Part 1. point-by-point response to the reviews

reviewer 1

Interactive comment on "The Effects of El Niño-South Oscillation on the Winter Haze Pollution of China" by Shuyun Zhao et al.

Anonymous Referee #1

Received and published: 24 July 2017

The manuscript explores the potential for the ENSO to have influenced the winter haze days and contents of anthropogenic aerosols in China. The observational data and the numerical simulations that was designed to isolate the impacts of ENSO point to a contrasting effect between El Nino and La Nina conditions on changes in the haze days and aerosol concentration over the southern China. However, the analysis and presentation shown in this paper, I think, have several flaws. Therefore, the revision is required to improve the reliability of the results reported.

General comments

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1.Statistical significance: The significance of results has not been discussed except for Fig.4. I recommend the authors show the significance in the figures. Because the authors have conducted multiple ensemble experiments, I think it is possible to do so.

Reply: We used the median instead of the ensemble mean of the differences of variables caused by El Niño or La Niña, as median is more robust and resistant to extreme values. And we also added markers to those grids where the differences of a specific variable between more than 70% pairs of ensemble members have the same sign with the median differences, as a reflection of significance. Please see the last paragraph of section 2.2.

2.Use of emission data for the year of 2010: The aerosol emissions had a long-term trend, the decadal variability, and regional deference. So, I would encourage the authors to check and argue the results for the other years.

Reply: Thank you for the suggestion. Actually, the effects of ENSO on the haze pollution of China under different emissions (even unrealistic large emissions) are parts of what we are considering to do in our next work.

3.My understanding is that the effects of ENSO on the haze pollution (or haze seasonal prediction) in southern China depend on the changes in aerosol transport and precipitation associated with changes in the SST patterns. However, I am very confused about the contrasting features of "haze days" and "aerosol concentrations" (Fig. 4 and Fig. 9, P1. L19-20, P12. L16-18). I have several questions. Which is a larger damage by haze pollution in El Nino years (less haze days but heavy) or La Nina years (many haze days but moderate) eventually? If it were less precipitation in El Nino years, one can assume that winter haze days in El Nino years increase?

Reply: It is very hard to tell which is more damaging, less haze days but some of them are very heavy or more haze days but they are generally moderate. This manuscript only discussed a phenomenon. The social and economic effects of haze pollution is a little beyond our research field. Personally, I think we cannot count on a natural circulation to release our suffering from haze pollution, instead we should reduce the emissions of aerosols and their precursors.

Precipitation enhances the wet deposition of particles, which is an important way of cleaning particles out of the atmosphere.

In southern China, it was found that there were more precipitation and less haze days in El Niño winters. The changes in precipitation and haze days correspond with each other very well. But this correspondence may not be good over other places. For example, in northern China, there are always a few clean days after an outbreak of a block in the high latitude, even without rain in winters, because strong northerly wind blows the haze pollutants away from northern China. That is why many studies related the winter haze events of northern and eastern China to Arctic sea ice and Eurasian snow cover. So, we can say that precipitation facilitates the reduction of haze days, but at the same time we must also check other meteorological variables for

Specific comments

a specific region.

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1. P3. L13: According to the CAM definition of the haze, relative humidity (<80%) is included in it. Dose this definition implies that the haze days are hard to be counted in the El Nino years because of the rainy years?

Reply: For a long time, meteorological stations use relative humidity to distinguish haze and fog when visibility is low (< 10 km), before the large-scale installation of precise instrument that can detect particle matters. The reason is that when humidity

is very high, it is easy for particles to activate as the cores of fog droplets. We do agree that there might be possibilities to mistake haze to fog, or vice versa, especially when the relative humidity is around the threshold (80%). That's why we mentioned in the manuscript that "Another reason probably cannot be neglected that drier conditions over southern China during La Niña winter can avoid mistaking haze to fog days". However, though maybe with some errors, the CMA haze-day dataset is still an authorized data set with long time period and large spatial coverage.

2. Figs. 2,3: I ask the authors to show separately the haze days during the El Nino or La Nina years.

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- Reply: In the last more than 50 years, the emissions of haze particles and their precursors have experienced dramatic variations, which can blanket the differences between the haze days in El Nino and La Nina winters. So, we selected 4 pairs of El Nino and La Nina winters, with the time interval of each pair ≤ 2 years, in order to diminish the influence of emission variations as much as possible. Please see the revision and the Figure S2 in the supplementary material.
- 3. Representation of geographical names in China (Fig.2, P5): The paper makes use of many area names in China. However, since general readers don't know their location, I would request the authors to plot the important locations in Fig.2.
- 15 Reply: Fig.2 is too crowed for other marks, so we marked the locations of the provinces mentioned in section 3.1 in Figure S1 in the supplementary material.
 - 4. P5. L17-19: The three regions (JJJ, JGZ, GG) are hard to grasp for me. These areas should be shown in the Figures. Reply: We also marked the areas of the three regions in Figure S1 in the supplementary material.
 - 5. Fig. 5: I don't understand why the authors represent the mean states in JJA. I think the analysis of the JJA is unnecessary because the paper focuses on the winter season.
 - Reply: Monsoon circulation is the key factor to understand the influences of ENSO over East Asia. Therefore, the first thing we thought about was to explore if the model could capture the general characteristics of the East Asian monsoon circulations.
- 25 We gave the monsoon circulations in winter as well as in summer in order to comprehensively explore this question. The

common problem in simulating the circulations of the East Asian winter monsoon and East Asian summer monsoon by the model might give some clues in the future works, e.g., improving the model or using the model in other studies.

6. P10. L7, Fig.10: The authors make a point that "the winter-average wet depositions of aerosol over southern China are decreased by La Nina (Fig.10b)". However, the wet depositions seem to be increased over the southern China during La Nina, although they are decreased over the south coast of China and the sea.

Reply: In many studies, people used "southern China" to represent the large area of China south of the Yangtze river. But in the climate category of the National Climate Center of China Meteorological Administration (NCC/CMA), southern China means the region that generally includes the provinces of Guangdong and Guangxi, very similar to the region of GG in our work (Figure S1).

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7. Fig. 12: I suggest that each number of days categorized by "moderate" or "heavy" aerosol concentrations are shown during El Nino and La Nina in addition to the PDF, because I have difficulty reading the small difference between the ENSO from the PDF.

Reply: We have changed the PDF to frequency distribution, so that the absolute number of days can be seen from the figure directly. And we also gave the number of days with the regional mean surface aerosol concentration over southern China (CONCsur) \leq 40, 40<CONCsur \leq 80, and CONCsur>80 μ g/m³. Please see the revised Figure 12. But it is necessary to note that here we used regional mean CONCsur subjected to the low spatial resolution of the model (2.8° x 2.8°). In the real operation, haze days and their degrees of severity are judged station by station. Therefore, Figure 12 is given just to explain a possibility that southern China can have less (more) haze days but more (less) mean aerosol concentration in El Nino (La Nina) winters, through analyzing the shifts of the curves in El and LA relative to the CLI.

8. P1. L20 and P12. L16-17: Do you use the "aerosol" as a synonymous term for "haze"? It is confusing for me because the authors do not mention the relationship between aerosol concentration and occurrence potential of haze days.

25 Reply: Haze pollution happens under stagnant weather conditions, because human-beings are emitting large amount of aerosol

particles and their precursors into the atmosphere. Therefore, in the part of model simulation, we used the aerosols that were mainly emitted by human-beings as a proxy of haze pollution. Comparing Figure 2 and 6, it can be seen that the distributions of the winter haze days and aerosol concentrations over China are consistent with each other. But, as we have discussed in the last paragraph of the manuscript that haze pollution is a very complicate problem, and works with more complicate experimental designs and chemistry schemes are still in need in the future.

reviewer 2

Interactive comment on "The Effects of El Niño-South Oscillation on the Winter Haze Pollution of China" by Shuyun Zhao et al.

10 M.-K. Kim

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mkkim@kongju.ac.kr Received and published: 26 July 2017

In this manuscript, the authors found observational evidence about the regional de- pendency of the relationship between winter haze days of China and ENSO SST, and tried to verify the mechanism about the effect of ENSO on the winter haze pollution of China based on the composite analysis for El Nino cases and La Nina cases which are simulated by using the aerosol-climate coupled model BCC_AGCM2.0_CUACE/Aero of the NCC, focusing both transport effect and scavenging effect which are related to the atmospheric circulation change and associated precipitation change induced by different SST forcing. The methodology is sound and the paper appears to be relatively well-organized. I believe that the paper provides a further insight into the regional dependency of the ENSO effect on haze pollution of China. However, in this version there are some scientific comments to be clarified.

