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

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Response to Reviewers "Aerosol particles in the Mexican east pacific part I: processing and vertical redistribution by clouds"

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We would like to address the questions, comments and suggestions of the two reviewers with the same response since their primary concerns with the paper were the same. We will, however, also respond to those comments from each reviewer that was not shared by the other. In those cases, we will preface our response with "To Reviewer 1 (2)".

Prior to presenting our response we would like to thank both of the reviewers for their scrutiny of our paper, their attention to detail, and their constructive comments. The clarity and quality of our manuscript is greatly improved as a result of their diligence.

To both Reviewers

1) The reviewers have expressed concern and confusion about our choice of particle composition when calculating dry diameter from the OPCs. Reviewer 1's concern is that we will dry the particles too much and underestimate the size if we assume pure NaCl. Reviewer 2 expressed similar concerns and asked that the details of the size correction be discussed.

We have added an additional description to the paper that clarifies the primary purpose of drying the aerosol particle, i.e. to minimize the possibility that any enhanced growth that is measured will be a result of cloud processing and not just deliquesced particles in the higher RH regions. The reviewers are correct that we will most likely remove too much water; however, this underscores the likelihood that the enhancement we see is probably an underestimate and would probably be even greater if we were to use a less conservative approach to the drying method. Here is what we have said in the new text:

"The alternative to removing water is to use the ambient RH to add water mass to the aerosol particles (AP) measured by the PCASP; however, as discussed in greater detail below, the objective of this study is to evaluate changes in AP mass that are a result of their passage through clouds. This evaluation is made in the cloud-free air near cloud boundaries where RH is usually quite high. Hence, in order to differentiate changes in mass due to deliquescence from changes that result from coalescence or absorption of gaseous species, we estimate the amount of water mass in each size category of the particle probes and remove it to obtain a new "dry" distribution. We have no measurements of the chemical composition of the AP, therefore we don't have an accurate relationship between mass change by deliquescence and RH. We

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make the most conservative estimate of the water mass, i.e. the most reasonable maximum amount is removed by assuming the particles are all deliquesced ammonium sulfate with the amount of water mass determined from a function that is fit to the results of previous laboratory studies (Tang, 1980). Clearly the particles can contain other chemical species; however, with this procedure we assure that any differences between near-cloud size distributions and those far from cloud will be a result of cloud processes and not from elevated humidities. Figure 2 illustrates the effect of applying this technique to one of the volume distributions measured near cloud in cloud system 1 (discussed below). The water removal method decreases each of the size thresholds of the FSSP-300 and 100 by a factor determined by the RH relationship."

2) The other mutual concerns of the reviewers were (a) the lack of information on the amount of variability in the data that was either attributable to inherent measurement uncertainties or that could be attributed to natural fluctuations, (b) insufficient quantitative analysis, and (c) limited information about our methodology for classifying the type of cloud processing.

In order to address these concerns, we have reprocessed and reanalyzed the measurements with a more quantitative approach that retains the critical information about the size distributions but incorporates a discussion of the natural variability. We have also changed our definition of “far from cloud” in order to provide a more robust comparison between the near-cloud far-from-cloud size distributions. We compute vertical profiles of the average far-from-cloud spectra from a complete flight, rather than a separate far-from-cloud spectra for each cloud case and level (Figure 3 in the new text). This provides more reasonable background spectra for a specific flight and decreases the variance as a result of more data points. Previously we had used visual inspection of the PSDs to determine the four types of cloud processing. We now calculate the ratios between the near cloud sub-micron and super-micron volumes and the same volumes at the 300 m level, far from cloud in order to determine any enhancement. In addition, we convert all volume concentrations to volume per mass of air so that the

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differences in altitude do not affect the results.

The cloud processing type previously referred to as “dilution” is now called “mixing” and is quantitatively estimated from the ratio between 300 m far cloud CN concentration and the near cloud CN concentration. We justify this in the text by explaining that CN, although not a conservative tracer, is dominated by the smallest particles that are unlikely to be removed by nucleation scavenging. Hence, detrained air will be a mixture of cloud base air that is brought from cloud base and environmental air. This incorporates Reviewer 1’s concerns about the possible mixing of aerosols of higher concentration aloft. This did turn out to be the situation for cases 3 and 4, cases that we had originally labeled maritime, based on the 300 mb CN, but when plotting the vertical profiles of CN and PCASP (now shown as figure 4 in the new text), we saw layer particles at a higher elevation, leading us to now classify cases 3 and 4 as continental due the effect on the clouds. In these cases, mixing did not produce dilution, but the near cloud volumes were impacted both by the cloud processing and the mixture with the upper level aerosol particles.

The suggestion by Reviewer 1 is also incorporated and discussed in which the ratio of the PCASP and CN concentrations are computed. This ratio is another very clear indication whether or not aerosol particles with sizes less than 0.1 um have acquired non-water mass and grown into the detection range of the PCASP. In the new text we discuss, in much greater detail, the physical mechanisms by which the AP properties are changed by cloud processing, the indicators of these processes in the PSDs, and representative vertical profiles that demonstrated the unambiguous signatures of processing mechanisms. We no longer talk about dilution, per se, but mixing as a mechanism for modifying the environmental AP characteristics at a given detrainment level. As shown in our new Figure 5, the variability is illustrated in the vertical profiles as standard deviations around the average values of submicron and supermicron volumes.

