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A size-segregated particle dry deposition scheme for an atmospheric aerosol module

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Abstract

A parameterization of particle dry deposition has been developed for the Canadian Aerosol Module (CAM).This parameterization calculates particle dry deposition velocities as a function of particle size and density as well as relevant meteorological variables. It includes deposition processes, such as, turbulent transfer, Brownian diffusion, impaction, interception, gravitational settling and particle rebound. Particle growth under humid conditions is also considered. Sensitivity tests show that the parameterization provides deposition velocities comparable with recent field observations, especially for sub-micron particles. The present parameterization has also been evaluated using two empirical bulk resistance models, which were originally developed from field observations. The present parameterization has been implemented in CAM, with meteorological input provided by the Canadian Regional Climate Model (RCM) to the eastern North America. A comparison of the modelled dry deposition velocities to a variety of recent measurements that have been reported in the literature demonstrated that the current parameterization produces reasonable results. The main improvement of the current parameterization compared to earlier size-dependent particle dry deposition models is that the current one produces more realistic deposition velocities for sub-micron particles and agrees better with recently published field measurements. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Particle dry deposition; Size-dependent; Model development and evaluation.

1. Introduction

Essential processes in CAM (Gong et al., 2000) include particle sources, transport and removal mechanisms. One of the removal processes is the particle dry deposition, which is a complex process depending on physical and chemical properties of the aerosol, the underlying surface characteristics and micro-meteorological conditions. The knowledge on particle dry deposition is far from complete due to the complex dependence of deposition on particle size, density, terrain, vegetation, meteorological conditions and chemical species. A variety of dry deposition parameterizations have been used in large and regional-scale transport models and are reported in a review by Ruijgrok et al. (1995). There are some models which calculate particle dry deposition velocity as a function of particle size (Bache, 1979; Davidson et al., 1982; Giorgi, 1988; Haynie, 1986; Ibrahim et al., 1983; Legg and Price, 1980; Peters and Eiden, 1992; Schack et al., 1985; Schmel and Hodgson, 1980; Slinn, 1982; Slinn and Slinn, 1980; Wiman and Agren, 1985). Some of the size-dependent dry deposition models apply only to one type of surface (Davidson et al., 1982; Peters and Eiden, 1992; Wiman and Agren, 1985; Slinn and Slinn, 1980), other models apply to any type of surface (Giorgi, 1988; Haynie, 1986; Schmel and Hodgson, 1980). Many of the size-dependent models were evaluated using wind tunnel measurements, which may not be representative of field observations. Estimates from some of these models (Ruijgrok et al., 1995) revealed that they differ from each other greatly and the largest uncertainty is for the

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0.1–1.0 μm particle size range, for which the deposition velocities can vary by 2–3 orders of magnitude. Theoretical studies by most of these models suggested that particles in the range 0.1–1.0 μm diameter should be subject to deposition velocities of $\leqslant 0.01\,\mathrm{cm\,s^{-1}}$ and this value seems comparable only to laboratory (wind tunnel) studies (Nicholson, 1988). Higher values have been obtained in many field studies investigating some trace species which are usually considered to be representative of particles in this size range. Aerosol sulphate has been frequently used as a convenient marker, due to its ubiquitous nature and chemically conservative behaviour. Recent observations for sulphate and other sub-micron particles showed that the dry deposition velocities are one to two orders of magnitude higher than previous studies (Allen et al., 1991; Everett et al., 1979; Gallagher et al., 1997; Hicks et al., 1982, 1989; Lamaud et al., 1994; Sievering, 1982, 1983, 1987; Wesely et al., 1983, 1985; Wyers and Duyzer, 1997; Wyers and Veltkamp, 1997). A review by Nicholson (1988) showed that some authors attempted to explain some of these higher values by invoking meteorological factors and sampling errors. However, Gallagher et al. (1997) demonstrated that the recent observed higher deposition velocity values, typically $1\,\mathrm{cm\,s^{-1}}$ or more, for sub-micron aerosols deposition to a forest, are consistent across the aerosol size spectrum, despite the very different techniques involved. Gallagher et al. (1997) stated that previous model studies significantly underestimated dry deposition velocities, especially for sub-micron particles, over rough vegetated surfaces.

In this study, a simple parameterization of particle dry deposition is developed for CAM. One reason for developing this new parameterization is that earlier models predict much lower deposition velocities for sub-micron aerosols, the most important part in CAM, compared to recent observations. The other reason is that other models apply to different land types than those used in CAM. The parameterization provides estimates of the particle dry deposition velocity as a function of particle size and density for different underlying surfaces and meteorological conditions. Since there are no data and no size-dependent models available to evaluate this parameterization, a review of measurements published in the literature and two bulk resistance models (Wesely et al., 1985; Ruijgrok et al. (1997)) are used to assess the current parameterization. Here, only measurements for fine particles over natural surfaces are included due to the reasons discussed above. Wesely et al. (1985) parameterization was derived from measurements of particulate sulphur deposition, where the particle mass mean size was "expected" to be typically 0.35–0.4 μm in diameter (although no size discrimination was available). This parameterization can produce sulphate deposition velocities are of the order of magnitude higher than earlier theoretical studies, but still lower than some measurements (e.g. Gallagher et al., 1997) and also lower than Ruijgrok et al. (1997) parameterization for sulphate. Ruijgrok et al. (1997) parameterization was derived from measurements over a needleleaf forest and can predict deposition velocities for NH_4, SO_4, NO_3 and Na^+. Mass mean diameter and geometrical standard deviation are provided in Ruijgrok et al. (1997) and this helps us evaluate the current parameterization. Here only the parameterization for Na^+ is used to evaluate the present parameterization for large particle size range.

