We thank the reviewers for their valuable comments and suggestions to improve our manuscript. We have made revisions accordingly. The point-to-point responses are provided below in Italic. The comparison of our manuscript between this version and the previous version is also provided.

5

Anonymous Referee #1:

This study explores the local and synoptic meteorological influences on summertime ozone variability in eastern China. The authors have used two different approaches, including MLR and SOM, and their MLR method includes the effects of synoptic

10 patterns. In general, I think this manuscript is well structured and the topic is suitable for ACP. The presentation quality is also very high. So I recommend publication in ACP after fixing only a few minor issues.

The reviewer's encouragement is much appreciated.

15 Minor comments.

L123. Please specify the value of dmax used in this study? 'd<sub>max</sub>' is specified as 1-degree distance in latitude-longitude grid. Please see Lines 132-133.

20 L283. It is worthwhile to mention the trends of anthropogenic emissions during this period. During the period of 2013-2017, the NOx emissions have declined (Zheng et al., 2018) and the VOC emissions almost remain constant (Zheng et al., 2018; Shen et al., 2019).

Thanks. Added. See Lines 293-295.

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Line 299. The correlation of temperature with ozone is higher in the north than in the south. I guess this is because the temperature gradient along the latitude is much larger in the north than in the south. So the synoptic activities (or jet wind) in the

north can be well represented by temperature. But in the south, the marine flux into

- 30 the continent won't result much changes in temperature but will definitely elevate the relative humidity. As a result, relative humidity displays much stronger correlation with ozone in the south. However, I never found any paper that discusses about the reason of this distinct north-south feature. Maybe the author can simply say like this.
- The correlation of temperature with ozone is higher in the north than in the south over eastern China (Figure 2c), which is consistent with the pattern found in the US (Camalier et al., 2007; Shen et al., 2016).
   *Thanks. Revised following the reviewer's suggestions. Please see Lines 317-319.*
- Line 305. Relative humidity can also be a strong indicator of transport in the southern China. So I won't use the words "mainly" here.
   *Thanks. Revised. Please see Lines 310-316.*

Figure 5. I would strongly recommend using the leave-one-out cross validation to

45 avoid overfitting if the authors haven't done this.
 Thanks. We have used the leave-one-out cross validation in this revision. Please see
 Lines 204-208.

Figure 6. I don't know if there is a way to separate the effects of local meteorology

50 and synoptic patterns. They are so closely related. I would suggest deleting this figure and also the text related.

Thanks for the suggestion. We have deleted the comparison of the weight between local meteorology and synoptic patterns and reserved the comparison between different local meteorological variables in this revision.

55

Anonymous Referee #2

The authors conducted an extensive analysis of local and synoptic meteorological

- 60 influences on daily variability in summertime surface ozone in eastern China for the time period of 2013-2018. They derived a multiple linear regression (MLR) equation for each grid within the eastern China domain to capture the linear relationships of daily average ozone concentrations as a function of 10 local meteorological variables and 2 synoptic factors, the latter derived using the singular value decomposition
- 65 (SVD) method. Not to be pedantic, it is an overstatement to call the MLR equation a model. They further examined synoptic weather patterns (SWPs) over eastern China using a self-organizing map (SOM) clustering technique. The MLR and SWPs provides a rich source of information but the authors were short of making a connection between the two. One interesting point from MLR was, as local
- 70 meteorological variables, relative humidity in the central and southern parts of eastern China and temperature in the BTH region showing the largest influence on surface ozone concentrations. The study would have been more in-depth should the authors have endeavored to understand the mechanism(s) driving that. Would it be possible to use their SWPs to further understand that point? The authors did use their derived
- 75 MLR to validate the calculated surface ozone concentrations under the 6 SWPs, but they only showed visual comparisons between the predicted and observed values. It'd make a stronger case if they could show some quantitative comparison. Most of Section 6 "Discussion and conclusions" repeated the results prior to it with the last paragraph suggesting the potential significance of the study. There was not really
- 80 much discussion but repetition. I suggest that the section be shortened and changed to "Summary". Figure 14 is missing from the manuscript. In the huge body of published work on surface ozone as a pollutant, the majority has used ppbv as units for ozone, and indeed in study of atmospheric trace gases mixing ratios have been used conventionally. The authors' use of mass units was a bit peculiar. I suggest that they
- 85 provide unit conversion upon the first appearance of the mass units if they insist upon

using them.

Thanks for the valuable comments. The following is our answers to the reviewer's questions.

- 90 By fitting a linear equation, the MLR predict the response variable by using several explanatory variables. As a simple and basic regression model, it has been widely used in the prediction of atmospheric pollutants (Kutner et al., 2004; Gao et al., 2019; Li et al., 2019). In this study, the MLR is applied to predict surface ozone in eastern China with the predictors of meteorological factors. In this revision, we
- 95 further used the leave-one-out cross validation (Section 2.2, Lines 204-208) to avoid overfitting of the MLR. The MLR shows strong performance with a regional mean coefficient of determination ( $R^2$ ) of 43% (Figure 5a).

In this revision, we combined the MLR and SOM to reveal the most important local

- 100 meteorological factor for ozone variability under each of the six SWPs (Figure S6). The MLR was conducted under each of the SWPs with the same procedures for the full summer. The most important meteorological variable for ozone over some areas in eastern China may vary with the prevailing SWP (Figure S6). The dominant driver in PRD is meridional wind at 850 hPa under PSW (P1), PS (P2), and PSWPSH (P5),
- 105 demonstrating the significant influences of marine air inflow. Controlled by the typhoon system, the most important factor over some coastal areas is zonal wind at 850 hPa under PTC (P6). The analysis has been added in Lines 486-492.

*Results from the MLR show that among local meteorological factors, relative* 

110 humidity is the foremost influential variable for summertime surface ozone over most locations in the center and south of eastern China including YRD and PRD, while temperature is more important in the north including BTH. Such a difference between the north and south were also found in the eastern United States by previous studies (Camalier et al., 2007; Porter et al., 2015). The difference is possibly related with

115 both ozone photochemistry and synoptic influences. The relative importance of relative humidity and temperature to ozone photochemistry may vary with location, because of different atmospheric environments. Moreover, the sensitivities of relative humidity and temperature to synoptic systems may change with the location as well. However, until now, there are no strong evidence to explain it.

120

We have added some statistical comparisons between the observations and predictions of averaged ozone anomalies under each of the SWPs in Table S1. The mean absolute error (MAE) ranges 1.0-2.2  $\mu$ g m<sup>-3</sup> and the root mean square error (RMSE) ranges 1.4-2.8  $\mu$ g m<sup>-3</sup>.

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We have renamed Section 6 as 'Summary' and shortened this section.

In the last version, Figure 14 showed the relative anomalies of observed surface ozone under the six SWPs, giving an additional explanation of Figures 8d, 9d, 10d,

130 11d, 12d, and 13d. In this revision, Figure 14 is moved into the supplement as FigureS5. We reserve the regional mean relative anomalies in Table 1.

We used the unit ' $\mu$ g m<sup>-3</sup>' to keep consistency with that for China national air quality standard. For ozone, 1  $\mu$ g m<sup>-3</sup> equals to 0.47 ppbv at 273 K and 1013.25 hPa.

Gao, M., Sherman, P., Song, S., Yu, Y., Wu, Z., and McElroy, M. B.: Seasonal
prediction of Indian wintertime aerosol pollution using the ocean memory effect, Science Advances, 5, eaav4157, 10.1126/sciadv.aav4157, 2019.

<sup>135</sup> *The unit conversion is added in Lines 116-117.* 

Kutner, M.H., Nachtsheim, C.J., Neter, J., Li, W., 2004. Applied Linear Statistical Models. McGraw-Hill/Irwin, New York, NY, USA.

Li, K., Jacob, D. J., Liao, H., Shen, L., Zhang, Q., and Bates, K. H.: Anthropogenic drivers of 2013-2017 trends in summer surface ozone in China, Proc. Natl. Acad. Sci. U. S. A., 116, 422, https://doi.org/10.1073/pnas.1812168116, 2019.

145

Some specific comments:

1. Line 36: The first sentence covered both human and vegetation health but the reference cited, Yue et al. (2017), was on vegetation.

Thanks. A reference for human health is added (Jerrett et al., 2009).

