<u>Hanna Pawlowska</u>¹ * and Wojciech W. Grabowski² ¹ Institute of Geophysics, Warsaw University, Poland ²NCAR, Boulder, Colorado, USA

1. INTRODUCTION

Radiative processes within clouds depend on cloud microphysical properties, especially for the shortwave (solar) radiation. For ice-free clouds, the relevant parameter is the effective radius, the ratio between the third and the second moment of the cloud droplets size distribution (e.g., Stephens 1978). Given the bulk cloud properties (i.e., the local cloud water mixing ratio), the effective radius depends on the mean volume radius (which in turn depends on the droplet number concentration) and on the spectral width of the cloud droplet spectrum (e.g., Pontikis and Hicks 1992; Martin et al. 1994). The spectral width was argued to increase with the droplet concentration (Liu and Daum 2002) and to partially compensate the expected decrease of the droplet effective radius, so that the optical thickness may increase less than predicted by the change of the droplet concentration alone. Moreover, spectral width does affect the development of drizzle and rain, and some bulk microphysics schemes require information about the spectral width to represent this process (e.g., the scheme of Seifert and Beheng 2001).

The key processes affecting microphysical properties of warm clouds include: i) the cloud-base nucleation of cloud droplets on cloud condensation nuclei (CCN); ii) the adiabatic growth of droplets above the cloud base; iii) changes of the cloud droplet spectrum due to entrainment and subsequent cloud dilution; iv) additional nucleation of cloud droplets above the cloud base due to either increasing updraft strength (e.g., Pinsky and Khain 2002) or entrainment (Paluch and Knight 1984; Brenguier and Grabowski 1993; Su et al. 1998; Lasher-Trapp et al. 2005), and v) collisioncoalescence. Some of these processes narrow the droplet spectra (i.e., adiabatic growth above the cloud base), whereas others tend to increase the spectral width (e.g., nucleation above the cloud base or collisioncoalescence). Although most of these processes are relatively well understood (i.e., cloud droplet nucleation, adiabatic growth, collision-coalescence), the effects of entrainment and mixing on cloud droplet spectra still lacks solid theoretical foundation; see discussions in Su et al. (1998), Andrejczuk et al. (2006), Burnet and Brenguier (2006), among many others. The interaction among all of these processes determines the shape of the spectrum at a given spatial location within a cloud.

This paper discusses observations of cloud microphysical properties collected in marine stratocumulus (Scu) in the subtropical Atlantic. The goal is to provide a better foundation for representations of microphysical properties of these clouds in various numerical models (e.g., LES, cloud-resolving, AGCMs, etc.). The next section discusses the data and data analysis procedures. Section 3 presents the results and their brief summary in section 4 concludes the paper.

2. THE DATA

The data used in this study come from ACE2 (Second Aerosol Characterization Experiment; Brenguier et al. 2000). Cloud microphysical properties are derived from measurements made by the Meteo-France Merlin IV aircraft using the Fast FSSP (Brenguier et al. 1998) for the droplet size distribution in radius range 1.3-18 μ m. Eight flights, each characterized by different aerosol conditions, were analyzed (Pawlowska and Brenguier 2000; 2003) ranging from clean maritime conditions (June 25 and 26), through partly polluted clouds (July 16, 17, and 19), to polluted clouds (July 8, 9, and 18). Table 1 in Pawlowska and Brenguier (2003) documents main characteristics of observed Scu clouds. On selected days the aircraft performed flights along a 60 km square flight track. Data presented below come from 10Hz data, i.e., averaged over about 10 m of the horizontal distance. Excluding from the analysis regions of drizzle (which was common in maritime clouds and less frequent in polluted clouds) changes little the overall conclusions, so the results below were analyzed without any consideration of the drizzle.

Data collected during ascents and descents through the cloud layer are used in the present analysis. They allow for a good vertical representation of cloud droplet microphysical properties. Each cloud sample is characterized by its location with respect to the cloud base. We represent parameters of the cloud droplet spectrum as a function of either the total droplet concentration (*N*) or the altitude above the cloud base (*h*). Cloud samples are grouped in bins of width of 20 cm⁻³ for the cloud droplet number concentration and of 10 *m* for the altitude above the cloud base. The Fast-FSSP misses small droplets that are common

^{*} Corresponding author address: Hanna Pawlowska, Institute of Geophysics, Warsaw University, ul. Pasteura 7, 02-093 Warszawa, Poland; e-mail: hanna.pawlowska@igf.fuw.edu.pl

near the cloud base and are often present near the cloud top due to entrainment. Therefore, altitudes close to the cloud base and cloud top are excluded from the analysis. To make a distinction between microphysical (condensation and collisional growth) and dynamical (entrainment and mixing) processes, we select samples in clouds with different values of the liquid water content with respect to the estimated adiabatic value at a given altitude.

