

## ***Interactive comment on “Aerosol optical properties relevant to regional remote sensing of CCN activity and links to their organic mass fraction: airborne observations over Central Mexico and the US West Coast during MILAGRO/INTEX-B” by Y. Shinozuka et al.***

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We thank both referees for their careful review and helpful comments. Our responses are given below in italic font. Response to the anonymous referee This is an important paper describing the observed relationship between CCN activity, bulk aerosol composition and aerosol optical properties using in-situ measurements over two separate regions in North America. The authors suggest that their results may be useful for

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predicting the indirect effect of aerosol on climate using remote-sensing observations. This reviewer recommends that the paper be published with minor revisions. Some people use  $d_{50}$ , some use  $D_d$ . I have not seen people use “ $D_{dc}$ ” to describe the threshold activation diameter. “ $D_{dc}$ ” has been replaced with “ $D_d$ ”. For clarity, “chemistry” should be replaced with “chemical composition” or just “composition”, where appropriate. “chemistry” has been replaced with “chemical composition” throughout the text. I think it would be helpful to include the time of day of the measurements and the altitude range. The following has been inserted to Section 2.1 Experiments. “Most measurements were made between 3 – 5 km GPS altitude in local afternoon.” (over Central Mexico) “Most measurements were made below 6 km GPS altitude in local late morning and afternoon.” (over the US West Coast). Introduction Pg 12522 Ln 10-21 : You explain how chemistry influences CCN, but cite no evidence for how it influences optical properties and remote sensing (as posited in the first sentence of the paragraph). In order to be concise, we have quitted positing. The first sentence now reads “Because aerosol chemical composition influences CCN activation it can impact the satellite remote sensing of CCN concentration.” The chemical composition does influence the aerosol optical properties through refractive index, but a bigger influence comes from its association with certain sizes (e.g., dust tends to be larger than sulfate particles). Pg 12522 Ln 20-21 : “: : :the uncertainty in the threshold diameter would hamper estimation of CCN concentration.” The threshold diameter is not the only necessary parameter for accurately predicting CCN. Aerosol mixing state should also be mentioned, especially in the context of a polluted environment like Mexico City. The last sentence of the third paragraph has been modified as following, to indicate that the threshold diameter is not the only parameter relevant to the CCN concentration. “ Thus, even if the size distribution of aerosols were accurately retrieved and mixing state well understood, the uncertainty in the threshold diameter would hamper estimation of CCN concentration.” Pg 12522 Ln 12-15 : “: : :determined by the solubility and surface tension of the particles: : : These chemistry-dependent parameters, expressed collectively as a single hygroscopicity parameter: : :” The way this is written makes it seem

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that surface tension is contained in the definition of the kappa-parameter. This is not correct. In actuality, these two properties are physically distinct from each other, and present themselves differently in the equations. I think it is important to be clear about this. If surface tension is not what you think it is, then your estimate for hygroscopicity will also be wrong. The reference to Petters and Kreidenweis [2007] has been moved to Section 3.2, and more explanations on our treatment of  $\kappa$  have been added. We have taken Sachin Gunthe's advice: In other words, the  $\kappa$  values derived this way have to be regarded as "effective hygroscopicity parameters" that account not only for the reduction of water activity of the solute ("effective Raoult parameters") but also for surface tension effects (Petters and Kreidenweis, 2007; Rose et al., 2008b; Pöschl et al., 2009; Gunthe et al., 2009). Experiments and Instruments Pg 12523 Ln 12: "Central Valley" add ": of California" The words added as suggested.

