Atmos. Chem. Phys. Discuss., 11, 11939–11957, 2011
www.atmos-chem-phys-discuss.net/11/11939/2011/
doi:10.5194/acpd-11-11939-2011

Atmospheric Chemistry and Physics Discussions

Seasonal differences in the vertical profiles of aerosol optical properties over rural Oklahoma

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Received: 30 March 2011 – Accepted: 4 April 2011 – Published: 18 April 2011

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Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

A small airplane made more than 450 aerosol optical property (light absorption and light scattering) vertical profile measurements (up to 4 km) over a rural Oklahoma site between March 2000 and July 2005. These profiles suggest significant seasonal differences in aerosol properties. The highest amounts of scattering and absorbing aerosol are observed during the summer, while the relative contribution of aerosol absorption is highest in the winter (i.e., single scattering albedo is lowest in winter). Aerosol absorption generally decreased with altitude below ~1.5 km and then was relatively constant above that. Aerosol scattering decreased sharply with altitude below ~1.5 km but, unlike absorption, also decreased at higher altitudes, albeit less sharply. The seasonal variability observed for aerosol loading is consistent with other aerosol measurements in the region including AERONET aerosol optical depth (AOD), CALIPSO vertical profiles, and IMPROVE aerosol mass. The column averaged single scattering albedo derived from in situ airplane measurements shows a similar seasonal cycle as the AERONET single scattering albedo inversion product, but a comparison of aerosol asymmetry parameter from airplane and AERONET platforms suggests differences in seasonal variability. The observed seasonal cycle of aerosol loading corresponds with changes in air mass back trajectories: the aerosol scattering was higher when transport was from polluted areas (e.g., the Gulf Coast) and lower when the air came from cleaner regions and/or the upper atmosphere.

1 Introduction

Understanding the temporal and spatial variability of aerosol optical properties is important for relating aerosols to their sources, quantifying the effects of transport and transformation on the aerosol and understanding the contribution of aerosol to such wide-ranging concerns as health, visibility and climate. The vertical variability of aerosol particles in the atmosphere is an aspect of aerosol spatial distribution which has been

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less well studied than the horizontal variability, in large part because it is more difficult to sample aloft than from a ground-based platform. Haywood and Ramaswamy (1998) observe that the magnitude and sign of the aerosol forcing effect are partly determined by the vertical distribution of the aerosol. More recently it has been shown that errors in the assumptions about the shape of aerosol profiles can cause errors in aerosol optical thickness retrieved from satellites (Rozwadowska, 2007). Of particular interest for climate change is the question of the vertical distribution of absorbing aerosol in the atmosphere (e.g., Haywood and Ramaswamy, 1998; Ramanathan et al., 2001). Ramanathan et al. (2001) showed that the vertical profile of absorbing aerosol could influence cloud formation and lifetime. Haywood and Ramaswamy (1998) showed that the atmospheric aerosol profiles could change calculated direct radiative forcing. Recent modeling studies utilizing a small set (55) of vertical black carbon (BC) aerosol profiles from multiple field campaigns at various locations have met with limited success (e.g., Koch et al., 2009; Vignati et al., 2010) due perhaps to coarse temporal resolution of the comparisons (2 to 10 measured profiles were compared with the modeled monthly means for the month the profiles were obtained) or to an oversimplified algorithm for describing transport and transformation of the black carbon. More recent model/measurement comparisons (Skeie et al., 2011) continue to suggest our ability to measured and/or model absorbing aerosol in the atmosphere needs improvement. These model/measurement comparison papers focused on absorbing aerosol and did not discuss how measured in situ aerosol scattering profiles compared with model results.

Lidar provides an alternative to in situ vertical profiling and can provide insight into the vertical loading and variability of aerosol particles; however, deriving profiles of aerosol absorption or other properties (e.g., single scattering albedo, asymmetry parameter) useful for radiative forcing calculations from lidar measurements is still in its infancy (e.g., Veselovskii et al., 2005; Müller et al., 2001). Measurements from ground-based sun-sky radiometers can be inverted to obtain aerosol size distributions, single scattering albedo, and asymmetry parameter (Dubovik and King, 2000) but these

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inversions are representative of the atmospheric column rather than any specific location aloft and are typically limited to clear sky conditions.

