Progress Report and Final report to NASA for

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Quantifying the Indirect Radiative Forcing of Sulfate Aerosol by a Hybrid Technique

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Third Year Progress Report:

During the third year of the project we carried out two parallel lines of work. The first was a more detailed study of a meteorological situation in which one would expect the Twomey effect of indirect radiative forcing by aerosol to be detectable. This was a follow on to the study reported in a publication to appear in the special GACP issue of JAS (Harshvardhan et al. 2002). At the same time, we have been investigating the limits to which meaningful cloud microphysical properties can be obtained from remotely sensed satellite data.

The Twomey effect of enhanced cloud droplet concentration, optical depth, and albedo due to anthropogenic aerosols is thought to contribute substantially to radiative forcing of climate change over the industrial period. However, present model-based estimates of this indirect forcing are highly uncertain. Satellite-based measurements provide global or near-global coverage of this effect, but previous efforts to identify and quantify enhancement of cloud albedo due to anthropogenic aerosols in satellite observations have been limited, largely because of strong dependence of albedo on cloud liquid water path (LWP), which is inherently highly variable. We examined satellite-derived cloud radiative properties over a one-week episode for which a chemical transport and transformation model indicates that sulfate aerosol in a remote area of the North Atlantic experienced a substantial excursion due to transport from Northern Europe. Despite the absence of discernible dependence of optical depth or albedo on modeled sulfate loading, examination of the dependence of these quantities on LWP readily permitted detection and quantification of increases correlated with sulfate loading which are otherwise masked by variability of LWP, demonstrating brightening of clouds due to the Twomey effect on a synoptic scale. Median cloud-top spherical albedo was enhanced over the episode, relative to the unperturbed base case for the same LWP distribution, by 0.04 to 0.15. The maximum change in albedo occurs at intermediate values of the liquid water path, as shown in Figure 1. At smaller values, the albedo is small and so is the change. At larger values of LWP, the albedo gets saturated and additional increase in cloud optical depth has a smaller effect.

The second study carried out during 2000-01 was a more detailed cloud microphysical study of maritime low-level clouds in the North Atlantic on 3 April 1987. The region is shown in the visible and infrared images of Figures 2 and 3, respectively, marked by the letters A through H, which identify 2.5° × 2.5° lat/long boxes. We concentrated on Box C for which Figures 4(a) and 4(b) show AVHRR GAC pixel statistics. The cluster of points at the foot of the "arch" represent cloud tops for pixels that are essentially completely cloud covered (Coakley and Bretherton 1982). Figures 4(c) and 4(d) show optical depth and effective radius retrievals for these cloud filled pixels (black) and also for partially filled pixels (gray). The figures illustrate the biases that affect retrievals of cloud microphysical properties from coarse resolution satellite measurements. Inclusion of all pixels will grossly overestimate the population of thin clouds and also affect the droplet distribution.

The panels in Figure 5 show scatter plots for the same region showing retrieved pixel effective radius for those pixels that have passed the cloud-filling criterion. Our interpretation of Figure 5 is that it shows the evolution of cloud top effective radius as parcels ascend to different levels. Figure 6 is a similar figure showing the retrieved optical depths. Recently, Brenguier et al. (2000) have shown that in an adiabatic stratified cloud, the optical depth, τ, and effective radius, r_e, obey the following relationship:

$$\tau \propto r_e^5. \quad (1)$$

The relationship also holds in non-adiabatic homogeneously mixed clouds (Szczodrak et al. 2001). Figure 7 shows the r_e vs. τ relationship on a log-log plot and the best-fit slope, b, for Boxes B, C, F, G and H. The slope and uncertainty so obtained suggests that one can use the adiabatic stratified or homogeneously mixed model to estimate other cloud microphysical quantities, such as the liquid water and cloud droplet concentration.

The integrated liquid water path (LWP), W, is related to the optical depth and effective radius at cloud-top by the following relationship (Szczodrak et al. 2001):

$$W \approx 5 \rho_w r_e \tau / 9, \quad (2)$$

where ρ_w is the density of liquid water. Figure 8 shows W for the various pixels plotted against the cloud top temperature, which acts as a surrogate for altitude. Superimposed are lines depicting the adiabatic LWP (solid) and 50% of adiabatic LWP (dashed). The cloud base has been estimated from the NCEP reanalysis (Kalnay et al. 1996) using conditions at 1000 hPa. The boxes for which there is a significant population appear to be filled with clouds that have LWP ranging from almost zero to about 40% of the adiabatic value.

