

## ***Interactive comment on “Aerosol particles in the mexican east pacific part I: processing and vertical redistribution by clouds” by J. C. Jiménez et al.***

**Anonymous Referee #2**

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This paper is an effort to characterize the influence of cloud processes on the physio-chemistry of the aerosol and is a suitable topic for ACP. This objective is an important but challenging task and the authors are to be given credit for tackling it. In order to do so they have resorted to statistically comparing humidity corrected wing probe aerosol data for so called “developing” and “dissipating” cloud regions in order to look for mass added, presumably as conversion of SO<sub>2</sub> to sulfate via cloud processing. However, there are numerous issues that are hidden in this type of analysis that are not revealed in the short paper presented here.

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I find the presentation interesting but rather superficial and weakly supported. I suspect that more information could be brought to bear to improve the analysis and clarify the uncertainties, including more complete analysis of the cases mentioned. This would be at the expense of a longer and more complex presentation but I would find it preferable to the grouped statistical approach used here. The issues of uncertainties (or variance implicit in the data) is one that needs greater attention. Results to 2 or 3 significant figures are often used but realistic uncertainties are not indicated.

In short, I find the paper topical and interesting but I feel the support for the observations incompletely developed and not as compelling as I would hope for. More could be done to support and confirm the claims presented (additional text and figures).

A few specific issues:

The PSAP, neph and CCN measurements indicated in Table 1 are not actually discussed in the text (why?) but they should be used as additional indications of polluted vs. marine cases (Table 2). More importantly, the “dry” nephelometer should be able to provide independent confirmation of estimated “dry” wing probe mass increases estimated in text and thereby strengthen arguments based up problematic humidity correction to the low-resolution wing probe size data. Periods of reasonably steady “dry” nephelometer values could imply a steady dry mass and wing probe variations during this time should reflect RH variations. Do they? This or a similar comparison should be demonstrated quantitatively as a test of performance.

These wing probes often exhibit uncertainties larger than those shown and can display differences in performance characteristics within a flight and in wet or dry air. Also, how long a sample is required to get a “stable” coarse size distribution for unperturbed conditions and how does this compare to sample times used here?

“Dilution” is mentioned as a major process and even calculated to 2 significant figures but the suggested process is never clearly described. What is assumed to be diluting what? If it is clean boundary-layer, above-cloud or FT air? Please show size distri-

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butions and demonstrate examples (size resolved) of how the resulting aerosol mix is consistent with mixing of two air masses based upon say differences potential temperature. How is dilution calculated and how robust the percent mixing that is claimed?

7799, I11 RH size correction:

This is based on NaCl and is only reasonable for particles larger than 1  $\mu\text{m}$  or so. A sulfate based correction is probably more reasonable [lower  $f(\text{RH})$ ] for smaller sizes. However, it should be noted that recent papers suggest that OC may be as important as sulfate in these settings. Moreover, wing probe size bins are coarse and the effects of this correction is really shown. For example, a doubling in mass would result in only a 25% diameter increase and this appears to be considerably less than the differences between adjacent size bins. Bin boundaries look like nearly a factor of 2 in diameter such that moving particles across a bin boundary (across the threshold on a pulse height amplifier) would effectively increase their mass about a factor of 8. The details of the size scheme used, assumptions made and actual impact on a measured size distributions (with uncertainties) should be illustrated since it drives the conclusions. Systematic differences in RH for growing and dissipating sides of cloud could lead to systematic size differences if these dry “adjustments” are not realistic. What were these differences, if any? Also, aircraft RH measurements are often a source of error unless carefully done and evaluated.

In short, the validity of the wet distributions, corrected dry distributions and uncertainty in this correction procedure deserves much more attention (including the assumption of NaCl only, ignoring the condensed sulfate discussed here) along with the influence of measured RH (or temp) uncertainty that is also involved. The size change  $g(\text{RH})$  is a function of both composition and mass mean diameter but this is not discussed. Also, were distributions from different altitudes corrected to STP?

What is variance on equivalent “cloudy” air and clear air over equivalent times? Include % variances in Table 3. Typically, the clear-air near surface layer and cloud layer can

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exhibit gradients in concentrations not unlike those suggested for Type 4. Show more examples of distributions being compared including those presumed as entraining from the free troposphere and/or the above cloud size distributions (also generally entrained into cloud).

OVERALL BASIC CONCERN - given the limited number of cases (Table 3). Are the differences discussed statistically significant or systematically biased? Representative aircraft sampling in convective environments is a difficult task and deserves greater discussion than provided here.

What are the sample periods for the presumably averaged data shown in Fig. 3. What is the variance in each size bin for the distributions shown? “Near” and “far” differences shown in Fig 3 are not great and can be expected (are often observed) for random differences in the so-called “far” samples. Also “far” samples away from cloud are presumably the subsidence region for air from aloft and should be smaller.

In this context, one would like to see vertical profiles of aerosol properties discussed and thermodynamic properties, water vapor mixing ratios discussed for the various cases or at least some case studies. The actual gradients in these properties are needed to put flight legs in context.

SOME “layer” homogeneity appears to be implicitly assumed here but even in remote oceanic regions there is significant natural variability. For example, in low wind regions locally high winds and sea-salt production at the surface can be induced by significant convective elements. This could lead to enhanced patchy supermicron and submicron aerosol from the sea-salt mode that may or may not get convected upward. In either case it could contribute to variability in either surface reference level or the outflow level concentrations depending upon how sampling was done.

Apart from the comparison to continental cases mentioned for the mass produced in cloud processed air, are the size changes claimed here consistent with expected photochemical production of condensable species from oceanic SO<sub>2</sub> over the time periods

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of the clouds studied for the expected sources in this remote oceanic region? I believe that SO<sub>2</sub> in continental air is often measured to be much more than a factor of two greater than remote oceanic values. What is expected here?

Coalescence is invoked for the larger mass increase in marine clouds. This would imply large reductions in the accumulation mode number that would also have to experience a large mass increase if it were to account for the coarse mode mass increase. However, this appears to be in clean air with low SO<sub>2</sub> compared to the polluted case. At the expected SO<sub>2</sub> levels, are results consistent with data or expectations? Are actual changes in number and volume distributions in coarse and submicron sizes consistent with size dependent coalescence?

#### Technical Concerns

P7798, I26 “Eaveraged into 1 sec intervals..” Is this appropriate? PSAP and other data often internally averaged over longer periods and is often not representative at 1sec. Nephelometer residence time is considerably longer than 1sec. etc. \*\*RR nephelometer sensitivity in table1 should be about  $1.0 \times 10^{-6}$  for about a 1 min average. Indicate the origin of the accuracy claimed in Table 1.? Any PSAP corrections for RH fluctuations and pressure changes (known artifacts).

Fig 3 Describe meaning of near, far and 300m in caption. Not clear that these differences are significant - uncertainty bars?. What is variance of these measurements? Other captions could be more complete also.

Fig 6 discussion appears confused with Fig 5 in text.

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