

Effects of different correction algorithms on absorption coefficient – A comparison of three optical absorption photometers at a boreal forest site

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Abstract.

We present a comparison between three absorption photometers that measured absorption coefficient (σ_{abs}) of ambient aerosol particles in 2012 – 2017 at SMEAR II, a measurement station located in a boreal forest in southern Finland. The comparison included an Aethalometer (AE31), a Multi Angle Absorption Photometer (MAAP), and a Particle Soot Absorption Photometer (PSAP). These optical instruments measured particles collected on a filter, which is a source for systematic errors, since in addition to the particles, also the filter fibers interact with light. To overcome this problem, several algorithms have been suggested to correct the AE31 and PSAP measurements. The aim of this study was to research how the different correction algorithms affected the derived optical properties. We applied the different correction algorithms to the AE31 and PSAP data and compared the results against the reference measurements conducted by the MAAP. The comparison between the MAAP and AE31 resulted in a multiple scattering correction factor (C_{ref}) that is used in AE31 correction algorithms to compensate for the light scattering by filter fibers. The C_{ref} varies between different environments and our results are applicable for a boreal environment. We observed a clear seasonal cycle in C_{ref} , which was probably due to variations in aerosol optical properties, such as the backscatter fraction and single-scattering albedo, and also due to variations in the relative humidity (RH). The results showed that the filter-based absorption photometers seemed to be rather sensitive to the RH even if the RH was kept below the recommended value of 40%. The instruments correlated well ($R \approx 0.98$) but the slopes of the regression lines varied between the instruments and correction algorithms: compared to the MAAP, the AE31 underestimated the σ_{abs} only slightly (the slopes varied between 0.96–1.00) and the PSAP overestimated the σ_{abs} only a bit (the slopes varied between 1.01–1.04 for a recommended filter transmittance > 0.7). The instruments and correction algorithms had a notable influence on the absorption Ångström exponent: the median absorption Ångström exponent varied between 0.93 – 1.54 for the different algorithms and instruments.

1 Introduction

Atmospheric aerosol particles have a notable effect on the Earth's radiative balance. The particles affect the Earth's climate directly by scattering and absorbing radiation from the sun, and indirectly through aerosol-cloud interactions (Stocker et al., 2013). According to the IPCC report (Stocker et al., 2013), one of the greatest uncertainties in determining the global radiative forcing is related to the atmospheric aerosol particles. Reasons for the large uncertainty are the complex nature of aerosol-cloud interactions and also the great spatiotemporal variation of the particles (Lohmann and Feichter, 2005). Since the number concentration, size distribution, chemical composition and the shape of the particles vary in both space and time, it is challenging to model and estimate the effect that the aerosol particles have on climate on a global scale (Stocker et al., 2013).

Generally, the direct effect of aerosol particles on climate is cooling, since most of the particles scatter radiation from the sun back in to space (Stocker et al., 2013). However, if particles that are dark (i.e., highly absorbing) are located above a bright surface (i.e., highly scattering), the particles have a warming effect on climate. The sign (i.e., negative sign for the cooling effect and positive sign for the warming effect) of the aerosol forcing efficiency depends on the darkness of the particles, which is described by a single scattering albedo (ω), and on the albedo of the ground below the aerosol layer (Haywood and Shine, 1995). To determine the direct effect of aerosol particles, in addition to the information about the albedo of the surface, we need measurements of aerosol optical properties (AOPs) like scattering, backscattering and absorption coefficients (σ_{sca} , σ_{bsca} and σ_{abs}). The σ_{sca} is a measure of light scattering by the particles in all directions, the σ_{bsca} is a measure of light scattering only in the backward direction, and the σ_{abs} is a measure of particulate light absorption. All these variables are wavelength dependent, which is why the measurements of AOPs are preferably conducted at multiple wavelengths.

Measuring the σ_{sca} and σ_{bsca} is rather straightforward and the measurements are typically conducted with an integrating nephelometer. Correction algorithms and coefficients to minimize the error sources and uncertainties of integrating nephelometers are systemically used (Anderson and Ogren, 1998; Müller et al., 2011b). However, for the σ_{abs} measurements there are still large uncertainties and the error sources are not as well defined as for the σ_{sca} and σ_{bsca} measurements. The main difference between the σ_{sca} and σ_{abs} measurements is that the σ_{sca} measurements are conducted for particles suspended in air, whereas the σ_{abs} is typically measured by filter-based techniques, where the aerosol particles are collected on a filter. The problem with the filter-based measurements is that in addition to the particles, also the filter fibers interact with radiation and thus influence the measurements.

One of the issues arising specifically with the optical filter-based measurements is a multiple scattering of light by the filter fibers. The multiple scattering is considered by the so-called multiple scattering correction factor (C_{ref}). Even though the C_{ref} should only depend on the properties of the filter, previous studies have shown that the C_{ref} also depends on the particulate matter suspended in the filter (Arnott et al., 2005; Collaud Coen et al., 2010; Weingartner et al., 2003). The C_{ref} has been

observed to vary from station to station and therefore, it has been studied in different environments. For example, Collaud Coen et al. (2010) studied the C_{ref} at very clean mountain sites, in a maritime site, and in urban areas; Schmid et al. (2006) did observations in Amazonia; Backman et al. (2017) studied the C_{ref} in Arctic sites; and Kim et al. (2019) ran measurements in a maritime, high altitude, and Arctic sites. Since there is no generally accepted method for deriving the C_{ref} values, the methods between different studies vary, which can also affect the results. In this study, we derived the C_{ref} by comparing two optical filter-based instruments with each other.

Another issue with optical filter-based measurements is related to the nonlinear response of the instruments as the filter is loaded with particles. When the filter is loaded with absorbing particles, the particle loading decreases the response of the instrument. Therefore, the instruments report a lower σ_{abs} for loaded filters compared to pristine filter measurements. Several studies have developed algorithms to overcome this problem that has been observed with different instruments (Arnott et al., 2005; Bond et al., 1999; Collaud Coen et al., 2010; Li et al., 2020; Müller et al., 2014; Ogren, 2010; Schmid et al., 2006; Weingartner et al., 2003; Virkkula et al., 2005; Virkkula et al., 2007; Virkkula, 2010). In general, after correcting the data for the multiple scattering and loading effects, the absorption instruments agree rather well with the reference measurements (Drinovec et al., 2015; Hyvärinen et al., 2013; Park et al., 2010; Segura et al., 2014). The outcome of the different algorithms, however, varies and they may affect, for example, the wavelength dependency of the σ_{abs} (Backman et al., 2014; Collaud Coen et al., 2010).

This study has two aims that address the variation in the C_{ref} and the differences between the correction algorithms. The first aim is to provide C_{ref} values that are suitable for a boreal forest site and to study how the C_{ref} varies for different correction algorithms. The second aim is to present how the different correction algorithms of the σ_{abs} affect the measured and derived AOPs.

The measurements presented in this study, were conducted in 2012 – 2017 at Station for Measuring Ecosystem-Atmosphere Relations II (SMEAR II; Hari and Kulmala, 2005), which is located in the middle of a boreal forest in southern Finland. During this period, the AOPs at SMEAR II have been measured by several instruments: an integrating nephelometer and three different absorption photometers (AE31, PSAP, and MAAP), which enabled determining the C_{ref} and an extensive comparison between the different instruments and correction algorithms. AOPs at SMEAR II have been extensively discussed by Virkkula et al. (2011) and Luoma et al. (2019), however, these studies focused on the temporal variation in the AOPs and they only discussed nephelometer and AE31 data. In this study, we focus on the technical side of the measurements and instrument comparison.

2 Measurements and methods

2.1 The field site

The measurements took place at SMEAR II (Station for Measuring Ecosystem-Atmosphere Relations; Hari and Kulmala, 2005). The measurement station locates in Hyytiälä, Southern Finland (61°51'N, 24°17'E). SMEAR II is a rural measurement station and it represents boreal forest environment. The area around the station is mostly forests that consists mainly of scots pine trees (Hari et al., 2013). The site is classified as rural and there are no significant sources of pollution nearby. The area is sparsely populated; in the nearby area there are few smaller towns and some scattered settlements. The closest bigger cities are Tampere (220 000 inhabitants) and Jyväskylä (140 000 inhabitants) and they are located 60 and 100 km away from the station.

2.2 Instrument set-up

The measurements of AOPs for PM₁₀ particles were started already on June 2006 with an integrating nephelometer (TSI model 3563) and an Aethalometer (Magee Scientific model AE31). Later on there has also been a Particle Soot Absorption Photometer (PSAP; Radiance Research model 3-λ; Virkkula et al., 2005) and a Multi-Angle Absorption Photometer (MAAP; Thermo model 5012; Petzold and Schönlinner, 2004).

The measurement arrangement of the instruments, which measured the AOPs, is presented in Fig. 1. The schematic figure represents the measurement line from a period when all the instruments mentioned before were measuring in parallel, which was during 2014 – 2015. In the start of the measurement line, a pre-impactor removed all the particles that were larger than 10 µm in aerodynamic diameter (i.e., PM₁₀ passed the pre-impactor). The air flow through another impactor, which removed all the particles larger than 1 µm (i.e., PM₁ passed the impactor) was controlled by two valves. The valves changed the direction of the flow in every 10 minutes, so that in a 20-minute measurement cycle the instruments were exposed 10 minutes to the PM₁₀ and then 10 minutes to the PM₁. To hinder the effect of changing inlets, the first minutes of the measurements after the inlet switch were omitted. For the absorption instruments the first three minutes and for the integrating nephelometer the first five minutes were omitted. The sample air was dried with Nafion dryers for the PSAP, AE31, and integrating nephelometer for the whole period and for the MAAP since March 2017. Also, a Cavity Attenuated Phase Shift extinction monitor (CAPS, Aerodyne Research; Kebabian et al., 2007) is marked in Fig. 1, since it was part of the measurement line. However, due to technical issues CAPS data was not applied in this study.

Even though the measurements of AOPs have been conducted at SMEAR II since 2006, in this study, we consider only data measured after January 2012 until December 2017. This period was selected to have at least two absorption instruments running in parallel: the AE31 stopped operating in December 2017, the PSAP operated from January 2012 to March 2016, and the

MAAP started operation in June 2013. Also, during this period there were only few changes in the measurement line: in March 2017 the MAAP flow was decreased from 18 lpm to 9 lpm and Nafion dryers were installed in front of MAAP; and in November 2017 one of the two Nafion dryers were removed in front of the nephelometer.

5 The instruments measured AOPs at different wavelengths: the integrating nephelometer at three wavelengths (450, 550 and 700 nm); the AE31, the PSAP and the MAAP measured at seven (370, 470, 520, 590, 660, 880 and 950 nm), three (467, 530 and 660 nm), and one wavelength (637 nm), respectively. Here, we reported the typically used AE31 and PSAP wavelengths, which are reported in the AE31 manual and by Virkkula et al. (2005), respectively. These reported wavelengths deviate slightly from the ones measured and reported by Müller et al. (2011a) (see their Table 6). For the MAAP, we decided to use the
10 wavelength reported by Müller et al. (2011a) since it more commonly used and it clearly deviated from the wavelength reported by the manual.

The data availability of all the instruments for the studied period sets are reported in Fig. S1. Some of the data were missing or invalidated due to instrument malfunctions, too high relative humidity (*RH*), or due to the absence of the instrument because
15 of workshops or campaigns. If the *RH* exceeded 40% in an instrument, the data was marked as invalid according to recommendations (WMO/GAW, 2016). Before the dryers were installed for the MAAP in March 2017, some of the MAAP data, especially from the summer, was invalidated due to too high *RH*. During the cold season, the indoor temperature at the measurement cottage was higher than outdoors and therefore the *RH* decreased when the sample air was warmed up to the indoor temperature (passive drying). However, in the summer the *RH* sometimes increased above the accepted limit since the
20 passive drying was not enough due to minimal difference between the indoor and outdoor temperature.

2.3 Absorption measurements

As mentioned above, the σ_{abs} of aerosol particles at different wavelengths at SMEAR II was measured with three different instruments: AE31, PSAP and MAAP. Each of these instruments measured the σ_{abs} by filter-based technique, which means that the measurements are conducted for aerosol particles that are collected on a filter. AE31 operated on quartz fiber filter
25 (Pallflex, type Q250F), PSAP on quartz fiber filter (Pall, type E70-2075W), and MAAP on glass fiber filter (Thermo Scientific, type GF 10).

The AE31 and the PSAP have a similar measurement principle (Bond et al., 1999). Before the σ_{abs} can be determined by using different correction algorithms, the instruments measure the attenuation coefficient (σ_{ATN}), which is the attenuation of light
30 through the sample collected on the filter. The equation for the σ_{ATN} is derived from the Beer-Lambert-Bouguer law

$$\sigma_{\text{ATN}} = \frac{A}{Q \Delta t} \ln \frac{I_{t-\Delta t}}{I_t} = \frac{A}{Q} \frac{\Delta \text{ATN}}{\Delta t}, \quad (1)$$

where A is the sample area on the filter, Q is the flow through the filter, and Δt is the length of the measurement period. $I_{t-\Delta t}$ and I_t are the measured and normalized light intensities through the filter in the beginning of the measurement period ($t - \Delta t$) and in the end of the measurement period (t). The intensities are normalized by comparing them to the intensity measured through a clean reference spot. Normalizing the intensities accounts for possible drifts and changes in the intensities of the LEDs. ΔATN is the change in attenuation (ATN), which is calculated from the ratio of light intensity through a clean filter (I_0) and through a loaded filter (I_t) as

$$ATN = -\ln\left(\frac{I_t}{I_0}\right) \cdot 100\% . \quad (2)$$

In addition to the ATN , the filter loading can also be described by transmittance (Tr)

$$Tr = I_t I_0^{-1}, \quad (3)$$

which can be also presented as a function of ATN ($Tr = \exp(ATN/100\%)$). The ATN and Tr represent essentially the same concept, but the way of expressing the change of intensity depends on the instrument used: the ATN is traditionally associated with Aethalometer data and Tr with PSAP data.