150

- Jerrett, M., Burnett, R. T., Pope, C. A., Ito, K., Thurston, G., Krewski, D., Shi, Y., Calle, E., and Thun, M.: Long-term ozone exposure and mortality, N. Engl. J. Med., 360, 1085-1095, https://doi.org/10.1056/NEJMoa0803894, 2009.
- 155 2. Lines 78-80: Shen et al. (2017a) is not the first and only reference for such a wellestablished point. There is a huge wealth of research on this point dating back to decades ago. This seems to be a fairly common problem nowadays, that for an extensively, long studied topic, only most recent few studies would be cited whereas a long list of monumental studies leading up to the recent works tend to be left out. In
- my opinion, we need to do due diligence to cite the references where credit is due. *Thanks for the points. Several reliable studies are added (Bloomfield e al., 1996;*Davis et al., 1998; Zanis et al., 2000, 2011; Ord óñez et al., 2005; Camalier et al., 2007).
- Bloomfield, P., Royle, J. A., Steinberg, L. J., and Yang, Q.: Accounting for meteorological effects in measuring urban ozone levels and trends, Atmos. Environ., 30, 3067-3077, https://doi.org/10.1016/1352-2310(95)00347-9, 1996.
  Camalier, L., Cox, W., and Dolwick, P.: The effects of meteorology on ozone in urban areas and their use in assessing ozone trends, Atmos. Environ., 41, 7127-7137,

170 https://doi.org/10.1016/j.atmosenv.2007.04.061, 2007.

Davis, J. M., Eder, B. K., Nychka, D., and Yang, Q.: Modeling the effects of meteorology on ozone in Houston using cluster analysis and generalized additive models, Atmos. Environ., 32, 2505-2520, https://doi.org/10.1016/S1352-2310(98)00008-9, 1998.

Ord óñez, C., Mathis, H., Furger, M., Henne, S., Hüglin, C., Staehelin, J., and Pr évôt,
A. S. H.: Changes of daily surface ozone maxima in Switzerland in all seasons from
1992 to 2002 and discussion of summer 2003, Atmos. Chem. Phys., 5, 1187-1203,
https://doi.org/10.5194/acp-5-1187-2005, 2005.

Zanis, P., Monks, P. S., Schuepbach, E., Carpenter, L. J., Green, T. J., Mills, G. P.,

- 180 Bauguitte, S., and Penkett, S. A.: In situ ozone production under free tropospheric conditions during FREETEX '98 in the Swiss Alps, J. Geophys. Res.-Atmos., 105, 24223-24234, https://doi.org/10.1029/2000JD900229, 2000.
  - Zanis, P., Katragkou, E., Tegoulias, I., Poupkou, A., Melas, D., Huszar, P., and Giorgi,F.: Evaluation of near surface ozone in air quality simulations forced by a regional
- 185 climate model over Europe for the period 1991-2000, Atmos. Environ., 45, 6489 6500, https://doi.org/10.1016/j.atmosenv.2011.09.001, 2011.

Line 167: What was "daily surface ozone" meant? Daily average or daily maximum
 8-hr average ozone concentrations? Also, the acronym for the latter would be

- 190 DM8(H)A; it's curious why the authors used "MD8A" instead. *Thanks. In this study, the ozone-weather relationship is examined using the daily mean ozone and meteorological data. We have clarified this in this revision (Lines 122-123).*
- 195 'Daily maximum 8-hour average' can also be stated as 'maximum daily 8 h mean' (Silver et al., 2018), 'daily maximum 8-hour running mean' (Fleming et al., 2018), or 'maximum daily average 8-h' (Lefohn et al., 2018). Conventionally, these expressions

can all be termed as 'MDA8'.

200 Silver, B., Reddington, C. L., Arnold, S. R., and Spracklen, D. V.: Substantial changes in air pollution across China during 2015-2017, Environ. Res. Lett., 13, 114012, https://doi.org/10.1088/1748-9326/aae718, 2018.

Fleming, ZL, et al. 2018 Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health. Elem Sci Anth, 6: 12.

205 *https://doi.org/10.1525/elementa.273.* 

Lefohn, AS, et al. 2018 Tropospheric ozone assessment report: Global ozone metrics for climate change, human health, and crop/ecosystem research. Elem Sci Anth, 6: 28. https://doi.org/10.1525/elementa.279.

4. Lines 298-299: Not clear where this came from.

Thanks. The sentence has been revised and the unclear statement has been removed.

5. Line 301-302: Was "higher meridional wind" enough to bring in "clean and humid marine air to the south" regardless of wind direction?

215 Thanks. In summer, the south-westerly monsoon wind prevails over eastern China (Figure S3). In most of the days in summer, the meridional wind blows from the south to the north. We added the explanation in Lines 320-321 in this revision.

6. Lines 305-306: How did the authors know that "the impacts of relative humidity on

- 220 surface ozone are mainly through the chemical processes"? The expressions have been revised (Lines 310-316). Relative humidity can influence ozone through various processes. Atmospheric water vapor can directly influence ozone concentrations by  $HO_x$  ( $HO_x=OH+H+peroxy$  radicals) chemistry with complicated regimes (Lu et al., 2019b). Moreover, a higher relative humidity is
- 225 usually associated with more fractions of clouds, which can slow the photochemical

production of surface ozone. In addition, higher relative humidity may somewhat be linked with larger atmospheric instability, favoring the dispersion of surface ozone (Camalier et al., 2007).

230 7. Line 321: why did R2=0.38 qualify to be "strong"?

The sentences related to  $R^2$  have been revised. In this revision, the leave-one-out cross validation is used to avoid overfitting of the MLR. The regional mean cross-validated  $R^2$  over eastern China is 43%, indicating strong performance of the MLR. Because of the large sample size (552 samples), the statistical results are at a very high

significant level, with p value being far below 0.01.

8. Lines 388: why is precipitation included in the indexes?

The air stagnation index used in this study is a common index to assess air mass stagnation (Wang and Angell, 1999; Horton et al., 2012). Precipitation is often

240 accompanied with deep or shallow convection. So, a day is considered to meet stagnation criteria, when daily total precipitation is less than 1 mm, which means a dry day.

Wang, J.X.L., and J.K. Angell, 1999: Air Stagnation Climatology for the United States

245 (1948-1998). NOAA/Air Resources Laboratory ATLAS, No.1.
Horton, D. E., Harshvardhan, and Diffenbaugh, N. S.: Response of air stagnation frequency to anthropogenically enhanced radiative forcing, Environ. Res. Lett., 7, 044034, https://doi.org/10.1088/1748-9326/7/4/044034, 2012.

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# Local and synoptic meteorological influences on daily variability of summertime surface ozone in eastern China

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## 260 Abstract

Ozone pollution in China is influenced by meteorological processes on multiple scales. Using multiple linear regression and weather classification, we statistically assess the impacts of local and synoptic meteorology on daily variability of surface ozone in eastern China in summer during 2013-2018. In this period, summertime 265 surface ozone in eastern China (110-130°E, 20-42°N) is among the highest in the world with regional means of 73.1 and 114.7  $\mu$ g m<sup>-3</sup>, respectively, in daily mean and daily maximum 8-hour average. By developing a multiple linear regression (MLR) model driven by local and synoptic weather factors, we establish a quantitative linkage between the daily <u>mean</u> ozone concentrations and meteorology in the study 270 region. The meteorology described by the MLR model can explain  $\sim 4643\%$  of the daily variability in summertime surface ozone across eastern China. The model showsthat synoptic factors contribute to ~37% of the overall meteorological effects on dailyvariability of surface ozone in eastern China. Among local meteorological factors, relative humidity is the most influential variable in the center and south of eastern 275 China including the Yangtze River Delta and the Pearl River Delta regions, while

temperature is the most influential variable in the north covering the Beijing-Tianjin-Hebei region. To further examine the synoptic influence of weather conditions

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explicitly, six predominant synoptic weather patterns (SWPs) over eastern China in summer are objectively identified using the self-organizing map clustering technique.

- The six SWPs are formed under the integral influence of the East Asian summer monsoon, the western Pacific subtropical high, the Meiyu front, and the typhoon activities. The results show thatOn regional mean, two SWPs bring about positive ozone anomalies (1.1 µg m<sup>-3</sup> or 1.7% and 2.7 µg m<sup>-3</sup> or 4.6%, respectively), when eastern China is under a weak cyclone system or under the impacts of each of
- theprevailing southerly wind. The impact of SWPs on the daily variability of surface ozone varyvaries largely inside the study area.within eastern China. The maximum impact can reach  $\pm 8 \ \mu g \ m^{-3}$  or  $\pm 16\%$  of the daily mean over<u>in</u> some subregions ineastern Chinaareas. A combination of the regression and the clustering approaches suggests a strong performance of the MLR model in predicting the sensitivity of
- 290 surface ozone in eastern China to the variation of synoptic weather. Our assessment highlights the important role of meteorology in modulating ozone pollution over China.

