Progress Report to NASA for

Grant NAG5 – 7727 entitled

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

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Second Year Progress Report:

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 previously (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.

Recent model developments are indicated schematically in Figure 1. The developments denoted model Version 2 incorporate representation of reactions in nonprecipitating clouds and improved representation of the oxidant chemistry. Aqueous-phase oxidation proceeds in all clouds where the liquid water volume fraction exceeds 10^{-9}. The cloud water pH is estimated assuming that all sulfate is present as ammonium bisulfate plus the small contribution of MSA, but is constrained to be between 2 and 5.6. The aqueous-phase oxidation of SO_2 by H_2O_2 and O_3 is explicitly calculated based on kinetic mechanisms. Representative daily average mixing ratios of HO_2, H_2O_2, O_3, and OH are obtained from the MOZART model of Brasseur and colleagues [Brasseur *et al.*, 1998]. H_2O_2 is depleted by reaction with SO_2; gas phase regeneration of H_2O_2 from HO_2 is explicitly represented, with the limitation that the calculated H_2O_2 mixing ratio not exceed the climatological value from the MOZART model. Additionally, the model incorporates estimation of time- and location-dependent oceanic emissions of DMS, and a variable mixed-layer height for vertical diffusivity. The model domain has been extended to the entire hemisphere (equator to 81° latitude). Vertical resolution is now enhanced to 27 levels from the surface to 100 hPa, and horizontal resolution has been enhanced slightly to 1° latitude × 1° longitude (approximately 111 × 111 km at the equator and 56 × 111 km at 60° latitude).

Output from the earlier version of the model has been used to identify a time period for investigating the indirect radiative effect of anthropogenic aerosols in the North Atlantic. The period chosen is April 1987 and Figure 2 shows the column amount of sulfate averaged over a particular grid area of 5° latitude × 5° longitude, as simulated every six hours by GChM-O. Time lapse loops of the output show that a mass of relatively polluted air from Northern Europe invaded the area from the east after April 3 and the region became relatively clean by April 8. Although there is more synoptic variability in sulfate column amount for the rest of the month, we have chosen the period April 2 – 8 for study. Neighboring areas also show the same pattern of influx.

In order to study the effect of sulfate aerosols on cloud microphysics we first identified cloudy regions in the study area that were essentially cloud covered. This is important in order to minimize the uncertainty in retrieving cloud properties from reflected visible radiances. This was done using the spatial coherence method of Coakley and Bretherton (1982). Since we used AVHRR GAC data for retrieval we used the technique described by Coakley and Davies (1986). Figure 3 shows an example for April 2, 1987. Pixels at the "foot" of the arch having brightness temperatures between 260 and 263 K were selected for further analysis. A further screening was applied to eliminate pixels having Ch. 1 (visible) reflectances less than 0.1. These two steps give us some confidence that the pixels chosen are most probably completely cloud covered although there is still inhomogeneity at the sub-pixel level.

Figures 4 and 5 show the retrieved effective radius and optical depth, respectively, for the screened pixels. We have used the two-channel scheme of Nakajima and Nakajima (1995) to obtain these results. There is an upper cutoff of 30 µm in effective radius beyond which retrievals are not made. Figures 6 and 7 show the derived quantities Liquid Water Path (LWP) and column concentration (N_C) following Han et al. (1998). These results are for the cleanest day during the week just prior to the incursion of sulfate rich air into the area. Similar analyses have been made for subsequent days and other areas in the North Atlantic. Results are being prepared for publication and presentation at the annual meeting of the American Meteorological Society in January 2001.

Third Year Statement of Work:

Future work involves exercising Version 2 of GChM-O for the period of the ACE-2 (Aerosol Characterization Experiment) field intensive campaigns (June 15 – July 25, 1997). For these runs, sulfur sources include volcanic emissions specific to the times and locations of the simulations and it is possible to follow the plumes of volcanic SO_2 and sulfate (maintained as separate variables in the model).

