Response to comments from Reviewer 2

Review of "Development of a source oriented version of the WRF/Chem model and its application to the California Regional PM10/PM2.5 Air Quality Study" by Zhang et al.

Synopsis

Zhang et al. present new developments within the regional scale online coupled chemistry transport model WRF/chem in which they include the possibility to explicitly describe the aging (coating) of particulate matter of different origins (emission source types). In contrast to the implicit assumption of instantaneous internal mixing of aerosols made in the aerosol modules available in WRF/chem so far, particulate matter emitted by several different source categories is tracked separately and mixing / coating processes are described explicitly. They apply their new model system over the domain of California and investigate the effects of mixing state on the radiation budget. A three week stagnation period during winter 2000/2001 serves as evaluation period.

General decision: Major revisions

Zhang et al. present a very interesting addition to the WRF/chem modeling system, and it appears the model is working correctly and the results seem reasonable. The way methods and results are presented, however, does not warrant publication in ACP in the current form, and I strongly suggest major revisions to be done before publication.

I applaud the efforts of the authors to describe in great detail how they changed the source code to implement the new module, however this does not have substantial scientific merit, and more important key scientific information about the implementation of the new module is lacking. The description of the results is sometimes confusing. The structure of the paper could be streamlined (see my suggestions below). The figures are presented in a sloppy manner and sometimes incomprehensible (e.g. Fig. 4).

This list of my perceived deficiencies of the manuscript should not deter the authors from updating their manuscript and submit it in revised form, as I do think it is a very useful addition to WRF/chem. In summary I suggest to accept this paper, but only after a substantial revision.

Response: The authors thank the reviewer for comprehensive suggestions to improve the manuscript. Changes and response are made according to all the comments. Specific responses are listed with each comment.

Major remarks

* The authors describe in detail how they wrote/changed the source code to make the module work, but are only vague about how each process is implemented scientifically. I suggest to move all descriptions of source code changes to the Appendix. See also my detailed comments below.

Response: Details about the model development were moved to appendix and the main text was revised to focus on the scientific contributions.

* This is a newly developed aerosol module. More convincing comparison against measurements (more stations, size distributions, highly time resolved chemical composition) or against and established aerosol module in WRFchem (MADE, MOSAIC) would be beneficial.

Response: The aerosol module employed in the current study is not new. This aerosol module has been used extensively in previous source oriented studies that employed off-line air quality models. Applications with a rigorous performance evaluation have been published for multiples studies in California and Texas with both prognostic (from both WRF and MM5) and diagnostic meteorology. The aerosol module has even been used in an off-line air quality model for the specific episode studied in the current paper, with overall good performance when compared to observations. References to these previous studies are included in the Introduction.

The purpose of this paper is to implement the aerosol module while calculating feedbacks to meteorology so that the effects of the fully source-oriented representation of particles can be quantified. The evaluation of meteorological parameters, particle scattering coefficients and pollutants concentrations in Tables 2, 3 and 4 validates the performance of the model features that are unique to the current study.

The source-oriented aerosol mixture model developed in this study is fundamentally different from other aerosol modules in WRF/Chem. A comparison against those internally mixed models does not help to quantify the effects of the particle mixing assumptions. It would be interesting to investigate the differences caused by mixing assumptions when MADE, MOSAIC, and other modules in WRF/Chem are employed, but this is beyond the scope of the current study.

* Immediate physical and chemical effects (settling, coagulation, condensation, gas phase concentrations) of the explicit representation of mixing state cannot be distinguished from radiative effects. I suggest to conduct two sets of simulations with instantaneous internal and explicit mixing (one without aerosol radiation feedbacks, and one including it) and try to separate these effects.

Response: Previous studies that used off-line air quality models cited in Introduction Section investigated the differences between external and internal representation without feedbacks and found that the differences are negligible. Thus, the differences in this study are from radiative rather than physical and chemical effects.

* A clear description of the configuration options of WRF/chem for which this new addition has been developed and tested is sorely lacking. WRF/chem offers a wide range of parameterizations for meteorological (convective transport, turbulence, advection, radiation...) and chemical (gas phase, dry deposition, wet deposition, photolysis rates). The authors need to clearly state firstly which options they chose, and secondly whether their module only works with these options, or, if so, with which other ones it does so.

Response: The authors thank the reviewer for this important suggestion. A table showing the configuration options was added (Table 1).