2. Model theory

The parameterization of the particle dry deposition presented in this paper is based on Slinn's (1982) model, which was developed for vegetated canopies. Slinn's (1982) model includes the deposition processes of Brownian diffusion, impaction, interception, gravitational settling and particle rebound. Particle growth under humid conditions was discussed but not included in his model. This model requires detailed canopy information, which is generally unavailable in regional scale transport models. In the present study, the same approach as Slinn's (1982) model is used for modelling particle dry deposition, but using simplified empirical parameterization for all deposition processes. Particle growth at high humidity is also included.

Following Slinn (1982), the dry deposition velocity V_d can be expressed as

$$V_d = V_g + \frac{1}{(R_a + R_s)}, \tag{1}$$

where V_g is the gravitational settling velocity, R_a is the aerodynamic resistance above the canopy, R_s is the surface resistance.

The gravitational settling is calculated as

$$V_g = \frac{\rho d_p^2 g C}{18\eta}, \tag{2}$$

where ρ is the density of the particle, d_p is the particle diameter, g is the acceleration of gravity, C is the correction factor for small particles and η is the viscosity coefficient of air.

The correction factor is calculated as

$$C = 1 + \frac{2\lambda}{d_p}(1.257 + 0.4\mathrm{e}^{-0.55 d_p/\lambda}), \tag{3}$$

where λ is the mean free path of air molecules and is calculated as a function of temperature, pressure and air's kinematics viscosity.

The aerodynamic resistance is calculated as

$$R_a = \frac{\ln(z_R/z_0) - \psi_H}{\kappa u_*}, \tag{4}$$

where z_R is the height at which the dry deposition velocity V_d is evaluated, z_0 is the roughness length, ψ_H is the stability function, κ is the Von Karman constant and u_* is the friction velocity. R_s depends on the collection efficiency of the surface and is determined by the various deposition processes, the size of the depositing particles, atmospheric conditions and surface properties. Here, R_s is parameterized as

$$R_s = \frac{1}{\varepsilon_0 u_* (E_B + E_{IM} + E_{IN}) R_1}, \tag{5}$$

where E_B, E_{IM}, E_{IN} are collection efficiency from Brownian diffusion, impaction and interception, respectively; R_1 is the correction factor representing the fraction of particles that stick to the surface. ε_0 is an empirical constant and is taken as 3 here for all land use categories (LUC).

For Brownian diffusion, there is evidence that E_B is a function of Schmidt number, *Sc*, given as

$$E_B = Sc^{-\gamma}. \tag{6}$$

The Schmidt number is the ratio of the kinematic viscosity of air, v, to the particle Brownian diffusivity, $D(Sc = v/D)$. γ usually lies between 1/2 and 2/3 with larger values for rougher surfaces. For example, Slinn and Slinn (1980) suggested γ, a value of 1/2 for water surfaces. Slinn (1982) suggest γ, a value of 2/3 for vegetated surfaces. In the present paper, Eq. (6) is used for calculating collection efficiency by Brownian diffusion with values of γ varying with land use categories.

The parameter governing impaction process is the Stokes number, *St*, which has the form $St = V_g u_*/gA$ for vegetated surfaces (Slinn, 1982) and $St = V_g u_*^2/v$ for smooth surfaces or surfaces with bluff roughness elements (Giorgi, 1988). "*A*" is the characteristic radius of collectors.

Slinn (1982) used a semi-empirical fit for smooth surfaces, for which the collection efficiency by impaction is

$$E_{IM} = 10^{-3/St}. \tag{7a}$$

Slinn (1982) then suggested another form for vegetative canopies, for which the collection efficiency by impaction is

$$E_{IM} = \frac{St^2}{1 + St^2}. \tag{7b}$$

Peters and Eiden (1992) use the following form for impaction efficiency over a spruce forest:

$$E_{IM} = \left(\frac{St}{\alpha + St}\right)^{\beta}, \tag{7c}$$

where α and β are constants. Using 0.8 for α and 2 for β, respectively, Peters and Eiden (1992) get the best fit for the data collected by Belot and Gauthier (1976).

Giorgi (1986) suggested two formulae for impaction efficiency, one for smooth surfaces and surfaces with bluff roughness elements,

$$E_{IM} = \frac{St^2}{400 + St^2} \tag{7d}$$

and the other one for vegetated surfaces

$$E_{IM} = \left(\frac{St}{0.6 + St}\right)^{3.2}. \tag{7e}$$

This form is the same as the one used by Peters and Eiden (1992), but using different constants α and β.

Davidson et al. (1982) applied the following form over grassland:

$$E_{IM} = \frac{St^3}{St^3 + 0.753 St^2 + 2.796 St - 0.202}. \tag{7f}$$

In the present study Eq. (7c) is used with α varying with LUC and β chosen as 2.

The collection efficiency by interception also exists if the particle passes an obstacle at a distance shorter than its physical dimensions. This is especially important for large particles over hairy leaves. Fuchs (1964) suggested a variety of forms for E_{IN} for viscous and potential flow above a sphere and a cylinder. All the forms are a function of particle diameter and the characteristic "radius" of the collectors. Slinn (1982) parameterized E_{IN} composing small and larger collectors. Giorgi (1988) uses the same approach as was used by Slinn (1982). It is very difficult to get data on the fraction of large and small collectors. Consequently, the following simple form is used for calculating collection efficiency by interception:

$$E_{IN} = \frac{1}{2}\left(\frac{d_p}{A}\right)^2. \tag{8}$$

The characteristic radius "*A*" in Eq. (8) is given for different land use and seasonal categories.

