Comments on:

Ultraviolet and visible complex refractive indices of secondary organic material produced by photooxidation of the aromatic compounds toluene and *m*-Xylene

In this paper Liu et al study the optical properties of anthropogenic SOA based on collection of the aerosols on a filter and deriving the optical constants using ellipsometry and UV-VIS spectrometry over a broad wavelength range. The study claims that anthropogenic precursors can lead to brown carbon formation at different NOx levels by forming nitrogen-containing species. As such this is an interesting and timely paper that sets a new paradigm regarding brown carbon formation, which has not been so far substantiated for biogenic or anthropogenic precursors. However, the method for deriving the optical constants used in this study is new and has not been well established. In addition, it follows a previous study by the same group in which the used solely ellipsometry. The question then arises why the difference between the two studies and if there are inherent issues with the experimental methods (i.e.; can ellipsometry alone be used for derivation of the refractive indices for films of collected aerosols of unknown thickness and density?). These issues, together with others, are mentioned below:

Page 20589 line 5: specify how was the gas phase precursor concentration was determined

Line 12-15: The experimental analysis assumes complete removal of O_3 from the reactor and conversion to OH, so that the only oxidant is hydroxyl radicals. Was this asumption validated? What is this assumption based on? Please include O_3 concentration measurement or theoretical rate constants calculation?

Line 24-30: Explain the method. Explain how the aerosols films were deposited, how they were characterized. How was the film thickness determined? how homogenous it is? How reproducible is the film and how many repetitions were made for each determination. What are the errors associated with this method. What validations were made to ensure that the method produces physically sound values.

Page 20590 line 1-4: in this study only the real part of the complex RI was retrieved using ellipsometry. In previous publication by the same group (also referenced in this work, Liu et al., 2013) the authors used ellipsometry to retrieve both the real and imaginary parts of the complex RI. Why this method was not used not this time? Is there a problem determining the imaginary part with this method? If yes, please discuss the problem. Why did you need to derive the imaginary part in a separate experiment using UV-VIS spectrometry of extracts? What are the possible artifacts using the UV-VIS methods? What is the absorption by the water soluble component of the SOA and by

the organic soluble component? How was this new approach verified? Did you measure standard materials to verify the validity of the method? What are the associated errors?

In the previous publication (Liu et al., 2013) the authors indicated that the film thickness measured by the ellipsometer should not vary by more than 10% and that the surface roughness should be less than 10% of the light wavelength. In both papers the film thickness is reported to be up to hundreds of nm and the wavelength range starts at about 300nm up to microns. This suggests that in order to satisfy the thickness uniformity and surface roughness, collected particles should be in the size range of less than 100nm. Unless there is evidence that particles become flat by impaction on the surface, this is probably not the case. Please explain how this was done, how it was verified, how film uniformity was assured and how the roughness was measured. What were implications of the film roughness on the associated errors.

Page 20590 section 2.3: The authors used an equation taken from sun et al., 2007. In that publication, the equation should be

$$k = \frac{\ln(10)}{4\pi}\frac{\rho\lambda}{cL}A(\lambda)\xi(n,k)$$

And not as presented in the manuscript, Equation 1.

In Sun et al 2007, ξ is a weak function of **BOTH** n and k with a values that ranges between 0.7 and 0.8. This factor can introduce an error (0.25-0.45% over estimation) and should not be neglected in the analysis as was done in this study.

Equation (1) in the current manuscript should not be equal to k but to k/ ξ(n,k). Using n from the ellipsometry measurements, if justified, one can then calculate k.

According to Chen and Bond 2010, the error introduced by selection of a density value, as was done here (1.4) is nulled out only when calculating the particulate absorption per mass using the same density. Because in this work the authors calculate the imaginary part, the uncertainty in the density should not be ignored.

In addition, the authors do not specify how the value of c (concentration) was obtained. Were the filters weighted and a 100% extraction efficiency was assumed? If not, what was assumed and how the error was calculated? This should be discussed.

Page 20594 line 4: "the spectra in the…" the IR spectra?

Page 20596 line 24-28: Do particle phase reactions depend on humidity? Does water play a role in these reactions? The experiment in this work started at 13% RH and water molecules were converted to hydroxyl radicals. These experiments should be conducted at several relative humidities to determine what is the potential role of water in the products formation.

Page 20598 line 1: sentence not clear, please rephrase.

Table 1. Please provide some information regarding the choice of the size distribution in table 2.

Table 2. Provide reference for the complex RI values of black carbon and sulfate?

Figure 3. Add error bars to figures 3c and 3d like you have done in figures 3a and 3b.

Figure 4. How were the k values for Suwannee river fulvic acid derived? With equation 1? If yes, what density was assumed? Please justify

Figure 6. The upper information segment is shifted relative to the figure.

Figure 8. Blue lines are missing in the legend.

Concluding remarks: the authors emphasize the decrease in UV power available for photochemistry as a result of light absorption in the atmosphere. It would be interesting to discuss also implications to the direct or semi-direct radiative effect. If this is the case, a quantitative estimation of the decrease (even only for a case study) and a qualitative estimation of its significance are needed.