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Numerical Study on Dry Deposition Processes in Canopy Layer

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ABSTRACT

A coupling model between the canopy layer(CL) and atmospheric boundary layer (ABL) for the study of dry deposition velocity is developed. The model consists of six parts: chemical species conservation equation including absorptive factor; the species uptake action including detailed vertical variation of absorptive element in CL; momentum exchange in CL which is represented by a first-order closure momentum equation with an additional larger-scale diffusive term; momentum exchange in ABL which is described by a complete set of the ABL turbulent statistic parameters; absorptivity (or solubility or reflection) at the surface including effects of the physical and chemical characters of the species, land type, seasonal and diurnal variations of the meteorological variables; and deposition velocity derived by distributions of the species with height in CL. Variational rules of the concentration and deposition velocity with both height and time are simulated with the model for both corn and forest canopies. Results predicted with the bulk deposition velocity derived in the paper consist well with experimental data.

I. INTRODUCTION

In many areas it is believed that one of the primary removal mechanisms is dry deposition onto the earth's surface as a result of ground absorption by the soil, forest, vegetation, body of water. The consequence of deposition of species to vegetable or forest surfaces is perceived by the many to be one of the major environmental problems of the times. The dry deposition has become the subject of widespread scientific attention over the past ten years (Chang et al., 1987 and Hicks et al., 1988). An important task confronting the atmosphere-surface exchange community is therefore to develop better methods to quantify flux densities of gaseous species to landscapes, both for inclusion in larger scale numerical models and for use in interpreting air chemistry observations made in areas of special interest.

The present model differs considerably in various aspects from earlier canopy models. First, momentum exchange in CL is based on the more realistic first-order closure momentum equation with an additional larger-scale diffusive term, the closure parameterizations are introduced not just for the sake of simplifying the computation of models using higher-order closure conditions, but also for enhancing the physical understanding of turbulent exchange processes in CL. Second, the model can simulate complete variations of uptake processes with height in CL, vertical structures of uptake factors are usually neglected by earlier models. Third, the model simulates combining effect of turbulent exchanges above and in CL, CL absorption, surface absorption and reflection, atmospheric stability, and diurnal variation of meteorological elements on dry deposition processes. Fourth, complete ABL turbulent statistic parameters are adopted.

II. COUPLING MODEL

1. *Chemical Species Conservation Equation*

Two-dimensional conservation equation for a scalar quantity that is transported through ABL and CL may be expressed by

$$\frac{\partial C}{\partial t}+U\frac{\partial C}{\partial x}=K_x\frac{\partial^2 C}{\partial x^2}+\frac{\partial}{\partial z}\left(K_z\frac{\partial C}{\partial z}\right)-\lambda C+Q\ , \tag{1}$$

where x and z refer to the downwind and vertical coordinates, K_x and K_z are horizontal and vertical eddy diffusive coefficients, respectively, U represents mean wind speed in x-direction, C is species concentration, and Q is source term. If $z \leqslant H$, $\lambda = B_a\ U$ and $z > H$, $\lambda = 0$. H is CL depth, B_a is an absorptive coefficient. Lower boundary condition is given by

$$K_z\frac{\partial C}{\partial z}=V_g C\ , \qquad z=Z_d\ . \tag{2}$$

The roughness length Z_o is not equal to mass sink height Z_d. V_g is local deposition velocity. The lower boundary condition at $z = Z_d$ shows that if $V_g(Z_d) = 0$, all material reaching the ground is reflected back into atmosphere, which is called total reflection. If $V_g(Z_d)$ is infinite, it is called total absorption and $C(Z_d) = 0$. For the intermediate case, the material of diffusion reaching ground is partly absorbed and partly reflected back into the atmosphere.

2. *Momentum Exchange in* CL

The momentum equilibrium equation in CL can be written as (Lei and Chang, 1992)

$$\frac{\partial U}{\partial t}+\frac{\partial}{\partial z}\left[L^2\left|\frac{\partial U}{\partial z}\right|\frac{\partial U}{\partial z}\right]+A_{ao}(U_h-U)z/(1+B_{ao}A_r)H=qC_d(U)A(z)|U|U\ , \tag{3}$$

where q is sheltering factor constant, $C_d(U)$ is effective drag coefficient, and $A(z)$ is foliage density. The third term on the left hand side of Eq.(3) is additional larger-scale diffusion term (Li, 1985). In a broad sense, the transfer process in CL may be thought of as a turbulent diffusive process. However, this diffusive process includes local turbulent diffusion proportional to local gradient of the mean property and larger-scale transport depending on difference of the property in a larger spatial scale such as the canopy height. U_h is a reference velocity at $z = H$, A_{ao} and B_{ao} are constants chosen as 0.04 and 0.8. A_r is a reference foliage density. For $z \leqslant H_{\max}$, $A_r = A_{\max}$, where $H_{\max}$ is the level of maximum leaf area density. When $z > H_{\max}$, $A_r = A(z)$. Mixing length L is given by Underwood (1987).

