

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 #1

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This paper examines aerosol mass scattering efficiencies using a chemical transport model and ground-based aerosol composition data. The work is important because aerosol mass scattering efficiencies are required to estimate aerosol radiative properties- both for surface and satellite-derived properties. Understanding model biases as a function of aerosol size and hygroscopic effects is necessary for accurate estimates of aerosol optical properties. The authors ‘tune’ the model to observations to determine the aerosol size and hygroscopic properties that most closely agree with observations. While the paper is fairly clearly written, it could benefit from clarifications in the method description, as described in the comments below. One of the major weaknesses of the paper is the lack of comparisons of observed and modeled mass

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concentrations and light scattering coefficients. Since mass scattering efficiencies are calculated by dividing light scattering by mass, it is important to understand if the model is accurately estimating either of those before attempting to minimize error in size or hygroscopicity. If the model cannot get those parameters correctly, adjusting size or hygroscopicity may be accounting for other errors. Does the model accurately estimate dry conditions? Are the errors in the observations accounted for? I recommend publication after addressing detailed comments below.

Comments

Pg 1:16, Does this reduction correspond to all of North America?

Pg 1:17-20, Are these reductions also reflecting the size shift or just the use of kappa?

Pg 2:4, Perhaps “expansion” is a more accurate term than “growth”?

Pg 2:7, Remove “fine” before “particulate mass concentrations” because this is followed by 10 um and add “PM2.5” before “chemical composition” to be clear that PM10 is not speciated.

Pg2:9, Replace “measurements” with “estimates” because mass scattering efficiency is not measured.

Pg3:8, Define coarse mass as “PM10-PM2.5”

Pg 3:5, bsp isn’t measured at all the sites. Perhaps add a line saying that bsp is measured at a subset of sites. This is stated in line 10, but it could be misunderstood here. Perhaps bsp and RH could be left off this list here and then it is clear from line 10 that they are measured at a subset of sites.

Pg3:10, bsp is “reported”, not “measured” hourly.

Pg3:14, It is important to consider the impacts of the truncation error for the open air nephelometer and how this can affect scattering where coarse mass is a major contributor.

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Pg3:16-17, Can the authors elaborate on why sea salt concentrations are not available before 2005? What parameter is being used to calculate sea salt?

Pg3:20, Do the authors also apply completeness criteria for mass? How many complete years are required for this time period?

Pg3:21, Does average bsp here correspond to ambient RH conditions? Are these the average of daily values or annual values?

Pg4:5, Have the authors considered the influence of this multiplier on mass scattering efficiency? Comparisons of mass and bsp at dry conditions would help show that these assumptions are also appropriate and not affecting efficiencies.

Pg4:10, Please note that the IMPROVE PM2.5 measurement is not a FRM measurement and after 2011 the laboratory RH varied significantly and could contribute a large bias to PM2.5 and PM10 measurements (they are not “dry”). See the data advisory at: http://vista.cira.colostate.edu/improve/Data/QA_QC/Advisory/da0035/da0035_IncreasedRH.pdf

Pg4:11, What RH are these calculated at- model or measured?

Pg4:15, Do the comparisons with the IMPROVE network correspond only to IMPROVE data from 2006 or from an average of 2000-2015? Given the yearly variability, it would more defensible to compare data and modeled values over the same time period.

Pg4: Section 2.3 I have several questions regarding how mass scattering efficiencies are calculated. As mentioned before, truncation errors for the open air nephelometer can impact measured bsp (See Malm and Hand, 2007), essentially removing some (up to half) of the CM scattering – resulting in a discrepancy between scattering and PM10 mass. In addition, dividing ambient bsp by a “dry” PM10 results in an ‘enhanced’ mass scattering efficiency if bsp and mass don’t have the same amount of associated water. PM10 is generally “dry”, but as mentioned earlier, after 2011 it can contain a significant amount of water based on the uncontrolled laboratory measurement RH which has nothing to do with ambient RH. If you’re comparing data only from 2006

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this won't be an issue. But missing the coarse mass contribution to bsp could result in an underestimate of mass scattering efficiency depending on location because in some regions CM is a major contributor to PM₁₀. It is unclear to me why IMPROVE mass is used to estimate the modeled mass scattering efficiency? This would seem to introduce more uncertainty and less consistency in the estimates, especially given the spatial resolution of the model.