Particles larger than 5 μm may rebound after hitting a surface. This process may be included by modifying the total collection efficiency by the factor of R_1, which represents the fraction of particles sticking to the surfaces, as is done in Eq. (5). Slinn (1982) suggested the following form for R_1:

$$R = \exp(-St^{1/2}). \tag{9}$$

Giorgi (1988) also adopted this form. Limited knowledge on particle rebound prevent us from estimating this term accurately, thus the same formula is used in the present study with the condition that no particles rebound from a wet surface.

Particles can grow in high humidity conditions. This effect is included here by replacing the dry particle radius with a wet one. The wet particle radius, r_w, is calculated using the dry particle radius, r_d, and the relative humidity

RH (Gerber, 1985) for sea-salt and sulphate aerosols

$$r_w = \left[\frac{C_1 r_d^{C_2}}{C_3 r_d^{C_4} - \log RH} + r_d^3 \right], \qquad (10)$$

where C_1, C_2, C_3, and C_4 are empirical constants using the values listed in Table 1. In CAM (Gong et al., 2000), particle growth is calculated for mixed aerosols rather than for individual particle species.

The particle dry deposition velocities can be calculated using Eqs. (1)–(6), (7c) and (8)–(10).

3. Land use category, seasonal category and parameters

The highest resolution LUC data available are the 1 km USGS (US Geological Survey) Global Land Cover Characteristics Data. This data set has been converted to several groups for different purposes. The one adopted here is the Biosphere Atmosphere Transfer Scheme (BATS; Dickinson, 1986). We regroup the BATS' original 20 LUCs into 14 land use categories and add "urban" as the 15th land use category for CAM's dry deposition model. It is necessary to point out that V_d for LUC 13, "inland water", and LUC 14, "ocean", are treated the same way. The reason for keeping these two LUCs separately is not for dry deposition considerations but for other purposes, such as sea-salt emission. V_d is calculated in CAM for all LUCs existing inside one grid and then averaging according to the area fraction of each LUC.

Since some parameters change with season of the year, it is necessary to define several different seasonal categories. The one used here is the same as in Brook et al. (1999), which was originally reported in Wesely (1989). Five seasonal categories are defined. The 15 land use categories and five seasonal categories used in CAM are listed in Table 2.

Several parameters mentioned above need to be defined for calculating the particle dry deposition. These include roughness length "z_0", characteristics radius "A", empirical constants "α" and "γ". These four parameters are listed in Table 3 for all LUCs. Roughness length z_0 and characteristic radius "A" depend on both LUC and seasonal categories.

Table 1
Constants for Eq. (10)

Aerosol model	C_1	C_2	C_3	C_4
Sea salt	0.7674	3.079	2.573×10^{-11}	− 1.424
Urbal	0.3926	3.101	4.190×10^{-11}	− 1.404
Rural	0.2789	3.115	5.415×10^{-11}	− 1.399
$(NH_4)_2SO_4$	0.4809	3.082	3.110×10^{-11}	− 1.428

Table 2
Land use categories (LUC) and seasonal categories (SC) used in Canadian Aerosol Module

Category	Description
Land use categories (*LUC*)	
1	Evergreen–needleleaf trees
2	Evergreen broadleaf trees
3	Deciduous needleleaf trees
4	Deciduous broadleaf trees
5	Mixed broadleleaf and needleaf trees
6	Grass
7	Crops, mixed farming
8	Desert
9	Tundra
10	Shrubs and interrupted woodlands
11	Wet land with plants
12	Ice cap and glacier
13	Inland water
14	Ocean
15	Urban
Seasonal categories (*SC*)	
1	Midsummer with lush vegetation.
2	Autumn with cropland that has not been harvested.
3	Late autumn after frost, no snow.
4	Winter, snow on ground and subfreezing.
5	Transitional spring with partially green short annuals.

4. Review of measurements and sensitivity tests

A review of published measurements for dry deposition velocities of fine particles are presented in Table 4. Sulphate particles are typically in this size range. Even through uncertainties may exist for the values presented in Table 4 due to the difficulties of the measurements and different methods and assumptions during different measurement studies, these values provide us some evidence for developing the current model. Earlier studies based on channel data found that fine particles with particle size in the 0.1–1.0 μm aerodynamic diameter range have very low deposition velocities (< 0.1 cm s^{-1}). However, recent field studies presented in Table 4 show that V_d can be larger than 1 cm s^{-1}. In general, the measurements summarized in Table 4 indicate that particles in this size range have higher deposition velocities over rough surfaces such as forests and lower deposition velocities over smooth surfaces such as snow. The typical mean deposition values are within 0.1–1.0 cm s^{-1} for vegetated surfaces.

