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Snow scavenging of ultrafine particles: field measurements and parameterization

Ella-Maria Kyrö[1)], Tiia Grönholm[1)], Henri Vuollekoski[1)], Aki Virkkula[1)], Markku Kulmala[1)] and Lauri Laakso[2)]

[1)] *Department of Physics, P.O. Box 64, FI-00014 University of Helsinki, Finland*
[2)] *School of Physical and Chemical Sciences, North-West University, Private Bag x6001, Potchefstroom 2520, Republic of South Africa*

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Snow scavenging of sub-micrometre aerosol particles is poorly known, even though it is an important aerosol removal mechanism in the polar and mountainous regions as well as in the upper troposphere and in mid-latitudes in winter. In our study, snow scavenging coefficients were calculated using four years of particle number concentration spectra measurements together with meteorological measurements. The obtained experimental median scavenging coefficients varied between 8.7×10^{-6} s^{-1} and 5.2×10^{-5} s^{-1} in the 10 nm to 1 μm size range. A parameterization of the results is presented and its functionality is tested using University of Helsinki Multicomponent Aerosol model (UHMA). The parameterization applies to snowfall types of slight continuous snowfall and snow grains with intensities of the order of 0.1 mm h^{-1}.

Introduction

The pollution transport from mid-latitudes is responsible for a significant fraction of the total air pollution in the polar regions (AMAP 2006). The transport pathways are controlled by both synoptic scale processes and large-scale meteorological phenomena such as NAO (North-Atlantic Oscillation) and in general, equator-to-pole transportation circulation. Since precipitation in the Arctic and Antarctica, and upper troposphere is mostly solid especially during wintertime, snow is an important scavenger in the atmosphere. Snow scavenging of aerosols is also important in the mountain regions surrounded by pollution sources, for instance the Alps, the Himalayas, and the Andes (e.g. Shrestha *et al.* 1997, Carrera *et al.* 2001).

Although there is some information on aerosol particle scavenging by falling raindrops (e.g. Nicholson *et al.* 1991, Volken and Schumann 1993, Laakso *et al.* 2003) scavenging by falling snowflakes and ice crystals is even less known both experimentally and theoretically. Due to the lack of suitable experiments only few field studies on scavenging by snow crystals have been reported. Jylhä (2000) studied scavenging of pollutants originating from a Finnish coal-fired power station unit during a wintertime precipitation event. In her study, the precipitation scavenging coefficient was estimated to be of the order of 10^{-6} s^{-1} or less. Some estimations of scavenging efficiency of snow based on observed differences in aerosol concentrations during snowfall have been made (Itagaki and Koenuma 1962, Reiter 1964, Carnuth 1967, Reiter and Carnuth 1969, Magono *et al.* 1974, 1975, Graedel and Franey 1975) but unfortunately all of these studies have not taken into account either the aerosol concentration differences in different air masses nor the changes in phoretic forces which may vary significantly with varying relative humidity. If these error sources are not taken into account, it is impossible to differentiate the effect of snow scavenging or change of air mass or relative humidity. Nevertheless, these studies show that snow scavenging is an important removal mechanism for aerosol particles and snow can scavenge aerosol particles up to 50 times more efficiently than rain, when based on equal equivalent water content of the precipitation.

Starr and Mason (1966) used tissue paper to simulate ice crystals. Hence, only the effects of inertial impaction and Brownian motion were taken into account. Another laboratory study was made by Knutson *et al.* (1976) who made experiments with natural snow crystals and aerosol particles of diameters 0.5–7 μm. Sauter and Wang (1988) made experiments with artificial aerosol particles of mean radius 0.75 μm and natural snow crystals of different shapes. They found that the collection efficiency of aerosol particles by snow crystals decreases with increasing crystal size.

More theoretical than experimental studies on snow scavenging exist (*see* e.g. Pruppacher and Klett 1997). Martin et *al.* (1980) determined collection efficiencies for ice crystal plates. The aerosol size range in their study was from 1 nm to 10 μm. They used theoretical models that took into account Brownian diffusion, thermophoresis, diffusiophoresis and inertial impaction and showed that the collision efficiency of aerosol particles exhibit a strong minimum around 100 nm and the difference increases with increasing relative humidity. Miller and Wang (1988) did calculations for columnar ice crystals and found similar results. Miller (1990) presented a theoretical model for the determination of scavenging of submicron aerosols by snow crystals. He found that of the parameters in this model, deposition was the most sensitive to variations of relative humidity. In addition, couple of attempts to implement the snow scavenging on global models exist (Stier *et al.* 2005, Tost *et al.* 2006). In these studies the below-cloud scavenging coefficient has been scaled by the snow flux.

The particle scavenging in the boundary layer depends on aerosol size, rainfall intensity, mixing processes between boundary layer and cloud elements, in-cloud scavenged fraction, in-cloud collection efficiency and in-cloud coagulation with cloud droplets (Andronache *et al.* 2006). The same study by Andronache *et al.* (2006) shows that the scavenging coefficient is very sensitive to parameters related to mixing and cloud microphysics.

This paper will focus on impaction snow scavenging in the 10 nm–1 μm size range. The measurements are carried out in the clean background station SMEAR II in Hyytiälä (Hari and Kulmala 2005) in southcentral Finland. Laakso *et al.* (2003) made similar calculations for water scavenging coefficient in Hyytiälä. Our results will be compared with theirs.

