

Interactive comment on “Real refractive indices and volatility of secondary organic aerosol generated from photooxidation and ozonolysis of limonene, α -pinene and toluene” by H. Kim and S. E. Paulson

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Please see attached supplementary file, which is formatted so as to be much more readable, and contains figures referred to here.

We appreciate the insightful and helpful comments of the referee concerning this manuscript. These comments have helped us clarify and substantially improve the manuscript. We have also conducted additional work requested by the reviewer, and have addressed each of the points raised by the reviewer as discussed in detail below.

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Reviewer 1: My primary concern with this work is that I find it somewhat difficult to follow the connections that are (or are not) being made between reaction conditions, particle size, particle mass loading and particle composition and how all of these influence the derived RI values. If I am understanding correctly, the authors are arguing that composition plays a controlling role on determining the derived RI values. However, they do not provide any direct measurements of particle composition, instead seeming to use particle size as a proxy for chemical composition. Yet the particle size that results from a given experiment can depend on myriad factors and may only loosely be related to particle composition. By presenting their derived RI values vs. particle size, they seem to be leading the reader to think that there is a one-to-one correspondence between particle size and composition, even if they don't explicitly make this claim. Although particle size may be related to composition, the relationship is most certainly not straightforward and highly subject to specific experimental conditions (including the amount of precursor VOC and the rate of oxidation). The authors do provide strong evidence for the optical properties of SOA being highly variable, but it is not clear to me that these results provide any particular elucidation of the controlling factors, at least not as presented. I would encourage the authors to look for ways they can present their data similar to how they did in Kim et al. (2012), where they showed derived RI vs. HC/NO_x. I believe that this would allow for more direct connections to be made between particle composition and particle RI, even when no composition measurements have been directly made. That said, most of the results presented in the current work are extensions or repeats of previous experiments: it is really only the results from the thermodenuding experiments that are particularly new.

Regarding this comment specifically, the reviewer is correct in that we believe properties such as refractive index arise from the chemical composition, including intermolecular interactions. We completely agree that particle size, in experiments such as these, is only indirectly related to particle chemical composition. We would love to have chemical composition, but unfortunately do not have that capability at the present time. However, because refractive index changes throughout the experiment (especially at

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the beginning) we can plot the refractive index at high resolution either as a function of time, particle size, or particle mass. Alternatively, we could plot it as a function of a highly averaged property such as the initial HC/NO_x ratio (now in Figure S5). We feel that a time variant property such as particle diameter is much more revealing than a highly averaged one. For the abscissa, plotting diameter allows experiments to be more easily compared than would time (experiments are done in an outdoor chamber, so there is experiment to experiment variability that impacts the speed of reactions). Mass concentration can also be used, but diameter produces cleaner results. We posit this may be because variation in chemistry results in more differences in nucleation vs. growth (which ultimately controls size) than in PM mass, but this needs investigation using chemical methods. As suggested, the refractive index vs. HC/NO_x ratio is plotted in Figure S5. It shows a similar relationship of refractive index vs HC/NO_x ratio as shown at Kim et al (2012) although in this case the relationship is weaker, mainly because many larger particles were generated at low NO_x conditions, and these have higher m_r. The main question we are exploring in this paper is how removing volatile material from particles influences SOA optical properties. The investigation of optical properties of SOA is a nascent area of study. We feel that further exploring the relationship between volatility and optical properties potentially provides interesting insights, and contributes to understanding the factors controlling the optical properties. Even SOA chemical composition still has many open questions, despite having been investigated by many research groups over many years in many chambers, including a few with thermodenuders.

Reviewer 1: Here, they find that there are some differences in how thermodenuding influences the derived RI values, with apparent differences between higher HC/NO_x and lower HC/NO_x conditions. This is interesting. They speculate that these differences suggest “significantly different chemical composition of SOA generated under low [vs. high] NO_x conditions,” but provide no supporting compositional evidence. How “significant” do they expect the differences to be based on the literature? Given that no direct composition measurements have been made, I suggest that a much greater link to the

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literature would be beneficial.

