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Below-cloud scavenging of aerosol particles by snow at an urban site in Finland

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Below-cloud scavenging of aerosol particles by snow is an important mechanism of wet deposition in mid-latitude, polar and mountainous regions. The study presents an analysis of below-cloud snow scavenging of aerosol particles in the 0.01–1 μm size range and 0.1–1.2 mm h^{-1} snowfall rate range for an urban environment. The calculated mean scavenging coefficients were in good agreement with those previously reported for a rural background site in Finland. The variation of scavenging coefficients across the size distribution clearly exhibited a Greenfield gap for particles 0.06–0.3 μm in diameter. It was shown that snow is a better scavenger of aerosol particles than rain per equivalent water content. The relative humidity (RH) was deemed the most important meteorological parameter affecting the efficiency of snow scavenging, where an increase in the RH clearly resulted in an increase in below-cloud scavenging coefficient values. A new parameterization equation was developed for scavenging coefficients with respect to both particle diameter and relative humidity for snowfall intensities up to 1.2 mm h^{-1}.

Introduction

Aerosols are omnipresent in the atmosphere, and due to their nature they exhibit great variation in both spatial and temporal scales. Atmospheric aerosol particles come in a variety of shapes and sizes, and their chemical composition, physical properties and fate in the atmosphere are different for various aerosol species. Since the advent of the aerosol science submicron particles suspended in the air have been shown to be involved in a great variety of atmospheric processes, both physical and chemical, and to have a noticeable effect both on the human health and the global climate (Hinds 1999, Seinfeld and Pandis 2006). More recently, the effect of aerosols on climate has garnered a great deal of attention, as the previously well-studied and analyzed direct effects of aerosols on climate have been found to be incomplete, and more attention was drawn towards the effects of aerosols on cloud microphysics (Forster *et al*. 2007). This signifies the importance of various aerosol removal mechanisms in the atmosphere and the necessity of their presence in the global climate models. Even though the majority of aerosol particles in the atmosphere are of natural origin, the localized character of anthropogenic emissions of aerosols in populated and industrial areas and their transport have also brought the attention to the definition and characteristics of what is known as urban aerosol, and the processes it is involved in.

This study presents an analysis of below-cloud snow scavenging of aerosols in the 0.01–1 μm size range and 0.1–1.2 mm h^{-1} snowfall rate range for an urban environment, where the levels of air pollution are typically higher than in background sites (Monn *et al.* 1995). The motivation behind this paper is to improve the knowledge of snow scavenging processes, determine the most important factors that influence the efficiency of snow scavenging at the specified geographical location, and compare snow scavenging efficiency at an urban site with that of the rural background setting, as well as with rain scavenging. The analysis presented here concentrates on several key issues, including the dependence of scavenging efficiency of snow on the diameter of scavenged aerosol particles, and the determination and quantification of the relationships between snow scavenging and various micro-meteorological parameters.

Theory

The process of wet deposition describes the removal of particles from the atmosphere by hydrometeors (cloud and fog drops, rain, snow) and the consequent deposition onto the Earth's surface (Seinfeld and Pandis 2006); it is one of the key aerosol particle-removal mechanisms in the atmosphere. The exact details of this mechanism vary for different geographical locations and are influenced by a variety of factors, with micro-meteorological conditions having the greatest effect (Miller 1990). Rain scavenging of atmospheric aerosols has been studied extensively, owing to the fact that raindrops have a predominantly uniform shape, thus somewhat simplifying theoretical and empirical modelling of rain scavenging. Snow scavenging is a more complicated process due to the variety of frozen precipitation types and their physical properties (Graedel and Franey 1975). Snow flakes, ice grains and ice pellets all have varying densities and shapes, resulting in different fall velocities and cross-sectional areas, which ultimately determine how efficient the falling flakes and crystals are at scavenging suspended aerosol particles. It is important to mention that aerosol particles are generally scavenged at the rim of ice crystals because of a strong horizontal flow component underneath the falling ice crystal (Pruppacher and Klett 1997). The process is not well understood, and only a few recent studies are available on this topic (e.g. Jylhä 1999, Jylhä 2000, Lei and Wania 2004, Feng 2009, Kyrö *et al.* 2009). Wet deposition by snow is an important scavenging mechanism in mid-latitude and polar regions, where precipitation during the winter months generally occurs in the frozen form, as well as in the mountainous regions, where precipitation might occur in the frozen form due to higher elevations (e.g. Carrera *et al.* 2001). During summer and at the lower latitudes scavenging by snow also takes place in the upper levels of the troposphere. It is important to note that the incorporation of aerosol particles into precipitation particles occurs both inside and below the cloud; however, this paper concentrates only on the below-cloud scavenging of aerosol particles by snow through the mechanisms of Brownian diffusion, interception and impaction.

The general aspect of scavenging by falling precipitation particles (both liquid and frozen) is the dependency of below-cloud scavenging rate on the collection efficiency of a falling precipitation particle (Andronache *et al.* 2006). In many studies collection efficiency is considered to be equal to collision efficiency, even though it is not the case if the aerosol particle size is close to that of the hydrometeor (Andronache *et al.* 2006). At $d_p << D_p$ (where d_p is the aerosol particle diameter and D_p is the droplet diameter) the statement above usually holds true, and considering the fact that typical below-cloud particles behave like small spherical particles and collide with falling hydrometeors, the assumption that an aerosol particle is scavenged at every collision is a legitimate one. Like with many other aerosol processes, below-cloud scavenging is size-dependent. The smallest particles ($\leq$ 10 nm) are scavenged efficiently by Brownian diffusion, while particles with diameters larger than 2 μm are efficiently scavenged due to their inertia (Andronache *et al.* 2006). Similar to the dry deposition, particles in the accumulation mode do not have strong removal mechanisms in the atmosphere, even in regards to below-cloud scavenging. This results in what is known as a "Greenfield gap" — a term that defines particles that are least efficiently scavenged by precipitation, both liquid and frozen. Various studies report different boundaries for the size range of the Greenfield gap, with margins as wide as 0.01 μm and 2 μm (Andronache *et al.* 2006, Henzing *et al.* 2006). Slinn and Hales (1971) and Tinsley *et al.* (2000) suggested that thermophoresis and electrical charge, respectively, may enhance the scavenging of particles in the Greenfield gap. It is agreed, however, that the gap is a result of Brownian motion dominating the scavenging of smaller particles and inertial impaction dominating the scavenging of larger particles (e.g. Kyrö *et al.* 2009).

One of the most important parameters, used in the estimation of precipitation scavenging efficiency, is the scavenging coefficient λ_s; it is usually evaluated with respect to particle diameter, and thus has a form $\lambda_s(d_p)$. λ_s represents a fractional amount of aerosol removed by precipitation in a unit time for a certain aerosol particle diameter (Henzing *et al.* 2006). If one assumes that removal of particles is the only mechanism that changes particle concentrations, then the change in particle concentration c over time t can be described as

$$\frac{dc\left(d_p\right)}{\mathrm{dt}} = -\lambda_s c\left(d_p\right) \tag{1}$$

where $c(d_p)$ is the concentration of particles with a diameter d_p, dt is the time interval and λ_s is the scavenging coefficient (Seinfeld and Pandis 2006). According to the same source, Eq. 1 can be rewritten with respect to the scavenging coefficient itself

$$\lambda_s\left(d_p\right) = \int_0^\infty \frac{\pi}{4} D_p^2 U_t\left(D_p\right) E\left(D_p, d_p\right) N\left(D_p\right) dD_p \tag{2}$$

where D_p is the rain droplet diameter, U_t is the velocity of the falling rain droplet, $E(D_p,d_p)$ is the collection efficiency between the falling droplet and aerosol particle, and $N(D_p)$ is the concentration of rain droplets as a function of droplet diameter. E is usually described by the collection kernel K_p — a volume swept out by a hydrometeor in a unit time (Pruppacher and Klett 1997). Because of the assumption that E is equal to collision efficiency, K_p is a valid parameter in determining E. Equation 2 is applicable for all types of precipitation; however, because of the difficulty in determining the radius of a snowflake, D_p is usually replaced with a product of surface area factor and equivalent radius — a term known as capacitance (Pruppacher and Klett 1997). The exact mathematical approach to determining λ_s has been described by Sperber and Hameed (1986) and further utilized by a number of studies (e.g., Laakso *et al.* 2003). Equation 3 presents this approach.

