

Review of “Drivers of Droplet Formation in East Mediterranean Orographic Clouds” by Foskinis et al.

**Summary:**

This paper investigates properties of aerosol and droplet residuals in orographic clouds measured with aerosol and cloud observations at a mountain site that varies between free tropospheric and boundary layer conditions. The authors employ a new method to separate interstitial aerosol and cloud droplet residuals from a single inlet system in the absence of two separate “total” and CVI inlets by combining effective diameter retrievals and in-cloud size distributions from a DMA. They note the relative uncertainty of the method and quantify penetration fractions of the aerosol. Further, aerosol composition measurements were combined with the size distributions to determine the hygroscopicity and effective supersaturation of interstitial, cloud free, and activated aerosol and these results are compared with a commonly used droplet parameterization where generally good agreement was found between the methods depending on the air mass conditions and activation regime.

Overall, the paper is a very interesting addition to the existing literature and provides an applicable method for distinguishing between droplets and residuals that can be used with previous and future measurements. I only have two major comments and a handful of minor comments, questions, and edits listed below. The paper is generally well written, and the figures provide mostly appropriate information, but I believe a few stray grammatical errors and clarifying captions were not checked prior to submission. I have pointed out a number of these, but I feel several remain, so I ask the authors to please give the paper, figures, and captions a careful re-read prior to resubmission. I believe this paper is appropriate for publication in ACP after these comments and edits are addressed.

**Major Comments:**

If I understand correctly, a Deff minimum threshold of 13.5  $\mu\text{m}$  was applied for all CALISHTO measurements in the “virtual filter” technique and Fig. 3 is only showing this for one case? This raises two questions for me:

- 1) For all the cloudy measurements considered, did the effective minimum threshold vary case-by-case or was 13.5  $\mu\text{m}$  always the most optimal? Are the authors able to provide a statistical analysis (maybe in the form of a histogram) showing the best minimum threshold (best agreement between integrated concentrations and in situ droplet number) for each case to support why 13.5  $\mu\text{m}$  was selected? This discussion should be included in the main text.
- 2) 30 min was used as the pre-/in-cloud sampling lengths. Given the scanning time of the SMPS, this would at most be 6 size distributions pre-/in-cloud. Were other sampling lengths tested, why or why not? Do the authors think a different sampling time, and therefore average, would change the results of the optimal minimum threshold?

The authors have done a convincing job arguing that aerosol concentration and type - not changes in cloud dynamics - drive droplet activation variations. Have the authors considered the impacts of the mean (or modal) sizes of the aerosol populations in the different airmasses and how they may impact hygroscopicity and supersaturation given that size has been shown to be a predominant factor controlling aerosol activation (Dusek et al., 2006; Ovadnevaite et al., 2017)? I would imagine the Hoppel minimum and hygroscopicity near that diameter could impact the activation in different conditions (Dedrick et al., 2024). Given the differences in airmass  $N_{\text{Total}}$ , I might also expect mean/modal size to be different for FTL and PBL conditions and impact hygroscopicity (Royalty et al., 2017; Wex et al., 2016). Were FTL aerosol, while less numerous, larger which also made them more hygroscopic? PBL aerosol were more numerous than FTL, where they also smaller which made them less hygroscopic? Illustrating median distributions in FTL and PBL conditions and highlighting these changes, if any, in the shape of the size distributions may provide this insight but may only warrant a brief discussion.

**Minor Comments/Edits:**

Lines 127-128: The authors reference Foskins et al. (under review) in these lines to describe PBLH. Can the authors please provide a brief description of how the PBLH was identified/determined? Was it from radiosondes, lidar, ceilometer, reanalysis, etc...?

Lines 127-135: Are the authors able to provide contextualization of the macrophysical properties of the clouds measured at the site (e.g. cloud type, base height, thickness, etc.) from CALISTHO measurements or previous work (climatology, surface observers)? Were the clouds encountered at the main site typically within or above the PBLH?

Section 2.2 Instrumentation: Have the authors quantified the particle losses to the aerosol measurements? Can these losses be discussed briefly in this section?

Line 238-239: Typo – delete the first “of” in the sentence reading: “...while the hygroscopicity [of] value of eBC was considered...”

Line 294: Typo – “...with [the] observed moisture content...”

Lines 300-301: does Figure 2 show that the winds from the east/southeast are associated with transported dust aerosol, or is this from previous work?

