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Progress Report Number three of DOE Contract with
Desert Research Institute

Linkage of Anthropogenic Aerosol to Clouds and Climate"

by

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During the first year of this project equipment was built to obtain direct droplet measurement data from a mobile platform in fog (or mountain impacted stratus) and to characterize CCN according to particle volatility (thermal processing). Since the start date, July 15, 1990, was during the California stratus season and it was necessary to complete the construction of equipment and allow time before a field project it was not possible to begin field work until the summer of 1991.

1. SEAHUNT (Shiptrail Evolution Above High Updraft Naval Targets)

As the 1991 field season approached there was an opportunity to make measurements on board ship in collaboration with Dr. William Porch of Los Alamos National Laboratory. Although this set of measurements did not satisfy all of the goals set forth in the original proposal it presented a number of advantages which could not have otherwise been achieved. The ship offered the opportunity to make longer term measurements within the important Pacific stratus regime. The ship also offered the possibility of going out to distances further from shore than could be done with the light aircraft (originally planned). The ship has the advantage of spending extended periods of time in unquestionably marine air. This is important in order to determine whether near coastal measurements made by light aircraft are representative of the entire eastern Pacific stratus regime.

Other advantages of this SEAHUNT project included the fact that the measurement area was nearly identical to that used for the airborne measurements in the FIRE project in 1987 which was also during the summer stratus season. Since there were some questions about the validity of the aircraft sampling technique and the representativeness of sporadic aircraft measurements, this offered the opportunity to in some sense "check" those measurements. At any rate there was an opportunity to collect a complementary data set, surface and airborne, in the same area at the same season. This was beneficial for the thermal characterization technique since it was being used for the first time.

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During the first of the three one-week legs of the SEAHUNT cruise we were fortunate enough to intersect a shiptrail cloud, July 13. These measurements resulted in the enclosed DOE research summary and the enclosed conference preprint (Hindman et al., 1992). These are the first measurements of CCN in association with shiptrail clouds; they show rather clearly that CCN are responsible for these clouds and that the very low background concentration is favorable to shiptrail formation. Moreover the concentration of CCN seemed to be inversely proportional to the solar energy which penetrated the cloud; this indicated that the CCN were indeed affecting cloud albedo on a small scale and producing higher albedo cloud parcels with higher CCN concentrations. A journal publication concerning this event is in final preparation in collaboration with Dr. Ward Hindman of CCNY, Dr. Phil Durkee of the Naval Postgraduate School, and Dr. Porch of Los Alamos.

Once this manuscript is submitted to the Journal of Geophysical Research another paper will be written by the P.I. and a graduate student concerning the CCN and CN measurements made throughout the three week cruise. Other results from SEAHUNT are presented in the other two preprints enclosed (Hudson, 1992 a & b). The concentrations under the stratus deck were generally similar to those observed from the aircraft in FIRE (1987) (Hudson and Frisbie, 1991). Hence it appears that those aircraft measurements were indeed probably valid and representative of the eastern Pacific stratus regime. The extremely low concentrations which were occasionally observed were extraordinary (fig. 1). During this 24 hour period prior to midnight and after 1200 on the 13th there was a solid stratus deck. The concentrations here were in the 100 to 200 cm^{-3} range. During the interim period between 00 and 12h on the 13th the concentrations were much lower. This was a regime of broken stratus and fog with drizzle. This is seen by the higher solar energy (for that time of day) and the highly variable solar energy trace for the forenoon of the 13th. Between 10 and 11h the shiptrail was intersected and the concentrations were elevated while the solar energy was much lower because of the solid stratus line (see enclosed preprints). Otherwise this period was one of unusually low particle concentrations.

The low concentrations which were observed on some occasions in and near drizzle (Hudson 1992a; fig. 5; enclosed) were significant indicators of the importance of coalescence scavenging (Hudson and Frisbie, 1991). The two enclosed preprints by Hudson (1992 a & b) show how the surface measurements in SEAHUNT fit in with the earlier maritime aircraft measurements.

Occasional bag samples were made at higher altitudes with a special grab-sampling device which could be attached to the tethered balloon of the Los Alamos group. Those measurements tended to show the somewhat higher concentrations which were observed above the cloud decks (Hudson and Frisbie, 1991) and the homogeneity of concentrations within the boundary layer.

Many successful particle volatility measurements were made during SEAHUNT (fig. 2). These indicated that more than 90% of the CCN behaved as though they were ammonium sulfate (volatile at about 180°C). This percentage seemed to be quite consistent throughout the project. There were not many opportunities to make droplet measurements as the cloud base was almost always well above the ship. Nevertheless problems were found with the newly-built

data system for the Royco optical counter used to measure ambient droplets (i.e. fog).