The final cloud microphysical quantity, number concentration, has been obtained for Boxes B, C, F, G and H. The number concentration, N, is obtained by first estimating the column cloud droplet concentration, N_c, (Han et al. 1998) and using the location of the cloud base mentioned earlier to calculate the height of the ascending parcel above the base. Histograms are shown in Figure 9. The median concentration is around 30 cm^{-3}, typical of unpolluted marine clouds in the North Atlantic (Brenguier et al. 2000).

Three-Year Summary of Work:

The project involved the melding of satellite remote sensing and chemistry/transport modeling. A key feature of the effort was the use of the Brookhaven National Laboratory (BNL) chemical transport and transformation model, described later. Output from this model was used in conjunction with a well-established procedure of inferring cloud microphysical properties from visible and near-infrared AVHRR radiances (Nakajima and Nakajima 1995). The first application of this hybrid method has been reported in a

publication to appear in the special GACP issue of JAS (Harshvardhan et al. 2002). The results are summarized below.

We used results from a chemical transport model driven by the output of a numerical weather prediction model, to identify an incursion of sulfate-laden air from the European continent over mid North Atlantic under the influence of a cut-off low-pressure system during 2-8 April 1987. Advanced Very High Resolution Radiometer (AVHRR) measurements of visible and near-infrared radiance were used to infer microphysical properties of low-altitude (T = 260-275 K) maritime clouds over the course of the event. Examination of the cloud optical depth, drop radius, and drop number concentration on the high- and low-sulfate days allowed identification of the increase in cloud droplet number concentration and decrease in cloud drop radius associated with the sulfate incursion. These observations are consistent with the Twomey mechanism of indirect radiative forcing of climate by aerosols.

In particular, we were able to estimate the adiabatic cloud droplet number concentration by identifying the cloud base from meteorological analyses and assuming that cloudy pixels represent the tops of air parcels ascending to different levels. The adiabatic cloud droplet number concentration ranged from 65 – 95 cm^{-3} on days when the chemistry model indicated that little sulfate was present, and was 300 cm^{-3} during the peak of the sulfate incursion. Mirroring this difference, it was found that the cloud drop effective radius ranged up to 20 μm on the low-sulfate days but was generally less than 12 μm on the high-sulfate days. The sulfate concentration in the lowest 2 km on 2 April, before the incursion, was 4 nmol m^{-3} and increased to 25-30 nmol m^{-3} on 5 April. The corresponding increase in number concentration is quantitatively similar to the model based on the Hegg et al. (1993) measurements quoted by Boucher and Lohmann (1995).

A key feature of this project has been the use of a chemistry model. The Brookhaven National Laboratory (BNL) chemical transport and transformation model, the **G**lobal **Ch**emistry **M**odel driven by **O**bservation-derived synoptic meteorological data (GChM-O), has been described in Benkovitz *et al*. (1994) and Benkovitz and Schwartz (1997). The model, a three-dimensional Eulerian transport and transformation model is driven by synoptic meteorological data assimilated from observations. The model represents emissions of the several sulfur species, chemical conversion of SO_2 to sulfate by H_2O_2 and O_3 in the aqueous phase and by OH in the gas phase, chemical conversion of DMS to SO_2 and MSA by OH, and removal by wet and dry deposition. The model tracks the several sulfur species according to the source type and region and conversion mechanism.

In using the model output to provide sulfate concentrations, we have, in effect, simulated a quantity that would be measured in a field experiment. We believe that this type of technique can have wide applications in determining global aerosol effects related to indirect radiative forcing. Details of our findings can be found in Harshvardhan et al. (2002), also available from http://gacp.giss.nasa.gov/specialIssue/harshvardhan.pdf, and will not be repeated here.

REFERENCES

Benkovitz, C.M., and Schwartz, S.E. Evaluation of modeled sulfate and SO_2 over North America and Europe for four seasonal months in 1986-87. J. Geophys. Res., **102**, 25,305-25,338 (1997).

Benkovitz, C.M., Berkowitz, C.M., Easter, R.C., Nemesure, S., Wagener, R., and Schwartz, S.E. Sulfate over the North Atlantic and adjacent continental regions: Evaluation for October and November 1986 using a three-dimensional model driven by observation-derived meteorology. J. Geophys. Res., **99**, 20,725-20,756 (1994).

Boucher, O., and Lohmann, U. The sulfate – CCN-cloud albedo effect: A sensitivity study with two general circulation models. Tellus, **47B**, 281-300. (1995).

Brasseur, G., Hauglustaine, D., Walters, S., Rasch, P., Müller, J., Granier, G., and Tie, X.X. MOZART: A global chemical transport model for ozone and related chemical tracers. 1. Model description. J. Geophys. Res., **103**, 28,265-28,289 (1998).

