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.

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.

* 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 WRF-chem (MADE, MOSAIC) would be beneficial.

* 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.

* 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.

* 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.

* 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.

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

Detailed remarks

p 16458

5-6: avoid repeating “The source-oriented approach”.
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”).
16-21: This description of your model setup is too detailed for an abstract. I suggest removing.
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.
26-28: rephrase: “Effects of a source-oriented representation of particles with meteorological feedbacks in WRF-chem on a number of meteorological parameters were identified: downward...”

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”)
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?
8: you do not show that mixing changes. Either remove, or show (see general remark).
14: why only nitrate? what about sulfate? SOA?
20: please add Fast et al., 2006 and Peckham et al., 2011 to the references for WRF-chem (see also here: http://ruc.noaa.gov/wrf/WG11/References/WRF-Chem.references.htm)
24: Do not repeat “stagnation events”.
26: make clear that this sentence is only true during stagnation events.

p 16460

1: again the atmospheric mixing argument - please add citation.
6: there are several aerosol modules implemented into WRF-chem (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.
6: see J. Fast comments about other work on explicit representation of mixing state. (I.e. cite Matsui et al., 2012)
10: time scales need references.
17-23: COSMO-ART (Vogel et al., ACP, 2009, http://www.atmos-chem-phys.net/9/8661/2009/acp-9-8661-2009.html) is comparable to WRF-chem and considers explicit aging of soot (Riemer et al., JGR, 2003, http://onlinelibrary.wiley.com/doi/10.1029/2003JD003448/abstract). They evaluate radiative effects.

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?
8: start a new paragraph after “hours to days”. Introduce it with something like “In this work”, “This paper is structured as follows”, ...
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.
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”.
21ff: the description of the study of Kleeman et al. is difficult to understand. What do you mean by “Na+ and SO42- that exist independently the real world environment”? Further I suggest using “particle” (instead of “one aerosol”) if you refer to one single physical entity.

p 16462

1: reason for bimodal plume development?
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 shell-core 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...
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.
24: Is there something citeable for CRPAQS? Overview paper?
28: “concentrations were measured continuously”, not “continuous concentrations” were measured.
22: United States

p 16463

1: exact dates of these IOPs?
7: replace “This” with “The.

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.

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

15: Appendix number/letter missing
16ff: remove source code description, but add information about number and range of size bins. State that you indeed do a bin-approach, not a modal approach. Do you do wet deposition? Or only dry? How, which scheme?

p 16464

7-22: this paragraph should be moved into the appendix.
24: the file format is scientifically not important. If you want to keep it, make sure to use proper capitalization (NetCDF) and add reference.

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

p 16465

1: Not of scientific interest, move to Appendix.
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?
14ff: Explain clearly which part APDC has, and which ISORROPIA, as they are overlapping. Why do you need both?

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?
11: remove “6-dimensional source oriented array”
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?
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?
16: “This operator...” remove - what is the purpose of this sentence if nothing follows?
19: “enhance vertical mixing” is misleading, talk about adding convective transport of species.
20ff: see J. Fast’s comment - is this an updated version of ACM2?

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end of Section 3: missing information to understand your model setup:
* How do you do photolysis? Do your aerosols affect photolysis rates?
* What is the meteorological forcing data, and do you do nudging?
* Is new particle formation happening (nucleation)? How?
* Do you do secondary organic aerosols? How?
* Do you have sea salt and dust? How do you do it?
* 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
* Can one choose to use a different option, and will your scheme still work? E.g. different PBL scheme, different advection operator...

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

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

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.

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.

19: “by _the_ external SOWC”
25: give equation for MFB
26: show location of these sites on map, and connect position with the abbreviations you use

p 16469

2: give a reason for differences in primary components
2-5: I do not really follow this argument - CO, NO and NO2 are reactive gas-phase compounds, while EC and POC are inert aerosols - explain, rephrase or remove.
5: State clearly (maybe even in section 4) how ozone can be affected by the aerosol mixing state in your simulations.
7-10: you are completely ignoring local O3 destruction through titration. Why? Not convincing.

Section 5.2 first paragraph: I do not understand Figure 4 (see below), so I cannot evaluate this paragraph.
21: Make clear that you will compare Figures 5 and 6 in the following.
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?

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.

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10: what’s the domain/land/California average?
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.
15-16: again: how does it look in other regions? Are these averages?
17-18: Here would be a nice spot to actually show how mixing changes (-> PBLH, TKE...).
20: Please do not evaluate wind components, but speed and direction.
25: again: averages? distribution?

p 16472

4: is 10% an average? Really? If not, add average and distribution
6: what “implications”? Or is this supposed to introduce the next section. Then add sth. like “as we will show in the next section”.

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.

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3-4: can you explain these differences?
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.
23: So how come? Can you explain this? Results like this might point to a technical issue (numerical instability, coding error...).

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1-3: so its only the radiation, not the mixing?
15-16: how do you know this?
21: this is quite a large number! Probably a singular event in a single grid cell (again)?
23ff: How do you explain PM differences over the ocean (Fig 12f) with predominant westerly flow patterns?

p 16475

1-10: this paragraph sounds like it is conclusions. Could be moved there.
7: again, you did not show that they are more accurate.

Section 6 in general: quantitative results would be great (“SWdown change by avg … Wm-2, PM2.5 increased by .. ug/m3 on avg. ...).

12: remove “The”

Figures

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

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 lat-lon, or plot lat/lon lines. Caption: "covers _the_ Central Valley".

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!

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

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

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

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.

Figure 10, 11, 12: see 7.