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Abstract. The influence of aerosols, both natural and anthropogenic, remains a major area of uncertainty when predicting the properties and behaviour of clouds and their influence on climate. In an attempt to better understand warm cloud formation in a tropical marine environment, a period of intensive measurements took place in December 2004 in Puerto Rico, using some of the latest developments in online instrumentation such as aerosol mass spectrometers, cloud condensation nuclei counters and a hygroscopicity tandem differential mobility analyser. Simultaneous online measurements of aerosol size distributions, composition, hygroscopicity and optical properties were made near the lighthouse of Cape San Juan in the north-eastern corner of the island and at the top of East Peak mountain (1040 m a.s.l.), the two sites separated by 17 km. Additional measurements of the cloud droplet residual and interstitial aerosol properties were made at the mountain site, accompanied by measurements of cloud droplet size distributions, liquid water content and the chemical composition of cloud and rain water samples.

Both aerosol composition and cloud properties were found to be sensitive to wind sector. Air from the east-northeast (ENE) was mostly free of anthropogenic influences, the submicron fraction being mainly composed of non-sea salt sulphate, while that from the east-southeast (ESE) was found to be moderately influenced by populated islands upwind, adding smaller

(<100 nm), externally mixed, carbonaceous particles to the aerosol that increased the number concentrations by over a factor of 3. This change in composition was also accompanied with a reduction in the measured hygroscopicity and fractional cloud activation potential of the aerosol. At the mountain site, the average cloud droplet concentrations increased from 193 to 519 cm⁻³, median volume diameter decreased from 20 to 14 μ m and the liquid water content increased from 0.24 to 0.31 g m⁻³ when the winds shifted from the ENE to ESE. Larger numbers of interstitial particles were recorded, most notably at sizes greater than 100 nm, which were absent during clean conditions. The average size of the residual particles and concentrations of cloudwater nitrate, sulphate and insoluble material increased during polluted conditions.

Previous studies in Puerto Rico had reported the presence of a significant non-anthropogenic organic fraction in the aerosols measured and concluded that this was a factor controlling the in situ cloud properties. However, this was not observed in our case. In contrast to the 1.00 ± 0.14 µg m⁻³ of organic carbon measured in 1992 and 1995, the organic matter measured in the current study of 0.17 ± 0.35 µg m⁻³ is many times lower, most of which can be attributed to anthropogenic sources. During clean conditions, the submicron aerosol was observed to be almost entirely inorganic, an observation supported by the hygroscopicity measurements. This suggests that organic aerosols from marine sources may not be completely ubiquitous (either spatially or temporally) in this environment and requires further investigation to quantify their true extent and implications, with more extensive, longer-term sampling in conjunction with wind field analyses.

■ <u>Final Revised Paper</u> (PDF, 1078 KB) ■ <u>Discussion Paper</u> (ACPD)

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