Brenguier, J.-L., Pawlowska, H., Schüller, L., Preusker, R., and Fischer, J. Radiative properties of boundary layer clouds: droplet effective radius versus number concentration. J. Atmos. Sci., **57**, 803-821 (2000).

Coakley, J.A., Jr., and Bretherton, F.P. Cloud cover from high-resolution scanner data: detecting and allowing for partially filled fields of view. J. Geophys. Res., **87**, 4917-4932 (1982).

Coakley, J.A., Jr., and Davies, R. The effect of cloud sides on reflected solar radiation as deduced from satellite observations. J. Atmos. Sc., **43**, 1025-1035 (1986).

Han, Q., Rossow, W.B., Chou, J, and Welch, R.M. Global variation of column droplet concentration in low-level clouds. Geophys. Res. Lett., **25**, 1419-1422 (1998).

Harshvardhan, Schwartz, S. E., Benkovitz, C.M., and Guo, G. Aerosol influence on cloud microphysics examined by satellite measurements and chemical transport modeling. J. Atmos. Sci., 2002 (to appear).

Hegg, D.A., Ferek, R.J., and Hobbs, P.V. Light scattering and cloud condensation nucleus activity of sulfate aerosol measured over the northeast Atlantic Ocean. J. Geophys. Res., **98**, 14,887-14,894 (1993).

Kalnay, E. et al. The NCEP/NCAR 40-year reanalysis project. Bull. Amer. Meteor. Soc., **77**, 437-471 (1996).

Nakajima, T.Y., and Nakajima, T. Wide-area determination of cloud microphysical properties from NOAA AVHRR measurements for FIRE and ASTEX regions. J. Atmos. Sci., **52**, 4043-4059 (1995).

Szczodrak, M, Austin, P.H., and Krummel, P.B. Variability of optical depth and effective radius in marine stratocumulus clouds. J. Atmos. Sci., **59**, 2912-2926 (2001).

GACP Bibliography:

Benkovitz, C.M., Schwartz, S.E., Mubaraki, M.A., Miller, M.A., and Bates, T. S. Simulating sulfur for the ACE-2 experiment: Preliminary results. Sixth Scientific Conference of the International Global Atmospheric Chemistry Project (IGAC), September 13-17, 1999, Bologna, Italy.

Harshvardhan, Wei, D., Green, R., Schwartz, S. E, and Benkovitz, C.M. An investigation of the effect of sulfate on cloud microphysics using a chemistry/transport model. A Millennium Symposium on Atmospheric Chemistry: Past, Present, and Future of Atmospheric Chemistry, Albuquerque, NM, Amer. Met. Soc., 152-159 (2001).

Harshvardhan, Schwartz S. E., Benkovitz, C.M., and Guo, G. Aerosol influence on cloud microphysics examined by satellite measurements and chemical transport modeling. J. Atmos. Sci., 2002 (to appear).

Schwartz, S. E, Benkovitz, C.M., and Harshvardhan. Aerosol influence on cloud optical depth and albedo over the North Atlantic shown by satellite measurements and chemical transport modeling. Workshop on Photooxidants, Particles, and Haze across the Arctic and North Atlantic: Transport Observations and Models, Center for International Earth Science Information Network, Lamont-Doherty Earth Observatory, Lamont-Doherty Earth Observatory, Palisades, New York, June 12-15, 2001.

Schwartz S. E, Benkovitz, C.M., and Harshvardhan. Aerosol Influence on Cloud Optical Depth and Albedo Over the North Atlantic Shown by Satellite Measurements and Chemical Transport Modeling, American Geophysical Union, Fall Meeting, 2001, submitted.

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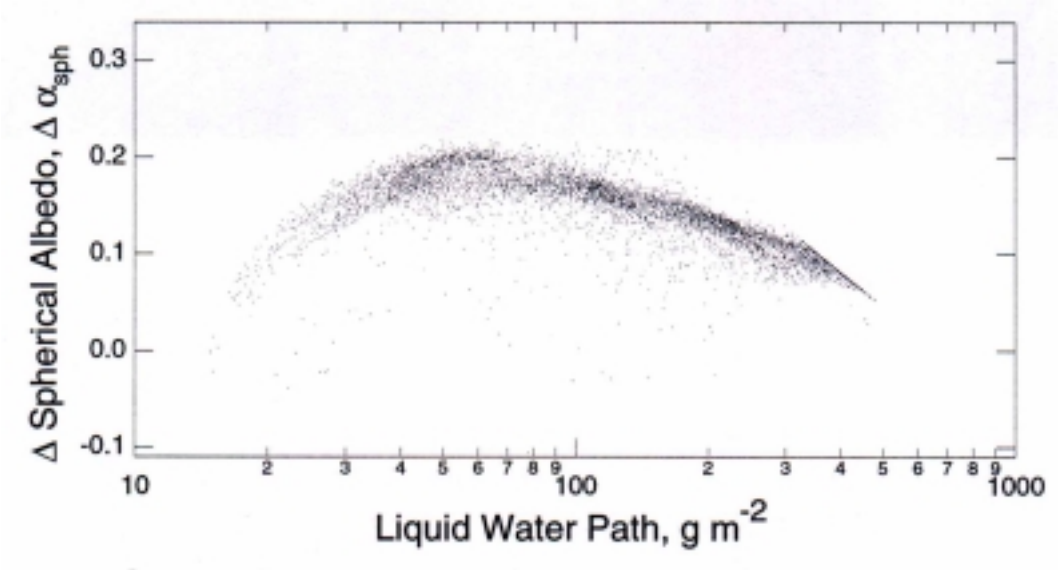

