

## ***Interactive comment on “Gas/particle partitioning of water-soluble organic aerosol in Atlanta” by C. J. Hennigan et al.***

**C. J. Hennigan et al.**

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We thank the reviewers for their thoughtful and detailed reviews. In our response to reviewer comments below, reviewer comments are stated and our response follows.

Anonymous Referee #1 Received and published: 15 January 2009 The authors present measurements of water-soluble organic carbon in both the particle (p) and gas phase (g) over 4.5 months in summer 2007 in Atlanta. Based on this huge data set, they correlate the WSOC(p) concentration as well as the partitioning ratio  $F_p$  ( $=\text{WSOC}(p)/(\text{total WSOC})$ ) and the concentration ratio (p/g) to various ambient parameters such as relative humidity, temperature, NO<sub>x</sub> and ozone levels. While previous studies of organic partitioning only focused on selected species (groups), the current approach gives an overall idea of the total WSOC budget and important parameters that affect its partitioning. This approach is certainly highly useful and complemen-

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tary to prior SOA studies in order to understand SOA formation, even though it only provides little information of the chemical processes and speciation of individual compounds. Thus, this study is of high interest to understand SOA formation and falls into the scope of Atmospheric Chemistry and Physics. I have a some thought and comments that should be considered before publication.

1) General comments Section 3.1: Do you have any means to compare also the total masses instead of just the carbon mass of WSOC? The comparison of the slopes of WSOC(p)/(g) to WS organic mass (p)/(g) would give information on the oxidation state of WSOC.

We agree with the reviewer that such an analysis would provide important insight into the nature of WSOC. However, this analysis is not possible with the data set we have collected. Following the work of Peltier et al. (2007) and Turpin and Lim (2001), it is likely that a reasonable estimate of the conversion from WSOCp carbon concentration to WSOCp mass concentration could be made. The problem lies with the conversion of the WSOCg measurement from carbon concentration to mass concentration: we simply do not have enough information about the aggregate species which constitute the total WSOCg to make this estimate without adding a degree of uncertainty which would render the estimation meaningless.

2) Section 3.2.2. What value for the Henry's Law constant would be required to explain the observed partitioning? Such a value should be easy to derive based on the measured WSOC(p), WSOC(g) and calculated aerosol water content. Is it in the order of magnitude as suggested by lab studies by e.g., Kroll et al., 2005; Volkamer et al., 2008, who observed  $KH > 10(7) \text{ M atm}^{-1}$ .

We have performed the calculation and have added to the text the following: This was attributed to the gas phase hydroxyl radical (OH) oxidation of C<sub>2</sub>H<sub>2</sub> to produce glyoxal and its subsequent uptake and reaction in the aqueous phase. Also of note, using 10 μg m<sup>-3</sup> liquid water concentration and an F<sub>p</sub> value of 0.3 (approximately

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equal to the observed  $F_p$  above 90% RH), we calculate an effective Henry's Law constant for the total WSOC of  $\sim 2 \times 10^9 \text{ M atm}^{-1}$ , which agrees well with the experimentally-derived value of Volkamer et al. [2008]. Overall, our ambient results showed a strong correlation;

3) Section 3.2. and Fig 6: What average inorganic aerosol composition is assumed here? How would the water mass change if also the uptake of water by WSOC would be considered? (Some comments should be added in the text).

For the predicted liquid water concentration, we assumed an inorganic composition equal to the average composition observed over an entire month of sampling in August, 1999 (Atlanta Supersite experiment). We used an average summertime composition because we didn't make speciated inorganic aerosol measurements throughout the summer. We have added a clarification to the text, referring the reader to the discussion in Hennigan et al. (2008a) for an explanation of the aerosol composition used in the ISORROPIA calculation. There are two main reasons for including Section 3.2.2, which essentially summarizes the major findings of Hennigan et al. (2008a). First, with the present study, we are attempting to provide as comprehensive an analysis as possible on all factors which affect WSOC partitioning. RH was a factor which had a large effect on  $F_p$ , and we extend the original RH analysis by incorporating the dual RH-WSOC effects (Figures 8, 9). Second, the results of Volkamer et al. (2008) were published after those of Hennigan et al. (2008a) but the similarity of the two studies warranted some discussion. So, a cursory discussion of the Hennigan et al. (2008a) findings is appropriate, but anything beyond that may be redundant. For the second part; how would the water mass change if the uptake of water by WSOC were also considered: based on the ambient concentrations of inorganic and organic aerosols observed in the Atlanta summertime [Weber et al., 2003; Solomon et al., 2003], we expect the water uptake to be controlled predominantly by the inorganic components (this point is made in the Hennigan et al. (2008a) study). This is in agreement with many studies, including Malm and Day (2001), Cruz and Pandis (2000), and