Preliminary results for ACE-2 suggest that volcanic emissions may be contributing to sulfate loading in substantially greater proportion than their contribution to sulfur emissions. Of the sulfur emissions in the modeling domain 84% are contributed by anthropogenic sources, 12% by biogenic sources, and 4% by volcanos whereas analysis of the sulfate in the model domain at a specific time, June 26, 1997 at 0 UTC, shows the contributions to sulfate from these sources as 66%, 6%, and 28%, respectively. An initial report of this work has been presented (Benkovitz et al., 1999).

Regions and times of significantly different aerosol loading will be studied for effects on cloud microphysics. The study periods will include, in addition to April 1987, the three months of July 1986, October 1986 and June/July 1997. AVHRR GAC data for the 1986-87 time period has already been obtained. Data for 1997 will be obtained from the standard archive.

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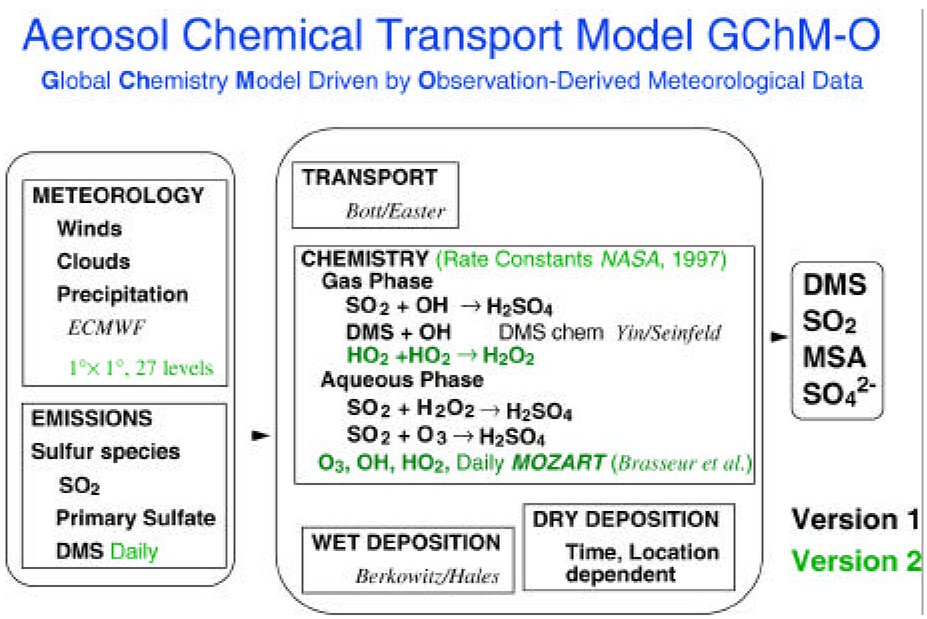

Figure 1. Key elements of the aerosol chemical transport model. Extensions beyond the original model are indicated in green (Version 2).

