10.2 THE RELATIONSHIP BETWEEN CLOUD DROPLET NUMBER AND AEROSOL EVOLUTION IN POLLUTED PLUMES

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1. INTRODUCTION

A number of flow-through experiments have been performed at sites around the world, in which a hill-cap cloud in contact with the ground has been used as a "natural flow through reactor" to examine the interaction between clouds, aerosols and gases. The aims have been to examine the effects of the properties of the aerosol and trace gases in the airmass flowing into the cloud on the microphysics and chemistry of the cloud and to investigate the role of the cloud in modifying the aerosol and trace gases emerging downwind. The overall goal has thus been to increase understanding of and reduce uncertainties in the direct and indirect radiative effects of aerosols on the earth's radiation balance, so that the role that clouds and aerosols will play in future global climate change may be better assessed. Results from an analysis of data from three such aerosol-cloud interaction experiments carried out at sites varying distances downwind of urban pollution sources will be presented and compared in this paper.

2. THE EXPERIMENTS

In Spring 1999 a EUROTRAC 2 (a EUREKA environment Project) PROCLOUD sub-project experiment was carried out at Holme Moss (a hilltop 30km east of the city of Manchester in the UK) to investigate the interaction of the Manchester urban plume with the cap cloud which forms at the site (Bower *et al.,* 2001(b)) (Figure 1). In summer 1997 a "HILLCLOUD" experiment was carried out on the island of Tenerife during ACE-2, the 2nd IGAC aerosol characterisation experiment. Tenerife (Figure 2), one of the Spanish Canary Islands, is in a maritime region influenced at times by pollution outflow from continental Europe (1-2 days upwind) and desert dust from the Sahara (Bower *et al.,* 2000(a)). More recently (Spring 2001), a aerosol-cloud interaction (ACI) experiment was carried out on the South Korean Island of Cheju Do (Figure 3) during the first intensive phase of ACE-Asia, the 3rd IGAC aerosol characterisation experiment (Bower *et al.,* 2001). Here, aerosols are comprised of substantial amounts of Asian desert dust, particles generated from the high levels of combustion of coal and

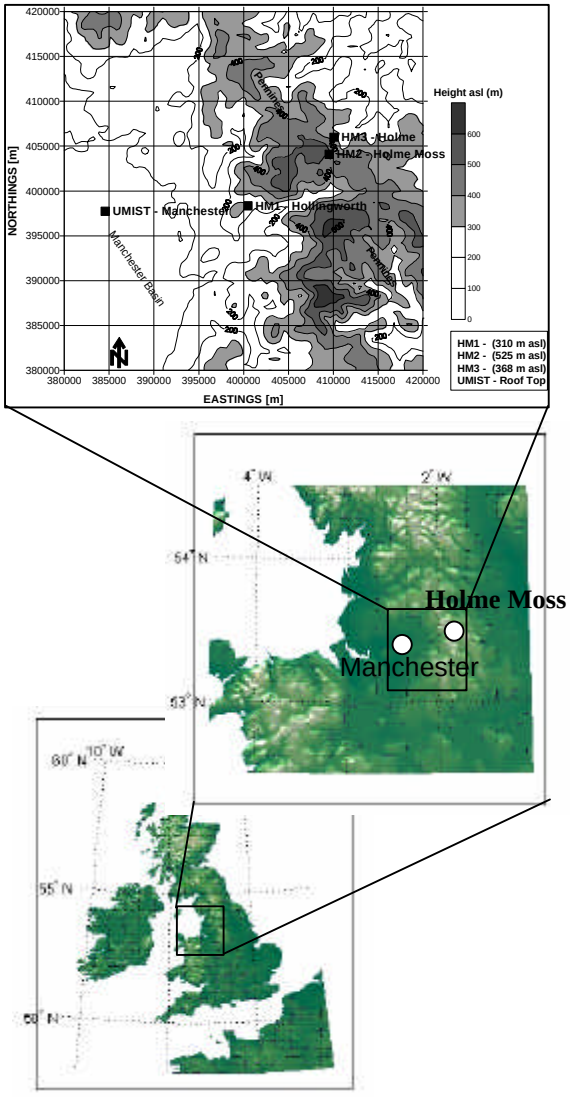

Figure 1: Location of the PROCLOUD Experiment at Holme Moss near Manchester in the UK, in 1999

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biomass, from industrial emissions, and from sea-salt picked up during transit across the ocean.

