ON THE OPTICAL PROPERTIES OF AEROSOL PARTICLES OBSERVED AT MT. ÅRESKUTAN, SWEDEN

Therese Gadd

SUPERVISORS: DR. PAUL ZIEGER, DR. MATTHIAS TESCHE, PROF. JOHAN STRÖM

DEPARTMENT OF METEOROLOGY
STOCKHOLM UNIVERSITY
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Abstract

In summer and fall 2014, the field campaign CAEsAR (Cloud and Aerosol Experiment at Åre) was carried out in Åre, Sweden, to study clouds and aerosols at an intercept of different air masses. Various remote sensing and in-situ instruments were installed at the peak of and around Mt. Åreskutan. The overall goal of this study is to explore the relationship between detailed aerosol properties observed in-situ with remote sensing lidar instrument, as this is a first step in order to generalize aerosol properties over larger regions compared to single measuring points. The aerosol light extinction at Åre was found to be dominated by contribution from scattering and the largest amount of particles was transported to the in-situ station at wind directions between 90°-190°. The effect of humidity on the ambient aerosol was found to be important to consider when validating in-situ measurements with lidar retrievals. The ambient extinction coefficient measured in situ were in correspondence with the lidar retrieval at the altitude-level of the in-situ station. However, discrepancies were observed which could be due to an incomplete correction of the lidar overlap effect or site specific orography as a result of the location of the measurement site.
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1 Introduction

Our atmosphere is made up predominantly of gases, but the air we breathe also contains suspensions of liquid and solid matter in the form of particles. These particles, called aerosols, impact the Earth’s climate, both directly and indirectly, by scattering and absorbing solar radiation and by altering the properties of clouds (IPCC, 2013a). However, as they are often highly variable in space and time, and complex when it comes to shape and chemical composition, it is difficult to achieve a thorough quantification of the aerosol effect on the Earth’s global energy budget. In order to develop and evaluate numerical models which are ultimately needed in order to make prediction of the state of a future climate, precise measurements of aerosol and cloud properties are essential. In summer and fall 2014, the Cloud and Aerosol Experiment at Åre (CAEsAR 2014) has been carried out in Åre, Sweden (63° 23' 55" N, 13° 4' 54" E), to study aerosols and clouds at an intercept of different air masses. Various remote sensing and in-situ instruments were installed at the peak of and around Mt. Åreskutan.

The following research questions motivate this study

1. How are aerosol optical properties characterized at Åre?
2. How do they change with air mass/origin?
3. What is the influence of relative humidity on the aerosol light extinction coefficient?
4. Are observations of remote sensing and in-situ instruments in correspondence?

The overall goal was to explore the relationship between detailed aerosol properties observed in-situ with remote sensing lidar instrument, as this is a first step in order to generalize aerosol properties over larger regions compared to single measuring points. Understanding detailed processes are fundamental in the quest to scale up very detailed information for ultimate use in climate models.

1.1 What are aerosols?

Aerosols are defined as suspensions of small solid and/or liquid particles in a gaseous medium (here: air) (Seinfeld and Pandis, 2006). The size of the particles varies greatly; from a few nanometers to several hundreds of micrometers. They can be classified into four different size modes. The nucleation mode is made up of the smallest particles with a diameter $D < 10$ nm. The next bigger modes are the Aitken mode, $10$ nm $< D < 0.1$ µm, the accumulation mode, $0.1$ µm $< D < 2.5$ µm, and the coarse mode, $D > 2.5$ µm.
The particles in the nucleation mode are usually formed in-situ from gas-to-particle conversion (secondary particles) whereas coarse mode particles mainly originate from direct emissions caused by mechanical processes such as wind or erosion (primary particles) (Seinfeld and Pandis, 2006). Particles in the accumulation mode are primarily formed by coagulation of smaller particles or condensation of vapors onto existing particles. Aside from coagulation and condensation, changes in particle size and composition can also be caused by evaporation, chemical reactions or water uptake which eventually turns aerosols into cloud droplets.

Aerosols originate either from natural or from anthropogenic activities. Examples of natural primary sources are deserts, oceans and volcanic eruptions. The primary particles, including mineral and volcanic dust, sea salt and biogenic particles such as pollen, are usually coarse and low in number concentration (Wallace and Hobbs, 2006). Secondary natural particles are sulfates, organic compounds which originate from the biosphere and volcanoes.

The residence times of aerosol particles in the atmosphere varies from less than a day up to a few weeks, depending on the size of the particles and precipitation patterns (Wallace and Hobbs, 2006). The accumulation mode particles tend to have longer residence times compared to larger and smaller particles, since their removal mechanisms are less efficient. The main sink (e.g. removal processes) for particles in the accumulation mode is wet deposition. Coarse mode particles are mainly removed from the atmosphere by sedimentation as a result of their larger size and mass (Wallace and Hobbs, 2006).

Contrary to the naturally emitted particles, anthropogenically derived particles are usually smaller in size and higher in number concentration. They typically arise from fuel combustion, industrial processes and transportation sources such as automobiles (Seinfeld and Pandis, 2006). In urban and industrialized locations, the concentration of particulate air pollution can become so large that it causes severe health effects and increased mortality. As we breathe, particulate phase pollutants may enter our body and deposit in different parts of the lungs. Smaller pollutants can penetrate deeper into the lungs, even into the alveolus where the gas-exchange takes place. One example of a severe pollution event that had devastating consequences is the Great Smog of London in 1952 (Wallace and Hobbs, 2006).

Besides the impact on air quality and human health, aerosols also affect our climate, as outlined in Sec. 1.2.
1.2 How do aerosols affect Earth’s climate?