2. Surface Measurements in 1991

During September some mobile surface measurements of CCN and CN were made on the California coast near Monterey. Thermal processing measurements here also showed the same 90% volatility at about 180°C. An example of some of the CCN measurements is shown in figure. 3. The concentrations at the coast were rather high during this period; they were more characteristic of continental air even at altitudes above cloud top. Although the problems with the Royco data acquisition system were corrected there were few fog episodes at this time of the year. Nonetheless a further problem was found with software which prevented recovery of some of the longer droplet data files.

3. ASTEX (Atlantic Stratocumulus Transition Experiment)

During the early part of the 1992 field season we had the opportunity to participate in the ASTEX (Atlantic Stratocumulus Transition Experiment) project near the Azores Islands (fig. 4). This was a stratocumulus multi-aircraft project similar to the FIRE stratus project in California in 1987. The CCN spectrometer was mounted on the NCAR Electra and flew through and in the vicinity of stratocumulus clouds for 17 research flights during the month of June (~115 hours). The CCN spectrometer operated successfully throughout the project and yielded valid data for every minute of research flying (data was missed only briefly for two takeoffs; before entering the research area).

Immediately at the conclusion of the ASTEX project it was revealed that DRI had received a grant from the DOE University Research Instrumentation Program to build another CCN spectrometer. Since a very successful program of measurements had just been completed and by the next field season two CCN spectrometers would be available for this measurement program we decided that it would be best to conserve resources and not to conduct any more field projects during the 1992 summer season. It would be best to use the new instrument in the spirit with which it was given; to further this DOE climate linkages project by making more efficient measurements.

Although our participation in the ASTEX field program was done with NASA support the results would be directly applicable to the climate linkages program, mainly a definition of CCN by comparing nearly simultaneous droplet (provided by the NCAR Electra FSSP) and the DRI CCN spectra. Since the NASA analysis funding is rather limited it seems prudent to put some of the DOE effort into extended analysis of the ASTEX data especially since it is similar to that which was called for in the original proposal. The ASTEX data set was in fact much more extensive than expected and the variability in CCN concentrations was much more than expected. There were extremes of very clean air (figs 5-8) and very polluted continental air (fig. 9 & 10) which produced a very rich data set. Figures 5-7 show the lower concentrations within the cloudy boundary layer which indicate the effects of coalescence scavenging (Hudson and Frisbie, 1991) where clouds remove CCN but not necessarily CN;

concentrations are somewhat constant because the boundary layer is usually very well-mixed. Figure 8 shows a rather constant concentration with altitude because there were no clouds to scavenge the particles. The very high concentrations of particles within the marine boundary layer shown in figures 9 and 10 were quite unexpected at these distances of hundreds of km from shore. They demonstrate very direct influence of anthropogenic CCN on maritime clouds over vast areas of the ocean. Complete analyses of these data including the cloud microphysics should show the great extent of the influence of anthropogenic CCN on climate. This manmade influence on clouds is not at all subtle. The great extent of CCN variability in the Atlantic will make a very wide range of concentrations for the comparisons between CCN spectra and droplet spectra.

Extensive CCN volatility measurements were made with the high temperature processor constructed under this grant (fig. 11). Therefore this DOE grant contributed specifically to the volatility measurements in ASTEX which were not a part of the NASA program analysis and would not otherwise be analyzed or published. Figure 12 shows displays the entire volatility data set from ASTEX. Again this shows that in general more than 90% of the CCN even in maritime air have a volatility similar to that of ammonium sulfate. There was very little difference in volatility with CCN concentration. That is the very clean and the very dirty air showed similar volatility characteristics. This might indicate common source regions for all maritime CCN but further research into this question is needed.

ASTEX also contained other data which was not part of the NASA plan or data analysis support. These were the measurements with the counterflow virtual impactor (CVI) (e.g. Noone et al. 1988). These measurements in fact are especially relevant to the original goals of this climate linkages project because they offer a direct identification of the CCN within cloud droplets. This is superior to the circumstantial comparisons of droplet and CCN spectra. Originally this task was going to be done with cloud interstitial measurements with a cyclone separator (Hudson and Rogers, 1986). Actually the CVI measurement provides a more direct identification of active nuclei because it actually separates the cloud droplets from the sample and then allows measurement of the CCN spectra of the residue of the evaporated droplets. Figure 13 shows examples of the shift to lower S_0 values for the CVI spectra. Thus the nuclei of droplet formation are the more soluble particles out of the available nuclei of the ambient spectrum. Further analysis of these data will be done with Dr. Kevin Noone of the University of Rhode Island. On the other hand the measurements with the cyclone separator require a subtraction of the interstitial spectrum from the total in-cloud spectrum in order to identify the nuclei of droplet formation. Two measurements then are required for the interstitial measurements but only one measurement is required for the CVI technique to determine the nuclei of cloud formation.

