Response to all the reviews

We thank the reviewers 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 #1 (Comments to Author):

This study attempts to quantify the influences of the aerosol feedbacks on meteorology and air quality during a severe haze episode which occurred in January 2013 over eastern China. Three scenarios of WRF-Chem simulations were conducted to further differentiate the aerosol radiative and indirect effects. In general, it is a worthwhile analysis which should be published.

Response: Thank you.

Major comments:

1. First, estimates of feedbacks were filtered in a statistical way, as shown in Figure 7-10. More discussion is needed to explain how the Student's t test was employed to distinguish the aerosol-induced changes from the system noises. To my understanding, aerosol indirect effects may not be that straightforward as direct effects which can be simply attributed to the aerosol loading.

Response: We've added more descriptions about how the Student's t test was used in the revised Section 4. The Student's t test is used to test the null hypothesis that the two sample means are the same (H0: $\overline{X}_1 = \overline{X}_2$). Rejection of the null hypothesis indicates that the two sample means are different; that is, the aerosol-induced changes are significant. For every meteorological and chemical variable in a given grid point, the sample size is 744 (24 hrs ×31 days). How the t statistic is calculated is shown in the revised Appendix. By employing the Student's t test, system noises are excluded and only significant changes are shown and discussed in the manuscript.

We agree with the reviewer that aerosol indirect effect is not as straightforward as direct effect. Inevitably, some aerosol-induced changes, especially those caused by aerosol indirect effect, are excluded. However, the remaining changes after statistic filtering are strong enough for use to make reasonable judgments.

2. Second, as claimed in the manuscript, the PM2.5 was wrongly calculated by the model after the inclusion of aerosol indirect effects. Assuming that is true, I don't think the PM2.5 in BASE scenario is appropriate to be used for comparisons against observations, as presented in Figure 11 and 13. Considering large uncertainties in the estimation of aerosol indirect effects which are also less important than radiative effects during this particular episode, I would recommend the authors just focus on the discussion of radiative effects.

Response: The model results in this study showed that model performance was not improved when including aerosol indirect effects. The model performance was the best when only aerosol radiative effects were included. Our finding was consistent with previous studies (Kong et al., 2014; Makar et al., 2014a). As for the model setting, the scenario including both aerosol radiative and indirect effects represents the most realistic situation. Aerosol indirect effects might have non-negligible effects on simulating clouds, which is quite important for the wetter South China and for other seasons.

We agree with the reviewer that our research should pay the most attention on radiative effects. However, considering aerosol indirect effects are very important and gain a lot of concerns (as shown in Reviewer #2's comments), we retained the discussion of the indirect effects.

Specific:

P 26089, L29: "The model's meteorology is re-initialized every five days based on NCEP", why need to be re-initialized? Any nudging technologies was adopted?

Response: We found that the 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 did not employ any nudging technologies, since aerosol effects were included.

P 26092, L13: any explanation about the high-bias? Such as land-use type? Lack of aerosol radiative effects?

Response: The overestimation of wind speeds possibly results from unresolved topographical features in surface drag parameterization and coarse resolutions of the domain (Cheng and Steenburgh, 2005; Yahya et al., 2014). This explanation has been added in the revised Section 3.1.

P 26092, L17: the figures for "00:00" and "12:00" look quite similar, might consider to combine them as one.

Response: Agree. We've combined "00:00" and "12:00" as one profile in the figure. Please refer to the revised Figure 3.

P 26093, L20: it might be better to do the analysis on daily-averages instead of hour averages, because of the diurnal pattern.

Response: Agree. We've obtained the enhancement ratio on the basis of daily average in the revised manuscript.

P 26094, L5: lack of the inclusion of aerosol feedback in traditional modeling might also contribute to such low-biases.

Response: Agree. We've added some discussion about this at the end of this paragraph.

P 26096, L7: "are less significant than solar radiation", how to qualify, in percentage?

Response: First, solar radiation is reduced by 21% over the regions where the change is determined as significant by the Student' t test. By comparison, wind speed and PBL height are reduced by 6% and 14%, respectively. Second, as shown in Figure 7, the regions with significantly reduced solar radiation are much larger than those with significantly reduced wind speed or PBL height. We've added the descriptions in the revised manuscript.

P 26096, L20: "aerosol indirect effects play a much more significant role in changing cloud proper ties", but mostly in the south, please clarify it.

Response: We've clarified this in the revised manuscript.

P 26096, L22-25: "The reduction over these relatively clean areas may be explained by the lower particle number concentrations in the BASE scenario than the default droplet number mixing ratio of 1.0×10^6 kg^{-1} in scenarios without aerosol indirect effect.", I don't understand the sentence, please clarify it.

Response: In WRF-Chem, if aerosol indirect effect is turned off, the cloud droplet number concentration is prescribed as 1.0×10^6 kg^{-1} in the cloud microphysics scheme. If aerosol indirect effect is turned on, the cloud droplet number is calculated based on aerosol number concentration. We've modified this sentence as: "The reduction over these relatively clean areas may be explained by the smaller droplet number mixing ratio which is derived from lower particle number concentrations in the BASE scenario. The scenarios without aerosol indirect effects adopt the default value droplet number mixing ratio of 1.0×10^6 /kg which does not vary with aerosol number concentrations."

P 26097, L4: “the most parts of the domain”, should be the north of the domain.
Response: Corrected.

P 26097, L10: “indicating similar sources of these pollutants”, not true, suggest to delete it.
Response: Done.

P 26097, L12: “CO is enhanced by up to 446 ppb”, is that domain-average? Pleaseclarify it.
Response: It is the maximum enhancement over the domain. We’ve clarified it in the revised manuscript.

P 26098, L17: “The reduction of PM2.5 in WRF-Chem simulations with aerosol indirect effects mainly comes from two aspects”. More intensive precipitations may also help reduce the particles, particularly in the south.
Response: Agree. We’ve added this explanation in the revised manuscript.

P 26111, Table 2: NMB for T2 is “-83.3%”, the number does not make any sense. Using “K” unit would be better.
Response: We’ve re-calculated NMB for T2 using “K” unit in the revised manuscript.

P 26114, Figure 2: lack of the label for x-axis.
Response: Figure 2 has been replaced with a revised one with x-axis included.

Editorial:
P 26088, L18: “the model” should be “the model’s performance”
Response: Corrected.

P 26089, L14: “such as” should be “including”
Response: Corrected.

P 26090, L8: “etc” should be deleted
Response: Corrected.

P 26090, L9: “(BASE-EMP)” should be “(i.e., BASE-EMP)”
Response: Done.

P 26093, L1: “evaluate” should be “evaluated”
Response: Done.

P 26096, L14: “is conductive for” should be “enhances”
Response: Done.

P 26097, L9: “the” should be “these”
Response: Corrected.

P 26097, L30: “Reduction” should be “Reductions”
Response: Corrected.

P 26098, L3: “respond” should be “responds”

Response: Corrected.

<u>Anonymous Referee #2 (Comments to Author):</u>

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 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. 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/m^3, 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.

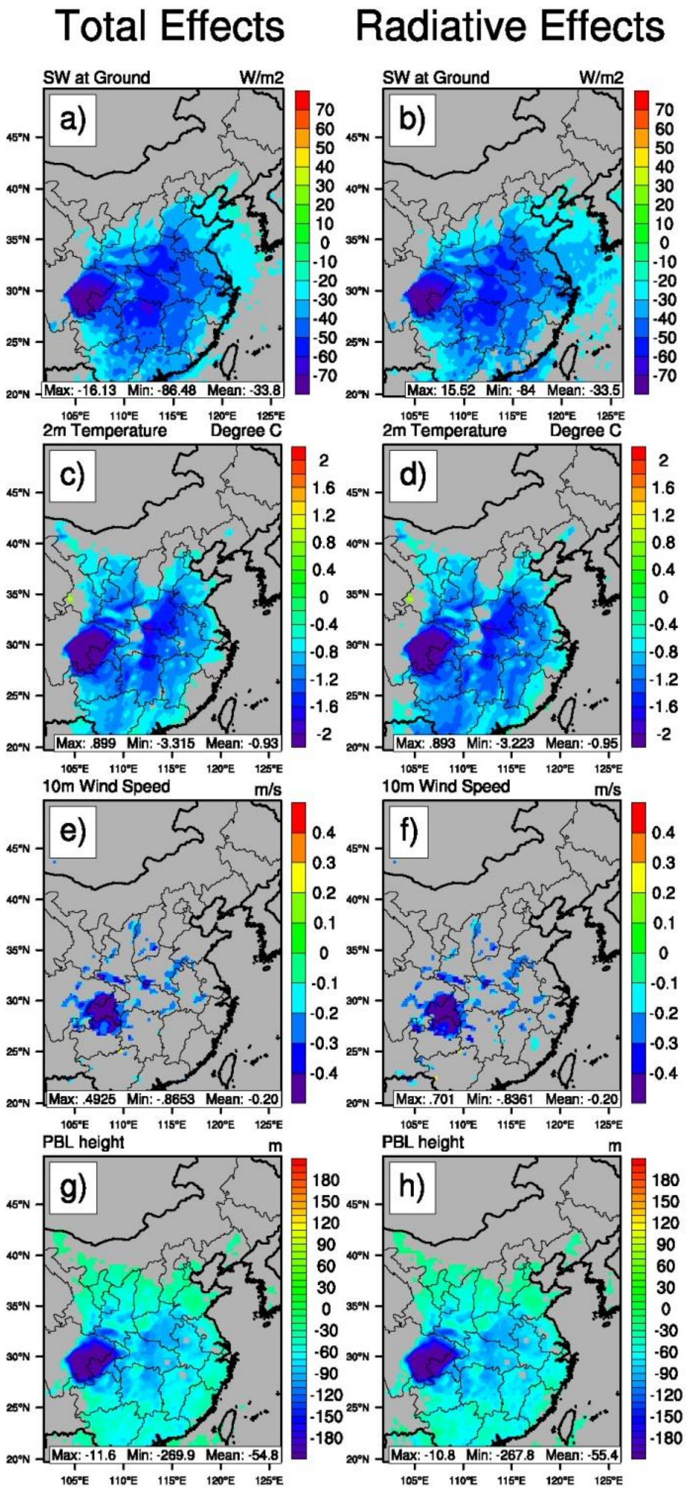
Page 26094: Line 12: “matters” ->→ “matter”

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/m^3, 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/m^3 to 70 ug/m^3.

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 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.

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.

All relevant changes:

L29
L39-40
L50
L69
L70-71
L84-94
L97
L98-99
L99-102

L110-111
L117-119
L120
L121-122
L130-131
L144-146
L150
L156
L159-160
L168-170
L173
L177
L181
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L380-387
L388-389

L393-394
L397
L404
L419
L426
L476-479
L482-483
L498-499
L515-518
L528-530
L541-553
L559-568
L584-586
L590-591
L614-616
L645-646
L648-652
L654-656
L662
L668
L702-705
L715-716

Simulating aerosol-radiation-cloud feedbacks on meteorology and air quality over eastern China under severe haze conditions in winter

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ABSTRACT

The aerosol-radiation-cloud feedbacks on meteorology and air quality over eastern China under severe winter haze conditions in January 2013 are simulated using the fully coupled on-line Weather Research and Forecasting/Chemistry (WRF-Chem) model. Three simulation scenarios including different aerosol configurations are undertaken to distinguish the ~~impact of~~ aerosol radiative (direct and semi-direct) and indirect effects ~~on meteorological variables and air quality~~. Simulated spatial and temporal variations of $PM_{2.5}$ are generally consistent with surface observations, with a mean bias of −18.9 μg/m^3 (−15.0%) averaged over 71 big cities in China. Comparisons between different scenarios reveal that aerosol radiative effects (direct effect and semi-direct effects) result in reductions of downward shortwave flux at the surface, 2 m temperature, 10 m wind speed and planetary boundary layer (PBL) height by up to 84.0 W/m^2, 3.2 ^{o}C, 0.8 m/s, and 268 m, respectively. The simulated impact of the aerosol indirect effects is comparatively smaller. Through reducing the PBL height and stabilizing lower atmosphere ~~wind speeds~~, the aerosol effects lead to increases in surface concentrations of primary pollutants (CO and SO_2). ~~and $PM_{2.5}$. The aerosol feedbacks on secondary pollutants such as surface ozone and $PM_{2.5}$ mass concentrations show some spatial variations.~~ Surface O_3 mixing ratio is reduced by up to 6.9 ppb due to reduced incoming solar radiation and lower temperature, while the aerosol feedbacks on $PM_{2.5}$ mass concentrations show some spatial variations. Comparisons of model results with observations show that inclusion of aerosol feedbacks in the model significantly improves ~~model's performances~~ model performance in simulating meteorological variables and improves simulations of $PM_{2.5}$ temporal distributions over the North China Plain, the Yangtze River Delta, the Pearl River Delta, and Central China. Although the aerosol-radiation-cloud feedbacks on aerosol mass concentrations are subject to uncertainties, this work demonstrates the significance of aerosol-radiation-cloud feedbacks for real-time air quality forecasting under haze conditions.