## **1** Introduction

- Surface ozone is a major air pollutant detrimental to human health (Jerrett et al., 2009) and vegetation growth (Yue et al., 2017). Ozone exposures are estimated to be associated with nearover 0.32 million premature deaths globally in one year (Cohen et al., 2017; Liang et al., 2018). The dominant source of surface ozone is the photochemical oxidation of volatile organic compounds (VOCs) and carbon
- monoxide (CO) in the presence of nitrogen oxides (NO<sub>x</sub>) (Monks et al., 2015). In the pastrecent decades, China has been suffering from severe ozone pollution, causing a worldwide concern (Verstraeten et al., 2015). High ozone concentrations exceeding China national air quality standard (200 and 160 µg m<sup>-3</sup>, respectively, for hourly and 8-hourly maximum values) occur frequently in major Chinese cities in the three most developed regions, the Beijing-Tianjin-Hebei (BTH) region (T. Wang et al., 2006a; G.

Li et al., 2017), the Yangtze River Delta (YRD) (Shu et al., 2016, 2019), and the Pearl River Delta (PRD) (Y. Wang et al., 2017; H. Wang et al., 2018). An increasing trend of 1-3% per year in surface ozone since 2000 is observed at urban and regional background sites in the three city clusters (Y. Wang et al., 2012; Zhang et al., 2014;

310 Ma et al., 2016; Sun et al., 2016; Gao et al., 2017) and at a global baseline station in western China (Xu et al., 2016).

Surface ozone concentrations in China largely depend on emissions and
meteorology (Han et al., 2018a, 2019); Lu et al., 2019a). Anthropogenic and natural
emissions from both native and foreign sources provide precursors for the formation
of high ozone levels in China (Ni et al., 2018; Han et al., 2019), while meteorology
can influence surface ozone variations from instantaneous to decadal scale through its
modulation of various chemical and physical processes (T. Wang et al., 2017). On a
decadal scale, both observations (Zhou et al., 2013) and simulations (S. Li et al.,
2018) show that surface ozone in southern China correlates positively to the strength
of the East Asian summer monsoon (EASM).

The daily variation of surface ozone in China is sensitive to synoptic weather systems, as illustrated by studies for BTH (Zhang et al., 2012; Huang et al., 2015),
325 YRD (Shu et al., 2016, 2019), PRD (Zhang et al., 2013; Jiang et al., 2015), and other regions of China (Tan et al., 2018). Frontal systems can drive the transboundary transport of ozone in northern China (Ding et al., 2015; Dufour et al., 2016<u>2015</u>). Downdrafts in the periphery circulation of a typhoon system can strongly enhance surface ozone before the typhoon landing in eastern or southern China (Jiang et al., 2016).

330 2015; Shu et al., 2016). Zhao and Wang (2017) suggested that a stronger western Pacific subtropical high (WPSH) can lead to lower surface ozone concentration over southern China and higher one over northern China in summer. Moreover, surface ozone concentrations also vary with mesoscale weather systems in hours (Hu et al., 2018), such as the mountain-valley circulation (T. Wang et al., 2006b) and the land-

335 sea breezes (H. Wang et al., 2018). Despite the valuablethese discussed mechanisms ofon how weather systems mentioned above influences-influence ozone concentrations in China-reported by previous studies, quantified ozone anomaliesresulted from these-, there is a lack of quantitative assessments on the influences of these weather systems are lacked on ozone pollution.

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Weather systems aton different scales bring about different changes in local meteorological variables and thus surface ozone through their impacts on, which, in turn, impact chemistry and physical processes- that modulate surface ozone concentrations. However, the relative importance of variouslocal meteorological 845 factors to surface ozone concentrations inin different regions of China are still unclear. Previous studies suggested the importance of temperature, relative humidity, and winds to surface ozone in different regions (Lou et al., 2015; Pu et al., 2017; Zhan et al., 2018). The key influential meteorological factors vary from cities region to citiesin Chinaregion (Gong et al., 2018; Chen et al., 2019). In general, high ozone episodes 350 commonly appear under weak wind, high temperature, low humidity, and clear conditions- (Bloomfield e al., 1996; Zanis et al., 2000, 2011; Ord óñez et al., 2005). These weather conditions can enhance stagnation and production of ozone (Camalier et al., 2007; Shen et al., 2017a). Variations of these local meteorological variables depend on the dominant weather systems (Davis et al., 1998; Han et al., 2018b; Leung 355 et al., 2018).

To have a comprehensive and quantitative understanding of how weather influences ozone pollution in China is the primary motivation of this study, in which, we aim to quantify the impacts of meteorology, specifically the dominant synoptic weather systems and the key <u>local</u> meteorological variables, on daily variations of surface ozone in eastern China, including the three representative megacity clusters, BTH,

13 / 37

YRD, and PRD. Surface ozone in China was not regularly and systematically monitored until 2012 when real-time hourly ozone data were available online from China Ministry of Ecology and Environment (MEE) (http://www.mee.gov.cn/) (T.

Wang et al., 2017). Owing to the limitation of in situ measurement, there is a lack of a

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long-term assessment on the synoptic influence on ozone pollution.

TheIn this study, the ground ozone observations from MEE covering 2013-2018period are used. First, we characterize the seasonal variations of surface ozone in370eastern China and the interannual changes during 2013-2018 in summer (June-August), which is the season of interest in this study. Second, we search for a linkagebetween the daily variation of surface ozone and the local and synopticmeteorological factors statistically and develop a multiple linear regression (MLR)model based on the linkage. Third, we examine the sensitivity of daily surface ozone

- 375 to the variation in synoptic weather systems. Considering the complexity of the synoptic meteorology in eastern China (Ding et al., 2017; Han et al., 2018b), we employ an objective clustering technique, the self-organizing map (SOM), to identify the predominant synoptic weather patterns (SWPs). In the following sections, we introduce the data and methods in section 2. The seasonal and interannual variations
- 380 of surface ozone in eastern China are characterized in section 3. Section 4 illustrates the linkage between ozone variability and meteorology on both local and synoptic scales, while section 5 describes sensitivity of surface ozone to various typical SWPs over the entire eastern China. Finally, we discuss our results and draw conclusions in section 6.

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#### 2 Data and methods

#### 2.1 Surface ozone observations and meteorological data

Hourly surface ozone measurements from the MEE observation network averaged over <u>the</u> stations in each city were used in the study. The measurements were

- downloaded from http://beijingair.sinaapp.com/, which were previously archived at http://pm25.in, a mirror of data from the official MEE publishing platform (http://106.37.208.233:20035/). The network covers 63 cities in eastern China (110-130°E, 20-42°N) in 2013, increasing to 118 in 2014 and 185 during 2015-2018. Locations of the 185 cities are shown in Figure 1, including 13, 26, and 9 cities,
- respectively, in BTH, YRD, and PRD. <u>The unit of ozone concentrations in the original</u> records and in this study is ' $\mu$ g m<sup>-3</sup>', with a conversion factor of 1  $\mu$ g m<sup>-3</sup> = 0.47 ppbv at 273 K and 1013.25 hPa.

The National Centers for Environmental Prediction (NCEP) Final (FNL)

- Operational global analysis data during the same period were acquired from https://rda.ucar.edu/datasets/ds083.2/. The data are available on 1°×1° latitude grids every 6 hours forat the surface and at 26 layers from 1000 to 10 hPa. We made daily averaged pollution and meteorological data in summer from 2013 to 2018. The ozone-weather relationship is examined using the daily mean data, unless stated otherwise.
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Using <u>an</u> inverse-distance weighting (Tai et al., 2010), we interpolated the dailyeitypollution measurements from the cities onto the FNL grid (1° latitude and  $\times 1^{\circ}$ longitude) to produce continuous gridded data. Ozone at each FNL grid was calculated with a weighted average of the concentration in the cities within a search distance ( $d_{max}$ ) from that grid, following the equation:

$$z_{j} = \frac{\sum_{i=1}^{n_{j}} (1/d_{i,j})^{k} z_{i}}{\sum_{i=1}^{n_{j}} (1/d_{i,j})^{k}}$$
(1)

where  $z_j$  is the calculated ozone at grid j,  $z_i$  is the observed ozone in city i,  $d_{i,j}$  is the distance between city i and the center of grid j,  $n_j$  is the number of the cities within  $d_{max}$  from grid j ( $d_{i,j} \leq d_{max}$ ), k is a parameter measuring the influence of distance on the target grid. We used 2 for k, and 1-degree distance in latitude-longitude grid for  $d_{max}$  in the interpolation. The generated gridded ozone data cover most of the mainland in eastern China (Figure 1c). The measurements interpolated to the grids 15/37

were used in this study, unless stated otherwise.

