 10%, when compared with estimates based on measured profiles of PAR. The parameterization of canopy stomatal resistance can be improved in a deciduous forest in several ways. For example, the estimation of vertical profiles of sunlit and shaded leaf area and the flux density of PAR on those leaves can be improved by considering: (a) penumbral effects; (b) the effects of clumped foliage, and (c) vertical variations in leaf inclination angles and leaf optical properties. Stomatal resistance is also sensitive to leaf temperature. Leaf energy lance considerations can also be incorporated into the model to simulate the leaf temperature effect on stomatal resistance more accurately (see Meyers, 1987). Measurements of leaf potential should also be provided as a model input to quantify the effects of water stress on the uptake of gaseous pollutants.

The sensitivity tests showed a great dependence of V_d on r_{sm} and b_{rs}. This result is somewhat disheartening since r_{sm} of many ecological groups can vary by an order of magnitude (Korner *et al.*, 1979) and few data on b_{rs} exist in the literature. Accurate computations of V_d with this model may need an on-site determination of r_{sm} and b_{rs} before the model is employed.

Computations of SO_2 V_d are also sensitive to cuticle resistance when this resistance is small, as when the canopy is wet. In order to accurately parameterize the cuticle resistance of a wet canopy we need to know how much of the canopy is wet, how much water is on the canopy and how saturated is the surface water with SO_2. Research on these topics is encouraged.

The treatment of the leaf boundary layer resistance is somewhat limited by use of engineering relation-

Table 2. Statistics on the comparison of SO_2 deposition velocities and flux measurements with values computed with the multi-layer, resistance analog model (MULTIVD). Data are representative of a deciduous oak forest near Oak Ridge, TN

	Measured V_d (cm s^{-1})	MULTIVD V_d (cm s^{-1})	Measured flux (ppb m s^{-1})	MULTIVD flux (ppb m s^{-1})
Mean	0.69	0.62	−0.054	−0.044
Standard error	0.076	0.021	0.018	0.011
Sample periods	12		12	

Paired Student's t-test	V_d	Flux
Degrees of freedom	11	11
Mean difference	−0.074	−0.010
Standard error	0.067	0.0087
Student's t-value	−1.09	−1.15
Significance level	0.15	0.14
Coeff. of determination	0.38	0.90

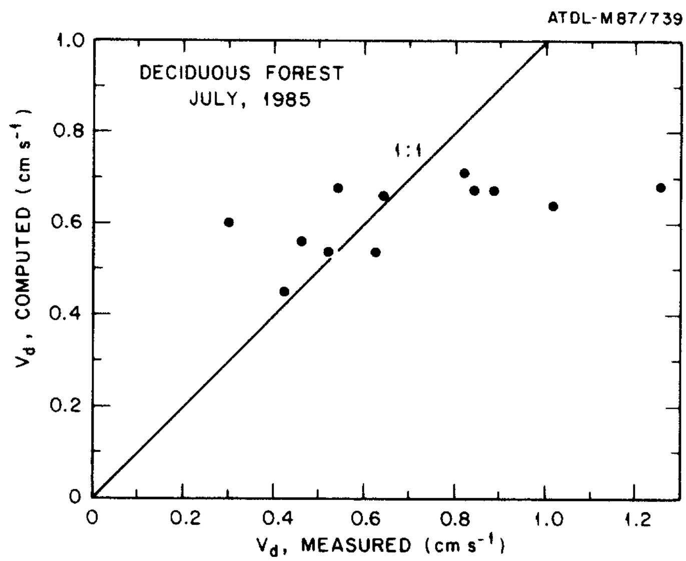

Fig. 14. Comparison between SO_2 deposition velocities measured and computed with the multi-layer, resistance analog deposition model (MULTIVD). These data are from a well-watered, fully-leafed deciduous forest, near Oak Ridge, TN. The data were obtained during July 1985.

ships developed for flow over flat plates. Natural variations in leaf size, orientation and pubescence, leaf flutter, the nature of the flow (i.e. turbulent vs laminar, forced vs free convection) and many other factors affect the leaf boundary layer (Grace and Wilson, 1976). Fortunately, this resistance is small in comparison with the other resistances and thus does not contribute to a large error in the estimate of SO_2 exchange to the forest. However, Bache (1986a) shows that its role is quite important when the surface resistance approaches zero.

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