Atmos. Chem. Phys. Discuss., 4, S3396–S3402, 2004
www.atmos-chem-phys.org/acpd/4/S3396/
European Geosciences Union

Atmospheric
Chemistry
and Physics
Discussions

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 #1

Received and published: 21 January 2005

General comments:

This paper is an attempt to classify the different kinds of modification of the aerosol spectrum observed in and around clouds in the ITCZ region over the ocean to the west of Mexico during EPIC. The authors attempt to ascribe the modifications observed to the effects of the interaction of these aerosol populations with the deep convective clouds which form along slantwise SW-NE lines in the region. They also consider the processes which may have led to the observed changes. This is a very useful exercise to carry out and is worthy of publication.

However, while I believe the authors have done a good job in reporting the observed aerosol modifications which they categorise into 4 general types of modification for their full set of C130 observations, I have some problems in accepting their explanations as to how these observed changes were brought about as being the only possible mechanisms for this. I feel that these ideas are incomplete, that the arguments they present are not always sufficient to arrive at their conclusions, that certain assumptions made have not been fully justified, and that the inclusion of more information (data) which I believe they have available to them could go some way to rule in or out some of these uncertainties. In addition, more information needs to be provided to illustrate the statistical significance of the case studies presented, and to appreciate the variability in the measured parameters in and around these ITCZ cloud systems. If these amendments can be made, then I believe this paper will be suitable for final publication in ACP.

Main points of concern and specific comments:

The authors generate their Particle Size Distributions PSD's from a combination of measurements from a heated PCASP (giving "dry" aerosol size distributions in the size range 0.1-3 um), and data from FSSP-300 and 100 probes which actually measure ambient (wet) aerosol over the size range 0.3-50 um (section 2.1 pp 7798-7799). Hence to derive the final "dry PSD"; the FSSP data are dried out and then averaged with PCASP data over the overlapping size range. The larger wet aerosol measurements are converted to equivalent dry sizes by assuming the ambient droplets they measure are deliquesced NaCl particles at the observed RH which they then dry by removing the water volume (using Tang 1980). However, as there is no information to the contrary, I guess the authors are assuming the particles to be composed 100% of this soluble material ie the presence of no internally mixed insoluble material or sparingly soluble, or soluble organic material is considered. There is much evidence to now suggest that even in the marine environment the organic aerosol fraction is significant. Even if this material is soluble it will still reduce significantly the growth of the ambient aerosol. If there is any insoluble material present this growth will be further diminished. Hence the derived dry sizes from the FSSPs assuming NaCl as the sole aerosol component will lead to an underestimate of the actual dry size. This will certainly be significant when later considering the volume PSDs. In addition, the assumption that the aerosol inorganic fraction is wholly composed of NaCl will further contribute to this underestimate. The contribution of NaCl to the soluble inorganic composition will be highly wind speed (and size) dependent, and other less hygroscopic inorganics (likely to be dominated by ammonium sulphate) will contribute significantly in lower wind speed conditions, and will almost certainly dominate at submicron sizes at all times. This will lead to a further underestimation of the dry particle size from the FSSPs. I would highly recommend the authors recalculate these PSDs taking these points into consideration.

In addition, there should be some measure of the uncertainty in the final dry aerosols sizes presented (eg error bars in Fig 3), arising as a result of the uncertainties associated with the measurements made by the different probes, as a result of the finite bin widths of all three PMS probes and as a result of the assumptions made in calculating the dry particles sizes.

The assumption that the aerosol are composed only of NaCl in the derivation of the PSDs is not consistently adhered to in the rest of the paper. For example, in the discussion section 4 (p7805) it is assumed the particles are ammonium sulphate when converting volume distributions to mass distributions, to examine the differences in particle mass concentrations between the near cloud regions and the 300m environment (P7805 and figures 6(a-d) - which incidentally are incorrectly referred to as figs 5(a-d) in paragraph 3), for comparison with the model results of Flossman et al., and when considering sulphate production as a means of modifying PSDs.

[Section 2.3, page 7801 paragraph 3 (lines 10-29)]: After categorising the four patterns of aerosol modification in the previous paragraph, in this paragraph the authors present four possible explanations for the increases in particle concentrations observed in various parts of the PSD. This is the section where I feel the authors have failed to fully explore a number of alternative explanations (or at least fail to tell us they have) and failed to make full use of all the data available to them which might help to clarify what is going on. For example, the authors consistently fail to consider a perfectly plausible (in the absence of evidence to the contrary) 5th explanation for increased concentrations, which is they have mixed aerosol rich air into the system from some other level. Mechanism (1), which is mixing in of ambient air whose particle concentrations are higher than at cloud base is considered , but only for mixing in of far-field or "far from cloud" air at the same altitude as the observation, and generally since it seems this air contains reduced aerosol concentrations it can't therefore account for the concentration enhancements. Hence in the example given in Figure 3a where there is both sub and super micron enhancement, this mixing mechanism is ruled out. There is never any discussion of what variations in aerosol number concentration or PSDs were observed in the vertical or of the variations encountered in the horizontal in the "far from cloud" regions. It would be extremely useful to present some variances of these concentrations and PSDs in the result tables.