Theory

The basic equation for the aerosol concentration (c) change due to precipitation (liquid or solid) is

$$\frac{dc(d_\mathrm{p})}{dt} = -\lambda c(d_\mathrm{p}), \qquad (1)$$

where d_p is the diameter of the particle and λ is the scavenging coefficient (Seinfeld and Pandis 2006). Since aerosol particles can be scavenged by rain droplets of any size, λ can be expressed as

$$\lambda(d_\mathrm{p}) = \int_0^\infty \frac{\pi}{4} D_\mathrm{p}^2 U_\mathrm{t}(D_\mathrm{p}) E(D_\mathrm{p}, d_\mathrm{p}) N(D_\mathrm{p}) dD_\mathrm{p}, \qquad (2)$$

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 drops as a function of droplet diameter (Seinfeld and Pandis 2006). For snow crystals, D_p is often replaced with the capacitance of the crystal — a product of surface area factor and equivalent radius — since the diameter can be somewhat difficult to define (Miller 1990, Pruppacher and Klett 1997). However, in our study neither the crystal size nor the capacitance is known.

Several models on snow scavenging exist, of which the most important are trajectory and flux models (Pruppacher and Klett 1997). Using trajectory models it has been shown that the aerosol particles are scavenged at the rim of ice crystals (Pruppacher and Klett 1997), which is due to the strong horizontal flow component underneath a falling ice crystal.

The collection efficiency is largest for ice crystals with diameter around 1 mm (Martin *et al*. 1980). For 6–30 mm sized snowflakes the collection efficiency is greater than for ice crystals of the same size. This is because of the particle filtering effect of the holes in the snowflakes; hence the collection efficiency is dependent on the flow through the aggregates rather than on the flow around the snowflake (Mitra *et al*. 1990).

In this paper, the snow scavenging coefficients are calculated using a semi-empirical approach similar to the one described in Mircea and Stefan (1997) and Laakso *et al*. (2003). The snow scavenging coefficient λ_s can be calculated by integrating Eq. 1 from time t_1 to time t_2 with corresponding particle concentrations of $c_1(d_p)$ and $c_2(d_p)$:

$$\lambda_s\left(d_p\right) = -\frac{1}{t_1 - t_0}\ln\left[\frac{c_1\left(d_p\right)}{c_0\left(d_p\right)}\right]. \tag{3}$$

Equation 3 is valid only when the snow scavenging is the only factor contributing to the particle concentration change. However, using only two consecutive measurements it is possible to minimize the changes in particle concentrations caused by other reasons. When taking all these error sources into account, Eq. 1 can be written as

$$\begin{aligned}\frac{dc}{dt} = -\Lambda_s c = -\lambda_s c &\pm \left(\frac{dc}{dt}\right)_{\text{instr}} \pm \left(\frac{dc}{dt}\right)_{\text{turb}} \\ &\pm \left(\frac{dc}{dt}\right)_{\text{adv}} \pm \left(\frac{dc}{dt}\right)_{\text{cond}} \pm \left(\frac{dc}{dt}\right)_{\text{nucl}} \\ &\pm \left(\frac{dc}{dt}\right)_{\text{coag}} \pm \left(\frac{dc}{dt}\right)_{\text{hygr}}\end{aligned}, \tag{4}$$

where Λ_s is the experimental snow scavenging coefficient. The sources changing the particle concentration, other than snow scavenging, $-\lambda_s c$, are instrumental errors (instr), turbulence (turb), advection (adv), condensation (cond), nucleation (nucl), coagulation (coag) and hygroscopic growth (hygr), respectively. In case these terms are zero or their average is zero, Eq. 4 reduces to Eq. 1, where $\lambda = \lambda_s$.

Relative instrumental errors usually increase with decreasing particle number concentration. Also low concentrations increase the statistical fluctuations. However, instrumental fluctuations are significantly smaller than those caused by advection and turbulence for the particle size range considered in this paper. Yet, they do affect the experimental snow scavenging coefficient if the particle concentration is very low, of the order of tens of particles per cubic centimetre. The data used in our study is selected so that all the other error sources in Eq. 1 can be neglected. The data selection criteria used will be discussed below.

Turbulence causes both temporal and spatial fluctuations in particle concentrations. This may affect the collection efficiencies between ice crystals and aerosol particles. In addition to turbulence caused by thermal differences and surface roughness, also falling snowflakes and ice crystals generate turbulence. Turbulence is rather difficult to remove from the data by applying any kind of selection criteria.

Since the time resolution of the measurements from which the Λ_s is calculated is ten minutes, it is possible that the air mass changes between measurements and thus, advection causes error in the result. In addition, falling snowflakes may cause downward advection.

Aerosol dynamical processes, such as condensation, nucleation and coagulation, can change the aerosol spectra and number concentration. However, nucleation has not been observed during snowfall. In addition, condensation and coagulation are supposed to be rather slow during precipitation. The validation of this statement is discussed further in the section "Data selection criteria".

Hygroscopic growth of aerosol particles changes the particle size spectrum so that the number of small particles decreases and large particles increases. Hence, this phenomenon causes an artificial increase of scavenging coefficient for small particles and decrease for large particles. However, in our study we used reasonable data selection criteria that reduce the significance of this problem.

