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Discussions

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Interactive comment on "Simulating aerosol–radiation–cloud feedbacks on meteorology and air quality over eastern China under severe haze conditions in winter" *by* B. Zhang et al.

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We thank the reviewer and the editor for their thoughtful and constructive comments to improve the analysis of the manuscript. The revisions/additions/edits are highlighted in BLUE in the revised manuscript. All the page/line/figure numbers in the response file refer to those in the revised manuscript with the changes noted.

Anonymous Referee #2 (Comments to Author): My overall rating of this paper is somewhere between minor and major revisions; I've marked it as major since I would like

a second look at the paper once the revisions are complete, not an option if they are marked as minor. The authors need to describe the potential impact of grid resolution vis-à-vis the cloud formation setup in their simulations, and (the most important point), they need to provide a solid justification for the absence of secondary organic aerosols in their model simulations and a quantitative discussion of the potential impact of its absence (or, better, repeat the simulationswith a SOA parameterization in place). Aside from those two concerns, there are a few minor issues where the text needs to be clarified, and a large number of cases where spelling or grammar needs to be corrected. The paper is basically sound and will be a useful addition to Atmospheric Chemistry and Physics once these issues are addressed.

Response: Thank you for the constructive comments. First, we have added discussions about the impacts of grid resolution on the indirect effect. Second, we have conducted new simulations using the aerosol mechanism including SOA and discussed the potential impact of that. Third, we have conducted thorough editing and corrected the spelling and grammatical errors.

Main issues: (1) I have concerns with regards to the authors' conclusions regarding the relative importance of the aerosol direct versus indirect effect. In their work, for the time period and region studied, they found that the indirect effect was relatively minor compared to the direct effect. This is contrary to other studies (the authors quoted Forkel's work, and there are others coming out in the Atmospheric Environment special issue on Phase 2 of the Air Quality Model Evaluation International Initiative (AQMEII-2) which the authors may wish to examine). One of the things that has come out of that multi-model comparison (where the indirect effect dominated across multiple models and domains in North America and Europe) is that the magnitude of the indirect effect response is very sensitive to the manner in which it is implemented and on the cloud microphysics parameterizations used. I agree with the authors' suggestion that this may be a special case in that the winter haze events simulated are for relatively low cloud conditions, and their check of the model response with the more sophisticated

Morrison et al microphysics scheme instead of the Lin scheme suggests a general low sensitivity to the microphysics helps in reducing the possibility that the microphysics itself is an issue. However, they should also discuss how the resolution of the model grid relates to the cloud formation in the model, and how this may affect the indirect effect response. That is, the authors are carrying out their simulations with a relatively coarse resolution of 27 km in the horizontal. This in turn requires the use of a cumulus parameterization (the authors make use of the Grell-Devenyi, 2002 scheme), since the cloud microphysics parameterizations are not capable of creating cumulus clouds at that resolution. It is not clear in their section 2.1 exactly how the model generated aerosols are incorporated into either the radiative code (direct effect) or the cloud formation parameterizations (indirect effect). For the direct effect, one needs a means of working out the particle radiative properties (optical depth, single scattering albedo, asymmetry factor) for incorporation into the radiative transfer. What was the means of doing that employed in this work (e.g. a Mie code incorporated into the model, a lookup table? What mixing assumption was used – heterogeneous mixture or core-shell?)? A few words of how this is set up is needed, beyond the references given on page 26089. For the indirect effect – how were the aerosols incorporated as cloud condensation nuclei into the parameterizations used? E.g. what was the means by which speciated aerosols were converted to cloud condensation nuclei numbers in the aerosol microphysics scheme (e.g. Abdul-Razzak and Ghan (2002), or some other scheme?)? Was the model modified to incorporate the effects of aerosols into the cumulus parameterization and if so, how? If not, then the authors should caveat their conclusion regarding the relative importance of the direct and indirect effect by noting that due to the resolution employed, only part of the indirect effect is incorporated, due to the need for a cumulus parameterization (which lacks a feedback connection to the aerosols) and the low resolution ofthe model, which requires the use of a cumulus parameterization.

Response: We have added descriptions of how physics modules are coupled with the aerosol direct and indirect effect in Section 2.1. For the aerosol direct effect, aerosol optical properties such as extinction, single scattering albedo, and asymmetry factor

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are calculated as a function of wavelength and three-dimensional position, and then transferred to Goddard shortwave scheme. Mie theory is used to estimate the extinction efficiency. In this study, refractive indices are calculated based upon a volume-averaging approximation. The first and second indirect effects are implemented in the model (Gustafson et al., 2007; Chapman et al., 2009). Activated aerosols serving as CCN are coupled with cloud microphysics. Activation of aerosols follows the method of Ghan and Easter (2006), which is derived from Abdul-Razzak and Ghan (2002). Through this coupling, aerosols alter cloud droplet number and cloud radiative properties, and aqueous processes and wet scavenging affect aerosols. In the model, aerosols change vertical profiles of meteorological variables by absorbing and scattering solar radiation, and further alter cumulus parameterization.

