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Observations on aerosol optical properties and scavenging during cloud events

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Abstract

Long term statistics of atmospheric aerosol and especially cloud scavenging were studied at the Puijo measurement station in Kuopio, Finland, during October 2010 – November 2014. Aerosol size distributions, scattering coefficients at three different wavelengths (450 nm, 550 nm, and 700 nm), and absorption coefficient at wavelength 637 nm were measured with a special inlet system to sample interstitial and total aerosol in clouds. On average, accumulation mode particle concentration was found to be temperature dependent with lowest average concentrations of 200 cm^{-3} around 0°C increasing to more than 800 cm^{-3} for temperatures higher than 20°C. From the in-cloud measurements, both scattering and absorbing material scavenging efficiencies were observed to have slightly increasing temperature dependence. At 0°C the efficiencies of scattering and absorbing matter were 0.85 and 0.55 with slopes of 0.005 $\frac{1}{°C}$ and 0.003 $\frac{1}{°C}$, respectively. Additionally, scavenging efficiencies were studied as a function of the diameter at which half of the particles are activated into cloud droplets. This analysis indicated that the is a higher fraction of absorbing material, typically black carbon, in smaller sizes so that at least 20-30% of interstitial particles within clouds consist of absorbing material. In addition, the PM1-inlet revealed that approximately 20% of absorbing material was observed to reside in particles with ambient diameter larger than ~1 µm at relative humidity below 90%. Similarly, 40% of scattering material was seen to be in particles larger than 1 µm. Altogether, this dataset provides information on size dependent aerosol composition that can be applied in evaluating how well large-scale aerosol models reproduce aerosol composition, especially with respect to scavenging in stratus clouds.

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1 Introduction

Aerosol particles in the atmosphere induce radiative forcing by absorbing and scattering light (direct effect) and by modifying cloud properties by acting as seeds for cloud droplets (indirect effect) (Lohmann and Feichter, 2005). The interaction of aerosols with radiation depends highly on their physico-chemical properties, such as size, morphology, and chemical composition. These properties are source-specific and evolve during the life-cycle of aerosol in the atmosphere. Aerosol particles can, for example, grow by condensation, coagulate with each other, exchange phase, or get cloud-processed, i.e. grow in clouds due to chemical processes and coagulation until they are finally removed from atmosphere through dry or wet deposition. If the atmospheric lifetime of an aerosol particle is long, it can travel long distances from the emission source region and cause radiative forcing over areas with low natural aerosol sources, like for example black carbon (BC) over Arctic areas (Chaubey et al., 2010, Latha et al., 2005). To estimate the radiative forcing of the anthropogenic aerosol, all these processes from aerosol formation to the removal must be understood and resolved.

The interaction of aerosols with clouds, especially cloud scavenging, changes the physical, chemical, and optical properties of aerosol particle populations (Browse et al., 2012; Berkowitz et al., 2011; Portin et al., 2009; Verheggen et al., 2007; Cozic et al., 2007; Komppula et al., 2005). Although aerosol properties evolve already at relative humidity (RH) below 100% due to microphysical processing, aerosol properties undergo a much faster change within clouds where a fraction of submicron aerosol particles is grown into cloud droplets with sizes of the order of 10 micrometres in diameter (Väisänen et al. 2016, Anttila et al. 2012). In order to be cloud-processed, an aerosol particle must be able to act as a cloud condensation nucleus (CCN). The CCN activity of an aerosol particle depends on its size and hygroscopicity (Swietlicki et al., 2008). Inorganic salts, such as sulphates, nitrates, and ammonium, are known to be hygroscopic but, for example, pure black carbon is highly non-hygroscopic (McMeeking et al., 2011). Typically, hygroscopic particles such as sulfate are known to be good scatterers of light, whereas insoluble compounds such as BC absorb light efficiently (e.g. Bergstrom et al. 2002). Therefore, in case of particles of the same size, the scattering aerosols are scavenged by cloud droplets more frequently. On average, this results in less numerous, more absorbing, and smaller aerosol particles if cloud scavenging leads to wet scavenging. With such an aerosol population the cooling effect is decreased compared to the warming effect of absorbing aerosols.

Optical and CCN properties are influenced by the size and the mixing state of the BC containing particles. Fors et al. (2011) and Väisänen et al. (2016) found that typically the accumulation mode contains a higher fraction of hygroscopic particles than Aitken mode in an externally mixed ambient aerosol. Due to aging processes, atmospheric BC particles contain a variable fraction of other chemical components (inorganics, water, water-soluble organics), and these compounds may enhance the light absorption of the particles (Bond et al., 2006). Coatings may also increase the CCN activity through elevated hygroscopicity (Liu et al., 2013) although the enhancement depends on the amount of coating and is not always extensive (Cappa et al., 2012). However, because of such a mixing, it is not straightforward to determine if the overall role of BC containing particles in the atmosphere is always warming, or if these particles actually cause negative forcing through the Twomey effect.

Experimental information about physical, optical, and chemical properties of aerosols is achieved from long-term in situ measurements which have been and are being conducted in several places throughout the world (e.g., Collaud Coen et al., 2013; Asmi et al., 2013). The effect of clouds or precipitation on aerosol optical properties has also been studied in a few campaigns (Zhang et al., 2012; Berkowitz et al., 2011; Hyvärinen et al., 2011; Chaubey et al., 2010; Marcq et al., 2010; Yamagata et al., 2009; Cozic et al, 2007; Latha et al., 2005) and model calculations (e.g. Browse et al., 2012; Croft et al., 2009). The wet scavenging of different compounds has great importance on the global aerosol composition. As scavenging of different compounds depends on where the compounds reside in the aerosol size spectrum, there is a need for

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measurements on how chemical composition is partitioned between interstitial aerosol and cloud droplets. However, such long term measurements are conducted only in a few locations related to low altitude clouds or mountain stations. For example, a handful of studies have been carried out in which cloud scavenging efficiency of black carbon has been studied as reviewed by Yang et al. (2019).

The partitioning of aerosol particles between the interstitial and activated (i.e. cloud droplet) phase can be studied by parallel measurements of the cloud interstitial and activated aerosols, for which different methods have been used: an interstitial inlet with a cut-off diameter low enough to remove cloud droplets, a counterflow virtual impactor (CVI) to extract cloud droplets (Noone et al., 1988) and ice particles from mixed phase clouds (Mertes et al., 2007), and a total air inlet, which samples both the interstitial and activated phases (Weingartner et al., 1999). In principle, the most suitable instrument to study the properties of activated aerosols is the CVI, which samples only large particles (or cloud droplets) with enough kinetic energy to pass through counterflow section, which removes small particles. A drawback of CVIs is the enrichment of the sample, as the input flow is larger than sample flow (Twohy et al. 2003). The total and interstitial inlets sample particles smaller than predetermined cut-off sizes which enables getting information about cloud scavenged and non-scavenged particles. In this work, we used a combination of interstitial and total inlets, and analysed the properties of activated particles as a difference between the inlets. Beyond reporting typical aerosol properties, our aim was to clarify how ambient conditions and properties of aerosol population influence the scavenged fraction of light absorbing and scattering aerosol particles.

2 Methods

2.1 Site description

The measurement site is located on the top of an observation and retransmitting tower at Puijo (62°54'34" N, 27°39'19" E) in Kuopio, Finland. The measurement height is 306 m above sea level and 224 m above the surrounding lake level. In the surroundings of the measurement site there are a few local pollutant sources, such as a paper mill, a district heating plant, traffic routes, and residential areas with biomass-fired combustion appliances. At this measurement site we are able to study how the properties of the fresh aerosol emissions from the local sources differ from aged aerosol observed at Puijo. Furthermore, the measurement site is approximately 8% of the time inside low-level clouds, which enables us to study the aerosol-cloud interactions in a continuous, long-term manner. The cloudiest period is during autumn (Fig. 1), when the tower is inside clouds about 13% of the time. A more detailed description of the measurement site, its surroundings and previous studies is given in Leskinen et al. (2009), Portin et al. (2009, 2014), Hao et al. (2014), Väisänen et al. (2016) and Romakkaniemi et al. (2017).