Pg 12523 Ln 16: "pass" replace with "transmit" Text modified as suggested. Pg 12523 Ln 20-22: ": : for a few exceptional cases with heavy dust or sea-salt concentrations encountered during the experiments, the data are more uncertain than for the rest of the periods." add ": : because a significant fraction of CCN could be lost in the inlet." or something to clarify this statement. The following has been added: "the scattering coefficient may be underestimated and its wavelength dependence overestimated". Because particles in the accumulation mode are barely affected by the inlet loss, the error in CCN concentration is much less significant than that in scattering. Pg 12524 Ln 1-2: "whose refractive index is 1.59" change to "with a refractive index of 1.59" Text has been modified as suggested. Pg 12528 Ln 3-5 "The "growth" factor for dry ( $\sim$ 20% RH) particles was usually found to be : :." A bit confusing as written, since the growth factor is always determined for initially dry particles. For clarity, write "for dry particles also exposed to  $\sim$ 20% RH in the conditioning section" or something like that. "for dry ( $\sim$ 20% RH) particles" has been replaced with "for dry particles exposed to  $\sim$ 20% RH in the conditioning section". Pg 12528 Ln 21: ": : Ddc is determined by seeking consistency between the simultaneous measurements of the total CCN concentration and the dry aerosol size distribution" Replace "determined" with "estimated". The only

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way to know Ddc for sure is by varying the dry particle size transmitted to the CCNc and simultaneously monitoring the total number of particles (both activated and unactivated) to get an activation curve. Externally-mixed nonhygroscopic particles will bias Ddc when estimated without size-resolved CCN data. "determined" has been replaced with "estimated". Pg 12528 Ln 23-24: "To illustrate this approach, Fig. 2a compares the CCN concentration at 0.15-0.20% supersaturation and the OPC number integrated from 100nm: : ." Is the DMA size distribution NOT used for this analysis? Since the smallest size the OPC reportedly measures is 100 nm, and the size distribution is often quite steep at this size, and the threshold diameter for CCN at 0.2% is also right around this size, it seems like a big mistake not to use the DMA size distributions to characterize the CCN. If I am correct that the DMA data was not used for this, can you explain why not? In that case, I am also unclear as to how Ddc is calculated from the CCN data (especially when Ddc is apparently smaller than 100 nm)... The following sentence has been added: "For threshold diameters smaller than 100 nm the DMA distribution was combined with the OPC." Because the threshold diameter at 0.2% is near 100 nm, we use the OPC only for the illustration. Pg 12529 Ln 4: ": : as if particles are homogeneously mixed" Explain what this assumption means in terms of the kappa-parameter. For instance, if there is a significant fraction of nonhygroscopic particles at 100 nm (which is quite possible, especially in a place like Mexico City, which has both dust and various types of local pollution) – then you will overestimate Ddc and thereby underestimate the amount of soluble material in the population of particles that does activate. Not everything about the aerosol composition can be lumped into one parameter, since, physically, different things can be happening at the same time. For instance, externally-mixed soot particles can have a fairly consistent diurnal variation controlled by primary emissions, while internally-mixed organic components can have a distinct diurnal variation based on photochemistry: : : Again, without identifying these independent factors, CCN prediction becomes much less precise, and more importantly, the mechanisms driving changes to the CCN distribution are much more difficult to tease out. The following has been added. "It should be noted that

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Dd is overestimated if there is a significant fraction of nonhygroscopic particles above the diameter. A better way to know Dd is by varying the dry particle size transmitted to the CCN counter and simultaneously monitoring the total number of particles (both activated and unactivated) to get an activation curve, though this is difficult to carry out on a fast-moving platform and with our available instrumentation. Meanwhile, as aircraft survey large regions, they encounter a diverse mix of aerosol characteristics. Our intent is to capture approximated features to characterize dominant tendencies.”

Pg 12529 Ln 13: “: : reflecting the fact that aerosols of identical chemical properties: : :” It should be mentioned that at higher supersaturations smaller particles will activate, and these particles may have a substantially different chemical composition and mixing-state, so this technique can only be applied for a small change in supersaturation. Since the 0.05% supersaturation difference is barely outside the supersaturation uncertainty, I’m not going to make a huge deal out of this. A note on the applicability of this technique has been added as advised.” Note this technique can only be applied for a small change in supersaturation. At higher supersaturations smaller particles will activate, and these particles may have a substantially different chemical composition and mixing-state.”