1. Introduction

Atmospheric aerosols are known to play a key role in the earth climate system. They absorb and scatter incoming solar radiation, referred to as the direct effect (Hansen et al., 1997). They also alter cloud properties by serving as cloud condensation nuclei (CCN), which is known as the indirect effect (Albrecht, 1989; Twomey, 1977; Rosenfeld et al., 2008). Absorbing aerosols in and under the cloud may heat the atmosphere, leading to evaporation of ~~also burn out the clouds~~ clouds (semi-direct effect) (Charlson and Pilat, 1969). It is of increasing ~~interests~~ interest to understand and quantify the complex impacts of aerosols on meteorology and air quality. The coupled “on-line” meteorology-air quality strategy with aerosol feedbacks is essential for real-time air quality forecasting using 3-D models. Negligence of aerosol feedbacks may lead to poor performance of the next hour’s meteorology and air quality forecasting, especially for high aerosol loading regions (Grell and Baklanov, 2011; Zhang et al., 2012).

Models simulating the ~~aerosol~~ direct, indirect, and semi-direct effects of aerosols on meteorology and chemistry need to couple aerosols with physical and chemical processes. The chemistry version of Weather Research and Forecasting (WRF-Chem) model (Grell et al., 2005) is a state-of-the-art meso-scale “on-line” atmospheric model, in which the chemical processes and meteorology are simulated simultaneously. This design makes WRF-Chem capable of simulating aerosol feedbacks on various atmospheric processes. Several studies employing WRF-Chem reveal that aerosols reduce downward solar radiation reaching the ground, inhibit ~~convections~~ convection, reduce the PBL, ~~and~~ hence make the lower atmosphere more stable (Fan et al., 2008; Forkel et al., 2012; Zhang et al., 2010). WRF-Chem results also indicate that aerosols can modify atmospheric circulation systems, resulting in changes in monsoon strength, precipitation distribution, and mid-latitude cyclones (Zhao et al., 2011; Zhao et al., 2012).

In January 2013, several severe and long-lasting haze episodes appeared in eastern China (Figure 1). Monthly mean mass concentrations of fine particulate matters ($PM_{2.5}$) exceeded 200 μg/m^3 in some cities in North China Plain. Meteorological conditions and chemical components of $PM_{2.5}$ during this month have been investigated by a number of studies in order to understand the chemical characteristics and formation mechanism of severe winter haze episodes (Bi et al., 2014; Che et al., 2014; Huang, K. et al., 2014; Sun et al., 2014; Wang, L. T. et al., 2014; Wang, Y. S. et al., 2014; Wang, Y. X. et al., 2014; Wang, Z. F. et al., 2014; Zhang, J. K. et al., 2014). Meanwhile, such high levels of PM concentrations are expected to exert impacts on meteorological conditions through the aerosol-radiation-cloud interactions. Few of current air quality forecasting systems for China include aerosol-meteorology interactions. The significance of this effect and the extent to which it feedbacks on air quality remains to be uncertain and needs to be quantified for better forecasting air quality in China in the future (Wang et al., 2013; Zhang, Y. et al., 2014; Wang, Z. F. et al., 2014).

In this work, the fully coupled “on-line” WRF-Chem model is employed to simulate the complex interactions between aerosols and meteorology and to characterize and quantify the influences of aerosol feedbacks on meteorology and air quality under severe winter haze conditions in January 2013 over eastern China. The aerosol direct, indirect and semi-direct effects are all included in the WRF-Chem simulation and analyzed separately. The WRF-Chem model configuration, scenarios setup, and observation data are described in Section 2. Section 3 evaluates the model performance in simulating meteorology and air quality. In Section 4, the aerosol feedbacks on meteorology and air quality are analyzed and discussed. Section 5 investigates the effects of including aerosol feedbacks ~~in the model to~~ on the ~~model's performances~~ model performance. The concluding remarks are given in Section 6.

2. Model and Observations Description

2.1 WRF-Chem Model and Scenarios Setup

The WRF model is a state-of-the-art meso-scale non-hydrostatic model, and allows for many different choices for physical parameterizations (http://www.wrf-model.org/). WRF-Chem is a chemical version of WRF that simultaneously simulates meteorologicaland chemical components. The version 3.3 of WRF-Chem released on April 6, 2011 is used in this study. A more detailed description of the model can be found in previous studies (Grell et al., 2005; Fast et al., 2006; Chapman et al., 2009).

The main physical options selected in this study include the Goddard shortwave radiation scheme coupled with aerosol direct effects (Chou et al., 1998), the Rapid Radiative Transfer Model (RRTM) longwave radiation scheme (Mlawer et al., 1997), the Noah Land Surface Model (Chen and Dudhia, 2001), the Yonsei University (YSU) boundary layer scheme (Hong et al., 2006), the Lin microphysics scheme coupled with aerosol indirect effects (Lin et al., 1983), and the Grell-Devenyi cumulus parameterization scheme (Grell and Dévényi, 2002).

For the aerosol direct effect, aerosol optical properties such as extinction, single scattering albedo, and asymmetry factor 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, while 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. When indirect effects are included, a more comprehensive in- and below-cloud aerosol wet removal module following the method of Easter et al (2004) is employed.

The Carbon Bond Mechanism version Z (CBMZ) (Zaveri and Peters, 1999) is used as the gas-phase chemistry scheme. The Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) (Zaveri et al., 2008) is applied as aerosol module. MOSAIC simulates aerosol species including ~~such as~~ sulfate, methanesulfonate, nitrate, ammonium, chloride, carbonate, sodium, calcium, black carbon (BC), organic carbon (OC), and other unspecified inorganic ~~matters~~ matter (OIN). Secondary organic aerosols (SOA) are not included in the version of MOSAIC used in this study. Section 3.3 discusses the impact of SOA on our analysis by replacing MOSAIC with the Modal Aerosol Dynamics Model for

Europe (MADE) (Ackermann et al., 1998) with the secondary organic aerosol model (SORGAM) (Schell et al., 2001) (referred to as MADE/SORGAM). MOSAIC in WRF-Chem uses a sectional approach to represent particle size distribution. In this study, four size bins (0.039–0.156 μm, 0.156–0.625 μm, 0.625–2.5 μm, 2.5–10.0 μm dry diameter) are employed, and aerosols are assumed to be internally mixed within each bin.

The simulated time period is the whole month of January 2013. Figure 1 illustrates the model domain, which covers eastern China (19°−51°N, 96°−132°E) and has a horizontal resolution of 27 km × 27 km. There are 28 vertical levels extending from the surface to 50 hPa. The initial and boundary conditions for WRF are provided by the 6-hourly 1° × 1° National Centers for Environmental Prediction (NCEP) Final Analysis (FNL). Chemical boundary conditions are provided by MOZART simulations (Emmons et al., 2010). In the initial spin-up process, the model is run with NCEP meteorological and MOZART chemical conditions for 48 hours, which are sufficient for the meteorological and chemical variables to reach equilibrium. Considering both accuracy and efficiency, ~~The~~ the model's meteorology is re-initialized every five days based on NCEP, while chemistry adopts the previous state.

In order to investigate the impact of aerosol feedbacks on meteorology and air quality, three WRF-Chem simulation scenarios are performed and compared. The first is the baseline scenario (BASE), including all aerosol effects on meteorology (i.e., direct, indirect, and semi-direct). The second scenario (RAD) focuses on the radiative effects by excluding aerosol indirect effects from the BASE scenario. The third scenario (EMP) does not contain any aerosol effects on meteorology. Aerosol radiative properties are connected with the shortwave radiation scheme in the BASE and RAD scenario but not in the EMP scenario. Cloud droplet number is prescribed to be 1.0×10^6 /kg in EMP and RAD, while it is calculated based on aerosol activation in BASE. Other than the differences in aerosols effects, the three scenarios are identical in input data (e.g., emissions and boundary conditions~~, etc~~) and model ~~setups~~ setup. The difference between BASE and EMP (i.e., BASE – EMP) is used to investigate the impact of total aerosol feedbacks, while the difference between BASE and RAD (i.e., BASE – RAD) and that between RAD and EMP (i.e., RAD – EMP) represents the influence of aerosol indirect effects and radiative (both direct and semi-direct) effects, respectively. Table 1 summaries the characteristics of the three scenarios.

2.2 Emissions

Anthropogenic emissions are taken from the Multi-resolution Emission Inventory of China (MEIC) (http://www.meicmodel.org/), which provides emissions of sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), ammonia (NH_3), BC, OC, PM_{10}, $PM_{2.5}$, and non-methane volatile organic compounds (NMVOCs) for China for the year 2010. NO_x emissions contain 90% of NO_2 and 10% of NO by mole fraction. PM emissions are assumed to be split into 20% in nuclei mode and 80% in accumulation mode, according to the recommended construction of anthropogenic emissions inventory in WRF-Chem user's guide.

Biogenic emissions are calculated on-line in the model based on the Model of Emissions of Gases and Aerosols from Nature (MEGAN) inventory (Guenther et al., 2006). Dust is included in the simulations, while sea salt or dimethylsulfide

(DMS) are not, and their impacts are expected to be small over eastern China in winter.

2.3 Observations

The simulation results are compared with meteorological and chemical observations. Daily meteorological observations at 523 stations are obtained from the National Climate Data Center (NCDC) of China Meteorological Administration (CMA), including 2 m temperature, 2 m relative humidity (RH), and 10 m wind speed. The radiosonde profiles at 20 stations are provided by the department of atmospheric science ~~in~~ at the University of Wyoming (http://weather.uwyo.edu/upperair/sounding.html).

The measurements of ~~real-time hourly~~ near surface $PM_{2.5}$ mass concentrations are obtained from China National Environmental Monitoring Center (CNEMC). The observation data have been ~~were~~ released since January 2013, including hourly concentrations of SO_2, NO_2, CO, Ozone (O_3), PM_{10} and $PM_{2.5}$ at 74 big cities in China with 71 of those cities covered by the study domain. Each city has several observation stations, containing both urban and rural stations. In this study, the $PM_{2.5}$ concentration for one city is the average of all the stations in the city, representing thus a more regional condition of the pollution level. This database allows for a spatially extensive evaluation of air quality simulations by atmospheric models.

3. Model Evaluations

Accurate representation of meteorology and ~~aerosols distributions~~ aerosol concentrations in the model provides the foundations of quantifying aerosol feedbacks. Therefore, in this section, the model ~~performances are~~ performance is evaluated by comparing model results with surface and radiosonde observations. If not otherwise specified, the model results presented in this section are from the BASE scenario, which represents the most comprehensive realization of different processes in the model. It should be noted that systematic differences may result from the comparison between grid-mean values and point measurements, since a model grid covers an area of 729 km^2 (27 km × 27 km).

3.1 Meteorology

Meteorology strongly affects the ~~formations, transportations, and eliminations~~ formation, transport, and elimination of atmospheric aerosols. The selected ~~near surface~~ meteorological variables for model evaluation are ~~2 m~~ temperature, ~~2 m~~ relative humidity, and ~~10 m~~ wind speed. ~~Vertical profiles of temperature, relative humidity and wind speed are also compared.~~ Figure 2 shows the time series of observed and simulated daily mean ~~meteorological variables~~ 2 m temperature and 2 m relative humidity, with the statistical summary of the comparisons shown in Table 2. The model reproduces temporal variations of these two meteorological variables.