Calculation of Λ_s from Eq. 3 by comparing two consecutive size distributions should in principle minimize the effects of the various error sources discussed above. However, even during snowfall the concentration does not decrease monotonically which leads to both positive and negative Λ_s values, as will be shown in the discussion of the data. Therefore, also another method was used: Λ_s was calculated by linear fitting to

$$\ln\left[\frac{c\left(t,d_p\right)}{c\left(0,d_p\right)}\right]=\Lambda_s\left(d_p\right)t \tag{5}$$

over selected snowfall periods for each d_p so that for each snowfall episode one $\Lambda_s(d_p)$ was obtained. This method reduces the effects of instrumental noise and real atmospheric fluctuations. The underlying assumption behind the linear fit method is that the particle concentration decreases only due to snow scavenging. Ideally, both the linear fit to Eq. 5 and the average of several consecutive $\Lambda_s(d_p)$'s from Eq. 3 yield equal values.

When measuring aerosols in the boundary layer, the changes in concentration depend on both below-cloud scavenging and in-cloud entrainment (Andronache *et al*. 2006). Both the averaging of several Λ_s's from Eq. 3 and the linear fit Eq. 5 yield an effective scavenging coefficient that does not differentiate between in-cloud and below-cloud scavenging. Yet, during wintertime the stratification of the boundary layer is stable and there is hardly any convection inside the boundary layer, and the particle mixing with cloud is negligible. Thus, it can be assumed that the decrease in the particle concentration depends on below-cloud scavenging only.

Instrumentation

The experimental snow scavenging coefficients presented in this paper were calculated using DMPS (Differential Mobility Particle Sizer, Aalto *et al*. 2001) and Vaisala FD12P automatic weather sensor (FD12P Manual 2002) data collected at the SMEAR II station (Station for Measuring Ecosystem–Atmosphere Relations) (Hari and Kulmala 2005) in Hyytiälä, in southern Finland (61°51´N, 24°17´E).

The Vaisala FD12P automatic weather sensor measures the amount and intensity of precipitation, visibility up to 50 kilometres, weather type and temperature (FD12P Manual 2002). With an optical forward scattering sensor it can distinguish between fog and different types of precipitation. The capacitive precipitation sensor measures the amount of precipitation. The weather type and visibility are calculated using all measured parameters. The device interprets the measured signals into general weather codes used by WMO (World Meteorological Organization) and NWS (National Weather Service). In comparison with another similar automatic weather sensor, FD12P has demonstrated better performance and reliability (Van der Meulen 1994).

The DMPS system measures particle size spectra with a time resolution of 10 minutes (Aalto *et al*. 2001). The measured particle spectrum ranges from 3 nm to 1 μm. The device classifies the particles in 38 logarithmically distributed size bins. The system consists of a neutralizer, two Hauke-type DMAs (Differential Mobility Analyzer) and two CPCs (Condensation Particle Counter) (Aalto 2004). The first DMA classifies particles between 3 nm and 50 nm and the second classifies particles between 15 nm and 1 μm. The sample air is dried before the classification.

Data selection criteria

For calculations of snow scavenging coefficient, only snowfalls with very little varying variables were selected. Also, all snowfalls occurring when the ambient temperature was above 0 °C were neglected. To obtain all possible changes in snowfall and present weather the data of FD12P with time resolution of one minute was used. The minimum length of selected snowfalls was two hours.

Only particle size data over 10 nm was used, since the concentration of particles below 10 nm during snowfall is usually very low. All cases with visible particle growth were discarded as well as cases with high concentration peaks in random size bins or other measurement errors.

To avoid particle concentration changes caused by phoretic forces the temperature and relative humidity were allowed to change ±3 °C and ±5%, respectively, during the snowfall. In addition to phoretic forces, significant changes in relative humidity may cause hygroscopic growth of aerosol particles. Other limitations in changes of meteorological variables were ±50° for the wind direction, ±3 m s^{-1} for the wind speed and ±5 hPa for pressure.

Particle concentration may vary by orders of magnitude in different air masses. Therefore snowfall episodes occurring when air mass was changing were neglected. The rejection was done using DMPS data and synoptic weather maps made by the German weather centre, Wetterzentrale.

We rejected all data with clear growth of particles. Thus the rate of apparent removal of particles from any selected size bin due to condensational growth to larger sizes can be assumed to be much lower than the rate of true particle removal by snow scavenging. This assumption was tested in a modelling study presented later in this paper. In addition, particle growth in Hyytiälä is observed to be related to photochemical reactions (Kulmala *et al*. 1998), which do not occur during snowfall.

Coagulation can also be neglected, since the particle sources are far away from the measurement site (Laakso *et al*. 2003), for air parcels of two consecutive measurements are equally affected by coagulation.

According to Eq. 4, the remaining factors that may change the particle concentration are instrumental errors, turbulence and snow scavenging. Instrumental errors may lead to both systematic and random errors in calculated scavenging coefficients, turbulence causes only random error. The flow field around falling snow crystals can be very turbulent due to the rapid difference in the flow direction around the crystal. However, for the particle size range studied, turbulence and instrumental errors are supposed to be much smaller than snow scavenging.