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

Equation 3 allows for a mathematical determination of the scavenging coefficient λ_s if the time interval and corresponding particle concentrations are known. It can be used when hydrometeor scavenging is the only mechanism of particle removal in the atmosphere. However, as mentioned above, particle number concentrations may change due to a variety of other reasons. Equation 1 can be rewritten to account for these other mechanisms:

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

where the various subscripts indicate change in measured concentration due to (in order of appearance) instrumental errors (instr), turbulence (turb), advection (adv), condensation (cond), nucleation (nucl), coagulation (coag) and hygroscopic growth (hygr). Changes in particle concentrations due to instrumental errors and turbulence are rather difficult to account for; however, their effect is considered small when compared with that of snow scavenging for the studied particle size range (Kyrö *et al.* 2009). The effects of advection and the hygroscopic growth can be avoided by monitoring changes in air mass and relative humidity (RH), respectively, during the snowfall. Condensation, nucleation and coagulation are all assumed to be slow, if present at all, during the snowfall, minimizing their effect (Kyrö *et al.* 2009). With the manipulation of the original dataset, strict selection criteria of data and certain assumptions, these sources can be generally considered negligible when compared with scavenging by Brownian diffusion, impaction and interception specifically. It is important to mention that the concentration of aerosol particles in the air during a precipitation event does not decrease monotonically, and there may be significant observed fluctuations. Thus, experimentally derived λ_s may have both positive and negative values.

Several studies previously attempted to mathematically derive the values of snow scavenging coefficients (Table 1). Estimated values of scavenging coefficients vary in some cases by several orders of magnitude, indicating the derivation uncertainties and importance of external parameters, such as precipitation rate, size and type of frozen hydrometeors, size and initial concentration of aerosol particles etc. In one of the earlier studies of snow scavenging Graedel and Franey (1975) reported that scavenging coefficients λ_s showed no systematic dependence on aerosol particle diameter for the size range studied. The analysis determined that average particle lifetime due to snow scavenging was approximately 0.7 hours, which is one to two orders of magnitude smaller than for dry deposition. This resulted in a conclusion that in areas with significant snowfall during the winter season, scavenging by snow is a much more efficient removal mechanism than dry deposition. Martin *et al*. (1980) studied collection efficiencies of ice crystal plates, and determined a clear collection efficiency minimum for particles around 0.1 μm in diameter. The same study showed that the highest collection efficiency was recorded for ice crystals with diameters around 1 mm. Miller (1990) used a theoretical model to calculate λ_s values. The model included parameters of Brownian diffusion, thermophoresis, diffusiophoresis and electrostatic attraction, and, therefore, is not exactly comparable to below-cloud snow scavenging studied in this paper. The study carried out parameterization of λ_s with respect to aerosol size, snow crystal capacitance, atmospheric temperature and RH, and showed that λ_s is indeed quite sensitive to the model input parameters. The study concluded that RH is the most important parameter affecting the value of λ_s, where a 5% change in RH resulted in one order of magnitude opposite change in λ_s due to phoretic forces (Miller 1990). A 50% change in snow crystal capacitance was reported to cause only a two-fold change in λ_s, indicating that λ_s might depend more on micrometeorological parameters than on the type of frozen hydrometeors. Jylhä (2000) used radar data to model the scavenging coefficient as a function of precipitation rate during snowfall events near a coal-fired power station. The study dealt specifically with sulphur emissions and the values of λ_s were the smallest for all studies presented (Table 1). The final conclusion of the study was that the percentage of emitted sulphur scavenged by snow within the first 10 km from the emission source was equal to or less than 0.7%. Kyrö *et al*. (2009) is one of the most recent studies attempting to quantify the below-cloud snow scavenging efficiency at a background site in southern Finland for snow intensities below 0.8 mm h^{-1}. This study showed that scavenging by snow is more efficient than by rain per equivalent water content. Also in this study the Greenfield gap was observed for particles of 0.1–0.3 μm in diameter, and snowfalls with higher intensities were determined to likely scavenge particles in the air more efficiently.

Table 1. An overview of snow scavenging coefficients from previous studies.

Author	Scavenging coefficient λ_s (s^{-1})	Particle size range (μm)	Comments
Kerker and Hampl (1974)	2.47×10^{-6}–1.69×10^{-5}	0.05–0.5	For AgCl particles
Graedel and Franey (1975)	4.82×10^{-4}–6.35×10^{-3}	0.29–1.48	
Miller (1990)	1.26×10^{-4}	0.03	Modelled results
Jylhä (2000)	$\leq 10^{-6}$	–	Freshly emitted sulphur
Feng (2009)	1.11×10^{-6}–4.17×10^{-4}	0.001–10	Modelled for planar ice crystals and a snowfall rate of 0.25 mm h^{-1}
Kyrö *et al.* (2009)	8.7×10^{-6}–5.2×10^{-5}	0.01–1	

Methodology

The measurements were conducted at the SMEAR III (System for Measuring Ecosystem-Atmosphere Relationships) station in Helsinki, southern Finland (Järvi *et al*. 2009). The geographic coordinates of the station, which is located on a rocky hill 26 m above sea level, are 60°12´N, 24°58´E. The station is within the limits of the City of Helsinki and is located 5 km northeast of the downtown area, on the grounds of the Kumpula Campus of the University of Helsinki. The area around the station can be described as a very heterogeneous urban setting. Notable features include buildings, parking lots, roads, patchy forest and low vegetation. The proximity to the buildings, roads and other urban infrastructure is indicative of the station being in a typical urban environment, with likely a significant effect of this environment on meteorology and air quality in the area. Weather patterns and air quality in Helsinki are also influenced by the proximity to the sea.

Time, amount and type of frozen precipitation were measured by the Vaisala Present Weather Sensor FD12P. It is ground-based and located in the immediate vicinity of the SMEAR III station. FD12P is a complex and multitasking instrument, which detects and measures visibility up to 50 km range, precipitation type and intensity, precipitation accumulation, water equivalent of frozen precipitation and snow accumulation (FD12P Manual 2002: Weather sensor FD12P user's guide, Vaisala Oyj, Helsinki, Finland). For the detection of fog and differentiation between types of precipitation the instrument utilizes an optical forward-scatter sensor that measures the scattering of light at 875 nm wavelength in a sample volume of roughly 0.1 dm^3, located at the intersection of transmitter and receiver beams (de Haij 2008). This volume of air is at the elevation of roughly 1.75 m above the ground. The complexity of the instrument allows it to distinguish between 11 precipitation types with a detection limit of 0.05 mm h^{-1} and with a range of intensities from 0 to 999 mm h^{-1}. For the purpose of this study, the precipitation codes of interest were given following the World Meteorological Organization code 4680 format, the name of which is "Present weather reported from an automatic weather station" (World Meteorological Organization 1995). FD12P has proven to be a reliable instrument when measuring both precipitation intensity and type (Mallama *et al*. 2000).

