

L.I. Kleinman et al., "Aerosol concentration and size distribution measured below, in, and above cloud from the DOE G-1 during VOCALS-REx, Atmospheric Chemistry and Physics Discussions

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## Overview

Kleinman et al. analyze measurements made by the Department of Energy aircraft during VAMOS Ocean Cloud Atmosphere Land Study (VOCALS). The main thrust is aerosol and cloud physics, with a secondary emphasis on trace gases (ozone and carbon monoxide). The manuscript has two parts. First, a statistical survey showing VOCALS observations stratified by longitude and altitude is presented. Second, Kleinman et al. analyze coincident measurements of cloud droplet (CAS probe), accumulation mode (PCASP probe within the G-1 cabin), and Aitken/accumulation mode properties (SMPS within the cabin). Much of the manuscript is devoted to an analysis of the CAS/PCASP/SMPS set. Data from ~ 100 flight legs, flown near the top of the cloudy marine boundary layer, are analyzed. Also, one of the cloud flight legs is presented as a case study.

In the second part, the emphasis is on cloud "interstitial aerosol." Kleinman et al. define these as "...particles observed in-cloud that either never grew in cloud droplets (i.e., were not activated) or having been activated are returned to the gas phase by cloud droplet evaporation" (P17292, L10). Implicit in their discussion is the notion that investigations of entrainment can be complimented by, and possibly refined by, a the concentration budget (N-budget) they present.

Factors confounding the Kleinman's N-budget analysis include particle concentration bias due to droplet and drop shattering, bias in the calibration of the droplet measurement sample volume (i.e., Sara Lance's comment), poor constraint on the vertical velocities that produce activation, uncertain mass accommodation coefficient, and etc. Furthermore, the N-budget approach was tried some years ago in a ground-based study, with the conclusion being that either particle chemistry or entrainment can explain the existence of unexpectedly large particles (dry diameter = 0.3  $\mu\text{m}$ ) remaining interstitial (Hallberg et al., Phase partitioning of aerosol particles in clouds at Kleiner Feldberg, J. Atmos. Chem., 107-127, 1994). When revisiting the latter, and comparing it to Kleinman's manuscript, I am struck by the additional complexity of an N-budget analysis based on aircraft observations. Most confounding, in my opinion, is the fact that shattering is enhanced because of the speed of the aircraft. In sum, I don't view the second part of Kleinman et al. as one that advances understanding of the entrainment problem.

## Recommendation

Having expressed my negative feelings about the manuscript, particularly the second part, I do feel it can be brought to a level meriting acceptance. Here are my recommendations:

1) Increase emphasis on the shatter-mode (Figure 12) and the cloud conditions that correlate with that mode (LWC, median volume droplet size, precipitation water content, etc). I feel you should explore the plausibility that the “unknown artifact” and “artifact source” is coming from right-tail of the shatter mode. Further, I note that very little is published on cloud conditions that produce shatter and on the size of particles produced subsequent to shatter, sampling and drying. Also, I feel that it is essential that the authors report on the inlet that was used to service the interior aerosol instruments (I comment on this below). The editor (Twohy) is expert in these issues.

2) Coincident sampling with the interior and exterior PCASP is a unique aspect of the G-1 data set. In my opinion, you missed an opportunity to report on how the PCASPs compare within regions identified as producing abundant shatter, how they compare in regions where shatter is not as dominating, etc. I encourage Kleinman et al to explore this. Reference to Gillani et al. (1995) and Gultepe et al. (1996), cited by Kleinman et al., underscore the significance of the shattering problem, and how coming to grips with shattering will help advance the airborne N-budget approach.

3) My reading of the manuscript was frustrated by the range of terminology used to describe aerosol within/nearby clouds. The following terminology is used to describe those particle: cloud free aerosol, pre-cloud aerosol, interstitial aerosol, and in-cloud particles. Related is the issue of pre-cloud layers defined as “below and within 15 km of cloud” (Table 3 and text). If I understand correctly you are only accepting as “pre-cloud” those measurements made within 15 km (horizontal) of a cloud exit. A cogent justification for the 15 km is required. Other parts of the text need clarification; these are mentioned below. I am also recommending that the revised manuscript be carefully vetted prior to resubmission.

4) Better articulation of the field of FT dew point is needed (P17300 and P17312). Isn't the tropical maritime atmosphere the suspected/dominant source region? I would start by stating that the key factor is the thermodynamic/chemical state of the air, prior to ascent. How far the air ascends is also important, as is the degree of subsidence subsequent to convective lofting. In very basic terms, you are describing the Hadley Cell circulation. Related to this, on P17312, I don't think you can assume a constant lapse rate. Also, in your introduction of Kleinman and Daum (1991), I wonder if you summarize that paper adequately by stating that the altitude-dependent decrease of absolute humidity represents a “cold trap” which prevents upward transport. In my view, it is the stratification of the atmosphere which determines the dew point and the maximum height of the ascent.

