Comparisons of remote sensing retrievals and in situ measurements of aerosol fine mode fraction during ACE-Asia

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Received 19 October 2005; revised 16 December 2005; accepted 23 January 2006; published 8 March 2006.

[1] We present sunphotometer-retrieved and in situ fine mode fractions (FMF) measured onboard the same aircraft during the ACE-Asia experiment. Comparisons indicate that the latter can be used to identify whether the aerosol under observation is dominated by a mixture of modes or a single mode. Differences between retrieved and in situ FMF range from 5–20%. When profiles contained multiple layers of aerosols, the retrieved and measured FMF were segregated by layers. The comparison of layered and total FMF from the same profile indicates that columnar values are intermediate to those derived from layers. As a result, a remotely sensed FMF cannot be used to distinguish whether the aerosol under observation is composed of layers each with distinctive modal features or all layers with the same modal features. Thus, the use of FMF in multiple layer environments does not provide unique information on the aerosol under observation. **Citation:** Gassó, S., and N. O'Neill (2006), Comparisons of remote sensing retrievals and in situ measurements of aerosol fine mode fraction during ACE-Asia, *Geophys. Res. Lett.*, *33*, L05807, doi:10.1029/2005GL024926.

1. Introduction

[2] Automated retrievals of aerosol optical depth (AOD) by spaceborne detectors have significantly improved our knowledge of the global distribution of aerosols [*Kaufman et al.*, 2002]. In addition, they have provided a measurement based verification of aerosol forcing derived from global aerosol models [*Penner et al.*, 2002]. However, passive remote sensing techniques have not had the same degrees of success in detecting aerosol size distribution properties. A proper global characterization of size distribution properties is important because it would improve the simulation of microphysical properties in global aerosol models [*Zhang et al.*, 2002]. The concept of fine mode fraction (FMF) has been introduced to describe columnar aerosol modal features using passive spectral detectors such as MODIS [*Tanré et al.*, 1997]. FMF is defined as the ratio of the accumulation mode OD to the total OD at 550 nm. It provides quantitative information on the nature of the aerosol size distribution. The FMF is defined such that it ranges from 0 to 1. The extreme values represent pure conditions where the total radiance can be modeled by a single accumulation mode (FMF = 1) or a single coarse mode (FMF = 0). For intermediate values, both modes contribute to the total radiance with each contributing to the total AOD in proportion to FMF and 1-FMF respectively [*Remer et al.*, 2005]. Because of its close association with modal features, the MODIS FMF product has been used for discriminating between natural and anthropogenic aerosols [*Kaufman et al.*, 2005]. Few studies have been dedicated to comparisons of FMF with corresponding ground retrievals or in situ measurements. Because it is rather difficult to carry out campaigns of aircraft aerosol measurements synchronized to satellite overpass times, comparisons with in situ measurements have been limited to case studies [*Gassó and Hegg*, 2003]. *Kleidman et al.* [2005] compared MODIS FMF retrievals with collocated AOD measurements made by the AERONET network using two retrieval techniques, one based on the existing operational retrieval [*Dubovik and King*, 2000] and the other using the *O'Neill et al.* [2003] technique. The latter method relies on AOD spectral derivatives to extract FMF whereas the former is employed to retrieve aerosol modal properties from the angular and/or spectral variation of sky radiances and solar extinction measurements. Unlike the Dubovik method, the O'Neill retrieval technique is used to derive FMF directly from OD spectra. It is attractive given the significantly greater frequency and weaker cloud contamination of AOD measurements (as opposed to the less frequent measurements in the Dubovik inversion). It also has the potential of being easily implemented as a MODIS FMF retrieval algorithm, and thus provides an alternative to the existing technique. The latest version of the model is employed here [*O'Neill et al.*, 2005].

[3] We applied the O'Neill technique to AODs retrieved by the AATS-6 sunphotometer onboard the NCAR C130 aircraft deployed during the ACE-Asia campaign [*Redemann et al.*, 2003]. The same aircraft carried a suite of in situ aerosol instrumentation [*Clarke et al.*, 2004]. Of particular interest are the measurements of total and accumulation mode extinction coefficients from which it is possible to integrate over the column and obtain an in situ FMF. The analysis and comparison between these measurements and the collocated AATS-6 retrievals are reported in this study.