In Eq. 1, A is typically a constant value defined by the manufacturer and Q is recorded and reported by the instrument. These values, however, might deviate notably from the real values, and therefore they should be measured and checked regularly. If these values differ from the reported ones, the Eq. 1 needs corrections for the A and the Q . At SMEAR II, the sample flow of each instrument was regularly measured with a gilliam flow meter and the Q reported by the instruments was corrected to match the gilliam measurements. For the PSAP and AE31 we used the A values of 18.1 and 54.8 mm² that deviated from the default ones, which were 17.8 and 50.0 mm², respectively. The A used by default in the MAAP matched the measured one and therefore it was not corrected.

In a filter, the light is attenuated because of the absorption and scattering by the particles, but also because of the scattering by the filter fibers, which is called multiple scattering. The scattering by the filter fibers increase the optical path of the light beam through the filter. Therefore, the probability for the light beam to be absorbed by a particle increases. Because of the scattering in the filter medium, the σ_{ATN} is larger than the σ_{abs} . Not only the filter fibers scatter light, but also the embedded aerosol particles scatter light and cause so-called apparent absorption, which is typically considered by subtracting a fraction of scattering from the σ_{ATN} . In addition to the scattering by the fibers and particles, also the increasing number of absorbing particles in the filter affects the instrumental response. The signal response caused by the particulate absorption decreases with increasing filter loading. Absorbing particles induce a so-called shadowing or a loading effect, which decreases the change in the intensity ($I_{t-\Delta t} I_t^{-1}$) as the filter gets more loaded (Weingartner et al., 2003). This means that the instrumental response is non-linear with increasing filter loading. The increasing filter loading has an opposite effect than the scattering of the filter fibers and particles: the absorbing particles collected on the filter decrease the optical path and therefore the reported σ_{ATN} for

a loaded filter is lower than for a pristine filter. This non-linearity is considered in the various correction algorithms presented in Sects. 2.3.1 and 2.3.2.

5 The measurement principle of the MAAP is different from that of the AE31 and PSAP (Petzold and Schönlinner, 2004). In addition to the light attenuation measurements, the MAAP also measures the backscattered light from the filter in two different angles. The σ_{abs} is then obtained by using a radiative transfer scheme where the measurements of the backscattering and light attenuation are taken into account (Petzold and Schönlinner, 2004). Because of the backscattering measurements, the MAAP does not suffer as much from the filter artefacts as the Aethalometer and the PSAP. However, in very polluted environments also the MAAP suffers from a measurement artifact that has to be corrected (Hyvärinen et al., 2013), which at SMEAR II, 10 however, is not the case.

At SMEAR II, the MAAP advanced the filter spot automatically once per day in 24 h intervals. The AE31 also advanced the spot automatically when the *ATN* reached 120 at 370 nm wavelength. The PSAP filters were changed manually and the aim was to be change the filter every second day, but due to weekends and holidays, the filters were sometimes changed only after 15 several days. On average the PSAP filters were changed once per three days.

The reported uncertainties of the MAAP, PSAP and Aethalometer are respectively 12, 13, and as large as 50 % (Amott et al., 2005; Ogren, 2010; Petzold and Schönlinner, 2004). Müller et al. (2011a) reported that the unit-to-unit variability of the PSAP, AE31 and MAAP were about 8%, 20% and 3%. It must be noted that the unit-to-unit variability is a lot smaller, about 2 % for 20 the new AE33 model (Cuesta-Mosquera et al., 2021). Since the uncertainty and unit-to-unit variability of the MAAP was a lot smaller than for the PSAP and AE31 we used the MAAP as the reference instrument for measuring σ_{abs} . However, even though the MAAP was used as the reference here, it must be remembered that like all the filter-based photometers, also MAAP suffers from the cross sensitivity to purely scattering aerosol and therefore it does not the best reference instrument (Müller et al., 2011a).

25 Each of the absorption photometer used in this study have their strengths and weaknesses that determine which instrument is the most useful in different situations. According to the uncertainty and unit-to-unit variability, the MAAP is the most precise instrument for monitoring σ_{abs} and black carbon (BC) concentration, which is typically derived from σ_{abs} measurements. Also, the backscattering measurements from the filter reduce the artefacts caused by the scattering aerosol particles and the filter loading effect making it a more accurate instrument. The MAAP changes the spot in a filter roll automatically and therefore it 30 does not require much assistance from the operator and the instrument can run at a remote station as well. However, it measures the σ_{abs} only at one wavelength, so it is not possible to do the source apportionment or interpretation on the chemical composition of the absorbing particles, which requires measurements on several wavelengths (see Sect. 3.1 and Eq. 16). The AE31 has a very wide range of wavelengths, which makes the seven-wavelength Aethalometers, the AE31 and the new model

AE33, widely used instruments. Like MAAP, the AE31 also operates the filter roll automatically so the instrument does not need that much assistance from the operator. Unfortunately, the problems with defining the errors caused by the filter material are not that well defined, and the instrument uncertainty and unit-to-unit variability of the AE31 is large. The uncertainty and noise of the PSAP is smaller than that of the AE31, which makes the PSAP a popular especially in areas with low concentrations. Even though the wavelength range is not as wide as with AE31, the PSAP measures the σ_{abs} at three wavelengths, allowing the use of applications that need the wavelength dependency of σ_{abs} . The PSAP filters have to be changed manually by the user so the instrument is not the best option to deploy at a remote site, but then again the leakage through the filter tape is lesser than for the MAAP and AE31.

2.3.1 AE31 correction algorithms

To determine the σ_{abs} from AE31 measurements, the σ_{ATN} needs to be corrected for the multiple scattering by the filter fibers and for the error caused by the filter loading, and in addition, the scattering of aerosol particles should also be taken into account

$$\sigma_{\text{abs}} = \frac{\sigma_{\text{ATN}} - a_s \sigma_{\text{sca}}}{C_{\text{ref}} R(\text{ATN})}. \quad (4)$$

The effect of the multiple scattering is corrected with a multiple scattering correction factor (C_{ref}) and it is larger than unity. For the filter loading correction ($R(\text{ATN})$) there are different kind of correction algorithms developed for example by Weingartner et al. (2003), Arnott et al. (2005), Schmid et al. (2006), Virkkula et al. (2007), and Collaud Coen et al. (2010). The R , which equals unity for unloaded filters is less than unity for loaded filters, depends on the filter loading, i.e., ATN defined in Eq. (2). The R can also depend on other factors, such as the ω , and some of the algorithms take also other parameters than ATN into account.

In Eq. (4), the scattering by the aerosol particles is considered by subtracting a fraction (a_s) of the measured scattering (σ_{sca}). However, the algorithms by Weingartner et al. (2003) and Virkkula et al. (2007) ignore the particle scattering subtraction, which makes it possible to apply the corrections algorithms without any σ_{sca} measurements. In Weingartner's algorithm, however, σ_{sca} is considered without the subtraction, as will be shown below. For a comparison, in this study we also present data, which was corrected only for the multiple scattering and not for the filter loading (i.e., the $R = 1$) or scattering by the particles. Below we present the different algorithms determined by Weingartner et al. (2003), Arnott et al. (2005), Virkkula et al. (2007), and Collaud Coen et al. (2010), which were selected to use in this study.

Current recommendation by the WMO and GAW is to assume the $R(\text{ATN})$ unity for the AE31 and to use a C_{ref} value of 3.5, which was determined by a comparison study of different AE31 instruments (WMO/GAW, 2016). Therefore, we also studied

“not-corrected” AE31 data for which we did not apply any $R(ATN)$ correction or particulate scattering reduction, but only the multiple scattering correction.

Weingartner et al. (2003) (hereon referred as W2003 and with a subscript WEI) derived an empirical correction algorithm based on laboratory measurements of mixed particles (soot, diesel exhaust, organic coating, ammonium sulfate). The W2003 correction algorithm interpolates the measurements at higher ATN values, to a point where ATN was 10%. When ATN is lower than 10%, the R is assumed unity. In W2003, the loading correction (R_{WEI}) is

$$R_{WEI}(ATN) = \left(\frac{1}{f} - 1\right) \frac{\ln(ATN) - \ln(10\%)}{\ln(50\%) - \ln(10\%)} + 1. \quad (5)$$

Weingartner et al. (2003) stated that the R depends on the single-scattering albedo (ω) and they found the following relation for the factor f

$$f = a(1 - \omega) + 1. \quad (6)$$

In Eq. 6, f is unity (i.e., R is unity), when ω is unity (i.e., the aerosol is purely scattering). Weingartner et al. (2003) determined that a in Eq. 6 was 0.87 and 0.85 at 450 and 660 nm, respectively. According to these two values, we interpolated a for all the seven wavelengths by assuming a linear wavelength dependency. Also, ω was interpolated to the seven AE31 wavelengths according to the mean σ_{abs} , σ_{sca} and scattering Ångström exponent (α_{sca}) values reported by Luoma et al. (2019; see their article Table 1) for PM10 particles. Using these values, we estimated f separately for each wavelength and we used those constant values in the correction values. The resulting a , ω , and f were slightly wavelength dependent and their values are presented in Table 1.

The correction algorithm does not apply the scattering correction by subtraction, so the $a_{s,WEI} = 0$ and therefore parallel scattering measurements are in principle not needed. However, the effect of the particulate scattering is considered in the f since it depends on the ω . If there are no parallel measurements of σ_{sca} , the ω cannot be determined. If there is no estimation on the ω , typically f values for different aerosol types determined by Weingartner et al. (2003) are used. The f values were close to the result Weingartner et al. (2003) acquired from measurements of ambient aerosols in a high alpine site and in a garage (f was 1.03 and 1.14 for a “white light” Aethalometer; AE10). For example, Collaud Coen et al. (2010) estimated an intermediate value $f = 1.10$ for Cabauw measurements site based on the study by Weingartner et al. (2003).

Arnott et al. (2005) suggested a correction algorithm, which is hereon we referred as A2005 and with the subscript ARN, based on a well-defined theoretical basis. One big difference to the W2003 is that there is a factor for scattering subtraction. Arnott et al. (2005) determined the scattering subtraction fraction $a_{s,ARN}$ from laboratory measurements using submicron ammonium sulfate particles and the values for different wavelengths are presented in Table 1, however, Arnott et al. (2005) noted that the values of $a_{s,ARN}$ could be different if supermicron aerosol particles are present. The loading correction R_{ARN} was defined as:

$$R_{\text{ARN}} = \left(\sqrt{1 + \frac{\left(\frac{V\Delta t}{A}\right) \sum_{i=1}^{n-1} \sigma_{\text{abs},i}}{\tau_{a,fx}(\lambda)}} \right)^{-1}, \quad (7)$$

where the n indicates the n^{th} measurement after a filter spot change. The correction takes in to account the cumulative σ_{abs} of the particles collected on the filter material. The $\tau_{a,fx}(\lambda)$ is the filter absorption optical depth for the filter fraction x that has particles embedded. To calculate the $\tau_{a,fx}(\lambda)$, we used the same a power law function $\tau_{a,fx}(\lambda) = \tau_{a,fx,521} \cdot (\lambda/521 \text{ nm})^{-0.754} = 0.2338 \cdot (\lambda/521 \text{ nm})^{-0.754}$. $\tau_{a,fx}$ as Virkkula et al. (2011) and the resulted values are presented in Table 1. The exponent -0.754 was obtained from a power function fitting to $\tau_{a,fx}$ vs. λ (Table 1 of Arnott et al., 2005), similar as in Virkkula et al. (2011). The $\tau_{a,fx,521} = 0.2338$ is the recommended $\tau_{a,fx}$ value for ambient measurements at 521 nm (Arnott et al., 2005).

Virkkula et al. (2007) proposed a correction algorithm, which is hereon referred as V2007 and with the subscript VIR, that utilizes the so-called compensation parameter (k). The k is determined by comparing the last measurements of a loaded filter to the first measurements conducted with a pristine filter. The compensation parameter is determined for each filter spot (fs) as follows:

$$k_{fs} = \frac{\sigma_{\text{ATN}}(t_{fs+1,\text{first}}) - \sigma_{\text{ATN}}(t_{fs,\text{last}})}{\text{ATN}(t_{fs,\text{last}})\sigma_{\text{ATN}}(t_{fs,\text{last}}) - \text{ATN}(t_{fs+1,\text{first}})\sigma_{\text{ATN}}(t_{fs+1,\text{first}})}, \quad (8)$$

where the “first” refers to the mean of three first values in a pristine filter (i.e., $fs + 1$) and the “last” refers to the mean of three last values in a loaded filter (i.e., fs). The k is then applied in the loading correction R_{VIR}

$$R_{\text{VIR}}(\text{ATN}) = (1 + k_{fs} \text{ATN})^{-1}. \quad (9)$$

This algorithm does not take into account the scattering correction so the $a_{s,\text{VIR}} = 0$.

Collaud Coen et al. (2010) applied this correction to data from several stations in Europe and found that it was highly nonstable and that it leads to large outliers. They correctly stated that the difficulty of applying this correction is due to the natural high variability of σ_{ATN} as a function of time, which is for most of the time greater than the σ_{ATN} decrease induced by filter changes. We therefore calculated the running average compensation parameter for all seven wavelengths in order to minimize these problems. Then we applied this averaged compensation parameter to correct the non-corrected AE31 data. In other words, the AE31 data were not averaged at this stage, just the compensation parameter.

