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Atmospheric Environment 37 (2003) 3605–3613

ATMOSPHERIC ENVIRONMENT

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Ultrafine particle scavenging coefficients calculated from 6 years field measurements

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Received 20 November 2002; accepted 10 April 2003

Abstract

Based on 6 years of outdoor measurements at a boreal forest site in Southern Finland, scavenging coefficients were calculated for aerosol particles having diameter between 10 and 510 nm. Median scavenging coefficients varied between 7×10^{-6} and $4 \times 10^{-5}\ \mathrm{s}^{-1}$ in this size-range. The dependence of scavenging coefficients on rain intensity was studied, and the scavenging coefficients were parameterized as a function of particle size for particle diameters of 10–500 nm and for rain intensities 0–20 mm h^{-1}.

PACS: 92.60.Mt; 92.60.Sz; 92.60.Jq

Keywords: Scavenging; Wet deposition; Rain wash-out; Deposition; Aerosol particles

1. Introduction

Wet deposition is an important aerosol particle removal mechanism in the atmosphere. In the submicron range particles are removed either by cloud processing, below-cloud scavenging or coagulation (Seinfeld and Pandis, 1998). A portion of the particles are removed by dry deposition. Cloud condensation nuclei activation during cloud formation as well as particle release during cloud dissipation are the principal mechanisms related to aerosol–cloud interaction to modify aerosol and cloud particle numbers. After the cloud is formed aerosol particles may collide with cloud droplets or be scavenged below cloud by falling rain droplets. Below-cloud scavenging is affected by rain droplet size distribution, rain intensity and collision efficiency between particles and rain droplets.

There are a number of studies (e.g. Dingle and Hardy, 1962; Cataneo, 1973; Harju and Jatila, 1973) where the rain intensity and the rain droplet number density were simultaneously measured. Several distributions (Slinn, 1977; Mircea and Stefan, 1997) have been fitted to these measurements, the most widely used is the Marshall–Palmer (MP) distribution (Marshal and Palmer, 1948). However, rain properties can show much variation (Slinn, 1983), so the MP-distribution is valid only for average conditions. Another problem is that the MP-distribution overestimates the number of small droplets (Mircea and Stefan, 1997).

The aerosol particle—rain droplet collision efficiency is another poorly known parameter. Several attempts (Slinn, 1977; Fenton, 1980; Levine and Schwartz, 1982) to measure or estimate collision efficiencies have been made but the difference between theory and measurements is still one or two orders of magnitude in the submicron range (Volken and Schumann, 1993). The main phenomena affecting collision efficiency between

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doi:10.1016/S1352-2310(03)00326-1

falling droplets and aerosol particles are inertial impaction (Pruppacher and Klett, 1978), Brownian diffusion, phoresis caused by thermal or concentration gradients, turbulent effects and electrical forces. Inertial impaction is most important when the aerosol particles are so large that they are not able to follow the stream lines around the falling droplet. This applies to particles with diameter more than about 1 μm. For particles smaller than 1 μm Brownian diffusion is the main removal mechanism. The effect of electrical forces is often (McGann and Jennings, 1991; Byrne and Jennings, 1993; Jaworek et al., 2002) assumed to be one of the major sources of uncertainty in the calculations of scavenging coefficients.

There are some studies (Davenport and Peters, 1978; Volken and Schumann, 1993; Nicholson et al., 1991) where scavenging coefficients have already been calculated and investigated for different size ranges. To our knowledge, no analysis of atmospheric scavenging coefficients for aerosols smaller than 0.28 μm exist.

In this article we focus on below-cloud scavenging in the 10–500 nm size range. All measurements are carried out in the clean background station of Hyytiälä (Vesala et al., 1998) in Southern central Finland.

Firstly, the theory and the statistical method used in our calculations are introduced. Next the measurement devices and the criteria used in the data selection are presented. The data rejection criteria are employed because for example advection or nucleation could otherwise affect the results. Together with the criteria, the reliability and the problems of the method are discussed. The experimental scavenging coefficients have been calculated from simultaneous aerosol particle number concentrations and rain intensity measurements. A six year period is used in our analysis. Finally, a parameterization of the results for the 10–500 nm size range is shown.

