

Response to the comments of Reviewer #1

Li et al. presented a study that examined the effects of temperature on the optical properties and chemical composition of secondary organic aerosols formed from the OH photooxidation of n-dodecane. The authors found that oligomers were formed at low temperatures, and these oligomers resulted in higher RI values being measured. This paper is potentially useful to the SOA community. However, there are some important issues that the authors need to address before the manuscript can be considered for publication.

Response: We thank Anonymous Referee #1 for the review and the positive evaluation of our manuscript. We have fully considered the comments and [responded to these comments below in blue text](#). The revisions in the manuscript are highlighted in yellow color. The response and changes are listed below.

Major comments:

1. *Why were experiments conducted under dry conditions?*

Optical properties of the SOA can be affected by many factors, in order to study the temperature affect, other factors must be kept unchanged, so the humidity of the experiments must be constant and cannot be changed. The experiments were conducted under dry conditions (RH < 5%). Because when the temperature changes, the saturated vapor pressure of water changes, that is to say, if the RH is consistent at different temperatures, the concentration of the water is not consistent; when the concentration of water is the same, the RH is different. So choosing other humidity (non-dry conditions) will introduce new problems, we can only choose dry conditions. And we have added the related statement in the main text (Page 3, Line 88-93).

2. *Table 1 showed used of 43 ppb at low temperature vs. 58 ppb at high temperature. Why wasn't the same amount of n-dodecane used?*

Actually, the concentration of *n*-dodecane is tested with PTR-QMS, and the calibration of the PTR-QMS's response to *n*-dodecane is achieved through permeation tubes. According to our experimental design, the expected concentration of *n*-dodecane is 50 ppb, which is introducing 2 μL liquid *n*-dodecane into 5 m^3 smog chamber. As the injection volume of *n*-dodecane is 2 μL , volume error during injection is inevitable, which will influence the concentration of *n*-dodecane in the chamber. Nevertheless, the relative small differences in *n*-dodecane concentration (43-50 ppb at low temperature and 52-58 ppb at high temperature) likely have little influence in SOA composition and optical properties. And we have added the related statement in the main text (Page 4, Line 100-105).

3. Do the authors know how the use of different temperatures will affect the loss rates of particles to the chamber walls? Is there a possibility that the observations of the SOA mass, composition and optical properties made by the authors can be explained partly by differences of particle wall loss rates at different temperatures?

We have measured the wall loss rates of particles under both room and low temperatures, and found that the wall loss rate under low temperature condition (0.0025 ~ 0.0028 min^{-1}) is larger than that under room temperature condition (0.0018 ~ 0.0020 min^{-1}). However, this difference in particle wall loss rate can only slightly change the SOA mass concentration, but not the particle chemical composition. Therefore, it is unlikely to change the optical properties.

Nevertheless, the difference in vapor wall loss rates may change the particle composition and optical properties. The low temperature can enhance the loss rates of higher-volatility compounds (while for lower-volatility compounds, their dominant fates are condensation so temperature may affect little on their losses), which may lead to their lower proportions in SOA particles. As their RIs are generally lower than lower-volatility compounds (Li et al., 2018), this proportion change can probably enhance SOA RI at low temperature. Therefore, the difference in wall losses and gas-particle partitioning of gas-phase products might partially contribute to the RI enhancement

under low temperature condition.

We have added the above contents in the revised paper (Page 10, Line 297-308).

Reference:

Li, K., Li, J., Wang, W., Li, J., Peng, C., Wang, D., and Ge, M.: Effects of gas-particle partitioning on refractive index and chemical composition of m-xylene secondary organic aerosol, J. Phys. Chem. A, 122, 12, 3250-3260, 10.1021/acs.jpca.7b12792, 2018.

4. In page 5 line 144, the authors state that “During this period, the optical properties of the particles tend to be stable and will not change much.” This sentence is ambiguous and needs to be clarified. What optical property is the authors referring to? RI value? Or are they referring to the absorption spectra?