Specific comments

1. Significant test: the most important procedure in composite analysis is to pro- vide readers with the significant test for the difference in target variables between two groups, at least 5% significant level. So, the authors should add the significant level for every difference field. The simulated changes caused by El Nino and La Nina forcing relative to climatology are not always symmetric each other. So, it is very difficult to understand the difference between El Nino effect and La Nina effect. In my

opinion the other approach to stress the effect of El Nino on the winter haze is to see the difference between two extremes (El Nino and La Nino). In this way, there will be a clear difference by offsetting the same sign and magnitude. Anyway, the authors should revise the manuscript based on the significant test. In addition, the authors should add some discussions about the dynamic link between circulation change and ENSO forcing through teleconnection pathway based on the significant test.

- Reply: About the significance. In the part of model results, we used the medians instead of the ensemble means of the differences of variables caused by ENSO, as median is more robust and resistant to extreme value that might happen in a specific experiment. And we also added markers to those grids where the differences of a specific variable between more than 70% pairs of ensemble members have the same sign with the median differences, as a reflection of significance. Please see the revision.
- According to the suggestion of the reviewer, we added discussions about the differences of variables between El Niño and La Niña winters. Please see the revision. Thanks for the suggestion.

About the teleconnection. We added discussions on how ENSO forcing influences the EAWM circulation through the Pacific-East Asian teleconnection, please see section 3.2.2 in the revised manuscript and the Figure S4 in the supplementary document.

2. Filtering of the monthly haze days: In Fig.4 the authors calculated the correlation coefficients between the winter-average monthly haze days and SST in winter, using data after applying a linear-trend removing and 2~8 years' band-pass filtering. Basically 2-8 years' band-pass filter also eliminates a linear trend because periods less than 2 years and over 8 years including a linear trend are removed. So, using both filters are redundant. Applying a linear-trend removing at first is obviously not valid because time series is not linear. Moreover, sudden jump in last year can make linear trend distorted toward large trend. Thus, the author should clarify this point. If it is so, the authors should re-calculate the correlation coefficients in Fig. 4 by considering the point suggested above.

Reply: Indeed, a 2-8 years' band-pass filtering has already eliminated a linear trend. Therefore, we replotted Fig. 4 only applying the band-pass filtering, please see the revision. Considering the abrupt increase of haze days after 2010, especially in 2013, we only used the data between 1960-2010. Please see the 4th paragraph of section 3.1.

3. Haze days and surface aerosol concentration in Southern China: Is less haze days in observation during El Nino winter shown in Fig. 3c matched with less surface aerosol concentration in model during El Nino winter shown in Fig. 9a? If it is true, why are surface aerosol concentrations over southern China decreased in both El Nino and La Nina winter? Please clarify this point. In addition, the authors should check whether Fig 9a and b are consistent with Fig. 12 over southern China. In my opinion, two pdf curves for El Nino and La Nina should be located below CLI curve on average because changes in both Fig. 9a and b are negative over southern China. Are pdf results dependent on the definition of southern China?

obvious in Fig. 9a. But if we compare the results between El Niño and La Niña winters, which we have added according to the suggestion of the reviewer, El Niño causes more aerosol CONCsur over southern China (revised Fig. 9c). And the increase in the winter atmospheric contents of aerosols over southern China caused by El Niño can be seen more clearly from the variable of aerosol loadings (revised Fig. 9, right panel). In this work, southern China mainly includes Guangdong and Guangxi provinces (the "GG" in Figure 3 and 4), according to the climate category of the National Climate Center of China Meteorological Administration. The scope for calculating regional mean aerosol CONCsur over southern China in Fig. 12 is given in the added supplementary document (Fig. S1), consistent with the southern China in Fig. 9.

Reply: The increase in winter surface aerosol concentration (CONCsur) over southern China caused by El Niño is not very

According to the suggestion of another reviewer, we have changed the PDF in Fig. 12 to frequency distribution. As we have explained in the last paragraph but two of section 3 that here we used regional mean aerosol CONCsur, which caused the curves in Fig. 12 to look close to each other. However, we can still find that the curve for La Niña (El Niño) generally shifts leftward (rightward) comparing with that for CLI. And it can also be reflected from the top right corner in Fig. 12 that southern China tends to have more moderate (heavy) but less clean and heavy (moderate) haze days in La Niña (El Niño) winter.

Technical corrections

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1. Line 9 page 8: "the weakness of EAWM" should be "the intensification of EAWM" because Siberian high is stronger than normal.

Reply: The changes in Siberian High can be seen more clearly from the sea level pressure (SLP) field (Figure R1). It is seen from the change in land-sea SLP gradient over East Asia-Pacific caused by El Niño (Figure R1 (a) and (c)) that El Niño causes

a weakness of EAWM. We also calculated the change in EAWM index (Li and Yang, 2010) caused by El Niño and La Niña, and also found that El Niño causes a weakness of EAWM. Please see the Figure S3 in the supplementary material.

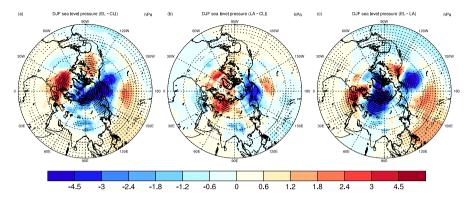


Figure R1. Medians of the simulated differences in DJF sea level pressure between (a) EL and CLI, (b) LA and CLI, and (c) EL and LA, with black dots indicating that the differences between more than 70% pairs of ensemble members have the same sign with the median differences.

- 2. Line 5-8 page 9: Over northeastern China winter CONCsur of aerosols are decreased in both El Nino and La Nina, indicating that the decrease over northeastern China is not irrelevant to ENSO. So, the authors should describe this point carefully.
- Reply: From the revised Figs. 9c and 9f, it seems that ENSO does not affects the atmospheric contents of aerosols over the north part of China (including northeastern China) as significantly as it does over southern China. As to this point, we have added a paragraph for discussion from the perspective of cold air frequency in China, please see section 4 in the revised manuscript.
 - 3. Line 1 page 24: "zonal flux" should be "horizontal flux".
 Reply: revised.

reviewer 3

Interactive comment on "The Effects of El Niño-South Oscillation on the Winter Haze

Pollution of China" by Shuyun Zhao et al.

Anonymous Referee #3

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Using observations and model simulations, the authors investigated the possible influences of ENSO events on aerosols

over China. Understanding the changes in aerosols is a relevant topic for improving our knowledge of relationship

between natural cycle and aerosols. Based on the observational data, they concluded that the haze days of southern

China tend to be less (more) than normal in El Niño (La Niña) winter, however the relationship is not significant.

However, inconsistent result is seen based on the simulated result. And they concluded it is due to the fact that heavy

haze days are more frequent in El Nino winter. I agree with the authors that the influence of ENSO events on aerosol

concentrations over China cannot be ignored. And it is an important interdisciplinary issue which needs more attention

and deep researches. However, there are many problems of this manuscript, and it cannot be accepted by ACP as it is

now. Some specific comments or suggestions are listed as follows.

1. The authors mainly focused on the influence of ENSO via EAWM on the haze over China, however, what is exact influences

of the ENSO influence on the EAWM? And the involved physical processes. The authors did not give a detailed explanation

regarding this point neither concluded the existed explanations. I suggest the authors do not avoid discussing this question, but

make complete statement about this point.

Reply: Thank you for the suggestion. We have added discussions on how ENSO influences the EAWM in section 3.2.2. It was

found that El Niño decreased the winter land-sea gradient in the sea level pressure field over East Asia and weakened the East

Asian jet stream, therefore caused a weakness of EAWM. This is to some extent in accordance with previous studies. According

to the work of Wang et al. (2000), the Pacific-East Asian teleconnection (PEAT) is the key bridge, which connects the ENSO

and its upstream climate in East Asia. Therefore, we discussed the PEAT from vorticity and stream function. Please see the revision and the Figures 3S and 4S in the supplementary document.

2. The influences of ENSO on the aerosols have been discussed in recent works. Regarding the impacts of El Niño and La Nina Modoki events on aerosol concentrations over eastern China, and the impacts of EASM on O3 over eastern China. The authors should include the relevant work into the present study, compare the differences, and further highlight the motivation of the present study.

Reply: We have cited more recent works involving the impacts of El Niño and La Niña Modoki events on aerosols over eastern China, and the impacts of EASM on O_3 . More discussions have also added. Please see the section 1 of the revised manuscript.

