Interactive comment on “Interpretation of Measured Aerosol Mass Scattering Efficiency Over North America Using a Chemical Transport Model” by Robyn N. C. Latimer and Randall V. Martin

Anonymous Referee #2

Received and published: 5 August 2018

The paper studies an important topic for chemistry transport modeling - the mass scattering efficiency that affects the conversion from aerosol mass to aerosol extinction and ultimately AOD. Mass scattering efficiency data from IMPROVE is used to constrain a global CTM (GEOS-Chem, GC). It is found that geometric mean radiuses for both dry secondary inorganic and organic aerosols in original GC configuration need to be decreased, and also the aerosol hygroscopicity curve needs to be adjusted to be less hygroscopic. Overall, the paper should be considered for publication after a major revision (and possibly another review).

General comments
1) there are lots of averaging done in the data analysis. For example, the captions for Figures 2, 3, 4, and 6 all start with the word ‘average’. But, how averaging is done is not described in the method section. In addition, how much is the standard deviation in the average? 2) only R and best fit are shown in all scatter plots. How about RMSE and bias? 3) some discussion of uncertainties here are needed. what are the uncertainties in the measurements of scattering coefficient bsp and surface PM2.5? Likewise, do GEOS fields have any systematic bias in simulating RH?

Specific comments.
P4L10. If IMPROVE PM2.5 is analyzed at 30-50% RH, there will be aerosol water in the aerosol mass measured by IMPROVE. What is the uncertainty here if we assume these are dry particle mass and used in the scattering efficiency calculation? Some consistence is needed or at least discussed between how PM2.5 dry mass is computed in GC vs. how aerosol optics (aerosol hygroscopic growth) are treated in GC. Later in the analysis, the cut off of RH is 35% to be considered as dry (solid) aerosol. Why not 40% as for dry PM2.5?
P4L23. Effectively, b_sp is averaged each day, and then daily averages of b_sp is used together with daily PM2.5 to compute scattering efficiency. How annual average of mass scattering efficiency is computed? How the averages are computed for RH in different ranges or for different compositions (e.g., dust dominated, SIA dominated, etc.)? RH does have a strong diurnal variation.
P6L20-24. The uncertainty of aerosol mass scattering efficiency can also come from the particle composition which essentially affect the particle hygroscopic growth factor, or particle size and density. the scattering efficiency of sulfuric acid vs. ammonium sulfate can be different even for the same size distribution at dry conditions. See “Table 1 in Sensitivity of sulfate direct climate forcing to the hysteresis of particle phase transitions, JGR, 2008”. The mass scattering efficiency can also be affected by the mixing state of the particle. The analysis here needs to discuss these uncertainties before
focusing on particle size.

P7L2-3. What is the seasonality of aerosol size distribution?

P7L13-15. Martin et al’s paper showed there can be aerosols in solid phase in RH larger than 40%. The phase transition depends on RH history of the particle, not just RH itself. Could this explain the part of overestimation in GC mass scattering efficiency? What is the fraction of solid SIA particles in U.S.? This paper might be helpful – “Global distribution of solid and aqueous sulfate aerosols . . . , JGR, 2008”.

P8L5. What is r_sp used in figure 8?

P11L20. Again, it is the composition that regulates the growth factor and density. The real index of refraction also has the effect on scattering efficiency. In addition, the paper didn’t mention effective variance at all. Is it important? References are needed here to support the statements.

P13, L7-8. The aerosol concentration is the same here for both figures. It is likely that the improvement with new optics is within the range of inter-annual variability of alpha_sp itself? It will be more convincing that the validation is done using other years data (say 2007 or 2008).

Figure 5. Caption. Is aerosol effective radius shown for dry (solid) or wet particles?

Figure 9. The GADS assume sulfate as 75% H2SO4, which often has the largest hygroscopic growth. For lab data, please provide reference. Also, lab data clearly shows that ammonium sulfate particles can be in solid phase in range of RH between 40-80%, which may explain why GC has an overestimation in scattering efficiency. How large is the uncertainty in assuming that all SIA are aqueous in RH of 40-80%? If particles are in solid phase in upper troposphere (as models suggest), the large improvement in surface with new optics may not be reflected in the AOD comparison.

Figure 12. Add RMSE, and also, how the spectral AOD (or Angstrom exponent) is compared against AERONET? This can be an interesting test for new optics that is a result of adjusting particle size. Particle size affects both scattering efficiency as well as the spectral AOD slope. Ideally, the new optics should provide an overall improvements for the model.