

Cover Letter

Dear Editor,

We are truly grateful to the reviewers' critical comments and comprehensive suggestions on our manuscript. Based on these comments and suggestions, we have made significant modifications on the original manuscript. **The whole manuscript has been carefully checked and a lot of paragraphs have been added or rewritten to better the structure. The sections of abstract, discussion, and conclusions are all rewritten. All changes made in the text are marked with red color in the revised manuscript.** The point-to-point responses to the reviewers' comments are listed as below. We hope the revised manuscript is now suitable for publication in *Atmospheric Chemistry and Physics*. Thank you for your consideration.

Best wishes

Sincerely yours

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Replies to reviewers' comments point by point

Referee #1

1. While this is an interesting case study, the motivation of the paper is not clear to me. Specifically, the authors did not say whether [a] they want to study how surface ozone responded to the tropical cyclone during their study period or [b] if they are interested in understanding whether emergency control measures put in place for the G20 meeting helped reduce ozone levels or not? If their objective is [a], this study lacks novelty because it is now well understood that clear-sky stagnant conditions favor photochemical ozone formation and cloudy-skies suppress it. Thus, ozone variations reported and modeled before, during, and after the cyclone are expected and there is no new knowledge gained here. If their objective is [b], the model experimental design is not appropriate. The authors did not modify their emission input to reflect emission control measures in their model simulations and no sensitivity experiment was performed to understand what would have happened in the absence of emergency emission control measures?

Reply: Thank you for this valuable comment. Accordingly, we have highlighted the motivation of the present study by rewriting the introduction with two additional paragraphs. The main objective of the present study is to understand the unique response of ozone increase to emission control measures during the 2016 G20 Summit in Hangzhou, while other pollutants had been significantly reduced (Li et al. 2019; Wu et al. 2019; Ji et al. 2018; Zheng et al. 2019). The title of the manuscript is also changed as “Spatial-temporal Variations and Process Analysis of O₃ Pollution in Hangzhou during the G20 Summit” to reflect this motivation. For this purpose, a regional air quality model, within the framework of the Model Inter-Comparison Study for ASIA phase III (Li et al., 2019), is used to investigate the spatial-temporal variations of ozone pollution in Hangzhou during the G20 Summit. Process analysis is conducted to understand the chemical and physical factors that contribute to the local O₃ abundance. It is worth noting that the base emission input has been modified to reflect emission control measures in our model simulations. Sensitivity experiments are not performed as

51 previous surface observations (Li et al. 2019; Wu et al. 2019; Ji et al. 2018; Zheng et al. 2019) have
52 suggested that the control measures took no immediate effect on local ozone formation, but
53 significantly reduced other pollutants. The added two paragraphs are attached as below.

54 “Hangzhou, the capital of Zhejiang Province, is located in the center of the Yangtze River Delta
55 which is one of the most developed areas in China. Resultant from local emissions (Wu et al. 2014,
56 Hu et al. 2015) and transboundary transport of aerosol and trace gases transport (Liu et al. 2015; Ni
57 et al. 2018; Zhang et al. 2018), air pollution in Hangzhou has become serious in the recent years. In
58 2016, Hangzhou city would host the 2016 G20 (Group of Twenty Finance Ministers and Central Bank
59 Governors) summit during September 4-6. To improve air quality for this event, 14-day temporarily
60 strict air pollution alleviation measures had been taken to reduce air pollutant emissions in Hangzhou
61 and surrounding areas from August 24 to September 6, 2016. The emission control scheme includes
62 a coal-fired power plant capacity 50% reduction since August 24, followed by an “odd-even” on-road
63 vehicle restriction since August 28, and further emergent VOC reduction from industrial sectors since
64 September 1 to 6 (Ji et al. 2018; Li et al. 2019; Wu et al. 2019). These short-term measures provide a
65 valuable opportunity to investigate the response of air quality to the emission reduction, understand
66 the formation mechanisms of air pollution, and explore effective policies for long-term air pollution
67 control in the local or regional scale.

68 The effects of emission control on air pollutants during this G20 Summit have been investigated
69 by several studies using field observations and numerical models. It is demonstrated that almost all
70 major air pollutants including SO₂, NO_x (Li et al. 2019; Wu et al. 2019), fine particles (Ji et al. 2018;
71 Li et al. 2019; Yu et al. 2018; Wu et al. 2019), and VOCs (Zheng et al. 2019) have been significantly
72 reduced during the 14-day control period, except O₃. Su et al. (2017) monitored the vertical profiles
73 of ozone concentration in the lower troposphere of Hangzhou during the control period by using an
74 ozone lidar. It was found that the ozone concentrations peaked near the top of the planetary boundary
75 layer, and the temporary measures took no immediate effect on ozone pollution. Wu et al. (2019)
76 investigated the variation of air pollution in Hangzhou and its surrounding areas during the G20

77 summit by using monitoring data from five sites, and reported that the air quality had been greatly
78 improved by the implementation of the emission control. However, the average O₃ concentration was
79 increased by 19% compared to the same periods of the five preceding years. This unique response of
80 ozone pollution to control measures is not well understood, and of great research interest for better
81 control of ozone pollution in the future.

82 To this end, a regional air quality model, within the framework of the Model Inter-Comparison
83 Study for ASIA phase III (Li et al., 2019), is used to investigate the spatial-temporal characteristics
84 of ozone pollution in Hangzhou during the G20 Summit in the present work. Process analysis is
85 conducted to understand the chemical and physical factors that contribute to O₃ abundance. It is found
86 that the serious ozone pollution happened, mainly resultant from the local photochemical reactions
87 which are not under good control by the emission reduction measures.”

88

89 2. There was no analysis of whether or not the observations at 96 sites violated the ozone standard
90 during the G20 meeting?

91 *Reply:* Following the reviewer’s suggestion, the observations at 96 sites are analyzed. Fig. 3c shows
92 that during the 14-day emission control period of G20 summit, 52% of the observed ozone samples
93 from the 96 sites are above the China’s national level-II standard (160µg/m³). This result confirms
94 that regional ozone pollution appears in the YRD region during the study period. Relevant statement
95 has been added into the revised manuscript (Line 309-312), and attached as below.

96 “This phenomenon is consistent with the satellite-derived tropospheric O₃ distribution in the area
97 (Su et al. 2017), and is also supported by the observed ozone data from the 96 sites in the YRD region
98 as shown in Fig. 3c. During the 14-day emission control period of G20 summit, 52% of the observed
99 ozone samples from the 96 sites are above the China’s national level-II standard (160µg/m³),
100 suggesting that regional ozone pollution appears in the YRD region during the study period.”

101

102 3. The choice of Hangzhou as the analysis site is also not clear. Authors say that they selected the site
103 based on evaluation but no evaluation metric was presented to justify their decision to focus on
104 Hangzhou. Why not use all the observations from 96 sites in your analysis to get a regional picture?

105 **Reply:** We have added two paragraphs (lines 84-122 see reply to the first comment) in the introduction
106 to indicate why Hangzhou is chosen as the focus. Basically, the main objective of the present study
107 is to understand the unique response of ozone increase to emission control measures while other
108 pollutants had been significantly reduced (Li et al. 2019; Wu et al. 2019; Ji et al. 2018; Zheng et al.
109 2019) during the 2016 G20 Summit which was held in Hangzhou. Observations from 96 sites are also
110 analyzed to give a regional picture (Fig. 3c), together with the model results, as attached below.

111 “This phenomenon is consistent with the satellite-derived tropospheric O₃ distribution in the area
112 (Su et al. 2017), and is also supported by the observed ozone data from the 96 sites in the YRD region
113 as shown in Fig. 3c. During the 14-day emission control period of G20 summit, 52% of the observed
114 ozone samples from the 96 sites are above the China’s national level-II standard (160µg/m³),
115 suggesting that regional ozone pollution appears in the YRD region during the study period.”

116
117 In addition to these major concerns, below are some other specific concerns that the
118 authors might find useful in their revision.

119 1) Section 2:3: Can you be a little more specific about the IPR here? Did you save the tendency
120 terms before and after the call to each process is made in the code? For example, did you save
121 ozone concentrations before and after the call the chemistry solver and used the difference in the
122 process analysis?

123 **Reply:** Yes, you are right. The IPR analysis is integrated into the WRF-Chem model and all the
124 tendency terms are saved before and after the call to each process. The difference is then used for
125 quantitative analysis of each process. For more details, please refer to the study of Jffries and
126 Tonnesen (Atmospheric Environment, 1994, 28(18): 2991-3003) and the user guide of WRF-Chem.

127 Following the reviewer's suggestion, we have added relevant description on the IPR analysis in lines
128 174-183. The description is also attached as below.

129 "To understand the underlying mechanism of O₃ formation, individual physical and chemical
130 processes of O₃ formation are investigated by using the integrated process rate (IPR) analysis in the
131 WRF-Chem model (Jffries and Tonnesen, 1994). The IPR analysis differentiates changes in pollutant
132 concentrations from individual atmospheric process which quantitatively elucidates the contributions
133 of each process, mainly including advection, diffusion, emission, deposition, clouds process, aerosol
134 and gaseous chemistry. The IPR analysis has been widely applied and demonstrated to be an effective
135 tool for investigating the relative importance of individual processes and interpreting O₃
136 concentrations (Goncalves et al., 2009; Tang et al., 2017; Shu et al., 2016). In the present work, we
137 consider gas chemistry, vertical diffusion, horizontal and vertical advections as the main atmospheric
138 processes for O₃ formation. Other processes, such as cloud process and horizontal diffusion, play
139 minor roles and are thus not considered."

140

141 2) Table S1: For some reason, the equations did not appear correctly in the Table. Please correct.

142 **Reply:** Revised as suggested.

143

144 3) Section 2.4: Are the observations from air quality monitoring network quality controlled or did
145 you apply any quality control procedure to the measurements before using those for evaluation?

146 **Reply:** The quality of all the observations from air quality monitoring network has been controlled by
147 the data provider.

148

149 4) Line 255: Change "supply raw material" to "transport ozone precursors"

150 **Reply:** Revised as suggested.

151

152 5) Figure 8 shows that horizontal advection contributes much larger to the ozone increase on most

153 of the days but in the abstract the authors say “vertical diffusion and chemical production” are the
154 main drivers. I did not understand how the authors concluded this in the abstract.

