Aerosol and cloud interaction observed from high spectral resolution lidar data

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Recent studies utilizing satellite retrievals have shown a strong correlation between aerosol optical depth (AOD) and cloud cover. However, these retrievals from passive sensors are subject to many limitations, including cloud adjacency (or three-dimensional) effects, possible cloud contamination, uncertainty in the AOD retrieval. Some of these limitations do not exist in High Spectral Resolution Lidar (HSRL) observations; for instance, HSRL observations are not affected by cloud adjacency effects, are less prone to cloud contamination, and offer accurate aerosol property measurements (backscatter coefficient, extinction coefficient, lidar ratio, backscatter Angstrom exponent, and aerosol optical depth) at a fine spatial resolution (~100 m) in the vicinity of clouds. Hence the HSRL provides an important data set for studying aerosol and cloud interaction. In this study, we statistically analyze aircraft-based HSRL profiles according to their distance from the nearest cloud, assuring that all profile comparisons are subject to the same large-scale meteorological conditions. Our results indicate that AODs from HSRL are about 8–17% higher in the proximity of clouds (~100 m) than far away from clouds (4.5 km), which is much smaller than the reported cloud three-dimensional effect on AOD retrievals. The backscatter and extinction coefficients also systematically increase in the vicinity of clouds, which can be explained by aerosol swelling in the high relative humidity (RH) environment and/or aerosol growth through in-cloud processing (albeit not conclusively). On the other hand, we do not observe a systematic trend in lidar ratio; we hypothesize that this is caused by the opposite effects of aerosol swelling and aerosol in-cloud processing on the lidar ratio. Finally, the observed backscatter Angstrom exponent (BAE) does not show a consistent trend because of the complicated relationship between BAE and RH. We demonstrate that BAE should not be used as a surrogate for Angstrom exponent, especially at high RH.


1. Introduction

Several satellite studies indicate that regions of the atmosphere with increased cloud cover are accompanied by increased aerosol optical depths (AODs) in the clear areas between the clouds, and that this phenomenon is occurring globally [Sekiguchi et al., 2003; Ignatov et al., 2005; Loeb and Manalo-Smith, 2005; Kaufman et al., 2005; Matheson et al., 2005, 2006; Loeb and Schuster, 2008]. Possible explanations for this observed trend include cloud contamination in the aerosol retrieval, aerosol swelling in the high-humidity cloudy environments [Clarke et al., 2002], increased illumination of the cloud-free columns by the nearby clouds (i.e., the cloud adjacency or three-dimensional effect [Podgorny, 2003; Wen et al., 2006, 2007]), increased particle production in the vicinity of clouds [Hegg et al., 1990; Hoppel et al., 1994; Clarke et al., 1998] and shift of aerosol size distribution to larger size because of cloud processing and cloud droplet evaporation in the vicinity of clouds [Lelieveld and Heintzenberg, 1992; Alkezewy, 1995; Lu et al., 2003; Hegg et al., 2004].

Wen et al. [2006, 2007] studied the three-dimensional cloud radiative effects on MODIS aerosol retrievals over Brazil. Their Monte Carlo simulations indicate that ignoring the cloud adjacency effect can cause an overestimation of 50–140% in AOD retrieval, with the overestimation more pronounced at shorter wavelengths. Large increase of AOD at shorter wavelengths is also shown in stochastic cloud model simulations [Marshak et al., 2008], who argue that the enhancement in radiance of the cloud-free column comes from enhanced Rayleigh scattering.
[4] In this study, we examine the aerosol properties both in the vicinity of clouds and far away from clouds using nadir-viewing aircraft-based High Spectral Resolution Lidar (HSRL) profiles of aerosol backscatter and extinction coefficients. The HSRL has many advantages over passive satellite instruments for observing aerosol and cloud interactions, including the following: (1) it provides accurate measurements of aerosol properties in the vicinity of clouds as opposed to the aerosol retrievals provided by satellites; (2) it is an active instrument with a narrow source, so it is not affected by cloud adjacency effects; (3) the backscatter coefficient of cloud droplets is much larger than that of aerosols, so cloud boundaries are readily distinguishable; and (4) the HSRL has a spatial resolution of <100 m, which makes the clear profiles less prone to cloud contamination than the much larger clear pixels (≥500 m) associated with satellite instruments. However, the disadvantage of HSRL measurement is that it has limited spatial coverage and can only provide cloud top height along the flight track.

