Response to Reviewer #1

Overall I am happy with how the authors have addressed my comments. Some of the overstatement about "seasonal prediction" and the assumption that the Ozone Weather Index (OWI) \approx surface ozone pollution have been removed. The authors have also dedicated more discussion to the meteorological links among the sea ice, Eurasia teleconnection pattern (EU), and the OWI. The addition of CAM5 numerical simulation experiments (and Figure 12) also adds confidence in the authors' findings on the links among sea ice, the EU pattern, and ozone-related weather conditions. Sections 4 and 5 have been extended and are much clearer. I am also pleased to see several of the supplementary figures moved to the main paper. This makes the paper easier to follow and makes more organizational sense.

Recommendation:

The authors have addressed my major concerns with the previous version. Aside from a couple specific comments and minor technical corrections, **I find this paper acceptable for publication in ACP.** The technical corrections were numerous so the English in the paper should be carefully reviewed once more by the authors and copy editor before publication.

I'll suggest a slight title change because the current one is a bit awkward, but I think the authors have the right idea: "Climate variability links among Arctic sea ice, Eurasia teleconnection pattern and summer surface ozone pollution in North China"

Reply:

Referring to the reviewer's comment, the title was revised to "Links of Climate variability among Arctic sea ice, Eurasia teleconnection pattern and summer surface ozone pollution in North China".

<u>Revisions:</u>

Links of Climate Variability among Arctic sea ice, Eurasia teleconnection pattern and

summer surface ozone pollution in North China

Specific/Minor Comments:

Line 243: What is "each September 1st?" Meteorological data from September 1st on each of the last 5 years? What 5 years? Please be clear here.

<u>Reply:</u>

Related presentations were modified.

<u>Revisions:</u>

.....During the control experiment (CTRL), the CAM5 model firstly integrated 20 years with climate mean initial and boundary conditions. Next, the data in 1st September of the last 5 years (i.e., 16–20 years) were designated as five slightly different initial conditions. With each initial condition, the CAM5 model integrated 10 years.....

Line 260: Could you explain your "generalized additive model" more in the text? How do you construct the model that gives you the red line in Figure 11?

<u>Reply:</u>

The "generalized additive model" is introduced in Section 2 and a necessary reference is added.

<u>Revisions:</u>

.....The generalized additive model, a data-driven method, is particularly effective at handling the complex nonlinear and non-monotonous relationships between the dependent variable and the independent variables (Hastieand Tibshirani, 1990). This approach used a smoothing function, determined by the independent variables themselves, to transform the expressions, and addressed the dependent variable with different probability distributions by the link function.....

Hastie, T. J. and Tibshirani, R. J.: Generalized Additive Models, Chapman & Hall, London, UK, 1990.

Technical Corrections:

Line 12: "Based on..."

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....Based on the close relationships.....

Line 12: Be sure to subscript the O3 in all instances.

<u>Reply:</u>

Throughout the manuscript, the neglects were corrected.

<u>Revision:</u>

.....Basing on the close relationships between the O_3 concentration and the meteorological conditions.....

Line 21: Change "specially" to "specifically"

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

 \dots variation of O₃ pollution, specifically the related meteorological conditions.....

Line 24: I preferred the original "increasing" to "serious." This sentence could be clarified by writing: "Over the past several decades, along with social and economic development, air pollution has been increasing in China."

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....Over the past several decades, due to fast economic development, air pollution has been increasing in China.....

Line 26-28: "haze pollution is being controlled..." "appearing as a sharp decrease in fine particulate matter."

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....haze pollution is being controlled in recent years (The environmental statistics unit of stat-centre in Peking University, 2018), appearing as a sharp decrease in fine particulate matter.....

Line 28: Change "always occurred on" to "always occurs on"

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

Revisions:

.....which always occurs on clear and sunny days.....

Line 29-30: It is unclear what you mean by "The negative effects of surface O3 pollution was not weaker than those of haze..." It is commonly known that particulate matter pollution is generally more harmful to human health than ozone pollution.

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....The negative effects of surface O_3 pollution, such as corroding human's lungs and destroying agricultural crops and forest vegetation, were not weaker than those of haze.....

Line 42: delete the comma on this line.

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....northern Europe through the anomalous atmospheric circulations.....

Line 43: Change "to" to "that"

Reply:

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....anomalous atmospheric circulations that influence regional photochemical processes.....

Line 46-47: Suggested rewrite "A strong positive correlation also exists between the East Asian summer monsoon and summer mean ozone in China."

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....A strong positive correlation between the East Asian summer monsoon and summer mean ozone were found by model simulations (Yang et al., 2014), illustrating that the changes in meteorological parameters, associated with East Asian summer monsoon, lead to 2–5% interannual variations of surface O_3 concentrations over central eastern China.....

Line 51: Suggested rewrite: "Local photochemical production was the main source of O3."

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....The photochemical reaction was the main local sources of O_3 (Sun et al., 2019).....

Line 52: Change "violent" to "intense"

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....the intense solar radiation.....

Line 66: Change "specially" to "specifically"

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....specifically the related meteorological conditions.....

Line 68-69: I think this sentence is unnecessary, or could be moved elsewhere to a more appropriate location in the text.

<u>Reply:</u>

According to the reviewer's comment, the sentence was moved to the ending of this paragraph.

Revisions:

The observation duration of the surface O₂ concentration was much shorter than the meteorological measurements and could not support the elimate analysis. The hourly O₃ concentration data from 2014 to 2017 in China were provided by the Ministry of Environmental Protection of China. As one of the three regional background air-monitoring stations in China, the hourly O₃ concentration data at the <u>Shangdianzi</u> station (SDZ: located at 40°39'N, 117°07'E and 293.3 m high) was continuously observed from 2006 to_2017 and were controlled by the National Meteorological Information Center, China Meteorological Administration. According to the Technical Regulation on Ambient Air Quality Index of China (the Ministry of Environmental Protection of China, 2012), the maximum daily average 8 h concentration of ozone (MDA8) was used to represent the daily O₃ conditions. The MDA8 was calculated as the maximum of the running 8 h mean O₃ concentrations during 24 hours in the day. <u>However, the systematic observation duration of the surface O₃ concentration was much shorter</u> than the meteorological measurements and could not support the climate analysis.^w

Line 71: Change "high" to "amsl" for above mean sea level.

<u>Reply:</u>

According to the reviewer's comment, the sentence was revised.

<u>Revisions:</u>

.....(SDZ: located at 40°39'N, 117°07'E and 293.3 m amsl).....

Line 72: Need a space between "to2017"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....from 2006 to 2017.....

Line 76: In your response to my review you indicated that you were using sea ice area, but here you cite sea ice "concentrations." Please be clear about which data set you are using.

<u>Reply:</u>

The downloaded data from Met Office Hadley Centre was sea ice concentration. When calculating the sea ice index, we first computed the sea ice area and then calculate the mean value.

Line 88: "Hadley Centre"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....Met Office Hadley Centre.....

Line 91: "and vice versa"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....and vice versa.....

Line 101: "Hadley Centre"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....Hadley Centre.....

Line 108: "severe"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....the threshold of the severe surface O₃ pollution in China.....

Line 119: Suggested rewrite: "..., the covariation of SDZ MDA8 and MDA8 in North China strengthens the representativeness of SDZ for North China." <u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....the covariation of SDZ MDA8 and MDA8 in North China strengthens the representativeness of SDZ for North China.....

Line 124: I still find "non-surface" to be confusing. Perhaps make this "non-polluted surface O3" (NPO) and "moderately-polluted surface O3" (MPO)? <u>*Reply*</u>:

We revised the definition into "non-O₃ polluted (NOP) level" at surface and "moderate-O₃ polluted (MOP) level".

Revisions:

.....we defined non-O₃ polluted (NOP) level at surface as the O₃ concentration $< 100 \ \mu g/m^3$ and moderate-O₃ polluted (MOP) level with O₃ concentration $> 215 \ \mu g/m^3$, respectively.....

Line 127: Replace "were not the NOP days" with "exceeded the NOP threshold," Also see my comment about this acronym.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

......75% of summer days exceeded the NOP threshold even at the regional background......

Line 133: Replace "During calculating" with "During the calculation of". The rest of this sentence is unclear and needs to be rewritten.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....The regional average meteorological elements were calculated as meteorological indexes, and here the selected regions determined on the most significantly different areas in the composites of MOP and NOP events in Figure 2.....

Line 139: Replace "was" with "were"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....the O₃ precursors in North China were dispersed.....

Line 142: Replace "the photochemical reaction" with "photochemistry"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....weakened the photochemistry by influencing exposure.....

Line 151: Replace "enlarged" with "enhanced"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....into the boundary layer enhanced the surface O3 concentration.....

Line 156: "(Figures S2 and S3)"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....(Figures S2, S3).....

Line 177: Delete ";even the MDA8 was the minimum"

Reply:

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

and could verify the performance of the OWI. The JJA mean OWI in 2006 successfully reflected the variation in observed MDA8; even the MDA8 in 2006 was the minimum, confirming the robustness of the OWI. Derived from two different

Line 179: Replace "kinds of reanalysis data." with "specific reanalysis data set." *Reply:*

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....did not depend on the specific reanalysis data.....

Line 213: Delete "a harsh and" and replace "reaction" with "production"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....supported efficient photochemical production of O3.....

Line 214: Replace "by" with "with"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

Revisions:

.....with the NCEP/NCAR data.....

Line 215: "The correspondence between the large-scale..."

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

Revisions:

.....The correspondence between large-scale EU teleconnection, and anti-cyclonic circulations were clear.....

Line 223: "responses like the EU pattern..."

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....responses like the EU pattern.....

Line 224: Replace "monthly checked" with "evaluated each month."

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....The correlation between the sea ice and JJA OWI was evaluated each month (Figure omitted).....

Line 228: Replace "anomalies may be" with "anomalies are"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....May SI anomalies are followed by.....

Line 241: Replace "contributed" with "contributing"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

Revisions:

.....the preceding May sea ice anomalies contributing to the subsequent.....

Line 242: Replace "firstly integrated 20 years" with "was first integrated for 20

years"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....the CAM5 model was first integrated for 20 years with.....

Line 246: Replace "i.e., totally 30 sensitive runs." With "i.e. a total of 30 sensitivity runs."

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....i.e., a total of 30 sensitivity runs......

Line 247: Replace "of 30 sensitive ensembles" with "of the 30 sensitivity runs"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

Revisions:

.....of the 30 sensitivity runs.....

Line 248: Replace "were the" with "represent the"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....The differences (LowASI minus CTRL) represent the responses of.....

Line 253: Replace "violent" with "intense"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....intense sunshine.....

Line 253-254: Replace "the photochemical reaction" with "photochemical production"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....the photochemical production was significantly decelerated.....