The statistical indices used here are mean observation (MEAN OBS), mean simulation (MEAN SIM), correlation coefficient (Corr. R), mean bias (MB), normalized mean bias (NMB), and root mean square error (RMSE). The definitions of these indices are given in the Appendix. The model reproduces 2 m temperature with a correlation of 0.97 and a cold bias of −1.0 ^{o}C, mainly due to underestimation in the first 9 days of the month. Relative humidity is simulated with a correlation of 0.47 and a negligible mean bias. The 10 m wind speed is systematically overestimated by the model by 105%. ~~This~~ A high positive bias in wind speed is also reported by several other studies using WRF-Chem (Matsui et al., 2009; Molders et al., 2012; Tuccella et al., 2012; Zhang et al., 2010). This high-bias probably results from unresolved topographical features in surface drag parameterization and the coarse resolution of the domain (Cheng and Steenburgh, 2005; Yahya et al., 2014).

Air pollution is influenced not only by surface meteorology, but also by vertical patterns of meteorological variables. In Figure 3, the monthly mean vertical profiles of simulated meteorological variables are compared with sounding observations (averaged at 00UTC and 12UTC). Generally, the model captures vertical variations of meteorological variables. The model well reproduces vertical variations of temperature with a small bias. The model underestimates relative humidity below 650 hPa and overestimates it in the upper levels. Wind speeds are overestimated in the lower atmosphere and underestimated in the upper atmosphere. The errors in meteorology may ~~have influences on the accuracies~~ influence the accuracy of simulating processes of aerosol formation, ~~transportation~~ transport and deposition. Overall, the evaluations presented here suggest that the model adequately simulates the spatial and temporal variations of meteorological variables of significant relevance to air quality.

3.2 $PM_{2.5}$

We first ~~evaluate~~ evaluated the spatial ~~distributions~~ distribution of simulated monthly mean $PM_{2.5}$ mass concentrations by comparing model results with observations at 71 big cities in the model domain in January 2013. As shown in Figure 4, the model ~~well~~ captures the spatial patterns of $PM_{2.5}$ during the month, including high levels of $PM_{2.5}$ over southern Hebei, Henan, Hubei Province, Sichuan Basin and three big cities (Harbin, Changchun, Shenyang) in Northeast China. The North China Plain (NCP), Central China (CC) area, and Sichuan Basin have the highest monthly mean $PM_{2.5}$ mass concentrations. $PM_{2.5}$ pollution is more severe over the Yangtze River Delta (YRD) than that over the Pearl River Delta (PRD).

Figure 5 presents the scatter plots of observed and simulated monthly mean $PM_{2.5}$ mass concentrations at 71 cities. The model has a low bias ranging from 25% to 70% for cities with monthly mean $PM_{2.5}$ exceeding 200 $\mu g/m^3$. The observed and simulated time-series of hourly surface $PM_{2.5}$ averaged over all the cities are compared in Figure 6. The model simulates hourly $PM_{2.5}$ with a temporal correlation of 0.67, and underestimates monthly mean $PM_{2.5}$ mass concentrations by 18.9 $\mu g/m^3$ (15.0%). The model generally reproduces the observed temporal variations of $PM_{2.5}$.

The enhancement ratio is employed to further evaluate the ~~model's performances~~ model performance in simulating $PM_{2.5}$ temporal variations in different regions. Within a given grid point, the ~~The~~ enhancement ratio is defined as the average of ~~hourly~~ daily $PM_{2.5}$ mass concentrations exceeding the median divided by that less than the median, representing changes of $PM_{2.5}$ from clean to polluted situations. As shown in Table 3, observed enhancement ratios are around ~~1.7~~ 1.6 over NCP, YRD, PRD and CC. The simulated enhancement ratios range from 1.8 to 2.0 over the four regions, which are close to observations. Since changes of hourly emissions ~~is~~ are not considered in this study, $PM_{2.5}$ enhancements mainly result from worsened meteorological conditions and more ~~productions~~ production of secondary aerosols. The consistency of the simulated enhancement ratios with the observed ones by region ~~indicate~~ indicates that the WRF-Chem model has some success in simulating the changes of ~~aerosol-related~~ meteorological and chemical processes from clean to polluted situations.

However, the model fails to capture the extremely high values of $PM_{2.5}$ during the haze episodes in January 2013, for example, January 13~15 and 18~20. Both positive and negative bias exists in simulated hourly $PM_{2.5}$. The model's underestimation during the severe winter haze episodes is consistent with previous studies (Liu et al., 2010; Wang, L. T. et al., 2014; Wang, Y. X. et al., 2014; Zhou et al., 2014). Possible reasons for this underestimation are: (1) the bias in simulating meteorological conditions during haze episodes; (2) uncertainties in emissions; (3) missing ~~secondary organic aerosols~~ SOA in the MOSAIC mechanism, which will be addressed below; and (4) the lack of formation mechanisms of secondary inorganic aerosols, like heterogeneous oxidation of SO_2 on the surface of particulate ~~matters~~ matter (Harris et al., 2013). By adjusting SO_2 and NO_x emissions according to surface observations and parameterizing the heterogeneous oxidation of SO_2 on deliquesced aerosols in the GEOS-Chem model, Wang, Y. X. et al. (2014) reported improvements of simulated $PM_{2.5}$ spatial distribution and an increase of 120% in sulfate fraction in $PM_{2.5}$. The traditional offline models do not include the aerosol feedbacks on meteorology, which may cause a low-bias of $PM_{2.5}$ for the severe pollution episodes. The inclusion of aerosol feedbacks in atmospheric models improves the model performance, which will be discussed in Section 5.

3.3 SOA

The MOSAIC aerosol module used in the three scenarios does not include SOA. Here we examine the sensitivity of modeled $PM_{2.5}$ to SOA by replacing MOSAIC with the MADE/SORGAM aerosol module in the BASE scenario. The test period is January 6~10, 2013. The mean SOA simulated from MADE/SORGAM is 0.3 ug/m^3, making up only 2.8% of OA (Table 4). This is comparable with the magnitude of simulated winter SOA concentrations reported by another study using WRF-Chem for the whole China (Jiang et al., 2012). The low SOA concentrations simulated by the model can be explained by low emissions of biogenic VOCs (key precursors of SOA) and low temperatures in wintertime. Recent observations have suggested much higher SOA concentrations at Chinese cities in winter, pointing to the importance of anthropogenic VOCs as SOA precursors in China (Huang et al., 2014), but a thorough investigation of this issue is outside the scope of this study. As shown in Table 4, the NMB of $PM_{2.5}$ with the MADE/SORGAM option is more than 3 times larger than that with MOSAIC. Since the modeled SOA has a small contribution to total $PM_{2.5}$, the MOSAIC aerosol module is chosen in this study and the omission of SOA in MOSAIC is not expected to affect our analysis of the aerosol effects on meteorology.

4. Aerosol Feedbacks on Meteorology and Air Quality

As seen in the previous section, the WRF-Chem model has shown some success in simulating meteorology and $PM_{2.5}$. Therefore, in this section, we aim to characterize and quantify the aerosol feedbacks on meteorology and air quality by comparing the three different scenarios described in Section 2.

In addition to different setups of the aerosol-radiation-cloud feedbacks, differences among the three scenarios can also result from model noise, such as errors in numerical computation and disturbances from discrete updating initial and boundary conditions. The Student's t-test is employed to identify statistically significant differences between the scenarios. The null hypothesis is that the two scenarios in comparison give the same simulation results. The sample size is 744 (24 hrs × 31 days) for each meteorological and chemical variable in a given grid box. Rejection of the null hypothesis indicates that the difference between the two scenarios is significantly different. The Appendix describes the calculation of the t statistic in more detail. We only present and discuss aerosol-induced changes of meteorological and chemical variables which exceed 95% confidence interval.

4.1 Feedbacks on Meteorology

The evolution of atmospheric aerosols is strongly influenced by meteorological variables, such as solar radiation, air temperature, and wind speed~~, etc~~. Figure 7 illustrates ~~aerosol impacts~~ the mean impact of aerosols on downward shortwave flux at the ground, 2 m temperature, 10 m wind speed and PBL height over eastern China in January 2013. Downward shortwave flux at the ground is strongly influenced by the existence of atmospheric aerosols, especially over high aerosol-loading regions. Aerosols affect shortwave radiation reaching the ground in two ways. First, particles scatter and absorb incoming solar radiation directly, resulting in surface dimming. Second, in-cloud particles change cloud lifetime and albedo, thus causing variations of shortwave radiation at the ground surface. As in Figure 7a, the downward shortwave flux at the ground is reduced over the vast areas of eastern China by up to −84.0 W/m^2, which mainly results from the aerosol radiative effects (Figure 7b). Consistent with our findings, ~~By employing ground-based measurements, aerosol optical and radiative properties over NCP during January 2013 were characterized in~~ Bi et al (2014) and Che et al (2014)~~. They~~ report~~ed~~ strong negative aerosol direct radiative forcing at the surface (with maximum daily mean exceeding −200.0 W/m^2) through analysis of ground-based measurements, aerosol optical and radiative properties over NCP in January 2013. ~~which our finding is consistent with our findings. Forkel et al. (2012) simulated aerosol direct and indirect effects over Europe, where aerosol concentrations were relatively low ($PM_{2.5}$=10-20 μg/m^3), and suggested that the aerosol indirect effects dominated in aerosol feedbacks on downward shortwave flux at the ground. Different from Forkel et al. (2012), we find that aerosol indirect effects have little influence on the downward shortwave flux at the ground (not shown here). This may be explained that cloud is not so important in winter over the continent.~~

When the downward shortwave flux at the ground is decreased due to aerosol interception, near surface energy fluxes are suppressed, leading to ~~a~~ weaker convection. Near surface air is heated mainly by longwave radiation emitted from the ground. In ~~that~~ the case of decreasing ~~, when~~ shortwave flux at the ground ~~is decreased~~, the near surface air is cooled and less longwave radiation is emitted from the surface ~~and thus the near surface air is cooled~~. Due to ~~a~~ weaker convection ~~resulted~~ resulting from less shortwave radiation reaching the ground, 2 m temperature is reduced by up to 3.2 ^{o}C, 10 m wind speed is reduced by up to 0.8 m/s, and PBL height is also reduced by up to 268 m, as shown in Figure 7c, 7e and 7g, respectively. Meteorological variables such as air temperature, wind speed, and PBL height could also be influenced by other factors like land surface properties (Zhang et al., 2010), ~~other than~~ in addition to solar radiations. ~~So that changes~~ The change of these variables, especially wind speed, ~~is~~ are less significant than that of solar radiation. First, solar radiation is reduced by 21% over the regions with significant changes, while wind speed and PBL height are reduced by 6% and 14%, respectively. Second, as shown by Figure 7, the regions with significant reduced solar radiation are much larger than those with significant reduced wind speed or PBL height. However, the spatial pattern of the changes of these variables is consistent with that of downward shortwave flux, which indicates that a more stable lower atmosphere resulting from less shortwave radiation plays an important role in aerosol feedbacks. The aerosol indirect effects during the severe haze episodes are found to be not significant in altering solar radiation, temperature, wind speed or PBL height over eastern China, which is not shown here. Overall, the near surface atmosphere is more stable when aerosol feedback is considered in the model, which ~~is conductive for~~ enhances pollution accumulation.

