BELOW-CLOUD SCAVENGING OF AEROSOL PARTICLES BY SNOW AT AN URBAN SITE IN FINLAND

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INTRODUCTION

Below-cloud scavenging of aerosol particles is an important mechanism of wet deposition, which can be accomplished by both liquid and frozen hydrometeors. The efficiency of this mechanism depends strongly on the nature of hydrometeors and aerosol particles, and, most importantly, on micro-meteorological parameters. Snow scavenging is a more complicated process when compared to rain scavenging due to the variety of frozen precipitation types and their physical properties. Raindrops are typically spherical or, at larger sizes, oblate, and, therefore, the modeling of their scavenging efficiency is simplified by this uniformity (Seinfeld and Pandis, 2006). The process of scavenging by snow is not well understood, and only a few recent studies are available on this topic (e.g., Jylhä, 2000 and 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; snow scavenging is also important in the mountainous regions, where precipitation might occur in the frozen form due to higher elevations.

This study presents an analysis of below-cloud snow scavenging of aerosols in the 0.01 to 1 μm size range for an urban environment, where the levels of air pollution are typically higher than in background sites. The main goals of this research are to determine how efficient the frozen precipitation is at scavenging aerosol particles at an urban site and how this compares to previously published results. The study also makes an attempt to demonstrate the dependence of scavenging efficiency of snow on various meteorological parameters.

METHODS

The measurements were conducted at the SMEAR III (System for Measuring Ecosystem-Atmosphere Relationships) station in Helsinki, Southern Finland. The station has the geographic coordinates of 60° 12’N, 24° 58’E and is located within the limits of the City of Helsinki, 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.

Time, amount and type of frozen precipitation were measured by the Present Weather Sensor FD12P. It utilizes an optical forward-scatter sensor that measures the scattering of light at 875 nm wavelength in a sample volume of roughly 0.1 dm³, located at the intersection of transmitter and receiver beams approximately 1.75 m above the ground. 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. Meteorological parameters of interest were air temperature, relative humidity (RH), wind speed and direction, as well as atmospheric pressure. All of these are continuously measured at the SMEAR III station.

The selection of snowfall episodes was based on their duration and gaps in precipitation, as well as on meteorological data and aerosol concentrations. Only the scavenging by Brownian diffusion, impaction and interception was of interest, and all other processes affecting particle number concentrations were minimized with the use of strict data selection criteria. Scavenging coefficients were calculated using the method described by Sperber and Hameed (1986) and further utilized by a number of studies (e.g., Laakso et al. 2003, Kyrö et al. 2009).

RESULTS AND DISCUSSION

A total of 22 episodes completely fulfilled the selection criteria and were used in the analysis of below-cloud scavenging efficiency of snow. The total duration of all episodes was equal to 85.5 hours, with an average episode lasting four hours. Almost 50% of the selected snowfalls occurred in February, which is in agreement with the fact that February is typically the coldest and snowiest month in the Helsinki region (Drebs et al., 2002). The examination of hourly distribution of the selected snowfalls reveals that snowfalls of interest occurred evenly throughout the day, with a clear increase in occurrence around midnight, more specifically from 23:00 to 03:00 FST. For all episodes mean values for air temperature, relative humidity, wind speed and air pressure were -4.22°C, 88.5%, 4.09 m/sec and 996.31 hPa, respectively. There was no prevailing wind direction during the selected snowfalls, however, most often the wind was coming from the NE sector.

The average snowfall rate value for the selected episodes was 0.43 mm/hour; the median value was 0.2 mm/hr. Almost 90% of the selected snowfalls had intensities of less than, or equal to, 1 mm/hr; more than 50% of the selected snowfalls had intensities of 0.1-0.2 mm/hr. The significant intensity range in this study is 0.1-1.6 mm/hr. Snowfalls of low intensities are typical in the Helsinki region (Drebs et al., 2002), and the median value reported here is equal to the one reported by Kyrö et al. (2009) for a rural environment in Southern Finland. Only five precipitation types were represented in the dataset, with an overwhelming majority being present as slight continuous fall of snowflakes. Other frozen precipitation types included snow mixed with rain, ice pellets and snow showers.