While in situ aerosol aircraft profiling has often been used for short field campaigns in the last three decades (e.g., Husar et al., 1977; Kim et al., 1988; Li et al., 1997; Osborne and Haywood, 2005; references in Koch et al., 2009), it can also be a relatively inexpensive method to measure aerosol properties in the vertical over the long term using a dedicated light aircraft. University of Maryland made aerosol and gas phase profiling measurements in a small airplane at various locations on the east coast of the US between 1997 and 2003, with a focus on pollution events and summertime air quality (e.g, Taubman et al., 2006; Hains et al., 2008). Similarly, NOAA and DOE/ARM developed an aerosol profiling platform for long term measurements of climatically important aerosol properties (e.g., Andrews et al., 2004). The DOE airplane measured climatically relevant aerosol property profiles over a site in rural Oklahoma between 2000 and 2007 and the first six years of these measurements are utilized in this paper.

Here we address the following scientific questions using measurements made by DOE's In situ Aerosol Profiles (IAP) airplane:

- Are there seasonal differences in vertical profiles of aerosol properties?
- Are the seasonal trends consistent with other aerosol measurements?
- Do these differences correspond with source region/transport?

2 Experimental approach

Profiling flight measurements were conducted over the Department of Energy's Southern Great Plains (SGP) site near Lamont, Oklahoma (36.6° N, 97.5° W, 315 m a.s.l.). The site is in north central Oklahoma, making it a location relatively remote from large sources of anthropogenic pollution. The three nearest large cities (Wichita, KS, Tulsa,

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OK, and Oklahoma City, OK) are all more than 100 km away. The airborne aerosol optical measurements reported here began March 2000 and observations from the first 2 yr of airborne measurements have been described previously (Andrews et al., 2004). Here, with a longer data set covering 6 yr and 458 profile flights, the seasonal variability of the aerosol optical property profiles is explored.

2.1 In situ aerosol measurements

The aerosol system on the IAP aircraft has been described in detail elsewhere (Andrews et al., 2004) so only a brief description is given here. The airplane, a Cessna 172XP, was originally instrumented with an integrating nephelometer (TSI-3563, three-wavelength (450, 550 and 700 nm), total and hemispheric-backscatter capabilities) and a filter-based light absorption instrument (Radiance Research PSAP, ($\lambda = 565$ nm adjusted to 550 nm). A heater upstream of the instrumentation ensured that measurements were made at low relative humidity (RH) conditions (RH<40%); the aerosol inlet ensured that only sub-micrometer aerosol were measured. This instrumentation package forms the basis of the observations reported in this paper. There have been several upgrades to the instrument package since the start of the program but because of a change in inlet cut size in August 2005, this paper only covers data from March 2000 through July 2005. While the data after the inlet change are not discussed here, the profiles from the new inlet are qualitatively consistent with the profile plots discussed below.

The airplane flew 2–3 profiles per week during daylight hours. The profile pattern consisted of a 9 level stair-step descent from ~3700 m a.s.l. down to ~460 m a.s.l. (flight levels at 3657, 3048, 2438, 1828, 1524, 1219, 914, 610 and 457 m). The plane spent approximately 10 min at the four highest levels and then 5 min at each of the five lower levels. Table 1 lists the monthly and seasonal breakdown of the flight data between March 2000 and July 2005. The absorption measurements were adjusted for instrument artifacts (e.g., filter spot size and flow) and interference by scattering aerosol

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deposited on the filter following the method described in Bond et al. (1999). The nephelometer light scattering measurements were corrected for instrument non-idealities (e.g., truncation angle) based on work by Anderson and Ogren (1998).