155 *Reply:* Sorry for the confusion. Although horizontal advection contributes much larger to the ozone
156 increase on most of the days, the contribution of vertical advection is also larger. The effects of these
157 two processes have been cancelled out during several circulations. As a result, photochemical
158 production and vertical diffusion from the upper-air background ozone are the main drivers for the
159 local ozone. To be clear, we have modified the relevant statements in the abstract and conclusions, as
160 attached below.

161 “Interesting horizontal and vertical advection circulations of O₃ are observed during several short
162 periods, and the effects of these processes are nearly cancelled out. As a result, the ozone pollution is
163 mainly attributed to the local photochemical reactions which are not obviously influenced by the
164 emission reduction measures. The ratio of reduction of Volatile Organic Compounds (VOCs) to that
165 of NO_x is a critical parameter that needs to be carefully considered for future alleviation of ozone
166 formation. In addition, the vertical diffusion from the upper-air background O₃ also plays an important
167 role in shaping the surface ozone concentration.”

168 “Horizontal and vertical advection circulations are captured in Hangzhou, with horizontal
169 advection the source and vertical advection the sink of the surface O₃ in Hangzhou. Consequently,
170 the serious ozone pollution is mainly resultant from the local photochemical reactions which are not
171 under good control by the emission reduction measures. As the surface O₃ formation in Hangzhou is
172 dominant by the VOCs-limited regime, the significant reduction of NO_x compared to that of VOCs is
173 unfavorable to chemical generation of O₃. The ratio of reduction of VOCs to that of NO_x based on the
174 O₃-NO_x-VOCs sensitivity analysis is a critical parameter for reduction of ozone formation from
175 photochemical reactions. In addition, it is found that the vertical diffusion from the upper-air notable
176 background O₃ also plays an important role in shaping the surface ozone concentration when the
177 photochemical reactions are weak. ”

178

179 **Referee #2**

180 1. The authors mentioned emergency emission control measures. Were emissions perturbed to
181 represent these measures? How did emission control measures contribute to the ozone episode?

182 *Reply:* Yes, emissions are perturbed to represent these measures. We have added two paragraphs
183 (please refer to the reply to the first comment of Referee #1) to introduce the background of
184 emergency emission control measures and the effects on pollutant emissions during the G20 summit.
185 Previous studies have demonstrated that almost all major air pollutants including SO₂, NO_x (Li et al.
186 2019; Wu et al. 2019), fine particles (Ji et al. 2018; Li et al. 2019; Yu et al. 2018; Wu et al. 2019),
187 and VOCs (Zheng et al. 2019) have been significantly reduced during the 14-day control period,
188 except O₃. It was found that the temporary measures took no immediate effect on ozone pollution (Su
189 et al. 2017), or even the average O₃ concentration was increased by 19% compared to the same periods
190 of the five preceding years (Wu et al. 2019). This unique response of ozone pollution to control
191 measures is not well understood, and of great research interest for better control of ozone pollution in
192 the future, which motivates the present work. To obtain the quantitative effect of emission control
193 measures on the ozone episode, scenario simulations and sensitivity analysis are required, which is
194 beyond the scope of the current work. However, the modification of the emission inventory to reflect
195 the control measures has been emphasized in the revised manuscript (line 169-171), as attached below.

196 “However, it is worth noting that these base inventories have been modified in the simulation to
197 reflect the realistic emissions according to the control measures taken in the period presented in the
198 introduction.”

199
200 2. The authors claimed that this study revealed notable background O₃ concentrations, but it is very
201 confusing how this conclusion was drawn. How much does it contribute to O₃ levels in the YRD?

202 *Reply:* Thank you for pointing out this issue. The background O₃ means the O₃ that vertically
203 distributes within the planetary boundary layer. High ozone concentrations, temporarily during most
204 day time of the emission control period and spatially from the surface to the top of the planetary

205 boundary layer, are observed in Hangzhou and even the whole YRD region. This can be seen from
206 Figs. 5, 7, and 8 in the revised manuscript. The background O₃ essentially influences the surface O₃
207 concentration through vertical diffusion. Its quantitative contribution to the surface O₃ level in
208 Hangzhou is different from day to day, as demonstrated in Figs 8 and 9.

209

210 3. It is not convincing that current categorization of process analysis can provide any useful
211 information. Concluding photochemistry dominated O₃ generation does not provide any indications
212 for O₃ pollution control. Which precursor or process are important? More in-depth analyses are
213 needed.

214 **Reply:** The IPR analysis differentiates changes in pollutant concentrations from individual
215 atmospheric process which quantitatively elucidates the contributions of each process, mainly
216 including advection, diffusion, emission, deposition, clouds process, aerosol and gaseous chemistry.
217 It has been widely applied and demonstrated to be an effective tool for investigating the relative
218 importance of individual processes and interpreting O₃ concentrations (Goncalves et al., 2009; Tang
219 et al., 2017; Shu et al., 2016). In the present work, to understand the underlying mechanism of O₃
220 formation, individual physical and chemical processes of O₃ formation are investigated by using the
221 IPR. The gas chemistry, vertical diffusion, horizontal and vertical advectons are considered as the
222 main atmospheric processes for O₃ formation. Other processes, such as cloud process and horizontal
223 diffusion, play minor roles and are thus not considered.

224 Through the IPR analysis, interesting horizontal and vertical advection circulations of O₃ are
225 observed during several short periods, and the effects of these processes are nearly cancelled out. As
226 a result, the ozone pollution is mainly attributed to the local photochemical reactions which are not
227 obviously influenced by the emission reduction measures. In addition, the vertical diffusion from the
228 upper-air background O₃ also plays an important role in shaping the surface ozone concentration.

229 Following the reviewer's suggestion, the discussion section has been rewritten and some more
230 in-depth discussions on the precursors of ozone formation have been added into the revised
231 manuscript, as attached below.

232 "Chemical generation of O₃ is the net effect of photochemical generation and titration
233 consumption. VOC oxidation (Jenkin et al., 1997; Sillman, 1999) in photochemical reactions provides
234 critical oxidants (i.e., RO₂) that efficiently convert NO to NO₂, resulting in further accumulation of
235 O₃ (Wang et al., 2017). The chemical generation of O₃ is controlled by NO_x and VOCs depending on
236 which substance is lack in the reactions. As a consequence, there are two sensitivity regimes of O₃
237 production, namely, the NO_x-limited and VOC-limited regimes. Previous studies have shown that the
238 sensitivity pattern of surface O₃ formation in Hangzhou is dominant by the VOCs-limited regime
239 (Yan et al. 2016; Li et al., 2017; Su et al., 2017). In this regime, if the regional reduction of VOCs is
240 much higher than that of NO_x, the O₃ concentration can be reduced, otherwise if the regional reduction
241 of VOCs is much less than that of NO_x, the inhibitory effect of NO_x on O₃ generation will be
242 weakened, and the O₃ concentration will increase remarkably (Wang et al. 2015). According to the
243 studies of Su et al. (2017), Zheng et al. (2019), and Wu et al. (2019), it can be deduced that NO_x has
244 been significantly reduced by about 60%, at least two times of the reduction of VOCs in Hangzhou.
245 The influence of stringent emission control measures on VOCs is not as immediate and effect as that
246 on NO_x, which is associated with the fact that there was a large amount of biogenic VOC emission in
247 Hangzhou and surrounding regions (Liu et al. 2018; Wu et al. 2020). In fact, the average temperature
248 during the study period is as high as around 31°C (Fig. 4c), which facilitates the biogenic VOC
249 emissions and photochemical reactions. As a result, the photochemical generation of O₃ was not under
250 control and high concentration of ozone appeared. However, it is worth noting that after the emergent
251 VOCs control measures had been implemented in the area during the third stage, the net generation
252 rate of O₃ gradually reduces since September 2, 2016, leading to a period of relatively low ozone
253 concentration together with other meteorological effects. These discussions implicate that to alleviate
254 ozone pollution, the ratio of reduction of VOCs to that of NO_x is the key parameter based on the O₃-

255 NO_x-VOCs sensitivity analysis. As the biogenic VOCs are important sources of the total VOCs in the
256 YRD region, it is necessary to balance the reduction of NO_x to make the ratio within effective regime
257 in the future.”

258

259 Minor comments:

260 1) Fig. 1a does not show domain 1.

261 **Reply:** Domain 1 has been marked in Fig. 1a.

262

263 2) Line 119: it is confusing if assimilation of meteorological variables were used or not, how

264 **Reply:** Assimilation of meteorological variables are not used in this study. To avoid confusion,

265 “assimilated” has been corrected as “mapped” in Line 145.

266

267 3) Line 143: In June, July, and August, biomass burning emissions are important in east China, why
268 do you ignore it?

269 **Reply:** Biomass burning emissions have already been included in the emission inventory we used

270 (2016 Multiresolution Emission Inventory for China (MEIC, 0.25° × 0.25°;

271 <http://www.meicmodel.org/>). Thus, their effect has been considered.

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Elucidating the ozone pollution **Spatial-temporal Variations and**
Process Analysis of O₃ Pollution in Yangtze River Delta-
region Hangzhou during the 2016 G20 summit for MICS-Asia-

III Summit

By

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Atmospheric Chemistry and Physics

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336 formation ~~and dispersion during tropical cyclone events~~ in Hangzhou, and support the Model
337 Intercomparison Study Asia Phase III (MICS-Asia Phase III),
338 **Keywords:** ~~Ozone, Tropical cyclone pollution,~~ WRF-Chem, ~~Spatial-temporal variation,~~ Process
339 analysis, Air quality.

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1. Introduction

Tropospheric ozone (O₃) is generated by a series of photochemical reactions involving volatile organic compounds (VOCs), nitrogen oxide (NO_x), and carbon monoxide (CO) (Wang et al., 2006).

