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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigating the sources and atmospheric processing of fine particles from Asia and the Northwestern United States measured during INTEXB" *by* R. E. Peltier et al.

R. E. Peltier et al.

Received and published: 27 February 2008

Anonymous Referee #1

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General Comments:

. . . is probably warranted is in the actual determination of differences between the North American and Asian plumes in Section 3.1. The authors compare median concentrations in plumes from the two continents and say that they are different, but they do not take into account differences in sample size and variance when making this claim. A non-parametric test could be used to show that

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the observed differences between plumes of Asian and North American origin during INTEX-B were indeed statistically significant.

We will address this by including the results from a Wilcoxon rank-sum test, which is a commonly-used equivalent to the parametric t-test statistic.

Specific Comments:

Page 17430, Line 12: Be sure to note that this is only true for the air masses of primarily Asian origin.

We will change this line to specify that this is only true for Asian air masses.

Page 17431, Line 6: Use e.g. in this citation. This is not an exhaustive list.

Agreed. We will add 'e.g.' to the citation list.

Page 17432, Line 10-11: Clarify what this means. Did GEOS-Chem predict correct maximum and minimum concentrations, but in the wrong location?

The GEOS-Chem model, on average, was in agreement with aircraft observations of WSOC in the Northeastern United States. However, the individual observations were poorly correlated (r = 0.21, Heald et al, 2006, JGR), and suggests the model may not capture all of the sources of WSOC. Hence, we will change the last line of this paragraph to:

For the eastern United States ICARTT dataset, the average OC mass (actually, the mass of water-soluble OC) was predicted fairly well by GEOS-Chem, but individual observations were poorly correlated {Heald, et al, 2006}, suggesting a poor understanding of atmospheric formation of WSOC.

Page 17432, Line 26-27: The phrase "Studies focused on WSOC can also reduce the complexity of the carbonaceous aerosol" doesn't make sense. Is this referring to chemical complexity? If so, then this is something that no amount of studying will change. Or is it referring to the seeming complexity of the aerosol

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due to lack of understanding?

We will delete this line in order to clarify the manuscript.

Page 17441, Line 20: Maybe mention here that you will support this claim with satellite data in Section 3.4.

We will amend the manuscript to state this.

Page 17444, Line 7: Can you speculate on why there is such a low minimum WSOC/sulfate ratio at 2 km altitude in the Northwestern U.S. profile in Fig. 3? Was this the boundary layer height?

While this is a possible explanation, we believe the most likely reason for this is found in Fig 2. From Fig 2 essentially WSOC drops off precipitously at about 2km, but we recorded a higher concentration region at 3 to 5 km due to the plumes we discuss later in the paper. Sulfate general trend is constant median concentrations below about 2.5 km and then a steady decreasing concentration above 4 km with increasing alt. Between 2.5 and 4 km the concentration is highly variable, mainly due to a limited number of measurements. This leads to high variability in the WSOC/SO4 ratio in this range.

Page 17444, Line 20: Could you comment here on the limitations of your assumption of cloud-free conditions for SOA and sulfate formation in the previous paragraph, given that conditions during trans-Pacific transport were often cloudy?

We do not claim for this study that SOA and sulfate formation conditions were cloud-free. We referenced Brock et al and de Gouw et al because they provide useful estimates of characteristic time constants for the formation of SOA and sulfate. Our intent in this passage is to convey that SOA and sulfate formation in Asian emissions occurred (either in cloudy or cloud free conditions), both SOA and sulfate aerosol was lost due to precipitation and cloud scavenging during transport across the Pacific Ocean, and

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finally that sulfate formation continued (and SOA formation was limited) which resulted in the observed WSOC/sulfate ratios during this study.

Page 17445, Line 7: Is this because lower-volatility organics have already been lost to particle formation?

This is the most likely explanation. But since measurements of VOCs, SVOCs, or organic aerosol were not conducted during the formation process (e.g. measurements closer to emission source), we won't speculation of specific chemical formation processes.

Page 17446, Line 16: Be sure to clarify that "each flight" only refers to the two mentioned in the previous paragraph.

