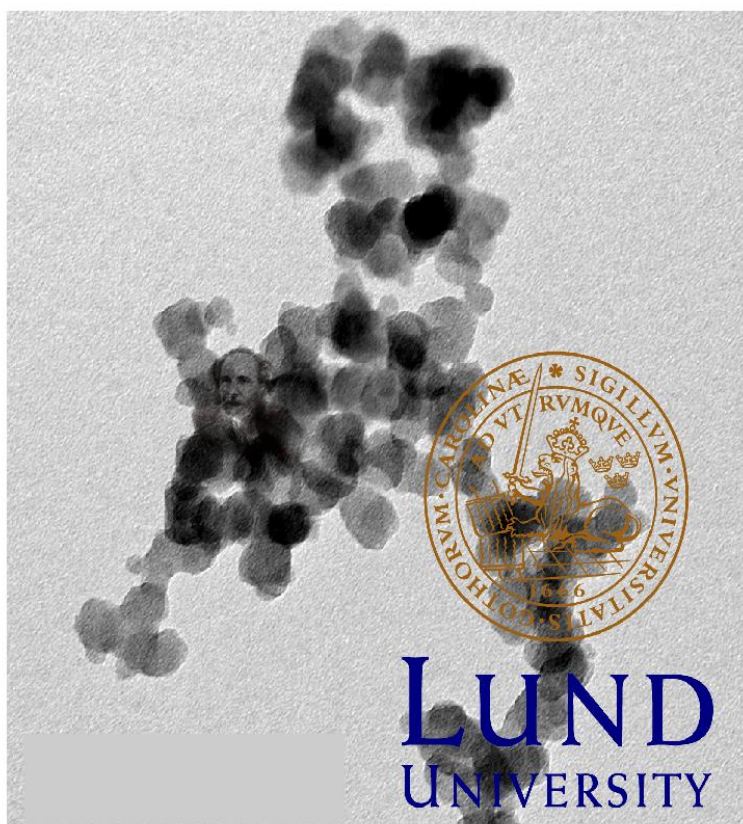


Investigation of scattering of soot with a three-wavelength integrating nephelometer

Master's thesis in Physics
by Maj Blomdell
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Abstract

Soot particles are one of the major contributors to climate change and global warming, mostly due to their absorbing effect. It is of great interest to gain better knowledge about soot particles optical properties, since there are great uncertainty how they affect the atmosphere. When soot particles are present in the atmosphere they disturb the sunlight albedo when absorbing light and by so heat up the atmosphere.

To be able to study the soot particles optical properties in this thesis an instrument that usually is used for monitoring air pollution has been used, i.e. a three-wavelength integrating nephelometer. The instrument measures the scattering of light by particles or gas. It performs the measurement with a LED light source that operates at three different wavelengths i.e. 635 nm, 525 nm and 450 nm, measuring the total scatter ($9 - 170^\circ$) for one second for each wavelength, then a shutter is turned on and the same measurement procedure is performed for the backscatter ($90 - 170^\circ$).

To be able to study the soot particles a soot generator (miniCAST) has been used. The soot generator mixes fuel, nitrogen and air in different combinations that produces soot with different characters. Five different soot types has been studied in this thesis and they are called OP-7, OP-6, OP-5, OP-3 and OP-1. Measurements have been made on both small primary particles (OP-7), and large aggregated soot particles (OP-1), hence different sized soot have different optical properties. // The results presented in this thesis are the first measurements on soot made with the nephelometer at the department of Combustion Physics in Lund, therefore some improvements to the set-up has to be made in future experiments. However, the focus of this thesis work has been to commissioning the nephelometer and to fully understand what functions the instrument has and how they can be applied to this type of experiments. Calibration of the instrument has been made several times. Typical measurements on outdoor air have been made, this to be able to see what the nephelometer is usually used for. The results from the soot experiments show that the scattering properties vary a lot for the different particles. Large particles, i.e. OP-3 and OP-1, results in a smaller SAE typically around 2, SAE stands for Scattering Ångström Exponent a parameter used in climate research models and explain the scattering parameter wavelength dependence. For small particles such as in OP-7 and OP-6 SAE=4. These two results corresponds well with what theory predicts about particles SAE's for different sizes, i.e. smaller particles results in larger SAE's whereas larger particles have smaller SAE's. A larger SAE's for backward scatter is obtained for all measurements made in this work, OP-experiments and outdoor measurements, why this is the case is still unclear and will be studied in the future and explained with mathematical modeling and analysis. Also the relative scattering, i.e. ratio between forward and backward scatter shows an increasing ratio and also an increasing separation between the three wavelengths when approaching larger particle sizes.

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Populärvetenskaplig sammanfattning

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Hur påverkar sotpartiklar atmosfären?

Sotpartiklars existens i atmosfären påverkar både vårt klimat och hälsa, partiklarnas närvaro påverkar hur solljuset sprids. Sotpartiklar är en restprodukt ifrån ofullständig förbränning, en sak som forskare jobbar kontinuerligt med för att reducera, detta genom att optimera förbränningsprocessen framförallt i motorer. I detta arbetet har sotpartiklars spridningsegenskaper studerats för att få bättre kunskap om hur sotpartiklars totala påverkan egentligen är på atmosfären.

Global uppvärmning och minskande fossila bränslereservoarer är dagsaktuella frågor. Trots att dessa resurser är ändliga så är förbränning fortfarande en av de största källorna till uppvärmning, matlagning och transport. Förbränning av fossilbränslen är inte riskfritt, det produceras restprodukter som har negativ inverkan på vår planet, sotpartiklar är en utav restprodukterna. Sotpartiklar har två primära egenskaper, antingen absorberar de ljus eller så sprider de ljus, båda egenskaperna påverkar klimatet. Det finns dock fortfarande ingen riktigt bra modell för att hantera sotpartiklarnas totala klimatpåverkan och detta är mest för att det är så många aspekter som spelar in och skall tas med i beräkningarna. Om partiklarna absorberar ljus så har de en värmande effekt och om de sprider ljus har de en avkylande effekt. Sotpartiklar är en restprodukt från ofullständig förbränning, detta är något som man ständigt försöker minimera i förbränningsmotorer genom att optimera förbränningen. Sotpartiklarna bildas i förbränningsprocesserna där ett överskott på bränsle finns i förhållande till syremängden (oxidationsmedlet). Bränslet som används när sotpartiklar bildas dissocierar de till mindre molekyler, vid bränsleöverskott reagerar dessa till större strukturer som så småningom bildar de första små sotpartiklarna. Sotpartiklar har många olika ursprungskällor, de härstammar inte enbart från förbränningsmotorer utan även från så som partikelutsläpp från motorfordon varav majoriteten från från dieselmotorer, uppvärmning av hus, tillredning av mat, utsläpp från flygplan och sjötransport. Alla har egentligen sett sot även om storleken på partiklarna är små, om man tittar på en flamma från ett ljus så ger denna ifrån sig ett gulaktigt sken detta sken är sotpartiklarna som genom sin höga temperatur strålar. Sot består till störst del av kol, 80-90 procent och de resten är väte, den höga andelen kol är varför partiklarna är mörka till färgen. Partiklarna är små vilket gör att de är skadliga för människans försvarssystem, storleken innebär att flimmerhåren i näsan inte kan filtrera bort partiklarna. När sotpartiklarna andas in tar de sig snabbt till lungorna för att sedan transporteras ut i blodomloppet. Eftersom att sotpartiklarna är cancerogena, är inte detta några partiklar som man vill ska komma i kontakt med de mänskliga organen, då de kan ställa till med stor skada. Genom att forska på sot och undersöka deras optiska egenskaper, framförallt

dess spridningsförmåga av ljus, kan vi komma närmre ett fastställande om hur stor påverkan partiklarna egentligen har. En reduktion av de mörka partiklarna i atmosfären hade bidragit till att färre partiklar fastnar på jordens snöbeksädda alptoppar- och isberg, solen dras till de mörka partiklarna och värmer lokalt upp is och snö som smälter. Som resultat av den smälta isen och snön så blottas barmark och vatten, som i sin tur absorberar ljus istället för att detta reflekteras bort från snö och is, denna absorption bidrar till den globala uppvärmningen. För att få bättre förståelse om hur sot påverkar klimatet, människan är forskning och bättre förståelse av sotets optiska egenskaper viktigt. För att undersöka hur sotpartiklar sprider ljus har ett instrument (nephelometer) använts, som brukar användas för att göra bakgrundsmätninga. Experimenten i examensarbetet har satts upp så olika sotkoncentrationer studerats med hjälp nephelometern.

För att kunna mäta på olika sorters sot har en sotgenerator använts (miniCAST), sotgeneratorn blandar bränsle (propan), kväve och syre som bildar en flamma. Man använder sedan kvävgas för att blåsa ur sotpartiklar från flaman. Olika kombinationer av bränsle och syre ger olika mängd och storlek på sot, som sedan leds vidare mot nephelometern. Innan sotpartiklarna leds in till nephelometern så späder man ut dem med luft, detta för att man inte vill leda in en för hög sotkoncentration i instrumentet. Partiklarna leds in i instrumentet där de belyses med ljus från en LED-källa, denna ljuskällan belyser sotpartiklarna med tre olika sorters ljus (rött, grönt och blått-ljus). Det ljus som sotpartiklarna sedan sprider registreras av en detektor som gör om detta till en elektronisk puls som skickas till en dator som nephelometern är kopplad till. Ju mer ljus som sprids från sotpartiklarna desto högre värde registreras av detektorn. Från mätningarna gjorda i detta mastersarbete kan vi visa hur olika storlekar av sotpartiklar sprider ljus. Detta eftersom fem olika typer av sotkoncentrationer har studerats: små färska partiklar, till många små sammanfogade partiklar i en enda stor partikelformation. Små partiklar sprider i princip lika mycket ljus i framåtriktning som bakåtriktning, medan stora aggregerade partiklar sprider helst ljus i framåtriktning. Dessa resultat stämmer väl överrens med den teorin som finns om hur partiklar helst vill sprida ljus.

Acronyms

Aerosol= Solid or liquid particle suspended in gas

Aurora 3000= The name of the nephelometer used in this thesis

BC= Black Carbon

MC= miniCAST, instrument that generates the soot in this thesis

ppm= Parts per million

PMT= Photo Multiplier Tube

OP-x: Operation Point (different soot studied, OP-7, OP-6, OP-5, OP-3 or OP-1)

Soot= Small particle that is a by-product from incomplete combustion of polyaromatic fuel

SAE= Scattering Ångström Exponent

SMPS= Scanning Mobility Particle Sizer

TEM= Transmission Electron Microscopy

α = Scattering Ångström Exponent

σ = Scattering parameter generated by the nephelometer [Mm^{-1}]

λ = Wavelength [nm]

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Introduction

The climate is changing continuously since the temperature is rising due to the greenhouse effect, because this an unwanted effect we therefore want to minimize the amount of particles and gases in the atmosphere that heats up the planet.

Soot particles are unwanted by-products from incomplete combustion processes of poly-aromatic hydrocarbons, i.e. a combination of carbon and hydrogen [1]. Combustion is vividly used for multi-purposes, such as heating of houses (mostly in developing countries), cooking and transportation etc.

Since soot is one of the main contributors to the greenhouse effect responsible for the climate change, it is therefore important to study the properties and behavior of the particles. This due to the today's great uncertainties regarding its climate effect.

When soot particles are formed the combustion process has an excess amount of fuel compared to oxidizer, the fuel is then decomposed into smaller molecules by pyrolysis. These small particles are highly reactive and unstable. These particles will most likely form particles and later on aggregates [2]. A combustion process is basically an exothermic process that involves two or more reactants. One of the reactants must be fuel and the other must be an oxidizer. When the oxidizer comes in contact with the fuel if ignition is made, it oxidizes the fuel and the chemical bond energy stored in the fuel is then released.

Soot or often called Black Carbon (BC) are carcinogenic particles, and are therefore hazardous to the human health. BC is also a major contributor to the climate change due to its positive climate forcing effect [3].

Soot particles consist mainly of carbon and some hydrogen. But what soot actually is, that is a question that has many answers. Different areas of research fields have their own definition of what soot actually is. Depending on the composition ratio of hydrogen and carbon in the soot particle, their properties and also the structure will be very different. The varying characteristics of the soot result in different optical properties. Soot particles are small: typically in the size range from a few nm up to some hundreds of nm. They are either aggregates, i.e. a combination of many primary particles (small particles with sizes from a few nm up to 50 nm), or just a single primary particle. Soot particles are often described and treated as spherical particles [1].

Soot particles are dark colored and will therefore absorb the sunlight which generates a heating. This absorption can cause massive negative effects, e.g. if soot particles stick to a glacier or an iceberg these particles will heat up and eventually melt the formations. When the formations melt, bare ground is exposed. The bare exposed ground will absorb sunlight that contribute to global warming [4].

There are many different ways to investigate how particles contribute to the climate change, one way is to gain some more knowledge about the optical properties of soot particles, which is a large contribution to the climate change. If we get more knowledge about aerosols impact on the radiation balance on our planet we might be able to do something about the increasing temperature on the planet.

The largest source to the increase of soot in the atmosphere is due to humans. This due to the fact that we are using combustion in several different ways [4]. The largest contribution comes from residential solid fuels for combustion processes, in Asia and China it stands for 60-80 percent of the soot emissions [4]. The use of residential coal is still being used in: eastern Europe, China and the former Soviet Union. The second largest contribution comes from diesel engines, they contribute up to 70 percent of the emissions in Europe, Northern America and Latin America [4].

It is important to have in mind that even combustion of biomass that results in negative climate forcing aerosols, can result in indirect positive climate forcing in e.g. when they stick to snow or ice, and

causes them to melt [4].

The main focus in this thesis is to observe the scattering properties of soot. The instrument used in this thesis to investigate the scattering of soot is a three-wavelength integrating nephelometer (Aurora 3000) [5]. It measures the scattering of light from a sample illuminated with an LED light source at three different wavelengths, 635 nm, 525 nm and 450 nm (red, green and blue light).

The nephelometer is an instrument commonly used for studying climate change, this because the instrument provides information about the aerosol asymmetry parameter, Ångström coefficient and scattering parameters [5], these parameters are used in most today's climate models. The nephelometer is usually placed at background locations such as in a forest or other site, or mounted onto an aircraft to perform background measurements.

1.1 Aim

The aim of this thesis was commissioning the nephelometer and explore the possibilities of using the nephelometer to gain knowledge about soot scattering properties. With the main focus to obtain reliable and accurate measurements.

Theory

2.1 Soot particles

Soot particles are classified as a aerosol particle, an aerosol is defined as a solid or liquid particle suspended in gas. If the aerosol is suspended in air the aerosol is an atmospheric aerosol [6]. Soot particles are carbonaceous particles, which have a very wide size range, they can either be small primary particles with a size of a few nm, or large aggregates with a size of some hundred micrometers. Soot particles originates from two major areas: i.e. industrial production of soot for coloring in e.g. tires and paint, and from incomplete combustion processes of hydrocarbons. The origin of soot particle emission is illustrated in figure 2.1.

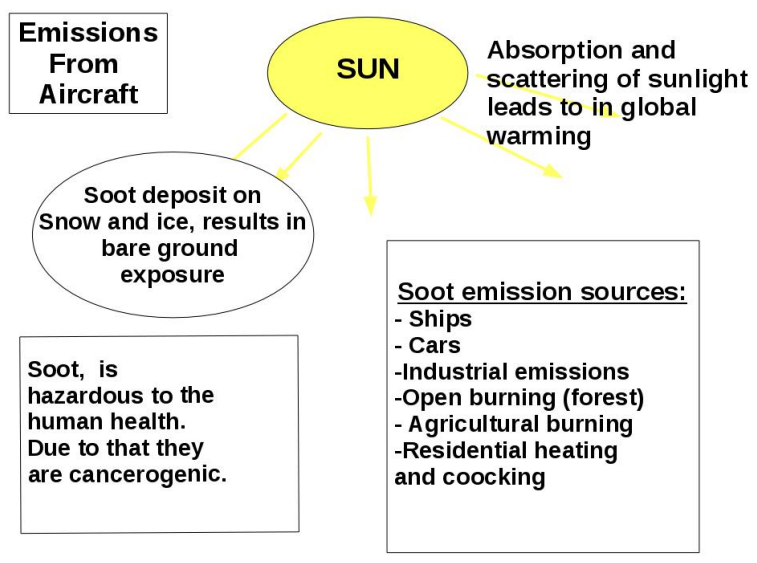


Figure 2.1: Illustration of the soot impact and emission on the planet.

2.1.1 Combustion process

Soot particles are generated when we have a non-ideal combustion process, i.e. the amount of fuel exceeds the amount of oxygen in the mixture. An ideal combustion process(stoichiometric reaction) is when the combustion process only creates carbon dioxide and water as products, is shown in an example of propane combustion in equation (2.1).



The ratio between fuel and oxidizer in a combustion process is often expressed as the equivalence ratio Φ or the carbon-oxygen ratio $\frac{C}{O}$. The equivalence ratio is expressed in equation (2.2).

$$\Phi = \frac{\left(\frac{n_{fuel}}{n_{oxidizer}}\right)}{\left(\frac{n_{fuel}}{n_{oxidizer}}\right)_{stoichiometric}} \quad (2.2)$$

When $\Phi = 1$ a reaction is stoichiometric, when $\Phi > 1$ it is rich, and when $\Phi < 1$ it is a lean combustion process. The carbon to oxygen ratio is simply $\frac{n_C}{n_O}$, n stands for the number of moles of the different reactants. Soot starts to form when this carbon oxygen ratio is 0.5 – 0.8 [7]

When the ratio between the number of carbon and oxygen molecules in a reaction increases and approaches a number higher than one then soot is formed. The mixture of fuel and oxygen is then said to be rich. The situation obtained is where the oxygen cannot oxidize all the fuel which basically results in more carbon left in the mixture. This excess of carbon is the basis for the soot formation.

2.1.2 Soot formation

For combustion processes when the amount of fuel exceeds the amount of oxidizer, it will result in polycyclic aromatic hydrocarbons. These will act as building blocks that form into larger and larger groups that eventually will form particles, i.e. soot particles, figure 2.2 explains the soot formation process. schematically step by step. In the first step we have the molecular zone, i.e. where fuel and oxidizer

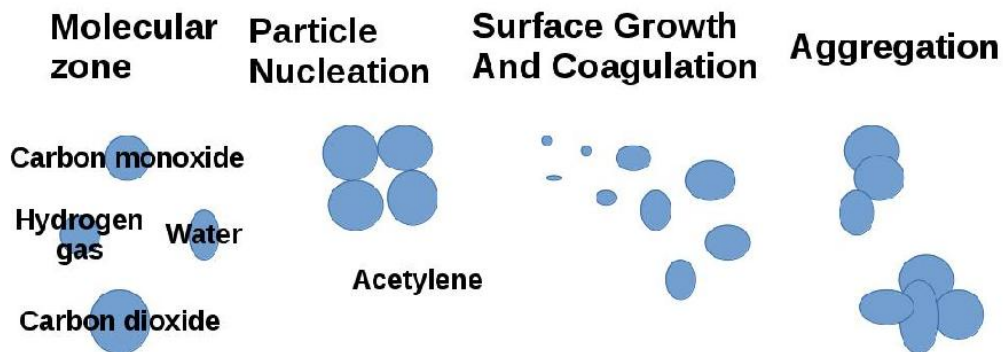


Figure 2.2: Soot-formation

mixed. In this first step the fuel is being decomposed into smaller building molecule blocks; this process is called pyrolysis. Some of these molecules are unstable and also have a certain chemical composition which makes them highly reactive, so they react with the other molecules present. The base for soot to form are primary poly-aromatic hydrocarbon particles that interacts with acetylene, see figure 2.2 in the second step [7]. When these two react with each other, they result in larger structures which will form a soot particle nuclei, this nuclei will in turn act as a building block which can continue to grow larger

through surface growth. In the fourth step the particles stick together and form larger compounds. These compounds are called soot aggregates, which will occur if the density of the particles is high enough.

2.1.3 Health effects

Health effects due to particulates is important, this because millions of people are dying prematurely due to the exposure of small particulates [4]. Air pollution that humans are exposed to is located both indoor and outdoor air, on the country side and in the cities. Since there are still a lot of questions that need to be answered in this research field, it is therefore of great importance for air pollution research to proceed.

Soot particles are highly reactive and unstable in the beginning of their growth process, which is also the main reason why they are hazardous [2].

Due to the size of soot particles, they can easily enter the human body. This because they are not filtered out from the human defense system, e.g. nose hair. When soot particles are inhaled or swallowed they can deposit in the lungs and enter the blood-stream. Since soot particles contain polyaromatic hydrocarbons, they are carcinogenic. So when the particles enter the human body they can cause tremendous harm [8][9].

People with an already existing problem with their respiratory system, diabetes or heart diseases will be more negatively affected when exposed to airborne particles. If a person exhibits lung dysfunction, then it is more harmful to inhale soot particles than for an healthy person.

If you have some chronically disease such as type two diabetes it is much more harmful to inhale the particles. There are many effects from exposure of soot particles, such as: coughing, breathing issues, decreased lung function, asthma, chronically bronchitis, hart beat irregularities, death for people whom lives with lung- or some kind of heart mal-function [10]. So, depending on whom that inhales these particles the effect will look differently.

Today there is not enough evidence of how airborne particles will affect a healthy human, but scientists expect that these small particles will have a long-term effect. Therefore this field of research is of great importance to improve and all new knowledge is valuable.

2.1.4 Atmosphere and climate

Aerosols and sunlight in combination create many optical effects such as rainbows and halos. However, increasing the amount of particulate in the atmosphere is not wanted phenomenon, as many of the particulates are unwanted highly absorbing particulates that heats the planet as and disturbs the solar radiation balance.

The first and also the most commonly spoken effect from soot, is the radiative forcing, i.e. the result from scattering and absorption of the incoming sunlight. Due to darken soot particles absorb the sunlight and will in turn heat the atmosphere where they are located, hence they contribute to global warming.

The second issue is the fact that soot affect clouds. Soot particles are volume absorbers, so when soot absorbs light they interfere with the clouds and change their composition. The soot particles will change the number of ice particles and number of liquid droplets. This cloud effect is a indirect effect from soot [4].