It would be helpful to see a comparison of modeled and measured mass concentrations and bsp. Do the predictions of SIA and OA generally agree with the observations? Can the model accurately predict measured bsp? It seems it would be important to know the model performance of the parameters that go into estimating mass scattering efficiency before trying to tune the model results using size and hygroscopicity. While perhaps the new revised estimates are more accurate, it is hard to know whether they are just accounting for other issues within the model or the measurements. Without the basic comparisons, the comparison is less convincing.

Pg5:4, What is “v” as the kappa subscript refer to?

Pg5:20, Include “annual” before “average”.

Pg5:22, It would help to at least mention that the figure also includes revised estimates that will be introduced later, otherwise the figures appear to be referred to out of numeric order (figure 3 is discussed before figure 2, right side- etc.).

Pg6:1, Is the PM_{2.5} here referring to observations or model estimates?

Pg6:1, Add “fine” before “dust”

Pg6:12, Can the authors reconcile studies that demonstrate that even at low RH particles will still have water associated with them? (e.g., Santarpia et al., 2004, 2005; Carrico et al., 2000, 2003, 2005; ten Brink et al., 2006; Malm and Day, 2001; Malm et al., 2003, 2005; Xu et al., 2002; Im et al., 2001; Eldering et al., 2002)

Pg8:20, The authors have seemingly tuned their model to the parameters that give the

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closest agreement with observations so it isn't surprising that the revised estimates agree more closely. However, they don't comment on why the model inherently had too large of sizes. Have other studies also observed this? Is it consistent with size distributions measured at surface sites? Given some of the issues with observations mentioned earlier, it would be important to understand whether the model adjustments are just accounting for some of the measurement biases. This could be partly answered if comparisons of modeled and measured mass concentrations and bsp were performed.

Pg9:18, Mass scattering efficiencies of 10-12 m²/g are exceptionally high unless they correspond to enhanced scattering efficiencies with higher RH bsp and dry mass. Providing the RH that these values correspond to is important.

Pg11:14, Again it would be helpful to show a comparison of measured and modeled bsp and of mass separately.

Pg12:4, Again, see the earlier advisory. IMPROVE PM_{2.5} and PM₁₀ measurements are not FRM. Pg12:8, Why is it necessary to partition IMPROVE OM? Unless I have missed something, hygroscopic calculations for IMPROVE mass weren't done as part of this paper?

Figures Pg19:4, Figure 1, Provide the wavelength and RH (or state ambient) for bsp. The sentence starting with "overlying circles" seems redundant.

Pg 20, Figure 2, please include the years included in the comparison, and the wavelength corresponding to the efficiency. It would help to also denote the parts of the figure as well.

Pg20, Figure 3: Similar comment regarding wavelength as other figures (this is helpful for readers if they are scanning figures and don't have to dig through the text for this information). Please include the years this comparison was made over. Also comment on "default" and "revised".

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Pg21, Figure 4: Include wavelength. Define SIA, OA, PMcoarse. Are these delin-
eations based on modeled or measured composition measurements? Please state in
caption. Also include RH ranges (ambient?) and years of comparison.

Pg 21, Figure 5: Please include wavelength. What kappa was used for OC and SIA?

Pg 22, Figure 6: Please include wavelength and years of comparison.

Pg 23, Figure 8: Please define kappa,s and kappa,o.

Pg23, Figure 9: Please define SIA and OA. “Hygroscopic” has a typo.

Pg24, Figure 10: Define SIA and OA.

Pg25, Figure 11: Please provide wavelength and RH (ambient?), define SIA and OA.
This color table makes it hard to see the sites in the Southwest.

Pg26: Figure 12: Please provide wavelength and RH (ambient?)

Pg 28, Figure B2: This color table makes it hard to see the comparisons.

Pg 30, Table A1, Define SIA and OA, kappa,s and kappa,o (use similar subscripts as
SIA and OA?)

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