Review of “Simulation of trace gases and aerosols over the Indian domain: evaluation of the WRF-Chem model” by Michael et al.

First of all I would like to make the following general remark: I have been asked to review the manuscript of Michael et al. for publication in Atmospheric Chemistry and Physics (ACP). I have not been involved in the access review. I would like to state that from my point of view this paper should not have been accepted for ACP. The manuscript is well written and the methods are sound, but it is a pure model evaluation effort without much scientific discovery apart from the fact that an existing model system sometimes agrees with measurements when applied to a different domain. This is not even the first time WRF-chem has been applied over India. The sister journal of Atmospheric Chemistry and Physics, Geoscientific Model Development, would have been a much better fit for this publication - like it has been used for the very similar publications by Kumar et al. (2012 a/b) referenced in this work.

Summary

Michael et al. present simulation results using the regional-scale chemistry model WRF-chem over the Indian subcontinent for three pre-monsoon periods. They compare simulations against surface measurements of trace gases and aerosol optical properties. Additionally, vertical profiles of several observation systems were used. They show reasonable agreement between model and measurements and indicate that an insufficient description of emissions and transport of dust emissions is responsible for a large part of the discrepancy. Performance to simulate black carbon is reasonable, and the authors suggest that previous findings in Nair et al., 2012, where large differences have been reported, can actually be remedied by using a different model system and a more regional emission inventory.
The paper is well written, methods and analyses are presented in an understandable manner, and conclusions are mostly reasonable. There are quite a number of rough edges that have to be removed before publication though, hence I suggest publish after major revisions.
I would like to see the revised manuscript before publication.

General comments

* The authors should replace or complement all instance of “compares well”, “very well”, “acceptable” and other non-scientific assessments of the level of agreement between model and measurements with quantitative assessments (e.g. “-14 ppbv MB”, “underestimated by 20% on average”). I have made the same mistake in the past and can assure you that this doesn’t help you to get reviewers in a favorable mood, nor does it help with understanding model performance.

* More clearly explain why your work is novel and different (or complementary) to Nair et al., 2012 and Kumar et al., 2012 ab. You are not the first one making a regional chemistry transport simulation over India. You were not the first simulating aerosols there. But you might be the first to present a comparison of AODs.

* AERONET data provides measurements at several wavelengths, hence allows to calculate Angstrom exponents. The same is true for WRF-Chem, which gives AOD at several wavelengths as well. Including this information in your analysis would provide valuable insights into the size distribution of aerosols without a lot of additional effort.

* Analyses in which the authors calculate correlation coefficients need to be redone considering autocorrelation in the data and applying appropriate decorrelation methods before.

* Inaccurate description of wet deposition is given as reason for mismatches compared to measurements, but it is not stated which schemes (gases and aerosols, resolved and parameterized precipitation) are used. Neither is it stated if and what aqueous-phase chemistry is used. This needs to be added in the model description section.

* Plotting multiple years in one figure makes it illegible, please show these in separate panels - and maybe consider moving some less interesting stations to the Appendix.

* Please add a figure showing the model domain and the location of the observations you use.

Detailed comments

Throughout the manuscript

* “overpredict” and “underpredict” is a single word, not two words.
* Do not start a sentence with “I. e.”, rephrase e.g. with “This means that” or similar

12288

5-9: how you set up your model is not of interest in the abstract, remove these technical descriptions. If you think that the use of the regional emission inventory is a substantial part for the agreements you find, than rephrase this sentence accordingly.
11: For readers not from India - explain what and where Kanpur is.
16: .. at _the_ Kanpur Micropulse ...
16: “in agreement” - replace with quantitative statement
16-21: add quantitative evalution (MB, R^2…)
19-20: remove “The” from “The vertical profiles”
21: from _an_ aircraft campaign
21: state which aircraft campaign this has been

22-24: this only holds true for one configuration of WRF-chem during a particular season compared against a certain set of observations. This statement is too strong and general. Rephrase.

12289

15: remove “the” from “from the Asian countries”
16: state why local emissions can contribute to global aerosol load, given that their lifetime is only several days. Is this only due to the large contribution to total aerosol mass, or also spatially true?