In summary, the more quantitative analysis of the PSDs in comparison with more repre-

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sentative background size distributions brings us to results that are much more robust and defensible with detailed descriptions that clarify technique and methodology. Both reviewers ask why we mention instruments that we did not use in the analysis, i.e. Nephelometer, CCN and PSAP. We believe that the composite size distributions are sufficient to show the cloud processing of aerosols and have removed any reference to the other three instruments. Reviewer 2 suggests that the PCASP and nephelometer measurements would reinforce our classification of clean versus polluted air, but both of the instruments sample air from inlets and the maximum AP size that can pass is about 1 um, hence, supermicron particles would be mostly under-sampled. The CN counter is very sensitive to anthropogenic particles and serves to differentiate air masses as well as show the growth of small particles when compared to the PCASP. There is nothing in this paper that requires CCN measurements; hence we leave those out of the discussion. We had listed all the particle instruments for completeness, but have now removed those from the table.

Individual response to reviewer 1 The inconsistencies between the use of NaCl and ammonium sulfate have been reconciled by using ammonium sulfate throughout. The growth factor as a function of salt or sulfate is practically indistinguishable between the two; hence, we did all the drying calculations with sulfate, also using the results from Tang, so that we could get a conservative estimate of water mass and also make the compute estimates of enhanced sulfate mass for comparison with previous measurements and models.

We have further differentiated between the sub and supermicron enhancements by evaluating the frequency of three events: submicron and supermicron enhancement together, submicron only and supermicron only. We have not added these three cases to the tables or graphs, however, but discuss these cases in the text with the implications (Discussion section).

We have corrected the error of showing area distributions rather than volume. The example PSDs are now all in terms of volume per mg of air.

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The description of how cloud systems were selected has been modified to remove the use of “recently processed particles” since we have no way of actually determining when a cloud might have dissipated. We acknowledge that we might be flying through air from a previous cloud but the way that the cloud formed suggest that this was probably never the case, i.e. these were not cloud fields but cloud lines.

We have expanded our discussion to include the possibility that the near cloud regions might contain detrained air that is not only from the same level but might possibly originate from other levels. We can’t rule this out, although our mixing argument and methodology for evaluating the mixing helps us determine that mixing with environmental air from other levels is unlikely, given the rather small vertical velocities in the near cloud regions, compared to those within the cloud that are transporting the particles.

The statement concerning the larger amount of sulfate in larger water droplets has been validated both with models and CVI measurements (from another project - not ours). The argument is that the larger water droplets provide more surface area for SO2 or SO3 to dissolve and add to the aerosol mass already in the droplet.

Part II of this paper addresses the question of comparing the measurements with a model. Part II takes the measured, near-cloud aerosol particle distributions and uses them as the new CCN for activating new cloud droplets. The model shows that the processing of aerosol particles and then recycling them produces large droplets and precipitation faster than the original CCN distribution. We also use a radiative transfer model to show that the optical depth of near cloud air is on average 10 times greater than the environment as a result of the AP enhancement.

The typographical errors have been corrected.

Individual response to reviewer 2

The use of the nephelometer as a quantitative test of the drying scheme is a good idea but the loss of most of the supermicron particles in the inlet limits its usefulness

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in this application. In addition, as the drying scheme is intended to give the most conservative estimate possible of any enhancement (discussed above), this type of validation is probably not essential for this paper.

The statistical sampling time to get representative supermicron concentrations is now discussed.

Representative aircraft sampling in a convective environment is clearly a difficult task and we present the data as statistical averages, with respect to the 78 near-cloud regions that we analyzed, but we are not trying to say that these 78 samples represent all convective maritime clouds. We have added additional discussion to emphasize this point. The variation within each near and far cloud sample, however, leads us to state that the enhancement in volume that we measure is greater than the natural variation during a single pass through the near cloud regions. We had, at a minimum, a kilometer of air sampled for each near cloud sample. This is discussed in somewhat greater detail now in the text.

The previous discussion on sulfate mass produced in cloud processed air included comparisons with ship and aircraft based measurements of SO2 and sulfate and our very rough estimates are within the expected values.

Coalescence does not necessarily entail large reductions in the accumulation mode, at least, with the lognormal distributions we see that the accumulation mode concentrations are several orders of magnitude larger than the supermicron. We haven't tried to quantify the ratios of super to sub micron mass in the same way that we added the ratios of PCAS to CN. It would be possible, but rather than add further to the length of the paper, that has grown substantially after addressing all of the reviewers' concerns, we will discuss this transfer of mass in Part II where it is most relevant to the activation of new cloud droplets. In this paper, the evidence for the enhancement of mass, in general, is the most important aspect that we are emphasizing, whereas in Part II we will show the relevant mechanisms, i.e. coalescence vs. uptake of SO2.

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As we have removed the references to the nephelometer and PSAP, the one second averaging period is sufficient since the particle probes and CN counter were sampled at 10 Hz.

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