3. The abbreviations of JJJ, JZH, and GG show no sense, the abbreviations should include geographic information and easy remember. I suggest using north China, southern China to replace.

Reply: replaced.

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4. The contradict description in the manuscript, for example, P5L21, "After the year of 2000, the winter-average monthly haze days over the three regions grew dramatically, especially over JZH and GG"; but in P5L25, "haze days over JZH and GG increased too abruptly after the year of 2010, especially in 2013".

Reply: contradict description has been modified.

5. The reasons for the applying a linear trend removing and a 2-8 years band-pass filtering showed be illustrated, and the valid freedom of significant should be applied to the significance test. In Figure 3, it is seen that the hazes days show nonlinear variations, even within 1950-2010. And whether the hazes days show a 2-8 years' periods is not clear, it makes me hard to understand why the authors removed the linear trend and a band pass filter.

Reply: As has been pointed out by another reviewer that by applying a 2-8 years band-pass filtering, the linear trend had already been removed. Therefore, we have modified Figure 4, by only applying the 2-8 years band-pass filtering. The reason

25 for applying the 2-8 years band-pass filtering is to remove the inter-decadal variabilities of both the winter haze days and SST.

We have illustrated the reason in the reviewed manuscript. Please see the section 3.1. The significance has been given when plotting the correlations between the winter haze days of China and SST, please see the figure caption of Figure 4.

We removed the linear trend before analyzing the relationship between the winter haze days of different regions of China and SST, because we wanted to remove the influences of global warming and the increasing anthropogenic emissions of aerosols and their precursors as much as possible. And we applied a 2-8 years band-pass filtering, in order to remove the inter-decadal variabilities, as it is known that ENSO is regulated by inter-decadal oscillations, e.g., PDO. And it can also be seen from Figure 3 that the winter haze days over northern and eastern China show an obvious peak around 1980, which also indicates an influence from some inter-decadal oscillation.

6. As to the influences of ENSO on the EAWM, many researchers have illustrated that the ENSO shows important role in impacting the intensity of EAWM, and the climate, including temperature, rainfall and winds over north China, central China are impacted by the EAWM. However, the result shows the role of ENSO on hazes over JJJ and JZH is not evident. I suggest the authors to separate the stronger and moderate ENSO events, or separate the eastern and central ENSO events to further explore the result, and it is unknown the exact years of El Nino and La Nina in the present work.

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Reply: Inspired by the question, we rethought about this chain of ENSO–EAWM–haze pollution of the large north part of China. In January 2013, when the unprecedented haze event happened in Beijing, there was no El Niño or La Niña event in the tropical pacific. These reminded one of our authors that in her previous operational work she had noticed that El Niño did not necessarily reduce the cold air frequency in China in winter, even though it could weaken EAWM. In winter, a few clean days in the north part of China always follow a breakout of cold air from Siberia or Mongolia. The relationship between ENSO and the cold air frequency makes the connection between ENSO and the haze pollution of the north part of China more complex than it is expected from the perspective of the strength of EAWM. We have added a paragraph in the part of conclusions and discussions. Please see the revision.

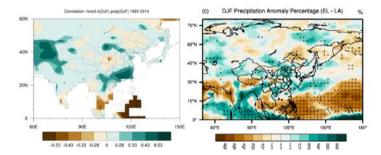
In this work, we did not use the SST for any real specific year, but the climatology SST and a typical ENSO mode scaled by average Niño3.4 index, in order to diminish the influences from other forcings (e.g., in the Atlantic Ocean and Arctic) as much as possible. In our future work, we will change the shape of the inter-monthly variation of Niño3.4 (Figure 1b) and/or other

ENSO indices by selecting different El Niño and La Niña events or change the shape artificially, to explore the influence of ENSO of different types and strengths on the haze pollution over the large north part of China. Thanks for the suggestion.

7. The performance of the model used to perform the simulations in reproducing ENSO patterns as well as its variations is unknown. Since ENSO is a complex air-sea interaction process, it is necessary to access the performance in simulating ENSO whereas given the circulations over eastern China during winter and summer. And the observed variations in rainfall should be shown while the simulated result for the poor simulations of rainfall in the models.

Reply: In this work, we used an AGCM instead of CGCM, with prescribed SST of climatology and superimposed ENSO signals. It can only explore one-way (ENSO-atmosphere-aerosols) influences, as we have also pointed out in the part of conclusions and discussions. Now, we've added more discussions on the simulated circulation responses to ENSO, including the responses of East Asian jet stream, EAWM index, and the Pacific East Asian teleconnection. Please see the revised manuscript and supplementary document.

The observational relationship between ENSO and the rainfall over East Asia and the globe has been given on a website of the NCC/CMA (Figure R2 left). We added the citation of the website in the revision. Thanks for the suggestion.



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Figure R2. left: Correlation coefficient between Nino3.4 and DJF precipitation given at http://cmdp.ncc-cma.net/pred/cn_enso.php?product=cn_enso_corr&season=DJF#corr; right: the revised figure 8c, differences in DJF precipitation in percentage between El Nino and La Nina winters.

- 8. Figure captions, the blank in figure 5a, b, should be mentioned, however not in c and d. Figure 7, shaded for hgt not contour. Figure 9, it is better if the relative changes is shown whereas the absolute values, considering that the climatology mean shows big differences as shown in Figures 1 and 6.
- 5 Reply: Captions of Figure 5 and 7 have been modified, please see the revised manuscript. Indeed, in Figure 6 the climatology mean aerosol concentrations and loadings show large geographical variations. However, over the areas east of 105°E and south of 45°N where we're interested in, their geographical variations are not very large. Therefore, we kept the absolute changes in Figure 9. Still very thank you for the suggestion.

Part 2. A list of relevant changes made in the manuscript

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- 1. We replotted Figure 4, by only applying a band-pass filtering of 2-8 years, as band-pass filtering has already done the linear-trend removing, which is redundant in the original manuscript. Corresponding, the figure caption was also modified.
- 2. We replotted Figures 7-11, by adding the comparisons between El Niño and La Niña winters. Correspondingly, we added relevant illustrations in the figure captions and discussions in the text.
 - 3. We added markers to Figures 7-11 where the differences between 70% pairs of experiments have the same sign with the median difference as a reflection of significance. Illustrations have also been added to the last paragraph of section 2.2.
- 4. We replotted Figure 12, by calculating the frequency distributions instead of the probability distribution functions of the winter daily surface concentrations (CONCsur), and we also added the number of days when the CONCsur $\leq 40\mu g/m^3$, $40 < CONCsur \leq 80\mu g/m^3$, and CONCsur $> 80\mu g/m^3$. Caption of Figure 12 was also modified. In the text section 3.2.3 and abstract, we also modified relevant discussions.
- 5. We added 5 more references, e.g., Feng et al. (2017), Kang and Lee (2017), Li and Yang (2010), Wu et al. (2013), and Yang et al. (2014) in the text and reference list.
- 6. We added a figure in the supplementary material (Figure S1) to illustrate the locations of the provinces and regions of China

mentioned in section 3.1.

- 7. We added the comparisons as to the winter haze days over southern China between 4 pairs of El Niño and La Niña winters in the supplementary material (Figure S2). And relevant discussions were also added in the text, e.g., section 3.1.
- 8. We modified some paragraphs in section 3.2.2, and added extensive discussions about how ENSO affected the East Asian
- Winter Monsoon (EAWM). We added a citation of the website of NCC/CMA (http://cmdp.ncc-

cma.net/pred/cn_enso.php?product=cn_enso_corr&season=DJF#corr) and two figures in the supplementary material (Figures

S3 and S4) to illustrate how ENSO affected the EAWM from the perspective of East Asian jet stream and Pacific-East Asian

teleconnection.

- 9. We added a paragraph in section 4 (conclusions and discussions) to illustrate why the connections between ENSO and the
- haze pollution over northern and eastern China are not as strong as we expected to be, as they both have some connections

with the EAWM. During illustration, we added two figures in the supplementary material (Figures S5 and S6).