12290

10: _the_ Weather Research and Forecasting …
13: using _the_ Regional Climate
13: need citation for RegCM4

12291

1-5: Kumar et al. actually did include aerosols in their simulation, but they did not compare them against observations.
5: you do mean “satellite” retrieval errors, right. Please add that.
7-8: please name these emission inventories.
10: “for 2006 satisfactorily” (no comma)
15: So Chung et al., 2010 already simulated aerosols over India on the regional scale?
20: they are not “undetected”, but rather “unresolved”.
27: a map of observation locations would be very helpful
28: period of _the_ TIGERZ

12292

4: explain what a CIMEL is and cite appropriately
4-8: it doesn’t become clear what that 50x50 km area should be - they have 3 AOD sites in there and several CIMELs? How can you say that this is a square of 50 km length?
6-7: you cannot “accomplish” temporal variation. Rephrase, e.g. using “captured” or “observed”.
8: it should be “complement”, not “compliment”

12293

1-14: I suggest adding the references to Table 1 and removing everything after “in this study are summarized in Table 1”. This is duplicate information.
24-25: this sentence doesn’t make sense - is TIGERZ a NASA AERONET intensive campaign? Reads like garbled text.
27: Explain how you do 2 months out of simulations of 15 days. Do you initialize chemistry /

aerosol fields with the result from the previous simulation and only re-initialize meteorology based on FNL? Also: are these 15 days free running (i.e. only forced at the boundaries) or do you do nudging?
If you do not nudge: this is too long a period, and will lead to considerable errors in meteorology!

12294

17: which grid mapping program? If this is not something you developed for this work, its not of importance and this sentence can be removed. If you created it - more information, please.

general questions regarding emissions:
* where do you get your VOC speciation from?
* do you have point sources, and if so do you do a plume rise?
* fire emissions? biogenic VOCs?
* do your emissions have a diurnal / weekly / seasonal cycle?

12295

1-7: nice to see that you use an independent dataset for validation. Could you also show the changes (improvements, hopefully) in meteorology between FNL and WRF?
8-16: Mentioning the models of the different instruments is a great thing, but please also add the manufacturer.
14: Note that your concentrations of SO2 (later section) is close / below the detection limit of the instrument.
15-16: Detection limit and response time for CO instrument?
23-next page: make the list of AERONET stations a table (including the references) and remove this paragraph. Also: show their locations on the overview map suggested above.

12296

14: manufacturer of aethalometer?
22: “2012). Vertical profiles” (no “The”)

12297

6: remove the comma after “defined as,”
15: quantify the agreement, this sentence doesn’t mean much.
15-16: rephrase: “Though the model can simulate parameters averaged over a large area and long timeframe”
18-21: rephrase, e.g. “Though monthly and seasonal averages can provide a general assessment of the meteorological situation in general, simulations at higher spatial and temporal resolution are required to represent small-scale meteorological phenomena.”

12298

5.1 in general

* in the last section you discuss the importance of high temporal and spatial resolution, now you start with 2 month averages. This doesn't read well. I think you can start with it, but should add a connecting sentence stating that you will go into more detail later.
* the description must become more quantitative. "well", "very well" and so on is of no scientific value!
* the comparisons in Figure 2 are statistically problematic: there is evident autocorrelation in the data which should be removed before calculating correlation coefficients. Unfortunately the authors are vague about how they averaged the data before comparison. Methods to decorrelate data can be found in statistical textbooks.
* please consider showing values from all three models (WRF-chem, FNL and ECMWF IFS) in each of the scatterplots in Figure 2 and calculate statistical measure for all three to show differences. This would also mean you could remove Figure S1.

5-6: "parameters show a good comparison over land". I disagree. This is badly worded ("compare well over land" would be better), its not quantitative ("compare with a mean bias of… over land" or similar), and finally it is not accurate - there are considerable discrepancies in RH and wind patterns also over land!
20: how and what exactly did you average?
21-22: I would like to see a map of the location of these stations. Or even a set of maps showing the time-averaged bias for the different parameters at the different locations to identify spatial patterns.
25: these correlation coefficients are overestimated because you have autocorrelation in the data!

12299

5: "about 151 stations"? It should be exactly "151 stations", no? Why "about"?

5.2 in general

* explain what your error bars mean - 1 sigma? 5-95%? quartiles?

