

FINAL REPORT ON PARTICIPATION IN GACP BY DEAN HEGG

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TITLE: "Investigation of the Relationships Between Aerosol Physical and Chemical Properties Derived From In-situ Measurements and Their Impact on Aerosol Remote Retrieval"

ORIGINAL ABSTRACT OF PROPOSAL:

Funding is requested for the PI to participate in the Aerosol Forcing Science Team (AFST) and for modest salary support to pursue analyses of data sets which are largely in hand. This funding will supplement support, which is currently in place from both ONR and NASA itself (in the form of a NASA Global Change Fellowship for one of the PI's graduate students). In addition to the obvious tasks associated with participation in the AFST, two objectives are proposed for the supplemental support. The first is the quantification of the role of water of hydration on the radiative properties of atmospheric aerosols. Current remote sensing algorithms cannot differentiate the impact of water from that of changes in dry aerosol mass, composition or size. Clearly this is undesirable. The second objective is to find a widely applicable quantitative relationship between aerosol mass and number concentration over the critical size range in which particles can act as cloud condensation nuclei (CCN). This will greatly facilitate remote retrieval of CCN concentrations—a key parameter for assessment of indirect radiative forcing of climate by aerosols.

GOALS AND OBJECTIVES:

The general issue which we have explored in the course of this study involves the impact of water of hydration on aerosol optical properties and thus on the top of the atmosphere radiance attributable to these aerosols. Currently, satellite aerosol retrievals inseparably convolute dry aerosol mass and water of hydration together. For example, there is no explicit RH dependence in such retrievals. This has adverse ramifications for elucidation of both direct and indirect aerosol forcing. Hence, the explicit goal of our study has been the formulation of parameterizations to relate water of hydration and "intrinsic" aerosol properties to aerosol optical properties as orthogonal factors, most likely as a function of aerosol type. This goal, in turn, has been broken down into several more specific goals. First, we have assessed the impact of aerosol hygroscopicity on satellite detected irradiances from the standpoint of remotely detecting this quantity. Second we have explored the feasibility of retrieving the dry aerosol volume in marine air since previous studies had suggested that this quantity may directly relate to the CCN number concentration at typical supersaturations in marine clouds and retrieval could permit remote retrieval of CCN concentration (e.g., Hegg and Russell, *J. Geophys. Res.*, 105, 15321-15328, 2000).

APPROACH:

The methodology, which we have employed to address the above issues, is a combination of analysis of in-situ aerosol measurements and numerical modeling to link the observed aerosol properties with satellite measurements (or other types of remote retrieval). For example, direct, in-situ measurements of aerosol hygroscopicity obtained during the ACE-2 experiment have been used to generate model prediction of the impact of this hygroscopicity on top-of-the-atmosphere radiances (see below).

RESULTS:

Based on measurements obtained in the region surrounding the Canary Islands during ACE-2, several different aerosol models were derived that incorporated a dependence of optical properties on the aerosol hygroscopicity, γ . Parameterizations were then derived that incorporated this dependence into the upwelling radiance detected by the Terra sensors MODIS and MISR. It was found that the MODIS near infrared bands are sensitive to models with different γ 's whereas the visible bands of both MODIS and MISR are not. On this basis, and with the assumption that estimations of ambient RH were (or would be) available, a modification to the operational MODIS aerosol algorithm was implemented to retrieve γ in addition to the standard aerosol products. It was found that for certain very specific conditions, a successful retrieval of γ could be achieved. However, in general, a retrieval was not possible. To explore this issue further, a Principle Component Analysis was performed on a correlation matrix made up of spectral measurements of radiation and RH. Between two and four principle components could be extracted (depending on selection criteria such as eigenvalue thresholds and the domain of aerosol characteristics examined) that accounted for more than 99% of the observed variance and that one of them was always uniquely associated with RH. Hence, at least in principle, successful retrieval of γ should be generally possible.

The retrieval of aerosol dry mass and cloud condensation nucleus (CCN) concentration were also studied. Comparison was made between MODIS operational products and an alternative method. This alternative method was essentially a parameterization to derive the columnar dry aerosol mass and columnar CCN concentration from the primary MODIS aerosol products. The method relies on the scaling between columnar mass and optical depth with a proportionality constant dependent on MODIS-derived values for the effective radius and the contribution of the accumulation mode radiance to the total radiance, ambient RH, and an assumed constant aerosol composition (for the retrieval scene). By comparing with in-situ data from ACE-2 and TARFOX, it was found that retrievals for relatively non-hygroscopic aerosols (such as dust) were significantly improved using the new method. However, for very hygroscopic aerosols in high humidity conditions there was no significant improvement, mainly due to current inability to take into account variability in vertical profiles of RH. Two detailed comparisons of dry aerosol mass and CCN concentration retrieval via the standard MODIS and new methods were undertaken using MAS data. Both methods yield dry mass values within an order of magnitude of in-situ measurements but the new method is somewhat closer to actual values. Similar results were obtained for CCN retrieval. The new method has also been exercised with actual MODIS data, producing plausible, though not yet validated, results.

PUBLICATIONS:

Two publications have been based on the work discussed here. The first, (Gasso et al, 2000), due to an oversight, did not acknowledge this particular grant but was in fact partially supported by it so I mention it here for your information. The other publication (Gasso and Hegg, 2002) did acknowledge this grant.

Gasso, S., D.A. Hegg, D.S. Covert, D. Collins, K.J. Noone et al., Influence of humidity on the aerosol scattering coefficient and its effect on upwelling radiance during ACE-2. *Tellus*, 52B(2), 546-567, 2000.

Gasso, S. and D.A. Hegg, On the retrieval of columnar aerosol mass and CCN concentration by MODIS, *J. Geophys. Res.*, In Press, 2002.