Line 254: Replace "On the other side," with "Additionally,"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....Additionally, sufficient moisture.....

Line 256: Replace "but" with "and"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....was weakened, and the wet deposition effect.....

Line 263: Replace "NOAA" with "NCEP/NCAR"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....the ERA-Interim and NCEP/NCAR reanalysis datasets.....

Line 268: Replace "seasonal look" with "understanding of seasonal variability"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....to the understanding of seasonal variability of O3 pollution.....

Line 272: "less low- and medium-altitude cloud cover"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....less low- and medium-altitude cloud cover.....

Line 273: Replace "reaction" with "reactions"

Reply:

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....photochemical reactions.....

Line 276: Replace "casually" with "causally"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

Revisions:

.....and was causally verified by.....

Line 283: Replace "the photochemical reaction" with "photochemistry"

Line 284: Replace "the atmospheric" with "atmospheric"

<u>Reply:</u>

According to another reviewer's comment, this sentence was removed.

Figure S1 Caption: Delete "were" in "were belonged"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

..... the first lines belonged to JJA 2007 and JJA 2008......

Figure S2 Caption: Replace "NOAA" with "NCEP/NCAR"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revisions:</u>

.....calculated using the NCEP/NCAR datasets.....

Figure S3 Caption: Replace "basing" with "based" and replace "NOAA" with "NCEP/NCAR"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

Revisions:

Figure S3. Differences in the boundary layer height between the MOP and NOP events during 2007–2014, based on the ERA-Interim (a) and NCEP/NCAR datasets (b). The black dots denote that the composite passed the 95% confidence level. The boxes represent the area to calculate the daily OWI. These composites were calculated using the NCEP/NCAR datasets.

Figure S4-S6 Caption: Replace "NOAA" with "NCEP/NCAR

<u>Reply:</u>

The neglects were corrected.

Response to Reviewer #2

The authors have greatly improved the manuscript responding to the three reviewers' comments. I believe this paper is suitable for publication after minor revisions.

1. My two main concerns are that (1) the NOP and MOP need to be clearly defined as a range of O3 concentrations. (2) This is not clear to me and thus how Figure 2 is calculated is not clear to me either.

<u>Reply:</u>

(1) The NOP and MOP was defined as a range of O₃ concentrations now, that is, defining non-surface O₃ polluted (NOP) level as the O₃ concentration $< 100 \ \mu g/m^3$ and moderate surface O₃ polluted (MOP) level with O₃ concentration $> 215 \ \mu g/m^3$.

(2) The composite results were calculated as the differences between MOP or NOP events with the rest events (i.e., all events but excluded MOP and NOP events). The calculations were illustrated in the caption of Figure 2 in the revised version.

<u>Revision:</u>

.....we defined non-O₃ polluted (NOP) level at surface as the O₃ concentration $< 100 \ \mu\text{g/m}^3$ and moderate-O₃ polluted (MOP) level with O₃ concentration $> 215 \ \mu\text{g/m}^3$, respectively.....

Caption of Figure 2The composite results were calculated as the differences between MOP or NOP events with the rest events (i.e., all events but excluded MOP and NOP events).....

I am troubled by two statements in the conclusion section:

(1) If "the processes how the weather conditions impact the photochemical reaction were not deeply discussed here" then I do not understand what was shown in this paper. (2) "However, the reason why the cooler high troposphere contributed to the surface ozone pollution was still an open question and needed further attention". If this is true, then why did the two temperature fields get

compared in the first place? What meteorological understanding would lead this to be shown in Figure 2?

<u>Reply:</u>

According to the reviewer's comment, the related sentences were removed to avoid unwanted confusions.

<u>Revision:</u>

The joint effects of the climate anomalies and the historical emissions should be lucubrated using the numerical models in the future. The processes how the weather conditions impacted the photochemical reaction were not deeply discussed here and have been analyzed in many previous studies by the atmospheric chemists. However, the reason why the cooler high troposphere contributed to the surface ozone pollution was still an open question and needed further attention. The EU pattern was a well-known continental <u>Rossby</u> wave train and could link the mid-high latitude climate with the change of the

Two additional references that have recently been published may be of interest to the authors to include in their introduction:

Int J Climatol.

Inter-annual variation of the spring haze pollution over the North China Plain: Roles of atmospheric circulation and sea surface temperature By Chen S, Guo J, Song L, Li J, Liu L, Cohen JB. https://rmets.onlinelibrary.wiley.com/doi/10.1002/joc.5842 ACP

Impacts of meteorology and emissions on summertime surface ozone increases over central eastern China between 2003 and 2015 by Lei Sun, Likun Xue, Yuhang Wang, Longlei Li, Jintai Lin, Ruijing Ni, Yingying Yan, Lulu Chen, Juan Li, Qingzhu Zhang, and Wenxing Wang https://www.atmos-chem-phys.net/19/1455/2019/

<u>Reply:</u>

The recommended publication related to O₃ pollution was cited in this manuscript.

The other paper, discussing the haze pollution, was also interesting and was cited in another manuscript of the authors, whose topic was the haze pollution.

Minor and Technical Comments:

Line 10: define how many years is meant by "recently"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....Summer surface O₃ pollution has rapidly intensified in China in the recent decade, damaging.....

Line 12: Make sure O3 has a subscript 3 everywhere

<u>Reply:</u>

Throughout the manuscript, the neglects were corrected.

<u>Revision:</u>

 \dots Basing on the close relationships between the O₃ concentration and the meteorological conditions.....

Line 14: To have both "major" and "significant" is too much. I recommend removing "significant"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....a major globally atmospheric teleconnection pattern.....

Line 24: This opening sentence does not make sense to me. Has social and economic development been serious in China?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....Due to fast economic development, air pollution has been serious in China.....

Line 25: Explain "haze" pollution. Is this from particulate matter or from a mix of other pollutants as well?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The major air pollution types in China are haze pollution (i.e., high-level fine particulate matter) in winter.....

Line 28: Change decreasing to decrease.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....appearing as sharp decrease in.....

Line 28: Why? Are the controls specific to winter or only targeting what might impact haze? I could imagine some controls might have also helped the O3 precursors.

<u>Reply:</u>

In the new publication (Li et al., 2018), Li and the co-authors pointed out "The most important cause of the **increasing ozone in NCP appears to be the decrease in PM2.5**, slowing down the sink of **hydroperoxy radicals** and thus speeding up ozone production". Thus, "Decreasing ozone in the future will require a combination of NO_x and VOC emission controls to overcome the effect of decreasing $PM_{2.5}$."

Ref: Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q., Bates, K. H.: Anthropogenic drivers of 2013–2017 trends in summer surface ozone in China, P NATL ACAD SCI USA., https://doi.org/10.1073/pnas.1812168116, 2018

Line 29: Be specific, are "the negative effects of surface O3 pollution" regarding human health, agriculture, economy, etc?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The negative effects of surface O_3 pollution, such as corroding human's lungs and destroying agricultural crops and forest vegetation, were not weaker than those of haze.....

Line 32: remove "i.e."

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....the surface O_3 concentrations exceeded the ambient air quality standard of China (100 μ g/m³) by 100–200 %.....

Lines 33-34: Why is this important? No explanation of ozone precursors, authors should consider explaining what those are before this statement.

<u>Reply:</u>

According to the reviewer's advice, the explanation on the ozone precursors was forward.

<u>Revision:</u>

.....Furthermore, the concentration of O_3 and its precursors, e.g. nitrogen oxides (NO_x) and volatile organic compounds (VOCs), in Beijing-Tianjin-Hebei.....

Line 35: Why are both O3 and the precursors large?

<u>Reply:</u>

In the cited references, the authors only stated the observed variations from the measurements, but did not talk about the reasons for both O_3 and the precursors became large.

In our opinion, due to the increasing of precursors emissions, the O_3 production enlarged by consuming the precursors. However, the consumption cannot counteract the fast emission by human activities.

Line 39: Define NOx and VOC. I would also suggest adding "with sunlight" after "react"

<u>Reply:</u>

According to the reviewer's advice, the explanation on the ozone precursors was forward, and the sentence was revised.

<u>Revision:</u>

.....Furthermore, the concentration of O_3 and its precursors, e.g. nitrogen oxides (NO_x) and volatile organic compounds (VOCs), in Beijing-Tianjin-Hebei.....

 \dots photochemically react with sunlight to generate O₃ under suitable weather conditions.....

Line 47: Add references for sentence ending in "mean ozone existed".

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....A strong positive correlation between the East Asian summer monsoon and summer mean ozone were found by model simulations (Yang et al., 2014), illustrating that the changes in meteorological parameters, associated with East Asian summer monsoon, lead to 2–5% interannual variations of surface O₃ concentrations over central eastern China.....

Line 51: The sentence "The photochemical reaction was the man local sources of O3" is in reference to which paper? And with NOx or VOCs?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The photochemical reaction was the main local sources of O_3 (Sun et al., 2019).....

Line 61: Do the authors mean "previous studies of O3 pollution in China mainly focused on....."? Consider adding "of O3 pollution in China" or some other description of these previous studies.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....previous studies of O_3 pollution in China mainly focused on observational analyses of several synoptic processes (e.g., Zhao and Wang, 2017),.....

Line 62: Is there a "lack of long-term surface O3 observations" at all or publicly available?

<u>Reply:</u>

At all. The network was only established in recent years.

Line 65: change "was" to "is"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....as a preceding and effective driver, is also analysed.....

Line 66: The authors may want to stress here that this may have possible seasonal forecast application.

<u>Reply:</u>

In the revised version, we toned down the contents about the seasonal prediction.

Line 67: Can I suggest adding subsections 2.1 Observation-based data (Line 68) and 2.2 Modelbased data (Line 78) or something like? And maybe a section 2.3 Teleconnection before Line 89

<u>Reply:</u>

Secondary headings are infrequent in the ACP format, thus the structure maintains itself.

Lines 68, 69 and 72: I am confused by the terminology of "observation duration of the surface O3 concentration was much shorter than the meteorological measurements". Is the "much shorter" in relation to "the hourly O3 concentration data from 2014 to 2017, " or the Shangdianzi station "observed from 2006 to 2017"? Eleven years of data versus four years of data is definitely a different story for short term vs long-term.

<u>Reply:</u>

What we wanted to present is that the network of ozone observation, not that in the single Shngdianzi site. The sentence was revised.

<u>Revision:</u>

.....The systematic observation duration of the surface O_3 concentration was much shorter than the meteorological measurements.....

Line 72: make sure to add a space between "to2017".

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....from 2006 to 2017.....

Line 76: The sea ice data should not be in the same paragraph as the ozone data. Can the authors make it a new paragraph and add more details about this dataset such as the time period it covers, is this a widely used data set at that resolution?