Figure 1. Change in spherical albedo from 2 April to 5 April 1987 at constant LWP.

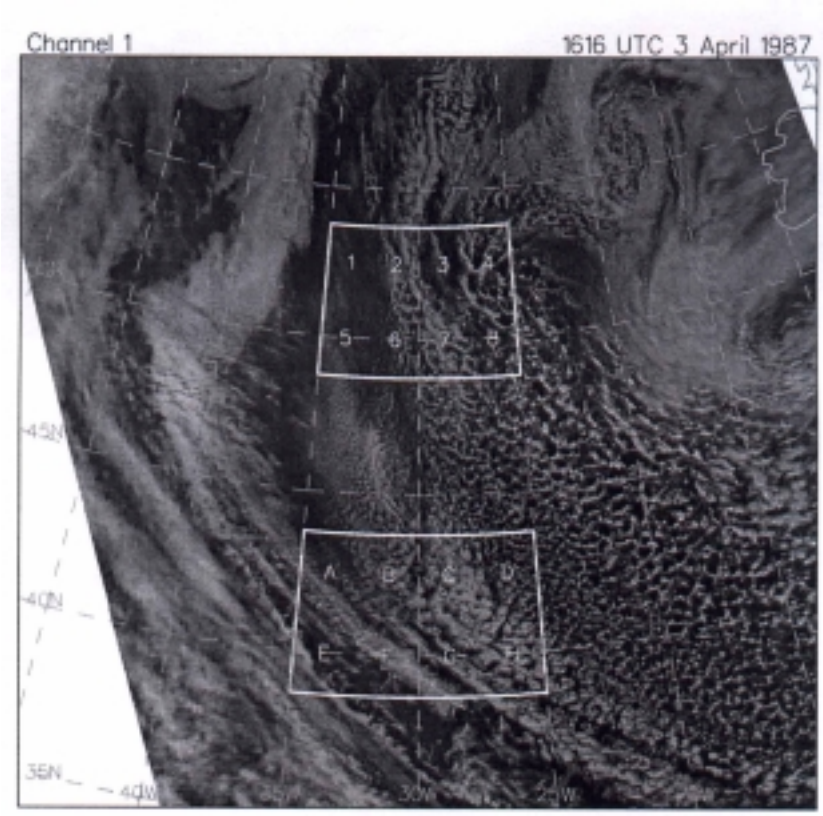

Figure 2. AVHRR Channel 1 image of study area.

