

Interactive comment on “WRF-Chem simulation of aerosol seasonal variability in the San Joaquin Valley” by Longtao Wu et al.

Anonymous Referee #3

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This paper shows the WRF-Chem simulation of aerosols in the SJV in California for one year and compares the results with observations of AOD from one AERONET site at Fresno and from MISR for a domain covering SJV, as well as measurements of aerosol mass concentrations of PM_{2.5}, PM₁₀, nitrate, sulfate, EC, OC, and dust from IMPROVE measurements. It tests the effects of using two different model resolution and two dust schemes, and attributes the model problems in matching observed AOD and PM₁₀ to mainly the poor simulation of dust. It is stated in the “Introduction” that the paper a) “serves as the first step for future investigation of the aerosol impact on regional climate and water cycle in California” and b) provides a priori input for remote sensing retrievals for air quality for the MAIA mission.

While this paper has clearly shown the WRF-Chem performance over SJV that provides useful information, it lacks the vigor and thoroughness in the analysis and in-

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terpretation, and the information presented in the paper is insufficient in helping understand the problems of the model. Given the goal of using such a model for MAIA retrieval and for climate study, much more in-depth analysis and vigorous diagnostics is necessary in order for the model improvements to be useful for those purposes. Although the content is suitable for ACP, major revisions are necessary before the paper can be considered again for publication.

General comments:

1. Dust simulations: The authors have concluded that the dust simulation is the major problem for model to capture the observed aerosol amount and variability in the warm months. Switching from GOCART to DUSTRAN just shows different problems but does not resolve the issue. However, there is no any explanation on the differences between the two schemes in terms of emission strength, source location, parameterization of dust mobilization, and deposition in order to understand why the dust amount and seasonal cycles are so different between the two schemes and yet none can capture the observations. Without understanding the cause of the problem, future improvement is not possible.

2. Non-dust aerosols: Figure 4 clearly shows that the model does not have much skill to simulate sulfate and OC, but the problem has not been investigated. The ammonium is completely left out, which is an important part of total aerosol mass. Also, large fraction of aerosol is classified as “other”, but it is not clear what the “other” aerosols are in both model and IMPROVE data.

3. Optical properties: It is also not clear how AOD and aerosol extinction are computed from the simulated aerosol mass. Is aerosol microphysics package used for calculating particle sizes and mixing state? How is mass-based aerosol converted to extinction and AOD? Is the relative humidity considered in these calculations?

4. Chemistry: Nitrate, sulfate, and a significant fraction of OC are secondary aerosols that are produced by chemical reactions of their gaseous precursors in the atmo-

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sphere. The authors attribute the high bias of model-simulated nitrate to “high bias in nitrate emission”, which is erroneous. The diagnostics should involve investigations of nitrate precursors such as NO_x and HNO₃, and also the formation of nitrate via heterogeneous reactions on dust and sea salt surfaces and homogeneous reactions in the sulfate-nitrate-ammonium system. It is not clear how WRF-Chem deals with nitrate formations and which is the major reaction pathway for nitrate aerosol production. Same as sulfate – it is formed via gas and aqueous phase reactions of SO₂. Better diagnostics of the problem is needed.

5. Other physical processes: Dry and wet depositions are the major removal processes for aerosols. The seasonal cycles of these processes also need to be investigated. For example, can the differences in seasonal variations between model and obs be partly explained by the differences in simulated and measured precipitation amount that determines the wet removal of aerosols? Or if the winds are realistically simulated in WRF-Chem that not only affect the dust emission, but also advection, both have profound effect on aerosol temporal and spatial distributions?

6. Meteorological fields: The only meteorological field compared in the paper is the equivalent potential temperature, which provides information on the atmospheric stability. Other important met fields, such as precipitation and wind speed/direction, as mentioned above, plays key roles in aerosol removal, transport, and wind-driven emissions of dust and sea salt but have not even mentioned in the paper. In addition, these fields and the physical processes driven by them are resolution-dependent, so the role of these met fields should be examined at different spatial resolutions.

7. Lateral boundary conditions: The effects of lateral boundary condition should be examined, or at least discussed, particularly because of SJV's geophysical locations that is susceptible to the transpacific transport. How much of the aerosol species and their precursor gases are regionally/locally produced vs. imported from the lateral boundary, and how they affect the seasonal cycle? In other words, are the features/problems mainly produced by WRF-Chem? How important is the lateral boundary conditions to

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different aerosol species?

8. Emissions: It seems the anthropogenic and biomass burning emissions used in this work are not up to date. For example, why the authors choose to use NEI05 emissions instead of more recent ones (e.g., NEI 2011 or NEI 2014) to better match the simulated time period (2012-2013)? Why GFEDv2 is preferred instead of GFEDv3 that was released a few years ago or GFEDv4 that has been available since 2015?

9. Model-data comparison: 1) For AOD, there is only one AERONET site in the study region, and MISR's spatial coverage is limited. Why not use MODIS, which has a much better spatial coverage to have a better representation of “monthly average”, in addition or even instead of using MISR? 2) Which months are defined as “cold” or “warm” months? 3) More statistical quantities are needed to mark the agreement between model and observations, including correlation coefficients and seasonal/annual bias. 4) The authors should avoid using the subjective adjectives, such as “good agreement”, “reasonably well”, etc., to describe the comparisons between model and observations. More objective and quantitative methods and presentations are needed. 5) Given that air quality changes quite a bit day to day and air quality forecasts are given on daily bases, why all the comparisons are done on monthly time scale instead of daily or sub-daily?