* Several times the argument is made that the changes to aerosol mixing state affect atmospheric mixing, but this is never shown directly. You can show this however, e.g. by comparing PBL heights or TKE.

Response: Comparison of PBL was added in Figure 6. It does show that atmospheric mixing is affected.

* In the evaluation the authors mostly give only 1 number for a certain change, usually picking a single very high value. This is not an accurate description of the results and a better representation of the range of changes (e.g. 25 50 75% values of change, or mean + 1 sigma, or median + 95% value) is needed. Plotting histograms of changes in Figures 7-12 could be useful for that.

Response: The overall averaged changes of each plot were calculated and shown on the plots.

* The authors should carefully address the questions raised in the scientific comment of J. Fast.

Response: Comments from J. Fast were addressed.

Detailed remarks

p 16458

5-6: avoid repeating "The source oriented approach".

Response: The manuscript was modified accordingly.

10: while this might be true, it is not a conclusion you can draw from this work. Please remove, or weaken ("explicit representation of mixing state should lead to a more accurate representation of radiative feedbacks").

Response: The manuscript was modified accordingly.

16-21: This description of your model setup is too detailed for an abstract. I suggest removing.

Response: The manuscript was modified accordingly.

20-22: (In case you keep the sentence in the abstract nonetheless) Split sentence starting with bin sizes and ending with domain resolution into 2 sentences.

Response: The manuscript was modified accordingly.

26-28: rephrase: "Effects of a source oriented representation of particles with meteorological feedbacks in WRFchem on a number of meteorological parameters were identified: downward..."

Response: The manuscript was modified accordingly.

p 16459

4: (and also on several occasions throughout the manuscript) if you use “central valley” as proper noun for the valley in the center of California you should use upper case. (“Central Valley”)

Response: The manuscript was modified accordingly.

5: Explain why does extinction goes down. Simplified: shouldn’t a black particle (uncoated BC) vs a white particle (coated BC) only affect the distribution between scattering and absorption?

Response: Both scattering and absorption are enhanced with coating of secondary pollutants on black carbon (Zhang et al., 2008).

8: you do not show that mixing changes. Either remove, or show (see general remark).

Response: The changes of PBL height are added in the paper, so this statement is retained.

14: why only nitrate? what about sulfate? SOA?

Response: Nitrate is the dominant secondary species in the SJV during winter stagnation events, with measured concentrations of PM2.5 nitrate reaching up to 50-60 μgm^{-3}. In contrast, measured concentrations of PM2.5 sulfate are less than 5 μgm^{-3}. SOA concentrations are also lower than nitrate concentrations in the winter due to low oxidant concentrations. Future applications of the SOWC model will study episodes where sulfate and SOA concentrations are dominant.

20: please add Fast et al., 2006 and Peckham et al., 2011 to the references for WRFchem (see also here: http://ruc.noaa.gov/wrf/WG11/References/WRFChem. references.htm)

Response: The references were added.

24: Do not repeat “stagnation events”.

Response: The manuscript was modified accordingly.

26: make clear that this sentence is only true during stagnation events.

Response: The manuscript was modified accordingly.

p 16460

1: again the atmospheric mixing argument please add citation.

Response: References were added.

6: there are several aerosol modules implemented into WRFchem(MADE, MOSAIC, GOCART), and at least both MADE and MOSAIC have a detailed description of particle size and composition. You should cite them all, not only Zaveri et al., 2008 (which is MOSAIC). Make clear that you add another one in this work.

Response: References were added for MADE and GOCART. And text was modified to clarify that a aerosol modules was added.

6: see J. Fast comments about other work on explicit representation of mixing state. (I.e. cite

Matsui et al., 2012)

Response: References were added.

10: time scales need references.

Response: References were added.

17-23: COSMO-ART (Vogel et al., ACP, 2009, http://www.atmoschemphys.net/9/8661/2009/acp986612009.html) is comparable to WRFchem and considers explicit aging of soot (Riemer et al., JGR, 2003, http://onlinelibrary.wiley.com/doi/10.1029/2003JD003448/abstract). They evaluate radiative effects.

Response: This paper was added to the references.

p 16461

7: clearly define what you will refer to in the following as "aging". Only coating? Or also coagulation. See J. Fast's comment: can particles of different source categories coagulate?

Response: Yes. Both coating and coagulation age particles. Fast's comment was addressed.

8: start a new paragraph after "hours to days". Introduce it with something like "In this work",

"This paper is structured as follows", ...

Response: The manuscript was modified accordingly.