Reply: Thanks for pointing out the need to elaborate here. We have expanded the links to previous literature and expanded the paragraph, as follows:

“(Nakayama et al. 2013) reported that refractive indices of toluene SOA increased with increasing O:C ratios, although Kim et al. (2013) show that for limonene and α -pinene, the opposite may be true, although the relationship between O/C and refractive index was weak. For these limonene and α -pinene SOA, Kim et al. (2013) found that H/C was strongly positively correlated to the refractive index. Several studies have investigated chemical composition of SOA generated at high and low NO_x. The most striking differences are in formation of organic nitrate (R-ONO₂) and hydroperoxide (ROOH); at high NO_x, organic nitrate (R-ONO₂) formation is favorable whereas at low NO_x conditions, hydroperoxide (ROOH) is more dominant ((Kroll and Seinfeld 2008) and references therein)). Recently Eddingsaas et al. (2012) showed that for α -pinene SOA, pinonic and pinic acid were only found under low NO_x conditions, although the total concentrations of organic acid are similar at all HC/NO_x ratios. While the refractive index is strongly correlated with changing chemical composition over the course of a photooxidation experiment (Kim et al., 2013), this relationship is in need of further study.”

Reviewer 1: It does appear that there are some differences in the volatility of the aerosol formed at higher vs. lower HC/NO_x that correlate with the RI behavior, but the authors do not refer to this in their discussion of the optical properties, and it is only in their discussion of the optical properties, and it is only indirect evidence in any case (although should nonetheless be exploited). Regardless, I find that the differences between different conditions are often difficult to discern, at least as presented. The authors might wish to try something like graphing the mean change in RI upon thermodenuding vs. HC/NO_x (for photooxidation experiments). Or the change in RI vs. the volume fraction remaining upon thermodenuding. Something like this, I believe, would help to make the relationship with composition stand out.

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We also expected that the volatility of the aerosol formed at high vs low NO_x would possibly correlate with mr behavior, but our result shows that volatility is not so different between high and low NO_x (shown in Fig 2) at least under the conditions of our experiments. Conceptually, volatility is likely a more blunt metric than optical properties especially considering the issues measuring it accurately (Khylstov and coworkers and others). Also, in an effort to present our results clearly (this type of plot can rapidly become overly busy), we finally settled on plotting each HC/NO_x condition separately. We have added a figure showing ΔRI vs. volume fraction remaining at the end of this response. That figure does not clearly show differences between different conditions, mainly because the HC/NO_x ratio is not the controlling factor for mass fraction remaining, at least for our conditions. In an attempt to address this, we added more discussion related to low NO_x conditions (see comment immediately above).

Other comments:

Reviewer 1: P1951, L21: The references given here seem biased towards the authors' own work.

Reply: We have changed it from "see summary [Kim et al.2010; 2012]" to a list of separate references.

Reviewer 1: P1952, L1: I find the statement "We note that smaller particles (e.g. <200 nm) do not scatter much light, which presents measurement challenges and in some cases requires high mass concentrations" to unduly put emphasis on techniques that do not have the sensitivity to make light scattering (or extinction) measurements. There are techniques that are quite capable of making measurements at low concentrations, such as cavity ringdown. See for example the sensitivity of the CRD spectrometer described in Langridge et al. (2011). This instrument can easily measure particle scattering for much smaller particles. Also, this statement seems inconsistent with the mention on P1957 that the authors can reliably retrieve Ri values for particles >100 nm when concentrations are >20 micrograms/m³.

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While light scattering is a strong function of the size parameter, such that scattering of red or green light by particles below 200 nm drops off rapidly, nevertheless, we have removed the sentence. Regarding CRD, it is correct that cavity ring-down measurements of aerosol extinction are quite accurate and have recently become popular due to their sensitivity to low particle concentrations. However, as far as we can find, studies of refractive index using CRD have not been performed at low particle concentrations. Additionally, in practice CRD applied to refractive index measurements seems to result in low time resolution. As far as we can tell this is due to the time required to size select aerosols to facilitate Mie retrievals, or to difficulty retrieving the refractive index from a rapidly changing polydisperse aerosol. In contrast, although the PN measurement is less sensitive than CRD, it is fast, and requires only the single simultaneous SMPS derived particle size distribution, resulting in three minute time resolution. Re: the later sentence, for particles >100nm when there aren't many particles and concentrations are >20 micrograms/m³, it is possible to retrieve the refractive index from our data.

