

Estimates of sulfate aerosol wet scavenging coefficient for locations in the Eastern United States

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Received 23 December 2002; received in revised form 20 May 2003; accepted 28 October 2003

Abstract

Scavenging of atmospheric aerosols by falling precipitation is a major removal mechanism for airborne particles. The process can be described by a wet scavenging coefficient (WSC), denoted L , that is dependent on the rainfall rate, R , and the collision efficiency between raindrops and aerosol particles, E . We report bulk average L values for location in the Eastern United States, estimated based on sulfate mass balance in the atmospheric domain of interest. Data used are taken from several observational networks: (a) the Atmospheric Integrated Research Monitoring Network (AIRMoN) which is part of the National Atmospheric Deposition Program/National Trends Network (NADP/NTN); (b) the Interagency Monitoring of Protected Visibility Environments (IMPROVE); and (c) the National Climatic Data Center (NCDC). The results are fitted relatively well by L values computed using a microphysical representation of the WSC process based on collision efficiency and precipitation size distribution. Such representation leads to a simple expression $L = f(R)$ for soluble aerosols, suitable for WSC description in regional scale models. The agreement between the bulk method and the microphysical representation is due in part to the predominant widespread precipitation, well represented by Marshall and Palmer raindrop distribution, and in part due to assumptions made in the bulk model. Results indicate that high-resolution rainfall rates and realistic vertical cloud structure information are needed to improve the accuracy of aerosol wet scavenging modeling for pollution studies.

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Keywords: Sulfate; Wet deposition; AIRMoN; Acid rain; Cloud

1. Introduction

The wet removal of atmospheric particles (AP) by precipitation is by far the most efficient atmospheric aerosol sink. This process influences the tropospheric aerosol turnover time and the magnitude of aerosol effects on environment (Rodhe and Grandell, 1972; Jaenicke, 1993). The detailed mechanism of the wet scavenging process involves microphysical interactions between AP and hydrometeors (Hales, 1995; Pruppacher and Klett, 1997; Jennings, 1998). For practical considerations, the APs wet removal is typically represented in current models by scavenging coefficients in aerosol

mass continuity equations. These scavenging coefficients are expressed as function of bulk quantities available from measurements, such as the precipitation rate at the ground. A review of published aerosol WSC determined from measurements and model calculations shows a large spread of data due in part to variability of raindrop size distribution, and collection efficiency between AP and raindrops (McMahon and Denison, 1979; Okita et al., 1996). Moreover, recent reports show that the representation of removal processes in current aerosol transport models remains a significant source of uncertainty (Rasch et al., 2000). Given the current importance of aerosol pollution and its implications at various scales, the study of the wet removal processes remains a crucial task for understanding the fate of airborne particulate matter (Slinn and Hales, 1971;

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Wang and Pruppacher, 1977; Grover et al., 1977; Wang et al., 1978; Dana and Hales, 1976; Radke et al., 1980; Scott, 1982; Chang, 1986; Jylha, 1991, 1999; Sparmacher et al., 1993; Hales, 1995; Okita et al., 1996; Mircea and Stefan, 1998; Jennings, 1998; Tinsley et al., 2000; Andronache, 2003).

An AP is removed from atmosphere by rainfall when it becomes incorporated into a raindrop that falls to the Earth's surface. Exceptions are weather situations in which fog and cloud droplets come in direct contact with ground surfaces (trees, buildings, hills, mountains) and these cases are not treated here. For the wet removal by precipitation, an AP can be incorporated into a raindrop following one of these scenarios: (a) AP initially at the cloud base becomes cloud droplet by the nucleation scavenging process. Some cloud droplets grow by collision and coalescence and reach the size of a raindrop (diameter ~ 0.1 – 5 mm). While the number concentration of raindrops is smaller than the number concentration of cloud droplets, the raindrops play a crucial role in the wet removal process because they collect other cloud droplets, interstitial aerosols as well as below cloud aerosols; (b) AP becomes cloud droplet by nucleation scavenging (diameter ~ 10 μm), and is collected in-cloud by a falling raindrop. This is the in-cloud scavenging (ICS) of aerosol present in cloud droplets (ICS_{drop}); (c) AP becomes in-cloud interstitial aerosol, remaining with a diameter typically less than 1 μm . Some of these interstitial particles are collected by collision with droplets and raindrops. This is the ICS of interstitial aerosol (ICS_{int}). The ICS process can be viewed as a cumulative effect of the ICS_{drop} and ICS_{int}; and (d) AP is present below the cloud base and is collected by falling raindrops by the below-cloud scavenging (BCS) process.