2.2 Instrumentation

We measured temperature, relative humidity, horizontal wind direction and visibility on the roof of the tower with a time resolution of 1 min, by using a temperature and humidity transmitter (Vaisala, Model HMT330MIK), an ultrasonic wind anemometer (Thies, Model UA2D), and a present weather sensor (Vaisala, Model FD12P). We also measured the cloud base height at the nearby Savilahti weather station (2 km south-west from Puijo) with a ceilometer (Vaisala CT25K).

Aerosol measurement instruments were placed on the upmost floor inside the tower. The sample air for the instruments was drawn through two separate sampling lines: the interstitial and total aerosol. The former is equipped with a 1.0-µm cutoff impactor which removes coarse particles and cloud droplets from the sample. The latter has a snow hood and a heated inlet, with a cut-off size of 40 µm (Weingartner et al., 1999). Water on the cloud droplets evaporates in the total aerosol inlet and sampling line. The resulting total aerosol contains therefore both the interstitial aerosol and cloud droplet residual. The

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properties of the cloud droplet residual can then be estimated, at least for additive parameters, by subtracting the property of the interstitial aerosol from that of the total aerosol. This so-called twin-inlet system makes it possible to compare the properties of non-activated and activated aerosols, making it a suitable method for studying how aerosol mixing state affects cloud droplet formation and aerosol scavenging.

We measured the aerosol scattering (at 450, 550, and 700 nm) coefficients by using a TSI Model 3563 integrating nephelometer (Anderson et al., 1996) and absorption (at 637 nm) with a Thermo Model 5012 multi-angle absorption photometer (MAAP; Petzold and Schönlinner, 2004). Both instruments took samples from both the interstitial and total line with the help of a controllable valve system (shown in Fig. 2). The system switched the sampling lines in 6-min intervals. In this configuration the flow rate through both instruments was 8.0 lpm, and we collected the data with a time resolution of 1 min from both devices.

Particle number size distributions were measured by a twin differential mobility particle sizer (DMPS; self-constructed) (Winklmayr et al., 1991; Jokinen and Mäkelä, 1997). The system was also used to get information about cloud activation of the particles and to determine the diameter at which half of the particles are activated into cloud (D_{50}). The measured size range was originally 7–800 nm and was extended to cover the range of 3–800 nm in February 2012. The DMPSs were also part of the controllable valve system like the nephelometer and MAAP. Throughout the measurements, we performed periodic flow checks and calibrations for the instruments to ensure the high quality of the measurements.

2.3 Data processing

The analysed data set covered the period 5 October 2010 – 30 November 2014, including meteorological variables, and, aerosol particles optical properties and size distributions for both the total and interstitial lines. All data is averaged for 12 minutes based on DMPS time resolution. First, we ruled out non-usable data during maintenance, calibrations, flow checks and autozeroing (i.e. periodic automated measurement to determine the background noise) from the optical variables. In addition, data with abnormal peaks and negative values was removed. As in our earlier work (Leskinen et al., 2012), we omitted the scattering data when relative humidity (RH) exceeds 50% in the integrating nephelometer inlet and corrected the scattering coefficient for truncation errors (Anderson and Ogren, 1998).

We categorized the data into three categories according to the cloudiness conditions and named them as “no cloud”, “cloud”, and “intermediate”. The criteria for “no cloud” were: RH < 80% and visibility > 8000 m or, if either RH or visibility data was not available, cloud base height (CBH) > 500 m. For “cloud” category: visibility limit was < 200 m or, if visibility data was not available, measured RH > 99%. The periods not meeting either of these criteria were labelled as “intermediate”. The fractions of each category from 5 October 2010 to 30 November 2014 were 44.6%, 8.0%, and 47.4% for “no cloud”, “cloud”, and “intermediate”, respectively.

We calculated scavenging fractions, i.e. the fractions of the absorbing and scattering material in the activated (cloud) phase with the following equations:

$$F_{scav,a} = \left(\frac{\sigma_{ap,\lambda,tot} - \sigma_{ap,\lambda,int}}{\sigma_{ap,\lambda,tot}}\right) \quad (1)$$

and

$$F_{scav,s} = \left(\frac{\sigma_{sp,\lambda,tot} - \sigma_{sp,\lambda,int}}{\sigma_{sp,\lambda,tot}}\right). \quad (2)$$

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In Equations (1) and (2) $\sigma_{sp,\lambda}$ is the scattering coefficient and $\sigma_{ap,\lambda}$ is the absorption coefficient at a given wavelength λ.

In this study we defined the accumulation mode number concentration (N_{acc}) as the number concentration of particles larger than 100 nm in diameter (electrical mobility diameter). N_{acc} is found to be a representative proxy for CCN in boundary layer stratified clouds in boreal conditions (Lihavainen et al. 2008). Naturally, total number concentration (N_{tot}) is defined as the number concentration of all particles. In addition we calculated volume concentrations (V_{tot} and V_{acc}) with the same particle diameter limits.

3 Results and discussion

3.1 Aerosol number concentrations

The aerosol emissions near the Puijo site have a large annual variability as the contribution of different emission sectors to the aerosol load is highly dependent on the season. To analyse the characteristics of size dependent aerosol properties in different weather conditions, we studied the behaviour of total and accumulation mode particle number concentrations as a function of temperature in all measured cases (Fig. 3a,b,c) and in-cloud cases (Fig. 3d,e,f). Panels a and d present total number concentrations, panels b and e present accumulation number concentrations, and finally, panels c and f present ratio of the accumulation and total number concentrations. Note the different concentration ranges in each panel and lack of cloud event data in temperature ranges from -20°C to -15°C and from 20°C to 25°C.

The total number concentration has a direct proportionality to air temperature as seen from all data case (Fig. 3a) and cloud cases only (Fig. 3d). However, at low temperatures the total particle number concentration increases (Fig. 3a) which can be caused by increased local anthropogenic emissions. The main reason during cold weather is the need of heating and electricity whereas the effect of biogenic emissions is no longer relevant (Geron and Arnts 2010, Tarvainen et al. 2005, Leaitch et al. 2011, Paasonen et al. 2013). In addition, it has been shown that vehicle emissions have higher impact in rather cold than warm temperature (Wang et al. 2017). While most of the time total number concentrations are relatively low, aerosol nucleation events with high total concentration occur, and this can be seen as a difference in median and average concentrations.

In the number concentration of accumulation mode particles presented as a function of temperature (all data case) we can see a parabolic curve with a minimum around 0°C (Fig. 3b). Similar effects have been reported earlier (Asmi et al. 2016, Paasonen et al 2013, Hussein et al. 2006). In the warm temperatures (above +10°C) accumulation mode particle number concentration increases rapidly which is likely caused by increase in biogenic emissions (Ahlm et al. 2013). Additionally, wet deposition tends to decrease in extremely warm temperatures as precipitation decreases. However, the low concentrations around 0°C are generally thought to be caused by lack of biogenic emissions as well as increased amount of precipitation, and thus, wet deposition. Interestingly, the number concentration begins to increase towards colder temperatures in the all data case but not in cloud event case. The increase could be explained by decreased amount of deposition and lower height of the boundary layer together with increasing amount of aerosol from local house heating and energy production.

