

Manuscript: egosphere-2023-2615

Title: Impact of Meteorology and Aerosol Sources on PM_{2.5} and Oxidative Potential Variability and Levels in China

We appreciate all the valuable comments from the reviewers and editor, which significantly improved the quality of the manuscript. We have studied the comments carefully and revised the manuscript accordingly. The comments and our responses point-to-point are listed below. Changes to the paper are shown in blue, so that the reviewers can easily review them again.

Replies to Reviewers and Editor

First of all, we thank both reviewers and editor for their positive and constructive comments and suggestions.

Reviewers' comments:

Anonymous referee #2:

I appreciate that the authors have addressed my comments well. There are a few minor word changes that I would like to suggest before the paper is accepted by the journal.

1. Thank you for providing information of high versus low values of OP. I recognize that there is not an agreed upon number above which is high or low, but it would help the reader if the paper gave some context of previously known values. Therefore, please revise the sentence in lines 28-29 of abstract to the following.

Furthermore, the probability density function revealed that about 40% of areas in China had an annual average PM_{2.5} concentrations exceeding the Chinese concentrations limit. For OP, 36% of the regions have OP below 1 $nmol\ min^{-1}\ m^{-3}$, 41% have OP between 1 and 2 $nmol\ min^{-1}\ m^{-3}$, and 23% have OP above 2 $nmol\ min^{-1}\ m^{-3}$, which are in line with previous measurement studies.

Response: Thank you very much for your constructive comments. Following your suggestion, we incorporated the recommended content into lines 28-29 of abstract.

The detailed revisions in the manuscript are shown below:

Abstract (Lines 28-29):

“Furthermore, the probability density function revealed that about 40% of areas in China had an annual average PM_{2.5} concentrations exceeding the Chinese concentrations limit. For OP, 36% of the regions have OP below 1 $nmol\ min^{-1}\ m^{-3}$, 41% have OP between 1 and 2 $nmol\ min^{-1}\ m^{-3}$, and 23% have OP above 2 $nmol\ min^{-1}\ m^{-3}$, which are in line with previous measurement studies.”

2. Line 47. I found the response to my first major comment, where the authors provide OP results from different locations and season to be very useful. However, I did not see those sentences in the revised manuscript. It would be good to add that information before the last sentence of the first paragraph of the Introduction.

Response: Thank you very much for your constructive comments. Following your suggestion, we have added the OP results from different locations and season in the first paragraph of the Introduction.

The detailed revisions in the manuscript are shown below:

Introduction (Lines 45-49):

“Liu et al. (2020) summarized OP measurements conducted in nine regions of China around 2014. The results

showed that the average OP content in northern Beijing was highest during the winter of 2016 ($\sim 14.0 \text{ nmol min}^{-1} \text{ m}^{-3}$), while the average OP level in Shanghai during the spring of 2016 was lowest ($\sim 0.15 \text{ nmol min}^{-1} \text{ m}^{-3}$). However, there is currently no exact threshold division of OP values.”

Liu, Q., Lu, Z., Xiong, Y., Huang, F., Zhou, J., Schauer, J.J. (2020). Oxidative potential of ambient PM_{2.5} in Wuhan and its comparisons with eight areas of China. *Sci Total Environ*, 701, 134844. <https://doi.org/10.1016/j.scitotenv.2019.134844>.

3. Line 168. I now understand that DEHM does not include aerosol impacts on radiation or clouds. Please state this in the sentence here. For example:

The current version of the DEHM model does not include wind-blown, resuspended dust emissions or road dust nor aerosol-radiation or radiation-cloud interactions.

As a side note, the high PM_{2.5} concentrations in the PBL affects the amount of solar radiation reaching the surface. Consequently, surface temperatures and PBL height are modified. Further, the aerosols affect the photolysis rates of the trace gas chemistry, impacting oxidant concentrations and therefore the yield of secondary aerosol formation. This information does not need to be included in the manuscript, but it illuminates areas of model improvement that can further refine estimates of OP.

Response: Thank you very much for your constructive comments. Following your suggestion, we have added the content that the current version of the DEHM does not include aerosol impacts on radiation or clouds.

The detailed revisions in the manuscript are shown below:

Materials and methods (Lines 172-174):

“The current version of the DEHM model does not include wind-blown, resuspended dust emissions or road dust nor aerosol-radiation or radiation-cloud interactions.”

4. Line 215. Please remove “abs represents the absolute value.”

Response: Thank you very much for your constructive comments. Following your suggestion, we have removed “abs represents the absolute value.”

The detailed revisions in the manuscript are shown below:

Materials and methods (Lines 222-224):

“Con(Emi) represents the impact of changing emission inventory on changes in PM_{2.5} and OP. NCon(Met) and NCon(Emi) represent the normalized contributions of meteorology and emission. In the equations, the abs function represents the absolute value of the quantity in parentheses.”

5. Line 219. Please remove “abs represents the absolute value.”

Response: Thank you very much for your constructive comments. Following your suggestion, we have removed “abs represents the absolute value.”