We determined the k as a 14-day running mean (± 7 days around the changing time of the filter spot), since without the averaging the k was very noisy (see time series for the non-averaged and averaged k in Fig. S6). Averaging the k was also recommended by Virkkula et al. (2007). On average, the 14-day-periods included about nine data points (i.e., the filter spot changed once per 1.6 days). Virkkula et al. (2015) used a similar approach for AE31 data from Nanjing, China, and calculated 24-hour running averages of the k including on the average six filter spot changes.

Collaud Coen et al. (2010), which is hereon referred as CC2010 and with the subscript COL, correction algorithm was based on the W2003 algorithm but here the reference ATN for the clean filter is 0 % instead of 10 %. They determined the a used in Eq. 6 a bit differently and got a mean value of $a = 0.74$ over different wavelengths and different experiments. The R_{COL} is defined as

$$R_{COL}(ATN) = \left(\frac{1}{a(1-\bar{\omega}_{0,n})+1} - 1 \right) \cdot \frac{ATN}{50\%} + 1. \quad (10)$$

Here the $\bar{\omega}_{0,fs,n}$ stands for the mean $\bar{\omega}_0$, that was calculated for the filter spot from the first measurements to the n^{th} measurement. The $\bar{\omega}_{0,fs,n}$ was determined by using the σ_{ATN} as the first estimate of the σ_{abs} . CC2010 differs from the W2003 also by considering the scattering correction. They suggested two different kind of ways to determine $a_{s,COL}$ and here we present the one that was determined in a manner similar to A2005. The difference to the $a_{s,ARN}$ is that the $a_{s,COL}$ is determined from the ambient scattering measurements so it is not constant. The $a_{s,COL}$ is defined similarly as by Arnott et al. (2005) (Eq. 8 in their article), but here they used measured scattering properties instead of constant values determined by laboratory measurements:

$$a_{s,COL} = \bar{\beta}_{sca,n}^{d-1} c \lambda^{-\bar{\alpha}_{sca,n}(d-1)}, \quad (11)$$

where $d = 0.564$ and $c = 0.329 \cdot 10^{-3}$. In Eq. 11 the over lined variables, $\alpha_{sca,n}$ and $\beta_{sca,n}$, stand for average properties of aerosols deposited in the filter i.e., mean values from the beginning of the filter measurements until the n^{th} measurement. The $\alpha_{sca,n}$ is the scattering Ångström exponent (see Sect. 3.1 and Eq. 16) and the β_{sca} is acquired from the power-law fit of the wavelength dependency of σ_{sca} :

$$\sigma_{sca} = \beta_{sca} \lambda^{-\alpha_{sca}}, \quad (12)$$

where the fit is calculated with λ and σ_{sca} in units nm and Mm^{-1} to acquire unitless β .

2.3.2 PSAP correction algorithms

Since the measurement principles of PSAP and AE31 are basically the same, the PSAP data needs similar kind of corrections as the AE31 data (Eq. 2). In this study the PSAP data was corrected with two algorithms: one described by Bond et al. (1999) and later specified by Ogren (2010), which is hereon referred as the B1999; the other determined by Virkkula et al. (2005) and later corrected by Virkkula (2010), which is hereon referred as the V2010. The algorithms of Müller et al. (2014) and Li et al. (2020) were not applied.

The B1999 correction algorithm revisited by Ogren et al., 2010 is given by

$$\sigma_{abs} = f(Tr) \sigma_0 - a_s \sigma_{sca}, \quad (13)$$

where

$$f_{Tr} = (1.5557 \cdot Tr + 1.0227)^{-1}. \quad (14)$$

In the V2010 correction algorithm, the σ_{PSAP} is determined in an iterative manner. The first estimation of absorption coefficient ($\sigma_{abs,0}$), which is determined by $\sigma_{abs,0} = (k_0 + k_1 \ln(Tr)) \sigma_{ATN} - s \sigma_{sca}$, where the k_0 and k_1 are constants presented in Table 1

in Virkkula (2010). The $\sigma_{\text{abs},0}$ is used to calculate the single scattering albedo ω (see Sect. 3.1 and Eq. 17), which is then again used to calculate the σ_{abs} again in an iterative manner, but now with a different kind of an equation

$$\sigma_{\text{abs}} = (k_0 + k_1 h(\omega_0) \ln(Tr)) \sigma_{ATN} - s \sigma_{\text{sca}}, \quad (15)$$

where the $h(\omega_0) = h_0 + h_1 \omega$. The ω is then calculated again with Eq. 17. These two steps are repeated until the change in the σ_{abs} is minor. Here, the iteration was stopped one the change was less than 1%. It must be noted that this correction algorithm is different from the V2007 determined for the Aethalometer data.

2.3.3 Differences between the algorithms

The W2003 algorithm only depends on the ATN , otherwise it applies constant values and it does not consider the scattering subtraction. The A2005 is not a function of ATN but it takes the filter loading into account by summing the σ_{abs} of the accumulated particles on the filter spot. It does not assume a constant for the scattering reduction but determines the fraction from the wavelength dependency of σ_{sca} . The CC2010 algorithm is similar to A2005 in a sense that it also defines its own scattering reduction factor and determines the filter loading correction by taking into account the properties of the particles accumulated in the filter. The V2007 only depends on the difference between the last and first measurements of two filter spots and it assumes no constant coefficients. The B1999 algorithm relies heavily on constants that describe the dependency on the Tr , whereas the V2010 algorithm is an iterative process that depends on the ω . Both B1999 and V2010 consider the scattering reduction with a coefficient.

2.4 Scattering measurements

The σ_{sca} data is needed to subtract a fraction of particulate scattering from the σ_{ATN} in A2005, CC2010, B1999 and V2010. Measurements of σ_{sca} and σ_{bsca} are also needed in determining the ω and the backscatter fraction (b , see Sect. 3.1 and Eq. 18), which are used to explain the observed variations in the results. The ω is also used in CC2010.

The σ_{sca} and σ_{bsca} were measured with an integrating nephelometer (TSI model 3565, Anderson et al., 1996). The integrating nephelometer measured σ_{sca} and σ_{bsca} at three wavelengths (450, 550 and 700 nm). Due to instrumental restrictions, the nephelometer can only measure σ_{sca} on the range of $7^\circ - 170^\circ$ and σ_{bsca} on the range of $90^\circ - 170^\circ$, and therefore a truncation correction is applied to σ_{sca} and σ_{bsca} measurements (Anderson and Ogren, 1998; Bond et al., 2009). The fractional uncertainty of the integrating nephelometer for PM10 has been reported to be $\pm 9\%$ (Sherman et al., 2015). Since scattering by aerosol particles is depends significantly on their size, the particulate light scattering is sensitive to hygroscopic growth. To prevent this, the integrating nephelometer operated with two Na fion-driers as shown in Fig. 1.

3 Data analysis

All the data were averaged to 1 h intervals. The PM1 and PM10 measurements were not separated in the data analysis and the PM1 and PM10 data were averaged together. Since all the instrument measured the same sample air, combining the PM1 and PM10 data caused no discrepancies between the instruments. Since this study discusses filter-based absorption photometers and the *ATN* in the filters decreases due to the accumulation of both PM1 and PM10, it would have been difficult to separate the effect of the different size cuts in the data analysis and therefore the data of different size cuts were combined.

3.1 Intensive properties

The intensive properties of aerosol particles are determined from the measurements of the extensive properties, which in our data are σ_{abs} , σ_{sca} , and σ_{bsca} . In addition to the chemical properties and size distribution, the extensive properties also depend on the number and volume concentration of particles. The intensive properties, however, are independent of the amount of particles and they depend only on the properties of the particles, such as the shape of the size distribution, chemical composition and shape of the particles. Therefore, intensive properties are useful parameters as they indirectly indicate the properties of the particle population. The intensive properties used in this article are the Ångström exponent (α), single scattering albedo (ω), and backscatter fraction (b) and they are presented below.

The Ångström exponent, (α) describes the wavelength dependency of the optical properties and it can be calculated for example for σ_{abs} and σ_{sca} to acquire the absorption Ångström exponent (α_{abs}) and scattering Ångström exponent (α_{sca}), respectively. The α is defined by

$$\alpha = -\frac{\ln \frac{\sigma_1}{\sigma_2}}{\ln \frac{\lambda_1}{\lambda_2}}, \quad (16)$$

where the σ_1 and σ_2 are the property for which the α is being calculated at wavelengths λ_1 and λ_2 , respectively. The α is typically used to interpolate or extrapolate optical properties to different wavelengths. This is useful for example in cases, when instruments measure optical properties at different wavelengths and the measurements between different instruments need to be compared. The wavelength dependency also gives information about the size distribution, chemical composition, and sources of the particles: α_{sca} depends on the size distribution of the particles and α_{abs} depends on both the chemical composition and size distribution. The α_{abs} is typically used in a set of empirical equations, that approximate the source of black carbon (BC) (Srandadewi et al., 2008; Zotter et al., 2017).

The single-scattering albedo (ω), which describes how big fraction of the total light extinction ($\sigma_{\text{ext}} = \sigma_{\text{abs}} + \sigma_{\text{sca}}$) is due to scattering:

$$\omega = \frac{\sigma_{\text{sca}}}{\sigma_{\text{sca}} + \sigma_{\text{abs}}}. \quad (17)$$

The lower ω is, the darker the aerosol particles are, which is typically caused by a higher content of black carbon (BC); ω close to unity indicates that the particles are high in scattering material like sulfates or sea salt. Therefore, ω is a rough indicator of the chemical composition of the particles.

- 5 The backscatter fraction (b) describes the fraction of backscattering coefficient (σ_{bsca} ; meaning that the light scatters in the backward hemisphere) of the total scattering coefficient (σ_{sca}):

$$b = \frac{\sigma_{bsca}}{\sigma_{sca}}. \quad (18)$$

The b is also size dependent. In molecular size range it is 0.5, which means that the particles scatter light evenly in the forward and in the backward direction. For larger particles the b decreases, so the particles scatter light more in the forward direction.

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3.2 Multiple scattering correction factor

As stated by Weingartner et al. (2003), the C_{ref} should in principle only depend on the instrument and the filter material used. The effect caused by different amounts of particles deposited in the filter material and their optical properties should be taken into account by the empirical filter loading correction functions $R(ATN)$. However, as shown by previous studies, the C_{ref} varies
15 both spatially and temporally (Backman et al., 2017; Collaud Coen et al., 2010) and therefore we determined the C_{ref} also at SMEAR II.

In this study, the multiple scattering correction factor (C_{ref}) was defined for the AE31 measurements by using the σ_{abs} measured by the MAAP as the reference absorption coefficient ($\sigma_{abs,ref}$). To determine the C_{ref} , the σ_{ATN} measured by the AE31 had to be
20 corrected for the artefact caused by the increased filter loading and then the measurements can be compared to the reference absorption ($\sigma_{abs,ref}$) measured by the MAAP

$$C_{ref} = \frac{\sigma_{ATN}}{R(ATN) \sigma_{abs,ref}}. \quad (19)$$

The C_{ref} was defined separately for data corrected using W2003, A2005, V2007, and CC2010 to obtain the C_{WEI} , C_{ARN} , C_{VIR} , and C_{COL} respectively. The C_{ref} was also determined for data, which were not corrected for the filter loading (C_{NC} , where
25 subscript NC stands for “not corrected”). Because the MAAP measures the $\sigma_{abs,ref}$ only on the wavelength 637 nm, the closest AE31 and nephelometer data were first interpolated to the same wavelength. The σ_{ATN} , ATN , and σ_{sca} were interpolated to 637 nm by applying the Ångström exponent explained in Eq. 16. Also, the wavelength dependent constants used in W2003 and A2005 were interpolated to 637 nm. The f used in W2003 at 637 nm was 1.12 and the $a_{s,ARN}$ and $\tau_{a,fx}$ used in A2005 were 0.0681 and 0.2009 at 637 nm, respectively.

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In the A2005, cumulative optical properties of the particles collected on the filter were needed, and thus the C_{ARN} was determined by iterating; the C_{ARN} was iterated for each filter spot until the median $\sigma_{abs,ref}$ and $\sigma_{abs,ARN}$ agreed within a 1% limit.

Because of the iteration, there is one C_{ARN} value for each filter spot. For other correction algorithms, the C_{ref} value was determined by two methods: 1) C_{ref} was determined as the slope of a linear regression for the whole data set (linear fit for a loading corrected σ_{ATN} vs. σ_{ref} -plot; Eq. 19); and by 2) simply using the Eq. 19 to acquire the C_{ref} value for each measurement point separately. In the A2005, the C_{ARN} depends on the wavelength. In this study the C_{ARN} was determined only at the 637 nm. Since we followed a similar procedure as presented by Amott et al. (2005), a fraction of the σ_{sca} was first subtracted from the σ_{ATN} , before determining the C_{ARN} , which is different to Eq. 19.

4 Results and discussion

4.1 Multiple scattering correction for the AE31

The different C_{ref} values were determined by a linear fit by comparing loading corrected AE31 data to the reference data from MAAP. Since the C_{ref} is described only by the slope of the fit, the intercept in the y-axis of the fit was forced to be zero. For the linear fit we used all the available parallel data from the AE31 and MAAP. The C_{ref} values were 3.00, 3.14, 2.99, and 2.77 for data corrected by W2003, V2007, CC2010, and for data that was not corrected, respectively. The results and their statistical variability are presented in Table 2. The relatively small standard error (SE) and the range of confidence interval (CI) indicate that the difference between the C_{ref} values were statistically significant. However, for example, the difference between C_{WEI} and C_{COL} was small.