In addition to total and accumulation particle number concentrations, we studied the fraction of accumulation sized particles from the total number. The ratio of accumulation to total particle number concentrations of all data (Fig. 3c) and cloud case data (Fig. 3f) show similar parabolic curve as in Fig. 3b. However, the curve is not strong in cloud case data, especially at

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cold temperatures. This backs up the notion made in previous paragraph about accumulation particles. Precipitation can be one of the explaining factors since precipitation events are most frequent around 0°C. Another interesting notion is that the ratio is slightly higher on average during cloud cases, excluding the below -5°C region. This would imply faster coagulations/coalescence removal of nucleation and Aitken mode particles than that of accumulation mode particles (Laakso et al., 2003).

3.2 Influence of environmental parameters on the aerosol optical properties during cloud events

3.2.1 Relative humidity and scavenged particle volume fraction

Information about the composition size distribution, especially with respect to scattering and absorbing compounds, can be inferred by how their sampling in the interstitial line changes at different humidities. For compounds which are dominantly in the largest particles, are "scavenged" (i.e. are too big to be sampled by the interstitial line but not technically cloud scavenged) at high relative humidities. Meanwhile, this scavenging of compounds in the smaller particle sizes is less affected by the increase in the relative humidity.

Here we present the scavenging efficiencies of scattering and absorbing material studied as a function of air relative humidity (Fig. 4 a,c) and scavenged total aerosol volume (Fig. 4 b,d). The box plots in Fig. 4 a and c illustrate how the scavenged fraction changes when there is a transition from cloud free to cloudy periods. The relative humidity subplots show an interesting effect of scavenging even at moderate air humidity. About 40% of scattering and 20% of absorbing material is scavenged by impactor already at 50% RH. However, there is a large variability in the observed scavenging fractions because of different aerosol size and chemical compositions during measurement period. This can be seen from the percentiles in the box plots. Interestingly, on average 20% of aerosol absorption is caused by particles close to or bigger than 1 µm. The origin or composition of this aerosol is not known as it is larger than typically analysed with mass spectrometry for example. However, the fraction is very similar to what reported previously by Luoma et al. (2019) as a fraction of absorbing material in particles between 1 µm and 10 µm in Hyytiälä, Finland. When reaching 100% air humidity both scavenging efficiencies increase. This is the transition zone to cloud cases. On average 70% of scattering material but only 30% of absorbing material is scavenged when the cloudy period begins.

While Figure 4 a and c showed the change in the fraction of scavenged scattering and absorbing material in transition from cloud free to cloudy conditions, Figures 4 b and d illustrate how the fraction further changes when the fraction of activated particles increases. Even in the wavering cloud cases 70% of scattering and nearly 30% of absorbing material is scavenged by clouds. As expected, in more prominent cloud cases (meaning how much of the total aerosol volume is scavenged) less scattering and absorption is measured from the interstitial sample line which leads to higher scavenging fractions. In practice, all of the scattering and nearly 80% of absorption material is scavenged by clouds when more than 90% of submicron aerosol volume is scavenged.

These results indicate a couple of notions regarding the sampling setup and the data analysis. First, sampling system with 1.0-µm diameter cutoff can lose relevant particles (measurement site specific). Depending on the ambient conditions hygroscopic and aged BC particles, that are expected to be sampled, are undetected by the interstitial line due to particle growth. Second, scattering is dominated by large particles, which is expected by scattering theory and is also reported in earlier studies (Anttila et al. 2012, Portin et al. 2014). However, scattering data can greatly help to explain absorption data. As mentioned earlier, about 40% difference between sampling lines even at moderate air humidity implies the presence of larger than 1 µm particles or growth of hygroscopic particles beyond 1 µm in size. The same applies to the absorbing material that has aged and is coated with hygroscopic coating (e.g. aged BC particles) and can be removed before cloud

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formation. However, typically BC resides in particles that are much smaller than 1 µm and thus the gradient in scavenging efficiency as a function of relative humidity is less steep than that for scattering fraction. Another indication for absorbing material to reside in smaller sizes is that more than 20% of the absorbing material is not scavenged after more than 90% of aerosol volume is scavenged by clouds.

It is clear from Fig. 4 that the dispersion of the data is fairly large and includes non-physical values such as scavenging efficiencies out of the range from 0 to 1. This cannot be avoided as there is a slight time difference between the measurement from the interstitial and total line, and this can introduce negative scavenging efficiency values when considering small absolute values of scattering and absorption coefficients. Another reason for the anomalous values is that the interstitial line values are practically zero in many of the cloud cases but, due to device measurement accuracies, can sometimes produce small negative values (especially for the scattering coefficient). This leads to situations where the estimated scavenging fraction is slightly over unity. Third possible source of dispersion resides in long term measurements. Since the dataset is large it is probable that similar kind of meteorological conditions occur multiple times during measurement period but with different kind of aerosol properties.

In comparison to Cozic et al. (2007) and Motos et al. (2019) we did not observe 1:1 line for scavenged absorption matter as a function of scavenged particle volume. It is worth to note that in our case the scavenged aerosol volume is calculated from the DMPS size distribution, thus including only submicron particles. However, while this has been taken into account, our results show that some amount of absorbing matter is left in the particle phase even with high particle volume scavenging efficiency. This difference is presumably caused by vicinity of the BC source regions, and relatively low supersaturation in the observed clouds which limits the activation of small particles.

3.2.2 Impact of air temperature

One of the main objectives of this study was to investigate how optical properties of aerosols change during cloud scavenging events at different temperatures. In Fig. 5 cloud scavenged fractions of scattering (a) and absorbing (b) material are presented as a function of temperature with boxplots. The boxes have a fixed width of 5°C ranging from -15°C to 20°C.

We can see from Fig. 5 that there is a slight dependency between temperature and scavenging efficiencies of both scattering and absorbing aerosols. Linear fits were calculated to estimate the connection. As Pitkänen et al. (2016) and Mikkonen et al (2019) showed, the errors in measured variables need to be taken properly into account in the linear fits. Thus, the slopes were calculated with Deming-regression (Deming, 1943). The slopes for scavenging efficiencies were calculated to be approximately 0.005 $°C^{-1}$ for scattering (Fig. 5a) and 0.003 $°C^{-1}$ for absorption (Fig. 5b). These correspond to approximately 5 percentage unit change for scattering and 3 percentage unit change for the absorbing material in the scavenged fraction at 10°C change in temperature, respectively. The efficiencies were observed to be 0.85 and 0.55 at 0°C for scattering and absorbing matter.

In both Fig. 5a and 5b a clear decrease in scavenging efficiency from 0°C to -15°C can be seen. In case of absorption, one explaining factor can be increasing amount of fresh BC emissions from local heating sources. Freshly emitted BC particles are known to be hydrophobic. However, since the effect is seen also in scattering data, it is likely that there are some effects that prevent cloud activation. For example, when there are mixed phase clouds, less numerous ice particles could cause more numerous liquid cloud droplets to evaporate due to lower saturation vapour pressure over ice. On the other hand, mixed phase clouds are not common in these temperatures. Also, it is possible that in colder conditions the radiative cooling from

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the cloud top is weaker and thus the negative buoyancy driving boundary layer mixing is also weaker. This would lead to lower updrafts at cloud base and thus limit the activation.

When considering temperatures above 0°C we can see a rather clear increase in the scavenging efficiency of scattering material. This could be explained by the increase of available water, increasing amount of hygroscopic material in particles e.g. from biogenic sources, and, general increase in particle size which can also likely be attributed to condensation of semi- and non-volatile biogenic compounds on pre-existing particles. Over the boreal region, biogenic compounds do have a significant effect on the aerosol load during the warm months and their emissions increase with increasing temperature (Liao, et al., 2014). Overall increase of biogenic, decrease of anthropogenic (BC) and aging of long-range BC increases scavenging efficiency.