Results

Data statistics

Our selected data consisted of approximately 160 hours of snowfall, which corresponds to approximately 960 particle size spectra. The average air temperature during the snowfalls was –6 °C, the minimum temperature was –20 °C and maximum was 0 °C. The average humidity for the selected snowfalls was 95%, average wind speed 6 m s^{-1} and the prevailing wind direction east.

Nearly half of the selected snowfalls occurred in 2007 (Fig. 1). The distribution of selected snowfalls was rather similar to the distribution of all occurred snowfalls. The selected snowfalls represented a slightly bigger fraction of total snowfalls in November. This could have been due to the more southern route of the midlatitude synoptic scale weather disturbances and thus larger amount of occlusion fronts of the total snowfalls. The air mass does not change in occlusion fronts and therefore snowfalls taking place in them could have been included into our analysis.

Most of the snowfalls had low intensities (median 0.2 mm h^{-1}), which was expected, since most represented snowfall type was slight continuous rainfall (Table 1). The intensities ranged from 0 to 0.8 mm h^{-1}. It was not possible, however, to calculate the intensity-dependence of the scavenging coefficient due to the small amount of data.

The particles decreased clearly during an example snowfall event on 21 January 2007 between 02:00 and 12:00 (+2 UTC) at the SMEAR II station (Fig. 2A). Also, the size segregated mean snow scavenging coefficients calculated from the shown spectra showed this, with the highest values reaching 3×10^{-5} s^{-1} (Fig. 2B). Also some negative values were observed, both for smallest and largest particles. This was due to the high relative instrumental errors for small particle concentrations. Note that in this case, the Greenfield gap (explained in the next subsection) in the scavenging coefficients could not be seen since Λ_s is calculated only from three particle spectra.

All the meteorological variables were very stable during the example snowfall event and a clear decrease in the total and 150 nm sized particle concentration was seen (Fig. 3). For the 50 nm particles the decrease in concentration was not as clear. The total particle concentration decreased from around 1500 cm^{-3} to around 1000 cm^{-3}.

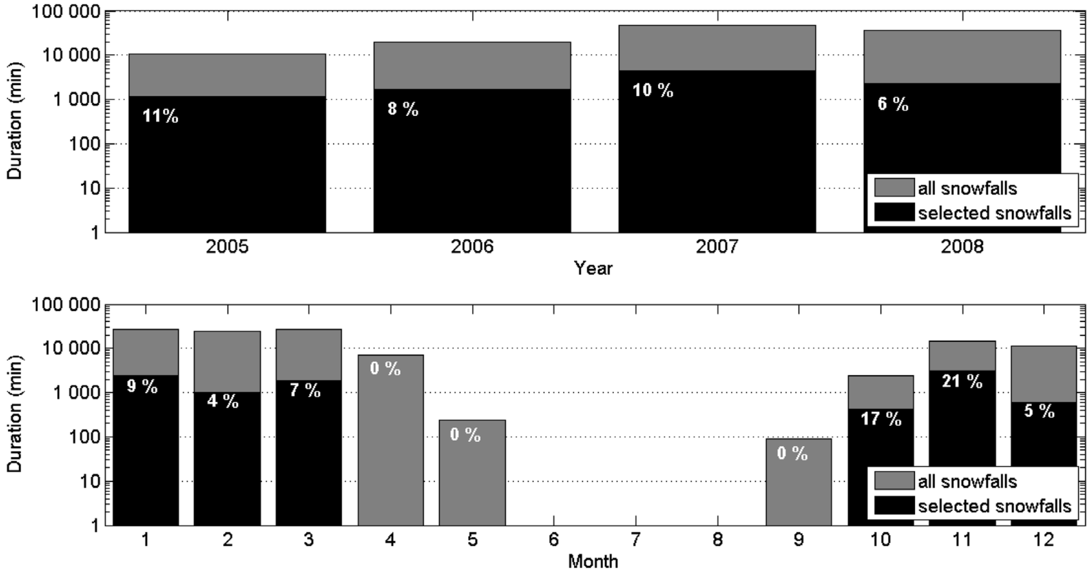

Fig. 1. Yearly and monthly distribution of the selected snowfalls (black bars) and all occurred snowfalls (grey bars). The percent of selected snowfalls *vs.* all snowfalls is also shown.

The distribution of calculated scavenging coefficients was wide as we expected (Fig. 4A) based on the statistical nature of our study. Also the distribution of the scavenging coefficients calculated using the linear fit was wide (Fig. 4B), but an order of magnitude narrower than using the method described by Laakso *et al*. (2003). However, in order to represent particle decrease by snowfall, the distribution should differ from a zero-centred Gaussian curve. Both distributions were plotted against a height adjusted Gaussian curve with $\mu = 0$ and $\sigma = 3 \times 10^{-5}$. The histogram in Fig. 4B differed from the above Gaussian curve at 88% significance level, hence one can say that the measured scavenging coefficients represent the snow scavenging process itself, not the noise. The median scavenging coefficient for all data using both methods was 1.8×10^{-5} s^{-1}. This was about an order of magnitude smaller than observed by Miller (1990) and Graedel and Franey (1975) but very close to the water scavenging coefficients calculated by Laakso *et al*. (2003) (Table 2).

Table 1. Type, duration and number of selected snowfalls. The most represented snowfall types were slight continuous snowfall and snow grains.