2. Theory

The basic equation for the change of aerosol particle concentration $c(d_\mathrm{p})$ due to rain scavenging is given by (Seinfeld and Pandis, 1998)

$$\frac{\mathrm{d}c(d_\mathrm{p})}{\mathrm{d}t} = -\lambda c(d_\mathrm{p}), \tag{1}$$

where d_p is aerosol particle diameter and λ is the scavenging coefficient given by

$$\lambda(d_\mathrm{p}) = \int_0^\infty \frac{\pi}{4} D_\mathrm{p}^2 U_t(D_\mathrm{p}) E(D_\mathrm{p}, d_\mathrm{p}) N(D_\mathrm{p})\,\mathrm{d}D_\mathrm{p} \tag{2}$$

where D_p is the rain droplet diameter, U_t the velocity of the falling droplet, $E(D_\mathrm{p}, d_\mathrm{p})$ the collision efficiency between the falling rain droplet and aerosol particle and $N(D_\mathrm{p})$ the concentration of rain droplets as a function of droplet diameter. The integral is due to the fact that an aerosol particle of certain size d_p can be captured by rain droplets of any size. λ can be calculated theoretically if the collision efficiency and rain droplet size distribution are known. However, usually they are not.

Our approach to the problem is somewhat similar to that presented by Sperber and Hameed (1986). If Eq. (1) is integrated from t_0 to t_1 with corresponding concentrations of $c_0(d_\mathrm{p})$ and $c_1(d_\mathrm{p})$ we get

$$\lambda(d_\mathrm{p}) = -\frac{1}{t_1 - t_0} \ln\left(\frac{c_1(d_\mathrm{p})}{c_0(d_\mathrm{p})}\right). \tag{3}$$

Using measured $c_0(d_\mathrm{p})$ and $c_1(d_\mathrm{p})$ and known $t_1 - t_0$, it is possible to calculate λ. Eq. (2) can be compared with Eq. (3) when the rain scavenging is the only particle source or sink.

Instead of using only two consecutively measured particle size spectra it would be possible to use all single measurements during one rain episode and obtain scavenging coefficients by logarithmic fitting (Volken and Schumann, 1993). The reason for our choice was the limited time resolution of particle measurements. By using only two consecutive measurements it is possible to minimize changes in particle concentrations caused by factors other than rain scavenging.

If the experimental errors and different phenomena affecting the particle concentrations are taken into account, Eq. (1) takes the form

$$\begin{aligned}\frac{\mathrm{d}c}{\mathrm{d}t} = -\bar{\Lambda}c = &-\lambda c \pm \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{instr.}} \pm \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{turb.}} \\ &\pm \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{adv.}} \pm \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{cond.}} \\ &+ \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{nucl.}} \pm \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{hygr.}} \pm \left[\frac{\mathrm{d}c}{\mathrm{d}t}\right]_{\text{coag.}},\end{aligned} \tag{4}$$

where $\bar{\Lambda}$ is the experimental scavenging coefficient including all contributions. If there are no other contributions than taken into account in Eq. (2) or their average is zero, $\bar{\Lambda}$ is equal to λ. The different terms on the right-hand side in Eq. (4) are explained in more detail below.

Instrumental errors $[\mathrm{d}c/\mathrm{d}t]_{\text{instr.}}$ come from different sources. One reason is the small number of collected particles which causes statistical fluctuation in the particle concentrations. These fluctuations can be large especially when the particles are smaller than 10 nm. Also, the collection efficiency of the instrument is a function of particle concentration which may lead to systematic errors in $\bar{\Lambda}$. However, in Aitken and accumulation mode range they are much smaller than fluctuations caused by advection and turbulence.

There can also be difficulties related to rain intensity measurements, e.g. the sensitivity of the instrument for light rains. It is also difficult to say how representative the rain measurements are since the location of the rain gauge can affect strongly measured rain intensity.

Turbulence $[\mathrm{d}c/\mathrm{d}t]_{\mathrm{turb.}}$ affects measured particle concentrations because concentration is not uniform but exhibits temporal and spatial fluctuations. It also affects collection efficiency between aerosol particles and rain droplets.