10. Other modifications relative to grammar corrections or making some sentences more clearly and properly.

Part 3. Marked-up manuscript version

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The Effects of El Niño-South Oscillation on the Winter Haze Pollution of China

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Abstract. It is reported in previous studies that El Niño-South Oscillation (ENSO) influences not only the summer monsoon,

but also the winter monsoon over East Asia. This contains some clues that ENSO may affect the winter haze pollution of China,

which has become a serious problem in recent decades, through influencing the winter climate of East Asia. In this work, we explore the effects of ENSO on the winter (from December to February) haze pollution of China statistically and numerically. Statistical results reveal that the haze days of southern China tend to be less (more) than normal in El Niño (La Niña) winter; whereas the relationships between the winter haze days of northern and eastern China and ENSO are not significant. Results from numerical simulations also show that ENSO influences the winter atmospheric contents of anthropogenic aerosols over southern China more obviously than it does over northern and eastern China. Under the emission level of aerosols for the year 2010, the winter atmospheric contents of anthropogenic aerosols over southern China are generally more (less) than normal in El Niño (La Niña) winter. It is because that the transports of aerosols from South and Southeast Asia to southern China are enhanced (weakened), which mask the better (worse) scavenging conditions for aerosols in El Niño (La Niña) winter. The frequency distribution of the simulated daily surface concentrations of aerosols over southern China indicates that the region tends to have less clean and moderate (heavy) haze days, but more heavy (moderate) haze days in El Niño (La Niña) winter.

1 Introduction

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Haze pollution, especially in winter, has become a very serious problem for China in recent decades (Ding and Liu, 2014; Tao et al., 2016). For example, in January 2013, most parts of central and eastern China experienced an extremely heavy and persistent haze pollution (Tao et al., 2014; Mu and Zhang, 2014; Zhang et al., 2014; Wang et al., 2014a; b; Zou et al., 2017). In the last decade, haze pollution in winter has received wide concerns from the scientific community, the government of China, and the public.

Haze pollution is a phenomenon mainly caused by human-emitted pollutants under stagnant meteorological conditions. Increasing anthropogenic emissions of aerosols and their precursors in recent decades are the main reasons for the worsening air qualities in China (Cao et al., 2007; Zhang et al., 2012; Zhu et al., 2012). In addition to the increase in anthropogenic emissions of aerosols and their precursors, climate changes caused by anthropogenic and/or natural forcings also exert great influences on the haze pollution in China, especially through changing the strength of East Asian Monsoon (Zhang et al., 2010; 2014; Liu et al., 2011; Yan et al., 2011; Chin, 2012; Zhu et al., 2012; Mu and Zhang, 2014; Chen and Wang, 2015; Li et al., 2016a; b; Cai et al., 2017). Studies generally showed that the wintertime haze days across central and eastern China had a close

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negative relationship with the strength of East Asian Winter Monsoon (EAWM) (Zhang et al., 2014; Mu and Zhang, 2014; Chen and Wang et al., 2015; Li et al., 2016a; Cai et al., 2017). In summer, the increase in surface aerosol concentration and optical depth over eastern and northern China always correlates with the weakening of the East Asian Summer Monsoon (EASM) (Zhang et al., 2010; Yan et al., 2011; Zhu et al., 2012). Whereas Yang et al. (2014) found that the gaseous pollutant of surface Oa over China had a positive relationship with the EASM.

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Wang et al. (2015) and Zou et al. (2017) revealed that the increasing winter haze pollution in eastern China in recent years was related to the decreasing Arctic sea ice in preceding autumn. Many studies suggested that under global warming, the future climate would be more stagnant and the weather conditions conducive to severe haze in eastern China would be more frequent (Jacob and Winner, 2009; Wang et al., 2015; Cai et al., 2017). But Jacob and Winner (2009) also pointed out that the effects of future climate change on particulate matter (PM) were complicated, as the projection of precipitation, wildfires, atmospheric chemistry, and natural emissions of aerosols by models were still in need of improvement.

ENSO is the dominant changing mode of the tropical sea surface temperature (SST) on interannual scale (Rasmusson and

ENSO is the dominant changing mode of the tropical sea surface temperature (SST) on interannual scale (Rasmusson and Carpenter, 1982), of which the climatic effects are global (Bjerknes, 1972; Huang and Wu, 1989; Zhang et al., 1999; Lau and Nath, 2003; Zhai et al., 2016, etc.). ENSO not only influences the EASM (Chang et al., 2000; Li et al., 2007; Zhao et al., 2017, etc.), but also influences the EAWM, especially over low latitudes (Chen et al., 2013; He and Wang, 2013; Kim et al., 2016, etc.), which means that ENSO may affect the haze pollution over East Asia through its influences on the monsoon circulation. Wu et al. (2013) found that the aerosol variations over the Maritime Continent and western North Pacific presented a biennial feature, which could be attributed to the impacts of ENSO. From the results of Wu et al. (2013), it seemed that ENSO only influenced the aerosols over eastern China (30°–40°N, 110°–120°E) around October of El Niño or La Niña developing years and July of El Niño or La Niña decaying years. Gao and Li (2015) revealed statistically that El Niño (La Niña) events were more likely to bring more (less) haze days in eastern China (25°–35°N, 105°–122.5°E). Feng et al. (2016) simulated the influences of the 1994/1995 El Niño Modoki event on the aerosol concentrations over southern China (20°–35°N, 105°–120°E), and found that the aerosol concentrations increased during the mature phase (in boreal winter) of the event. Feng et al. (2017) simulated the influences of the 1998/1999 and 2000/2001 La Niña Modoki events on the aerosol concentrations over eastern China during the winters of the

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two events were opposite with each other. Most previous studies discussed the effects of ENSO on the winter haze pollution of China based on observation or case simulation separately. In this study, we try to explore the effects of ENSO on the winter haze pollution of China statistically and numerically. ENSO is a recurring climate pattern, and many climate centers of the world monitor it systematically and use it in climate prediction extensively. Therefore, exploring the effects of ENSO on the winter haze pollution in China may provide useful information in the prediction of haze for the country.

The article is organized as follows: methodology is given in section 2, including data and model introduction; results including statistical and model results, are presented in section 3, followed by conclusions and discussions in section 4.

2 Methodology

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2.1 Data used in statistical analysis

We firstly analyze the relationship between the winter haze days of China and global SST in section 3. The monthly haze days from the data set for haze project version 1.0 of the National Meteorological Information Center, China Meteorological Administration (CMA) are used. The CMA defines haze using visibility (<10 km) and relative humidity (<80%) (Tao et al., 2014). The time-span of the data set is January 1954 July 2014, but only the data after 1960 are actually used in this study, as the data set has steadily included more than 2000 stations since 1960. A merged monthly SST data (Hurrell, et al., 2008) formed by the SST data of the Hadley Centre and the National Oceanic and Atmospheric Administration (NOAA) is used in the correlation analysis. The SST data together with a merged sea ice (SI) data of the same sources are used in the following numerical simulation. The SST and SI data are both from 1870 to 2012, with a horizontal resolution of 1°×1°.

2.2 Model description and experimental set-up

The aerosol-climate coupled model BCC_AGCM2.0_CUACE/Aero (Zhang et al., 2012) of the National Climate Center (NCC), CMA, is used in the numerical study of the effects of ENSO on the atmospheric contents of aerosols. The coupled model composes of the NCC/CMA climate model (BCC_AGCM2.0, Wu et al., 2010) and the CMA Unified Atmospheric Chemistry Environment/Aerosol model (CUACE/Aero, Gong et al., 2002, 2003). The coupled model employs a horizontal T42 spectral resolution (about 2.8°×2.8°) and a hybrid vertical coordinate with 26 levels, the top of which is located at about 2.9 hPa. Five types of aerosols (including their emissions, gaseous chemistry, transports, coagulations, and removals): sulfate (SF), black carbon (BC), organic carbon (OC), dust, and sea salt are considered in the model. The emissions of the first three types of

