# **Spectral Signature of Solar Radiation Absorption**

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### Introduction

Many studies have addressed the issue of the absorption of solar radiation within the atmosphere, in both cloud-free and cloudy conditions (e.g., Stephens and Tsay 1990). Comparisons between modeled solar radiation absorption and observations suggest that the models underestimate the amount of radiation absorbed by the atmosphere for both clear and cloudy skies (Ramanathan et al. 1995; Cess et al. 1995; Pilewskie and Valero 1995; and Kato et al. 1997).

The Atmospheric Radiation Measurement Enhanced Shortwave Experiment (ARESE) was conducted in the fall of 1995 to address the issue of enhanced absorption in a cloudy atmosphere from an observational perspective. Measurements were obtained above and below clouds, with both spectral and broadband radiometers. No in-cloud observations were, however, collected. An analysis of broadband observations for one day by Zender et al. (1997) suggests a suspiciously large (nearly 100Wm<sup>-2</sup>) difference between modeled and observed absorption.

In an attempt to elucidate the possible causes for such large absorption, this paper describes the observed spectral characteristics of the absorption for the day analyzed by Zender and compares them with those produced by a model designed to simulate the observations and provide physical insight.

## **Spectral Observations**

Two aircraft, flying above and below clouds, were equipped with an identical Radiation Measurement System (RAMS) and a Total Direct Diffused Radiometer (TDDR), measuring solar irradiance at seven spectral bands (approximately 10 nm wide) centered at 0.500, 0.862, 1.064, 1.249, 1.501, 1.651 and 1.750  $\mu$ m. Nadir viewing spectral reflectance (0.418 - 1.096  $\mu$ m) was also obtained from observations made by the Scanning Spectral Polarimeter (SSP) on the highest aircraft. Only the spectral data are used in this study.

## **Model Simulations**

To evaluate the consistency between the different sets of spectral observations and diagnose the physical processes involved in enhanced absorption, simulated fluxes were computed for a synthesized cloud field. To ensure that the cause of the enhanced absorption was not three-dimensional (3-D) effects and to match the spatial variability of the diffused upwelling flux, the 3-D Monte Carlo radiative transfer model described in O'Hirok and Gautier (1998) was used. The variability of the cloud liquid water distribution field was derived from downwelling flux observations obtained from the TDDR at

0.500  $\mu$ m. The field is presented on Figure 1. The mean liquid water path is 302 g m<sup>-2</sup>. The cloud droplet radius distribution has an average  $r_e$  of 7.3  $\mu$ m and follows a modified gamma size distribution. The corresponding mean cloud optical depth,  $\tau$  is 63. The cloud droplet single-scattering albedo, extinction efficiency, and phase function were computed directly from Mie theory for each spectral calculation. Pressure, temperature, and water vapor vertical profiles were derived from soundings at the Cloud and Radiation Testbed (CART) site, while ozone amount was obtained from surface measurements.



Figure 1. Synthetic cloud liquid water concentration, effective radius cross sections and vertically integrated liquid water path.

### **Model and Observations Comparisons**

#### **Spectral Variations of Albedo and Transmission**

Comparisons of model computations with measurements were made (but are not shown here) for the seven spectral channel TDDR at the level of both aircraft and with the SSP. In general, there was very good agreement (better than 5%) with the TDDR downwelling spectral radiation flux and the SSP nadir upwelling spectral flux. Comparisons between upwelling flux for most channels provided fair results for most channels (within 10%), except for 1.06  $\mu$ m, and a general deterioration for the longer wavelength channels. Comparisons between modeled and SSP-observed upwelling flux around 1.06  $\mu$ m also indicate a significant discrepancy.

To reduce the noise in the instantaneous measurements, they were averaged over the length of the flight. The results from this comparison are presented on Figure 2 (a, b) for albedo and transmission. On both figures the vertical bars indicate the magnitude of the standard deviation (from the flight average). For albedo, the model results and the SSP observations are in exact agreement at 0.500  $\mu$ m, suggesting the reasonableness of the model results and the input cloud field. Differences between model results and observations exist for most other TDDR channels. A large discrepancy exists between model computations and SSP observations at 1.06 $\mu$ m. For transmission, there is a much better agreement between model and observations, except for 1.06 $\mu$ m.

#### **Spectral Variations of Absorption**

The comparisons discussed above suggest that the model provides a reasonable description of the cloud environment that existed on the analyzed day. Therefore, it can be used to interpolate between discrete measurements and to compute the column spectral radiation absorption for that flight. The flight-averaged spectral distribution of atmospheric column absorption has therefore been computed from the model and is compared with values computed from the aircraft observations on Figure 3. Significant differences exist between the computed and the observed absorptance, reflecting the differences in the albedo and transmission discussed above. In general, the agreement is best for the shorter wavelengths regions (0.500  $\mu$ m) and deteriorates for longer wavelengths. A large difference exists at 1.06  $\mu$ m and for the three longest wavelengths of the TDDR (1.501, 1.651, and 1.750  $\mu$ m).