The amount of precipitation is low in January 2013 for most regions in China (Wang and Zhou, 2005). Cloud and precipitation ~~formations~~ formation mainly ~~occur~~ occurs over areas in the south and over the ocean (Figure 8a and 8d). In this month, the changes of cloud and precipitation due to aerosol radiative effects are not significant (Figure 8h). Aerosol indirect effects directly alter cloud properties such as effective radius, cloud lifetime, and precipitation rate. As shown in Figure 8i, aerosol indirect effects play a much more significant role in changing cloud properties, mostly in the south. Cloud water path is greatly reduced by up to 5.7 kg/m^2 over the junction of Yunnan and Guizhou Province and the ocean around Taiwan. ~~The reduction over these relatively clean areas may be explained by the lower particle number concentrations in the BASE scenario than the default droplet number mixing ratio of 1.0×10^6 /kg in scenarios without aerosol indirect effect.~~ The reduction over these relatively clean areas may be explained by the smaller droplet number mixing ratio which is derived from lower particle number concentrations in the BASE scenario. The scenarios without aerosol indirect effects adopt the default value droplet number mixing ratio of 1.0×10^6 /kg which does not vary with aerosol number concentrations. Reduced cloud droplet number results in accelerating auto-conversion to rain droplets. Thus, simulated monthly precipitation is increased by almost 100% over these areas (Figure 8j). Similar results are found when we replace the Lin microphysics scheme by the two-moment Morrison scheme (Morrison et al., 2009). Previous model assessments also showed that the inclusion of aerosol indirect effect reduced cloud water content over South Pacific ocean and ~~made~~ improved model ~~simulations match better~~ comparison with aircraft observations (Yang et al., 2011). The relatively small precipitation, as well as small changes of precipitation due to aerosol feedbacks over the north ~~most parts~~ of the domain, suggests that precipitation has a minor effect on near surface aerosols in January 2013.

Several studies under the Air Quality Model Evaluation International Initiative (AQMEII) project suggested that the aerosol indirect effect dominated in aerosol feedbacks on solar radiation, temperature, and PBL height (Forkel et al., 2012; Forkel et al., 2014; Kong et al., 2014; Makar et al., 2014a). Different from these studies, we find that aerosol indirect effects have little influence on the downward shortwave flux at the ground, 2 m temperature, 10 m wind speed, and PBL height (not shown here). In order to investigate how grid resolution relates to aerosol indirect effect, we have conducted another three groups of nested grid runs, using the same scenarios (BASE, RAD, and EMP). The outer domain is the same with previous domain, but a 9-km nested domain covering North China Plain is added. In the 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, suggesting grid resolution might not be the reason why aerosol indirect effect is minor in our study.

The discrepancy relating to aerosol indirect effect between those studies under AQMEII and our work may be explained by three reasons. First, we focus 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. Studies under AQMEII focused on summertime, when aerosol indirect effects on solar radiation, temperature, and PBL height were 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 aerosol indirect effect. The aerosol indirect effect was found to dominate over clean ocean and near ocean land (Forkel et al., 2014; Kong et al., 2014). Third, we do find some regions dominated by aerosol indirect effect. However, the Student's t test filters the values over those regions, because the changes due to aerosol indirect effect are not statistically significant.

Ice crystals can be formed by activation of Ice Nuclei (IN). Recently, increasing evidence has suggested that aerosols, especially dust, black carbon, and organic matters, can influence cloud physical process by acting as IN (Tao et al., 2012). The WRF-Chem version in this work does not include direct interactions of aerosol number concentration with ice nuclei, which may be another reason for the possible underestimation of aerosol indirect effects.

4.2 Feedbacks on Air Quality

Through moderating meteorological variables, aerosols exert feedbacks on air quality. Figure 9 shows spatial distributions of CO, SO_2, and O_3 and the ~~aerosol~~ feedbacks of aerosols on ~~the~~ these three gas pollutants in January 2013. Spatial patterns of CO and SO_2 are similar with that of $PM_{2.5}$~~, indicating similar sources of these pollutants~~. The near surface CO and SO_2 concentrations are increased when aerosol feedbacks are included. Over the domain, CO is enhanced by up to 446 ppb, while SO_2 is increased by as much as 28 ppb. Large increases of CO and SO_2 are found over areas with high aerosol loading. This phenomenon may mainly result from lower PBL and a more stable atmosphere near the surface due to aerosol radiative effects, as discussed in Section 3.2. We also found aerosol indirect effects do not have significant influence on gas pollutants.

The formation of O_3 is directly related to solar radiation and temperature in regions with sufficient NO_x and VOCs. The lower air temperature and reduced incoming solar radiation as a result of aerosols radiative effects lead to reduced photolysis rate of NO_2 and consequently reduce O_3 concentrations. The largest suppression of surface ozone by aerosols is found to be up to −6.9 ppb in the warmer southern China. Changes in northern China are relatively small. These findings are similar to those in Zhang et al. (2010), ~~and~~ Forkel et al. (2012), Kong et al. (2014) and Makar et al. (2014b).

The aerosol-radiation-cloud feedbacks on near surface aerosol mass concentrations are illustrated in Figure 10. As shown in Figure 10a, both increases and decreases of $PM_{2.5}$ are found in the domain. Enhanced $PM_{2.5}$ mass concentrations are simulated over Henan, Hubei, Guangxi Province, and Sichuan Basin with the maximum enhancement of 69.3 μg/m^3. ~~Reduction~~ Reductions in $PM_{2.5}$ as much as −38.2 μg/m^3 are simulated over the Bohai Sea surrounding area, Northeast China and the conjunction area of Yunnan, Guangxi, and Guizhou Province (Southwest China).

In order to better understand the mechanisms of how $PM_{2.5}$ ~~respond~~ responds to aerosol feedbacks, the aerosol effects are divided into aerosol radiative effects (Figure 10b) and indirect effects (Figure 10c). $PM_{2.5}$ can be influenced by the changes in various atmospheric processes due to aerosol radiative effects. For example, lower temperature may suppress the formation of sulfate, and reduced solar radiation may inhibit the oxidations of precursors of secondary aerosols. Among the various changes in atmospheric processes, the reduced PBL height and the stabilized lower atmosphere ~~is the most important.~~ may be the most important factors to explain the increase of $PM_{2.5}$ caused by the aerosol radiative effects, since primary gas pollutants also increase when aerosol radiative effects are included. From Figure 11b, we can see that $PM_{2.5}$ is greatly increased by aerosol radiative effects over the region where solar radiation and PBL height are significantly reduced during winter haze (Figure 7a and 7g). The mechanism involved is that aerosol radiative effects stabilize the lower atmosphere and suppress the dilution and ventilation of $PM_{2.5}$~~, which is the same situation and also for~~ as well as primary gas pollutants.

The comparison between Figure 10a and 10c indicates that aerosol indirect effects are the main reason of the suppression of $PM_{2.5}$. The reduction of $PM_{2.5}$ in WRF-Chem simulations with aerosol indirect effects mainly comes from ~~two~~ three aspects. First, once aerosol indirect effects are included in the model, the cloud droplet number is based on simulated atmospheric aerosol number other than what is prescribed in the model ~~scenarios without indirect effects~~. This coding strategy allows interstitial air-borne aerosols to become cloud-borne aerosols after activation. Therefore, air-borne aerosols are reduced in simulations including aerosol indirect effects, especially over cloudy regions like southwestern China (Figure 8a). Second, enhanced precipitation may also help reduce the air-borne particles, especially in the south. Third, in the simulations including aerosol indirect effects, a more comprehensive in- and below-cloud aerosol wet removal module following the method of Easter et al. (2004) is employed, while in the simulations without aerosol indirect effects, this module is not activated. In this aerosol wet removal mechanism, the removal processes are assumed to be irreversible, and aerosol re-suspension is not considered, even when precipitation is weak. This leads to a stronger removal of atmospheric aerosols when ~~including~~ aerosol indirect effects are included. It should be noted that in this work, the enhancement of aerosol wet removal process, when including aerosol indirect effects in the model, mainly results from WRF-Chem model configurations used here, not from aerosol-induced changes in cloud properties or precipitation.

The above discussion is based on model results temporally averaged during the whole month. In order to better understand $PM_{2.5}$ variations on a day to day basis, 4 cities with significant $PM_{2.5}$ enhancements and 4 cities with significant $PM_{2.5}$ reductions are selected. Figure 11 shows the time series of observed and simulatedhourly surface $PM_{2.5}$

mass concentrations in the selected 8 cities in January 2013. The four cites with increasing monthly mean $PM_{2.5}$ due to aerosol feedbacks are Zhengzhou, Wuhan, Changsha, and Chengdu (left panels in Figure 11). Major enhancements of $PM_{2.5}$ are simulated when $PM_{2.5}$ levels are high, for example, during the period of January 15~17. The changes in $PM_{2.5}$ ~~has~~ have a moderate negative correlation with the changes in PBL height (correlations coefficient ≈−0.3) at the four ~~cites~~ cities, suggesting that the $PM_{2.5}$ enhancement is partly caused by decreased PBL height in these regions. ~~Suppression~~ The reduction of $PM_{2.5}$ in Qinhuangdao, Tianjin, Shenyang, and Changchun (right panels in Figure 11), which are the four cities with decreased monthly mean $PM_{2.5}$, mainly ~~happen~~ happens in the last 5 days of the month (January 26~31).

In summary, aerosol radiative effects reduce the downward shortwave flux at the ground, decrease near surface temperature and wind speed, and further weaken convection, all leading to a more stable lower atmosphere. In a more stable lower atmosphere due to aerosol radiative effects, primary gas pollutants (CO and SO_2) and $PM_{2.5}$ are enhanced, while O_3 is decreased because of less incoming solar radiation and lower temperatures. $PM_{2.5}$ ~~are~~ is ~~suppressed~~ reduced when aerosol indirect effects are included, mainly due to the transition from air-borne aerosol to cloud-borne aerosol and the activation of a more comprehensive aerosol wet removal module. The underestimations at the higher end indicate that some key mechanisms are missing in the model, especially the productions of secondary aerosols.

5. Effects of Including Aerosol Feedbacks on ~~Model's Performances~~ Model Performance

~~In previous sections, the model results are evaluated, and the aerosol feedbacks on meteorology and air quality are characterized and quantified. Atmospheric aerosols during severe winter haze episodes bring along changes of incoming solar radiation, near surface temperature, PBL height and lower atmosphere stability, which is discussed in Section 4. Furthermore, these changes of meteorological variables increase or decrease near surface $PM_{2.5}$ concentrations through direct or indirect influences. For example, reductions of PBL height and stabilized lower atmosphere increase near surface PM2.5 concentrations, while lower temperature inhibits productions of secondary aerosols. Therefore, whether or not inclusions of aerosol feedbacks in the model improves model's performances in simulating severe winter haze episodes is not obvious and need to be investigated.~~

In this section we address the question whether including aerosol feedbacks within the model improves ~~model's performances~~ model performance in simulating severe haze episodes. Model results from the BASE (with all aerosol feedbacks) and EMP (without any aerosol feedbacks) scenarios are compared with observations to evaluate which scenario is more consistent with reality.

As an example to shown the extent to which simulated meteorological variables are affected by including aerosol feedbacks, Figure 12 compares downward shortwave radiation at the ground and 2 m temperature ~~between the BASE and EMP scenario~~ among the three scenarios over NCP, where $PM_{2.5}$ pollution is most severe in January 2013. ~~Both~~ All scenarios have a high bias in daily total shortwave radiation at the ground, mainly due to the overestimation of maximum shortwave radiation at noon (Wang, Z. F. et al., 2014). However, the inclusion of all aerosol feedbacks (BASE) leads to a 22% reduction of the normalized mean bias. Simulated shortwave radiation in the RAD scenario has the smallest bias.

The model prediction of 2 m temperature is also improved in the scenario with aerosol feedbacks during haze episodes, such like January 12~15 and 19~24. These findings are consistent with the ~~results in~~ those from Wang, Z. F. et al (2014), indicating the importance of including aerosol feedbacks in simulating meteorology under high aerosol loading conditions.

Figure 5 and Figure 6 compare simulated $PM_{2.5}$ over 71 big cities in January 2013 in the BASE and EMP scenarios averaged temporally and spatially, respectively. However, no significant improvements are found when aerosol feedbacks are included, partially due to ~~the missing of smaller scale~~ temporal and spatial ~~information~~ averaging. So we further investigate the ~~model's performances~~ model performance in simulating $PM_{2.5}$ over several important regions. Box plots of monthly mean $PM_{2.5}$ mass concentrations in January 2013 over NCP, YRD, PRD and CC are displayed in Figure ~~12~~ 13. Over all the four ~~areas~~ regions, the median values of hourly $PM_{2.5}$ are underestimated in the EMP scenario, in which aerosol feedbacks are excluded. Biases of the median values in the EMP scenario are −29.1%, −16.8%, −10.7%, −5.3% over NCP, YRD, PRD, and CC, respectively. Through including aerosol feedbacks, the BASE scenario improves the simulation ~~Simulation~~ of hourly $PM_{2.5}$ mass concentrations ~~distributions are improved when aerosol feedbacks are included in BASE scenario~~ in two aspects. First, biases of the median values are reduced to −22.0%, −12.0%, −6.7%, +2.6% over NCP, YRD, PRD, and CC, respectively. Second, the distribution of the middle 50% (ranging from 25th percentile to 75th percentile) hourly $PM_{2.5}$ mass concentrations is more consistent with observations than without aerosol feedbacks in the model. We also find a positive feedback for $PM_{2.5}$; that is, aerosols increase $PM_{2.5}$ through meteorological and chemical processes.