2. Method

[5] A standard backscatter lidar measures attenuated backscatter; retrieving extinction profiles from a backscatter lidar requires the assumption of extinction-to-backscatter ratio (i.e., lidar ratio). Unfortunately, the actual value of lidar ratio for tropospheric aerosols can vary over a wide range (from 20 to 100 sr) depending on their optical properties [Ansmann et al., 1990; Ackermann, 1998; Anderson et al., 2000; Ferrare et al., 2001; Cattrall et al., 2005], which leads to large uncertainty in the retrieved aerosol extinction. Unlike a standard backscatter lidar, the HSRL provides measurements of both aerosol extinction and backscatter, which enables the robust computation of the lidar ratio.

[6] In this section, first we describe the NASA Langley Research Center (LaRC) airborne HSRL, then we describe our statistical analysis of HSRL data. We compare each HSRL clear profile to a clear profile next to clouds (i.e., reference profile). Comparisons are limited to clear profiles that are within 5 km from the reference profile, which ensures that both the clear profiles and the reference profile are subject to the same large-scale meteorological conditions. For a given flight, there are multiple reference profiles; aerosol properties of all clear profiles and differences between clear profiles and respective reference profiles are binned by clear profiles’ distances to the nearest clouds. For each bin, average differences are presented and possible causes for the differences are analyzed.

### 2.1. Description of LaRC HSRL

[7] The LaRC HSRL instrument and its measurement technique is described in Hair et al. [2008], which we briefly review here. The basic concept of HSRL measurements is to obtain the lidar return signal with high spectral resolution (~75 MHz laser bandwidth), which enables the separation of aerosol and cloud returns from molecular returns. This separation is possible because the spectrum of the molecular backscatter is Doppler broadened by the thermal motion of the molecules, whereas Doppler broadening of the aerosol and cloud backscatter is negligible because of the much slower thermal velocities of the aerosol/cloud particles. The separation of molecular from aerosol/cloud backscatter enables the independent retrieval of aerosol/cloud backscatter and extinction profiles. Another key feature of the LaRC HSRL is the ability to calibrate the instrument internally, thereby eliminating systematic errors associated with vicarious calibration in regions that are assumed to have negligible aerosol loading.

[8] The LaRC HSRL includes three measurement channels at the 532 nm wavelength and two measurement channels at the 1064 nm wavelength; parallel and perpendicular scattering channels are included at both wavelengths, and an additional channel for molecular scattering is included at the 532 nm wavelength. The molecular channel makes it possible to derive reliable aerosol backscatter (β) and extinction (σ) profiles at 532 nm, and hence the aerosol lidar ratio: \( S_\lambda = σ/β \). A lidar ratio is assumed to derive extinction profiles for the 1064 nm wavelength (although a more sophisticated algorithm incorporating the 532 nm lidar ratio could be implemented in the future).

[9] Two additional parameters can be derived from the HSRL measurements. The aerosol depolarization ratio can be calculated from the perpendicular and parallel backscatter at both wavelengths: \( δ = \frac{β^p - β^\parallel}{β^p} \). We also define the backscatter Angstrom exponent (BAE, analogous to the Angstrom exponent):

\[
BAE = -\frac{\ln(β_{1064}/β_{532})}{\ln(2)},
\]

where the denominator represents the logarithm of the wavelength ratio. The absolute uncertainties of these HSRL observed variables used in this study are listed in Table 1.

#### 2.2. Statistical Analysis Method

[10] The LaRC HSRL was deployed on the LaRC’s King Air B-200 aircraft during the CATZ/CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) AERONET (Aerosol Robotic Network) Twilight Zone campaign over the Eastern United States (26 June to 29 August 2007). One of the objectives of this campaign was to investigate the nature of particles in the “twilight zone” between clouds and aerosols [Koren et al., 2007]. The aircraft flew at an altitude of 9 km above sea level, and measurements were averaged over 100 shots (0.5 sec) in the lidar electronics before being transferred to a laptop computer. Within 0.5 sec, HSRL usually covers a spatial resolution of 50–75 m, depending upon the flight speed of the aircraft (which varies somewhat during each flight).