The scenarios presented above involve one or both of the following processes: (1) nucleation scavenging, and (2) impaction scavenging. In the nucleation scavenging, APs serve as cloud condensation nuclei (CCN) or ice nuclei (IN) in the initial stage of cloud formation. These processes are controlled largely by the ambient water vapor supersaturation and to a less extent by the chemical composition of aerosols. It has been shown that the nucleation scavenging is a dominant process at the beginning of cloud formation and this process can deplete the original aerosol population in air up to 75–90% (Flossmann et al., 1985, 1987; Ten Brink et al., 1987; Schumann, 1991; Pruppacher and Klett, 1997). Thus, the role of nucleation scavenging is to transfer aerosol mass from “dry” form into cloud droplets. In contrast, the impaction scavenging transfers aerosol mass from cloud droplets, interstitial aerosol and below-cloud aerosol into falling raindrops. The impaction scavenging depends on the net action of various forces influencing the relative motion of APs and hydrometeors in-cloud and below-cloud. Similar mechanisms act

during ICS and BCS. These mechanisms are: convective Brownian diffusion, interception, inertial impaction, thermophoresis, diffusionphoresis, airflow turbulence and electrostatic attraction (Pruppacher and Klett, 1997; Seinfeld and Pandis, 1998). Analysis of field measurements show that by nucleation scavenging most of the aerosol mass is transferred into cloud droplets and a small fraction of aerosol mass remains as interstitial aerosol. Thus, the ICS_{drop} has a much larger contribution to the wet scavenging than ICS_{int} because: (1) raindrops collect very efficiently cloud droplets with diameters of ~ 10 μm ; (2) less aerosol mass is present in the interstitial aerosol with diameters less than ~ 1 μm . This conclusion is well supported by recent field experimental data which show that the sulfate aerosol particles with diameter greater than 0.1 μm becomes activated, and overall, the fraction of the particle volume, that is incorporated in cloud droplets is 70–92% (Hallberg et al., 1994; Svenningsson et al., 1997; Martinsson et al., 1999). Similarly, BCS is less significant than ICS_{drop} because the efficiency of collision between falling raindrops and below cloud aerosol (most with sizes in the accumulation mode) is much smaller (Greenfield, 1957; Dana and Hales, 1976; Byrne and Jennings, 1993; Pruppacher and Klett, 1997). Exceptions are cases with coarse particles in the BL (such as marine aerosol at high wind intensity and dust storms, but these cases are not characteristic for the region studied here and for the aerosol type considered). In this sense, for most cases, WSC is well approximated by ICS_{drop} coefficient and this approach will be followed in this paper when we present a microphysical representation of the wet scavenging coefficient (WSC).

The methods used to determine the aerosol WSC, as reported in previous work, are: (1) microphysical model (MM) based on prescribed hydrometeor size distribution and collection efficiency between raindrops and aerosols (Scott, 1982; Chang, 1986; Mircea and Stefan, 1998; Jylha, 1991; Pruppacher and Klett, 1997), and (2) bulk average estimations of the scavenging coefficient based on the scavenging ratio determined from observations (Hales, 1995; Okita et al., 1996). Ultimately, a scavenging coefficient based on prescribed collision efficiency, E , and rainfall rate, R , is needed in mesoscale and large scale models of aerosol transport as well as for rapid assessments of aerosol fate in local pollution studies. The goals of this work are: (1) calculate bulk average values of L based on mass balance of sulfate at several AIRMoN sites in the Eastern United States; (2) compare L bulk values with estimations based on a MM of L for sulfate aerosol for hourly precipitation rates, R , at the same locations. A discussion of the statistics of the scavenging coefficients in the Eastern US and implications for the 0.5-folding time of sulfate aerosols with respect to the wet removal process are also presented.

2. Method

2.1. Bulk method

The bulk method (BM) estimates of the average value of L are based on the mass balance of the aerosol of interest in an imaginary atmospheric box above a station of observation. The box covers a horizontal area A and extends from surface to a height h_2 (Bolin and Rodhe, 1973; Hales, 1995; Okita et al., 1996). For the discussion presented here, we refer to SO_4 aerosol, the choice being dictated by the availability of data and by the fact that sulfate is a soluble aerosol. Under these conditions, we can compare the bulk values of L derived from sulfate and precipitation observations with the model described in Section 2.2, which is valid for the removal of soluble aerosols. Thus, $L = FM^{-1}$ where F is the flux of aerosol at the Earth’s surface and M the mass of aerosol in the considered box. The flux F can be expressed as, $F = C_wRA$, where C_w is the SO_4 concentration in precipitation water recorded at the Earth’s surface, and R is the precipitation rate. The aerosol mass in the box is $M = \langle C_a(z) \rangle Ah$, where $\langle C_a(z) \rangle$ is the SO_4 vertical average concentration in the box, and h is the height of the cloud layer with precipitation that significantly contributes to the wet removal of aerosol (assumed mainly between the cloud base level and the freezing level). Thus

$$L = \frac{F}{M} = \frac{C_wR}{\langle C_a(z) \rangle h}, \tag{1}$$

where

$$\langle C_a(z) \rangle = \frac{1}{(h_2 - h_1)} \int_{h_1}^{h_2} C_a(0) \exp\left(-\frac{z}{H}\right) dz \tag{2}$$

with the solution

$$\langle C_a(z) \rangle = \frac{C_a(0)H}{h} \exp\left(-\frac{h_1}{H}\right) \left[1 - \exp\left(-\frac{h}{H}\right) \right], \tag{3}$$