Since the start of the industrialization (~1750), humans have contributed to climate change through anthropogenic releases of greenhouse gases and aerosol particles (IPCC, 2013a). These atmospheric constituents alter the amount of incoming (shortwave) and outgoing (longwave) radiation and thus influence the Earth’s energy balance. The contribution from each agent is often given in terms of its radiative forcing (RF) in units watts per square metre (Wm\(^{-2}\)). RF is defined as the change in net energy flux at the tropopause or at the top of the atmosphere (TOA) caused by the introduction of a forcing agent (IPCC, 2013a). If the RF is positive, more energy is entering the tropopause (or TOA) compared to what is exiting. Thus, a positive RF tends to have a warming effect on the global mean surface temperature whereas a negative RF indicates a surface cooling.

Figure 1 shows a summary of the main drivers of climate change today (2011) with corresponding RF estimates relative to preindustrial conditions (1750). The total anthropogenic RF is positive (warming effect) with carbon dioxide (CO\(_2\)) as the largest contributor. As indicated by Fig. 1, the effect of aerosols and their precursors on climate change can be grouped into two processes: aerosol-radiation interactions and aerosol-cloud interactions.

Firstly, aerosols directly interact with radiation though scattering and absorption (see Sec. 2.1). Aerosol scattering of solar radiation generally increases the planetary albedo which tends to cool the climate system whereas absorption has the opposite effect and tends to warm the climate (IPCC, 2013a). The main factors that determine the sign of the forcing is the size of the particles, their chemical composition and the surface properties below the aerosol layer.

A subset of aerosol particles also serve as centers upon which cloud droplets and ice particles can form (IPCC, 2013a). If a liquid phase cloud is influenced by an increase in aerosol number concentration it tends to have more but smaller droplets (assuming a fixed water content). More droplets with smaller diameters make the cloud brighter, and thus, more reflective. This leads to a cooling effect. Beside making the cloud brighter, more and smaller droplets in a liquid cloud tend to increase cloud lifetime by reducing the precipitation efficiency (Albrecht, 1989).
Aerosol-radiation interactions may also cause changes in cloud properties. Absorption due to aerosols such as black carbon (BC), causes heating in the aerosol layer by the release of thermal radiation. The increased temperature may lead to evaporation of cloud droplets and, with that, a decrease in cloud cover. Additionally, the heating may change the stability of the boundary layer and the free troposphere which in turn can prohibit convection. Together with the life-time effect, these cloud changes are referred to as rapid adjustments. Rapid adjustments are separated from climate feedbacks, since they are not dependent on changes in surface temperature, and taken into account in the RF estimates due to cloud-aerosol interactions (IPCC, 2013a).

Overall, the total RF from aerosol changes is negative with an effect of -0.9 [-1.9 to 0.1] Wm$^{-2}$ (values in brackets correspond to the 5 to 95% uncertainty range) (IPCC, 2013a). As a result, aerosols masks some of the positive global mean forcing caused by well-mixed greenhouse gases. However, they are associated with the largest uncertainty when it comes to the total RF estimate. The uncertainty is mainly due to their high spatial and temporal variability together with a short observational record, difficulties when it comes to measurements and the lack of observations of relevant parameters (IPCC, 2013a).
2 The physical basis

2.1 Interaction of particles with light

When radiation passes through the atmosphere it is attenuated through scattering and absorption by aerosols, gas molecules, cloud and rain droplets, or ice crystals (Wallace and Hobbs, 2006). Briefly, the electromagnetic wave hits the particle and excites its internal degrees of freedom, causing the particle to spontaneously undergo a transition to a higher energetic state. The excited charges may re-emit the energy (scattering) or partly convert it to thermal energy (absorption) (Wallace and Hobbs, 2006). If the wavelength of the scattered radiation is the same as the incident radiation, the process is referred to as elastic. Contrary, a scattering process in which the wavelength changes is referred to as inelastic. Most atmospheric scattering are a result of elastic processes (Wandinger, 2005).

The combined effect of scattering and absorption is called extinction. The Beer-Bouger-Lambert law describes the exponential attenuation of light through a certain medium (here: air) and is defined as

\[ I_t(s, \lambda) = I_0(\lambda) e^{-\int_0^s \sigma_{\text{ext}}(\lambda) ds} \]  

where \( I_0 \) and \( I_t \) are the spectral radiance of the incident and transmitted light, respectively, \( s \) is the length of the attenuating medium, \( \sigma_{\text{ext}} \) is the so-called extinction coefficient and \( \lambda \) is the wavelength.\(^1\) (Gumbel et al., 2009).

For a population of monodisperse, spherical, particles, the extinction coefficient is given as

\[ \sigma_{\text{ext}} = Q_{\text{ext}} \cdot A \cdot N \]  

where \( A \) is the cross-sectional area of the individual particles (in units of \( m^2 \)), \( N \) is the number density of the medium (in units of \( m^{-3} \)) and \( Q_{\text{ext}} \) is the dimensionless extinction efficiency. The product \( Q_{\text{ext}} \cdot A \) is the single-particle extinction cross-section, \( C_{\text{ext}} \) (in units of \( m^2 \)) (Sienfield and Pandis, 2006).

To obtain \( \sigma_{\text{ext}} \) for a population of different sized particles, Eq. (2) has to be integrated over the entire size spectrum,

\[ \sigma_{\text{ext}} = \int_{D_{\text{min}}}^{D_{\text{max}}} C_{\text{ext}} \frac{dN}{dD} dD \]  

where \( D_{\text{max}} \) and \( D_{\text{min}} \) represents the upper and lower particle diameter, respectively.

\(^1\)For simplicity, the wavelength denotation will be omitted from here on. However, it is important to keep in mind that all optical properties are wavelength dependent.
Due to conservation of energy \( C_{\text{ext}} \) can be described as the sum of the scattering and the absorption cross section:

\[
C_{\text{ext}} = C_{\text{sca}} + C_{\text{abs}}
\]  

(4)

with similar relations to the dimensionless extinction efficiency \( Q_{\text{ext}} = Q_{\text{scat}} + Q_{\text{abs}} \) and the extinction coefficient \( \sigma_{\text{ext}} = \sigma_{\text{scat}} + \sigma_{\text{abs}} \).