Reviewer 1: P1952, L13: The statement "By generating aerosols at higher concentration and thermodenuding them, we may obtain aerosols with the full range of volatilities, and then thermodenude them to obtain more atmospherically relevant lower volatility species" implicitly assumes that particle phase reactions do not occur that modify the particle properties, making thermodenuding a potentially inefficient method of generating atmospherically relevant particles. Note that this is true whether such reactions are enhanced by heating or not. In other words, if particle phase reactions are already leading towards the production of low volatility material, then lower volatility species may already be generated at "high" SOA concentrations. The question is then not one of volatility, but of compositional representativeness.

There are many issues with making particles in chambers. Experiments performed at low precursor concentrations can lose to the walls the low volatility material that should be in the particles. Experiments performed at high precursor concentrations produce semi-volatile material that may not be on particles in the atmosphere. We feel

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that thermodenuding particles may make some of the most representative chamber particles possible. It is correct that it is impossible with the current data to rule out the possibility of irreversible in-particle reactions. While the time scales seem relatively short (1-3 hours in the chamber and 16s in the thermodenuder) for these to play a major role, this is a topic worthy of further study.

P1952, General: If the point of the thermodenuding is to push the particle composition back towards ambient, then why not just do experiments at lower initial concentrations? One cannot get around the issue of instrument sensitivity just by using a thermodenuder.

Several reasons: 1) doing experiments at low precursor concentrations does not guarantee production of model atmospheric particles because the low volatility material that should be in the particles preferentially (compared to more volatile species) deposits to the walls. One can argue (as we have in the manuscript) that generation at slightly higher concentrations and thermodenuding may produce the most representative chamber particles. 2) We wanted to explore the relationship between volatility and optical properties, which is, in our view, a piece of the puzzle in the question of what controls refractive index.

Reviewer 1: P1955, L24 and Fig. 1a: Looking at this figure, it appears that it took more like 1 hour to stabilize at a new temperature, yet the authors' state it takes 6-9 minutes. It would be helpful if clarification was provided.

In Fig1a (the new version is also shown below), each dot represents 3 minutes. The white dots indicate the transition period (6-9 minutes) and the black dots indicate the period for which we consider the temperature to be stabilized. It does vary by a few degrees during this latter period.

Reviewer 1: P1959, L3: It is not entirely clear why "Reliable refractive indices were first obtained when the particles had grown to about 344nm at 13:07." What specifically limited the ability to extract refractive indices prior to this? Presumably not concentra-

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tion, because this would then suggest that the thermodenuded results are unreliable. Is it the rapidly evolving size distribution during this period? Also, on P1957 the authors state that reliable measurements can be made for particles >100 nm when concentrations are >20 micrograms/m³.

Thanks for pointing out the need for more explanation here. Actually, the problem was that for this experiment, when particles were smaller than 344nm, the SMPS was frozen for about 18 minutes, so we don't have the particle data needed to retrieve the mrs. This can be seen in fig 1; 18 minutes (6 mrs retrieval points) are missing. Also because of that issue, we couldn't fully re-zero the polar nephelometer so that fitness of the initial value wasn't good. We revised the paragraph to read: "The phase functions of particle were first obtained when the particles had grown to about 194 nm (21 $\mu\text{g}/\text{m}^3$) at 12:40 but reliable refractive indices (fitness>0.95) were first obtained when the particles had grown to about 344 nm at 13:07 (Because of the malfunction of SMPS for a while and re-zeroing issue with polar nephelometer)."

Reviewer 1: General: I am not entirely convinced that the characteristic shape (i.e. the up/down behavior) that the authors observe for SOA RI values vs. particle diameter is not a result of changing instrument sensitivities. This is natural to wonder about, and it is something we spent considerable effort verifying ourselves. We have written multiple papers to verify our instrument, including this issue, and cannot go into all of the details here. Perhaps the simplest verification is that for any given particle size, we observe very different values for different precursor systems.

Reviewer 1: Figure 2 and associated discussion: Khlystov and co-workers have shown that "mass fraction remaining" is potentially an unreliable metric because it explicitly depends on the associated mass loading for a given experiment. This is a result of recondensation at higher mass loadings influencing the net evaporation. Given this, it could be helpful if the authors were to color code the data by mass concentration, using a consistent color scheme through all 3 panels. This is especially important given that, based on the information in the table 1, the mass loadings for Limonene

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SOA were systematically higher than those for α -pinene, and the mass loadings for toluene SOA appear to have been even higher. How can the authors ensure that the differences are not simply due to different mass loadings?