Pg 12529 Ln 25-27: These last two sentences are confusing and do not seem to add to the discussion of measurement uncertainties: : : The following sentence has been brought forward for a more orderly description of our variability assessment. “The variability in the aerosol number during the course of each 30 second measurement time period is similar or smaller [than the effect on the aerosol number of sizing error]. The CN counts indicate that it is 20% in number, which corresponds to a ~15% change in Ddc.” The overall precision including this effect is 25 – 30%, increased from 20 – 25%. “indeed not significantly smaller than 40%” has been replaced with “about 30%” to be more precise. The subsequent sentence now reads: “This implies that, any fixed value assumed for CCN critical dry diameter over the wide geographical region without consideration of the chemical composition is associated with a typical error of no less than about 30%.”

Pg 12529 Ln 29: “This indicates that the natural variability in the critical diameter: : :” Is it possible that some of the variability in Ddc

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is due to the intermittent presence of externally-mixed nonhygroscopic particles, which are not taken into account? Yes, it is. And this effect is smaller than our measurement uncertainty (~20%).” the possible intermittent presence of externally-mixed nonhygroscopic particles“is now mentioned in Section 3.3. Also see our response regarding Pg 12535 Ln 13-15 below. Pg 12531 Ln 12-13: “For very hygroscopic aerosol similar to sodium chloride the hygroscopicity parameter [kappa] is 1: : :” 1.0 is not a limit for kappa (e.g. kappa for NaCl is 1.3). See Petters and Kreidenweis [2007]. Also, many people would say that ammonium sulfate is a “very hygroscopic aerosol”, but kappa for AS is only 0.6. True. This part of text now reads “For very hygroscopic aerosol similar to ammonium sulfate and sodium chloride the hygroscopicity parameter  $\kappa$  is near 1 (Dd ~ 70 nm) and for low hygroscopicity typical of many organics  $\kappa$  is near 0.1 (Dd ~ 151 nm).”.

Pg 12531: It should be mentioned that Ddc estimated from hygroscopicity studies can be biased because of slightly soluble compounds (like Calcium Sulfate, found in dust, and some organics) that may not dissolve until the aerosol particle grows nearer to its critical diameter. Also surface active species behave differently under dilute and concentrated conditions. Therefore determination of kappa from CCN measurements is typically preferred for prediction of CCN.

The following has been added at the beginning of Section 3.2: “, though slightly soluble compounds and surface active species may behave differently between the two humidity domains”

Pg 12532 Ln 7-8: “: : black carbon and dust comprised only a small fraction of the submicron range in and around Mexico City”. Although the mass fraction of black carbon for submicron aerosol may be small for this study, I suspect that the number of black carbon particles can be significant (since the mass distribution for black carbon peaks at 200 nm). Even if on average only 10-20% of the particles at 100 nm are soot or dust particles, this could directly translate into a significant bias (not a random error) in the estimated Ddc. High loadings during dust events or interception of primary

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emission plumes may also contribute to the observed variability in Ddc.

This point on the number concentration is true, and is now noted in the first paragraph of Section 3.1, after this reviewer's advice (see above). In the text here regarding the mass, "a small fraction" has been modified to "a small mass fraction" to be clear.

Pg 12533 Ln 7-8: " : :the lack of systematic biases means the average Ddc is neither significantly overestimated nor underestimated" I'm not convinced of this, for the reasons outlined previously (externally-mixed particles and slightly soluble compounds would both tend to overestimate Ddc).

"The geometric mean Dd may be systematically biased because of our inexact assumption of homogeneous internal mixing as mentioned before, but hardly because of instrument errors."

Pg 12533 Ln 16: "chemical component" Change to "chemical composition"

Changed as suggested.

Pg 12533 Ln 16-19: "This perhaps reflects the more diverse sources of particles (Central Valley pollution of urban and agricultural mix, Asian fossil fuel and biomass combustion, and possibly ocean surface), sampled over and off the US West Coast." Does not Mexico City also have urban, Asian and marine influences: : :

True, but over Central Mexico, Asian and marine influences are minor compared with urban/industrial/biomass combustion processes. "compared to Central Mexico's air predominantly influenced by local combustion processes"

Pg 12533 Ln 16-19: "In fact, the hygroscopicity tends to be higher for samples from California than those from the State of Washington for OMF greater than 0.6." The point of this statement escapes me. Please clarify.