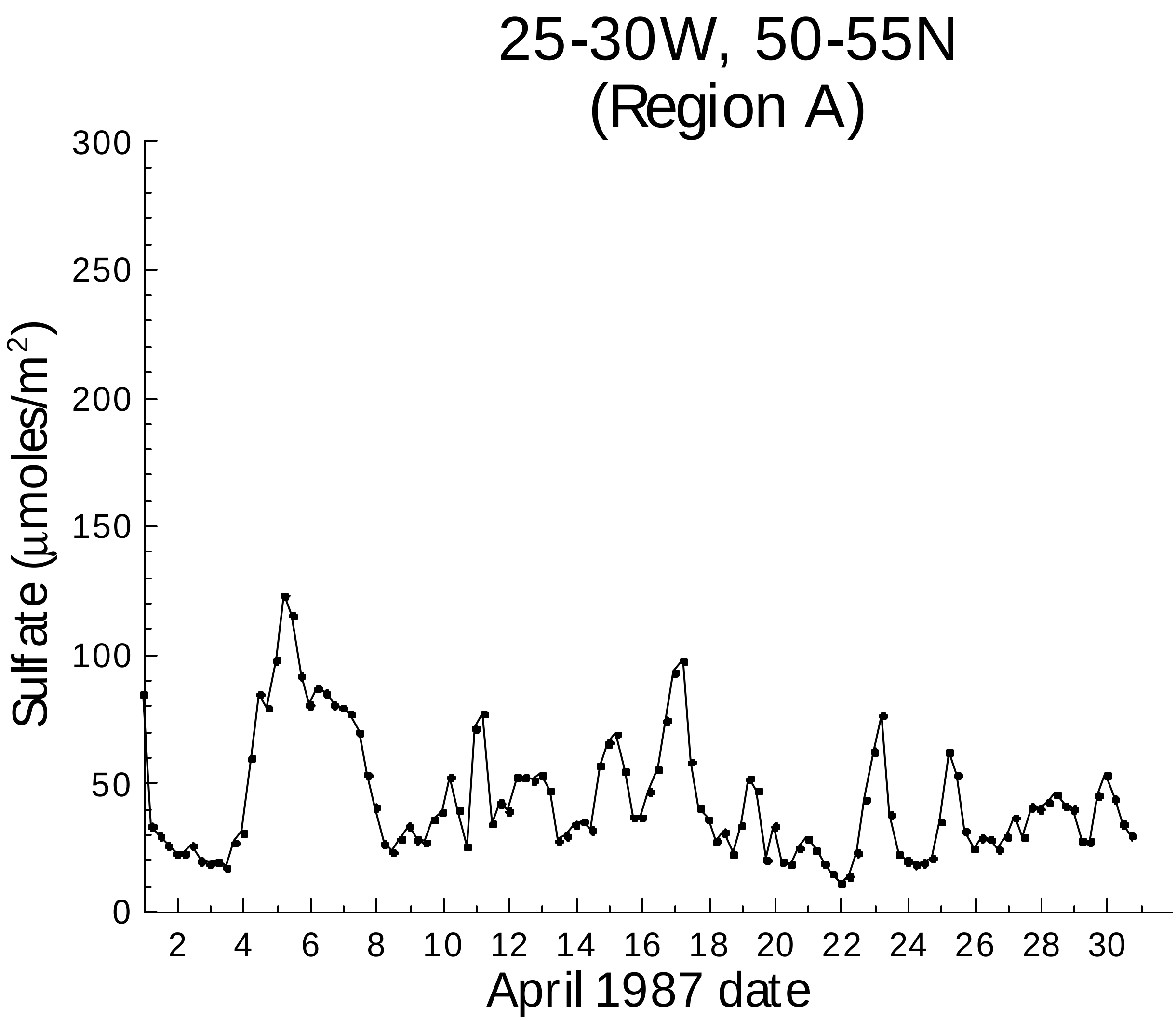

Figure 2. Column sulfate in micromoles per square meter averaged over a 5º × 5º grid box for the month of April 1987.