Sensitivity tests are shown in Figs. 1 and 2. Fig. 1 is an example showing how V_d varies among surface types and

Table 3
Parameters for 12 land use categories (LUC) and five seasonal categories (SC)[a]

LUC		1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Z_0 (m)	SC 1	0.8	2.65	0.85	1.05	1.15	0.1	0.1	0.04	0.03	0.1	0.03	0.01	$f(u)$	$f(u)$	1.0
	SC 2	0.9	2.65	0.85	1.05	1.15	0.1	0.1	0.04	0.03	0.1	0.03	0.01	$f(u)$	$f(u)$	1.0
	SC 3	0.9	2.65	0.80	0.95	1.15	0.05	0.02	0.04	0.03	0.1	0.02	0.01	$f(u)$	$f(u)$	1.0
	SC 4	0.9	2.65	0.55	0.55	1.15	0.02	0.02	0.04	0.03	0.1	0.02	0.01	$f(u)$	$f(u)$	1.0
	SC 5	0.8	2.65	0.60	0.75	1.15	0.05	0.05	0.04	0.03	0.1	0.03	0.01	$f(u)$	$f(u)$	1.0
A (mm)	SC 1	2.0	5.0	2.0	5.0	5.0	2.0	2.0	na	na	10.0	10.0	na	na	na	10.0
	SC 2	2.0	5.0	2.0	5.0	5.0	2.0	2.0	na	na	10.0	10.0	na	na	na	10.0
	SC 3	2.0	5.0	5.0	10.0	5.0	5.0	5.0	na	na	10.0	10.0	na	na	na	10.0
	SC 4	2.0	5.0	5.0	10.0	5.0	5.0	5.0	na	na	10.0	10.0	na	na	na	10.0
	SC 5	2.0	5.0	2.0	5.0	5.0	2.0	2.0	na	na	10.0	10.0	na	na	na	10.0
α		1.0	0.6	1.1	0.8	0.8	1.2	1.2	50.0	50.0	1.3	2.0	50.0	100.0	100.0	1.5
γ		0.56	0.58	0.56	0.56	0.56	0.54	0.54	0.54	0.54	0.54	0.54	0.54	0.50	0.50	0.56

[a] Note: $f(u)$ represents a function of wind speed (u) and na represents not applicable.

particle size for seasonal category 1. In this example, V_d values were calculated for particles with density of 2000 kg m^{-3}, wind speed of 5 m s^{-1} at a height of 20 m and a neutral stratification. It is seen that particle dry deposition velocities are highly dependent on surface types. For small particles ($d_p = 0.1$ μm in Fig. 1), the V_d values were controlled by Brownian diffusion and the aerodynamic resistance. Parameter u_* is an important variable as can be seen in Eqs. (4) and (5). For forests and urban surfaces with larger roughness lengths and thus bigger friction velocity u_*, the surface resistance and aerodynamic resistance are smaller and the V_d values are higher than that for the other surface types. V_d for LUC 2 has a much higher value than all other surface types since z_0 for LUC 2 is much higher than that for the other LUCs. For particles with radius of 0.5 μm (Fig. 1), V_d are still larger over rougher surfaces but the differences of V_d between different surface types are not as large as V_d for smaller particles (e.g. $d_p = 0.1$ μm). Values for particles with radius of 0.1 and 0.5 μm are well within the range of measurements presented in Table 4.

For larger size particles ($d_p = 5$ μm in Fig. 1), impaction and interception become important. Needleleaf forests (LUCs 1 and 3) not only have larger roughness lengths, which result in larger u_*, but also smaller collection radii "A", which result in larger Stokes numbers. This subsequently results in a larger impaction collection efficiency. Therefore, LUCs 1 and 3 have the highest V_d values. LUC 6 (grass) and LUC 7 (crops) also have small collection radius, so the V_d for LUC 5 and 6 are also higher than that for LUCs 8, 9, 10, 11, 12, 13, 14, 15. For LUC 15 (urban) the roughness length is larger than most canopies (except forest). In urban areas there could be a considerable percentage area of trees and grasslands, thus the V_d could be large. So, here, V_d for the urban LUC is parameterized as having slightly higher values than smoother surfaces, but not as high as that for forests, grass and agricultural lands. For particles with a radius of 1 μm (Fig. 1), V_d is higher over rougher surfaces with smaller collector radii. However, neither Brownian diffusion nor impaction is very effective for this size range of particles, thus V_d has smallest values compared to other particle size ranges.

Fig. 2 shows how V_d changes with wind speed for a particle having a density 2000 kg m^{-3}, neutral stratification and seasonal category 1. For both LUCs 1 and 7, it is seen that V_d has higher values at high wind speeds for all particle size ranges. A higher wind speed causes higher friction velocity, thus smaller aerodynamic resistance and surface resistance. Particles in the size range of 0.1–2 μm have the smallest V_d value since none of the Brownian diffusion, impaction or interception is very effective for this size range. V_d for this size range can change by an order of magnitude when the wind speed changes from 2 to 15 m s^{-1}. The deposition values shown in Figs. 1 and 2 are reasonable when compared to limited observed data and some other deposition models.

5. Model assessment

Due to the lack of experimental data, it is not possible to compare the present model results with observed data directly. For verification, two empirical models are chosen to compare with the present parameterization. One is the model developed by Wesely et al. (1985) for sulphate. This model is an empirical fit to the field experiment data over grassland in California. This model is adopted in the air quality model referred to as RADM