### 420 **2.2 Development of a prediction model of surface ozone**

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MLR is an effective and widely-used way to describe the relationship between meteorology and air quality and thus to help prediction of air quality (Shen et al., 2015; Otero et al., 2016; K. Li et al., 2019). MLR establishes a linear function between a scalar response and the explanatory variables. In this study, we applied a stepwise MLR model to quantitatively correlate daily surface ozone in eastern Chinaand meteorology in summer. Considering the combined effect of meteorology at various scales, we used both local meteorological variables and synoptic circulation factors as predictors following Shen et al. (2017b), who showed that, comparing with regression models only considering local meteorology, adding the synoptic factors in a MLR can significantly improve the model performance. The MLR-model takes the following form:

 $\hat{Y} = b + \sum_{i=1}^{K_1} \alpha_i X_i + \sum_{j=1}^{K_2} \beta_j S_j \qquad (2)$ 

where  $\hat{Y}$  is the predicted value of surface ozone, *b* is the intercept term, *X<sub>i</sub>* is the local meteorological variables with a total number of *K<sub>1</sub>*, *S<sub>j</sub>* is the synoptic meteorological factors with a total number of *K<sub>2</sub>*, and  $\alpha_i$  and  $\beta_j$  are the regression coefficients. We used 10 local meteorological variables (*K<sub>1</sub>*=10), i.e.,including relative humidity at 2 m (RH2m), cloud fraction (CF), temperature at 2 m (T2m), planetary boundary layer height (PBLH), zonal wind at 850 hPa (U850), meridional wind at 850 hPa (V850), vertical wind at 850 hPa (W850), wind speed at 850 hPa (WS850), geopotential height at 850 hPa (HGT850), and sea level pressure (SLP), all of which were identified significantly (p<0.05) correlated to the daily variations of surface ozone in part of eastern China, as shown in Figure 2. Cloud fraction retrievals at 1°×1° grids were from the spaceborne Atmospheric Infrared Sounder (AIRS) instrument

(AIRS3STD daily product, https://disc.gsfc.nasa.gov/). The other 9 local meteorology

445 were from FNL data (section 2.1). We computed the anomalies of meteorological 16/37

variables and ozone on a given day by taking the difference between the value of a given meteorological variable (or ozone) on that day and the mean value of the <u>meteorological</u> variable (or ozone) in that month. Thus, all the data were detrended and the influences of meteorology on the ozone variability on longer time scales (trends, and annual and seasonal variations) were generally removed. Any anomaly of a variable (or ozone) divided by its corresponding monthly mean is referred as relative anomaly of that variable (or ozone).) with a unit of %.

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We For  $S_i$  in equation (2), we also used identified two synoptic factors generated from through the singular value decomposition (SVD) of the spatial correlations 455 between surface ozone and local meteorological variables in eastern China (Shen et al., 2017b). The SVD approach effectively extracted representative signals from the spatial distribution of the correlation coefficients. The extracted information was then used to characterize the spatial patterns of the meteorological variables aton a 460 synoptic scale by inversing SVD. For each of the FNL grids in eastern China, we constructed the synoptic circulation factors as follows. First, we calculated the correlation coefficients between daily mean surface ozone at that given grid and each of the 10 meteorological variables at all the grids in eastern China in summer during 2013-2018. For example, the correlations for the grid of Nanjing are shown in Figure 465 S1, which indicates that surface ozone in Nanjing is correlated to the meteorology in the surrounding regions. We made a matrix A that consists of the correlation coefficients for that grid with elements of 21 (numbers of grids in longitude)  $\times 23$ (numbers of grids in latitude)  $\times 10$  (numbers of the local meteorological variables). Second, to fit the decomposition, we aligned the dimension of longitude-latitude into 470 one column and reshaped matrix A into a 483 (longitude  $\times$  latitude)  $\times$  10 two-

dimensional matrix *F*. The SVD decomposed *F* used the equation:

$$F = ULV^T$$
 (3)

where U is  $483 \times 10$  matrix, L is a  $10 \times 10$  diagonal matrix with non-negative numbers

on the diagonal, V is also a  $10 \times 10$  matrix. The columns of the three transformations

- 475 together characterize SVD modes, with 10 modes in total. Each column of U represents the spatial weights of the SVD mode and each column of V represents the variable weights in the SVD mode. The spatial and variable weights of the first two SVD modes for the Nanjing grid are shown in Figure S2. The pattern of the spatial weight of the first SVD mode for the Nanjing grid (Figure S2a) is similar to the
- pattern of the correlations between surface ozone and relative humidity (Figure S1a) and cloud fraction (Figure S1b). The first SVD mode is more correlated to relative humidity and cloud fraction than other variables (Figure S2b). Therefore, the first SVD mode for <u>the Nanjing grid is related to chemical processes of ozone</u>. In contrast, the second SVD mode for <u>the Nanjing grid is more related to transport than chemical processes (Figure S2d)</u>. Third, we assigned the anomalies of the daily mean values of the 10 local meteorological variables in eastern China to a 552 (days in summer of 2013-2018) ×21 (longitude)×23 (latitude)×10 (meteorology) four-dimensional matrix *M*. At each grid, we normalized the time series of each variable to zero mean and unit standard deviation. Then, the magnitude of each SVD mode for every day *t* was calculated by inversing SVD:

$$S_{k,t} = \boldsymbol{U}_k^T \boldsymbol{M}_t \boldsymbol{V}_k \qquad (4)$$

where  $U_k$  and  $V_k$  respectively are the  $k^{th}$  columns of U and V, respectively.  $S_{k,t}$  is a scalar depicting the magnitude of the  $k^{th}$  SVD mode.  $S_{k,t}$  refers to a newly produced meteorological field and reflects that represents the influence of synoptic factorsrelated tometeorology on ozone variability. We implemented the procedure at every-FNL grid in eastern China. The first two SVD modes can generally explain 55-85% of the total variance. They can respectively reflect the dynamical or thermal characteristics in theof synoptic meteorology (Shen et al., 2017b). Therefore, we applied the primary two SVD modes in the MLR model ( $K_2$ =2).

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We used the leave-one-out cross validation to avoid overfitting of the MLR for each

grid. Data during the study period (summer over 2013-2018) included 552-day
observations. Each time, one observation in the time series was reserved as the test set
and the remaining ones were used as the training set. The process was repeated until
all observations had been predicted. Every observation was to be a test set once and a
training set 551 times.

We measured the relative importance of each of the meteorological variables to ozone by its relative contributions to the total explained variance of the MLR-model. The weight of each predictor ( $w_i$ ) was calculated from the normalized MLR coefficient ( $z_k$ ):

$$w_i = \frac{z_k^2}{\sum_{k=1}^{12} z_k^2}$$
 (5)

where  $z_k$  is:

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$$z_k = \frac{s_k}{s_y} c_k \qquad (6)$$

and <u>12 is</u> the number of all the predictors <u>is 12</u>, including 10 local and 2 synoptic\_ <u>meteorological</u> factors (section 2.2).  $c_k$  is the regression coefficient, referring to  $\alpha_i$  or  $\beta_j$  in equation (2).  $s_k$  is the standard deviation of a predictor, <u>i.e.</u>,  $X_i$  or  $S_j$  in equation (2).  $s_y$  is the standard deviation of the observed daily surface ozone.

# 520 **2.3 Classification of the synoptic weather patterns**

Weather classification is a well-established tool to characterize atmospheric processes aton multiple scales and further to study air pollution-weather relationship (Han et al., 2018b). The methods for weather classification can be generally categorized into three groups: subjective, mixed, and objective, depending on the automatic degree during

525 the classification process (Huth et al., 2008). The methods can also be categorized in more detail according to the basic features of each classification algorithm (Philipp et al., 2014). Depending on the study domain and research objectives, different meteorological variables including geopotential height, mean sea level pressure, and zonal and meridional winds are used for the classification.

