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"Linkage of Anthropogenic Aerosol to Clouds and Climate"

By

James G. Hudson

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The equipment construction called for in the original proposal has now been competed. These instruments are the high temperature processor, the data acquisition system for the direct Royco optical particle counter (OPC), and modifications to the formvar replicator. The main field effort during the past year has been the shipboard experiment SEAHUNT (Shiptrail Evolution Above High Updraft Naval Targets) in collaboration with Dr. William Porch of Los Alamos National Laboratory. There were also some laboratory and local ambient particle measurements and a surface field program on and near the California coast. The shipboard project was not anticipated in the original proposal but the laboratory and surface measurements were along the lines suggested in the original proposal.

The high temperature processor was acquired and modified for our laboratory and field usage during the spring. It was used in several laboratory experiments while it was being optimized for field use. Figure 1 shows the extent of the capabilities which are available by using this in conjunction with the CCN spectrometer. When a sample aerosol is volatilized it does not simply disappear but the particles get progressively smaller as the temperature is increased. This allows greater information which can be used to better characterize the particles. Laboratory test particles consisting of NaCl and ammonium sulfate were used to determine the evaporation temperatures of known particles. An internally mixed aerosol of these two substances was also tested. Results of this experiment showed that the ammonium sulfate evaporated separately from the rest of the particles. This thus resulted in an appropriate shrinkage to the size of the NaCl component of the particles. This was an important finding because it showed that the two components of the same particle acted independently with respect to volatility. These results dismissed the possibility that the more volatile component would either be hidden within the less volatile material or that the evaporation of only the most volatile material would result in shattering and thus the production of more particles. These results mean that the volatility technique is viable for identifying mixed particles. Ambient measurements in Reno showed a rather large non-volatile aerosol component; apparently much of the CCN aerosol was not sulfate.