where, $C_a(0)$ is the SO_4 concentration measured at surface, h_1 the cloud base height, H the scale height of SO_4 aerosol, h_2 the height of the box, and $h = h_2 - h_1$. We define a factor, f that depends on the characteristic heights h_1 , h , and H

$$f = H \exp\left(-\frac{h_1}{H}\right) \left[1 - \exp\left(-\frac{h}{H}\right) \right] \tag{4}$$

and L becomes

$$L = \frac{C_wR}{C_a(0)f} \tag{5}$$

which is applied to measurements taken at stations from the networks described below (Table 1). The dependence of f on h for several values of the scale height of sulfate is discussed in Section 2.2. We note that with this formulation, L can be calculated for observations made at a time scale of the order of 1 h similar to the approach taken by Okita et al. (1996).

2.2. Microphysical model

The MM used to estimate WSC follows the concepts introduced by Scott (1982) and is illustrated here for removal of spherical APs by falling spherical raindrops inside clouds. Similar considerations apply for removal of APs by snowfall. Typically, the aerosol removal by snowfall is less efficient than the removal by rainfall and scavenging coefficient estimations are subject to more uncertainties due to the complex shapes of hydrometeors (Scott, 1982; Pruppacher and Klett, 1997; Jennings, 1998). Moreover, the rainfall removal is the dominant

Table 1

AIRMoN network stations used in this study and average $\langle \rangle$, and standard deviation, Std, of the number of rainy hours per day, n_h , the precipitation rate, R , and bulk average wet scavenging coefficients for sulfate aerosol, L

Station	$\langle n_h \rangle$ (h)	Std(n_h) (h)	$\langle R \rangle$ (mm h ⁻¹)	Std (R) (mm h ⁻¹)	$\langle L \rangle$ (h ⁻¹)	Std (L) (h ⁻¹)	a	b
DE02 Lewes	4.8	4.2	2.7	2.8	4.56	10.89	1.42	0.83
FL18 Tampa Bay	3.0	3.0	4.7	5.7	7.79	10.3	2.49	0.71
IL11 Bondville	3.7	3.0	1.6	1.9	2.08	2.65	1.26	0.93
MD15 Smith Island	4.5	3.8	2.5	3.5	2.17	2.45	1.06	0.74
NY67 Ithaca	5.0	4.0	1.6	1.8	2.17	2.47	1.18	0.81
PA15 Penn State	4.6	4.0	2.0	3.1	1.80	2.24	1.03	0.80
TN00 Walker Branch W.	4.5	4.0	2.6	3.0	4.13	5.39	1.69	0.72
VT99 Underhill	4.8	3.9	1.6	2.9	2.42	3.46	1.37	0.68
WV99 Caanan Valley	5.0	4.0	1.5	2.1	1.55	2.4	0.92	0.87
All stations					3.9	7.4	1.43	0.81
Model							1.26	0.78

The coefficients a and b of the least squares fit of the form $L = aR^b$ with L (h⁻¹) and precipitation rate R (mm h⁻¹) based on the bulk method are shown in the last two columns. The last row shows a and b corresponding to the microphysical model.

process for the locations of interest here. As shown in Section 1, the WSC is approximated by the ICS_{drop} coefficient and the contributions of the scavenged interstitial aerosol mass and the below cloud aerosol to the wet removal can be neglected. APs enter the cloud by nucleation scavenging and grow rapidly by condensation of water vapor to sizes of about 10 μm . Consider an AP of diameter a collected by raindrops of diameter D . The number of collisions per unit time experienced by a falling raindrop is

$$\frac{\pi D^2}{4} V(D)E(a, D)n(a) da, \quad (6)$$

where $V(D)$ is the raindrop fall velocity, $E(a, D)$ the collision efficiency of the raindrop of diameter D with an AP of diameter a ; and $n(a)da$ is the number concentration of collected particles with diameters between a and $a + da$. The rate of accumulation of particle mass experienced by a single raindrop is

$$\frac{\pi D^2}{4} V(D)E(a, D)x(a) da, \quad (7)$$

where $x(a)$ is the mass of AP with diameter a . Integrating (7) over all sizes of collecting raindrops, the result is the total collected particles of diameter a by precipitation

$$\frac{\delta x(a) da}{\delta t} = -x(a) da \int_0^\infty \frac{\pi D^2}{4} V(D)E(a, D)N(D) dD, \quad (8)$$

where $N(D)dD$ is the precipitation size distribution. By definition

$$L(a) = \int_0^\infty \frac{\pi D^2}{4} V(D)E(a, D)N(D) dD \quad (9)$$

is the ICS coefficient, and this is a good approximation for the WSC (Scott, 1982). Scott considered a typical AP diameter inside cloud of 10 μm (after the growth of AP from activated aerosol particle to a cloud droplet size) and selected a value $E = 0.65$ which will be used for calculations in this work. This E value corresponds to soluble aerosols and will be used here for sulfate aerosols. Experimental data show cases where L is compatible with model calculations for $E = 0.1$ – 0.2 which will correspond to less soluble aerosols (and possibly to different hydrometeors size distribution). Following Scott (1982), a is assumed 10 μm , and we drop the dependence of L on a . L is linked to the rainfall rate, R , defined as (in units mm h^{-1})