The semi-direct effect is basically when light is absorbed by the soot particles and this absorption results in a temperature change from before, both underneath and above the clouds location [4]. Snow- and ice effect is when the soot particles is stuck to a ice- or snow surface. When this occurs the surface start to melt, this due to the dark surface of the particles that absorbs the sunlight. This heating is a typical positive climate forcing.

The soot particles positive climate forcing results in an warmer troposphere, and also a warmer cryosphere, resulting in global warming. The troposphere is the lowest part of the atmosphere i.e. 0-20 km from earth surface and it is in this part of the atmosphere where we have weather. The troposphere consists of 75 percent of the atmospheric mass[11]. The cryosphere is defined as all the non liquid phases of water on the planet i.e. glaciers, sea ice and snow [12].

The impact induces some climate responses, e.g. surface temperature, formation of clouds, circulation and change in precipitation. Soot particles are short-lived compared to other climate forciers, so they will only stay in the atmosphere for a few weeks and be reactive. This is short-lived compared to e.g. CO_2 , which stays in the atmosphere for a much longer time [4].

2.2 Optical properties

Optical properties of airborne particles are responsible for many different phenomenon around us, e.g. rainbows or the color of the sky[8]. The scattering and absorption of light is responsible for how good the visibility is and also for the solar balance on earth [8]. The change in intensity of incoming light that passes a volume of aerosol is called extinction. This phenomenon describes the attenuation of light when it interacts with the aerosol by scattering and absorption [5]. This is shown in equation (2.3), where σ_{ext} is the extinction coefficient, σ_{scat} is the scattering coefficient of light, and σ_{abs} is the absorption coefficient.

$$\sigma_{ext} = \sigma_{scat} + \sigma_{abs} \quad (2.3)$$

The phenomena can neatly be described by the Beer-Lambert law equation (2.4).

$$I = I_0 e^{-\sigma_{ext} x} \quad (2.4)$$

Where I_0 is the initial intensity of light on the particle, I is the intensity of light after traveling the distance x , σ_{ext} is the extinction coefficient or attenuation and x is the the path length that the light travels through the aerosol.

The attenuation in the atmosphere can be from either particles or molecules, therefore the scattering and absorption of light in the atmosphere can be expressed as in equations (2.5) and (2.6).

$$\sigma_{scat} = \sigma_{scat_{gas}} + \sigma_{scat_{part}} \quad (2.5)$$

$$\sigma_{abs} = \sigma_{abs_{gas}} + \sigma_{abs_{part}} \quad (2.6)$$

This approximation will be used when studying the aerosol particles with the nephelometer.

2.2.1 Scattering of light

There are many processes that are types of scattering, e.g. diffraction, reflection and refraction. Scattering of light by a system, e.g. particle, liquid or gas is directly related to its heterogeneity. The scattering principles are the same in all systems no matter what size the system has [13].

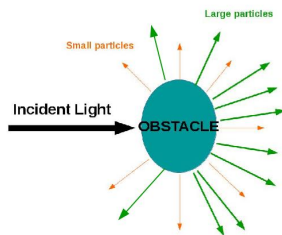


Figure 2.3: Scattering of light.

It is the systems electrons which start to oscillate when in contact with light i.e. electromagnetic illumination. The illuminating wave forces the charges in the system into an oscillating motion [13]. Scattering consists of two processes i.e. excitation and re-radiation. Excitation i.e. the incoming electromagnetic wave sets the electron of the system into motion. Whereas re-radiation is the emitted radiation from electrons in motion due to them being accelerated. It is this radiation that is the scattered light by the particle.

There are three types of scattering, i.e. elastic, inelastic and quasi-elastic scattering. Elastic scattering is scattering where the wavelength is the same as the incident light before the scattering process (Mie- and Rayleigh scattering are elastic scattering processes). Inelastic scattering refers to that the wavelength shifts in the process, thus we do not have the same wavelength after the scattering as the incident radiation. The quasi-elastic scattering is when we have a shift in the wavelength, this due to matter in movement (Doppler effects). In this thesis the scattering processes observed and treated are the elastic ones. There are three things that affects what kind of scatter that will occur, the wavelength of the incoming light, the size of the scattering particle and the particles optical properties relative to the surroundings [13].

Using the equation (2.7), scattering from aerosol particles can be explained with either Mie theory or

Rayleigh theory, this is $x \leq 1$ or $x \ll 1$. Mie theory explains the scatter from particles in the same size as the incident wavelength. Whereas Rayleigh theory handles the much smaller particles than the incident wavelength [14].

$$x = \frac{2\pi r}{\lambda} \quad (2.7)$$

λ is the wavelength, r is the radius for the particle and x is the non-dimensional size parameter.

Scattering by a dipole array describe the scattering by particles, when they are in the same size range as the incident wavelength of light (Mie regime) they scatter preferably in the forward direction. Whereas for smaller particles (Rayleigh regime), where the phase differences are smaller, so the scattering is roughly the same for forward and backward scattering [14].

2.2.1.1 Mie scattering

Mie theory explains the scattering by aerosol particles, i.e. it basically explains the scattering of electromagnetic radiation by spherical particles by solving the Maxwell equations [14]. Mie theory is applied for particles in the same size range as the illuminating wavelength of light [8]. The theory is a fully analytic solution to the Maxwell equation. Equation (2.8) explains the intensity, when illuminating a particle with unpolarized light.

$$I(\theta) = \frac{I_0 \lambda^2 (i_1 + i_2)}{8\pi^2 R^2} \quad (2.8)$$

Where $I(\theta)$ is the scattered intensity, I_0 is the incoming unpolarized light, i_2, i_1 are the Mie parameters for intensity for scattered light for parallel and perpendicular polarization [8].

When the light is perpendicular polarized equation (2.9) is used.

$$I_1(\theta) = \frac{I_0 \lambda^2 i_1}{4\pi^2 R^2} \quad (2.9)$$

And for parallel polarization equation (2.10) is used.

$$I_2(\theta) = \frac{I_0 \lambda^2 i_2}{4\pi^2 R^2} \quad (2.10)$$

2.2.1.2 Rayleigh scattering

Rayleigh scattering is a theory that is applied for particles that are much smaller than the wavelength of the incoming light.

The theory is an simplified approximation of a quite complex description for scattering of light, i.e. Maxwell's equations. Hence for small particles and gas molecules, we have that the electromagnetic field from the incident light is the same at all points on the particle. This generates the particle illuminated to oscillate in the same way as the incident light does, this because when the particle is illuminated it becomes an oscillating dipole [8]. When an dipole is oscillating it re-radiates energy, and this in all different directions.

The intensity of the Rayleigh scattered light is given by equation (2.11).

$$I(\theta) = \frac{I_0 \pi^4 d^6}{8R^2 \lambda^4} \left(\frac{m^2 - 1}{m^2 + 2} \right)^2 (1 + \cos^2 \theta) \quad (2.11)$$

Equation (2.11) valid for small particles i.e. particles that are much smaller than the incident wavelength of incoming light ($\lambda = 500nm$). Where $I(\theta)$ is the total intensity, R is the distance from the particle, θ is the scattering angle [8].

2.2.2 Scattering Ångström Exponent (SAE)

The scattering Ångström exponent is a way of explaining the scatter parameter and its dependence on wavelength, this exponent is included in most of the today's climate models [5].

Scattering Ångström exponent is expressed as in equation (2.13), which is valid when observing the scattering dependence for two wavelengths.

$$\sigma_{scattering} \propto \frac{1}{\lambda^\alpha} \quad (2.12)$$

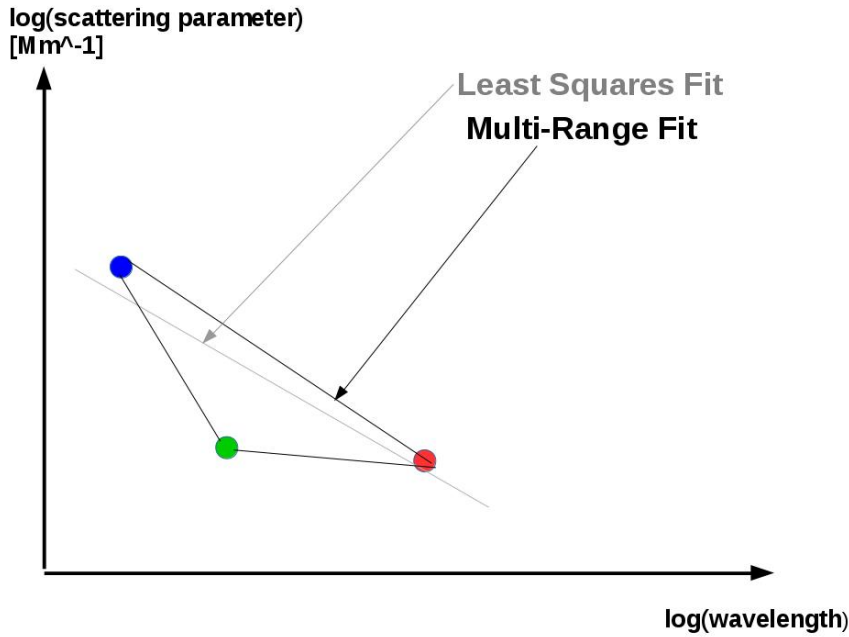


Figure 2.4: Least squares fit and multi-range fit illustration.

$$\alpha = -\frac{\log \frac{\sigma_1}{\sigma_2}}{\log \frac{\lambda_1}{\lambda_2}} \quad (2.13)$$

Where σ is the scattering parameter and λ is the wavelength [15].

For gas molecules and small particles (Rayleigh scattering) we have that SAE=4 and also wavelength dependence. For larger particles the SAE is smaller and the wavelength dependence is decreasing with particles size, whereas large particles approaching a smaller value this is shown in equation (2.12). The smaller the particles are the larger the SAE will be, large particles have small SAE [16].

In this thesis two ways of calculating and expressing SAE will be evaluated and compared, illustrated in figure 2.4 . Least-squares methods adapt a line that fits the three wavelengths for each time-step (this is done for forward- and back scatter, providing two points for each time step). The method gives an indication how well the adapted or fitted the line is compared to the measuring values. The second method is called multi-range fit, where SAE is calculated for the three wavelength pairs $(\frac{450}{635}, \frac{450}{525}, \frac{525}{635})$, calculated with equation (2.13). These two ways of expressing SAE will be compared in the result as , Least square figures and multi-range method figures, the two methods are compared in tables.

2.2.3 Absorption

Absorption is when a particle or gas absorbs the incoming light. Absorption only occur for particles that consists of a absorbing material. Soot particles are the most absorbing particle in the atmosphere, whereas most of all the other atmospheric particles are mainly scattering.

Soot particles act as black bodies, which means that they act as an ideal particle absorber, and absorbs "all" of the incoming electromagnetic radiation. When soot emit radiation it does so according to Planck's law, and the radiation emitted is due to particle temperature dependence [13]. Planck's law is described in equation (2.14).

$$I(\lambda) = \frac{2\pi hc^2}{\lambda^5} \frac{1}{e^{hc/\lambda k_B T} - 1} \quad (2.14)$$

Where as the total intensity increases with temperature according to Stefan Boltzmanns law

$$I = \sigma T^4 \quad (2.15)$$

Even though the scattering for soot particles often can be neglected in comparison with absorption, since the absorption for these particles are so much greater. In this thesis though the opposite thing is made , this because the instrument used in the thesis only measures the scattering of the particles and not the absorption, some compensation due to absorption will be made in future mathematical models.

Instrumental

The main diagnostic instrument used in this thesis project was a three wavelength integrating nephelometer with back scatter, from the manufacturer Ecotech, model Aurora 3000 [5]. The source of soot particles was a miniature combustion aerosol standard (miniCAST) model 5201C [17].

3.1 Nephelometer

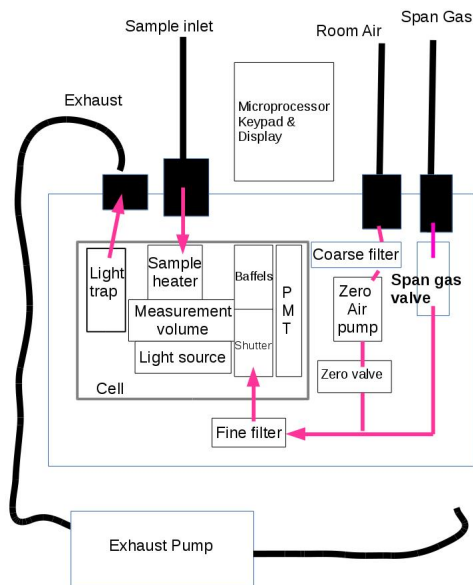


Figure 3.1: Illustration of the Aurora 3000 nephelometer.

scatters light smoothly and gives the light a lambertian distribution. The lambertian makes the light look the same at all angles. Due to the diffuser there will be no glare nor reflections or change in the light brightness [5].

One major benefit with this instrument is that it performs calibration automatically. This multi-wavelength integrating nephelometer makes it possible to obtain a deeper analysis of particles [5].

The instrument is a commonly used one for climate research. This because it provides information about some useful parameters used to describe the climate models. Some of these parameters are the

Aurora 3000 measures and provides the scattering coefficient (σ_{sp}) of light of particles at three different wavelengths in both total $9 - 170^\circ$ and back direction $90 - 170^\circ$, i.e. wavelengths i.e. 450 nm (blue), 525 nm (green) and 635 nm (red).

The scattering coefficient generated by the Aurora 3000 is given in inverse mega meters (Mm^{-1}) [5]. The instrument provides scattering parameters in total- and back direction. To obtain the forward scattering is just a simple calculation, by subtracting the back scatter from the total scattering parameter. The higher value that one obtains for the scattering coefficient (σ_{sp}) from a measurement the higher concentration (or more scattering media) of particles is present in the measuring cell.

The light sources in the instrument are LED lights, there are nine LED's in the instrument i.e. three LED's for each wavelength (635, 525 and 450 nm) [16]. These LED's will simultaneously illuminate the sample, and then the photo-multiplier tube (PMT) measures the scattered light from the sample [5].

A LED-light source has many benefits e.g. it is low power consuming and will last for a long time. The LED's have also a great signal-to-noise ratio, with a band width of 40 nm. The light source in this instrument has a opal diffuser. That is an optical device that

aerosol asymmetry parameter, the Ångström coefficient and the scattering parameter.

3.1.1 Optics

The measurement volume inside the nephelometer is the most interesting part of the instrument, since it is where all the scattering occurs. The volume consists of the PMT (Photo Multiplier Tube), the light source i.e. nine LED's, three for each wavelength (635, 525 and 450 nm). Aurora 3000 is a nephelometer with backscatter function, i.e. therefore there is a backscatter shutter mounted inside the measurement volume just below the light source. The region where the measurement occurs is the narrow cone illustrated in figure 3.2. The cone's area increases with r^2 whereas the scattering intensity decreases with $\frac{1}{r^2}$, so this is basically why the optics is formed as neat as it is.

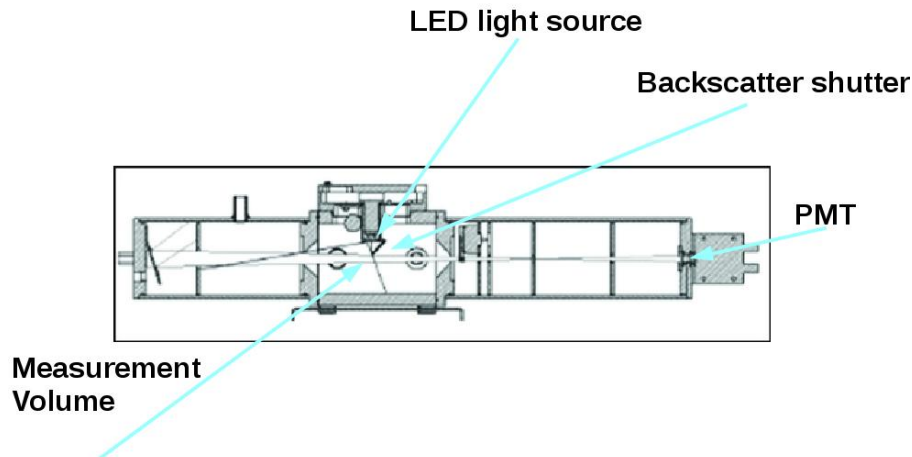


Figure 3.2: Illustration over the measurement volume inside the nephelometer.

3.1.2 Detailed description of operation

Sample air is drawn into the measuring volume (10.5 cm^3) through the sample inlet at the top of the instrument and then exits through the sample outlet, at a certain rate given by the external exhaust pump (5 l/minute in this work).

Inside the measurement volume the LED's pulses at the three wavelengths: 450 nm, 525 nm and 635 nm. When the particles/gas in the sample air are/is illuminated they will scatter light of which some will be detected by the PMT.

Inside the optical chamber, some baffles are mounted, with the purpose to limit the scattering angles into a narrow cone that spans from an angle of 9 to 170 degrees. From this narrow scattering cone of light, the scattered light from particles/gas molecules travels and hits the PMT. Due to the positioning of the baffles, it is not likely for multiple scattered events to be detected by the photo-multiplier tube. Inside the instrument there are some light traps and more baffles, and their purpose is to minimize the amount of unwanted reflections coming from the LED light source, and parts of the cell where no measurements are performed [5].

When the scattered light hits the photo-multiplier tube it creates an electrical signal that is proportional to the scattered light intensity. This gives us the data that is interesting for us i.e. σ_{scatt} .

This procedure above is conducted for both total (9-170 degrees) and backscatter (90-170 degrees), when the back scatter measurement is performed a shutter is blocking the light source from the opposite

side of the light source.

In order for the PMT signal to be converted to a scattering factor, certain calibration procedures have to be done before and during measurement series. These are shortly presented in table 3.1.

Table 3.1: Nomenclature for the nephelometer events.

Nomenclature	Explanation
Span	During calibration: A gas with known Rayleigh-factor is passed through the nephelometer and this works as a reference
Zero	During calibration: Air without particles are passed through the instrument and used as reference
Shutter Count	During measurements: Measurement of the light shone through a dark colored glass, with known transmittance. Used as reference, for intensity- and PMT measurement
Dark Count	During measurement: Background light scatter measurement, when the LED is off. This is subtracted from the measurement to get rid of background noise
Measure Count	Raw measurement of the light scattering of particulates in the inlet sample with air, in the measure volume
Backscatter	Measurement of the scattered light at angle (90-170 degrees)
Total scatter	Measurement of the scattered light at angle (9-170 degrees)

3.2 miniCAST (MC)

The miniCAST is the source that generates soot in the experimental set-up. It mixes propane, nitrogen and air in different combination, which generates different kinds of soot. Inside the miniCAST a diffusion flame is present, the fuel to generate this flame is propane, and the oxidant is air. These two are mixed together with nitrogen, ignited and a flame is formed. See schematic illustration in figure 3.3. A quench gas (N_2) is present and blows from the side of the flame onto the diffusion flame, this forces the soot particles to exit from the flame before they are fully combusted (oxidized). When the particles then leave the flame they are diluted with air, to obtain a good mix of air and particles, which suits our purpose [17].

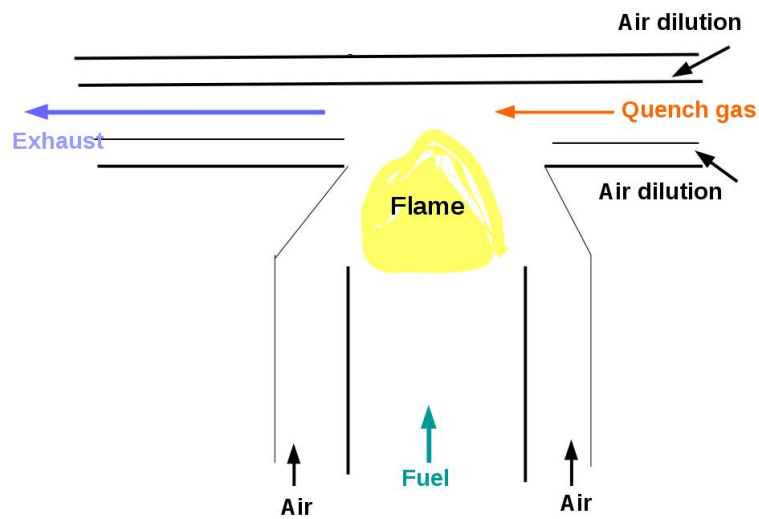


Figure 3.3: Schematic illustration over the miniCAST soot generator.

3.2.1 Operation Points (OP)

In this thesis five different operation points were used i.e. OP-7,OP-6,OP-5,OP-3 and OP-1. The different operation points are used in order to generate different kinds of soot particles with the miniCAST. The operation points are different settings of fuel-, oxidizer- and nitrogen/fuel dilution flows which will generate flames with different characteristics. In this way the quenched particles will have different properties depending in the operation point. Where OP-7 is the case where we have small primary particles, and in the OP-1 case we have mature soot particles in form of aggregates see TEM (Transmission Electron Microscopy) images in the figures 3.4a to 3.4c.

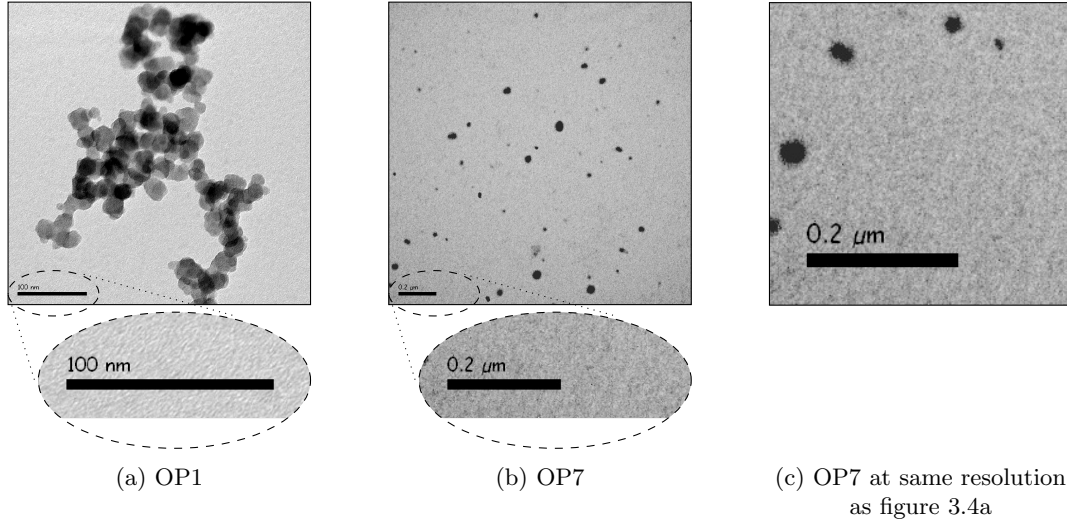


Figure 3.4: Illustration of soot aggregates sampled from OP-1 and OP-7 conditions studied using transmission electron microscopy (TEM), figure c is scaled so that figure a and figure c can be compared and have the same scale.(TEM imaging a-b by Sandra Török)

Table 3.2: Operation point mixture description of the miniCAST soot generator [17].

