November 03, 2017

Dear Editor,

We have received the comments from the two reviewers of the manuscript. Below are our responses and the revisions that we have made in the manuscript.

Thank you for your efforts on this manuscript. We look forward to hearing from you.

Best Regards,

Guohui Li

Reply to Anonymous Referee #1

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

This manuscript addresses a topical and important issue in aerosol researches, i.e. how to improve the simulation and forecast of pollution levels in severe haze events. The influence of meteorological conditions in the regional WRF-CHEM model is explored by employing an ensemble simulation approach with perturbed initial and boundary atmospheric fields. The finding of the substantial sensitivity of the simulated $PM_{2.5}$ concentrations to meteorological conditions on at the city scale highlights the importance of the ensemble approach in model assessment of air quality. Overall, the paper is well written, and I only have some minor comments for the authors to address.

1 Comment: The ensemble method needs to be clarified, as some technical details are not described clearly.

Response: We have clarified the ensemble method in Section 2 as suggested.

1.1 Comment: For example: What variables are perturbed in this method? A list of perturbed fields can be more informative. Are those variables equally important in the contribution to the uncertainty?

Response: We have clarified in Section 2: "*The perturbed variables include the horizontal wind components, potential temperature, perturbation pressure, and mixing ratio of water vapor. Other prognostic variables such as vertical velocity and mixing ratios of hydrometeors are not perturbed. In general, the perturbation in horizontal wind components constitute the most important uncertainty in those variables (Bei et al., 2008).*"

1.2 Comment: L101, the word "perturbations" comes from nowhere. Please clarify how to obtain them.

Response: We have changed "perturbations" to "members".

1.3 Comment: L102-105, it seems that the boundary conditions are different in each ensemble member as well. However, the authors only mention "initial meteorological uncertainties" in the later discussions.

Response: We have removed "initial" in the later discussions because we have perturbed both the meteorological initial and boundary conditions, as the reviewer pointed out.

1.4 Comment: L111, what is the "average spread", and what is the "mean of ensemble spreads" in Fig. 2? To my understanding, the spread is calculated as the standard deviation of the perturbations imposed on each ensemble member's initial field.

Response: We agree to the reviewer's comment and have clarified in Section 2: "*Figures 2a–d show the vertical distribution of the average initial ensemble spread which is calculated as the standard deviation of the perturbations imposed on each ensemble member's initial field.*"

2 Comment: Readers may remain curious about how important the meteorological impacts on PM simulations are, compared to other uncertainties in the WRF-CHEM model, e.g. emission, or different chemistry/aerosol schemes. Is there a way to quantify the relative importance of each uncertainty source in the model?

Response: We have clarified in Section 4: "*It is worth noting that aside from meteorological fields, uncertainties in emissions or various chemistry/aerosol schemes, also considerably influence the WRF-CHEM model simulations. The extended response surface modeling (ERSM) technique can be used to quantify the relative importance of each uncertainty source in the WRF-CHEM model (Zhao et al, 2017).*"

3 Comment: The quality of figure can be further improved: - Figure 2, better to provide the percentage information as well. An ideal way to illustrate the ensemble experiment is to show the PDF of each quantity using color-coded contour plot. - Figure 3, too busy. Suggest having one panel for each city.

Response: Since the mean of ensemble meteorological fields might be close to zero, causing the very large percentage change, we have included the standard deviation of the initial ensemble spread and replotted Figure 2. For Figure 3, considering more than 10 cities, we have divided the cities in BTH into megacites and non-megacities and replotted it.

4 Comment: A grammar check is needed: - L354, “A climatological . . .”; remove “the” before “initial” - L356, “are generally in good agreement”, as ENSM here should refer to the means for each aerosol species. - L359, is it “primary aerosols”? - L372, “measurements” - L375, remove “the” before “ensemble”; replace “avoid” by “minimize”.

Response: We have corrected the grammatical errors and typos as suggested.

