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1. INTRODUCTION

The symbiotic relationship between aerosol particles and clouds is complex because of the many feedback loops inherent in the processes by which cloud particles form on aerosols. This relationship is fundamental to the development of clouds, removal and transformation of atmospheric aerosols and changes in the optical properties of aerosols and clouds particles. A detailed analysis of this symbiosis is needed if we are to understand how clouds and aerosols impact climate and other atmospheric processes.

In December, 2004, a field program was conducted in Puerto Rico, the Puerto Rico Aerosol and Cloud Study (PRACS) with the focus on evaluating aerosol processing by clouds. Puerto Rico was selected because of its location in the Caribbean Sea and its topography. Puerto Rico is an island with no large sources of anthropogenic emissions nearby, other than those produced on the island itself by industry and motor vehicles. The large scale flow is generally maritime from the north east and has passed over a large stretch of the Atlantic before arriving at the ocean. As seen in Figure 1, the major axis of the island is oriented approximately east to west and there is a range

that runs along the center of the island. On the most northeastern point of the island is located a lighthouse and on the eastern end of the mountain range is one of the highest points, called East Peak.

The lighthouse and East Peak were the locations where instruments were installed for measuring the properties of atmospheric particles. These locations were selected for the experimental stage of PRACS because of their relative position with respect to one another and because the East Peak is frequently enveloped in clouds.

As shown schematically in Figure 2, the assumption was that air arriving from the northeast would pass over both research sites so that measurements of the aerosol properties would provide the means to evaluate changes in the particle properties during their passage from the lighthouse to the mountain peak and, when clouds were present, to evaluate the relationship between the cloud microphysical properties and

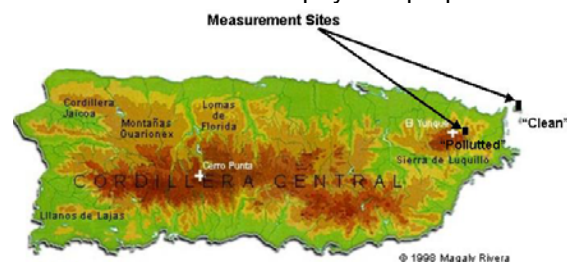


Figure 1

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those of the aerosols prior to, during and after the formation of clouds.

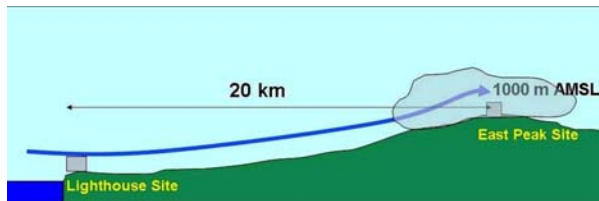


Figure 2

Figure 3 is a photograph that shows the view of the lighthouse site from the installation at the mountain top. The remainder of this presentation focuses on the measurements at this site.



Figure 3

2. MEASUREMENT METHODOLOGY

The instrumentation at the mountain site was installed in a trailer located directly at the top of an overlook facing the lighthouse. Figure 4 shows schematically the instruments that were installed at the mountain site and how they were connected with respect to the three inlets that brought air into the trailer. The “total” inlet was connected to a heater that was maintained at 50° C in order to dry the aerosol particles and to evaporate any water droplets. When in cloud the aerosols measured in this air stream should include interstitial particles as well as the cloud droplet nuclei. The interstitial inlet separated water droplets from the air stream so that in cloud only non-activated particles would be measured. The aerosol particles that entered via the counterflow virtual impactor (CVI) were those that remained after cloud droplets had been evaporated. The results discussed in the

following section will only be those related to measurements made of aerosols that were sampled from the “total” inlet.