<table>
<thead>
<tr>
<th>Variable</th>
<th>Uncertainty</th>
</tr>
</thead>
<tbody>
<tr>
<td>β (Mm⁻¹ sr⁻¹)</td>
<td>0.36</td>
</tr>
<tr>
<td>σ (km⁻¹)</td>
<td>0.055</td>
</tr>
<tr>
<td>( S_\lambda ) (sr⁻¹)</td>
<td>14</td>
</tr>
<tr>
<td>δ</td>
<td>0.009</td>
</tr>
<tr>
<td>BAE</td>
<td>0.007</td>
</tr>
<tr>
<td>AOD</td>
<td>0.05</td>
</tr>
</tbody>
</table>

*β, backscatter; σ, extinction; \( S_\lambda \), lidar ratio; δ, depolarization; BAE, backscatter Angstrom Exponent; AOD, aerosol optical depth.*
The vertical resolution of backscatter and extinction are 30 m and 300 m, respectively. [11] As our objective is to understand changes in aerosol optical properties in the vicinity of boundary layer clouds, we must first determine the proximity of the lidar beam to a cloud boundary. We identify cloud top altitudes for clouds located at or near the top of the planetary boundary layer using background subtracted, range-square-corrected profiles from the HSRL. A three point Haar wavelet covariance transform [Gamage and Hagelberg, 1993] is used to detect the sharp gradients in these HSRL profiles associated with cloud boundaries. [12] Thus the location of clouds along the flight track can be easily determined using HSRL data, but the HSRL cannot provide information about clouds that are not directly underneath the aircraft. The ideal flight tracks for our aerosol and cloud interaction study are those that have almost no clouds on either side of the clear portions on the tracks. We visually inspected Geostationary Operational Environmental Satellites (GOES) images obtained during the CATZ campaign to identify those ideal cases, and chose segments of flights on 4, 7, and 9 August for our study. Note the spatial resolution of GOES visible images is 1 km; therefore subpixel clouds may still be nearby. Backscatter profiles of the time periods used in this analysis for the 3 days are shown in Figure 1. The 4 August flight segment covers a distance of about 250 km from Maryland to Virginia with AOD around 0.4, the 7 August flight segment covers a distance of about 560 km off the coast of North Carolina with AOD around 0.7, and the 9 August flight segment covers a distance of about 200 km off the coast of South Carolina with AOD around 0.9. [13] Next, we choose a reference altitude of the low clouds ($H_c$) for each flight segment ($H_c = 2000$ m, 500 m, and 500 m for 4, 7, and 9 August, respectively), and analyze all of the clear HSRL profiles at this altitude. We define a profile as being “clear” if it does not have a valid cloud top height but has valid backscatter and extinction measurements over the entire altitude range. We define a profile as being “cloudy” if it has a valid cloud top height between $H_c$ and $H_c + 200$ m, assuming that the geometrical thickness of the boundary layer clouds is about 200 m [Bennartz, 2007]. However, we also tested cloud geometrical thickness of 500 m and 1000 m, which did not change the general trends of our results. [14] Then we save the aerosol optical properties ($\beta$, $\sigma$, $S_o$, $BAE$) in each clear profile ($x_i$), and search the temporal record to identify the closest cloudy profile ($x_j$) that occurs before or after $x_i$. Since $x_j$ is the nearest cloudy profile, any profile (such as $x_{i+1}$ and $x_{i+2}$) between it and $x_i$ is clear. We are interested in how aerosol properties in the near-cloud environment differ from the aerosol properties in the far-cloud environment, so we also note the aerosol optical properties in a clear profile $x_{i+2}$, which is $\sim 100$ m from $x_i$. Hereafter, $x_{i+2}$ is referred to as reference profile. We choose $x_{i+2}$ instead of $x_{i+1}$ to reduce the possibility of cloud contamination that could occur in profiles that are too close to clouds ($x_{i+1}$ is about $\sim 50$ m away from $x_i$). Nonetheless, our analysis produces statistically equivalent results when using $x_{i+1}$ or $x_{i+2}$ for the near-cloud reference profile, which is a testament to the robustness of the cloud screening technique used for the HSRL data. [15] We calculate the difference in aerosol optical properties between profile $x_i$ and reference profile $x_{i+2}$ for example, $\delta \beta = \beta_i - \beta_{i+2}$, where $\beta_{i+2}$ is the near-cloud reference value) and the difference is binned according to the distance between $x_i$ and $x_j$. Here we consider seven bins, which are listed in Table 2, along with the total number of samples in each bin for the 3 days analyzed in this study. For each bin, the averaged difference ($\delta \beta$, $\delta \sigma$, $\delta S_o$, $\delta BAE$) and the standard error for each aerosol property are calculated. Note that all the differences are calculated relative to reference profiles that are no more than 5 km away, which ensures that both $x_i$ and $x_{i+2}$ are under the same large-scale meteorological conditions. 3. Results [16] Figure 2 shows the averaged differences for 532 nm backscatter coefficient ($\delta \beta$, in unit Mm$^{-1}$ sr$^{-1}$) for the 3 days. The averaged near-cloud reference backscatter coefficients ($\overline{\beta}_{i+2}$) are also included. The error bars for each bin in Figure 2 are given by the standard error of that bin, calculated as the standard deviation of the differences divided by the square root of the number of samples of that bin. Figure 2 indicates systematic increases in $\beta$ as clear profiles get closer to clouds. For example, on 4 August, $\overline{\beta}_{i+2}$ is about 1.2 Mm$^{-1}$ sr$^{-1}$ (23%) higher than the averaged backscatter coefficient in the second distance bin (501–1000 m away from nearest clouds); and $\overline{\beta}_{i+2}$ is about 1.4 Mm$^{-1}$ sr$^{-1}$ (27%) higher than the averaged backscatter coefficient in the seventh distance bin (4001–5000 m away from nearest clouds). The other 2 days show similar increasing trends of $\beta$ as the clear profiles get closer to clouds. $\overline{\beta}_{i+2}$ is about 30% and 26% higher than the averaged backscatter coefficient in the seventh bin for 7 and 9 August. [17] The uncertainty of the averaged difference in each bin associated with the instrumental error can be calculated by dividing the instantaneous uncertainty, listed in Table 1, by the square root of the total number of samples of that bin. For example, the instrumental error in $\delta \beta$ is about 0.06 Mm$^{-1}$ sr$^{-1}$ for the first bin on 4 August. The $\delta \beta$ shown in Figure 2 is much larger than the instrumental error for all the bins on the 3 days, which indicates that the systematic trend is not affected by random instrumental error. [18] Depolarization ratios at 532 nm during these 3 days are less than 0.02 and exhibit nearly no changes in the vicinity of clouds, indicating that there are very few nonspherical dust particles. [19] The extinction coefficients at 532 nm for the 3 days are qualitatively similar to backscatter coefficients (not shown). For clear profiles in the second bin, the averaged extinction coefficients are about 0.10, 0.06, and 0.10 km$^{-1}$ lower than those near-cloud reference values and correspond to 24%, 10%, and 13% decreases. The differences are 0.17, 0.10, and 0.17 km$^{-1}$ for clear profiles in the seventh bin and correspond to 42%, 17%, and 23% decreases. Again, the averaged extinction differences in all bins for the 3 days are much larger than the instrumental errors. [20] Aerosol optical depth (AOD) can be computed by integrating the HSRL measured extinction profiles between the surface and the flight altitude of 6.5 km. Differences between AODs that are near and far from clouds are
Figure 1. Backscatter coefficient profiles at 532 nm for the periods used in this analysis on 4, 7, and 9 August 2007. Black dashed lines are the reference altitudes of low clouds for these three flights.