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Prenni et al. (2003). Many studies have investigated the ability of organics to take up water, and the effect can be highly dependent upon the specific organic compounds present. This is illustrated nicely in a study by Saxena et al. (1995), who observed an enhancement of water uptake due to organics in a rural location and a suppression of water uptake due to organics in an urban location (Los Angeles). Because Atlanta is a unique environment, in that it is an urban center where biogenic VOC emissions are dominant, it is reasonable to assume that overall, the impact of organics on water uptake is modest. Given the observations of Saxena et al. (1995), an increase in water uptake on the order of 10-15% due to organics may be expected, though it is important to note that the shape of the water uptake curve is not expected to change appreciably.

4) p. 647, l. 7 ff: The main products of terpene oxidation are less water-soluble (e.g., pinonaldehyde) than those of isoprene (some of its major oxidation products are glyoxal and methylglyoxal). This might be the reason of the different dependencies on liquid water. Something about these facts should be added in order to clarify that the lab studies are not contradictory.

We have added the following to clarify: The variable effects of liquid water observed between smog chamber studies may be due to differing experimental conditions, or to differences in the water solubilities of the oxidation products from different precursor VOCs;

5) Section 3.2.3: It is actually not the absolute NO<sub>x</sub> level but the ratio of organic C/NO<sub>x</sub> that determines the fate of RO/RO<sub>2</sub> radicals. How does Fig. 7b change if you plot F<sub>p</sub> vs the NO<sub>x</sub>/WSOC(g) ratio?

The reviewer is correct that the VOC/NO<sub>x</sub> ratio is the actual determinant of the fate of the RO/RO<sub>2</sub> radicals. As such, we have updated the text to reflect this: In general, for a given precursor VOC concentration, SOA yields from the reaction of hydrocarbons with ten or fewer carbon atoms are significantly higher at low NO<sub>x</sub> levels compared to high NO<sub>x</sub> levels [Kroll and Seinfeld, 2008]. It is important to

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note that the absolute NO<sub>x</sub> concentration is not a critical parameter in VOC oxidation; rather, it is the VOC:NO<sub>x</sub> ratio which impacts the oxidation product distribution [Kroll and Seinfeld, 2008]. For this study, parent VOCs were not co-measured with WSOC, and thus an analysis of F<sub>p</sub> as a function of the VOC:NO<sub>x</sub> ratio is not possible. This may partly explain the large spread observed in Figure 7b, as the parent VOC concentrations were likely highly variable for a given NO<sub>x</sub> concentration. Plotting the WSOC<sub>g</sub>/NO<sub>x</sub> ratio vs. F<sub>p</sub>, though, is not the same as the VOC/NO<sub>x</sub> ratio analyzed in smog chamber studies since the VOC in question is the parent hydrocarbon (in the Southeast, isoprene and monoterpenes) while WSOC<sub>g</sub> represents gas-phase products of VOC oxidation.

6) p. 648, l. 25ff: Can you perform an estimate of how much sesquiterpenes could contribute to total SOA (e.g., product of emission flux and SOA yield)? I assume that despite their high yields, the absolute SOA mass would be small as compared to monoterpenes and isoprene.

According to a recent modeling study by Henze et al. (2008), sesquiterpenes contribute ~8% of the total biogenic SOA on a global scale. By contrast, isoprene and terpenes account for 54% and 32%, respectively (alcohols account for another 6%). Certainly, emissions in the Southeast will vary from global averages, but it is still likely that isoprene and monoterpenes provide the greatest contribution to the SOA budget.

7) p. 649, l. 1ff: It is true that under high NO<sub>x</sub> conditions the formation of aldehyde compounds is favored. However, they are more water-soluble (e.g., glyoxal) than the low-volatility hydroperoxides. Thus, should not be expected that you also see an increased SOA mass at high NO<sub>x</sub> conditions and high RH since water-soluble products are readily taken up and eventually even oxidized to other species (e.g., Ervens et al., 2008)?