To address the reviewer's concern on how the grid resolution relates to the indirect effect, we have conducted another three groups of nested grid runs with a 9-km resolution, using the same scenarios (BASE, RAD, and EMP). The outer domain is the same with previous domain, and a 9-km nested domain covering North China Plain (NCP, shown in Fig. 4) is added. In the 9km nested domain, cumulus parameterization is turned off. We find that with the finer resolution simulation, aerosol direct effect still dominates over NCP and aerosol indirect effect is not significant. This is similar to the conclusion we reached using the coarse-resolution simulation, but contrary to several studies under AQMEII Phase 2 as mentioned by the reviewer which suggested that aerosol indirect effect is non-negligible (Kong et al., 2014; Forkel et al., 2014; Makar et al., 2014).

Considering that the above mentioned studies under AQMEII Phase 2 used similar grid resolution (23-km and 27-km) as our study, the grid resolution may not be the main reason why aerosol indirect effect was only minor in our study. There are three reasons to explain our result. First, we focused on a low-cloud-cover winter case, especially in North China. It is confirmed by both the more sophisticated Morrison scheme and the finer-resolution (9 km) nested grid simulation that cloud cover is low in North China.

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Studies under the AQMEII Phase 2 focused on summertime, when aerosol indirect effects on solar radiation, temperature, and PBL height are found to be most pronounced (Forkel et al., 2014). Second, under high aerosol loading conditions (for example, fire conditions in Kong et al. (2014) and haze conditions in our study), aerosol direct effect is found to dominate over the aerosol indirect effect. The aerosol indirect effect is found to dominate over the clean ocean and near ocean land (Forkel et al., 2014; Kong et al., 2014). Third, we did find some regions dominated by aerosol indirect effect. However, the Student's t test filtered the values over those regions, because the changes due to aerosol indirect effect were not statistically significant. We've added more discussions about aerosol indirect effect and did a more comprehensive comparison with studies under the AQMEII Phase 2.

Please refer to the revised Section 2.1 and 4.1 for details.

(2) The authors mention in a single line in the text (page 26089, line 16-17) and once in the conclusions that secondary organic aerosol formation was not included in their model. This choice was not explained or justified in the text. This is a potentially major problem, in that organics often make up the bulk of the PM2.5 mass, especially in urban areas. The authors' PM2.5 comparisons to observations may be therefore be flawed – had secondary organic aerosols been included, they would potentially have much higher PM2.5 values, and a larger impact of aerosols on direct and indirect effect radiative transfer. The authors need to explain why this choice was made: WRF-CHEM comes with secondary organic aerosol formation parameterizations – why was one of these not used in their work? Ideally, they should repeat these runs and analysis with asecondary organic aerosol formation algorithm in place. Failing that, the authors need to provide a justification for the lack of this source of aerosol in their simulations, as well as quantifying the potential impact of its absence on their results, using estimates of speciated PM2.5 for their study area.

Response: We agree with the reviewer. We have conducted another model simulation with the same settings with the BASE scenario, but replacing the MOSAIC aerosol

module with MADE/SORGAM, which contains SOA. The new model results show that SOA is about 0.3 ug/m3, averaged in urban cities, making up only 2.8% of OA and 0.4% of PM2.5. This result is comparable with other modeling studies of SOA in China (Jiang et al., 2012). The reason why the simulated SOA is low is probably because of low temperature and low biogenic VOCs emissions in winter time. Considering the modeled SOA is small, we do not expect it to significantly change the direct and indirect effects.

The reason why we chose the MOSAIC aerosol module other than MADE/SORGAM is because PM2.5 concentrations simulated with MOSAIC have a lower bias (15.0%) than that using MADE/SORGAM (49.7%) .

A recent paper (Huang et al., 2014) indicated that SOA at four China cities (Beijing, Shanghai, Guangzhou, and Xi'an) in January 2013 is comparable with secondary inorganic aerosol. However, present models have difficulties in representing SOA, especially during haze episodes in winter. While a thorough evaluation of the model's SOA simulation is out of the scope of this paper, we have added a discussion on the SOA uncertainty.

Please refer to the revised Section 3.3 for details.

Minor Issues:

Page 26086, line 20: "improves model's performances" should be "improves model performance".