Our results are qualitatively similar to the findings of Cozic et al. (2007) about the effects of temperature. At 0°C scavenging efficiency of black carbon was found to be 0.6 and to decrease towards colder temperatures. However, in our case the decrease of the scavenging efficiency is not as strong as the decrease reported by Cozic et al. (2007). In another study (Schneider et al. 2017) an average scavenging efficiency of 0.24 was observed over 14 campaigns with mean ambient temperatures varying from -3.0°C to 9.2°C. On the other hand, in each study the impactor for differentiating interstitial particles were different. In our study we used PM1 impactor while Cozic et al. (2007) and Schneider et al. (2017) used PM2.5 and PM5, respectively. These could slightly affect the results as higher cut-off could lead to smaller scavenging efficiency on average when smallest cloud particles might be counted as aerosol particles.

3.2.3 Impact of particle activation size

To obtain more information about the cloud activation potential of different sized particles in different weather conditions, we studied how the mean diameter D_{50} at which half of the particle population is activated as cloud droplets behaves at different conditions. Fig. 6 illustrates D_{50} as a function of temperature (panel a) and fraction of scavenged total volume (panel b). Additionally, a fraction of scavenged scattering (panel c) and absorption (panel d) are presented as a function of D_{50}. D_{50} has a local minimum at approximately 0°C (average) while more extreme values are present especially at low temperatures. The local minimum of D_{50} further supports the idea of BC being in relatively small particles which is seen as an increase in scavenging efficiency of absorbing matter in Fig. 5b.

Considering Fig. 6b we can see a kind of saturation of D_{50} value at low fractions of scavenged aerosol volume. This effect can also be seen in Fig. 4b and 4d. Excluding the saturated part, D_{50} value decreases rather linearly. This is logical; as more of the aerosol population volume is scavenged, the smaller and smaller particles get scavenged into the cloud droplets. Also the measurement setup with 1 µm cut-off in interstitial line affects the analysis. It is likely that within clouds a hygroscopic interstitial aerosol particle larger than 250 nm grows larger than 1 µm due to uptake of water. Hence the setup is not suitable to study such conditions. Similar effects have previously been observed for example in case of radiation fogs, where activation diameters as big as 364 – 450 nm has been observed (Hammer et al. 2014). Also in our station some of the observed clouds can actually be well developed fogs. In our case, however, only a small fraction of observations is in the range in which it is unclear if D_{50} is affected by the interstitial inlet and thus the statistics over long time period are not affected.

Scavenging efficiencies of scattering and absorbing material as a function of D_{50} support the idea that absorbing matter tends to be in relatively small sized particles and most of the scattering is caused by large particles as mentioned in 3.2.1. This can

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be seen as high scavenging efficiency of scattering material even at D_{50} of >300 nm while scavenging efficiency of absorbing material is low. In addition, only 70% of absorbing material is scavenged on average with D_{50} values higher than 50 nm. Although qualitatively the finding is similar to Motos et al. (2019) in the sense that more material is scavenged with low D50, which corresponds to high supersaturation. However, there is a clear difference as in our study some of the BC is too small to be scavenged. This can be due to both fresher and less CCN active aerosol emissions close to Puijo, or, lower supersaturations typical for low altitude clouds observed at Puijo station.

These observations of D_{50} offer a useful dataset for large scale aerosol modellers to evaluate the aerosol-cloud interactions in the models as they provide size dependent information about aerosol activation to cloud droplets. Data for such validation is scarce and for example aerosol-cloud interaction parameters from satellite observations are highly derived and include large uncertainties (e.g. Grosvenor et al., 2018). Especially, wet deposition of absorbing material in global scale models can be evaluated using this dataset. The methods used in this study could be applied at different in situ sites.

4 Conclusions

We investigated the size dependence of scattering and absorbing material in different conditions with a measurement setup which samples hydrated aerosol from two inlets. One inlet samples all sized aerosol droplets and the other samples particles in sizes which can be assumed to remain unactivated within a cloud (i.e. interstitial particle). The main findings can be summarized as:

1) The measurements showed that aerosol number concentrations were temperature dependent. The total number concentrations increased with temperature likely due to an increase in biogenic aerosol. Accumulation mode number concentrations, however, had a parabolic type dependence on temperature, with a minimum at approximately 0°C. The increase in the concentration with temperature is due to increase in biogenic aerosol, and the increase in sub-zero temperatures is caused by increase in local sources due to residential heating and inversion conditions limiting the vertical mixing.

2) We observed in our measurement setup that a fraction of scavenged scattering and absorbing material stays constant until up to 80% to 90% relative air humidity, above which their scavenged fraction increases when RH approaches 100%. For scattering aerosol, this increase in scavenged fraction starts at lower RH compared to absorbing material, indicating that higher fraction of absorbing material resides in smaller particles. However, on average 20% of absorbing material is found in particles larger than 1 μm in diameter.

3) In cloud cases the measurements of the scavenged fraction of scattering and absorbing material showed the same tendency of absorbing material to reside in the smaller particles. The scavenged fraction of scattering material was found to be high for the whole range of D_{50} values, while the scavenged fraction of absorbing material reached 70% only for the smallest D_{50} with diameter of approximately 50 nm. Noticeable but slight linear temperature dependence was observed for average scavenging efficiencies of both scattering and absorbing aerosol. At 0°C the efficiencies were 0.85 and 0.55 for scattering and absorbing matter, respectively. Slopes were approximately 0.005 $°C^{-1}$ and 0.003 $°C^{-1}$.

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The data presented is especially useful for evaluating the cloud scavenging parameterisations in atmospheric models as it provides information on size dependent cloud scavenging efficiency of both absorbing and scattering material. It also provides long term statistics on the concentration of CCN relevant particles at different temperatures.

Data availability

Data is available upon request from the authors.

Author contribution

AR, AL, SR, and MK conceived the study. AR, AL and SM carried out data analysis. AR prepared the manuscript with contributions from all co-authors.

Competing interests

The authors declare that they have no conflict of interest.

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References

Ahlm, L., Shakya, K. M., Russell, L. M., Schroder, J. C., Wong, J. P. S., Sjostedt, S. J., Hayden, K. L., Liggio, J., Wentzell, J. J. B., Wiebe, H. A., Mihele, C., Leaitch, W. R. and Macdonald, M.: Temperature-dependent accumulation mode particle and cloud nuclei concentrations from biogenic sources during WACS 2010, Atmos. Chem. Phys., 13, 3393-3407, 2013.

Anderson, T. L., Covert, D. S., Marshall, S. F., Laucks, M. L., Charlson, R. J., Waggoner, A. P., Ogren, J. A., Caldow, R., Holm, R. L., Quant, F. R., Sem, G. J., Wiedensohler, A., Ahlquist, N. A., and Bates, T. S.: Performance characteristics of a high-sensitivity, three-wavelength, total scatter/backscatter nephelometer, J. Atmos. Ocean. Technol., 13, 967–986, 1996.

Anderson, T. and Ogren, J.: Determining aerosol radiative properties using the TSI 3563 integrating nephelometer, Aerosol. Sci. Technol., 29, 57-69, 1998.

Anttila, T., Brus, D., Jaatinen, A., Hyvärinen, A. -., Kivekäs, N., Romakkaniemi, S., Komppula, M. and Lihavainen, H.: Relationships between particles, cloud condensation nuclei and cloud droplet activation during the third Pallas Cloud Experiment, Atmos. Chem. Phys., 12, 11435-11450, 2012.

Asmi, A., Collaud Coen, M., Ogren, J. A., Andrews, E., Sheridan, P., Jefferson, A., Weingartner, E., Baltensperger, U., Bukowiecki, N., Lihavainen, H., Kivekäs, N., Asmi, E., Aalto, P. P., Kulmala, M., Wiedensohler, A., Birmili, W., Hamed, A., O'Dowd, C., Jennings, S. G., Weller, R., Flentje, H., Fjaeraa, A. M., Fiebig, M., Myhre, C. L., Hallar, A. G., Swietlicki,

https://doi.org/10.5194/acp-2020-464
Preprint. Discussion started: 27 July 2020

E., Kristensson, A., and Laj., P.: Aerosol decadal trends – Part 2: In-situ aerosoconcentrations at GAW and ACTRIS stations, Atmos. Chem. Phys., 13, 895–916, 2013.