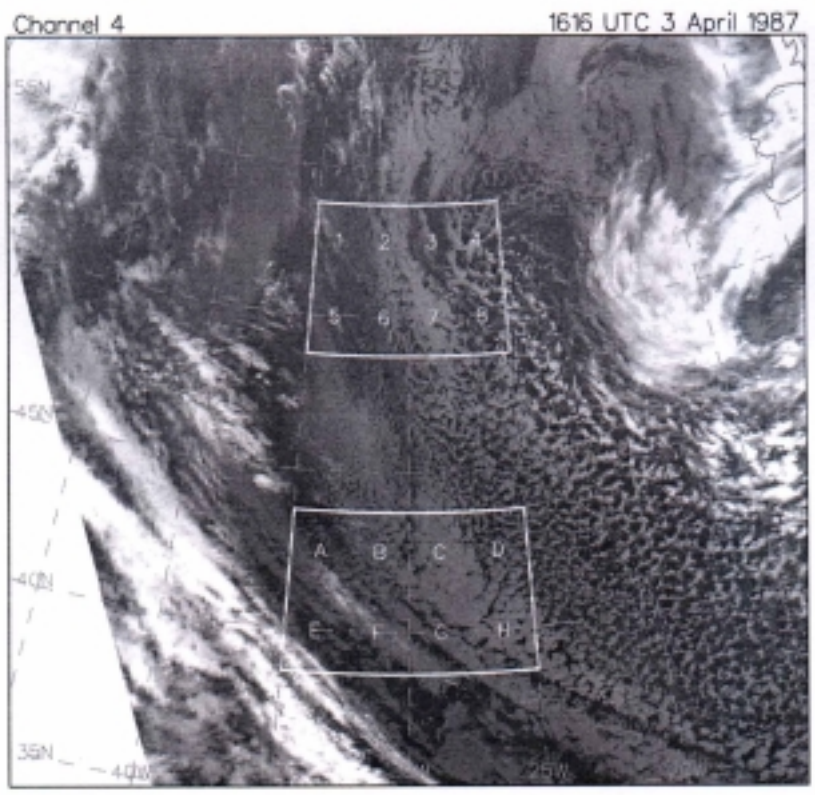

Figure 3. AVHRR Channel 4 image of study area.

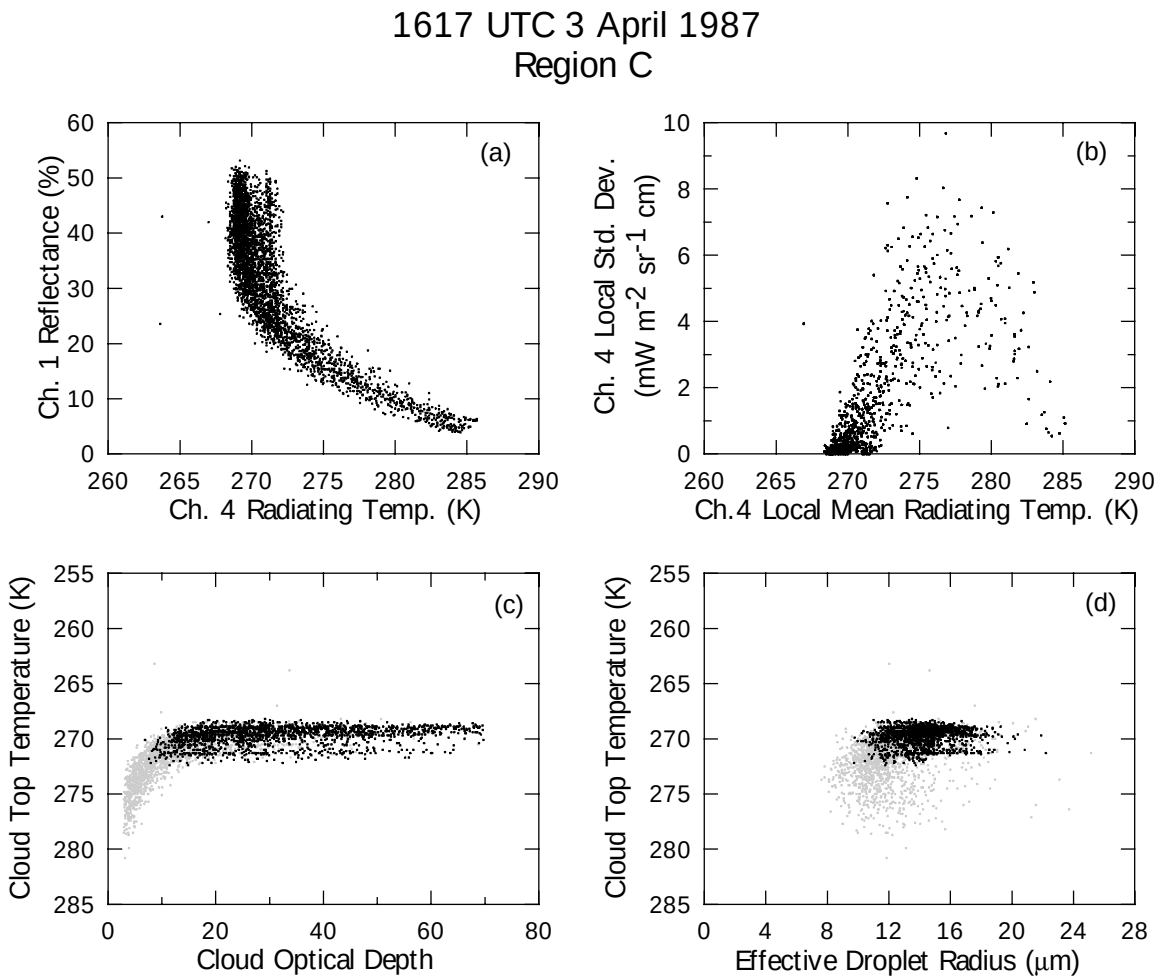

Figure 4. (a) Ch. 1 reflectance versus Ch. 4 radiance for AVHRR GAC pixels in Box C; (b) Local standard deviation of Ch. 4 radiance versus local mean radiance of Ch. 4; (c) Cloud optical depth versus cloud top temperature for cloud-filled pixels (black) and partly filled pixels (gray); (d) As in (c) but for effective radius.