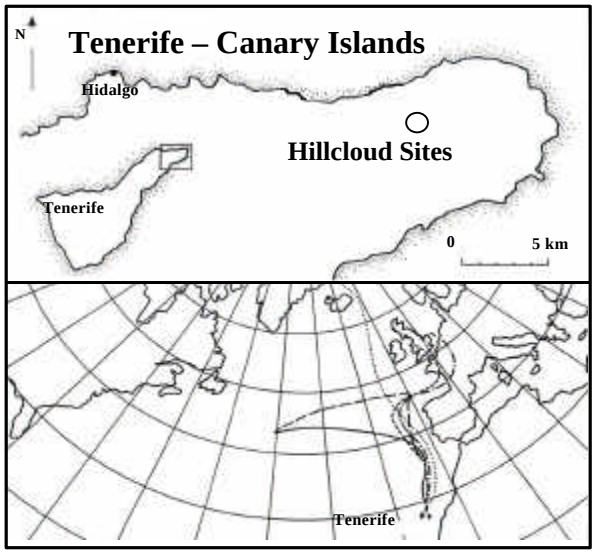

Figure 2: Location of the ACE-2 Hillcloud experiment on Tenerife in the Canary Islands, during June-July 1997.

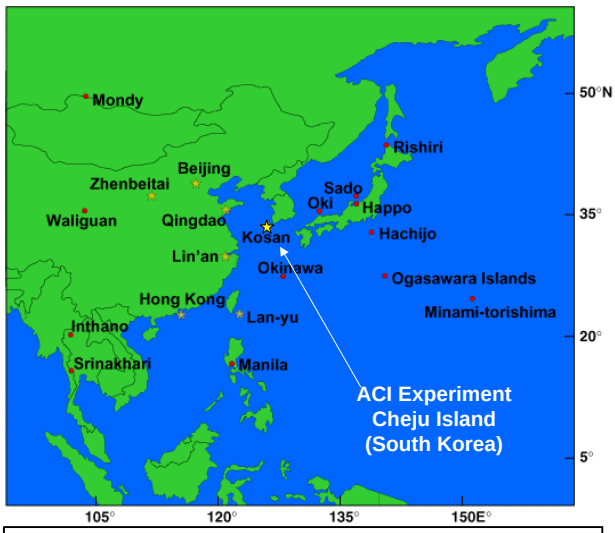

Figure 3: Location of the ACE-Asia Aerosol-Cloud Interaction experiment on the South Korean Island of Cheju Do during spring 2001

In each case, the philosophy of the experiment was the same. Measurements were made of the properties of the aerosol (number, size distribution, hygroscopic growth properties, chemical composition) and trace gases at an out of cloud site upwind of a cap cloud covered hill prior to their interaction with the cloud system.

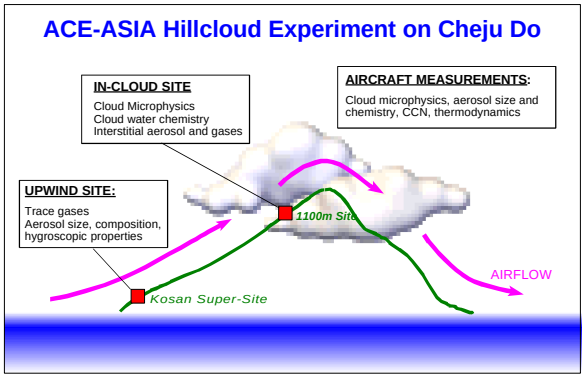

Figure 4: A schematic view of the flow through experiment concept set up to investigate aerosol–cloud interactions on Cheju Do during ACE-Asia. A similar "natural flow through reactor" concept using a cap cloud in contact with the ground was used in ACE-2 and PROCLOUD experiments, but with additional downwind out-of-cloud measurement sites.