<u>Reply:</u>

According to the reviewer's advice, the sea ice data was carefully introduced.

<u>Revision:</u>

.....The monthly sea ice concentrations $(1^{\circ} \times 1^{\circ})$ were downloaded from the Met Office Hadley Center (Rayner et al. 2003), which are widely used in sea ice-related analysis. The sea ice fields are made more homogeneous by compensating satellite microwave-based sea ice concentrations for the impact of surface melt effects on

retrievals in the Arctic, and by making the historical in situ concentrations consistent with the satellite data. The gridded sea ice data was available from 1870 to date, and those during 1979 to 2018 were extracted here.....

Line 78: Give more detail about the ERA-Interim dataset. It is available from 1979-present. Besides the horizontal resolution, can the authors provide the vertical resolution of the data used.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....air temperature from 1000 hPa to 100hPa.....

.....The daily mean and monthly mean ERA-Interim data from 1979 to present were directly downloaded from the ERA-Interim website.....

Line 83: Give more detail about the NCEP/NCAR dataset. It is available from when to when, vertical resolution of the dataset, etc?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....air temperature at from 1000 hPa to 100hPa.....

.....were downloaded, which was available from 1948 to present......

Line 86: The BLH is not from the NCEP/NCAR dataset but from a different reanalysis altogether. Describe the NOAA-20CR reanalysis (horizontal and vertical resolution, period of coverage, how it is different to NCEP/NCAR).

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The BLH dataset was only available from 1979 to 2014 in the website of the NOAA-CIRES 20th Century Reanalysis version 2c.....

Line 88: Consider moving daily precip data to the 2.1 Observation section if it is an observation data set and not a model dataset.

<u>Reply:</u>

Secondary headings are infrequent in the ACP format, thus the structure maintains itself.

Line 91: I do not care for the use of "vice versa" used throughout the paper instead of writing it out what the alternative conditions may be. Perhaps this is a technical edit to discuss with ACP.

<u>Reply:</u>

After checking some published paper in ACP, the "vice versa" was maintained.

Line 93: Consider adding "for summertime" following "the calculation procedure for the EU index here"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....the calculation procedure for the EU index for summertime was consistent with.....

Line 95: Give all equations numbers. This would then be Equation 1 and the authors can reference it in the paper as such.

<u>Reply:</u>

According to the reviewer's advice, it was revised.

Revision:

$$EU index = [-1 \times H500_{(70-80^{\circ}N,60-90^{\circ}E)} + 2 \times H500_{(45-55^{\circ}N,90-110^{\circ}E)^{+}}]$$

 $-1 \times \overline{\text{H500}}_{(35-45^{\circ}N, 120-140^{\circ}E)}]/4$ (1)

Line 100: What is the top of the atmosphere for this model?

<u>Reply:</u>

.....CAM5.3 uses vertical hybrid δ -pressure coordinates including 26 layers with the top located at about 3.5 hPa.....

<u>Revision:</u>

.....CAM5.3 uses vertical hybrid δ -pressure coordinates including 26 layers with the top located at about 3.5 hPa.....

Line 107: I think change "The observations, with" to "Observations with"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....Observations, with maximum MDA8.....

Line 108: Change "server" to "severe"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....threshold of the severe surface.....

Line 109: I do not agree fully with the statement "the ozone polluted region has expanded" as over time the measurement coverage has increased spatially so it is possible the other areas were already polluted but just not observed. Can the authors defend this statement?

<u>Reply:</u>

(1) In Figure 1, the number of sites was almost the same in 2015, 2016 and 2015.(2) Some overlapping sites were not polluted in 2015, but became polluted in 2016 and 2017.

Line 111: In the figure the crosses to indicate the ozone is above the severe threshold of 265 make the circles actually look like a darker shade of red. Is there a reason the authors chose not to use colors based on the thresholds?

<u>Reply:</u>

The colors were used to show the mean MDA8. We want to include more information without a new Figure, thus the crosses was selected.

Line 115: Can SDZ still be considered one of the "background monitoring stations" as the authors argue it represents urban centers?

<u>Reply:</u>

The SDZ station was the background monitoring station. The reason for using the data in SDZ is the long time rang of observation.

Line 121: I do not understand why the large daily difference in MDA8 contradicts the quasiconstant emission of ozone precursors.

<u>Reply:</u>

To avoid the confusion, the sentence was removed.

Line 125: Are NOP levels anything above 100 micrograms per meter cubed or less than 100? Or are NOP levels between 100 to 215 and then MOP is anything above 215? The definition of NOP and MOP are not clear to me and this is VERY IMPORTANT for Figure 2.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....we defined non-O₃ polluted (NOP) level at surface as the O₃ concentration $< 100 \ \mu\text{g/m}^3$ and moderate-O₃ polluted (MOP) level with O₃ concentration $> 215 \ \mu\text{g/m}^3$, respectively.....

Line 129: Why is NOP in brackets?

Reply:

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....Both of the interannual variation in MOP and NOP days was significant at the 95% confidence level.....

Line 129: Do the authors have enough data for a trend analysis?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....without an obvious trend.....

Lines 131-133: This sentence does not make sense. Maybe remove "although" and add "and" after "(Figure 2),"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....Due to the significant covariation between the SDZ MDA8 to the MDA8 in North China, the meteorological conditions were composited for the MOP and NOP days in SDZ (Figure 2) and the results were also appropriate for those in North China.....

Line 132: If the composites are for MOP and NOP conditions, then what are they differenced from to get positive and negative values? It is very important to make sure the NOP and MOP definitions are made abundantly clear. If NOP is anything up to 100 and MOP is anything above 215, then are the authors using the conditions between the two (100 to 215) as the control?

<u>Reply:</u>

(1) The NOP and MOP was defined as a range of O_3 concentrations now, that is,

defining non-surface O₃ polluted (NOP) level as the O₃ concentration $< 100 \ \mu g/m^3$ and moderate surface O₃ polluted (MOP) level with O₃ concentration $> 215 \ \mu g/m^3$.

(2) The composite results were calculated as the differences between MOP or NOP events with the rest events (i.e., all events but excluded MOP and NOP events).

<u>Revision:</u>

.....we defined non-surface O_3 polluted (NOP) level as the O_3 concentration < 100 µg/m³ and moderate surface O_3 polluted (MOP) level with O_3 concentration > 215 µg/m³, respectively.....

Caption of Figure 2The composite results were calculated as the differences between MOP or NOP events with the rest events (i.e., all events but excluded MOP and NOP events).....

Line 133: significant by which test?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The local and surrounding weather conditions were significantly different (t-test).....

Line 135: Insert references to which Figure 2 panel the reader should look at. For example, "The anomalous southerlies (Fig. 2a), higher BLH (Fig. 2c), less rainfall (Fig. 2e), warmer surface air temperature (Fig. 2g), and cooler temperature in the high troposphere (Fig. 2g) favored surface O3 pollution (i.e., MOP conditions)." Remove the vice versa.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The anomalous southerlies (Figure 2a), higher BLH (Figure 2c), less rainfall (Figure 2e), warmer surface air temperature, and cooler temperature in the high

troposphere (Figure 2g) favored surface O₃ pollution.....

Line 139: change was to were and add "in North China" after "surface O3 concentration".

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....the O_3 precursors in North China were dispersed, and the surface O_3 concentration in North China was reduced.....

Line 141: I'd like to suggest that the OWI calculation in line 159 becomes Equation 2 and then wherever the authors "denote" a variable for this equation then Equation 2 is referenced. Such as here would become "denoted as V10mI, Eqn. 2".

<u>Reply:</u>

We tried to modify the paragraph like the comment, but it became disordered. Thus it is maintained.

Line 141: I would also suggest saying the region given in the brackets here is the box in Fig 2a.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....(35–50 N, 110–122.5°E, black box in Figure 2a, denoted as V10mI).....

Line 141: The authors switched gears and started describing the NOP with the sentence "The cloudy skies...." I would suggest consider keeping the MOP description together separate from the NOP.

<u>Reply:</u>

The physical mechanisms for MOP and NOP were the same, just opposite. To be concise, they were analyzed together, instead of separate discussion.

Line 145, 149, 153: state boxed regions are from which panel in Fig 2 and add the Eqn. 2 reference.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....(37.5–42.5 N, 112–127.5 E, black box in Figure 2e, denoted as PI).....

.....(37.5–47.5 N, 110–122.5 E, black box in Figure 2g, denoted as DTI).....

.....(37.5–47.5 N, 112.5–120 E, black box in Figure 2c, denoted as BI).....

Line 148: Explain why one would look for this "difference in the temperature at the surface and 200 hPa". Is it a good coincidence or what are the links in the meteorology for cold air aloft to be expected with clear conditions instead of cloudy conditions? What led the authors to make this plot?

<u>Reply:</u>

In sunny days, no more long-wave radiations were reflected by the cloud, this might be a possible reason.

Line 151: The authors could look at ozone from ERA-Interim, as this dataset has assimilated satellite-retrieved ozone. Otherwise, the authors should state clearly that this is "not shown" in the paper and then reference An et al., 2009. There are other papers which also show this such as Ott et al., 2016, JGR.

<u>Reply:</u>

It is really new work for us to analyze the assimilated satellite-retrieved ozone, but indeed a meaningful suggestion. In our next job, we will try to read the data and analyzed them.

Line 158: Is "long-term" meaning a 10 year period here?

<u>Reply:</u>

It means more than 30 years.

Line 159: As stated before, make OWI as a separate Equation from the text. <u>*Reply:*</u>

Reply to **Line 141:** We tried to modify the paragraph like the comment, but it became disordered. Thus it is maintained.

Line 168: "the historical period before 2007 and the projected future" assumes the O3 emissions are the same as for the 11 year period from 2007 to 2017? *Reply*:

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....Thus, it is reasonable to analyse the variation in **surface O₃-related atmospheric circulations** in North China using the OWI.....

Line 170: This first sentence should be a general one so I would suggest removing the "the" before reanalysis and the "was" before improved.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....the quality of reanalysis data improved......

Line 175: Is it possible to add 2006 to Table S1?

<u>Reply:</u>

The data in 2006 were independent samples, thus we did not analyzed it.

Line 180: "Before the mid-1990s" but looking at the figure is it also after 1983?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....During mid-1980s to the mid-1990s.....

Line 181 and 183: Does "insignificant" in line 181 contradict the "strong" in line 183?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

..... The strong interannual variation in the OWI after mid-1990s,.....

Line 182: Over what time period do the "emissions of O3 precursors increase persistently and linearly"?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....increased persistently and linearly due to the steady economic development after 1978 in China.....

Line 189: Can the authors be more definitive than "appeared to be the positive phase of the EU pattern". Can the authors provide a reference if they can't be certain.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....appeared to be the positive phase of EU pattern (Wang and He 2015).....

