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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Regional and global modelling of aerosol optical properties with a size, composition, and mixing state resolved particle microphysics model" *by* F. Yu et al.

Anonymous Referee #1

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This paper uses a sophisticated sectional aerosol microphysics module (APM) in the global chemical transport model GEOS-Chem to explore how secondary aerosol mass produced in the atmosphere is distributed among secondary (nucleated sulphate) and primary (e.g. sea-salt, dust, carbonaceous) particles. The paper further usees GEOS-Chem-APM to quantify the contribution to the global aerosol optical depth of these different particle types. The paper then presents a brief evaluation of the GEOS-Chem-APM simulated annual-mean global distribution of total AOD against AERONET, MODIS and MISR measurements. Finally, analysis of the temporal evolution of APM simulated aerosol optical properties within the regional WRF-Chem model is presented, with comparison to AERONET measurements.

Both global (GEOS-CHEM) and the regional (WRF-Chem) APM simulations suggest that secondary (nucleated) particles provide the dominant contribution to aerosol optical depth in continental regions (except in regions with strong dust and biomass burning sources). The regional simulations suggest that North America experiences frequent plumes of high-number-concentration arising from nucleation events.

The paper is certainly within the scope of ACP and the role of mixing state and particle formation mechanisms on simulated aerosol optical properties will be of considerable interest to the aerosol-climate modelling community.

Although there are now many global aerosol microphysics models developed, there are very few studies which have attempted to quantify the contributions of primary and nucleated particles to aerosol optical properties.

The paper is quite well written, although the written style could be shortened in many places to more succinctly summarize discussions/findings. In particular, section 2 needs to be considerably re-worked – it is too long and some of the text needs substantial attention – needs to be reduced to the minimum.

Some of the descriptions of the model methods were also a little vague in places and this section (3) also needs to be sharpened up a little (see specific comments).

However, the results are presented well and the findings are very interesting. The implementation of the aerosol-radiation module for the APM model into the global and regional modelling frameworks also represents a substantial model development. I therefore recommend the paper be published once the specific (minor) revisions I list below have been attended to.

Specific revisions

1. Abstract, Line 8: The sentence beginning "According to GEOS-CHEM simulations.." is slightly confusing as it is unclear what the "secondary species" refers to – to be clear I would say "secondary species mass" – so suggest to say:

According to GEOS-Chem-APM simulations, in most parts of the globe, the mass of secondary species resides mainly within secondary particles (60-90%)..." and continue as currently.

2. Abstract, Line 18: The sentence "the model captures the absolute values as well as the variations of AOD at the AERONET sites" needs to be made more quantitative – the authors should derive mean-bias and correlation coefficient for the comparison to the AERONET measurements. I also suggest to reword to say "the model *mostly* captures the absolute values..." so as not to over-state how well the model compares.

3. Introduction, page 95, lines 5-6: the sentence beginning "Modeling of these aerosol parameters...." is difficult to read – suggest put commas between "parameters" and "as" and between "variations" and "by".

4. Introduction, page 95, lines 9-10: suggest to delete this sentence which begins "In addition to the.." as the information in that sentence is given again straight afterwards.

5. Introduction, page 95, lines 11-12 – need to give a clearer sentence here – the part of the sentence which states "treat BC particles either as externally or internally mixed" should be rewritten to emphasise that most aerosol schemes in climate models treat BC as externally mixed (give some references) but refer to some global aerosol microphysics models (e.g. Stier et al., 2005; Bauer et al., 2008; Spracklen et al., 2008) which do resolve the internal mixtures.

6. Introduction, page 95, lines 14-18 – the sentence would be made easier to read by adding commas between "particles" and "within" and between "sizes" and "are". Also suggest to add "internally" between "are" and "mixed" in that sentence. Suggest also to change the comma after "BC particles" to a full-stop and then start a new sentence replacing "and showed that.." with "In particular they showed that...". Also suggest to replace "occurs" with "is resolved", replace "where as" with "whereas".

7. Introduction, page 95, lines 24-29 to page 96 lines 1-2– this para can be shortened

substantially – and where the authors should give some reference for the importance of resolving water uptake online.

8. Introduction, page 96, line 3 – suggest to delete the sentence beginning "It is clear from above discussions that....". The para stands on its own without that sentence - suggest to remove.

9. Introduction, page 96, line 12 replace "aerosol climate" with "aerosol-climate".

10. Introduction, page 96, lines 18-19 – need to clarify what is meant by "similar approximations"

11. Introduction, page 97, line 9 – replace "look into" with "investigate".

12. Suggest to add sentence at the end of the Introduction stating that although many papers have investigated the role of nucleated particles on CCN/indirect forcing (Spracklen et al, 2008; Merikanto et al, 2009; Wang and Penner; 2010) this is (to my knowledge) the first to attempt to quantify the contribution to aerosol optical depth of the secondary (nucleated) particles.