In the numerical simulation, a Gaussian relationship for the vertical foliage distribution is used. The Bache's (1986a) expression of drag coefficient is adopted. K_z, K_x, and the velocity scale (U^*) are specified by

$$K_z=L(k)^2\left|[U(k+1)-U(k)]/[z(k+1)-z(k)]\right|\ , \tag{4}$$

$$K_x=K_x(H)K_z(z)/K_z(H)\ , \tag{5}$$

$$U^*(z)=\{K_z(k)|[U(k+1)-U(k)]/[z(k+1)-z(k)]|\}\ . \tag{6}$$

where $K_z(H)$ and $K_z(H)$ are given by Lei (1988).

3. *Species Absorbing Processes in CL and at Surface*

The local deposition velocity can be represented by the following transformation from basic definition (Bache, 1986b)

$$V_g = B_a U(z) / A(z) = \lambda / A(z) \ . \tag{7}$$

Clearly V_g depends on wind speed, physiological responses and chemical reactivity. It is the function of height and demonstrates that the assumption of constant V_g is in general incorrect. In fact, V_g of vegetation may vary by an order of magnitude depending on the canopy environment. There were hourly, daily and seasonal changes in V_g largely due to change in surface properties. From expression (7), λ can be represented as follows:

$$\lambda = V_g A(z) = A(z) / R_g \ . \tag{8}$$

V_g can also be shown by local resistance R_g. In order to analyze CL vertical structure affection, the model layer is subdivided into four regions. First, ABL area, the constant flux zone, where $\lambda = 0$ and turbulent diffusive processes dominate the flux of pollutant downward; Second, upper CL (UCL), where occur three uptake processes: bulk canopy stomatal uptake (R_s); bulk mesophyll uptake (R_m); bulk uptake of the outer surfaces of leaves(R_{lu}); Third, lower CL (LCL), there are two uptake processes: uptake pathways at leaves twig, bark (R_{cl}), and effects of mixing forced by buoyant convection due to solar heating and by penetration of wind into canopies on the sides of hills (R_{dc}); Fourth, surface layer (SL), there are two uptake factors: uptake at the ground by soil, leaf litter , snow, water (R_{gs}), and effect of different land types and seasons (R_{ac}).

By analogous to expression (8), λ can be represented in the every sublayer as follows:

$$\lambda_{ucl} = A(z)[1 / (R_s + R_m) + 1 / R_{lu}] = A(z) / R_u \ , \tag{9a}$$

$$\lambda_{lcl} = A(z) / (R_{cl} + R_{dc}) = A(z) / R_l \ , \tag{9b}$$

$$\lambda_{sl} = A(z) / (R_{ac} + R_{gs}) = A(z) / R_{sl} \ , \tag{9c}$$

$$R_c = 1 / [1 / R_u + 1 / R_l + 1 / R_{sl}] \ . \tag{10}$$

where R_c, R_u, R_l and R_{sl} are bulk resistances in CL, UCL, LCL and SL, respectively. Parameters R_s, R_m, R_{lu}, R_{dc}, R_{cl}, R_{ac}, and R_{gs} have been given by Wesely (1988) for different gaseous substances, land types, seasons, and meteorological conditions.

4. *Parameterizations of ABL Variables*

The required ABL parameters for the model have been deduced by Lei (1988). The parameterization expressions among internal parameters of ABL, parameters of near surface layer and external parameters are tested by field experimental data. The parameters vary with both height and atmospheric stability.

According to corresponding relationships between atmospheric stability classifications and time of 24 hours during a clear day and night, variations of the parameter with stability are changed into ones with time. Both surface air temperature, and solar radiation for 72 hours in OSCAR–4 case (NCAR, 1987) originate from a mesoscale model (MM4)(Anthes et al., 1987).