Response: Corrected.

Page 26087

Line 6-7: "burn out the cloud" is not a particularly clear description of the meteorological process concerned. Please explain in more detail.

Response: Absorbing aerosols cause atmospheric heating, leading to evaporation of

clouds. More detailed explanation has been added.

Line 7: "increasing interests"→"increasing interest". Line 16: "version of Weather"–>"version of the Weather" Line 21: "convections"–>→"convection" ;"the PBL, and" might be better as"the PBL, hence"

Response: Done.

Page 26089:

Line 12: "as gas-phase"–>→"as the gas-phase" Line 16: "matters"–>→"matter"

Response: Done.

Lines 28-29: There needs to be an explanation as to why this particular forecast cycle was used (i.e. why not 2 days, or 1 day, or 7 days)?

Response: 48 hours are sufficient for meteorological and chemical variables to reach equilibrium. We found that biases in meteorological variables would become larger when the model was run continuously for more days. So, re-initialization for every five days is a reasonable strategy given consideration to both efficiency and accuracy. We've added explanations of why this particular forecast cycle was used in the revised manuscript.

Section 2.1 also needs mention of the aerosol assumptions used in the model for the EMP scenario(e.g. any default aerosol direct effect radiative properties, any default assumptions regarding cloud droplet numbers or cloud liquid water content).

Response: We've added descriptions of aerosol radiative properties and cloud droplet numbers assumptions in the revised manuscript. In EMP, aerosol radiative properties are not coupled with shortwave scheme, whereas they are coupled in BASE and RAD. In EMP and RAD, cloud droplet numbers are prescribed, while they are calculated based on aerosol activation in BASE.

Page 26090:

Line 4: "the radiative"→–>"the direct radiative"

Response: Direct and semi-direct effects could not be separate in WRF-Chem. So RAD scenario includes both direct and semi-direct effects.

Line 8: "model setups"–>→"model setup"

Response: Done.

Line 20: What was the basis for this particular split in PM mass between nucleation (not nuclei mode) and accumulation mode? Give a reference or an explanation.

Response: We split PM mass following recommendations in WRF-Chem user's guide. We've add an explanation of this.

Page 26091:

Line 12: I have some concerns that all of the observation stations are incities – the observations may thus be controlled by very local sources, which may not be captured that well in the model emissions at that resolution. Do the authors have rural stations that could be used for comparison as well?

Response: Each city has several observation stations, containing both urban and rural stations. In this study, the PM2.5 data in one city was averaged among all stations in the city, representing regional average. We've added the above description in the revised manuscript Section 2.3.

Line 14: "aerosols distributions"–>→"aerosol size distribution" Line 16: "performances are"–>→"performanceis"

Response: Done.

Page 26092:

Line 4: "meteorological variables."–>→"2 m relative humidity and temperature." Others have not been shown, so the statement should be limited to the analysis presented.

Response: Corrected.

Line 22-23: "may have influences on the accuracies"–>→"may influence the accuracy" Mention, in the discussion of Figure 2, that a later analysis is made for all three scenarios (and include the RAD run results in Figure 12).

Response: Done. The RAD results have been included in Figure 12 and discussed in the text.

Page 26093:

Line 1: "distributions"–>→"distribution"

Response: Done.

Line 4: "well captures": this sort of qualitative phrasing should be avoided in favor of a qualitative statement of the model biases, etc. For example, from Fig 4, the model apparently has a negative bias of 80 ug/m3 in the cities to the north-west. The authors also mention later (line 12) that the model has an overall low bias for cities with high PM2.5 levels, which seems to contradict the "well captures" statement made on line 4.

Response: We meant to say that the model simulates the spatial patterns. We've change "well captures" into "captures".

Line 18: "performances"–>→"performance" Line 24: "emissions is"–>→"emissions are" Line 26: "productions of"–>→"production of"

Response: Done.

Page 26094: Line 12: "matters"–>→"matter"

Response: Done.

Page 26095 Figure 7: Note that the "blue to red" colour scales make it difficult to distinguish contour levels that are on the low or high end of the scales. Suggest using a rainbow scale for the difference plots as well.

Response: We tried to use a rainbow scale (shown below). We found that it was hard to distinguish whether the value was positive or negative. The "blue to red" color scales are much easier to distinguish positive or negative values.

Line 16-17: "which our finding is consistent with."–>→"which is consistent with our findings."

Response: Done.

Line 21-22: The authors should add a few lines discussing the potential impact of ice nuclei on clouds here.

Response:We've added some lines talking about ice nuclei in the last paragraph of Section 4.1.

Line25: "a weaker"→–>"weaker" Line 28-line 1 next page: "Due to a weaker convection resulted from"–>→"Due to a weaker convection resulting from".