Table 4
Review of measured dry deposition velocities for fine particles

Surface	Method	Species	Size	Deposition velocity (cm s^{-1})	Reference
Agricultural land	Gradient		0.15–0.3	0.1–1.2 mean 0.5	Sievering (1982)
	Eddy correlation		0.2	1.2 (unstable) 0.37 (stable)	Sievering (1987)
	Eddy correlation		0.09–2.5	< 0.0–0.7 Mean 0.05	Sievering (1983)
Coniferous forest	Gradient	Sulphate		0–4.0 Mean 0.7	Wyers and Duyzers (1997)
	Gradient	^{214}Pb	< 1.5	Mean 0.73	Wyers and Veltkamp (1997)
	Eddy correlation		0.10–0.18	0.02–0.4	Gallagher et al. (1997)
			0.18–0.24	0.02–1.0	
			0.24–0.30	0.02–2.0	
			0.30–0.50	0.03–1.8	
	Gradient		0.5–1.0	Mean 0.43	Lorenz and Murphy (1989)
			1.0–2.0	Mean 0.78	
	Eddy correlation	Sulphate		0.41–1.44 Mean 0.7	Hicks et al. (1982)
Deciduous forest		Sulphate		0.0 to > 1.0 Mean 0.6	Hicks et al. (1989)
Grass	Gradient	Sulphate	0.1–2.0	−0.33–0.57 Mean 0.10	Allen et al. (1991)
	Eddy correlation	Sulphate		0.18	Wesely et al. (1983)
	Gradient	Sulphate		< 0.1	Atkins and Garland (1974)
	Gradient		0.05–1	< 0.1	Garland and Cox (1982)
	Gradient	Sulphate		< 0.3–0.4	Doran and Droppo (1983)
	Eddy correlation		0.1–0.5	< 0.05	Neumann and den Hartog (1985)
	Eddy correlation		0.05–0.1	0.1–1.0	Wesely et al. (1977)
Grass (rough)	Eddy correlation	Sulphate		0.0–0.5 mean 0.22	Wesely et al. (1985)
	Gradient	Sulphate		1.0–2.0	Everett et al. (1979)
	Gradient	Sulphate		−0.53 to 0.57 Mean 0.07	Nicholson and Davies (1987)
Semi-arid	Eddy correlation		< 1.0	< 0.1–0.6	Lamaud et al. (1994)
Snow	Tracer		0.7	Mean 0.04 Mean 0.1	Ibrahim et al. (1983)
	Eddy correlation		0.15–0.30	0.034	Duan et al. (1988)
			0.50–1.0	0.021	

(Chang, 1987) and a routine deposition model Brook et al. (1999). The other one is developed by Ruijgrok et al. (1997) for sulphate, nitrate, K$^+$ and Na$^+$. This model is based on the field experimental data over forests (mostly needleleaf forest and a small portion of mixed forests). The sub-model for Na$^+$ from Ruijgrok et al. (1997) is used here to test the present model for relatively large particles. Since these two models are based on data sets collected in field experiments, they are expected to predict reasonable particle dry deposition velocities.

The CAM dry deposition model calculates the V_d as a function of particle size and density. The two benchmark models used for the comparison include a range of particle sizes for particular particle species. Thus, only the size-averaged V_d from present parameterization is compared to the V_d calculated directly from the two empirical models. In order to accomplish this comparison, a size distribution of aerosol particles for the two measurement campaigns has to be assumed and used in the CAM dry deposition model to calculate the size-averaged V_d. Here, log-normal size distributions are assumed requiring mass mean diameter (MMD), geometric standard deviation (GSD) and a number of size bins. The geometric diameter and area fraction for each size bin can be calculated. For each size bin, V_d is calculated and then averaged according to its area fraction. We then compare the averaged V_d with V_d calculated from Ruijgrok's and Wesely's model.

Two sets of comparisons have been conducted. The first one uses a stand-alone 1-D version of the present model and compared with Ruijgrok's and Wesely's models. The input meteorological data are assigned arbitrarily. The wind speed "u" at 20 m varies from 1 to 15 m s^{-1} at an interval of 2 m s^{-1}. Temperature at the surface "T_s" varies from 280 to 300 K at interval of 10 K. Temperature at 20 m "T_R" is varied from $T_\mathrm{s} - 1$ to $T_\mathrm{s} + 1$ at interval of 0.5. Relative humidity "RH" changes from 45 to 95% at interval of 10%. The second one is to compare V_d calculated from the present model with Ruijgrok's and Wesely's models for several selected grids which contain only one LUC in CAM with meteorological input provided by RCM, the Canadian regional climate model. CAM has been run for 2 days at 10 min time step. The selected region is set at the eastern North America with 50 × 40 grids at 40 km horizontal resolution. A total of 288 V_d samples are available for comparison.

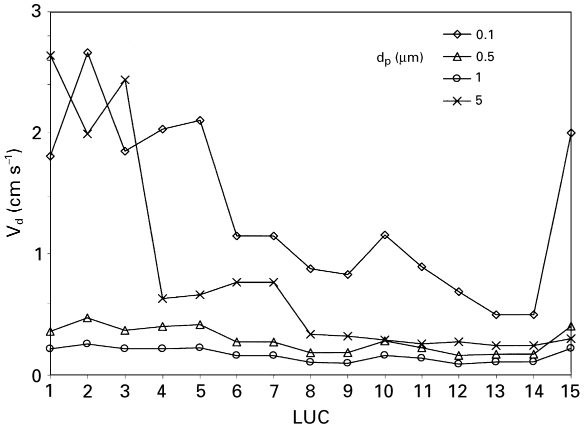

Fig. 1. Sensitivity tests for dry deposition velocity depending on LUC for particles with a diameter of 0.1, 0.5, 1 and 5 μm.

5.1. Comparison with Wesely's sulphate dry deposition model

The dry deposition velocity for sulphate from Wesely et al. (1985) model is calculated using Eq. (1) with the gravitational settling velocity omitted. The inverse of the surface resistance R_s is defined as the surface deposition velocity V_ds, which is parameterized as

Unstable:

$$V_\mathrm{ds} = \frac{u_*}{500}\left[1 + \left(-\frac{300}{L}\right)^{2/3}\right], \quad \left(\frac{\mathrm{PBL}}{L} \geqslant -30\right),$$

$$V_\mathrm{ds} = 0.0009u_*\left(-\frac{\mathrm{PBL}}{L}\right)^{2/3}, \quad \left(\frac{\mathrm{PBL}}{L} < -30\right), \tag{11a}$$

Neutral or stable:

$$V_\mathrm{ds} = \frac{u_*}{500} \quad (L \geqslant 0), \tag{11b}$$

where PBL is the planetary boundary layer height and L is Monin–Obukhov length.