Aerosol particle concentrations were measured with a Twin Differential Mobility Particle Sizer (TDMPS) system, the main components of which are a neutralizer, two Hauke-type Differential Mobility Analyzers (DMAs) and two Condensation Particle Counters (CPCs). The main principle of the DMA is sorting the particles according to their electrical mobility (Aalto 2004). CPC consequently determines the number of particles in each size bin by subjecting them to conditions of supersaturation with respect to water or alcohol vapour, and measuring the number with a simple optical particle detector. At the SMEAR III station, TDMPS is located in the container at the bottom of the tower, and it samples air through the inlet in the roof at a height of four metres above the ground. The sample inlet is shielded with a cover to protect it from falling precipitation. The TDMPS system in question classifies all particles into 38 logarithmically distributed size bins, with the first and last size bins having a mean diameter of 0.003 μm and 0.812 μm, respectively. Particle size distributions are measured with a time resolution of ten minutes, and the output format of particle number in each size bin is given as $dN/d\log D_\mathrm{p}$. The size distribution is calculated using a pseudo-inversion algorithm presented by Golub and Van Loan (1996).

Meteorological parameters of interest were air temperature, RH, wind speed and direction, atmospheric pressure, visibility and cloud base height. All of these are continuously measured at the SMEAR III station.

All abovementioned parameters were obtained for the period from October 2006 to April 2010. To analyze the below-cloud snow scavenging of aerosol particles by Brownian diffusion, interception and impaction only, strict selection criteria were applied to the data. Four precipitation types were of interest in this research, expressed as the WaWa WMO code 4680 (Table 2). Eligible episodes had to contain only the precipitation of types of interest and last for at least 120 minutes. Gaps in precipitation were allowed as long as they were under 60 minutes long and as long as the total duration of precipitation accounted for at least 75% of the total episode duration. Each episode was then examined with respect to meteorological parameters, with selection criteria set stricter than in a similar study by Kyrö *et al*. (2009), since weather patterns in seaside urban Helsinki are more heterogeneous and local sources of aerosol particles more abundant than in an inland rural background setting (Drebs *et al*. 2002). To avoid the effect of phoretic forces, the temperature was allowed to change by at most 3 °C throughout the episode, and the temperature above 0 °C was allowed as long as the detected precipitation was of frozen nature; additionally, to avoid the possibility of phoretic forces and hygroscopic growth of particles, RH was allowed to change by at most 10% throughout the episode. Other meteorological limitations included ±25° for the wind direction, ±1.5 m s^{-1} for the wind speed and ±2.5 hPa for atmospheric pressure. Any episode that did not fulfil all of these requirements was discarded. Two more selection criteria based on observed particle concentrations were then applied. First, each episode was analyzed with respect to particle concentration peaks due to rush hour traffic. Based on the day of the week and presence of peaks between 7:00–9:00 and 15:30–17:30, episodes were discarded accordingly. Second, episodes with clear growth of particles or erratic particle concentrations were also deleted. This was done by calculating the increase in particle concentrations over one or more consecutive time intervals. If over any number of consecutive time steps particle concentrations increased more than five-fold, the episode was discarded. The remaining number of episodes was the final group, for which the further analysis was carried out. All scavenging coefficients were then calculated using Eq. 3.

Results

Data statistics

A total of 51 episodes fulfilled all selection criteria and were selected for the analysis. The episodes ranged in duration from 120 to 600 min, with an average duration of 225 min; the total duration of all selected episodes is 11 470 min, equivalent to approximately 191 hours. More than half of the selected episodes, both by number and total duration, occurred during the winter of 2009–2010. Taking into account precipitation gaps within episodes, which were allowed to last for a maximum of 60 min and account for a maximum of 25% of each episode's total duration, the percentage of time that it was actually snowing accounted for 91.35% of the total duration of all episodes. Snowfalls of interest occurred from October to March, with almost half of the snowfalls both by number and duration taking place in February (Fig. 1A), which is in correlation with the fact that February is generally the coldest and snowiest month in the Helsinki area (Drebs *et al*. 2002). No

Table 2. WaWa codes of frozen precipitation.

Type	WaWa code	Description
Snow + rain	67	Rain (or drizzle) and snow, slight
	68	Rain (or drizzle) and snow, moderate or heavy
Snow	71	Snow, slight
	72	Snow, moderate
	73	Snow, heavy
Ice grains	74	Ice pellets, slight
	75	Ice pellets, moderate
	76	Ice pellets, heavy
	77	Snow grains
	78	Ice crystals
Snow showers	85	Snow shower(s) or intermittent snow, slight
	86	Snow shower(s) or intermittent snow, moderate
	87	Snow shower(s) or intermittent snow, heavy

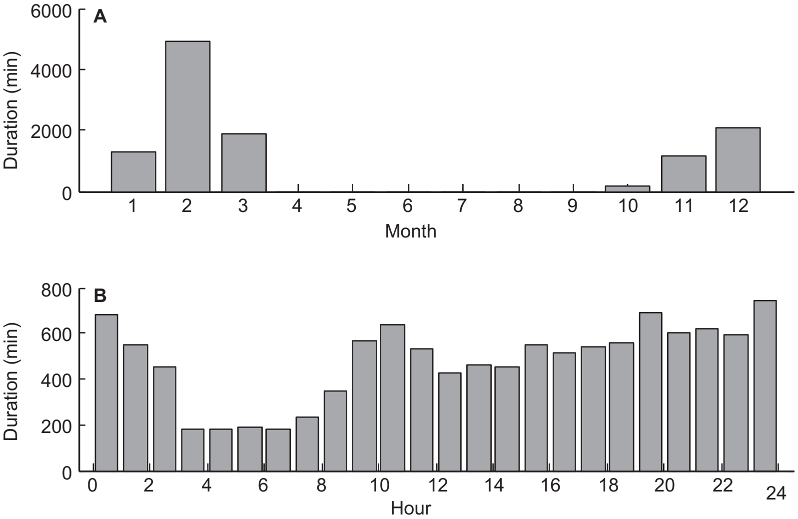

Fig. 1. (**A**) Monthly and (**B**) hourly distribution of all selected snowfall episodes.

particular trend in the hourly distribution of the selected snowfalls was observed, with the exception of a minimum from 3:00 to 9:00 (Fig. 1B). The selected snowfalls took place most often when wind was coming from the NE sector and least often when the wind was coming from SW sector (Fig. 2). This is an interesting observation since, on average, a typical wind pattern in Helsinki and the whole of Finland is the exact opposite (Drebs *et al*. 2002). It is, however, well-known that air masses coming from the SW sector (over the sea) usually have a moderating effect and bring warmer temperatures with them, which would hinder the occurrence of snowfalls in winter. Air masses from the NE are typically cold and dry — this explains the occurrence of suitable snowfalls with respect to temperature, but not to moisture content. It is assumed that a possible scenario of a snowfall of interest is that of an advancing north-easterly cold front, which results in frontal lifting of the warmer air and subsequent precipitation. Additionally, the observed snowfalls during the ENE winds may be explained by the coastal convergence, moisture advection and convection over the ice-free Gulf of Finland (e.g. Solantie and Drebs 2001). In order to thoroughly clarify the meteorological conditions during the snowfall cases, synoptic and meso-scale analyses would be required, which is beyond the scope of the present paper. More than 50% of all snowfalls had intensities of 0.1 and 0.2 mm h^{-1}; more than 75% of all snowfalls had intensities of 0.4 mm h^{-1} or less; the highest observed precipitation rate was 5.2 mm h^{-1} (Fig. 3A). The mean and median values of the snowfall rate were 0.38 and 0.2 mm h^{-1}, respectively. The 5th and 95th percentiles for the snowfall rate for the whole dataset were 0.1 and 1.2 mm h^{-1}, respectively. An overwhelming majority of the selected snowfalls occurred as the type “Snow”, which is continuous fall of snowflakes, with the whole dataset being largely dominated by the slight continuous fall of snowflakes (WaWa code 71) (Fig. 3B). Based