13. Section 2, page 97, line 23 – make it clearer that kr and ki refer to the shell here.

14. Section 2, page 98, lines 15-30 and page 99 lines 1-9 – these two paragraphs essentially describe the information contained within Tables 1 and 2 – suggest to reduce the text to one of two sentences and also refer to combined Tables 1 and 2 (see later comments)

15. Section 2, page 99,lines 12-19 – I don't understand this sentence – please clarify what is meant here – don't you just mean that you evaluate monochromatic aerosol optical depth for at several wavelengths to compare against AERONET, MODIS and MISR?

16. Section 2, page 99, lines 18-30 and page 100 lines 1-3– again this para could be shortened substantially – pleae remove "it should be noted that" and replace "For

particles of same sized (dcore+dshell), the particles with and without BC and dust cores" with "For same size particles (dcore+dshell), the 3 types of 400nm particle....".

17. Section 2 page 100 lines 4-7 – suggest to delete this first sentence completely (and remove in the Figure the dots showing the interpolation which just makes the Figure more difficult to read).

18. Section 2 page 100 lines 8-9 – suggest to remove the sentence "For particles without solid cores, dshell is their diameter." and replace "It is clear from Figure 2a that coating..." with "Coating..." and put "(Figure 2a)" after "dshell«dcore".

19. Section 2 page 100 lines 8-24 and page 100 lines 25-30 and page 101 lines 1-3— I think this Figure is confusing because the changes are combining size effects and mixing effects – it is not clear what you are trying to show here – and I don't understand some of what is written here – these paras need re-writing.

20. Section 2 page 101 lines 8-10 – be clear what is meant by that ki value – does this refractive index refer to the whole (internally mixed) particle or to the shell only? Also Alexander et al (2008) is missing from References.

21. Section 2 page 101 line 18 – replace "A number of recent studies" with specific references.

22. Section 3 page 101 lines 29-30, page 102, lines 1-5 – shorten existing text here but add some references to other bin-resolved models that have used similar approaches (Pierce et al, 2007)

23. Section 3 page 102 line 17 – So the APM approach emits primary BC and primary POC as externally mixed particles? – many models treat the BC and POC as internally mixed at emission – suggest to add sentence here clarifying the difference approach and any limitations on the findings in the study.

24. To check– in Yu and Luo (2009) – section 2.3.2.3 the description explains that the model does indeed co-emit the BC with OC within internally-mixed BC/OC particles.

Why is the different approach taken here?

25. Section 3, Page 102 line 24 – "The condensation of LV-SOA on various particles..." is vague – is condensation to all the particles types considered? Also it is stated that "the uptake of SV-SOA and MV-SOA is calculated based on partition theory" with reference given as Chung and Seinfeld (2002) – please clarify the approach used here. Also page 103 lines 7-8— reword that similarly.

26. Page 103, lines 12-15 – suggest to delete these 3 sentences – not needed for para. Then delete first half of following sentence "Because of..." and start sentence "We use GEOS-Chem-APM....."

27. Page 103, lines 22-23 – suggest to remove "ageing" from "successive oxidation ageing"

28. Page 104, line 7 – replace "It is clear that.." with "Figure 3b shows that..." and delete "(Fig 3b)" later in sentence – also suggest to make reference to this being because SPs are the dominant contribution to the condensation-sink.

29. Page 104 line 16 – replace "ratio of total mass... " to "ratio of total dry mass...." and then can delete the following sentence "It should be noted that aerosol water....."

30. Page 104, line 21 – delete "For SP, the core component is considered to be sulphate and it is clear from" and start sentence "Fig 4a shows that....."

31. Page 104, line 22 – replace "The SP is dominated by" with "The SP mass is dominated by.."

32. Page 104, lines 24-25 – suggest to move the sentence "Over the oceans and polar regions" to be before the sentence starting "The SP" as that statement follows on more closely from the overall picture given for continental regions given in the 1st sentence.

33. Page 104, line 26 – replace "it becomes <0.5 because of the dominant of" with "where" and add "dominate" at the end of the sentence.

34. Page 105, lines 12-15 – Suggest to add a sentence here stating that that particular finding is strongly dependent on the treatment of the BC and OC at emission –> this "coated OC" is considered not to contain BC because of the external mixing assumption at emission. Please comment on this. Also please refer to range of kappa values used in the literature for OC.

35. Page 105, line 24 – replace "DRF" with "climate" –> the sink of SPs affects CCN and aerosol indirect effects as well as direct effects.

36. Page 105, lines 24-27 – Suggest to re-word this sentence – the word "properly" is not really appropriate here –> also suggest to replace "necessity to treat" with "importance of treating".