Table 2. Distances of the Seven Bins From the Nearest Clouds and Total Number of Samples (N) of Each Bin for the 3 Days

<table>
<thead>
<tr>
<th>Bin No.</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>Distance (m)</td>
<td>1–500</td>
<td>501–1000</td>
<td>1001–1500</td>
<td>1501–2000</td>
<td>2001–3000</td>
<td>3001–4000</td>
<td>4001–5000</td>
</tr>
<tr>
<td>N (08/04)</td>
<td>37</td>
<td>29</td>
<td>24</td>
<td>20</td>
<td>26</td>
<td>27</td>
<td>25</td>
</tr>
<tr>
<td>N (08/07)</td>
<td>362</td>
<td>200</td>
<td>108</td>
<td>60</td>
<td>90</td>
<td>70</td>
<td>62</td>
</tr>
<tr>
<td>N (08/09)</td>
<td>246</td>
<td>120</td>
<td>75</td>
<td>52</td>
<td>55</td>
<td>25</td>
<td>24</td>
</tr>
</tbody>
</table>
analyzed using the same method. Figure 3 shows the averaged differences for AOD at 532 nm ($\delta_{\text{AOD}}$). Averaged near-cloud reference AODs and their standard deviations of the seven bins are $0.47 \pm 0.007$, $0.73 \pm 0.009$, and $0.93 \pm 0.027$ for the 3 days. These standard deviations show that the reference AODs changed by $1$–$3\%$ along the flight tracks. AODs from profiles in the second distance bin are $0.03, 0.02$ and $0.07$ lower than those near-cloud reference values, on 4, 7, and 9 August, respectively. These correspond to $6\%$, $3\%$, and $8\%$ decreases relative to the reference AODs. AODs from profiles in the seventh distance bin are $0.04, 0.06$, and $0.16$ ($9\%$, $8\%$, and $17\%$) lower than the near-cloud reference AODs. The averaged AOD differences in all bins for the 3 days are much larger than the instrumental errors.