19ff: The background section is not exhaustive enough to justify a complete section. It could be removed, and be partially merged into the introduction (where you talk about other modeling studies), and you could move the CRPAQS description to Section 4.

Response: The manuscript was modified to move the CRPAQS description to Section 4, but the remainder of the background was maintained in Section 2.

20: if they are so numerous then you should cite some more, and then highlight (only) their main findings. In the manuscript you do this in too much detail for (only) two that seem "cherry-picked".

Response: A total of 23 papers using the source-oriented air quality model have been published by the Kleeman research group. Several additional examples are now cited in the manuscript, but we prefer to not cite the entire list since this does not add any value to the paper and appears self-serving. For the Reviewer's benefit, a synopsis of the 8 most recent is provided below.

1) 2012 A. Mahmud, M. Kleeman. Quantifying Population Exposure to Airborne Particulate Matter during Extreme Events in California Due to Climate Change. Atmospheric Chemistry and Physics, 12:16, pp7453-7463. Climate change makes extreme PM2.5 concentrations higher in California.
2) 2012 M. Kleeman, C. Zapata, J. Stilley, and M. Hixson. PM2.5 Co-benefits of Climate Change Legislation Part 2: California Governor's Executive Order S-3-05 Applied to the Transportation Sector. Climatic Change, 117:1, pp399-414. Climate change measures have the co-benefit of reducing PM concentrations from multiple sources.
3) 2012 C. Zapata, N. Muller, and M.J. Kleeman. PM2.5 Co-benefits of Climate Change Legislation Part 1: California's AB32. Climatic Change, 117:1, pp377-397. Climate change measures have the co-benefit of reducing PM concentrations from multiple sources.
4) 2012 Hixson, M., A. Mahmud, J. Hu, and M.J. Kleeman. Resolving the Interactions between Population Density and Air Pollution Emissions Controls in the San Joaquin Valley, USA. Journal of the Air and Waste Management Association, 62:5, pp566-575. Smart growth measures in the SJV will increase population exposure to common sources of PM2.5 such as diesel engines, gasoline engines, and food cooking unless control strategies are simultaneously implemented.
5) 2010 Mahmud, A., M. Hixson, J. Hu, S. Chen, and M.J. Kleeman. Climate Impact on Airborne Particulate Matter Concentrations in California Using Seven Year Analysis Periods. Atmospheric Chemistry and Physics, 10(22), pp 11097-11114. Climate change does not alter annual average population weighted concentrations to PM2.5 associated with different sources in California.
6) 2009 Chen, J., Ying, Q., and Kleeman, M.J. Source Apportionment of Visual Impairment during the California Regional PM10/PM2.5 Air Quality Study. Atmospheric Environment, 43, 6136-6144. Secondary nitrate from diesel engines accounts for a major fraction of visibility reduction during winter stagnation events in the SJV.
7) 2009 Chen, J., Ying, Q., and Kleeman, M.J. Source Apportionment of Wintertime Secondary Organic Aerosol during the California Regional PM10/PM25 Air Quality Study. Atmospheric Environment, 44, 1331-1340. Solvent use is a major source of winter SOA production in the SJV.
8) 2009 Ying, Q., Kleeman, M.J. Regional Contributions to Airborne Particulate Matter in Central California during a Severe Pollution Episode. Atmospheric Environment, 43, 1218-1228. Two thirds of the PM2.5 nitrate formed during a winter stagnation event in the SJV originates as NOx emissions in the SJV. Upwind sources do not dominated winter nitrate formation during a severe winter stagnation event.

and references to 15 additional papers using the source-oriented model are provided below:

9) 2009 Ying, Q. Lu J., Kaduwela, A. and Kleeman, M.J. Modeling Air Quality during the California Regional PM10/PM2.5 Air Quality Study (CPRAQS) using the UCD/CIT Source Oriented Air Quality Model – Part III. Regional Source Apportionment of Secondary and Total Airborne PM2.5 and PM0.1. Atmospheric Environment, 43, pp419-430.