As a primary component of photochemical smog, ground-level O₃ pollution exhibits detrimental effects on human health (Ha et al., 2014; Kheirbek et al., 2013) and the ecosystem (Landry et al., 2013; Teixeira et al., 2011). The contribution of outdoor air pollution sources to premature mortality may increase globally in the coming decades (Lelieveld et al., 2015). However, O₃ pollution is a challenging problem worldwide. O₃ levels in cities in the United States and Europe are increasing more than those in the rural areas of these regions, where peak values gradually decreased during 1990–2010 (Paoletti et al., 2014). Nagashima et al., (2017) reported that long-term (1980–2005) trends of increase in surface O₃ over Japan may be primarily attributed to the continental transport that have contributed to photochemical O₃ production. Urban O₃ pollution events can also be observed in developing countries, such as Thailand (Zhang and Kim Oanh, 2002) and India (Calfapietra et al., 2016).

Air quality has been deteriorating in China as urbanization and motorization have progressed. Many field monitoring and modeling studies have investigated the photochemical characteristics of near-surface O₃ pollution (Tang et al., 2009, 2012; Wang et al., 2013, 2014), the photochemistry of O₃ and its precursors (Xie et al., 2014), the interactions of between O₃ with and PM_{2.5} (Shi et al., 2015), and the urban O₃ formation (Tie et al., 2013). It is clear that in addition to anthropogenic emissions of O₃ precursors, uncontrollable physical and chemical processes involved in meteorological phenomena critically significantly modulate changes in O₃ concentration (Xue et al., 2014). In the Yangtze River Delta (YRD) region of China, high O₃ concentrations are associated with pollutant transport and diffusion from surrounding areas have been observed (Gao et al., 2016; Jiang et al., 2012). Synoptic patterns related to tropical cyclones may be conducive to one reason for such high O₃ concentrations (Huang et al., 2005). Jiang et al. (2015) reported that enhanced stratosphere–troposphere exchange (STE) driven by a tropical cyclone abruptly increased O₃ concentrations (21–

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389 42 ppb) in the southeast of China during June 12–14, 2014. ~~STE, which~~ has been highlighted as a
390 ~~significant~~ ~~another~~ contributor to near-surface O₃ concentrations ~~under certain conditions~~ (Lin et al.,
391 2012, 2015). ~~Because relevant data are limited~~ ~~However,~~ the complex dynamics in atmospheric
392 processes related to O₃ formation are ~~so~~ difficult to ~~evaluate, and the main processes that account for~~
393 ~~high O₃ concentrations are challenging to~~ identify; ~~that the~~ O₃ pollution characteristics and underlying
394 causes have not ~~been sufficiently investigated in China, especially in relation to extreme~~
395 ~~meteorological conditions. The lack of relevant data may influence urban pollution prevention~~
396 ~~efforts~~ ~~yet been well understood.~~

397 Hangzhou, the capital of Zhejiang Province, is located in the center of the Yangtze River Delta
398 which is one of the most developed areas in China. Resultant from local emissions (Wu et al. 2014,
399 Hu et al. 2015) and transboundary transport of aerosol and trace gases transport (Liu et al. 2015; Ni
400 et al. 2018; Zhang et al. 2018), air pollution in Hangzhou has become serious in the recent years. In
401 2016, Hangzhou city would host the 2016 G20 (Group of Twenty Finance Ministers and Central Bank
402 Governors) summit during September 4-6. To improve air quality for this event, 14-day temporarily
403 strict air pollution alleviation measures had been taken to reduce air pollutant emissions in Hangzhou
404 and surrounding areas from August 24 to September 6, 2016. The emission control scheme includes
405 a coal-fired power plant capacity 50% reduction since August 24, followed by an “odd-even” on-road
406 vehicle restriction since August 28, and further emergent VOC reduction from industrial sectors since
407 September 1 to 6 (Ji et al. 2018; Li et al. 2019; Wu et al., 2019). ~~— In this study~~ ~~These short-term~~
408 ~~measures provide a valuable opportunity to investigate the response of air quality to the emission~~
409 ~~reduction, understand the formation mechanisms of air pollution, and explore effective policies for~~
410 ~~long-term air pollution control in the local or regional scale.~~

411 The effects of emission control on air pollutants during this G20 Summit have been investigated
412 by several studies using field observations and numerical models. It is demonstrated that almost all
413 major air pollutants including SO₂, NO_x (Li et al. 2019; Wu et al. 2019), fine particles (Ji et al. 2018;
414 Li et al. 2019; Yu et al. 2018; Wu et al. 2019), and VOCs (Zheng et al. 2019) have been significantly

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415 reduced during the 14-day control period, except O₃. Su et al. (2017) monitored the vertical profiles
416 of ozone concentration in the lower troposphere of Hangzhou during the control period by using an
417 ozone lidar. It was found that the ozone concentrations peaked near the top of the planetary boundary
418 layer, and the temporary measures took no immediate effect on ozone pollution. Wu et al. (2019)
419 investigated the variation of air pollution in Hangzhou and its surrounding areas during the G20
420 summit by using monitoring data from five sites, and reported that the air quality had been greatly
421 improved by the implementation of the emission control. However, the average O₃ concentration was
422 increased by 19% compared to the same periods of the five preceding years. This unique response of
423 ozone pollution to control measures is not well understood, and of great research interest for better
424 control of ozone pollution in the future.

425 To this end, a regional air quality model, within the framework of the Model Inter-Comparison
426 Study for ASIA phase III (MICS-ASIA III) (Li et al., 2019), was used to elucidate the
427 spatial-temporal characteristics of ozone pollution in Hangzhou during the G20 Summit in the present
428 work. Process analysis is conducted to understand the chemical and physical factors that
429 contributed to O₃ abundance during the G20 (Group of Twenty) summit. The summit was
430 held in Hangzhou, China, and the focus of the summit was. It is found that the sustainable and healthy
431 development of serious ozone pollution happened, mainly resultant from the world economy.
432 Emergency local photochemical reactions which are not under good control by the emission
433 control reduction measures (e.g., industrial stoppages, limitations of vehicle movement) were
434 implemented over an area with a diameter of approximately 600 km to improve the air quality from
435 24 August to 06 September 2016. Because of severe concerns regarding O₃ concentrations and the
436 summer cyclonic weather pattern, the aforementioned pollution control event attracted wide policy-
437 related interest. The rest of this paper is organized as follows. Section 2 outlines the methodology and
438 configuration of the model system. Section 3 describes the synoptic weather conditions as
439 well as individual O₃ formation model evaluation, the spatial-temporal characteristics of ozone

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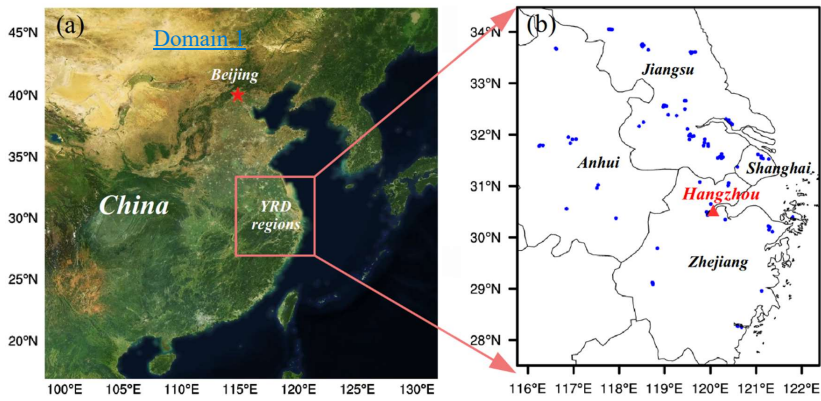
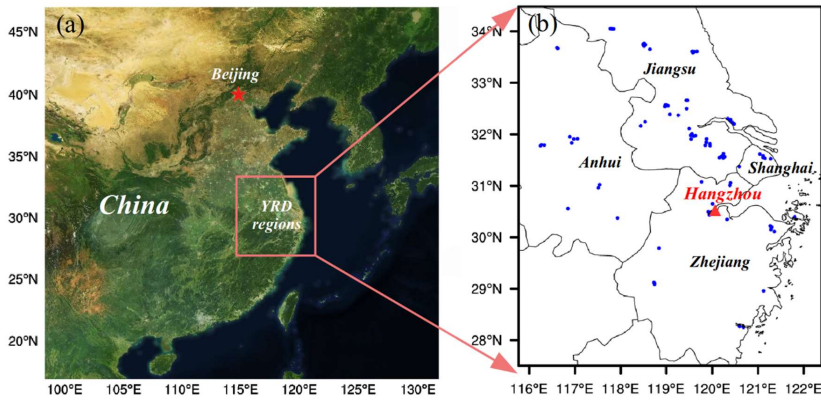


Fig. 1. Double-nested simulation domains. (a) Domain 1: 30 km in East China with 102 (W–E) × 111 (S–N) × 31 (vertical layers) grids; (b) Domain 2: 6 km in the Yangtze River Delta (YRD) region with 100 (W–E) × 115 (S–N) × 31 (vertical layers) grids. Blue dots denote the air quality monitoring sites. The copyright of the background map belongs to Google Map © Microsoft.

The meteorological boundary and initial conditions were determined from the global objective final analysis (FNL) data of the National Centers for Environmental Prediction (Kalnay et al., 1996). The FNL data were also assimilated and mapped to domain 1 (East China), and the grid-nudging method (Stauffer et al., 1991) was used to reduce the meteorological integral errors. The chemical initial and boundary conditions were dynamically downscaled from the simulation results of model for ozone and related chemical tracers, version 4 (MOZART4) (Emmons et al., 2010)

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472 simulation results; the relevant data are available at <https://www.aocom.ucar.edu/wrf-chem/mozart.shtml>.