$$R = 3.6 \int_0^\infty X(D)V(D)N(D) dD. \quad (10)$$

Defining the total water concentration M (in units g m^{-3})

$$M = \int_0^\infty X(D)N_0 \exp(-\lambda D) dD, \quad (11)$$

where $X(D)$ is the mass of one raindrop, and assuming a Marshall and Palmer (1948) precipitation size distribution

$$N(D) dD = N_0 \exp(-\lambda D) dD \quad (12)$$

with $N_0 = 1 \times 10^8 \text{ m}^{-4}$ and $\lambda = 2367 M^{-0.25} (\text{m}^{-1})$, and considering the relationships $V(D) = 130 \times D^{0.5} (\text{m s}^{-1})$ and $M = 0.074 R^{0.89}$ (M in g m^{-3} and R in mm h^{-1}) (Kessler, 1969), it can be shown that

$$L = 1.26 \times R^{0.78}, \quad (13)$$

where L is in units of h^{-1} and R is in units of mm h^{-1} .

L estimations using this model are in general good agreement with values determined from field studies (Okita et al., 1996; Jylha, 1991) while significant variability was observed from case to case. Such variability was attributed to changes in aerosol solubility, precipitation type, vertical distribution of the precipitating system and raindrop size distribution. Nevertheless, the model presented here provides a reliable framework for calculations of L for soluble aerosols. Based on results from Jylha (1991) and Okita et al. (1996) and from data reviewed by McMahan and Denison (1979) it can be inferred that WSC can be expressed as $L = aR^b$ with a in the range [0.36–1.28] and b in the range [0.5–1.0] (with most recent values in the range [0.64–0.78]). To estimate L using the above model, we use average hourly rainfall rates R based on the NCDC data analysis for the stations listed in Table 1 and we use E value valid for soluble aerosols.

3. Results

3.1. Estimations of sulfate bulk average wet scavenging coefficients

Calculations of average bulk values of L are made using data from several networks. Thus, C_w and R are measured on a daily basis in the Atmospheric Integrated Research Monitoring Network (AIRMoN). The AIRMoN program is part of the National Atmospheric Deposition Program/National Trends Network (NADP/NTN) (Hales et al., 1987; Gilliland et al., 2002). This network was formed for studying precipitation chemistry trends with greater temporal resolution. Precipitation samples are collected daily from a network of nine sites and analyzed for the same constituents as the AIRMoN network see Hicks et al., 2001). Daily values of $C_a(0)$ are taken from the Interagency Monitoring of Protected Visibility Environments (IMPROVE) program. The IMPROVE monitoring network consists of aerosol, light scatter, light extinction and scene samplers in several National Parks and Wilderness areas (Trijonis et al., 1990; Malm et al., 1994a, b, 1996). Hourly values

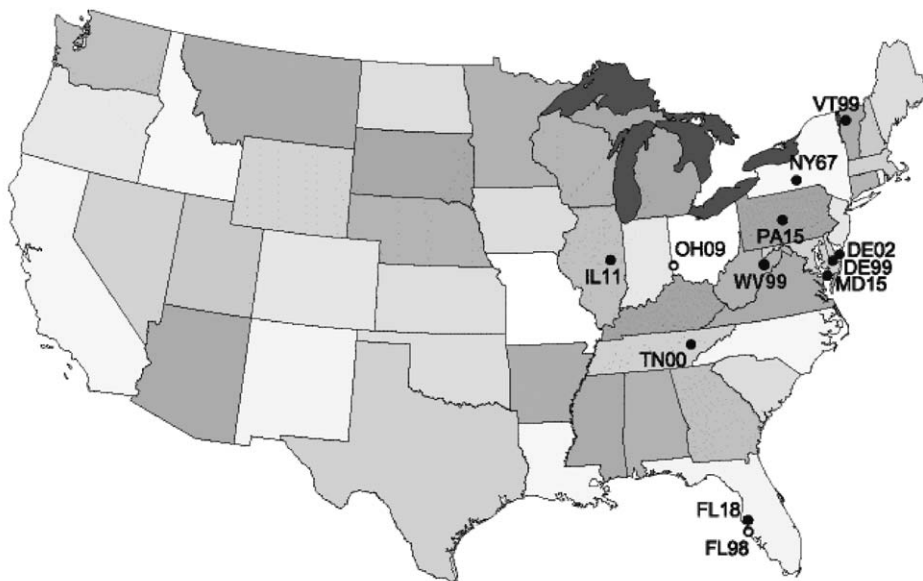


Fig. 1. AIRMoN sites. The full name of the stations used in this work is given in Table 1.