The smallest determined C_{ref} value was the C_{NC} , which was expected. Since the σ_{ATN} decreases for a loaded filter and the filter loading correction was not applied, the C_{NC} had to be smaller than for the corrected data. Since the values of the C_{WEI} and C_{COL} were almost the same, the result suggested that the loading corrections R_{WEI} and R_{COL} had on average a similar effect on the data. The highest value was determined for the C_{VIR} , which suggests that on average, the value of the R_{VIR} was the lowest (i.e., the effect of filter loading correction in V2007 was stronger).

Since the C_{ARN} was determined in an iterative manner for each filter spot, the C_{ARN} was calculated as the median of all the filter spots and the resulted value was 3.13, which is also shown in Table 2. This result is not directly comparable to the other C_{ref} values that were derived as a linear fit. Also, unlike the other algorithms, the A2005 assumed a wavelength dependent C_{ARN} . Here, we were only able to determine the C_{ref} at one wavelength by comparing the interpolated AE31 data to the MAAP measurements at 637 nm. To acquire the C_{ARN} at all the seven wavelengths of the AE31, we used the power law function $C_{ARN}(\lambda) = C_{ARN,637nm}(\lambda/637\text{ nm})^{0.181} = 3.13 \cdot (\lambda/637\text{ nm})^{0.181}$, where the exponent 0.181 was obtained from a power function fitting to C_{ref} vs. λ in Table 1 of Amott et al. (2005), similar to Virkkula et al. (2011). $C_{ARN,637nm} = 3.13$ is the value determined above at $\lambda = 637$ nm. The results of the wavelength dependent C_{ARN} values are presented in Table 3.

According to Collaud Coen et al. (2010), who studied the C_{ref} of different algorithms for ambient measurements in different environments, the higher C_{ref} values were typically measured in polluted areas. Observations in our study support this claim. For example, they determined mean C_{WEI} of 2.81, 2.81, 3.05, and 4.09, at Hohenpeissenberg, Jungfraujoch, Mace Head, and Cabauw, respectively. Segura et al. (2014) obtained a C_{ref} value of 4.22 measured in Granada, Spain at 637 nm for the correction algorithm by Schmid et al. (2006). Compared to their study the C_{ref} values at the SMEAR II were obviously lower than the mean C_{ref} values at the Cabauw and Granada measurement stations. Cabauw station is located near populated and industrial areas and the station in Granada is located close to a highway. At SMEAR II, the average C_{ref} values were somewhat higher than in the clean mountain stations in Hohenpeissenberg and Jungfraujoch. The closest values were defined for the Mace Head station, which observes mostly marine air. The C_{ref} values by Collaud Coen et al. (2010) and Segura et al. (2014) were determined similar to our study, so by comparing AE31 measurements against MAAP.

Backman et al. (2017) determined C_f (Backman et al., 2017, used the symbol C_f instead of C_{ref} to mark that the comparison was not conducted with a reference instrument) values for ambient data at several Arctic sites. They also derived the C_f optically by comparing Aethalometer measurements against MAAP, PSAP and CLAP (Continuous Light Absorption Photometer; Ogren et al., 2017). They ran the comparison for Aethalometer data that were not corrected for the filter loading error. The median C_f values at 637 nm were 1.61, 3.12, 3.42, 4.01, and 4.22 measured at Summit, Barrow, Alert, Tiksi, and Pallas, respectively. Backman et al. (2017) did not find any clear explanation for the very low C_f at Summit. At the other sites, the C_f values were rather high compared to the C_{NC} observed at SMEAR II ($C_{NC} = 2.77$), which is unexpected if we would assume that the C_{ref} was lower at clean environments, such as Arctic, compared to sites closer to pollution sources.

In laboratory runs, Amott et al. (2005) determined $C_{ref} = 2.076$ (at 521 nm) for kerosene soot by comparing AE31 against photoacoustic instrument and Weingartner et al. (2003) observed $C_{ref} = 2.14$ (averaged over wavelengths) for not-coated soot particles by subtracting scattering from extinction measurements. Compared to the C_{ref} determined in laboratory studies by Weingartner et al. (2003) and Amott et al. (2005), the ambient measurements in our study yielded higher values. This was also observed by Amott et al. (2005), who suggested $C_{ref} = 3.688$ (at 521 nm) for ambient measurements, which is closer to our observations. In addition to not-coated soot, Weingartner et al (2003) determined the C_{ref} for coated particles as well and the resulted C_{ref} was higher, about 3.6. This is also closer to our observations, which is explained by the fact that at SMEAR II, the observed soot particles are likely aged and coated since there are no significant local emission sources. In these studies, however, the reference instruments were not filter-based photometers and that can have an effect on the results.

The report 227 by WMO and GAW (World Meteorological Organization and Global Atmosphere Watch) recommends to determine the σ_{abs} from Aethalometer measurements by using C_{ref} value of 3.5 and not applying any filter loading correction or particle scattering reduction in the data. The C_{ref} was determined as an average over several datasets collected from different GAW stations. Comparing this value to the C_{NC} , using the recommended $C_{ref} = 3.5$ would systematically underestimate the σ_{abs} at SMEAR II by ~20%. It must be also noted, that due to the lack of $R(ATN)$ correction, the BC concentration or σ_{abs} may

differ as much as 50% even if the true σ_{abs} would stay constant (Arnott et al., 2005). Therefore, in some cases, the data user may want to take the error caused by the filter loading into account and to use different correction algorithms. For example, when studying shorter time periods (e.g., few days of data, which may fit few filter spot changes causing apparent variation in the measured concentration).

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There are both studies where constant C_{ref} has been used and studies where wavelength dependent C_{ref} has been used. Others observed no significant dependency for the C_{ref} on the wavelength (Backman et al., 2017; Bernardoni et al., 2021; Collaud Coen et al., 2010; Weingartner et al., 2003; WMO/GAW, 2016) and other studies observed the opposite and showed that the C_{ref} is wavelength dependent (Arnott et al., 2005; Kim et al., 2019; Schmid et al., 2006). These studies suggested that C_{ref} increased with wavelength (i.e. filter fibers scattered more light at longer wavelengths). Interestingly, even though the wavelength dependency was not statistically significant, Weingartner et al. (2003) reported that the C_{ref} obtained for internal mixtures of Diesel soot and ammonium sulfate and coated Palas soot yielded $C_{\text{ref}} = 3.9 \cdot (\lambda/660 \text{ nm})^{0.18}$ and $C_{\text{ref}} = 3.66 \cdot (\lambda/660 \text{ nm})^{0.23}$, respectively, as can be calculated from their Table 3. The exponents are very close to the value 0.18 obtained from the fittings to the Arnott et al. (2005) Table 1. Kim et al. (2019) found that C_{ref} depended on wavelength even more strongly, a fitting to their Table 2 yielded $C_{\text{ref}} = 4.48(\lambda/532 \text{ nm})^{0.48}$. Because the results between the different studies vary, it is difficult to conclude whether the C_{ref} is wavelength dependent or not. To study the wavelength dependency of C_{ref} , it would be ideal to use a photoacoustic or $\sigma_{\text{ext}} - \sigma_{\text{sca}}$ -methods as the reference measurements, since they are independent from the filter artefacts.

Newer model of the Aethalometer, AE33, applies the so-called dual-spot correction so the instrument operators do not need to apply the correction algorithms themselves. However, the value of C_{ref} is an open question for the AE33 also but since its filter material is different from the one used in the AE31, the results of the present study are not applicable to it. The filter material in AE33 is Teflon-coated glass filter tape (Pallflex type T60A20), but also the “old” filter tape (Q250F) has been used with AE33 and the recommended C_{ref} values to use with these filters are 1.57 and 2.14, respectively (Drinovec et al., 2015).

The different C_{ref} values were not only determined as a linear fit that considered the whole time series. In addition to the results from linear fits, the Table 2 presents the median, mean, and standard deviation of different C_{ref} values that were determined separately for each data point according to Eq. 19. Determining the C_{ref} separately for each data point enabled studying the temporal variation in C_{ref} ; for example, the times series of the different C_{ref} values are presented in Fig. S2. The median and mean values differed somewhat from the slopes of the linear fits, which were about 10% lower than the median values. Comparing the median and mean values shows no large difference, meaning that the C_{ref} values were rather normally distributed. The variation in median C_{ref} values between the different correction algorithms was small compared to the relatively large standard deviation (see Table 2).

The C_{ref} , determined separately for each data point, was not stable over time (see time series presented in Fig. S2) and we observed seasonal variation for the C_{ref} , which is presented in Fig. 2 for the C_{NC} as an example. The seasonal variation was not observed only for the C_{NC} , but also for the C_{WEI} and C_{COL} , which is presented in Fig. S3. Figure 2 shows that the C_{NC} was clearly above the median during the summer and below the median in winter and early spring. The C_{NC} reached its maxima in July and its minima in February. For C_{VIR} and C_{ARN} , the seasonal variation was much less pronounced (Fig. S3).

Since the C_{WEI} and C_{COL} had a similar seasonal variation, it is unlikely that the seasonal variation observed for C_{NC} was caused by the lack of filter loading correction. There was a seasonal variation for the C_{COL} as well and for example, the seasonal variations between the C_{WEI} and C_{COL} were rather similar, even though we applied constant f values in W2003. The algorithm by CC2010 considers the wavelength dependency of scattering and the ω of the accumulated particles. It is rather surprising that taking these parameters, which have a seasonal variation at SMEAR II (Luoma et al., 2019; Virkkula et al., 2011), did not seem to reduce the seasonality of C_{ref} .

The seasonal variations in the C_{ARN} and C_{VIR} were less obvious than in C_{WEI} , C_{COL} , and C_{NC} . The lesser seasonal variation for the C_{ARN} might be explained by the subtraction of the scattering fraction before the loading correction was applied and the C_{ARN} was determined. The fact that the C_{ARN} has less data points than the other the C_{ref} values, might also explain part of the lesser seasonality. For C_{VIR} , the lack of seasonal variation was probably caused by the very strong seasonal variation in the compensation parameter (k ; see Fig. 9a) as will be discussed below in Sect. 4.4. The algorithm by V2007 does not assume any coefficients but depends only on the difference between the last and first measurements of the filter spots. Therefore, it seems to adjust to seasonal changes whereas the other algorithms apply coefficients. According to our results, the V2007 and A2005 accounted well the variations in the optical properties of the particles embedded in the filter and therefore the seasonal variations in the C_{VIR} and C_{ARN} were reduced.

As indicated by the seasonal variation, the C_{ref} was not a constant value, but it depended on the optical properties of the particles embedded in the filter. As stated before, Weingartner et al. (2003) and Arnott et al. (2005) observed different C_{ref} values for different types of aerosols so that the C_{ref} was lower for “pure” soot (no coating) and higher for coated soot or ambient aerosol particles. This suggests that the C_{ref} increases with increasing ω . This supports our observations, since at SMEAR II, the ω is the highest in summer and lowest in winter (Luoma et al., 2019; Virkkula et al., 2011). However, Collaud Coen et al. (2010) observed a decreasing trend for C_{ref} with increasing ω when they compared the average conditions at several stations.

The ω , however, is not the only optical property of aerosol particles that had a clear seasonal variation (Luoma et al., 2019; Virkkula et al., 2011). For example, the size dependent b and α_{sca} reached their maxima in summer and minima in winter, which indicated that in summer the fraction of smaller particles increased. Luoma et al. (2019) showed that the seasonal variation in b and α_{sca} is explained especially by the differences in accumulation mode (particles in the size range of 100 nm –

1 μm) particle concentration and size distribution: in summer, the volume concentration peaks around 250 nm and in winter around 350 nm. The size distribution affects the penetration depth of the particles as smaller particles penetrate deeper in the filter (Moteki et al., 2010). Scattering particles that penetrate deeper in the filter increase the multiple scattering in the filter, and that could be one explanation for higher C_{ref} values observed in summer.

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The differences in the scattering properties of differently sized particles might also explain the observed seasonal variation in C_{ref} . The correction algorithms only consider the amount of scattering, but not the direction of scattering. Smaller particles scatter relatively more light in the backward direction, which increases the optical path of the light ray through the filter (i.e. C_{ref} should increase). Therefore, this effect may cause the observed increase in the multiple scattering correction factor C_{ref} in summer. This could also explain why the C_{VIR} had no seasonal dependency; the compensation parameter seemed to depend also on the b (see Sect. 4.4) and that would make the V2007 the only algorithm that takes the direction of the particulate scattering into account. Note that the V2007 does not take the b into account directly but it seems to influence the calculated compensation parameter (see Sect. 4.4).

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15 However, only very weak correlation was found between the C_{NC} and ω ($R = 0.17$, $p\text{-value} < 0.05$), and C_{NC} and b ($R = 0.23$, $p\text{-value} < 0.05$), so the ω and b do not necessary explain the observed seasonal variations in the C_{ref} values. For the C_{WEI} and C_{COL} , the results were similar, but for C_{VIR} , the R -values were even lower and even insignificant for ω .

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We observed slightly higher correlation ($R = 0.30$, $p\text{-value} < 0.05$) between the C_{NC} and relative humidity (RH), which is presented in Fig. 3 (the correlation was similar for C_{WEI} and C_{COL} , but weaker, about 0.09, for the C_{VIR}). Therefore, one possible reason for the observed seasonal variation in the different C_{ref} values could be caused by changes in the instrumental RH and the RH differences between the MAAP and AE31. The RH presented in Fig. 3 was measured in the MAAP and it varied between 5 – 40% since the periods when the filter of the MAAP was exposed for RH equal or larger than 40% were excluded from this study. Because the AE31 was equipped with Nafion-dryers, the RH in the AE31 varied less and was in the range of 5–20%. The RH can influence filter-based optical measurements by affecting to the optical properties of the aerosol particles and the filter fibers as well as by affecting the penetration depth of particles in the filter medium. The effect of rate of change in RH on the C_{ref} was also studied, but the rate of change in RH did not show any correlation between C_{NC} .