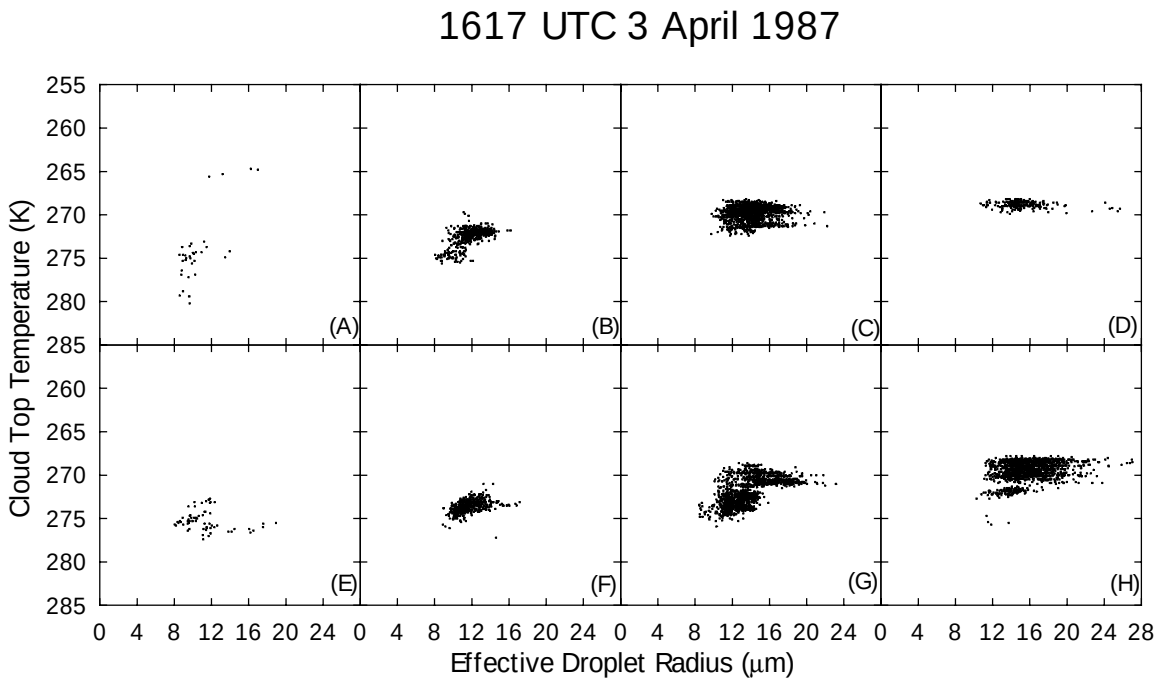

Figure 5. Effective droplet radius versus cloud top temperature for Boxes A, B, C, D, E, F, G and H. Only cloud filled pixels have been plotted.