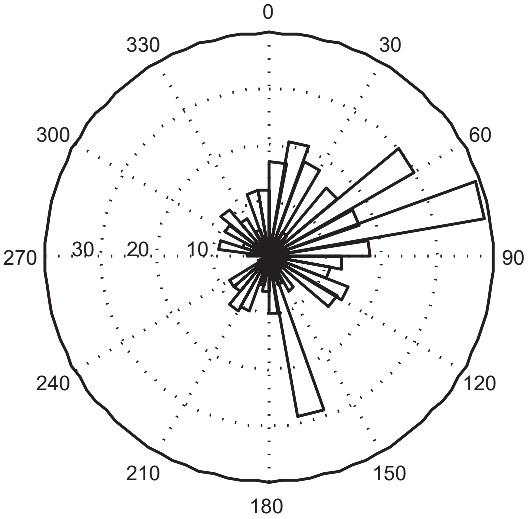

Fig. 2. Frequency distribution wind rose diagram for all selected snowfall episodes.

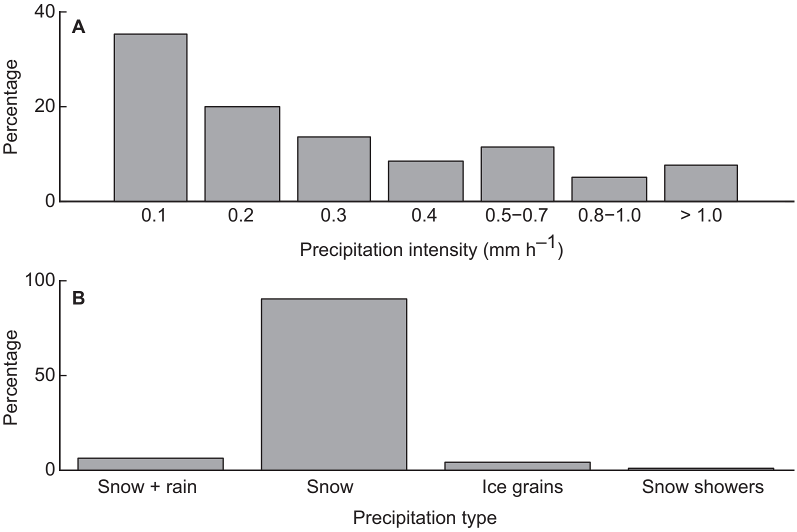

Fig. 3. Fractional distribution of (**A**) precipitation rate, and (**B**) precipitation type for all selected snowfall episodes.

on precipitation type classification (Table 2), 29 episodes contained only precipitation of the type "Snow" and 22 episodes contained precipitation of different types; from here on, these episodes will be referred to as "Snow" and "Mixed" episodes, respectively. For all selected episodes mean values for air temperature, RH, wind speed and air pressure were –6.07 °C, 87.03%, 3.81 m s^{-1} and 1001.58 hPa, respectively.

Scavenging coefficients

The calculated mean scavenging coefficients varied between 1.87×10^{-6} s^{-1} and 4.20×10^{-5} s^{-1} in the 0.01 to 1 μm size range. Across the whole size distribution the mean, median and standard deviation values were 1.42×10^{-5}, 7.75×10^{-6} s^{-1} and 7.08×10^{-4} s^{-1}, respectively. These statistical parameters are based on as many as 1146 scavenging coefficient values in each size bin. The values of scavenging coefficients determined in this research are in a reasonable agreement with some of the previous and most recent studies, although they fall slightly below the ranges reported by Kyrö *et al*. (2009) (8.7×10^{-6} to 5.2×10^{-5} s^{-1} for snow scavenging) and by Laakso *et al*. (2003) (7×10^{-6} to 4×10^{-5} s^{-1} for rain scavenging). The results here are also an order of magnitude smaller than those reported in some earlier studies (e.g. Graedel and Franey 1975, Miller 1990). The mean snowfall rate of 0.38 mm h^{-1} corresponds to the scavenging coefficient of 3×10^{-5} s^{-1} according to Chang (1986) and to 1.3×10^{-5} s^{-1} for D_p of 0.3–0.9 μm according to Jylhä (1999), with both of these values falling within the range reported in this paper. Several functions were tested to see which one would fit the dataset of mean scavenging coefficients better. For the sake of simplicity and the ability to draw meaningful comparisons, the function proposed by Laakso *et al*. (2003) for rain scavenging and further utilized by Kyrö *et al*. (2009) for snow scavenging was used (Fig. 4A). The function proposed here is written as:

$$\log\left(\frac{\lambda}{\lambda_0}\right) = a + d\left[\log\left(\frac{D_p}{D_0}\right)\right]^{-2} + e\left[\log\left(\frac{D_p}{D_0}\right)\right]^{-1} \quad (5)$$

where λ is the scavenging coefficient in s^{-1} with λ_0 set to 1 s^{-1}, D_p is the particle diameter in m with D_0 set to 1 m, and a, d and e are fitting parameters equal to 28, 1550 and 456, respectively. The function given by Eq. 5 yielded the R^2 value of 0.60, indicating a better fit of this function when compared with the dataset presented in Kyrö *et al*. (2009). Pearson IV function was also fit to the dataset of mean instant scavenging coefficients, and its R^2 value was 0.79 (Fig. 4B). The detailed description of the Pearson IV function can be found in Kyrö *et al*. (2009). It can be noted that several consecutive size bins exhibit clearly lower values than the rest of the dataset; the trough can be estimated to extend