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474 2.2. Emissions

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475 The 2016 Multiresolution Emission Inventory for China (MEIC, $0.25^\circ \times 0.25^\circ$;
476 <http://www.meicmodel.org/>) was used for the outer domain (Fig. 1a)
477 with a spatial resolution of 30 km (Li et al., 2017), including species of SO₂, NO_x, CO, NH₃, PM_{2.5},
478 and VOCs from the power, industrial, residential, transportation, and agricultural sectors. Inventories
479 of finer anthropogenic emissions for the YRD region (Fig. 1b) over the year of 2014 were compiled
480 based on the bottom-up method by Shanghai Academy of Environmental Sciences. are used for the
481 inner domain (Fig. 1b). These inventories have been well documented in detail in previous studies
482 (Huang et al., 2011; Li et al., 2011; Liu et al., 2018). Thus, only brief discussions of these inventories
483 are presented herein. The fine emission inventories include major sectors such as large point sources,
484 industrial sources, mobile sources, and residential sources. The anthropogenic emissions over the
485 YRD region are mainly located over the industrial and urban areas along the Yangtze River as well
486 as over Hangzhou Bay. In this study, the emission inventories for the two domains were projected
487 into horizontal and vertical grids as hourly emissions, with temporal and vertical profiles obtained
488 from Wang et al. (2011). VOCs emissions were categorized into modeled species, according to
489 von Schneidemesser et al. (2016). In addition, biogenic emissions were generated offline using the
490 Model of Emission of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006). Dust
491 emissions were calculated online from surface features and meteorological fields by using the Air
492 Force Weather Agency and Atmospheric and Environmental Research scheme (Jones et al., 2011).
493 Other emissions (i.e., such as those from biomass burning, aviation, and sailing ships), accounting
494 for very small fractions during this period, were therefore not considered here. However,
495 it is worth noting that these base inventories have been modified in this study the simulation to reflect
496 the realistic emissions according to the control measures taken in the period presented in the
497 introduction.

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2.3. Atmospheric processes analysis

To understand the ~~mechanism~~ underlying ~~mechanism of~~ O₃ formation, individual physical and chemical processes of O₃ formation ~~were~~ investigated by using the integrated process rate (IPR) analysis in the WRF-Chem model. ~~(Jffries and Tonnesen, 1994). The IPR analysis differentiates changes in pollutant concentrations from individual atmospheric process which quantitatively elucidates the contributions of each process, mainly including advection, diffusion, emission, deposition, clouds process, aerosol and gaseous chemistry. The IPR analysis has been widely applied; this method has been proven and demonstrated to be an effective tool for demonstrating investigating the relative importance of individual processes and for interpreting O₃ concentrations (Goncalves et al., 2009; Tang et al., 2017; Shu et al., 2016). The~~In the present study investigated atmospheric processes involved in O₃ formation, including work, we consider gas chemistry, vertical diffusion, and horizontal and vertical advection. ~~advections as the main atmospheric processes for O₃ formation, Other processes (i.e., such as cloud processes and horizontal diffusion) that either, play minor roles or result in the formation of a sink (i.e., dry and wet deposition) were and are thus not considered in this study.~~

2.4. Evaluation ~~method~~ metrics

To increase the confidence in interpretations of model results, model outputs should first be evaluated based on observations. Accordingly, ~~in this study,~~ the model results derived from domain 2 ~~were~~ compared with hourly surface observational data obtained from 96 air quality monitoring sites in the YRD region (blue dots, Fig. 1b). ~~These data were downloaded from <http://www.pm25.in>. The air pollutants assessed were O₃ and NO₂. Model performance was evaluated using statistical measures, namely mean fractional bias (MFB), mean fractional error (MFE), and correlation coefficient (*R*), following the recommendation of the US Environmental Protection Agency (US EPA; 2007). The formula used in this evaluation is presented in Table S1. Additionally, the meteorological parameters were evaluated based on observational data including temperature at 2 m (*T*₂), relative humidity at 2 m (*RH*₂), and 10 m wind speed (*WS*₁₀) and direction (*WD*₁₀) from the~~

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524 Meteorological Assimilation Data Ingest System (b) in this study. These observational data are
 525 downloaded from <http://www.pm25.in>, and O₃ as well as its precursor NO₂ are evaluated, in terms of
 526 statistical measures, namely the mean fractional bias (MFB), the mean fractional error (MFE), and
 527 the correlation coefficient (*R*), following the recommendation of the US Environmental Protection
 528 Agency (US EPA, 2007). Additionally, the meteorological parameters are evaluated based on the
 529 observational data, including temperature at 2 m (T2), relative humidity at 2 m (RH2), 10 m wind
 530 speed (WS10) and direction (WD10), from the Meteorological Assimilation Data Ingest System.

531 **Table 1.** Discrete statistical indicators used in the model evaluation

Metrics	Definition	Range
Mean Fractional Bias (MFB)	$MFB = \frac{2}{N} \sum_{i=1}^N \frac{S_i - O_i}{S_i + O_i} \times 100\%$	-200% to 200%
Mean Fractional Error (MFE)	$MFE = \frac{2}{N} \sum_{i=1}^N \frac{ S_i - O_i }{S_i + O_i} \times 100\%$	0 to 200%
Correlation Coefficient (<i>r</i>)	$r = \frac{\sum_{i=1}^N (S_i - \bar{S})(O_i - \bar{O})}{\sqrt{\sum_{i=1}^N (S_i - \bar{S})^2 \sum_{i=1}^N (O_i - \bar{O})^2}}$	0 to 1
Mean Bias (MB)	$MB = \frac{1}{N} \sum_{i=1}^N (S_i - O_i)$	$-\infty$ to $+\infty$
Gross Error (GE)	$GE = \frac{1}{N} \sum_{i=1}^N S_i - O_i $	0 to $+\infty$
Root Mean Square Error (RMSE)	$RMSE = \sqrt{\frac{1}{N} \sum_{i=1}^N (S_i - O_i)^2}$	0 to $+\infty$

N is the number of samples. *S_i* and *O_i* are values of simulations and observations at time or location *i*, respectively.

532
 533 (<https://madis.noaa.gov>). Following the study of Zhang et al. (2014), commonly used mean bias (MB),
 534 gross error (GE), and root mean square error (RMSE) were calculated as the statistical indicators;
 535 corresponding equations are denoted. All used statistical indicators are summarized in Table S1.
 536 Besides the above evaluation of single-point based time series results, the vertical spatial
 537 distribution of modeled O₃ in Hangzhou was also evaluated based on comparisons with observed
 538 differential absorption LiDAR (DIAL) data (Su et al. 2017). In the DIAL technique, the mean gas
 539 concentration over a certain range interval is determined by analyzing the LiDAR backscatter signals

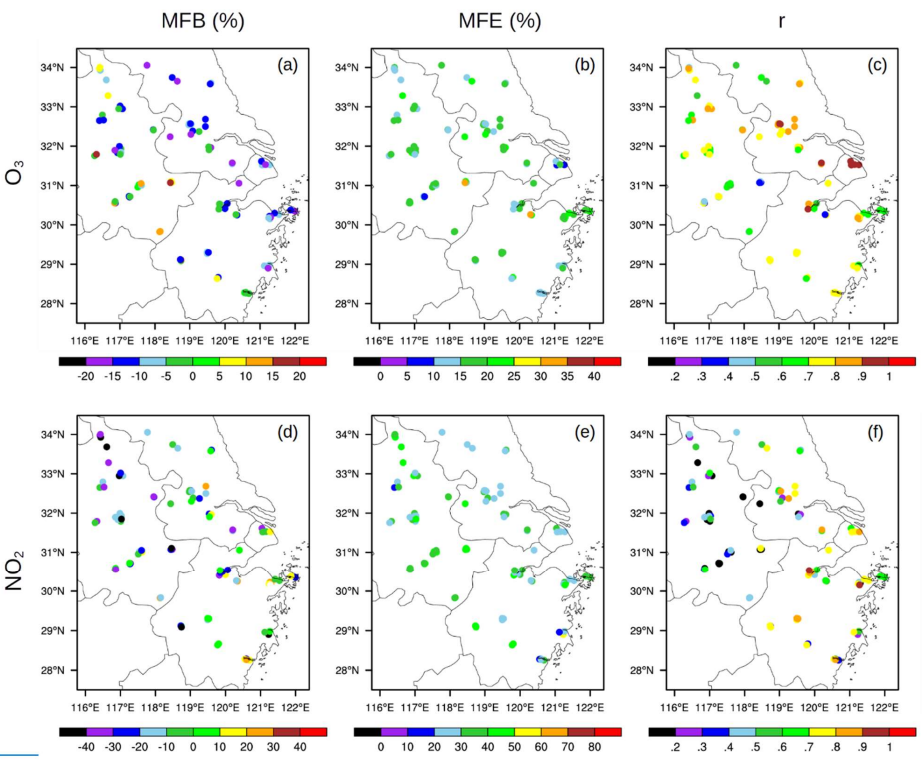
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540 for laser wavelengths tuned “on” (λ_{on}) and “off” (λ_{off}) in a molecular absorption peak of the gas under
 541 ~~investigation (Browell et al., 1998). The DIAL technique can be used to measure O₂ concentrations~~
 542 ~~above or near a specific location (Browell, 1989). In our DIAL datasets, the vertical height available~~
 543 ~~was from 0.3 km to 3 km due to the limitations of the signal-to-noise ratio and detection range.~~

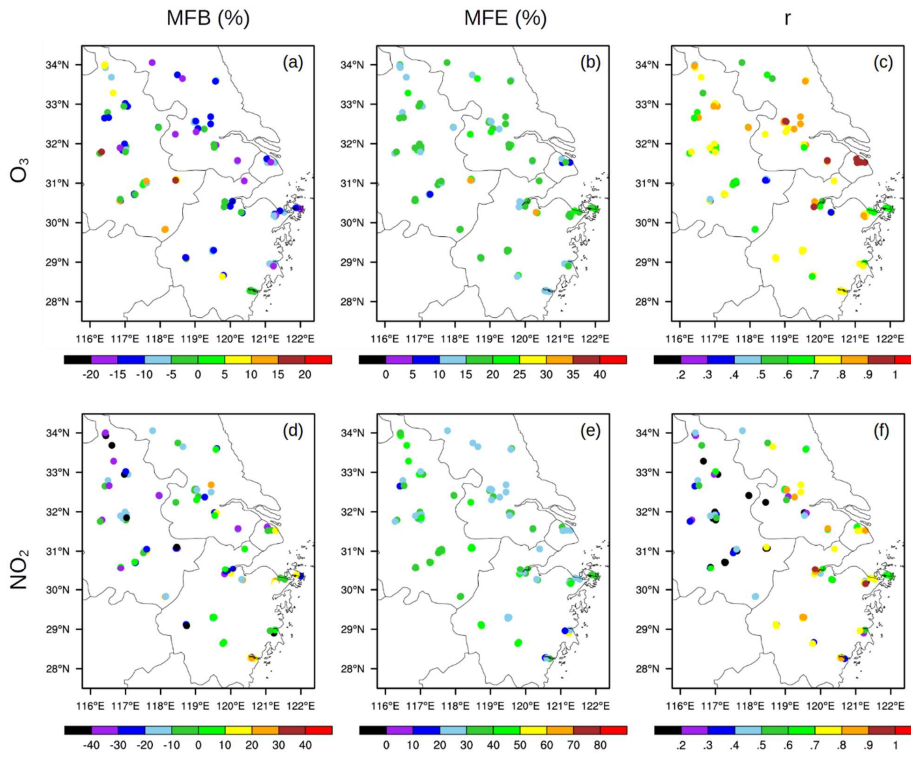
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544 **3. Results**

545 **3.1. Model performance evaluation**



546



547
548 **Fig. 2.** Comparison of modeled air pollutant concentrations against measurements in 96 monitoring sites
549 over YRD region during August 24–September 6, 2016: Mean fractional bias (MFB), mean fractional error (MFE)
550 and Pearson's correlation coefficient (r) of O_3 (a–c) and NO_2 (d–f), respectively.