Also, from an atmospheric point of view, \( \sigma_{\text{scat}} \) and \( \sigma_{\text{abs}} \) can be decomposed into contributions from molecules \( (\sigma_{\text{sg}} \text{ or } \sigma_{\text{ag}}) \) or particulate matter \( (\sigma_{\text{sp}} \text{ or } \sigma_{\text{ap}}) \):

\[
\sigma_{\text{scat}} = \sigma_{\text{sg}} + \sigma_{\text{sp}}
\]

(5)

\[
\sigma_{\text{abs}} = \sigma_{\text{ag}} + \sigma_{\text{ap}}
\]

(6)

From this we define the extinction coefficient caused by aerosol particles only (the particle light extinction coefficient):

\[
\sigma_{\text{ep}} = \sigma_{\text{ap}} + \sigma_{\text{sp}}.
\]

(7)

The incident wavelength (or frequency), the particle’s size and the chemical composition of the particle are the main parameters that govern the scattering and absorption of light by a particle. The size of a spherical particle is usually expressed in its relation to the incident wavelength by the dimensionless size parameter \( x \) as

\[
x = \frac{2\pi r}{\lambda}
\]

(8)

where \( r \) is the radius of the particle and \( \lambda \) is the wavelength of the incident radiation, and the chemical composition of the particle is defined by the complex refractive index \( m \) as

\[
m = m_r + im_i
\]

(9)

where \( m_r \) and \( m_i \) denotes the scattering and absorbing component of the particle (referenced to that of air), respectively (Seinfeld and Pandis, 2006).

Another important property is the angular scattering phase function, \( P(\theta) \), which describes the angular dependency of light scattered by a particle. It is defined as

\[
P(\theta) = \frac{I(\theta, x, m)}{\int_0^\pi I(\theta, x, m) \sin(\theta) d\theta}
\]

(10)

where \( I(\theta, m, x) \) is the scattered light intensity at a scattering (zenith) angle \( \theta \) relative to the incident light beam (Gumbel et al., 2009).
Figure 2 shows the normalized phase function for four different sized particles in polar coordinates. If the spherical particles are small compared to the incident wavelength (i.e. \( x << 1 \)), the phase function is symmetric in the forward and backward direction (Rayleigh scattering). However, as the particle increases in size (\( x >> 1 \)), the light is scattered more in the forward scattering lobes (Mie scattering).

The fraction of backscattered light to the total scattered light is given by the backscattered ratio \( b \) as

\[
b = \frac{\int_{\pi/2}^{\pi} P(\theta) \sin(\theta) d\theta}{\int_{0}^{\pi} P(\theta) \sin(\theta) d\theta}
\]  

and can be calculated as the ratio of the backscattered coefficient (\( \sigma_{bsp} \)) to the total scattering coefficient (\( \sigma_{sp} \)):

\[
b = \frac{\sigma_{bsp}}{\sigma_{sp}}
\]

The wavelength dependence of the aerosol extinction coefficient can be expressed as a power law function:

\[
\sigma_{ep} = \beta \lambda^{-\alpha}
\]

where \( \beta \) is a concentration dependent constant and \( \alpha \) the Ångström exponent (analogously for \( \sigma_{sp} \) and \( \sigma_{ap} \)) (Sienfield and Pandis, 2006).

The Ångström exponent can be calculated using measurements of the extinction coefficient at two discrete wavelengths,

\[
\alpha \approx -\frac{\log(\sigma_{ep}(\lambda_1)/\sigma_{ep}(\lambda_2))}{\log(\lambda_1/\lambda_2)}
\]

or by fitting a power law function to the extinction coefficient measured at various wavelengths. For small exponents (\( \alpha < 1 \)), the measured aerosol is dominated by coarse
mode particles (diameter \( D \gtrsim 1 \mu m \)) whereas a large exponent (\( \alpha > 2 \)) points toward a size distribution dominated by fine mode particles (\( D \lesssim 1 \mu m \)).

Another useful quantity is the aerosol single scattering albedo \( \omega_0 \), which describes the relative importance of scattering and absorption to the total light extinction as

\[
\omega_0 = \frac{\sigma_{sp}}{\sigma_{sp} + \sigma_{ap}} = \frac{\sigma_{sp}}{\sigma_{ep}}.
\]

Values of the single scattering albedo vary from 1 (extinction is entirely caused by scattering) to below 0.5 for strongly absorbing particles (Wallace and Hobbs, 2006). A value of \( \omega_0 = 0 \) represents the unrealistic case that all light extinction is due to absorption.

### 2.3 Relative humidity effect on aerosol light scattering

As mentioned in Sec. 1.1, aerosol particles can grow by condensation of water vapor or shrink by evaporation. The ability of an aerosol particle to take up water (hygroscopicity) depends on its size, chemical composition and the ambient relative humidity (RH):

\[
RH = \frac{p_{H_2O}(T)}{p_{H_2O}^0(T)}
\]

where \( p_{H_2O} \) is the partial pressure over water, \( p_{H_2O}^0 \) is the saturation water vapor pressure and \( T \) is the temperature (Seinfeld and Pandis, 2006).

Since aerosol light scattering is highly dependent on the size of the particles, the change in size due to an increase or decrease in \( RH \) may have a strong impact on the amount of scattered radiation. The ratio between the wet to dry particle scattering, or the scattering enhancement, \( f(RH) \), is a key parameter that describes the \( RH \)-dependency of the aerosol light scattering:

\[
f(RH) = \frac{\sigma_{sp}(RH_{amb})}{\sigma_{sp}(RH_{dry})}.
\]

If the contribution from light absorption is small, Eq. 17 can be approximated by:

\[
f(RH) \approx \frac{\sigma_{ep}(RH_{amb})}{\sigma_{ep}(RH_{dry})}.
\]

Due to the dominance of light scattering at Åre, \( f(RH) \) will be referred to as the light extinction enhancement factor from here on.