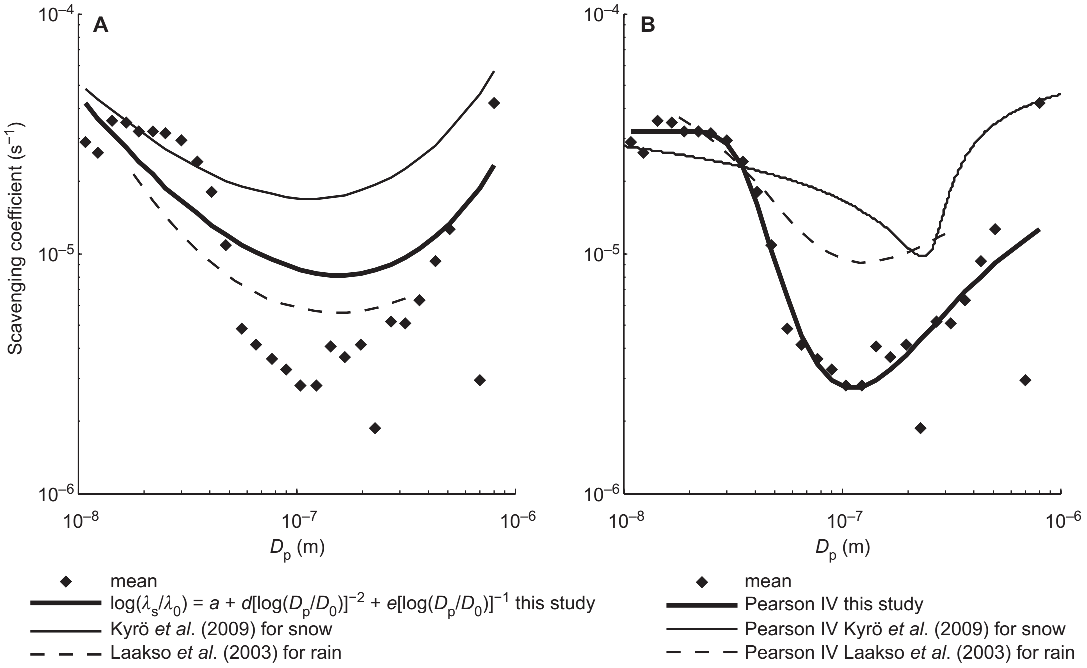

Fig. 4. Mean snow scavenging coefficients with (**A**) parameterization according to Laakso *et al.* (2003) function, and (**B**) Pearson IV function. Included in both figures are parameterizations from two previous studies.

from 0.06 to 0.3 μm (Fig. 4). This size range is in a very good agreement with the definition of a Greenfield gap. If some of the earlier studies reported the Greenfield gap being centred around larger particles (1 μm in diameter) (e.g. Radke *et al.* 1980), then some of the more recent ones indicated that the gap is located around 0.1 μm in diameter (e.g. Laakso *et al.* 2003, Andronache *et al.* 2006, Kyrö *et al.* 2009). It can, therefore, be concluded that this study further solidified the claim that particles around 0.1 μm are least efficiently scavenged by frozen hydrometeors, with this efficiency increasing for particles on either side of this minimum.

Comparing the results of this study with those reported by Kyrö *et al.* (2009) for a rural background setting reveals that snow scavenges particles slightly more efficiently in a rural background setting than in the urban environment of the current study (Fig. 4). However, while both fitted functions and the mean scavenging coefficients themselves support this claim, the observed pattern might simply represent the difference between the two sites in question, not necessarily differences in scavenging between rural and urban settings. Moreover, differences in scavenging might have arisen due to different chemical and physical properties of aerosol particles themselves, which were not analyzed. Nevertheless, it will be shown later that higher temperature and precipitation intensities result in higher rates of scavenging. Kyrö *et al.* (2009) opted not to include snowfalls occurring at temperatures above 0 °C, meanwhile approximately 16% of selected snowfalls in this study occurred at temperatures above 0 °C. Furthermore, the significant snowfall rate range in this analysis is wider than in the aforementioned study (0.1–0.8 mm h^{-1} and 0.1–1.2 mm h^{-1} in Kyrö *et al.* (2009) and in this analysis, respectively). Therefore, based on higher temperatures and precipitation intensities, it was expected that the current study would result in higher scavenging coefficient values than in the study by Kyrö *et al.* (2009), which was not the case. It will also be shown later that relative humidity is the most important meteorological parameter affecting the scavenging efficiency of snow, where higher RH results in higher rates of scavenging. Average RHs in the study by Kyrö *et al.* (2009) and in this analysis are 95% and 87%, respectively, indicating that of all meteorological parameters taken into account relative humidity might be the most likely reason for the observed differences in scavenging efficiency. An analysis of chemical and physical properties of aerosol particles, as well as a larger dataset are required to accurately describe differences in snow scavenging between rural and urban settings.

Differences in aerosol particle scavenging by snow and by rain may be inferred from the fitted parameterizations, as well as from scavenging coefficient values themselves; however, certain inconsistencies are evident (Fig. 4). While the log-function parameterization indicates that snow is a better scavenger of aerosol particles than rain (Fig. 4A), the Pearson IV parameterization exhibits the opposite trend (Fig. 4B). If the actual observed values are compared, scavenging coefficients are similar for both studies. It is necessary to mention, however, that the study by Laakso *et al*. (2003) used only episodes where minimum rain intensity was equal to or greater than 0.4 mm h^{-1}; their studied intensities were up to 20 mm h^{-1} with a mean intensity of 0.94 mm h^{-1}. The snowfall rates in this study included mostly those equal to or smaller than 1 mm h^{-1}, with the mean and median values of the snowfall rate of 0.38 and 0.2 mm h^{-1}, respectively. Since the difference in scavenging coefficient values for both studies is relatively small and since the study by Laakso *et al*. (2003) used larger precipitation intensities, it is suspected that snow is, indeed, a better scavenger of aerosol particles than rain per equivalent water content, which has to do with various shapes and sizes of frozen hydrometeors. This agrees with earlier studies by Magono *et al*. (1975) and Graedel and Franey (1975), as well as with more recent ones by Croft *et al*. (2009) and Kyrö *et al*. (2009). It is well-known that the scavenging efficiency of snowflakes is largely increased by the filtering effect of the holes within the snowflake, which creates a flow through rather than around the snowflake (Mitra *et al*. 1990).

Parameterization

Once the relationship between the particle diameter and scavenging coefficients was approximated, several parameterizations with respect to additional external parameters were carried out. Note, that the function fitted in all parameterization figures in this section is a Pearson IV function. Each figure consists of two parts, each showing the exact same dataset, with log-linear axes on the left-hand side and log-log axes on the right-hand side.

The parameterization with respect to precipitation type was impeded by the absence of sufficient amount of snowfalls types other than "Snow" (Fig. 3B). It will be recalled, however, that 29 episodes contained only precipitation of the type "Snow" and 22 episodes contained precipitation of mixed types (*see* Table 2). Therefore, to indirectly investigate the difference in scavenging efficiency of various frozen precipitation types, the parameterization with respect to episode type was conducted (Fig. 5). The mean scavenging coefficient values across the whole size distribution were 9.05×10^{-6} and 2.57×10^{-5} s^{-1} for the "Snow" and "Mixed" episodes, respectively. Both mean values and fitted functions indicate that particles are scavenged more efficiently when snow is mixed with some other type(s) of frozen hydrometeors, implying that these other types of frozen hydrometeors might be more efficient scavengers (Fig. 5). This statement is true for particles smaller than 0.06 μm and larger than 0.2 μm in diameter as in the 0.06–0.2 μm size range the two fitted functions converge, indicating similar scavenging efficiencies. Only three types of precipitation other than "Snow" were present in the dataset. "Snow + rain" accounted for almost 6% of the data and indicated that liquid hydrometeors were present together with frozen ones. Even though, as discussed and shown above, snow is a better scavenger of aerosol particles than rain, presence of raindrops at probably higher temperatures makes snowflakes stickier and larger, resulting in better scavenging due to increased size and terminal velocity (Eq. 2). "Ice Grains" accounted for almost 4% of the data and occurred mostly during warmer episodes. Martin *et al*. (1980) noted that ice crystals with the diameter around 1 mm have the highest collection efficiency amongst other frozen hydrometeors of this size. While the precise size of ice pellets in question is not known, it can be assumed that the higher abil-