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Due to the hygroscopic growth, the aerosol particles scatter more light in humid conditions compared to dry conditions. The enhanced scattering induced by higher RH could then increase the scattering and optical path in a particle-laden filter medium. However, at SMEAR II, increasing RH should have caused a decrease in C_{NC} , since hygroscopic growth would have increased the particulate scattering especially in the reference instrument MAAP. The hygroscopic growth may also affect the penetration depth of the particles in the filter (Moteki et al., 2010). When particles are penetrated deeper in the filter, the effect of the multiple scattering is higher increasing the measured σ_{ATN} . Because the RH in the MAAP was higher than in the AE31, the

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particles directed in the AE31 may have penetrated relatively deeper in the filter than the particles directed in the MAAP filter, in summer, larger difference in the RH between the instruments could have increased the measured C_{ref} . However, the hygroscopic growth should not be significant in RH conditions below 40%, which is why the effects related to hygroscopic growth seem unlikely explanations.

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Also, the optical properties of the filter may change if the filter is exposed to high RH conditions. The aerosol particles may take up water even below super saturation and when the liquid particles collide on the filter the moisture is taken up by the filter. Kanaya et al. (2013) compared the MAAP against Continuous Soot Monitoring System (COSMOS; Miyzaki et al., 2008) and actually observed a slight dependency in the σ_{abs} measured by MAAP so that at low RH ($< 40\%$) the σ_{abs} increased with increasing RH , which is contrary to our results as we observed that MAAP observed relatively lower σ_{abs} at higher RH . However, they also observed opposite behavior at higher RH ($> 50\%$). They suggested that the RH affected the surface roughness of the filter, which is used in the radiative transfer scheme (Petzold and Schönlinner, 2004), and therefore could have affected the C_{ref} .

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The results showed that even though we excluded the high RH data, the instruments seemed to be sensitive to variations in RH even below the recommended 40%. However, the reason for the sensitivity remains unclear and would require more research and measurements and therefore further analysis is omitted from the scope of this article.

4.2 Performance of the correction algorithms

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In this section, we included data from June 2013 to February 2016 to have all the three absorption instruments running in parallel to prevent any differences caused by different periods.

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Since the σ_{abs} derived from AE31 measurements used the C_{ref} values determined here, the σ_{abs} measurements of AE31 and MAAP were expected to agree well, which is shown in Fig. 4. The AE31 data in Fig. 4 was produced by applying the C_{ref} values determined from the linear fits (Table 2 column “fit”). The correlation coefficients and slopes of the linear fits presented in Fig. 4 were close to unity. The AE31 correction schemes underestimated the σ_{abs} only slightly and the slopes varied from 0.96 to 1.00. The AE31 data corrected with the A2005 and CC2010 underestimated the σ_{abs} the most (slopes of the linear fits were 0.97 and 0.96, respectively). The reduction of particulate scattering in CC2010 after applying the multiple scattering correction (i.e., C_{ref}) could explain the slight underestimation in CC2010 derived data. For the underestimation in A2005 derived data, the reason is probably the different way of determining the C_{ARN} compared to other C_{ref} values. The iterative manner of determining the C_{ARN} separately for each filter spot and then taking the median from these values was not as successful as the linear fit -method, which was used for the other algorithms. However, the underestimation for A2005 and CC2010 are only minor.

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Surprisingly, the not-corrected (NC) AE31 data (Fig. 4e) did not seem to have a significant difference in correlation coefficient compared to, for example, the data corrected with W2003 or CC2010 (Figs. 4a and d, respectively). However, the relation between the $\sigma_{\text{abs,NC}}$ and σ_{ref} depended more on the *ATN* than for any filter loading corrected data, which is shown by the color coding (*ATN*) of the data points and in Table 3, which presents the slopes of the linear fits and R^2 values for different *ATN* intervals. If only data from highly loaded filter (*ATN* > 60 at 660 nm) were taken into account, the slopes of the linear fits were 0.97, 1.06, 0.99, 0.95, and 0.93 for W2003, A2005, V2007, CC2010, and NC, respectively. The smallest decrease in the slope with increasing *ATN* determined for the loaded filter was observed for data that was corrected by V2007. Interestingly, the slopes for the loaded filter increased for data that was corrected by A2005. This different behavior is probably caused by the fact that the A2005 algorithm did not consider the loading through *ATN* but applied a cumulative σ_{abs} , which apparently at SMEAR II seemed to overestimate the loading and loading correction, thus leading to an increasing slope with the *ATN*. The biggest decrease in the slope determined for a highly loaded filter was observed for the NC data, as expected.

According to the R^2 values presented in Table 3, the precision of the AE31 decreased with increasing *ATN*. For example, the data corrected with the A2005 algorithm, the R^2 decreased from 0.96 for a clean filter (*ATN* < 20) to 0.90 for loaded filter (*ATN* > 60). However, the decrease in R^2 was quite minor. Miyakawa et al. (2020) observed also rather high R^2 values between Aethalometer (model AE51) and a reference instrument (single particle soot photometer and COSMOS) when the *ATN* was below 70, but when the *ATN* exceeded 70, the R^2 decreased more rapidly. Unlike for the AE31, the loading on the filter did not seem to affect to the precision of the PSAP at all as the R^2 values did not decrease with increasing loading.

As presented in Table 3, the linear fits for the AE31 and PSAP data against the reference did not have an intercept of zero. This could be caused by the scattering artifact and the fact that the correction algorithms failed to take the scattering artefact partly into account. The intercept is the smallest for the B1999-corrected PSAP data and the largest to AE31 data. A fraction of σ_{sca} is subtracted in the AE31 algorithms by A2005 and CC2010. However, the data corrected with these algorithms still have a higher or similar intercept as with the not-corrected data and the data corrected by the W2003 and V2007 algorithms. Considering the intercept, the V2007-corrected data performs the best in the AE31 vs. MAAP comparison, which is slightly surprising, since it does not take the scattering subtraction into account. For the V2010-corrected PSAP data, the intercept is negative suggesting that the V2010 algorithm overestimates the apparent absorption by scattering particles.

The comparison between the MAAP and the PSAP is presented in Figs. 5a and b and in Table 3 for both the correction schemes B1999 and V2010, respectively. Figure 5b shows that V2010 overestimated the σ_{abs} especially when the loading was high (*Tr* was low) and the linear regression was 1.25. Also the B1999 overestimated the σ_{abs} slightly, but in general it performed better in comparison with MAAP and the slope was 1.07 (Fig. 5a). The linear fits in Figs. 5a and b include all the data, but Table 3 presents the slopes of the linear fits for data with different *Tr* limits. It is actually recommended to use PSAP data with *Tr* >

0.7 and if only this data is taken into account, especially the data corrected with the V2010 algorithm performs much better and has a slope of 1.01, but also the slope for the data derived with the B1999 algorithm yields a smaller slope of 1.04.

If all the data were included in the comparison, as in Figs. 5a and b, the overestimation of σ_{abs} would suggest deriving the C_{ref} values also for the PSAP data. Here, we did not derive the C_{ref} values for the PSAP since they are not typically used in a similar way as for deriving the σ_{abs} from the AE31 measurements. In general, the multiple scattering does not cause such a big artefact in filter material typically used in PSAP compared to the thicker AE31 filters. However, if we considered only the data below $Tr < 0.7$ the PSAP and MAAP agree well for both correction algorithms. This result then suggests that there is no need for deriving a new C_{ref} for PSAP. Svensson et al. (2019) studied the multiple scattering in quartz filters and they derived the equations that can be used in determining the C_{ref} value for PSAP. Differently to AE31 correction algorithms, the C_{ref} used in PSAP algorithms is included in the coefficients of Eqs. 13 – 15 and therefore determining the C_{ref} for PSAP is not as straightforward.

The differences between these two correction algorithms are studied in more detail in Fig. 5c, which shows how the algorithms perform with different Tr and ω values. As discussed before, the V2010 produces notably higher σ_{abs} values when the filter is highly loaded ($Tr < 0.5$). However, the difference between the algorithms does not only depend on the Tr but also on the ω so that at high ω and Tr , $\sigma_{\text{abs,PSAP,VIR}}/\sigma_{\text{abs,PSAP,BON}} < 1$ and when ω decreases the $\sigma_{\text{abs,PSAP,VIR}}/\sigma_{\text{abs,PSAP,BON}}$ -ratio grows. The reason is that the V2010 algorithm is a function of ω .

The dependency of the σ_{abs} on the ATN and Tr is presented in supplementary material (Figs. S4 and S5). On average, the decrease in the σ_{abs} , which was not corrected for the filter loading ($\sigma_{\text{abs,NC}}$ and $\sigma_{\text{abs,PSAP,ATN}}$), with the increasing ATN and decreasing Tr was not clear. This effect is better seen in the results presented in Table 3. However, Fig. S5 shows that especially for the PSAP, the use of correction algorithms decreased the variation, which is a strong recommendation for using the correction algorithms. This is also seen in the AE31 data, but the effect was less notable (Fig. S4).

Because it is impossible to separate the effect of different size cuts from a loaded filter, here the PM1 and PM10 measurements were combined and averaged together. In general, PM1 accounted for about 90% of the PM10 σ_{abs} ; for the σ_{sca} the fraction of PM1 was about 75% (Luoma et al., 2019). Because absorbing particles, which is considered to consist mostly of black carbon, are typically in the fine mode (diameter $< 1 \mu\text{m}$), the σ_{abs} is not expected to deviate much between the different size cuts. However, the differing size cuts, which causes more deviation in the σ_{sca} , could have affected the σ_{abs} measurements since the particulate scattering causes apparent absorption and affect the multiple scattering in the filter. For example, the coarse particles (diameter $> 1 \mu\text{m}$) do not penetrate as deep in the filter as the fine mode particles, which could possibly influence on the C_{ref} values. In an ideal situation the PM1 and PM10 absorption would have been measured by separate instruments.

Our observations underline the need for filter loading correction, especially if one studies shorter time periods. For longer time periods (e.g., trend analysis or studies of seasonal variation), the effect of the ATN on the variation smooths out, but for shorter time periods (e.g., case studies) the changing ATN can have a notable effect on the results if no filter loading correction is applied. However, not correcting for the filter loading effect, the precision of the instrument and the σ_{abs} or BC concentration on average are reduced, which is why applying a filter loading correction on filter-based photometers is always recommended.

4.3 Absorption Ångström exponent for different correction algorithms

The effect of the correction algorithms on α_{abs} were studied and the average α_{abs} for different correction algorithms of the AE31 and PSAP are presented in Fig. 6. This figure includes only parallel data from both AE31 and PSAP in order to avoid any differences caused by different time periods. For a comparison, the α_{abs} was also determined for the “raw” PSAP data that was not corrected by any algorithms (i.e., σ_{ATN} , see Eq. 1). To have comparable α_{abs} from the different instruments, Fig. 6 included only overlapping AE31 and PSAP data from 2011 – 2015. Since the PSAP operates at three wavelengths (467, 530, and 660 nm), we determined the AE31-related α_{abs} in Fig. 6 by using only the wavelengths 470, 520, 590, and 660 nm of the AE31. The rest of the AE31 wavelengths were omitted from this comparison to minimize the effect of different wavelength ranges have on α_{abs} (for example, see Luoma et al. (2019) Table 1). The α_{abs} was determined as a linear fit over all the selected wavelengths according to Eq. (16). Since Luoma et al. (2019) did not observe a big difference between the PM1 and PM10 α_{abs} , we included both measurements in this comparison.

According to Fig. 6, the median values of α_{abs} varied notably between the different instruments and correction algorithms: the lowest median value of α_{abs} was 0.93 and it was measured by AE31 and corrected by the CC2010; and the highest median value of α_{abs} was 1.54 and it was measured by PSAP and corrected by the V2010. The difference between the highest and lowest median values of α_{abs} was about 1.7-fold. The correction algorithms were applied to each wavelength separately and therefore the correction algorithms affected the wavelength dependency of the derived σ_{abs} . The scattering and loading corrections are different for each wavelength because for example the σ_{sca} , ω , ATN , and Tr , which are used in the algorithms, are wavelength dependent. For the AE31, we studied the same five correction algorithms as in Sect. 4.1. The lowest median α_{abs} values were observed for the not corrected data ($\alpha_{abs,AE,NC}$) and for data that were corrected with the CC2010 and W2003 algorithms ($\alpha_{abs,AE,COL}$ and $\alpha_{abs,AE,WEI}$). The median α_{abs} values for the data corrected with A2005 and 2007 algorithms ($\alpha_{abs,AE,ARN}$ and $\alpha_{abs,AE,VIR}$) were higher, 1.20 and 1.19, respectively.

The A2005 was the only algorithm that assumed a wavelength dependent C_{ref} . Since the C_{ARN} increased with wavelength (i.e., bigger correction due to multiple scattering at higher wavelengths), taking the wavelength dependency of the C_{ref} into account increases the $\alpha_{abs,AE,ARN}$ compared to other algorithms. The correction factor of the V2007 algorithm depended on the difference between the ATN of loaded and clean filter spots. Most of the time ATN increased faster at short wavelengths than at long wavelengths, so the difference between the ATN of the loaded and clean filter spots was higher than for longer wavelengths.

Therefore, the filter loading correction was bigger for shorter wavelengths and after the correction the difference between the σ_{abs} at different wavelengths increased, increasing the α_{abs} as well.