From the measured aerosol absorption (σ_{ap}), scattering (σ_{sp}) and backscattering (σ_{bsp}) coefficients, several climatically important aerosol optical parameters can be calculated. These include: single scattering albedo $\omega_o = \sigma_{sp}/(\sigma_{sp} + \sigma_{ap})$ and asymmetry parameter, estimated with the empirical relationship $g = -7.1439b^3 + 7.4644b^2 - 3.9636b + 0.9893$, where b is the ratio of hemispheric backscattering to total backscattering (σ_{bsp}/σ_{sp}) (Andrews et al., 2006). Additionally, for qualitative comparison with model output and chemical measurements, equivalent black carbon (EBC) concentrations were derived from light absorption measurements made by the PSAP by assuming that all light absorption was caused by black carbon with a mass absorption coefficient of $7.5\,m^2\,g^{-1}$ at 550 nm wavelength, as recommended by Bond and Bergstrom (2006) for fresh black carbon. We call this EBC because if the aerosol absorption was caused by other species such as organic carbon or dust, or if aging influenced the optical properties of the absorbing aerosol then the value of $7.5\,m^2\,g^{-1}$ would be inappropriate (Bond and Bergstrom, 2006).

2.2 AERONET measurements

AERONET sunphotometer measurements are also made at the SGP site. Because we are focused on seasonal properties rather than comparing on a flight-by-flight basis, the "Level 2.0" aerosol optical depth (AOD) and almucantar inversion products for the years 1994 thru 2008 were downloaded from the AERONET site (http://aeronet.gsfc.nasa.gov). The almucantar inversion products include column-average ω_o and g (Dubovik and King, 2000). The aerosol properties obtained from AERONET differ in wavelength, altitude range covered, humidity conditions and particle size cut from those observed using the in situ instruments on the aircraft. Nonetheless, by adjusting the in situ measurements to ambient conditions (i.e., using estimates of

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hygroscopic growth, supermicrometer scattering and absorption fractions from the SGP surface site (as described in Andrews et al., 2004)), accounting for wavelength differences using an Ångström correction for σ_{sp} and assuming a $1/\lambda$ dependence for σ_{ap}, and averaging over the column we can quantitatively compare seasonal medians of column properties derived from the in situ measurements with those for the AERONET data products.

3 Results and discussion

There are strong seasonal differences in the profiles of median sub-micron aerosol optical properties measured over the SGP site (Fig. 1). Figure 1a, b depicts aerosol absorption and scattering at low RH (<40%) and standard temperature and pressure (STP, $T_{standard} = 0\,^{\circ}C$, $P_{standard} = 1013.25\,hPa$). More aerosol (as indicated by absorption (and EBC) and scattering) is observed during summer than for any other season, while the least amount of aerosol is observed in the winter. Near the surface, summertime scattering and absorption are approximately a factor of two higher than the scattering and absorption observed in winter while at higher altitudes summertime scattering may be a factor of 10 larger than that observed in the winter and summertime absorption may be 3–4 times larger than the wintertime absorption. Median EBC ranges from a maximum of approximately $250\,ng\,m^{-3}$ in the summer at low altitudes (<1 km) to $100\,ng\,m^{-3}$ over the same altitude range in winter. At higher altitudes (>1.5 km) EBC is less than $100\,ng\,m^{-3}$ for all seasons except summer and is relatively invariant with altitude. Additionally, the shapes of the aerosol profiles shown in Fig. 1a, b suggests that increased amounts of aerosol extend higher in the atmosphere in the summer than at other times of year, reflecting changes in the boundary layer height and/or enhanced vertical mixing in the summer.

The Mann-Whitney U-test (Fay and Proschan, 2010) suggests that the differences observed between summer and winter scattering and summer and winter absorption profiles are significant to greater than the 0.99 confidence level. Spring, fall and annual

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profiles of scattering and absorption are not statistically different from each other at the 0.99 confidence level. These profile plots show that at the highest altitudes very little aerosol is measured. This can lead to outliers in the calculation of derived aerosol properties such as ω_o and g. The seasonal variability (high in summer/low in winter) and general shape of the light scattering profiles is consistent with the eastern United States seasonal average extinction profiles from CALIPSO reported by Yu et al. (2010). The CALIPSO profiles are ambient nighttime extinction measurements made between June 2006–November 2007 over a very large region (~4 million km^2) so a closer comparison with the daytime, sub-micron, dry, STP aircraft scattering profiles obtained over a single site in Oklahoma between March 2000 and July 2005 is inappropriate. A more detailed statistical comparison of seasonal profiles for CALIPSO and these airplane measurements is in progress.