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SOM, an artificial neural network method with unsupervised learning (Kohonen, 1990; Michaelides et al., 2007), is widely used in cluster analysis in atmospheric sciences (Jiang et al., 2017; Liao et al., 2018; Stauffer et al., 2018) because of its superiorities over other algorithms (Liu et al., 2006; Jensen et al., 2012). SOM

- 535 performs a nonlinear projection from the input data space to a two-dimensional array of nodes objectively. Each node is representative of the input data. SOM allows missing values in the input data and can effectively visualize the relationships between different output nodes (Hewitson and Crane, 2002).
- 540 The FNL geopotential height fields (section 2.1) at 850 hPa can well capture the synoptic circulation variations over eastern China (Han et al., 2018b). In this study, we used geopotential height at 850 hPa in 2013-2018 as the input for SOM. Each of the SOM output nodes corresponds to a cluster of SWPs. Finally, we identified six predominant SWPs over eastern China in summer. All days in summer of 2013-2018 545 were included in the clustering results.

#### 3 Seasonal and interannual variations of surface ozone in eastern China

Figure 3 and Figure 4, respectively, show the seasonal and interannual variations of
the regional mean surface ozone concentrations in eastern China and the three
subregions (BTH, YRD, and PRD) during 2013-2018. Among *n* cities with air quality
monitoring in a given region, if ozone levels exceed the national air quality standard
in *m* cities, we defined the ratio of *m* to *n* as the regional exceedance probability of
ozone (Figure 3c). Higher regional exceedance probability implies ozone pollution
over wider surface areas in that region. Primary pollutant (Figure 3d) is defined in the
Air Quality Index (AQI) system, in which, AQI for an individual air pollutant is
calculated based on the concentrations of thethat pollutant. When the individual AQI

20 / 37

of a pollutant on a day is both above 50 and the largest among all the pollutants-on, that day this pollutant is defined as the primary pollutant on that day.

560 On-the regional average, the seasonality of daily mean ozone is similar to that of daily maximum 8-hour average (MDA8) ozone in eastern China, as well as in the three subregions (BTH, YRD, and PRD) (Figures 3a and 3b). In BTH, both daily mean and MDA8 have a unimodal seasonal pattern and peak in June, being 99.5 and 158.4  $\mu$ g m<sup>-3</sup>, respectively. The extremely high ozone in June leads to a simultaneous 565 seasonal maximum in both probability of the regional exceedance (46.9% of the cities with ozone measurements in BTH) and primary pollutant (68.7% of the days in June) (Figures 3c and 3d). The seasonal peak of surface ozone in BTH mainly results from enhanced photochemistry due to stronger solar radiation and lower humidity (Hou et al., 2014). Surface ozone over YRD reaches a seasonal maximum in May (82.6 and 570 127.7 µg m<sup>-3</sup>, respectively, for daily mean and MDA8 ozone), earlier than that over BTH. While the seasonal peak over PRD occurs the latest in October (71.5 and 118.1 µg m<sup>-3</sup>, respectively, for daily mean and MDA8 ozone). Although the temperature is higher in summer than in the other seasons, the EASM brings more cloudy weather, stronger convection, and clearer air from the oceans, weakening the production and 575 accumulation of surface ozone over YRD and PRD (Hou et al., 2015; S. Li et al., 2018). The pre-monsoon and post-monsoon peak of surface ozone were also found in YRD and PRD, respectively (He et al., 2008; T. Wang et al., 2009).

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On the regional and seasonal averageaverages, daily mean and MDA8 ozone over eastern China in summer are 73.1 and 114.7 µg m<sup>-3</sup>, respectively. Among the three city clusters subregions, summertime surface ozone is highest in BTH (88.3 and 143.7 µg m<sup>-3</sup>, respectively, for daily mean and MDA8 ozone), second highest in YRD (72.9 and 114.7  $\mu$ g m<sup>-3</sup>), and lowest in PRD (51.0 and 91.9  $\mu$ g m<sup>-3</sup>) (Figures 3a and 3b). These regional differences among the three *city clusters* subregions appear similar to

21/37

585 these in the Ozone Monitoring Instrument (OMI) tropospheric column ozone (Figure 1). The regional exceedance probability of ozone over eastern China reaches 17.7% in summer, accompanied with a high percentage (45.6%) of ozone being the primary pollutant. Among the three subregions, **<u>BTH has the highest</u>** regional exceedance probability of ozone (35.1%) and the probability of ozone being the primary pollutant (55.8%) are largest in BTH.%).

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A rapid increasing trend in summertime surface ozone over China after 2012 was observed in recent studies (Lu et al., 2018; Silver et al., 2018; Shen et al., 20192019a; K. Li et al., 2019). We examine the regional mean trendtrends over eastern China in 595 daily, daytime (7:00-18:00), and nighttime (19:00-6:00) means (Figure 4). Significant (p<0.05)-summer increasing trends (p<0.05) of approximately 3-6 µg m<sup>-3</sup> or 4-8% per year are found over eastern China, BTH, and YRD during 2013-2018, while the increasing trend over PRD during the period is insignificant (p>0.05). Silver et al. (2018) found the annual mean MDA8 ozone has increased significantly (p<0.05) at 600 around ~50% of the over 1000 stations across China from 2015 to 2017, with a median rate of 4.6 µg m<sup>-3</sup> year<sup>-1</sup>. The increasing trend over eastern China was also captured by the OMI satellite records of tropospheric ozone, reported by Shen et al.  $(\frac{20192019a}{2019a})$ . The absolute increasing trend (in a unit of  $\mu g m^{-3}$ ) in daytime is higher than that in nighttime, whereas the relative increasing trend (in a unit of %) in daytime 605 is lower than that in nighttime (Figures 4e-4h vs. Figures 4i-4l). The increasing ozone trend over China may result from both meteorology and anthropogenic emissions. During 2013-2017, the anthropogenic emissions of  $NO_x$  in China declined (Zheng et al., 2018) but the anthropogenic emissions of VOCs changed little (Zheng et al., 2018; Shen et al., 2019b). K. Li et al. (2019) suggested the ~40% decrease of fine particulate 610 matter (PM<sub>2.5</sub>) is the primary reason for the increasing trend of surface ozone in summer during 2013-2017, as the aerosol sink of hydroperoxy radicals was weakened

and thus ozone production was enhanced. Figure 44b demonstrates a strong increase

in summertime surface ozone over BTH from 2016 to 2017, which is probably related to the hot extremes in 2017 (Herring et al., 2019). The sudden decline in summertime

615 surface ozone over PRD from 2016 to 2017 (Figure 4d) is likely associated with the extremely heavy precipitation in 2017 (Herring et al., 2019).

#### 4 Meteorological drivers for summertime surface ozone in eastern China

- Meteorological factors can individually or integrally modulate surface ozone 620 concentration through their impacts on relevant chemical, dynamical, and thermal processes in the atmosphere. Figure 2 shows a simple way to examine the overall effect of each of the meteorological variables statistically by correlating surface ozone with a selected set- of local meteorological variables during 2013-2018 summer.
- 625 correlation with surface ozone in eastern China on regional mean-, (r=-0.39). Relative humidity can influence ozone through various processes. Atmospheric water vapor can directly influence ozone concentrations by HO<sub>x</sub> (HO<sub>x</sub>=OH+H+peroxy radicals) chemistry in complicated ways (Zanis et al., 2002; Jacob et al., 2009; Lu et al., 2019b). Moreover, a higher relative humidity is usually associated with more fractions

Among all the meteorological variables, relative humidity shows the highest

- of clouds, which can slow the photochemical production of surface ozone. Higher
   relative humidity may also somewhat be linked with larger atmospheric instability,
   favoring the dispersion of surface ozone (Camalier et al., 2007). The correlation map
   of cloud fraction is similar to that of relative humidity (Figures 2a and 2b). The
   correlation of temperature with ozone is higher in the north than in the south over
- eastern China (Figure 2c), which may dueis similar to lower humidity in the north.pattern found in the eastern United States (Camalier et al., 2007; Shen et al., 2016). Meridional wind at 850 hPa is positively correlated to surface ozone positively in the north but negatively in the most areas of the south (Figure 2f). In summer, the south-westerly monsoon wind prevails over eastern China (Figure S3). Higher
- 640 meridional wind brings clean and humid marine air to the south, while it transports

ozone and its precursors from the south to the north. All the meteorological variables are not independent with each other. For example, relative humidity is stronglycorrelated with cloud fraction. A higher relative humidity is usually associated with more fractions of clouds, which can slow the photochemical production of surfaceozone. The impacts of relative humidity on surface ozone are mainly through thechemical processes. In addition, higher relative humidity may somewhat be linkedwith larger atmospheric instability, favoring the dispersion of surface ozone (Camalier et al., 2007). Overall, the meteorological variables that are related to photochemistry processes (relative humidity, cloud fraction, and temperature) have more significant correlation than transport-related variables (zonal, meridional, and vertical winds and wind speed) (Figure 2), implying greater effects of chemical process is the uppermost factor controlling surface ozone levels over eastern China in summer.