Figure 4 shows the averaged differences for the 532 nm lidar ratio ($\delta_{\text{L}}$) for the 3 days. Unlike the systematic decreases seen in $\beta$, $\sigma$, and AOD as clear profiles get farther away from clouds, we do not see any consistent changes in $\delta_{\text{L}}$ (except for those between 2000 and 5000 m away from the nearest clouds on 4 August). Note the instrumental error in $\delta_{\text{L}}$ for some bins with small number of samples can be up to 3 sr, which could be responsible for some of the variability seen in Figure 4. We also notice that the averaged near-cloud reference lidar ratios are $\sim 80$ sr for the 3 days that we investigated. Since the lidar ratio for water clouds is significantly lower ($\sim 18$ sr) [Pinnick et al., 1983; O'Connor et al., 2004], this once again shows that our results are not affected by significant cloud contamination.

Figure 5 shows the averaged differences for the BAE, and the averaged near-cloud reference BAEs. The trends of BAE are mixed. On 4 August, no systematic changes of BAEs are observed within 1500 m of clouds. Thereafter the BAEs decrease by about $0.05$ to $0.14$ ($5\%$ to $13\%$ relative to the near-cloud value) as the clear profiles get farther away from clouds. On 7 August, BAEs exhibit systematic increases as the clear profiles move farther away from clouds. BAEs increase by about...
0.15 to 0.09 (12% to 8%) for clear profiles that are between 2000 and 5000 m away from the nearest clouds. On 9 August, there is no systematic changes in BAEs.

4. Discussion

[23] The increased backscatter and extinction coefficients observed with the HSRL in the vicinity of clouds are not caused by cloud adjacency effects or cloud contamination; we know this because of the advantages of HSRL measurements outlined in section 1 (i.e., narrow field of view and high sensitivity to differences in scattering caused by aerosols and cloud drops). Other possible causes include (1) new particle production in the vicinity of clouds, (2) aerosol growth through in-cloud aqueous oxidation of SO$_2$ to sulfate and collision/coalescence, and (3) aerosol swelling as relative humidity (RH) increases near clouds.

[24] New particle production in the vicinity of clouds alters the aerosol size distribution. Hegg et al. [1990] noted that aerosol number size distribution in the vicinity of clouds consists of more smaller particles ($r < 0.05 \, \mu m$) than the nearby clear air aerosol size distribution, and suggested that this is due to homogeneous heteromolecular nucleation. Hoppel et al. [1994] observed a large number of small particles ($r < 0.01 \, \mu m$) above the cloud top and interpreted this as evidence of new particle formation. Clarke et al. [1998] also observed significant increase of ultra fine particles ($0.002 < r < 0.005 \, \mu m$) at the edges of clouds. However, given the very small size of these particles, they are unlikely to affect the measured extinction and backscatter coefficients [Schuster et al., 2006].

[25] Aerosol growth near clouds has been observed both in situ measurements and model simulations. Alkezweeny [1995] measured a decreased aerosol number concentration for optically active aerosols with radii below 0.2 \, \mu m, and an increased aerosol number concentration for aerosol radii between 0.2 and 1.5 \, \mu m in the processed clear air. He argued that the in-cloud chemical conversion of SO$_2$ to sulfate adds new material to droplet. Since every droplet generates only one aerosol particle upon evaporation [Mitra et al., 1992], the new size is therefore larger. Although Alkezweeny [1995] did not measure particle sizes...
greater than 1.5 μm, we note that the increase in particle number concentrations altered the particle size distribution of the coarse mode \((r \gtrsim 0.5 \mu m)\) as well as the accumulation mode.