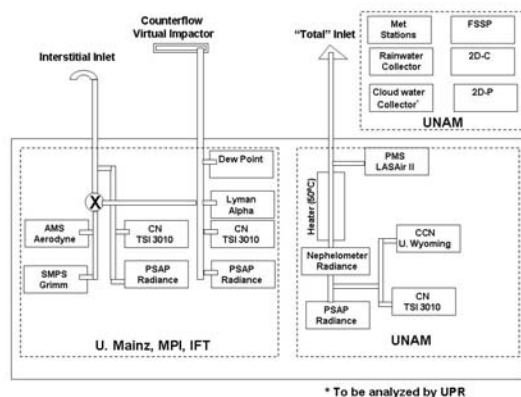


Figure 4

3. RESULTS

The measurements were made from December 2 to 16, during which time the general circulation changed from a northeasterly to easterly/southeasterly flow, as shown in the back trajectory analysis shown in Fig. 5, estimated using the NOAA HYSPLIT model.

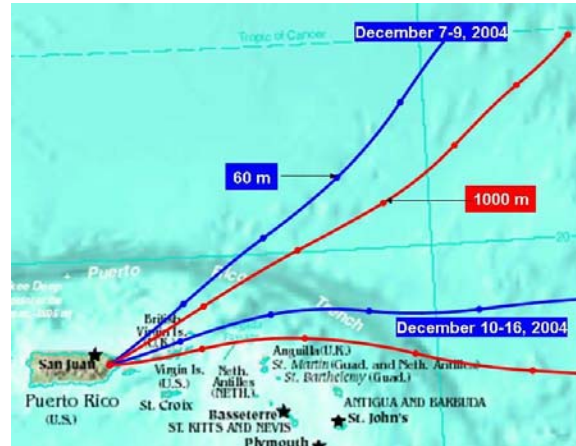


Figure 5

As mentioned previously the air that arrives from the northeast should be free of anthropogenic particles as there are no major land masses for several thousand kilometers up wind. To the east and southeast of Puerto Rico are a number of small islands that are moderately populated and are possibly a source of anthropogenic particles at the mountain site when the air comes from this direction. The largest source of pollution in Puerto Rico is the metropolis of San

Juan; however, it is located on the north-central coast of the island and did not influence the measurements at the mountain site during the measurement period.

As seen in the wind rose drawn in Fig. 6, the winds were primarily from the NNE or ESE during research period and were usually strong with an average wind speed between 5 and 10 ms^{-1} .

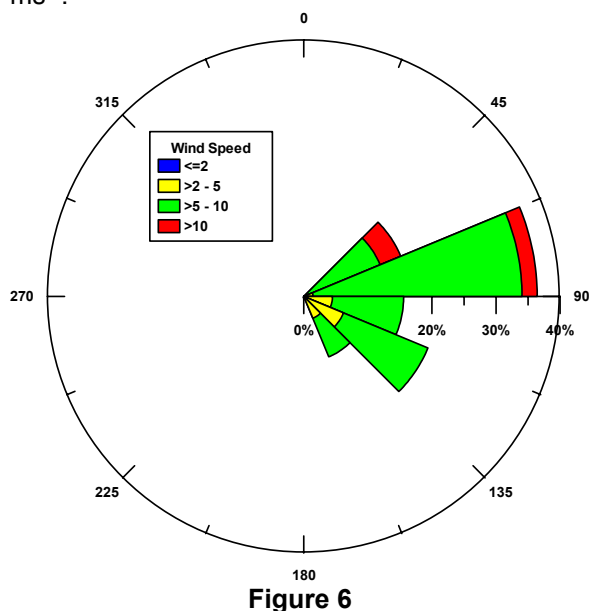


Figure 6

In Figure 7 we compare some of the properties of the aerosol particles, stratified by wind direction and whether or not the cloud was present, determined from the droplet number concentration measured with the Forward Scattering Spectrometer Probe (FSSP). Number concentrations greater than 10 cm^{-3} were arbitrarily used as an indicator that a cloud was present. The graphs in the figure are frequency histograms of the concentrations of CN (condensation nuclei with diameters $> 0.15 \mu\text{m}$), OPC (optical particle counting of particles $> 0.3 \mu\text{m}$), CCN (cloud condensation nuclei activated at 0.5 %) and BC (black carbon derived from the absorption coefficient). The CN were measured with a TSI Model 3010 particle counter, the OPC was the PMS LASAir Model 310, CCN were measured with the Wyoming thermal diffusion chamber, and the absorption coefficient was measured with the Radiance Research Particle Soot Aerosol Photometer (PSAP). The frequency histograms with hatched and solid bars were accumulated when out of cloud and in cloud, respectively. The blue and red bars denote winds from the NE and SE, respectively.