37. Section 4, Page 106 lines 5-6, replace "(each chemistry time step)" with "in the model"

38. Page 106, lines 6-7 replace "It should be pointed out that while..." with "While..."

39. Page 106, lines 13 – insert comma between "particle wet size" and "which"

40. Page 106, line 15 – replace "average" with "volume-average"

41. Page 106, lines 16-17 – delete sentence "The kappa values for various....." – already said in that in previous sentence!

42. Page 107, lines 17-18 - delete "hydrated (i.e. wet)" – and later in that sentence delete "set the core size to zero and".

43. Page 107 lines 5-6, replace "various particles to global AOD" with "various particle types to AOD globally".

44. Page 107 line 8 replace "generally confined to" with "present in highest concentrations in"

45. Page 107 line 14 replace "most of particle extinction" with "most particle extinc-

tion" and insert "the" between "above" and "surface". Suggest to delete the following sentence beginning "High extinction coefficient..." and start the next as "However, the vertical gradient....."

46. Page 108 lines 3-4 Delete the sentence "The model calculation does not have this limitation" and start the next sentence as "Consequently.., " rather than "Nevertheless.."

47.Page 108 line 20 replace "AEORNET" with "AERONET" – and also please give some quantitative mean-bias and correlation values for the comparison of the model and AERONET AOD.

48. Page 108 line 21 replace "It can be seen from Figs 6 and 7 that" with "Figs 6 and 7 show that..."

49. Page 108 lines 26-28 suggest to replace the sentence "Our comparison of model...." This is obvious from the next sentence.

50. Page 109 line 1 replace "yet" with "with" and replace "is" with "even".

51. Section 4.2 Page 109, line 7-8 give reference after "WRF-Chem" and delete the text that follows "and the details of..... can be found in Luo and Yu (2011)."

52. lines 8-10 Replace "the model using the relevant" with "WRF-Chem-APM" and replace "measurements obtained during..." with "against measurements from...", replace "in the summer of 2004" with "(summer, 2004)" and give a reference for the INTEX-A field campaign – and replace "WRF-Chem-APM" with "the model".

53. Page 110, line 5-8 – The comparison to the obs at station 7 is only moderate whereas the other sites look much better. The authors should comment on the potential cause of the high AOD events on days 10-11 and 13-14 that are missed by the model.

54. Page 110 line 17 – Please state briefly what the conditions are that make the IMN nucleation rate so much higher on that particular day compared to the other days.

55. Page 111 line 14 – delete "it is clear from Figs 9 and 10 the drop" and replace with

"the simulation results (Figs 9 and 10) suggest that...."

56. Pages 117-118 Tables 1 and 2 Suggest to combine Tables 1 and 2 into 1 and remove the "Values at each point" column –> instead just put (geometric) or (arithmetic) after total # of points. And to delete the "No core" row under "d-core". Also a range of 0-1 is given for dshell for BC-core and Dust-core respectively –> is this really 0-1 micron??? it doesn't seem to correspond to the "values of each point". Also in the text referring to the Figure please explain why 91 and 61 points are considered needed in the No-core-dshell and dust-core-dcore dimension table whereas only 21/32 are needed for the others. Also - -is there no dependency of d-shell on d-core here? Also typo in d-core-dust-core –> says 0.02 in RH column but should be 0.05.

57. Page 119 caption to Fig.1 –> add "400nm" between "types of" and "particles".

58. Page 120 – "Dependenc" –> "Dependence" and suggest to remove interpolated lines from the Figures as this does not add anything and just makes the Figure harder to read.

59. Page 124 – Figure 6 – hard to pick out white AERONET sites in (a) and (b) – thicker lines?

60. Page 127 and 128– Figures 9 and 10 – need to add a), b), c) labels to caption as referred to in text. Also put stations numbers in white rather than black so they can be seen more clearly.

References:

Bauer et al., 2008; MATRIX (Multiconfiguration Aerosol TRacker of mIXing state): an aerosol microphysical module for global atmospheric models, Atmos. Chem. Phys., 8, 6003–6035, 2008

Pierce et al, 2007; Contribution of primary carbonaceous aerosol to cloud condensation nuclei: Processes and uncertainties evaluated with a global aerosol microphysics model, Atmos. Chem. Phys., 7, 5447–5466, 2007,

Merikanto et al., 2009; Impact of nucleation on global CCN, Atmos. Chem. Phys., 9, 8601–8616, 2009

Spracklen et al., 2008; Contribution of particle formation to global cloud condensation nuclei concentrations, Geophys. Res. Lett., vol. 35, doi:10.1029/2007GL033038.

Stier et al., 2005; The aerosol-climate model ECHAM5-HAM, Atmos. Chem. Phys., 5, 1125–1156, 2005

Wang, M. and Penner, J. E.: Aerosol indirect forcing in a global model with particle nucleation, Atmos. Chem. Phys., 9, 239–260, 2009,

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