An increase in particle size due to hygroscopic growth (water uptake of aerosol particles) also reduces the visibility. The *Koschmeider equation* relates the extinction coefficient
with the visual range, $x_v$ (Seinfeld and Pandis, 2006):

$$x_v = \frac{3.912}{\sigma_{ext}}. \quad (19)$$
3 Experimental setup

3.1 Site and campaign description

The Cloud and Aerosol Experiment at Åre (CAEsAR) took place from 25 June to 10 October, 2014 in Åre (63° 23’ 55’’ N, 13° 4’ 54’’ E), Sweden (see also campaign website: http://www.aces.su.se/itm/page.php?pid=1126). A range of remote-sensing and in-situ instruments were installed in the Åre valley, 420 m above sea level (a.s.l.), and at the research station located on Mt. Åreskutan (1250 m a.s.l). Figure 3 gives an overview of the distribution of the main measurement sites set up for CAEsAR 2014. Detailed views of the research cabin on Mt. Åreskutan and the remote-sensing site at the valley station of the cable-car line are presented in Fig. 4. For the in-situ retrievals used in this study, air was sampled through a custom-built total inlet and measurements were performed under dry conditions ($RH < 30\%$). No specific size cut was used. In addition to the in-situ instruments that will be presented in the following sections, a weather station measured meteorological parameters (e.g. $T$, $RH$ and wind direction).

![Figure 3: Measurement set-up during the CAEsAR 2014 campaign performed in Åre, Sweden. Shown are the different measurement sites which data is used within this study. Figure reproduced from Google Earth.](image-url)
3.2 In-situ instrumentation at Mt. Åreskutan

3.2.1 Light scattering coefficient at dry conditions

The aerosol light scattering, and backscattering coefficient was measured with an integrating nephelometer (model Aurora 3000, ECOTECH Pty Ltd, Knoxfield, Australia). A detailed description of the instrument is given by Müller et al. (2011a). Briefly, gaseous and particulate components are brought from the inlet to the nephelometer. Inside the instrument, the sample volume is illuminated by a LED light source that emits short light pulses at the wavelengths of $\lambda = 450$ (blue), 525 (green) and 635 nm (red), one at a time. Light is scattered onto a photomultiplier tube (PMT) which produces electrical signals which are proportional to the scattering coefficient $\sigma_{\text{scat}}$ of the sample air. In the backscatter mode, a shutter blocks the light scattered in the forward direction so that the PMT only detects the backscattered light, $\sigma_{\text{bscat}}$. Zero calibrations are performed where the Rayleigh scattering component $\sigma_{\text{sg}}$ of $\sigma_{\text{scat}}$ and $\sigma_{\text{bscat}}$ is subtracted. During this so-called zero adjust, only particle free air is being sampled. Additionally, span calibrations are made using CO$_2$ as a reference gas, with known optical properties.
Due to instrumental restrictions, the nephelometer only measures at angles between $10^\circ-171^\circ$ ($90^\circ-171^\circ$ for backscatter). In order to correct for angular and illumination non-idealities a correction factor $C$ was introduced by Andersson and Ogren (1998):

$$ C = \frac{\sigma_{\text{true}}}{\sigma_{\text{Neph}}} \quad (20) $$

where $\sigma_{\text{true}}$ represents the true particle light scattering coefficient and $\sigma_{\text{Neph}}$ the scattering coefficient measured by the nephelometer within the scattering angles of $10^\circ-171^\circ$ or $90^\circ-171^\circ$.

Müller et al. (2011a) provide parameterized values of the correction factor for total scatter and backscattering. For total scatter, the correction factor ($C_{sp}$) is obtained using the linear relationship:

$$ C_{sp} = a + b \cdot \alpha_{sp} \quad (21) $$

where $\alpha_{sp}$ is the dimensionless total scattering Ångström exponent derived from three different wavelength pairs (450/525 nm for correction at 450 nm, 450/635 nm for correction at 525 nm and 525/635 nm for correction at 635 nm, respectively) using the uncorrected nephelometer scattering coefficients. Values of $a$ and $b$ are listed in Tab. 1 together with the correction factors for the backscatter coefficients $C_{bsp}$. Values of $C_{bsp}$ are derived from calculations based on Mie theory (Bohren and Huffman, 1983) since no correlation between $C_{bsp}$ and the scattering Ångström exponent has been found (Andersson and Ogren, 1998). This method is more robust since it requires knowledge on particle number size distributions and complex refractive indices.

Table 1: Correction factors for total scatter, as a linear function of the Ångström exponent (see Eq. 13), $C_{ts} = a + b \cdot \alpha_{sp}$, and backscatter ($C_{bsp}$) (values from Müller et al. (2011a))

<table>
<thead>
<tr>
<th>$\lambda$</th>
<th>correction factors</th>
<th>$a$</th>
<th>$b$</th>
<th>$C_{bsp}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>450 nm</td>
<td></td>
<td>1.455</td>
<td>-0.189</td>
<td>0.936</td>
</tr>
<tr>
<td>525 nm</td>
<td></td>
<td>1.434</td>
<td>-0.176</td>
<td>0.971</td>
</tr>
<tr>
<td>635 nm</td>
<td></td>
<td>1.403</td>
<td>-0.156</td>
<td>0.968</td>
</tr>
</tbody>
</table>
3.2.2 Measurements of extinction coefficient at ambient conditions

The extinction coefficients at ambient conditions were obtained by using the data of a visibility sensor (model AEVIS 6500). The instrument measures the extinction coefficient as light is projected by a LED transmitter at a wavelength $\lambda = 880$ nm into a sample volume and provides the visibility as output. The measured visibility was recalculated to extinction using the Koschmeider equation (Eq. 19). The contribution from molecular (Rayleigh) scattering ($\sigma_{sg}$) was removed so that only the extinction caused by aerosol particles remained. Calculations of $\sigma_{sg}$ were made according to Bodhaine et al. (1999).