The optical property here refers to the extinction coefficients (α_{ext}) of the particles. During the last 1 h of the experiments, the extinction coefficients (α_{ext}) measured by the CRDS at 532 nm and PAX at 375 nm tend to be stable; at the same time, the surface mean diameter (D) of the particles tend to be stable and does not change much. When the D and α_{ext} are stable, the extinction efficiency (Q_{ext}) will be stable (as shown in the following equation). A fixed set of D and Q_{ext} results in a fixed RI value.

$$Q_{ext} = \frac{4\alpha_{ext}}{N\pi D^2}$$

We have rephrased this sentence to “During this period, the surface mean diameter and the extinction coefficients (α_{ext}) of the particles tended to be stable and will not change much” (Page 7, Line 210-211)

5. Explain the rationale behind tracking the absorption at 532 and 375 nm.

The CRDS measures the total optical extinction coefficient (α_{ext}) of the SOA, and $\alpha_{ext} = \alpha_{abs}$ (absorption coefficient) + α_{sca} (scattering coefficient). In order to calculate the real (n) and imaginary (i) part of RI values, both the α_{abs} and the α_{sca} need to be known. So the absorption at 532 nm is measured with a UV-Vis light spectrometer (Avantes 2048F). With the measured α_{ext} and α_{abs} , the α_{sca} will be calculated.

For the absorption at 375 nm, the rationale is as following: The photoacoustic extinctions (PAX-375, Droplet Measurement Technologies) directly measures in-situ light absorption and scattering of aerosol particles at 375 nm, from which the α_{abs} and α_{sca} can be derived.

6. The authors did not use seed aerosols in this study to promote gas-to-aerosol partitioning. Hence, I expect substantial vapor wall loss in these experiments, and the extent of vapor wall loss is likely to be different at 5 °C vs. 25 °C. Is it possible that the authors are not detecting some products (due to their loss to the chamber walls) that can contribute to SOA optical properties?

We agree that the gas-particle partitioning can influence the particle composition and optical properties. As shown in our previous study (Li et al., 2017), the presence of seeds can promote the condensation of low-molecular-weight products and decrease the real part of the RI of *n*-dodecane SOA under low NO_x conditions. The experimental conditions in that study are very similar to those of room temperature condition in this study. Therefore, we can expect that similar results can be found in this study at room temperature. However, even if we perform the experiments with seeds in this study, the gas-particle partitioning can also be different at different temperature. In other words, we cannot rule out the contribution of different gas-particle partitioning at different temperatures.

As we have discussed in our response to comment #3, the difference in gas-particle partitioning of products might partially contribute to the RI enhancement under low temperature condition.

Reference:

Li, J., Li, K., Wang, W., Wang, J., Peng, C., and Ge, M.: Optical properties of secondary organic aerosols derived from long-chain alkanes under various NO_x and seed conditions, *Sci. Total Environ.*, 579, 1699-1705, 10.1016/j.scitotenv.2016.11.189, 2017.

7. In section 3.5, the authors tried to relate their results to observations made during

*winter haze episodes in China. I advise the authors to be more circumspect in the extrapolation of their results to ambient observations since NO_x concentrations are likely substantial during winter haze episodes in China. The authors performed a study under low-NO_x conditions. Under high NO_x conditions, I expect the reaction mechanism of *n*-dodecane OH photooxidation to be different. For example, more fragmentation will likely happen, which will result in the formation of more volatile products. If this is the case for both 5 °C and 25 °C conditions, there may not be significant differences between SOA composition and their RI values under high NO_x conditions, which would imply that temperature does not play a big role in the DRF of SOA formed under areas with significant NO_x concentrations, like China.*

We agree that the reaction mechanism of *n*-dodecane under high NO_x conditions is different from that under low NO_x conditions. The temperature effects in the presence of NO_x are indeed very important and will be investigated in future studies. Therefore, we have revised this part in the manuscript “The enhancement in light extinction of SOA and oligomer compositions formed under low temperature condition might provide some possible inspiration for the regional visibility issues, especially the suburban areas. It had been reported that UV-scattering particles in the boundary layer could accelerate photochemical reactions and haze production (Sun et al., 2014). The observations above showed that the scattering property of formed SOA increased under low temperature condition, which might provide one possible reason for the rapid occurrence of haze in suburban areas in winter. According to field observations, haze occurred frequently in winter, especially in China (Cheng et al., 2016; Huang et al., 2014; Guo et al., 2014; Parrish et al., 2007). When haze occurred, it was often accompanied by high NO_x, especially in urban areas. The temperature effects under high-NO_x conditions are also important and need to be investigated in future studies.”

(Page 11, Line 317-324)

Minor comments:

8. *Inconsistent tenses. The authors switch between using past and present tenses in some parts of the manuscript. Please correct this.*

Thank you for the helpful comments. We have corrected this in the manuscript.

Page 7, Line 209: are → were; reaches → reached;

Page 7, Line 210: are → were;

Page 7, Line 211: tend → tended;

Page 7, Line 215: are → were;

Page 7, Line 216: have → had;

Page 8, Line 223: indicate → indicated;

Page 8, Line 249: are → were;

Page 9, Line 257: changes → changed.