551 [investigation \(Browell et al., 1998\). In the DIAL data of \$O_3\$, the vertical height available is from 0.3](#)
552 [km to 3 km due to the limitations of the signal-to-noise ratio and detection range.](#)

553 [3. Results](#)

554 [3.1. Model performance](#)

555 We first [evaluated](#) the overall performance of WRF-Chem for the YRD region by
556 incorporating data from the 96 air quality monitoring sites. Specifically, the maximum daily 8 h (Θ_3 -
557 $8hMDA_8$) ozone and daily mean NO_2 concentrations at the surface [were](#) used. [The spatial](#)
558 [distributions of MFB and MFE for \$O_3\$ and \$NO_2\$ are illustrated in Fig. 2. In general, the model-](#)

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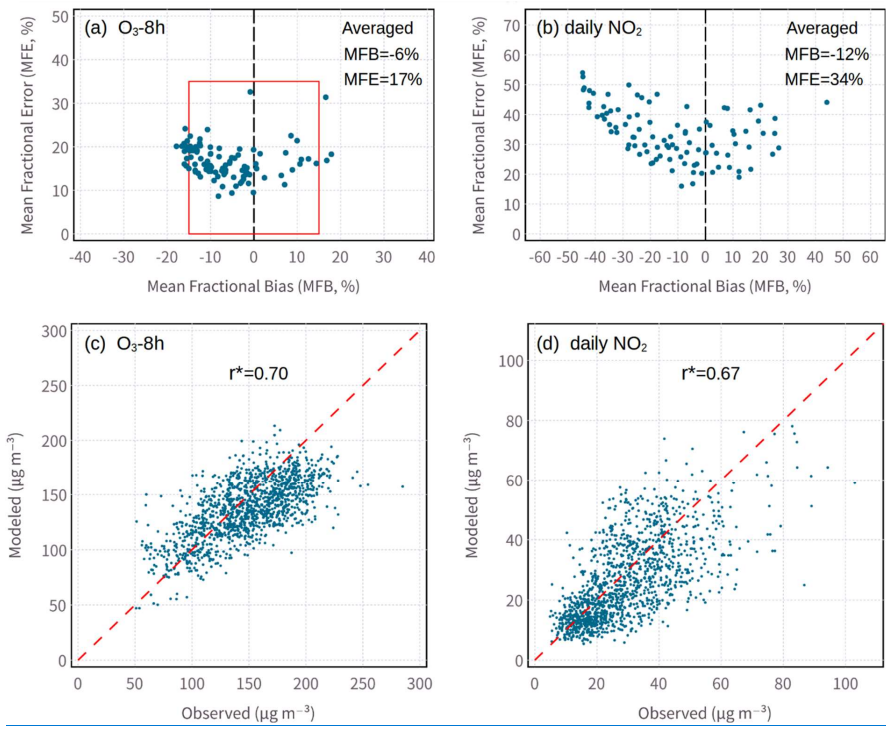
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559 simulated air pollutant concentrations agreed well with the observations. The spatial
 560 distributions of MFB and MFE for O₃ and NO₂ at the 96 observational sites over the YRD region are
 561 illustrated in Fig. 2. The results reflected reasonable performance, with MFB and MFE for most of
 562 the sites meeting

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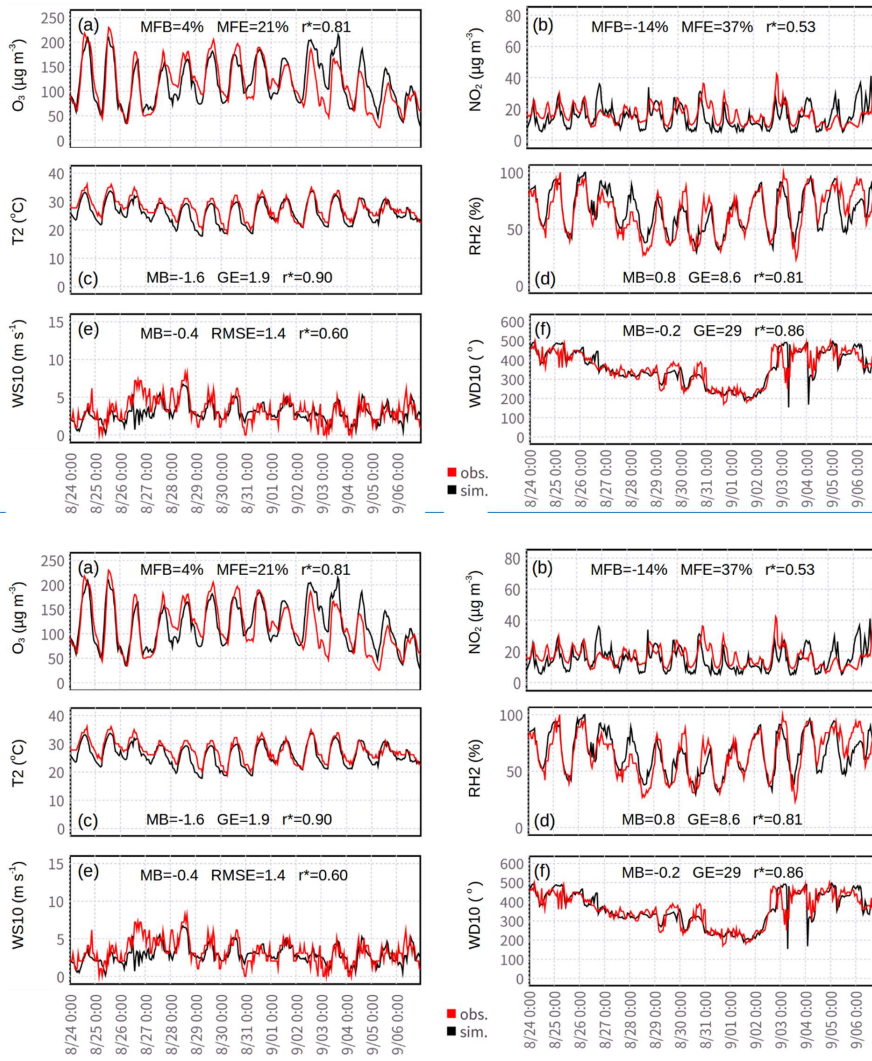


563
 564 **Fig. 3.** Comparisons of modeled and observed concentrations of the air pollutants from 96 air quality monitoring
 565 sites across the YRD from 24 August to 06 September 2016 (1,344 pairs). Scatter plots for MFB and MFE of (a) O₃
 566 and (b) NO₂. Performance goals (red box) for O₃ are the benchmarks. Scatter plots for daily observed and modeled
 567 (c) O₃ and (d) NO₂.
 568 the benchmarks (MFB: $\pm 15\%$; MFE: $\pm 35\%$) proposed by the US Environmental Protection Agency
 569 (US EPA, 2007). A scatter plot of MFB/MFE was shown in Fig. S1a,b in the supporting
 570 information, further hinting demonstrating the ability capability of the present model in reproducing
 571 the observations, which is also reflected supported by the high correlation between model and
 572 observations (observation (Fig. 3c, d), Fig. S1c,d in the supporting information). Other model

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573 evaluations with satellite retrievals during this period can be seen in our previous study (Ni et al.,
 574 2019).

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577 **Fig. 3-4.** Modeled air pollutants and meteorological parameters compared with measurements at the Hangzhou
 578 monitoring site from 24 August to 06 September 2016. Surface concentrations of (a) O_3 and (b) NO_2 , (c) temperature
 579 at 2 m (T2), (d) relative humidity at 2 m (RH2), (e) wind speed at 10 m (WS10) and (f) wind direction at 10 m
 580 (WD10).

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581 Following After the above overall evaluation of the greater present model in the whole YRD
 582 region, the site of Hangzhou was will be focused on for further analysis, and WRF-Chem simulations
 583 of the site's air quality and meteorological conditions were assessed. The time series of hourly
 584 simulated and observed air pollutants (O₃, Fig. 3a4a; NO₂, Fig. 3b4b) and meteorological factors (T2,
 585 Fig. 3c4c; RH2, Fig. 3d4d; WS10, Fig. 3e4e; and WD10, Fig. 3f4f) at Hangzhou are presented in
 586 Fig. 3-AH4. It is found that all modeled data were are statistically significantly correlated with the
 587 observed data at the 95% level. Overall, WRF-Chem well represented the observed diurnal variations.
 588 For example, the The MFB and MFE for both O₃ and NO₂ were near the benchmarks in particular of
 589 O₃ levels (MFB/MFE: 4%/21%), and were are well below the benchmarks (MFB/MFE: 15%/35%;
 590 US EPA, 2007).
 591 For evaluation of-) and the observed diurnal variations are well reproduced. For meteorological
 592 parameters, Emery et al. (2001) proposed benchmarks, including 2 m air temperature (MB $\leq \pm 0.5^\circ$
 593 C, GE $\leq 2.0^\circ$ C), 10 m wind speed (MB ≤ 0.5 m/s, RMSE ≤ 2.0 m/s) and 10 m wind direction
 594 (MB $\leq \pm 10^\circ$, GE $\leq 30^\circ$). McNally (2009) suggested a relaxed benchmark for 2 m temperature
 595 (MB $\leq \pm 1.0^\circ$ C). In this study, the 10 m wind speed and wind direction (Fig. 3e, f) results were are
 596 well within the proposed limits benchmarks. The GE of 2 m air temperature (1.9°C; Fig. 3c) also
 597 satisfied satisfies the criteria; however, but the MB was is slightly higher (-1.6°C), and a slightly high
 598 temperature bias was) which has also been noted in a previous study (Zhang et al., 2014). Overall,
 599 favorable performance was noted for These comparisons further demonstrate that the present model
 600 is able to correctly predict the simulation time series of both meteorological parameters and air
 601 pollutants of O₃ and NO₂ in comparison with observations Hangzhou.

