"CCN Spectra Measurements as an Active Tracer of Stratocumulus Mechanisms"

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A paper "Transport and Mixing Processes in the Lower Troposphere over the Ocean" was submitted to the Journal of Geophysical Research (J.G.R.). This was done in collaboration with Ilga Paluch and Donald Lenschow of the National Center for Atmospheric Research and Richard Pearson of NASA Ames. This manuscript concerned data obtained in the 1987 FIRE stratocumulus project which was a precursor of ASTEX. Comparisons and contrasts along these lines between the two projects will be most beneficial. This will be particularly useful since the FIRE regime was much more stratified than the ASTEX regime where stratocumulus clouds are much more common. This paper examined the consequences of the severe stratification of the atmosphere off the California coast.

In contrast to both of these regimes the HaRP project was conducted in marine cumulus clouds in the mid Pacific near Hawaii. Analysis of these data has been proceeding and a paper presenting the extensive set of CCN data is in preparation. This manuscript compares and contrasts the measurements about the marine stratus clouds of FIRE with the measurements near the cumulus clouds in HaRP. The CCN measurements in FIRE were recently published in J.G.R. (Hudson and Frisbie, 1991). An example of the vertical distribution of CCN is given in figure 1. In both regimes the concentration of CCN was higher above the cloud layers, that is above the marine inversion. In Hawaii there was usually not the layer of high concentrations just above the clouds (just above the inversion) as was seen in FIRE. This is probably because of the
longer distance from the continents. Measurements of lower concentrations within the boundary layer are consistent in both projects and appear to be due to coalescence scavenging which was in fact much more rapid in Hawaii due to the larger cloud droplets and a more active collision-coalescence process which resulted in substantial precipitation. In fact extremely low particle concentrations were found very close to some of the clouds, this was often the case just above the cloud tops where concentrations of less than 5 cm⁻³ were often observed (Fig. 1).

In both projects very high total particle concentrations (CN - condensation nuclei) were usually found within the clouds. Unlike FIRE, high concentrations of CCN were also found within the clouds. We believe that this gives further evidence that the in cloud aircraft measurements are artifacts as we suggested in the earlier paper (Hudson and Frisbie, 1991). The fact that the CCN as well as the CN concentration was so affected in HaRP is probably due to the larger cloud drops in Hawaii which produce even more splashing in the inlet system. Even further evidence of this point is revealed by the fact that very high CCN and CN concentrations were also found in the measurements obtained in precipitation below the HaRP clouds. Apparently the even larger precipitation drops result in yet greater splashing and an even greater artifact. These interpretations are in sharp contrast with those of the University of Washington group (e.g. Hegg et al., 1990, 1991) which has made similar aerosol observations in and near maritime clouds. They maintain that the high particle concentrations in and near clouds are a result of particle production processes associated with phytoplankton emissions of dimethylsulfide (DMS). We believe that our measurements have fine enough resolution that we can distinguish between the in cloud and out of

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Figure 1. Typical vertical distribution of CCN (dashed line) in Harp. Cloud droplet concentrations are denoted by the solid line (FSSP concentration). Note the very low CCN concentrations near cloud, the high concentrations in cloud (artifacts), and the higher concentrations above cloud.
cloud measurements. Since we find lower and sometimes extremely low concentrations just outside of the clouds we suggest that in-cloud particle production seems quite unlikely because some of the newly produced particles should spill out into the out of cloud environment. The higher concentrations which we do find above cloud especially off the California coast are clearly separated (at least in our measurements) from the clouds and they are associated with high ozone concentrations. Those higher concentrations of particles and ozone are more likely to be the result of continental and even polluted air.

Furthermore the consistently lower CCN and CN concentrations in the boundary layer are certainly not indicative of particle production processes. The lower concentrations are a result rather of cloud scavenging processes. Many have referred to this as nucleation scavenging. However this is not a good descriptive term since the nucleation process itself is reversible; when a droplet evaporates it yields back to the atmosphere nearly the same particle as were nucleated in the first place. However drops which have undergone the coalescence process upon evaporation yield nuclei which are the product of the nuclei of the coalesced drops. Collision-coalescence is the process of differential gravitational settling which results in larger drops overtaking and combining with smaller droplets. Therefore even without precipitation to the surface the aerosol scavenging occurs by coalescence. Liquid water clouds are in fact the only place where such gravitational coagulation of particles can occur. Lower CCN concentrations are known to produce lower cloud droplet concentrations. Measurements in these projects confirm this and go on to show that the lower droplet concentrations also induce larger drops which in turn
make coalescence, precipitation, and lower CCN concentrations. This cloud feedback process is the subject of a paper, "Cloud-CCN Feedback" (by J.G. Hudson) which has just been accepted for oral presentation at the International Conference on Clouds and Precipitation in Montreal, Canada, August 17-21, 1992. The mechanisms outlined here are very important in the global cycle of CCN.