The detailed revisions in the manuscript are shown below:

Materials and methods (Lines 222-224):

“Con(Emi) represents the impact of changing emission inventory on changes in PM_{2.5} and OP. NCon(Met) and NCon(Emi) represent the normalized contributions of meteorology and emission. In the equations, the abs function represents the absolute value of the quantity in parentheses.”

6. Line 231. Please revise the sentence to say the following.

The emissions of both primary aerosols and tracegases from each individual source are reduced by 30%.

Response: Thank you very much for your constructive comments. Following your suggestion, we have modified "The emission from each individual source is reduced by 30%." to " **The emissions of both primary aerosols and**

tracegases from each individual source are reduced by 30%." (Please see Lines 235-236)

7. Line 311. Please revise the sentence to read more easily.

Additionally, this study evaluated the model performance in scenarios C₂ and C₃ (Figure S2).

Response: Thank you very much for your constructive comments. Following your suggestion, we have modified " Additionally, this study also evaluated the model performance in scenarios C₂ and C₃, as illustrated in Figures S2." to " **Additionally, this study evaluated the model performance in scenarios C₂ and C₃ (Figure S2).** " (Please see Lines 314-315)

8. Line 312-313. Please remove the following sentence.

Figures S2a (c) and S2b (d) depicted density scatter plots of model performance and evaluation in scenarios C₂ (C₃) based on annual mean observations and the Dalhousie dataset, respectively.

Response: Thank you very much for your constructive comments. Following your suggestion, we have removed "Figures S2a (c) and S2b (d) depicted density scatter plots of model performance and evaluation in scenarios C₂ (C₃) based on annual mean observations and the Dalhousie dataset, respectively."

The detailed revisions in the manuscript are shown below:

Results and discussion (Lines 314-317):

"Additionally, this study evaluated the model performance in scenarios C₂ and C₃ (Figure S2). It was found that under scenarios C₂ and C₃, the model performance in terms of R and NME, calculated based on both annual mean observations and Dalhousie dataset met the performance criteria suggested by Emery et al. (2017)."

9. Line 318. The sentence needs a plural verb.

Therefore, the simulated annual mean PM_{2.5} concentrations in scenarios C₁, C₂ and C₃ are considered reliable.

Response: Thank you very much for your constructive comments. Following your suggestion, we have modified " Therefore, the simulated annual mean PM_{2.5} concentrations in scenarios C₁, C₂ and C₃ is considered reliable." to " **Therefore, the simulated annual mean PM_{2.5} concentrations in scenarios C₁, C₂ and C₃ are considered reliable.**" (Please see Lines 319-320)

10. Figure 4. I realize that my confusion before on the labeling of the Figure 4 panels, i.e. the difference of simulation minus observations, was caused by red colors being negative and blue being positive. Normally it is the other way.

Response: Thank you very much for your constructive comments. Thank you for your reminder, and to avoid any misunderstandings, we have swapped the color bars in Figure 4 so that red colors show high values and blue colors show low values.

The detailed revisions in the manuscript are shown below:

Results and discussion (Lines 343-345):

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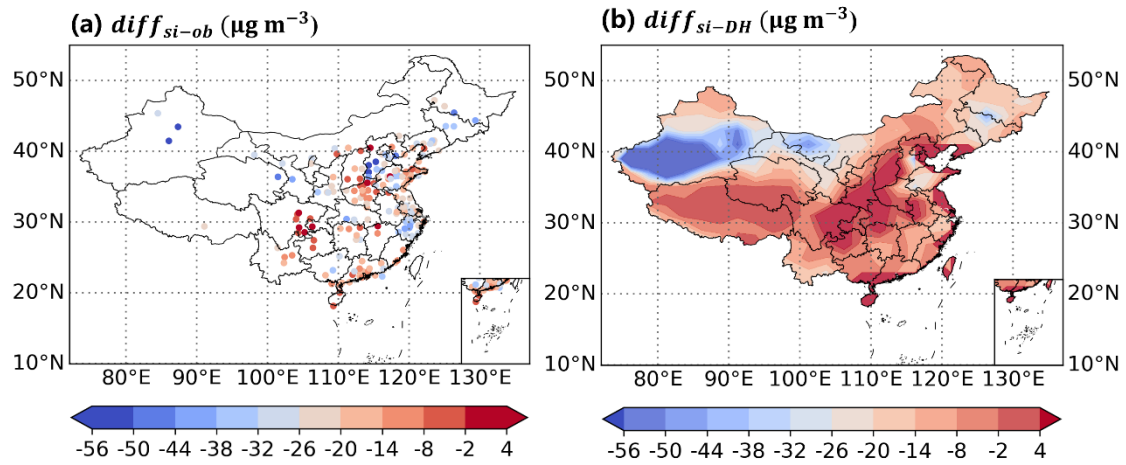


Figure 4. Spatial distribution of the annual mean simulated minus annual mean observed values (a), as well as the spatial distribution of the annual mean simulated values minus the Dalhousie dataset (b) for China in 2014 under scenario C₁.

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