Line 196: What does "After 2007, the EU index and the observational SDZ MDA8 synchronously changed" mean? During period with data, one can see good agreement between the JJA EU and O3?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....the EU index and the observational SDZ MDA8 showed good agreement.....

Line 198: Consider changing "intensified" to "large". Also, "homodromous" is an odd choice of word.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....the large EU pattern anomalies (i.e., the |EU pattern index $| > 0.8 \times its$ standard deviation) always induced in-phase surface ozone pollution.....

Line 202: Add (Fig. 7c) after "300 hPa"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....below 300 hPa (Figure 7c).....

Line 210: Add "likely" before facilitated

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....likely facilitated.....

Line 218: Check spelling of NCAR

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....NCEP/NCAR data.....

Line 227: Insert "(SI)" after "sea ice" as SI is used later in that sentence.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....sea ice (SI) area in May.....

Line 232: "could induce EU-like pattern"....so are they the EU or are they something else? Be clear what we see in Fig 10c. Line 236-237 the authors sound very confident that it is the EU pattern.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....These positive sea ice anomalies could induce EU pattern responses in the subsequent summer.....

Line 246: Is ASI for Arctic Sea Ice?

<u>Reply:</u>

In section 1, the abbreviation ASI was pointed to Arctic sea ice.

<u>Revision:</u>

..... The role of May Arctic sea ice (ASI), as a preceding and effective driver.....

Line 253: Should the reference be Fig 12c not 12b?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....cooled the air in the boundary layer (Figure 12c).....

Line 253-254: I think the statement "the photochemical reaction was significantly decelerated and the generation of surface O3 was rather weak" is speculative since this was not specifically shown. It should be stated as such.

<u>Reply:</u>

This statement was linked with Section3.

Line 255: I also think the statement "The wet deposition effect was also significantly enhanced" is speculative since not shown specifically.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....The wet deposition effect might be enhanced......

Line 256: Change "but" to "and"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....and the wet deposition effect.....

Line 257: At the end of this paragraph I am left wondering, What scientific question did this section answer. Make this clear.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....That is, the positive relationship and associated physical mechanisms (i.e., climate links among ASI, EU pattern and summer surface ozone pollution in North China) were causally verified

Line 260: Change "Spatially" to "In general"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

 \dots In general, the O₃ concentrations \dots

Line 261: Is the EU predictable on seasonal time-scales?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

 \dots . To reveal the climatic driver of summer surface O₃ pollution in North China.....

Line 262: Does "long-term" mean 11 years or is it reference to the reanalysis datasets?

<u>Reply:</u>

It is reference to the reanalysis datasets.

Line 262: Add "and ozone" before "observations"

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....was constructed based on meteorological and ozone observations.....

Line 264: Add "which may help for seasonal forecasting" after "climatic driver". <u>*Reply:*</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....a preceding and efficient climatic driver, which may help for seasonal forecasting......

Line 267: This final sentence in this paragraph is what I was looking for at the end of Section 5 (line 257).

<u>Reply:</u>

Line 257 was modified.

<u>Revision:</u>

Line 257:That is, the positive relationship and associated physical mechanisms (i.e., climate links among ASI, EU pattern and summer surface ozone pollution in North China) were causally verified

Line 270: Can the authors pick one "induce" or "enhance" or can the accumulated sea ice in May do both?

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

..... The accumulated sea ice in May could induce the positive EU phase

Line 273: "was accelerated" sounds too bold to me. Maybe "supported" instead? *Reply:*

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....photochemical reaction to produce surface O3 was supported.....

Line 274: "fairly" in fairly high seems weak for something that is over an air quality standard. Can the authors be less vague and more quantitative, possibly linking back to the thresholds used in MOP.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....were continuous to achieve a high concentration.....

Line 275: Perhaps the authors want to add "and O3" before "measurements"

<u>Reply:</u>

Here, the sentence was maintained.

Line 276: "casually verified". I don't think the authors want to describe their work as casual.

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

.....was causally verified.....

Line 277: Describe How the OWI extended the time range of this study.

<u>Reply:</u>

It is needless to expand how the OWI extended the time range, which was detailedly explained in the main text.

Line 282: "lucubrated" is a bit of an odd word

<u>Reply:</u>

According to the reviewer's advice, the sentence was revised.

<u>Revision:</u>

...should be studied using...

Line 283: if "the processes how the weather conditions impact the photochemical reaction were not deeply discussed here" then what did the authors show in this paper?

Line 284: Add references for "many previous studies"

<u>Reply:</u>

According to the reviewer's comment, the related sentences were removed to avoid unwanted confusions.

<u>Revision:</u>

The joint effects of the climate anomalies and the historical emissions should be lucubrated using the numerical models in the future. The processes how the weather conditions impacted the photochemical reaction were not deeply discussed here and have been analyzed in many previous studies by the atmospheric chemists. However, the reason why the cooler high troposphere contributed to the surface ozone pollution was still an open question and needed further attention. The EU pattern was a well-known continental <u>Rossby</u> wave train and could link the mid-high latitude climate with the change of the

Figure 2: Can the authors add MOP and NOP headers above the left and right panels, respectively? Also what are the units for each variable?

Line 474: Remove "s" at the end of "ERA-Interim datasets"

<u>Reply:</u>

(1) the MOP and NOP headers were already indicated by >215 and <100. (2) The units were added in the captions.

<u>Revision:</u>

Figure 2. Composite of the meteorological conditions associated with different O_3 events during 2007–2017. Results for MOP (a, c, e, g) and NOP (b, d, f, h) events included (a–b) surface wind (m/s, arrow) and v-wind (m/s, shading), (c–d) BLH (m), (e–f) precipitation (mm), (g–h) SAT (°C, shading), and temperature at 200 hPa (°C, contour). The black dots denote the composite results passed the 95% confidence level. The boxes represent the area used to calculate OWI. These composites were calculated using the ERA-Interim dataset. The green triangle in panel (a-b) illustrates the location of the Shangdianzi site. The composite results were calculated as the differences between MOP or NOP events with the rest events (i.e., all events but excluded MOP and NOP events).

Figure 7: Shading temp in (b) and (c) might have matched shading in (a) better?

<u>Reply:</u>

Primitively, we plotted the Figures like this comment, but the distributions of relative humidity were not clear in contours. Thus, we changed the relative humidity into shading.

Figure 8: What are the units for the Y-Axis?

<u>Reply:</u>

According to the reviewer's advice, the caption of Figure 8 was revised.

<u>Revision:</u>

Figure 8. The variation in the JJA mean observational SDZ MDA8 ($\mu g/m^3$, blue) and EU index (gpm, red) from 2007 to 2017.

Links of Climate Variability among Arctic sea ice, Eurasia teleconnection pattern and summer surface ozone pollution in North China: in terms of climate

variability

Zhicong Yin^{12*}, Huijun Wang¹², Yuyan Li¹, Xiaohui Ma³, Xinyu Zhang¹

¹Key Laboratory of Meteorological Disaster, Ministry of Education / Joint International Research Laboratory of Climate and Environment Change (ILCEC) / Collaborative Innovation Center on Forecast and Evaluation of Meteorological Disasters (CIC-FEMD), Nanjing University of Information Science & Technology, Nanjing 210044, China ²Nansen-Zhu International Research Center, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, China ³Institute of Urban Meteorology, CMA / Environmental Meteorology Forecast Center of Beijing-Tianjin-Hebei

10 **Corresponding author:* Zhicong Yin (yinzhc@163.com)

Abstract. Summer surface O_3 pollution has rapidly intensified in China recentlyin the recent decade, damaging human and ecosystem health. In 2017, the summer mean maximum daily average 8 h concentration of ozone was greater than 150 µg/m³ in North China. Baseding on the close relationships between the O_3 concentration and the meteorological conditions, a daily surface O_3 weather index was constructed, which extends the study period to the historical period before 2007 and the projected future. Here, we show that in addition to anthropogenic emissions, the Eurasia teleconnection pattern (EU), a major globally significant atmospheric teleconnection pattern, influences surface O_3 pollution in North China on a time scale of climate. The local meteorological conditions associated with the EU positive phase supported intense and efficient photochemical reactions to produce more surface O_3 . The associated southerlies over North China transported surrounding O_3 precursors to superpose local emissions. Increased solar radiation and high temperature during the positive EU phase dramatically enhanced O_3 production. Furthermore, due to the close connection between the preceding May Arctic sea ice and summer EU pattern, approximately 60% of the interannual variability of O_3 -related weather conditions was attributed to Arctic sea ice to the north of Eurasia. This finding will aid in understanding the interannual variation of O_3 pollution, specifically specially the related meteorological conditions.

Keywords: ozone pollution, photochemical reaction, Eurasia teleconnection pattern, climate change, Arctic sea ice

25 1 Introduction

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Along with social and<u>Over the past several decades, due to fast</u> economic development, air pollution has been <u>increasing</u> serious in China (Chen, 2013; Watts et al., 2018). The major air pollution types in China are haze pollution <u>(i.e., high-level fine</u> particulate matter) in winter (Yin et al., 2015; Wang, 2018) and surface ozone (O₃) pollution in summer (Ma et al., 2016; Tang et al., 2018). Due to drastic air pollution control in China since 2013, haze pollutions are is being controlled in recent years (The