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aerosols and/or their precursors are prescribed, and the last two types of aerosols are emitted online (Gong et al., 2002). The particle radii of each type of aerosol are divided into 12 size bins from 0.005 to 20.48 um. All types of aerosols are assumed to be externally mixed with each other. Sulfate, organic carbon, and sea salt are assumed to be hygroscopic, and the other two types of aerosols are assumed to be non-hygroscopic. The coupled model has been introduced, evaluated, and used in many studies of the radiative forcing and climatic effects of aerosols (e.g., Wang et al., 2015; Zhao et al., 2014; Zhang et al., 2016). Three groups of experiments are conducted (Table 1), named CLI, EL, and LA, with each group including 20 members by altering initial conditions. To get different initial conditions, a preparation experiment is run firstly with the model's default setting (Zhao et al., 2014). Three types of files (initial, restart, and history files) of the preparation experiment are output, and the output frequency is set to daily. 20 initial files output from the preparation experiment are then used as the different initial conditions for different ensemble members. The group of CLI uses the climatological-mean (from 1981 to 2010) monthly SST and SI, which have been introduced in the section 2.1 but interpolated to the model's resolution, as boundary conditions. In the groups of EL and LA, the climatological-mean monthly SST is superposed by El Niño and La Niña SST perturbations, respectively, and the SI is identical to that in CLI. The El Niño and La Niña SST perturbations are obtained by scaling a typical ENSO mode (Figure 1a) with the average monthly Niño 3.4 indices of 21 El Niño and 18 La Niña events (selected from 1951 to 2015, Figure 1b), respectively. The Niño3.4 indices from January 1951 to date can be downloaded from the website of NCC/CMA (cmdp.ncc-cma.net/download/Monitoring/Index/M Oce Er.txt). The typical ENSO mode is obtained through the regression between the monthly Niño3.4 index and the SST field after removing their linear trends. The running period of the group of CLI is from October to the next August, to testify if the model can capture the general features of the circulations of the East Asian winter and summer monsoons. In the testifying process, the geopotential height and wind from the National Centers for Environmental Prediction (NCEP) reanalysis data are used. Whereas the running periods of EL and LA are both from October to the next February. The results in boreal winter (December, January, and February, or DJF for short) of the three groups are used in analysis, allowing prior two months for the atmosphere to response to SST perturbations. The emission data of SF, BC, OC, and/or their precursors used in all experiments are from the Representative Concentration

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changes in the atmospheric contents of SF, BC, and OC caused by different SST perturbations are analyzed, as the three types 18

Pathway 4.5 (RCP 4.5) of the Intergovernmental Panel on Climate Change (IPCC) for the year of 2010. In this study, only the

of aerosols are mainly emitted by anthropogenic activities and important components of haze pollutants. <u>In the analyzing</u> process, the median instead of the average of the changes of a specific variable between different groups of experiments (e.g., <u>EL – CLI</u>) is used, as median is more robust and resistant to extreme large or small values that may happen in some ensemble members. And we also mark the grids where the differences of a specific variable between more than 70% pairs of ensemble members have the same sign with the median differences, as a reflection of significance.

3 Results

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3.1 Statistical results

In this section, we firstly present the geographical distribution of the winter haze days in mainland China over the past about 50 years and since 2000. Then, three typical polluted regions are selected, and the correlations between their respective winter haze days and global SST are analyzed.

It's seen from Figure 2a that Beijing, the southwest part of Hebei, the central and south parts of Shanxi, the central part of Shaanxi, and the north part of Henan suffered winter haze pollution more frequently than elsewhere in China during J 960–2013, with the largest value of DJF mean monthly haze days among 10–20. The diffusive conditions of these areas are not good because of the influences of the Tai-hang and Qin-ling mountains. Besides the above areas, stations with more than 5–10 DJF mean monthly haze days during 1960–2013 distributed densely in the provinces of Hubei, Hunan, Jiangxi, Zhejiang, Guangxi and Guangdong. Compared with 1960–2013, winter haze pollution in mainland China generally became more frequent during 2000–2013 (Figure 2b). For example, in the Yangtze River Delta and Pearl River Delta, stations with 5–10 and 10–20 DJF mean monthly haze days during 2000–2013 were much more than that during 1960–2013. At some stations of

the Chinese provinces mentioned above and in the following, can be found in Figure S1 in the supplementary document.

Three representative regions: JJJ (Beijing, Tianjin, and Hebei province; accounting for 179 stations), JZH (Jiangsu and Zhejiang provinces, and Shanghai; accounting for 164 stations), and GG (Guangdong and Guangxi provinces; accounting for 178 stations) are selected to represent northern, eastern, and southern China, respectively. It is seen from Figure 3 that the DJF mean monthly haze days of these three regions were generally less than 3 before the year of 2000, with a small peak around

Shanxi province, the DJF mean monthly haze days during 2000-2013 were even more than 20 (Figure 2b). The locations of

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grew rapidly. The DJF mean monthly haze days of northern China increased much later than the other two regions after the year of 2000. Actually, the DJF mean monthly haze days over northern China, were relatively few until 2012, which was consistent with the result of Chen and Wang (2015).

Considering that the increases in the DJF mean monthly haze days over castern and southern China increased too abruptly after the year of 2010, only the winter haze days of the three regions from 1960 to 2010 are used in analyzing their relationships with winter SST. In order to remove the inter-decadal variabilities, we applied a 2-8 years band-pass filtering to both the data of haze days and SST. It is seen from Figure 4 that only the winter haze days of southern China have significant negative relationships with the equatorial SST over central and eastern Pacific and central Indian Ocean, and positive relationships with the equatorial SST over western Pacific. The geographical distribution of the correlation coefficients between the winter haze days over southern China and SST is generally an opposite phase of the typical ENSO mode shown in Figure 1a, indicating that southern China tends to suffer more (less) haze days than normal in La Niña (El Niño) winter. This can also be seen from the comparison of the DJF mean monthly haze days between several selected pairs of La Niña and El Niño winters over southern China (Figure S2). In order to avoid the influences of the emission variations of haze particles and their precursors, each pair of La Niña and El Niño winters are selected with their interval not longer than 2 years.

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The relationships between the winter haze days of northern and eastern China and equatorial SST are not significant (Figures 4a and 4b), indicating that ENSO does not influence the winter haze days of these two regions significantly. It's probably because that as a tropical phenomenon, ENSO affects the climate over southern China more directly than that over northern and eastern China, especially in winter when the Western Pacific Subtropical High (WPSH) is generally weaker and located more south than other seasons. Zou et al. (2017) linked the extreme winter haze events over East China Plains (112°-122°E, 30°-41°N, including JJJ and most parts of JZH) to Arctic sea ice loss in the preceding autumn and extensive Eurasia snow fall in early winter. Gao and Li (2015) found that the winter haze days of eastern China had positive relationship with the SST over eastern equatorial Pacific. But the "eastern China" in Gao and Li (2015) included most regions between the Yangtze and Yellow rivers east of 105°E, which was much larger and more west than the representative region of eastern China in this work, and more south and west than the concerned region in Zou et al. (2017).

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3.2 Model results

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3.2.1 Winter and summer circulations, and the atmospheric contents of aerosols

First of all, the simulated winter and summer circulations over East Asia in the group of CLI are testified by comparing with NCEP reanalysis data (Figure 5). The model can capture the general features of the winter and summer circulations over East Asia. For example, in winter, the deepening of East Asian Trough (EAT), the overwhelming of northwesterly over the east part of China, and the strengthening of the easterly north of the equator are all depicted by model results (Figure 5a); In summer, the northward shift of WPSH, and the strengthening of the cross-equatorial westerly over the Indian Ocean and Maritime Continent are generally captured by the model (Figure 5b). But the simulated EAT in winter by the model is weaker and narrower, and locates more west than NCEP reanalysis data. In summer, the simulated WPSH is weaker and locates more east than NCEP reanalysis data. It seems that the simulated cross-equatorial flow is stronger than reanalysis data both in winter and summer, which is probably the reason for the weakness of the simulated EAT in winter and WPSH in summer. In the model results of the group of CLI, the stronger cross-equatorial westerly over the Indian Ocean and Maritime Continent obstructs the westward stretch of the WPSH in summer, resulting in positive precipitation biases over South Asia and Southeast Asia, and negative precipitation biases over South Asia and Southeast Asia, and

The simulated winter surface concentrations (CONCsur) of aerosols and aerosol loadings in the group of CLI (Figure 6) show that central and eastern China (about east of 105° E) are the most haze polluted regions of the country, in line with the observational distribution of winter haze days shown in Figure 2. The maximum of the winter CONCsur of aerosols centers in Henan province, and is about $20 \mu g \text{ m}^{-3}$. Comparing with other observational studies (e.g., Cao et al., 2007; Zhang and Cao, 2015; Cai et al., 2017), the simulated CONCsur of aerosols shown in Figure 6a are underestimated by about $1\sim2$ orders of magnitude. It should be illustrated that the aerosol CONCsur in this study are the aerosol concentrations at the lowest level of the model. As has been introduced in section 2.2, the model used in this study has 26 levels in the vertical hybrid σ -pressure axis, and the mid height of the lowest level is about $50\sim70$ meters above the surface in China (not shown). Therefore, the aerosol concentrations at the lowest level of the model actually reflect the mean of the aerosol concentrations from the surface to a height of about $100\sim140$ meters (or maybe even higher) above the surface. This certainly brings about underestimations as to aerosol CONCsur. Another reason for the underestimation is that we exclude all nature-emitted aerosols, in order to focus

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our attentions on haze, as another meteorological disaster occurring frequently in late winter and spring over northern China – sandstorm – has quite different weather conditions with haze. The exclusion of nature-emitted aerosols (mainly dust aerosol) is also the reason why the northwestern China is much cleaner than it is expected to be.