", indicating a wide variety of organic particles present over the West Coast" has been added.

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Pg 12533 Ln 24-25: "The organics sampled in Asian plumes over the West Coast were essentially all aged and oxygenated (OOA-I): : ." You should cite here the paper that describes these terms and the technical detail behind them.

Aiken et al. [2008], DeCarlo et al. [2008], Dunlea et al. [2008] and Ulbrich et al. [2009] are now cited. Also, in the AMS community we have recently abandoned the terminology "OOA-I" and "OOA-II" in favor of "OOA-1" and "OOA-2" (so Arabic instead of Roman numerals). This change has been implemented in the manuscript.

Pg 12533 Ln 26-28: "It is possible that this is due to some potassium in the particles in Mexico due to biomass burning, which is excluded from the calculation of OMF." I suspect that "this" is referring to the apparent discrepancy between the type of organics measured in the two regions and the hygroscopic growth estimated for the two regions. But this whole paragraph could be rewritten/reorganized to be clearer. Now it reads: "It is possible that the apparent discrepancy in the hygroscopic growth for the 50-nm particles between the two regions is due to some potassium in the particles in Mexico due to biomass burning, which is excluded from the calculation of OMF." Pg 12534 Ln 19: "decreases more slowly" The time-rate of change of hygroscopicity is an important subject. However, this is not what is being presented in the current study. I suggest using the words "is less sensitive to". Changed as suggested. Pg 12535 Ln 7-8: If the "natural variability" in  $\kappa$  is 0.1-0.2, and  $\kappa$  is on average 0.2-0.4, then that translates into as much as 100% variability on average, which translates into a 26% variation in Ddc (at lower OMFs). It is the RMS of the difference in  $\log_{10}(\kappa)$ , not in  $\kappa$ , that turned out to be 0.1 – 0.2. Our original manuscript did state our calculation correctly, but in a diffuse manner. To be more concise, our revised text gives numbers for the derived relative variability only. "For the calculation of relative variability, fit lines for  $\log_{10}\kappa$  instead of  $\kappa$  were made against OMF (see Figure S4 and S5, Table S2 of the online supplement). Using the root mean square of the differences in  $\log_{10}\kappa$  between this new fit and the individual data, we represent the apparent relative variability in  $\kappa$  by  $(10\text{RMSlog-1})^*100\%$ . The variability is 25 – 50% depending on the dry particle

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diameter ( $\sim 35\%$  for 100-nm particles), a range expected from the RMS of the linear fit (Table 1). This  $\kappa$  variability translates into an 8 – 14% variation in  $D_d$ ." Similar modification has been made in the second last paragraph of Section 3.3, the discussion on the variability of  $\kappa$  derived from the direct CCN measurements. Pg 12535 Ln 13-15: "In other words, the CCN activity of particles does not noticeably vary among ionic species and among organic species, or with their state of mixing." You have not shown this to be true. For instance, you do not measure the mixing-state, and therefore cannot say whether or not it is changing during the times that you are sampling. Some results from Mexico City show a strong diurnal cycle in externally mixed nonhygroscopic particles, with highest fractions late in the evening and early morning. If the flights always took place during the mid-day, then this variability may have been missed. It should also be mentioned that these results may not apply at higher supersaturations, with activation of smaller particles that are more likely to vary in concentration and composition. This sentence has been modified, and more explanations added: " That means, as far as the aerosols sampled from our aircraft are concerned, the CCN activity of particles does not noticeably vary among the ionic species, organic species or mixing states (e.g., the possible intermittent presence of externally-mixed nonhygroscopic particles). As detailed in DeCarlo et al. [2008], our aircraft sampled pollution particles of a wide age range, including fairly fresh ( $<1$  day old) urban pollution, aged (1 – 5 days old) outflow and biomass burning particles. The mass ratio of sulfate to all non-refractory ionic species spanned widely (between 0.1 and 0.8 for the time periods assessed in the present paper), owing to the various sulfate sources including volcanoes and petrochemical/power plants compared to mainly urban sources of nitrate. Existence of numerous organic species of different origin has been revealed by the AMS. The mixing state, though not directly measured throughout the experiment, might also be highly variable. It is very useful that the OMF constrains  $D_d$  to such a small range of values for a collection of aerosols of unknown detailed chemical properties and state of mixing. The results presented here may not apply at higher supersaturations with activation of smaller particles that are more likely to vary in concentration and composition." Pg