10) 2008 Ying, Q., J. Lu, P. Allen, P. Livingstone, A. Kaduwela, and M.J. Kleeman. Modeling Air Qualitying during the California Regional PM10/PM2.5 Air Quality Study Using the UCD/CIT Source-Oriented Air Quality Model – Part I. Base Case Model Results. Atmospheric Environment, 42, pp 8954-8966.
11) 2008 Ying, Q., J. Lu, A. Kaduwela, and M.J. Kleeman. Modeling Air Qualifying during the California Regional PM10/PM2.5 Air Quality Study Using the UCD/CIT Source-Oriented Air Quality Model – Part II. Regional Source Apportionment of Primary Airborne Particulate Matter. Atmospheric Environment, 42, pp8967-8978.
12) 2008 Kleeman, M.J. A Preliminary Assessment of the Sensitivity of Air Quality in California to Global Change. Climatic Change, 87 (Suppl 1), pp273-292.
13) 2007 Kleeman, M.J., Q. Ying, J. Lu, M.J. Mysliwiec, R.J. Griffin, J. Chen, and S. Clegg. Source Apportionment of Secondary Organic Aerosol during a Severe Photochemical Smog Episode. Atmospheric Environment, Vol 41, pp576-591.
14) 2007 Ying, Q., M.P. Fraser, R.J. Griffin, J. Chen, and M.J. Kleeman. Verification of a Source-Oriented Externally Mixed Air Quality Model During a Severe Photochemical Smog Episode. Atmospheric Environment, Vol 41, pp1521-1538.
15) 2006 Ying, Q. and M.J. Kleeman, Source contributions to the regional distribution of secondary particulate matter in California. Atmospheric Environment, 40: 736-752.
16) 2005 Kleeman MJ, Ying Q, Kaduwela A. Control strategies for the reduction of airborne particulate nitrate in California's San Joaquin Valley. Atmospheric Environment 39: 5325-5341.
17) 2005 Held T., Q. Ying, M.J. Kleeman, J.J. Schauer, M.P. Fraser. A comparison of the UCD/CIT air quality model and the CMB source-receptor model for primary airborne particulate matter. Atmospheric Environment. 39: 2281-2297.
18) 2004 Held T, Q. Ying, A. Kaduwela, M.J. Kleeman. Modeling particulate matter in the San Joaquin Valley with a source-oriented externally mixed three-dimensional photochemical grid model. Atmospheric Environment 38(22): 3689-3711.
19) 2004 Ying Q, M. Mysliwiec, M.J. Kleeman. Source apportionment of visibility impairment using a three-dimensional source-oriented air quality model. Environmental Science and Technology 38(4) 1089-1101.
20) 2003 Ying Q, M.J. Kleeman. Effects of aerosol UV extinction on the formation of ozone and secondary particulate matter. Atmospheric Environment 37 (36): 5047-5068.
21) 2003 Aw J, M.J. Kleeman. Evaluating the first-order effect of intraannual temperature variability on urban air pollution. Journal of Geophysical Research – Atmospheres 108 (D12) Art. No. 4365.
22) 2003 Foresman E.L., M.J. Kleeman, T.P. Kear, D.N. Niemeier. PM10 conformity determination using the equivalent emissions method. Transportation Research Part D – Transport and the Environment. 8: 97-112.
23) 2001 Kleeman M.J., G.R. Cass. A 3D Eulerian source-oriented model for an externally mixed aerosol. Environmental Science and Technology, 35: 4834-4848.

21ff: the description of the study of Kleeman et al. is difficult to understand. What do you mean by “Na+ and SO42that exist independently the real world environment”? Further I suggest using “particle” (instead of “one aerosol”) if you refer to one single physical entity.

Response: The referenced study showed that sulfate ($SO4^{=}$) does not preferentially condense onto sea salt particles containing sodium (Na+) in Los Angeles. Internal mixture representations of the system artificially combine sulfate and sodium into the same particles. This skews the calculated hygroscopic behavior of the internally mixed particles.

p 16462

1: reason for bimodal plume development?

Response: The internal mixture assumption uniformly distributes the hygroscopic secondary components (nitrate, sulfate, ammonium, etc.) over all particles. In reality, hydrophobic particle cores do not readily accumulate these hygroscopic secondary components. High humidity events therefore activate a subset of the hygroscopic particles into droplets while leaving the hydrophobic particles relatively unchanged. This results in a bimodal distribution.

16ff: this sentence needs cleanup UV and visible are different things, and the way you write it here throws them in the same category. Also, with the shellcore difference you refer to the way you do your Mie calculations, I guess. Then you should say what the core is considered to be made out of in the case SOA > 5%. But in the end I would not go into detail here and just state that optical properties change at 5% mass coating or similar...

Response: Sentence has been revised to clearly describe shell and core assumptions for UV and visible wavelengths as stated in the referenced paper. The sentences here only describe previously work, not aiming to introduce the way we do in the Mie calculation as discussed in Section 3.5.