Advection $[\mathrm{d}c/\mathrm{d}t]_{\mathrm{adv.}}$ is one possible cause of errors (Flossman, 1991; Volken and Schumann, 1993). When the particle concentrations are measured with e.g. 10 min resolution, air mass may change between measurements. In addition to horizontal advection, it is possible that falling rain droplets cause downward advection.

Condensation $[\mathrm{d}c/\mathrm{d}t]_{\mathrm{cond.}}$, nucleation $[\mathrm{d}c/\mathrm{d}t]_{\mathrm{nucl.}}$ and coagulation $[\mathrm{d}c/\mathrm{d}t]_{\mathrm{coag.}}$ can also change the particle size distribution and thus change the results. However, they are supposed to be quite slow during rain. The justification of this assumption is discussed in Section 4.

The situation is different for hygroscopic growth $[\mathrm{d}c/\mathrm{d}t]_{\mathrm{hygr.}}$, because smaller particles can grow to the bigger sizes. In this case the scavenging coefficients for smaller particles are increased and for larger particles decreased. In our study we have solved this problem by applying proper data selection criteria explained in Section 4.

3. Measurements

In this study, data from 6 years (1996–2001) have been used. The measurements have been carried out at SMEAR II Station (Station for Measuring Forest Ecosystem-Atmosphere Relations), Hyytiälä, Southern Finland (61°51′N, 24°17′E, 181 m asl) (Vesala et al., 1998).

3.1. Particle size spectrum measurements

Particle size distributions between 3 and 510 nm have been measured by two differential mobility particle sizers (DMPS) with time resolution of 10 min. The first device classifies particles between 3 and 10 nm and the second classifies the particles between 10 and 500 nm (Aalto et al., 2001).

For each 10 min period, the particles are classified in 29 (28 before 9 October 1998) logarithmically distributed size channels between the diameters of 3 and 510 nm. During each DMPS measurement run, each size class is measured twice and an average of these two measurements is calculated. Thus one concentration measurement of each channel takes about 20 s.

3.2. Rain measurements

Rain is measured by the tipping bucket method with an ARG100 rain gauge (ARG100 User Manual, 2001). Precipitation is collected by a funnel and is passed to one of the two buckets situated at both ends of a short balance arm. When the first bucket is full, the balance arm tips, empties the bucket and moves the second bucket under the funnel. At each tip the moving arm forces a magnet past a reed switch, causing contact to be made for a few milliseconds. The number of pulses are counted and saved with time resolution of 15 min.

4. Data selection criteria

Only the period from 1 May to 31 October during the years 1996–2001 was used. These months were chosen in view of the objective to avoid scavenging by snow and errors in rain measurements caused by frozen measurement devices.

We limited the analysis to particles larger than 10 nm because the concentration of smaller particles is normally too low and the instrumental errors too high. Another reason for the lower diameter limit was the possible growth of nanometer-sized particles. For the same reason, the cases with visible particle growth were rejected.

The analysis of the data started with selecting rain events lasting at least 0.5 h and having rain intensity more than 0.4 mm h^{-1}. Smaller rain intensities were rejected because of the possible inaccuracy of our rain gauge at low rain intensities.

The limitation in duration is due to the accuracy of rain measurement and also because the method presented here requires spatially large enough rains to be applicable. Normally the rain clouds are at a height of a few kilometers, whereas particle concentrations are measured in the boundary layer representing, say the lowest 1 km. There are several possible combinations of how rain clouds move: they can move in the same direction with boundary layer flow or they can be totally independent of it. Normally the clouds move faster than the boundary layer air.

The problem arises when the homogeneous cloud layer covers only a limited area. For reliable scavenging coefficient estimation, two consecutively measured air parcels must have been initially exposed to rain at the same moment in time (beginning of rain). If boundary layer flow is in the same direction as cloud movement, two consecutively sampled air parcels (i.e. particle concentration measurements) have been exposed for different periods to the rain since the beginning of rain, allowing to estimate scavenging coefficient. If, however, a small cloud stays over the measurement station for a longer period, say a few hours, in absence of local sources all air parcels sampled at different moments have the same concentration, and thus the method gives a scavenging coefficient of zero. In view of the large quantity of measurements the possible influence of such situations on average scavenging coefficients is minimized. Also the influence of inhomogeneity of cloud cover, leading to uncertainty and wider distribution of values, is removed by using large number of observations for estimation of average behavior of scavenging coefficients.