Asmi, E., Kondratyev, V., Brus, D., Laurila, T., Lihavainen, H., Backman, J., Vakkari, V., Aurela, M., Hatakka, J., Viisanen, Y., Uttal, T., Ivakhov, V. and Makshtas, A.: Aerosol size distribution seasonal characteristics measured in Tiksi, Russian Arctic, Atmos. Chem. Phys., 16, 1271-1287, 2016.

Bergstrom, R. W., Russell, P. B. and Hignett, P.: Wavelength dependence of the absorption of black carbon particles: Predictions and results from the TARFOX experiment and implications for the aerosol single scattering albedo. J. Atmos. Sci., 59(3), 567-577, doi:10.1175/1520-0469(2002)0592.0.CO;2, 2002.

Berkowitz, C. M., Berg, L. K., Yu, X.-Y., Alexander, M. L., Laskin, A., Zaveri, R. A., Jobson, B. T., Andrews, E., and Ogren, J. A.: The influence of fog and airmass history on aerosol optical, physical and chemical properties at Pt. Reyes National Seashore, Atmos. Environ., 45, 2559–2568, 2011.

Bond, T. C., Habib, G., and Bergstrom, R. W.: Limitations in the enhancement of visible light absorption due to mixing state, J. Geophys. Res., 111, D20211, doi:10.1029/2006JD007315, 2006.

Browse, J., Carslaw, K. S., Arnold, S. R., Pringle, K., and Boucher, O.: The scavenging processes controlling the seasonal cycle in Arctic sulphate and black carbon aerosol, Atmos. Chem. Phys., 12, 6775–6798, 2012.

Cappa, C. D., Onasch, T. B., Massoli, P., Worsnop, D. R., Bates, T. S., Cross, E. S., Davidovits, P., Hakala, J., Hayden, K. L., Jobson, B. T., Kolesar, K. R., Lack, D. A., Lerner, B. M., Li, S. M., Mellon, D., Nuaaman, I., Olfert, J. S., Petaja, T., Quinn, P. K., Song, C., Subramanian, R., Williams, E. J., and Zaveri, R. A.: Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon, Science, 337, 1078–1081, 2012.

Chaubey, J. P., Moorthy, K. K., Babu, S. S., Nair, V. S. and Tiwari, A.: Black carbon aerosols over coastal Antarctica and its scavenging by snow during the Southern Hemispheric summer, J. Geophys. Res. D Atmos., 115, 2010.

Collaud Coen, M., Andrews, E., Asmi, A., Baltensperger, U., Bukowiecki, N., Day, D., Fiebig, M., Fjaeraa, A. M., Flentje, H., Hyvärinen, A., Jefferson, A., Jennings, S. G., Kouvarakis, G., Lihavainen, H., Lund Myhre, C., Malm, W. C., Mihapopoulos, N., Molenar, J. V., O'Dowd, C., Ogren, J. A., Schichtel, B. A., Sheridan, P., Virkkula, A., Weingartner, E., Weller, R., and Laj, P.: Aerosol decadal trends – Part 1: In-situ optical measurements at GAW and IMPROVE stations, Atmos. Chem. Phys., 13, 869–894, doi:10.5194/acp-13-869-2013, 2013.

Cozic, J., Verheggen, B., Mertes, S., Connolly, P., Bower, K., Petzold, A., Baltensperger, U., and Weingartner, E.: Scavenging of black carbon in mixed phase clouds at the high alpine site Jungfraujoch, Atmos. Chem. Phys., 7, 1797–1807, doi:10.5194/acp-7-1797-2007, 2007.

Croft, B., Lohmann, U., Martin, R. V., Stier, P., Wurzler, S., Feichter, J., Posselt, R. and Ferrachat, S.: Aerosol size-dependent below-cloud scavenging by rain and snow in the ECHAM5-HAM, Atmos. Chem. Phys., 9, 4653-4675, 2009.

Deming, W. E.: Statistical adjustment of data. Wiley NY, 1943.

Fors, E. O., Swietlicki, E., Svenningsson, B., Kristensson, A., Frank, G. P., and Sporre, M.: Hygroscopic properties of the ambient aerosol in southern Sweden – a two year study, Atmos. Chem. Phys., 11, 8343–8361, doi:10.5194/acp-11-8343-2011, 2011.

Geron, C. D. and Arnts, R. R.: Seasonal monoterpene and sesquiterpene emissions from Pinus taeda and Pinus virginiana, Atmos. Environ., 44, 4240-4251, 2010.

https://doi.org/10.5194/acp-2020-464
Preprint. Discussion started: 27 July 2020

Atmospheric Chemistry and Physics Discussions

Grosvenor, D. P., Sourdeval, O., Zuidema, P., Ackerman, A., Alexandrov, M. D., Bennartz, R., et al.: Remote sensing of droplet number concentration in warm clouds: A review of the current state of knowledge and perspectives. Rev. Geophys., 56, 409– 453. https://doi.org/10.1029/2017RG000593, 2018.

Hammer, E., Gysel, M., Roberts, G. C., Elias, T., Hofer, J., Hoyle, C. R., Bukowiecki, N., Dupont, J.-C., Burnet, F., Baltensperger, U., and Weingartner, E.: Size-dependent particle activation properties in fog during the ParisFog 2012/13 field campaign, Atmos. Chem. Phys., 14, 10517–10533, https://doi.org/10.5194/acp-14-10517-2014, 2014.

Hao, L. Q., Kortelainen, A., Romakkaniemi, S., Portin, H., Jaatinen, A., Leskinen, A., Komppula, M., Miettinen, P., Sueper, D., Pajunoja, A., Smith, J. N., Lehtinen, K. E. J., Worsnop, D. R., Laaksonen, A. and Virtanen, A.: Atmospheric submicron aerosol composition and particulate organic nitrate formation in a boreal forestland–urban mixed region, Atmos. Chem. Phys., 14, 13483-13495, 2014.

Hussein, T., Karppinen, A., Kukkonen, J., Härkönen, J., Aalto, P. P., Hämeri, K., Kerminen, V. and Kulmala, M.: Meteorological dependence of size-fractionated number concentrations of urban aerosol particles, Atmos. Environ., 40, 1427-1440, https://doi.org/10.1016/j.atmosenv.2005.10.061, 2006.

Hyvärinen, A. -., Raatikainen, T., Komppula, M., Mielonen, T., Sundström, A. -., Brus, D., Panwar, T. S., Hooda, R. K., Sharma, V. P., De Leeuw, G. and Lihavainen, H.: Effect of the summer monsoon on aerosols at two measurement stations in Northern India-Part 2: Physical and optical properties, Atmos. Chem. Phys., 11, 8283-8294, 2011.

Komppula, M., Lihavainen, H., Kerminen, V.-M., Kulmala, M., and Viisanen, Y.: Measurements of cloud droplet activation of aerosol particle at a clean subarctic background site, J. Geophys. Res., 110, 06204, doi:10.1029/2004JD005200, 2005.

Laakso, L., Grönholm, T., Rannik, Ü, Kosmale, M., Fiedler, V., Vehkamäki, H., & Kulmala, M.: Ultrafine particle scavenging coefficients calculated from 6 years field measurements, Atmos. Environ., 37, 3605-3613, https://doi.org/10.1016/S1352-2310(03)00326-1, 2003.