In order to resolve mean wind and turbulence, vertical grid levels are determined according to the following relationship

$$z = 5.779 \times 10^{-3}[(k+1)^3 - 1] \ . \quad \text{(m)} \tag{11}$$

The horizontal and vertical domains are taken as 80 km and 428 m, and the vertical grid number is 41 with $dx = 2$ km and time step $dt = 6$ s.

5. *Bulk Deposition Velocity*

If we consider vertical transport through an absorbing CL of height Z_r and neglect the transport at the base, the bulk deposition velocity $V_d(Z_r)$ is defined by

$$V_d(Z_r) = V_g(z)A(z)C(z)dz \,/\, C(Z_r) \ , \tag{12}$$

where $C(Z_r)$ and $C(z)$ have been known for given downwind distance. For different canopy conditions, $V_g(z)$ can be derived by expression (7). It may be shown from expression (12) that bulk deposition velocity depends on variational rules of pollutant concentration, absorbing factors and the foliage density with height. In order to make comparative analyses, the following V_g governing equation is solved

$$\frac{dV_g}{dz} + \frac{V_g}{K_z} = \lambda \ . \tag{13}$$

Table 1. Numerical Analyzed Cases and Parameters

Case	1	2	3	4	5	6	7	8
Z_o (m)	1	1	1	1	0.2	0.2	0.2	0.2
H (m)	10	10	10	10	3	3	3	3
λ	0	$B_a U$	$B_a U$	$B_a U$	0	$B_a U$	$B_a U$	$B_a U$
$U,\ U^*,\ K_z,\ K_y$	ABL	ABL	CL	CL	ABL	ABL	CL	CL
for $z \leqslant H$	par.	par.	par.	par.	par.	par.	par.	par.
Diff. term	/	/	no	have	/	/	no	have

Eq.(13) is called the Ricatti equation. For completely general forms of K_z and λ, it must be solved numerically.

III. EFFECTS OF CONTROL FACTORS ON V_d

In order to clarify control factors affecting CL deposition velocities, eight different kinds of cases (Table 1) are simulated with the model.

1. *Effects of CL Vertical Structures and Atmospheric Stabilities*

Using vertical distributive data of both concentration obtained with the model and absorbing parameters in CL, V_d are predicted by Eq.(12) for the eight cases. In order to obtain quantitative expression between V_d and control factors, correlation analyses between V_d of different cases (given downwind distance and hourly time) are done, the results show the following linear relationship

$$V_{d5}(XX1) = aa1 + bb1 \ V_{d5}(XX2) \ , \tag{14}$$

where $XX1$ and $XX2$ represent different cases, $aa1$ and $bb1$ are parameters dependent on atmospheric stabilities and the canopy vertical structures. MM, average values of $V_{d5}(XX1) / V_{d5}(XX2)$ for both the forest and corn canopies, are listed in Table 2. The correlation coefficients of expression (14) are greater than or equal to 0.97 in all the cases. As long as deposition velocity due to one control factor is known, effect of the other factor on deposition velocity can be derived conveniently by (14).

For total circumstances, MM is always greater than 1.00. This shows that the total effect of all the control factors increases V_{d5}. The effect of CL absorption is the largest and larger-scale diffusion is the least among them. Although R_{sl} and R_c of forest canopy are much larger than ones of the corn (*if vertical structure of CL does not considered, then surface absorption of case 2 condition is less than case 6*), MM of the forest canopy is much larger than ones of corn. In fact, R_u and R_l for forest canopy are much less than ones of corn. The results show that consideration of variations of absorptive factors with height in CL is very important for correct calculation of V_{d5}.

It can be seen from comparison between case 2 (or case 6) and case 1 (or case 5) that MM increases with decrease of atmospheric stability. A specially noticeable problem is that V_{d5} (case 6, or case 8) / V_{d5} (case 5) is less than 1.00 for stable condition. Because in the nighttime the absorptive action of corn canopy is less than ground (R_{sl} is less than R_u or R_l). If effect of atmospheric stability on V_{d5} is not considered, then V_d will be overestimated during the night, and underestimated during the day.

Average effect of the larger-scale diffusion term on V_{d5} is less than 4%. Although the effects of the factor on wind and concentration distribution in CL appear some important, its effect on V_{d5} can be neglected. So that we can make conclusion that exponential wind profile (Underwood, 1987) can apply to calculating V_{d5}.