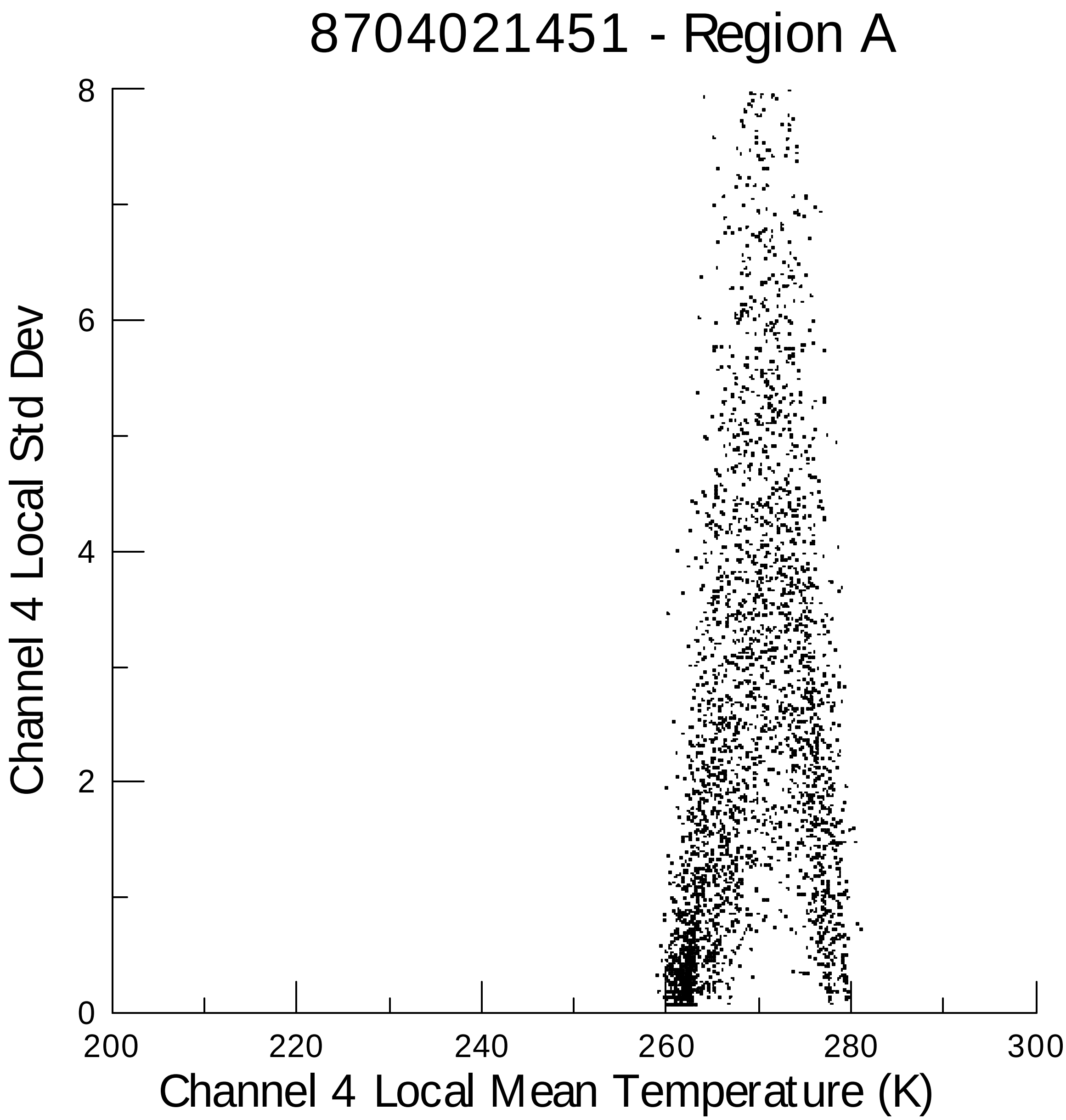

Figure 3. Channel 4 (11 μm) local mean brightness temperature and local standard deviation for 2 × 2 arrays of GAC data points constructed for region A on April 2, 1987.