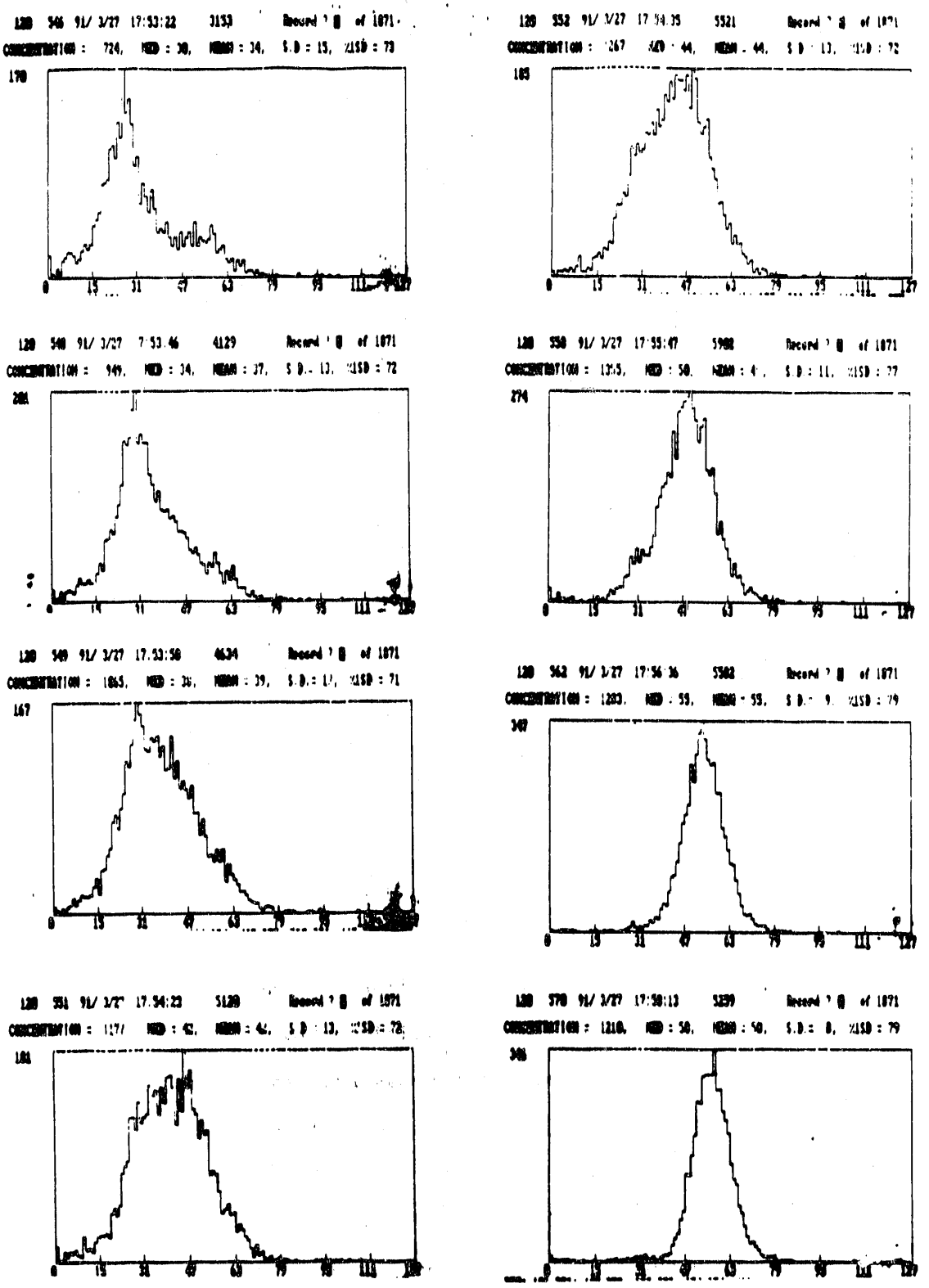


Figure 1. Several CCN spectra for an ammonium sulfate aerosol which was passed through a high temperature processor in which the temperature was being lowered through the temperature of volatility. Higher channels correspond to lower S_c or larger particles. The particles are smaller at the higher temperatures where they are partially vaporized.

The shipboard experiment was not contemplated in the original proposal but this possibility was discussed in the report submitted last year because it was to have taken place within the first year. The cruise actually straddled the first and second years of this project as it lasted for three weeks in July. The shipboard project turned out to be extremely successful; it satisfied all of the objectives which were outlined in the report submitted last year and produced some unexpected bonuses. Some of our results from the cruise were presented at the IUGG meeting in August, the AMS meeting in January (a preprint was prepared for the collaborative paper with Dr. Porch), and will be published as a DOE Research Summary (Feb. 1992). Fortunately we were able to obtain data on all three legs of the cruise. Actually the CCN spectrometer, and the CN counter operated almost continuously throughout the entire period. In addition to this we brought a Dasibi ozone monitor and a solarmeter which also operated throughout the cruise. The ozone measurement proved to be a sensitive marker of the plume from our own ship. Thus it precluded the problem of self pollution. The purpose of the solarmeter was simply to keep a record of whether we were under cloud or in fair weather during the daylight hours.

As pointed out in the last report the continuous surface record will offer a compliment to the aircraft data which was obtained in the same area during the 1987 FIRE project. These measurements have largely verified the rather low CCN concentrations which were measured below cloud in 1987. They tend to verify the arguments which were recently made in a paper about the aircraft CCN measurements in FIRE (Hudson and Frisbie, 1991). We suggested that coalescence scavenging reduces the concentration of CCN in the marine boundary layer. This process can occur only in liquid water clouds. Many have spoken of cloud scavenging as nucleation scavenging but this does not describe the process because nucleation is reversible; nucleation itself does not change the aerosol. However when two (or more) cloud droplets coalesce the aerosol is irreversibly changed. Even if that cloud drop evaporates the resulting residue is a nucleus made up of the two (or more) nuclei of the cloud droplets which formed the coalesced drop. We suggest that this is the mechanism by which aerosol concentrations are reduced in cloudy marine environments.

A notable observation during this cruise was the fact that lower CCN concentrations seemed to be associated with the presence of drizzle. We did not have any specific measurements of drizzle so these observations were subjective. Drizzle was either felt on the skin or it could be seen falling below the nearby clouds. Concentrations were typically 100 cm^{-3} but near drizzle the concentrations would be reduced to less than 30 cm^{-3} (Fig. 2). However during one part of the cruise the concentrations were reduced to below 10 cm^{-3} for more than 12 hours. The CCN and CN (total) particle concentrations were in fact usually below 1 cm^{-3}