This model was originally based on field sulphate deposition data over grassland. Since there is no grid in which grassland is dominant within the current model domain, the Grid (05,12), contains of 100% LUC 7 (crops and mixed farmland) is chosen as an alternative. We assume that the farmland has aerodynamic characteristics similar to grassland since both of these two LUCs have relatively low canopies. The observed size range of sulphate was not reported in Wesely et al. (1985) and here

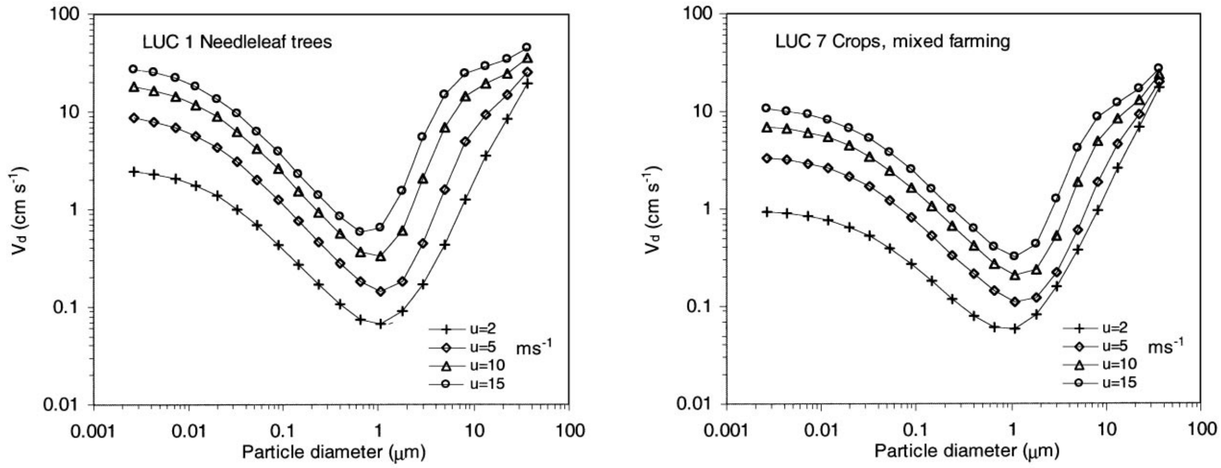

Fig. 2. Sensitivity tests for dry deposition velocity depending on wind speed for LUC 1 and 7.

the MMD and GSD for sulphate are chosen as 0.35 μm and 2.0, respectively.

Fig. 3 shows a comparison of V_d from the present model and Wesely et al. (1985) model using assigned arbitrary meteorological inputs for LUCs 2 and 14. For LUC 2, the broadleaf trees, the two models predict very close values. The correlation coefficient is also high (0.88). For LUC 14, the water surface, the current model also predict very close values, even through slightly higher, comparing to wesely's model. The correlation coefficient is 0.73. Most models for particle dry deposition usually differ from each other by more than one order of magnitude for this particle size range (Ruijgrok et al., 1995). By comparison, the present model performs better.

Fig. 4 shows a comparison of V_d for three selected model grids from the present model and Wesely's model under same meteorological conditions produced by RCM. It is seen that, for LUCs 4 and 7 in unstable conditions (daytime), V_d from the present model is lower than those from Wesely's model. This can be explained from the inclusion of "L", the Monin–Obkohov length in Wesely's model for unstable conditions. "L" could be very small at weak wind conditions and thus, V_d predicted from Wesely's model could be higher. Under stable conditions, V_d predicted from present model is higher than those from Wesely's model. This is not always true for LUC 14 (an ocean surface), since the stability over a water surface does not have the variability which occurs over a land surface. The wind speed is also larger over the ocean surface. These physical features cause near neutral conditions over an ocean surface which lead to large "L" values. However, the daily averaged V_d for all three LUCs from present model agree well with those from Wesely's model (not shown in figure). Comparing the values shown in Fig. 4 with measurements shown in Table 4, it is found that the current parameterization predict very reasonable deposition velocities.

5.2. Comparison with Ruijgrok's model

The Ruijgrok et al. (1997) model is an empirical fit based on extensive field date over forest (mostly needleleaf forests with some mixed forests). The model also uses Eq. (1) to calculate V_d but the surface deposition velocity V_ds is parameterized differently, using the following formula:

$$V_\mathrm{ds} = E\frac{u_*^2}{u_h}, \tag{12}$$

where, u_h is the wind speed at the top of the canopy and E is the total collection efficiency with which the canopy captures particles. The collection efficiency is parameterized as

$$E = \begin{cases} A_1 u_*^{A_2}, & \mathrm{RH} \leqslant 80, \\ A_1 u_*^{A_2}\left[1 + A_3 \exp\dfrac{\mathrm{RH} - 80}{20}\right], & \mathrm{RH} > 80, \end{cases} \tag{13}$$

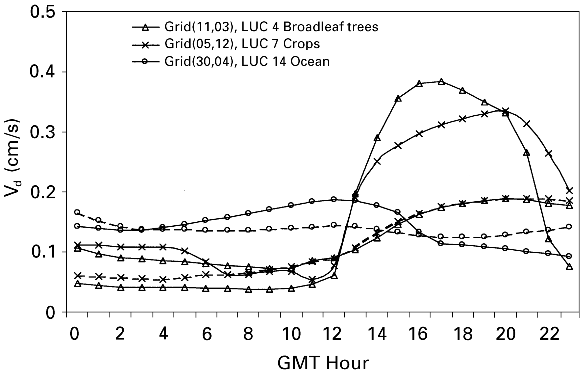

Fig. 4. Comparison of hourly averaged dry deposition velocity of sulphate between the present model (dashed lines) and Wesely's model (solid lines) for three model grids representing LUC 4, 7 and 14 from CAM model.