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12538 Ln 24-26: "In contrast, that calculated for OPC particles up to 750 nm, which roughly corresponds to 1  $\mu\text{m}$  aerodynamic diameter, varies between 2.3 and 2.5 (thin triangles in Fig. 5a), a range narrower than observed. Hence the Angstrom exponent in this geographical region is determined by coarse particles: : :". I do not understand this. From the caption of Fig 5a, the "thin triangles" represent calculations for the sub-micron pollution particles only, whereas the "thin squares" represent the total aerosol (including both submicron and coarse particles). Yet from the text it appears that the "thin triangles" represent the full size range (0.1-1  $\mu\text{m}$ ): : : Please clarify. 0.1 – 1  $\mu\text{m}$  is submicron, not the full size range. To clarify this, we have added "(particles larger than 1  $\mu\text{m}$ )" in the paragraph. Pg 12540 Ln 23-24: "Satellite retrievals of these optical properties would be helpful". I think it would useful to add "in cloud-free, but not necessarily dry, regions" or something like that. "in cloud-free regions" has been added. We could certainly also add "not necessarily dry", but this statement would have to be accompanied by detailed analyses on humidity responses of aerosol scattering, a topic this paper did not elaborate. Acknowledgements Pg 12542 Ln 25: "PDF" should be "PFD" Corrected. Figures. Pg 12554 It is not completely clear whether or not only a subset of the HTDMA-derived kappa values (only at high angstrom coefficients) are plotted in Figs 3c and 3d (as in Fig 3b). In the caption, "tabulized" should be "tabulated", and I believe "horizontal axes" describing the Petters and Kreidenweis tabulated values should be "vertical axes". Is this data an average for all flights? The referee is correct on all counts. For Figure 3c, the following has been added: ", for all data regardless of Angstrom exponent", "horizontal" has been replaced with "vertical", and "tabulized" has been replaced with "tabulated". If "this data" refers to our aircraft-based data as opposed to Petters and Kreidenweis's, then, yes, the large dots represent the average from all flights where valid AMS and TDMA data are available (i.e., Flights #3, 4, 7, 9, 11 and 12 of MILAGRO (Central Mexico), all 12 flights of IMPEX (the West Coast)).

Pg 12555 Can we get more tick labels and/or ticks on the right axes (preferably in the range of the data presented), so that we can see what  $D_{dc}$  we're looking at?

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As suggested, tick labels have been added on the right axes of Figures 2(c,d), 3(c,d) and 4 as well as the supplementary figures.

Response to Dr. Gunthe General Comments: In the present work authors have examined relationship of chemical composition to cloud condensation nuclei (CCN) activities and optical parameters during the MILAGRO/INTEX-B aircraft campaign. The measurements were carried out over the Central Mexico City and the west coast of United States. The manuscript is carefully prepared and well written considering the size of the data set reported. The data appear to be of high quality and of high relevance for atmospheric science studies and manuscript is within the scope of Atmospheric Chemistry and Physics (ACP). I recommend publication in ACP after the following points have been addressed.

Specific comments: 1) Introduction 1.1) I would suggest to include more references to other recent studies of CCN and aerosol optical properties applying similar and complementary methods (e.g. Wang et al., 2008, Sorooshian et al., 2008, Kuwata et al., 2008, Bougiatioti, et al., 2009, Rose et al., 2008b, Garland et al., 2008, Andreae, 2009, Kinne, 2009 and references therein)

All suggested references are now included in our manuscript.

1.2) Could you strengthen your argument on page 12522, line10 “Because aerosol chemistry: : : : . impact the satellite remote sensing of CCN” with appropriate references?

That part of the sentence was neither defensible nor relevant, and has been deleted.