Within the cap-cloud, an incloud site was set up to measure the microphysical properties (drop size distribution, liquid water content, droplet-aerosol relationship) and chemistry of the clouds as a function of the aerosol and trace gases flowing into and interacting with the cloud. Here, measurements were also made of the aerosol remaining unactivated and interstitial to the cloud drops, as well as some gases. In PROCLOUD and ACE-2 experiments, out of cloud sites were set up downwind of the cloud system to re-measure the properties of the aerosol and trace gases emerging from the aerosol-cloud system, following any modification that might have occurred through physical or chemical processing within the cloud. Figure 4 shows the concept of the cloud-aerosol interaction experiment as set up for ACE-Asia

3. RESULTS

3.1 *Upwind Aerosol Properties*

Investigation of the diameter growth factor (GFD) distribution of atmospheric aerosol as measured by the hygroscopicity tandem differential mobility analysers (HTDMAs) deployed at upwind out-of-cloud sites in ACE-2 and PROCLOUD experiments, reveals sharp discrete banding of GFD (defined here as the ratio of the aerosol diameter when subjected to a relative humidity of around 90% to that of its dry size) into more- and less-hygroscopic (MH and LH or hydrophobic) modes as described by Swietlicki *et al.,* (1999). The contour plots in figure 5 compare the aerosol GFD distributions obtained at the upwind site some 30 to 40 minutes downwind from

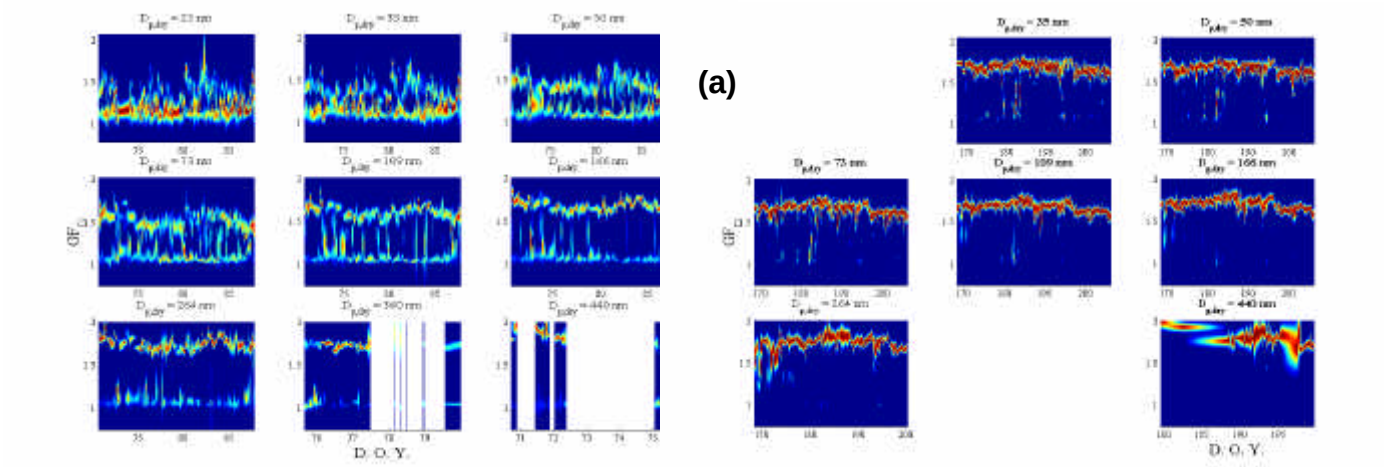

Figure 5: Growth factor (GF_D) distributions from (a) PROCLOUD (left) and (b) ACE-2 (right) field projects. Each panel shows the GF_D values observed as a function of time (the duration of each project) for a particular dry aerosol size cut. Dry size cuts increase from 25 nm (top left panel) to 440 nm (bottom right panel). Dry size cuts on subsequent panels are 25, 35, 50, 73, 109, 166, 264, 360 and 440 nm respectively.

the strong urban source of Manchester during the PROCLOUD project with those measured on Tenerife during polluted periods during ACE 2. In both cases, narrow GFD bands are observed at all sizes. However, closer to the source, the average MH GFD increases with dry aerosol size, whilst the LH mode reduces in fractional contribution with size but has a relatively constant low GFD. During ACE 2, a few days downwind of the pollution sources, the LH mode is frequently absent, but at no time is there a GFD continuum and the MH GFD mode remains relatively constant with size at any particular time.