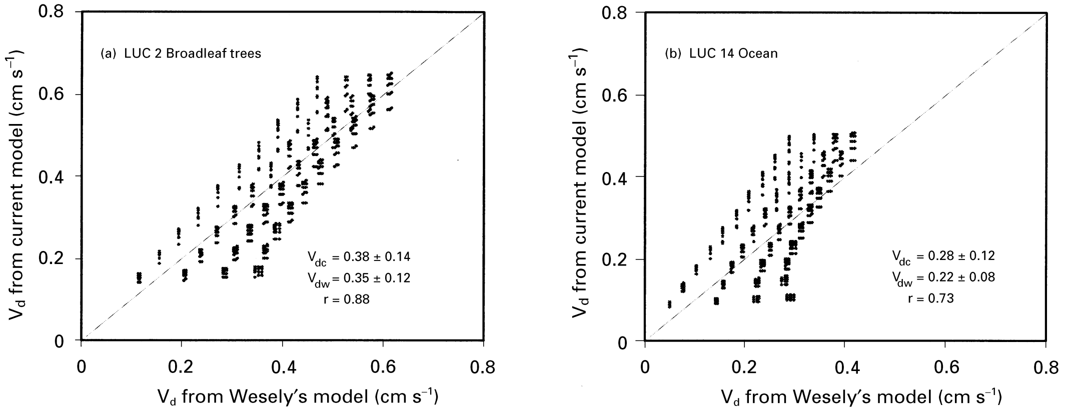

Fig. 3. Comparison of dry deposition velocity of sulphate between the present model and Wesely's model using arbitrary given meteorological input for LUC 2 and 14.

where A_1, A_2 and A_3 are empirical constants and have the following values for Na^+

$$A_1 = 0.14,\quad A_2 = 0.12,\quad A_3 = -0.09 \quad \text{dry surface}$$

$$A_1 = 0.14,\quad A_2 = 0.12,\quad A_3 = 0.37 \quad \text{wet surface}$$

The MMD and GSD for Na^+ are taken as 5.12 and 2.64 μm, respectively, as is reported in Ruijgrok et al. (1997).

Fig. 5 shows a comparison of V_d from the present model and the Ruijgrok et al.'s (1997) model using assigned meteorological inputs for LUC 1, the needleleaf trees. It is shown that, the V_d predicted from the two models agrees well, although the current model underpredicted V_d when V_d is small and overpredicted V_d when V_d is larger. The averaged deposition velocities from two models are very close and the correlation coefficient is very high (0.99). Since Ruijgrok et al. (1997) model is based on a needleleaf forest (although they may use it for other forests), the parameterization of the present model gives V_d over needleleaf forests (LUCs 1 and 3) with comparable values to Ruijgrok et al. (1997) model. For broadleaf forests and mixed forests (LUCs 2, 4 and 5), the characteristic radius is assigned to be larger than that for needleleaf forest, thus, V_d over broadleaf forests from present model is expected to be smaller than that from Ruijgrok et al. (1997) model. It is probably reasonable to expect the needleleaf forests to capture large particles more effectively than broadleaf forest (Pleim et al., 1984). However, this is difficult to quantify.

Fig. 6 shows a comparison of V_d for two selected model grids from the present model and the Ruijgrok et al.'s model, under the same meteorological conditions produced by RCM. It is seen that, for LUCs 1, V_d from the present model agrees very well with Ruijgrok's model. While for LUC 4, V_d from the present model is lower than from Ruijgrok's model. In some other models (e.g. Giorgi, 1988), the characteristic radii for needleleaf forest are assigned very small values (0.5 mm) and for broadleaf forest, they are relatively large (10 mm). In the present model, the characteristic radii are given as 2 and 5 mm for needleleaf and broadleaf forest, respectively (for SC 1). The present model shows that the V_d (shown in Fig. 5) for a needleleaf forest can be a few times larger than for a broadleaf forest for Na^+. Since particles at this size range are controlled by impaction, which is a function of Stokes number (*St*), and *St* is very sensitive to the characteristic radii ("*A*"), it can be expected that V_d for a needleleaf forest could be more than ten times larger than for a broadleaf forest if very smaller "*A*" is chosen for needleleaf forest and much larger "*A*" is chosen for broadleaf forest, as was done in Giorgi (1988). There is not enough evidence to indicate that V_d for needleleaf forest can be that much larger than for broadleaf forests. Besides, V_d for broadleaf forests with hairy leaves can also be very large (Pleim et al., 1984). Thus, in the present model, V_d for broadleaf forest is parameterized to give just slightly smaller values than for needleleaf forests.