2. Data Set

[4] In this study, we employed the same profiles utilized by *Redemann et al.* [2003] with an additional criterion in the data selection: particle size distribution measurements had to be simultaneously available with the nephelometer and sunphotometer data. In this way, distinction of aerosol type could be made based on number and volume concentration. As a result, some of the profiles were discarded because the optical particle counter was not functioning or

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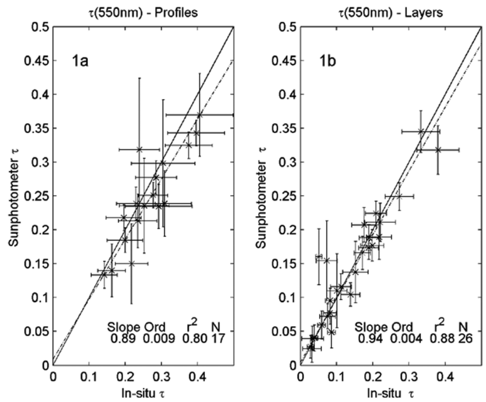

Figure 1. Retrieved and in situ AOD (550 nm) for (a) profiles and (b) individual layers within profiles.

too few size distribution measurements were made. An interesting feature of the aerosol vertical structure during ACE-Asia was the presence of multiple aerosol layers [*Kahn et al.*, 2004]. The individual aerosol layers were identified by the change in the total number and volume concentration measurements. In most of the cases, the layer edges were easily identified by the abrupt change in concentration. In cases where the change in concentration was smooth, an arbitrary threshold based on number concentration was used.

[5] The derivation of AODs, FMF and respective uncertainties from in situ and AATS-6 data followed the same procedure as described by *Redemann et al.* [2003] and *Anderson et al.* [2003]. The AATS-6 data and in situ data were selected by matching the time period when the plane was sampling the layers or profiles. In situ extinction coefficients were measured using a four-nephelometer system. Two nephelometers (two PSAPs) measured the dry scattering (absorption) coefficients for particles smaller than (nominally) 10 and 1 um in aerodynamical diameter. The extinction coefficient was derived after applying a correction to ambient conditions with an optical humidity factor measured by two nephelometers set at different RHs. All the measurements were made at or interpolated to 550 nm. Coarse mode extinction coefficients are reduced by 10% to account for plumbing and probe effects [*Anderson et al.*, 2005]. The fine mode and total AODs are computed as the integral of the respective ambient extinction coefficient measured at 550 nm, then the in situ FMF is calculated as the ratio of fine to total AODs. The AATS-6 AODs (380.1, 450.9, 525.7 and 1021.3 nm) are selected by taking the differences between the bottom and the top of the selected profile or layer and propagating the respective errors.

3. Comparison of Optical Depth Retrievals With Measurements

[6] We tested the consistency of the in situ data in reproducing columnar optical data. The objective of this exercise is to verify that our data selection method captures all the necessary information needed to reproduce an optical ambient parameter and minimize any systematic error that may affect the in situ and retrieved FMF because of the data selection procedure. The AATS AODs at 550 nm are linearly interpolated from the closest two wavelengths. Figure 1 shows (a) profile and (b) layer AODs derived from the AATS and in situ data. The figure includes the fit parameters computed using a model II least squares bisector regression [*Redemann et al.*, 2003]. Both panels show very good agreement with high correlation coefficients, slopes near 1 and small offsets. The selection of data by layers expands the range of optical depths at low values. In addition, the analysis by layers reproduces the ambient AODs significantly better as indicated by the linear regression coefficients. For data from layers, the error bars associated with both the sunphotometer and in situ data are smaller. For the in situ data, layers have better signal-to-noise ratios than whole profiles. The reason is that data from whole profiles include layers of clean air and the nephelometers measure very close to the noise level resulting in low signal-to-noise ratios. When propagated over the column, it results in large uncertainties. Because of the customized nature of the selection process, these gaps of clear air are not included in the layer data. In the sunphotometer data, the smaller error bars in layers are a direct consequence of the smaller horizontal variability of the ambient aerosol since the plane flies a shorter horizontal distance between the top and bottom of the layer. Although by selecting layers it is more likely that the AATS ODs will be closer to the detection threshold of the instrument, it is clear in Figure 1b that the smaller layer optical depths match the in situ data very well. These results are in agreement with *Redemann et al.* [2003], who obtained a similar AOD comparison with a slightly different data set. Figure 1 confirms that the selected in situ data successfully reproduce an extensive ambient optical property such as the optical depth at 550 nm.