For the PSAP data, the α_{abs} were generally a bit higher compared to the AE31 derived α_{abs} . The lowest PSAP derived median value for $\alpha_{\text{abs,PSAP,NC}}$ was 1.01, which resulted from data that was not corrected by any algorithm. B1999 resulted for median $\alpha_{\text{abs,PSAP,BON}}$ value of 1.04, and the V2010 produced the overall the highest $\alpha_{\text{abs,PSAP,VIR}}$, which was 1.48. Similar order of the average α_{abs} from different algorithms was observed by Backman et al. (2014) at an urban station in Elandsfontein, South Africa. For a dataset measured off the east coast of the United States on a research ship, Backman et al. (2014) reported also the highest α_{abs} for the V2010. These results are consistent with each other. The explanation is that in V2010 all constants are wavelength dependent, contrary to the B1999.

The differences between the correction algorithms could possible be decreased by adding or reducing the wavelength dependency of the used constant values. Since we did not have reference measurements at several wavelengths, it is impossible to say which one of the correction algorithms yielded the most truthful value for the α_{abs} . This could be done with several MAAPs operating at different wavelengths, by measuring the particles suspended in the air by photoacoustic method (Kim et al., 2019), by a polar photometer (Bernardoni et al., 2021), or by a Multi-Wavelength Absorption Analyzer (MWAA; Massabò et al., 2013). According to the comparison between AE31 and MWAA by Satumö et al. (2017), the best agreement for the α_{abs} was achieved with uncorrected AE31 data and also the AE31 data corrected by CC2010 agreed well with the reference measurements.

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We also studied if the α_{abs} values were affected as the filter got more loaded with particles. Figure 7 presents the α_{abs} derived from AE31 data corrected with different algorithms as a function of ATN and Fig. 8 presents the α_{abs} derived from PSAP data as a function of Tr . The α_{abs} derived from corrected PSAP data (Fig. 8a and b) did not seem to depend on loading at $Tr > 0.4$ but for higher filter loadings the α_{abs} still increased with decreasing Tr with both B1999 and V2010 corrections. In comparison, for the not corrected PSAP data (i.e., σ_{ATN}), the α_{abs} decreased with increasing Tr (Fig. 8c). The α_{abs} derived from the AE31 data was also studied; the $\alpha_{\text{abs,AE,WEI}}$, $\alpha_{\text{abs,AE,ARN}}$, $\alpha_{\text{abs,AE,COL}}$, and $\alpha_{\text{abs,AE,NC}}$ clearly decreased with increasing ATN . If the ATN increased from 5 to 70, the decreases in $\alpha_{\text{abs,AE,WEI}}$, $\alpha_{\text{abs,AE,ARN}}$, $\alpha_{\text{abs,AE,COL}}$, and $\alpha_{\text{abs,AE,NC}}$ were rather linear and the decreases were around -22, -23, -33, and -27%, respectively.

30 The $\alpha_{\text{abs,AE,VIR}}$, derived from data corrected with V2007, did not seem to depend on the ATN , if not taking into account very high filter loadings (ATN at 660 nm > 70 ; on average the filter changed when the ATN at 660 nm ≈ 90). In V2007, the k was determined for each wavelength separately. The k is often larger for the shorter wavelengths, which means that the nonlinearity caused by the increased filter loading is relatively stronger at the shorter wavelengths (Drinovec et al., 2017; Virkkula et al.,

2007; Virkkula et al., 2015), which was also observed by this study (discussed in the next chapter). According to these results, the other algorithms but V2007 do not seem to account enough the wavelength dependency of the $R(ATN)$.

4.4 Variations in the compensation parameter

- 5 The variation in k at SMEAR II has already been studied by Virkkula et al. (2007) who used AE31 data from December 2004 to September 2006. During this period, the AE31 was operating without any cut-off and there were no scattering measurements available, and this period was not included in our study. Here, we repeated the analysis for a longer time series and included the σ_{sca} measurements, so we could also determine the b and ω .
- 10 The average values of the k are presented in Table 4. The mean values of the k varied from $4.6 \cdot 10^{-3}$ at 370 nm to $2.0 \cdot 10^{-3}$ at 950 nm. The wavelength dependency of k is described by a_k , which is the slope of a linear fit of the k over different wavelengths ($k_\lambda = a_k \lambda + k_0$, see example in Fig. 9b). Negative a_k means that on average the filter loading correction was greater at shorter wavelengths. The light attenuation is stronger at shorter wavelengths due to higher absorption and scattering by the particles and therefore the shorter wavelengths are prone to bigger error caused by the filter loading. At longer wavelengths, the standard
- 15 deviation of the k was higher, meaning that the k was more sensitive to the particle properties at longer wavelengths. The same observation was noted by Virkkula et al. (2015) as well.

At SMEAR II, we observed that the k and the a_k had a very strong seasonal variation so that the k and a_k were the lowest in summer, which was also noted by Virkkula et al. (2007). The seasonal variation was observed at all wavelengths, but the

20 variation was more pronounced at longer wavelengths. The seasonally averaged k and a_k are presented in Table 4 and an example of the seasonal variation in k at 880 nm is presented in Fig. 9a. Similar seasonal pattern for the k were also observed by Virkkula et al. (2007), Wang et al. (2011), and Song et al. (2013). In summer, the mean k values at the longer wavelengths (660–950 nm) were negative, meaning that without the correction, the AE31 would actually overestimate the σ_{abs} at longer wavelengths.

- 25 Previous studies (e.g., Virkkula et al., 2007; Wang et al., 2011; and Song et al., 2013) suggested that the seasonal variation in k could be due to variations in ω , so that lower ω induced higher k . This behaviour is observed at SMEAR II as shown in Figs. 9a and d; the ω peaks in summer as the k has its minima and the correlation coefficient between the ω and k is -0.47. The variation in ω also explains the observed negative k values. Virkkula et al. (2007) stated that the negative values are associated
- 30 with the response of the Aethalometer to scattering aerosols as the negative k are observed when the ω is high. The effect of ω was taken into account for example in the AE31 correction algorithms suggested by Weingartner et al. (2003), Arnott et al. (2007), and Collaud Coen et al. (2010). Virkkula et al. (2015) presented a theoretical explanation to the ω dependency of k , which our analysis supports.

Also, the effect caused by the sizes of the particles has been suggested. The sizes of the particles affects their scattering properties and also on their penetration depth in the filter that again could affect the k . The size distribution of the particle population is described by the b so that higher b indicates smaller particles. Müller et al. (2014), for example, showed that the effect of asymmetry parameter, which is a function of b (Andrews et al., 2006), had an effect on the PSAP data.

The dependency of k on both b and ω was investigated more closely by Virkkula et al. (2015) at SORPES, an urban station located in Nanjing, China. The study showed positive correlation between k and b , and negative correlation between k and ω . At SMEAR II, we also observed negative correlation between k and ω (Fig. 9c). However, contrary to the results by Virkkula et al. (2015), we observed negative correlation between k and b (Fig. 9d). Virkkula et al. (2015) discussed difficulties to show whether the b or the ω was the dominant property in determining the k . At SMEAR II, the ω varies in a wider range compared to the observations at SORPES, which could explain some of the observed differences. The mean and standard deviation of ω at SMEAR II were 0.87 ± 0.07 (at 550 nm; Luoma et al., 2019) and at SORPES 0.93 ± 0.03 (at 520 nm; Shen et al., 2018). However, a clear reason for the negative correlation between the k and b at SMEAR II was not found.

5 Summary and conclusions

In this study, we presented a comparison of three different absorption photometers (AE31, PSAP, and MAAP), which measured ambient air at SMEAR II, a rural station located in middle of a boreal forest in southern Finland. We also compared different correction algorithms that are used in determining the absorption coefficient (σ_{abs}) from the raw absorption photometer data. We studied how the algorithms affected the derived parameters and determined multiple scattering correction factor (C_{ref}) applicable at SMEAR II.

To get more reliable AE31 measurements, the AE31 data were compared against the MAAP data to acquire the C_{ref} that is used in the processing of the AE31 data. Previous studies observed that the C_{ref} varied between different types of environments and stations and here it was determined for the SMEAR II station, which represents the atmospheric conditions in a boreal forest. The resulted C_{ref} values were 3.00, 3.13, 3.14, and 2.99 for the algorithms suggested by Weingartner et al. (2003), Amott et al. (2005), Virkkula et al. (2007), and Collaud Coen et al. (2010), respectively. The C_{ref} determined at the SMEAR II can be applied to other boreal forest sites as well and even though the AE31 is an older model and no longer in production, the results can be used in post-processing older data sets or at sites that still operate the older AE31.

We also observed a clear seasonal cycle associated with C_{ref} , which was probably due to the variations in the optical properties of the aerosol particles, such as the b and ω . We also observed some correlation between the C_{ref} and RH even though the RH

in the instruments were kept below 40%. These results show that the filter measurement methods seem to be rather sensitive to the RH even if the RH is below the recommended value of 40%.

The results obtained for data corrected with the algorithm by Virkkula et al. (2007) were in many ways different from those obtained by Collaud Coen et al. (2010) who applied the Virkkula et al. (2007) correction to data from several stations in Europe. They found that the compensation parameter (k) used in the algorithm was highly nonstable and that it led to large outliers. They correctly stated that the difficulty of applying this correction is due to the natural high variability of σ_{ATN} as a function of time, which is for most of the time greater than the σ_{ATN} decrease induced by filter changes. We therefore calculated 14-day running average compensation parameters (± 7 days around each filter spot) in order to minimize these problems. The approach was obviously successful. It can be recommended that users of this method calculate running averages of k . The suitable period for the running average at each site depends on the rate of change in the ATN , which determines how often the filter spots are changed. According to this study and to the study by Virkkula et al. (2015) time period that includes about 6 – 9 filter spot changes on average seems to yield good results. At SMEAR II, a relatively clean site, this period was 14 days and at SOPRES, a rather polluted site, the period was 24 hours.

The results showed a great variation between the α_{abs} derived from differently corrected σ_{abs} data and at SMEAR II the median α_{abs} for different algorithms varied in the range of 0.93 – 1.54. We also observed that most of the correction methods did not prevent the change in the wavelength dependency as the filter got more loaded and therefore the α_{abs} decreased notably with increasing attenuation (ATN). The correction algorithm by Virkkula et al., (2007) was the only AE31 correction algorithm, which produced a stable α_{abs} for the increasing filter loading. For example, the α_{abs} derived from Aethalometer measurements is often used to describe the chemical properties of the particles and to describe the source of black carbon. Not taking the used correction algorithm and the effect of increasing filter loading into account, could lead to wrong interpretation of the results. According to our results, applying the Virkkula et al. (2007) correction algorithm could help solving if the changes in α_{abs} were due to real variation or due to increased filter loading.

In general, at SMEAR II, the effect of the filter loading on average did not seem to cause major difference in the measured σ_{abs} . However, a strong effect of increased filter loading was seen in the derived parameter α_{abs} , which encourages to apply a filter loading correction to filter-based absorption data. Even though on average, the σ_{abs} did not seem to be greatly affected by the filter attenuation (ATN), the filter loading effect can have a great effect when studying shorter periods and, for example, different seasons, which also justifies applying a correction to the data. According to our study the correction algorithms by Virkkula et al. (2007) and Arnott et al., (2005) performed the best in taking the seasonal variations of the aerosol particles into account. Also, the algorithm by Virkkula et al. (2007) produced the most stable α_{abs} that did not depend on the ATN , which was not the case for the other algorithms.

When applying a correction algorithm to AE31 data, it is important to report which algorithm, C_{ref} values and other coefficients were used to acquire the final data product, since the algorithms can have a notable effect on the results, especially on the α_{abs} . Our results showed that in general, it is a good practice to perform the analysis of AE31 data by using few different correction algorithms, to see if the results vary notably for different algorithms.

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Author contributions

KL performed the data analysis and wrote the paper together with AV. PA and AV have set up the aerosol optical measurements at SMEAR II. All authors reviewed and commented on the paper.

10 Acknowledgements

We thank SMEAR II staff for taking care that all the measurements were running.

Financial support

This research has been supported by the European Union's Horizon 2020 research and innovation program via projects
15 ACTRIS-2 (grant no. 654109) and iCUPE (grant no. 689443). Additional financial support was received through the Academy of Finland (Center of Excellence in Atmospheric Sciences) under the projects PROFI-3 (decision no. 311932), NANOBIOMASS (decision no. 307537). This research was also supported by Academy of Finland via project NABCEA (grant no. 29664) and by Business Finland via project BC Footprint (grant no. 49402-201040). Financial support of University of Helsinki to ACTRIS-FI is gratefully acknowledged.

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Table 1. All the wavelength dependent coefficients used in the AE31 correction algorithms proposed by Weingartner et al. (2003) and Arnott et al. (2005). The extrapolated values of the multiple scattering correction factor used in the Arnott et al. (2005) correction algorithm (C_{ARN}) at different wavelengths.