3.2.3 Measurement of the aerosol absorption coefficient

The absorption coefficients were measured with a multi-angle absorption photometer (MAAP, model 5012) and an aethalometer (Model AE30). The instruments were used synergistically to exploit their different advantages; the MAAP has higher precision but only measures at a single wavelength whereas the aethalometer operates at several wavelengths.

The MAAP measures the attenuated and backscattered light from a particle-loaded fibre filter at $\lambda = 637$ nm (Müller et al., 2011b). By applying a radiative transfer scheme, the fraction of light absorbed by the deposited particles is obtained (Petzold and Schönlinner, 2004). The measured absorption coefficients are converted to black carbon mass concentration by the instrument using a specific absorption coefficient $Q_{BC}$. By multiplying the measured BC concentration with the instrumental set value $Q_{BC} = 6.6 \text{ m}^2\text{ g}^{-1}$ the absorption coefficient was obtained.

The aethalometer measures the attenuation of light as it passes through a quartz fiber filter in which light-absorbing particles (black carbon) are loaded over time (Weingartner et al., 2003). It operates at seven wavelengths ($\lambda = 370, 470, 520, 590, 660, 880, 950$ nm) and provides the concentration of black carbon ($M_{BC}$) as output. From this, the absorption coefficient was calculated in accordance with Weingartner et al. (2003) as

$$\sigma_{ap} = \frac{Q_{BC}}{C \cdot R(ATN)} M_{BC}$$

(22)

where $C$ and $R(ATN)$ are two empirical calibration factors correcting for filter artifacts. The constant $C$ (accounting for multiple scattering within the filter and the deposited particles) was assumed to be 2 which is typical for low absorbing continental aerosol (Collaud et al., 2010). $R(ATN)$ (accounting of shadowing effects as the particles deposited on the filter may overlap) was assumed to be unity. It should be noted that the choice of these empirical correction factors is not critical within this work since only the Ångström
exponent of the aethalometer will be used (see Sec. 3.4). The instrumental set values \( Q_{BC} = 39.5 \) (\( \lambda = 370 \text{ nm} \)), 31.1 (\( \lambda = 470 \text{ nm} \)), 28.1 (\( \lambda = 520 \text{ nm} \)), 24.8 (\( \lambda = 590 \text{ nm} \)), 22.2 (\( \lambda = 660 \text{ nm} \)), 16.6 (\( \lambda = 880 \text{ nm} \)) and 15.4 (\( \lambda = 950 \text{ nm} \)) were used.

3.3 Lidar

The compact aerosol Raman lidar (light detection and ranging) Polly of the Leibniz Institute for Tropospheric Research in Leipzig, Germany, was installed at the remote sensing site for the time period of the campaign. Briefly, the instrument emits lasers pulse at 532 nm wavelength into the atmosphere and detects the elastically (by particles and air molecules at 532 nm) and Raman (by nitrogen molecules at 607 nm) backscattered light with a two-channel receiver. This enables the retrieval of particle extinction and backscatter coefficient profiles independent of each other during nighttime (Althausen et al., 2009). However, during daytime, the bright daylight background interferes with the measurements of Raman-scattered light. Hence, only the elastically backscattered signal can be used to determine particle backscatter coefficient profiles which is made according to Klett’s method (Althausen et al., 2009; Klett, 1981). A detailed description of the instrument is given by Tesche et al. (2007) and Althausen et al. (2009). The extinction profiles used in this study were obtained using Klett’s method and a lidar ratio (LR) of 50 sr. This is a reasonable assumption as it represents a value which is common for continental aerosols (Müller et al., 2007).

3.4 Data processing

The data of all in-situ instruments use in this work was averaged into 5 min values. Only cloud-free measurements were used (\( RH < 95 \% \)) to avoid biases due to scavenging of aerosol particles and possibly precipitation. Also, the inlet was not characterized for cloud periods due to deposition of cloud droplets onto the inlet. The extinction coefficient and the aerosol intensive properties (\( \omega_0 \), \( b \) and \( \alpha \)) were calculated from the measured scattering, backscattering and/or absorption coefficients using Eq. 7, 15, 12 and 13, respectively.

All aerosol optical properties will be presented and discussed for the three nephelometer wavelengths. The absorption coefficients therefore have to be recalculated to fit those wavelengths using the Ångström law (in analogy to Eq. 14)

\[
\sigma_{ap}(\lambda) = \sigma_{ap}(637 \text{ nm}) \left( \frac{\lambda}{637 \text{ nm}} \right)^{-\alpha_{ap}}
\]

where \( \alpha_{ap} \) was derived by fitting a power law function to the the absorption coefficients measured at various wavelengths by the aethalometer (in analogy to Eq. 13). The same
procedure was used to transform the scattering coefficient (and absorption coefficient) to the wavelengths of the lidar (532 nm) and visibility sensor (880 nm). As mentioned before, the MAAP was preferably used to provide the particle light absorption coefficient due to its higher precision.

