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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical composition and sources of coastal marine aerosol particles during the 2008 VOCALS-REx campaign" *by* Y.-N. Lee et al.

Y.-N. Lee et al.

phdaum@bnl.gov

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Major Comments: The paper is filled with interesting data and conclusions. It contains useful information about aerosol particles in the region that should be published. I applaud the authors on collection and analysis of the data set. However, the presentation needs work, and the paper as written is very long. It is organized in small chunks, which is helpful, but please be more concise to decrease the overall length. Several of the 26 figures could likely be removed also. Some of the arguments are hard to follow; most of these are mentioned in the Minor Comments. There is also some awkward grammar and there are many spelling errors. Some, but not all of these are flagged be-

low. The whole manuscript would benefit greatly from running a spelling and grammar check.

Reply: The manuscript has been subjected to numerous minor changes to correct spelling errors, improve grammar and to make the manuscript read more clearly. The number of Figures has been reduced from 26 to 16 and the manuscript has been shortened from 73 pages including figures to 50 pages including figures.

The Abstract is too long and reads like rapid-fire bullets of every specific thing that was observed, rather than a summary of the work and results. It needs to be rewritten.

Reply: The abstract has been shortened.

Section 4.6 and elsewhere: Many inferences about processes are made based on small differences in vertical profiles of various chemical components from LOWESS fits (a form of smoothing). There is considerable scatter in the actual data, however, and the "goodness" or significance of the LOWESS fits do not seem to be evaluated–so how do we know that these small differences are actually meaningful? Please evaluate and discuss.

Reply: Lowess fits are simply lines that represent fits to the local median of the data. They are meant to reveal trends in the data that may not be obvious from scatterplots. No assumptions are made in Lowess fits about the underlying dependencies (unlike a linear least squares fit) and it is thus not possible to say anything about the "goodness" of a lowess fit.

Minor Comments: Abstract Line 13-15: Awkward text: "Sea-salt aerosol (SSA) particles, represented by NaCl, showed Cl− deficits caused by both HNO3 and H2SO4, and were externally mixed with SO42− particles as the AMS detected no NO3− whilst uptake of HNO3 occurred only on SSA particles." If the Cl- deficit is partly due to H2SO4, then sea-salt and sulfate must be internally mixed. I think you mean they are externally mixed as well? If you break this up into two sentences, it might be more

clear.

Reply: We agree with the reviewer, see the revised abstract.

There is jargon in the Abstract without defining it: for ex, “LOWESS fits”, HOA, OOA p 26045

Reply: Acronyms defined before used.

Line 23: aerosol is misspelled.

Reply: Corrected

p 26046 Line 1: suggest inserting “originally” or “predominately” before “zonal”.

Reply: Suggestion taken

For clarity, some of the very long sentences should be broken upâĂŤfor example, the one starting “Since satellite data. . .”on line 5.

Reply: Revised text is as follows- “Satellite data shows a strong increase in the cloud droplet effective radius with distance from the coast [Wood, et al., 2011]. This implies a higher aerosol number concentration near the coast and it is important to understand whether the source of this aerosol is anthropogenic or biogenic.”

Commas are needed throughout between phrases–for example, after “upwelling” on line 3 of this page. Another example is on p. 26058, line 8 after “species”, and another on p. 26059, line 17 (after “bursting”), but there are many more.

Reply: Commas added as appropriate

Line 23: insert “from” before “shipboard measurements”. p. 26050

Reply: Suggestion taken

Line 6: SI abbreviation for gram is “g” not “gm”

Reply: Corrected

p. 26051 Line 7: “flight” is misspelled.

Reply: Corrected

Lines 11-12: Are these percentages without the 100 m legs typically made on the eastbound return, then? Or are these counted as “below cloud” even though the clouds may have dissipated?

Reply: The percentages include the eastbound legs.

Lines 21-22: I am not sure that you really are making this assumption, since, for example, some non-sea-salt sulfate is internally mixed on sea-salt, which tends to be the larger of the sub-micron particles. What you are doing (I think) is simply scaling the data based on their non-sea-salt sulfate concentrations. Recommend just deleting the phrase “assuming that nss-SO42- was present only in particles of Dp < 440 nm”.

Reply: Phrase deleted

p. 26053

Lines 15-18: The FT is also likely cleaner because of fewer aerosol (surface) sources directly supplying the layer, which should be mentioned.

Reply: Added the phrase “and because fewer aerosol sources directly supply the layer” at the end of the paragraph.

Line 25-27: This is confusing. If some AC samples are below the inversion, how can they be FT? Do you mean they are within the inversion layer?

Reply: The vast majority of the “above cloud” data was taken in the free troposphere, but there are a few instances where data was taken above cloud, but below the inversion. Since this was a very small amount of data, we did not take the trouble to define an addition class of data “above-cloud, but below the inversion”, but simply included it with the free-troposphere data. Such classification does not materially affect any of our conclusions.

p. 26054 Line 9: “Whiskers” is misspelled.

Reply: Corrected.

p. 26056 Line 25: Not really “total mass concentration”âĂŤqualify size range. Now it seems you are talking about not just the sub 440 nm size range, but the size range measured by the PILS. Please clarify.

Reply: “total mass concentration” has been changed to “mass concentration”. How this is computed is already described “(the sum of all the ionic species measured by the PILS and the Org measured by the AMS)

p. 26057 Line 8: Define Dg.