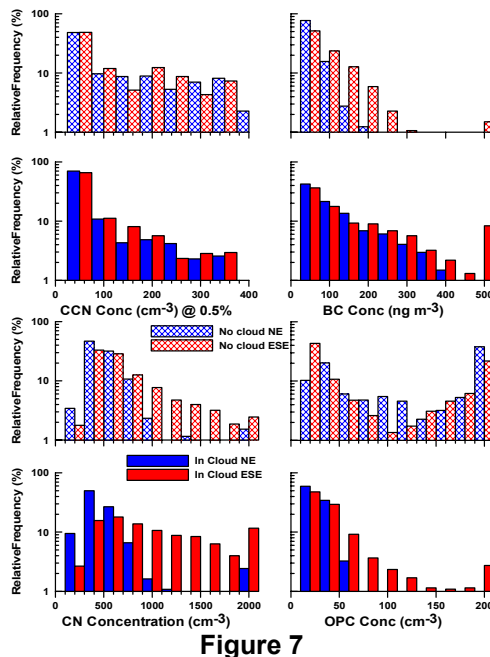


Figure 7

The CN and OPC distributions are clearly different when winds are from the NE and SE during cloud conditions. Both show that there are higher frequencies of larger concentrations when winds are from the SE. When out of cloud, however, the OPC shows no noticeable change with wind direction whereas the CN were slightly higher during periods of wind from the SE. The CCN concentrations are not sensitive to wind direction, regardless of the presence of cloud. The BC concentrations are distributed nearly identically when in cloud but have a slightly higher frequency of larger mass concentrations when in cloud free air and when the air is from the southeast.

The relative shapes of the CN, CCN and BC frequency distributions are quite similar when comparing in and out of cloud conditions. The OPC distribution is the exception in that the more concave shape of the cloud free distribution indicates that the highest frequency occurs for the lowest and the highest concentrations. This concavity disappears under cloud conditions.

Figure 8 displays four frequency distributions of the cloud microphysical bulk parameters, number concentration, liquid water concentration (LWC), mass weighted diameter (Dmvd) and precipitation. The concentration, Dmvd and LWC were derived from the FSSP measurements.

The precipitation was measured with a tipping bucket gauge on the Davis weather station.

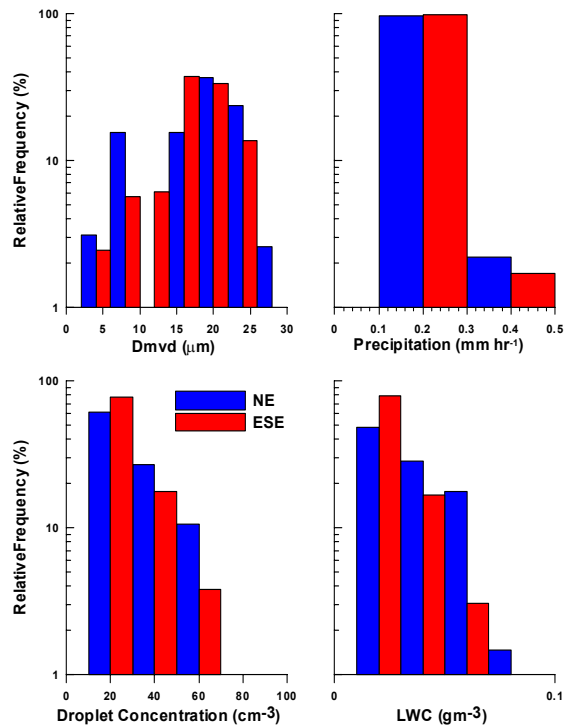


Figure 8

As in Fig. 7, the cloud properties are stratified by wind direction. None of the cloud characteristics show a significant sensitivity to the wind direction.