of precipitation rate, R , are taken from the National Climatic Data Center (NCDC). A map with the location of all stations from the AIRMoN network is shown in Fig. 1. Nine of the total of the 12 AIRMoN network stations are used in this study and are listed in Table 1. Three AIRMoN stations (indicated on map as DE99, OH09 and FL98) were not considered because they do not have sufficient data for the analysis performed here. For each analyzed station we selected data for cases with concurrent measurements of $C_a(0)$ from the IMPROVE network. The time series of daily values of C_w and $C_a(0)$ at the nine analyzed stations are shown in Fig. 2. While the full data sets for each variable is much larger, only the data from days with concurrent measurements of air and rainwater sulfate concentrations are shown. Data points for the sulfate in rainwater, C_w , are connected by a solid line to improve the clarity of the figure, and the line does not necessarily reflect changes from one point to another. Generally, we found a significant positive correlation between C_w and $C_a(0)$. This is consistent with the physical mechanism by which larger sulfate concentration in air leads to more activated particles as cloud droplets and therefore more sulfate collected by raindrops. Note also that the AIRMoN network provides daily values, thus R from this network is a daily accumulated precipitation. However, our analysis on the NCDC hourly precipitation data for these stations indicates that on average the number of hours with precipitation is about 3–5 h/day (with a standard deviation in the range 3–5 h). Fig. 3 illustrates histograms of the number of rainy hours per day indicating that it is more frequent to have only a few

rainy hours per day. As these plots show, it is very unlikely to have rain events that persist more than several hours. We use this statistics to infer hourly averages of L to be compared with the model-estimated values.

To calculate average bulk L , the value of factor f in Eq. (4) must be determined. We consider a typical height scale of SO_4 , $H = 2$ km (Hegg et al., 1984), and h_1 is taken as the top of the BL (1 km). A typical value of h is about 1 km, a value supported by the following considerations: (1) precipitation rate decreases with height and becomes less intense above 2–3 km from ground (Scott, 1982); (2) radar observations of frontal systems show predominance of the top of convective cells in the range 2–3 km (Okita et al., 1996); and (3) observations show that the melting height is at about 2–3 km altitude and the rainfall rate is more significant below the melting height where the main wet removal occurs in cloud (Mittermaier and Illingworth, 2003). Thus, h is the thickness of the cloud layer most active in wet scavenging and is taken 1 km for the calculations reported here, while variability in h is expected from case to case. The factor f is plotted in Fig. 4 for h varying between 0 and 5 km and for three values of H . We note that f depends on h but the sensitivity of f to h is also a function of the sulfate vertical distribution. For sulfate confined mainly in the BL, f has little dependence on h , while for sulfate well mixed in the troposphere, f tends to increase with h at a higher rate. For the base calculation we choose $f = 0.48$ corresponding to $H = 2$ km and $h = 1$ km. We will illustrate the sensitivity of f values to h when we present the variations of

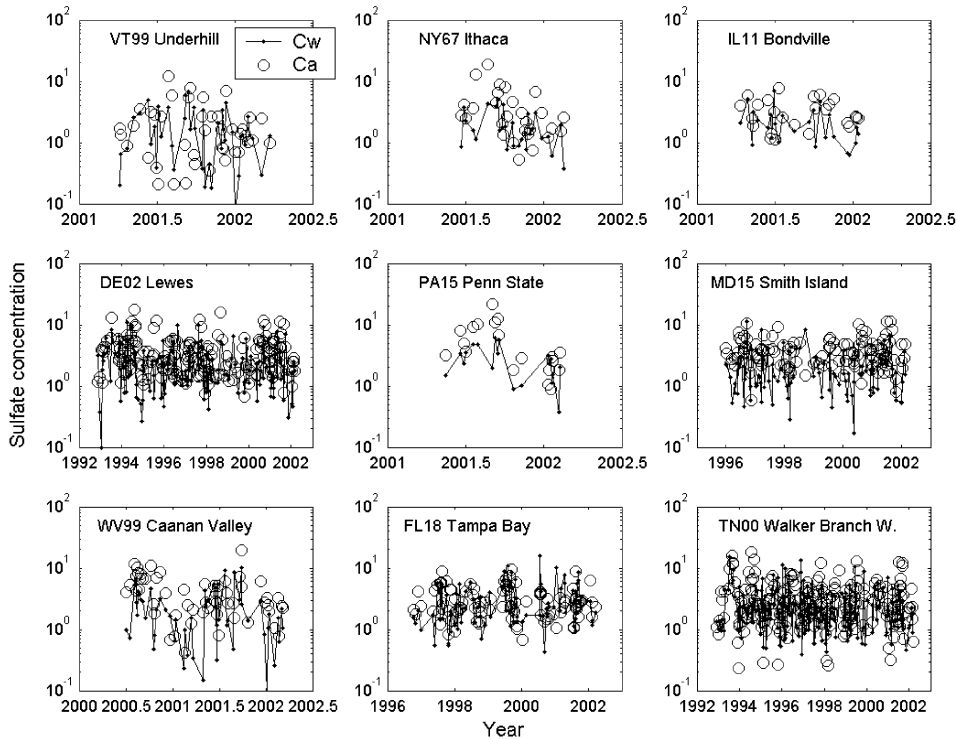


Fig. 2. Time series of daily rainwater sulfate concentration (C_w in units of mg l^{-1}) and in sulfate concentration in air near the ground ($C_a(0)$ in units of $\mu\text{g m}^{-3}$) from the nine AIRMoN stations analyzed. Data corresponding to rainwater sulfate concentration are linked by a solid line for clarity of the figure. Only days with concurrent measurements of sulfate in rainwater and air are shown.