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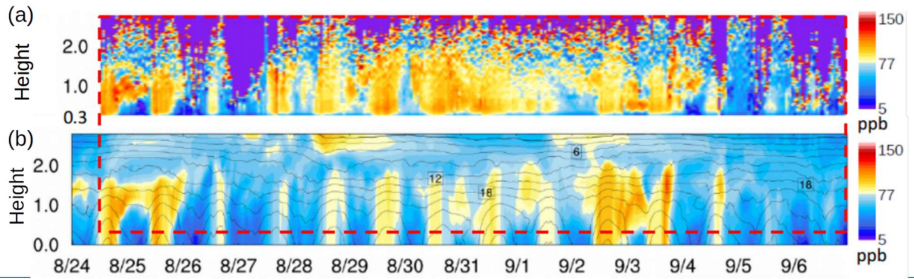


Fig. 4. Upper-level

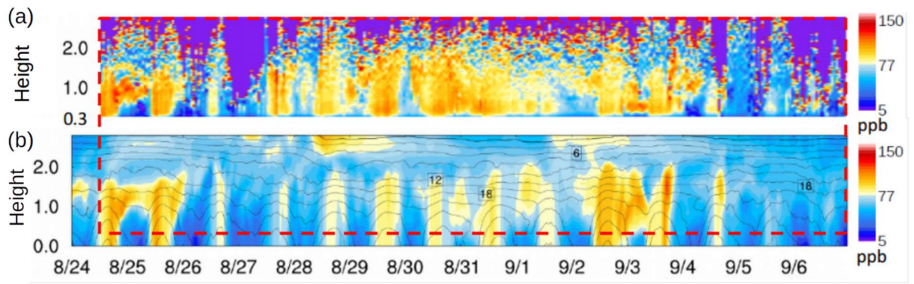


Fig. 5. Vertical comparison of hourly (a) observed (from differential absorption LiDAR) and (b) simulated O_3 concentrations (ppb) in Hangzhou from 24 August to 06 September 2016. Purple regions in the top panel denote invalid data with a low signal-to-noise ratio. To facilitate direct comparison, the red dashed line indicates the ozone level recorded for the same time periods (starting from 12:00, 24 August) and vertical heights (0.3–3 km) in the observations and simulation results.

To further evaluate the ability of the model to reproduce the vertical structure of ozone concentration, the vertical distribution of the modeled O_3 in Hangzhou from 24 August to 06 September 2016 is qualitatively compared with the DIAL datasets (data, as shown in Fig. 4). The comparison revealed that it is interesting to find that the present model can successfully predict the spatial-temporal distribution of ozone in Hangzhou. All observed major features of ozone are well captured by the model. This gives us high confidence and lays a solid foundation for further exploring the pollution characteristics and influencing factors of ozone in Hangzhou during the G20 summit.

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618 **3.2. Spatial-temporal variations of O₃ pollution**

619 To discuss spatial-temporal characteristics of O₃ pollution in Hangzhou, the whole emission
620 control period can be divided into three stages according to the reduction intensity of the measures.

621 August 24-27, 2016 is the first stage (S1) during which industrial and construction emission controls
622 were implemented. During the second stage (S2, August 28-31), traffic restrictions were further added.
623 September 1-6 2016 is the third stage (S3) with the emergent VOCs control further implemented.

624 Figs. 4(a) and 4(b) in the above section also present the temporal evolution of O₃ and its precursor
625 NO₂ in Hangzhou city during the emission control period of G20 summit. It is evident that the NO₂
626 has been significantly reduced by the emission control measures and the concentration is well below
627 the national level-II standard of 200 µg/m³. However, the concentration of O₃ keeps high levels for
628 the whole 14 days, with 7 days of MDA8 are above and 4 days are close to the national level-II
629 standard (GB-3095–2012) of 160µg/m³. This serious O₃ pollution indicates that the emission control

630 measures seem to make no obvious effect on ozone, which is consistent with the previous observations.

631 Diurnal O₃-variations were mainly observed within the planetary boundary layer (approximately <2
632 km). Notably, the model captured a nocturnal O₃-rich mass, which exhibited an n-shaped distribution
633 in the upper air (approximately 1 km) on 25 August 2016. (Su et al. 2017; Wu et al. 2019). The diurnal
634 variation of O₃ is similar for the

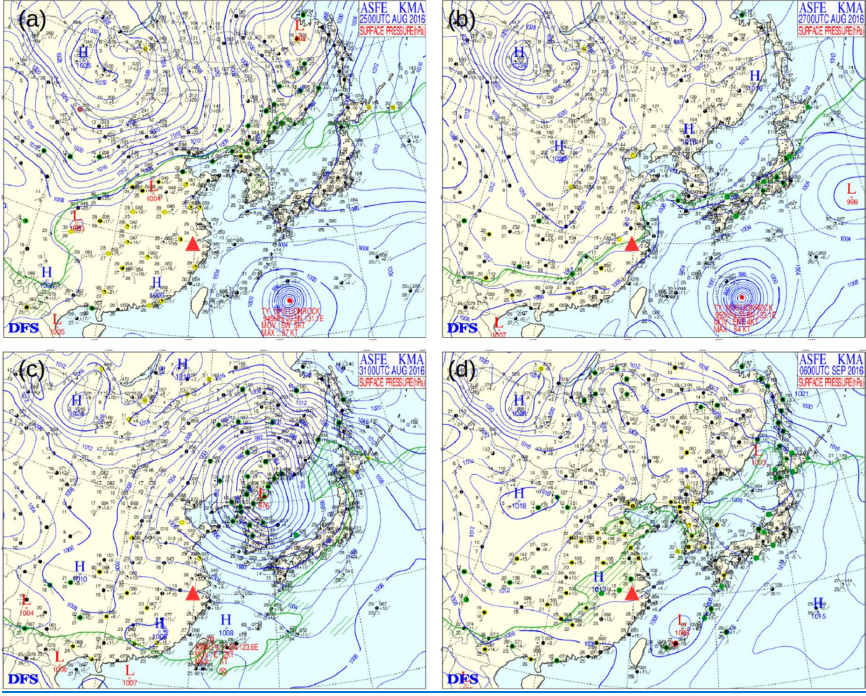
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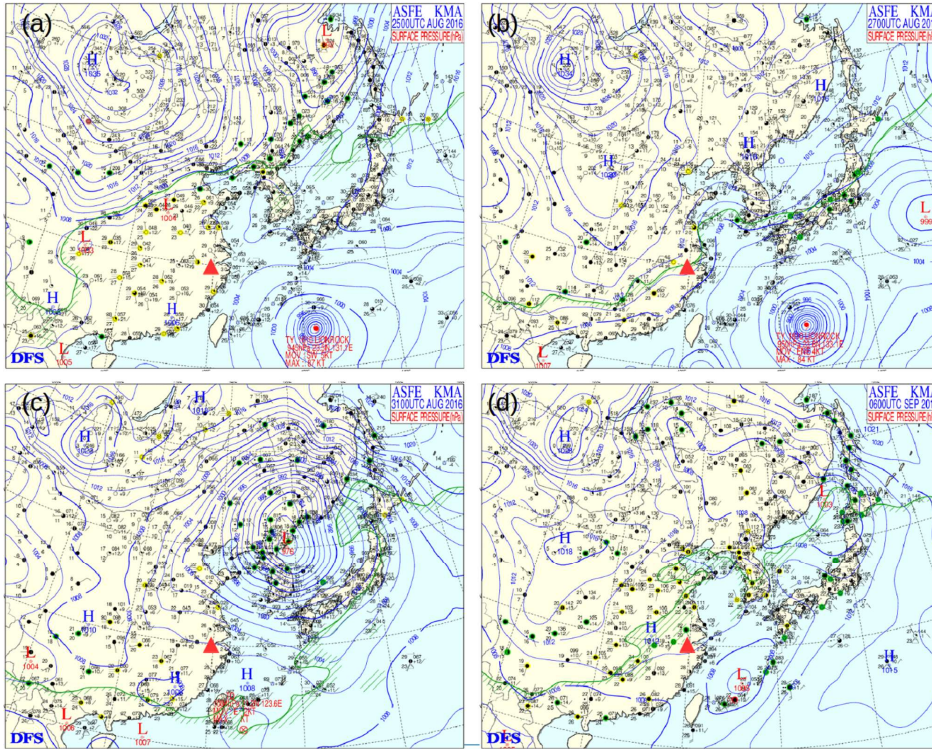


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Fig. 3.26. Synoptic weather system

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circulation. Fig. 5. Tropical cyclone evolution in East Asia during the 2016 G20 summit. Weather charts for four representative periods at 08:00 LST on (a) 25 August, (b) 27 August, (c) 31 August, and (d) 06 September 2016. LST: Local Sidereal Time; H: High-pressure system; L: Low-pressure system. The red triangle denotes the location of Hangzhou city.

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three stages with a peak value at the time around 16:00 and a valley value at the time around 8:00 of each day. However, the variation magnitude in Stage 2 is obviously lower than those of other stages, which will be further discussed later.

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Fig. 5 also clearly shows this diurnal variation of O_3 in the ground level. However, nocturnal O_3 -rich mass is observed during certain periods in the upper air (approximately 1 km), such as August 25, August 31, and September 3, which makes an n-shaped distribution pattern of the O_3 in the upper air. This kind of spatial distribution of ozone will promote vertical exchange of O_3 in the area. In

648

649 general, high concentrations of O₃ appear vertically until the top of the planetary boundary layer (PBL,
650 approximately <2 km), suggesting the ozone pollution is not a local but a regional phenomenon in
651 the whole low-level (from surface to close to the PBL height) region.