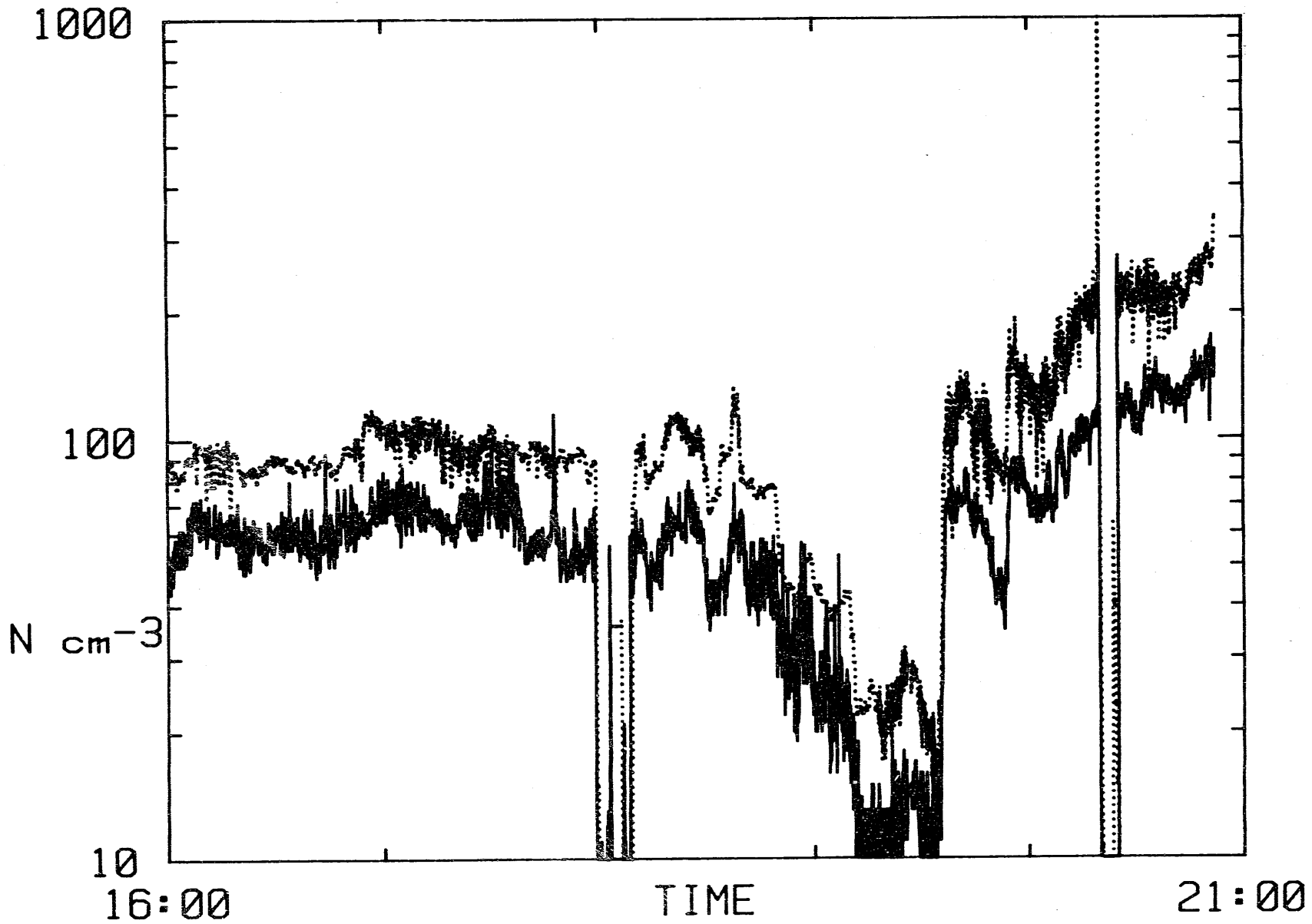


Figure 2. CCN concentration (solid line) and CN (total particles) concentrations as a function of time on board the Egabrag III research vessel on July 18, 1991. Drizzle was experienced at about 1900 hours. The low concentrations at 1800 and 2020 were filter checks of the system.

throughout most of this period (Fig. 3a). Unlike the usual solid stratus deck which was ubiquitous throughout the cruise this area was characterized by broken low level clouds which were often fogs. The variability in the clouds is exhibited in the modulations of the solar energy measured on the ship (Fig. 3b). The clouds were often rather transparent but they frequently produced drizzle. This seemed unusual for such thin transparent clouds. During the daylight fogbows were usually apparent. The fogbows which are just uncolored rainbows appeared to be arcs of cloud at the rainbow angle; the drops are not large enough to cause the diffraction patterns of rainbows but they intensify the internally reflected light. This was an unusual phenomena in that the clouds with which they were associated were essentially invisible. Thus the "clouds" were visible only at the rainbow angle. This was presumably because they were composed of low concentrations of large cloud droplets which is the type of cloud microphysics which is consistent with the very low CCN concentrations which were observed.

While still within this regime the concentrations suddenly increased by two orders of magnitude, to 100 to 200 cm^{-3}, for about 20 minutes (Fig. 3a). This occurred in conjunction with the passage under a distinctly visible cloud line which we latter found to be visible on satellite photos as a definite ship trail. This is seen by the consistently suppressed solar energy between 1000 and 1100 in figure 3b. The decrease in concentration on the other side of the cloud line was just as abrupt. These results indicate that ship trails form in very clean air and that they appear to be associated with high CCN concentrations. Moreover figure 4 shows that the variations in the CCN concentration under the cloud seemed to be correlated with the amount of solar energy received. This is as if the surface CCN concentrations were reflective of the variations which were occurring in cloud microphysics. The higher CCN concentrations seemed to be associated with less surface solar energy and vice versa. At the suggestion of our contract monitor we have ordered a factory calibration of the solarmeter in order to make a more accurate investigation of the connection with the variations in CCN concentrations. We did not anticipate the opportunity to make such precise use the solarmeter.