For the figure, we changed the colors to be consistent and changed the color coding of the legends to indicate the pre-thermodenuder mass concentration range for each experiment. Our thermodenuder has a reasonably long residence time (16s), and uses some activated charcoal in the cool-down region, which may limit mass effects for most of the range we studied.

Figure S6 shows the same data plotted as a function of the mass concentration prior to denuding. There is little or no indication of influence of the mass concentration for the photochemical oxidation experiments for all three hydrocarbons, which had mass concentrations prior to thermodenuding of 25 to $\sim 300 \mu\text{g}/\text{m}^3$. A sentence to this effect, with reference to Khlystov and co-workers and the supplementary figure has been added to the manuscript: "Initial mass concentration (Saleh et al., 2011) also did not appear to be a factor (Figure S6)." There does appear to be some mass concentration effect for the ozonolysis experiments, especially for α -pinene. The upper end of the initial mass concentration for these experiments was higher, up to $550 \mu\text{g}/\text{m}^3$, but a contribution from mass concentration may play a role throughout most of the mass range ($80 - 550 \mu\text{g}/\text{m}^3$) for this data set. This has now been pointed out in the text, reading " α -Pinene SOA from ozonolysis had about 15% lower MFRs than limonene ozonolysis SOA, however this could have a contribution from the high mass concentration of particles prior to denuding between the experiments (Figure S6, Saleh et al., 2011)." Importantly, the focus of the discussion is really on whether or not there are observable differences for the different HC/NO_x ratios. For these comparisons, the mass concentrations are in similar ranges, so a mass effect doesn't seem to be a significant concern.

Reviewer 1: P1959, L20: The authors mention that higher f₄₄ values for toluene SOA compared to biogenic SOA might indicate aromatic SOA is less volatile than biogenic

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SOA. However, f₄₄ is never defined to let the reader know what this means. In other words, the authors are using Aerodyne aerosol mass spectrometer (AMS) jargon without defining it or assuming that the reader knows that f₄₄ is thought to correspond more to organic acids. In any case, the reference they cite (Chhabra et al.) says nothing directly about volatility of SOA. A specific reference is needed to support the contention that higher f₄₄ should equal lower volatility.

Thanks for pointing out this oversight. We have clarified this in the manuscript and have added a reference to Ng et al. (Ng et al. 2010), which suggested that the low volatility associated with xylene SOA is a result of its high f₄₄ (from that paper: "products formed from the photooxidation of m-xylene are generally less volatile, consistent with the higher f₄₄ values in aromatic experiments.(compared to biogenics experiments).") We have adjusted the paragraph to read: "There are no investigations available in the literature concerning the volatility of toluene SOA, however, AMS measurements of chemical composition show that SOA from aromatic HCs, including toluene, has consistently higher values compared to biogenic SOA of f₄₄ (ratio of m/z 44 (CO₂⁺) to total signal in the component mass spectrum). Because f₄₄ is associated with organic acids, which are associated with lower volatility, this suggests aromatic SOA may be less volatile (Ng et al. 2010; Chhabra et al. 2011)."

P1960: Regarding the various discussion about RH effects: do the authors mean to imply that particle phase water is leading to differences, or are they assuming that there are differences in the gas-phase formation of SOA condensables? This should be stated. At these low RH's the amount of water uptake for these types of SOA are typically very, very low.

Both our study and the Johnson et al. study posit that there are differences in the gas phase formation of SOA, that influence to the volatility, although there isn't definitive proof that we know of. For the clarification, at the end of section 3.3.1, we have expanded our discussion, as below: "Consistent with this result, Jonsson et al. (2007) and Lee et al. (2011) also reported moderate increases in volatility with increasing RH,

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since the presence of more water in the gas phase during oxidation possibly increases the fraction of volatile material present on the particles. “ We note that we do think water in the particles themselves is important at the smallest sizes, and contributes to why we observe rather low refractive indices for the freshest particles. This is consistent measurements of water uptake by recently nucleated particles (please see references in Kim et al. 2012). This issue comes up later in the manuscript.