Type of snowfall	Duration (min)	Number
Snow (slight, continuous)	8357	156
Snow (moderate, intermitted)	9	3
Snow (heavy, intermitted)	45	12
Ice precipitation	19	5
Snow grains	806	134
Snow crystals	157	24

Parameterization

The snow scavenging coefficients (Λ_s) and their medians and means were calculated as a function of particle size (Fig. 5). The median scavenging coefficients varied between 8.7×10^{-6} s^{-1} and 5.2×10^{-5} s^{-1}. Also the mean error,

$$\Delta\Lambda = \frac{\sigma}{\sqrt{N}} \quad (6)$$

is shown in Fig. 5. The $\Lambda_s(D_p)$ exhibited a minimum for around 200 nm sized particles, as expected.

Theoretical studies have shown that the snow scavenging coefficient as a function of aerosol particle size exhibit a strong minimum for particles around 100 nm–1 μm in diameter (Martin *et al*. 1980, Miller and Wang 1988). This minimum is often referred to as “Greenfield gap”. The gap

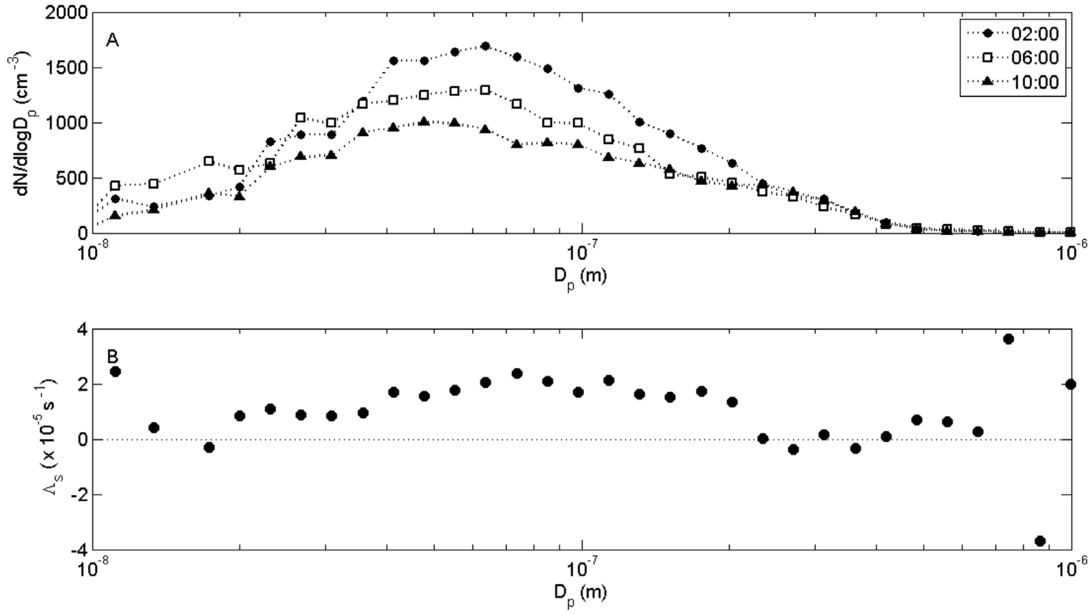

Fig. 2. (**A**) Particle size spectra during a snowfall event occurred on 21 January 2007 between 02:00 and 12:00 (+2 UTC) at the SMEAR II station, and (**B**) mean snow scavenging coefficient Λ_s calculated from them.

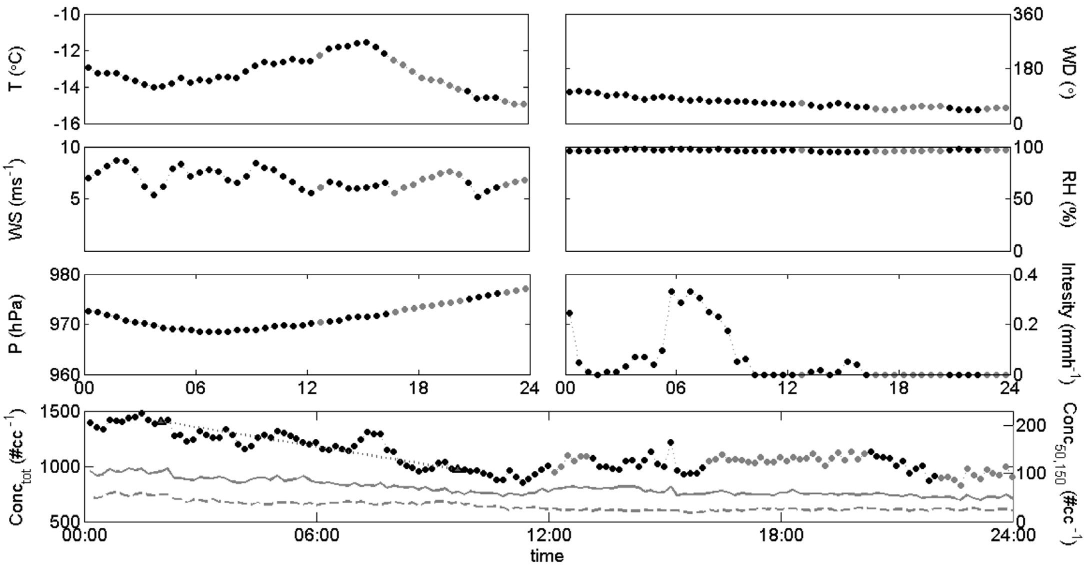

Fig. 3. Meteorological variables during 21 January 2007. Time of snowfall is indicated with black circles, the rest of the data is marked with grey circles. In addition, the total particle concentration and concentrations for 50 (dashed grey line) and 150 nm (solid grey line) sized particles are shown. For the time period from 02:00 to 10:00 am a linear fit to the total concentration is shown (solid black line).

is a result of Brownian diffusion dominating particle capture for smaller particles and inertial impaction dominating capture for bigger particles and it is the stronger the higher the relative humidity is (Martin *et al*. 1980). The gap was first noticed by Greenfield (1957), who took inertial impaction, Brownian diffusion and turbulent shear into account in his calculations.