Lines 309-310: Can the authors please check and clarify which figure is being referenced in this discussion? The authors talk about aerosol content (presumably  $N_{\text{Total}}$ ),  $N_d$ , and  $De_{\text{eff}}$  changes but the figure they are referencing is Fig. S2 which has wind rose plots not  $N_d$  or  $De_{\text{eff}}$ . Are the authors referencing (main text) Figure 2 (please correct the referenced panel letters if so) and/or Figure S3? Further, please be consistent and specific with the terminology of aerosol concentration using  $N_{\text{Total}}$ ; “aerosol content” is vague.

Figure 2 caption: Because SO<sub>4</sub><sup>2-</sup> was also used a proxy for FTL vs. PBL, did the authors see a correlation between SO<sub>4</sub><sup>2-</sup> and PBLH? This relationship is a bit difficult to distinguish comparing the time series.

Figure 2: Why are CCN concentration time series not included in this figure? Can the authors please provide this time series either in this plot or as a supplement?

Figure 3: Panel A shows the size distributions averaged for the different Deff thresholds, correct? I don't believe this is clearly specified in the caption or referenced in the text.

Figure 3: In the main text you state that the Deff threshold sensitivity starts at a cutoff of 10 μm (lines 346-347), but the color scale shows cutoffs starting at 0 μm with distributions in panel A and symbols in panel B showing a cutoff Deff <10 μm was used in some cases (blue colors from color bar). Please clarify this discrepancy. Were these lower cut offs shown for comparison/completeness? If so, please state in text and caption.

Figure 3: Please specify in the caption what the solid and dashed/dashed-dotted black lines represent in this figure.

Lines: 324-325: Where in Figure 3 are the authors showing CCN-active aerosol “of order 100 nm” that “grow at least 10-fold”?

Lines 325-328: When the authors state: “when the station was fully covered by clouds...”, do they mean the station was “in-cloud” (i.e. LWC>0.02)? In these lines there is a reference to Fig. 2, but that figure does not show “cloudiness” or what is meant by “fully covered by clouds.” Fig. S3, however, does separate “cloudiness” and statistics described in this discussion. Did you mean to also reference this figure (Fig. S3)? If Fig. 2 is to be included in this discussion, it would be appropriate for the LWC time series to be added in one of the panels of Fig. 2 (likely panel D). The phrase “fully covered by clouds” is used several times in the manuscript; can this be clarified as “in-cloud” instead?

Lines 329-330: The authors have not shown CCN (timeseries or statistics), therefore the reader cannot glean that the “FTL has fewer CCN” as discussed here. Please provide these metrics.

Line 343: Typo: “We select the periods during we were sampling...” correct to-> “...periods when we were sampling...”?

Line 369: Typo: delete the first “is”: “...for this [is] conclusion is...”

Line 374: Typo: check this sentence: “... , include during the...”. I think the “include” was left over?

Line 428: Typo: change the first “that” to “what”?

Line 455: Typo: correct “was” to “were” in: “...distributions that [was] measured.”

Section 3.4 Closure study of Nd and  $s^*$ : can the authors please place the retrieved/parameterized Nd and  $s^*$  in context with previous observations or modelling results? Were these values consistent with previously reported orographic clouds and/or representative of the regional aerosol conditions and air masses or more similar to other conditions and retrievals?

#### References

- Dedrick, J., Russell, L., Sedlacek, A. I., Kuang, C., Zawadowicz, M., & Lubin, D. (2024). Aerosol-Correlated Cloud Activation for Clean Conditions in the Tropical Atlantic Boundary Layer During LASIC. *Geophysical Research Letters*, 51(3). Article.
- Dusek, U., Frank, G., Hildebrandt, L., Curtius, J., Schneider, J., Walter, S., et al. (2006). Size matters more than chemistry for cloud-nucleating ability of aerosol particles. *Science*, 312(5778), 1375-1378. Article.
- Ovadnevaite, J., Zuend, A., Laaksonen, A., Sanchez, K., Roberts, G., Ceburnis, D., et al. (2017). Surface tension prevails over solute effect in organic-influenced cloud droplet activation. *Nature*, 546(7660), 637-641. Article.
- Royalty, T., Phillips, B., Dawson, K., Reed, R., Meskhidze, N., & Petters, M. (2017). Aerosol Properties Observed in the Subtropical North Pacific Boundary Layer. *Journal of Geophysical Research-Atmospheres*, 122(18), 9990-10012. Article.
- Wex, H., Dieckmann, K., Roberts, G., Conrath, T., Izaguirre, M., Hartmann, S., et al. (2016). Aerosol arriving on the Caribbean island of Barbados: physical properties and origin. *Atmospheric Chemistry and Physics*, 16(22), 14107-14130. Article.