Reply to Anonymous Referee #2

We thank the reviewer for the careful reading of the manuscript and helpful comments. We have revised the manuscript following the suggestion, as described below.

This paper evaluates the impact of uncertainties in aerosol simulations due to the meteorological initial conditions in WRF-Chem. The study appears to be thorough and the subject is timely. I am happy to recommend publication in Atmospheric Chemistry & Physics subject to minor revisions.

Minor Comments: The paper would benefit from additional proof reading. In particular, the abstract could be clarified and improved.

Response: We have polished the English of the manuscript as suggested.

1 Comment: Line 16: "have shown" suggests previous work – I don't think that is what is intended here?

Response: We have changed "have shown" to "show".

2 Comment: Line 20-23: This seemed confusing. What does the 30% refer to?

Response: The 30% refer to the ratio of the ensemble spread to ensemble mean. We have rephrased the sentence as: "*and the RMSE for simulated $PM_{2.5}$ concentrations can be up to 30% at the region scale*"

3 Comment: Line 24: Do you mean "evaluation" rather than "implementation" – the uncertainties are to do with analysis, not with actually doing something on the ground.

Response: Yes, "implementation" should read "evaluation", and we have changed "implementation" to "evaluation".

4 Comment: Line 26: Needs rephrasing: we cannot avoid the impact, but we can take it into account.

Response: We have rephrased the sentence as suggested: "*Therefore, our results suggest that the ensemble simulation is imperative to take into account the impact of the initial meteorological uncertainties on the haze prediction.*"

5 Comment: Line 98: Please give a summary of the method so that the reader can get a sense without looking up the references.

Response: We have clarified in Section 2: "*The ensemble initialization method used in the present study is called "climatological ensemble initialization method" (Zhang et al., 2007; Bei et al. 2012). In the approach, dynamically consistent initial and boundary conditions are statistically sampled from a seasonal meteorological data set. In order to represent the wintertime climatological statistics, a data set during the period from 1 November 2013 to 28 February 2014 is generated using NCEP-FNL 1°×1° reanalysis data. The perturbed variables include the horizontal wind components, potential temperature, perturbation pressure, and mixing ratio of water vapor. Other prognostic variables such as vertical velocity and mixing ratios of hydrometeors are not perturbed. In general, the perturbation in horizontal wind components constitute the most important uncertainty in those variables (Bei et al., 2008, 2010). Thirty ensemble members are randomly chosen from this climatological data set. Similarly, boundary conditions for each ensemble member are generated from the same data set beginning at the randomly selected initial time of the given member, and extended for the same length of time as the simulated episode. Deviations of the initial and boundary condition data for each member from the climatological mean for the entire period are then scaled down to be 20% to reduce the ensemble spread to be less than typical observation error magnitudes (Nielsen-Gammon et al., 2007) and added to the unperturbed initial and boundary conditions derived directly from the NCEP-FNL analyses valid at 12:00 UTC on 12 January 2014, which are used for the 6-km domain ensemble simulation.*"

6 Comment: Line 181: the "clod" clean air?? Line 374 and 375: rephrase, as for Line 26.

Response: We have changed "clod" to "cold" and rephrased the sentence as suggested: "*Therefore, the ensemble simulation is needed to take into consideration the impact of the meteorological uncertainties on the haze prediction.*"

6 Comment: Fig 2: Could you show the spread of the ensemble initial conditions, for example with a boxplot?

Response: Since there are thirty-four layers in the vertical direction, when the spread of the ensemble initial conditions is shown using the boxplot, the figure looks rather busy. Therefore, we have included the standard deviation of the initial ensemble spread and replotted Figure 2.

7 Comment: Fig 3: This is rather busy. I think a legend and possibly splitting the graph by geographical area would help.

Response: We have divided the cities in BTH into megacites and non-megacities and replotted Figure 3 as suggested.

Author's changes in the manuscript

We have included several co-authors in the manuscript as follows:

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