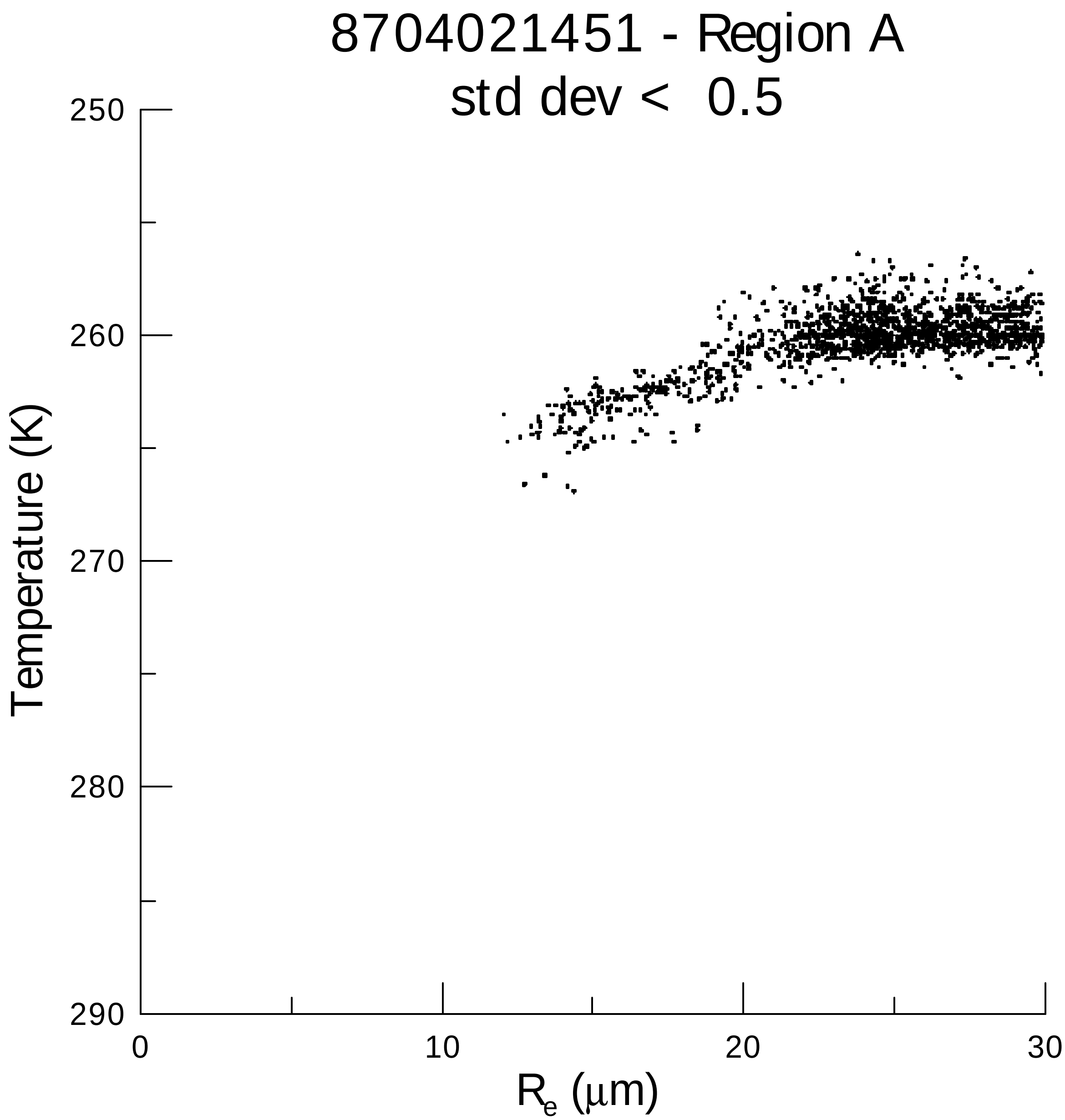

Figure 4. Scatter plot of droplet effective radius in micrometers for each pixel passing the complete cloud cover and reflectance threshold screening and Channel 4 brightness temperature.