3. RESULTS

Figure 1 summarizes results from the analysis of all eight flights. It shows the mean radius \overline{r} (top panels), standard deviation σ of cloud droplet spectra (middle panels), and the relative dispersion $d = \sigma/\overline{r}$ (bottom panels) as a function of the droplet concentration N. The left column shows results for cloud volumes close to adiabatic (adiabatic fraction, AF, the ratio between the observed cloud water and its adiabatic value, larger than the 0.9), the middle one - for 0.5 < AF < 0.9, and the right one - for 0.1 < AF < 0.5. The figure combines observations from various cloud levels (between 0.3 and 0.7 of the nondimensional cloud depth).

As expected, the mean \overline{r} (top row) decreases as N increases in near-adiabatic cloud volumes. This is true for the average values between the flights (i.e., for different CCN characteristics) as well as within the individual flights (i.e., for the same CCN; note that we implicitly assume here that horizontal variability of CCN can be neglected for each of the flights). The same applies for the diluted cloud volumes, but the range of droplet sizes is smaller. The decrease of \overline{r} with increasing N reflects a simple fact that, for a given liquid water content, the mean volume radius cubed is inversely proportional to droplet concentration. For a given flight, different droplet concentrations in near-adiabatic cloud volumes can only result from variations of the strength of cloud-base updraft that affects the number of nucleated droplets.

The standard deviation σ (middle panels in Fig. 1) shows considerable variations among different flights as well as within each flight for the near-adiabatic cloud volumes (left panel in the middle row), with σ ranging from less than 1 to above 3 μ m. Contrary to expectations based on classical differences between maritime and continental clouds, there is no clear trend among various flights (i.e., polluted and pristine clouds have similar σ). This is likely the result of the range of cloud base updraft strengths in a given flight, which affects not only the number of nucleated droplets, but the initial width of the spectrum as well. However, for each flight the relationship between σ and N is clear, with larger N resulting in smaller σ . This applies to near adiabatic as well as diluted cloud volumes. Since larger N corresponds to smaller \overline{r} , the decrease of σ with the increase of N seems to imply the *increase* of σ with the *increase* of \overline{r} . Such a result seems counterintuitive because an opposite relationship is anticipated based on the classi-

cal argument concerning the parabolic growth of cloud droplets by diffusion of water vapor, which predicts narrowing of the spectrum as the mean size increases. However, the analysis presented here includes both the horizontal variability of \overline{r} at a given level above the cloud base (due to horizontal variability of N) and the vertical variability for a given N. In other words, considering current results in the context of a single adiabatic air parcel is not appropriate. Moreover, spatial variability of σ at the cloud base (again due to spatial variability of cloud-base updraft strength) can be responsible for some of the observed variability. Finally, it is feasible that collision-coalescence complicates the picture. In the diluted cloud volumes, the spread of the standard deviations and their mean values for each flight are smaller, which again might be considered counterintuitive. In general, a robust σ -N relationship exists neither in near-adiabatic nor in diluted cloud volumes.

The lower row in Fig. 1 shows results for the relative dispersion d, which is consistent with the pattern shown for \overline{r} and σ . Relative dispersion ranges from about 0.1 to about 0.5. For near-adiabatic cloud volumes, flightaveraged d seems to increase with N, which is consistent with some previous observations (e.g., Martin et al. 1994) and theoretical predictions that consider diffusional growth only (Liu et al. 2006). However, the opposite is true for points within each flight, which is also in agreement with theoretical predictions for diffusional growth with given aerosol characteristics and at different vertical velocities (Liu et al. 2006). The main point is that the overall pattern seems to result from the strong dependence of \overline{r} on N combined with the weak dependence of σ on N. This is especially true in diluted cloud volumes.

Results shown in Fig. 1 imply that the variability of *mean* microphysical parameters among various flights is typically smaller than the variability within each flight. This is especially true for the standard deviation σ and the relative dispersion *d*. Since only these mean values are used in cloud parameterizations (e.g., Liu and Daum 2002; Rotstayn and Liu 2003), the presence of large spatial variability has important implications for the representation of clouds in large-scale models of weather and climate. To the authors' knowledge, none of such representation explicitly includes the variability illustrated in Fig. 1.

Since the data presented in Fig. 1 are collected at various cloud heights (i.e., with different cloud water contents), the data include variability as a function of height. It is thus instructive to stratify the data for each flight as a function of height. Martin et al. (1994) showed that the relative dispersion *d* was quite uniform across the depth of clouds they investigated. In the case of clouds considered here, however, the data fail to provide a consistent picture. This is illustrated in Figs. 2 and 3, which show N, \overline{r} , σ , and *d* (and standard deviations of their horizontal variability) as a function of height for two selected flights (marine case of June 26 and polluted case of July 18).