1.3) Page 12522, line 12-16: Following up on the comment of Referee#1 (page C3176, point number 3 under specific comments) I would suggest to clarify the use and distinction of  $\kappa$  as an “effective hygroscopicity parameter” (assuming surface tension of pure water) as opposed to an “effective Raoult parameter” (with variable surface tension). For more information see Gunthe et al. (2009 Sect. 3.3, page 3836), Pöschl

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et al. (2009, page 65) and references therein. Note that the following clarifying statement will be added in the revised version of Gunthe et al. (2009. Sect. 2.2). “The kappa values derived from CCN measurement data through Köhler model calculations assuming the surface tension of pure water have to be regarded as “effective hygroscopicity parameters” that account not only for the reduction of water activity of the solute (“effective Raoult parameters”) but also for surface tension effects (Petters and Kreidenweis, 2007; Rose et al., 2008b; Pöschl et al., 2009)”.

The text has been modified following this suggestion. See our response to the anonymous referee above.

2) Experiment/Instrumentation 2.1) Please specify how pressure compensation was managed for all the instruments, in particular for the CCN which is highly sensitive to pressure changes (Lance et al., 2006, Rose et al., 2008a)

For all instruments the pressure was “near-ambient pressure”, which is now noted in the first sentence of Section 2.2. Our record shows the difference from the ambient air was almost always 0 – 10%.

For the CCN counter, since there was no constant pressure controller, CCN data was filtered using only straight and level flights when ambient pressure was constant. This was noted in the instrument section. Additionally, the following sentence is now included for clarification: “The instrument supersaturation has been calibrated over the full range of pressures experienced during the campaigns.”

2.2) Please specify exactly which type of CCN counter was used (commercially available instrument from DMT)?

“manufactured by Droplet Measurement Technologies (Boulder, CO)” has been inserted in Section 2.2.

2.3) Page 12526, line 29: 0.18% supersaturation is not at the lower end of the range of the supersaturation in tropospheric clouds. In stratus clouds much lower values are

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typically found. Please reframe the statement.

We now say “. . . $0.18 \pm 0.09\%$ , which corresponds to the lower end of the peak supersaturation in convective clouds”.

3) Results 3.1) I would suggest including also the numbers of the estimated critical dry diameter from the US west coast measurements for orientation (page 12530, line 7)

The revised text gives the numbers of the estimated critical dry diameter from this region. “This conclusion from our data over Central Mexico appears to apply to those over the US West Coast as well, though the smaller number of data points makes us somewhat less confident. The largest 90% of  $D_d$  derived for the US West Coast forms a lognormal distribution centered near 100 nm with a geometric standard deviation near 1.4. The rest forms a prolonged tail over small diameters (15 – 40 nm), a feature not evident in the Central Mexico data (Figure 2b), which further discourages the use of a single fixed value of critical dry diameter.”

3.2) Page 12531, line 13, I would suggest saying  $\beta$  is close to 1 or something similar. We have modified the text as suggested. See above.

3.3) Page 12534, line 10, Please correct that AMAZE-08 tower was  $\beta$ 39 m tall (not 110 m) Corrected. 3.4) I would suggest testing the  $\beta$ -OMF analysis/regression also on linear scale (in analogy to Gunthe et al., 2009). According to Petters and Kreidenweis, 2007,  $\beta$  should depend (near) linearly on chemical composition (mass or volume fractions). It may be worthwhile to compare the goodness of fit ( $R^2$ ) on linear and log scales.

We have made the vertical scale of Figure 2(c,d), 3(c,d) and 4 linear for the discussion of the OMF dependence. The fit was made through the bin averages, and its goodness against the individual data points was measured by the RMS of their difference (Table 1). Our previous note on our preference of logarithmic scales (section 3.3) has been deleted. We keep the previous, log-based analysis in the online supplementary mate-

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rial, noting the following in the main text: “For the calculation of relative variability, fit lines for  $\log_{10}\kappa$  instead of  $\kappa$  were made against OMF (see Figure S4 and S5, Table S2 of the online supplement). Using the root mean square of the differences in  $\log_{10}\kappa$  between this new fit and the individual data, we represent the apparent relative variability in  $\kappa$  by  $(10^{\text{RMS}\log_{10}\kappa} - 1) * 100\%$ . The variability is 25 – 50% depending on the dry particle diameter ( $\sim 35\%$  for 100-nm particles), a range expected from the RMS of the linear fit (Table 1),”