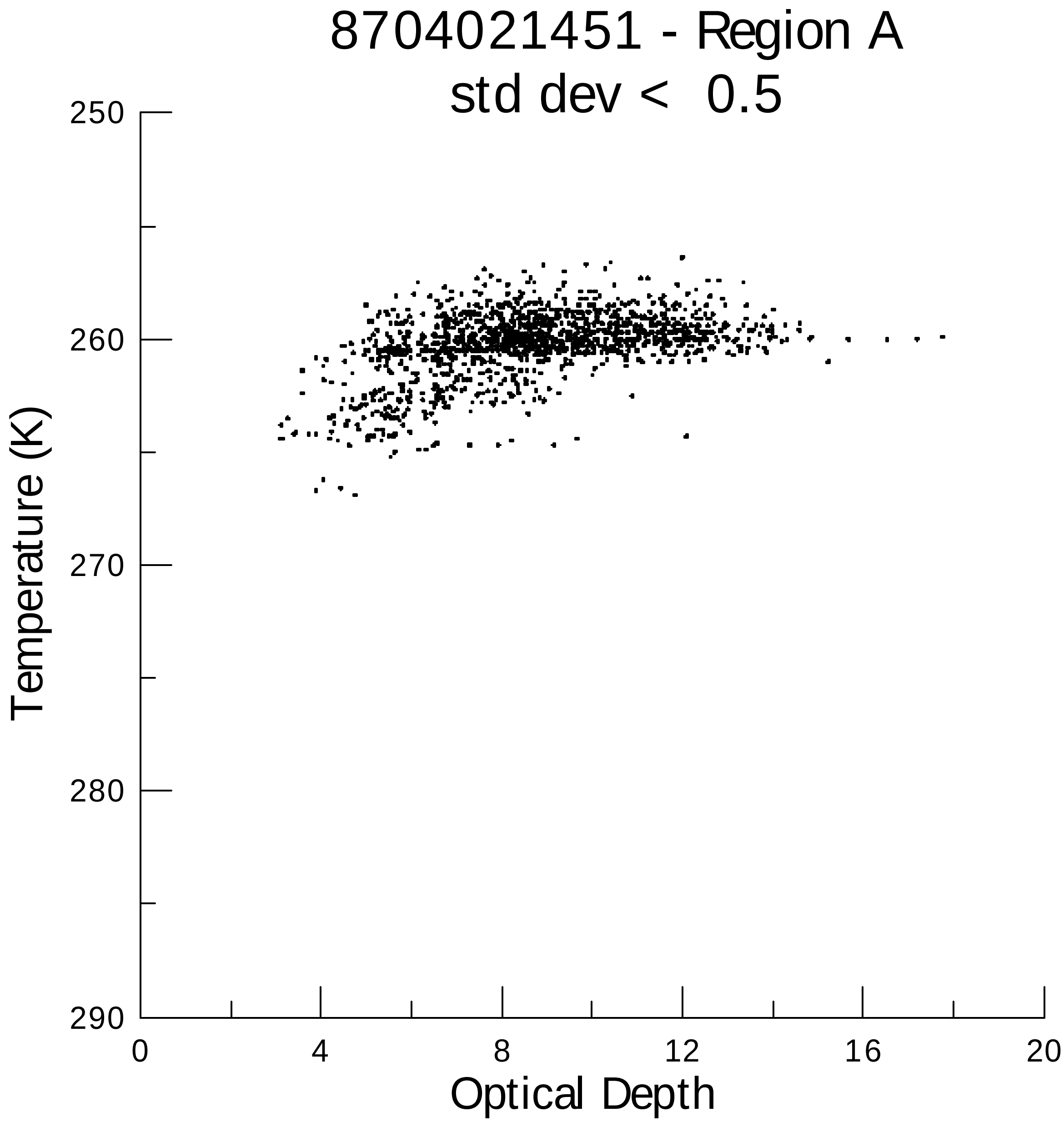

Figure 5. Scatter plot of cloud optical depth for each pixel passing the complete cloud cover and reflectance threshold screening and Channel 4 brightness temperature.