Figure 1 shows the mean instant scavenging coefficients as a function of particle diameter for the studied size range, including two fitted functions and parameterizations from two previous studies. Calculated mean scavenging coefficients varied between 6.65×10^-6 s^-1 and 5.14×10^-5 s^-1, which is in a good agreement with those previously reported for a rural background environment (Kyrö et al., 2009), and one to two orders of magnitude greater than those reported for sulphur emissions particles (Jylhä, 2000). The variation of scavenging coefficients across the size distribution clearly exhibited a trough of lower values, known as the Greenfield gap, for particles of 0.09 to 0.3 µm in diameter. If some of the previous studies reported the Greenfield Gap to be centered around 1 µm in diameter (e.g., Radke et al., 1980), this study further solidified the claim proposed by e.g., Laakso et al. (2003) and Andronache et al. (2006) that particles around 0.1 µm in diameter are least effectively scavenged by hydrometeors. Direct comparison of scavenging efficiency between two different sites in Finland reveals that particles are scavenged by snow with very similar efficiency in rural background and urban environments. Figure 1 also shows that snow is a better scavenger of aerosol particles than rain, per equivalent water content.

Several parameterizations were carried out with respect to scavenging coefficients, but their quantitative nature was hindered by a fairly small dataset. Snow scavenged particles more efficiently when it was mixed with some other type of frozen precipitation, indicating that these other types (snow mixed with rain, ice pellets and snow showers) scavenge particles more efficiently than snow alone. It was not possible to show this difference directly since frozen types other than snow were present in the dataset in insufficient amounts.
Snow was found to scavenge particles more efficiently at temperatures 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. Parameterization with respect to snowfall intensity did not exhibit any significant difference in scavenging efficiency for the studied snowfall rate range. Relative humidity was deemed as the most important meteorological parameter in determining the scavenging efficiency of aerosol by snow, where an 8% increase in RH resulted in one order of magnitude increase in below-cloud scavenging coefficient values. This trend is attributed to the increase in the snowflake capacitance at higher RH. Some of the earlier studies reported an opposite trend (e.g., Martin et al., 1980 and Miller, 1990), however in their analysis phoretic forces were taken into account. In this study these forces were meant to be minimized using appropriate data selection criteria, so the positive correlation between scavenging coefficients and RH is true only when scavenging by Brownian diffusion, impaction and interception is considered. A new parameterization equation was developed for scavenging coefficients with respect to both particle diameter and relative humidity.

\[
\Lambda_j = 10^{a+d\log(D_p / D_{p0})^{-1} + e\log(D_p / D_{p0})^{-1} + g(RH) - h}
\]

where \(\Lambda_j\) is a scavenging coefficient in s\(^{-1}\), \(D_p\) is particle diameter in m with \(D_{p0}\) set to 1 m, \(RH\) is relative humidity expressed from 0 to 1, and \(a, d, e, g, h\) are fitted parameters. Figure 2 shows all modelled data with \(g\) and \(h\) terms omitted, as well as observed and modeled results for two RH classes. The produced correlation between observed and modeled results was not as good as anticipated; however, 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 relative humidity.

Cases where snow did not produce a significant scavenging effect on aerosol concentrations were shown to be attributed to non-uniform precipitation around the measurement site with the use of the radar data.

Figure 1. Instant mean scavenging coefficients with two parameterizations. Parameterizations from two previous studies are also shown.
CONCLUSIONS

Calculated scavenging coefficients agreed well with those previously published and once again indicated that snow is a better scavenger of aerosol particles than rain, per equivalent water content. No significant difference was observed in the scavenging efficiency of snow between rural and urban environments in Finland. The Greenfield Gap was observed for particles in the size range of 0.09-0.3 µm in diameter. Scavenging efficiency of snow was shown to depend strongly on precipitation type and meteorological parameters, with relative humidity exhibiting the greatest effect.

Figure 2. Observed values and parameterization of scavenging coefficients with respect to particle size and relative humidity.

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REFERENCES