0.5-folding time with rainfall rate in Section 3.2. With this reference value for h , the bulk estimates of L are shown in Fig. 5 for all nine stations. The least-squares fit lines are also plotted and the parameters are given in Table 1 for each station. For all stations, there is significant positive correlation between L and the rainfall rate, R . Note that the fit compares well with the model described above by Eq. (13) and shown in Fig. 6 where data from all nine stations are combined. The MM estimations for $E = 0.65$ agree quite well with the bulk estimations. To illustrate the upper limit of MM estimations, we plotted L for $E = 1$. These estimations are consistent with other reported results obtained from other cases studies (McMahon and Denison, 1979; Okita et al., 1996; Jylha, 1991) while we must keep in mind many uncertainties related to parameters involved. The BM average value of L and the standard deviation for each station are also listed in Table 1. Thus, the average L is in the range $[2\text{--}8]\text{h}^{-1}$ with a standard deviation in the range $[2\text{--}10]\text{h}^{-1}$. These results show that overall the estimated bulk average values of L are statistically consistent with the estimations based on the MM for soluble aerosols. These L values can provide a rapid estimation of the residence time of aerosol after the rain starts, analyzed in Section 3.2.

3.2. Aerosol wet removal 0.5-folding time

The 0.5-folding time, $t_{0.5}$ can be used to characterize the decay of aerosol concentration after the start of precipitation in the domain of interest. It is defined as the time required to decrease the aerosol concentration to half of the initial concentration at the beginning of rain, assuming that during this process the rainfall rate R remains constant and that no other processes contribute to aerosol concentration in the domain of interest (no advection, no sources or other sinks of aerosol mass). This is an idealized parameter but valuable for a rapid estimation of how effective is the wet removal for a given rainfall event of intensity R . Considering that sulfate aerosol concentration is $C_a(t)$ at time t and $C_a(0)$ at $t = 0$ (as the beginning of rain), the time evolution of $C_a(t)$ is described by

$$\frac{dC_a(t)}{dt} = -LC_a(t) \quad (14)$$

with the solution $C_a(t) = C_a(0) \exp(-Lt)$. For $C_a(t_{0.5}) = C_a(0)/2$, we get $t_{0.5} = \ln(2)/L$.

Estimations of L show that for the reference calculations with $f = 0.48$ ($h = 1$ km) we find that $t_{0.5} \sim 0.5$ h for $R = 1$ mm h^{-1} (Fig. 7). For weaker precipitation

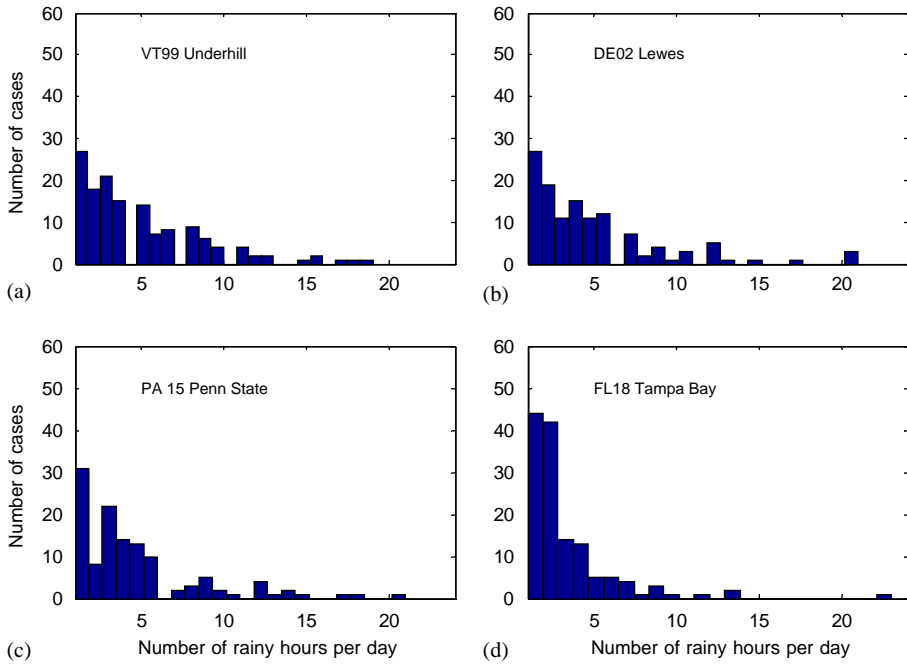


Fig. 3. Histograms of the number of rainy hours per day at four stations from Eastern United States.