[26] Hegg et al. [2004] observed enhanced light-scattering efficiency in the vicinity of clouds, and attributed the enhancement to a shift in the aerosol size distribution toward a more effective scattering size range. Feingold and Kreidenweis [2002] used a large eddy simulation to demonstrate that aqueous chemistry increased the aerosol number concentration for radii between 0.1 and 1.5 μm (no data were shown beyond 1.5 μm). Kerkweg et al. [2003] used an air-parcel model to demonstrate that aqueous chemistry and coalescence of cloud droplets enhanced the number of larger aerosol particles \((r >2 \mu m)\) in the processed aerosol size distribution.

[27] Swelling of aerosols in the high RH environment can alter their optical properties and possibly their size distribution depending on their composition. We investigate the observed near- and far-cloud differences in aerosol optical properties by simulating aerosol hygroscopic growth, following the method outlined by Loeb and Schuster [2008]. That is, we assume that the aerosols in our study are an equilibrium mixture [Tang, 1996] of ammonium sulfate and water in the fine mode and sea salt (or dust) in the coarse mode, and that the size distribution is represented by the GSFC climatology of Dubovik et al. [2002]. Then we use Mie theory [Wiscombe, 1980] to compute the backscatter, extinction, lidar ratio, and backscatter Angstrom exponent.

[28] The backscatter coefficient \((\beta)\) and lidar ratio \((S_a)\) are plotted in Figures 6 and 7 for four dry fine volume fractions, assuming sea salt occupies the coarse mode. The results for dust in the coarse mode are qualitatively similar, except that the sensitivity of the lidar ratio to dry fine volume fraction is smaller at high RH. Since the hygroscopic growth factor is different for the fine and coarse modes in our simulations (owing to the different composition of the two modes), the wet fine volume fraction changes along the curves in Figures 6 and 7, even though the dry fine volume fraction remains the same. Hereafter, all fine volume fractions in our discussion refer to dry fine volume fractions. If aerosol swelling in the vicinity of clouds is the only mechanism altering the \(\beta\) and \(S_a\), the aerosol properties are expected to follow a single curve. However, if aerosol growth through in-cloud processing alters the relative distribution of the fine and coarse modes, then \(\beta\) and \(S_a\) will move from one curve to another in Figures 6 and 7.

[29] Figure 6 shows the simulated backscatter coefficient \((\beta)\) as a function of RH for four dry fine volume fractions; \(\beta\) increases by a factor of two as RH increases from 55% to 90%, and thereafter it increases sharply. In fact, \(\beta\) at 99% RH is more than an order of magnitude larger than at 55% RH. Also note that \(\beta\) is not sensitive to fine volume fraction for RH less than 97%, especially for aerosols dominated by the fine mode (like pollution). The relationship between extinction coefficient and RH is qualitatively similar to our backscatter coefficient simulation (not shown); the extinction coefficient is also not sensitive to the fine volume fraction when RH is less than 97%.

[30] Figure 6 clearly indicates that aerosol swelling is consistent with the unambiguous increase of backscatter and extinction coefficients observed in the vicinity of clouds (Figure 2). In addition, aerosol growth through in-cloud processing (which enhances the aerosol light-scattering efficiency), can also increase backscatter and extinction in the vicinity of clouds.

[31] The unambiguous increase of extinction coefficients also translates to the increase of column aerosol optical depths at 532 nm in the vicinity of clouds. AODs increase by about 8% to 17% in the vicinity of clouds than far away from clouds (4.5 km). Koren et al. [2007] found that AOD at 440 nm decreased by about 13% ± 2% on average from...
the first sample measured near a cloud to the second sample (less than 15 minutes later). In their study, multiple years of data from 15 AERONET stations were used, and the boundary layer wind speed data were not always available at these stations to translate the time interval between these two samples to geometrical distance. However, if we assume a boundary layer wind speed of 4.96 m s\(^{-1}\) (the globally averaged daytime 80-m wind speed from Archer and Jacobson [2005]), we can approximate the distance between these two samples to be \(\sim 4.5\) km. Although the wind speed we use here is a crude approximation, it nonetheless shows that AOD increases in the vicinity of clouds at nearly the same magnitude for both the Koren et al. [2007] study and our study. The AOD increase in the vicinity of clouds from these two studies (which are not affected by cloud adjacency effects) is much smaller than the cloud adjacency effects on AOD retrieval (50 to 140% overestimation) [Wen et al., 2006, 2007].