Reviewer 1: S3.4.1: I find the discussion regarding water uptake to be confusing, and the authors appear to be conflating chemical changes with size changes. Size is not important, except to the extent that it reflects that the reactions have been run for longer times and greater amounts of SOA formed, potentially with a different composition than earlier in the experiments. It is composition that matters. I find the emphasis on size to be misplaced and suggest that the authors focus on the compositional differences. As such, I suggest that the authors adopt their approach from Kim et al. (2010), in which they consider their observed RI values vs. mass concentration, instead of the approach from Kim et al. (2012), in which they also use particle size as the key metric.

Thank you for the suggestion; please see response to the first few comments above. One can argue that for a similar set of experiments, size is a better proxy for chemical composition than mass, because chemical composition of condensing vapors controls nucleation vs. condensational growth. Mass tells the total amount of condensable species, while size tells something about the ratio of higher to lower volatility material.

Reviewer 1: S3.4.1 Toluene SOA: The values observed here do not actually seem to me to be consistent with the previous observations, despite the statement that they are “consistent with previous studies.” In Kim et al. (2010) the RI for toluene SOA formed at the same VOC/NO_x (=15) as here ranged from ~1.45–1.55. This is substantially higher than the 1.35 values reported in the current study. What is the reason for this difference? It is somewhat difficult to make direct comparisons between this work and Kim et al. (2010) because here the authors report their results relative to particle diameter while in Kim et al. (2010) the report them vs. mass concentration

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(which, in my opinion, is the physically more meaningful quantity in any case; see above comment). The data from the two studies should be compared directly, perhaps in the supplemental material like the aP and limonene results.

Thanks, at the end of this response, comparisons with 2010 results are attached. Since the wavelengths that we use were different (670 vs 532nm), and mass concentrations were much higher in the 2010 paper, they are difficult to compare. The lower value of 1.35 is for young particles. In 2010 we were not able to retrieve comparable data due to the much lower sensitivity of the earlier version of the polar nephelometer. Other than that, if you look at the RI vs mass, overall the 2013 values are higher by ~0.03 than 2010. This difference is the expected difference due to the difference in wavelengths of the two measurements. On a more detailed level, the consistency is poor (actually the 2010 data isn't completely consistent with itself, but we cannot completely explain the reasons) but the overall range is still consistent.

Reviewer 1: S3.4.1 Toluene SOA: Nakayama et al. report an imaginary RI of 0.007, not 0.037 as stated. This is a very, very important distinction as Mie theory is very sensitive to changes in the imaginary RI when it is around this range. In any case, the authors should state what they believe to be their detection limit for imaginary RI values. To state that their GA retrievals give values O(10⁻⁵) is not helpful if they cannot distinguish between 0.1 and 0.05 (as a random example).

We really appreciate catching these errors, and have corrected the reference and revised our statement to better reflect the uncertainty of our own measurements. There is no expectation of absorption at the wavelength used here (532 nm) ((Kanakidou et al. 2005), Figure 7, and Myhre and Nielsen (2004). In the past we have been asked by reviewers to report a rough value for the imaginary part of the refractive index, and that is what we have provided. We have changed phrasing to better reflect our best estimate of our ability to measure this value, to read “below our very rough detection limit of 10⁻⁴ (Barkey et al., 2011);”.

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Reviewer 1: P1964, L9: As far as I can tell, Salo et al. (2011) report a temperature of formation influence on the volatility of the formed SOA, but they in no way identify 20°C as “an important transition temperature.” The authors need to elaborate as to what exactly leads them to this conclusion. Also, here the difference in temperature between the Nov. 18 experiment and the other experiment ranges from 2–5°C, which is much smaller than the 20°C steps in the Salo et al. (2011) experiments. Given this, I am not convinced by the argument that the difference between, in particular, the Nov. 18 and Nov. 10 experiments is explainable by the different temperatures.

Thanks for pointing this out. You are correct that the Salo et al. (2011) experiments show differences between 0, 20 and 40 °C, with the result that somewhere between 20 and 40 may be a transition point, so it is going too far to infer that the transition temperature is close to the lower end of this range. We have revised the section to read “The Nov 18 α -pinene data has lower refractive indices (1.39-1.45) than the other experiments, a phenomenon we consistently observe for particles generated at lower temperatures (Kim et al. 2010). Several other researchers have noted significant differences in chemical composition of α -pinene SOA generated at different temperatures, including in the range studied here (Warren et al. 2009; Wang et al. 2011; Salo et al. 2012).” In our opinion, this is another area that warrants further investigation.