## Measurements

- 1) The inlet leading to the interior PCASP is not discussed. If this is forward facing then some of the droplets will get in, evaporate (provided the distances and flows allow for sufficient for thermal equilibration), and these will supplement the interstitial fraction with a subset of the activated (and subsequently evaporated) particles. There are other complexities, for example, when droplet size is large (~15  $\mu\text{m}$ ; I am not certain where this point is), shattering occurs and the interstitial fraction will be increased by shattered hydrometeors (droplets and drops). The same can happen when flying through sub-cloud and in-cloud drizzle. Data screening strategies are needed to minimize these effects – they need to be described – and the reader also needs to know about the inlet.
- 2) Given that dew point temperature is used for much of the analysis, I was surprised that there isn't discussion of the dew point measurement. I do see, from the PNL link ([http://www.pnl.gov/atmospheric/programs/raf\\_capabs.stm](http://www.pnl.gov/atmospheric/programs/raf_capabs.stm)) that the device is a chilled mirror. Response time is a known limitation, particularly flying within a broken cloud layer where intrusions of air from above the inversion can create low Tdp (-10 C) patches surrounded by high Tdp (~10 C) air at scales ranging from 0.1 to 1000s of meter.
- 3) Given the inability of the AMS to detect the O/S ratio of the Aitken mode particles (P17303, L21), I worry that results in Figure 7 are being overstated. In particular, I am concerned that there may not be sufficient signal in the AMS-derived organic mass to conclude that there is tendency for sulfate to decrease faster with altitude compared to organic. You can allay my skepticism by reporting what the averaging interval is for each data point (Figure 7), and what the detection limit is for an average performed at representative altitudes within the MBL and FT.
- 4) CPC and CN measurements are reported having a 10 nm minimum detectable size (P17294, L13) and having a 15 nm minimum detectable size (P17299, L12). How are these minimum sizes established?
- 5) In your response to Sara Lance, you seem to be asserting that that an underestimate of CDNC, due to coincidence, will also manifest as a decrease in LWC. In fact, it is possible that coincidence could also increase droplet size, and thus compensate within the LWC/probe comparison you show in your response.
- 6) About the size for the PCASP. A common technique is to pass PSL ( $n=1.59$ ) into the probe, but you are referencing to calibration particles with  $n=1.55$  (P17294, L26). The difference between 1.59 and 1.55 is likely inconsequential, yet, I am curious why your reference is not PSL.
- 7) With regard to PCASP low detection efficiency below  $D=160$  nm. The paper by Liu et al. suggests that this is not low detection efficiency, rather just that the high gain threshold/size table is set incorrectly (factory) such that particles are oversized (Liu et al., AST, 16, 83-95, 1992).

## Other

It is not clear to me how Figure 12 provides secondary evidence in support of the assertion that “..a substantial fraction of particles measured in-cloud....represent evaporated cloud droplets or particles that did not activate..” (P17309, L21).

Absorption measurements are mentioned but we are not told what is absorbing or how the measurement is performed.

Figures 5, 6 and 7. The span of altitude/Tdp is not a constant. Can you show the plots with actual altitude/Tdp range of the boxes?

Twohy (2005) is Twohy et al. (2005)

I suggest that terminology non-refractory/refractory be used at the expense of non-volatile/volatile. There appears to be a mistake with the use of “non-volatile” at the bottom of P17294.

Molality (molal) is not a very informative unit. Please express the aerosol composition as weight fraction (P17293, L20).

Solar heating as a driver of cloud thinning. There may be other drivers, for example, MBL decoupling due to drizzle.

Reflectivity, dimming, and the adjective “brighter”, are used conceptually in regard to the first indirect effect. I recommend that these concepts be defined.

SPMS

“Fragmentation” is used for shattering

Your assessments of the “fraction of aerosol”, “activation fraction”, “the fraction of aerosol not activated”, and “high activation fraction” all use the PCASP N as the maximum possible. Since particles are abundant at dry sizes smaller than 0.11  $\mu\text{m}$ , this terminology needs to be qualified.

Schwartz and Slingo (1996) are short on measurements of the dependence of reflectance on cloud depth (or LWP), but there is observational data supporting this in Brenguier et al., JAS, 2000.

You have NO<sub>x</sub> measurements (P17301, L16)?

Solubility, when first introduced, needs definition

What is “reduced major axis least squares regression”?

Droplet mode and accumulation mode are used for the same thing

Measurement uncertainty equal to 20% for CCN-to-aerosol closure. I agree, but there is a lot of scatter about the 20% as seen in Tables 6 and 7 of McFiggans et al. (2006) (The effect of physical and chemical aerosol properties on warm cloud droplet activation, *Atmos. Chem. Phys.*, 6, 2593-2649, 2006, [PDF](#))

## Gasses

The terminology “were CCN”, can imply that they were measured with a CCN instrument. Rather, you are interested in whether (or not) particles nucleated condensation (acted as cloud droplet nuclei).

TSI and DMT footnotes are incorrect (Table 1)

The nominal PCASP size range is different in footnote (Table 1) and text

Why (Table 1) no footnote for the CAS, CIP, AMS or PILS?

CD (Table 2) is CDNC?

The BAE interval (Table 2) is 12:45 to 15:51?

P17313-L30 – The vertical velocity was only 0.4 cm/s?

The BL, AMS, PILS, POC, CTM acronyms are not defined.

“..particles..returned to the gas phase..by evaporation.” Since the evaporated droplet remains as a particle, i.e. the particle does not evaporate, I suggest that this be reworked. There are a few uses of this terminology.

Haller et al. (2006) is Hallar et al. (2006).

About the PCASP (A and B). I understand (footnote to Table 1) that these have the SPP200 electronics upgrade. It is my opinion that this information should go up in the text, where you describe the probe.

In my opinion, and in contrast to the 20 °C value quoted in Hallar et al. (2006), the following two references do a better job of examining the issue of particle heating/drying within the PCASP:

1) Strapp, J. W., W. R. Leaitch and P. S. K. Liu. Hydrated and dried aerosol-size-distribution measurements from Particle Measuring Systems FSSP-300 probe and deiced PCASP-100X probe, *J Atmos Oceanic Technol*, 9(5), 548-555, 1992

2) Snider, J.R. and M.D.Petters, Optical particle counter measurement of marine aerosol hygroscopic growth, *Atmos. Chem. Phys.*, 8, 1949-1962, 2008, [PDF](#)