Although the Electra did not penetrate any shiptrail clouds during ASTEX it did penetrate some ship exhaust plumes. Figure 14 shows an example of one such penetration. This shows that this plume had CCN but not especially active CCN. Comparisons with the plumes in SEAHUNT will yield more information about ship-produced CCN.

Analysis of ASTEX data awaits the meteorological and cloud data from the NCAR Electra which is due for release any day.

4. Surface Measurements in 1992

Figure 15 shows stationary measurements at our Reno Stead laboratory using the high temperature processor. When the concentrations are higher earlier in the morning there seems to be a greater non-volatile component to the CN; the CCN are much more volatile than the CN. Later in this figure and in figure 16 (for rural Utah) the non-volatile component of the CCN is much lower (3% compared to 10% in the urban air) while the non-volatile CN represent 5% as compared to as much as 40% in the urban air. Figures 2, 11, and 12 show that about 10% of CCN in maritime air are non-volatile. The non-volatile component may be elemental carbon which can remain to temperatures of several hundred degrees (Countess, 1990) is a product of combustion and not expected to be soluble. This may explain why the urban air has a greater non-volatile component. This means that it represents a greater external mixture than rural air.

Figures 17-19 display measurements of dry CCN size vs. critical supersaturation, S_c . These show that in urban areas the CCN have substantial insoluble components as the particles are much larger than they would be if they were pure ammonium sulfate. Calibrations were done with artificially prepared ammonium sulfate particles of similar sizes as those used in the ambient samples. Downtown Reno and San Francisco showed rather similar results but Sacramento showed even more insoluble material for the larger particles. Further measurements such as these will be made in other rural areas and at other times of the year.

Since CCN and CN concentrations are definitely quite different in urban air and they also exhibit differences in volatility properties it is apparent that there is at least an externally mixed aerosol in these source regions. These aerosols are composed of volatile, soluble CCN and non-volatile, insoluble CN. Since the concentrations of these particles in urban air are often very high (several thousand cm^{-3} and much higher right at some of the sources) it is likely that there is significant coagulation between the two sets of particles and/or condensation of various material on other types of particles. Numerous measurements near diesel trucks show that aerosol mixtures form immediately after source emission (Hudson, 1991). Outside of the urban areas concentrations are only a few hundred or a thousand cm^{-3} which means that the coagulation half life of the particles is more than a week (Twomey, 1977). However Twomey (1965) and Fitzgerald et al. (1982) showed that most CCN have substantial insoluble components. Whether or not coagulation is responsible for this or if material actually condenses upon preexisting particles it appears that there are substantial portions of the aerosol which are internal mixtures. It might be expected that the primary natural particles might be pure substances which do not exhibit this internally mixed characteristic. They would be external mixtures if they are produced in the presence of already existing particles or if they become mixed with other particles. However it is unlikely that they would become an internal mixture away from the source region unless very high concentrations are encountered.

Although natural sources of CCN are very difficult to directly identify we have been able to detect pure natural sources in the northern Nevada area in the form of geothermal hot springs. With our mobile measuring platform (a four wheel drive Chevrolet suburban) we have been able to measure upwind and downwind to distinctly determine that the emissions from local hot springs are direct sources of CCN. From all indications we expect that these particles are sulfate aerosol. During the next few months this will be confirmed by the use of our high temperature processor. Furthermore we will use the Electrostatic Classifier (Differential Mobility Analyzer; DMA) with the spectrometer to determine the relationship between size and S_c ; this is for instance unique for pure ammonium sulfate (figs. 17-19). One distinguishing characteristic of this emission is that there was no increase in CN particles beyond the numbers which accounted for the CCN, the hot springs were a source of CCN alone and not a small particle or non-soluble particle source. If these particles are rather pure soluble, volatile material they could be distinguished in two ways from the urban aerosol. We expect that natural particles in this case to be internally unmixed and to remain so throughout the atmosphere. The urban particles should be internally mixed and remain as such even through long range transport.