21ff: The description of CRPAQS study is not part of “2 Background” in my point of view. I suggest merging it with the beginning of Section 4.

Response: CRPAQS background information has been merged with Section 4 as requested.

24: Is there something citeable for CRPAQS? Overview paper?

Response: References were added.

28: “concentrations were measured continuously”, not “continuous concentrations” were measured.

Response: The manuscript was modified accordingly.

22: United States

Response: The manuscript was modified accordingly.

p 16463

1: exact dates of these IOPs?

Response: Dates were added.

7: replace “This” with “The.

Response: The manuscript was modified accordingly.

Section 3 in general:

*it is of low scientific interest how the code of WRF(chem) is structured internally. However, we applaud the effort of the authors to try to clearly describe their developments. I suggest that everything that deals with which routines were changed, what changes to the Registry were necessary, and how the array containing the aerosol properties looks like should be moved into the Appendix.

Response: Details about the model development were moved to appendix. The revised text in the manuscript focuses on scientific results.

* you need to make clear at the beginning of this section that you take parts of your previous sourceoriented aerosol model developments and hook them up with the WRFchem model. I had a hard time understanding that you do not try to present a from scratch development here.

Response: Clarification was added.

15: Appendix number/letter missing

Response: The manuscript was modified accordingly.

16ff: remove source code description, but add information about number and range of size bins.

Response: The manuscript was modified accordingly.

State that you indeed do a bin approach, not a modal approach. Do you do wet deposition? Or only dry? How, which scheme?

Response: The information was added. We do have dry deposition. The scheme used is listed in Table S1. The CRPAQS episode did not include a rain event and so wet deposition has not yet been added to the model, but this can easily be done in future applications.

p 16464

7-22: this paragraph should be moved into the appendix.

Response: The manuscript was modified accordingly.

24: the file format is scientifically not important. If you want to keep it, make sure to use proper capitalization (NetCDF) and add reference.

Response: The manuscript was modified accordingly.

25: multiple inventories?

Response: Inventories for different sources. The manuscript was modified.

26: What is UCD? This “UCD emissions preprocessing system” has to be cited, or explained.

Response: The manuscript was modified. The acronym stands for “University of California at Davis (UCD)”.

p 16465

1: Not of scientific interest, move to Appendix.

Response: The manuscript was modified accordingly.

8: Please state briefly if they are resolved in time or in the vertical. Probably only minor influence, but which source category are the aerosol species attributed to?

Response: Initial conditions are only for the start point, they are not resolved in time, but they are needed for the whole domain including every vertical layer. Both initial conditions and boundary conditions are grouped to the unresolved “other” sources. The manuscript was modified accordingly.

14ff: Explain clearly which part APDC has, and which ISORROPIA, as they are overlapping. Why do you need both?

Response: APDC does the gas-particle conversion calculation (how much conversed at each time step) while ISORROPIA only calculates the vapor pressures of gases above the particle surface.

p 16466

1ff: see J. Fast’s comment: can particles of different source origin coagulate? You say “the source origin of larger particles is preserved” does this hold true also for particles of very similar size? So a small “diesel” particle which collides once with a slightly larger “agriculture” particle becomes an agriculture particle, and stays like that, even if all subsequent collisions (of smaller particles) are with “diesel”? Or if it then grows by condensation of (anthropogenic) NOx?

Response: J. Fast’s comment was addressed and manuscript was modified accordingly. Yes, the smaller particles will lose their source origin. The fastest coagulation rates occur between very small particles with high Brownian velocity and very large particles that provide a large target size for collisions. These types of collisions are well-characterized by the algorithm that preserves the source-origin of the larger particles. Coagulation rates between particles with similar sizes are orders of magnitude slower, and effectively negligible under the conditions studied.

11: remove “6dimensional source oriented array”

Response: The manuscript was modified accordingly.

13: “standards” are “default settings” here which can change over time. State exactly which advection operator is used, whether others could be used as well. The “default” advection operator is not positive definite did you change that? Or did you use the alternative operator?

Response: All advection options work. Positive definite advection scheme was used with 3rd order. The source code was modified to allow the source oriented species to be transported.

13ff: the paragraph heading states “diffusion”, but nothing is said about it. This paragraph should answer the following question: compared to water vapor (QVAPOR) or any other meteorological scalar quantity in the WRF core are these aerosol species subject to the same processes (except for cloud microphysics, of course)? Can we use all possible options?