Although this was the only encounter with a ship trail there were several ship encounters which resulted in measurements of ship plumes at closer ranges (Fig. 5). The background concentration for these ship plumes was much higher than in the ship trail of July 13. Measurements such as these enabled an estimate of the production rate of CCN from ships (Fig. 6). This estimate was not too different from the estimate for the CCN production within the ship trail itself.

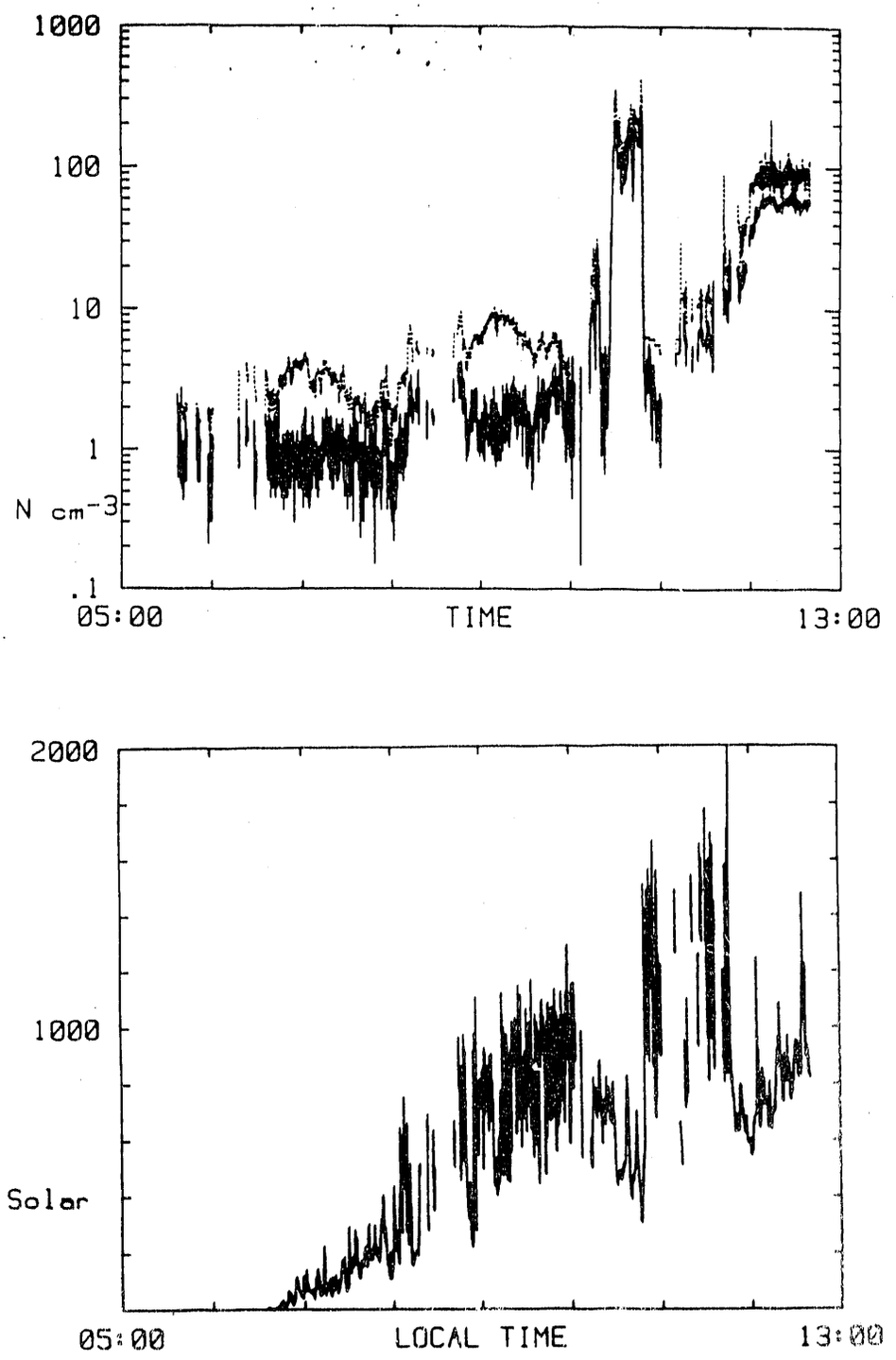


Figure 3. Surface measurements of the CN (total particles) (dotted line) and CCN (solid line) (panel a), and solar energy (panel b) with local time during a northward traverse off the coast of Baja, California. These measurements were obtained on board the Egabrag III oceanographic vessel on July 13, 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.