- 30 environmental statistics unit of stat-centre in Peking University, 2018), appearing as <u>a</u> sharp decrease ing in fine particulate matter. However, surface O₃ pollution, which always occurroccursed on clear and sunny days (Wang et al., 2017), has not improved (Li et al., 2018). The negative effects of surface O₃ pollution-, such as corroding human's lungs and destroying agricultural crops and forest vegetation, was were not weaker than those of haze (Liu et al., 2018), but the impacts of climate variability on surface O_3 pollution in China (Yang et al, 2014) have not been sufficiently studied. In the major urban 35 agglomerations in China, such as Beijing-Tianjin-Hebei, Yangtze River delta and the Pearl River delta, the surface O_3 concentrations exceeded the ambient air quality standard of China (i.e., 100 µg/m³) by 100–200 % (Wang et al., 2017). In the Yangtze River delta, the inter-annual variations of NO and O₃ levels generally presented decreasing and increasing trends respectively, from 2012 to 2015 at both urban and suburban sites (Tong et al., 2017). Furthermore, the concentration of O_3 and its precursors, e.g. nitrogen oxides (NO_x) and volatile organic compounds (VOCs), in Beijing-Tianjin-Hebei was significantly 40 larger than that in other regions of China (Wang et al., 2006; Shi et al., 2015). Revealed by the datasets from Shangdianzi Station, the long-term trend of O_3 concentrations in North China indicated that the O_3 pollution has undergone a significant increase in the period of 2005–2015, with an average rate of 1.13 ± 0.01 ppb year⁻¹ (Ma et al., 2016).
- Surface O_3 is a secondary pollutant. The precursors of O_3 (e.g. NOx and VOC) photochemically react with sunlight to generate O₃ under suitable weather conditions, i.e., hot-day and sunny environments (An et al., 2009). Surface deposition, dynamic 45 transport and dispersion of O₃ are also closely related to atmospheric circulations. For example, the prevailing positive phase of the North Atlantic Oscillation contributed to the increasing ozone concentration in western and northern Europe, through the anomalous atmospheric circulations to that influence regional photochemical processes (Christoudias et al., 2012; Pausata et al., 2012). The summer surface O₃ variability in North America is significantly modulated by the position of the jet stream (Lin et al., 2014). Barnes and Fiore (2013) pointed out jet position may dynamically modulate surface ozone variability in eastern 50 North America and other northern mid-latitude regions. A strong positive correlation between the East Asian summer monsoon and summer mean ozone-existed were found by-The-model simulations (by-Yang et al., (2014), illustratinged that the changes in meteorological parameters, associated with East Asian summer monsoon, lead to 2-5% interannual variations of surface O₃ concentrations over central eastern China. Focusing on the dataset in 2014, a significantly strong west Pacific subtropical high resulted in higher relative humidity, more clouds, more rainfall, less ultraviolet radiation and lower air temperatures, which 55 were unfavourable for the formation of O_3 (Zhao and Wang, 2017). The photochemical reaction was the main local sources of O₃ (Sun et al., 2019). The hot and dry environments and the violent-intense solar radiation could accelerate the chemical conversion from the precursor to O₃ (An et al., 2009; Tong et al., 2017). In 2013, a severe heat wave, with highest temperature 41.1 °C, contributed to the high O₃ concentration in the Yangtze River Delta (Pu et al., 2017). The frequency of large-scale, extreme heat events is closely related to atmospheric patterns, such as the Eurasia teleconnection pattern (EU; Pu et al., 2017; 60 Li and Sun, 2018) and aerosol effective radiative forcing (Liu and Liao, 2017). The winds from a polluted area also transport

 O_3 and its precursors downwind (Doherty et al., 2013). Due to the close relationship between surface O_3 and meteorological conditions, the impacts of climate change on O_3 have been projected by various numerical models (Doherty et al., 2013; Melkonyan and Wagner, 2013; Zhu and Liao, 2016; Gaudel et al., 2018). Over eastern China, the surface ozone concentration and possibility of severe ozone pollution may both increase in the future (Wang et al., 2013).

However, previous studies of O₃ pollution in China mainly focused on observational analyses of several synoptic processes (e.g., Zhao and Wang, 2017), rather than long-term climate diagnostics, because of the lack of long-term surface O₃ observations. The goal of this study is to examine the large-scale atmospheric circulations associated with the interannual variation of summer surface O₃ pollution in North China based on long-term meteorological observations. The role of May Arctic sea ice (ASI), as a preceding and effective driver, was-is also analysed. The outcomes of our research, in terms of climate variability, may provide a basis for understanding the interannual variation of O₃ pollution, <u>specifically specially</u>-the related meteorological conditions.

2 Data and Method

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not support the climate analysis. The hourly O₃ concentration data from 2014 to 2017 in China were provided by the Ministry of Environmental Protection of China. As one of the three regional background air-monitoring stations in China, the hourly O₃ concentration data at the Shangdianzi station (SDZ: located at 40°39'N, 117°07'E and 293.3 m <u>amslhigh</u>) was continuously observed from 2006 to_2017, and were controlled by the National Meteorological Information Center, China Meteorological Administration. According to the Technical Regulation on Ambient Air Quality Index of China (the Ministry of Environmental Protection of China, 2012), the maximum daily average 8 h concentration of ozone (MDA8) was used to represent the daily O₃ conditions. The MDA8 was calculated as the maximum of the running 8 h mean O₃ concentrations during 24 hours in the day. <u>However, the systematic observation duration of the surface O₃ concentration was much shorter than the meteorological measurements and could not support the climate analysis.</u>

The observation duration of the surface O₃-concentration was much shorter than the meteorological measurements and could

The monthly sea ice concentrations (1°×1°) were downloaded from the Met Office Hadley Centrer (Rayner et al. 2003), which are widely used in sea ice-related analysis. The sea ice fields are made more homogeneous by compensating satellite microwave-based sea ice concentrations for the impact of surface melt effects on retrievals in the Arctic, and by making the historical in situ concentrations consistent with the satellite data. The gridded sea ice data was available from 1870 to date, and those during 1979 to 2018 were extracted here.

The $1^{\circ} \times 1^{\circ}$ ERA-Interim data used here included the geopotential height (Z), zonal and meridional wind, relative humidity,

90 vertical velocity, air temperature at different pressure levels from 1000 hPa to 100hPa, boundary layer height (BLH), surface air temperature (SAT) and wind, downward UV radiation, downward solar radiation, low and medium cloud cover and precipitation (Dee et al. 2011). The daily mean and monthly mean ERA-Interim data from 1979 to present were directly downloaded from the ERA-Interim website in this study. Furthermore, the daily mean and monthly reanalysis datasets supported by the National Oceanic and Atmospheric Administration were also employed and denoted as NCEP/NCAR
95 (National Center for Environmental Prediction and the National Center for Atmospheric Research) data. The 2.5°×2.5° geopotential height (Z), zonal and meridional wind, relative humidity, vertical velocity, air temperature at from 1000 hPa to 100hPadifferent pressure levels, SAT and wind, downward UV radiation, downward solar radiation, low and medium cloud cover were downloaded, which was available from 1948 to present (Kalnay et al. 1996). The BLH of NCEP/NCAR datasetdataset was only available from 1979 to 2014 in the website of the NOAA-CIRES 20th Century Reanalysis version 2c (Giese et al., 2016). The daily precipitation data was from the CPC global analysis of the daily precipitation dataset (Chen et al., 2008).

The EU pattern is a major teleconnection pattern in the Northern Hemisphere and appears in all seasons. Wang and Zhang (2015) used the method defined by Wallace and Gutzler (1981) to calculate the EU pattern index in winter and pointed out that the positive EU phase is associated with a cold-dry climate in East China, <u>and vice versa</u>. Meanwhile, Wang and He (2015) regarded the summer EU pattern as the main reason for the severe summer drought in North China in 2014. Considering the seasonal change of the EU pattern's location, the calculation procedure for the <u>summertime</u> EU index here was consistent with that in Wang and He (2015).

EU index = $[-1 \times \overline{\text{H500}}_{(70-80^{\circ}N,60-90^{\circ}E)} + 2 \times \overline{\text{H500}}_{(45-55^{\circ}N,90-110^{\circ}E)}]$

$$-1 \times \overline{\text{H500}}_{(35-45^{\circ}N, 120-140^{\circ}E)}]/4 \tag{1}$$

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110 where H500 represents the geopotential height at 500 hPa, and overbars denote the area average.

The generalized additive model, a data-driven method, is particularly effective at handling the complex nonlinear and non-monotonous relationships between the dependent variable and the independent variables (Hastieand Tibshirani, 1990). This approach used a smoothing function, determined by the independent variables themselves, to transform the expressions, and addressed the dependent variable with different probability distributions by the link function. To verify the connection
 between the Arctic sea ice and the O₃ pollution, the Community Atmosphere Model version 5.3 (CAM5, Meehl and Washington 2013) was employed to design numerical experiments. The spatial resolution employed was 0.9°×1.25°, with 30 vertical hybrid sigma-pressure levels. CAM5.3 uses vertical hybrid δ-pressure coordinates including 26 layers with the top located at about 3.5 hPa. The climatological mean sea surface temperature and sea ice taken from the Hadley Centre# were

used to force the control run.

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120 **3** Summer ozone pollution and associated weather conditions

Due to increased surface O_3 pollution in China, the number of O_3 measurement stations has dramatically increased since 2014 (Figure 1 a, c, e, g). During 2006–2014, O_3 concentrations were only observed in the most developed regions in China. Since 2015, O_3 concentrations have been measured in most areas in eastern China. O_3 concentrations in the high-mid latitudes were higher than those in the lower latitudes, which appeared to be separated by the Yangtze River. The O_3 concentrations in North China were already high in 2014; the summer mean MDA8 in North China was higher than 120 µg/m³. The oObservations⁷ with maximum MDA8 higher than 265 µg/m³ (i.e., the threshold of the serverg surface O_3 pollution in China) existed in the south of Hebei Province and the north of Shandong Province (Figure 1a). Since that time, the O_3 polluted region has expanded. In 2017, the areas with summer mean MDA8 > 120 µg/m³ were visibly enlarged. In North China, the summer mean MDA8 observations were larger than 150 µg/m³, and the maximum MDA8 was nearly 265 µg/m³. South of the Yangtze River, the O_3 concentrations were distinctly lower and decreased progressively towards the Pearl River Delta.

The time span of O₃ observations (i.e., 2015–2017 for most of the sites) limited the possibility of determining the role of climate variability in the interannual O₃ variations in North China. Thus, we examined the representativeness of the O₃ measurements at SDZ (one of the three regional background air-monitoring stations in China, with observations from 2006–2017). The correlation coefficients between SDZ MDA8 and the observed MDA8 at the other sites were calculated and are

135 shown in Figure 1 (b, d, f, h). The distribution of correlation coefficients is similar to the MDA8 on Figure 1 (a, c, e, g). The SDZ MDA8 significantly covaried with the MDA8 in North China in summer. Along with the increasing of the surface O₃ pollution, the covariation and the representativeness of SDZ MDA8 to the MDA8 in North China was strengthened. the covariation of SDZ MDA8 and MDA8 in North China strengthens the representativeness of SDZ for North China. However, the correlation coefficients between SDZ MDA8 and MDA8 in the south of China were negative, indicating opposite variation 140 (Zhao and Wang, 2017). The variation of summer SDZ MDA8 is presented in Figure S1. The daily difference in MDA8 was large, which contradicts the quasi constant emission of ozone precursors. Therefore, we speculated the impacts of meteorological conditions were significant. According to the Technical Regulation on Ambient Air Quality Index in China (The Ministry of Environmental Protection of China, 2012), we defined non-O₃ polluted (NOP) level at surface as the O₃ concentration $< 100 \ \mu g/m^3$ and moderate-O₃ polluted (MOP) level with O₃ concentration $> 215 \ \mu g/m^3$, respectively the 145 thresholds of non-surface O₃ polluted (NOP) level and moderate surface O₃ polluted (MOP) level are 100 µg/m³ and 215 µg/m³, respectively. The upper and lower quartile of SDZ MDA8 was 188 µg/m³ and 114 µg/m³ indicating that more than 75% of summer days exceeded the NOP thresholdwere not the NOP days even at the regional background air-monitoring station. During the years 2007–2017, there were 126 NOP days and 155 MOP days in summer at SDZ station. The maximum number

of MOP days was 26 days in 2015, and the mean number of MOP days was 14 days (Table S1). Both of **T**the interannual