The extinction enhancement factor was calculated using the extinction coefficient from the visibility sensor and the dry extinction coefficient at 880 nm (in analogy to Eq. 18):

$$f(RH) = \frac{\sigma_{ep}(RH, 880 \text{ nm})}{\sigma_{ep}^{\text{dry}}, 880 \text{ nm}}.$$  \hspace{1cm} (24)

In order to compare the lidar measurement to the in-situ measurement, the extinction coefficient measured in-situ was recalculated to ambient conditions as

$$\sigma_{ep}^{\text{amb}} = f(RH) \left( \sigma_{sp}^{\text{dry}} + \sigma_{ap}^{\text{dry}} \right),$$  \hspace{1cm} (25)

where $f(RH)$ was determined by fitting 5-min averaged humidograms ($f(RH)$ versus $RH$) with an empirical two-parameter equation:

$$f(RH) = a(1 - RH)^{-\gamma}$$  \hspace{1cm} (26)

where $a$ is the intercept and $\gamma$ is the so called hygroscopicity parameter.
4 Results and discussion

4.1 Characterization of aerosol optical properties

The top panel in Fig. 5 shows an overview of the measured scattering ($\sigma_{sp}$) and absorption ($\sigma_{ap}$) coefficients at 525 and 637 nm, respectively. The data is given as 5-min averages at standard temperature and pressure (STP; $T_{\text{standard}} = 273.15$ K and $P_{\text{standard}} = 1013.25$ hPa). All timescales are Coordinated Universal Time (UTC). A significant increase in light scattering and absorption was observed during the first week of August. The elevated $\sigma_{ap}$ measured by the MAAP implied that the air surrounding the site contained a substantial amount of absorbing particles. This period is presented in more detail in the bottom panel of Fig. 5. Intermediate periods of low $\sigma_{sp}$ and $\sigma_{ap}$ are explained by clean air masses and/or wash-out due to clouds and precipitation.

The possibility that the measurements during the first week of August was influenced by the massive wildfire that broke out in the forests of Västmanland, Sweden, on July 31, 2014, was investigated using air mass trajectories calculated from the NOAA (National Oceanic and Atmospheric Administration) HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model (http://ready.arl.noaa.gov/HYSPLIT.php; 10-day backward calculations with a time resolution of 1 hour). The 2014 Västmanland wildfire is the greatest fire in Swedish modern history (see e.g., http://lup.lub.lu.se/student-papers/record/7471628).

Figure 6 shows the measured extinction coefficient ($\sigma_{ep}$) versus the scattering Ångström exponent ($\alpha_{sp}$). The dots are color coded according to time. In the side panels, typical air mass trajectories responsible for distinct patterns are shown. Generally, $\sigma_{ep}$ increased with increasing $\alpha_{sp}$. Thus, when the amount of particles was higher, the particles were smaller in size and vice versa. As mentioned in Sec. 1.1, the former feature is typical for anthropogenic particles and the latter generally describes particles of natural origin.

Like expected, the model results implied that air reaching the in-situ site around August 4, 2014 (see Fig. 6d) had been advected over areas affected by the wildfire. Hence, the air probably picked up large amount of elemental and organic particulate matter that had been released into the air as a result of the fire. This probably explains the peak observed in Fig. 5 around August 4, 2014. On August 3, 2014, air was instead advected over Great Britain before reaching the in-situ site (Fig. 6b). By passing over populated areas the air probably picked up increased amounts of anthropogenic aerosol (e.g. organic material).
The features observed as (b) and (d) in Fig. 6 were likely linked to the pollution and forest fire plume. The separation between the two are indicated by the greater $\alpha_{sp}$ found at (b) which points towards a domination of finer mode particles and, thus, a more anthropogenic influence. Particles found at (a) and (c) were less numerous, which is an indication of cleaner air masses.

Figure 5: Time series of 5-min averaged values of the light scattering ($\sigma_{sp}$; blue) and absorption coefficient ($\sigma_{ap}$; red) measured during the entire campaign (top panel) and the week of the 2014 Västmanland wildfire (bottom panel), see text for details.
In order to investigate how the optical properties changed with air mass/origin, they were separated according to wind direction. Certain sectors were more common than others, which could influence the interpretation. In particular two main wind directions could be observed, as is clearly depicted in Fig. 7. A detailed view of the measured optical properties with respect to wind direction in intervals of 10° is given in Fig. 8. The extensive parameters (left panel) exhibited similar variations with lower magnitude at directions between 20°-70° and 260°-360°, and higher at 90°-190°. The higher values observed in the latter sector points toward a continental (polluted) influence on these air-masses and a cleaner influence in the two former sectors. The intensive parameters (right panel) showed less variation, although a slight tendency of finer particles being transported from the continental sector was observed. One indication is the increase of the scattering Ångström exponent and the decrease in the single-scattering albedo. The backscatter ratio showed a tendency of being higher at directions 20-70°, which agrees well with the findings showed in Fig. 6 with finer particles being advected from the Arctic region (Fig. 6b).
Figure 7: Number of data points versus wind direction at ambient $RH < 95\%$ (blue line) and ambient $RH > 95\%$ (red line), respectively. Each point represents the average number of points in wind sectors of $10^\circ$.

Figure 8: Median and percentile values of (a) the aerosol scattering coefficient ($\sigma_{sp}$), (b) backscattering coefficient ($\sigma_{bsp}$), (c) absorption coefficient ($\sigma_{ap}$), (d) scattering angström exponent ($\alpha_{sp}$), (e) single scattering-albedo ($\omega_0$) and (f) backscatter ratio ($b$) versus wind direction (at dry conditions, no clouds). Each point represents the average parameter in wind sectors of $10^\circ$. 
Table 2 shows the statistics of the measured parameters, including means, standard deviations (SD) and percentile values (5, 25, 50, 75 and 95). The mean ambient temperature and relative humidity during the campaign were 8 ± 5 °C and 85 ± 16 % (mean ± SD), respectively. The $\sigma_{sp}$ (525 nm), $\sigma_{bsp}$ (525 nm) and $\sigma_{ap}$ (637 nm) were 19.4 ± 21.5 Mm$^{-1}$, 3.4 ± 2.6 Mm$^{-1}$ and 0.6 ± 0.8 Mm$^{-1}$, respectively. Generally, the aerosol light absorption coefficient was very low compared to the scattering coefficient; indicated by the high single-scattering albedo. $\omega_0$ (525 nm) had a mean value of 0.97 ± 0.02 during the measurement period. However, daily variations in the absorption coefficient were also observed. The scattering Ångström exponent had a campaign mean value of 1.26 ± 0.73. Compared to literature values (e.g. Titos et al., 2014a; Titos et al., 2014b; Fierz-Semidhauser et al., 2010), $\alpha_{sp}$ lies in the lower range which could be a result of the maritime aerosol influence at Åre.