Although it has not yet been possible to distinguish the natural maritime CCN postulated by Charlson et al. (1987) we would expect that this aerosol would be pure sulfate material. We would expect it to be similar to the sulfate aerosol from geothermal hot springs at least from the volatility and size vs. S_c perspective. The relationships between CCN and CN can be used to evaluate the validity of the assumption of Bigg and Turvey (1978) that their finding that most CN being anthropogenic applies to CCN.

Figure 20 shows a composite of the volatility data obtained during October 1992 in Nevada and northern California. This shows similar results to those in ASTEX that more than 90% of the particles behaved like ammonium sulfate. However many of the particles were even more volatile; they evaporated at even lower temperatures than 180°C. These measurements were obtained in urban areas and are indicative of acid aerosol which has a lower volatility temperature than ammonium sulfate. This might indicate that these particles are not chemically neutralized by ammonium but are primary sulfuric acid particles. This makes sense in that these particles are freshly produced from urban combustion sources.

During the 1993 season which includes part of the third year of this project there should be some surface and light aircraft measurements on the California coast as promised in the original proposal. These would await the second CCN spectrometer for maximum efficiency.

5. Future

In future projects it will be much more efficient to use the two CCN spectrometers together. This will allow such measurements as interstitial, CVI, volatility, and size vs. supersaturation measurements to be made without concern for changes in ambient aerosol. One instrument can make the ambient measurement while the other instrument monitors the processed aerosol; either

from the cyclone separator, the CVI, the thermal processor, or the DMA (Differential Mobility Analyzer). This technique will be especially useful during flights when the concentrations change rapidly.

6. References

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Fig. 1. a. CN (dotted line) and CCN (solid line) (active at 0.7%) at the surface during SEAHUNT on July 12-13. Panel b shows the solar energy trace for this period.

Fig. 3. a. Concentration of CCN (solid line) (at 0.7% S_0) and CN (dotted line) with pressure altitude over land near the California coast. These measurements were obtained at the surface while driving up Freemont Peak. These show low concentrations above the cloud deck and highly variable concentrations within the cloud layer. The concentrations even above cloud are higher than typical maritime values. Panel b displays the CCN concentration only.

Project ASTEX

Fig. 4. Map of ASTEX study region. The NCAR Electra was based at Santa Maria Island in the southeast corner of the Azores which is also the northwest apex of the research triangle.

Fig. 5. Vertical sounding of CCN (0.7% S₂) and CN in ASTEX. This shows the lower concentrations of CCN in the cloudy boundary layer and the rather constant concentrations above the cloudy boundary layer.

Fig. 6. As fig. 5.

Fig. 7. As fig. 5.

Fig. 8. As fig. 5 for a non-cloudy boundary layer.

Fig. 9. As fig. 5 but with an outbreak of polluted air from Europe in the boundary layer. Note the sharp change in concentration above the inversion base where concentrations are typical of the marine free troposphere (~200 cm⁻³).

Fig. 10. As fig. 9.

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Fig. 11. CN (dotted line) and CCN (solid line) ($0.7\% \pm 8\%$) with time during level flight during ASTEX. The CCN only were sent through the high temperature processor during the designated times. The temperatures here were about 300°C and the concentrations were reduced by about one order of magnitude.

Fig. 12. All CCN-volatility data during ASTEX. Above 200°C less than 10% of the CCN survived.

Fig. 13. Two examples of the shift in the CCN spectrum for the counterflow virtual impactor (CVI). The CVI median measurements with the counterflow virtual impactor (CVI). The CVI median S_c is lower than the ambient S_c , showing that the lower S_c nuclei are within the cloud droplets.

Fig. 14a. CN (dotted) and CCN concentration at 0.7% supersaturation from the Electra during ASTEX during a transect of a ship plume.

Fig. 14b. As Fig. 14a but the CCN supersaturation here is 0.1%.

Fig. 15 As Fig. 2 for stationary measurements as Reno Stead for March 19, 1992. Panel B shows the temperature of the processor which was periodically used to condition the sample before entering the CN and CCN instruments.

Fig.16. As Fig. 2 for rural Utah on May 10, 1992. Measurements were made from the mobile platform.

Fig. 17. Critical supersaturation, S_c , vs. dry particle size for Reno, Nevada. Note the larger particles for the same S_c from the DMA for the ambient particles compared to the calibration particles of ammonium sulfate. This shows that the particles have insoluble components.

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Fig. 18. As fig. 17 for San Francisco.

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* ammonium

+ ambient

Fig. 19. As fig. 17 for Sacramento.

Fig. 20. As fig. 12 for northern Nevada and California.

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