We are told profiles were carried out in clear air regions far from cloud (used to determine the environment thetaE profile - p7800 line 6), but were any profiles carried out nearer to the clouds - or above clouds in FT air? It would very useful to know how the PSD total number concentration, the CN concentration and the PSD itself varied during a profile. Were any elevated concentration levels ever encountered, and if so were these increases comparable to those observed in the modification cases where aerosol concentrations increased in some part or all of the spectrum? Were any PSD shapes measured during the profile similar to the modified spectra observed allowing for "dilution" and mixing with the cloud base level spectra? Inclusion of such data, or at least discussion of the presence or absence of such variations in the vertical is required to rule in or rule out this mixing explanation (if no such layers of enhanced concentration existed). Otherwise if there were levels where elevated concentrations were observed above clouds, it is possible that entrainment at cloud tops (even previously in now dissipated clouds) and transport in downdraught regions associated with the convective systems could introduce these enhanced aerosol concentrations into the system at the observation levels. This could then account for the "modifications" observed.

The above information (variability in CN and PSD concentrations and PSD shapes in the vertical and horizontal) is also required in order to properly consider mechanism (4) "dilution". To understand the effects of dilution, there is a need to consider the aerosol size distribution in the air which is carrying out this dilution with the cloud base level air or air at the observation level.

Another mechanism which is considered that might lead to the enhanced concentrations in PSDs at submicron sizes, is the growth of particles smaller than 0.1um (outside the psd region) into the PSD size range from below (mechanism 2). No evidence is presented to support this kind of modification even though some information to support this might be available. For example, little use is made of the CN concentration data which includes a measurement of concentrations at sizes less than the 0.1um PSD lower size cut-off. How do these CN concentrations compare with the summed up concentrations in the PSD up to the size range where they stop overlapping (which the authors suggest is around 3um) in the different cases (where there is and there is not any aerosol modification)? In observations where enhancement at submicron PSD sizes is observed, how does the ratio of this summed up PSD concentration to the CN concentration vary? If the aerosol smaller than 0.1um are simply growing into the PSD (in the absence of new particle formation) the ratio might not be expected to change. There are a number of other simple tests like this that could be tried to help to illuminate the processes occurring.

In this context, it might also be helpful to consider a 5th kind of modification pattern, where both sub and super micron parts of the spectrum are seen to be enhanced simultaneously. Later in the paper (p7804, lines 22-24) it is said that this kind of modification of the spectrum is often observed, and further that sub-micron enhancement almost never occurred without enhancement in the super micron particle volume.

The authors emphasise the importance of analysing the volume concentrations when examining PSD modifications (section 2.3, pp 7800 and 7801) for a number of important reasons. However, when illustrating the four modification patterns observed the authors present PSDs as area distributions (dA/dD) in Figure 3. Why is this?

Section 3 – Results, (line 23 page 7802): the criterion for selecting cloud systems for study was that they should be no less than 10km apart so that far field observations contained no "recently" processed particles. How recent is "recent"? How long might it have been since a cloud dissipated, and how long might it take air to travel between systems?

Page 7804, line 6: re: size distributions classified as homogeneous mixing with boundary layer air. What sort of variation was observed in CN, PSD concentrations and PSDs in far field "ambient" or background air?

Section 4, Discussion, page 7804, paragraph ending on line 21: regarding interactions with a layer of AP advected from a different source than the cloud base AP. This is ruled out as a source of modification because the far cloud environments always have lower AP concentrations. As in earlier comments, is this only a comparison with concentrations at similar altitudes or is some consideration given to variations in concentrations in the vertical? There really needs to be some comment about variations in the vertical, and about vertical transport, even if this just states there were no such variations

P7805, line 1: "larger water droplets will have more sulphate in them as a result of the scavenging of SO2" - yes, but most probably relative to the original sulphate mass present this may not be as significant as the mass gain in drops with smaller initial sulphate mass, where the absolute gain will be less, but the fractional gain more. This will be especially important in terms of their activation properties.

P7805, paragraph 1, lines 4-6: Flossmann's theoretical result might be supported by observational results from EPIC, but has any attempt been made to model the observed changes in the EPIC spectra using observed data as input (with some sensible guesses of the chemistry and gas phase loadings - have measurements of aerosol composition and gas phase species concentration been carried out in the EPIC region before or since, which could be used for this purpose?)? To do this there would also need to be information about the aerosol spectrum in the size range $<$ 0.1um (from where activated particles will grow into the observed PSD, or be scavenged from) which I guess is not available. However, this information is also needed in order to be able to be sure that the EPIC observations are supported by the previous modelling exercises.

P7805, paragraph 2 lies 7-10: here estimated mass enhancements are calculated assuming particles are ammonium sulphate, while earlier it was assumed they were wholly NaCl - which is it to be ?

Section 5, p7806, line 17: "sulphate mass gain by scavenging sulphate particles" if this is the cause for enhancements at submicron sizes, then this must refer to scavenging of aerosol $<$0.1um ie from outside the PSD range, else there would be no net sulphate gain.

Paragraph 3, page 7799, last sentence: The common inlet cut-off size is approximately 1um. Is this the 50% cut-off size or what? Clarify.

Section 2, page 7799: Details are presented about instruments which are not discussed elsewhere eg CCN, PSAP and nephelometer. Are these details left over from the earlier incarnation, before the paper was split into two papers.

Technical corrections:

Typo: page 7803, line 22, final word: "particle" should be plural "particles".

Typo: P7805, line 11 : figures 6(a-d) are incorrectly referred to as figs 5(a-d) in this paragraph

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