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Interactive comment on "Sensitivity of aerosol optical properties to the aerosol size distribution over central Europe and the Mediterranean Basin" *by* Laura Palacios-Peña et al.

Anonymous Referee #2

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This manuscript summarizes an interesting and potentially useful study to examine the effect of varying the aerosol size distribution parameters within the WRF-Chem model on aerosol-radiation interactions. This work is an examination of findings in the literature that aerosol optical depth and radiative effects are sensitive to modal parameters such as geometric mean diameter and standard deviation, and that descriptions in models need to more closely align with observational evidence for narrower size distributions than are typically prescribed.

I am an experimentalist, so my understanding of the use of modal representations of the aerosol size distribution in the WRF-Chem model is limited. From the manuscript,

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the model uses the GOCART scheme, in which (lines 108-111) " bulk mass and number is converted into an assumed log-normal modal distribution, then dividing the mass into sections or bins." So from this I gather that the mass for different aerosol components is carried in the model, and then different components are divided into different lognormal distributions (e.g., dust and sea-salt mass would go in the coarse lognormal mode, sulfate into the accumulation and Aitken lognormal modes, etc.). These lognormal distributions have prescribed geometric mean diameter Dg and geometric standard deviation Sg, while the mass varies with the abundance of each component as predicted by GOCART. The number follows from the mass; it is not explicitly predicted by GOCART. Once this mass apportionment has been done, the lognormal distributions are chopped into discrete bins, the number of particles in each bin is calculated, optical and hygroscopic properties for each component are assigned, and then the Mie parameterization is applied to calculate optical parameters such as extinction and absorption.

Assuming this is correct, I see some substantial issues with the manuscript as currently written. My major concerns are as follows: 1) There seems to be a fundamental lack of understanding of the lognormal function. For example, lines 43-44, it is stated that the 3rd moment is mass and the 4th is volume. In fact, mass is just the 3rd moment (volume), multiplied by particle density. Furthermore, there are (at least) two variations of the lognormal function found in the aerosol literature. One is:

F=(N*ln(10)/(sqrt(2*pi)*ln(Sg)))*exp(-.5*(ln(D/Dg)/ln(Sg))ˆ2), where N is the particle number (or volume if the lognormal describes the volume distribution). This is the differential function, describing dN/dlogDp.

The other common formulation is:

F=N*exp(-1*(ln(D/Dg)/ln(Sg))ˆ2)

In the one I'm most used to, the first, the geometric standard deviation represents the multiplicative standard deviation. For example, for a lognormal distribution with a Dg

of 1 μm and a Sg of 1.5, 63% of the distribution would lie with diameters between 1/1.5 and 1*1.5 μm. In this formulation, Sg can never be <1; a Sg of 1 represents a monomodal aerosol. This is the formulation that is used in most, but not all, of the values for Sg in Tables 2 and 3. In the second formulation, I believe Sg can be <1. This is likely the formulation used in Remer et al., 1998, listed in Table 2.

For the current manuscript the values for Sg used for the base case are 1.7, 2.0, and 2.5 for the Aitken, accumulation, and coarse modes, which would imply the first formulation was used. However, if this is the case, the sensitivity case for L50_Sgai is run with a value of 0.85, which is physically impossible. The sensitivity case for L50_Sgac has a value of 1.00, which would imply a monomodal size distribution.

2) The values of Dg for the accumulation mode listed in Table 1, "found through a comprehensive review", are implausibly small. The accumulation mode is often defined as having diameters from 0.1 to 1.0 μm, yet all the values except the "H50" in this table list a Dg of <0.084 μm. I have a strong suspicion that radius has been confused for diameter.

3) At lines 225-227, the case of "L50_SGacc", which I assume means a reduction in 50% in the geometric standard deviation of the accumulation mode relative to the base case, is described as leading to a reduction in both number concentration and in mass. This is nonsensical; reduction of the standard deviation of a lognormal distribution does not change the integrated amount of the parameter; it merely changes the width of the PDF. If the lognormal is describing the distribution of mass in the mapping of GOCART mass to diameter, then the mass should be held constant as the standard deviation changes (number would change, however). Alternatively, if the number is held constant, then the mass will change. But both should not change if just the standard deviation is changed.

There are a number of more minor issues with the manuscript, including no description of the coding used for the different cases ("L50_Sgac", "H20_Dgai", etc.). Figure cap-

tions do not describe the contents of the figures (for example, in Figure 2 there is no explanation in the caption for the curves that are plotted.) And there are many unexplained parameters and typographical errors. I recommend a thorough proofreading.

To be acceptable for publication, I recommend the following: 1) Clearly define the lognormal function that is being used. Please read J. Heintzenberg, "Properties of the Log-Normal Particle Size Distribution" in Aerosol Science and Technology, 1994, https://doi.org/10.1080/02786829408959695. Also, the Seinfeld and Pandis textbook has a substantial section on the lognormal distribution that should be reviewed.

2) Clearly explain how the GOCART masses are mapped to the lognormal functions (i.e., is mass being conserved during the sensitivity tests, or is mass?). How are the different components–dust, sea-salt, secondary sulfate–being mapped to the different lognormal modes? Probably a separate section with equations and a thorough verbal description of this mapping is warranted.

3) Ensure that the range of literature values are all defined using the same definition of the lognormal function. My preference would be to use the more common definition listed above, wherein all values of Sg are >1.

4) Make sure that diameter, rather than radius, is used consistently.

5) Make sure all figure captions describe the contents of the figures clearly. For example, Fig. 1 caption currently says, "AOD at 550nm and differences for simulations of sensitivity test at 50%". It should say something like, "Map showing the difference between the base case and sensitivity tests using 50% changes in parameters. a) Base case showing AOD. b) Aitken mode with 50% reduction in Sg. c) Accumulation mode with 50% reduction in Sg." etc. Or have much larger, clearly defined labels on the columns and rows.

6) Perform a very thorough proofreading, making sure all sentences are logical and complete and looking for grammatical errors. Perhaps Dr. Fast could help with this.

It is a very useful exercise to investigate how uncertainties in assumptions about aerosol size distributions are reflected in AOD, and this manuscript has good potential. However, the fundamental confusion about the definition of the lognormal function being used and the appropriate range of lognormal parameters for sensitivity tests makes me concerned that there are substantial errors embedded in the calculations and results. These, and issues of presentation, must be dealt with before the manuscript can be considered again for publication in GMD.

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