3.5) Page 12534, line 28 I would suggest calculating CCN number concentrations for different supersaturation levels assuming different values of  $\beta$ . It would be interesting and helpful to see if you get a stronger dependence on  $\beta$ , or if the variability is still totally being dominated by the variability of aerosol particle number concentration and size distribution. I feel this test will help to strengthen your claim. In addition please keep in mind that apart from number and size the available conditions of the CCN activation (amount of water vapor) tend to reduce the influence of  $\beta$  even further as demonstrated by Reutter et al., 2009. DeCarlo et al., 2008 have shown size distributions obtained from AMS measurements. It is apparent from those size distributions that variations in modal size are in the range of factors of 2-10. Clearly such variations would have a much stronger effect on CCN efficiency than composition variations of a factor of 2-3. The only way to really make a quantitative statement is to do a test along the lines of Dusek et al., 2006 where one compares the effects of using a fixed mean kappa and varies the size distribution using observations, and on the other hand, applies a fixed mean size distribution and varies kappa. You can then compare which better explains the observed variability.

The present manuscript does not compare size and chemistry. It compares our aircraft observations and previous ground-based ones, focusing on the chemistry (without a mention of the variation in size distribution). This may be different from the impression our reference to “size matters more than chemistry” may have given. That mention has been deleted.

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We agree that varying the size distribution and  $\kappa$  independently within the observed range of values is a proper way to discuss “size vs. chemistry”. We leave analyses on the variation in size distribution for a separate paper [in preparation]. (The first author originally included them in the present manuscript, but some of the co-authors advised him not to for enhanced readability.)

The second full paragraph after equation (4) now notes: “The wider range of OMF, 0 – 0.8, for our research areas (particularly the US West Coast) means that aerosol composition will be more critical for estimation of CCN concentration than at the fixed sites previously studied (Dusek et al. 2006; Gunthe et al., 2009). As a crude estimate of the sensitivity of CCN concentration to aerosol chemical composition, the integral of our US West Coast size distributions from Dd is typically twice as large for Dd = 90 nm ( $\kappa \sim 0.5$ ) as that for Dd = 140 nm ( $\kappa \sim 0.1$ ). These critical diameters correspond to OMF of 0 and 0.8, respectively, for the dry 100-nm particles. The variation in the CCN concentration estimated in this manner for the US West Coast would not be as large as a factor of 2 but near 30%, if the OMF did not go below 0.6 (Dd = 120 nm,  $\kappa \sim 0.2$ ).”  
Technical corrections: Page 12521, line 7: Did you mean ..100-nm particles decreased with increasing organic: : : Yes. Rewritten as suggested.

Page 12521, line 8: OMF is accurately described here, on page 12532, line 4: Replace organic mass fraction (OMF) with OMF Replaced as suggested.

Page 12522, line 23: Replace US with United States Replaced with “United States (US)”.

Voluntary changes Section 1. Introduction The fourth paragraph. “a potential remote-sensing method” has been replaced with “potential remote-sensing methods”. Section 2. Methods. The seventh paragraph. The first two sentences have been connected for brevity. “A prototype 3-wavelength Radiance Research particle soot absorption photometer (PSAP) continuously measured aerosol light absorption by monitoring the change in transmittance across a filter using an alternating 3-wavelength (470,

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530 and 660 nm) LED.” “<” has been inserted to “0.02” in describing the estimated uncertainty in measured SSA. 4.1.1 The first paragraph. “groud” has been replaced with “ground-based”. 5. Conclusions To be concise, “weaker dependencies recently found for” was deleted from the last sentence of the first bullet, right before “fixed-point studies elsewhere”. The last paragraph. “reasons” has been replaced with “regions”.  
Acknowledgments We now thank several more scientists for their recent reviews. References Citations for Rogers et al. [2009] and Yokelson et al [2009] have been updated.

Please also note the Supplement to this comment.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 12519, 2009.

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