Fig. 1: Results from eight flights plotted as a function of cloud droplet number concentration N. Top, middle, and bottom rows show the mean droplet radius \overline{r} , the standard deviation of cloud droplet spectrum σ , and the relative dispersion σ/\overline{r} , respectively. Left, middle, and right columns are for near-adiabatic (AF > 0.9), diluted (0.5 < AF < 0.9) and strongly diluted (0.1 < AF < 0.5) cloud volumes, respectively. Maritime, partly polluted, and polluted cases are shown using blue, green, and red colors, respectively. Lines show results binned as explained in text, whereas the large big symbols show the averages for the entire flight.

Droplet concentration N (upper rows in Figs. 2 and 3) is relatively uniform across the cloud depth (except near the cloud base and cloud top, where presence of small droplets may lead to the underestimate of N by the FSSP). This is true for near-adiabatic as well as diluted cloud volumes, with N typically smaller in the diluted volumes. The mean droplet radius \overline{r} (second row in Figs. 2 and 3) increases with height, in agreement with the increase of the cloud water mixing ratio.

Change of the standard deviation σ with height (the third row) is different in the two cases selected. For the marine case (Fig. 2), σ decreases with height in the near-adiabatic cloud volumes. However, this trend is reversed when highly diluted volumes are considered (see the right panel in the third row). For the polluted case (Fig. 3), σ increases with height for both near-adiabatic and diluted cloud volumes. Large standard deviation of horizontal variability of σ is also worth pointing out. Comparisons with other flights show that the changes of σ with height shown in Figs. 2 and 3 provide limits for all cases considered: typically, σ varies little with height for the near-adiabatic cloud volumes and increases with height for the diluted volumes. Typical values of σ are in the 1 to 2 μ m range with no clear trend between maritime and polluted cases. As a result of the variability of $\overline{\tau}$ and σ , the relative dispersion *d* (bottom rows in Figs. 2 and 3) varies between 0.1 for marine clouds (especially in the upper half of the cloud) to 0.3 for the polluted clouds. These values are typical for other flights as well (not



Fig. 2: Results for the marine case of June 26. The 1st, 2nd, 3rd, and 4th row shows the mean droplet concentration N, the mean radius \overline{r} , the mean standard deviation σ , and the mean relative dispersion d, respectively, at different heights above the cloud base. Left, middle, and right columns are for near-adiabatic (AF > 0.9), diluted (0.5 < AF < 0.9) and strongly diluted (0.1 < AF < 0.5) cloud volumes, respectively. Horizontal lines represent one standard deviation around the mean value. The dashed line shows the mean height of the cloud top.

shown). In summary, it appears that the increase of droplet size with height is the only systematic impact on the relative dispersion d, and its large horizontal variability results from the variability of the standard deviation σ .

4. CONCLUSIONS

In this paper, the variability of cloud microphysical parameters in marine Scu was investigated in order to provide a guidance for cloud parameterizations and to compare the results with theoretical predictions (e.g., Liu and Daum 2006) and previous observations (e.g., Martin et al. 1994). The emphasis was on the spectral width of cloud droplet spectra, an important parameter affecting radiative properties of clouds and develop-



Fig. 3: As Fig. 2, but for the polluted case July 18.

ment of drizzle and rain. The results presented here paint a rather complex picture as far as the width of cloud droplet spectra in Scu is concerned. This comes from a combination of various factors. For given CCN characteristics (i.e., for a given flight), local droplet concentration varies considerably both within adiabatic and diluted volumes, and reflects both the impact of spatially varying cloud base updraft (which affects the spectrum of cloud droplets just after nucleation) as well as the spectral changes due to entrainment and mixing. It appears that the main factor affecting the relative dispersion $d = \sigma/\overline{r}$ is the mean size of cloud droplet radii \overline{r} , which is larger in maritime clouds at the same height within a cloud (or, alternatively, at the same liquid water content). For all flights, d either decreases with height or does not change significantly. This comes mostly from the increase of \overline{r} with height, with σ varying differently (and unpredictably) with height in different flights. Moreover, σ tends to be higher in diluted cloud volumes, as one might expect.

A significant, but not surprising conclusion of this study, is that spatial variability of the cloud droplet number concentration, size, and spectral dispersion in Scu are all significant on a given day, presumably due to the spatial variability of the cloud base updraft (which affects the cloud droplet nucleation) and spatial/temporal variability resulting from entrainment and mixing (which affects macroscopic, e.g., cloud water content, as well as microphysical cloud properties). Such variability needs to be accounted for in large-scale models of weather and climate.

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