Dry aerosol size distributions were measured by differential mobility particle sizer (DMPS) instruments over the size range 3 to (at least) 500nm at the out of cloud sites in all 3 projects. Remapping HTDMA GFD data onto DMPS size distributions measured concurrently has enabled dry and wet aerosol distributions to be assembled, separated according to hygroscopicity. Figure 6 shows the diurnal variation in the average of all

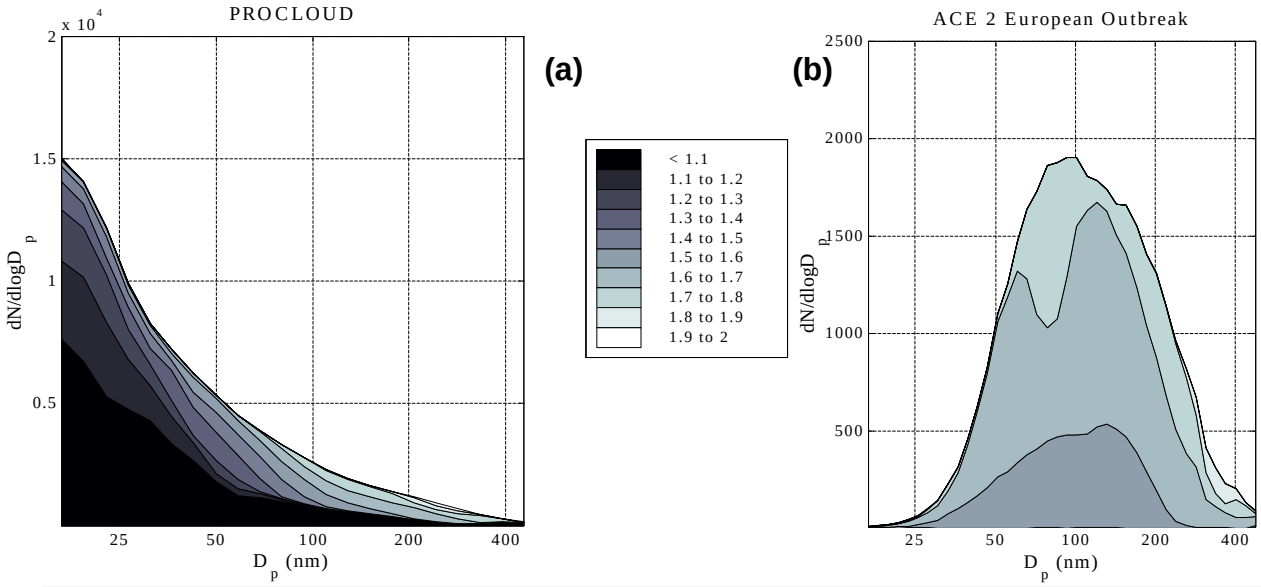

Figure 6: Averaged mid-morning dry aerosol size distributions during (a) PROCLOUD and (b) ACE-2. The distributions have been separated into component growth factor bands of 0.1 GF_D. In (a) the distribution is relatively-local source dominated and externally-mixed, in (b) the distribution is internally-mixed and predominantly hygroscopic. The smaller particles and lower growth factors are almost entirely absent.

such assembled spectra, and clearly highlights the differences between young plumes (PROCLOUD) and aged European Outbreak airmasses measured during ACE 2. No LH aerosols are present in the ACE 2 spectra, although aerosol (and cloud droplet) numbers are significantly higher than in clean airmasses. This infers that, if the PROCLOUD distribution properties are representative of airmasses downwind of urban source regions, either the LH mode has been processed to MH aerosol or it has been preferentially removed from the airmass and been repopulated with MH aerosol. The latter appears unlikely.

Factors affecting aerosol hygroscopicity, and possible processes leading to the evolution of the observed aerosol properties have been considered and will be discussed in the presentation. In addition, calculations based on the theory of water uptake to aerosol of differing chemical composition have been carried out to try to predict the observed aerosol growth factor behaviour, based on the measured size dependent inorganic aerosol composition.