Comparisons of the CCN spectra with cut sizes used in the counterflow virtual impactor (CVI) which was also a part of HaRP have been proceeding. These measurements were made periodically throughout HaRP during various cloud penetrations. The CVI is a discriminating sampler which allows only particles above certain cut sizes to enter; smaller cloud droplets and interstitial particles are rejected. The large cloud droplets which do enter the CVI are evaporated and then passed to the CCN spectrometer. This allows identification of the critical supersaturations ($S_c$) of the particles which had formed those cloud droplets. When these measurements are compared with ambient out of cloud CCN spectra one can determine which of the nuclei out of the available CCN spectrum actually formed droplets (Fig. 2). This then allows firm identification of the CCN of droplet formation. This example which was typical of most of the data so far analyzed shows a significant shift in the CCN spectrum when the CCN spectrum of the large cloud drops is considered. This shows that the larger nuclei seem to form the larger cloud droplets as would be the case for homogeneous mixing processes (Hudson and Rogers, 1986--as outlined in the original proposal). This information can be used to determine the relationship between cloud dynamics and cloud microphysics. The P.I. has had the opportunity to go to NCAR and meet with Cindy Twohy who operated the CVI in HaRP and will interpret the CVI results.
Figure 2(a). Ambient CCN spectrum observed in Hawaii. Note this is a unique differential spectrum which is only possible with the DRI instantaneous CCN spectrometer.

Figure 2(b). As figure (a) but measured downstream of the CVI which passed only cloud droplets above about 20 micrometers. This spectrum contains the nuclei which formed cloud droplets. Note that there is a considerable shift to lower Sc values as compared with (a).
We have been able to identify several periods when useful data were obtained and are currently working on the analysis of these data. We should be able to produce a publication concerning CCN-CVI comparisons this year.

During July we had the opportunity to put our equipment on board a research ship which cruised in the Pacific ocean out of San Diego in the same area where the FIRE project had taken place. This gave the opportunity to make longer term surface measurements of CCN. The object of the mission was to investigate ship trails. On July 13, 1991 our research ship actually passed through a ship trail. There was a sharp two order of magnitude increase in the CCN concentration under the ship trail cloud (Fig. 3). The ship trail occurred within a very clean air regime where the particle concentration was below $10 \text{ cm}^{-3}$ over a very large area for a long period of time. This appeared to be a favored zone for ship trail formation. The consistently lower solar energy between 1000 and 1100 indicates the ship trail cloud. The variations in solar energy before this time reveal the broken cloud regime which was characteristic of the clean air with low concentrations of very large drops. Moreover the very low particle concentrations and the drizzle which was observed in this area appeared to be brought about by the same coalescence scavenging process which was observed in FIRE and HaRP. Therefore coalescence scavenging appears to be an important atmospheric process.

Plans are proceeding to mount the CCN spectrometer on the NCAR Electra for measurements throughout ASTEX. The P.I. will attend the planning meeting in Denver in February. The CVI will also be on this aircraft and there will be the same kind of interactions of the two devices as were done in HaRP. The
Figure 3. Surface measurements of the CN (total particles) (dotted line) and CCN (solid line) (panel a), and solar energy (panel b) with local time during a northward traverse off the coast of Baja, California. These measurements were obtained on board the Egabrag III oceanographic vessel on July 13, 1991 during project SEAHUNT (Shiptrail Evolution Above High Updraft Naval Targets) conducted by Dr. William Porch of Los Alamos National Laboratory. Prior to 1200 the ship was in patchy cloud and fog. Between 1000 and 1100 the ship passed under a solid line of cloud, a shiptrail. At 1200 the ship went under a conventional marine stratus deck.
CCN spectrometer has been improved since that time so that it has a much shorter response time. It can operate at rates better than 1 Hertz whereas in the earlier projects it could only obtain data every few seconds. This will greatly improve the spatial resolution and will be especially important for the CVI intercomparisons. Interpretations of the continuous measurements especially the interactions of the CCN spectrometer and the CVI should enable us to determine important effects of entrainment on the droplet spectra as outlined in the original proposal and earlier. The availability of the stratus (ASTEX) and cumulus (HaRP) regimes should help in determining the different effects of the two types of clouds and should aid in interpreting the transition from stratus to cumulus clouds which is intended to be an important aspect of ASTEX.