- 50 variation in MOP <u>and (NOP)</u> days was significant at the 95% confidence level, without an obvious long term-trend.
 - Due to the significant covariation between the SDZ MDA8 to the MDA8 in North China, although the meteorological conditions were composited for the MOP and NOP days in SDZ (Figure 2), and the results were also appropriate for those in North China. The local and surrounding weather conditions were significantly different (t-test). The regional average meteorological elements were calculated as meteorological indexes, and here the selected regions determined on the most
- 155 significantly different areas in the composites of MOP and NOP events in Figure 2. During calculating the correlation coefficients with the meteorological conditions, the averaging area for meteorological indexes were the regions with most significantly different elements in the composites of MOP and NOP events. The anomalous southerlies (Figure 2a), higher BLH (Figure 2c), less rainfall (Figure 2e), warmer surface air temperature, and cooler temperature in the high troposphere (Figure 2g) favored surface O₃ pollution and *vice versa*. Anomalous southerlies from the Yangtze River transported O₃ precursors (that were emitted in the economically developed Yangtze River Delta) and superposed them with the local high emissions in North China (Figure 2a). When the anomalous winds reversed, i.e., northerlies, the O₃ precursors in North China was reduced (Figure 2b). The correlation coefficient
- between the SDZ O₃ concentration and the area-averaged meridional wind at 10 m (35–50 N, 110–122.5°E, <u>black box in</u>
 Figure 2a, denoted as V10mI) was 0.39, exceeding the 99% confidence level. The cloudy skies and precipitation weakened the
 photochemistryphotochemical reaction by influencing exposure to ultraviolet rays. In addition, precipitation was also an important indicator of the wet removal efficiency (Figure 2f). In summer, a day without rain represents efficient solar radiation, in favor of the occurrence of surface O₃ pollution (Figure 2e). The correlation coefficient between the area-averaged precipitation (37.5–42.5 N, 112–127.5 E, <u>black box in Figure 2e</u>, denoted as PI) and the SDZ O₃ concentration was –0.35 (above the 99% confidence level), indicating that precipitation was connected with more NOP days.
- In contrast, high SAT enhanced the photochemical reactions and resulted in higher surface O₃ concentrations (Figure 2 g). The correlation coefficient between the area-averaged difference in the temperature at the surface and 200 hPa (SAT minus temperature at 200 hPa, 37.5–47.5 N, 110–122.5 E, black box in Figure 2g, denoted as DTI) and the SDZ O₃ concentration was 0.49. Furthermore, due to the strengthening of solar radiation, the near-surface turbulence was enhanced, and the boundary layer was lifted (Figure 2c). The entrainment of atmospheric ozone from the upper air into the boundary layer enlarged the surface O₃ concentration (An et al., 2009). The correlation coefficient between the SDZ O₃ concentration and the area-averaged BLH (37.5–47.5 N, 112.5–120 E, black box in Figure 2c, denoted as BI) was 0.40. Therefore, the anomalous southerlies, high surface temperature, above average BLH, and sunny skies were favorable environments for severe surface O₃ pollution. To confirm the robustness of the link between meteorological conditions and the MOP and NOP days over North China, the above composite analysis was repeated with NCEP/NCAR reanalysis data, and identical results were obtained

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To assess the interannual variation of surface O_3 pollution and its relationship with climate variability (Cai et al., 2017), we tried to fit an O_3 weather index (OWI) based on long-term meteorological observations. Here, we defined the OWI as OWI=normalized V10mI+normalized BI–normalized PI+normalized DTI. For comparison, the multiple regression equation was built between the MDA8 and associated weather indices (Figure 3). Our analysis indicated that the observed MDA8 was well fit by the multiple regression equation (Figure 3). The correlation coefficient was 0.61 between the fit and daily measured MDA8 during 2007–2017 (i.e., 92 days ×11 years). The correlation coefficient between the observed MDA8 and daily OWI was also 0.61 for the 11 year period. Thus, the OWI was easily constructed by accumulating the normalized weather index and was selected to represent the variation in surface O_3 pollution. A total of 90.3% of the MOP events were in the range of OWI > 0, and correspondingly, 90.5% of the NOP events were linked with OWI < 0 (Figure 4). The correlation coefficients between the OWI and observed MDA8 at the other sites were calculated (Figure 5). The significantly positive correlations were distributed in North China (Figure 5 b-d). Thus, it is reasonable to analyse the variation in surface O_3 -pollution i

4 Impacts of EU pattern on the interannual variation of surface ozone

195 After the assimilation of satellite data, possible in 1979, the quality of the reanalysis data was improved. Here, the daily OWI was calculated with both ERA-Interim and NCEP/NCAR reanalysis data from 1979. According to the above analysis, the daily OWI could largely represent the variation in MDA8 in North China. The monthly OWI was computed as the monthly mean of the daily OWI. During 2007-2017, the constructed JJA (June-July-August) mean OWI varied similarly with the observed MDA8 and captured the extremes (Figure 6). Although the range of the SDZ MDA8 was 2006–2017, only the data from 2007 200 to 2017 were used in the above OWI construction processes. Thus, the datasets in 2006 were independent samples (i.e., test set), and could verify the performance of the OWI. The JJA mean OWI in 2006 successfully reflected the variation in observed MDA8; even the MDA8 in 2006 was the minimum, confirming the robustness of the OWI. Derived from two different reanalysis datasets, the OWI-ERA and OWI-NCEP varied consistently. The above independent verifications proved that the performance of the summer OWI did not depend on the specifickinds of reanalysis data. In the following study, the monthly 205 OWI from ERA-interim data and associated physical mechanisms were analysed. Before-During mid-1980s to the mid-1990s, the OWI was below zero, with a slightly decreasing trend and insignificant interannual variation. Since then, the OWI has increased; furthermore, the intensity of interannual variation has strengthened. The emissions of O₃ precursors increased persistently and linearly due to the steady economic development after 1978 in China (Wang 2017). The strong interannual variation in the OWI after mid-1990s, representing the impacts of meteorological conditions on O₃ concentrations, contributed

- 210 to the interannual fluctuations of the surface O₃ pollution. Thus, the impacts of the large-scale atmospheric circulations on the summer O₃ pollution, specially the related OWI, were analyzed.
- The atmospheric circulations associated with summer mean OWI, indicated by the correlation coefficients, are displayed in Figure 7. In the mid-upper troposphere, cyclonic and anticyclonic anomalies were alternately distributed over the north-central Siberian Plateau (-), North China and Mongolia (+), and the Yellow Sea and Japan Sea (-) (Figure 7a). These three 215 atmospheric centers, propagated from the polar region to the mid-latitudes, appeared to be the positive phase of EU pattern (Wang and He 2015). This Rossby wave-like train, i.e., the EU pattern, could also be recognized in the surface air temperature. The correlation coefficient between the EU pattern index and OWI was 0.44 (after detrending and above the 99% confidence level), indicating that the strengthening of the EU positive phase contributed to the severe surface O_3 pollution in North China. More precisely, the positive phase of EU pattern could modulate the local meteorological conditions to enhance 220 the photochemical reactions. The EU pattern is considered to be the main reason for the variability of the severe drought in North China, i.e., resulting in hot and dry climate extremes (Wang and He, 2015). To a certain extent, the severe drought environment promoted the formation of surface ozone. After 2007, the EU index and the observational SDZ MDA8 showed good agreementsynchronously changed (Figure 8). More than 80% of the SDZ MDA8 anomalies showed the same mathematical sign as the anomalous EU pattern index. Furthermore, the intensified-large EU pattern anomalies (i.e., the |EU 225 pattern index $| > 0.8 \times its$ standard deviation) always induced homodromous-in-phase surface ozone pollution.
- Under barotropic anticyclonic circulation over North China, i.e., one of the active centerscentres of the positive EU pattern, the significant descending air flows indicated efficient adiabatic heating (resulting in high temperatures near the surface) and dry air (i.e., less cloud cover) below 300 hPa (Figure 7c). Furthermore, over North China, the air temperature (relative humidity) anomalies were negative (positive) at 200 hPa but positive (negative) below 300 hPa (Figure 7c). The barotropic anticyclonic 230 circulation associated with surface ozone pollution (Figure 7b) was similar to the positive EU pattern (Figure 7c) and led to sunny days, i.e., hot temperatures (Figure 7a), strong downwards solar radiation and UV radiation (Figure 9c-d), less low and medium cloud cover (Figure 9d), and dry conditions (Figure 9b-c). Without the cover of low and medium clouds, the short wave solar radiation, especially the UV radiation, penetrated straight to the land surface. The photochemical reaction of the O_3 precursor was enhanced, generating more O₃ near the surface. The dry atmosphere near the surface, i.e., less precipitation and 235 lower relative humidity, accelerated the photochemical reaction but restricted the wet clearing of the stocked O_3 in the atmosphere. A higher BLH (Figure 9b), resulting from the strengthening of solar radiation, likely facilitated the downward transportation of O_3 from aloft. Near the surface, the western part of these anticyclonic anomalies manifested as significant southerlies (Figure 9a), which transported the O_3 precursors from the economically developed Yangtze River Delta. The extraneous O_3 precursor, superposed with local emissions, supported a harsh and efficient photochemical reaction-production of O₃. To confirm the robustness of the atmospheric circulations and associated physical mechanisms, the above analysis was

repeated <u>by-with</u> the NCEP/NCAR data and identical results were obtained (Figure S4–S5). The <u>correspondence between</u> large-scale EU teleconnection₇_-and anti-cyclonic circulations were clear. Local meteorological conditions, such as hot land surface (Figure S4), violet solar radiation (Figure S5c–d), clear sky (Figure S5d), less precipitation (Figure S5c) and lower relative humidity (Figure S5b) were also clearly recognized. Thus, the impacts of the atmospheric circulations were confirmed by both the ERA-Interim and NCEP/NACAR data, i.e., the analyses and conclusions were independent of data sets.