We will change this line to read:

'For each of these flights, . . . '

Page 17446, Line 17: What you have called the correlation coefficient here is actually the square of the correlation coefficient (r2), otherwise known as the coefficient of determination. It is good to be precise when using these terms, because, although both contain information about the strength of the correlation, only the correlation coefficient, r, contains information about the sign of the relationship. If you did not also include the scatter plots in Fig. 6 or the regression coefficients of those scatter plots in later sentences, the reader would not know whether there was a negative or positive relationship between WSOC and CO or water vapor from the r2 values alone.

This terminology was an oversight. In this paper, we generally discuss the coefficient of determination (or r^2 value), and will change the manuscript to more precisely name the statistic.

Page 17447, Line 1: Section 3.4.3 doesn't exist. Looks like it should be a reference to Section 3.5.3?

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This is a copy-edit issue, which will be resolved during the editorial processing of the manuscript.

Page 17447, Line 9: The phrase "In order to further identify WSOC sources" is probably stretching it a bit... so far the analysis has only identified relationships between WSOC and different variables.

We agree that this statement might be overly presumptuous. We will change the line to read:

'In order to further investigate WSOC sources. . . '

Page 17448, Line 25: Here (and elsewhere in this section) "coefficients" are referred to, with no clarification as to what kind of coefficients these are. It would be good to remind the reader that the discussion is still focused on the _-coefficients from the multivariate linear regression.

In order to clarify this, we have added the term 'multivariate linear regression' before the term 'coefficient' in several locations throughout the manuscript.

Page 17449, Line 5-7: Good! Although your analysis has provided much insight on relationships, it is wise to not over-interpret the results.

Page 17450, Line 23: Be sure to remind readers that "North American air masses" only refers to those that impact the Western U.S. Throughout this section, there are places where "Asian" and "North American" plumes are referred to in general; it would be good to remind the reader that these are still restricted to ones sampled during INTEX-B.

We have clarified this in the text to emphasis that these findings are for plumes that impact the Western US.

Page 17450, Line 24-25: This sentence is too vague. Be more specific by saying something like "variability in acetonitrile contributed little to WSOC variability"

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rather than "acetonitrile contributed no significant influence".

We have changed the line to be more direct:

In the North American data set, WSOC variability was not significantly affected by acetonitrile, indicating a limited influence on WSOC by biomass burning.

Page 17451, Line 19: Refer to Table 4 here so the reader remembers that the coefficient in the first set of parentheses (0.97 $\pm$ 0.13) is referring to the coefficient for methanol.

Reference to Table 4 is added.

Page 17452, Line 7: The phrase "during this plume" doesn't really make sense. Perhaps change to "in the vicinity of this plume" or something similar.

We will change the line to read:

The model accounts for 92.6% of the variability of WSOC during observations downwind of clouds

Page 17453, Line 2: The INTEX-B campaign isn't mentioned anywhere in the conclusions. Maybe reference it in this first sentence.

We will reference the overview paper to the INTEX-B campaign in the introduction. When this manuscript was submitted, the overview paper reference had not yet been issued.

Page 17453, Line 16: Remind the reader what the "particular altitude ranges" were.

We change this line to read:

WSOC concentration was highest at lowest altitude ranges in both North American and Asian air masses, though several enhancements at $\sim$1km and $\sim$2-3km for Asian air masses and at $\sim$ 4km in North American air masses, were observed.

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Page 17453, Line 23-24: This sentence doesn't really describe an exception to the previous sentence, because these plumes represented less-aged air masses compared to the Asian ones. Make sure to clarify that.

Page 17454, Line 6: The phrase "meteorological transformations" could be more specific here, since your previous conclusions focused on cloud processing and scavenging by precipitation.

We will replace '. . . possible meteorological transformations. . . ' to '. . . possible cloud scavenging or precipitation loss. . . '.

Page 17454, Line 10-12: This sentence doesn't make sense. The data, not the air masses themselves, were further refined, followed by a comparison of the cases left in the newly-restricted dataset.