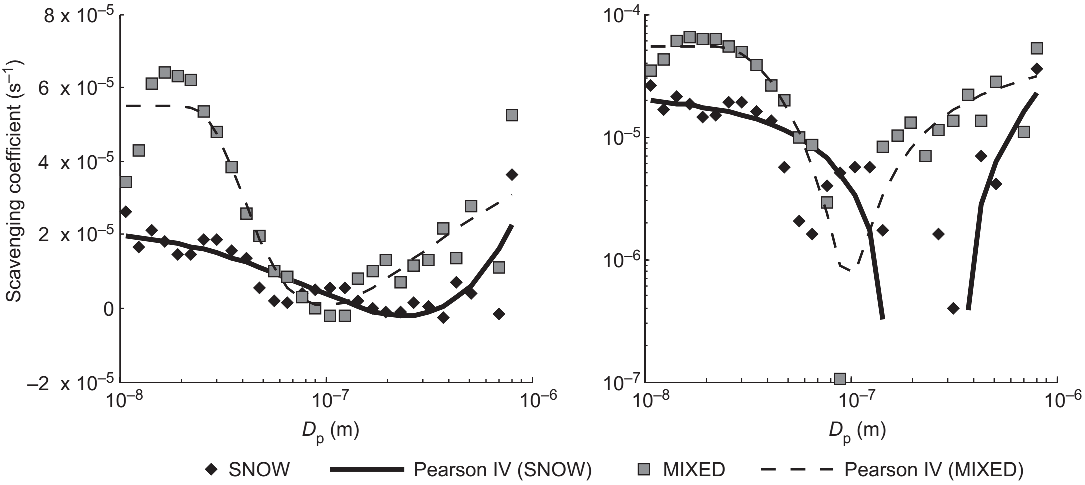

Fig. 5. Parameterization of scavenging coefficients with respect to particle diameter and episode type (left-hand side: log-linear axes; right-hand side: log-log axes).

ity of ice crystal rim to capture particles together with higher average air temperature resulted in higher rates of scavenging. “Snow showers” were present in only seven episodes with a total duration of just 100 minutes, and are similar to precipitation types of the category “Snow” but with higher intensities. It has already been mentioned that higher snowfall rates result in better scavenging, so this also partially explains higher scavenging coefficients for episodes with mixed types of frozen precipitation. Any assumptions about how different types of frozen precipitation might differ from each other with respect to their scavenging efficiency can only be based on this parameterization.

The parameterization of scavenging coefficients with respect to particle diameter was carried out for three temperature categories (Fig. 6). Percentage breakdown of instant coefficients was 39.6%, 44% and 16.4% for the $T < -7.5$ °C, -7.5 °C $< T < 0$ °C and $T > 0$ °C categories, respectively. Similarly, their respective mean scavenging coefficients across the whole size distribution were 6.34×10^{-6}, 1.92×10^{-5} and 3.63×10^{-5} s^{-1}. Evidently, scavenging of particles by snow is more efficient at higher temperatures, which is also supported by the aforementioned mean values (Fig. 6). For episodes occurring at above 0 °C, the snowflakes pass through the melting layer near the ground and start slowly melting and accumulating liquid water at the outer perimeter, becoming stickier and heavier. According to Eq. 2, the collection of aerosol particles by hydrometeors increases with increasing hydrometeor size and terminal velocity. Increased weight results in increased terminal velocity and better scavenging efficiency. The hydrometeor size distribution also changes during melting process: the total number of snowflakes decreases while their average size increases, which results in larger cross sectional areas. The opposite is true for the coldest episodes: at lower temperatures the air is typically very dry and ice crystals are small and lightweight. It is speculated that it is the small size and weight of ice crystals at lower temperatures, as well as lower rates of collision and growth, that result in lower rates of scavenging. Apart from the function of the coldest category, which reaches its minimum at 0.694 μm probably due to an outlier, the other two functions reach their minimum at around 0.1 μm, indicating the presence of the Greenfield gap at this particle size for different temperatures. The observed scavenging coefficients for the -7.5 °C $< T < 0$ °C category exhibit their greatest trough in the 0.123–0.231 μm size range, while for the above freezing category the trough is clearly in the 0.056–0.123 μm size range. While concluding that the Greenfield gap shifts towards smaller particles for snow scavenging at above freezing temperatures might be an overstatement, the differences in how frozen and melting

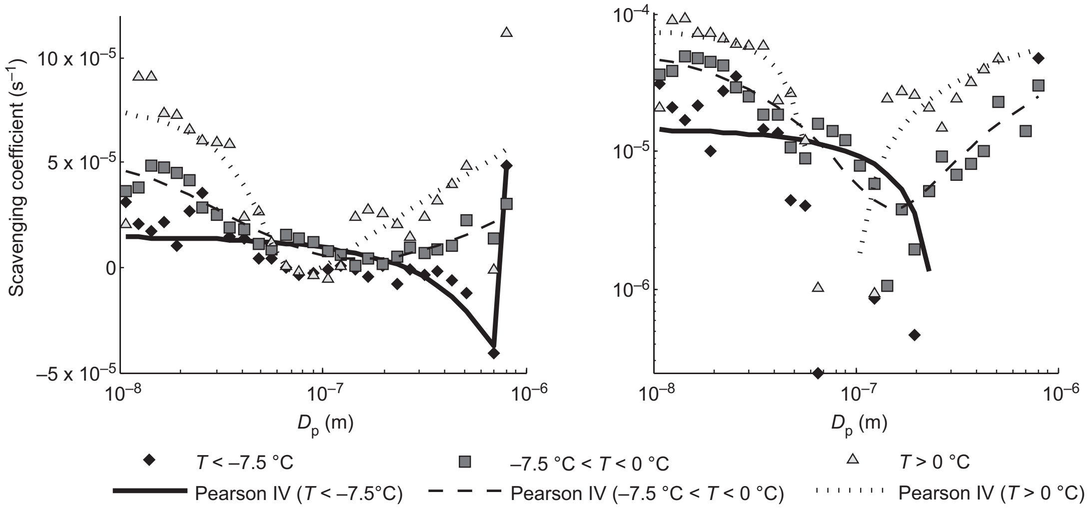

Fig. 6. Parameterization of scavenging coefficients with respect to particle diameter and temperature (left-hand side: log-linear axes; right-hand side: log-log axes).

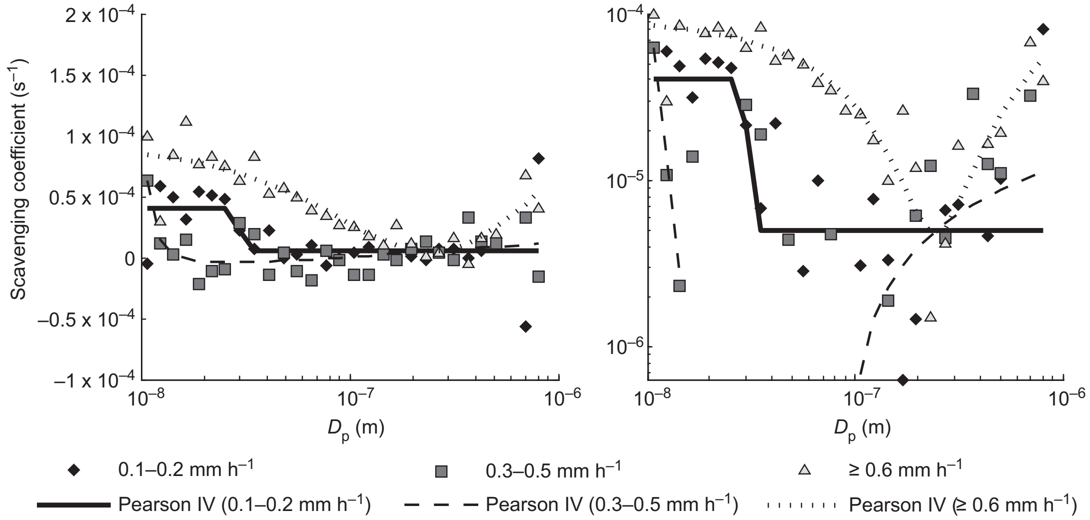

Fig. 7. Parameterization of scavenging coefficients with respect to particle diameter and precipitation intensity (left-hand side: log-linear axes; right-hand side: log-log axes).

hydrometeors interact with Aitken and accumulation mode particles is definitely of interest.