Vertical profiles of ω_o (low RH<40%) also vary by season (Fig. 1c), particularly above 1000 m a.s.l. in the winter. Above 1000 m, the median wintertime ω_o is lower (more absorbing) than at any other time of year and decreases substantially (by more than 10%) between 1000 m a.s.l. and 3700 m a.s.l. In contrast, the summertime ω_o is consistently higher (i.e., the aerosol is less absorbing) and is also fairly invariant with altitude. As with the scattering and absorption profiles, the spring and fall ω_o profiles lie in between the winter and summer profiles and are similar to the annual median ω_o profile. All seasons show ω_o values in the lower column (<1.2 km) that agree well with the long term surface value at SGP of 0.94–0.95 (Sheridan et al., 2001; Delene and Ogren, 2002). The Mann-Whitney U test suggests summer and winter ω_o values are different at the 0.99 confidence level above 1.2 km.

Asymmetry parameter vertical profiles (low RH<40%) also show statistically significant seasonal differences (Fig. 1d). Summer and winter are statistically different from each other at the 99% level below 1.8 km, while fall is statistically different (lower) than the annual profile below 2.0 km. Smaller particles (lower g) are observed during the fall and winter than during the summer (except at or above 3000 m during the winter). The increase in g observed above 3000 m a.s.l. during the winter is likely due to noise

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associated with using b, the ratio of two small numbers, to calculate g. Springtime asymmetry profiles are representative of the median annual profiles.

The observed seasonal variations in AOD derived from the in situ measurements adjusted to ambient conditions and 673 nm are consistent with seasonality of AERONET AOD (see Fig. 2a), although the AOD derived from in situ measurements tends to be lower than the AERONET AOD as has been frequently observed in direct comparisons (e.g., Schmid et al., 2009 and references within). This is attributed to limitations in adjusting the in situ measurements to ambient conditions (in particular, the humidity adjustment and accounting for aerosol not sampled by the in situ measurements). The observed seasonality in AOD is also consistent with other measures of aerosol loading in the region, including aerosol extinction profiles obtained from Raman lidar described in Turner et al. (2001), CALIPSO profiles for the eastern US (Yu et al., 2010) and surface measurements of aerosol optical properties at SGP (Delene and Ogren, 2002). IMPROVE network measurements of aerosol mass in the central US also typically show the most mass during the summer (Malm et al., 2004) primarily due to increases in the sulfate aerosol mass. Malm et al. (2004) suggest that the amount of organic aerosol, another potentially large scatterer, is fairly invariant throughout the year and region.

Figure 2b shows a comparison of column average ω_o for the in situ and AERONET measurements. Unfortunately the ω_o retrievals from AERONET are severely limited by the constraint that aerosol optical depth must be greater than 0.4 at 440 nm such that out of a possible 2870 valid level 2.0 AERONET almucantur inversions between 1994 and 2008, only 115 ω_o values were retrieved and, of those, only three were for the winter season. There are several things to notice from this figure: (1) the AERONET and in situ column average ω_o show similar seasonal variation, with ω_o being higher in spring and (for the limited number of points) winter, (2) the median AERONET ω_o values tend to be quite a bit higher (~0.05) than the values derived from the in situ profiles (perhaps related to differences in AOD shown in Fig. 2a) and (3) the seasonal variation of the ambient column average ω_o has a slightly different pattern than that

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suggested by the dry profiles ω_o shown in Fig. 1c. This difference in seasonal patterns is driven in part by the adjustment to 673 nm. The inverse wavelength correction for in situ absorption data is constant over the year, while the in situ scattering adjustment to 673 nm utilizes scattering Ångström exponents which vary seasonally (not shown).

A comparison of the column average asymmetry parameters (Fig. 2c) shows that the values calculated from in situ measurements are lower than those provided in the AERONET inversion products, consistent with the in situ measurements not being correctly adjusted for the presence of larger aerosol particles. The in situ values are more variable than observed for AERONET and suggest the asymmetry parameter is lower in the fall than for the other seasons (0.56 ± 0.1). In contrast, the g derived from AERONET measurements show less seasonal variation (0.64 ± 0.05) than the in situ values.