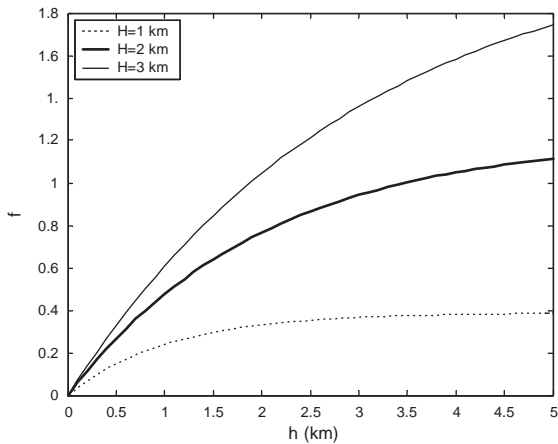


Fig. 4. The factor f versus the height, h , of the cloud layer most active in aerosol scavenging by liquid precipitation. H is the height scale of sulfate aerosol concentration in air.

rates, $R \sim [10^{-2} - 10^{-1}] \text{ mm h}^{-1}$, we determine $t_{0.5}$ in the range [3–20] h. We note that the removal of aerosol is less efficient for weak R , considering that the probability of a number of rainy hours greater than 5 h/day is small. In contrast, for intense precipitation, $R \sim [10 - 100] \text{ mm h}^{-1}$, $t_{0.5}$ is of the order of minutes. Thus, a short intense rainfall can be the most effective process for aerosol removal. Variations of f between 0.3 and 1.0 (corresponding to $h = 0.5$ and 3.5 km) caused variations

in $t_{0.5}$ as shown in Fig. 7. For $R = 1 \text{ mm h}^{-1}$, $t_{0.5}$ is in the range [0.3–1] h, and for $R = 0.1 \text{ mm h}^{-1}$, $t_{0.5}$ is in the range [2–6] h. Sensitivity calculations of these results to changes of limits of integration in the factor f from Earth surface to the freezing level show that conclusions do not change significantly. Integrating from the Earth’s surface instead of the cloud base height leads to potentially more aerosol removal below-cloud. However, as argued in Section 1, the efficiency of collisions, E , between BL aerosol and raindrops is much smaller than in-cloud E values and therefore, we expect that below-cloud precipitation has less contributions to the overall removal of aerosol in most cases.

4. Conclusions

Two distinct methods were used to determine the wet scavenging of sulfate aerosol for the Eastern United States locations of the AIRMoN. One method is the mass balance of sulfate aerosol and is based on observations of sulfate concentrations in air and rainwater as well as on rainfall rates. The second method is a MM based on the interactions between falling raindrops and sulfate aerosols. The relatively good agreement between the BM and the MM can be attributed in part to the predominant widespread precipitation type in the Eastern United States that are well represented by the MM, and in part due to assumptions made in the bulk model for the calculations of average values of

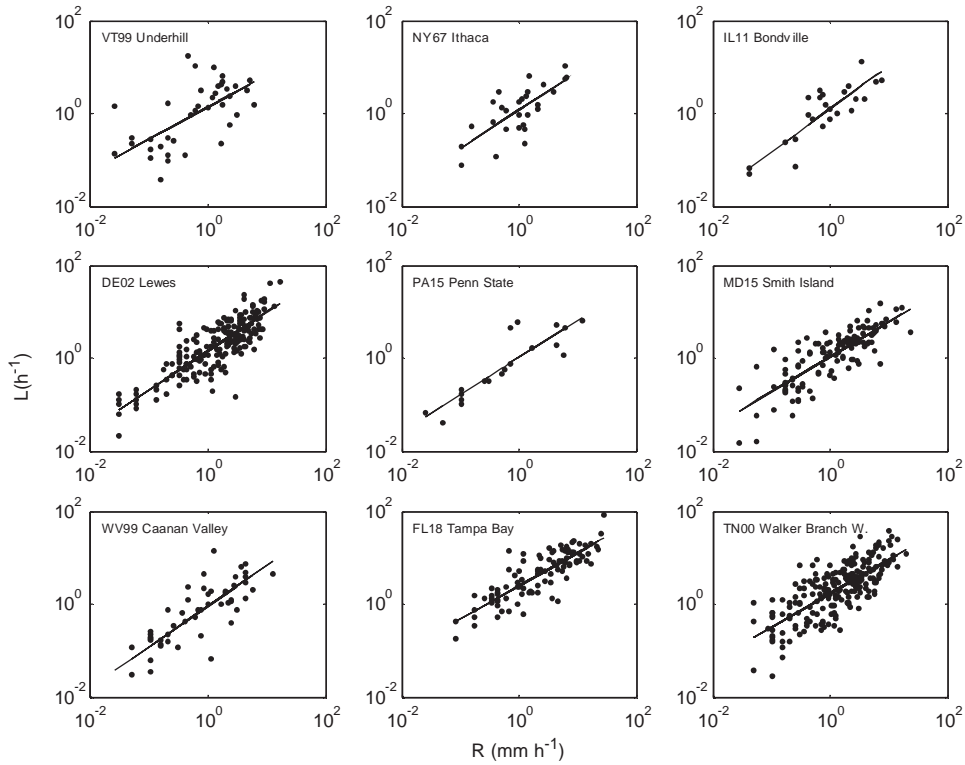


Fig. 5. WSC of SO₄ aerosol, L , versus precipitation rate, R , at nine locations from Eastern United States. The dots represent the bulk values of L estimated from observations of sulfate concentration in air and rainwater, and the lines are least-squared fit of the data. The parameters of the curve fit are given in Table 1.