Response: Yes, all advection and diffusion options are functional.

16: “This operator...” remove what is the purpose of this sentence if nothing follows?

Response: The manuscript was modified accordingly.

19: “enhance vertical mixing” is misleading, talk about adding convective transport of species.

Response: Wording was changed to “more realistically represent the effects of mixing caused by turbulent eddies of multiple sizes”.

20ff: see J. Fast’s comment is this an updated version of ACM2?

Response: We implemented the standard version of ACM2 in the WRF model.

p 16467

end of Section 3: missing information to understand your model setup:

* How do you do photolysis? Do your aerosols affect photolysis rates?

Response: Information of photolysis was added to Section 3.3 and details can be found in Appendix. Photolysis is calculated based on the cosine of the solar zenith angle, grid square latitude and longitude, and the solar declination angle. Photolysis rates are affected by aerosols.

* What is the meteorological forcing data, and do you do nudging?

Response: No nudging was done. Please find following sentence in model application section: “SOWC simulations were conducted without four dimensional data assimilation (FDDA) to minimize artificial forcing so that the effects of chemistry feedback effects on meteorology would be most apparent.”

* Is new particle formation happening (nucleation)? How?

Response: No nucleation was included. Nucleation is not an important process in the highly polluted SJV during a winter stagnation event.

* Do you do secondary organic aerosols? How?

Response: SOA formation was not included in the present application since in this winter episode, the concentrations are very low compared to primary organic aerosol and secondary nitrate. The offline versions of the source-oriented air quality model have implemented numerous SOA schemes over the past decade (see some of the references above) and any of these schemes could be adopted in the future in the SOWC model for simulation of episodes where SOA is an important species.

* Do you have sea salt and dust? How do you do it?

Response: Dust was processed with the UCD emission processor based on the inventory from CARB. Sea salt was included using an on-line emissions subroutine that is based on correlations between emissions and surface wind speed.

* a table of major options chosen for selected WRF/chem processes (PBL scheme, Land model, Radiation scheme, Cloud microphysics, Cumulus param., Meteorological forcing data, Photolysis scheme...) would be really helpful

Response: Table 1 was added with the configuration information.

* Can one choose to use a different option, and will your scheme still work? E.g. different PBL scheme, different advection operator...

Response: Different PBL scheme works and all advection operators under the EM_CORE also work.

21: Just to make sure: you do mean 100 hPa here?

Response: The typo was corrected.

p 16468

11: explain how you “conceal” the feature is it still your aerosol module and you only emit into one category?

Response: The user can define how many source types should be tracked. If the number of source types is specified as 1 then all the emissions from different sources are grouped into an internal mixture. This effectively conceals or disables the source-oriented mixing features of the model. Further explanation was added to the manuscript to clarify this point.

Before Section 5: It would be helpful to have a short summary here of the type, number and location of the measurements you will use.

Response: A summary was added as the first paragraph in Section 5.1.

Section 5 in general: do not only report the maximum value you find in a single grid cell, but also average values or some kind of statistics.

Response: The averages over 4km domains with standard derivations were calculated and used together with the maximum values.

19: “by _the_ external SOWC”

Response: The typo was corrected.

25: give equation for MFB

Response: Equations of MFB, RMSE, and MAE were given with the tables.

26: show location of these sites on map, and connect position with the abbreviations you use

Response: Locations of the sites were added to Figure 1.

p 16469

2: give a reason for differences in primary components

Response: The meteorological variables such as wind speed and PBL height were influenced by the mixing state of the particles. This leads to changes in the concentrations of primary PM component concentrations. This section only shows the model performance – the full explanation of results is given in Section 5.4.

2-5: I do not really follow this argument CO, NO and NO2 are reactive gasphase compounds, while EC and POC are inert aerosols explain, rephrase or remove.

Response: The sentence was rephrased.

5: State clearly (maybe even in section 4) how ozone can be affected by the aerosol mixing state in your simulations.

Response: The manuscript was modified to be clear about why ozone concentrations were affected by a small amount. Surface O3 in the SJV in winter time is mostly associated with vertical mixing from upper layers to the surface to the surface during the day. This finding has been documented in several previous studies. When the meteorological conditions (for example, PBL) are changed, the transport of O3 to the surface is changed.

7-10: you are completely ignoring local O3 destruction through titration. Why? Not convincing.