This is an excellent point, and one that we have analyzed, however a clear answer to this question is not apparent from our data. When we analyzed F<sub>p</sub> as a function

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of NO<sub>x</sub> at high (all data >70%) and low (all data <70%) RH, their behaviors were nearly identical. That is, higher NO<sub>x</sub> levels were associated with lower F<sub>p</sub> values, regardless of the RH. This may seem to indicate a mechanism that is different from cloud processing (as the reviewer indicates, at higher VOC:NO<sub>x</sub> ratios, Ervens et al. (2008) predict significantly higher SOA yields from isoprene through a cloud processing mechanism). The confounding aspect of this argument, however, is that the parent VOC concentrations (i.e. isoprene and monoterpenes) may not be independent of RH. There is evidence that the factors which induce biogenic VOC emissions (light, T) also influence the emission of water by vegetation and the evaporation of water from soils [e.g., Makarieva and Gorshkov, 2007]. In our own work, we have observed periods of high correlation between WSOC<sub>p</sub> concentrations and water vapor levels that may be due to this co-emission phenomenon [Hennigan et al., 2008b].

8) p. 649, l. 16: As for NO<sub>x</sub>, also the absolute ozone level might not be the key parameter but the ratio of WSOC/ozone. Is there any correlation of these concentrations?

See, also, our response to item 5 above. We agree that the VOC/ozone ratio may be revealing, however plotting the WSOC<sub>g</sub>/ozone ratio may not be as telling, since WSOC<sub>g</sub> is likely made of the oxidation products of VOC oxidation.

9) p. 653, l. 7ff: The conclusion that the combination of (i) the presence of liquid water and (ii) the subsequent reaction with WSOC(p) leads to enhanced SOA formation. Indeed it is in agreement with recent laboratory studies. In such lab studies, however, no consistent conclusion can be drawn about the (ir)reversibility of such heterogeneous reactions. Do you have evidence about that from your measurements? E.g., have you encountered periods of high RH followed by low RH where the total WSOC(p) was higher than in periods in the opposite order? (This could be a hint of irreversibility).

We cannot comment with any degree of confidence on the reversibility of the heterogeneous reactions. In simply analyzing the time series of RH and the absolute WSOC concentration (either WSOC<sub>g</sub> or WSOC<sub>p</sub>), other factors are likely to confound the anal-

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ysis (similarly, no Fp-Temperature relationship was apparent because of other factors). We do know that absolute concentrations of both WSOCp and WSOCg show overall negative correlations with RH (opposite of the relationships shown with Temperature in Figure 5), but this is likely due to the effect of temperature on WSOCp and WSOCg shown in Figure 5 and discussed in section 3.2.1. The question of reversible or irreversible partitioning is one of the most important to come out of our results, and will be addressed in future studies designed to investigate this question specifically.

10) Section 3.3: These considerations of controlling SOA formation by the means of NO<sub>x</sub> are highly useful. However, by changing the NO<sub>x</sub> level the yields of biogenic and anthropogenic precursors would change with opposite signs. So, could you comment on what would be expected in terms of the anthropogenic vs biogenic SOA fraction by controlling NO<sub>x</sub> levels?

This is a valid question. To reflect this concern, we have added discussion so the text now reads: "This potential feedback to NO<sub>x</sub> control is noteworthy and should be examined in more detail. (Note that decreasing NO<sub>x</sub> may also decrease anthropogenic SOA yields [Kroll and Seinfeld, 2008], though the overall contribution of anthropogenic compounds to the SOA budget is small in the Southeast [Weber et al., 2007]. Thus, the feedback potential is likely greater with biogenic compounds, though this question should be examined as well.) There is evidence";

11) Specific comments p. 636; l. 18/19: What is meant here by "phase state";?

We have changed the text so it now reads: "There was, however, a relationship between WSOC partitioning and the WSOCp concentration, suggesting a compositional dependence between partitioning semi-volatile gases and the absorbing organic aerosol.";

12) p. 646, l. 24: What is meant here by "water uptake";? Uptake of water or uptake by water?

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We have clarified the text, so it now reads: Seinfeld et al. [2001] predict a substantial influence of liquid water on SOA formation due to the effect of the uptake of water on two terms;

13) p. 650, l. 2: An absorbing medium always implies a volume process (as opposed to adsorption that occurs on a surface).

We have deleted surface area; from the text to reflect this correction.

14) p. p. 655, l. 22: I think it is misleading to say that there is no dependence on temperature;. There is no net effect; since in reality there are two effects (with opposite signs) that obviously just cancel each other (higher emission rates and less partitioning due to higher volatility).

Done. We have changed the text to read Partitioning, analyzed through the fraction of total WSOC in the particle phase,  $F_p$ , was found to have no net dependence on temperature;

15) Fig. 1: Why does the regression line have an intercept? It implies that WSOC(g) could become zero. I think it would be more reasonable to force it through the origin which would also be consistent with your approach in Figs. 8b and 9b.