We tested thousands of functions with our dataset; of them Pearson IV (*see* Appendix) gave

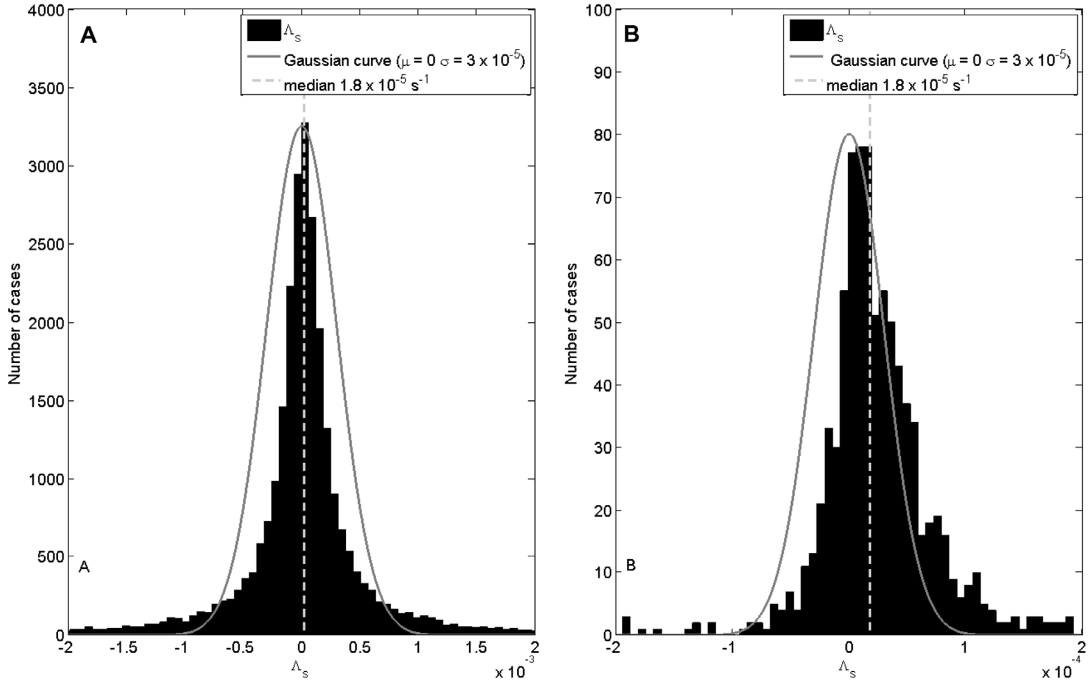

Fig. 4. All measured snow scavenging coefficients calculated using the method described by (**A**) Laakso *et al.* (2003), and (**B**) linear fitting. In addition, the Gaussian curve with μ = 0 and $\sigma = 3 \times 10^{-5}$ is shown in both panels. The median of the data (dashed grey line) was 1.8×10^{-5} s^{-1} using both methods.

the best, physically reasonable fit with the R^2 value and fit standard error being 0.73 and 0.12, respectively. Yet the function is long and complicated to use in modelling studies. In order to get rid of this problem, we tried several simple functions to parameterize the results. However, none of them was significantly better than the other, so we decided to use the same equation as Laakso *et al*. (2003) used for water scavenging,

$$\log_{10}\left(\frac{\Lambda}{\Lambda_0}\right) = a + bd_p^{-4} + cd_p^{-3} + dd_p^{-2} + ed_p^{-1} + f\left(\frac{p}{p_0}\right)^{0.5}, \quad (7)$$

but with the difference that the coefficients *b*, *c* and *f* were chosen to be zero:

$$\log_{10}\left(\frac{\Lambda}{\Lambda_0}\right) = a + dd_p^{-2} + ed_p^{-1}. \quad (8)$$

The coefficients *a*, *d* and *e* were fitted and their values were 22.7, 1321 and 381, respectively (Fig. 5). The fitting was done for size segregated median scavenging coefficients using least squares method with Gaussian elimination. The fit standard error was 0.15 and R^2 value 0.49.

Median snow scavenging coefficients were slightly higher than those of water scavenging. Compared with the water scavenging parameterization, snow scavenging parameterization gave similar values to water scavenging during

Table 2. Λ values.