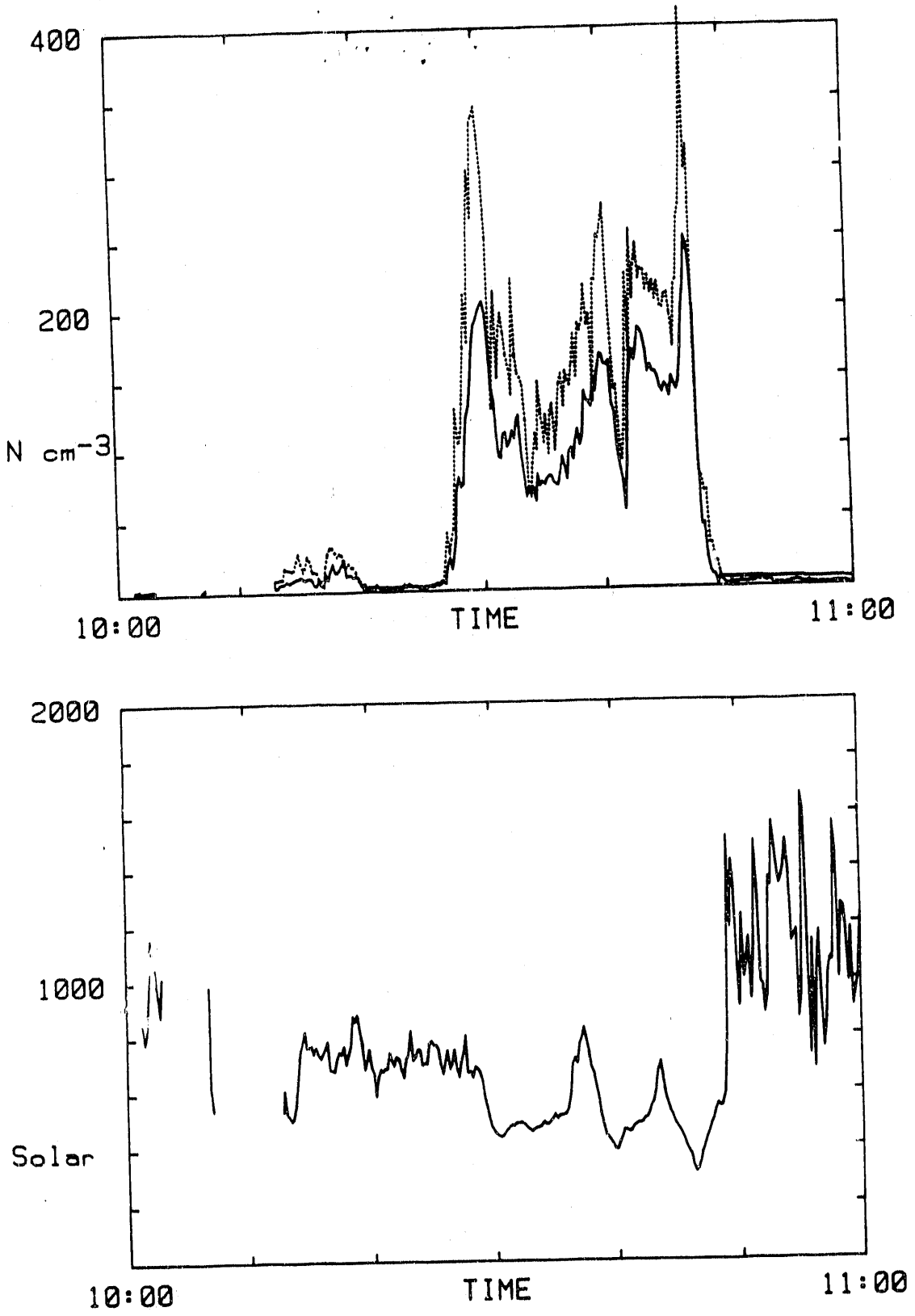


Figure 4. Enlargement of the shiptrail portion of fig. 3.