OP	C_3H_8 [l/min]	N_2 [ml/min]	Oxidation air[l/min]	Quench gas[l/min]	Dilution air[l/min]
1	0.06	0	1.55	7.0	20
3	0.06	100	1.24	7.0	20
5	0.06	200	1.47	7.0	20
6	0.06	250	1.42	7.0	20
7	0.06	300	1.36	7.0	20

Methods

4.1 Set-up

The instrumental set-up used is the nephelometer and the miniCAST (MC) soot generator and a dilution system. The dilution system is connected to the MC, so that the soot particles generated by the MC can be diluted. Since it is unclear how fast the measurements will contaminate the nephelometer and especially the measure volume, a too high concentration of soot entering the measuring volume is undesirable, hence dilution is performed on the soot generated before it is fed into the instrument. Figure 4.1 shows the instrumental set-up for the nephelometer, miniCAST and the dilution system.

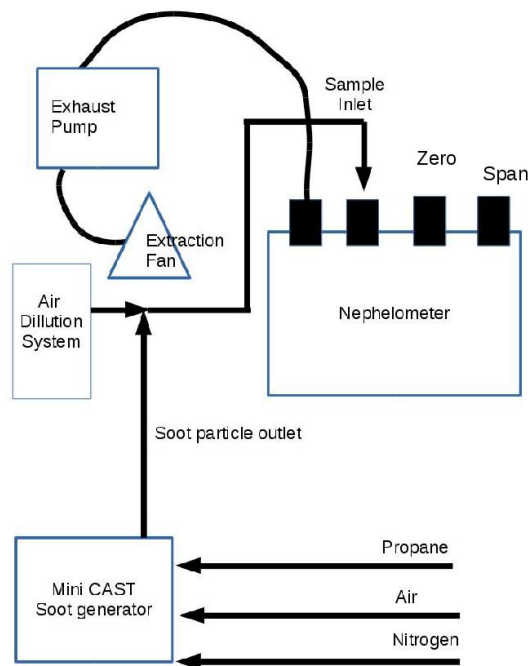


Figure 4.1: Schematic illustration over the instrumental set-up used in the laboratory measurements.

4.2 Calibration of the nephelometer

The calibration of the instrument is straightforward as the instrument performs the calibration automatically. A reference gas is used for the calibration, in this thesis carbon dioxide is used with high purity of 99.9992 percent. The calibration of the instrument is performed in two steps i.e.

- i). Calibration with a known gas (e.g. CO_2) (*span-gas*)
- ii). Calibration with particle free air (*zero-check*)

These two calibration parts produces a point for the span-gas and one for the zero-check. These two points makes up the calibration curve illustrated in figure 4.2 [5]. This calibration curve will then later be used in all measurements.

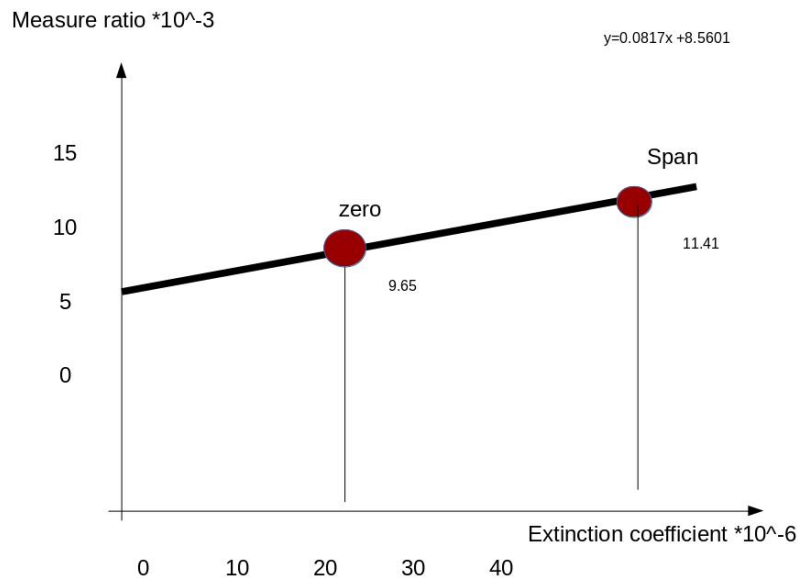


Figure 4.2: Illustration over the calibration curve.

4.3 Program for handling the data

The manufacturer of the nephelometer provided a program for downloading data (Airodis). This program is operating in Windows based environments, and sometimes it did not work, so it was not reliable. Therefore a program for downloading data had to be made, the program for downloading the data in a Linux environment directly from the instrument is presented in appendix B. To be able to write this downloading program a switch was build, i.e. a device to be able to eavesdrop what kind of information the nephelometer and the Airodis program sent to each other. After this eavesdropping a program for downloading the data from the nephelometer was written (see Appendix B). If this program would not have been written it would not have been possible to collect the data and analyze them in a reliable way.

4.4 Laboratory work

The measurements made in this thesis conducted according to the same schedule during each experiment. It started with turning on the nephelometer. The instrument needs some warm-up time, this before calibration can be made (30 minutes, warm-up time). Then calibration is made, and after that the experimental measurements.

4.4.1 Measurement procedure

The measurement procedure was the same for all laboratory days, i.e. first the nephelometer needs 30 minutes of warm-up time, the nephelometer is turned on and indoor air is measured during the warm-up time see first part in figure 4.3. After warm-up time the calibration is made, first calibration with CO_2 ("span" inlet) is made for 20 minutes and directly after calibration with air (via the "zero inlet", and filters all the particles away) is made for the same amount of time, see figure 4.3. After the calibration the first thing is to set the air dilution system to 5.0 liters/minute, as when we have 5.0 liters/minute we do not get any soot particles into the nephelometer at the time of turning on the miniCAST. Secondly the miniCAST soot generator (at OP-1) is started. Measure on tube air will be performed for a while, after that the soot measurements begin (third part in figure 4.3). After the three steps are finished,

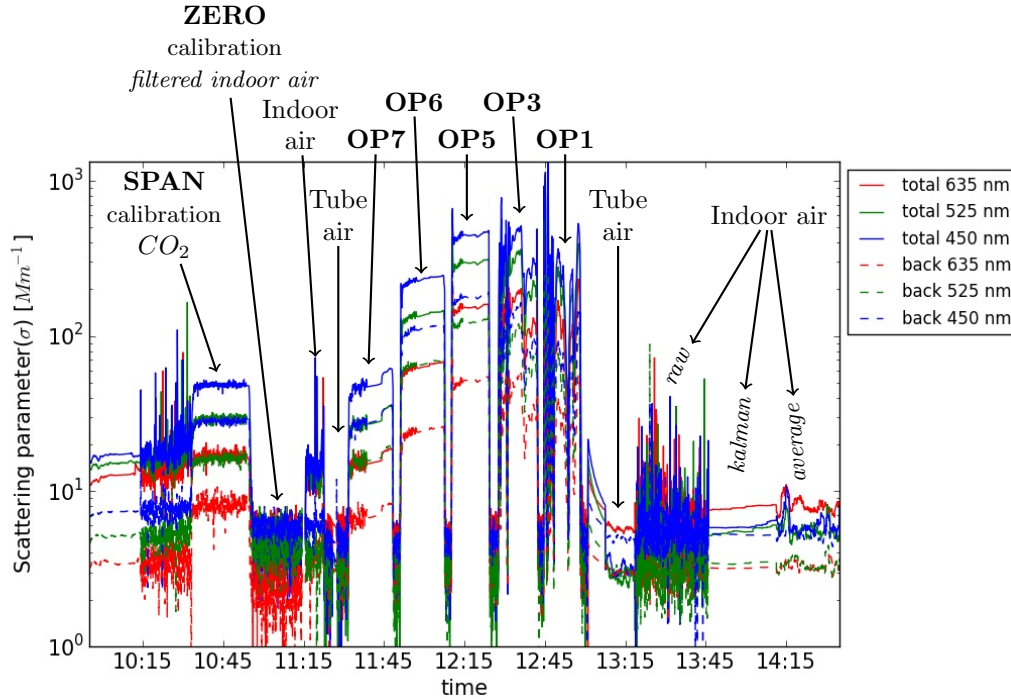


Figure 4.3: Illustration over a measurement procedure.

measurements on actual soot can be performed.

The experimental measurements are performed in the same way at all times, starting with the operation point with small soot/not aggregated soot particles and then performing measurements on the five operation points towards operation point one i.e. the largest/aggregated soot particles. Why you go from the least sooty case to the sootiest case is to eliminate the possibility to contaminate the instrument.

- The first measurement is on OP-7 i.e. the smallest particles investigated in this thesis, illustrated in figure 3.4b . The measurement for this condition will be for a time determined in advance (minimum measurement time is 5 minutes up to 15 minutes). The air dilution flow is set to a value that has been experimentally set where we see that a stable signal is obtained. After 5-15 minutes tube air is lead into the nephelometer again at 5.0 liters/minute. This tube air after the OP-measurement is to empty the nephelometer on particles (if we obtain the same value for tube air as we had before measurements was made, then we know that the soot do not contaminate the nephelometer).
- Then the second measurement is on OP-6 and the same procedure as just explained is repeated.
- Then the procedure is repeated for OP-5, OP-3 and OP-1. After these measurements has been made, tube air is lead into the nephelometer again for the same amount of time as before the experimental measurements started. This is done to verify that the same level of signal is reached as before we made the measurements. This is a way of knowing that we do not ruin or contaminate anything in the nephelometer when we the measurements are performed.

4.4.2 Improvements and corrections

4.4.2.1 Filters

There are three modes valid in the nephelometer:

- Kalman filter
- Moving average filter
- No filter (raw data)

From the manual [5] the Kalman filter is set by default, there is no information in the manual that the filter function can be turned off, the kalman filter is used when performing background measurements i.e. when no fast changing events occurs. When doing the preliminary measurements in this thesis and analyzing the data, I found out that these results were not as expected nor how they were desired to be represented. So therefore some investigation on weather this filtering function could be turned off was set into motion.

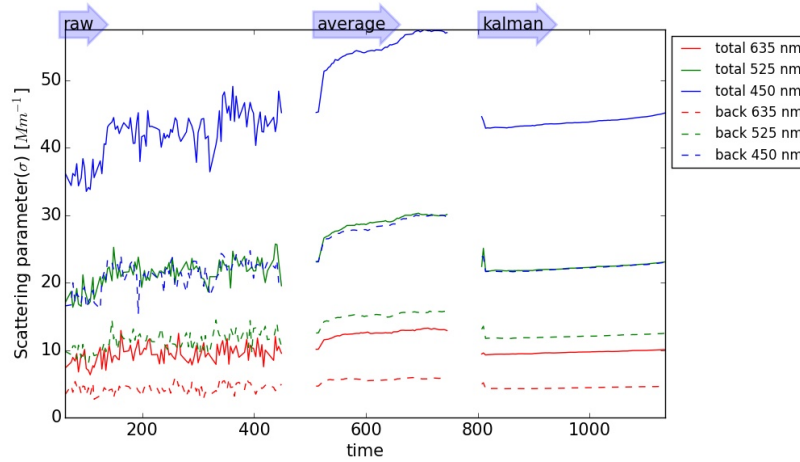


Figure 4.4: Illustration over the three filters in the nephelometer .

In figure 4.4 the scattering data values had a certain response time before it reached stable values, this indicated on some kind of digital filter in the instrument. Therefore David Logan, Electronic Engineer at ECOTECH were contacted for some answers weather the filtering function could be turned off, (E-mail contact with David Logan at Ecotech, 25/10 2016). After the correspondence with David, new experiments without filter was designed and made. A illustration over the three different filters are illustrated for OP-7 in figure 4.4

The sampling time in the instrument is set to five minutes per default, this can be changed to one minute averaging according to the manual [5], since these measurement on soot contains rapid changes we want to record everything that happens, therefore David Logan were contacted once again to see if it was possible to obtain better sampling values. It was possible to change the averaging time to five or one minute to "All" which provided 10 sampling values/minute.

4.4.2.2 Correstions of negative data values

In figure 4.5 the calibration process with the corresponding environment figure are displayed. It is shown here that the relative humidity decreases and therefore the absolute humidity increases and the temperature in the laboratory room is raised during the warm-up time of the instrument presence of human body in the room. The assumption is that the negative values are due to the breathing exhale (CO_2 and H_2O).

During the warm-up phase, (10.00 - 10.30) see figure 4.6 the absolute humidity is increasing. It is assumed that this increase is due to operator exhaled H_2O , and it is not unreasonable to assume that the CO_2 concentration follows a similar trajectory.

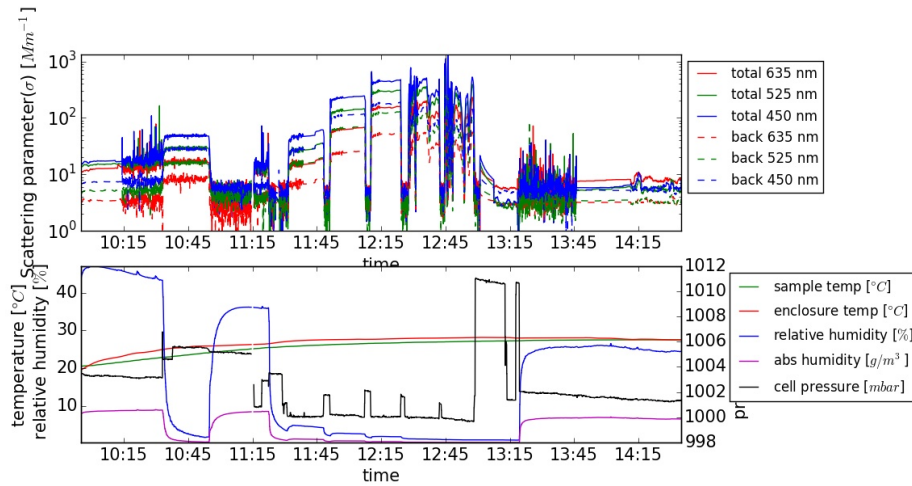


Figure 4.5: Illustration over scattering parameter data with environment in the subplot.

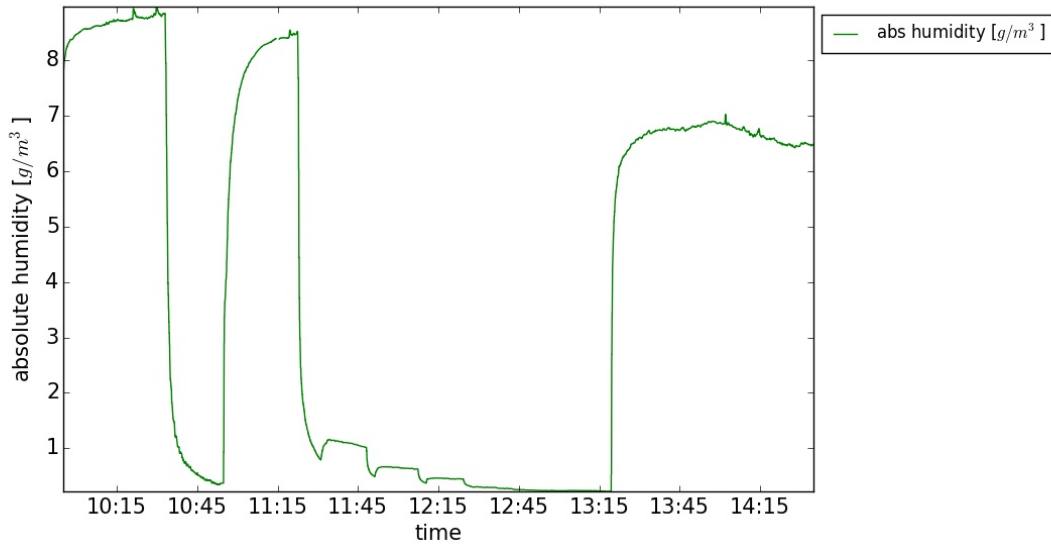


Figure 4.6: Illustration of absolute humidity for 2016-11-01.

Calibration of the nephelometer is made with room air and hence tube air consists of less CO_2 than the laboratory room air, this therefore results in some negative values in some measurements. This since tube air is used in the measurements between the five different OP-cases. The negative values due to the increasing amount of CO_2 in the room are small, but they are still there.

The temperature in the room increases as a human body is being present during measurements, this temperature increase affects the absolute humidity in the room, i.e. a temperature increase of 4 °C corresponds to:

20°C: 45% relative humidity corresponds to $5.85g/m^3 H_2O$ and

24°C: 40% relative humidity corresponds to $8.44g/m^3 H_2O$

i.e. the total amount of water in the room increases as a operator of the instrument is being present in the room during the measurements.

There are no measurements for the CO_2 amount in the laboratory though, however since the operator sits next to the nephelometer during all calibration and warm-up of the instrument, it is unlikely that the ventilation in the room can reduce all CO_2 in the room air before entering the nephelometer. A human generates a CO_2 amount of 17 l/hour, according to [18] in rooms at the University where one or more people are present at the same time the CO_2 amount shall not exceed 800 ppm (CO_2 amount outside is 350-400 ppm), whereas in auditorium 1000 ppm CO_2 is accepted. A rough estimation of the

CO_2 increase generated by one person in the room during a one hour calibration is shown in

$$\frac{17}{1000} \cdot \frac{1}{5 \cdot 5 \cdot 3} = 0.000227 = 227ppm \quad (4.1)$$

The calculation in equation (4.1) results in a 230 ppm increase of CO_2 per hour, if no ventilation is present in the room (there is ventilation in the room, thus this will not be able to remove all the CO_2 , H_2O before entering the nephelometer since I was located 30 cm from the nephelometer, the ventilation is not that great).

The corrections made due to negative values: $\frac{(Highest-lowest)value}{10^{-6}}$, when we do this we get rid of the negative values. In the future calibration should be made with tube air instead of room air.

Results

In this section the results from the soot measurements are presented, the first section will explain the OP-7 measurement i.e. the Operation Point that generates the the case with clusters of small primary particles. In the second part OP-1 measurement will be presented i.e. the Operation Point producing aggregates. In the third section all five operation points will be evaluated and compared. The structure of the result sections will be as follows:

- Scattering Ångström Exponent divided into four separate parts :
 - i) Least-squares method
 - ii) Forward scattering multi-range method
 - iii) Backward scattering multi-range method
 - vi) Comparison table for least-squares method and multi-range method.
- Ratio between forward and backward scattering.

In the comparing tables four main columns are presented: N= number of data points, Least-Squares Fit (forward and backward) comparison, Multi-Range Ångström calculated for each time step for all three wavelengths and the last column comment(explaining if the data is OK or Bad, since some data in OP-1 are non trustworthy, this will be explained in section 5.2). In the last section the outdoor measurements will be presented and discussed.

5.1 OP-7

In this section the results for the Operation Point that generates small particles that are not aggregated yet.

5.1.1 Scattering

In figure 5.1 the scattering signal from two different measurements is shown. An increasing trend in the signal is present in both measurements, whereas different scattering intensities between the three wavelengths are present as well. The signal are similar for the two measurements and the same separation features can be seen in both measurements displayed in the figure.

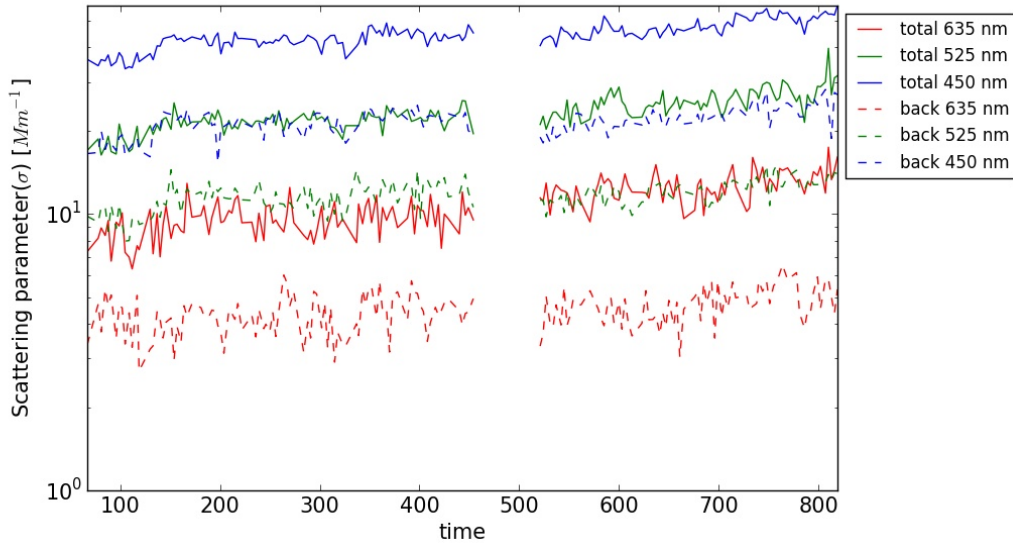


Figure 5.1: Illustration of OP-7 scattering.

5.1.2 SAE

In this section the Scattering Ångström Exponent will be evaluated using the two different methods explained in figure 2.4, least-squares method and multi-range method.

5.1.2.1 Least-squares method

In figure 5.2 the SAE is presented for forward and backward scatter. The variation of SAE over time is kept and the strength of the color for the calculated SAE is reduced, a mean value with standard deviation is included for each measurement. The reason the calculated values are kept in the figures is that this gives the figure a new dimension and it is also easier to see how SAE varies over the measurement. This figure provides more information than just a mean value figure over the two measurements, which would only have generated two values per measurement. For forward scattering values of SAE varies from 4.14 ± 0.95 and 3.43 ± 0.65 . Whereas for the backward scattering 4.67 ± 0.52 and 4.56 ± 0.44 is obtained. This tell us that we obtain larger values of SAE in the backward scattering direction. SAE=4 corresponds to small particles and also a strong wavelength dependence. Why we obtain larger SAE values in the backward direction will be explained in the future with complementary mathematical simulation and analysis.