Reviewer 1: P1962, L22: The authors need to advance a stronger theoretical reason for the increase/decrease behavior of the real RI for both limonene and α -pinene SOAs. I find the arguments put forth on the top of the next page to be weak, given the lack of actual composition data here. Their own calculations in Kim et al. (2012) indicate a monotonic increase in RI with mass concentration (and presumably particle size, but this is not shown). Is this behavior simply being driven by the particulars of whether particles are more likely to nucleate fewer, bigger particles or more, smaller particles given a particular set of reaction conditions?

In Kim et al. (2012), we suggest that the HC/NO_x ratio can help determine the size distribution of SOA generated in chambers. Generally, high NO_x conditions gener-

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ate smaller numbers of large particles because the ozone concentration builds up late relative to low NO_x (high HC/NO_x ratio) experiments. In contrast, under low NO_x conditions, ozone builds up rapidly, and nucleation occurs earlier in the experiment, relative to both time and the amount of HC reacted. That creates a situation where there are many small nucleated particles. And the amount of condensing species seems to influence the refractive index. To clarify this relationship between *m_r* and chemical composition (size), we have added the paragraph below (as also indicated above). “We have consistently observed that the smallest, newest particles have significantly lower refractive indices than larger particles. HC/NO_x ratio-dependent differences in higher and lower volatility condensing species and their contributions relative to one another appear to contribute to determine size distributions as well as the *m_r*.”

Reviewer 1: P1963, L9: The authors need to provide a reference (or references) in support of the statement: “The observed TD behavior is consistent with a simple model for aerosol growth in which more volatile species are deposited on lower volatility species as the experiments progress; heating appears to simply reverse the process. This behavior is consistent with the notion that condensed species do not mix effectively, but instead layer material on as they grow,”

Thanks for pointing this out. (Cappa and Wilson 2011) and references therein have been added.

Reviewer 1: Figure 6: I find the presentation in this figure to be somewhat confusing. In particular, it is not clear to me what is determining the vertical height of the boxes as compared to the $\pm 15\%$ error bars. Are these $\pm 15\%$ on top of the total observed range? Further, I would suggest that this figure is potentially misleading in terms of the atmospheric relevance. The authors include results from all of their experiments, which cover an extremely wide range of organic mass loadings, most of which are well above the atmospherically relevant range. What would this figure look like if constrained simply to the atmospherically relevant range of concentrations? I suppose that this, then, limits the ability to make this figure to the thermodenuder

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measurement here, as they are the only ones that measure optical properties for SOA anywhere close to ambient concentrations. Nonetheless, the point is well made that SOA optical properties cannot be considered a constant.

Thank you for pointing out the need for more clarity. We have added more explanation to the figure caption. The vertical height is the range of m_{rs} that we observe for each set of oxidation conditions (e.g., high NO_x α -pinene, low NO_x Limonene and so on) and error bars indicate the ± 0.03 from the highest and lowest value of m_r (± 0.03 is the absolute error of m_r retrieval). The main point of this figure is to show the wide range of m_{rs} , which persist after thermodenuding. Also as Nakayama commented, we should not have combined measurements at different wavelengths, so we separated our 670 nm value from 532 values, and have indicated other's work separately depending on the wavelength. If reviewers think it is too busy (it is busy) we could remove the 670 data. We also note that while it is appropriate to question high mass loading values (noting that the atmosphere does experience a remarkably wide range of values when plumes are included) results so far do not support the notion that mass loading is a major factor in determining the real part of the refractive index, with the caveat that we don't currently have data below 10 $\mu\text{g}/\text{m}^3$.

Reviewer 1: P1954, L7: "gasses" should be "gases".

You are correct that "gases" is the more common form, although "gasses" is also acceptable. We changed it. Reviewer 1: P1957, L18: "Detailed" should be "Details"

This has been corrected.

Reviewer 1: P1962, L10: Figure 4S should be Figure S4.

Thanks for catching this; it has been corrected.

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