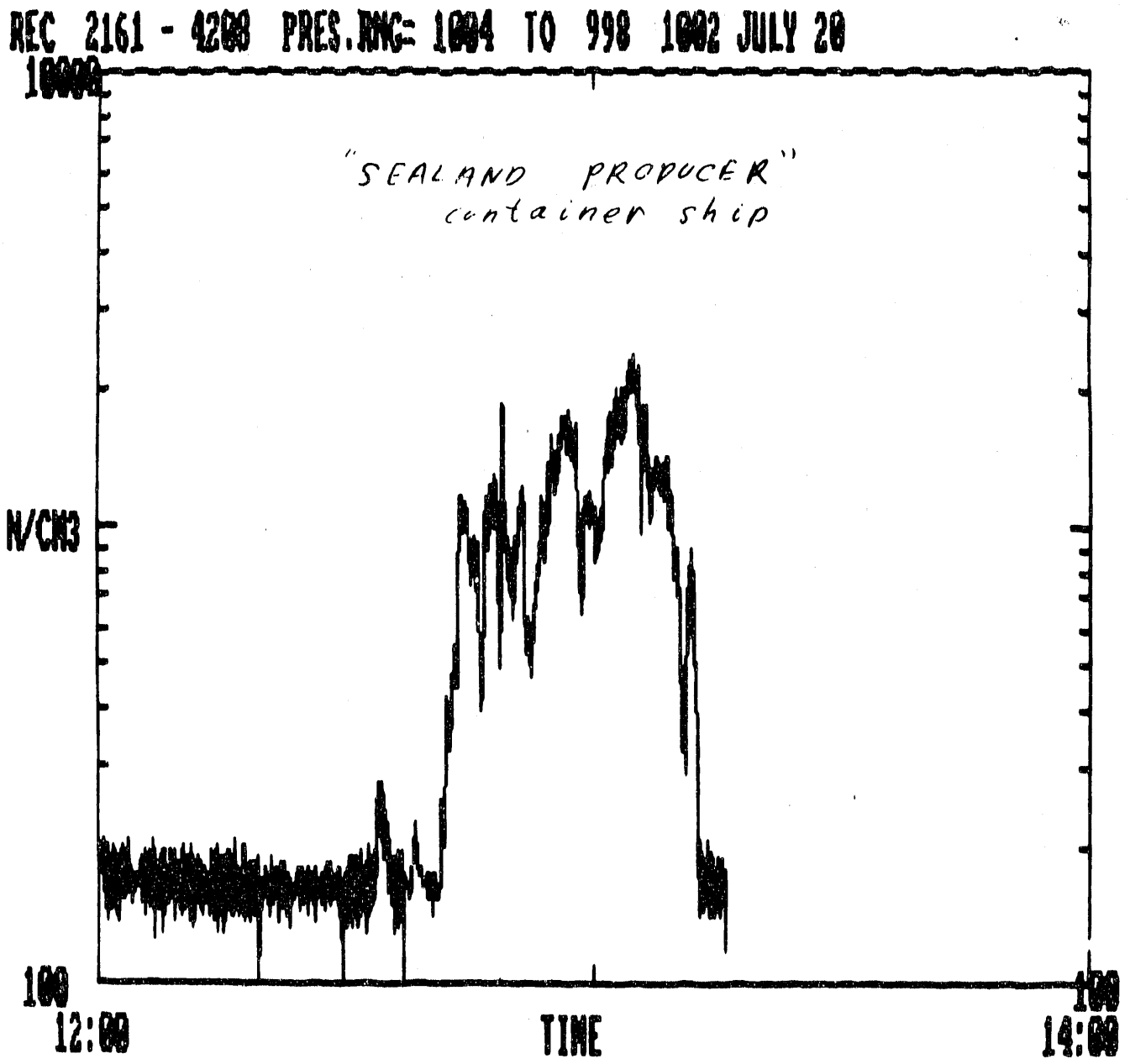
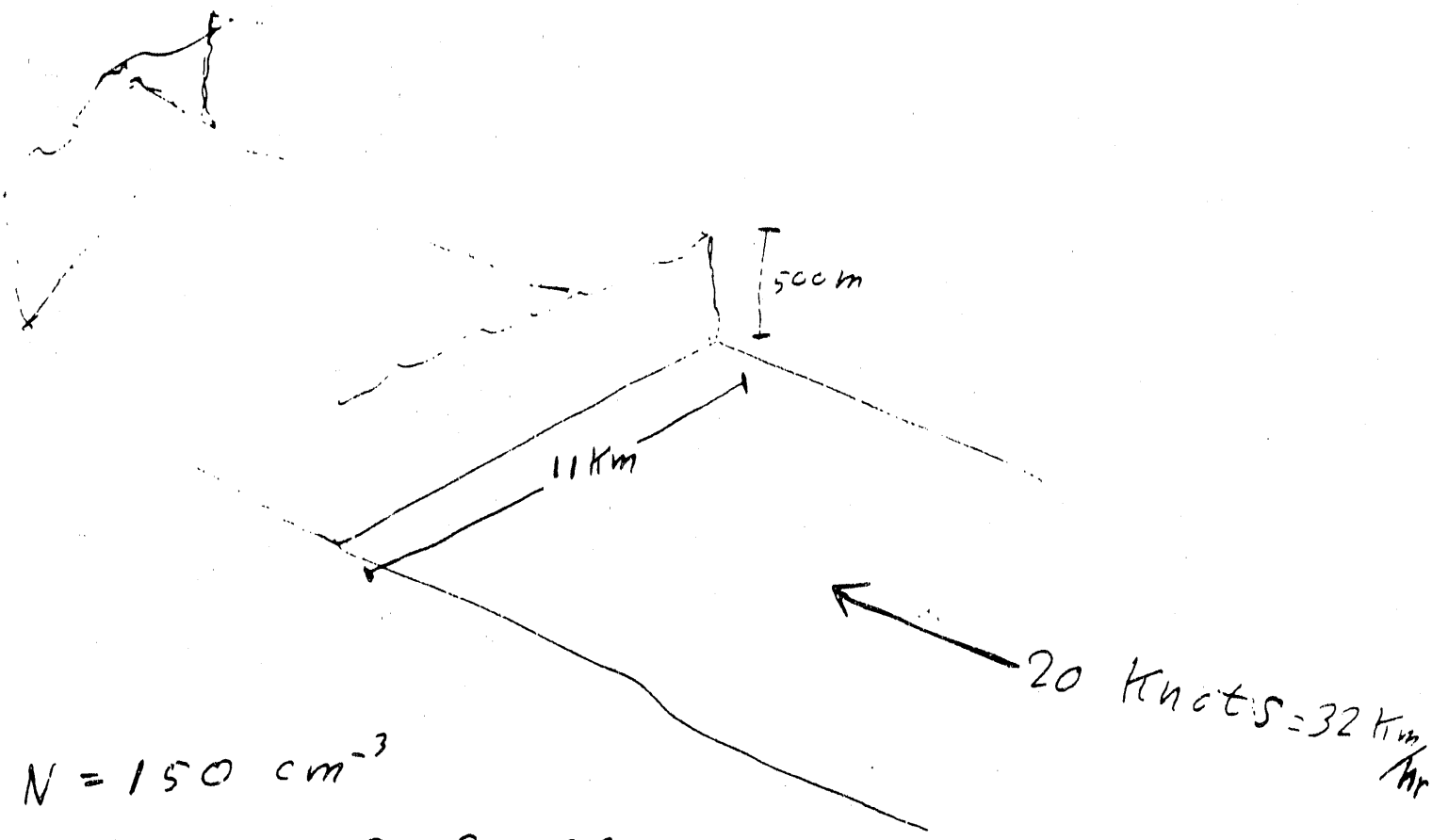