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Combining the effects of different meteorological variables, we applied the MLR
model (section 2.2) using predictors of both local and synoptic factors (section 2.2) to simulate summertime daily surface ozone in eastern China. The MLR model-was developed using the observation data in 2013-2017 and evaluated with observation-data in 2018. using the leave-one-out cross validation to avoid overfitting. The MLR
model-performs strongly as it can explain 3014-65% variations in the observed surface ozone concentrations in 2013-2017, yielding a regional mean coefficient of determination (R<sup>2</sup>) of 4643% (Figure 5e). 5a). The mean absolute error (MAE) and the root mean square error (RMSE) of regional mean ozone anomalies in eastern China between observations and predictions by the MLR are 12.0 and 7.1 μg m<sup>-3</sup>,
respectively (Figure 6a). Geographically, the model performs better in the south (R<sup>2</sup>=0.5251 in YRD and R<sup>2</sup>=0.5449 in PRD) than in the north (R<sup>2</sup>=0.4442 in BTH)

(R<sup>2</sup>=0.38 in eastern China) (Figure 5d). Moreover, we simulated surface ozone

(Figure 5c). In the validation period, the model also shows strong performance-

24 / 37

considering 5a). Compared with the simulation that only considers the local

- 670 meteorological variables (Figure 5a) and only the synoptic factors (Figure 5b)-in the MLR-model. Compared with the two simulations (Figures 5a and 5b),, the model performance is overall improved in areas in eastern China when both local and synoptic meteorological factors are considered (Figure 5eFigures 5a vs. 5b). Shen et al. (2017b) found that, compared with the<u>a</u> MLR model describing that describes
- monthly PM<sub>2.5</sub> in the United States driven by only the<u>using</u> local
  meteorologymeteorological factors, the inclusion of the synoptic meteorological
  factors in the MLR model increases R<sup>2</sup> from 34% to 43%. In addition, we We also
  conducted the stepwise MLR model using local and synoptic meteorology without
  detrending the input data. The results show that meteorology can explain 3918% of
  the increasing trend in the regional mean of summertime surface ozone over eastern
  China from 2013 to 2018, and the explained variance is 23%, 5316%, 41%, and
  5744% for BTH, YRD, and PRD, respectively (Figure \$354).
- We applied the MLR model to identify the dominant meteorological drivers for 685 ozone variability (section 2.2). Synoptic factors diagnosed by SVD are the mostpronounced drivers in ~45% areas of eastern China and contribute to 30-60% of the meteorological effects on surface ozone over these locations (Figure 6b). The regional mean contributions of the synoptic factors are 37% over eastern China and 41% over-BTH, YRD, and PRD (Figure 6b). Among the local meteorology, relative humidity is 690 dominant over  $\sim$ 4851% areas of eastern China, mainly in the central and the southern regions including YRD and PRD (Figure 6c). Relative humidity is estimated toaccount for ~30% of the meteorological impacts on daily surface ozone variation in-YRD and PRD on the regional scale (Figures 7c and 7d), although at 5c), although on a city scale in PRD, Zhao et al. (2016) suggested that sea level pressure is the most 695 significant variable for MDA8 ozone in Hong Kong. Air temperature is the most important local meteorological variable in  $\sim \frac{1517}{9}$ % areas of eastern China,

specifically in the north including BTH (Figure <del>6c). Temperature is estimated to account for 20% of the meteorological impact in BTH (Figure 7b<u>5c</u>). The importance of temperature to surface ozone over BTH was also suggested by Chen et al. (2019).</del>

Previous studies found that temperature and relative humidity showed pronounced impact on ozone in the north and south of the eastern USUnited States, respectively (Camalier et al., 2007; Porter et al., 2015). The difference of the most influential variables between the south and north in eastern China is similar to that in the eastern United States. In Europe, Otero et al. (2016) suggested that temperature is the most important local meteorological driver over a major part of Europe. On regional average, the second most important meteorological variable for the daily surface ozone variation in eastern China, BTH, YRD, and PRD is temperature, relative humidity, geopotential height at 850 hPa, sea level pressure, and meridional wind at

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#### 5 Synoptic impacts on summertime surface ozone in eastern China

850 hPa, respectively (Figure 76).

In the last section, we have shown that both local and synoptic meteorological factors are important forto surface ozone <u>variations</u> in eastern China. The synoptic factors used there were extracted via an inversing SVD process and do not stand for specific weather systems. In this section, we <del>will</del> further <del>showinvest</del> how the specifiedifferent synoptic weather systems influence surface ozone in eastern China by looking into the typical SWPs. Atmospheric circulations over eastern China in summer are largely regulated by the evolution of the components of EASM, for instance, the <del>western</del> Pacific subtropical high (WPSH), WPSH, the subtropical westerly jet, the Meiyu front, and the Southwest Vortex (Ding and Chan, 2005). Among these systems, the WPSH can largely modulate the seasonal migration of the rain belt over eastern China. Typhoon is also an influential weather system, especially on the southeast coastal regions. The main features of the synoptic circulations over eastern China during 2013-2018 can be represented by six predominant SWPs (Figures <u>8-137-12</u>), which

- were identified by an objective approach, SOM (section 2.3). The occurrence
  frequency of these SWPs is shown in Figures 8-137-12. We name the six SWPs by
  their dominant weather systems or prevailing wind, including Pattern 1 featured
  southwesterly wind (P1 or PSW), Pattern 2 featured Southerly wind (P2 or PS),
  Pattern 3 featured Northeast Cold Vortex (P3 or PNECV), Pattern 4 featured a weak
  cyclone (P4 or PWC), Pattern 5 featured strong WPSH (P5 or PSWPSH), and Pattern
- cyclone (P4 or PWC), Pattern 5 featured strong WPSH (P5 or PSWPSH), and Patter
  6 featured typhoon systems (P6 or PTC) (Table 1).

To compare the differences of meteorological conditions among the six SWPs, we calculated the daily EASM index (EASMI) and WPSH index (WPSHI) representing
the strength of EASM and WPSH respectively. The two indexes were normalized to zero mean and unit standard deviation. The averaged anomalies of the normalized indexes under each SWP are shown in Figures 8-137-12 and Table 1. The EASMI is a shear vorticity index defined as the difference of the regional mean zonal wind at 850 hPa between 5-15°N, 90-130°E and 22.5-32.5°N, 110-140°E in B. Wang and Fan

- 740 (1999) recommended by B. Wang et al. (2008). The WPSHI is defined by the accumulative enhancement of geopotential height above the WPSH characteristic isoline (5880 gpm at 500 hPa) averaged over the area north to 10°N. The WPSHI is adopted by the National Climate Center in China (https://cmdp.ncc-cma.net) in the monitoring and diagnosis of the atmospheric circulation. Using the WPSHI, Zhao and
- Wang (2017) found a significant correlation between the WPSH and the first empirical orthogonal function (EOF) pattern of surface ozone in China. Moreover, we used the averaged anomalies of the meteorological variables in a SWP to describe that SWP. We used the averaged ozone anomaly (in µg m<sup>-3</sup>) (Figures 8-137-12) and the averaged relative ozone anomaly (the ozone anomaly divided by the monthly ozone
- mean, in %) (<u>Table 1 and Figure 14S5</u>) under a SWP to assess the influence of that SWP on ozone (Han et al., 2018b). Furthermore, a common index for air stagnation (Horton et al., 2012) is used to assess the impact of air stagnation on surface ozone.

For each FNL grid, when <u>the</u> daily average wind speed at 10 m, daily average wind speed at 500 hPa, and the daily total precipitation <u>on a day</u> are respectively less than 3.2 m s<sup>-1</sup>, 13 m s<sup>-1</sup>, and 1 mm, the day is considered as a stagnant day <u>forat</u> that grid. The National Oceanic and Atmospheric Administration (NOAA) Climate Prediction Center (CPC) precipitation data

(https://www.esrl.noaa.gov/psd/data/gridded/data.cpc.globalprecip.html) were used in the calculation of the air stagnation index.