Response: The chemical mechanism does include a description of titration by NOx. This is actually the dominant path for nitrate formation, and it explains why ozone concentrations decrease during the nighttime hours. The text has been modified to make this point clear.

Section 5.2 first paragraph: I do not understand Figure 4 (see below), so I cannot evaluate this paragraph.

Response: The description of Figure 4 (Figure 2 in new version) was clarified.

21: Make clear that you will compare Figures 5 and 6 in the following.

Response: Clarification was added.

21-28: please add an explanation to why we see things, e.g. 3 modes, and do not only describe what we see. What about SOA?

Response: Figure 6 (Figure 4 in the revised manuscript) provides an explanation for the 3 modes. The smallest mode is dominated by particles from diesel engines, wood burning, and food cooking. The intermediate mode is dominated by nitrate formation on particles from high sulfur fuel combustion and "other sources". The largest mode is dominated by dust sources (encoded in the "other sources category"). Past studies have shown that predicted SOA concentrations are relatively small compared to POA and nitrate in the current episode.

p 16470

20: First you say that other people found no influence on the NH3+HNO3 product, and then you find something new. Hence the sentence "The current study" needs to start with a "However", "But" or some word like that to make clear this is a new finding.

Response: The sentence was modified.

p 16471

10: what's the domain/land/California average?

Response: Averages for land and the SJV covered by 4km domain with standard deviation were added to each plot and were reported in corresponding positions in the manuscript. The focus was on the SJV and California was not fully covered, so the California average was not shown.

11: misleading sentence: it is not the "first" variable, as aerosols are aloft, and the surface is below, so the radiation budget at the location of the aerosols will probably be the "first" variable. Rephrase.

Response: The sentence was rephrased.

15-16: again: how does it look in other regions? Are these averages?

Response: The focus of this study is the SJV. The value here is the maximum. Averaged over land within 4km domain was calculated and added.

17-18: Here would be a nice spot to actually show how mixing changes (> PBLH, TKE...).

Response: Change in PBL height was added.

20: Please do not evaluate wind components, but speed and direction.

Response: Wind components instead of wind speed and direction were shown because this simplified the comparison to available observations. This point is a question of style, and the authors respectfully prefer to present the results in the indicated format.

25: again: averages? distribution?

Response: Averaged values over land and the SJV within 4km domain were calculated and presented in the manuscript.

p 16472

4: is 10% an average? Really? If not, add average and distribution

Response: Averaged over land within 4km domain was calculated and added.

6: what "implications"? Or is this supposed to introduce the next section. Then add sth. like "as we will show in the next section".

Response: A sub-summary was added to the end of Section 5.3. And the sentence was rephrased accordingly.

9: You did not show this in your work. More complex is not necessarily better. To draw this conclusion I would want to see a comparison with a lab study.

Response: "More realistic" is definitely true for this study. Furthermore, we included a comparison of particle scattering coefficients with observation and it does indeed show that the source-oriented external representation is more accurate than the internal representation.

p 16473

3-4: can you explain these differences?

Response: These differences are consequences of differences in meteorological conditions such as PBL and wind, and differences in dry deposition due to changes of absorption, partitioning and coagulation resulting from the external representation and internal representation. Explanation was added in the manuscript.

18-21:

I do not think this is very apparent it takes time to form HNO3 and then to condense it on particles. That can happen downwind of urban centers.

Response: The reviewer may misunderstand this sentence. It says "**fresh** wood smoke particles emitted in urban centers …do not account for". It does not conflict with the statement that aged woodsmoke particles can accumulate a significant fraction of nitrate.

23: So how come? Can you explain this? Results like this might point to a technical issue (numerical instability, coding error...).

Response: In this study, the sources types tracked are based on the characterization of particles. The "other sources" are a lump group of all sources that are not tracked by one of the explicit source types. The "other sources" category has more available particle surface area acting as nitrate condensation cites. This is a consequence of the emissions and the choice of explicitly tracked sources. It has nothing to do with technical issue like numerical instability or coding error.

p 16474

1-3: so its only the radiation, not the mixing?

Response: Redistribution of solar energy or radiation is the start point, mixing and other meteorological parameters are subsequent. We are sorry about the confusion. The sentence was modified to clarify this issue.

15-16: how do you know this?

Response: The main sources in emission inventory are the air force bases.

21: this is quite a large number! Probably a singular event in a single grid cell (again)?

Response: The stated concentration is a maximum, but the concentrations gradients are not sharp. Food cooking particles are a major contributor to PM2.5 concentrations during the current study.