Figure 5. CCN concentration as a function of time on July 20, 1991 while passing through the plume of a ship.



$N = 150\ cm^{-3}$

Lost Rate = $N \delta x \delta z v$

$150\ particles\ cm^{-3} \times 11 \times 10^5\ cm \times 500 \cdot 10^2\ cm \cdot 32 \cdot 10^5\ cm\ hr^{-1} \frac{1\ hr}{3600\ s}$

$= 7 \times 10^{15}\ particles\ s^{-1}$

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$1000\ particles\ cm^{-3} \times 3 \times 10^5\ cm \times 300 \times 10^2\ cm \times 32 \times 10^5\ cm\ hr^{-1} \frac{1\ hr}{3600\ s}$

$= 8 \times 10^{15}\ particles\ s^{-1}$

Figure 6. Diagram showing the method of calculating the flux of particles from a ship.

The high temperature processor was used several times during the cruise. Figure 7 shows that the sample concentrations were reduced by about an order of magnitude when temperatures of more than 150 degrees C were used. This indicates that most of the CCN were certainly not NaCl but probably sulfate.

A surface field measurement program was carried out in September. This consisted of measurements from the DRI mobile laboratory at and near the California coast as planned in the original proposal. The four-wheel drive Chevrolet Suburban contained the CCN spectrometer, the TSI CN counter, a temperature sensor, a wind sensor (speed and direction; the compass direction is also recorded so that the wind direction can be determined even when the vehicle changes orientation), the solarmeter, the optical particle counter (Royco with data acquisition system), the ozone monitor, the high temperature processor, and the formvar replicator. All of these instruments operated on the direct current provided by the battery and generator system in the truck. Measurements were carried out at the Big Sur coast and up on some of the high cliffs near the coast. A series of measurements was made all the way up and down Freemont Pk. (elevation 3900 ft.). This was done through a solid stratus deck so that we were able to make measurements considerably above the clouds. The aerosol and ozone showed a substantially layered structure.

The CN and CCN concentrations were considerably higher than were found with the shipboard measurements. When the high temperature processor was applied to these measurements there seemed to be a much higher non-volatile component than had been found on board ship. This does not necessarily mean that there is more NaCl in the coastal or continental aerosol as the processor temperature could not be raised above 300 degrees with the lower voltage (28 volts) available with the mobile direct current. A temperature of more than 800 degrees C is required to evaporate NaCl. These results indicate a substantial aerosol component which is much less volatile than ammonium sulfate. These results are somewhat similar to the measurements made in Reno and may indicate that this aerosol component is of continental origin.