16: rephrase: "During the pre-monsoon period of 2010 ozone, carbon monoxide and sulfur dioxide were observed hourly ..."
18: ". Long-term measurements" (remove The)
20: consider making subsections (5.2.1, 5.2.2, 5.2.3) for O3, CO and SO2

12300

11-14: need to state which schemes are used. Note that you need to provide information on gas as well as aerosol scavenging parameterizations for both resolved and parameterized

(convective) precipitation. Finally, you need to discuss aqueous-phase chemistry used (but maybe better in the model description section, not here).
15 “Ozone concentrations have a …”

12301

4: this looks very odd - explain where all your variability in CO after June 21.
7-9: you have no proof for this hypothesis. Either put forward more evidence or remove sentence.
14: note that 1.76 is below the detection limit of the SO2 instrument.

SO2 in general: explain why it is difficult to model SO2

21 - next page 3: This paragraph needs to be completely reworked, or even omitted. How does wet scavenging affect OH? O3 is about 2 orders of magnitude less soluble than SO2, and CO is an order of magnitude less soluble than O3. Why would lacking aqueous-phase chemistry overestimate O3? All three, O3, CO, and SO2 are reduced by wet scavenging. However, the chemistry there is non-linear, so the outcome is difficult to assess, and this is clearly not shown in a brief paragraph like this one. I would want to see a box model study to believe that.
Further down: why is it a “low rate of conversion”? Low compared to what? How do you even do this in the model? Why would this lead to overestimation of SO2?
In summary - please make this paragraph much more clear or remove.

12302

6-7: not true and apparently only there to suggest you did novel work. Unfortunately this is not the novel part of your work: all simulations of WRF-chem that consider aerosols calculate AOD (in various ways), and they do so necessarily at each call of the radiation scheme. You’re just one of the first to present comparison against AOD measurements. Remove.
8-9: “integrating it over altitude”
9-11: again, a map of station locations would be helpful.

AOD comparison in general:

Figures need to be split into different panels, one for each year. Overplotting several years makes them illegible.

24-25: “Though, most of the days, … within error bars”. This is not supported by the figure as far as I can see it. Overstated. Rephrase.

12303

5: citation for HYSPLIT needed.

9: Just because Bian et al., 2011 showed that transport of dust is not done well doesn't necessarily mean by inference that local dust emissions are well represented. Remove/rephrase/support with evidence.
13: _the_ Thar desert
14: do not repeat Jaipur (use "It")
27: relatively clean (not cleaner)

12304

1: there are only scarce observations for this period, be cautious.

12305

26-27: no, they do not compare very well. They don't compare "well" at all. Plot a difference of AOD(meas) - AOD(mod) and look at the time series - from what one can see in Figure 12 they differ often by a factor of 2 or more!

12306

6-8: You do not have data for April, and there is no trend in the simulated extinction coefficients shown. This claim is unsupported and should be removed. (Or properly justified)

12307

5: does you emission inventory have a diurnal cycle in BC emissions?
8ff: be more cautious with your description as you are showing a log-scaled BC concentration here.
"forenoon" is old-fashioned english - maybe replace with "morning"?
21: "are in agreement" - quantify.

12308

6-9: Description of when and how the monsoon happens should go into the introduction.
18ff: quantify results.
26: no, this is not really shown, just inferred. Too strong a conclusion, rephrase e.g. to "it is hypothesized".

12309

3: "very well" -> quantify
11: "WRF-chem" (capitalized WRF)
11: which various atmospheric processes? You only showed this for one configuration, and you only evaluated concentrations - you could very well be right for the wrong reasons. Just because concentrations agree with measurements doesn't mean the underlying processes are

understood. Rephrase.

13-17: remove. Purely speculative and not scope of this work.

Figure 1

x/y axis labels missing
units of temperature? length of wind barbs meaning?
did you correct U/V to be in geographical coordinates before plotting wind barbs?

Figure 2

decorrelate
add data for ECMWF and FNL
add overall titles on top (“Temperature”, “RH”) and the right hand side (“IGP”, “Central India”, “Coastal India”)
strange scaling of RH in Coastal India - how come?

Suggestion: make 1 multi panel figure out of Figures 3, 5 and 6

Figure 3, 4, 5, 6

Color coding would be nice to see

Figure 4

is this local time - if so, state that in the x-axis label (e.g. “local time (UTC+X)”)

Figures 7 to 12

do not overplot several years / stations. Please make 1 panel for each year.
state that this is AOD at 550nm

Figure 13

lots of error bars, maybe plot envelope lines instead?

Figure 15

local time? adapt x-axis label, see comment for Figure 4.

Figure 16

separate panels for different years would be helpful.