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The characteristics of the six SWPs and their impacts on surface ozone are briefly summarized in Table 1. PSW (P1) is the most common circulation pattern occurring in 25% days of summer during 2013-2019 (Figure <u>8b7b</u>). Characterized with weak EASM conditions, PSW is dominated by an anomalous anticyclone located in the southeast of eastern China (Figure 8e7e). In PSW, the enhanced meridional wind 765 brings clear marine air to the south of eastern China (Figure 8i7i), where the meridional wind is significantly correlated to surface ozone (Figure 2f). The enhanced zonal wind from the anomalous anticyclonic circulation (Figure 8e7e) increases the ozone export from the south of eastern China (Yang et al., 2014). The negative 770 anomalies of temperature (Figure  $\frac{8g7g}{g}$ ), and positive anomalies of relative humidity (Figure  $\frac{8f7f}{1}$ ) and cloud fraction (Figure  $\frac{8h7h}{1}$ ) in the south are <u>all</u> unfavorable for photochemical processes. In consequence, PSW reduces ozone levels in the south (Figure <u>8e7c</u>) by enhancing the dispersion and suppressing the production of ozone. Negative anomalies of -1.5 (-2.4%) and -6.6  $\mu$ g m<sup>-3</sup> (-13%) in the regional mean 775 ozone are respectively observed over YRD and PRD (Figures 8c, respectively (Figure 7c and 14aTable 1). In contrast, the lower cloud fraction (Figure 8h7h) and higher temperature (Figure  $\frac{8g7g}{g}$ ) in the north stimulate ozone production. Surface ozone over BTH increases by 3.4  $\mu$ g m<sup>-3</sup> (3.6%) from the regional mean in PSW (Figures-<u>8cFigure 7c</u> and <u>14aTable 1</u>).

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PS (P2) is the second frequent SWP (Figure 9b8b), characterized with strong EASM and weak WPSH (Figure 9a). In8a). Under PS, the FNL meteorological data illustrateshow frequent stagnation events (Figure 9181), low humidity (Figure 9f8f), and low cloud fraction (Figure 8h) over most of eastern China. In contrast to PSW, the zonal wind has negative anomalies (Figure 9e8e) in PS, reducing ozone export from the south of eastern China. Overall, an increase of 1.1  $\mu$ g m<sup>-3</sup> (1.7%) in the regional mean ozone concentrations is resulted in eastern China is observed inunder PS (Figures 9eFigure 8c and 14bTable 1).

PNECV (P3) is a typical pattern for Meiyu, an important climate phenomenon over the middle and lower reaches of the Yangtze River duringfrom early June to mid-July (Figure 10a),9a). PNECV is characterized by persistent rainfall (Ding and Chan, 2005). Under a combined effect of the Northeast Cold Vortex and the WPSH, Meiyu front forms and maintains over YRD (He et al., 2007). Meiyu in PNECV increases
relative humidity (Figure 10f9f) and decreases air stagnation (Figure 10l9l) over YRD. Consequently, PNECV reduces surface ozone concentrations by 1.3 µg m<sup>-3</sup> (1.7%) over YRD (Figures 10eFigure 9c and 14eTable 1). Meantime, more sunny days with high temperature (Figure 10g9g) and low moisture (Figure 10f9f) occur in the north to YRD, affected by the northwesterly and downward airflows from the Northeast Cold Vortex (Figure 10a9a). As a result, positive ozone anomalies are observed in the regions north of YRD (Figure 10e9c).

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PWC (P4) features the weakest WPSH, when a weak extratropical cyclone locates over the east of the mainland China (Figure <u>11a10a</u>). The extratropical cyclone is probably formed by <u>thean</u> eastward movement of the Southwest Vortex or <u>thea</u> transition from <u>a</u> typhoon. Pushed by the cyclone, the WPSH retreats (Y. Li et al., 2018). The weak pressure gradient over the mainland of eastern China (Figure <u>11a10a</u>) in PWC results in more stable weather conditions. The anomalies of the

29 / 37

meteorological variables in PWC show opposite spatial patterns to those in PSW

(Figure <u>87</u> vs Figure <u>110</u>). With the favorable meteorological conditions except temperature, PWC enhances ozone over the south, with. PWC statistically increased regional mean values of ozone by 5.2 μg m<sup>-3</sup> (7.5%) over YRD and 6.7 μg m<sup>-3</sup> (11.8%) over PRD (Figures <u>11eFigure 10c</u> and <u>14dTable 1</u>). Mean negative ozone anomalies of -4.8 μg m<sup>-3</sup> (-5.1%) are observed over BTH in PWC (Figure 10c and Table 1).

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PSWPSH (P5) occurs in late summer (Figure 12a11b), when Meiyu breaks in the Yangtze River and the rain belt jumpsshifts to North China (Ding and Chan, 2005). In PSWPSH, the WPSH is the strongest and extends westward the-mostly (Figure 12a11a). Thus, relative humidity is lower than the seasonal mean over YRD and higher than the seasonal mean over BTH (Figure 12f11f). Meantime, stable weather conditions occur more frequently over YRD (Figure 12f11f). Therefore, ozone accumulates over YRD in PSWPSH with a regional mean enhancement of 1.8 μg m<sup>-3</sup> (2.5%) (Figures 12eFigure 11c and 14eTable 1). Surface ozone decreases by 0.8 (1.4%) and 5.0 μg m<sup>-3</sup> (8.9%)%), respectively, over BTH and PRD under this SWP (Figures 12eFigure 11c and 14eTable 1).

PTC (P6) is a typical typhoon weather pattern that is over the southeast coast of the mainland China (Figure 13a12a). Forced by <u>a</u> typhoon <u>system</u>, the WPSH in PTC migrates further north than under the other SWPs. The typhoon <del>system</del> brings clear and moist marine air to coastal <u>eitiesareas</u> in eastern China, reducing surface ozone by 6.8  $\mu$ g m<sup>-3</sup> (9.2%) over YRD (Figures 13cFigure 12c and 14fTable 1). Shu et al. (2017) identified that SWPs like PTC can lead to clean PM<sub>2.5</sub> episodes in YRD. However, the cyclonic circulation enhances ozone transport from the central part of eastern China to the downwind regions in the south including PRD. The collective effect of higher temperature, lower humidity, and heavier downdrafts, PTC increases surface ozone in PRD by 7.9  $\mu$ g m<sup>-3</sup> (15.5%) (Figures 13cFigure 12c and 14fTable 1).

Lam et al. (2018) found ozone increases by 16.8  $\mu$ g m<sup>-3</sup> at urban stations in Hong Kong of PRD, when the synoptic circulation controlling PRD is featured typhoon in the vicinity of Taiwan, similar to PTC. They also suggested that this SWP is associated with the interannual variations of ozone pollution in Hong Kong.

We further compared the SWPs analysis with that from the MLR model-discussed in section 4. We evaluate the performance of the MLR model under the six SWPs based on the predicted (Figures 7d, 8d, 9d, 10d, 11d, 12d, and 13d) and observed (Figures 7c, 8c, 9c, 10c, 11c, <del>12c,</del> and <del>13c</del>12c) ozone anomalies. The comparison 845 shows that the ozone anomalies predicted by the MLR have spatial variations and magnitudes similar to those in the observations under each of the SWPs. The MAE of averaged ozone anomalies under each of the SWPs ranges  $1.0-2.2 \ \mu g \ m^{-3}$ , and the RMSE ranges 1.4-2.8 µg m<sup>-3</sup> (Table S1). The MLR model can well capture the ozone 850 anomalies under the six predominant SWPs (Figures 8-137-12). For example, the negative ozone anomaly over PRD under PSW (P1) featured weak EASM (Figures 8e7c vs. 8d7d), the negative ozone anomaly over YRD under PNECV (P3) featured Meiyu (Figures 10c9c vs. 10d)9d), and the positive ozone anomaly over PRD caused byunder PTC (P6) featured typhoon (Figures 13c12c vs. 13d12d). Since the MLR-855 model only considers the meteorological influence on surface ozone, the consistency between the regression and the clustering results suggests that the mean observed ozone anomalies under a SWP can adequately reflect the sensitivity response of daily ozone variation to meteorology. The noise of day-to-day variations of chemistry and emissions in the surface ozone data can be largely removed by long-term average of 860 ozone anomalies under a SWP from the big data set of surface ozone (Han et al., 2018b).