We will replace this line to read:

The North American air mass data observed during this study were further refined to include only Central Valley outflow, and compared a relatively fresh plume (aged 1-2 days) with a somewhat aged (2-3 days) plume that appeared to be recently advected through clouds

Page 17454, Line 13: Change "by multivariate regression analysis" to something like "multivariate regression analysis revealed that both biogenic..." The sentence currently doesn't make sense as is.

We will replace this line to read:

Multivariate regression analysis revealed that both biogenic and fossil fuel combustion VOCs were nearly equally responsible for WSOC variability in relatively fresh plumes.

This reviewer has noted a number of technical changes. Rather that address each one, we will summarily accept all suggestions and make the modifications to the manuscript. There is one exception, addressed below.

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Page 17430, Line 7: change *was* to *were* (subject-verb agreement)

Page 17430, Line 19: change *were* to *was* (subject-verb agreement)

Page 17431, Line 7: delete 306 after 1999

Page 17431, Line 13: *ASIA* should not be in all capital letters

Page 17432, Line 7: *ASIA* should not be in all capital letters

Page 17433, Line 26-27: there are two sets of references here, combine?

Page 17435, Line 22: change *this* to *the*

Page 17436, Line 12: *take off* should be one word or hyphenated

Page 17438, Line 11: comma should be a semicolon

We believe the use of a comma here is appropriate, as we are not introducing a new idea.

Page 17440, Line 5: *it* should be changed to a plural pronoun (to agree with subject)

Page 17441, Line 26: capitalize first letter in *western*

Page 17442, Line 2 and 3: *ASIA* should not be in all capital letters

Page 17442, Line 8: *Trace* should be in all capital letters

Page 17442, Line 16: need comma before at *altitudes*

Page 17442, Line 20: delete extra s before sm-3

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Page 17442, Line 24: change *is* to *was*

Page 17443, Line 12: change *was* to *were*(subject-verb agreement)

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Page 17443, Line 15: hyphenate *WSOC sulfate*

Page 17443, Line 20: need comma before *relative*

Page 17443, Line 22: capitalize first letter in *northeast*

Page 17443, Line 26 and 28: *ASIA* should not be in all capital letters

Page 17444, Line 1: capitalize first letter in *northwestern*

Page 17444, Line 6: *ASIA* should not be in all capital letters

Page 17445, Line 4: capitalize first letter in *northwestern*

Page 17446, Line 14: capitalize first letters in *central valley*

Page 17446, Line 15: capitalize first letter in *aqua*

Page 17446, Line 17: need a period after 0.52

Page 17446, Line 20: change *are* to *were*

Page 17446, Line 29: capitalize first letter in *northern*

Page 17447, Line 22: change *on* to *of*

Page 17449, Line 3: change *includes* to *include* (subject-verb agreement)

Page 17449, Line 11: need comma before *as of yet*

Page 17449, Line 18-19: the sentence beginning with *For example* is a fragment

Page 17450, Line 2: change *is* to *are* (subject-verb agreement)

Page 17450, Line 20: delete comma after *include*

Page 17450, Line 28: capitalize first letter in *northwestern*

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Page 17451, Line 2: capitalize first letter in *northwestern*

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Page 17451, Line 23: capitalize first letter in *northern*

Page 17451, Line 4: capitalize first letters in *central valley*

Page 17452, Line 6: change *that* to *than*

Page 17452, Line 10: insert *the* before *Central Valley*

Page 17453, Line 2: capitalize first letter in *spring*

Page 17453, Line 3: change *attitude* to *altitude*

Page 17453, Line 3: insert *the* before *Western*

Page 17454, Line 5: *can not* should be one word

Page 17467, Table 4: R2 should either be less than one, or expressed as a percentage

Page 17468, Table 5 caption: first letter of *northern* should be capitalized

Page 17468, Table 5: R2 should either be less than one, or expressed as a percentage

Page 17471, Figure 3 ACE-Asia data label: *ASIA* should not be in all capital letters

Page 17472, Figure 4, 4th text box from the left: *tranport* should be *transport*

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