In order to be able to avoid changes in particle concentration due to advection in frontal zones, rain events with strongly changing meteorological parameters like temperature, pressure, wind speed and wind direction were rejected. Particle concentrations may either decrease or increase due to advection. This changes the calculated scavenging coefficients like turbulent mixing, but the median value of $\bar{\Lambda}$ stays approximately constant. The possible effect of downward advection of cleaner air caused by falling droplets could not be analyzed.

Significant changes in relative humidity was also a criterion for rejecting rain events as it may cause hygroscopic growth of particles.

There was no way to avoid turbulent fluctuations in concentration. Because each concentration measurement lasts only 20 s, turbulent fluctuations are not averaged out and lead to variations in concentration. However, particle concentrations are assumed to change symmetrically in both negative and positive directions. In this case, the mean value of the scavenging coefficient does not change, only the variation is larger.

As there are no local sources and nucleation has been observed only during sunny conditions (Mäkelä et al., 2000), nucleation does not change the situation. Nucleation was never observed during the selected rain events.

Condensational growth of particles is assumed to be much smaller than the scavenging rates during each rain episode. There are two reasons for this assumption: (1) Data used in our calculations does not include days when growth was clearly observed. (2) Observed growth episodes in Hyytiälä have been always related to photochemical reactions that do not occur during the rain.

Coagulation does not change the size distribution between two measurements because particle sources are far from the measurements site so the concentration in both "air-parcels" measured is similarly affected by coagulation and thus the effect cancels out.

By using the data selection criteria explained above, the remaining factors affecting the particle concentration are assumed to be turbulent mixing, instrumentation errors and scavenging. Instrumental errors affect measured particle concentrations which may lead to systematic or random errors, turbulent fluctuations only contribute to random errors.

One concentration measurement of each channel lasts about 20 s. This is certainly not enough to obtain good enough counting statistics nor average over a turbulent time series and results in a significant uncertainty in scavenging coefficient derived from two subsequent 10 min periods. However, our approach is based on a large amount of observations and calculation of statistically significant ensemble average values. The uncertainty related to our statistical method is illustrated in Fig. 1 where a histogram for all $\bar{\Lambda}$, regardless of particle size is shown. Mainly because of the turbulent fluctuations, these scavenging coefficients have both negative and positive values across a very large range. This also makes the use of percentiles meaningless in the subsequent analysis.

For this reason, the validity of our method was tested statistically. The error of the mean $\Delta\Lambda$ can be calculated from

$$\Delta\Lambda = \frac{\sigma}{\sqrt{N}}, \tag{5}$$

where σ is the standard deviation and N is the number of the observations. Variations for 95% confidence level for the overall scavenging coefficient, $\bar{\Lambda} - 2\Delta\Lambda$ and $\bar{\Lambda} + 2\Delta\Lambda$ were 1.69×10^{-5} and 2.05×10^{-5}, respectively. Thus, based on the large enough number of observations, average results presented here have reasonably small error intervals and differ statistically significantly from zero (corresponding to the case where particles are not scavenged by rain).

This result also applies when scavenging coefficients are calculated as a function of size for at least three or four particle size channels together rather than for only one channel. N for three channels correspond, depending on particle size, to 13 000–14 000 observations. In Fig. 2 median and mean scavenging coefficients for different rain intensity classes are shown, the latter with

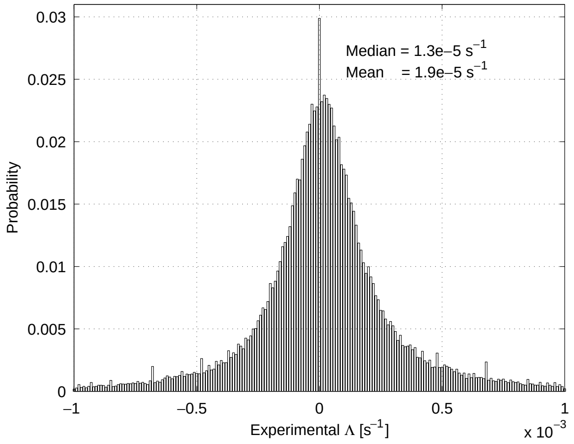

Fig. 1. All experimental scavenging coefficients, in particle size range 10–500 nm.