### **Enhanced Absorption**

The results presented above are indicative of the possible existence of some absorption not represented by the model. Some of this absorption could result from uncertainties in the measurements, but the accuracy noted by the instrument providers is smaller than the unexplained absorption. In an attempt to minimize the difference between modeled and observed absorption, we have modified the input parameters to our model in such a way as to maximize the modeled absorption, while keeping the input data within realistic bounds. Since the absorption results are relatively insensitive to atmospheric parameters such as temperature and water vapor profiles, and ozone amount, we have limited our modifications to the aerosol and cloud droplet properties. The results of these changes are presented in Figure 4. The best fit between modeled and observed absorption has been obtained by slightly changing the aerosol optical depth from 0.12 to 0.15, the single-scattering albedo from 0.938 to 0.82, and the asymmetry factor from 0.67 to 0.61. For the cloud droplets, the co-albedo increased by a factor 3. The agreement between modeled and observed absorption with these tuned parameters is now very good for almost all wavelengths with an exception at 1.06  $\mu$ m, where the modeled absorption is still much smaller than that observed.



**Figure 2**. Modeled average Egrett flight level spectral albedo and Otter flight level transmission along flight path (gray line). Modeled (plus) and observed (asterisk) albedo average for TDDR channels. SSP (diamond) albedo average and standard deviation.



Egrett - Otter atmospheric column

**Figure 3**. Modeled average Egrett-Otter atmospheric column absorptance along flight path (gray line). Modeled (plus) and observed (asterisk) absorptance average for TDDR channels.

### Discussion

The decrease of the aerosol single-scattering albedo and asymmetry factor required to reconcile the shorter wavelengths modeled values with the observations suggest a type of aerosol more absorbing than typical urban aerosol. This result is not too surprising in view of other results (Ricchiazzi et al. 1999) that indicate that to match clear-sky diffuse irradiance observations with theory requires highly absorbing (soot-like) small particles. In fact, when such small particles are included in the present computations, a better agreement between model and observations is obtained for the spectral distribution of absorption at the shortest wavelengths.

The more puzzling aspect in the closure between observed and modeled absorption is the requirement for very absorbing cloud droplets (co-albedo 3 times that of pure water). This conclusion is entirely based on near infrared measurements of the TDDR. If we exclude purely instrumental problems, the only physically sound explanation that quantitatively matches these results corresponds to a small amount of drizzle in the cloud layer. A sensitivity study indicates that a layer of drizzle of optical thickness 2 is sufficient, and that the effects are only slightly dependent upon the location of that layer.



**Figure 4**. Modeled average Egrett-Otter atmospheric column absorptance along flight path (gray line). Modeled (plus) and observed (asterisk) absorptance average for TDDR channels. (Same as Figure 3, but for adjusted cloud droplet co-albedo and aerosol.)

However, the amount of liquid water required for this drizzle is inconsistent with Microwave Radiometer observations. Soot-containing particles were not an acceptable solution since they would produce a spectral signature dramatically different than that observed at the shorter wavelengths.

The other mystery, and the one that may require some modifications to our understanding of the physical processes underlying the observations, is the enhanced absorption at 1.06  $\mu$ m. This feature is present in both spectral data sets analyzed (TDDR and SSP), which gives us more confidence in its validity. This spectral region corresponds to the absorption by O<sub>2</sub>-O<sub>2</sub> dimers. A modification of the absorption cross section based on the most recent results from Susan Solomon (personal communication) produced too small an absorption value to explain the enhanced absorption derived.

# **Summary and Conclusion**

The results presented here have shown that the spectral signature of absorption in a cloudy layer could be duplicated (except for the 1.06-µm region) with a rather sophisticated radiative transfer model if the absorption by both aerosol and cloud droplets was enhanced. In the case of aerosol, highly absorbing

(imaginary part of refractive index between 0.1 and 0.01) small (2 - 5 nm) particles dramatically improved the match between observations and model computations. Duplication of the observed cloud absorption requires a three-fold increase in cloud-droplet single-scattering albedo. The only feature remaining unexplained at this time is the enhanced absorption at 1.06 µm.

These results are only based on one day of observations and therefore they need to be verified. This study suggests the need for additional collocated broadband and spectral observations in clear and cloudy sky conditions in different atmospheric regimes. In situ aerosol and cloud droplet microphysical measurements will be crucial to unravel the role of these particles in the "enhanced absorption" issue. Finally, accurate absorption measurements are needed at  $1.06 \,\mu\text{m}$  to understand observed absorption in that spectral region.

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