We have performed the analysis 1) forcing the intercept through zero and 2) allowing it to be a non-zero value and have indicated both results in the text. Overall, the importance of this analysis to the findings of the study is minor (especially when one compares the slopes of the two analyses and considers the relatively small magnitude of the intercept in the one analysis).

16) Fig. 7: The dimension of x-axis seems odd. Replace it by (ppb). Strictly speaking, ppm (or ppb) is not a concentration but a mixing ratio. This should be replaced in the legend and text.

Done

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17) Fig. 9b. Could this figure be combine with Fig. 8b? It might be nice to see in one plot the average as well as the individual RH regimes.

Done

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Warneke, C.; Brock, C.; Holloway, J. S.; Atlas, E. L.; Edgerton, E., A study of secondary organic aerosol formation in the anthropogenic-influenced southeastern United States. *Journal of Geophysical Research-Atmospheres* 2007, 112, (D13).

Anonymous Referee #2 Received and published: 6 March 2009 This study investigated the gas/particle partitioning of water-soluble organic aerosols in Atlanta in summer 2007. An extensive dataset is obtained and the relationships between the fraction of total WSOC in the particle phase ( $F_p$ ) and various parameters such as temperature, RH, NO<sub>x</sub>, organic aerosol mass, WSOC<sub>p</sub>, and ozone are examined in detail. While many of previous studies on gas/particle partitioning of SOA have been done in the chambers, this paper gives an excellent overview on gas/particle partitioning based on ambient data. By measuring the WSOC<sub>g</sub> and WSOC<sub>p</sub> simultaneously and carefully examining the various parameters that may affect the partitioning of WSOC, this work provides a powerful test of our understanding of the gas-particle partitioning in the atmosphere, particularly on the role of fine-particle water and heterogeneous reactions in SOA formation and growth. This paper is well-written; the results are original and would be of great interest for the community, I recommend it to be published in ACP. I only have a few minor comments.

Specific comments: 1. Page 642, line 16 onwards. It was found that while the WSOC<sub>g</sub> have another maximum increase at night, there was no corresponding increase in the WSOC<sub>p</sub>. This is an interesting observation. The authors wrote "Smog chamber studies by Hallquist et al. (1999) observed significantly lower SOA yields from the reactions of NO<sub>3</sub> with apinene, compared to reactions of NO<sub>3</sub> with either b-pinene, delta3-carene, or limonene. Additionally, nighttime chamber studies by Griffin et al. (1999) observed significantly lower SOA yields from the ozonolysis of b-pinene than either the ozonolysis of a-pinene or the photooxidation of b-pinene". I understand that the author are citing these studies to suggest that it is possible that oxidation is taking place at night but not forming condensable products. However, the citations should be clarified. a. For instance, the first sentence is saying Y of NO<sub>3</sub>+a-pinene is less than Y of NO<sub>3</sub>+others,

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and the second sentence is saying  $Y$  of  $O_3+b$ -pinene is less than  $Y$  of  $O_3 + a$ -pinene and  $Y$  of  $OH$  and/or  $O_3+b$ -pinene. Without knowing all these SOA yields, it is difficult to get an idea of what the authors are trying to get to. (These sentences make it sound like the authors trying to imply that "NO<sub>3</sub>+a-pinene" and "O<sub>3</sub>+b-pinene" may be the most important reactions here, and since their SOA yields are lower than other reactions, this may explain the lack of increase in WSOCp). b. At night, the monoterpenes can react with NO<sub>3</sub> and/or O<sub>3</sub>. What is the typical ozone concentration at night? Do the authors think that the ozonolysis of monoterpenes at night is occurring at a significant extent in this study? Ozonolysis of monoterpenes typically have pretty high SOA yields, it is surprising that an increase in WSOCp is not observed with such an increase in WSOCg at night. c. There are several recent studies pointing to the formation of SOA from reactions with NO<sub>3</sub> radicals (e.g. isoprene with NO<sub>3</sub> by Ng et al. 2008 and bpinene with NO<sub>3</sub> by Fry et al. 2009). Perhaps it is unlikely that the reaction of isoprene and NO<sub>3</sub> is leading to the increase in WOSCg (as it is unlikely that isoprene mixingratio increases that late into the night?), but what about the reactions of NO<sub>3</sub> and bpinene? Could the authors comment on why an increase in WSOCp is not observed with respect to this reaction? d. What is the cause of the decrease in WSOCg from midnight to 6am?