In all of these AERONET/in situ comparisons the differences are likely due to a combination of any number of issues including differences in altitude range covered, differences in size cut, ambient relative humidity adjustment and/or the assumptions inherent in the AERONET inversion algorithm. An in-depth analysis of these differences will be pursued in a separate paper utilizing both measurements discussed here and those from a similarly instrumented airplane which flew profiles over an AERONET site in Illinois.

The NOAA HYSPLIT model (www.arl.noaa.gov/ready/hysplit4.html; Draxler and Hess, 1998, 1997) was run in batch mode using NCEP reanalysis data to generate two-day back trajectories for every day of the year between 2000 and 2006 for air arriving over SGP at 500, 1500 and 3000 m a.s.l. These calculated back trajectories were clustered on the basis of latitude, longitude and altitude for each season. Trajectories were grouped using k-means clustering. This type of clustering minimizes variability within a cluster and maximizes the variability between clusters. There were significant differences in source regions for the different seasons and altitudes (Fig. 3) and these differences appear to correspond with the seasonal differences observed for aerosol optical properties.

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Approximately 90% of the 500 m summertime trajectories arriving originated from the south with the medians of several of the clusters passing near the Houston and the Dallas-Fort Worth conurbations prior to arriving at SGP. These trajectories tend to be confined within the boundary layer (Turner et al., 2001 noted the summer time boundary layer height over SGP is ~1500 m a.s.l.) and would presumably contain polluted air from the various sources over which they passed. In contrast, 70% of the wintertime trajectories arriving at 500 m a.s.l. came from the northwest, passing over relatively unpopulated regions and descending from altitudes above the wintertime boundary layer (~600 m a.s.l. (Turner et al., 2001)) on their way to the site. Spring and fall trajectories (not shown) were similar to each other and tended to be evenly split between northwesterly higher altitude trajectories (i.e., winter-like) and southeasterly low altitude trajectories (i.e., summer-like). Back trajectories arriving at 1500 m (not shown) were similar to those at 500 m. These back trajectory patterns are consistent with the amount (i.e., σ_{ap} and σ_{sp}) of aerosol observed.

At 3000 m the back trajectories reflect the prevailing westerly winds. Trajectories come from NW, W, and SW for all four seasons. The clusters indicate (a) most of the air masses start at 2000 m or above and (b) they tend to travel over relatively unpolluted/unpopulated regions of the western US. This is consistent with the low amounts of aerosol (as indicated by low scattering coefficients) observed at the higher flight levels. Sources of aerosol arriving at the 3000 m level in the in situ profiles are difficult to determine. The back trajectory clusters suggest that many of these air masses descend from high altitudes and may have been aloft a long time. The long time aloft provides another explanation for the low aerosol amounts observed at higher altitudes as various removal mechanisms will have opportunity to affect the aerosol properties. A specific example is cloud scavenging which, followed by precipitation, removes aerosol from the atmosphere. Cloud scavenging may also explain the slight decrease in single scattering albedo observed with altitude as scattering aerosol (which tends to be more hygroscopic) would be preferentially removed by cloud scavenging.

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4 Conclusions

Temporal analysis of 6 yr of flight data (458 profiles) show that profiles of aerosol optical properties differ as a function of season over the DOE CART site in central Oklahoma. The profiles differ both in amount of aerosol and, to a lesser extent, shape as a function of season. Aerosol absorption decreases sharply from the surface to approximately 1.5 km and then is approximately constant above that altitude for all seasons but summer. Summertime aerosol absorption continues to decrease above 1.5 km. In contrast, aerosol scattering decreases with increasing altitude for all seasons but the decrease is sharpest below 1.5 km. The observed seasonal differences are consistent with independent measurements of aerosol loading made by other platforms in the region (e.g., AERONET, CALIPSO, IMPROVE) and single scattering albedo (AERONET). However, the AERONET asymmetry parameter inversion product does not show the same seasonal cycle or as much seasonal variability as the column averaged value calculated from the in situ measurements. Much of the seasonal variability observed in aerosol optical properties is likely due to differences in source regions of the air masses over the site. Summertime observations of high levels of aerosol scattering coincide with air being transported within the boundary layer from populous and polluted regions in Texas (Houston and Dallas), while the cleaner conditions (lower scattering) observed in the wintertime correlate with northwesterly air mass back trajectories originating at higher altitudes. This multi-year collection of aerosol vertical profiles should be useful for validation of chemical transport models.