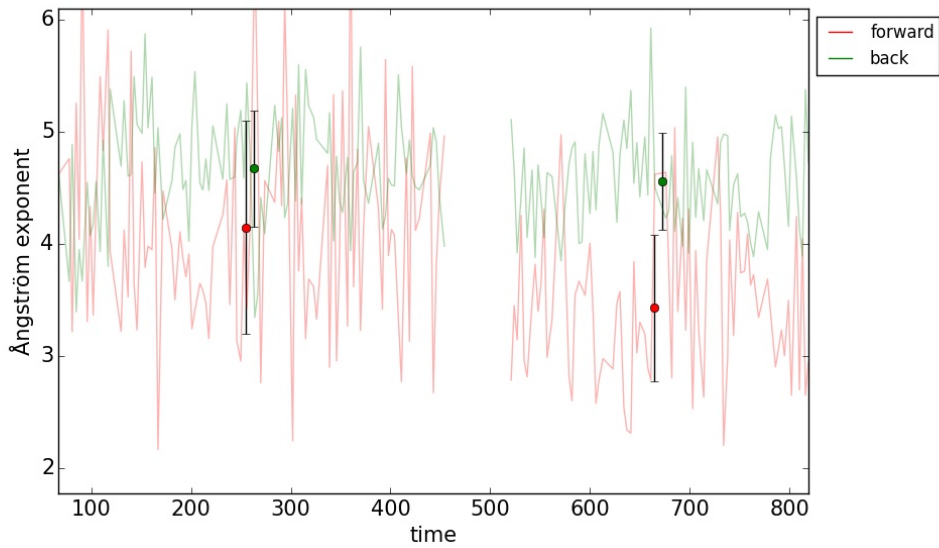


Figure 5.2: Illustration of OP-7 least squares method.

5.1.2.2 Forward scattering, multi-range method

In figure 5.3 the two events with OP-7 are displayed, a clear separation trend between the three wavelengths i.e. 635 nm, 525 nm and 450 nm can be observed. Where 525/635 nm has the lowest SAE in both cases, second highest SAE is the 450/635 nm and the highest wavelength couple is 450/525 nm. In table 5.1 the variations in SAE are presented, there is a variation in SAE that lies around 4, which corresponds to small particles being present in the measurement volume. From TEM images over OP-7 and OP-1 we know that OP-7 is not yet aggregated particles, they are most likely small ones, i.e. are hence expected to have SAE of 4.

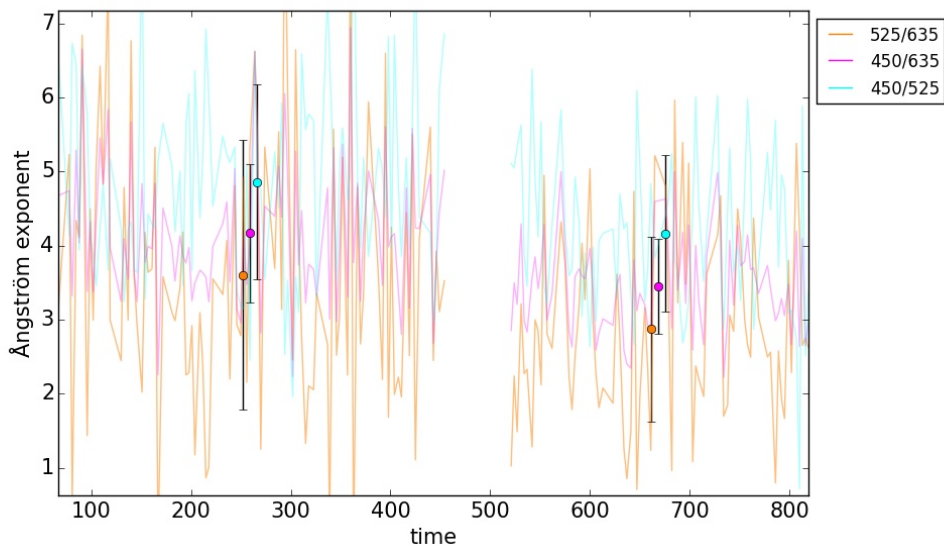


Figure 5.3: Illustration of OP-7 SAE forward scattering.

5.1.2.3 Backward scattering, multi-range method

In figure 5.4 an opposite trend in SAE compared to figure 5.3 is observed. The highest SAE value is for 525/635 nm, then 450/635 nm and the lowest 450/525 nm. In table 5.1 the variation of SAE is presented.

SAE varies from 2.87 to 4.16 in the backward scatter direction, which basically corresponds to small particles scattering light.

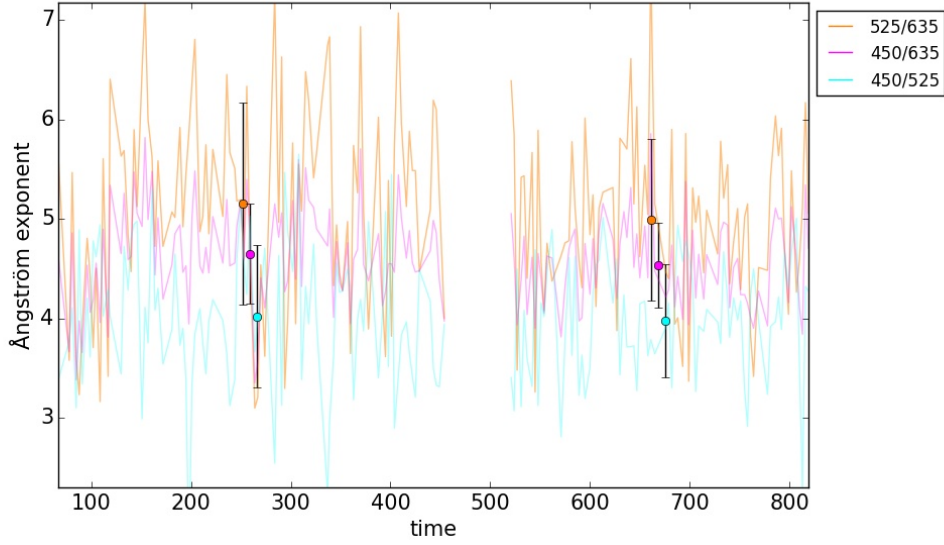


Figure 5.4: Illustration of OP-7 SAE backward scattering.

Table 5.1: Comparison of OP-7 Ångström Exponent variation.

N	Least Squares Fit		Multi Range Ångström			Comment
	Forward	Back	Wavelengths	Forward	Back	
98	4.14 ± 0.95	4.67 ± 0.52	450/525	4.86 ± 1.31	4.02 ± 0.72	OK
			450/635	4.17 ± 0.93	4.65 ± 0.50	
			525/635	3.61 ± 1.82	5.16 ± 1.01	
79	3.43 ± 0.65	4.56 ± 0.44	450/525	4.16 ± 1.06	3.98 ± 0.57	OK
			450/635	3.45 ± 0.64	4.54 ± 0.43	
			525/635	2.87 ± 1.24	4.99 ± 0.81	

5.1.3 Ratio

In figure 5.5 the relative scattering between forward and backward is shown for the two OP-7 measurements. We see that a separation between the 635 and 525, 450 nm is present, this corresponds to a slightly larger amount of scattering in the forward direction, in the first case the ratio for 635 nm is 1.25, whereas for the second event it is 1.75.

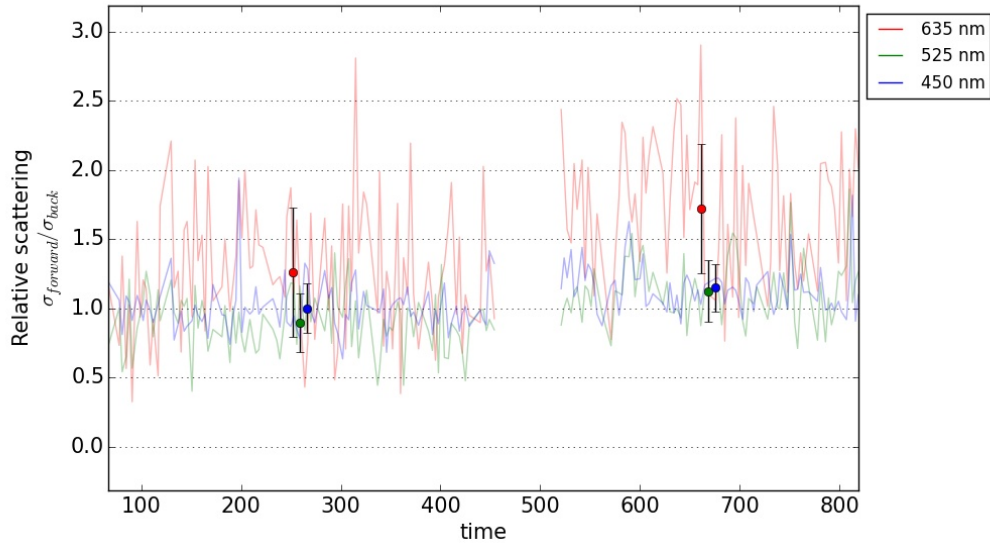


Figure 5.5: Illustration of OP-7 ratio of forward and backward scattering.

5.2 OP-1

In this section the results for the most sooty case is presented, i.e. OP-1. This Operation points consist of aggregated soot particles i.e. large particles.

5.2.1 Scattering

In figure 5.6 the scattering for two OP-1 cases are presented. Since this is the sootiest case it was difficult to obtain a stable signal during a longer period of time, this because the dilution system is not optimal for this measurement, it do not provide a continuous and stable flow into the nephelometer. Since the concentration at some times increased very rapidly from low concentration to very high this might contaminate the nephelometers measuring volume. Due to these difficulties keeping a stable signal the air dilution was changed at some times during the measurements, which is indicated in figure 5.6 with segmented measuring values. The first three values corresponds to the first measurement (0-210 seconds), whereas the next five corresponds to the second measurement (220-450 seconds). In table 5.2 in the comment field a notation over if the measurement is Ok or Bad is explained. If the measurement is denoted as ok this will be used when analyzing and calculating SAE for the OP-case. Whereas if it is denoted as bad it will not be included in the evaluation between all five OP-cases in section 5.3.

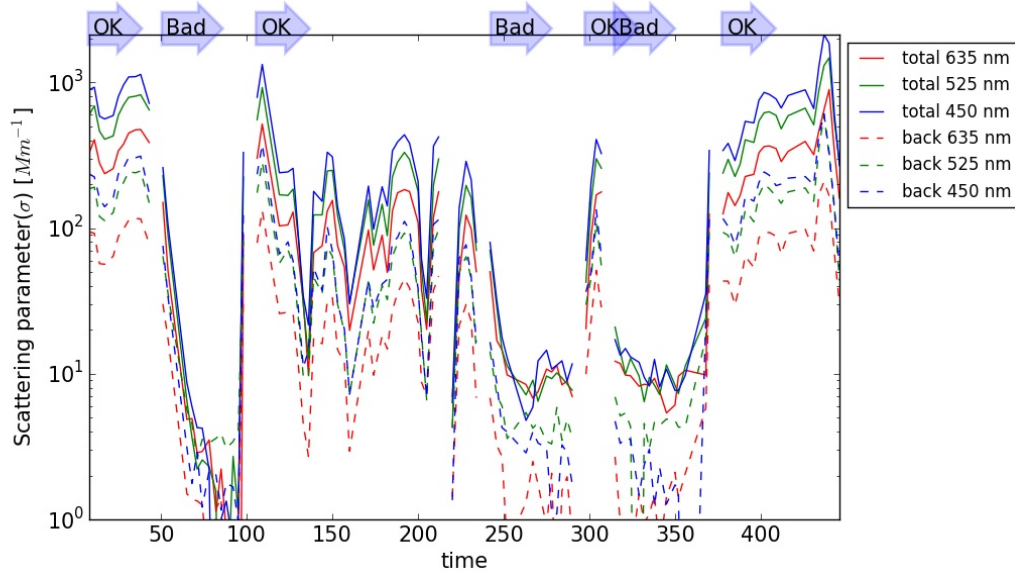


Figure 5.6: Illustration of OP-1 scattering.

5.2.2 SAE

In this section the Scattering Ångström Exponent will be evaluated using the two different methods explained in figure 2.4, least-squares method and multi-range method.

5.2.2.1 Least-squares method

In figure 5.7 the OP-1 soot SAE's are presented and calculated least-squares method. The SAE variation is presented in table 5.2 SAE in the forward direction varies between 1.87 and 2.28 for the "ok" data, whereas for the backward scattering SAE varies between 2.5 and 2.78. The SAE values are around 2 and according to theory the SAE decreases with increasing particle sizes, we know that for small particles $SAE=4$.

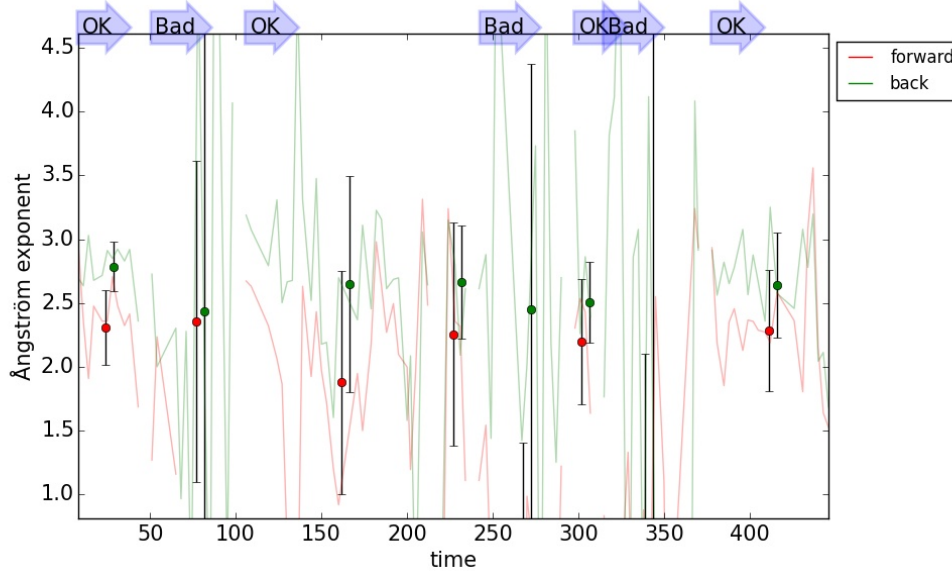


Figure 5.7: Illustration of OP-1 least squares method.

5.2.2.2 Forward scattering, multi-range method

In figure 5.8 the variation of SAE for forward scatter is shown. For the data that is labeled with ok in the comment field in table 5.2 similar appearance with low standard deviations. Whereas the data with "bad" label have large standard deviations, and are not trustworthy data. The data that is ok has SAE around 2 which according to theory corresponds to large particles or aggregates, this correspond to what we know about OP-1 from figure 3.4a.

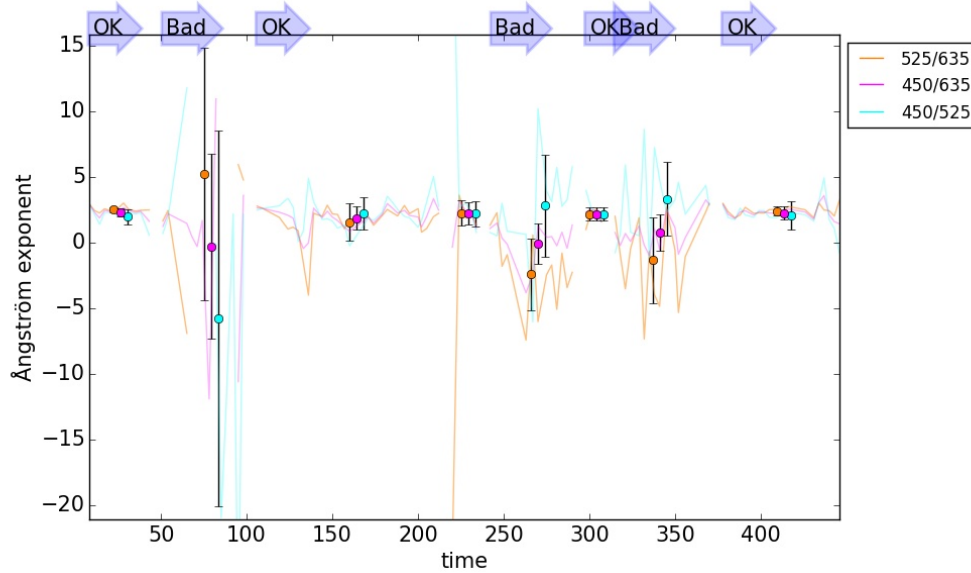


Figure 5.8: Illustration of OP-1 SAE forward scattering.

5.2.2.3 Backward scattering, multi-range method

In figure 5.9 the SAE variation for backward scatter are shown. It is clear that the bad data have large standard deviations and large error bars in the figure 5.9. Whereas the ok data have smaller error bars, the variation in SAE is shown in table 5.2. The variation is large, between 1.16 and 3.96. This large span of SAE might indicate both small non aggregated particles (SAE=4) and aggregated particles (lower SAE).

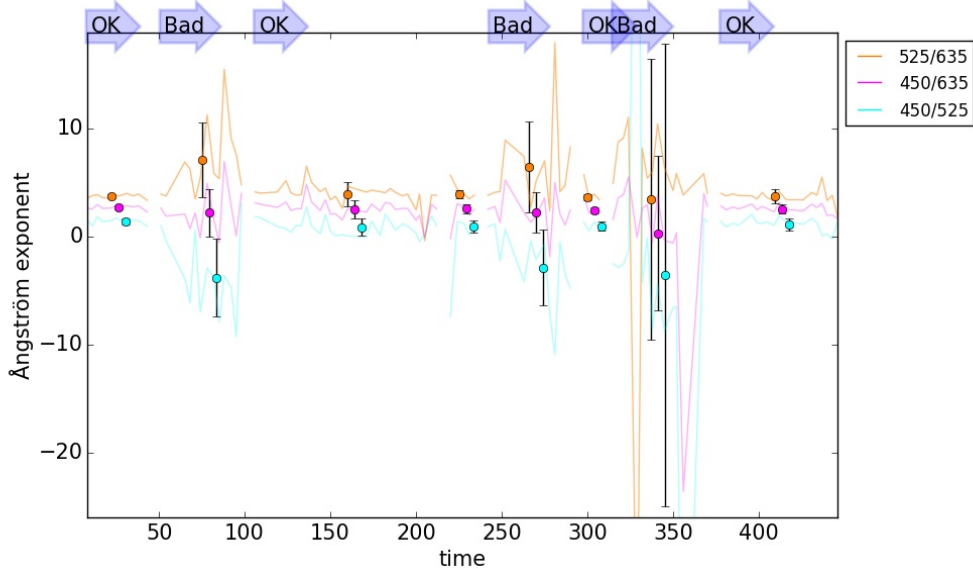


Figure 5.9: Illustration of OP-1 SAE backward scatter.

Table 5.2: Comparison of OP-1 Ångström Exponent variation.

N	Least Squares Fit		Multi Range Ångström			Comment
	Forward	Back	Wavelengths	Forward	Back	
10	2.30 ± 0.30	2.78 ± 0.19	450/525	2.00 ± 0.57	1.46 ± 0.27	OK
			450/635	2.30 ± 0.30	2.74 ± 0.20	
			525/635	2.54 ± 0.20	3.78 ± 0.19	
12	2.35 ± 1.26	2.44 ± 2.23	450/525	-5.77 ± 14.33	-3.79 ± 3.59	Bad
			450/635	-0.26 ± 7.05	2.25 ± 2.22	
			525/635	5.25 ± 9.64	7.14 ± 3.44	
27	1.87 ± 0.88	2.65 ± 0.85	450/525	2.26 ± 1.22	0.90 ± 0.77	OK
			450/635	1.89 ± 0.87	2.59 ± 0.84	
			525/635	1.58 ± 1.43	3.97 ± 1.14	
4	2.25 ± 0.88	2.66 ± 0.44	450/525	2.22 ± 0.98	0.95 ± 0.54	OK
			450/635	2.25 ± 0.88	2.61 ± 0.45	
			525/635	2.28 ± 0.98	3.96 ± 0.39	
12	-0.15 ± 1.56	2.45 ± 1.93	450/525	2.83 ± 3.89	-2.86 ± 3.51	Bad
			450/635	-0.06 ± 1.57	2.29 ± 1.88	
			525/635	-2.41 ± 2.75	6.45 ± 4.20	
3	2.20 ± 0.49	2.50 ± 0.32	450/525	2.20 ± 0.52	0.97 ± 0.43	OK
			450/635	2.20 ± 0.49	2.46 ± 0.32	
			525/635	2.20 ± 0.50	3.66 ± 0.28	
14	0.69 ± 1.41	0.46 ± 6.98	450/525	3.36 ± 2.79	-3.53 ± 21.47	Bad
			450/635	0.77 ± 1.37	0.34 ± 7.18	
			525/635	-1.33 ± 3.25	3.47 ± 13.05	
18	2.28 ± 0.48	2.64 ± 0.41	450/525	2.10 ± 1.07	1.16 ± 0.59	OK
			450/635	2.28 ± 0.49	2.59 ± 0.41	
			525/635	2.43 ± 0.36	3.75 ± 0.67	

5.2.3 Ratio

In figure 5.10 the ratios between forward and backward scattering are shown for the OP-1 measurements. For the ok data a clear trend between 635, 525 and 450 nm is present. For 635 nm the ratio varies between 3 and 3.5. For 525 nm the ratio is 3.2 to 3.6. Whereas for 450 nm the ratio varies between 2.1 and 2.25 i.e. more scattering in the forward direction than backwards. The order of wavelengths are unexpected, 635 nm is at the top then 525 nm and last 450 nm, the 635 nm was thought to be at the

bottom. Perhaps this is connected to the larger values of SAE's in backward direction as well.