The maximum of the simulated winter aerosol loadings in China is $21\sim25$ mg m⁻², and heavy aerosol loadings (≥18 mg m⁻²) in China locate east of 105° E, and between the Yellow and Yangtze rivers (Figure 6b). The median of 10 models that participated in the Aerosol Comparisons between Observations and Models (AeroCom, http://aerocom.met.no/cgibin/aerocom/surfobs_annualrs.pl) showed that the maximums of the loadings of SF, BC, and OC in January 2000 in China were $15\sim30$, $2.5\sim5$, and more than 10 mg m⁻², respectively. The model used in this study is also a member of AeroCom. Zhao et al. (2014) have compared the simulated loadings of five kinds of aerosols (SF, BC, OC, sea salt, and dust) by the model

with AeroCom median, and found that the model generally simulated the distributions and magnitudes of aerosol loadings well, though with some underestimation $\underline{\underline{s}}$ in BC and OC.

3.2.2 The effects of ENSO on winter circulation and precipitation

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The effects of ENSO on the winter circulation and precipitation over East Asia are discussed firstly, as these meteorological fields determine the transports, diffusions, removals, and consequently the atmospheric contents of aerosols.

Two important features are apparent in the winter anomalous circulation field caused by El Niño (Figure 7a). Firstly, negative

and positive anomalous geopotential heights at 500 hPa are seen near Ural Mountains and Lake Baikal, respectively, Secondly, two anomalous anticyclones at 850 hPa are seen over western North Pacific, with one in the Philippine Sea and the other one in the mid latitude. It is expected that the anomalous southwesterly in the northwest of the Philippine Sea anticyclone brings more water vapor and also more aerosols to southern China, as Indo-China Peninsula and South Asia are both areas with heavy aerosol loadings (Figure 6b). Whereas the anomalous geopotential height at 500 hPa caused by La Niña are positive and negative near Ural Mountains and Lake Baikal, respectively, and two anomalous cyclones are caused by La Niña over western North Pacific (Figure 7b).

The differences in circulation between El Niño and La Niña winters are shown in Figure 7c. It is seen in Figure 7c that the negative and positive anomalies in 500 hPa geopotential height caused by El Niño are more obvious than that in Figure 7a over Ural Mountains and Lake Baikal, respectively. And the anomalous anticyclones in the western North Pacific are also

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that El Niño causes a weakness of the EAWM. The weakness of the EAWM caused by El Niño can also be seen from the weakness of East Asian jet stream and the decrease in EAWM index caused by El Niño (Figure S3). The EAWM index used in Figure S3 is the one defined by Li and Yang (2010). It has been suggested in previous studies that the weakness of East Asian jet stream and the Philippine Sea anticyclonic anomalies in winter caused by El Niño connected with each other by local Hadley circulation over East Asia (Kang and Lee, 2017). Previous studies have also found that the EAWM tended to be weak in El Niño winter (e.g., Chen et al., 2000; Huang et al., 2012; Wang and Chen, 2014; Kang and Lee, 2017). Wang et al. (2000) has explored how ENSO influenced its upstream climate in East Asia, and found that the Pacific-East Asian teleconnection (PEAT) was the key bridge. PEAT is a vorticity wave pattern that starts from the central Pacific and extends poleward and westward to East Asia. In Figure 7c, PEAT can be recognized from the 850 hPa wind anomalies with a cyclonic vorticity over the central Pacific, the anticyclonic vorticities over western North Pacific, and a cyclonic vorticity over northeast Asia. The PEAT can be seen more clearly from the differences in the stream function at 500 hPa between El Niño and La Niña winters (Figure S4), which has an opposite sign with vorticity. Corresponding to the anomalous anticyclones at 850 hPa over western North Pacific caused by El Niño, winter precipitation decreases over Indo-China Peninsula and porthwestern Pacific, and increases over southern China (Figure 8a). In contrast, precipitation increases over Indo-China Peninsula and northwestern Pacific, and decreases over southern China during La Niña winter (Figure 8b). The opposite effects of El Niño and La Niña on the winter precipitation over southern China can be seen more clearly in Figure 8c, which is in accordance with the observational relationship between ENSO and China precipitation in winter (http://cmdp.ncc-cma.net/pred/cn enso.php?product=cn enso corr&season=DJF#corr). The decrease in winter precipitation over southern China caused by La Niña is very likely the reason for the more-than-normal haze days over the region during La Niña winter (Figure 4c), as less precipitation means slower cleaning particles out of the atmosphere (Figures 10b and 10c), which will be discussed in the next section. Another reason probably cannot be neglected that drier conditions over southern China during La Niña winter can avoid mistaking haze to fog days.

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more obvious than that in Figure 7a. Wang et al. (2000) found that the anomalous anticyclones over western North Pacific formed in the boreal autumn of a developing El Niño, attained its peak in winter, and persisted into the following spring and early summer. In the sea level pressure field, the land-sea gradient in winter is decreased by El Niño (not shown), indicating

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3.2.3 The effects of ENSO on winter atmospheric contents of aerosols

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In this section, the changes in the winter aerosol CONCsur and loadings over China caused by ENSO are presented, and then the mechanism how ENSO affects the winter atmospheric contents of aerosols is analyzed from the perspective of wet and dry depositions and interregional transports of aerosols.

It is seen from Figure 9a that the winter CONCsur of aerosols are decreased by El Niño over northeastern China and eastern China, The winter aerosol CONCsur are also decreased by La Niña over northeastern China and eastern China, as well as the north part of northern China and most areas south of the Yangtze river (Figure 9b). Over southern China, although the increases in the winter CONCsur of aerosols caused by El Niño are not very clear in Figure 9a, they are obvious by comparing with La Niña winter (Figure 9c), which is generally in line with the simulation result of Feng et al. (2016). It is found from Figure 9d. that the winter aerosol loadings are increased by El Niño over most areas east of 105°E and south of 40°N, and decreased over northeastern China and the north part of northern China. From Figure 9e, it is seen that the anomalous winter aerosol loadings caused by La Niña present a meridional "- + -" pattern over the east part of China. The different influences of El Niño and La Niña on the winter aerosol loadings are the most obvious over the large areas south of the Yangtze river (Figure 9f). From Figures 9c and 9f, it is found that the differences in the atmospheric contents of aerosols between El Niño and La Niña winters are more obvious over southern China than the other areas of the country. This is to some extent in line with the results in Figure 4 that only the winter haze days over southern China have significant relationship with ENSO Therefore, in the following analysis, we focus on the mechanism how ENSO affects the winter atmospheric contents of aerosols over southern China. It has been discussed in section 3.1 that the haze days over southern China tended to be less (more) than normal in El Niño

(La Niña) winter, which is to some extent contradictory to the increase (decrease) in the winter atmospheric contents of aerosols over southern China caused by El Niño (La Niña). In the following of this section, we firstly explore the reasons for the increase (decrease) in the winter atmospheric contents of aerosols caused by El Niño (La Niña) over southern China. Then, we try to explain why the changes in the winter haze days and atmospheric contents of aerosols caused by ENSO over southern China are not consistent with each other.

As the emissions of aerosols are kept the same in all experiments (section 2.2), how quickly aerosols are removed from the

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atmosphere, especially through wet deposition, can affect the atmospheric contents of aerosols greatly (Zhang et al., 2016). It is found that ENSO influences the winter wet depositions more obviously than the winter dry depositions of aerosols over China (Figure 10). The winter dry depositions of aerosols are decreased both by El Niño and La Niña over southern China (Figures 10e–f). Comparing with dry deposition, wet deposition is a faster process. The winter wet depositions of aerosols over southern China are increased (decreased) by El Niño (La Niña) (Figure 10a–c), corresponding to the changes in winter precipitation (Figure 8). Comparing Figure 9 and 10, it is found that the winter atmospheric contents and wet depositions of aerosols are both increased (decreased) by El Niño (La Niña) over southern China. It seems that the changes in the winter wet depositions of aerosols are the results rather than the reasons of the changes in the winter atmospheric contents of aerosols over southern China.