5 Roles of the Arctic sea ice

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The positive EU pattern enhanced the local anticyclonic circulation over North China and facilitated the photochemical processes leading to the formation of surface ozone. The EU pattern originated from the Arctic region. The preceding sea ice anomalies could stimulate atmospheric responses like <u>the</u> EU pattern in summer (Wang and He, 2015) Thus, the role of Arctic sea ice on the OWI was also explored in this study. The correlation between the sea ice and JJA OWI was <u>evaluated each</u> <u>monthmonthly checked</u> (Figure omitted), and we found the interannual variation of OWI was significantly correlated with May sea ice conditions to the north of Eurasia, especially near the Gakkel Ridge, the Canada Basin and the Beaufort Sea (Figure 10a). The averaged (green boxes in Figure 10a) sea ice <u>(SI)</u> area in May was calculated as the SI index, whose linear correlation coefficient with JJA OWI was 0.67 (after detrending) from 1979 to 2017. During 2007–2017, 73% of the May SI anomalies may beare followed by observational SDZ MDA8 anomalies with the same mathematical sign (Figure 10b). Furthermore, the linear and nonlinear relationships were both introduced using the generalized additive model (Figure 11), and the contribution of May sea ice to the interannual variability of OWI was approximately 60%.

These positive sea ice anomalies could induce EU-like pattern responses in the subsequent summer (Figure 10c). The excited atmospheric and thermal centerscentres were located over the Central Siberian Plateau, North China and Mongolia, and the Yellow Sea. Similarly, the local meteorological responses, such as anomalous southerlies and less precipitation (Figure 10d), less cloud and strong solar radiation (Figure 10e) were also closely connected with the positive sea ice anomalies in May. Thus, the preceding May sea ice positively modulated the EU pattern, and then, this Rossby wave train transported the impacts from the polar region and strengthened the anti-cyclonic anomalies over North China. Finally, suitable meteorological conditions, including hot-dry air, anomalous southerlies and intense sunshine, were induced to intensify the photochemical production of surface ozone pollution. To confirm the roles of Arctic sea ice and associated physical mechanisms, the above analysis was repeated with the NCEP/NCAR data, and identical results were obtained (Figure S6).

The causality, i.e., the preceding May sea ice anomalies contribut<u>inged</u> to the subsequent JJA OWI in North China, was also confirmed by CAM5. During the control experiment (CTRL), the CAM5 model <u>was</u> firstly integrated <u>for</u> 20 years with climate mean initial and boundary conditions. Next, the data in 1st September of the last 5 years (i.e., 16–20 years) were designated as five slightly different initial conditions. With each initial condition, the CAM5 model integrated for 10 years.

integrated 10 years with each 1st September of the last 5 years (i.e., five slightly different initial conditions). The JJA mean results of the last 6 years (i.e., 6 years ×5 groups = 30 ensembles) were employed as the output of the CTRL. On the basis of CTRL, the May sea ice concentration in the two boxes of Figure 10a was separately reduced by 10% (denoted as LowASI experiments), i.e., a total of 30 sensitivity runs totally 30 sensitive runs. Similarly, the JJA mean results of the 30 sensitive ensembles runs were employed as the output of the LowASI. The differences (LowASI minus CTRL) were represent the responses of atmospheric circulations and meteorological conditions to the declining May sea ice.

It was evident that an EU-like Rossby wave train was induced on the mid-troposphere (Figure 12a), which propagated from the Taymyr Peninsula (–), Northeast China (+), to east of China and the west Pacific (+). Under such large-scale atmospheric anomalies, the anomalies of relative humidity were significantly positive and resulted in denser low and cloud cover in North China (Figure 12d). Furthermore, the cover of cloud efficiently prevented the solar radiation from reaching the land surface, meanwhile, cooled the air in the boundary layer (Figure $12b_{12c}$). Without hot-dry air and violent-intense sunshine, the photochemical reaction-production was significantly decelerated and the generation of surface O₃ was rather weak. Additionally,On the other side, sufficient moisture and clouds caused more rainfall (Figure 12c). The wet deposition effect might be was also significantly enhanced. Thus, corresponding to less Arctic sea ice in May, the photochemical process to generate O₃ was weakened, but-and the wet deposition effect to decrease O₃ was enhanced. That is, the positive relationship and associated physical mechanisms (i.e., climate links among ASI, EU pattern and summer surface ozone pollution in North

6 Conclusions and discussions

China) were causally verified.

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Recently, the summer surface O₃ concentrations and the number of O₃ observation stations have steadily increased in China. In generalSpatially, the O₃ concentrations in North China were substantially higher than those in South China. To reveal the climatic driver and improve the potential of seasonal prediction of summer surface O₃ pollution in North China, a daily OWI (i.e., surface O₃ weather index) was constructed based on long term-meteorological and ozone observations. The robustness of this index (i.e., OWI) was verified by the ERA-Interim and NCEP/NCARNOAA reanalysis datasets and surface O₃ measurements. May Arctic sea ice was found to be a preceding and efficient climatic driver, which may help for seasonal forecasting. In the historical period, variation in Arctic sea ice can explain approximately 60% of the interannual variability of the summer OWI in North China, which was closely associated with the surface O₃ pollution. Currently, the Arctic region has been warming approximately twice as much as the global average (Huang et al., 2017; Zhou, 2017), indicating accelerated change in the sea ice. Thus, understanding the role of Arctic sea ice may contribute to the <u>understanding of seasonal variabilityseasonal look</u> of O₃ pollution.

³⁰⁰ The EU pattern acted as an atmospheric bridge to link May Arctic sea ice and the summer surface O₃ pollution in North China.

The accumulated sea ice in May could induce or enhance-the positive EU phase. The anticyclonic circulation over North China, i.e., one of the active <u>centers_centres</u> of the EU pattern, was connected with high surface temperature, strong downward solar radiation, less low__ and medium<u>-altitude</u> cloud cover, and drought over North China. Under such local meteorological conditions, the photochemical <u>reaction to produce surface O₃ wasreactions to produce surface O₃ were supported accelerated. Generally, these anticyclonic anomalies over North China were barotropic and could persist for a long time; thus, the processes</u>

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that produce surface O_3 were continuous to achieve a fairly high concentration. The connections revealed in this study were based on long-term meteorological measurements and was <u>causally casually</u> verified by well-designed numerical experiments.

In order to extend the time range of this study, the OWI was constructed in North China. Although the feasibility of the construction approach was strictly examined, the OWI was still a substitution focusing on the impacts of the weather 310 conditions. When discussing the impacts of atmospheric circulations, the linear trend was removed to weaken the signal of anthropogenic emissions. Thus, the results in this study concentrated on and emphasized the meteorological and climate factors. However, there is no doubt that the polluted emissions are the fundamental inducement of the surface O_3 pollution. The joint effects of the climate anomalies and the historical emissions should be lucubrated studied using the numerical models in the future. The processes how the weather conditions impacted the photochemical reaction were not deeply 315 discussed here and have been analyzed in many previous studies by the atmospheric chemists. However, the reason why the eooler high troposphere contributed to the surface ozone pollution was still an open question and needed further attention. The EU pattern was a well-known continental Rossby wave train and could link the mid-high latitude climate with the change of the Arctic. Although the connection between the Arctic sea ice and the ozone pollution was revealed, the separate roles of the sea ice near the Gakkel Ridge, and the Canada Basin and Beaufort Sea should be intensively studied in the 320 future.

Author contribution

Yin Z. C. and Wang H. J. designed the research. Yin Z. C., Li Y. Y. and Ma X. H. performed research. Yin Z. C. and Zhang X. Y. analysed data. Yin Z. C. prepared the manuscript with contributions from all co-authors.

325 The authors declare no conflict of interest.

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Table and Figures captions

Figure 1. The distribution of the JJA mean MDA8 (a, c, e, g) and the correlation coefficients (b, d, f, h) between the daily MDA8 and SDZ MDA8 from 2014 to 2017. The black cross in panels a, c, e, and g indicate that the maximum daily MDA8 was larger than 265 μ g/m³. The black cross in panels b, d, f, and h indicates that the CC was above the 95% confidence level. The green triangle in panel (a) illustrates the location of the Shangdianzi site.

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Figure 2. Composite of the meteorological conditions associated with different O₃-events during 2007 2017. Results for MOP (a, c, e, g) and NOP (b, d, f, h) events included (a b) surface wind (arrow) and v-wind (shading), (c-d) BLH, (e-f) precipitation, (g-h) SAT, and temperature at 200 hPa. The black dots denote the composite results passed the 95% confidence level. The boxes represent the area used to calculate OWI. These composites were calculated using the

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events (i.e., all events but excluded MOP and NOP events).

Figure 3. The variation in the daily (a) observational SDZ MDA8 (black), (b) fitting SDZ MDA8 (red), and (c) OWI (blue) from June to August during 2007–2017. The numbers are the correlation coefficients between the observational SDZ MDA8 and fitting SDZ MDA8 (red) and OWI (blue).

455 Figure 4. The OWI for MOP (red) and NOP (blue) events during 2007–2017.

Figure 5. The correlation coefficients between the daily MDA8 and OWI from 2014 to 2017. The black crosses indicate that the CC was above the 95% confidence level.

Figure 6. The variation in the JJA mean observed SDZ MDA8 (green) from 2006 to 2017, OWI calculated from ERA-interim datasets during 1979–2017 (blue) and OWI calculated from NOAA datasets (red) during 1979–2014.

460 Figure 7. The associated atmospheric circulation. (a) The correlation coefficients between the JJA mean OWI and surface air temperature (shading), wind (arrow) at 200 hPa and geopotential height at 500 hPa (contour) from 1979 to 2017. The black dots indicate that the CC with surface air temperature was above the 95% confidence level. The cross-section (110°–125°E mean) correlation coefficients between JJA mean OWI (a), EU pattern index (b) and relative humidity (shading), temperature (contour), wind (arrow, vertical speed multiplied by 100) from 1979 to 2017. The black dots indicate that the CC with relative humidity exceeded the 95% confidence level (t test). The data used here are ERA-Interim datasets.

Figure 8. The variation in the JJA mean observational SDZ MDA8 (blue) and EU pattern index (red) from 2007 to 2017. Figure 8. The variation in the JJA mean observational SDZ MDA8 (µg/m³, blue) and EU index (gpm, red) from 2007 to 2017.

- 470 Figure 9. The associated meteorological conditions. (a) The correlation coefficients between the JJA mean OWI and v wind at 10 m (shading), surface wind (arrow), (b) relative humidity near the surface (shading), boundary layer height (contour), (c) precipitation (shading), downward UV radiation at the surface (contour), (d) downward solar radiation at the surface (shading), sum of low and medium cloud cover (contour) from 1979 to 2017. The black dots indicate that the CC with temperature was above the 95% confidence level. The contours plotted in panel (b–d) exceeded the 95% confidence level.
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Figure 10. The role of the Arctic sea ice. (a) The correlation coefficients between the JJA mean OWI and May sea ice, (b) The variation of the May SI index (red bar, area-averaged sea ice of the green boxes in panel a), JJA mean EU pattern index (blue bar) and JJA mean observational SDZ MDA8 (black bar) from 2007 to 2017. (c) The correlation coefficients between the May SI index and surface air temperature (shading), geopotential height at 500 hPa (contour) from 1979 to 2017. The

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Figure 12. Composite results of the LowASI experiments (LowASI minus Ctrl) by the CAM5 model: (a) geopotential height at 500 hPa, (b) precipitation, (c) net radiative flux at the top of the atmosphere (shading) and temperature at 925 hPa (contour), and (d) sum of low and medium cloud fraction (shading) and relative humidity at 925 hPa (contour). The black hatching denotes the differences with shading were above the 95% confidence level (t-test).