In both ACE-2 and PROCLOUD experiments the size dependent aerosol composition was obtained from analysis of samples obtained using low pressure cascade impactors (Berner impactors) deployed at upwind and downwind out of cloud sites. In ACE-Asia a similar impactor was deployed upwind. However, in addition, a state-of-the-art quantitative Aerosol Mass Spectrometer (AMS) (manufactured by Aerodyne Research Inc.) was also deployed giving similar information at much higher time and size resolution (compared to the minimum 3 hour time resolution and 5 or 6 size stages of the impactor data). These data from ACE-Asia will be presented and discussed, and compared briefly with results obtained more recently using the AMS deployed at measurement sites within Manchester, and elsewhere near to and further afield from urban pollution sources in the UK and USA. These investigations indicate that near to the source, a mode of small newly generated particles generally exists, comprised largely of hydrophobic organic components. Further afield this mode is much reduced and there are a greater proportion of larger aerosols. These are comprised of a mixture of soluble inorganic species such as sulphates and nitrates together with organic species which appear to be of a more oxidised form than in the small mode aerosol (closer to the source). Results from ACE-Asia indicate that the kind of aerosol observed on Cheju Do varied depending on the trajectories taken by the airmass prior to arriving at the island (ie whether it had originated over the China, spent many of few days over the ocean or had recently crossed over mainland Korea).

3.2 *Cloud Microphysics*

The consequence of the very different hygroscopic properties of the aerosol seen in the near and far field to urban pollution sources is dramatically illustrated in Figure 6. This shows the relationship between the observed cloud droplet number concentration and upwind aerosol number concentrations in the PROCLOUD and ACE-2 studies. At Tenerife, the increased numbers of more hygroscopic aerosol arriving at the island during polluted episodes, leads to a corresponding increase in the observed cloud droplet number. The majority of these aged aerosol are obviously sufficiently hygroscopic to enable them to act as highly efficient cloud condensation nuclei (CCN)

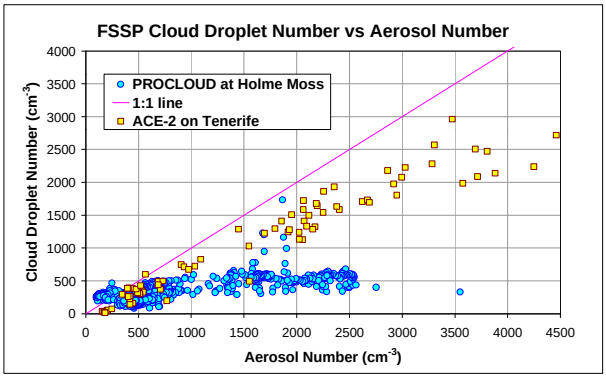

Figure 6: Comparison of the cloud droplet number versus upwind aerosol number relationship observed in the PROCLOUD and ACE-2 experiments

upon which cloud droplets activate in the right circumstances. This leads to an almost linear relationship of increasing cloud droplet number with increasing aerosol number at this site, generating extremely high concentrations of cloud droplets (>2500 cm^{-3}) in the most polluted conditions. Conversely, downwind of the urban source in Manchester, only a few aerosol arriving at Holme Moss are sufficiently hygroscopic to act as CCN. This means that cloud droplet numbers do not continue to increase as aerosol numbers increase because the majority of the additional aerosol in the spectra are small and of the LH variety. Hence, the peak in cloud droplet number concentration rarely exceeds around 600-700 cm^{-3} and is generally 500 cm^{-3} or less close to the source region. In ACE-Asia it appears that a mixture of both regimes is observed, depending on

the recent history of the airmass advecting to the island, however, a shortage of cloud data during this experiment means this is not conclusive.

4. CONCLUSIONS

In many experiments performed at sites around the world an almost universal observation is that atmospheric aerosol populations inhabit one or more narrow, distinct and separate hygroscopic growth factor regimes at all sizes, never exhibiting a hygroscopicity continuum. This observation has important implications, providing boundaries for the timescales of operation of possible processes dominating aerosol property evolution. The effect this subsequently has on the microphysics of clouds interacting with, and forming on these aerosol can be particularly dramatic. In an aged airmass, in which losses due to processes such washout by rain are low, an almost linear relationship between cloud droplet number and aerosol number is generated. This can result in the production of very high droplet number concentrations in the most polluted conditions. These observations have important consequences for the indirect radiative effects of aerosol on the earth's radiation budget near to and several days downwind of pollution sources, particularly when losses due to processes like washout by rain are low.

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