Hence more scattering in the forward direction for all three wavelengths, this corresponds well with theory which state that large particles preferably scatters in the forward direction.

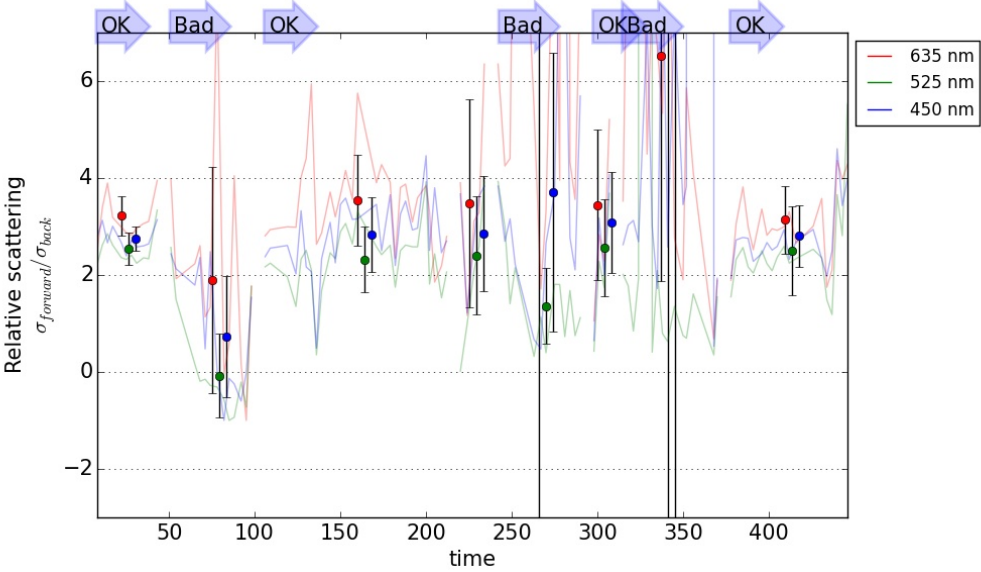


Figure 5.10: Illustration of OP-1 ratio of forward and backward scatter.

5.3 Comparison OP-cases

In this section all five Operation Points will be compared. The data presented in all figures in this section will start with OP-7 i.e. the least sooty case and also the smallest particles and then with increasing size resulting in OP-1 i.e. the sootiest and also the OP-case with largest particles.

5.3.1 SAE comparison

In this section the scattering ångström exponent will be compared and evaluated between the different OP-cases.

5.3.1.1 Least-squares method

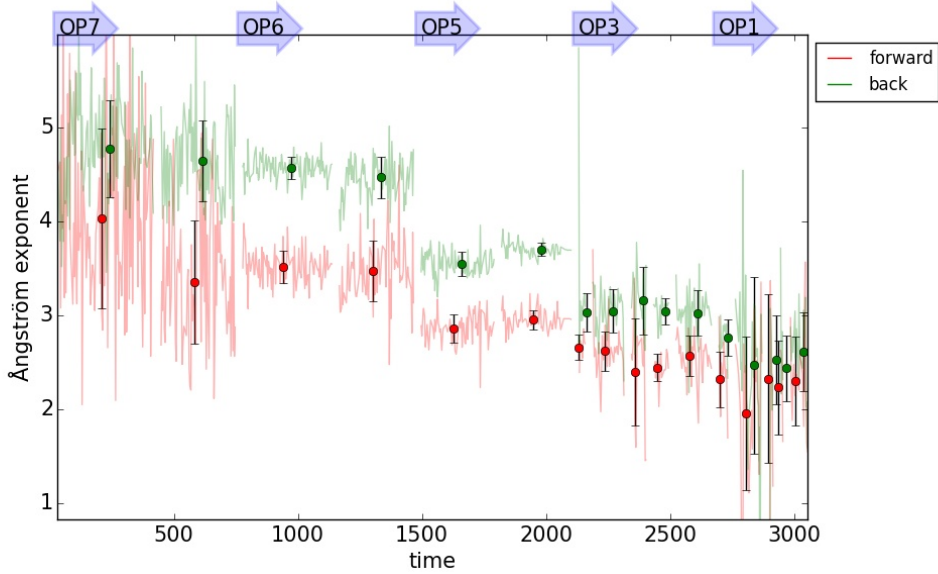


Figure 5.11: Illustration of all OP-cases SAE with least squares method.

In figure 5.11 the SAE calculated with least-squares method for OP-7, OP-6, OP-5, OP-3 and OP-1 are presented for forward and backward scatter. A decreasing trend with increasing particle size is present in both the forward and the backward scattering direction. This trend corresponds well with theory that says that small particles have large SAE's typically SAE=4 and large particles have lower SAE's. Hence from TEM images we know that OP-1 consist of large aggregates and OP-7 of small particles and OP-6 is similar with OP-7. OP-3 is similar to OP-1 i.e. large particles, OP-5 is in between small particles and aggregates. However the backward scattering have larger SAE than the forward scattering, which will be evaluated in the future. A clear separation between forward and backward scatter is present for all five OP-cases.

5.3.1.2 Forward scattering, multi-range method

In figure 5.12 SAE for forward scattering calculated with multi-range are shown for OP-7, OP-6, OP-5, OP-3 and OP-1. The variation of SAE for the OP-cases are shown in table 5.3. A clear decreasing trend in SAE with increasing particle size is present here as well as in figure 5.11. For the small particles in OP-7 SAE=4 and for OP-1 SAE around 2. From table 5.3 we can see that OP-7 is the OP-case with the largest standard deviation: an explanation of that could be that during one measuring day the flame went out at several times (not in the data presented in this thesis), this indicates that the combustion of this point is not optimal nor stable. Hence various concentration of particles are lead into the nephelometer and measured on, which results in a larger standard deviation in the calculated SAE's.

In figure 5.12 there is a clear wavelength dependence, i.e. for OP-7 and OP-6 we have the same trend in the figure i.e a stronger wavelength dependence for small particles (from bottom to top: $\frac{525}{635}$, $\frac{450}{635}$, $\frac{450}{525}$). . This corresponds well with theory. For OP-5 on the other hand we see that the three wavelengths lie in

a row i.e. no wavelength dependence, the soot particles are about to change at this operation point and approaching aggregates instead of small individual particles as in OP-7 and OP-6. Whereas for OP-3 and OP-1 the inverse trend from OP-7 and OP-6 is observed (bottom to top: $\frac{450}{525}, \frac{450}{635}, \frac{525}{635}$).

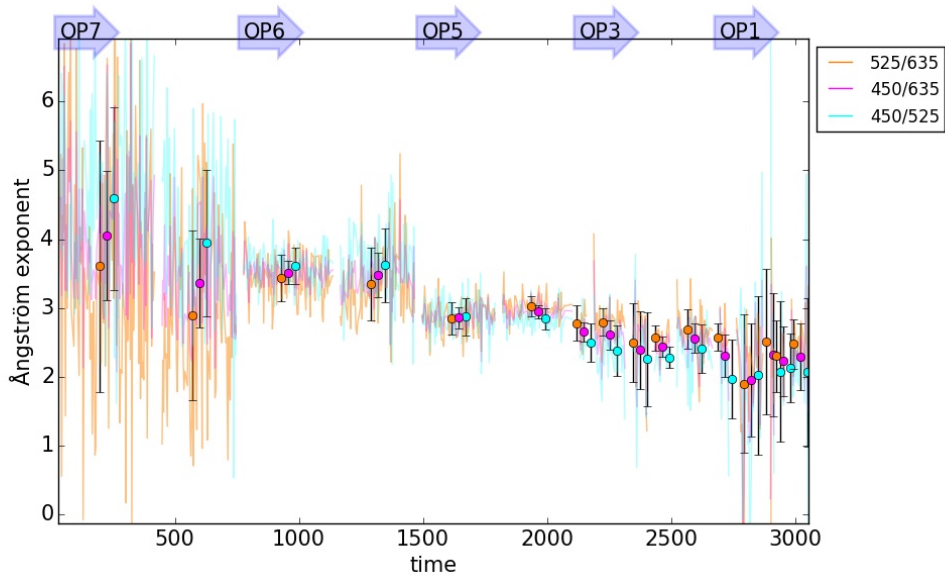


Figure 5.12: Illustration of all OP-cases SAE with multi range method for forward scattering.

5.3.1.3 Backward scattering, multi-range method

In figure 5.13 the variation of all OP-cases in SAE in backward direction are shown. An decreasing trend in SAE-values for increasing particle size is also present here. Although a separation between the three wavelengths 635 nm, 525 nm and 450 nm is present. Anyhow for OP-7 a larger SAE is present than for OP-1 which indicates smaller particles in the first case and larger ones in the last. In backward scattering direction there are the same wavelength trends for all OP-cases (bottom to top: $\frac{450}{525}, \frac{450}{635}, \frac{525}{635}$) there is no change at OP-5 as there is for the forward scattering.

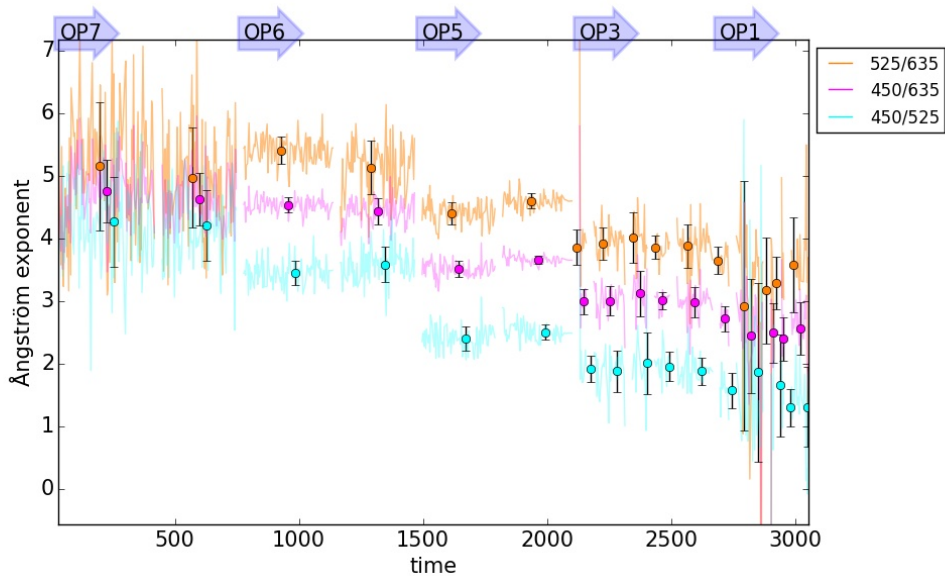


Figure 5.13: Illustration of all OP-cases SAE with multi-range method for backscattering.

Table 5.3: Comparison of all OP-cases Ångström Exponent variation.

N	Least Squares Fit		Multi Range Ångström			Comment
	Forward	Back	Wavelengths	Forward	Back	
96	4.03 ± 0.96	4.77 ± 0.52	450/525	4.59 ± 1.32	4.27 ± 0.71	OP7
			450/635	4.05 ± 0.93	4.76 ± 0.50	
			525/635	3.61 ± 1.83	5.15 ± 1.02	
77	3.35 ± 0.66	4.65 ± 0.43	450/525	3.95 ± 1.06	4.21 ± 0.56	OP7
			450/635	3.37 ± 0.64	4.63 ± 0.42	
			525/635	2.90 ± 1.23	4.97 ± 0.80	
89	3.51 ± 0.18	4.57 ± 0.12	450/525	3.61 ± 0.26	3.46 ± 0.19	OP6
			450/635	3.52 ± 0.17	4.54 ± 0.12	
			525/635	3.44 ± 0.33	5.41 ± 0.22	
74	3.47 ± 0.33	4.47 ± 0.22	450/525	3.62 ± 0.53	3.59 ± 0.28	OP6
			450/635	3.47 ± 0.32	4.44 ± 0.21	
			525/635	3.35 ± 0.53	5.13 ± 0.43	
75	2.86 ± 0.15	3.55 ± 0.13	450/525	2.87 ± 0.28	2.41 ± 0.20	OP5
			450/635	2.86 ± 0.15	3.51 ± 0.13	
			525/635	2.85 ± 0.24	4.41 ± 0.18	
71	2.95 ± 0.10	3.70 ± 0.07	450/525	2.85 ± 0.15	2.51 ± 0.12	OP5
			450/635	2.95 ± 0.10	3.66 ± 0.07	
			525/635	3.03 ± 0.15	4.60 ± 0.12	
8	2.66 ± 0.14	3.03 ± 0.20	450/525	2.50 ± 0.28	1.92 ± 0.21	OP3
			450/635	2.65 ± 0.14	2.99 ± 0.20	
			525/635	2.78 ± 0.26	3.86 ± 0.28	
33	2.62 ± 0.21	3.04 ± 0.23	450/525	2.38 ± 0.37	1.88 ± 0.33	OP3
			450/635	2.61 ± 0.22	3.01 ± 0.23	
			525/635	2.79 ± 0.20	3.92 ± 0.26	
15	2.40 ± 0.57	3.16 ± 0.36	450/525	2.26 ± 0.69	2.02 ± 0.49	OP3
			450/635	2.39 ± 0.57	3.12 ± 0.36	
			525/635	2.50 ± 0.57	4.02 ± 0.40	
15	2.44 ± 0.15	3.04 ± 0.14	450/525	2.28 ± 0.15	1.96 ± 0.23	OP3
			450/635	2.44 ± 0.14	3.01 ± 0.14	
			525/635	2.56 ± 0.18	3.86 ± 0.18	
35	2.57 ± 0.21	3.02 ± 0.25	450/525	2.40 ± 0.35	1.89 ± 0.22	OP3
			450/635	2.56 ± 0.21	2.99 ± 0.24	
			525/635	2.69 ± 0.29	3.88 ± 0.35	
10	2.32 ± 0.30	2.76 ± 0.20	450/525	1.97 ± 0.57	1.58 ± 0.28	OP1
			450/635	2.31 ± 0.30	2.73 ± 0.20	
			525/635	2.58 ± 0.20	3.65 ± 0.22	
27	1.95 ± 0.82	2.47 ± 0.94	450/525	2.02 ± 1.15	1.87 ± 1.42	OP1
			450/635	1.96 ± 0.82	2.45 ± 0.91	
			525/635	1.90 ± 1.00	2.93 ± 1.99	
4	2.32 ± 0.90	2.52 ± 0.48	450/525	2.08 ± 1.01	1.66 ± 0.82	OP1
			450/635	2.32 ± 0.89	2.50 ± 0.47	
			525/635	2.51 ± 1.06	3.18 ± 0.84	
3	2.23 ± 0.50	2.43 ± 0.35	450/525	2.13 ± 0.50	1.31 ± 0.30	OP1
			450/635	2.23 ± 0.50	2.40 ± 0.35	
			525/635	2.30 ± 0.52	3.29 ± 0.42	
18	2.30 ± 0.48	2.61 ± 0.42	450/525	2.07 ± 1.08	1.32 ± 0.63	OP1
			450/635	2.29 ± 0.49	2.57 ± 0.41	
			525/635	2.48 ± 0.35	3.58 ± 0.75	

5.3.1.4 Ratio

In figure 5.14 the ratio between forward and backward scattering for the five OP-cases are shown. From left to right i.e. small to aggregated particles an increasing trend is detected. A separation between 635

nm and the other two wavelengths is present. As the particle size increases the ratio increases as well, when a ratio above 1 is reached, this means that the forward scattering is higher than the scattering in the backward direction.

Hence a clear increase in forward scatter with increasing particle size is displayed in the figure 5.14. This corresponds well with theory that says that small particles scatters equally in forward and backward direction and larger ones scatter preferably in the forward direction.

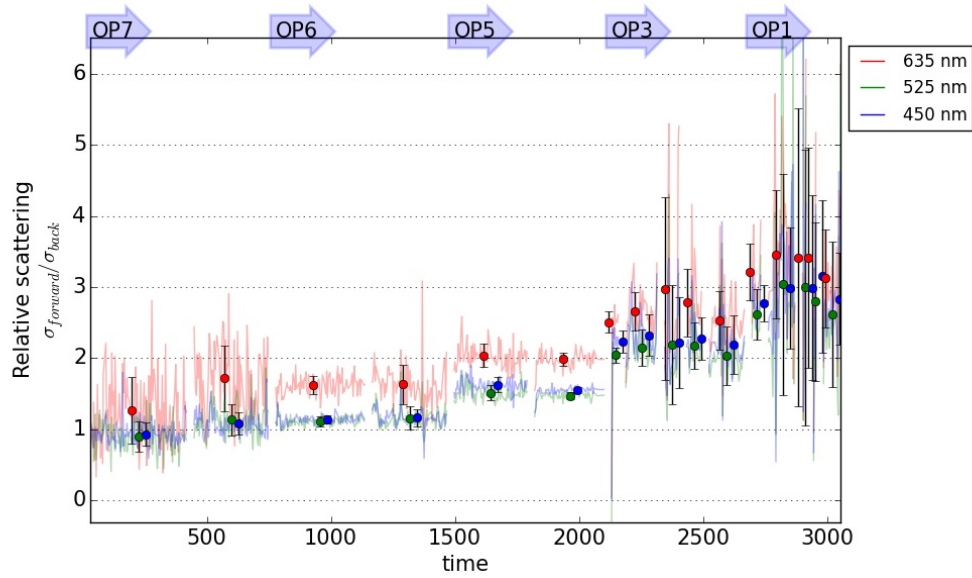


Figure 5.14: Illustration of all OP-cases ratio between forward and backward scattering.

5.4 Outdoor measurement

In this section two independent outdoor measurements are shown. These measurements are performed with Kalman filter, this to show how the Nephelometer is typically used. The two measuring days were completely different from each other, during the first day of measurements it was a lot of fog outside, whereas the second measurement day was a sunny day without clouds present.

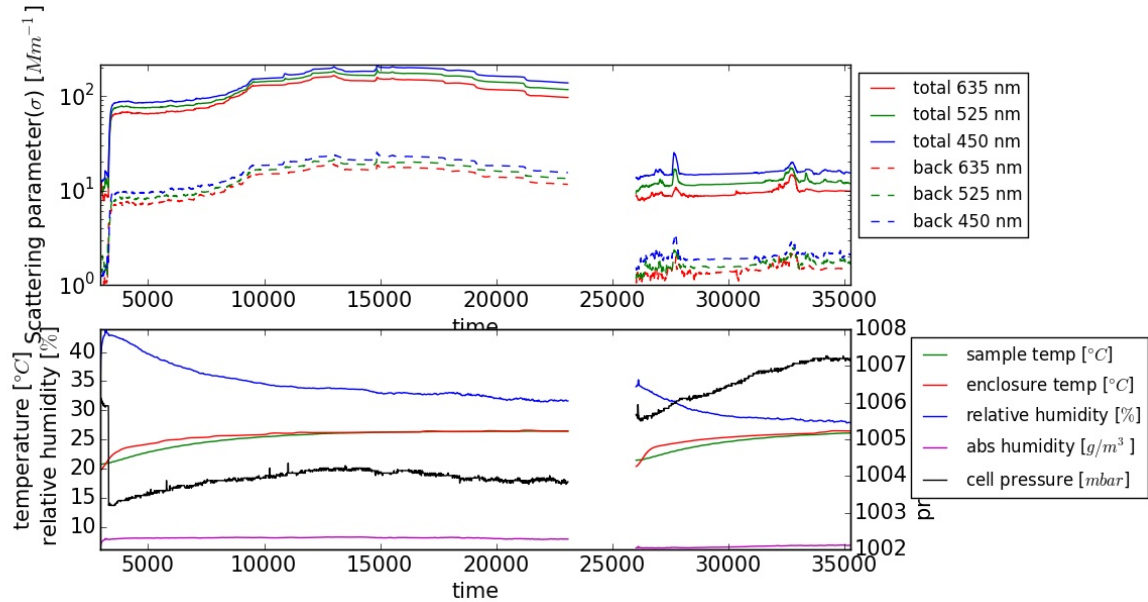


Figure 5.15: Illustration of outdoor measurement.

In figure 5.15 the two days of outdoor measurements are shown. It is clear that the first measurement has much higher scattering signal than the second one. During the first measurement there were a lot of fog outside and the visibility were almost zero, due to all fog. In this figure the scale is logarithmic, to show the wavelength separation between total and backscattering for the two events. However since if the scale is not in logarithmic it is hard to see the small variations when the signal strength between the two events have large deviations. The same separation between total and backscatter is present for the two measurements. The environment is included in a subplot for this measurement, this for clarification on how the weather actually was during the measurement.

However, since the measurements were made with Kalman filter, which basically filters away any quick or large deviation, the data from this measurement looks smooth and even.

In figure 5.16 the scale is linear instead of logarithmic as in figure 5.15, the separation between total and backscatter for the two events compared to each other is much harder to see in this figure, due to the scale. It is though much clearer in the first event how much fog that was present that day, the signal that looked slightly increasing in figure 5.15 is instead very increasing in figure 5.16 instead. Therefore it is of great importance to use the right scale to be able to see everything. Different scales show different features.

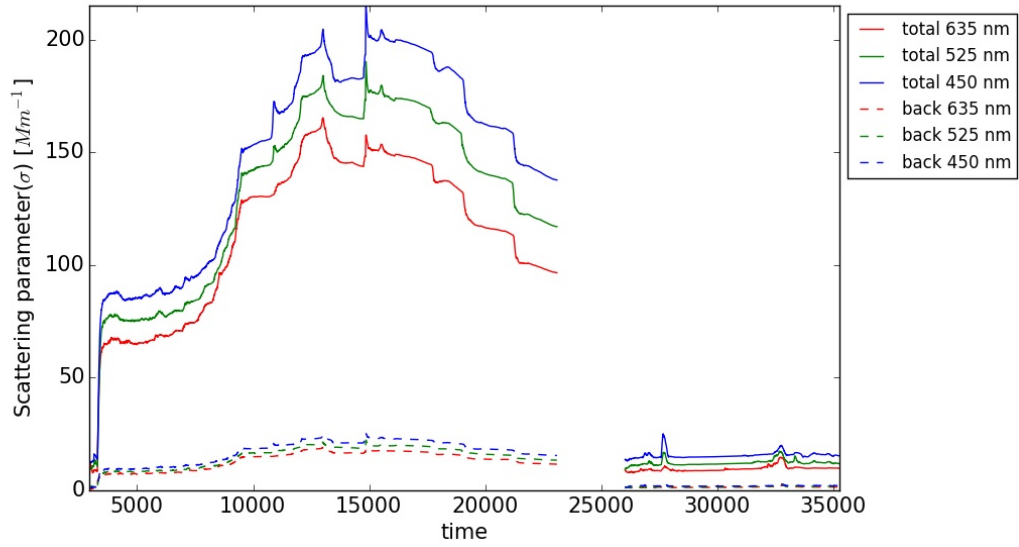


Figure 5.16: Illustration of outdoor measurement.