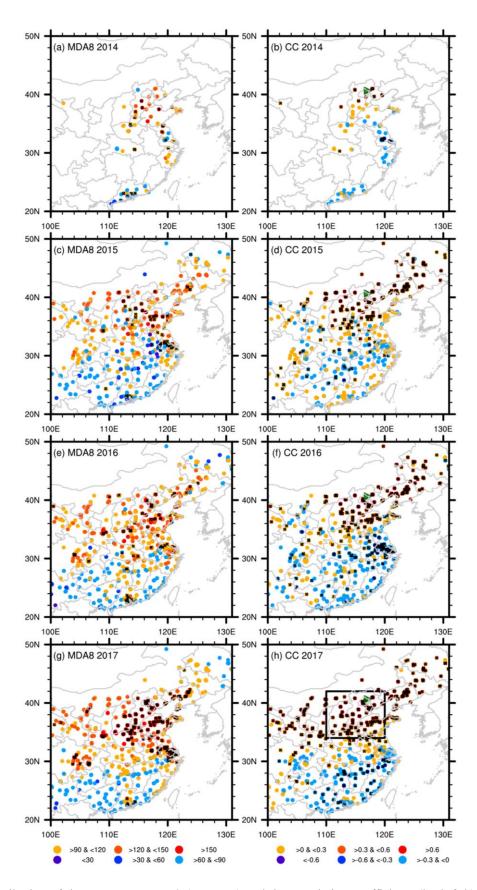


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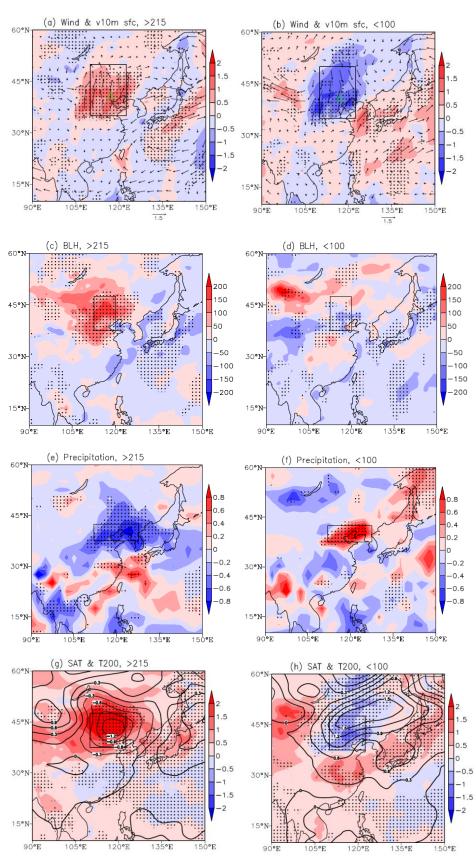


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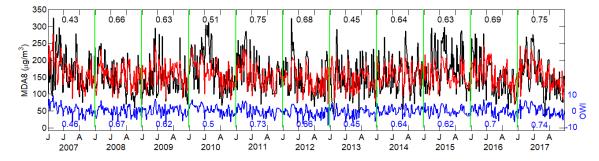
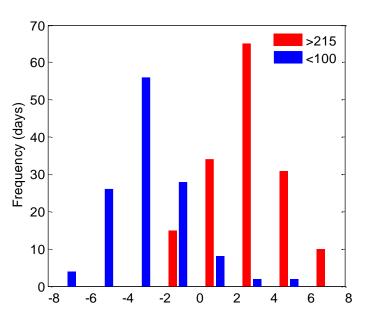


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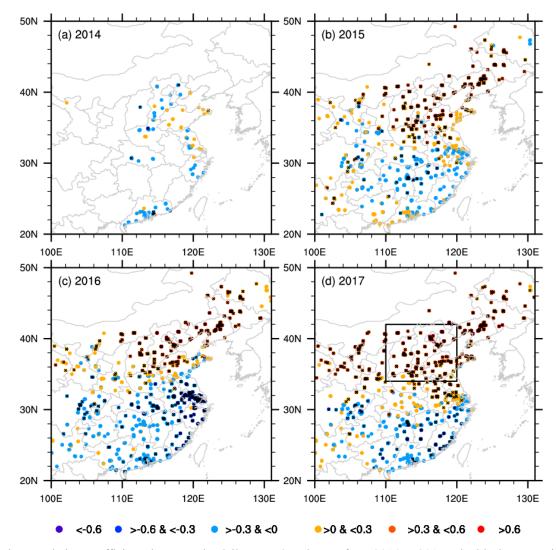


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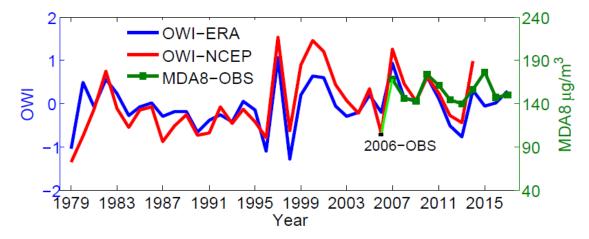
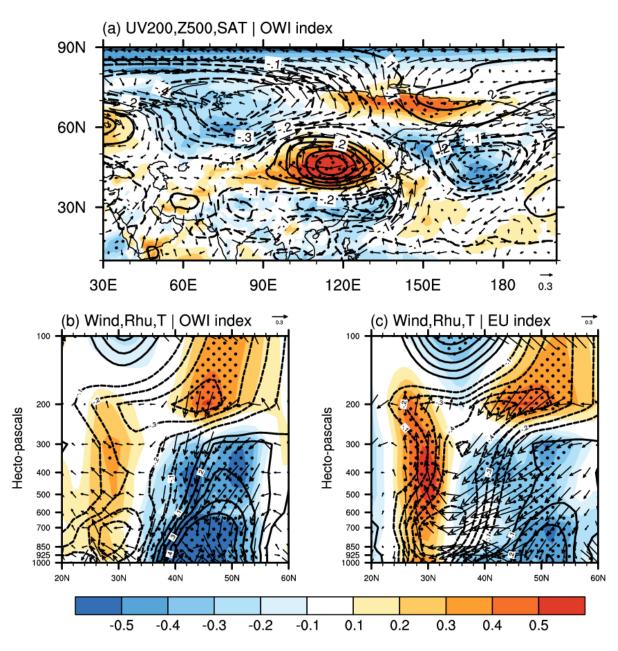


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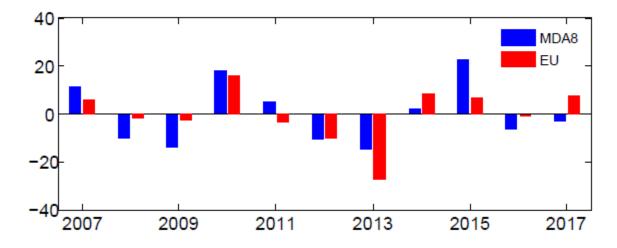


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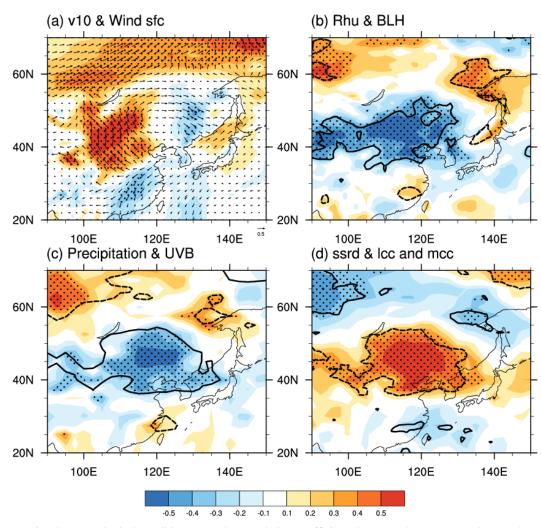
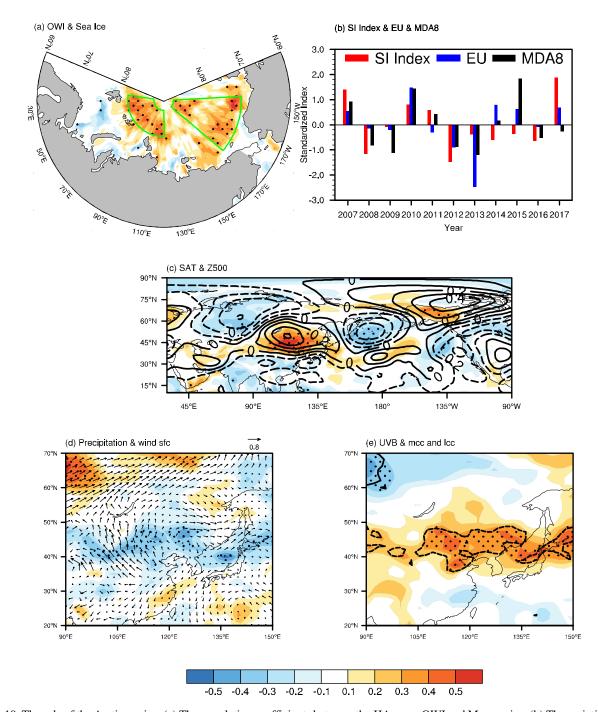


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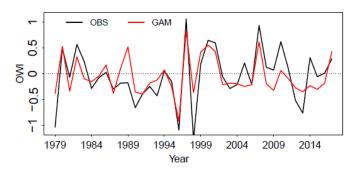


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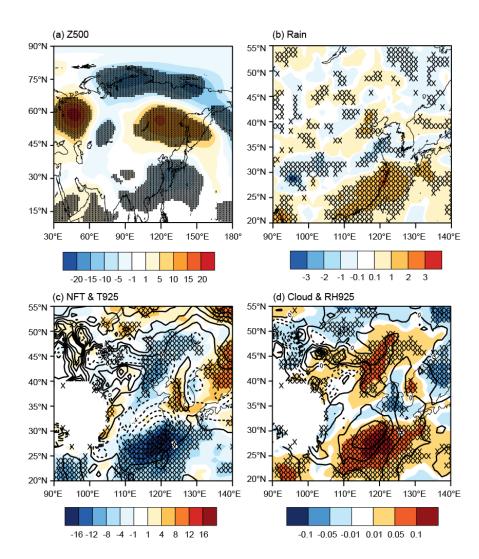


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