10. The most important step forward is to understand the causes of deficiencies in the model and suggest/incorporate improvements for better results. However, the current paper does not offer those aspects.

Specific comments

Page 5, line 72-82: I wonder why Fast et al 2014 and Zhao et al 2013 were able to “reasonably” represent the observations with the same WRF-Chem model, either in the warm months (Fast) or on annual bases (Zhao), but this work has difficulties to do the same?

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Page 5, line 83: I don't think the word "extend" is appropriate – this study only focuses on SJV while Fast and Zhao showed large regions in CA.

Page 6, line 102-104: I don't get it – why simulation for SJV is critical to MAIA? Is MAIA only focuses on SJV?

Page 7, line 116: Why are the original wavelength(s) from AEORNET that you used to interpolate to 550 nm?

Page 8, line 146: What does "speciated" mean here? There is no aerosol species information from the CALIOP data. Marine, polluted continental, etc. provided by CALIOP are aerosol types, not species.

Page 9, line 179-180: How is convective transport (and removal) of aerosols simulated in 4-km resolution?

Page 9-10, line 183-184: Was the overestimation by MOZART in the free troposphere a factor of 2 such that the concentrations had to be divided by 2? If the overestimation was only in the free troposphere, why the concentrations in the lower atmosphere and BL were also divided by 2?

Page 10, line 198: Are the dust emissions in the GOCART and DUSTRAN also available in 20 and 4 km resolutions? What are the major differences between GOCART and DUSTRAN schemes?

Page 11, first paragraph in section 4.1: What PM_{2.5} species and precursor gases are emitted? Have you checked the domain budget between 4 and 20 km resolution to ensure the total emission for all species are identical with these different resolutions?

Page 11, line 215: How was AOD calculated without having information of PM_{2.5} composition? For example, dust and BC have very different mass to extinction conversion factor, known as mass extinction efficiency (MEE). There is no single MEE for a generic PM_{2.5} or PM₁₀.

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Page 12, line 237: As I said earlier, nitrate is not emitted but chemically produced. The precursor emission/concentration/transport/chemistry have to be examined to explain the nitrate.

Page 12, line 238: Why is simulation over Texas relevant here?

Page 12, line 242: Be specific on what "SOA processes" is referred here.

Page 12, line 244 and 246: Be quantitative – what is the standard of "good agreement"?

Page 12, line 250: How large is the "large low bias"?

Page 13, line 253-254: "The 4km simulation has better agreement...", but only in the cold season.

Page 13, line 254-255: "The 4km simulation captures seasonal variability of PM_{2.5} and its speciation": From Figure 4, the seasonal variability for the PM_{2.5} species are very similar between the 4- and 20-km simulations, only the concentrations are higher from the 4km simulation. The seasonal variability of PM_{2.5} sulfate and OC are not capture by both 4 and 20 km simulations.

Page 13, line 267-268: The 4km_D2 overestimates PM_{2.5} by 52%, but it overestimates the PM_{2.5_dust} by up to a factor of 4 in the warm season!

Page 13, line 270-272: As I suggested earlier, please show correlation coefficients on all comparisons (in addition to the bias), which indicates how model and data agree on seasonal variations.

Page 14, line 285-286: How much better does 4km_D2 agree with MISR than other simulations? Visually, JAS is still nowhere near MISR, and AMJ is higher than MISR. Please quantify the degree of agreement.

Page 14, line 290-292: I don't understand the statement of "reasonably capture the vertical distribution", even though the model has "low biases in the boundary layer and high biases in the free troposphere". To me, this is rather "unreasonable".

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Page 15, line 298-299: “. . .suggesting relative good performance. . .”: How good? Figure shows poor agreement between obs and model for sulfate and OC, so they are not “good” at all.

Page 15, line 303: How to explain that dust from 4km_D2 is way too high but the extinction in the boundary layer is still way too low?

Page 15, line 313 and 316: If the model has weak vertical mixing, the aerosols should be trapped within the BL and not transported to high altitudes. But the model actually overestimates the aerosol at high altitude – what is the source of high altitude aerosol?

Page 16, line 321-322: This precisely indicates the need to quantify the role of chemical boundary conditions.

Page 16, line 323-324, “good performance. . .”: But in JFM the model results are much higher (by a factor of infinity?) at above 1.5 km! How can that be evaluated as “good”?

Page 16, line 330: “reasonable simulation”, “good representation” – what are the measures of reasonable and good here?

Page 16, line 337: Please explain what “climatological fire emissions” mean.

Page 16, line 339-340: Why can Wu et al do it right for South America fire but cannot do it for California? What are the major obstacles?

Page 17, line 371-372: No need to spell out what GOCART and DUSTRAN stand for at the last part of the paper, since they have been introduced and used many times earlier in the text.

Page 17, line 383-385: Unfortunately, I cannot see how the evaluation in this study can be apply to other regions to ensure that aerosols are simulated correctly for the right reasons. This paper has shown the problems but has not shown how to solve the problems with what approach.

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