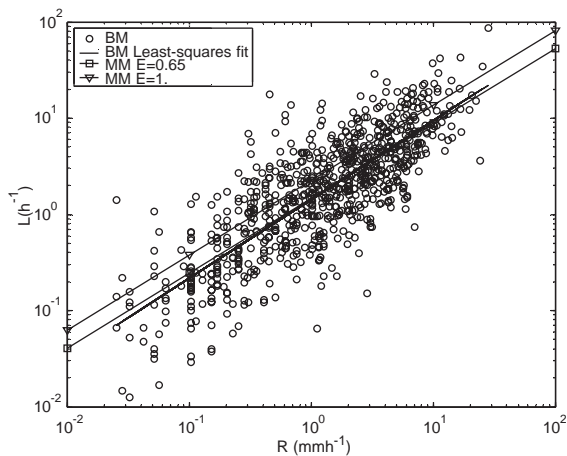


Fig. 6. WSC of SO₄ aerosol, L for all data combined from the nine stations analyzed versus rainfall rate R . Shown are: estimations from observations using the BM (open circles), least-squares fit of the BM estimations (solid line), and MM estimations corresponding to the collection efficiency $E = 0.65$ between raindrops and aerosol particles (line with squares). The MM estimations for $E = 1$ are shown by a solid line with triangles.

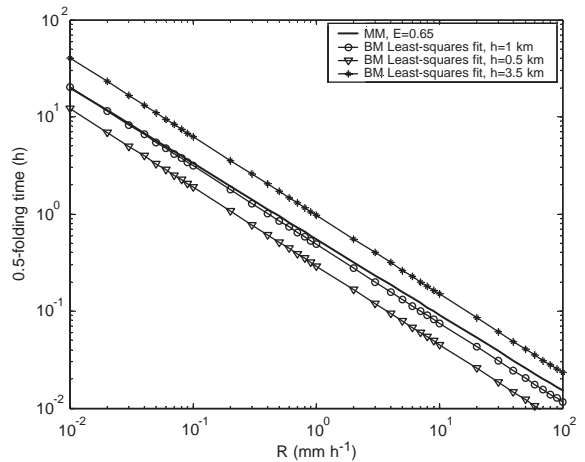


Fig. 7. The 0.5-folding time of sulfate concentration due to wet scavenging versus rainfall rate, R : MM calculations for $E = 0.65$ (solid line) and least-squares fit of the BM estimations for $h = 1$ km (solid line with circles). The solid lines with triangles and stars correspond to least-squares fit of BM data for $h = 0.5$ and 3.5 km and reflect variability caused by possible uncertainties of h , the cloud thickness with precipitation significant for the wet removal process.

scavenging coefficients. Significant differences between the two methods are expected for short precipitation events associated with deep convective systems, mixed precipitation, and for cases when aerosol is less soluble than sulfates.

We found that L varies between 2 and 8 h^{-1} , with a standard deviation in the range $[2\text{--}10]\text{ h}^{-1}$ for the stations analyzed. We found that for $R = 1\text{ mm h}^{-1}$, the 0.5-folding time of aerosol concentration, $t_{0.5}$, is about 0.5 h. Even for large uncertainties in the vertical extent of precipitation, $t_{0.5}$ is of the order of hours for weak precipitation, and is of the order of minutes for very intense rain ($R \sim 100\text{ mm h}^{-1}$). The use of high-resolution rainfall rate data reveals that $t_{0.5}$ is a useful parameter to characterize the decay of aerosol after the beginning of rain, which can be valuable in pollution studies.

The study also suggests that higher resolution data of aerosol concentrations, precipitation rate (such as hourly measurements), and simultaneous information on the vertical structure of precipitating clouds (that can be obtained from radars and regional weather modeling systems) can reduce the uncertainties in the BM and provide further insight for the microphysical modeling approach. Such methodology can be used to obtain more details of aerosol precipitation scavenging process with application to air pollution studies at local and regional scales.

Acknowledgements

The author acknowledges the use of data from the National Climatic Data Center (NCDC), from the Interagency Monitoring of Protected Visibility Environments (IMPROVE) and from the Atmospheric Integrated Research Monitoring Network (AIRMoN). The AIRMoN program is part of the NADP/NTN. The author thanks Jan Peter Muller, Robin Hogan, and Marion Mittermaier for kindly providing information and a manuscript on cloud vertical structure observations. The comments from two anonymous reviewers are very appreciated.

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