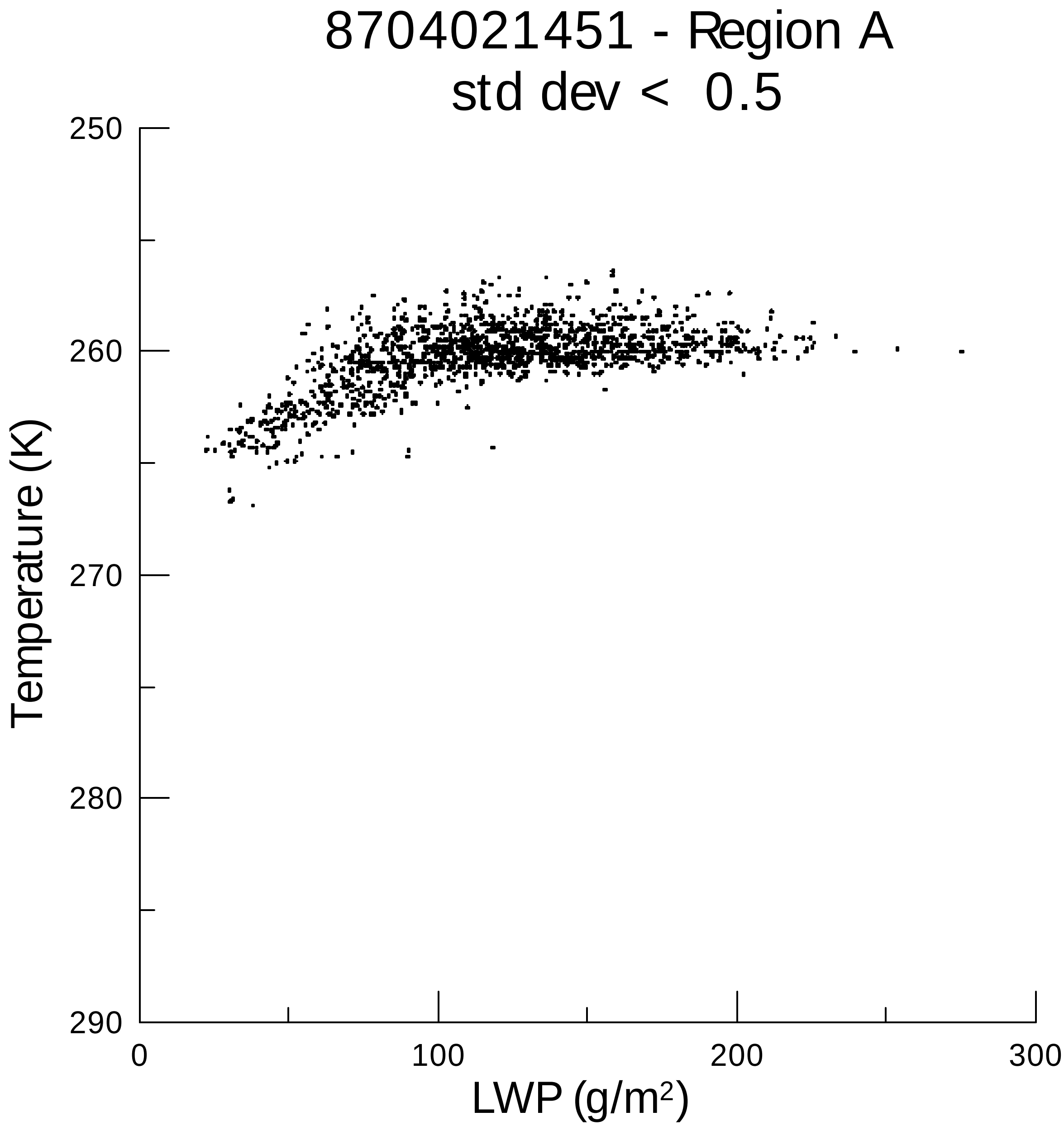

Figure 6. Scatter plot of derived liquid water path in grams per square meter for each pixel passing the complete cloud cover and reflectance threshold screening and Channel 4 brightness temperature.