Overall in this section, we demonstrate the significance of including aerosol feedbacks in the model. Inclusions of aerosol feedbacks in the model reproduce aerosol effects on solar radiation and temperature. Thus, biases of simulated meteorology are reduced. Though the responses ~~reactions~~ of $PM_{2.5}$ to aerosol feedbacks are complex, the inclusion of aerosol feedbacks improves ~~model's performances~~ the model performance to some extent in simulating $PM_{2.5}$ in winter haze conditions.

6. Conclusions

In this work, the fully coupled on-line WRF-Chem model is applied to investigate aerosol-radiation-cloud feedbacks on meteorology and air quality over eastern China in January 2013, in which month China experienced the most severe haze pollution in history. Three simulation scenarios including different aerosol configurations are undertaken and compared.

~~Results in~~ The evaluation of the baseline simulation shows that the model ~~well~~ captures temporal and vertical variations of meteorological variables, except for overestimating lower atmosphere wind speed which is a common issue for the WRF-Chem model. The model reproduces spatial distribution of monthly mean $PM_{2.5}$ mass concentration, with high aerosol concentrations over southern Hebei, Henan, Hubei Province, Sichuan Basin and three big cities (Harbin, Changchun, Shenyang) in Northeast China. Monthly mean $PM_{2.5}$ averaged over 71 big cities is underestimated by 15%.

The model tends to underestimate $PM_{2.5}$ at the high ends, which is a common problem for current models ~~are facing with~~ in simulating severe haze conditions. Further studies ~~improving model abilities in simulating high aerosol pollution~~ are needed to improve model abilities in simulating high aerosol pollution.

Previous work indicated that the influences on air quality meteorology of aerosol indirect effects are larger than radiative effects, but this was derived under conditions with much lower aerosol loadings than those in our study. In this work we find that under winter haze conditions, aerosol radiative effects (direct effect and semi-direct effects) play a dominant role in modulating downward shortwave flux at the ground surface, lower atmosphere temperature, wind speed and PBL height. These four meteorological variables are reduced by up to 84.0 W/m^2, 3.2 ^{o}C, 0.8 m/s, and 268 m, respectively. However, aerosol indirect effects are more important than radiative effects in altering cloud properties and precipitation.

The lower PBL and ~~smaller wind speed~~ stabilized lower atmosphere result in increases of near surface CO and SO_2 concentrations. Higher aerosol loading reduces solar radiation and temperature at the surface, which results in a reduction of NO_2 photolysis rate and subsequently a reduction in a reduction of O_3 mixing ratios by up to 6.9 ppb. The aerosol feedbacks on $PM_{2.5}$ concentrations exhibit large spatial variations. Both increases and decreases of $PM_{2.5}$ are found in the domain. The enhancements of $PM_{2.5}$ over Henan, Hubei Province, and Sichuan Basin by up to 17.8 μg/m^3 are mainly due to large reduction of PBL height in these areas. The ~~suppressions~~ reduction of $PM_{2.5}$ over Bohai Sea surrounding area, Northeast China, and Southwestern China are resulted from the transition from air-borne aerosol to cloud-borne aerosol and the activation of a more comprehensive aerosol wet removal module.

The inclusion of aerosol feedback improves the model's ability in simulating downward shortwave radiation and temperature. Simulations of hourly $PM_{2.5}$ mass concentration distributions over NCP, YRD, PRD, and CC, are also improved when aerosol feedbacks are included. These indicate the importance of involving aerosol-radiation-cloud interactions in modeling air quality meteorology.

There are a number of limitations in this work. The relative coarse grid (27 km), the uncertainty of emission inventory, and the lack of secondary organic matters all contribute to the uncertainties in simulating aerosols. Also, one month length simulation could not represent a full view of aerosol-radiation-cloud feedbacks. Better understandings in the future are expected by applying more comprehensive aerosol treatments and a longer time period. Previous studies mainly focus on mechanisms of severe winter haze formation. Different from them, this work demonstrates the importance of aerosol feedbacks on meteorology and air quality during severe winter haze periods.

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Appendix

The statistical indices used in this study are defined as following.

(1) Mean Bias (MB)

$$MB = 1/N\sum_{i=1}^{N}(M_i - O_i)$$

(2) Normalized Mean Bias (NMB)

$$NMB = \frac{\sum_{i=1}^{N}(M_i - O_i)}{\sum_{i=1}^{N}O_i} \times 100\%$$

(3) Correlation Coefficient (Corr. R)

$$Corr.R = \frac{\sum_{i=1}^{N}(M_i - \overline{M})(O_i - \overline{O})}{\sqrt{\sum_{i=1}^{N}(M_i - \overline{M})^2}\sqrt{\sum_{i=1}^{N}(O_i - \overline{O})^2}}$$

(4) Root Mean Square Error (RMSE)

$$RMSE = \sqrt{1/N\sum_{i=1}^{N}(M_i - O_i)^2}$$

where $\overline{M} = 1/N\sum_{i=1}^{N}M_i$, $\overline{O} = 1/N\sum_{i=1}^{N}O_i$, $\overline{M}$ and $\overline{O}$ are model result and observation for sample i, respectively. N is the number of samples.

(5) The Student t statistic

$$t = \frac{\overline{X}_1 - \overline{X}_2}{S_{X_1X_2}\sqrt{1/n_1 + 1/n_2}}$$

$$S_{X_1X_2} = \sqrt{\frac{(n_1 - 1)S_{X_1}^2 + (n_2 - 1)S_{X_2}^2}{n_1 + n_2 - 2}}$$

Here, $S_{X_1X_2}$ is the grand standard deviation, $S_{X_1}^2$ and $S_{X_2}^2$ are the variances of the two samples.

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Table 1. Summary of three simulated scenarios.

CASE name	Characteristics
BASE	With all aerosol feedbacks
RAD	Only with aerosol direct and semi-direct effects
EMP	Without any aerosol feedbacks

Table 2. Statistical Performance of baseline simulations for meteorology.

	T2 (°C)	**RH2 (%)**	**WS10 (m/s)**	**PM2.5 (μg/m³)**
N of stations	523	523	523	71
Mean OBS	−1.8	66	1.9	129.2
Mean SIM	−2.8	66	3.9	111.5
Corr. R	0.96	0.47	0.47	0.67
MB	−1.0	0	2.0	−18.9
NMB	~~−83.3%~~ −0.4%*	<0.1%	105%	−15.0%
RMSE	3.4	16	2.7	30.7

* Calculated in K.

Table 3. Observed and simulated enhancement ratios of $PM_{2.5}$. The enhancement ratio is defined as the average of ~~hourly~~ daily $PM_{2.5}$ larger than the median value divided by that of hourly $PM_{2.5}$ less than the median value during the month. NCP, YRD, PRD, and CC represent the North China Plain, the Yangtze River Delta, the Pearl River Delta, and Central China, respectively.

	NCP	YRD	PRD	CC
Observations	~~1.8~~ 1.7	~~1.7~~ 1.6	~~1.7~~ 1.5	1.7
WRF-Chem (BASE)	~~1.8~~ 1.6	~~2.0~~ 1.7	~~1.8~~ 1.6	~~2.0~~ 1.7

Table 4. Modeled SOA, OA and $PM_{2.5}$ using MADE/SORGAM and MOSAIC in comparison with observations. The data shown are spatially averaged over 71 big cites and temporally averaged during January 6~10, 2013.

	SOA (μg/m^3)	**OA (μg/m^3)**	**SOA/OA (%)**	**$PM_{2.5}$ (μg/m^3)**	**NMB of $PM_{2.5}$ (%)**
MADE/SORGAM	0.3	9.4	2.8	68.3	49.7
MOSAIC				115.4	15.0
Observations				135.7	

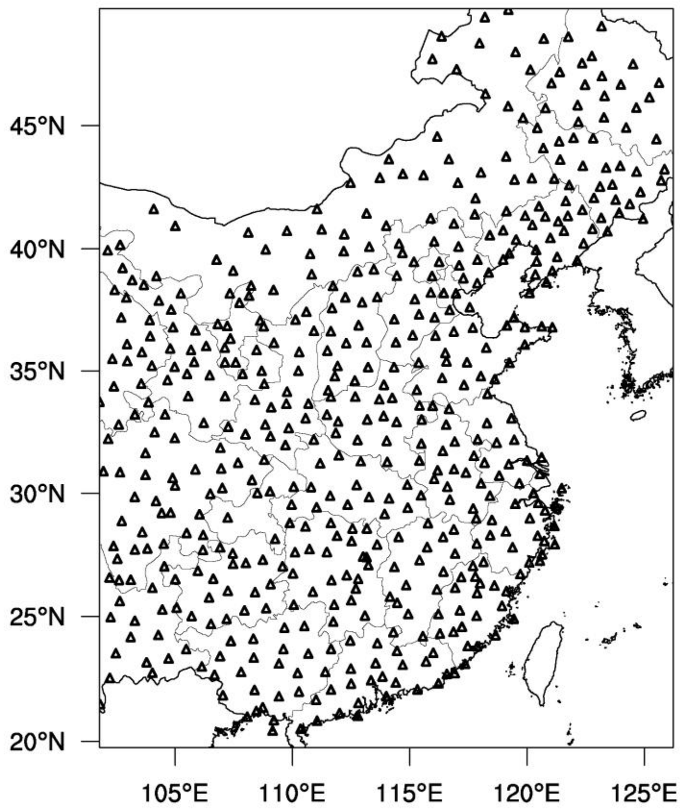

Fig 1. WRF/Chem modeling domain with grid resolution of 27 km. The domain covers eastern parts of China. The triangles indicates the location of 523 meteorology stations used for evaluations in this work.

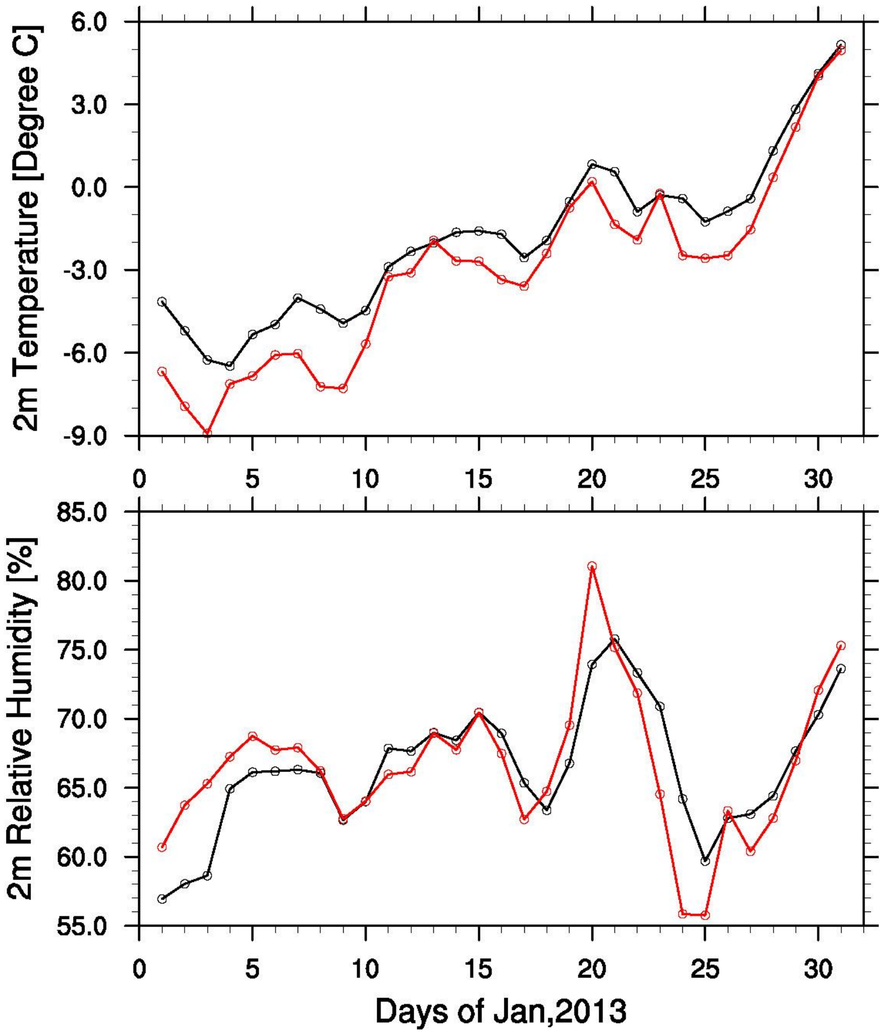

Fig 2. Time series of observed (black line) and simulated (red line) daily meteorological variables averaged over 523 meteorology stations in January 2013.