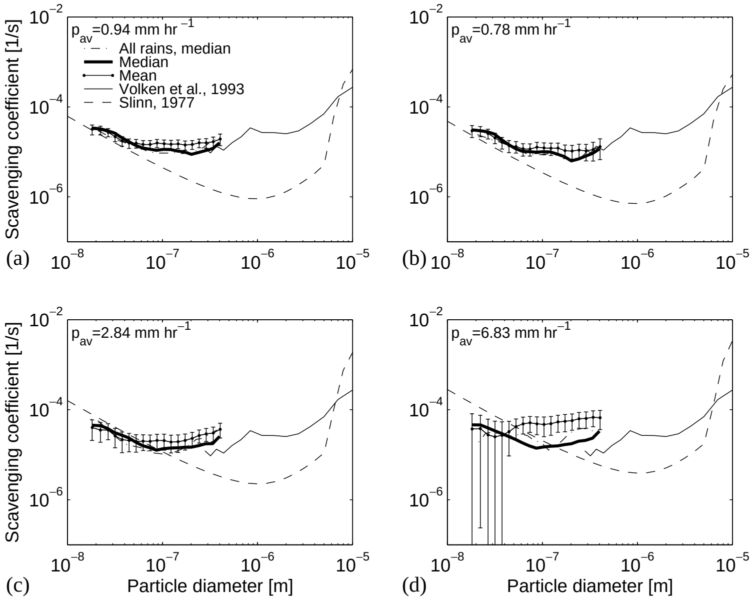

Fig. 2. Mean and median scavenging coefficient for different rain intensity classes. (a) $p_{av} = 0.94$ mm h^{-1}, (b) $p_{av} = 0.78$ mm h^{-1}, (c) $p_{av} = 2.84$ mm h^{-1}, (d) $p_{av} = 6.83$ mm h^{-1}.

error bars $\bar{\Lambda} \pm 2\Delta\Lambda$. As we can see median and average do not differ from each other systematically with lower rain intensities (Figs. 2a–c). With higher rain intensity, Fig. 2d, the agreement was worse due to the limited amount of heavy rain episodes.

To check the reliability of our results, we also used all rain events (i.e. selection criterion mentioned earlier were not applied) during the period 1996–2001 and repeated the calculation. The results were similar to those calculated using the selection criteria. This is not surprising since our carefully selected rain events are included in all rain events and also since disturbances do not affect median statistics significantly. This can also be seen from Fig. 2, where the curves representing the medians of all rains are very close to the coefficients calculated from selected rain events. The standard errors for each size bin characterize the uncertainty of mean values. Since median has less variation, it will be used further instead of mean as ensemble mean statistic for scavenging coefficients. In addition, the median is less sensitive to possible sudden changes of air masses. If for example the air mass changes during the rain, using an average or logarithmic fit on the data would lead to an erroneous result, whereas the median is much less affected.

For comparison, we have also calculated values for scavenging coefficients for the period 1.6.–30.10. in years 1997–1999 for the periods without rain. The mean and median artificial scavenging coefficients for this period were 3.512×10^{-6} s^{-1} and 2.301×10^{-6} s^{-1} respectively, which are an order of magnitude lower than the mean and median scavenging coefficients during the rain. Thus we can assume, that the combined effect of coagulation, condensation, nucleation and dry deposition is much smaller than the effect of rain scavenging during the rain.

5. Results

In total 3530 particle spectra were accepted according to selection criteria and this corresponds to approximately 588 h (or 25 days) of measurements during rain. The average durations of selected rain episodes in each month are shown in Fig. 3. For comparison, the average durations of all rain episodes during each month are also shown. In this section, "all rain measurements" refer to all rains during the chosen period whereas "selected rains" refer to the data sets selected according to the criteria presented in Section 4, from which the scavenging coefficients are calculated. The probability distributions of wind speed, rain intensity, temperature and relative humidity are shown in Fig. 4. Distributions both for selected episodes and all rain events are shown. The average wind speed was 5 m s^{-1} (Fig. 4a) and the most frequent rain intensities (Fig. 4b) were between 0 and 1 mm h^{-1}. If the selected and all rains are compared, it can be seen that both light and heavy rains are undersampled by applying the selection criteria. This is related to our data selection criteria which reject short and light rains. Light rains were rejected because of the lower limit of allowed rain intensity. A part of heavy rains were rejected because they are normally short and they are often related to fronts and rapidly changing temperatures and relative humidities. The average temperature during the selected events was 9.2° (Fig. 4c). Relative humidities for both selections were quite similar, Fig. 4d, and they were close to 100% as can be expected. Relative humidities over 100% are due to instrumental inaccuracies.