Table 2. Variations of MM with Stability

Stability		*Forest*			Corn	
classes	case	MM	case	case	MM	case
	4	1.04	3	8	1.01	7
Stable	3	1.59	2	7	1.00	6
	2	3.45	1	6	0.32	5
	4	5.51	1	8	0.33	5
	4	1.00	3	8	1.01	7
Neutral	3	1.44	2	7	0.94	6
	2	8.17	1	6	1.02	5
	4	11.6	1	8	1.10	5
	4	1.00	3	8	1.00	7
Unstable	3	1.37	2	7	1.09	6
	2	15.8	1	6	2.06	5
	4	21.2	1	8	2.25	5
	4	1.01	3	8	1.01	7
Total	3	1.48	2	7	1.01	6
	2	8.71	1	6	1.08	5
	4	12.3	1	8	1.17	5

2. *Variation of Deposition Velocity with Height and Time*

Dry deposition velocity is a velocity equivalent to covariance between vertical turbulence and species concentration divided by the mean concentration. The covariance and mean concentration are functions of both time and height. Therefore V_g is necessarily the function of both time and height.

For a typical clear day and night, there could be a significant diurnal variation of $V_g(H)$ in CL (Fig.1). The values during the daytime are considerably more than at night. During the transitional periods (06–08 or 18–20 LST), they exhibit jump. After the transition, the V_g during the day and night does not mainly vary with time. The variational laws are very similar to mean wind speed and are of opposite to the concentrations and resistances of CL.

It can be seen from expressions (9)–(10) that variations of V_g with time mainly depend on factor λ (or bulk resistances of the sublayer). The effect of stability due to insolation is particularly apparent. $V_g(H)$ varies with time by almost a factor of 4.8, indicating that use of single diurnal average $V_g(H)$ would make the amount of removed material overestimated at night time.

Under a typical clear day and night condition, there could be considerable variation of V_g with height in CL (Figs. 2– 3), and it reaches the largest at Z_r. The V_g varies with height by almost a factor of 6.5 at 02 LST, a factor of 24.4 at 14 LST. When $z < 0.2H$, the variation of V_g with height is very small. In the very dense UCL, the absorbing action of the canopy dominates deposition processes, therefore, the V_g in UCL is higher than one in LCL. The variation of V_g with height is quite similar to the foliage density. At lower layer, the variation of $A(z)$ with height is not considerable, the removal rate gradually increases with continuous height increase until the height of the peak is attained, which usually occurs at maximal $A(z)$.

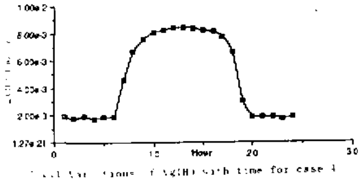


Fig.1. Variation of $V_g(H)$ with time for case 4.

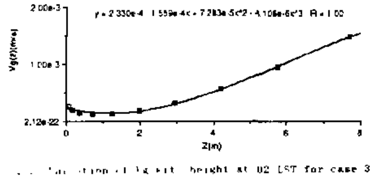
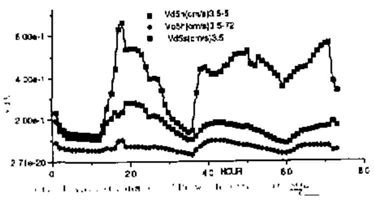


Fig.2. Variation of V_g with height at 02 LST for case 3.



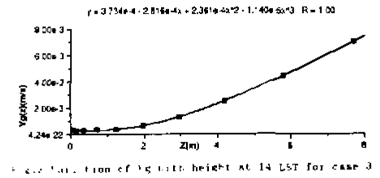


Fig.3. Variation of V_g with height at 14 LST for case 3.

Fig.4. Variation of V_{d5} with time for SO_2.

The elevation of the lowest computation grid point in the regional pollutant models is typically much higher than the reference height used to establish the pollutant bulk deposition velocities (Walcek et al., 1987). When coupled with the observation data that V_d varies with height ($Z_r \geqslant H$), there is a need to develop an effective deposition velocity, V_{de}. Following one way to develop such a model is to assume that most of the lowest grids of regional model are in the constant flux layer. If this is the case then we can obtain the following expression (Janssen, 1988)

$$V_{de}(Z_e) = V_d(Z_r) / [1 + V_d(Z_r) \quad dz / K_z] \ . \qquad (15)$$

One implication from Eq.(15) is that if $V_d(Z_r)$, rather than $V_{de}(Z_e)$, is used in a practical model calculation then the surface removal flux would be considerably overestimated.

