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

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.

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.

## Minor Comments:

Abstract Line 13-15: Awkward text: "Sea-salt aerosol (SSA) particles, represented by NaCl, showed Cl<sup>-</sup> deficits caused by both HNO3 and H2SO4, and were externally mixed with  $SO_4^{2^-}$  particles as the AMS detected no  $NO_3^-$  whilst uptake of HNO3 occurred only on SSA particles." If the Cl<sup>-</sup> deficit is partly due to H<sub>2</sub>SO<sub>4</sub>, 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.

There is jargon in the Abstract without defining it: for ex, "LOWESS fits", HOA, OOA

p 26045

Line 23: aerosol is misspelled.

p 26046

Line 1: suggest inserting "originally" or "predominately" before "zonal".

For clarity, some of the very long sentences should be broken up—for example, the one starting "Since satellite data…"on line 5.

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.

Line 23: insert "from" before "shipboard measurements".

p. 26050

Line 6: SI abbreviation for gram is "g" not "gm"

p. 26051

Line 7: "flight" is misspelled.

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? 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-SO4<sup>2-</sup> was present only in particles of  $\mathbf{D}_{p} < 440$  nm".

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.

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?

p. 26054

Line 9: "Whiskers" is misspelled.

p. 26056

Line 25: Not really "total mass concentration"—qualify 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. p. 26057

Line 8: Define Dg.

Line 12: Swap "undetected" with "components" for clearer meaning.

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

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.

p. 26058

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

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

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

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.

Line 16: significant is misspelled.

p. 26062

Line 9: gradient is misspelled.

Line 15: Replace "against" with "of". Also, I'm confused—aren't these mass concentration for Na+ based on dry mass?

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.

Line 7: "no uptake" should probably be "little uptake", since it could be below the detection limit. Lines 7-9: I don't follow this—sulfate 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?

Line 28: "but none for  $SO_4^{2-}$ ". None of what? Awkward. Do you mean no PILS samples were below LOD for sulfate?

p. 26065

Line 2: Twohy is misspelled.

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. p. 26066 Line 2: Respectively is misspelled. p. 26067 Line 14: "1 ppb attains"?? Awkward. Line 26: gas misspelled. Line 29: considered misspelled. p. 26069 Lines 7-9: This is not a complete sentence. p. 26072 Lines 3-20: Some of this discussion of sea-salt variability is repetitive from elsewhere in the paper-can it be combined? p. 26075 Line 21: "lessed cloud processing"? p. 26076 Line 21: refractory misspelled. p. 26078 Aitken, not Aitkin. Lines 22: Why? p. 26079 Line 11: Insert "Estimated" before "number concentration of SSA", as SSA was not measured directly. Figures:

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