652 Considering the synoptic circulation is closely related to regional O₃ abundance, four
653 representative surface weather charts obtained from the Korea Meteorological Administration were
654 used to track the tropical cyclone (are presented in Fig. 5)-6. In the early stage of the tropical cyclone
655 during 24 and 25 August 2016 (Fig. 5a), strong and uniform high-pressure fields covered vast regions
656 of southeastern China. A, and a tropical cyclone moved northeastward over the East China Sea. (Fig.
657 6a). In the middle stage (Fig. 5b6b), the tropical cyclone approached the YRD region, bringing strong
658 north wind fields to this area. TheAs a result, the long narrow rain band arrived in Hangzhou (red
659 triangle) on 27 August 2016. In the later stage (Fig. 5e6c), the cyclone continuously moved toward
660 Japan and eventually hit the land. The, and the tropical high in the YRD region recovered gradually.
661 Finally, the cyclone faded, and a rainstorm appeared over most of the YRD region. This rainstorm
662 continued from approximately 02 through 07 September 2016 (Fig. 5d, for clarity, only the data for
663 06 September are presented (Fig. 6d).

664 3.3. O₃ pollution episode

665 In Fig. 6. The typical hourly vertical and horizontal O₃ distributions and wind fields in the YRD
666 region are further presented in Fig.7. The wind fields are also included for three representative
667 episodes according to the movement of the tropical cyclone. better understanding. For stagnation days
668 with weak wind fields (i.e., 25 August and 02 September) and strong radiation before or after the
669 tropical cyclone, meteorological conditions wereare unfavorable for pollutant dispersion. As a result,
670 O₃ pollution wasis more regional and intense, with an hourly peak O₃ concentration of 250 μg·m⁻³
671 below the high layer (2 km) around Hangzhou (Fig. 6a,e). As the cyclone approached (on 27 August),
672 a large belt of O₃-rich mass (>160 μg·m⁻³)/m³ appeared in the upwind direction and moved toward
673 Hangzhou under a prevailing north wind field (Fig. 6b). Transboundary pollutant transport played a
674 critical role within the planetary boundary layer in the whole YRD region, as shown in Figs. 7a and

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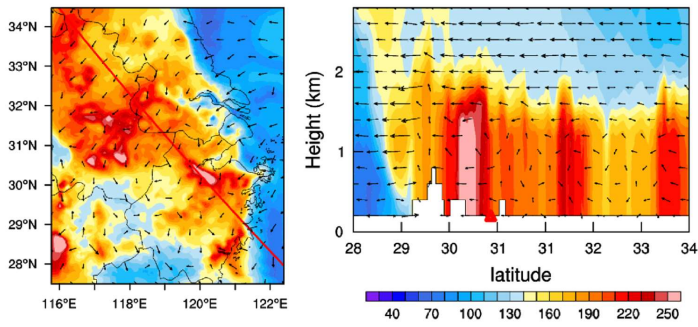
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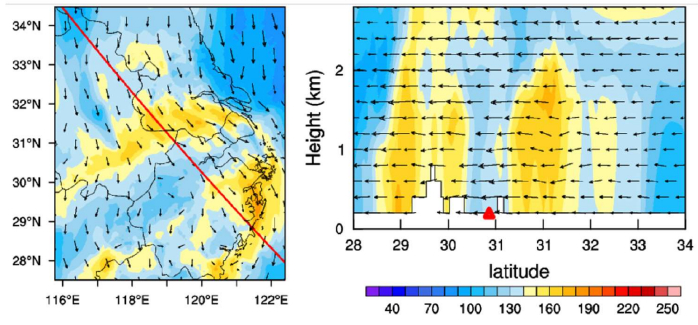
675 7c. In these processes, and this finding was consistent with the atmospheric trajectories from North
676 China (Fig. S2). This southward transport of pollutants may supply raw materials for conditions,
677 photochemical O₃-generation.

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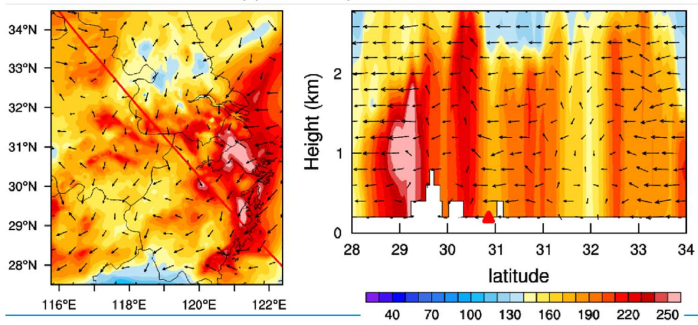
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(b) 14:00 Aug. 27, 2016

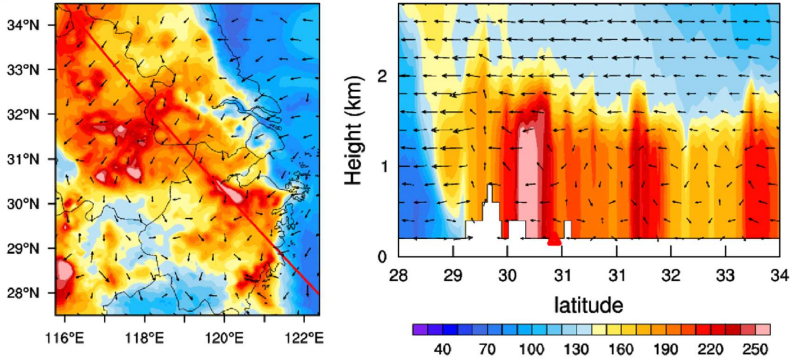


(c) 16:00 Sep. 02, 2016

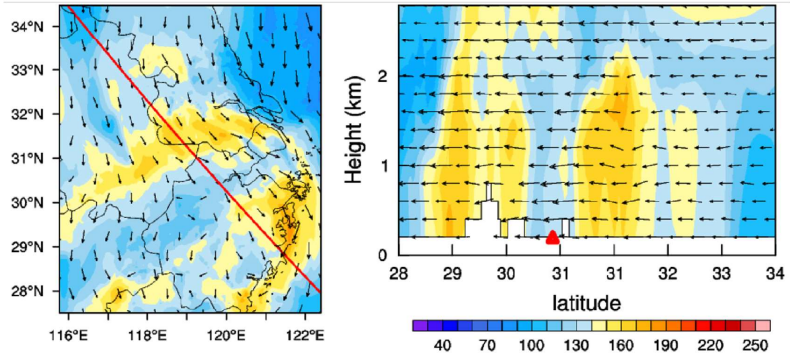


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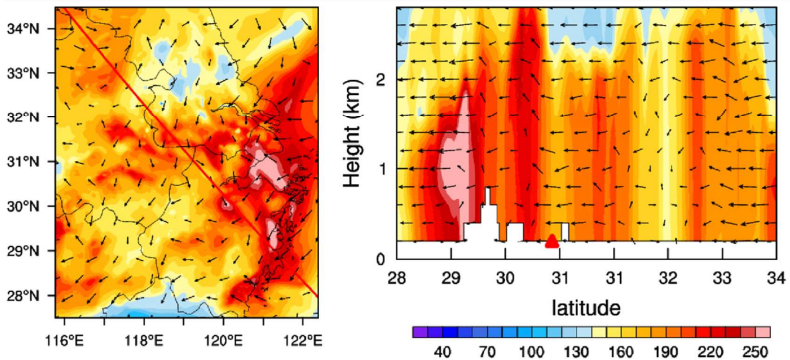
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Fig. 67. Surface and upper-level O_3 distributions ($\mu\text{g m}^{-3}$) and wind fields (vectors, m s^{-1}) for representative episodes. (a) Stagnant weather before the tropical cyclone, (b) pollutant transport when the tropical cyclone approached, and (c) stagnant weather after the cyclone. The red line denotes the cross-section line of upper-level O_3 distributions. The red triangle denotes the location of Hangzhou.

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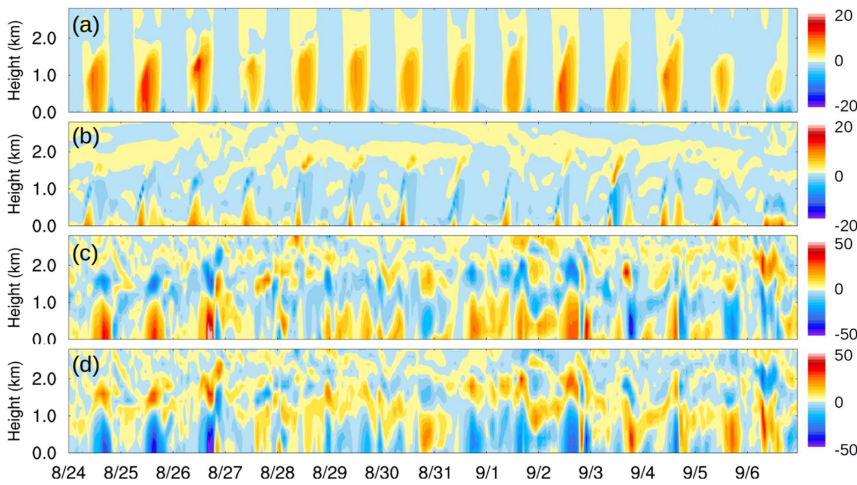
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684 [reactions dominate the ozone formation and accumulation. This phenomenon is consistent with the](#)
685 [satellite-derived tropospheric O₃ distribution in the area \(Su et al. 2017\), and is also supported by the](#)
686 [observed ozone data from the 96 sites in the YRD region as shown in Fig. 3c. During the 14-day](#)
687 [emission control period of G20 summit, 52% of the observed ozone samples from the 96 sites are](#)
688 [above the China's national level-II standard \(160μg/m³\), suggesting that regional ozone pollution](#)
689 [appears in the YRD region during the study period. As the cyclone approached on 27 August, a large](#)
690 [belt of O₃ mass appeared in the upwind direction and moved toward Hangzhou under a prevailing](#)
691 [north wind field \(Fig. 7b\). Regional pollutant transport may play an important role under this](#)
692 [condition. However, because of the rain and cooling effects from the cyclone, the ozone concentration](#)
693 [is relatively low in the whole YRD region.](#)