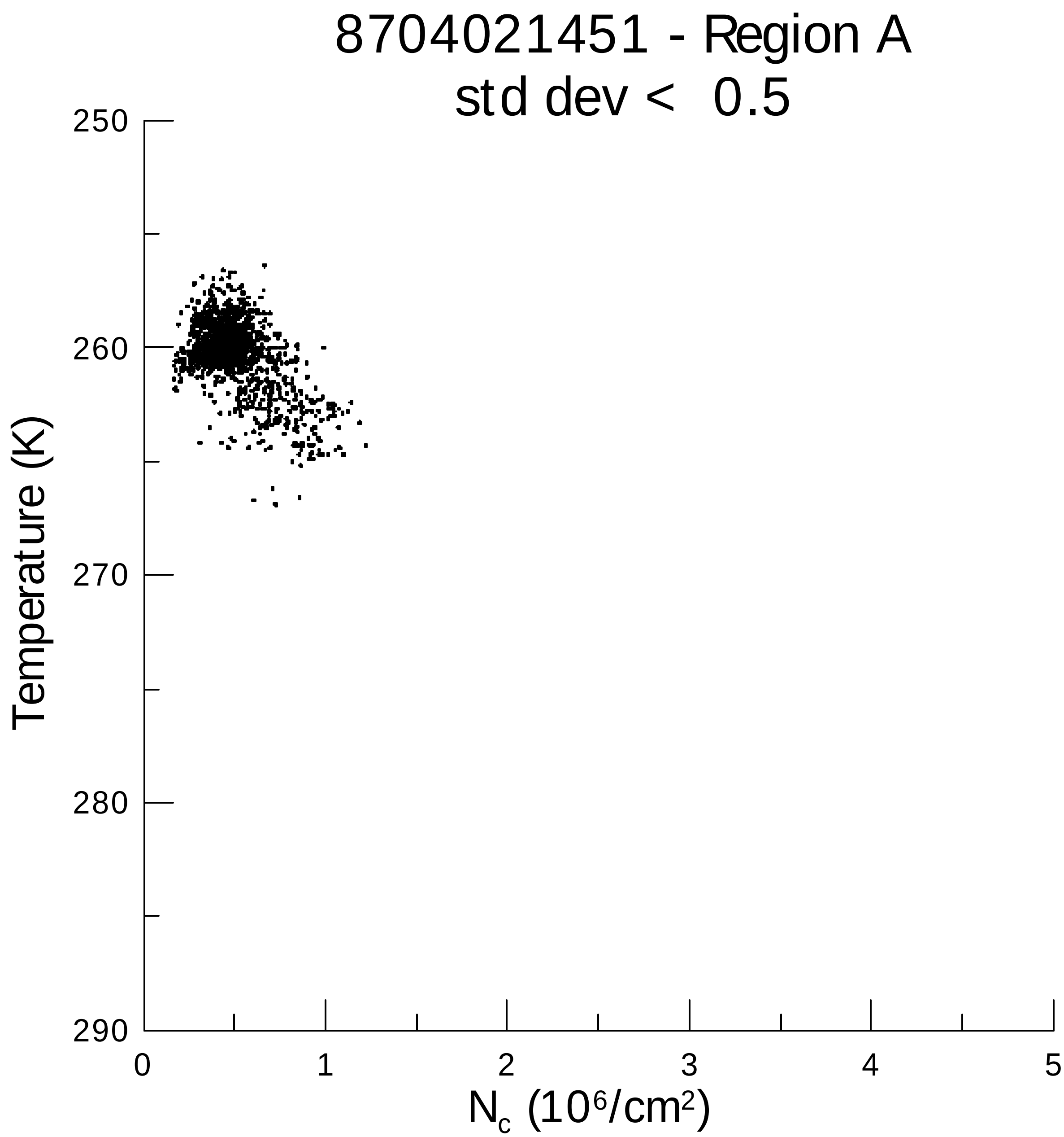

Figure 7. Scatter plot of derived column droplet concentration in 10^6 per square centimeter for each pixel passing the complete cloud cover and reflectance threshold screening and Channel 4 brightness temperature.