A parameterization of scavenging coefficients with respect to precipitation intensity was attempted despite very low absolute values of snowfall rates during the selected episodes. Three intensity classes were considered in this parameterization (Fig. 7). For 0.1–0.2 mm h^{-1}, 0.3–0.5 mm h^{-1} and ≥ 0.6 mm h^{-1} intensity categories, percentage of instant scavenging coefficients was 54%, 27.5% and 18.5%, and the mean scavenging coefficients across the whole size distribution were 1.46×10^{-5}, 4.29×10^{-6} and 4.33×10^{-5} s^{-1}, respectively. The difference in scavenging efficiencies for 0.1–0.2 mm h^{-1} and 0.3–0.5 mm h^{-1} classes is inconclusive as shown by the inappropriateness of the fitted Pearson IV function and the mean scavenging coefficient values for these two categories. Both Laakso *et al*. (2003) and Kyrö *et al*. (2009), as well as Chang (1986),

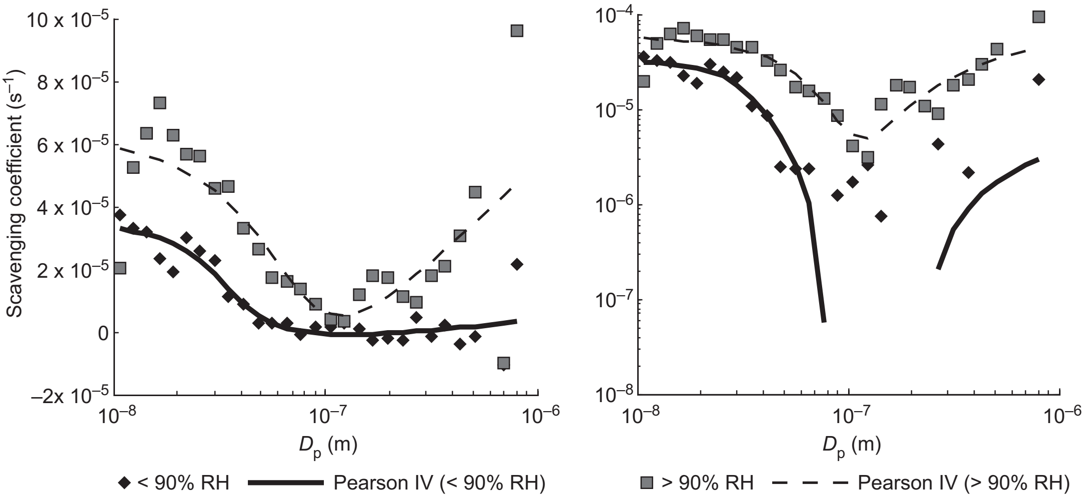

Fig. 8. Parameterization of scavenging coefficients with respect to particle diameter and relative humidity (left-hand side: log-linear axes; right-hand side: log-log axes).

Sparmacher *et al*. (1993), Jylhä (1999) and Feng (2009), studied the dependence of scavenging efficiency on the precipitation rate, reporting a positive correlation, justifying the expectation of higher scavenging coefficient values for the 0.3–0.5 mm h^{-1} category. Both 0.1–0.2 mm h^{-1} and 0.3–0.5 mm h^{-1} classes, however, exhibit a large scatter of the data in a seemingly random pattern, rendering their comparison ineffective. On the other hand, the third intensity class of ≥ 0.6 mm h^{-1} clearly exhibits higher scavenging coefficient values on both sides of the Greenfield gap, which is located around 0.24 μm for this intensity class. The effect of higher precipitation intensity is more pronounced for the particles in nucleation and Aitken modes. This idea is also supported by the highest mean scavenging coefficient value across the whole size distribution for this intensity class. It is speculated by the authors of this paper that the positive correlation between the scavenging efficiency of snow and the snowfall rate becomes more pronounced only when fairly different snowfall rates are compared. Snowfalls of higher intensities are needed to further solidify these claims.

The last parameterization was attempted to see whether RH has any effect on the scavenging efficiency of snow. Two RH classes were considered in this parameterization (Fig. 8). The 90% RH division value breaks down the whole dataset of instant coefficients into 64% and 36% fractions for below and above 90% RH, respectively, with their respective mean scavenging coefficients across the whole size distribution being 9.24×10^{-6} and 3.10×10^{-5} s^{-1}. It is easily seen that scavenging efficiency of snow is higher for higher RH, with mean coefficients indicating a three-fold increase in the scavenging efficiency. In fact, when the average scavenging coefficients are examined for every particle size bin, it is found that for all, except one, of the 28 size bins average scavenging coefficient in the above 90% RH category is higher than the corresponding value in the below 90% RH category. The particle diameter, at which both functions reach a minimum, is similar for both datasets: 0.123 μm and 0.118 μm for below and above 90% RH, respectively. Out of all parameterizations carried out, RH seems to have the largest and most discernible effect on scavenging efficiency of snow with a directly proportional relationship between the two parameters. It should be recalled that several studies in the past have demonstrated the exact opposite relationship between RH and scavenging efficiency, all based on theoretical model studies (e.g. Martin *et al*. 1980, Miller and Wang 1989, Miller 1990, Feng 2009). These studies have reported that the most efficient scavenging of aerosol particles by snow occurs in the driest conditions, attributing it to the increased thermophoresis and diffusiophoresis at lower RH values. While this does hold true, the goal of the current study is to examine the scavenging of particles by Brownian diffusion, impaction and interception only. It is anticipated that the physical properties of snowflakes are changed at high RH due to the water vapour condensation onto the existing surfaces. Field *et al.* (2006), Westbrook *et al.* (2008) and several other references have reported an increase in snowflake capacitance as a result of high RH values. Whether altering the shape or the size of the snowflake, the condensing water vapour molecules will inevitably make a snowflake bigger, which, as discussed earlier, will increase its cross-sectional area and terminal velocity and, hence, the scavenging efficiency. The condensation of water vapour onto the snowflake surface and the resulting increase in size, cross-sectional area and terminal velocity at high RH is deemed as the main reason for increased scavenging efficiency of snow at higher RH.

After determining that higher RH resulted in better scavenging of aerosol particles, cloud base height and visibility measurements were used to investigate whether the conditions of the measurement site being below the cloud are met when the relative humidity is high. Even though a negative correlation between the cloud base height and relative humidity was observed, the absolute minimum value observed during all selected episodes was 30 m above the station, indicating that below-cloud conditions were met at all times. Visibility data was examined as well, revealing that for 3% of all analyzed snowfall time visibility was below 1 km; however no significant correlation was found between visibility and relative humidity, as well as between visibility and scavenging coefficients. It is, therefore, speculated that even though visibility of less than 1 km might be indicative of fog conditions, it had no significant effect on the efficiency of scavenging of aerosol particles by snow.