## **6 Discussion and conclusions**

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In addition, we applied the MLR to reveal the most important local meteorological

- <u>factor for daily ozone variability under each of the six SWPs (Figure S6). The MLR</u> <u>was conducted under each of the SWPs with the same procedures in the full summer.</u> <u>The most important meteorological variable for ozone over some areas in eastern</u> <u>China may vary with the prevailing SWP (Figure S6). The dominant driver in PRD is</u> <u>meridional wind at 850 hPa under PSW, PS, and PSWPSH, demonstrating the</u>
- 870 <u>significant influences of marine air inflow. Controlled by the typhoon system, the</u> <u>most important factor over some coastal areas is zonal wind at 850 hPa under PTC.</u>

## **6 Summary**

Meteorology can influence surface ozone variability on different time scales, from 875 long-term trends to sub-daily scalevariations. Based on surface ozone observations-ineastern China during 2013-2018 from MEE, we characterized the seasonal and interannual variations of surface ozone in eastern China. The measurements show that surface ozone pollution in the study region is severest in summer and the severity goesis in a rapid increasing trend during the study period. We then focused on the 880 meteorological influence on the daily variability of summertime surface ozone in eastern China. We took daily anomalies of meteorological and ozone values to remove the variabilities on scales longer time scales than daily variations in these datasets. We estimated the local and synoptic meteorological impacts on daily variability of surface ozone using a MLR model and a SOM clustering technique. Synoptic The MLR is 885 driven by local meteorological variables and synoptic weather factors identified by the SVD analysis were combined with local meteorological variables to drive the MLR model. The regression model.

<u>The MLR</u> suggests that on regional average, meteorology can explain 4643%
 variations in the summertime daily surface ozone in eastern China, with an explained variance of up to 65% over some locations (Figure 5e5a). The regression model also shows that meteorology contributes to 3918% of the increasing trend in the regional

mean of summertime surface ozone over eastern China from 2013 to 2018. <u>Exploiting</u> the MLR, we also identified the key meteorological variables that are mostly

- 895 responsible for daily variations of summertime surface ozone in eastern China during 2013-2018. Among the local meteorological variables, relative humidity is the foremost over most areas in the center and south of eastern China including YRD and PRD, while temperature is the foremost in the north including BTH (Figure 5c).
- Exploiting the MLR model, we also identified the key meteorological variables that are mostly responsible for variations of summertime surface ozone in eastern China during 2013-2018. On regional average, the synoptic circulation factors constructed by the SVD analysis were estimated to contribute to 37% of the meteorological impact over eastern China and 41% over BTH, YRD, and PRD (Figure 6b). Among the local meteorological variables, relative humidity is the foremost over most-locations in the center and south of eastern China including YRD and PRD, while temperature is more important in the north including BTH (Figure 6c).
- \_\_We assessed the impacts of the dominant synoptic weather systems on surface
   ozone using cluster analysis. Employing the SOM, the summer synoptic circulations over eastern China during 2013-2018 were objectively classified into six predominant SWPs (Figures 8-137-12). The six SWPs control the variations of the key meteorological variables and thus impact the transport and production of ozone-\_
   regionally. Among the six SWPs, the SWP (PS) featured southerly wind, strong\_
- EASM and weak WPSH (Figure 8), and the SWP (PWC) featured a weak
   extratropical cyclone and the weakest WPSH (Figure 10) tend to increase the regional
   mean surface ozone in eastern China. In contrast, the other four SWPs (namely, PSW,
   PNECV, PSWPSH, and PTC) tend to reduce regional mean surface ozone in eastern
   China (Figures 7, 9, 11, and 12). As the predominant meteorological controlling
- variables of surface ozone vary greatly in space (Figures 2 and  $\frac{65}{5}$ ), strong differences

are found in surface ozone concentrations under every SWP between northern and southern parts of eastern China or between eastern and western parts of eastern China (Figures <u>8-14</u>). Daily<u>7-12</u>). Under the dominant SWP, daily mean surface ozone overin some regions inareas of eastern China can maximally increase or decrease maximally by 8 µg m<sup>-3</sup> or 16% impacted byof the dominant SWP.mean (Table 1).

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Among the six SWPs, the SWP (PS) featured southerly wind, strong EASM and weak WPSH (Figure 9a), and the SWP (PWC) featured a weak extratropical cyclone and the weakest WPSH (Figure 11a) tend to increase the regional mean surface ozone in eastern China (Figures 9c and 11c). For specific regions in eastern China, PS and PWC statistically enhance ozone in YRD and PRD. However, mean negative ozone anomalies are observed over some locations in PS and PWC, such as BTH in PWC.

In contrast, the other four SWPs (namely, PSW, PNECV, PSWPSH, and PTC) tendto reduce regional mean surface ozone in eastern China. PSW is a SWP featuredsoutheasterly wind and weak EASM (Figure 8a) and it leads to overall ozonereduction in the south of eastern China including YRD and PRD (Figure 8c). Wheneastern China is influenced by the Northeast Cold Vortex, the WPSH, and the Meiyufront (Figure 10a), PNECV tends to reduce ozone over YRD (Figure 10c). Whereas,
ozone likely increases over some subregions in eastern China in the four SWPs.
PSWPSH featured the strongest and the most extensive WPSH (Figure 12a) can
enhance ozone over YRD, although it may reduce regional mean ozone over eastern
China (Figure 12c). When the atmospheric circulation is controlled by the typhoon
systems with their centers around Taiwan, southeast to the mainland of China (Figure 13c).-

This study provides some new insights on the relationship between meteorology and air pollution, by untangling the complex response of surface ozone to different SWPs and local meteorological variables. The most significant meteorological

- 950 variables for surface ozone in eastern China were identified regionally, which was rarely investigated by previous studies (Gong et al., 2018; Zhan et al., 2018; Chen et al., 2019). Extending from previous studies, we quantified ozone anomalies in eastern China resulting from the prominent synoptic weather systems such as the WPSH (Shu et al., 2016; Zhao and Wang, 2017), the extratropical cyclones (Zhang et al., 2013;
- Liao et al., 2017), the Meiyu front, and typhoon (Jiang et al., 2015; Lam et al., 2018). These systems are important drivers for variations of air pollutants over eastern China (Ding et al., 2017). The relationship between weather and ozone is examined in one specific season, summer. Averagedmean ozone anomalies under a SWP in a relatively long term (six years over 2013-2018) was were used to represent the ozone sensitivity to that SWP. This method is also applicable for a full year, as it can remove the seasonal differences in the pollutant concentrations of pollution and the frequency of SWPs (Han et al., 2018b). No consideration of seasonal differences in ozonepollutant concentrations and meteorology can lead to biases in addressing daily variations of a pollutant (e.g. Zhang et al., 2013, 2016; Liao et al., 2017).

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In this study, the developed MLR and cluster techniques can well describe the meteorological impacts on the surface ozone variation in eastern China. Both regression and clustering analyses show strong performance, so they can be effective tools for air quality forecast. Many previous studies have reported the significance oflocal meteorology to the prediction of daily ozone in China (Zhao et al., 2016), however, few have included the meteorology at a synoptic scale. Here, we emphasized the synoptic role in the meteorological effects on surface ozone. The constructed synoptic factors by the SVD analysis can be a useful predictor for shortterm forecast of surface ozone. Regarding the time scale, this study focused on the day to day variations of surface ozone. Investigating the meteorological influences on a shorter time scale, such as diurnal variations, should be one of the directions for future work. The regression and clustering approaches can also be applied to project the potential effects of climate change on ozone variations in the future (Shen et al., 2017b). In the MLR regression analysis, we focused on the meteorological effects-

- 980 without direct consideration of variations in emissions, assuming emissions in a season are more or less constant. Here, we emphasize the importance of synoptic meteorology to the daily variations of surface ozone. The constructed synoptic factors by the SVD analysis can be a useful predictor for forecasting such daily variations. As ozone responses nonlinearly to variations in meteorology, emissions, and chemistry
- (Wu et al., 2009), the developed MLR model cannot fully describe the importance of meteorology to surface ozone predict daily ozone variations yet. Therefore, future-work is needed to address the the nonlinearity issue, need to be addressed in the future. Future work can also be conducted on the sensitivity of the diurnal ozone variation to meteorology and on the impact of climate change on future surface ozone
   levels regionally and globally (Shen et al., 2017b).

#### Data availability

Surface ozone measurements were obtained from the public website of MEE (http://beijingair.sinaapp.com/). The FNL meteorological data were acquired from

995 NCEP (https://rda.ucar.edu/datasets/ds083.2/). The OMI tropospheric column ozone monthly data were from NASA Goddard Space Flight Center (https://acdext.gsfc.nasa.gov/Data\_services/cloud\_slice/).

#### **Author contributions**

1000 H. Han designed the study and performed the research. H. Han and L. Shu analyzed the data and developed the model. H. Han and J. Liu wrote the manuscript with inputs from L. Shu, T. Wang, and H. Yuan.

## **Competing interests**

1005 The authors declare that they have no conflict of interest.

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