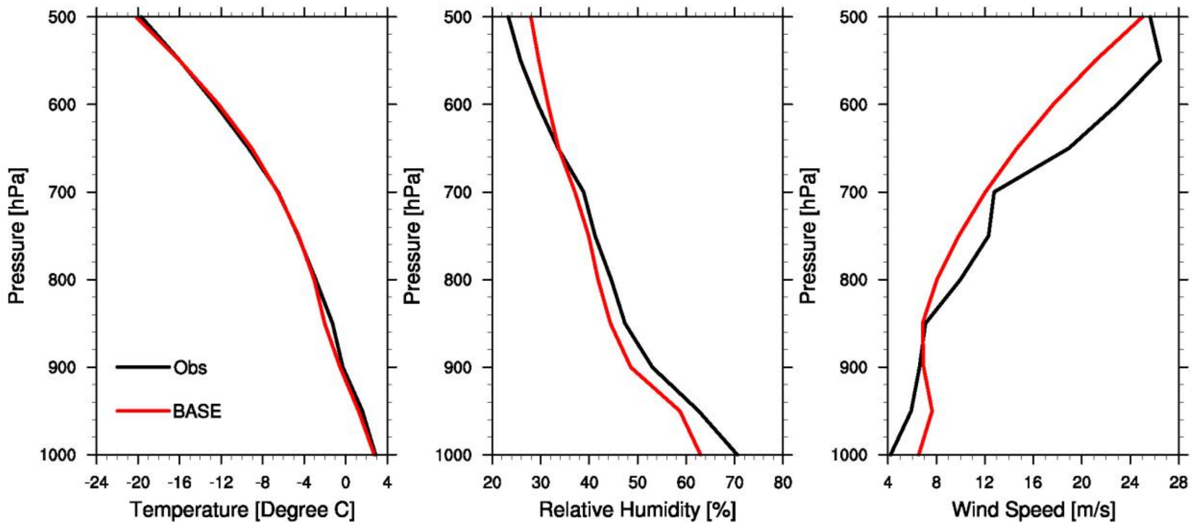

Fig 3. Monthly mean vertical profiles of observed (black line) and simulated (red line) meteorological variables averaged over 36 meteorology stations.

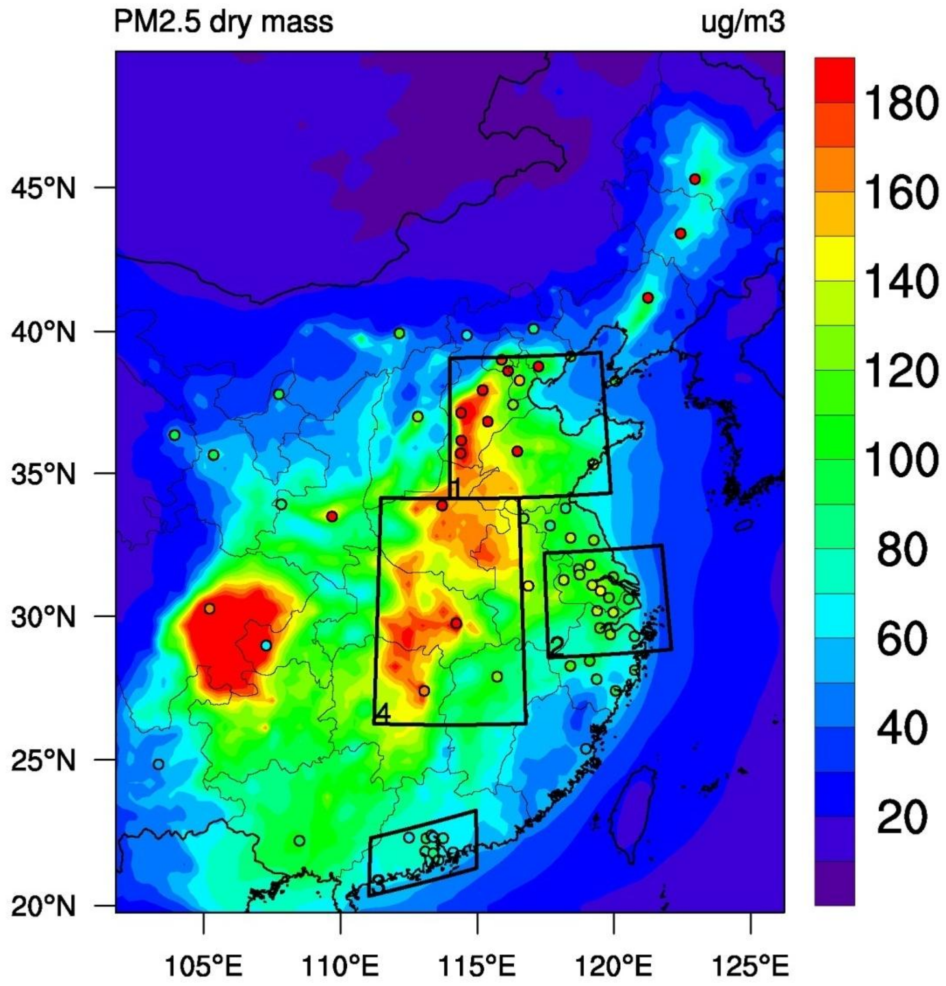

Fig 4. Simulated and Observed (circles) monthly mean $PM_{2.5}$ mass concentration over eastern China in January 2013. The four polygons stands for the North China Plain (NCP) (#1), the Yangtze River Delta (YRD) (#2), the Pearl River Delta (PRD) (#3), and Central China (#4).

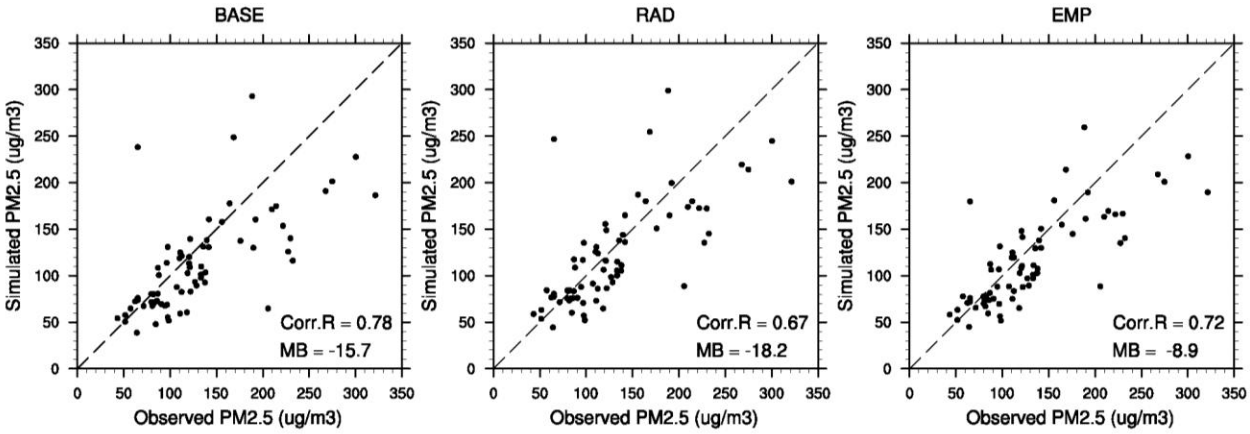

Fig 5. Scatter plots of monthly mean $PM_{2.5}$ mass concentrations in 71 cities in January 2013.

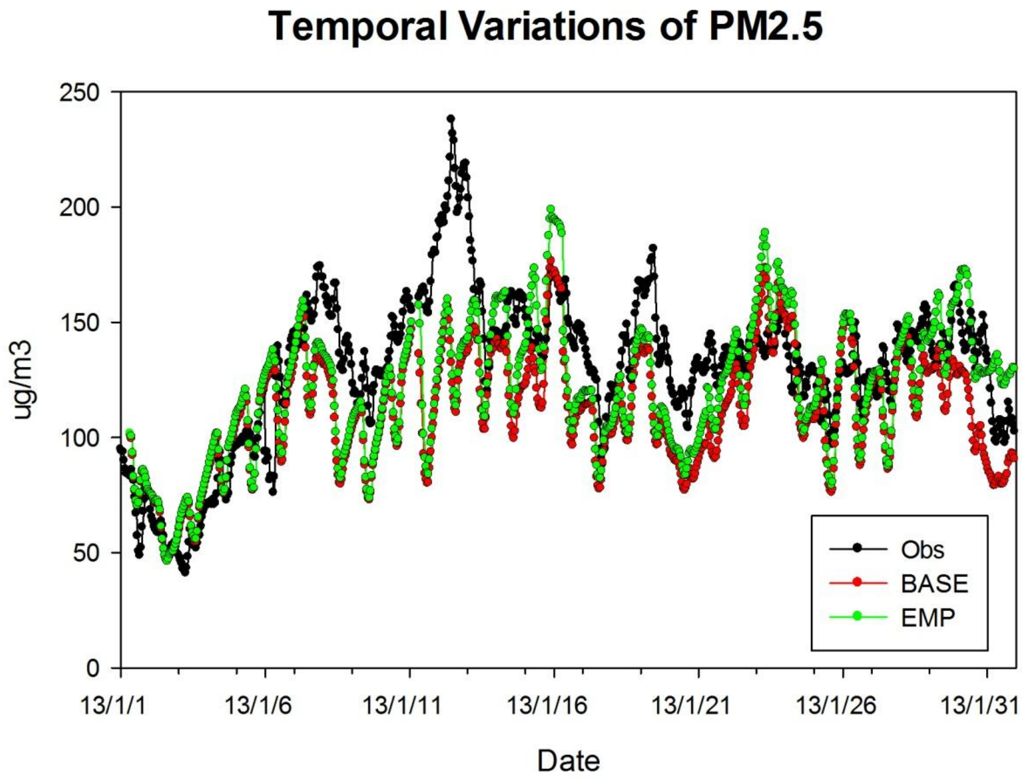

Fig 6. Comparison of observed (black) and simulated (red for the BASE scenario and blue for the EMP scenario) hourly near surface $PM_{2.5}$ mass concentrations averaged over 71 cities in China in January 2013.