Leaitch, W. R., Macdonald, A. M., Brickell, P. C., Liggio, J., Sjostedt, S. J., Vlasenko, A., Bottenheim, J. W., Huang, L., Li, S., Liu, P. S. K., Toom-Sauntry, D., Hayden, K. A., Sharma, S., Shantz, N. C., Wiebe, H. A., Zhang, W., Abbatt, J. P. D., Slowik, J. G., Chang, R. Y. -., Russell, L. M., Schwartz, R. E., Takahama, S., Jayne, J. T. and Ng, N. L.: Temperature response of the submicron organic aerosol from temperate forests, Atmos. Environ., 45, 6696-6704, 2011.

Liao, L., Kerminen, V.-M., Boy, M., Kulmala, M., and Dal Maso, M.: Temperature influence on the natural aerosol budget over boreal forests, Atmos. Chem. Phys., 14, 8295–8308, https://doi.org/10.5194/acp-14-8295-2014, 2014.

Lihavainen, H., Kerminen, V. -., Komppula, M., Hyvärinen, A. -., Laakia, J., Saarikoski, S., Makkonen, U., Kivekäs, N., Hillamo, R., Kulmala, M. and Viisanen, Y.: Measurements of the relation between aerosol properties and microphysics and chemistry of low level liquid water clouds in Northern Finland, Atmos. Chem. Phys., 8, 6925-6938, 2008.

Leskinen, A., Portin, H., Komppula, M., Miettinen, P., Arola, A., Lihavainen, H., Hatakka, J., Laaksonen, A., Lehtinen, K. E. J.: Overview of the research activities and results at Puijo semi-urban measurement station, Boreal Env. Res., 14, 576–590, 2009.

Leskinen, A., Arola, A., Komppula, M., Portin, H., Tiitta, P., Miettinen, P., Romakkaniemi, S., Laaksonen, A. and Lehtinen, K. E. J.: Seasonal cycle and source analyses of aerosol optical properties in a semi-urban environment at Puijo station in Eastern Finland, Atmospheric Chemistry and Physics, 12, 5647-5659, 2012.

https://doi.org/10.5194/acp-2020-464
Preprint. Discussion started: 27 July 2020

Atmospheric Chemistry and Physics Discussions

Liu, D., Allan, J., Whitehead, J., Young, D., Flynn, M., Coe, H., McFiggans, G., Fleming, Z. L., and Bandy, B.: Ambient black carbon particle hygroscopic properties controlled by mixing state and composition, Atmos. Chem. Phys., 13, 2015–2029, 2013.

Lohmann, U. and Feichter, J.: Global indirect aerosol effects: a review, Atmos. Chem. Phys., 5, 715–737, 2005.

Luoma, K., Virkkula, A., Aalto, P., Petäjä, T., and Kulmala, M.: Over a 10-year record of aerosol optical properties at SMEAR II, Atmos. Chem. Phys., 19, 11363–11382, https://doi.org/10.5194/acp-19-11363-2019, 2019.

Madhavi Latha, K., Badarinath, K. V. S. and Manikya Reddy, P.: Scavenging efficiency of rainfall on black carbon aerosols over an urban environment, Atmos. Sci. Let., 6, 148-151, 2005.

Marcq, S., Laj, P., Roger, J. C., Villani, P., Sellegri, K., Bonasoni, P., Marinoni, A., Cristofanelli, P., Verza, G. P. and Bergin, M.: Aerosol optical properties and radiative forcing in the high Himalaya based on measurements at the Nepal Climate Observatory-Pyramid site (5079 m a.s.l.), Atmos. Chem. Phys., 10, 5859-5872, 2010.

McMeeking, G. R., Good, N., Petters, M. D., McFiggans, G., and Coe, H.: Influences on the fraction of hydrophobic and hydrophilic black carbon in the atmosphere, Atmos. Chem. Phys., 11, 5099–5112, doi:10.5194/acp-11-5099-2011, 2011.

Mertes, S., Verheggen, B., Walter, S., Connolly, P., Ebert, M., Schneider, J., Bower, K. N., Cozic, J., Weinbruch, S., Baltensperger, U. and Weingartner, E.: Counterflow virtual impactor based collection of small ice particles in mixed-phase clouds for the physico-chemical characterization of tropospheric ice nuclei: Sampler description and first case study, Aerosol. Sci. Technol., 41, 848-864, 2007.

Mikkonen, S., Pitkänen, M. R. A., Nieminen, T., Lipponen, A., Isokääntä, S., Arola, A., and Lehtinen, K. E. J.: Technical note: Effects of uncertainties and number of data points on line fitting – a case study on new particle formation, Atmos. Chem. Phys., 19, 12531–12543, doi:10.5194/acp-19-12531-2019, 2019.

Motos, G., Schmale, J., Corbin, J. C., Modini, R. L., Karlen, N., Bertò, M., Baltensperger, U., & Gysel-Beer, M.: Cloud droplet activation properties and scavenged fraction of black carbon in liquid-phase clouds at the high-alpine research station Jungfraujoch (3580 m a.s.l.). Atmos.Chem.Phys., 19(6), 3833-3855. 10.5194/acp-19-3833-2019, 2019.

Noone, K. J., Ogren, J. A., Heintzenberg, J., Charlson, R. J. and Covert, D. S.: Design and calibration of a counterflow virtual impactor for sampling of atmospheric fog and cloud droplets, Aerosol. Sci. Technol., 8, 235-244, 1988.

Paasonen, P., Asmi, A., Petaja, T., Kajos, M. K., Aijala, M., Junninen, H., Holst, T., Abbatt, J. P. D., Arneth, A., Birmili, W., van, d. G., Hamed, A., Hoffer, A., Laakso, L., Laaksonen, A., Richard Leaitch, W., Plass-Dulmer, C., Pryor, S. C., Raisanen, P., Swietlicki, E., Wiedensohler, A., Worsnop, D. R., Kerminen, V. and Kulmala, M.: Warming-induced increase in aerosol number concentration likely to moderate climate change, Nat. Geosci., 6, 438-442, 2013.

Petzold, A. and Schönlinner, M.: Multi-angle absorption photometry – a new method for the measurement of aerosol light absorption and atmospheric black carbon, J. Aerosol Sci., 35, 421–441, 2004.

Pitkänen, M. R. A., Mikkonen, S., Lehtinen, K. E. J., Lipponen, A. and Arola, A.: Artificial bias typically neglected in comparisons of uncertain atmospheric data, Geophys. Res. Lett., 43, 10,003-10,011, 2016

Portin, H., Mielonen, T., Leskinen, A., Arola, A., Pärjälä, E., Romakkaniemi, S., Laaksonen, A., Lehtinen, K. E. J., and Komppula, M.: Biomass burning aerosols observed in Eastern Finland during the Russian wildfires in summer 2010 – Part 1: In-situ aerosol characterization, Atmos. Environ., 47, 269–278, 2012.

Portin, H., Leskinen, A., Hao, L., Kortelainen, A., Miettinen, P., Jaatinen, A., Laaksonen, A., Lehtinen, K. E. J., Romakkaniemi, S. and Komppula, M.: The effect of local sources on particle size and chemical composition and their role in aerosol–cloud interactions at Puijo measurement station, Atmos. Chem. Phys., 14, 6021-6034, 2014.

Romakkaniemi, S., Maalick, Z., Hellsten, A., Ruuskanen, A., Väisänen, O., Ahmad, I., Tonttila, J., Mikkonen, S., Komppula, M. and Kühn, T.: Aerosol–landscape–cloud interaction: signatures of topography effect on cloud droplet formation, Atmos. Chem. Phys., 17, 7955-7964, 2017.