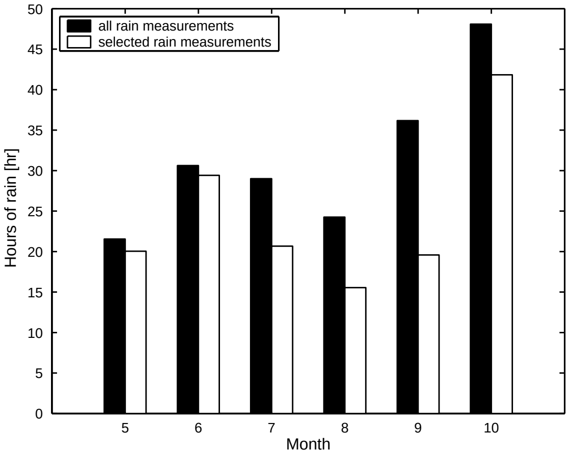

Fig. 3. Duration of all rain events and selected rain events during the summer months. Numbers are averages for years 1996–2001.

Fig. 5 shows median scavenging coefficients calculated from the measurements. Median is used instead of mean because it is less sensitive to extreme values resulting from uncertainties. Each point corresponds to a running median of three channels next to each other. Scavenging coefficients measured in a field campaign at Mt. Rigi, Switzerland (Volken and Schumann, 1993) are also shown. In those measurements the average rain intensity was about 0.8 mm h^{-1} whereas in our studies it was about 0.9 mm h^{-1}.

The thick solid line is an average of all data over all rain intensities. In the overlapping area our data and the data of Volken and Schumann (Volken and Schumann, 1993) agree quite well. In our data there is a minimum in scavenging coefficients at about particle diameter of 0.2 µm.

The rain intensities were also divided in three different intensity classes, light rain corresponding to rain intensities between 0 and 2 mm h^{-1}, moderate rain between 2 and 5 mm h^{-1} and heavy rain > 5 mm h^{-1} as indicated in Fig. 5. A clear difference between different rain intensities was found. Median rain intensities for these three groups were 0.78, 2.84 and 6.83 mm h^{-1}, respectively.