During the remainder of the second year data analysis will continue. When supporting data becomes available from Dr. Porch of Los Alamos we should prepare and submit a journal paper on the ship trail incident. Moreover we will also prepare a paper on the other aerosol measurements from the ship experiment. Analysis of the September surface measurements will be completed before another surface measurement program is conducted. This may or may not be completed before the end of the second year. Laboratory experiments with the high temperature processor and ambient high temperature processed measurements will be done in Reno. We will try to find aerosol substances which duplicate the performance of the ambient samples; that is substances which show the same volatility as the ambient samples. Local measurements will also include measurements with the electrostatic classifier which obtains the relationship between dry particle size and nucleus critical supersaturation, S_c.

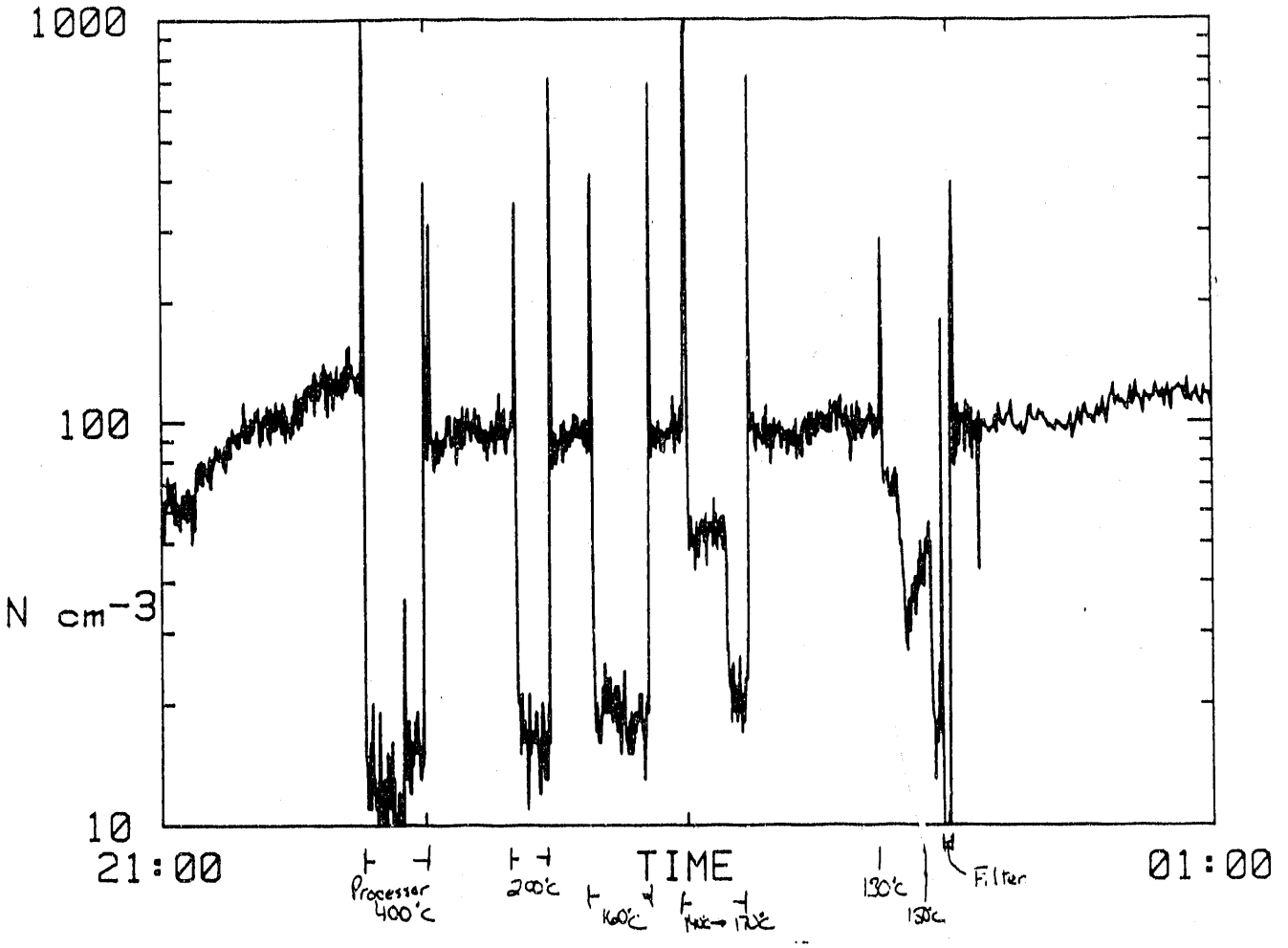

Figure 7. Concentration of CCN for a four hour period on board ship off the California coast. Several times, as noted, the aerosol was heated to very high temperatures. The order of magnitude decrease in concentration when heated indicates that this aerosol is composed of material which behaves like ammonium sulfate.

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