The simulated lidar ratio \((S_o)\) also increases as dry fine volume fraction and RH increase, as shown in Figure 7. If aerosol swelling is the only mechanism affecting our HSRL measurements in the vicinity of clouds, then \(S_o\) should increase near clouds, which is not consistent with our observations; 2 out of 3 days shown in Figure 4 do not exhibit any systematic trend in \(S_o\). One possible explanation is aerosol growth caused by oxidation of \(\text{SO}_2\) and collision/coalescence increases the relative fraction of coarse mode aerosols in the vicinity of clouds. Since \(S_o\) is smaller for coarse mode aerosols than for fine mode aerosols, this counteracts the enhancement in \(S_o\) caused by aerosol swelling; hence, the nonsystematic trend in Figure 4. Unfortunately, we do not have simultaneous aerosol size and RH measurements to verify this hypothesis.

The simulated BAE increases as fine volume fraction at a given RH (as expected), but the relationship between RH and BAE is not monotonic (not shown). BAE first decreases as RH increases, until a certain RH (94% for fine volume fraction of 0.65), then BAE increases. Therefore depending on the RH values next to clouds and away from clouds, and the aerosol properties along the flight tracks, the BAE differences shown in Figure 5 can be explained by aerosol swelling and growth. This once again stresses the importance of concurrent RH and aerosol property measurements.

To investigate if BAE could be used as a surrogate for Angstrom exponent (AE), we present BAE as a function of AE for four dry fine volume fractions as RH increases from 55% to 99% in Figure 8, using sea salt for the coarse mode. The AE is defined for wavelengths 553 and 855 nm, which are the same wavelengths used for one of the MODIS’ AEs over the ocean [Remer et al., 2005]. For RH less than \(\sim 85\%), BAE and AE are linearly correlated for aerosol size distributions dominated by the fine mode (BAE decreases as AE decreases). As aerosols continue to swell, however, the BAE increases while the AE decreases. Also, the BAE and AE are anticorrelated for coarse mode dominated aerosols over the RH range considered here. Therefore the humidification trends of BAE and AE are not similar, and BAE should not be used as a surrogate for Angstrom exponent. Also, the information content about the aerosol size distribution in the BAE is very limited, as also shown by Feingold and Grun [1994].

5. Summary

We used aircraft HSRL measurements to study aerosol and cloud interaction during the CATZ campaign. Unlike satellite retrievals, HSRL observations are not affected by cloud adjacency effects, and offer accurate measurements of aerosol properties in the vicinity of clouds. The aerosol properties investigated in this study include aerosol backscatter and extinction coefficients, lidar ratios, backscatter Angstrom exponents, and column aerosol optical depths. The HSRL also provides robust cloud screening, and we used GOES images to select observational periods with very few low clouds on both sides of the flight tracks, thereby minimizing the effects of clouds that do not appear in the lidar beam.

We examined the aerosol properties of HSRL profiles in the vicinity of clouds according to the distance from the nearest clouds, observing differences between clear profiles that are adjacent to clouds (\(\sim 100\) m from clouds) and clear profiles that are up to 5000 m away from clouds. The short distance between these profiles (<5000 m) assures the same large-scale meteorological condition for both profiles, which is important for studying aerosol and cloud interactions [Loeb and Manalo-Smith, 2005; Mauger and Norris, 2007; Loeb and Schuster, 2008].

Results from three observations in August 2007 reveal unambiguous increases of backscatter coefficients, extinction coefficients, and aerosol optical depths in the vicinity of clouds. These increases are possibly caused by aerosol swelling and aerosol growth in the vicinity of clouds. However, we do not observe any systematic lidar ratio changes in the vicinity of clouds. We hypothesize that changes in \(S_o\) are neutralized by the opposite effects of aerosol swelling associated with a high relative humidity environment (which increases \(S_o\)), and aerosol growth associated with cloud processing (which decreases \(S_o\) if the aerosol size distribution shifts to the coarse mode). We do not observe systematic changes of BAE in the vicinity of
clouds, either. Theoretical simulations reveal that the relationship between BAE and RH is rather complicated, and that it would be rather difficult to infer changes in the aerosol size distribution from BAE. Furthermore, BAE should not be used as a surrogate for Angstrom exponent, especially at high relative humidity.

References