Comparisons between with and without height correction are given in Fig.4 and Table 3 respectively, where V_{d5s} is value at 3.5 m. V_{d5h}(3.5–72) and V_{d5h}(3.5–5) represent corrected V_{d5s} by expression (15) to 72 and 5 m, respectively.

Regarding Fig.4 and Table 3, the following results should be obtained: action of the height correction decreases V_d; after the height correction, variational amplitude of V_d with time becomes small and height of the lowest model grid point is higher, the amplitude is smaller; differences between V_{d5h}(3.5–72) and V_d(72) without height correction are very large, variational amplitudes of V_d(72) with time are more considerable than V_{d5h}(3.5–72) and the variational rule of V_{d5h} (3.5–72) is more consistent with practice than V_d(72), which shows that the revised method adopted in the study is better.

Table 3. Statistics of Ratios between Different V_d

	Max	Mean	Min
$V_{d5s}(3.5) / V_{d5h}(3.5\text{–}72)$	10.27	5.67	2.25
$V_{d5s}(3.5) / V_{d5h}(3.5\text{–}5)$	4.37	2.36	1.16

3. *Effects of Variation of K_z and U^* with Height on V_d*

Practical test of effect for the two K_z patterns (Lei,1988 and NCAR, 1987) on deposition velocity shows that Lei's formula is better than NCAR's. The comparative results only due to different K_z formulas are described in Fig. 5 and Table 4, where subscripts n and r represent K_z formula of Lei and NCAR, respectively. Regarding Fig. 5 and Table 4, the following points should be noted: effect of Lei's formula decreases aerodynamic resistance, $R_{an} < R_{ar}$, except for a few points, ranges of variation of R_{ar} with time are much greater than R_{an}; the coefficient of correlation between V_{dn} and V_{dr} is greater than and equal to 0.99; V_{dn} are greater than V_{dr}, the mean ratios from 3.5 to 72 m obviously increase,

Table 4. *Statistics of Ratios between V_{dn} and V_{dr}*

	Max	Average	Min
$V_{dn} / V_{dr}(3.5)$	1.76	1.09	0.97
$V_{dn} / V_{dr}(72)$	2.86	1.28	0.65
$R_{an} / R_{ar}(3.5)$	0.69	0.45	0.19
$R_{an} / R_{ar}(72)$	10.31	0.47	0.02

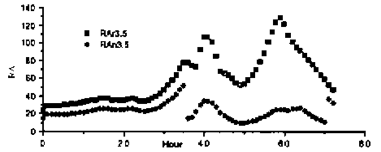


Fig.5. Variation of R_a with time at $z = 3.5$ m.

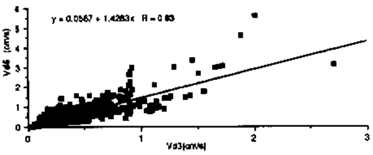


Fig.6. Comparison between V_{d5} and V_{d3}.

average value is about 1.3 at 72 m, but differences between maximum and minimum values are by far larger than the mean value, it appears that action of K_z is very obvious. Thus the choice of K_z profile is important for correct calculating V_d.

Among many factors of influencing dry deposition velocity, U^* is one of the most important factors. Because U^* in surface boundary layer above CL is a constant, and both U^* and V_d at Z_r are used in early regional models, then V_d / U^* =constant. But in deep forest canopy, U^* decreases with height decrease. $V_g / U^*(z)$ is a function of height. General relationship between $V_g(z)$ and $U^*(z)$ in CL can be described by the following linear form

$$V_g(z) = V_a(z) + \exp[V_b(z)]U^*(z), \tag{16}$$

where $V_a(z)$ and $V_b(z)$ are complex functions of height. If Z_r = 10m, U^* = 16.5 cm / s, it can be obtained from expression (16) that $V_g(10) / U^*(10) = 0.002$. This result is the same as the relationship, $V_d / U^* = 0.002$, given by Wesely et al.(1985) for the neutral condition, which is only a specific condition of expression (16).