The sensitivity of aerosol particle size to wind direction and the presence of cloud was evaluated by comparing the size distributions measured by the OPC under these four conditions. The OPC measures in six size channels that range from 0.3 to 25 μm . A seventh size channel from 0.015 to 0.3 was added to the size distribution by taking the difference in total OPC and CN concentrations. The left and right panels of Figure 9 compares the size distributions when winds are from the NE (left panel) and from the SE (right panel) when in and out of cloud (blue and red curves). There is only a marginal difference in the distributions when winds are from the NE and no difference when they are from the SE.

Figure 10 is a similar comparison of the droplet size distributions averaged over conditions of winds from the NE or SE (solid and dashed curves), only when the total number concentrations exceeded 100 cm^{-3} . The size

distribution measured during northeasterly winds is slightly broader than when winds were from the SE but peak concentrations are almost the same.

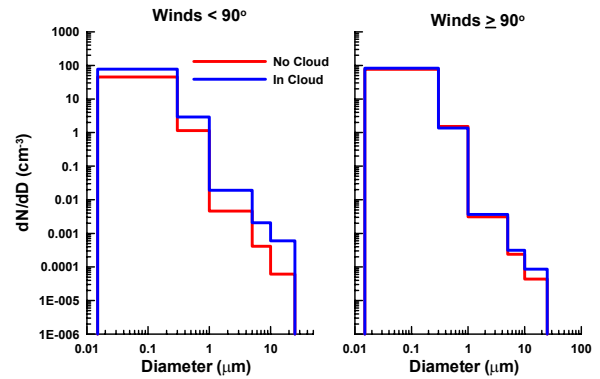


Figure 9

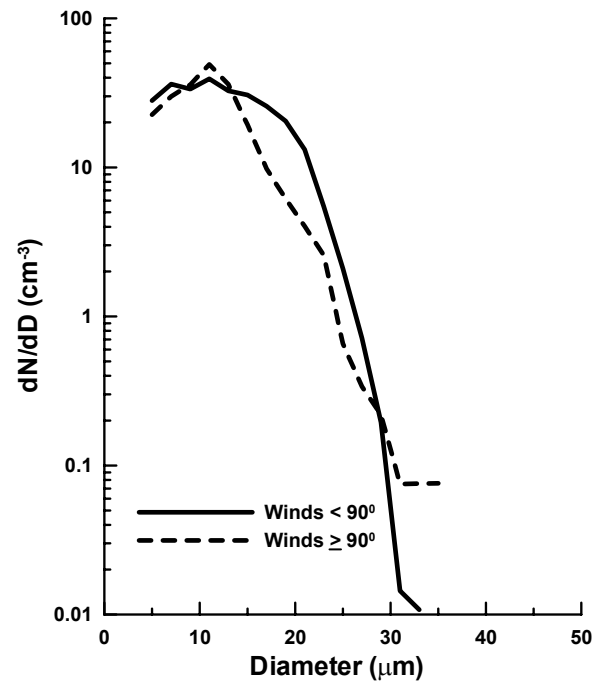


Figure 10

Cloud and precipitable water samples were taken throughout the ten day period and analyzed after the project to evaluate the acidity (PH) and the mass concentration of inorganic ions. The time series in Figure 11 are of the hourly averages of the wind direction, the PH and the mass concentrations of sodium, chloride, sulfate, nitrate and ammonium ions. The PH decreases from the beginning to the end of the project, indicating an increasing acidity in

the cloud water. All of the ions show somewhat of a decrease, although there is a lot of variance from day to day. There does not appear to be an observable correlation between changes in wind direction from NE to SE.