Schneider, J., Mertes, S., van Pinxteren, D., Herrmann, H., and Borrmann, S.: Uptake of nitric acid, ammonia, and organics in orographic clouds: mass spectrometric analyses of droplet residual and interstitial aerosol particles, Atmos. Chem. Phys., 17, 1571–1593, https://doi.org/10.5194/acp-17-1571-2017, 2017

Swietlicki, E., Hansson, H. C., Hameri, K., Svenningsson, B., Massling, A., McFiggans, G., McMurry, P. H., Petaja, T., Tunved, P., Gysel, M., Topping, D., Weingartner, E., Baltensperger, U., Rissler, J., Wiedensohler, A., and Kulmala, M.: Hygroscopic properties of submicrometer atmospheric aerosol particles measured with H-TDMA instruments in various environments – a review, Tellus B, 60, 432–469, doi:10.1111/j.1600-0889.2008.00350.x, 2008.

Tarvainen, V., Hakola, H., Hellén, H., Bäck, J., Hari, P. and Kulmala, M.: Temperature and light dependence of the VOC emissions of Scots pine, Atmos. Chem. Phys., 5, 989-998, 2005.

Twohy, C. H., Strapp, J. W. and Wendisch, M.: Performance of a Counterflow Virtual Impactor in the NASA Icing Research Tunnel, J. Atmos. Oceanic Technol., 20, 781-790, 2003.

Verheggen, B., Cozic, J., Weingartner, E., Bower, K., Mertes, S., Connolly, P., Gallagher, M., Flynn, M., Choularton, T., and Baltensperger, U.: Aerosol partitioning between the interstitial and condensed phase in mixed-phase clouds, J. Geophys. Res., 112, D23202, doi:10.1029/2007JD008714, 2007.

Väisänen, O., Ruuskanen, A., Ylisirniö, A., Miettinen, P., Portin, H., Hao, L., Leskinen, A., Komppula, M., Romakkaniemi, S., Lehtinen, K. E. J. and Virtanen, A.: In-cloud measurements highlight the role of aerosol hygroscopicity in cloud droplet formation, Atmos. Chem. Phys., 16, 10385-10398, 2016.

Wang, J. M., Jeong, C., Zimmerman, N., Healy, R. M., Hilker, N. and Evans, G. J.: Real-World Emission of Particles from Vehicles: Volatility and the Effects of Ambient Temperature, Environ. Sci. Technol., 51, 4081-4090, 2017.

Weingartner, E., Nyeki, S., and Baltensperger, U.: Seasonal and diurnal variation of aerosol size distributions (10 < D < 750 nm) at a high-alpine site (Jungfraujoch 3580 m asl), J. Geophys. Res.: Atmos., 104, 26809–26820, 1999.

Yamagata, S., Kobayashi, D., Ohta, S., Murao, N., Shiobara, M., Wada, M., Yabuki, M., Konishi, H. and Yamanouchi, T.: Properties of aerosols and their wet deposition in the arctic spring during ASTAR2004 at Ny-Alesund, Svalbard, Atmos. Chem. Phys., 9, 261-270, 2009.

Yang, Y.; Fu, Y.; Lin, Q.; Jiang, F.; Lian, X.; Li, L.; Wang, Z.; Zhang, G.; Bi, X.; Wang, X.; Sheng, G.: Recent Advances in Quantifying Wet Scavenging Efficiency of Black Carbon Aerosol, Atmosphere, 10, 175, 2019.

Zhang, X., Huang, Y. and Rao, R.: Aerosol characteristics including fumigation effect under weak precipitation over the southeastern coast of China, J. Atmos. Sol. -Terr. Phys., 84-85, 25-36, 2012.

https://doi.org/10.5194/acp-2020-464
Preprint. Discussion started: 27 July 2020

Atmospheric Chemistry and Physics Discussions

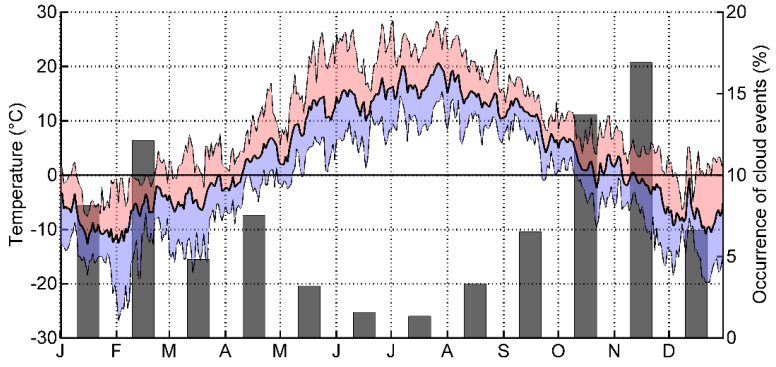

Figure 1: Average daily temperatures with minimum and maximum values over the studied period (left vertical axis) with occurred cloud events frequency (right vertical axis).

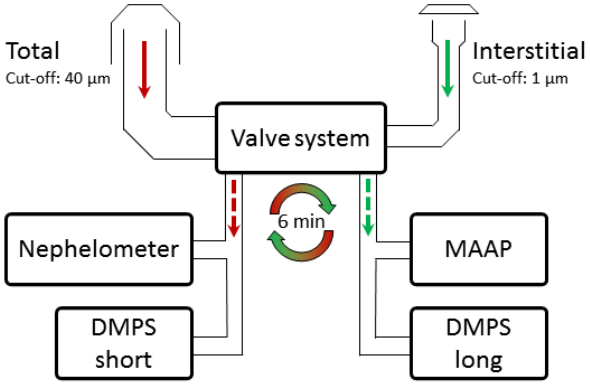

Figure 2: Schematic figure of the sampling line and valve system. Total (red) and interstitial (green) samples reach valve system, where the samples are directed to the devices. Valve system switches the lines between two stages: in stage one a pair of devices sample from total line and in stage two the same pair samples from interstitial line. Naturally the other pair of devices samples vice versa.

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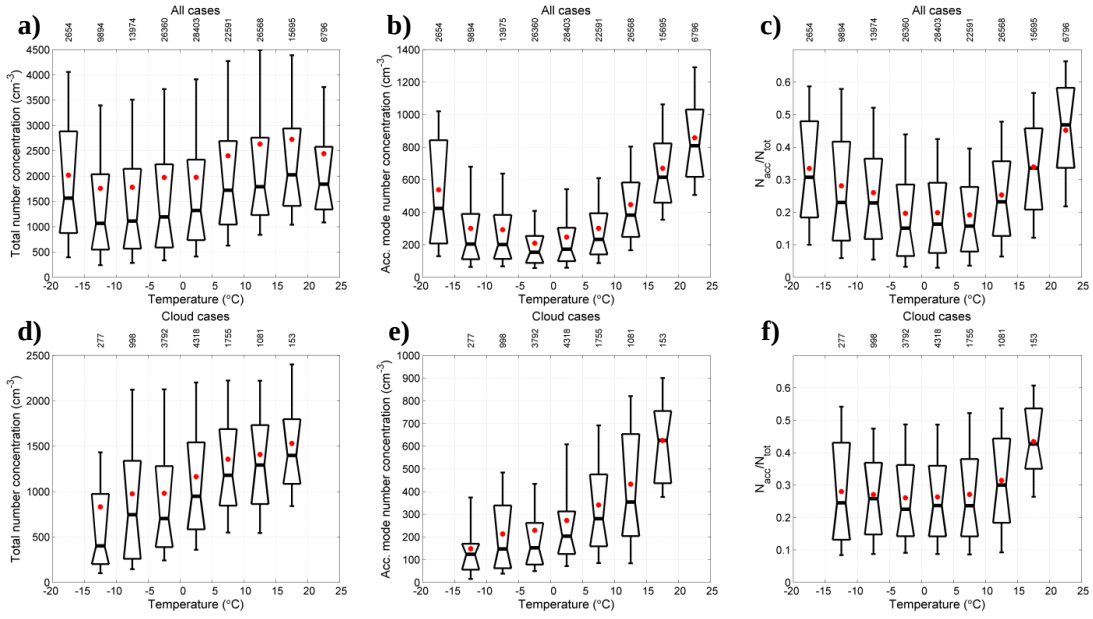

Figure 3: Box plots of (a) total number concentration, (b) accumulation mode number concentration, and (c) ratio of accumulation and total number concentrations in all measured cases. Similarly presented plots of (d) total number concentration, (e) accumulation mode number concentration, and (f) ratio of accumulation and total number concentrations in cloud cases. The boxes have 10th, 25th, 50th, 75th, and 90th percentile marked with black lines. Mean of each box is marked with red circle. Data points per box are presented above the boxes. All subfigures (a)-(f) are presented as a function of temperature.