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Besides local emissions and removals, the interregional transports of aerosols can also influence the atmospheric contents of aerosols over a specific region. It was mentioned in Zhang et al. (2016) that South and Southeast Asia had become the important source areas of anthropogenic aerosols in 2010, which was also seen in the simulated aerosol CONCsur and loadings over these regions (Figure 6). A low-level anomalous anticyclone (cyclone) is caused by El Niño (La Niña) over the Philippine Sea in winter (Figure 7). The southwesterly (northeasterly) at the northwest of the anomalous anticyclone (cyclone) leads to an enhanced (weakened) transports of aerosols from South and Southeast Asia to southern China in El Niño (La Niña) winter (Figure 11). As the changes in the winter atmospheric contents of aerosols over southern China caused by ENSO cannot be explained by local emissions or removals, it seems can only be attributed to the changes in the transports of aerosols from South and Southeast Asia to southern China. Zhu et al. (2012) has also pointed out that in determining aerosol concentrations, the changes in monsoon circulation were more dominant than that in precipitation (or wet deposition of particles) in East Asia. It has been revealed statistically in Figure 4c that southern China tended to suffer more (less) haze days than normal in La Niña (El Niño) winter, which was also seen in the 4 selected pairs of El Niño and La Niña winters (Figure S2) with one of them near 2010 (in numerical simulations, the aerosol emissions are fixed in 2010). However, numerical results show that La Niña (El Niño) causes a decrease (increase) in the winter atmospheric contents of aerosols over southern China (Figure 9). Is it possible for southern China to have more (less) haze days but less (more) atmospheric contents of aerosols than normal in La Niña (El Niño) winter? To answer this question, the frequency distributions of the simulated winter daily aerosol CONCsur Deleted: that t

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averaged over southern China (21°N-27°N, 104°-118°E, see Figure S1) in the groups of CLI, EL, and LA are plotted in Figure 12. It has been illustrated in section 3.2.1 that the simulated aerosol CONCsur in this study were smaller than observational studies by 1~2 orders of magnitude. Therefore, for calibration, the simulated winter daily aerosol CONCsur, over southern China are amplified by 10 times before plotting the frequency distributions. It is seen from Figure 12 that the frequency distribution of the simulated winter daily aerosol CONCsur over southern China is a little right-skewed in the group of CLI, reaching peak at around 65 µg m⁻³. The frequency distribution of the simulated winter daily aerosol CONCsur over southern China in the group of LA is a little left-shifting compared with that in CLI when aerosol CONCsur is larger than 40 µg m⁻³. The frequency distribution of the simulated winter daily aerosol CONCsur over southern China in EL is a little right-shifting compared with that in CLI. The average numbers of days in winter with aerosol CONCsur bellow 40, between 40~80, and above 80 µg m⁻³ in CLI, EL, and LA are also given in the top right corner of Figure 12. It is found that the number of days in winter with aerosol CONCsur between 40~80 µg m⁻³ is the largest in LA and smallest in EL. Whereas, the number of days in winter with aerosol CONCsur above 80 µg m⁻³ is the largest in EL and smallest in LA. Figure 12 indicates that southern China tends to have less clean and heavy but more moderate haze days than normal in La Niña winter. Whereas, in El Niño winter, southern China tends to have more heavy but less clean and moderate haze days than normal. This explains why southern China has less (more) haze days but more (less) atmospheric contents of aerosols than normal in El Niño (La Niña) winter. It should be noted that here we use regional mean aerosol CONCsur over southern China subjected to the low model spatial resolution, which is the reason why the curves in Figure 12 are close to each other. In the real practice, however, haze days and their severities are judged station by station (e.g., Figure 2), and the thresholds are not necessarily the same with what we use in Figure 12. Therefore, we can get some rough information about the effects of ENSO on the winter haze days over southern China from the shifts of curves in Figure 12, but cannot compared it with observational data directly.

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It is depicted in Figure 11 that under the emission level of aerosols for the year 2010, the enhanced (weakened) transports of aerosols from South and Southeast Asia to southern China caused by El Niño (La Niña) is the main reason for the increase (decrease) in the atmospheric contents of aerosols over southern China in El Niño (La Niña) winter. Therefore, it is expected that when the emissions of aerosols over South and Southeast Asia diminish in the future, the contradiction between the

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influences of ENSO on the winter haze days and atmospheric contents of aerosols over southern China will also disappear.

4 Conclusions and discussions

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The effects of ENSO on the winter haze days and atmospheric contents of aerosols over China are discussed statistically and numerically. Statistical results show that southern China tends to have less (more) haze days than normal in El Niño (La Niña) winter, which is in line with the simulated more (less) winter precipitation over southern China caused by El Niño (La Niña). Statistical results indicate that the relationships between the winter haze days over northern and eastern China and ENSO are not significant. Numerical results also reveal that the influences of ENSO on the winter atmospheric contents of aerosols over northern and eastern China are not that obvious as over southern China. As a tropical phenomenon, it seems that ENSO affects the winter haze pollution over southern China more significantly than it does over northern and eastern China.

Numerical results indicate that the atmospheric contents of aerosols over southern China are more (less) than normal in El

Niño (La Niña) winter, which is to some extent not in line with the effects of ENSO on the winter haze days of the region. In 2010, South and Southeast Asia have become important source areas of anthropogenic aerosols (Zhang et al., 2016). The enhanced southwesterly (northeasterly) at the northwest of the winter anomalous anticyclone (cyclone) over the Philippine Sea caused by El Niño (La Niña) enhanced (weakened) the transports of aerosols from South and Southeast Asia to southern China. The frequency distributions of the simulated daily surface concentrations of aerosols in winter over southern China indicate, that the region tends to have more heavy (moderate) haze days, but less clean and moderate (heavy) haze days than normal in El Niño (La Niña) winter. But it should be noted again that the emission data of aerosols used in our study are fixed in 2010, and if the emissions of aerosols change the story may be different.

Many studies have found that the haze pollution over northern and eastern China was influenced by the EAWM, as we have cited in section 1. And previous studies also found that ENSO could influence the strength of the EAWM, as we have discussed in section 3.2.2 that El Niño can weaken the EAWM. It is expected that there will be some connection between ENSO and the winter haze pollution over northern and eastern China. However, the connection between ENSO and the winter haze pollution over northern and eastern China is not strong, e.g., the correlations between the winter haze days of the two regions and ENSO are not significant. This makes us to think the way how haze pollutants over the north part of China are cleared away in winter.

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Usually, a few clean days in the north part of China follow a breakout of a block over high latitude Asia, during which a stream of cold air sweeps southwardly and rapidly from the Siberia or Mongolia. Therefore, we can get some information about the influence of ENSO on the winter haze pollution over the north part of China from the frequency of cold air recorded in China in El Niño and La Niña winters. In the Decembers of 1986 (during the 1986/1987 El Niño event) and 2010 (during the 2010/2011 La Niña event), cold air happened in China both for 7 times, the maximum of the same month during 1960–2011 (Figure 5S). This indicates that El Niño (La Niña) does not necessarily reduce (increase) the cold air frequency of China in winter, even though it can weaken (strengthen) the EAWM. To the contrary, it seems that El Niño (La Niña) is in favor of (not in favor of) the cold air frequency of China in winter (Figure 6S). The relationship between ENSO and the cold air frequency of China more complicated than it is expected simply from the perspective of the EAWM. The cold air frequency of China in winter is also influenced by the upstream conditions in the Atlantic Ocean, the Arctic ice, and the Eurasian snow cover (not shown). This emphasizes the importance of exploring the comprehensive effects of ENSO and the extratropical systems on the haze pollution of China.

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Haze pollution is a very sophisticated problem, because it is the comprehensive result of human activities and weather conditions. Weather conditions determine the dispersions and removals of haze particles over a specific region, influence the complex chemistry reactions among different components, and also connect the haze pollution of a specific region with the emissions of neighboring regions. To be more complicated, haze pollution and weather conditions interact with each other closely over some areas. This work explores the effects of ENSO on the winter haze pollution over China under relatively simple experimental settings, with prescribed SST and fixed aerosol emissions, which means that SST does not response to aerosols. Besides, the chemistry reactions in the model we used is also simplified, especially without the complex reactions related to nitrate aerosols. Therefore, studies with more sophisticated experimental designs (e.g., with atmosphere and ocean coupled models) and chemistry schemes are still in need in the future as to the topic discussed in this work.

Author contributions. The work was done under the guidance of Hua Zhang. Shuyun Zhao and Hua Zhang designed the experiment, and prepared the manuscript. Bing Xie conducted model simulations. The analysis of results and preparation of all figures and tables were done by Shuyun Zhao.