Since the parameterization of the scavenging coefficients with respect to the particle diameter and RH turned out to be the most consistent and legible, and since the observational data proposed a pattern previously not observed, it was decided to further pursue this parameterization and to attempt to derive factual quantitative conclusions. However, for the purpose of this task the Pearson IV function was not the best choice due to its complexity, length and the resulting challenge in its applicability. It was, therefore, decided to use the function described by Eq. 5 and see whether that function can be modified to allow for a quantitative analysis of the dependence of scavenging efficiency on both particle size and RH. A clear positive correlation was observed between scavenging efficiency of snow and RH with a good R^2 value of 0.73 for six RH bins (Fig. 9); note that RH is expressed as a unitless number from 0 to 1. From Fig. 9 the correlation has the form

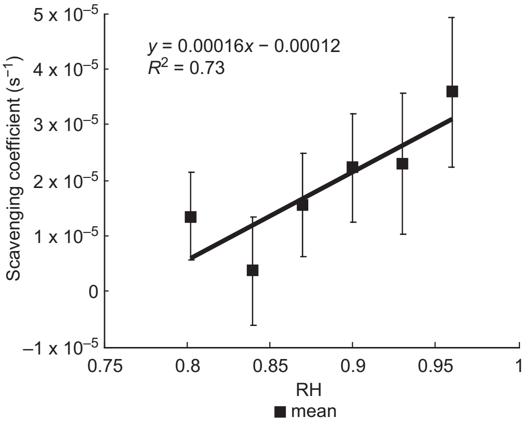

Fig. 9. Scavenging coefficients with their standard errors as a function of relative humidity; average over particle diameters 0.1–1 μm.

$$\lambda_s \sim g(\mathrm{RH}) - h \tag{6}$$

where RH is the relative humidity expressed as a unitless number from 0 to 1, and g and h are the fitted parameters. Rearranging Eq. 5 and adding the term in Eq. 6 results in an empirical relationship, which would be capable of estimating scavenging coefficients based on both particle diameter and RH.

$$\lambda_s = 10^{a+d\left[\log\left(\frac{D_p}{D_0}\right)\right]^{-2} + e\left[\log\left(\frac{D_p}{D_0}\right)\right]^{-1}} + g(\mathrm{RH}) - h \tag{7}$$

This function was fitted to the whole dataset of scavenging coefficients and the values of the fitted parameters a, d, e, g and h were found to be 28, 1550, 456, 0.00015 and 0.00013, respectively. The validity and applicability of this function was then examined for the two RH classes discussed in the original RH parameterization:

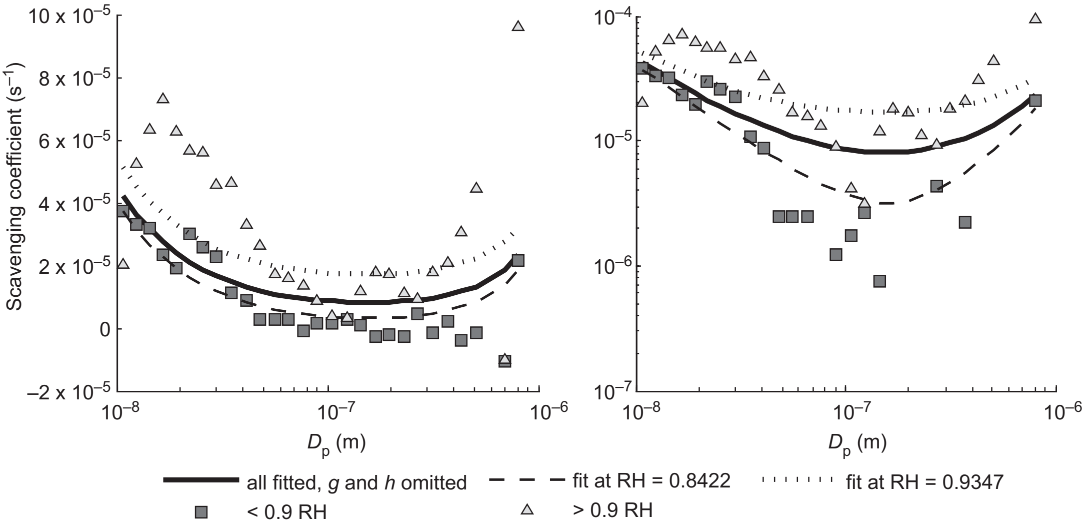

Fig. 10. Observed values and parameterization of scavenging coefficients with respect to particle size and relative humidity using Eq. 7 (left-hand side: log-linear axes; right-hand side: log-log axes).

below and above 0.9 RH. The mean RH values for these two classes were 0.842 and 0.935, respectively; standard deviations for these two RH classes were 0.039 and 0.021, respectively. As expected, at higher RH input the function indicates higher scavenging efficiency (Fig. 10). It is visible that functions based on Eq. 7 behave very similarly to the function based only on particle diameter (Eq. 5), with all exhibiting a minimum at 0.158 μm. However, the examination of R^2 values, which were 0.69 and 0.22 for the below and above 90% RH, respectively, reveals that differences between the observed and modelled data when RH is taken into account are large. It should be remembered, however, that in all previous parameterization, the fitting parameters were varied in order to find the best fit to the data and, therefore, as such they were different in any particular scenario in question. Equation 7 and the values of a, d, e, g and h mentioned above, on the other hand, present a comprehensive approach to determining the scavenging coefficient values based on the known RH in question and the desired particle size. It is believed that this is the first parameterization of its kind based on observational data. The parameterization shown in Eq. 7 may be used with moderate success to estimate the scavenging efficiency of snow based not only on particle size, but also on RH for snowfall intensities in the range of 0.1–1.2 mm h^{-1}.

Conclusions

The calculated mean scavenging coefficients varied between 1.87×10^{-6} s^{-1} and 4.20×10^{-5} s^{-1} in the studied size range, which is in good agreement with, and somewhat smaller than, those reported by Kyrö *et al.* (2009) for a rural background environment, and one to two orders of magnitude greater than those reported by Jylhä (2000) for sulphur emissions particles within 10 km from the source. The variation of scavenging coefficients across the size distribution clearly exhibited a Greenfield gap for particles of 0.06 to 0.3 μm in diameter. Direct comparison of scavenging efficiency between two different sites in Finland reveals that particles are scavenged by snow with a higher efficiency in rural background than in an urban environment; however the scavenging efficiency of snow is very sensitive to micrometeorological parameters and physical and chemical properties of aerosol particles at different geographical locations. The claim that snow is a better scavenger of aerosol particles than rain per equivalent water content was also shown to be a legitimate one. This is attributed to snowflakes being of different shapes and sizes and exhibiting a filtering effect due to the presence of holes and cavities within a snowflake. It was shown that snow scavenges the air more efficiently when it is mixed with other types of precipitation, indicating that these other types of frozen hydrometeors are more efficient scavengers. Snow was found to scavenge particles more efficiently at temperatures slightly above 0 °C, which is attributed to the fact that frozen hydrometeors start developing a liquid coating, become stickier, larger and heavier, thus increasing their terminal velocities and shifting the hydrometeor size distribution to the right. Similarly, the lowest rates of scavenging were observed at the coldest temperatures. Parameterization with respect to snowfall intensity indicated that scavenging efficiency might be positively correlated with the snowfall rate, although this trend is more pronounced when comparing very low and very high snowfall rates. RH was deemed as the most important meteorological parameter affecting the efficiency of snow scavenging, where an increase in RH clearly resulted in an increase in below-cloud scavenging coefficient values. The increase in scavenging efficiency at higher RH is due to the increase in snowflake capacitance. A new parameterization equation was developed for scavenging coefficients with respect to both particle diameter and RH. While it has not produced the desired correlation between the observed and modelled results, it is the first parameterization of its kind based on observational data, and it may be used with moderate success to estimate the scavenging efficiency of snow based not only on particle size, but also on RH.

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