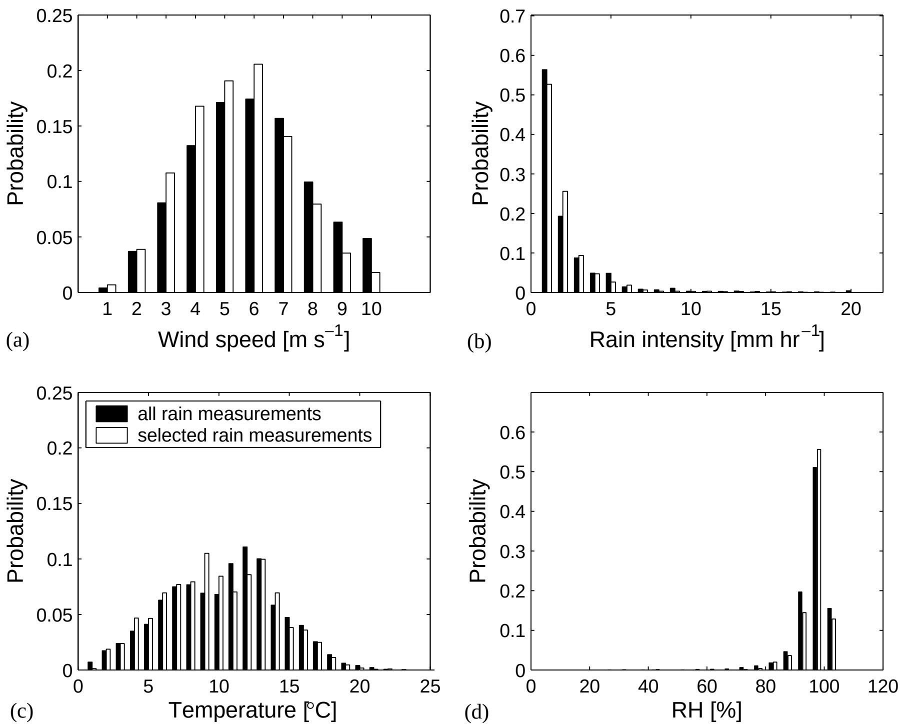

Fig. 4. Rain event properties during selected rain events (white) and all rains (black). (a) Wind speed at 50.4 m height, (b) Histogram for rain intensities, (c) Temperature, (d) Histogram about RH.

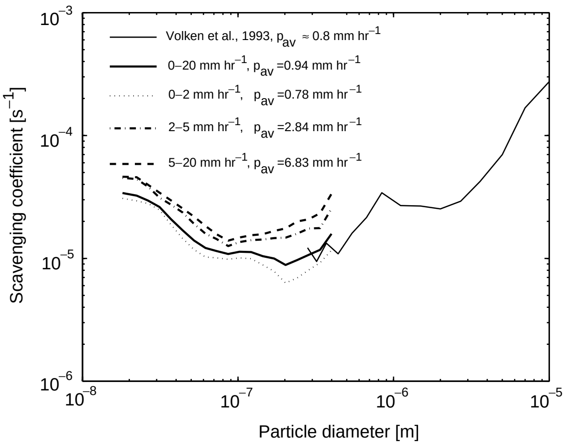

Fig. 5. Median scavenging coefficients for all data and for three different rain intensity classes.

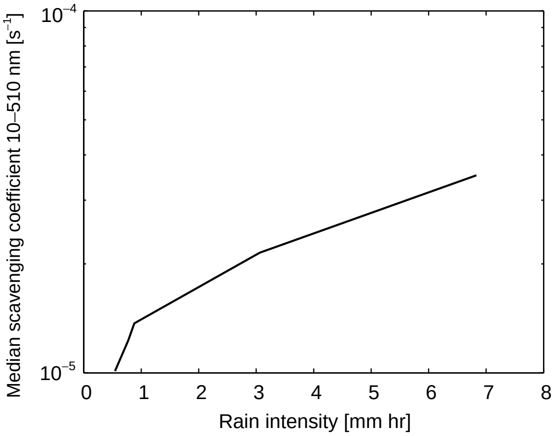

Fig. 6. Scavenging coefficients as a function of rain intensity, average over particle sizes 10–500 nm.

As theoretical analysis of the Marshall–Palmer rain droplet size distribution (Marshal and Palmer, 1948) shows that the number of rain-droplets increases with increasing rain intensity, so the results presented here agree with theory. In the accumulation size range, our results are higher than those suggested by theories presented by e.g. Slinn (1983) (see Fig. 2), Schumann (1989) and McGann and Jennings (1991). One reason for discrepancy can be the electrical interaction between charged rain droplets and aerosol particles (Jaworek et al., 2002). An extensive analysis of the possible reasons for this discrepancy is given by Volken and Schumann (1993).

In Fig. 6 the average scavenging coefficient for the range 10–500 nm is shown as a function of precipitation intensity. When the rain intensity increases from 0.5 to 9 mm h^{-1}, the corresponding scavenging coefficient changes from 1×10^{-5} to 4×10^{-5} s^{-1}.

6. Parameterization

We have also parameterized scavenging coefficients as a function of particle size and rain intensity. Firstly, scavenging coefficients as a running median of three channels, regardless of rain intensity were calculated and a function of the form

$$\log\frac{\lambda}{\lambda_0} = a + \frac{b}{\log_{10}(D_p/D_{p0})^4} + \frac{c}{\log_{10}(D_p/D_{p0})^3} + \frac{d}{\log_{10}(D_p/D_{p0})^2} + \frac{e}{\log_{10}(D_p/D_{p0})^1} \qquad (6)$$

was fitted to the data. a, b, c, d and e are fitting parameters. λ_0 was set to 1 s^{-1} and D_{p0} to 1 m. The scavenging coefficients measured by Volken and Schumann (1993) were also supposed to agree with the parameterization presented here. Because the conditions and the type of the rain may be different in their case, we do not recommend the use of our parameterization out of our measurement range. However, we wanted our parameterization behave physically also outside our range because of all-too-common extrapolation of the different kind of parameterizations.