Study	Λ_s (s^{-1})	Size range (μm)
This work	8.7×10^{-6} s^{-1}–5.2×10^{-5} s^{-1}	0.01–1
Graedel and Franey (1975)	4.82×10^{-4}–6.35×10^{-3}	0.29–1.48
Jylhä (1990)	~10^{-6}	(Scavenging of sulphur)
Miller (1990)	1.05×10^{-4}	0.03
Laakso *et al.* (2003)	7×10^{-6} s^{-1}–4×10^{-5} s^{-1}	0.01–0.51 (water scavenging)

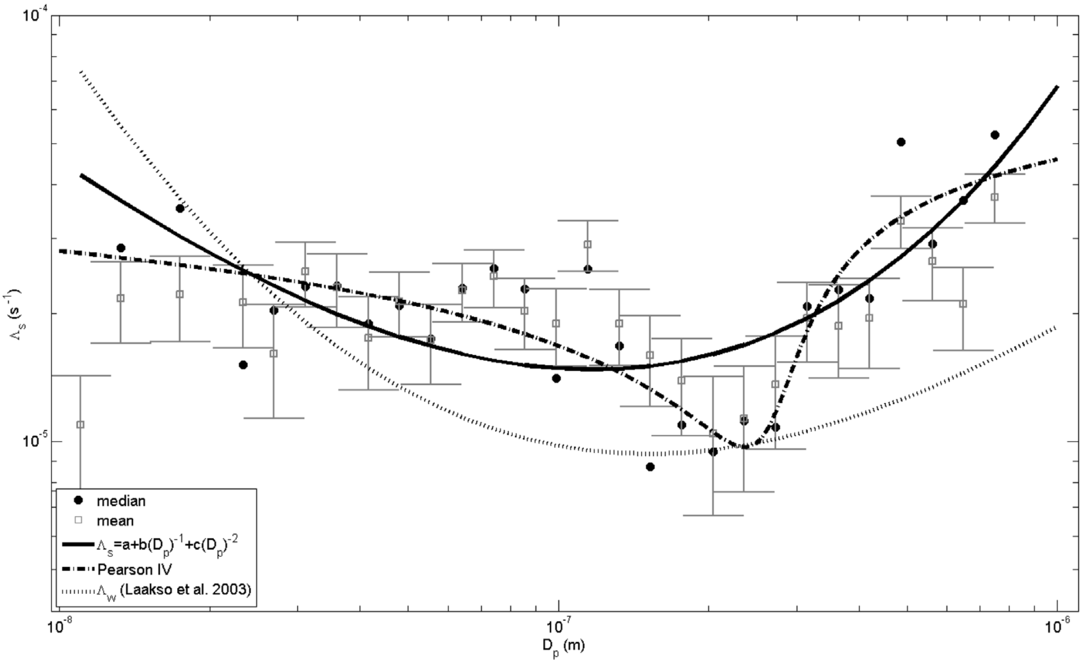

Fig. 5. Mean snow scavenging coefficients with their mean error and median snow scavenging coefficients. Two parameterizations for median Λ_s are also shown. In addition, the parameterization by Laakso *et al.* (2003) for water scavenging is shown.

rain intensities around 3 mm h^{-1}, although snowfalls were dominated by intensities below 0.2 mm h^{-1}. It is expected, that snowfalls with higher intensities scavenge the air more efficiently. This is assumption was affirmed by a case study of a snowfall that occurred in Hyytiälä between 16:00 and 22:00 on 23 November 2008 and had intensities higher than 5 mm h^{-1}. The median and mean Λ_s for the snowfall were 8×10^{-5} s^{-1} and 2×10^{-4} s^{-1}, respectively.

Case studies with the UHMA model

In order to test the obtained parameterization, we performed a set of simulations using UHMA (University of Helsinki Multicomponent Aerosol model, Korhonen *et al*. 2004), which is a size segregated aerosol dynamics box-model that was developed for studies of a multi-component tropospheric aerosol particle population. The original model has all basic aerosol dynamical mechanisms for clear sky conditions implemented: nucleation, condensation, coagulation and dry deposition.

In this study, no new particle formation was assumed and hence nucleation could be neglected. A new routine was written to include snow scavenging. Evolution of the particle number distribution *N* was then solved by using the 4th order Runge-Kutta method from the general dynamic equation (e.g. Seinfeld and Pandis 2006) with a five-second time step. The particle size distribution was represented using the hybrid sectional method (Jacobson and Turco 1995) with 38 sections distributed uniformly in logarithmic size coordinate over the (spherical) particle diameter range from 3 to 1000 nm. As input for the initial particle distribution in the model, we used data from DMPS measurements. Ambient conditions and vapour concentrations of sulphuric acid (10^4–10^5 cm^{-3}) and a condensing generic organic one (10^5–10^6 cm^{-3}) were chosen to represent winter time.