5.4.1 SAE

5.4.1.1 Forward scattering, multi-range method

In this section data presented for outdoor measurement SAE calculated with multi range method are shown. In the first measurement in figure 5.17 we see that this SAE approaching the same trend as

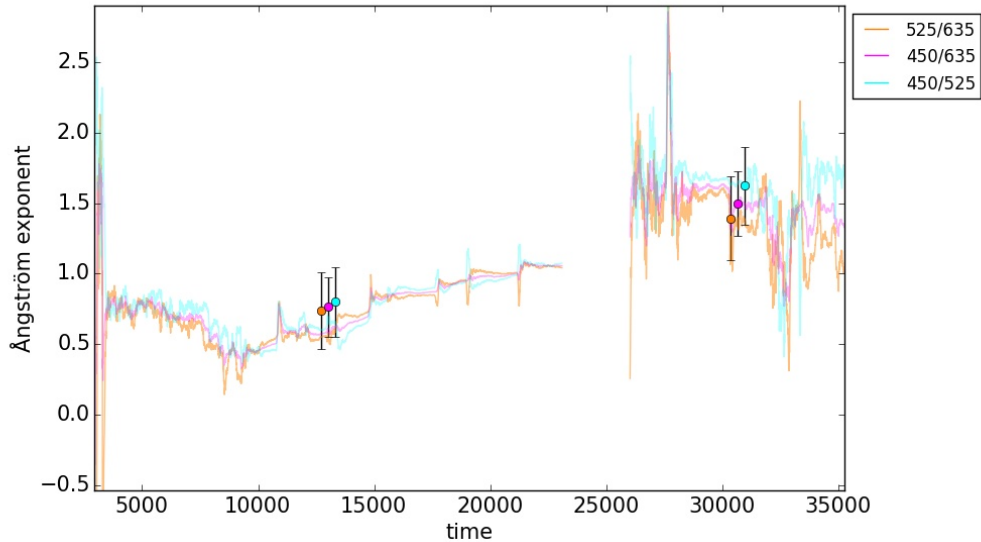


Figure 5.17: Illustration of SAE forward scattering outdoor measurement.

OP-5 event measurement in figure 5.12. Hence approaching larger particles or aggregated ones, and the wavelength dependence is decreasing. In the second outdoor event the trend is similar to the trend observed for OP-7 and OP-6 figure 5.12 i.e. there are smaller particles present than in the first outdoor measurement than in the second measurement. From the values of SAE we know that larger particles have lower SAE, i.e. in the first outdoor measurement there are larger particles present than in the second one. However since $SAE \neq 4$ we know that the particles are not small in that sense, but still smaller than in the first outdoor event.

5.4.1.2 Backward scattering, multi-range method

In this section data presented for the outdoor measurement SAE calculated with multi-range method are shown.

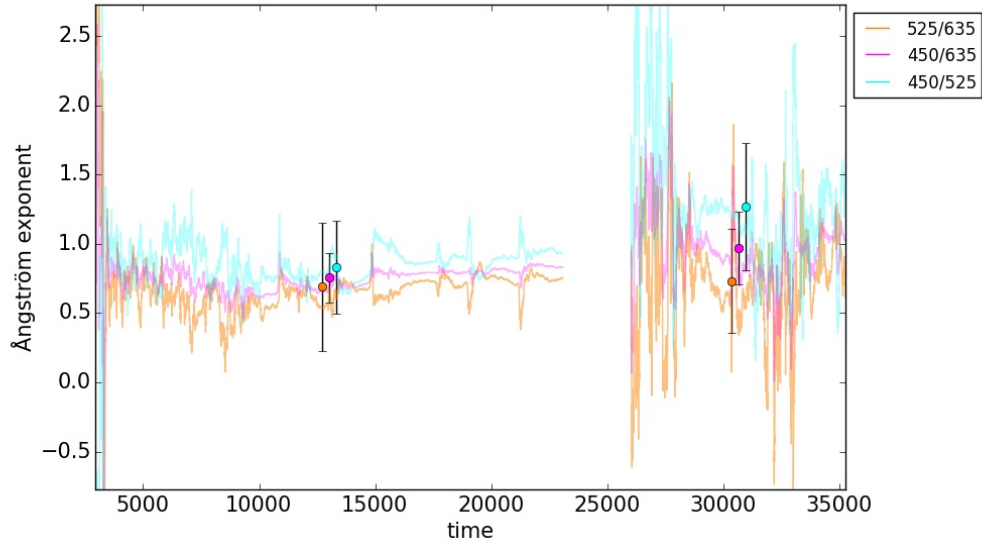


Figure 5.18: Illustration of SAE backward scatter outdoor measurement.

In figure 5.18 a larger separation between the three wavelengths is present. Otherwise there are no extreme difference in size from the SAE shown in figure 5.17. There are though larger fluctuations in the second event in backward direction that were not as distinct in the forward SAE.

However we can see that SAE is almost the same for forward and backward direction and that corresponds well to theory i.e. small particles scatters equal in forward and backward direction.

Discussion

The focus in this thesis has been to commissioning the nephelometer and perform measurements on soot particles generated by a miniCAST soot generator. Where five different types of soot have been studied: OP-7 small particles, OP-6 soot of same type as OP-7, OP-5 in between the single particles going towards aggregated states, OP-3 and OP-1 generates aggregated soot.

OP-7

From the SAE results of OP-7 a value of SAE=4 is obtained, this means small particles according to theory. From TEM imaging we know that OP-7 soot consists of small clusters of primary particles much smaller than the illuminating wavelengths used in this thesis. The ratio of this operation point is equal to 1, which indicates that OP-7 are small particles and lie in the Rayleigh regime. What is still unclear and will be evaluated by mathematical models in the future and theory is why a larger SAE in the backward direction is obtained, typically SAE=4.02-5.2. Small particles such as them in OP-7 scatters accordingly to theory an equal amount in forward and backward direction.

OP-1 and OP-3

The OP-1 measurements along with OP-3 were the most difficult measurement for obtaining smooth and stable signals. When they were first lead into the nephelometer the instrument generated a smooth increasing signal and then suddenly the signal increased massively, this due to a non optimal air dilution system. When there was an increase the air dilution was increased not to contaminate the nephelometer with aggregated soot particles. This is the reason why there are so many fluctuations in the OP-3 and OP-1 measurements. All OP-1 and OP-3 data presented in this thesis, however it should be noted that some time intervals generated non-trustworthy data due to rapid concentration changes. This should be taken into account when making future measurements. In the analysis the non-trustworthy data is not included.

Outlier removal and unexpected results Instead of removing the values where the air dilution flow has been changed I choose to keep all the data in my results, but instead divide it into "ok" and "bad" parts. In this way the data is not ruined. When the results are divided like this SAE's for OP-1 and OP-3 resulted in good values i.e. SAE=2, when first analyzing and observing the OP-1 data the SAE results were not that great. Hence after this reconstruction of dividing the data into ok and bad data improved for the final result.

From TEM imaging of OP-1 we know that this operation point produces aggregated soot particles, i.e. large particles in that scene, for larger particles the SAE's decrease, therefore SAE=2 corresponds well with theory. However a larger value than expected in the backward scattering direction is obtained for OP-1 as well as for OP-7, SAE=1.5-3.8.

Comparison of least squares method and multi range method

In this thesis the SAE's have been calculated with two different methods: least squares method and multi range method. Least squares method basically builds on a specific type of function: $\frac{1}{\lambda^\alpha}$, where a line is fitted to the three wavelengths in this thesis work, i.e. 635 nm, 525 nm and 450 nm, for each time step where the nephelometer measure and generate data. If the three wavelengths have a large spread and are not located at at the fit, then this method provides an unwanted result.

On the other hand the multi-range method as it is call in this thesis the calculate SAE in each time step

for all possible wavelength pairs are presented, i.e. $\frac{450}{635}, \frac{450}{525}, \frac{525}{635}$. This method is better than the least squares method since it takes into account the wavelength dependence in the different spectral regions. The wavelength pair $\frac{450}{635}$ is basically just a mean value of the two other wavelength pairs and basically corresponds to the least squares methods value of the measuring points.

Comparison of all OP-cases

Five different OP-cases have been presented and evaluated. In figure 5.11 an decreasing trend in SAE is present for increasing particle size, which is accordingly to theory, i.e. larger particles equals lower SAE's. Data from the backward scattering results in a higher SAE than the forward direction. There is a separation between backward and forward scatter that is present for all the five different OP-cases and it looks like this separation is the same for all five cases.

In figure 5.12 the SAE for all five OP-cases in the forward direction are shown, a clear decreasing trend in SAE for increasing particle size is observed. From the figure we can see that OP-7 and OP-6 looks the same whereas something happens in OP-5, SAE is constant. Probably this OP-case is where the particles starts to create aggregates and will look more like OP-3 and OP-1, this state is somewhere in between single particles and aggregated ones. Hence for the next two OP-cases i.e. OP-3 and OP-1 another decreasing trend is observed. This trend follows theory very well, for small particles SAE=high=4, whereas for larger particles SAE decreases, in our case SAE=2. For the OP-cases containing small particles the separation between the SAE wavelength pairs order: bottom to top $\frac{525}{635}, \frac{450}{635}, \frac{450}{525}$, and for OP-3 and OP-1 the following order $\frac{450}{525}, \frac{450}{635}, \frac{525}{635}$, for OP-5 the OP-case that might have with mixed sizes and aggregates both of the explained trends above can be observed.

In figure 5.13 SAE's for backward scatter are shown, where there is a clear wavelength separation in all five OP-cases. The separation is larger than the separation present in figure 5.12 for forward scatter. The order of the wavelength separation is the same for all OP-cases no matter if the particle sizes are small or large, the order is $\frac{450}{525}, \frac{450}{635}, \frac{525}{635}$ (i.e. the same order as for OP-3 and OP-1 forward scatter, bottom to top). The SAE's are slightly higher for all OP-cases in the backward direction, this will be explained in the future.

In figure 5.14 the ratio between forward and backward scatter for the five OP-cases are shown, i.e. the relative scattering. An increasing trend for increasing particle size is observed in the figure, whereas the separation between 635 nm (red wavelength) and 525 nm and 450 nm is more distinct when approaching aggregated large particle sizes such as in OP-3 and OP-1. This trend is a nice result, since it corresponds well with theory that tells us that larger particles such as those in OP-3 and OP-1 scatters light preferably in the forward direction. From the figure we can see that the ratio for OP-7 is 1.25 (for 635 nm and 1 for 525 nm and 450 nm), whereas for OP-1 is 3.5 (for 635 nm, and 3 for 525 nm and 450 nm).

Outdoor measurements

In figure 5.16 and figure 5.15 typical measurements performed with a nephelometer are shown. These types of measurements are the usual way of doing measurements with the instrument, background measurements. The outdoor scattering are present with two different scales i.e. linear scale in the second one and then logarithmic scale in the first figure. This because different features are observed when using different scales, when using logarithmic scale comparison between two highly different measurements in strength can be compared and evaluated, this is much more difficult at a first attempt if the scale is linear. The second figure showing the scattering parameter has linear scale, this is now okay since the features of the measurements have already been observed in the first figure, so now comparison in strength can be made.

A clear separation trend between total scatter and backward scatter are shown in figure 5.15 for the two separate outdoor measurements. The separation is equal between the two events as well. Whereas in figure 5.16 it is much more clear to see the details in the measurements, i.e. in the first event we can see more features what happens that we do not see in figure 5.15. We see an increase in signal in the first event that later on decreases clearly, this is not as clear in figure 5.15, logarithmic scale, as it is in figure 5.16 i.e. the figure with linear scale. So, different scales shows different features.

For SAE in forward direction for outdoor measurements in figure 5.17 for the first event the same trend as observed for OP-5 in figure 5.12 is observed, i.e. the three wavelengths are placed horizontally after each other (SAE=0.8). For the second event the same wavelength dependence as observed for small particles (OP-7 and OP-6) in figure 5.12 is observed (SAE=1.6). This basically means that in the first outdoor event larger particles are present while the second event smaller particles are present. The first event was made during a foggy day whereas the second one was made on a sunny non cloudy day.

For the SAE's in backward direction figure 5.18 the first event SAE=0.8 whereas the second one has

SAE=1.3. A larger separation between the wavelengths are present in the outdoor measurements just as observed in the OP-measurements for backward scatter. Larger fluctuations in SAE are also present in the second outdoor event.

Important results in the thesis work

Since the main purpose of this thesis was commissioning the nephelometer and perform some soot measurement generated by the miniCAST soot generator, gaining as much knowledge about the instrument was the priority. To obtain as good data as possible with the instrument possible was a huge part of the thesis work. Discovering that the filter function could be turned off made huge improvements on what we actually can see in the data generated by the nephelometer. The filter function should only be on while doing background measurements when basically no rapid fluctuations do any good or harm to the measurement itself. In soot measurement we want all the fluctuations and rapid changes to be recorded and displayed in the data. When it comes to the sampling time in the instrument, this is so important for generating good data in soot measurement, this since the soot concentrations can change fast, therefore we want the sampling time to be as short as possible. Per default the nephelometers sampling time is set to five minutes averaging, but can be changed to one minute as well. After correspondence with David Logan on Ecotech it was discovered that this averaging time could be set to ALL, i.e. 10 values/minute, generated by the nephelometer. These two discoveries made the measurements so much better than it would have been with kalman or moving average filter and the sampling time default.

Conclusion

The commissioning of the nephelometer and doing the first soot measurements with the instrument has been made. Important results of this during the thesis work that improved the data was to stop using the Kalman filter and by so obtaining the raw data generated by the nephelometer. The second thing was changing the sampling averaging time from five minutes to one minute and finally to all values, which resulted in 10 values/minute.

From the measurements in this thesis we can deduct that OP-7 the least sooty case as shown in figure 3.4b produces small clusters of primary particles, which is strengthened by the result of $SAE \sim 4$. Whereas for OP-1 the most sooty case illustrated in figure 3.4a produces aggregates with certain specific optical properties, $SAE \sim 2$, where theory predicts that larger particles have smaller SAE's. Why the backward scattering results in larger SAE's than in the forward direction will be explained in the future. The ratio between forward and backward scattering increases with increasing particle size, and a separation between 635 nm and the other two wavelengths i.e. 525 nm and 450 nm is more prominent when approaching larger particles. Also the order of the wavelengths in the ratio figures are unexpected.

Outlook- Future work and improvements

There are several things that can be added to the set-up to obtain improved results in the future, this due this was an experimental set-up.

- For starters a SMPS (scanning Mobility Particle Sizer Spectrometer) should be added to the set-up, to be able to measure the size distribution of the soot particles generated by the miniCAST. If this instrument is connected right after the miniCAST the size of the particles can be measured before they are lead into the nephelometer.
- A second SMPS should be added to the set-up after the sample outlet on the nephelometer i.e. connected to the exhaust pump, then some measurements and comparison with the first SMPS can be made to see if the soot particles age and grow larger and form aggregates inside the nephelometer.
- A flow-meter should be connected before the sample inlet on nephelometer, this to be able to know the exac flow rate that the sample is entering the nephelometer with.
- Calibration should be made with tube air instead of room air, this because the soot experiments do not use lab room air, it only uses tube air. This set-up uses tube air between the operation points (different soot cases studied) to "empty" the optical measuring volume of particles in the nephelometer, If the calibration of the nephelometer is made with tube air that consists of less CO_2 than the laboratory room air the negative values obtained in the data will be reduced.
- Soot particles are strongly absorbing particles in comparison to their scattering ability, so maybe some compensation might have to be made due to this in future measurements.
- All measurements shall be made during a much longer period of time, to obtain better results.
- The dilution has to be optimized to generate a more stable signal, this especially for the sootiest cases OP-3 and OP-1. These plots looks like the most varying and noisy measurements (due to manually air dilution changes). Instead of having a manual air dilution system, one should have a automatic one. This is because the repeatability increases and to reduce the source of error due to human, automatic dilution systems with digital choices are more reliable than a human that sets analog dilution.
- Some reference experiment/measurements should be made from gases with known molecular size such as: $[H_2, He, N_2, O_2, CO_2]$, their molecular size is $\ll \lambda$ (the wavelength of the light source). The data from these measurements can be analyzed and compared with the soot experiments.
- The department cooperates with a mathematician who is doing a mathematical description over how the nephelometer operates and how particles typically behave as they scatter light, the data in this thesis could be a complement to his modeling.

Appendix A

Measurements 1/11 2016

Appendix with information from the measurements performed 1/11 2016. In table A.1 the exact measurement done is presented in detail.

Table A.1: Notation over the events in measurement 1/11 2016.

Time	Event	Filter	Air dilution flow [l/min]
10:15	Room air measurement	No filter	0
10:35	Calibration	No filter	0
11:22	Tube air	No filter	5.0
11:25	Tube air	Kalman	5.0
11:27	Tube air	No filter	5.0
11:31	OP 7	No filter	4.5
11:32	OP 7	No filter	4.45
11:33	OP 7	No filter	4.4
11:38	OP 7	Kalman filter	4.4
11:43	OP 7	Moving average	4.4
11:48	Tube air	No filter	5.0
11:51	OP 6	No filter	4.8
11:57	OP 6	Kalman	4.8
12:02	OP 6	Moving average	4.8
12:07	Tube air	No filter	5.0
12:09	OP 5	No filter	4.75
12:14	OP 5	Kalman	4.75
12:19	OP 5	Moving average	4.75
12:24	Tube air	No filter	5.0
12:27	OP 3	No filter	4.82
12:32	OP 3	Kalman	4.82
12:37	OP 3	Moving average	4.82
12:42	Tube air	No filter	5.0
12:43	OP 1	No filter	4.85
12:48	OP 1	Kalman	4.85
12:53	OP 1	Moving average	4.85
12:58	Tube air	No filter	5.0
13:01	Tube air	Kalman	5.0
13:07	Tube air	Moving average	5.0
13:19	Room air	No filter	0
13:47	Room air	Kalman	0
14:12	Room air	Moving average	0
14:35	End of measurement		

In figure A.1 the environment plot is presented, here the cell temperature, relative humidity, sample temperature and pressure can be observed. Here an entire measurement series is presented, used in

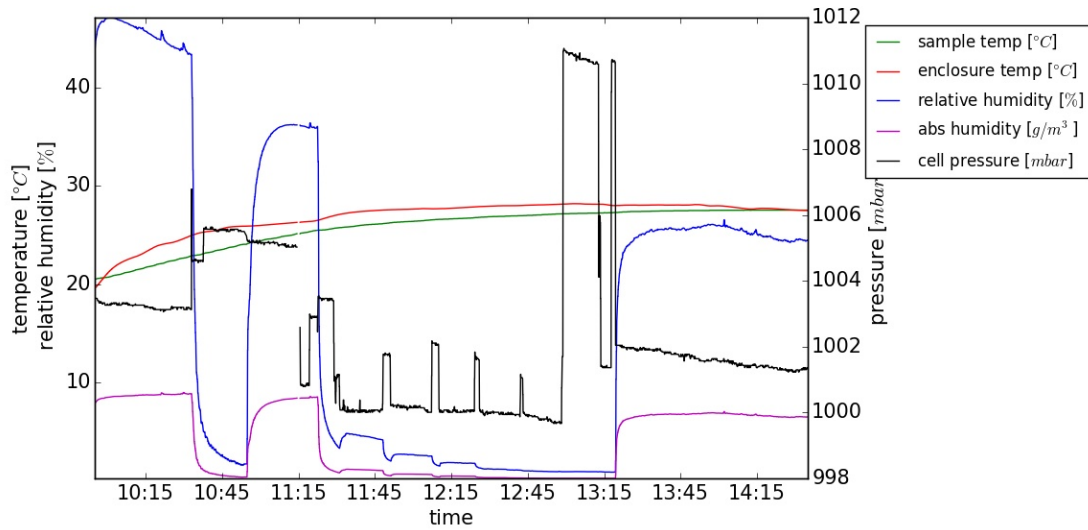


Figure A.1: Illustration over the environment for the entire measurement 1/11 2016.

section 4.4.1 (Measurement procedure). First warm-up time 30 min, then calibration with gas and air followed by OP-cases are presented starting with the first peak OP-7 then OP-6, OP-5, OP-3 and last OP-1.

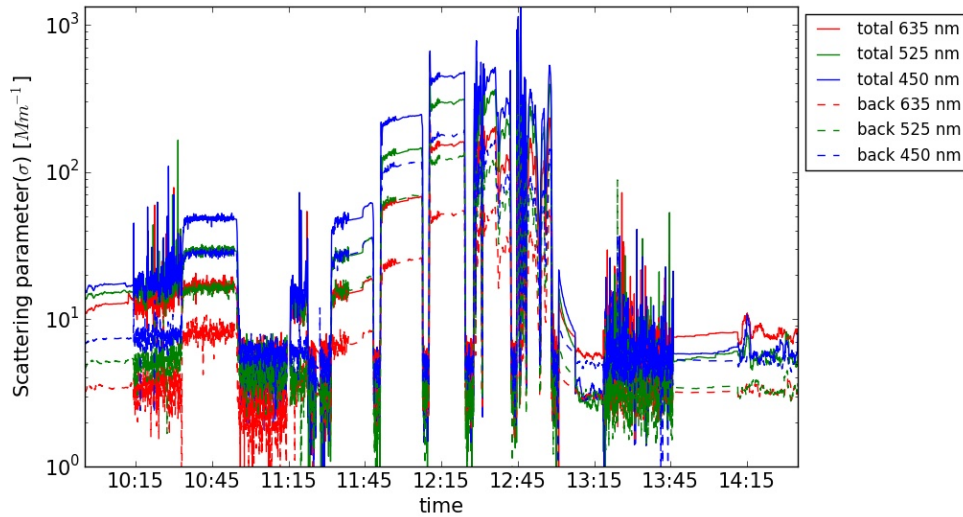


Figure A.2: Illustration over the scatter for the entire measurement 1/11 2016.