23ff: How do you explain PM differences over the ocean (Fig 12f) with predominant westerly flow patterns?

Response: First, the peak color represents very small concentration differences over the ocean. Furthermore, the stagnation event is characterized by a high pressure system situated over central California with clockwise rotation of winds. Thus, changes over California can be re-circulated over the ocean towards the western boundary of the modeling domain.

p 16475

1-10: this paragraph sounds like it is conclusions. Could be moved there.

Response: This is a sub-conclusion of Section 5.

7: again, you did not show that they are more accurate.

Response: The sentence was modified to reflect the results of a comparison between source-oriented and internally mixed optical properties relative to measurements.

Section 6 in general: quantitative results would be great ("SWdown change by avg … Wm2, PM2.5 increased by .. ug/m3 on avg. ...).

Response: Quantitative results are shown in abstract. Here only shows a general conclusion.

12: remove “The”

Response: “The” was removed.

Figures

Figures 1 & 2: move to Appendix together with technical description

Response: The manuscript was modified accordingly.

Figure 3: Black text on blue background is not readable, change. Round numbers for legend, and make sure they don’t overlap. Indicate measurement sites instead of major cities. State that the coloring is elevation. Regrid to latlon, or plot lat/lon lines. Caption: “covers _the_ Central Valley”.

Response: The figure was modified accordingly. The tics are not changed to lat/lon because we want to show the distance more clearly. The simulation region is small, lat/lon is not necessary to show.

Figure 4: I could not understand this figure. What are the different particle compositions in the external mixing case? Can’t you show the mass contributions of each source type? What does the size of a pie mean? I suggest to use a commonly used color scheme, e.g. like AMS people do (sulfate is red, ammonium orange, nitrate blue, organics green, BC black). “noon” means local time? Is this a snapshot or an average? In its current form this figure is very bad start for reading Section 5!

Response: More description was added to the caption to explain the Figure: “Each of the pie charts illustrates the composition of a single particle from one of the independent particle groups tracked. The size of each pie chart is proportional to the log of the actual particle diameter. The pie charts in this figure are ordered so that the particles with the highest number concentration of each size bin are at the top”. “noon” means average between 12:00 pm to 13:00 pm local time. It was modified to avoid confusion. The mass contributions are shown in Figures 5 and 6 (new Figures 3 and 4). The AMS color scheme is not a universally accepted standard and is not relevant. We did not change the color scheme because our legend in the Figure clearly shows what color corresponds to each species.

Figure 5: Why are these figures black and white, while the others are in color? Use the same colors. Why is there almost no SOA? This is not realistic! How come there are several bins with exactly the same amounts? Is this again at noon on the 24? Instantaneous?

Response: Color is reserved for figures where it clearly makes the results more readable. Black and white is preferred when possible. This is a question of style and we prefer to present results in the indicated format.

Offline model calculations show that predicted SOA concentrations are very small compared to POA and nitrate in the winter time due to low oxidant concentrations. Thus, SOA formation was not enabled in the present study. The legend of SOA was removed.

The plot shows a set of default size bins that the model sizes were mapped to for presentation purposes. When the model size bins span more than one of the standard plotting size bins, then the concentrations appear identical. Again, this is a style feature (admittedly not optimal) but it does not impair the readers understanding of the results.

Figure 6: see Figure 5 comments. Also, state that the y axes have different scales.

Response: The figure was modified accordingly.

Figure 7: again, black on dark blue is not readable. Maybe plot histogram of changes (and maybe plot these in % original value)?

Response: The figure was modified accordingly. The averages over land and the SJV covered by 4k domain were added.

Figure 8: see 7, and also: how do these single negative points happen? This looks fishy.

Response: These negative points are due to changes in cloud fraction within one grid cell, thus, the changes are not continuous.

Figure 9: see 7, and also: plot wind speed instead of components. You do not assess changes in wind direction, hence speed is sufficient and much more easy to read.

Response: The figure was modified accordingly. But, as discussed previously, wind components are plotted as a matter of author preference.

Figure 10, 11, 12: see 7.

Response: The figures were modified accordingly.

References:

Zhang, R., Khalizov, A.F., Pagels, J., Zhang, D., Xue, H., McMurry, P.H., 2008. Variability in morphology, hygroscopicity, and optical properties of soot aerosols during atmospheric processing. Proceedings of the National Academy of Sciences of the United States of America 105, 10291-10296.