We compared four case studies of experimental snow scavenging observations against the evolution of modelled particle distribution in the UHMA model. According to our analysis, the model was capable of predicting the decrease in size dependent particle concentration

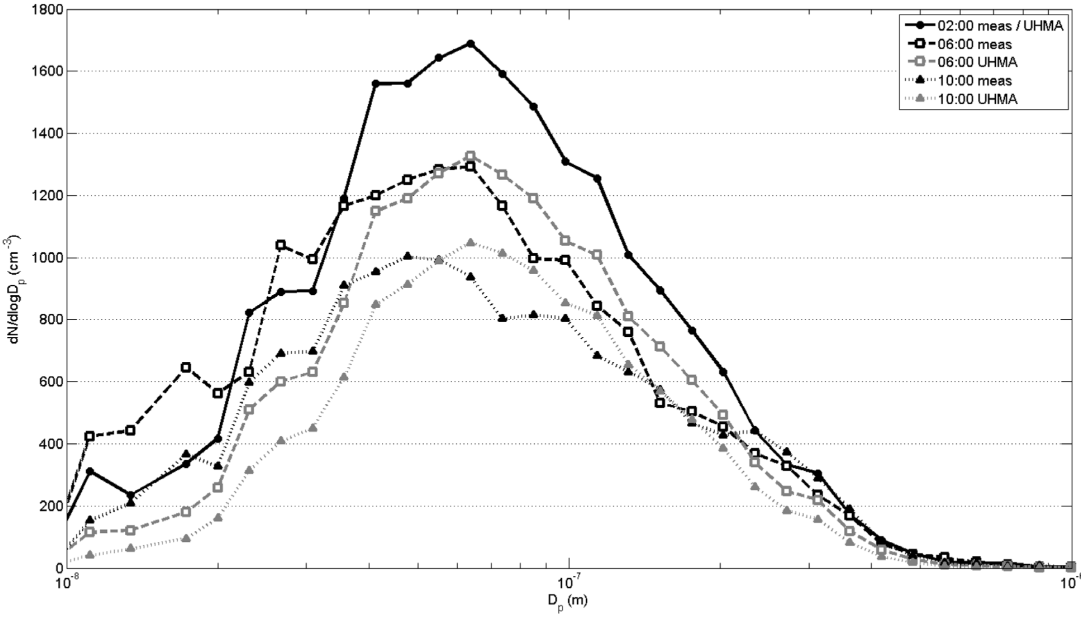

Fig. 6. Measured particle size spectra on 21 January 2007 between 02:00 and 10:00 (+2 UTC) (black lines) and the spectra modelled using University of Helsinki Multicomponent Aerosol Model UHMA (grey lines).

surprisingly well throughout the studied diameter range (Fig. 6). Because the size of particles affects their probability of being scavenged, particle growth during the scavenging episode is an important factor. Indeed, our modelling studies showed that by increasing the concentrations of condensing vapours, the resulting growth in particle mode diameter results in increased scavenging probability.

We also conducted a modelling study of the relative effect of the aerosol dynamical processes by neglecting all processes besides snow scavenging. Apart from the apparent condensational growth observed in some cases, the evolution of particle distribution did not differ much from the more complete studies. This result demonstrates the great relative importance of the snow scavenging process.

Conclusions

In our study, snow scavenging coefficients were calculated for 10 nm–1 μm sized aerosol particles from 4 years of measurements. The median snow scavenging coefficient for Hyytiälä was found to be 1.8×10^{-5} s^{-1} and it varied between 8.7×10^{-6} s^{-1} and 5.2×10^{-5} s^{-1}. The obtained snow scavenging coefficients were not significantly different from the coefficients for rainfall. However, most of the chosen snowfalls had very low intensities (median of 0.2 mm h^{-1}), whereas the parameterization for rainfall is for 3 mm h^{-1}.

The parameterization of snow scavenging coefficient applies to snowfall types of slight continuous snowfall (WMO code 71) and snow grains (WMO code 77). Even though it is clear that Λ_s depends on snowfall rate, we could not determine this dependency because in our dataset snowfalls were practically always similar with 95% of the intensities between 0 and 0.8 mm h^{-1}. Hence, new measurements are needed at locations where the snowfall rates are orders of magnitude higher, and preferably at a location where also pollution concentrations are high so that clear parameterizations can be obtained.

A modelling study was conducted, and it agreed very well with the experimental observations. Analysis showed the importance of particle diameter to its probability of being scavenged. The relative importance of the snow scavenging process was also proven to be great by a comparative study. Therefore, it is suggested to use our parameterization in the future modelling studies.

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Appendix. xFortran code for our Pearson IV fit.

```
!------------------------------------------------------------
REAL*8 FUNCTION pearson_fit(x)
!------------------------------------------------------------
!
! X=
! Y=
! Eqn# Pearson IV(a,b,c,d,e,f)
! r2=0.7292350327047953D0
! r2adj=0.6480055425162339D0
! StdErr=0.1158375634021132D0
! Fval=11.3116078788025D0
! a= -3.656286475684101D0
! b= -1.357231572081016D0
! c= -6.626850958802669D0
! d= 0.07659953772639075D0
! e= 0.101596949507848D0
! f= 0.1555922386809117D0
! Constraints: d>0,e>0
!------------------------------------------------------------
REAL*8 x,y
REAL*8 n
n=(x-0.07659953772639075D0*0.1555922386809117D0/(2.0*&
&0.1015969495078480D0)-(-&
&6.626850958802669D0))/0.07659953772639075D0
y=(-3.656286475684101D0)+(-1.357231572081016D0)*(1.0+&
&n*n)**(-0.1015969495078480D0)*DEXP(-&
&0.1555922386809117D0*(DATAN(n)+&
&DATAN(0.1555922386809117D0/(2.0*0.1015969495078480D0))))/(1.0+&
&0.1555922386809117D0*0.1555922386809117D0/(4.0*&
&0.1015969495078480D0*0.1015969495078480D0))*&
&*(-0.1015969495078480D0)
pearson_fit=y
RETURN
END
```