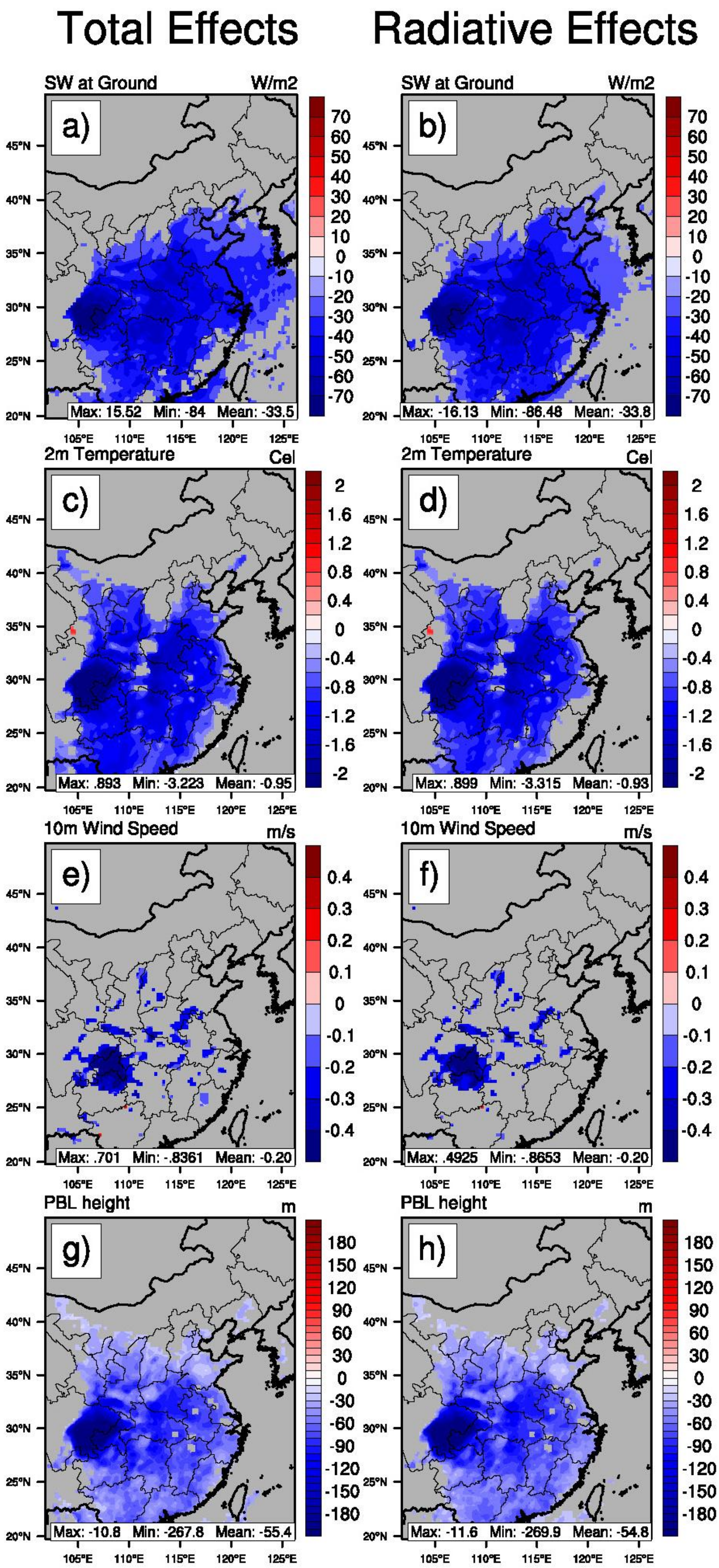

Fig 7. Simulated aerosol total effects (BASE − EMP) and radiative effects (RAD − EMP) on downward short wave flux at ground, 2m temperature, 10 m wind speed and PBL height in January 2013.The aerosol indirect effects on these four meteorological variables are not shown here, since the induced changes are not significant according to Student's t-test. Grey shaded areas indicate regions with less than 95% significance.

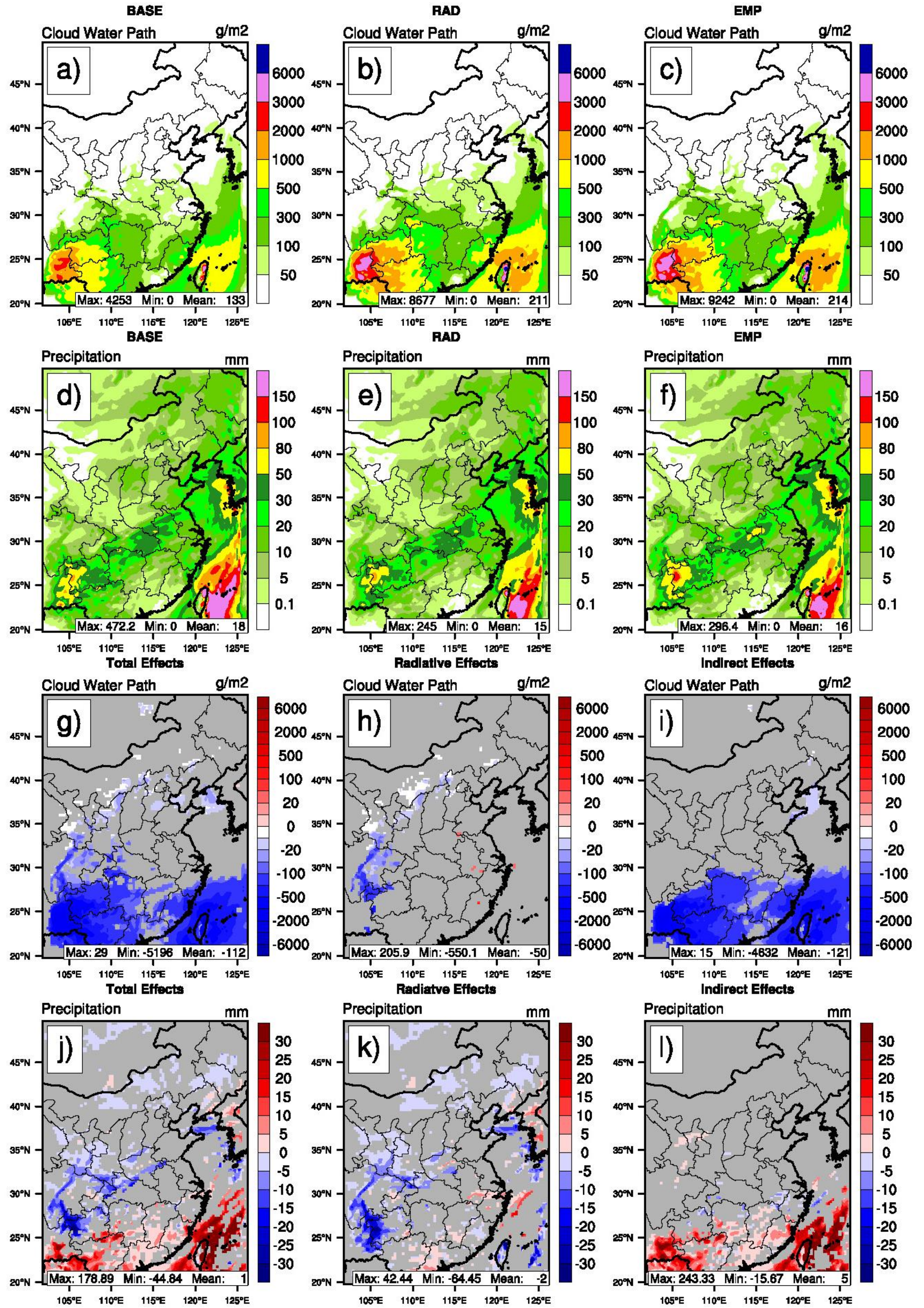

Fig 8. Simulated cloud water path (a-c) and precipitation (d-f) for the three scenarios and aerosol total effects (BASE − EMP) (g and j), radiative effects (RAD − EMP) (h and k), and indirect effects (BASE − RAD) (i and l) over eastern China in January 2013. Grey shaded areas indicate regions with less than 95% significance.

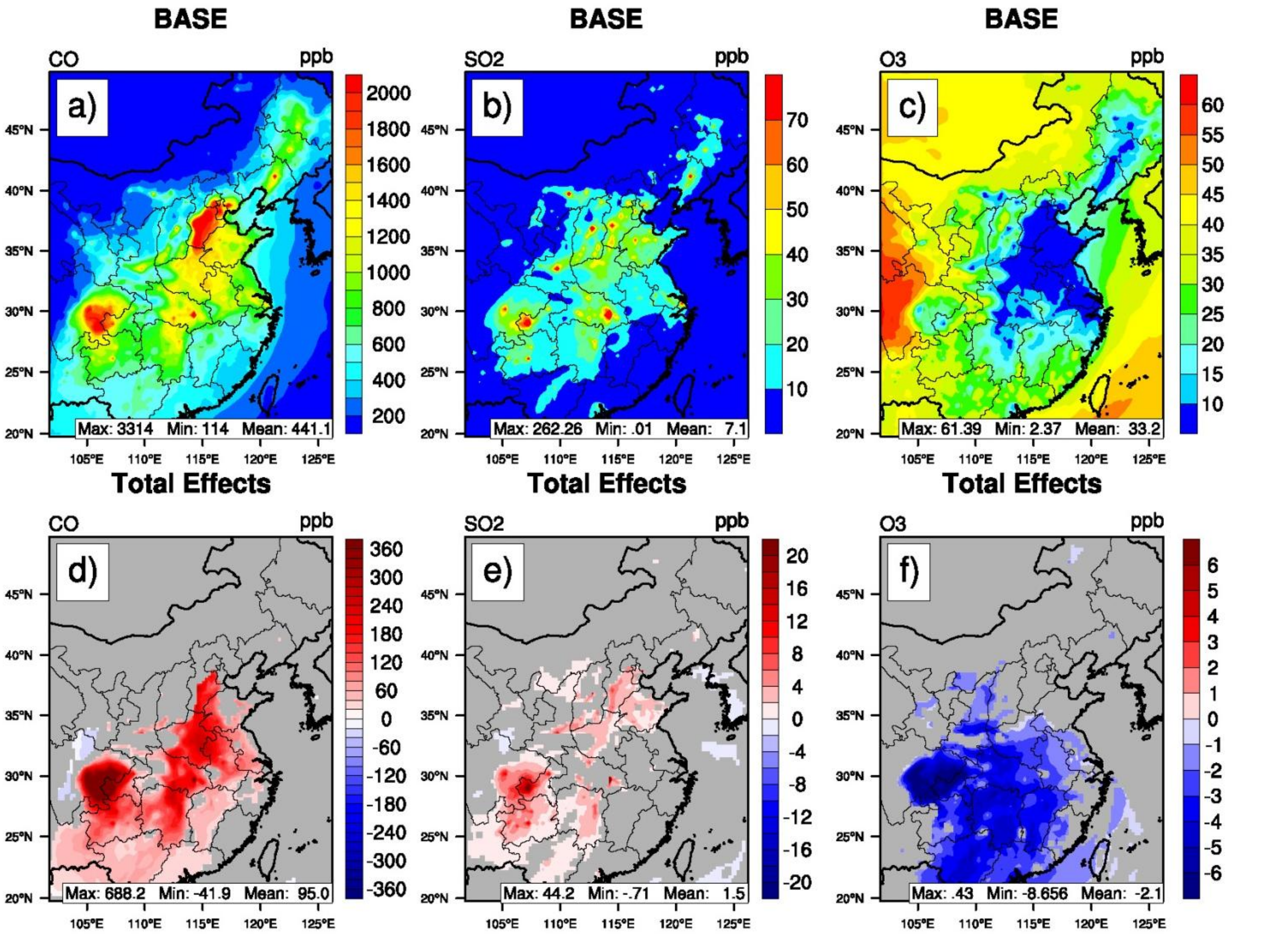

Fig 9. Simulated monthly mean CO, SO_2, and O_3 mixing ratios and aerosol feedbacks (BASE − EMP) on the three gas pollutants over eastern China in January 2013. Grey shaded areas indicate regions with less than 95% significance.