Reply: Text using Dg deleted, but Dg, physical diameter, is related to Dva by particle density and shape factor.

Line 12: Swap “undetected” with “components” for clearer meaning.

Reply: Suggestion taken

Line 19: un-neutralized is misspelled. Line 24: caused is misspelled.

Reply: Spelling corrected

Last paragraph: This is not clear. Are we talking dry aerosol mass, or ambient, or at the measurement humidity? If you are trying to determine dry mass and the DMA particles are retaining water, then the PILS and AMS do not “underestimate” mass, the DMA over-estimates it. Trying to correct things to the 15% RH of one of the measurements (if that is what you are doing) doesn’t seem meaningful. I suggest you rewrite this discussion, taking the time to make it very specific and to flow properly.

Reply: Discussion in section 4.1 has been rewritten.

p. 26058 Line 26: added to the calculated what?? Line 27: Observed what? I understand what you mean, but please include the nouns. . .

Reply: Changed to- “was added to the calculated conductivity so that the lowest conductivity matches the conductivity observed in the FT”

Section 4.3.1: This section is very longâĂŤcan it be condensed with just main points included? p. 26060

Reply: Section 4.3.1 reduced by $\sim$50%

Lines 24-27: Is particle density included here? Presumably the estimated inlet cut size is aerodynamic diameter.

Reply: Yes, aerodynamic

p. 26061

Line 4-5 “exceeded” is not the right term, since you are talking about the number concentration of SSA vs the total concentration (sub 2 microns). Rephrase to mean that SSA is only 5% of total aerosol, if that is what you mean. I am not sure that this is a “tiny” fraction–but a small fraction, yes.

Reply: Section deleted

Line 16: significant is misspelled.

Reply: Corrected

p. 26062 Line 9: gradient is misspelled.

Reply: Corrected

Line 15: Replace “against” with “of”. Also, I’m confusedâĂŤaren’t these mass concentration for Na+ based on dry mass?

Reply: Section deleted

p. 26064 Line 5-6 has awkward phrasing: “This divergent observation is consistent with not only that the AMS is oblivious of the refractory. . .”. Please rephrase.

Reply: Changed to - " This observation confirms that the AMS does not detect the refractory NaNO3 on SSA and that no detectable HNO3 uptake has occurred on the acidic sulfate aerosols."

Line 7: "no uptake" should probably be "little uptake", since it could be below the detection limit.

Reply: See response to line 6 above.

Lines 7-9: I don't follow thisâĂŤsulfate can become internally mixed with sea-salt through various aerosol or aqueous-phase processes–as discussed on the next page. Perhaps you mean no "primary" sulfate aerosols are internally mixed with sea-salt?

Reply: Changed the sentence starting on line 8 to read- "These observations suggest that SSA and sulfate aerosol remain mostly externally mixed in the MBL consistent with their different production mechanisms (i.e., sea salt is mechanically produced, whereas sulfate is produced by a combination of gas and aqueous phase processes and while aqueous phase production will occur in cloud droplets formed on SSA aerosol particles, the number of SSA particles is very small)."

Line 28: "but none for SO4 2-". None of what? Awkward. Do you mean no PILS samples were below LOD for sulfate?

Reply: Paragraph deleted

p. 26065 Line 2: Twohy is misspelled.

Reply: Reference deleted

Section 4.5: This section is somewhat rambling and should be condensed and reorganized, perhaps starting with the myriad assumptions made and following with the most important results for each species.

Reply: Text has been revised to make it read more smoothly and was considerably shortened.

p. 26066 Line 2: Respectively is misspelled.

Reply: Corrected

p. 26067 Line 14: "1 ppb attains"?? Awkward.

Reply: Sentence deleted

Line 26: gas misspelled. Line 29: considered misspelled.

Reply: Corrected

p. 26069 Lines 7-9: This is not a complete sentence.

Reply: Corrected

p. 26072 Lines 3-20: Some of this discussion of sea-salt variability is repetitive from elsewhere in the paperâĂŤcan it be combined?

Reply: Paragraph shortened to- "Distinct from SO42- and Org, Na+ (representing SSA with Dp $\leq$ 2 μm) exhibited a much more uniform vertical distribution with a small decreasing trend with altitude consistent with a surface source. Unlike continentally derived aerosols which exhibit a strong land to sea gradient, SSA loading in the MBL remained relatively constant with longitude and for this reason the composite vertical profile is less prone to sampling biases. The Na+ vertical profile exhibited a small minimum at $\sim$260 m that corresponded to a small maximum in the profiles of both water mixing ratio and potential temperature, but not in that of SO42- and Org. We believe this apparent "inversion" is an artifact due to the sampling pattern, and does not reflect a true physical layering."

p. 26075 Line 21: "lessed cloud processing"?

Reply: Changed to "a lesser degree of cloud processing"

p. 26076 Line 21: refractory misspelled.

Reply: Corrected

p. 26078 Aitken, not Aitkin.

Reply: Corrected

Lines 22: Why?

Reply: Text added-“ because all things equal, larger particles will be activated before the smaller ones and lost through precipitation scavenging”

p. 26079 Line 11: Insert “Estimated” before “number concentration of SSA”, as SSA was not measured directly.

Reply: Suggestion taken

Figures: Fig 4&5: Individual plots are too smallâĂŤsuggest you lay these figures out in rows of 2 or 3 larger plots to better use the space on the page.

Reply: Suggestion will be taken in final version

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 26043, 2013.