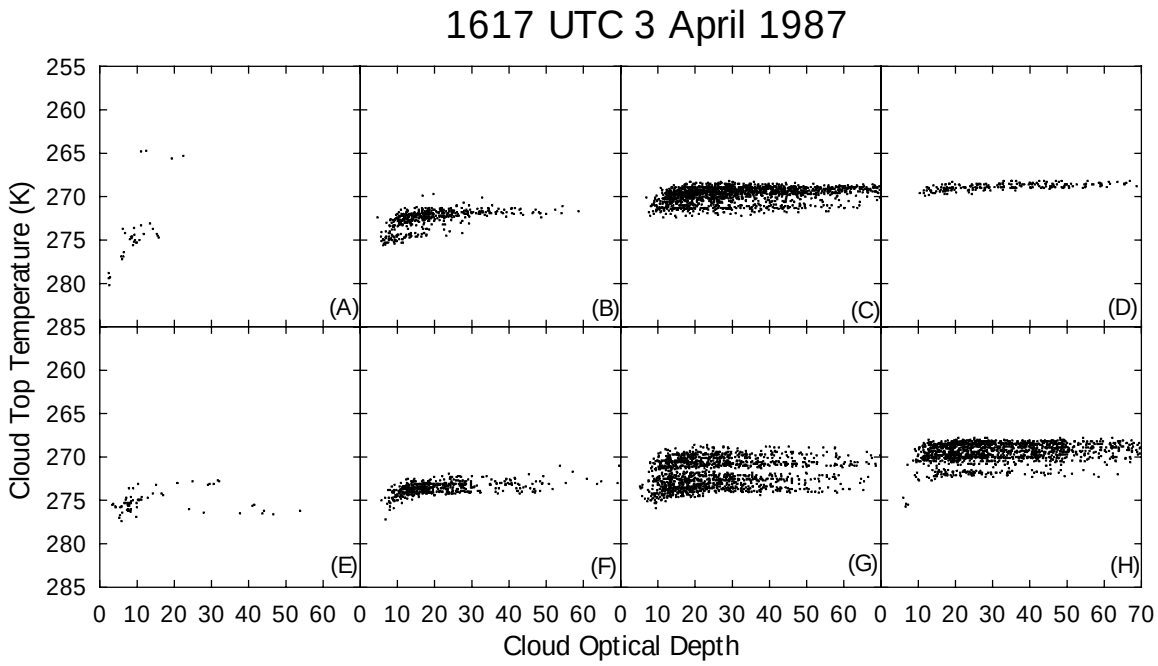

Figure 6. As in figure 5 but for optical depth.

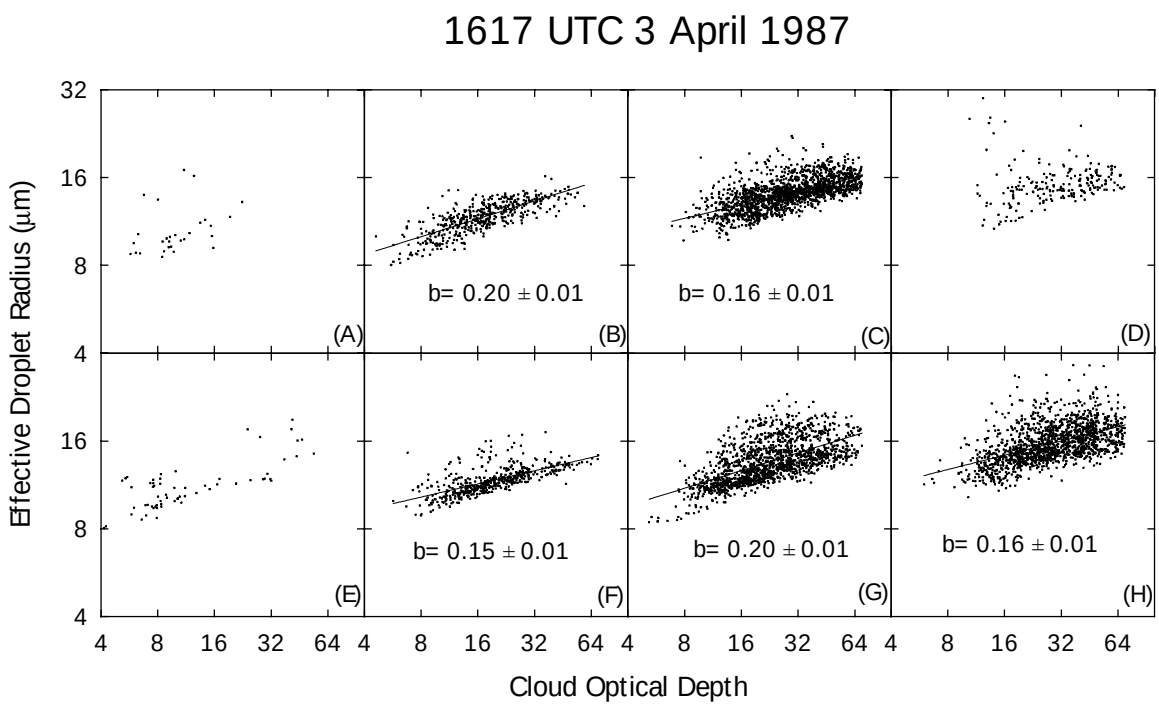

Figure 7. Scatter plot of effective radius and optical depth from figure 6. The slope and its uncertainty are marked on the panels for boxes with sufficient data.