The next step was to find any functional dependence on rain intensity. For this purpose, scavenging coefficients were calculated as a functions of particle diameter for four different rain intensity ranges having median rain intensity of 0.78, 0.94, 2.84 and 6.83 mm h^{-1}. These data sets were used to fit the dependence of parameterized scavenging coefficient on rain intensity. Since Fig. 6 suggests that the dependence should be

$$\log\frac{\lambda}{\lambda_0} \sim \sqrt{\frac{p}{p_0}},$$

where p is rain intensity and p_0 is 1 mm h^{-1}, we added a term

$$f\left(\frac{p}{p_0}\right)^{0.5}, \qquad (7)$$

where f is the fitted parameter, to Eq. (6). The combination of these two equations forms our parameterization. In Fig. 7 the data used in parameterization is shown with corresponding parameterization.

The parameterization is valid for particles having diameters between 10 and 500 nm and for rain intensities 0–20 mm h^{-1}. Error in parameterization compared to our data is about ±6%. When the particles have a diameter less than about 10 nm, our parameterization probably overestimates the scavenging coefficients.

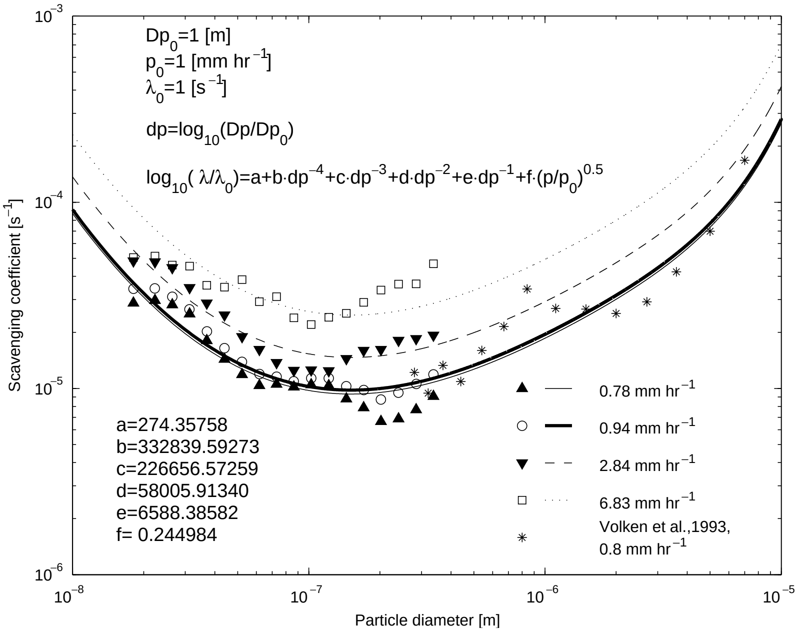

Fig. 7. Measurements and parameterization of scavenging coefficients as a function of particle size and rain intensity.

7. Conclusions

In this paper the below-cloud scavenging coefficients were calculated for aerosol particles having diameters between 10 and 510 nm. The statistical calculations were based on 6 years of ambient measurements at a boreal forest site in central Finland.

Median scavenging coefficients varied between 7×10^{-6} and 4×10^{-5} s^{-1}. Also a dependence of scavenging coefficients on rain intensity was found. In addition to calculated scavenging coefficients, the scavenging coefficients were also parameterized as a function of particle size and rain intensity for particle sizes 10–500 nm and 0–20 mm h^{-1}, respectively.

Acknowledgements

The authors want to thank Ph.D. Kirsti Jylhä for useful comments and discussions and Pasi P. Aalto for carrying out the DMPS-measurements. Robert Flanagan is acknowledged for improving the language of the article. The authors want also to thank the referees for their critical comments which improved the manuscript notably.

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