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Tables and Figures

Table 1. Simulation set-up.

Group name	SST	Running time	Output frequency
CLI	Climatologic SST	Oct. ⁰ -Aug. ¹	Monthly & Daily
EL	Climatologic SST + Δ SST _{El Niño}	Oct. ⁰ -Feb. ¹	Monthly & Daily
LA	Climatologic SST + Δ SST _{La Niña}	Oct. ⁰ –Feb. ¹	Monthly & Daily

The superscripts of the 3rd column: 0 and 1 represent the 1st and 2nd model year, respectively.

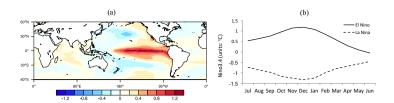


Figure 1. (a) Typical ENSO mode (units: °C/°C) and (b) average monthly Niño3.4 (units: °C) of 21 El Niño and 18 La Niña events from 1951 to 2015.

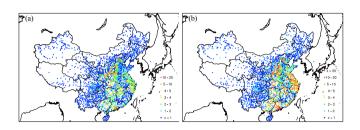


Figure 2. The winter-average monthly haze days (units: days/month) during the years of (a) 1960-2013 and (b) 2000-2013 over main

land China.

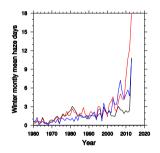
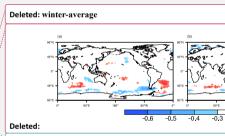


Figure 3. Time series of the winter-average monthly haze days (units: days/month) of JJJ (black), JZH (red), and GG (blue).

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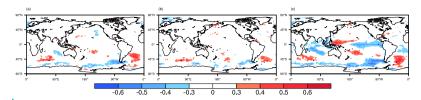


Figure 4. Correlation coefficients (unitless) of the monthly haze days of (a) JJJ, (b) JZH, and (c) GG with SST in winter, after applying a band-pass filtering of 2-8 years to both the data of haze days and SST. Shade denotes that results pass 95% significance level.

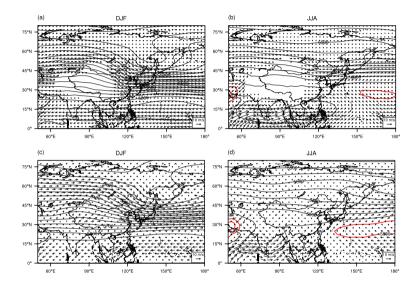


Figure 5. Comparisons between (a-b) the simulated and (c-d) reanalysis winter and summer geopotential height at 500 hPa (contour, units: gpm) and wind at 850 hPa (vector, units: m s⁻¹), with the blank places in (a) and (b) are because of the influence of the Tibetan Plateau. Model results are from the group of CLI, and reanalysis data are from NCEP.

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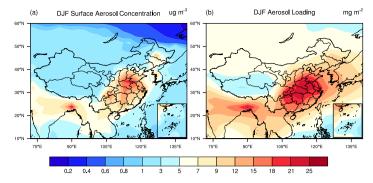


Figure 6. The simulated <u>winter-average</u> (a) surface aerosol concentrations (units: µg m⁻³) and (b) aerosol loadings (units: mg m⁻²) in

the group of CLI.

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differences.

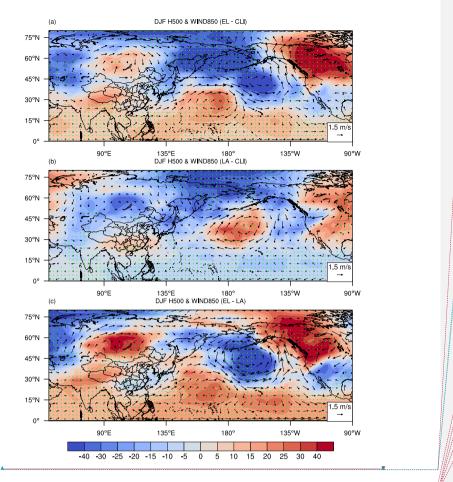
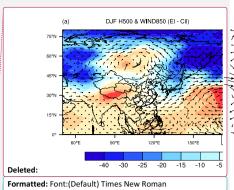


Figure 7. Medians of the simulated differences in winter-average geopotential height at 500 hPa (H500, shaded, units: gpm) and wind at 850 hPa (WIND850, vector, units: m s⁻¹) between (a) EL and CLL (b) LA and CLL and (c) EL and LA, with green dots indicating that the differences of H500 between more than 70% pairs of ensemble members have the same sign with the median

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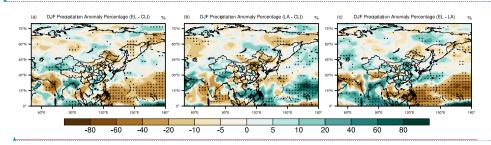


Figure 8. Medians of the simulated differences in winter-average precipitation in percentage (units: %) between (a) EL and CLL_(b)

LA and CLI, and (c) EL and LA, with black dots indicating that the differences between more than 70% pairs of ensemble members

have the same sign with the median differences.

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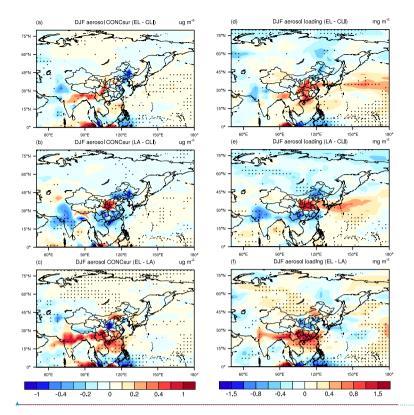


Figure 9. Medians of the simulated differences in winter-average aerosol surface concentrations (left panel, units: µg m-3) and aerosol loadings (right panel, units: mg m,2) between (a)-(d) EL and CLI, (b)-(e) LA and CLI, and, (c)-(f) EL and LA, with black dots indicating that the differences between more than 70% pairs of ensemble members have the same sign with the median differences.

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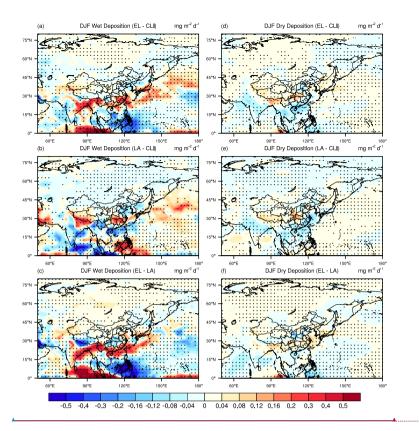


Figure 10. Medians of the simulated differences in winter-average wet depositions (left panel) and dry depositions (right panel) of aerosols (units: mg m⁻² d⁻¹) between (a)-(d) EL and CLL (b)-(e) LA and CLI, and (c)-(f) EL and LA, with black dots indicating that the differences between more than 70% pairs of ensemble members have the same sign with the median differences.

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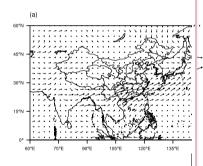
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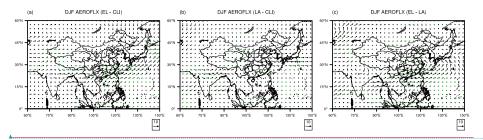
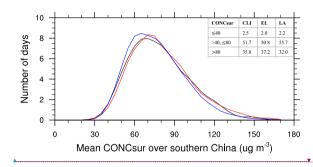


Figure 11. Medians of the simulated differences in the winter-average vertical integral of aerosol horizontal fluxes (AEROFLX, units:

kg m⁻¹ s⁻¹) between (a) EL and CLL_w(b) LA and CLI, and (c) EL and LA_w with green dots indicating that the differences of the zonal components of AEROFLX between more than 70% pairs of ensemble members have the same sign with the median differences.



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Figure 12. Frequency, distributions of the simulated winter daily surface aerosol concentrations (CONCsur, units: μg m_s⁻³) averaged over southern China (after being amplified by 10 times), with black, red and blue lines representing the results from CLI, EL, and LA, respectively; The average numbers of days in winter with CONCsur ≤ 40, 40 <CONCsur ≤ 80, and CONCsur>80 in CLI, EL, and LA are given in the table at the top right corner,

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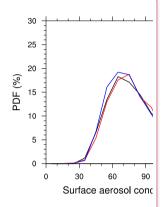
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