Coefficients for AE31 correction algorithm by Weingartner et al. (2003)							
λ (nm)	370	470	520	590	660	880	950
a	0.878	0.868	0.863	0.857	0.850	0.829	0.822
ω	0.91	0.89	0.89	0.88	0.85	0.83	0.82
f	1.079	1.096	1.095	1.103	1.128	1.141	1.148
Coefficients for AE31 correction algorithm by Arnott et al. (2005)							
λ (nm)	370	470	520	590	660	880	950
$100 \cdot a_{s,ARN}$	3.35	4.57	5.23	6.16	7.13	10.38	11.48
$\tau_{a,fx}$	0.3026	0.2527	0.2338	0.2129	0.1956	0.1575	0.1486
C_{ARN}	2.70	2.82	2.87	2.94	3.00	3.16	3.20

5 **Table 2. Average values for the multiple scattering correction factor (C_{ref}) for the different correction algorithms. The values are reported at 637 nm. The slope of the fit, standard error of the fit (SE), and 95 % confidence interval (CI) were determined by a linear regression applied for the whole data set. The median, mean, standard deviation (SD), as well as the 5th and 95th percentile range were determined from the C_{ref} values that were calculated for each data point separately.**

	fit	SE	95 % CI	median	mean	SD	5 th & 95 th percentiles
C_{WEI}	3.00	0.003	[2.99; 3.00]	3.34	3.29	0.57	[2.59; 4.26]
C_{ARN}				3.13	3.13	0.45	[2.49; 3.81]
C_{VIR}	3.14	0.002	[3.13; 3.14]	3.30	3.28	0.56	[2.53; 4.18]
C_{COL}	2.99	0.003	[2.98; 2.99]	3.28	3.32	0.57	[2.55; 4.23]
C_{NC}	2.77	0.003	[2.76; 2.77]	3.09	3.06	0.55	[2.32; 3.95]

10 **Table 3. Linear fits between the AE31 vs. reference absorption ($\sigma_{abs,ref}$ measured by MAAP) for different ATN intervals (at 660 nm) as well as between the PSAP vs. $\sigma_{abs,ref}$. The value in parenthesis is the coefficient of determination (R^2).**

ATN	0 - 20	20 - 40	40 - 60	60 - 80
W2003	$1.05 \cdot x + 0.07$	$0.99 \cdot x + 0.15$	$0.95 \cdot x + 0.15$	$0.97 \cdot x + 0.12$
	(0.98)	(0.97)	(0.96)	(0.94)
A2005	$0.93 \cdot x + 0.06$	$0.93 \cdot x + 0.16$	$0.97 \cdot x + 0.15$	$1.06 \cdot x + 0.10$
	(0.96)	(0.95)	(0.92)	(0.90)

V2007	$1.02 \cdot x + 0.05$ (0.98)	$0.98 \cdot x + 0.10$ (0.98)	$0.97 \cdot x + 0.08$ (0.97)	$0.99 \cdot x + 0.05$ (0.96)
CC2010	$1.01 \cdot x + 0.06$ (0.98)	$0.95 \cdot x + 0.15$ (0.97)	$0.92 \cdot x + 0.16$ (0.95)	$0.95 \cdot x + 0.13$ (0.92)
Not corrected	$1.12 \cdot x + 0.07$ (0.98)	$1.01 \cdot x + 0.15$ (0.97)	$0.93 \cdot x + 0.14$ (0.96)	$0.93 \cdot x + 0.12$ (0.95)
<i>Tr</i>	1 – 0.7	1 – 0.4	0.4 – 0.7	0 – 0.4
B1999	$1.04 \cdot x + 0.01$ (0.97)	$1.06 \cdot x + 0.02$ (0.97)	$1.06 \cdot x + 0.07$ (0.97)	$1.11 \cdot x + 0.02$ (0.98)
V2010	$1.01 \cdot x - 0.02$ (0.96)	$1.12 \cdot x - 0.07$ (0.94)	$1.17 \cdot x + 0.01$ (0.95)	$1.46 \cdot x - 0.20$ (0.96)

Table 4. The mean compensation parameters (k) and the wavelength dependency of k (a_k) for the AE31 correction algorithm suggested by Virkkula et al. (2007). The average values are calculated over all the seasons, but also separately for each season. The seasons were classified as: spring (March – May), summer (June – August), autumn (September – November), and winter (December – February).

Season	370 nm ($\cdot 10^{-3}$)	470 nm ($\cdot 10^{-3}$)	520 nm ($\cdot 10^{-3}$)	590 nm ($\cdot 10^{-3}$)	660 nm ($\cdot 10^{-3}$)	880 nm ($\cdot 10^{-3}$)	950 nm ($\cdot 10^{-3}$)	a_k ($\cdot 10^{-6} \text{ nm}^{-1}$)
all	4.6 ± 7.0	3.6 ± 7.2	3.5 ± 8.0	3.4 ± 8.9	2.7 ± 9.5	2.1 ± 10.6	2.0 ± 10.8	-4.2
spring	4.4 ± 7.0	3.5 ± 6.5	3.4 ± 7.6	3.4 ± 8.8	2.8 ± 9.2	2.2 ± 10.5	2.2 ± 10.2	-3.5
summer	3.0 ± 6.7	1.5 ± 6.3	1.1 ± 6.9	0.5 ± 9.0	-0.4 ± 9.6	-1.7 ± 10.3	-2.5 ± 11.3	-8.8
autumn	4.6 ± 7.7	3.6 ± 9.1	3.6 ± 9.7	3.5 ± 10.3	2.8 ± 10.2	2.5 ± 10.9	2.2 ± 12.1	-3.6
winter	5.5 ± 6.3	4.8 ± 6.4	4.8 ± 7.0	4.8 ± 6.9	4.3 ± 8.4	4.4 ± 9.9	4.3 ± 9.1	-1.7

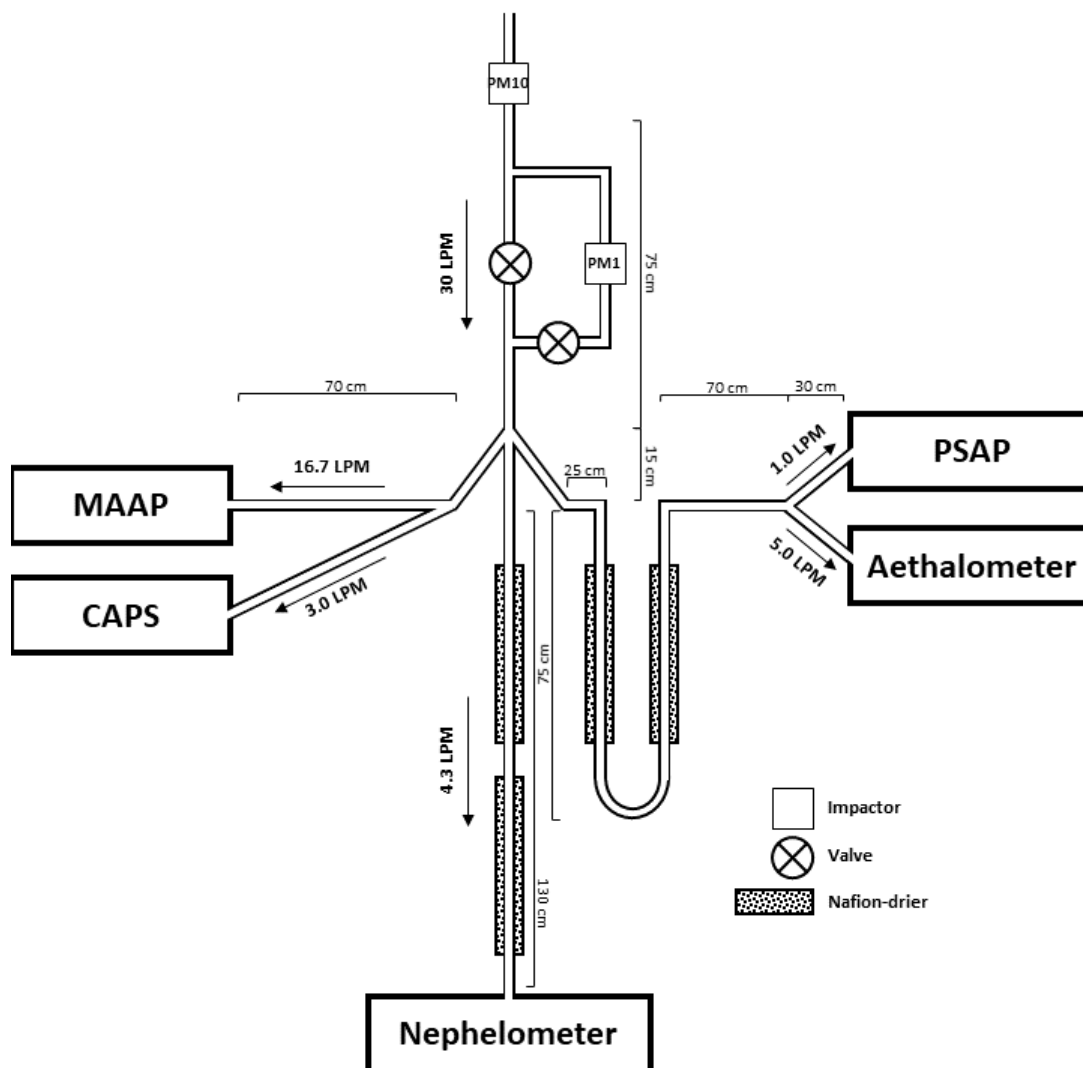


Figure 1: Measurement scheme for the instruments that measured the aerosol optical properties at the SMEAR II station. This setup was running during 2014–2015, when all the instruments were operating in parallel.

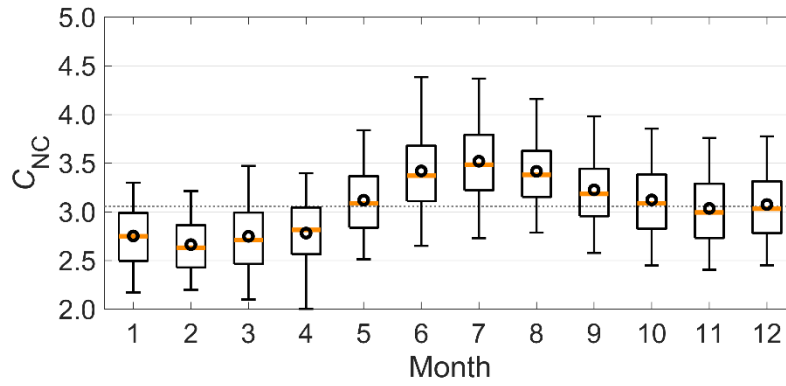


Figure 2. The seasonal variation in the multiple scattering correction factor for not corrected data (C_{NC}). The orange line in the middle of the box is the median, the black circle is the mean, the edges of the boxes represent the 25th and 75th percentiles, and the whiskers represent the 10th and 90th percentiles of the data. The dashed line is the median for all data

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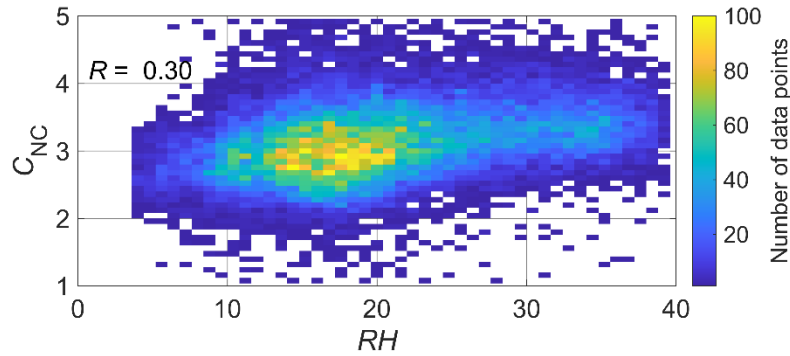


Figure 3. The dependency of the multiple scattering correction factor for not corrected data (C_{NC}) on the instrumental relative humidity (RH) in the MAAP. The colored grid points represent the number of data points in each grid point. There is 50 grid points in x- and y-directions so in total there are 2500 grid points.

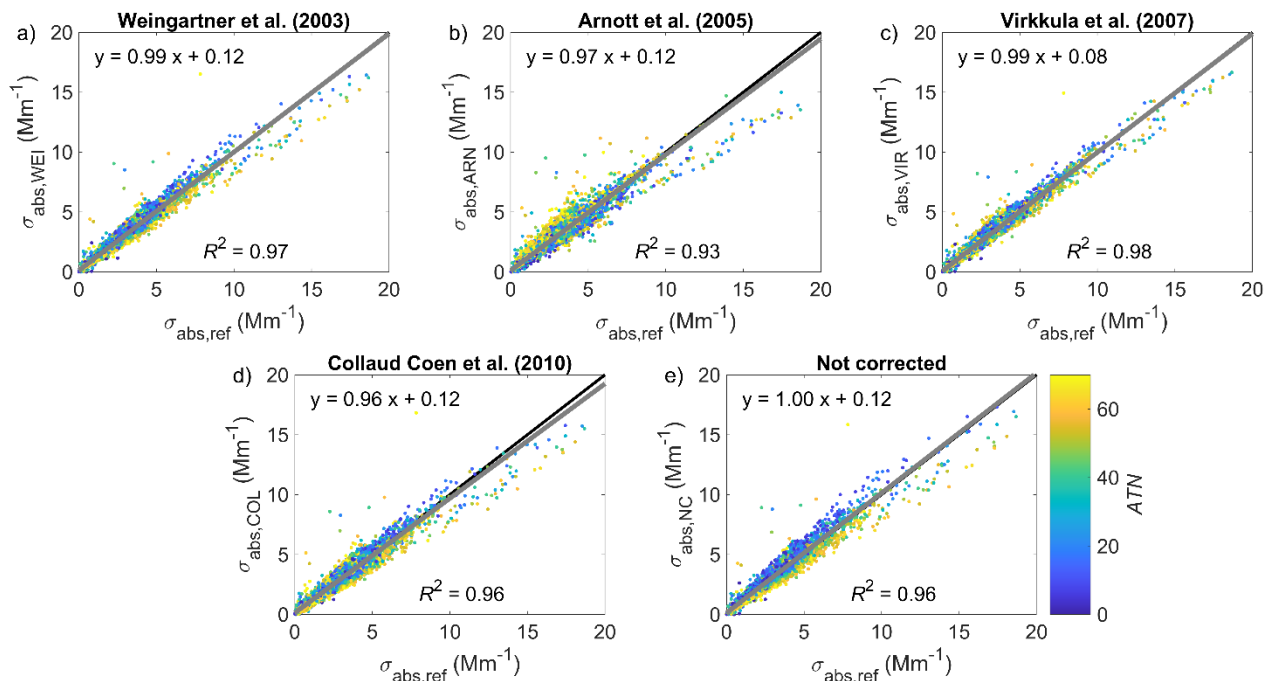


Figure 4. Comparison of the AE31 and MAAP measurements for all the different AE31 correction algorithms. The corrected AE31 data has been interpolated to the same wavelength with MAAP (637 nm). The data points are colored by the AE31 filter attenuation (ATN; at 660 nm). The fit to the data is presented with a grey line and the equation and the coefficient of determination (R^2) are shown in the subfigures. One-to-one line is shown with black color.