Table 2: Statistics (mean, standard deviation (SD) and percentile values) of aerosol optical properties derived from 5-min mean values for the entire campaign. Data were normalized to STP. Only could-free periods were taken into account (RH<95%). $\sigma_{sp}$, $\sigma_{bsp}$ and $\sigma_{ap}$ are the aerosol light scattering, backscattering and absorption coefficients, respectively. $b$ is backscatter ratio, $\omega_0$ the single-scattering albedo, $\alpha_{sp}$ and $\alpha_{ap}$ are the scattering and absorption Ångström exponent, respectively.

<table>
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<tr>
<th>Parameter</th>
<th>$\lambda$ [nm]</th>
<th>Mean</th>
<th>SD</th>
<th>Median</th>
<th>Percentiles</th>
<th>5</th>
<th>25</th>
<th>75</th>
<th>95</th>
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<td>28.1</td>
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<td>9.7</td>
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<td></td>
<td>525</td>
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<td>21.5</td>
<td>14.2</td>
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<td>7.7</td>
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<tr>
<td></td>
<td>635</td>
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<td>15.0</td>
<td>11.5</td>
<td>3.7</td>
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<td>2.5</td>
<td>0.6</td>
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<tr>
<td></td>
<td>525</td>
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<tr>
<td></td>
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<td>$\sigma_{ap}$ [Mm$^{-1}$]</td>
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<td>0.1</td>
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<td>0.13</td>
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<td>0.11</td>
<td>0.14</td>
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<tr>
<td></td>
<td>525</td>
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<td>0.10</td>
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<td>0.02</td>
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<tr>
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<td>$\alpha_{sp}$ [-]</td>
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<td>0.73</td>
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<td>1.77</td>
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<td>0.15</td>
<td>0.80</td>
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4.2 Influence of relative humidity on aerosol light extinction

One main task of this study is to compare and validate the in-situ measurements with the lidar observations. In order to do so, the humidity effect of the aerosol light extinction coefficient had to be corrected for. Figure 9 shows the measured aerosol light extinction enhancement factor \( f(RH) \) and corresponding two-parameter fit function (Eq. 26) as a function of ambient \( RH \). Values were normalized to 1 at 0% ambient \( RH \) (assuming an intercept of \( a = 1 \)). \( f(RH) \) increases with increasing \( RH \) implying that the humidity effect becomes larger at elevated \( RH \). At 80% \( RH \), \( f(RH) \) was found to be 1.7 with values of 1.5 and 1.9 for the 25th and 75th percentile, respectively. At this ambient \( RH \) the extinction coefficient is approximately twice as large as at dry condition.

As mentioned in Sec. 3.4, \( f(RH) \) was used to convert the dry aerosol extinction coefficient to ambient conditions and describes the increase in aerosol scattering due to hygroscopic growth. Since the visibility sensor only operates at a wavelength of 880 nm, \( f(RH) \) was strictly speaking only valid for \( \lambda=880 \) nm. However, the \( \gamma \)-value found in this study lies in the same range as values measured at other sites at different wavelengths (see Tab. 3). Additionally, previous studies (e.g. Zieger et al., 2010) have found that the wavelength dependency of \( f(RH) \) is weak. Thus, \( f(RH) \) at 880 nm was used, together with the scattering and absorption coefficients at 532 nm, to obtain the ambient extinction coefficient from the in-situ measurements, in analogy to Eq. 23.

![Figure 9: Aerosol light-extinction enhancement factor \( f(RH) \) at 880 nm versus ambient relative humidity \( RH \). Each point represents the average \( f(RH) \) in 2% \( RH \) size bins. The error bars represent the standard deviation of the measurements.](image-url)
Table 3: The hygroscopicity parameter $\gamma$ derived within this work for Åre and comparisons from literature. Values in brackets corresponds to the 25th and 75th percentile.

<table>
<thead>
<tr>
<th>Observation site</th>
<th>$\lambda$ [nm]</th>
<th>$\gamma$ [-]</th>
<th>Origin</th>
<th>Reference</th>
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<td>Åre</td>
<td>880</td>
<td>0.34 [0.28 0.40]</td>
<td>Continental</td>
<td>This work</td>
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<tr>
<td>Hyytiäälä</td>
<td>635</td>
<td>0.30</td>
<td>Boreal</td>
<td>Zieger et al., 2015</td>
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<tr>
<td>Granada</td>
<td>550</td>
<td>0.40</td>
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<td>Titos et al., 2014b</td>
</tr>
<tr>
<td>Cape Cod</td>
<td>550</td>
<td>0.40</td>
<td>Anthropogenic</td>
<td>Titos et al., 2014a</td>
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<tr>
<td>Cape Cod</td>
<td>550</td>
<td>0.70</td>
<td>Marine</td>
<td>Titos et al., 2014a</td>
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</tbody>
</table>

4.3 Comparison of in-situ to lidar retrieved aerosol extinction coefficient

Figure 10 and 11 shows extinction profiles as measured by the lidar on August 3, 2014 and August 12, 2014, respectively, together with the findings from in-situ measurements performed in the cabin on Mt. Åreskutan. The dry extinction coefficient was recalculated using the minimum and maximum relative instrumental errors; assumed to be 12% in the MAAP, 20% in the aethalometer and 10% in the nephelometer. The difference between the measurement and the recalculated values were used as lower and upper error limits, respectively, and are represented by the errorbars in Fig. 10 and 11. Regarding the ambient in-situ measurement, an additional uncertainty was introduced as a result of $f(RH)$ (the mean value for the entire campaign was used, thus it was assumed that the hygroscopic properties of the air mass did not change during the campaign). In this case, the 25th and 75th percentile values was used as upper and lower boundaries, respectively.