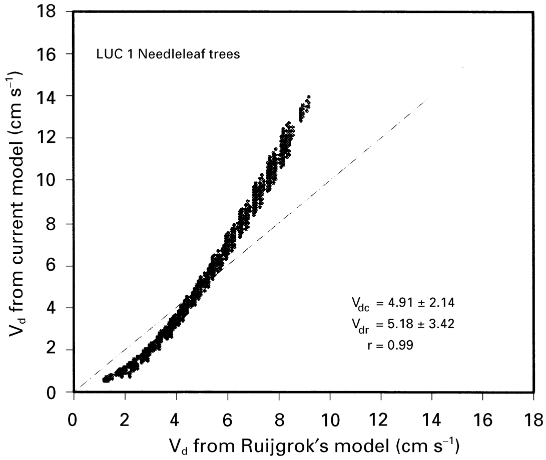

Fig. 5. Comparison of dry deposition velocity of Na^+ between the present model and Ruijgrok et al. (1997) model using arbitrarily assigned meteorological input for LUC 1.

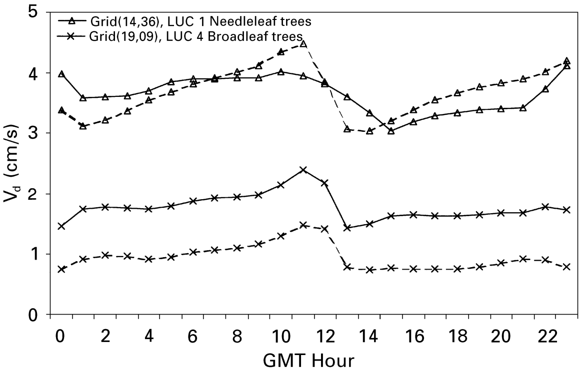

Fig. 6. Comparison of hourly averaged dry deposition velocity of Na^+ between the present model (dashed lines) and Ruijgrok et al. (1995) model (solid lines) for two model grids representing LUC 1 and 4 from CAM model.

5.3. V_d *for eastern–northern American*

Fig. 7 shows an example of modelled V_d at time 20:00 GMT, 20 August 1995, from CAM, in which the present dry deposition model is implemented. The particles modelled here are mixed aerosols composing of sulphate and sea-salt, for which the densities are 1769 and 2170 kg m^{-3}, respectively. For particles with a size of 0.24 μm as shown in Fig. 7a, V_d is in the range of up to 0.75 cm s^{-1}. It has relatively small values (< 0.35 cm s^{-1}) over smooth land surfaces and water surfaces (lakes and oceans); larger values (0.3–0.75 cm s^{-1}) over forest areas;

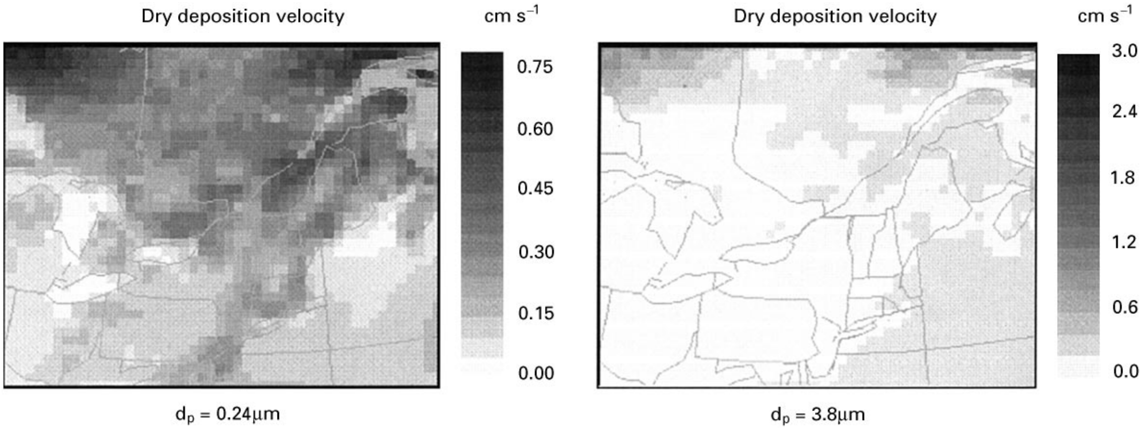

Fig. 7. Modelled V_d from CAM at 20:00 GMT, 20 August 1995 over east North America for particle with a diameter of (a) 0.24 μm and (b) 3.8 μm.

and 0.1–0.5 cm s^{-1} for the other surface types. It is noticed that V_d over ocean is higher than that over lakes. This is probably because the wind speed is usually higher than that over lakes. Comparing the value in this figure to the measurements presented in Table 4, it is seen that the model results are well within the range of observations.

For particles with a size of 3.8 μm, V_d can be up to 3.0 cm s^{-1}. The highest V_d values are over needleleaf area. It is seen that V_d over ocean are much higher than some land surfaces. This is because the particles grow very quickly at a humidity near 100%. In Fig. 2, we can see that for particles larger than 2 μm, V_d increase rapidly with the increase of particle size. The spatial distribution of V_d shown in Fig. 7 seems reasonable. The range of V_d values are within the range of field experiments (Schmel, 1984; Nicholson, 1988; Davidson and Wu, 1990; Hofschreuder et al., 1997).

6. Conclusions

A parameterization for particle dry deposition has been developed in the present study. The dry deposition velocity is calculated as a function of particle size and density, as well as meteorological conditions. Sensitivity tests and comparison with published measurements show that the parameterization can predict reasonable deposition velocities for a wide particle size range over different surface types. Model assessment shows that the present parameterization can predict deposition velocities for different particle size ranges which are comparable to some empirical models based on field data. This model is suitable for use in aerosol models like CAM. Model application to eastern North America shows that it can compute reasonable spatial patterns of particle dry deposition velocities. However, due to limited knowledge on particle dry deposition, the present parameterization with empirical and simplified formula needs further improvements.

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