Response: Done.

Page 26096:

Line 6: "radiations. So that changes"–>→"radiation. Changes"

Response: Done.

Line16: "formations mainly occur"–>→"formation mainly occurs" Note that the caption for Figure 8 needs to mention that a-f are model values and g-l are differences (and for thelatter, which differences correspond to which panels needs to be identified).

Response: Done.

Page 26097

Lines 15-16: There should be some mention in the text whether the feedbacks improved the forecast for the pollutants. I may have missed this.

Response: In Section 5, we discussed the improvement of simulating radiation, temperature, and PM2.5.

Lines 22-23: Might be worth comparing to the AQMEII-2 Atmospheric Environment Special Issue papers, if you want a more recent and multi-model comparison (see their on-line page– there are a few comparing the equivalent of the authors' RAD, EMP and BASE simulations, for North American and European domains).

Response: Yes. We've compared our results with Kong et al. (2014) and Makar et al. (2014a and 2014b).

Line 28: Max enhancement is 69.3 ug/m3, but the colour scale in the figure only goes to 28: suggest that the entire max to min range is included in the scale.

Response: Agree. We've changed the range into -70 ug/m3 to 70 ug/m3.

Page 26098

Line 1: "Bohai Sea"–>→"the Bohai Sea". I suggest that one of the starting figures show the locations of placenames. Readers unfamiliar with the study region will not know where any of the places mentioned in the description are located.

Response: Done. We've defined the Bohai Sea surrounding area in the revised Figure 10.

Line 3: "respond to"–>→"responds to"

Response: Done.

Line8-9: There needs to be a justification for why the authors feel that the reduced PBL height and stabilized lower atmosphere is "the most important". Why?

Response: We found primary gas pollutants are suppressed due to aerosol radiative effects, and they are very sensitive to PBL height and stability of lower atmosphere. So we thought the reduced PBL height and stabilized lower atmosphere might be the most

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important. We've changed this expression into "the reduced PBL height and stabilized lower atmosphere explain a lot of the PM2.5 suppression". And we've added more descriptions.

Line 13-14:"which is the same situation for"–>→"and also for"

Response: Done.

Line 25: reference to Easter et al should also appear in section 2.1.

Response: Done.

Page 26099: Line 2: "from WRF-CHEM model configurations"–>→"from the WRF-CHEM model configurations used here" Line 10: "Chengdu."–>→"Chengdu (left column of panels in Figure 11)." Line 11: "January The"–>→"January. The" Line 12: "has a"–>→"have a" Line 13: "cities" is misspelled. Line 14: "Suppressions"–>→"Suppression" Line 15: "Changchun,"–>→"Changchun (right column of panels in Figure 11),"Line 23: "are suppressed"–>→"is suppressed"

Response: Done.

Page 26100

The first paragraph merely restates what has already appeared in thepaper and is unnecessary: delete.

Response: Done.

Figure 12 and the text associated with it should include the RAD run.

Response: We've included the RAD scenario in Figure 12 and added some text in Section 5.

Line 14: "model's performances"–>→"model performance".

Response: Done.

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Line 20: I'm not sure if NCP has been defined earlier in the text. Please define it, if not.

Response: NCP was defined in Section 3.2.

Page 26101

Line 5-6: "partially due to the missing of smaller scale temporal andspatial information averaging". This portion of the sentence is not clear, please rewrite.

Response: We've changed this into "partially due to temporal and spatial averaging"

Line 11: "Fig 12" should be "Fig 13" here, I think.

Response: Agree. Has been corrected.

Line 25: I suggest you quantify the last statement by including some bias values for the entire grid for each of the runs, to show how the overall performance changed.

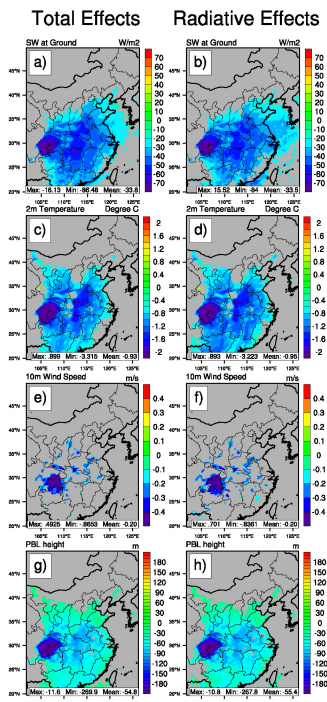Response: For some regions, we found improved model performance as shown in Figure 13. However, we did not see improved overall performance.

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Fig. 1. rainbow color scale