We have completely re-written this section, to include new references and a more definitive analysis. The reviewer is right in questioning the role of O<sub>3</sub> in the nighttime formation of WSOCg. Due to its rapidly decreasing concentration as WSOCg is increasing (which we have added to Figure 2), and its overall low evening concentration (~25ppb average), it is unlikely that O<sub>3</sub> was prominently involved in the nighttime spike of WSOCg. More likely is that nitrate radical was reacting with either alpha-pinene or isoprene. It is possible that either mechanism would form significant WSOCg and minimal WSOCp. We cannot distinguish between the two with our data, but we have added significant discussion as to why either may be possible.

2. Page 646, line 17. What is the Henry's Law constant calculated in this study? Kroll

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et al (2005) determined an effective Henry's Law constant of  $2.6 \times 10^7$  M/atm for glyoxal uptake on aqueous aerosols. How does the value from this study compare to that?

We have performed the calculation and have added to the text the following: This was attributed to the gas phase hydroxyl radical (OH) oxidation of C<sub>2</sub>H<sub>2</sub> to produce glyoxal and its subsequent uptake and reaction in the aqueous phase. Also of note, using  $10 \mu\text{g m}^{-3}$  liquid water concentration and an F<sub>p</sub> value of 0.3 (approximately equal to the observed F<sub>p</sub> above 90% RH), we calculate an effective Henry's Law constant for the total WSOC of  $\sim 2 \times 10^9$  M atm<sup>-1</sup>, which agrees well with the experimentally-derived value of Volkamer et al. [2008]. Overall, our ambient results showed a strong

3. Page 647, line 7. It does not look like there is a definite conclusion from lab studies on the effect of RH on SOA formation. There are some other studies suggesting that higher humidity can result in higher SOA yields in monoterpene ozonolysis (e.g. Jonsen et al., 2006; Stenby et al., 2007). These works should also be cited. Certainly more laboratory work is needed in determining the effect of humidity on aerosol formation.

We have added the references, as suggested, and have included a more thorough discussion of the laboratory results.

4. Page 649, line 21. If O<sub>3</sub> is abundant, one would expect that ozonolysis should be contributing to SOA formation at night. This goes back to the question on why the WSOC<sub>p</sub> does not seem to be increasing with the increasing WSOC<sub>g</sub> at night?

See discussion for item 1 above.

5. Page 663, figure 1. The full scale should be shown (i.e. show the origin, also, the maximum of WSOC<sub>g</sub> observed is  $73.1 \mu\text{g/m}^3$ , but the x-axis is only up to about  $54 \mu\text{g/m}^3$ ).

We have changed the figure, as suggested.

6. Page 664, figure 2. What is the average daily temperature profile? From this figure it

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looks like  $F_p$  varies over the course of the day (highest during the middle of the day). Is there a correlation between  $F_p$  throughout the day and the average daily temperature profile?

Yes,  $F_p$  peaks around 12:00-2:00pm, however there is no correlation with the average daily temperature profile (for least-squares regression analysis,  $r = 0.34$ ). On average, the peak temperature occurred later (3:00-4:00pm). Also, there were several periods (from midnight &#8211; 8am, and then again from 1pm &#8211; 6pm) where the two may have been weakly anti-correlated.

7. Page 668, figure 6. What is the aerosol composition used in performing the ISOR-ROPIA calculations? And why? Also, what is the temperature used in the calculations?

For the predicted liquid water concentration, we assumed an inorganic composition equal to the average composition observed over an entire month of sampling in August, 1999 (Atlanta Supersite experiment). We have updated the text to reflect this detail. We used an average summertime composition because we didn't make speciated inorganic aerosol measurements throughout the summer. There are two main reasons for including Section 3.2.2, which essentially summarizes the major findings of Hennigan et al. (2008a). First, with the present study, we are attempting to provide as comprehensive an analysis as possible on all factors which affect WSOC partitioning. RH was a factor which had a large effect on  $F_p$ , and we extend the original RH analysis by incorporating the dual RH-WSOCp effects (Figures 8, 9). Second, the results of Volkamer et al. (2008) were published after those of Hennigan et al. (2008a) but the similarity of the two studies warranted some discussion. So, a cursory discussion of the Hennigan et al. (2008a) findings is appropriate, but anything beyond that may be redundant. The temperature used in the calculations was 298 K. For an average calculation, this seems reasonable since the average temperature for our entire study was 299K +/- 4.4K (mean +/- 1sigma).

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