4. Comparison of Retrieved and Measured FMF

[7] The retrieved FMF is derived at 550 nm by using the O'Neill algorithm. Because the retrieval method is sensitive to errors in the spectral dependence of the AOD and the effects of interband AOD errors increase in severity as the AOD decreases, the inversion was applied only to those profiles with $\tau(1020\ \mathrm{nm}) > 0.04$. We note that the AATS optical depth errors consist essentially of two types of uncertainties. The first is related to calibration, filter response and other instrumental corrections. The second is an estimation of the variation due to the aerosol change while the plane is descending through the column (horizontal variability error) [*Redemann et al.*, 2003]. The retrieved FMF errors were accordingly estimated using a combination of the stochastic error model defined by *O'Neill et al.* [2003] and the FMF variance due to the horizontal variability. For the former, the AOD measurement errors were input directly into the stochastic error model; in the latter, a simple model of FMF variation due to systematic spectral changes induced by natural (horizontal) AOD variations across a flight line was employed to estimate a corresponding FMF variance. The two contributions (effectively representing incoherent and coherent variations) were then summed quadratically to achieve a final total FMF variance for each retrieved FMF data point.

[8] Figure 2 shows retrieved and in situ FMFs (with the fitting parameters) derived from (a) profiles and (b) indi-

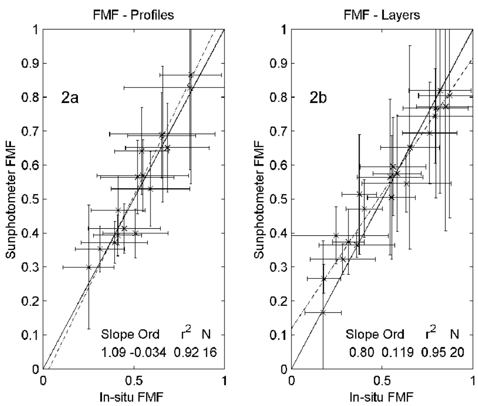

Figure 2. Retrieved and in situ FMF (550 nm) for (a) profiles and (b) individual layers within profiles.

vidual layers. The slope of the linear regression indicates that the retrieved FMF is underestimated with respect to the in situ data in the layers and overestimated in the profiles. Closer inspection of the point distribution shows that in the case of layer data, the retrieved FMF tends to be underestimated at high values (above $\sim$0.5) whereas at lower values, the retrieved values are overestimated with respect to in situ data. Although it is possible that the in situ measurements did not sample all accumulation mode particles at low values of FMF, previous studies with this same data set do not seem to support this hypothesis. For example, *Anderson et al.* [2003] reported excellent efficiencies in the measuring system, indicating that transmission losses were unlikely for small particles (<2 um diameter). However, coarse mode particles are more difficult to measure in an airborne platform. Even though the aircraft had an inlet probe optimized to perform best with particles <10 um diameter [*Huebert et al.*, 2003], post deployment studies pointed out that there were still some inefficiencies in the sampling of large particles. *Clarke et al.* [2003] were able to successfully model measured scattering coefficients from size distributions using the same set of measurements. They pointed out that particle losses were minor in fine mode dominated environments whereas during dust events the simulations disagreed by 10%. They suggested that transmission efficiencies and calibration in the sampling instruments could be the reason for the observed differences. This suggests that the in situ FMF will be affected most where the coarse mode contribution is a significant proportion of the total extinction. The FMF error bars computed for the O'Neill method retrieval were dominated by the errors in the first and second spectral derivatives at 550 nm (α and α'). These errors were not effectively reduced by the filtering action of a second order polynomial fit to the AOD spectra since four wavelengths represents minimal redundancy above the three degrees of freedom of the fit (which is not the case for typical second order AERONET fits to 6 or 7 wavelengths). Other sources of difference not incorporated in the stochastic error model include the possible effects of water vapor absorption at 1020 nm and differences due to the O'Neill method employing an optical discrimination of fine and coarse mode versus an effective mechanical discrimination for the in situ estimates of FMF (see *O'Neill et al.* [2003] for specific examples).

[9] It is clear the FMFs retrieved in layers span a larger range of values than the FMFs retrieved from profile data. This is reasonable since selection by layers is more likely to capture an aerosol with definite modal and composition features such as dust or pollution dominated.

[10] *Kleidman et al.* [2005] compared MODIS retrievals with AERONET fine mode fractions derived by integrating the Dubovik inverted size distributions and by the O'Neill method. When comparing with MODIS retrievals, they found that the satellite retrievals tended to overestimate at low values of FMF with respect to the O'Neill method. At high values, the MODIS retrievals tended to underestimate slightly with respect to the O'Neill method. Our comparisons show exactly the opposite trend. It is possible that our FMF retrievals are sensitive to the relatively high OD uncertainties (compared to AERONET) in the AATS retrievals and the lack of built in data redundancy due to the smaller number of spectral bands in the latter case.