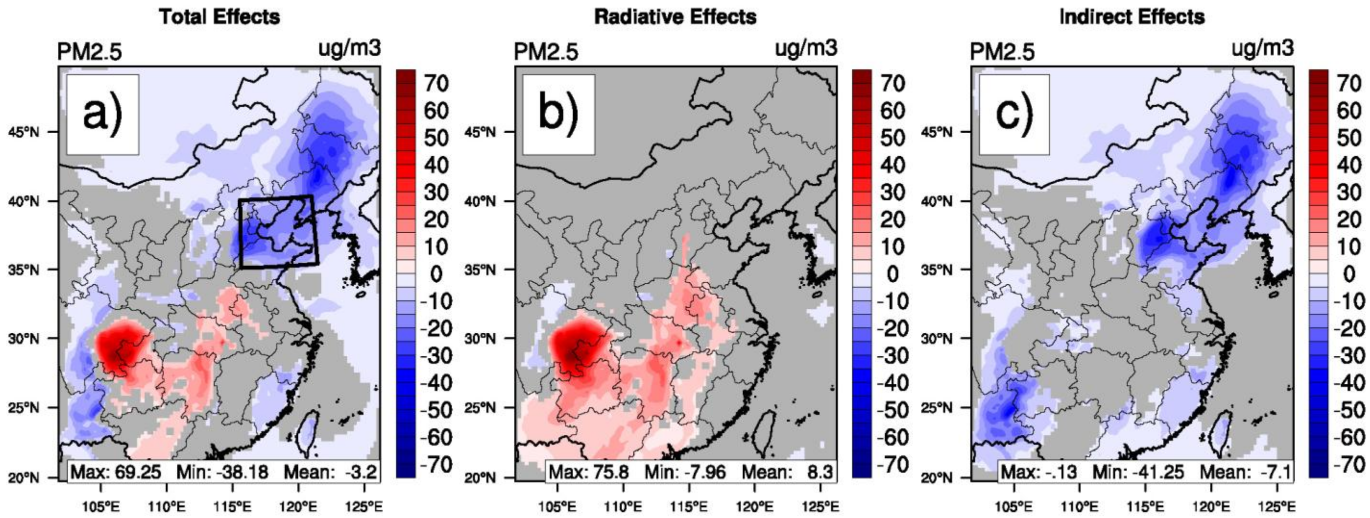

Fig 10. Simulated aerosol total effects (BASE − EMP), radiative effects (RAD − EMP), and indirect effects (BASE − RAD) on monthly mean $PM_{2.5}$ over eastern China in January 2013. Grey shaded areas indicate regions with less than 95% significance. The black polygon defines the Bohai Sea surrounding area.

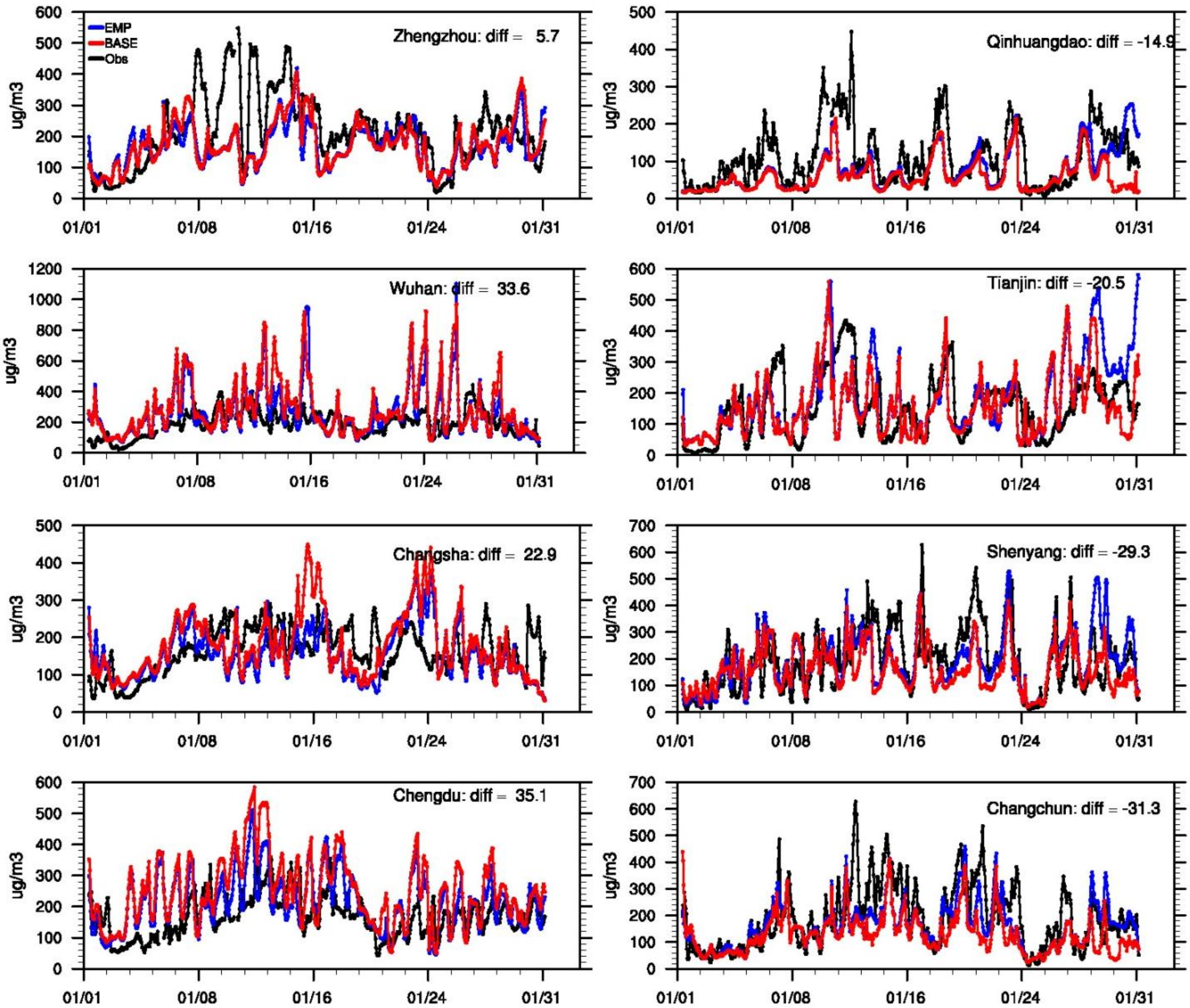

Fig 11. Time series of observed (black) and simulated (red for BASE scenario and blue for EMP scenario) hourly surface $PM_{2.5}$ mass concentrations in 8 cities in January 2013. Monthly mean $PM_{2.5}$ are enhanced in the four cities (Zhengzhou, Wuhan, Changsha and Chengdu) in the left column. Cites in the right column have suppressed monthly mean $PM_{2.5}$ mass concentrations. "Diff" indicates aerosol feedbacks (BASE − EMP) on monthly mean $PM_{2.5}$ mass concentrations.

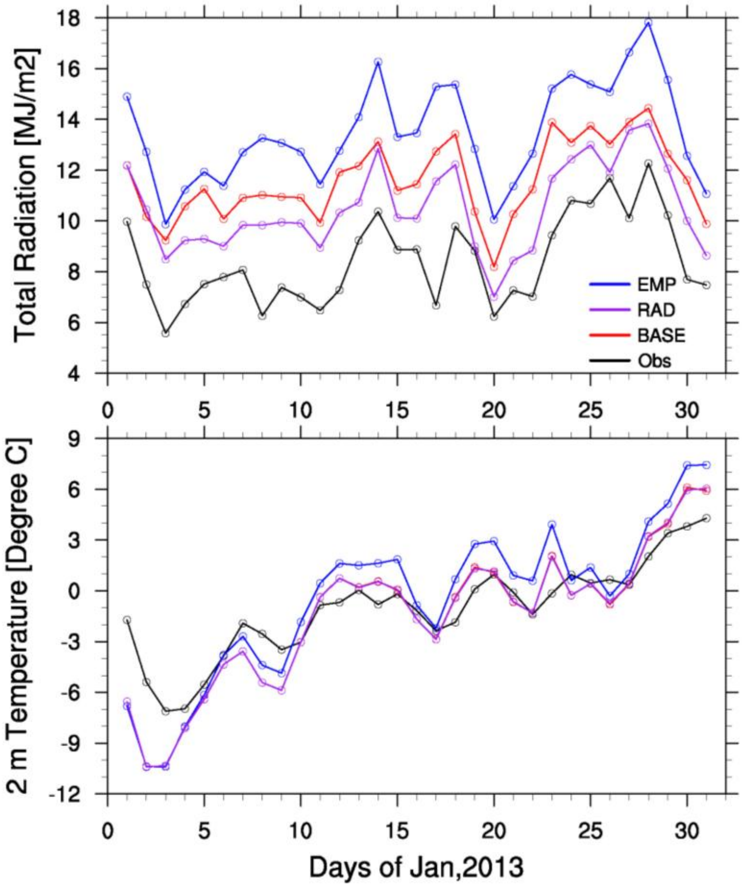

Fig 12. Time series of observed (black) and simulated (red for the BASE scenario, purple for the RAD scenario, and blue for the EMP scenario) daily total radiation and 2 m temperature averaged over North and central China in January 2013.

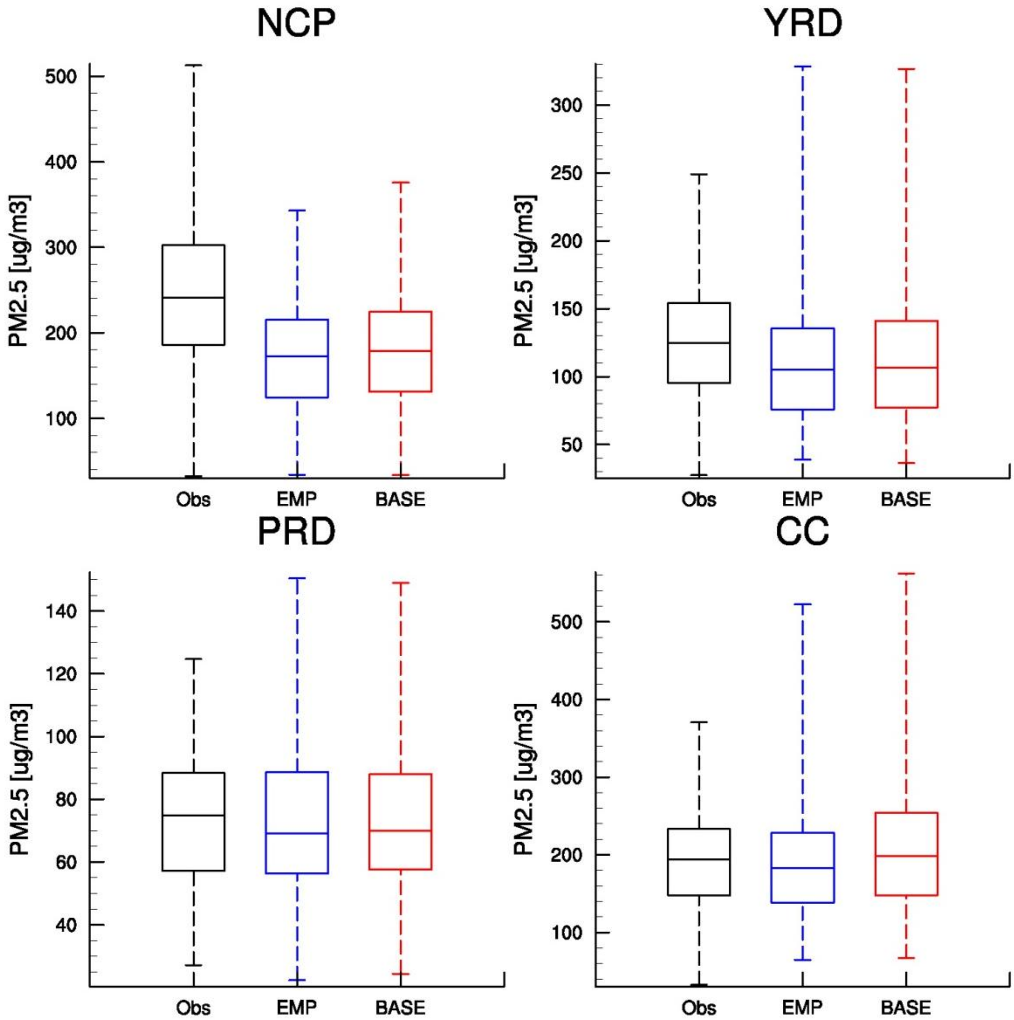

Fig 13. Observed (black) and Simulated (red for the BASE scenario and blue for the EMP scenario) monthly mean $PM_{2.5}$ mass concentrations in January 2013 over the North China Plain (NCP), the Yangtze River Delta (YRD), the Pearl River Delta (PRD) and Central China (CC). The dashed lines indicate the maximum and minimum value. The solid lines in the box indicate the median value (the central line), the 25th and 75th percentiles.