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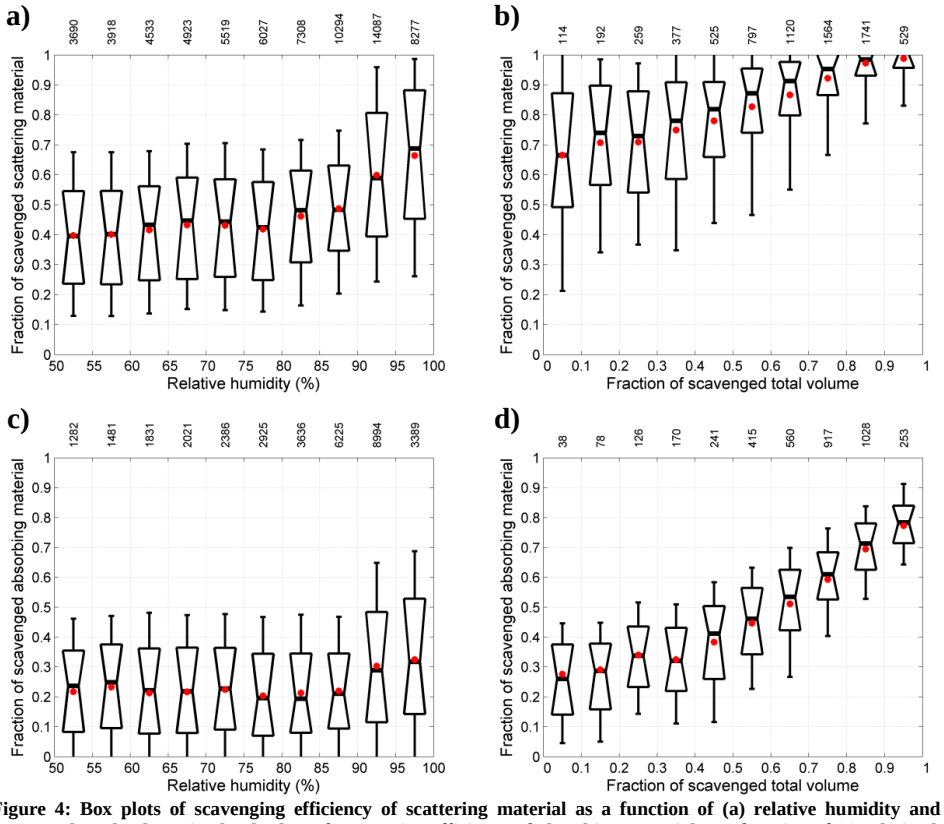

Figure 4: Box plots of scavenging efficiency of scattering material as a function of (a) relative humidity and (b) fraction of scavenged total volume in cloud. Plots of scavenging efficiency of absorbing material as a function of (c) relative humidity and (d) fraction of scavenged total volume in cloud. Boxes similarly as in Fig. 3.

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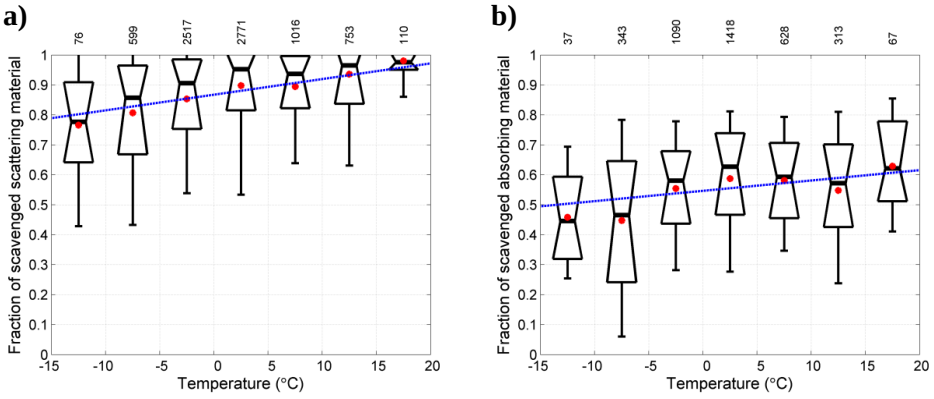

Figure 5: Box plots of in cloud (a) scavenging efficiency of scattering material, and, (b) scavenging efficiency of absorbing material as a function of temperature. Boxes similarly as in Fig. 3.

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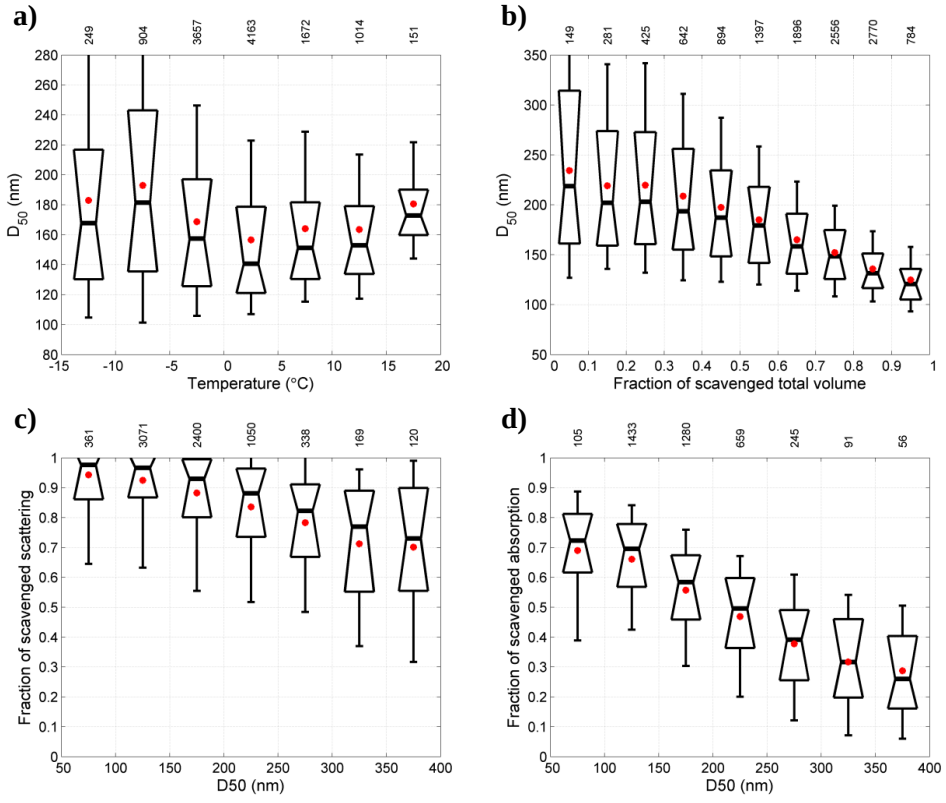

Figure 6: D_{50} as a function of (a) temperature and (b) fraction of scavenged total volume. Additionally fraction of scavenged scattering (c) and absorption (d) as a function of D_{50}. All plots during in cloud. Boxes similarly as in Fig. 3.