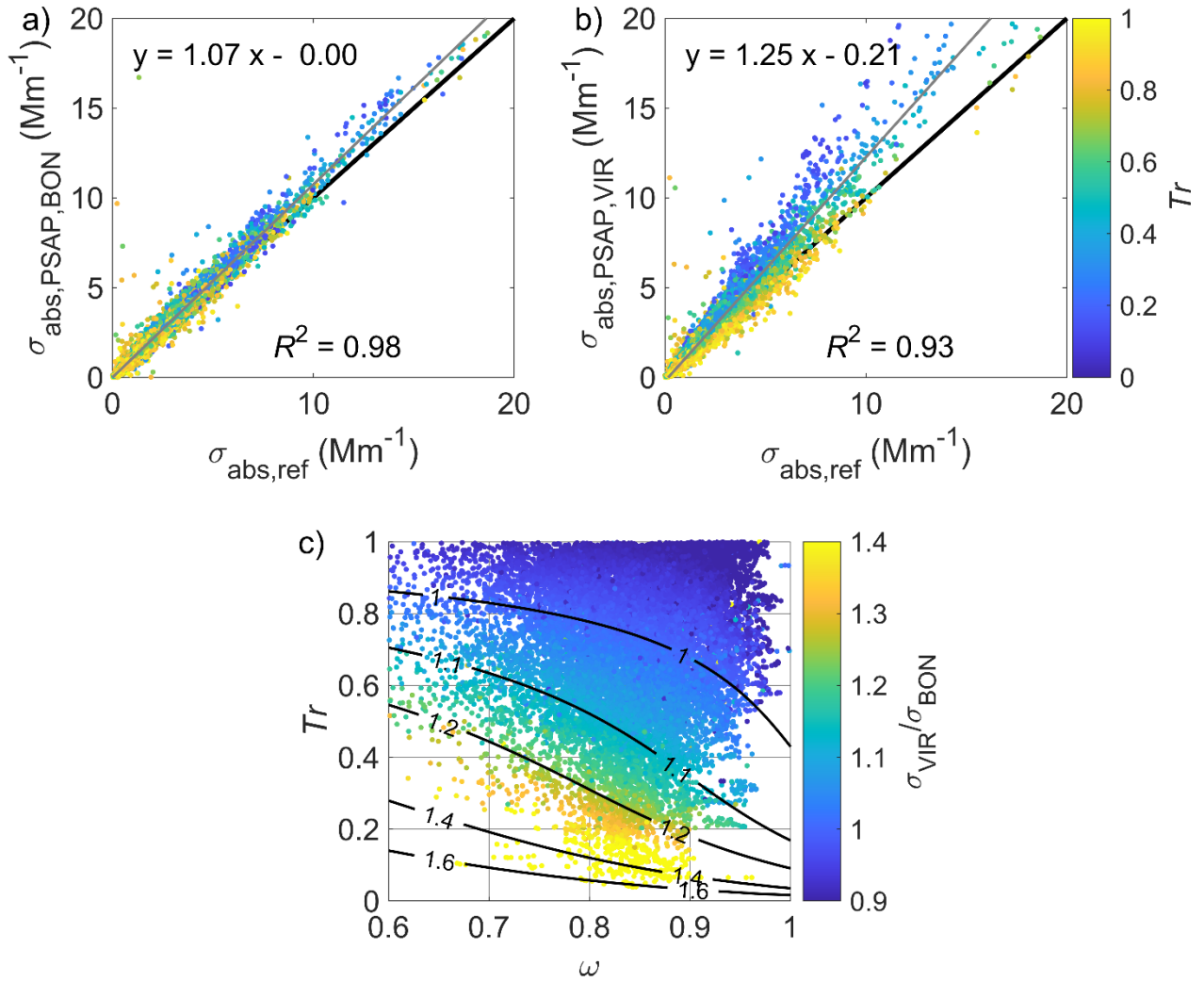


Figure 5. The a) and b) subfigures present the comparison of the PSAP and MAAP measurements for both B1999 and V2010 correction algorithms, respectively. The data points are colored by the PSAP filter transmittance (Tr), the fit to the data is presented with a grey line, and the equation and the coefficient of determination (R^2) are shown in the subfigures. One-to-one line is shown with black color. The subfigure c) presents the relation of the PSAP derived absorption coefficients corrected with the V2010 algorithm ($\sigma_{\text{abs,PSAP,VIR}}$) and the B1999 algorithm depends on the Tr and single scattering albedo (ω). The contour lines show the theoretically determined $\sigma_{\text{abs,PSAP,VIR}}/\sigma_{\text{abs,PSAP,BON}}$ ratio. The ω was determined from nephelometer and MAAP measurements at 637 nm.

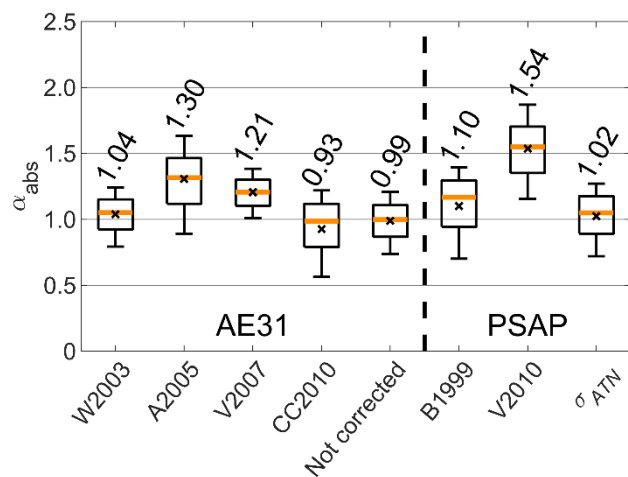


Figure 6. The absorption Ångström exponent (α_{abs}) for all the different AE31 and PSAP correction algorithms. The orange line in the middle of the box is the median, the black circle is the mean, the edges of the boxes represent the 25th and 75th percentiles, and the whiskers represent the 10th and 90th percentiles of the data. The values given above each box show the according median values.

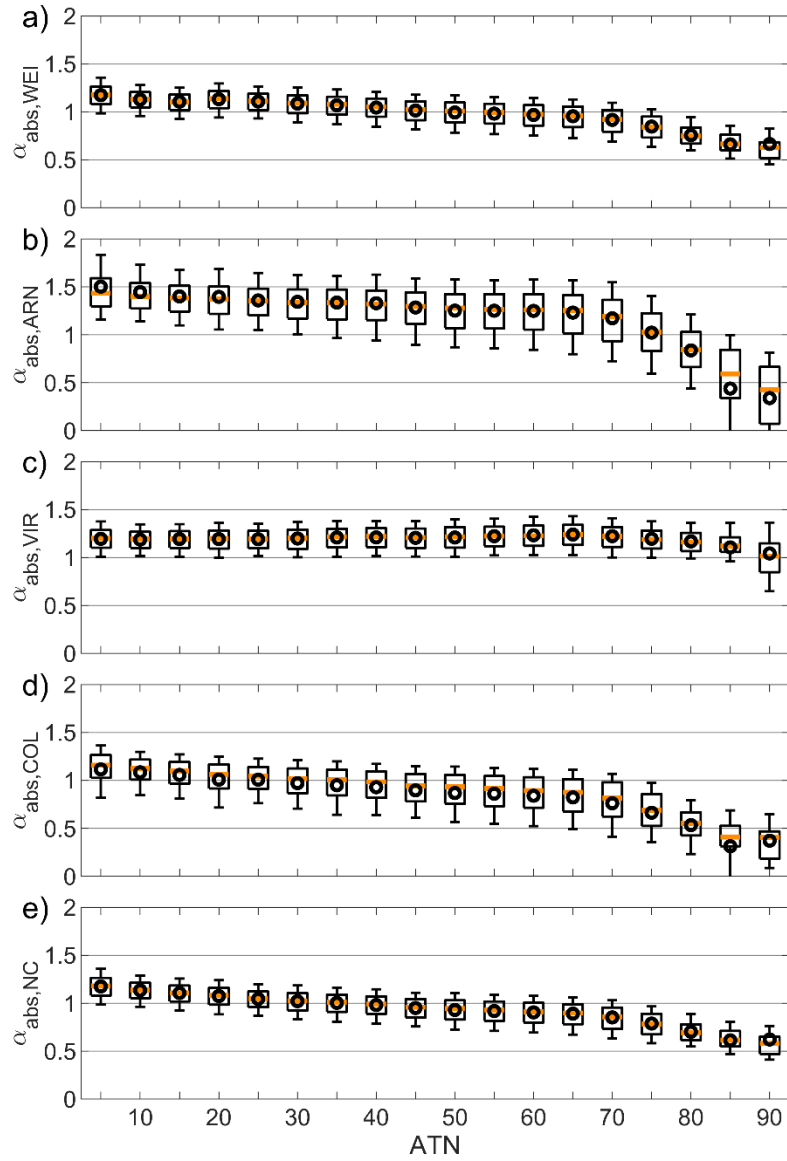


Figure 7. The dependency of the absorption Ångström exponent (α_{abs}) on the AE31 filter attenuation (ATN; at 660 nm) for different correction algorithms. The orange line in the middle of the box is the monthly median, the black circle is the mean, the edges of the boxes represent the 25th and 75th percentiles, and the whiskers represent the 10th and 90th percentiles of the data.

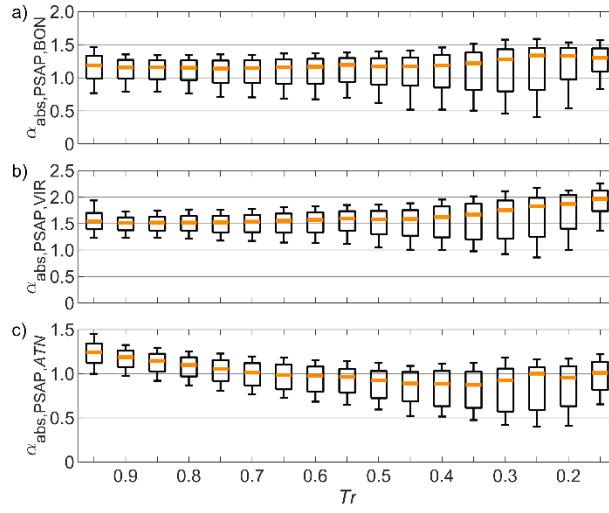


Figure 8. The dependency of the absorption Ångström exponent (α_{abs}) on the PSAP filter transmittance (Tr) for the a) B1999, b) V2010, and c) not corrected σ_{ATN} . The explanation for the boxplots is the same as in Fig. 5.

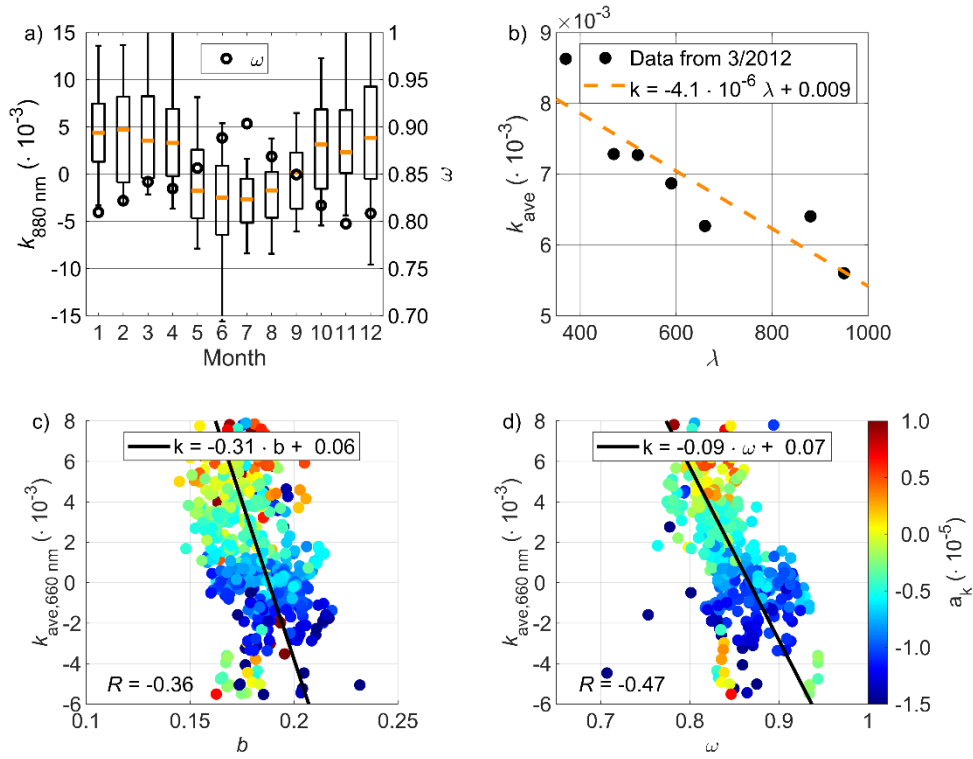


Figure 9. a) The seasonal variation in the compensation parameter (k). b) An example of calculating the wavelength dependency of the k (a_k). c) The dependency of the k on the backscatter fraction (b). The data points are coloured by a_k . d) The dependency of the k on the single scattering albedo (ω). The data points are coloured by a_k .