IV. COMPARISON BETWEEN COMPUTATIVE AND EXPERIMENTAL RESULTS

For comparison, the formula used in RADM (NACR, 1987) is shown as

$$V_{d3} = 100 / (R_a + R_b + R_c) \ , \qquad \text{(cm / s)} \tag{17}$$

where R_a is aerodynamic resistance, and R_b is sublayer resistance.

Expression (14) most completely represents relationship of dry deposition velocity between with and without CL. Its advantages are: effects of vertical structure of both absorbing factor (or resistance factor) and the foliage density on V_d are considered and the assumption of constant flux layer is not taken. The physical meaning of expression (14) is clearer than expression (17). But they will be still tested by experimental data.

In order to test reliability of expressions (17) and (14), 1022 measured data on V_d, which are obtained at 20 different literatures and reports over the past ten years, have been collected. There are six species (SO_2, O_3, HNO_3, NO_X, SO_4, and Particulate Sulfur) in the data. A wide range of surfaces is included in these data ranging from soil surfaces through short grass to forest. Their magnitudes are probably correct to better than a factor of two. The data are not perfect, even so, they still can provide the study widely experimental bases.

Formulas (17) and (14) have been tested by 1022 data (Table 5 and Fig.6), where subscript s represents effect of both atmospheric stability and CL vertical structure, t is only the effect of CL vertical structures. Several results are noted. First, although standard deviations (CGM) are largish, all of them are less than the mean ratios (maximum mean ratio is 1.11, minimum is 0.81), results predicted by formulas (17) and (14) consist well with experimental data. Second, the best formula is V_{d5s} among computative formulas, it is the closest to the experimental data. Third, V_{dis} are closer to practical values than V_{di}, showing that the corrections of both canopy vertical structure and atmospheric stability improve predicted effects. Fourth, Fig.6 shows results of correlation analyses between V_{d5} and V_{d3} for 1022 experimental data, there is a better linear relationship between them, based on the current observational technology, this agreement is well. Fifth, effects of CL vertical structure increase deposition velocities ($[V_{dit} - V_{di}]/V_d > 0$), the increment of V_{d3} is the largest, it shows that correction of the canopy vertical structure for V_{d3} is more important than for V_{d5}. Sixth, effects of the stability also increase deposition velocity($[V_{dis} - V_{di}]/V_d > 0$) , the increments are greater than 14.11%.

Table 5. Comparison between Computative and Experimental V_d

V_{d5}/V_d	CGM	V_{d3}/V_d	CGM	V_{d5s}/V_d	CGM	V_{d3s}/V_d	CGM
0.92	0.71	0.81	0.61	1.06	0.82	1.11	0.83
$(V_{d5t} - V_{d5})/V_d$		$(V_{d3t} - V_{d3})/V_d$		$(V_{d5s} - V_{d5})/V_d$		$(V_{d3s} - V_{d3})/V_d$	
0.99%		19.74%		14.11%		29.67%	

V. CONCLUSIONS

The detailed vertical structure of dry deposition velocity in CL can be well presented by the coupling model. The results show that consideration of variations of absorptive factors with height in CL is very important.

The correlation analysis for effect of different control factors on deposition velocity shows that there are very well linear relationship between V_d of different cases. The effect of canopy absorption is the largest among them. The average effect of larger–scale diffusion in CL is less than 4%.

There are significant diurnal variations of V_d in CL. The V_d varies by almost a factor of 5. By use of a single diurnal average V_d, the amount of material removal would be overestimated at night time.

There is obvious height variation of V_g in CL, reaching the largest at Z_r. The V_g varies with height by almost a factor of 6.5 at 02 LST, by factor of 24.4 at 14 LST. When $z < 0.2$ h, variation of V_g with height is very small. For height correction of deposition velocity above CL, the variation of V_d with height is decreased. It is very important to make height correction in regional model with a large height of the lowest grid point. U^* decreases with height decrease in CL. $V_g / U^*(z)$ is not a constant, but a complex function of height. If the constant $V_d(Z_r) / U^*(Z_r)$ is used into CL, large errors would appear.

Comparative results between expressions (17) and (14) show that correlation between them is better for all the their inputting data, but there are large differences for their magnitudes. The best formula is V_{d5s} among the computative formulas, which is the closest to the experimental data.

In a word, many processes influencing dry deposition are represented completely in the study. The space–time distributions of dry deposition velocity can be predicted by the coupling model. The expression (14) is closer the experimental data than the formula (17) used in RADM.

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