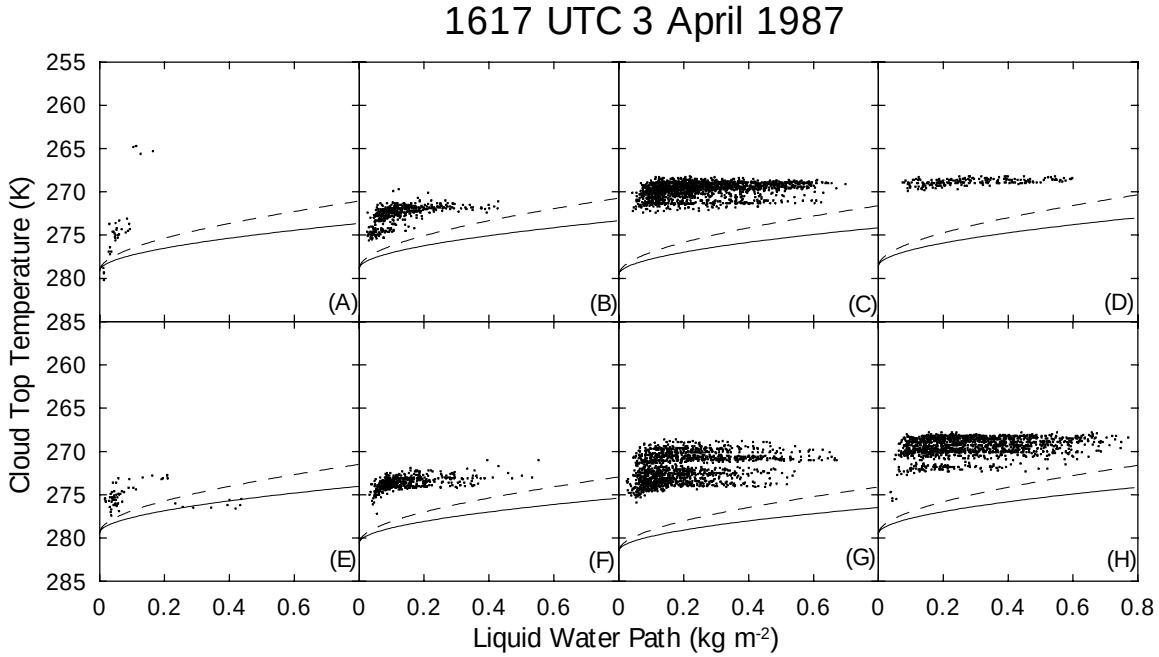

Figure 8. Calculated liquid water path for each pixel plotted versus the cloud top temperature. Also shown are adiabatic and 50% of adiabatic LWP.

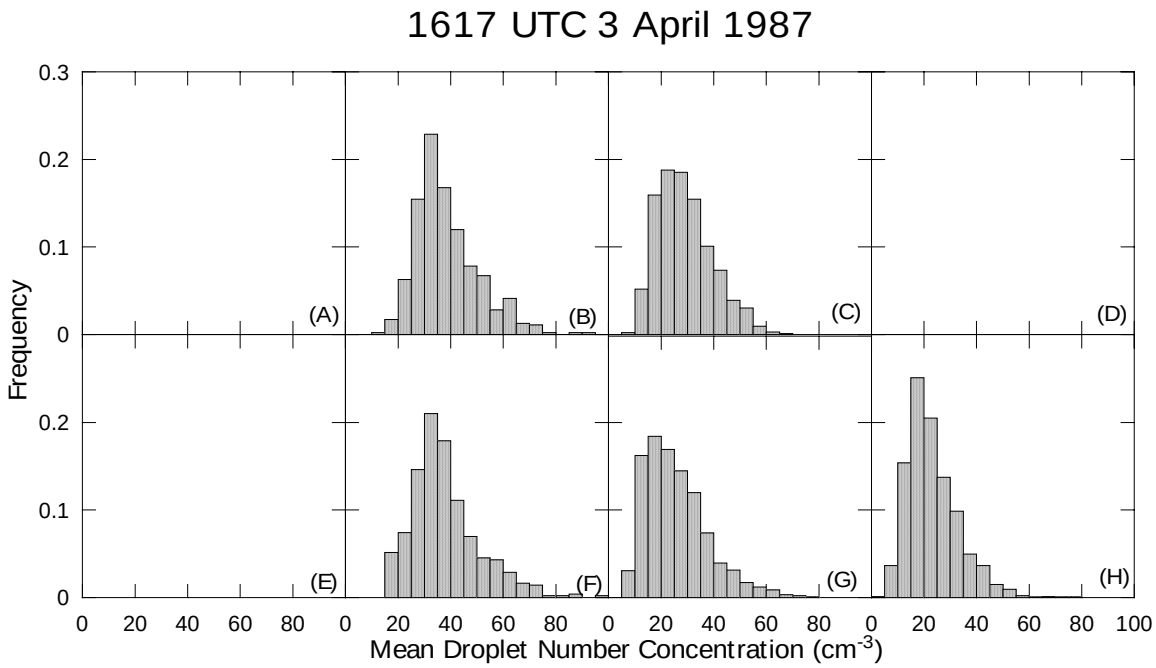

Figure 9. Histogram of cloud droplet number concentration for Boxes B, C, F, G and H.