In all cases, the $\sigma_{ep}$ values of the lidar profiles and the ambient in-situ measurement $\sigma_{ep}^{in-situ}(amb)$ are in better agreement compared to uncorrected dry extinction coefficients $\sigma_{ep}^{in-situ}(dry)$. This clearly shows the importance of the humidity effect on the ambient aerosol. The temporal evolution of $\sigma_{ep}$ as measured by the lidar on August 3, 2014 was similar as for the in-situ and a totally 4 of 6 comparisons agreed within their errorbars; which is a very good sign. At low ambient $RH$ the humidity effect was less pronounced as shown in Fig. 11b.

The worse correspondence between the different measurements during the afternoon on August 12, 2014 could be a result of the lower aerosol loadings. One also needs to take into account the horizontal distance (approximately 2870 m) between the remote sensing and in-situ instruments (Fig. 3) which could cause differences in the result.
Figure 10: Profiles of the ambient aerosol extinction coefficients measured by the lidar (solid lines). The corresponding dry and ambient extinction coefficients measured at Mt. Åreskutan are shown as red and blue bullet points, respectively. A time-height display of the range-corrected lidar signal is shown in the upper panel.
Figure 11: Profiles of the ambient aerosol extinction coefficients measured by the lidar (solid lines). The corresponding dry and ambient extinction coefficients measured at Mt. Areskutan are shown as red and blue bullet points, respectively. A time-height display of the range-corrected lidar signal is shown in the upper panel.
Figure 12 shows the occurrence frequency in the ratio of the ambient and dry extinction coefficients measured with the in-situ instrumentation and the ambient extinction coefficient derived from the lidar observations at an altitude of 827 m above ground level (agl). The peak is close to a ratio of unity when the in-situ measurements are transformed to ambient conditions and lower for comparison with the dry measurements. As expected, this points toward a better correspondence between the lidar and the measurements corrected for ambient conditions. However, the width of the distribution is larger for the comparison with ambient measurements indicating that the uncertainty in these measurements is higher.

The ratio was also computed using \( \sigma_{ep} \) of the lidar as measured at several altitudes. Each value was separately compared to the in-situ value measured at ambient and dry conditions, respectively. The result is shown in Fig. 13. The ratio was found to be close to unity (ratio \( \approx 0.9 \)) at the altitude of the in-situ station when the ambient aerosol extinction coefficient was taken into account and ratio \( \approx 0.6 \) when the dry measurement was considered. It is also worth noticing that a ratio of unity appears at approximately 100 m above the in-situ station. This discrepancy will be further discussed in the next section.

![Figure 12: Frequency of the ratio of the extinction coefficient measured by the in-situ instruments (\( \sigma_{ep}^{in-situ} \)) and the extinction coefficient measured by the lidar (\( \sigma_{ep}^{lidar} \)) at an altitude of 827 m agl (for cases with an absence of clouds).](image-url)
Figure 13: Ratio of the extinction coefficient measured by the in-situ instruments ($\sigma_{ep}^{\text{in-situ}}$) and the extinction coefficient measured by the lidar ($\sigma_{ep}^{\text{lidar}}$) versus height (for cases with an absence of clouds). The vertical line marks the height of the in-situ station.

Figure 14 shows the squared correlation coefficient ($R^2$) versus the altitude above the remote sensing site. Again, the $\sigma_{ep}$ of the lidar at each altitude was separately compared to the in-situ value measured at ambient conditions. When all data was taken into account the correlation coefficient $R^2 \approx 0.85$ around the height of the in-situ station which points towards a strong link between the lidar and the in-situ measurement. The difference between the westerly and easterly flow shows the orographical effect on the height of the planetary boundary layer which was found to be much higher for easterly winds (wind directions smaller than 180°) compared to westerly winds (wind directions larger than 180°).

It is speculated that the observed shift in altitude of $R^2$ compared to the location of the measurement station, is a result of an insufficient treatment of the laser-beam receiver-field-of-view overlap effect of the lidar or can be related to influence on the flow field by the Mt. Åreskutan itself. However, a more detailed study on this interesting aspect is out of scope of this thesis.
Figure 14: Squared correlation coefficients ($R^2$) obtained from comparisons between the aerosol extinction coefficient retrieved from the lidar measurements and in-situ measurements at ambient conditions versus height (for cases with an absence of clouds).
5 Conclusions

The following is concluded from this study

1. The aerosol light extinction coefficient measured at Mt. Åreskutan was dominated by contribution from light scattering. However, diurnal variations, where the magnitude of the light absorption coefficient increased, were observed. Compared to literature values, the scattering Ångström coefficient was in the lower range which is possibly due to the maritime influence at Åre.

2. The air advected to Mt. Åreskutan can be divided into three main sectors; maritime (260°-360°), continental (90°-190°) and Arctic (20°-70°). The aerosol extensive properties showed similar variations with lower magnitudes in the Arctic and maritime sectors and higher in the continental. The intensive properties showed less variation with wind direction, although a tendency of finer particles being transported in the continental sector was observed.

3. The light extinction coefficient was found to be approximately twice as large at an ambient relative humidity of 80 % as at dry conditions. The effect of humidity on the ambient aerosol is important to consider when validating in-situ measurements with lidar retrievals.

4. The remote sensing and in-situ instruments were in correspondence. However, there was an observed shift in altitude of the correlation compared to the location of the measurement station. The reason for the discrepancy could be due to an insufficient treatment of the laser-beam receiver-field-of-view over lap effect of the lidar or caused by site specific orography. Also, the agreement seemed to be worse at low aerosol loadings which could be an indication of the sensitivity of the instruments at low aerosol loadings.
Acknowledgements

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References