Acknowledgements. This research was funded by DOE-ARM. Greenwood Aviation in Ponca City operated the plane. We thank the Brent Holben and Rick Wagener for their effort in establishing and maintaining the CART Site AERONET data discussed in this paper.

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Table 1. Temporal statistics for 458 profile flights between March 2000 and July 2005*.

Season	# flights/season	# flights	month
Spring	135	41	March
		31	April
		63	May
Summer	111	23	June
		42	July
		46	August
Fall	105	38	September
		32	October
		35	November
Winter	107	35	December
		37	January
		35	February

* Only flights with all 9 level legs are included in this table.

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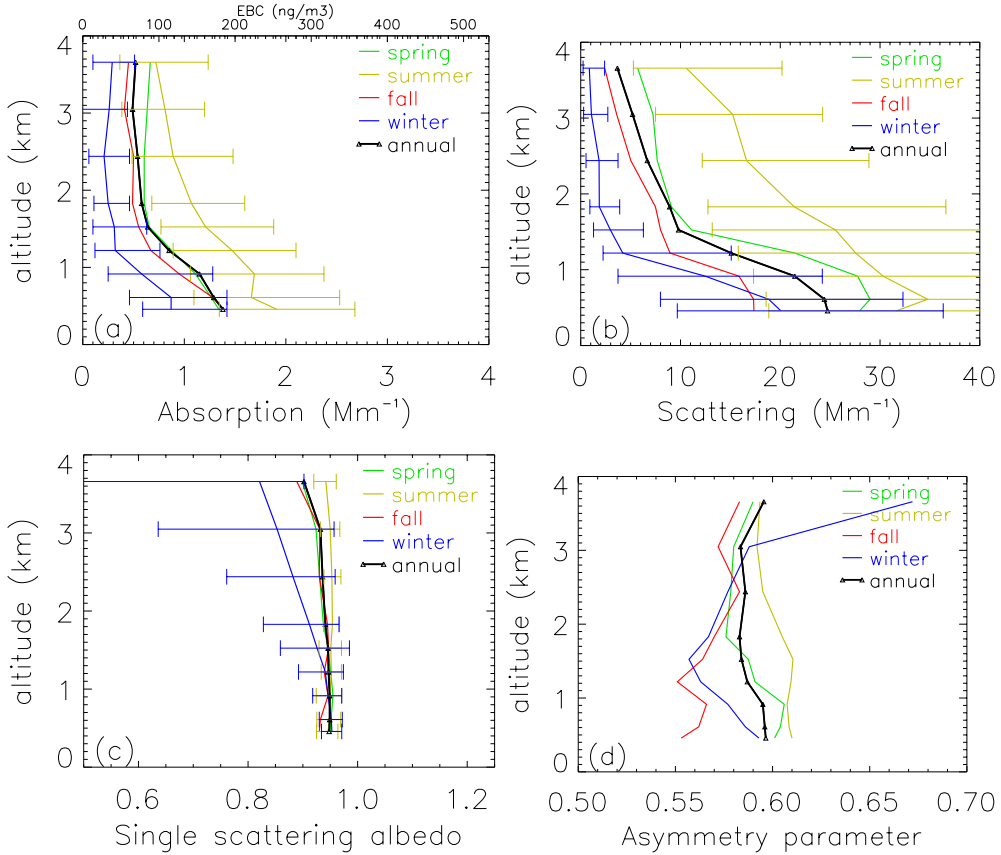

Fig. 1. Seasonal profiles of median aerosol optical properties ($Dp < 1\mu m$; RH<40%, wavelength = 550 nm) **(a)** light absorption and EBC (corrected to STP); **(b)** light scattering (corrected to STP); **(c)** single scattering albedo; and **(d)** asymmetry parameter. Horizontal lines indicate 25th and 75th percentiles for winter (blue) and summer (gold). Plots only include flights with all 9 flight levels.