Appendix B

Downloading program

In this Appendix the code for downloading the data from the nephelometer to a computer is presented.

grab.py

```
#!/usr/bin/python3

import serial
import threading
import time
import sys
import calendar

class Grabber:

    def __init__(self, device, baudrate, start, end):
        self.serial = serial.Serial(device, baudrate)
        self.range = (start + end).encode('ascii')
        self.running = True
        self.cond = threading.Condition()
        t = threading.Thread(target=self.reader)
        t.daemon = True
        t.start()
        t = threading.Thread(target=self.writer)
        t.daemon = True
        t.start()

    def send_and_match(self, send, match=None):
        self.serial.write(send)
        with self.cond:
            if match == None:
                self.match = send
            else:
                self.match = match
            self.cond.wait()

    def writer(self):
        # Get instrument ID
        def match_id(data):
            if data.find(b'\r\n*\r\n') != -1:
                print(data)
                return True
            return False
```

```

self.send_and_match(b'\x14', match_id)
time.sleep(1)
# Select instrument
self.serial.write(b'VI16-019564\r')
time.sleep(10)

# Select sample range
def match_range(data):
    return data[-1] == ord('\n')
#self.send_and_match(b'***ED16092618461609261857\r',
self.send_and_match(b'***ED%s\r' % self.range,
                    match_range)

time.sleep(1)

def match_sample(data):
    # print(len(data), data)
    if data[0] == 4:
        self.running = False
        return True
    elif len(data) < 2:
        return False
    elif len(data) == 132 and data[0] == 1 and data[-2] == ord('\n'):
        values = data[3:-3].decode('ascii').strip().split(',')
        date = time.strptime('%Y-%m-%d:%H:%M:%S',
                             time.gmtime((calendar.timegm(
                                 [2000, 1, 1, 0, 0, 0]) +
                                 int(values[0]))))
        print(date, ', ', ', ', ', , '.join(values[0:]))
        # print(list(data[0:3]), data[3:-3].decode('ascii'))
        return True
    else:
        return False

# Get first sample
self.send_and_match(b'\x15', match_sample)

# Grab all samples
while True:
    # time.sleep(0.5)
    self.send_and_match(b'\x06', match_sample)

def reader(self):
    data = b''
    while True:
        data += self.serial.read()
        with self.cond:
            if self.match(data):
                # print('GOT', data)
                data = b''
                self.cond.notify_all()

"""
/dev/ttyUSB0\b'\x14'
/dev/ttyS0\b'Ecotech_Aurora_3000_Nephelometer_v1.27.004,_ID_#160195\r'
/dev/ttyS0\b'\n'
/dev/ttyS0\b'*\r'

```



```

/dev/ttyS0_b'\n'
/dev/ttyUSB0_b'VI16-019564\r'
/dev/ttyUSB0_b'***ED16092600001609261543\r'
/dev/ttyS0_b'***ED16092600001609261543\r'
/dev/ttyS0_b'\n'
/dev/ttyUSB0_b'\x15'
/dev/ttyS0_b'\x01'
/dev/ttyS0_b'\x01'
/dev/ttyS0_b'\xfe0528211440 ,_0,_04,_0.000 ,_0.000 ,_0.000 ,_0.000 ,_0.000 ,_0.000 ,_0.000 ,_294.636 ,_2
/dev/ttyS0_b'\n'
/dev/ttyUSB0_b'\x06'
/dev/ttyS0_b'B\x01'
/dev/ttyS0_b'\x02'
/dev/ttyS0_b'\xfd0528211501 ,_0,_04,_10.416 ,_14.692 ,_19.684 ,_2.723 ,_2.453 ,_3.524 ,_294.650
/dev/ttyS0_b'\n'
/dev/ttyUSB0_b'\x06'
/dev/ttyS0_b'\xd8\x01'
/dev/ttyS0_b'\x03'
/dev/ttyS0_b'\xfc0528211561 ,_0,_04,_10.744 ,_19.506 ,_21.454 ,_2.797 ,_2.600 ,_3.753 ,_294.650
"""

```

```

if __name__ == '__main__':
    grabber = Grabber('/dev/ttyUSB0', baudrate=19200,
                      start=sys.argv[1], end=sys.argv[2])
    while grabber.running:
        time.sleep(1)

```

Appendix C

Plot and table generating program

In this Appendix all the different plots and automatically generated tables are presented.

plot.py

```
#!/usr/bin/python3

import argparse
import matplotlib.pyplot as plt
import matplotlib.dates as mdates
from matplotlib.ticker import MaxNLocator, ScalarFormatter
import numpy as np
import sys
import datetime
import math
import os
import time

#GR = '#D0D000'
GR = '#FF8000'
BG = '#00FFFF'
BR = '#FF00FF'

BeginTable = """
\\begin{tabular}{cccccc}
\\multirow{2}{*}{N}&&
\\multicolumn{2}{|c|}{Least_Squares_Fit}&&
\\multicolumn{3}{|c|}{Multi_Range_Ångström}&&
\\multirow{2}{*}{Comment}&&
&\\multicolumn{1}{|c|}{Forward}&&Back&
&\\multicolumn{1}{|c|}{Wavelengths}&&Forward&&\\multicolumn{1}{|c|}{Back}&&
\\hline\\hline
"""

EndTable = "\\end{tabular}\\n"

def get_keywords(s):
    if len(s)== 0:
        keywords = {}
    else:
        keywords = dict([ e.split('=') for e in s.split(':') ])
    return keywords

class Segment:
```

```

def __init__(self, start, line, end=None):
    self.start = start
    self.keywords = get_keywords(line[2:].split(', ')[0])
    if end != None:
        self.end = end

def last(self, end, line):
    self.end = end
    keywords = get_keywords(line[2:].split(', ')[0])
    if keywords != self.keywords:
        raise Exception('Keywords differs %s %s (%s)',
                        self.keywords, keywords, self)
    self.N = self.end - self.start

def keyword(self, keyword):
    return self.keywords.get(keyword, '')

def __getattr__(self, key):
    return self.keywords.get(key, '')

def __repr__(self):
    return "Segment(%d, %d, %s)" % (self.start, self.end, self.keywords)

class SegmentList:
    def __init__(self, segment):
        self.segment = segment

    def keyword(self, keyword):
        if len(self.segment) == 0 or keyword == None:
            return SegmentList([])
        result = []
        start = 0
        for i in range(1, len(self.segment)):
            if (self.segment[start].keyword(keyword) !=
                self.segment[i].keyword(keyword)):
                result.append(
                    Segment(self.segment[start].start,
                            '*+%s=%s' % (keyword,
                                           self.segment[start].keyword(keyword)),
                            end=self.segment[i-1].end))
                start = i
        result.append(
            Segment(self.segment[start].start,
                    '*+%s=%s' % (keyword,
                                   self.segment[start].keyword(keyword)),
                    end=self.segment[-1].end))
        return result

    def __len__(self):
        return len(self.segment)

    def __iter__(self):
        return self.segment.__iter__()

    def __repr__(self):
        return "SegmentList(%s)" % (self.segment)

```

```

def read_data(path, byseconds=False):
    value = []
    timestamp = []
    segment = []
    last_t = None

    for l in [ l.strip() for l in open(path) ]:
        if l.startswith('*'):
            # Files generated by data_select.py, scale in seconds
            byseconds = True
            if l[1:2] == '+':
                # Start of segment (neither marker nor value saved yet)
                new_segment = Segment(len(value) + 2, 1)
            elif l[1:2] == '-':
                # End of segment (value is saved)
                new_segment.last(len(value), 1)
                segment.append(new_segment)

    tmp = list(map(lambda l: l.strip(), l.split(',') ))
    if byseconds:
        ts = float(tmp[1])
    else:
        ts = datetime.datetime.strptime(tmp[0], "%Y-%m-%d:%H:%M:%S")
    # Check if we have a timejump
    t = float(tmp[1])
    if last_t != None and t > last_t + 60:
        nan = float('NaN')
        timestamp.append(timestamp[-1])
        value.append([last_t ,
                     nan, nan, nan, nan, nan, nan,
                     nan, nan, nan, nan, nan, nan])
        timestamp.append(ts)
        value.append([float(tmp[1]),
                     nan, nan, nan, nan, nan, nan,
                     nan, nan, nan, nan, nan, nan])

    last_t = t
    timestamp.append(ts)
    value.append(list(map(float, tmp[1:])))

    return timestamp, value, SegmentList(segment), byseconds

```

```

class ErrorBar:

```

```

    def __init__(self):
        self.sample = []

    def add(self, index, segment, wavelength, mean, stddev):
        if len(self.sample) <= index:
            self.sample.append([])
            self.sample[index].append((segment, wavelength, mean, stddev))

    def __repr__(self):
        return ('ErrorBar(%s)' % self.sample)

```

```

class ErrorBarTable:

```

```

    def __init__(self, path, **ebar):
        self.path = path

```

```

self._ebar = ebar

def ebar(self, key):
    if key in self._ebar:
        return self._ebar[key]
    return None

def rows(self, keys):
    length = 0
    for ek in keys:
        length = max(length, len(self._ebar[ek].sample))
    return length

def get(self, keys):
    for i in range(self.rows(keys)):
        rows = 0
        segment = None
        result = []
        for ek in keys:
            column = self._ebar[ek].sample[i]
            rows = max(rows, len(column))
            for c in column:
                if segment == None:
                    segment = c[0]
                if segment != c[0]:
                    raise Exception()
            result.append(column)
        yield [rows, segment] + result

def write(self):
    if self.rows(['lstsq', 'forward', 'back']) == 0:
        return
    with open(self.path, 'w') as fd:
        fd.write(BeginTable)
        for rows, segment, l, f, b in self.get(['lstsq', 'forward', 'back']):
            for i in range(rows):
                if i == 0:
                    fd.write("\multirow{%d}{*}{%s} \&\n" %
                               (rows, segment.N))
                    fd.write("\multirow{%d}{*}{$_%0.2f \pm %0.2f $} \&\n" %
                               (rows, l[0][2], l[0][3]))
                    fd.write("\multirow{%d}{*}{$_%0.2f \pm %0.2f $} \&\n" %
                               (rows, l[1][2], l[1][3]))
                else:
                    fd.write('&&\n')
                    fd.write("$_%s \&\n" % (f[i][1]))
                    fd.write("$_%0.2f \pm %0.2f $ \&\n" % (f[i][2], f[i][3]))
                    fd.write("$_%0.2f \pm %0.2f $ \&\n" % (b[i][2], b[i][3]))
                if i == 0:
                    fd.write("\multirow{%d}{*}{%s} \n" % (
                        rows, segment.comment))
                    fd.write('\\\\\n')
                    fd.write("\hline\n")
            fd.write(EndTable)

def absolute_humidity(v):
    # Absolute humidity
    #

```

```

# From http://www.vaisala.com/Vaisala%20Documents/Application%20notes/
# Humidity-Conversion-Formulas-B210973EN-F.pdf
#
# (6):
#  $Pws = A * 10^{(m*T/(T+Tn))}$ 
#
# Table 1: (-20...+50 deg C)
#
#  $A = 6.116441$ 
#  $m = 7.591386$ 
#  $Tn = 240.726$ 
TC = v[:,9] - 273.15
pws = 6.116441 * np.exp(np.log(10) * (7.591386 * TC) / (TC + 240.726))

# (17)
#  $A = C * Pws * Rh / T$ 
#  $C = 2.16679 \text{ gK/J}$ 
Rh = v[:,11]
TK= v[:,9]
A = 2.16679 * pws * Rh / TK
return A

```

class Data:

```

def __init__(self, path, byseconds=False, zeroadjust=True):
    self.path = path
    timestamp, value, segment, byseconds = read_data(path, byseconds)
    t = timestamp
    v = np.array(value)
    self.segment = segment

    if byseconds:
        self.major_x_formatter = ScalarFormatter()
    else:
        self.major_x_formatter = mdates.DateFormatter('%H:%M')

    if zeroadjust:
        for i in range(3,9):
            # Make all measurements slightly positive (zero adjustment)
            print(i, np.min(v[~np.isnan(v[:,i]),i]))
            v_min = np.min(v[~np.isnan(v[:,i]),i])
            if v_min <= 0:
                v_max = np.max(v[~np.isnan(v[:,i]),i])
                v[:,i] += -v_min + (v_max - v_min) / 1e6
            print(i, np.min(v[~np.isnan(v[:,i]),i]))
        self.v = v
        self.t = t

    def keyword_text(self, fig, keyword):
        ymin, ymax = fig.get_ylim()
        for s in self.segment.keyword(keyword):
            fig.text(self.t[s.start], ymax, s.keyword(keyword),
                    bbox=dict(boxstyle="rarrow", lw=2,
                              color='b', alpha=0.2))

    def do_scatter_plot(self, fig, t, v, keyword=None):
        fig.set_ylabel('Scattering parameter ( $\sigma$ ) [ $Mm^{-1}$ ']
        fig.set_xlabel('time')

```

```

fig.xaxis.set_major_formatter(self.major_x_formatter)
fig.semilogy(t, v[:,3], 'r', label='total_635_μm')
fig.semilogy(t, v[:,4], 'g', label='total_525_μm')
fig.semilogy(t, v[:,5], 'b', label='total_450_μm')
fig.semilogy(t, v[:,6], 'r—', label='back_635_μm')
fig.semilogy(t, v[:,7], 'g—', label='back_525_μm')
fig.semilogy(t, v[:,8], 'b—', label='back_450_μm')
fig.set_ylim(1.0)

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = fig.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))

def do_scatter_lin_plot(self, fig, t, v, keyword=None):
fig.set_ylabel('Scattering_parameter( $\sigma$ ) [ $Mm^{-1}$ ]')
fig.set_xlabel('time')
fig.xaxis.set_major_formatter(self.major_x_formatter)
fig.plot(t, v[:,3], 'r', label='total_635_μm')
fig.plot(t, v[:,4], 'g', label='total_525_μm')
fig.plot(t, v[:,5], 'b', label='total_450_μm')
fig.plot(t, v[:,6], 'r—', label='back_635_μm')
fig.plot(t, v[:,7], 'g—', label='back_525_μm')
fig.plot(t, v[:,8], 'b—', label='back_450_μm')
fig.set_ylim(0.0)

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = fig.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))

def do_environment_plot(self, fig, t, v, keyword=None):
fig.set_ylabel('temperature [ $^{\circ}C$ ] \n relative_humidity [%]')
fig.set_xlabel('time')
fig.xaxis.set_major_formatter(self.major_x_formatter)

fig.plot(t, v[:,9] - 273.15, 'g', label='sample_temp [ $^{\circ}C$ ]')
fig.plot(t, v[:,10] - 273.15, 'r', label='enclosure_temp [ $^{\circ}C$ ]')
fig.plot(t, v[:,11], 'b', label='relative_humidity [%]')
fig.plot(t, absolute_humidity(v), 'm', label='abs_humidity [ $g/m^3$ ]')
right_axis = fig.twinx()
right_axis.autoscale(enable=True, axis='x', tight=True)
right_axis.set_ylabel('pressure [ $mbar$ ]')
right_axis.set_ylim(int(np.min(v[~np.isnan(v[:,12]),12]) - 1),
                    int(np.max(v[~np.isnan(v[:,12]),12]) + 1))
right_axis.get_yaxis().get_major_formatter().set_useOffset(False)
right_axis.get_yaxis().set_major_locator(MaxNLocator(integer=True))
right_axis.plot(t, v[:,12], 'k', label='cell_pressure [ $mbar$ ]')

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = right_axis.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])

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right_axis.set_position([box.x0, box.y0, box.width * 0.8, box.height])
lines, labels = fig.get_legend_handles_labels()
lines2, labels2 = right_axis.get_legend_handles_labels()
right_axis.legend(lines + lines2, labels + labels2,
                  loc='upper_left',
                  bbox_to_anchor=(1.07, 1))

def do_absolute_humidity_plot(self, fig, t, v, keyword=None):
    fig.set_ylabel('absolute_humidity_[$g/m^3$]')
    fig.set_xlabel('time')
    fig.xaxis.set_major_formatter(self.major_x_formatter)

    fig.plot(t, absolute_humidity(v), 'g', label='abs_humidity_[$g/m^3$]')
    self.keyword_text(fig, keyword)

    # Legend outside shrunk plot
    box = fig.get_position()
    fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
    fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))

def do_lstsq_plot(self, fig, t, v, plot_alpha=None, with_ebar=None,
                 keyword=None):
    fig.set_ylabel('Ångström_exponent',)
    fig.set_xlabel('time')
    fig.xaxis.set_major_formatter(self.major_x_formatter)

    A=np.array([[math.log(635), math.log(525), math.log(450)],
                [1,1,1]]).T
    y_forw = np.log(v[:,3:6] - v[:,6:9]).T
    k_forw = -np.linalg.lstsq(A, y_forw)[0][0]
    y_back = np.log(v[:,6:9]).T
    k_back = -np.linalg.lstsq(A, y_back)[0][0]
    def pa(key):
        if not plot_alpha: return None
        if not key in plot_alpha: return None
        return plot_alpha[key]
    fig.plot(t, k_forw, 'r', label="forward", alpha=pa('forward'))
    fig.plot(t, k_back, 'g', label="back", alpha=pa('back'))

    ymin = None
    ymax = None
    if with_ebar:
        display_to_data = fig.transData.inverted()
        for i,seg in zip(range(len(self.segment)), self.segment):
            w = (display_to_data.transform([4, 0])[0] -
                 display_to_data.transform([0, 0])[0])
            x = np.mean(t[seg.start:seg.end])
            # There might be NaN's in alpha...
            y_forw = np.nanmean(k_forw[seg.start:seg.end])
            y_back = np.nanmean(k_back[seg.start:seg.end])
            err_forw = np.nanstd(k_forw[seg.start:seg.end], ddof=1)
            err_back = np.nanstd(k_back[seg.start:seg.end], ddof=1)
            fig.errorbar(x - w, y_forw, yerr=err_forw,
                        color='r', ecolor='k', fmt='o')
            fig.errorbar(x + w, y_back, yerr=err_back,
                        color='g', ecolor='k', fmt='o')
            ymax = max(filter(None, [ymax, y_forw + err_forw]))
            ymax = max(filter(None, [ymax, y_forw + err_forw]))

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        ymin = min(filter(None, [ymax, y_forw - err_forw]))
        ymin = min(filter(None, [ymax, y_forw - err_forw]))
        with_ebar.add(i, seg, 'forw', y_forw, err_forw)
        with_ebar.add(i, seg, 'back', y_back, err_back)

if ymin == None or ymax == None:
    ymin = -5.0
    ymax = 5.0
fig.set_ylim(ymin - 1, ymax + 1)

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = fig.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
leg = fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))
for l in leg.get_lines():
    l.set_alpha(1.0)

def do_alpha_plot(self, fig, t, v, plot_alpha=None, with_ebar=None,
                 keyword=None):
    fig.set_ylabel('Ångström_exponent')
    fig.set_xlabel('time')
    fig.xaxis.set_major_formatter(self.major_x_formatter)

    alpha_525_635 = -np.log(v[:,1]/v[:,0])/math.log(525/635)
    alpha_450_525 = -np.log(v[:,2]/v[:,1])/math.log(450/525)
    alpha_450_635 = -np.log(v[:,2]/v[:,0])/math.log(450/635)
    def pa(key):
        if not plot_alpha: return None
        if not key in plot_alpha: return None
        return plot_alpha[key]
    fig.plot(t, alpha_525_635, GR, label='525/635', alpha=pa('525/635'))
    fig.plot(t, alpha_450_635, BR, label='450/635', alpha=pa('450/635'))
    fig.plot(t, alpha_450_525, BG, label='450/525', alpha=pa('450/525'))

    ymin = None
    ymax = None
    if with_ebar:
        display_to_data = fig.transData.inverted()
        for i, seg in zip(range(len(self.segment)), self.segment):
            w = (display_to_data.transform([7, 0])[0] -
                 display_to_data.transform([0, 0])[0])
            x = np.mean(t[seg.start:seg.end])
            # There might be NaN's in alpha...
            y_525_635 = np.nanmean(alpha_525_635[seg.start:seg.end])
            y_450_525 = np.nanmean(alpha_450_525[seg.start:seg.end])
            y_450_635 = np.nanmean(alpha_450_635[seg.start:seg.end])
            err_525_635 = np.nanstd(alpha_525_635[seg.start:seg.end],
                                   ddof=1)
            err_450_525 = np.nanstd(alpha_450_525[seg.start:seg.end],
                                   ddof=1)
            err_450_635 = np.nanstd(alpha_450_635[seg.start:seg.end],
                                   ddof=1)
            fig.errorbar(x - w, y_525_635, yerr=err_525_635,
                        color=GR, ecolor='k', fmt='o')
            fig.errorbar(x + w, y_450_525, yerr=err_450_525,
                        color=BG, ecolor='k', fmt='o')

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```

fig.errorbar(x, y_450_635, yerr=err_450_635,
             color=BR, ecolor='k', fmt='o')
ymax = max(filter(None, [ymax, y_525_635 + err_525_635]))
ymax = max(filter(None, [ymax, y_450_525 + err_450_525]))
ymax = max(filter(None, [ymax, y_450_635 + err_450_635]))
ymin = min(filter(None, [ymin, y_525_635 - err_525_635]))
ymin = min(filter(None, [ymin, y_450_525 - err_450_525]))
ymin = min(filter(None, [ymin, y_450_635 - err_450_635]))
with_ebar.add(i, seg, '${450}/{525}$', y_450_525, err_450_525)
with_ebar.add(i, seg, '${450}/{635}$', y_450_635, err_450_635)
with_ebar.add(i, seg, '${525}/{635}$', y_525_635, err_525_635)

if ymin == None or ymax == None:
    ymin = -5.0
    ymax = 5.0
fig.set_ylim(ymin - 1, ymax + 1)

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = fig.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
leg = fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))
for l in leg.get_lines():
    l.set_alpha(1.0)

def do_alpha_ratio_ebar_plot(self, fig, t, v, keyword=None):
    fig.set_ylabel('Ångström_exponent_ratio_(forward/back)')
    fig.set_xlabel('time')
    fig.xaxis.set_major_formatter(self.major_x_formatter)

    forw = v[:,3:6] - v[:,6:9]
    alpha_forw_525_635 = -np.log(forw[:,1]/forw[:,0])/math.log(525/635)
    alpha_forw_450_525 = -np.log(forw[:,2]/forw[:,1])/math.log(450/525)
    alpha_forw_450_635 = -np.log(forw[:,2]/forw[:,0])/math.log(450/635)
    alpha_back_525_635 = -np.log(v[:,7]/v[:,6])/math.log(525/635)
    alpha_back_450_525 = -np.log(v[:,8]/v[:,7])/math.log(450/525)
    alpha_back_450_635 = -np.log(v[:,8]/v[:,6])/math.log(450/635)
    fig.plot(t, alpha_forw_525_635/alpha_back_525_635,
            GR, label='525/635', alpha=0.5)
    fig.plot(t, alpha_forw_450_525/alpha_back_450_525,
            BG, label='450/525', alpha=0.3)
    fig.plot(t, alpha_forw_450_635/
            alpha_back_450_635, BR, label='450/635', alpha=0.3)

    ymin = None
    ymax = None
    display_to_data = fig.transData.inverted()
    for seg in self.segment:
        w = (display_to_data.transform([7, 0])[0] -
            display_to_data.transform([0, 0])[0])
        x = np.mean(t[seg.start:seg.end])
        # There might be NaN's in alpha...
        y_525_635 = np.nanmean(
            (alpha_forw_525_635/alpha_back_525_635)[seg.start:seg.end])
        y_450_525 = np.nanmean(
            (alpha_forw_450_525/alpha_back_450_525)[seg.start:seg.end])
        y_450_635 = np.nanmean(

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        (alpha_forw_450_635/alpha_back_450_635)[seg.start:seg.end])
err_525_635 = np.nanstd(
    (alpha_forw_525_635/alpha_back_525_635)[seg.start:seg.end], ddof=1)
err_450_525 = np.nanstd(
    (alpha_forw_450_525/alpha_back_450_525)[seg.start:seg.end], ddof=1)
err_450_635 = np.nanstd(
    (alpha_forw_450_635/alpha_back_450_635)[seg.start:seg.end], ddof=1)
fig.errorbar(x - w, y_525_635, yerr=err_525_635,
             color=GR, ecolor='k', fmt='o')
fig.errorbar(x + w, y_450_525, yerr=err_450_525,
             color=BG, ecolor='k', fmt='o')
fig.errorbar(x, y_450_635, yerr=err_450_635,
             color=BR, ecolor='k', fmt='o')
ymax = max(filter(None, [ymax, y_525_635 + err_525_635]))
ymax = max(filter(None, [ymax, y_450_525 + err_450_525]))
ymax = max(filter(None, [ymax, y_450_635 + err_450_635]))
ymin = min(filter(None, [ymin, y_525_635 - err_525_635]))
ymin = min(filter(None, [ymin, y_450_525 - err_450_525]))
ymin = min(filter(None, [ymin, y_450_635 - err_450_635]))

if ymin == None or ymax == None:
    ymin = -5.0
    ymax = 5.0
fig.set_ylim(ymin - 1, ymax + 1)

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = fig.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
leg = fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))
for l in leg.get_lines():
    l.set_alpha(1.0)

def do_ratio_plot(self, fig, t, numerator, denominator,
                 numerator_name, denominator_name,
                 plot_alpha=None, with_ebar=None,
                 keyword=None):
    fig.set_ylabel('Relative_scattering\n'
                  '$\\sigma_{%s}/\\sigma_{%s}$' %
                  (numerator_name, denominator_name))
    fig.set_xlabel('time')
    fig.xaxis.set_major_formatter(self.major_x_formatter)

    ratio_635 = numerator[:,0] / denominator[:,0]
    ratio_525 = numerator[:,1] / denominator[:,1]
    ratio_450 = numerator[:,2] / denominator[:,2]
    def pa(key):
        if not plot_alpha: return None
        if not key in plot_alpha: return None
        return plot_alpha[key]
    fig.plot(t, ratio_635, 'r', label='635_μm', alpha=pa('635'))
    fig.plot(t, ratio_525, 'g', label='525_μm', alpha=pa('525'))
    fig.plot(t, ratio_450, 'b', label='450_μm', alpha=pa('450'))
    fig.yaxis.grid(True)

    ymin = None
    ymax = None