5. Interpretation of FMF in Multiple Layer Environments

[11] In passive remote sensing measurements, the retrieved parameter is representative of the entire atmospheric column. If the parameter is an extensive property such as the optical depth, it can be interpreted as the sum of the contributions of each layer in the column. When retrieving an intensive property such as FMF, it is reasonable to expect that the retrieved parameter is representative of that particular aerosol in the column if the aerosol under observation has uniquely defined modal features such as dust or pollution/smoke. However, if multiple layers of different aerosol types are present in the column, the resulting FMF will be an optical mean of the pure FMFs representing each layer. In the absence of some layer altitude discrimination, it is not possible to ascertain what the modal features of the aerosol are from a satellite-based retrieval of FMF. This concept can be illustrated with the current data set. Figure 3 shows sunphotometer FMFs from profiles with more than one layer present. It shows the derived FMF for complete profiles (dark bars) and for individual layers in the same profiles (light bars) for five profiles. Size distribution data

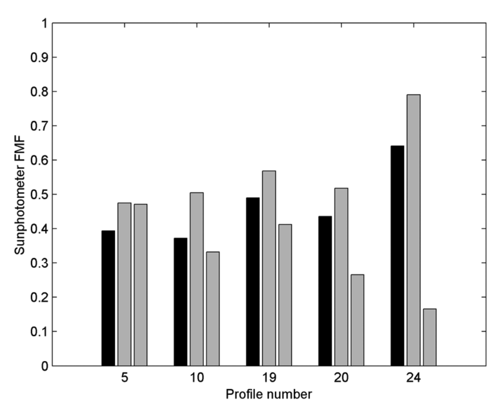

Figure 3. Retrieved FMF for profiles including two aerosol layers. Whole profile FMFs are in dark and FMF for the individual layers within the same profile are in grey. The first grey bar of each pair of grey bars represents a lower layer.

indicated that in all five cases the profile contained an aerosol layer in the boundary layer and another in the free troposphere. In the case where the layers have very similar modal features (profile 5), the corresponding columnar FMF is close to both layer values. In the case of significant modal differences between the layers (profiles 10, 19, 20 and 24), it is clear that the FMF has an intermediate value. Given a satellite measurement of an intermediate FMF value, the result is indistinguishable from a single aerosol with artificially averaged loadings in both accumulation and coarse modes versus two or more aerosol layers with different modal features in each layer. The example shows that FMF cannot be unequivocally associated with a unique aerosol type unless both layer values are similar.

6. Final Comments

[12] We have shown simultaneous and collocated retrievals of aerosol fine mode fraction with corresponding in situ measurements collected during the ACE-Asia campaign. The retrievals employed an alternative technique, which is easy to implement as long as aerosol optical depth spectral measurements are available. We analyzed the in situ and sunphotometer data in terms of total column profiles (the defacto standard in sunphotometer–satellite comparisons) and in terms of layers within the same profiles. The latter approach expanded the range of FMF variability and captured aerosols with more purely defined features. We showed that the retrieved FMF can be a good indicator of the modal features of the columnar aerosol size distribution. The difference between an in situ and retrieved FMF at low FMFs (<0.5) ranges from 10% to 20% whereas at high FMFs (>0.5) the differences range from 5% to 15%. Quantitatively, the differences tend to decrease as FMF approaches 1. When multiple aerosol layers are present, the column FMF retrievals yield values intermediate to the FMFs of the individual layers in the same column. This comparison indicates that a columnar retrieval of FMF cannot provide a unique characterization of the aerosol size distribution in the column unless all layers contain aerosols with similar modal features. This suggests that in multiple layer environments such as pollution and dust in East China, columnar retrievals of FMF do not provide unique information on the aerosol under observation.

[13] Because the O'Neill method relies exclusively on spectral AOD retrievals, it can be applied to spaceborne retrievals. In particular, the range of uncertainties of MODIS AOD retrievals over ocean is comparable to those of the AATS sunphotometer during ACE-Asia. Given the good results shown in this study, it is suggested the O'Neill inversion method can be used as an alternative method for the derivation of FMF using MODIS AODs.

[14] **Acknowledgments.** We thank Tad Anderson and Jens Redemann for their comments on the in situ and sunphotometer data, respectively. This research was funded under the auspices of the NASA Radiation Program (NASA grant NNG04GM36G).

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