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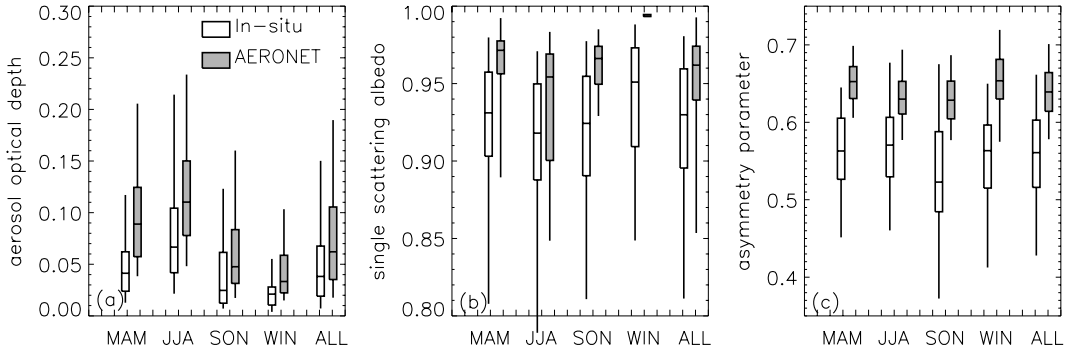

Fig. 2. Comparison of in situ aerosol optical properties adjusted to ambient T, P, RH and wavelength = 673 nm with AERONET values at 673 nm **(a)** aerosol optical depth, **(b)** single scattering albedo, **(c)** asymmetry parameter.

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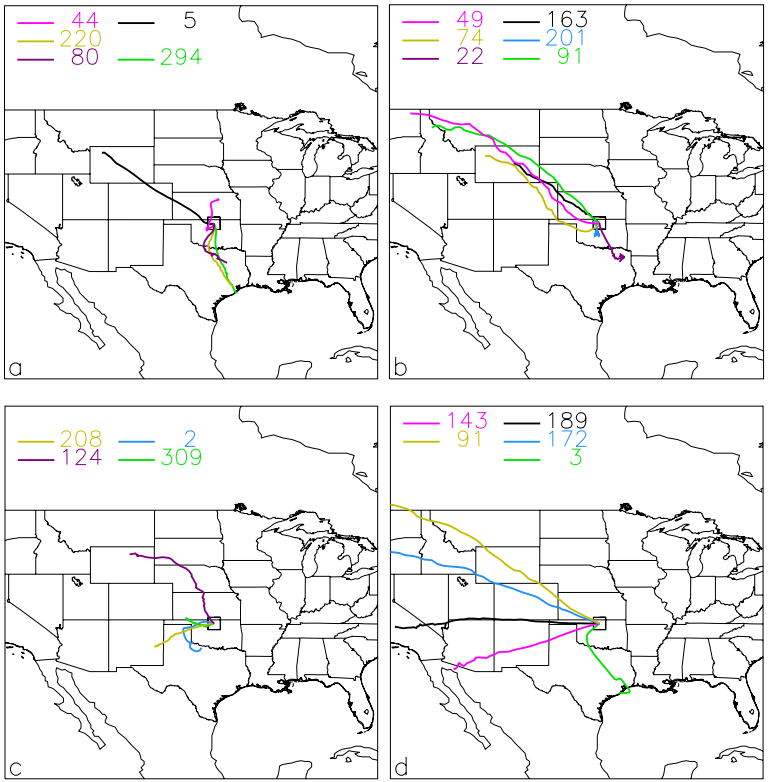

Fig. 3. Plots of air mass back trajectory clusters to the profile site (indicated by the square) calculated using NOAA/HYSPLIT_4[1]; **(a)** Summer, 500 m, **(b)** Winter, 500 m, **(c)** Summer, 3000 m, **(d)** Winter, 3000 m. Numbers indicate how many trajectories are in each cluster. A maximum of 6 clusters was allowed[2].

[1] Trajectories were generated using HYSPLIT_4 downloaded from: www.arl.noaa.gov/ready/hysplit4.html.

[2] Trajectories that look similar in the x-y plane (e.g., most of the 500 m winter trajectories) were clustered based on their vertical motion.

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