```

```

if with_ebar:
    display_to_data = fig.transData.inverted()
    for i, seg in zip(range(len(self.segment)), self.segment):
        w = (display_to_data.transform([7, 0])[0] -
             display_to_data.transform([0, 0])[0])
        x = np.mean(t[seg.start:seg.end])
        # There might be NaN's in alpha...
        y_635 = np.nanmean(ratio_635[seg.start:seg.end])
        y_525 = np.nanmean(ratio_525[seg.start:seg.end])
        y_450 = np.nanmean(ratio_450[seg.start:seg.end])
        err_635 = np.nanstd(ratio_635[seg.start:seg.end], ddof=1)
        err_525 = np.nanstd(ratio_525[seg.start:seg.end], ddof=1)
        err_450 = np.nanstd(ratio_450[seg.start:seg.end], ddof=1)
        fig.errorbar(x - w, y_635, yerr=err_635,
                    color='r', ecolor='k', fmt='o')
        fig.errorbar(x, y_525, yerr=err_525,
                    color='g', ecolor='k', fmt='o')
        fig.errorbar(x + w, y_450, yerr=err_450,
                    color='b', ecolor='k', fmt='o')
        ymax = max(filter(None, [ymax, y_635 + err_635]))
        ymax = max(filter(None, [ymax, y_525 + err_525]))
        ymax = max(filter(None, [ymax, y_450 + err_450]))
        ymax = min(filter(None, [ymax, 6]))
        ymin = min(filter(None, [ymin, y_635 - err_635]))
        ymin = min(filter(None, [ymin, y_525 - err_525]))
        ymin = min(filter(None, [ymin, y_450 - err_450]))
        ymin = max(filter(None, [ymin, -2]))
    if ymin != None and ymax != None:
        # Reset ylim to contain errorbars
        fig.set_ylim(ymin - 1, ymax + 1)

if ymin == None or ymax == None:
    ymin = -1.0
    ymax = 5.0
fig.set_ylim(ymin - 1, ymax + 1)

self.keyword_text(fig, keyword)

# Legend outside shrunk plot
box = fig.get_position()
fig.set_position([box.x0, box.y0, box.width * 0.8, box.height])
leg = fig.legend(loc='upper_left', bbox_to_anchor=(1, 1))
for l in leg.get_lines():
    l.set_alpha(1.0)

def savename(self, suffix, keyword=None):
    result = "plots/%s_" % (os.path.basename(self.path))
    result += suffix
    if keyword != None:
        result += "_%s" % keyword
    result += ".jpg"
    return result

def scatter_environment_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.canvas.set_window_title("Scatter/Environment_(%s)" % self.path)
    scatter = fig.add_subplot(211)
    scatter.autoscale(enable=True, axis='x', tight=True)

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self.do_scatter_plot(scatter, self.t, self.v, keyword=keyword)
environment = fig.add_subplot(212)
environment.autoscale(enable=True, axis='x', tight=True)
self.do_environment_plot(environment, self.t, self.v, keyword=keyword)
fig.savefig(self.savename("scatter_environment", keyword))

def scatter_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Scatter_␣(%s)" % self.path)
    self.do_scatter_plot(fig.gca(), self.t, self.v, keyword=keyword)
    fig.savefig(self.savename("scatter", keyword))

def scatter_lin_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Scatter_␣(%s)" % self.path)
    self.do_scatter_lin_plot(fig.gca(), self.t, self.v, keyword=keyword)
    fig.savefig(self.savename("scatter_lin", keyword))

def environment_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Environment(%s)" % self.path)
    self.do_environment_plot(fig.gca(), self.t, self.v, keyword=keyword)
    fig.savefig(self.savename("environment", keyword))

def absolute_humidity_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Absolute_␣Humidity(%s)" % self.path)
    self.do_absolute_humidity_plot(fig.gca(), self.t, self.v,
                                   keyword=keyword)
    fig.savefig(self.savename("absolute_humidity", keyword))

def lstsq_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Least_␣Squares_␣Fit_␣(%s)" % self.path)
    self.do_lstsq_plot(fig.gca(), self.t, self.v, keyword=keyword)
    fig.savefig(self.savename("lstsq", keyword))

def lstsq_ebar_plot(self, table, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Least_␣Squares_␣Fit_␣(%s)" % self.path)
    self.do_lstsq_plot(fig.gca(), self.t, self.v,
                       plot_alpha={'forward': 0.3,
                                   'back': 0.3 },
                       with_ebar=table and table.ebar('lstsq'),
                       keyword=keyword)
    fig.savefig(self.savename("lstsq_ebar", keyword))

def total_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ångström_␣total_␣(%s)" % self.path)
    total = self.v[:,3:6]

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self.do_alpha_plot(fig.gca(), self.t, v=total, keyword=keyword)
fig.savefig(self.savename("total", keyword))

def total_ebar_plot(self, table, keyword=None):
    if not self.segment:
        return
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ångström_total_(%s)_[with_errorbar]" %
                                self.path)

    total = self.v[:,3:6]
    self.do_alpha_plot(fig.gca(), self.t, v=total,
                        plot_alpha={'525/635': 0.5,
                                    '450/525': 0.3,
                                    '450/635': 0.3 },
                        with_ebar=table and table.ebar('total'),
                        keyword=keyword)
    fig.savefig(self.savename("total_ebar", keyword))

def forward_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ångström_forward_(%s)" % self.path)
    forward = self.v[:,3:6] - self.v[:,6:9]
    self.do_alpha_plot(fig.gca(), self.t, v=forward, keyword=keyword)
    fig.savefig(self.savename("forward", keyword))

def forward_ebar_plot(self, table, keyword=None):
    if not self.segment:
        return
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ångström_forward_(%s)_[with_errorbar]" %
                                self.path)

    forward = self.v[:,3:6] - self.v[:,6:9]
    self.do_alpha_plot(fig.gca(), self.t, v=forward,
                        plot_alpha={'525/635': 0.5,
                                    '450/525': 0.3,
                                    '450/635': 0.3 },
                        with_ebar=table and table.ebar('forward'),
                        keyword=keyword)
    fig.savefig(self.savename("forward_ebar", keyword))

def back_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ångström_back_(%s)" % self.path)
    back = self.v[:,6:9]
    self.do_alpha_plot(fig.gca(), self.t, v=back, keyword=keyword)
    fig.savefig(self.savename("back", keyword))

def back_ebar_plot(self, table, keyword=None):
    if not self.segment:
        return
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ångström_back_(%s)_[with_errorbar]" %
                                self.path)

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back = self.v[:,6:9]
self.do_alpha_plot(fig.gca(), self.t, v=back,
                  plot_alpha={ '525/635': 0.5,
                              '450/525': 0.3,
                              '450/635': 0.3 },
                  with_ebar=table and table.ebar('back'),
                  keyword=keyword)
fig.savefig(self.savename("back_ebar", keyword))

def alpha_ratio_ebar_plot(self, keyword=None):
fig = plt.figure(figsize=(12, 6))
fig.gca().autoscale(enable=True, axis='x', tight=True)
fig.canvas.set_window_title("Alpha_ratio_[with_errorbar]_(%s)" %
                             self.path)
self.do_alpha_ratio_ebar_plot(fig.gca(), self.t, self.v,
                              keyword=keyword)
fig.savefig(self.savename("alpha_ratio_ebar", keyword))

def ratio_plot(self, keyword=None):
fig = plt.figure(figsize=(12, 6))
fig.gca().autoscale(enable=True, axis='x', tight=True)
fig.canvas.set_window_title("Ratio_(%s)" % self.path)
forward = self.v[:,3:6] - self.v[:,6:9]
back = self.v[:,6:9]
self.do_ratio_plot(fig.gca(), self.t,
                  numerator=forward, denominator=back,
                  numerator_name='forward', denominator_name='back',
                  keyword=keyword)
fig.savefig(self.savename("ratio", keyword))

def ratio_ebar_plot(self, keyword=None):
fig = plt.figure(figsize=(12, 6))
fig.gca().autoscale(enable=True, axis='x', tight=True)
fig.canvas.set_window_title("Ratio_[with_errorbar]_(%s)" % self.path)
forward = self.v[:,3:6] - self.v[:,6:9]
back = self.v[:,6:9]
self.do_ratio_plot(fig.gca(), self.t,
                  numerator=forward, denominator=back,
                  numerator_name='forward', denominator_name='back',
                  plot_alpha={ '635': 0.3,
                              '525': 0.3,
                              '450': 0.3 },
                  with_ebar=True,
                  keyword=keyword)
fig.savefig(self.savename("ratio_ebar", keyword))

def ratio_total_plot(self, keyword=None):
fig = plt.figure(figsize=(12, 6))
fig.gca().autoscale(enable=True, axis='x', tight=True)
fig.canvas.set_window_title("Ratio_total(%s)" % self.path)
total = self.v[:,3:6]
back = self.v[:,6:9]
self.do_ratio_plot(fig.gca(), self.t,
                  numerator=total, denominator=back,
                  numerator_name='total', denominator_name='back',
                  keyword=keyword)
fig.savefig(self.savename("ratio_total", keyword))

```

```

def ratio_total_ebar_plot(self, keyword=None):
    fig = plt.figure(figsize=(12, 6))
    fig.gca().autoscale(enable=True, axis='x', tight=True)
    fig.canvas.set_window_title("Ratio_total [with_errorbar] (%s)" %
                                self.path)

    total = self.v[:,3:6]
    back = self.v[:,6:9]
    self.do_ratio_plot(fig.gca(), self.t,
                      numerator=total, denominator=back,
                      numerator_name='total', denominator_name='back',
                      plot_alpha={ '635': 0.3,
                                   '525': 0.3,
                                   '450': 0.3 },
                      with_ebar=True,
                      keyword=keyword)
    fig.savefig(self.savename("ratio_total_ebar", keyword))

if __name__ == '__main__':
    optParser = argparse.ArgumentParser(usage="(prog) s [options]")
    optParser.add_argument("path", nargs='+',
                          help="files to plot")
    optParser.add_argument("--nodisplay",
                          action="store_true",
                          help="Only generate files, don't show")
    optParser.add_argument("--nozeroadjust",
                          action="store_true",
                          help="plot without zero adjustment")
    optParser.add_argument("--seconds",
                          action="store_true",
                          help="plot with seconds on x-scale")
    optParser.add_argument("--keyword",
                          action="store",
                          help="Generate plots with given keyword")
    options = optParser.parse_args(sys.argv[1:])

    SIZE = 15
    SIZE2 = 12
    plt.rc('font', size=SIZE) # controls default text sizes
    plt.rc('axes', titlesize=SIZE) # fontsize of the axes title
    plt.rc('axes', labelszize=SIZE) # fontsize of the x and y labels
    plt.rc('xtick', labelszize=SIZE) # fontsize of the tick labels
    plt.rc('ytick', labelszize=SIZE) # fontsize of the tick labels
    plt.rc('legend', fontsize=SIZE2) # legend fontsize
    for name in options.path:
        d = Data(name,
                byseconds=options.seconds,
                zeroadjust=not options.nozeroadjust)
        table = ErrorBarTable('tables/%s.tex' % (os.path.basename(name)),
                             lstsq=ErrorBar(),
                             total=ErrorBar(),
                             forward=ErrorBar(),
                             back=ErrorBar())
        d.scatter_environment_plot(keyword=options.keyword)
        d.scatter_plot(keyword=options.keyword)
        d.scatter_lin_plot(keyword=options.keyword)
        d.environment_plot(keyword=options.keyword)
        d.absolute_humidity_plot(keyword=options.keyword)
        d.lstsq_plot(keyword=options.keyword)

```



```

d.lstsq_ebar_plot(table, keyword=options.keyword)
d.total_plot(keyword=options.keyword)
d.total_ebar_plot(table, keyword=options.keyword)
d.forward_plot(keyword=options.keyword)
d.forward_ebar_plot(table, keyword=options.keyword)
d.back_plot(keyword=options.keyword)
d.back_ebar_plot(table, keyword=options.keyword)
d.alpha_ratio_ebar_plot(keyword=options.keyword)
d.ratio_plot(keyword=options.keyword)
d.ratio_ebar_plot(keyword=options.keyword)
d.ratio_total_plot(keyword=options.keyword)
d.ratio_total_ebar_plot(keyword=options.keyword)
table.write()
if options.nodisplay:
    plt.close('all')
if not options.nodisplay:
    plt.show()

```

Appendix D

Data selection program

In this Appendix all the program for the selected files used in the plot.py program is explained.

```
#!/usr/bin/python3

import sys
import os
import time
import calendar

if __name__ == '__main__':
    op = os.path.basename(sys.argv[1]).split('.')[0]
    Y2K = time.mktime(time.gmtime(calendar.timegm([2000, 1, 1, 0, 0, 0])))
    segment = []
    for l in open(sys.argv[1]):
        # print(l)
        if l.strip().startswith('#'):
            continue
        tmp = l.strip().split(',')
        print(tmp)
        if len(tmp) < 2:
            print('No_selection_interval"%s"' % l.strip())
            continue
        if len(tmp) >= 2:
            t1,t2 = tmp[0:2]
        if len(tmp) >= 3:
            description = tmp[2]
        else:
            description = ''

        # print(t1,t2)
        # Get selection interval
        s1 = time.mktime(time.strptime(t1, '%Y-%m-%d:%H:%M:%S')) - Y2K
        s2 = time.mktime(time.strptime(t2, '%Y-%m-%d:%H:%M:%S')) - Y2K
        # Select interval from data files
        interval = {}
        for filename in sys.argv[2:]:
            for l2 in open(filename):
                # Match on field with seconds from Y2K
                s3 = int(l2.strip().split(',')[1])
                if s1 <= s3 and s3 <= s2:
                    interval[s3] = l2.strip().split(',')
            pass
        pass
```

```

        pass
    if len(interval) == 0:
        print('No_data_in_%s_-%s' % (t1, t2))
        pass
    else:
        segment.append([ [k,description] + interval[k] for k in
                           sorted(interval.keys()) ])
    pass

# Find total data timespan
t_total = 0
for s in segment:
    t_total += s[-1][0] - s[1][0]
time_jump = int(t_total * 0.2 / len(segment))

# Output selection suitable for plot program
outfile = open('selected/%s' % op, 'w')

# Empty space at start of plot
print("*,%d,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan" % 0,
      file=outfile)
t_last = time_jump
t_offset = 0
for s in segment:
    t_offset = s[0][0] - t_last
    print("*,%s,%d,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan" %
          (s[0][1],
           s[0][0] - t_offset - 1),
          file=outfile)
    for row in s:
        t_new = row[0] - t_offset
        print("%s,%s,%s" % (row[2], t_new, ",".join(row[4:])),
              file=outfile)
    pass
    print("*,%s,%d,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan,nan" %
          (s[-1][1],
           s[-1][0] - t_offset + 1),
          file=outfile)
    t_last = s[-1][0] - t_offset + time_jump
    pass
pass

# Output selection suitable for matlab/octave
if not os.path.exists('matlab'):
    os.mkdir('matlab')
if not os.path.exists('matlab/%s' % op):
    os.mkdir('matlab/%s' % op)
for s,i in zip(segment,range(len(segment))):
    outfile = open('matlab/%s/%03d-%s' % (op, i, s[0][1].strip()), 'w')
    for row in s:
        print("%s" % (",".join(row[5:11])),
              file=outfile)

```

Appendix E

In this Appendix all the selected files used by the plot.py program are shown.

E.1 OP7.select

2016-11-01:11:32:00,2016-11-01:11:38:30,filter=raw:comment=OK
2016-11-28:11:39:00,2016-11-28:11:44:00,filter=raw:comment=OK

E.2 OP1.select

2016-11-01:12:44:40,2016-11-01:12:45:20,filter=raw:comment=OK
2016-11-01:12:45:20,2016-11-01:12:46:10,filter=raw:comment=Bad
2016-11-01:12:46:10,2016-11-01:12:48:10,filter=raw:comment=OK
2016-11-28:12:18:05,2016-11-28:12:18:19,filter=raw:comment=OK
2016-11-28:12:18:19,2016-11-28:12:19:10,filter=raw:comment=Bad
2016-11-28:12:19:10,2016-11-28:12:19:30,filter=raw:comment=OK
2016-11-28:12:19:30,2016-11-28:12:20:28,filter=raw:comment=Bad
2016-11-28:12:20:28,2016-11-28:12:21:36,filter=raw:comment=OK

E.3 raw.select

2016-11-01:11:32:00,2016-11-01:11:38:30,filter=raw:comment=OP7
2016-11-28:11:39:00,2016-11-28:11:44:00,filter=raw:comment=OP7
2016-11-01:11:51:20,2016-11-01:11:57:20,filter=raw:comment=OP6
2016-11-28:11:48:00,2016-11-28:11:53:00,filter=raw:comment=OP6
2016-11-28:11:58:00,2016-11-28:12:03:00,filter=raw:comment=OP5
2016-11-01:12:10:20,2016-11-01:12:15:00,filter=raw:comment=OP5
2016-11-01:12:27:39,2016-11-01:12:28:18,filter=raw:comment=OP3
2016-11-01:12:28:35,2016-11-01:12:30:44,filter=raw:comment=OP3
2016-11-28:12:07:40,2016-11-28:12:08:40,filter=raw:comment=OP3
2016-11-28:12:09:20,2016-11-28:12:10:20,filter=raw:comment=OP3
2016-11-28:12:11:05,2016-11-28:12:13:30,filter=raw:comment=OP3
2016-11-01:12:44:40,2016-11-01:12:45:20,filter=raw:comment=OP1
2016-11-01:12:46:10,2016-11-01:12:48:10,filter=raw:comment=OP1
2016-11-28:12:18:05,2016-11-28:12:18:19,filter=raw:comment=OP1
2016-11-28:12:19:10,2016-11-28:12:19:30,filter=raw:comment=OP1
2016-11-28:12:20:28,2016-11-28:12:21:36,filter=raw:comment=OP1

E.4 outdoor.select

2016-10-19:07:35:00,2016-10-19:13:11:00
2016-10-20:07:56:00,2016-10-20:10:30:00

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