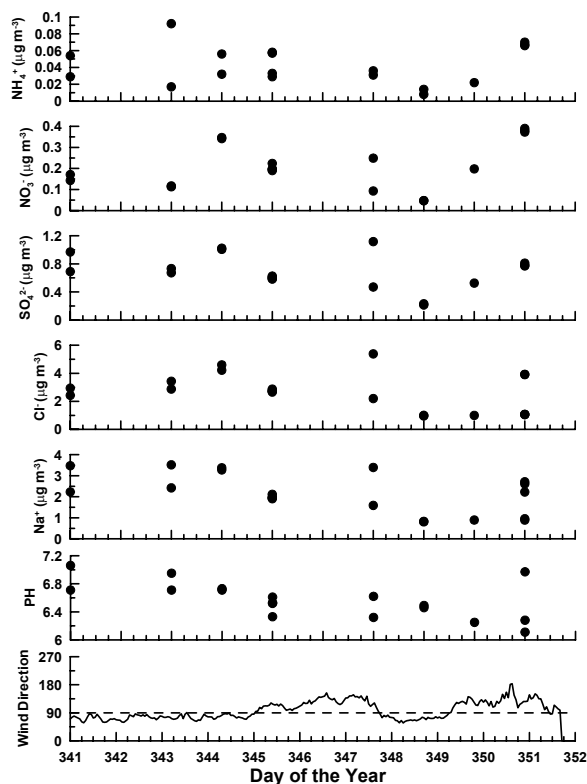


Figure 11

Figure 12 is a pie chart that illustrates what fraction of the total inorganic mass is represented by the eight ions that were derived from the analysis. The mass concentrations of each of the ions were divided by their respective molecular weights in order to get mass equivalents.

The cloud water residual aerosol mass is dominated by sodium and chloride that make up 71% of the mass. The sulfate and ammonium ions make up another 17%. This indicated that the CCN are primarily sea salt and ammonium sulfate.

4. SUMMARY

The preliminary analysis of measurements made in a clear and cloudy environment show that the aerosol particle size distributions were independent of the air mass origin, i.e. clean

maritime air from the NE or moderately polluted air from the SE. Likewise the CCN

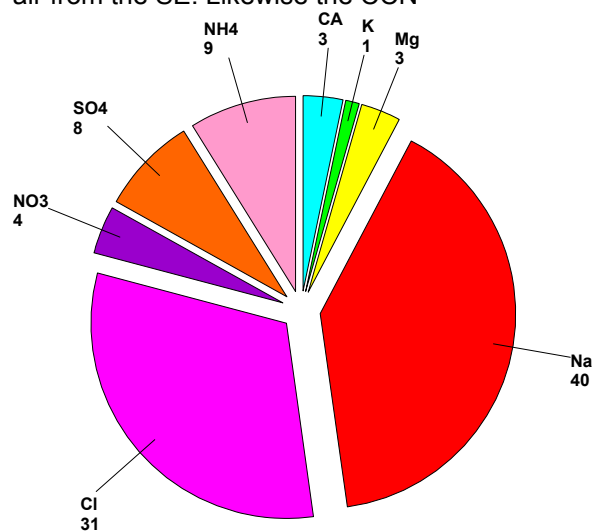


Figure 12

concentrations were insensitive to the wind direction. The CN number and BC mass concentrations were somewhat higher when winds were from the SE, suggesting that the majority of the anthropogenic particles were very small, non-hygroscopic soot.

The cloud properties were virtually unaffected by the source of the air and the amount of rain was the same regardless of wind direction. This indicates that the anthropogenic particles do not increase droplet concentration or impact the efficiency of precipitation.

Finally, the dominance of sea salt in the cloud water suggests that the clouds that form over the East Peak of Puerto Rico are generated primarily from CCN that originate from natural sources.

5. ACKNOWLEDGEMENTS

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allowing the use its facilities in the natural reserve of Cabezas de San Juan.