694 **3.53. Process analysis of O₃ formation**

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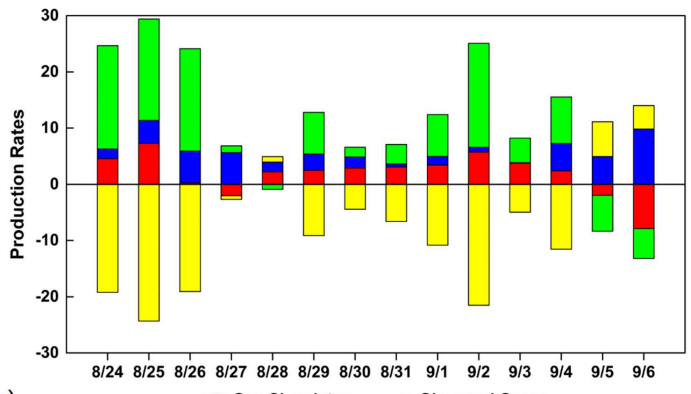
714 through vertical advection. The mass subsequently reversed and traveled away from Hangzhou in the
 715 highAt this higher layer through horizontal advection in a circular manner. Urban heat island
 716 circulations caused the upward and downward flows, the mass

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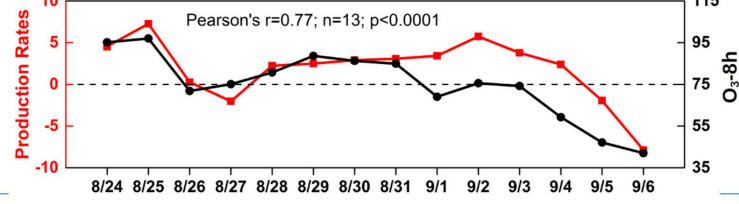
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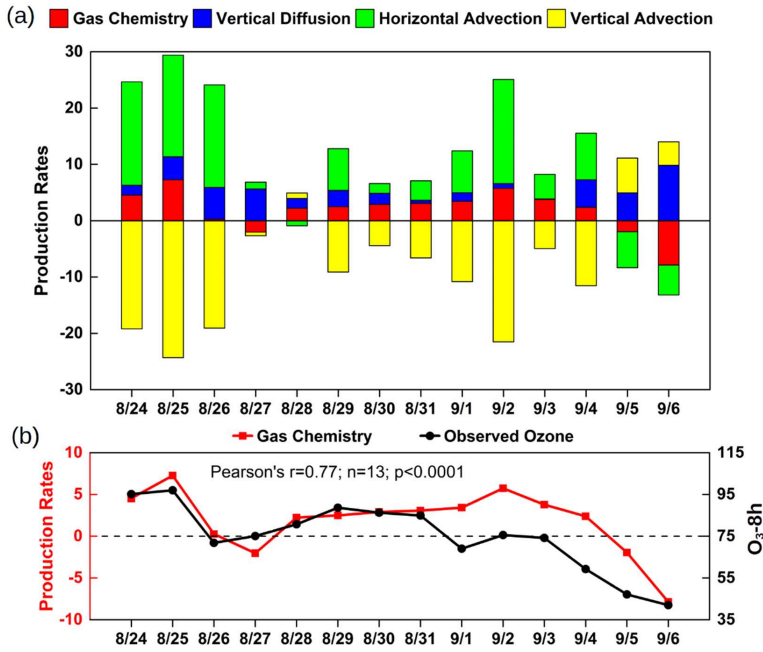
(a) Gas Chemistry Vertical Diffusion Horizontal Advection Vertical Advection



(b) Gas Chemistry Observed Ozone



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 719 **Fig. 89.** (a) Daytime mean (08:00–17:00 LST) variations in change rate of simulated surface O_3 change rate (ppb
 720 h^{-1} ; left y axis) resulting from gas chemistry, vertical diffusion, and horizontal and vertical
 721 advection in Hangzhou. (b) Comparison Correlation of daytime mean gas chemistry generation ($ppb h^{-1}$;
 722 left y axis) and observed surface-level maximum for daily 8-h8h concentration of O_3 (O_3 -8h; ppb; right y axis) in
 723 Hangzhou. China's national level-II standard is approximately 75 ppb ($160 \mu g m^{-3}$),
 724 — subsequently travels away from Hangzhou to other places through horizontal advection in a
 725 circular manner. This phenomenon might be associated with the urban heat island circulation (Lai
 726 and Cheng, 2009). However, during the period of September 3–6 2016, similar circulation
 727 phenomenon is observed, but the flow direction is reverse. The O_3 -rich mass travels downward to the
 728 ground through vertical advection and is then transported to surrounding regions through horizontal
 729 advection. This downward circulation is also related to the meteorological conditions after the
 730 cyclone. In addition, the horizontal and vertical advectons of O_3 take on a chaotic status during
 731 August 27–30, 2016, suggesting that complicated variable meteorological conditions happened in the
 732 time. This is also the reason for the lower magnitude of diurnal variation in Stage 2.

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Fig. 9 shows the daytime mean variationschange rate of simulated O₃ at the ground level resulted from various atmospheric processes (Fig. and the correlation of gas chemistry generation and observed maximum for daily 8h concentration of O₃. As a whole, the main sources of local surface ozone in Hangzhou are (8a). Quantification of these variations revealed major positive contributions from gas chemistry, vertical diffusion, and horizontal advection to surface O₃ formation, with mean production rates of 1.9, 3.3, and 6.7 ppb h⁻¹, respectively, from 24 August to 06 September 2016. Firstly, the trends between August 24 to September 6, 2016, and the major sink is vertical advection. However, during some days such as September 5-6, the gas chemistry consumes O₃ while the vertical advection increases it. In general, strong net horizontal and vertical advections of O₃ are observed for most days of the period, except August 27-28 during

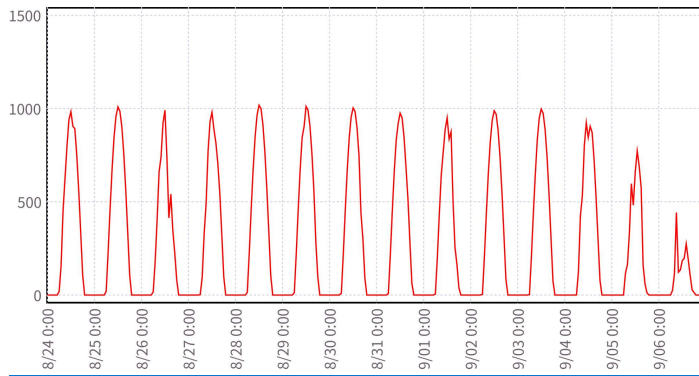


Fig. 10. Simulated hourly downward short wave flux at ground surface in Hangzhou (W m⁻²) during August 24 to September 6, 2016. which the strongest northwest cold winds (Fig. 4e) occurred and made the net advections of O₃ negligible. Similar to Fig. 8, dynamic O₃ circulations are observed for the periods of August 24-26, August 31 to September 2, and September 5-6. Particularly, the circular direction is reverse during September 5-6 and the net gas chemistry is to consume ozone due to weak solar radiation in the days as shown in Fig. 10.

In addition, the variation trend of the daytime mean values associated with production rate of gas chemistry and observed O₃-8h concentration were consistent (Fig. with the observed MDA8

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753 concentration and the local chemical generation has large positive correlation (Pearson's $r = 0.77$)
754 with the observed MDA8 concentrations (Fig. 8b), indicating). This indicates a trade-off effect among
755 vertical diffusion and horizontal advection, and vertical advection. High O_3 concentrations (i.e., 25
756 August 2016 O_3 -8hMDA8: 98 ppb) were always accompanied by strong radiation and prolific
757 generation of gas chemicals. Local chemical generation was found to have large positive correlation
758 (Pearson's $r = 0.77$) with O_3 concentrations. Secondly, chemical reactions. It is also interesting to find
759 that vertical diffusion may have partially compensated for gas chemistry when the
760 chemical reaction rate was relatively low or negative. For example, on 26 and 27 during August 26-
761 27 and 05 and 06 September 2016, most of 5-6, the vertical diffusion rates were greater are higher than
762 the chemical production rates. The low O_3 episode on these periods mainly resulted may result from
763 local chemical consumption. Finally, advection processes were essential and integral to air circulation:
764 horizontal advection exerted remarkably positive effects on surface O_3 concentrations in Hangzhou,
765 and vertical advection exerted dispersion effects.

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766 4. Discussion

767 The above results demonstrate that high ozone concentrations are observed, temporarily during
768 most day time of the emission control period of G20 summit, and spatially in Hangzhou and even the
769 whole YRD region, from the surface to the top of the planetary boundary layer. Strong horizontal and
770 vertical advectons appear, but they form circulations due to special meteorological conditions so that
771 the effects of them almost cancel each other out. As a result, the serious ozone pollution in Hangzhou
772 is mainly resultant from the local photochemical reactions. When the photochemical reactions are
773 weak, the vertical diffusion from the upper-air notable background O_3 further compensates for the
774 local surface ozone concentration. Therefore, it is of great importance to understand why the strict
775 emission control measures make no obvious effect on the local photochemical reactions of ozone
776 generation.

777 Chemical generation of O_3 is the net effect of photochemical generation and titration
778 consumption. VOC oxidation (Jenkin et al., 1997; Sillman, 1999) in photochemical reactions provides

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779 critical oxidants (i.e., RO₂) that efficiently convert NO to NO₂, resulting in further accumulation of
780 O₃ (Wang et al., 2017). This study revealed notable background O₃ concentrations in the upper air
781 layer in the YRD region. Peripheral downdrafts in large-scale cyclone circulation can transport an
782 O₃-rich mass in the upper troposphere or lower stratosphere downward to the surface (Tang et al.,
783 2011; Hsu and Prather, 2014). This type of O₃ intrusion during this period was reported in southeast
784 China (Ni et al., 2017). The chemical generation of O₃ is controlled by NO_x and VOCs depending on which
785 substance is lack in the reactions. As a consequence, there are two sensitivity regimes of O₃
786 production, namely, the NO_x-limited and VOC-limited regimes. Previous studies have shown that the
787 sensitivity pattern of surface O₃ formation in Hangzhou is dominant by the VOCs-limited regime
788 (Yan et al. 2016; Li et al., 2017; Su et al., 2017). In this regime, if the regional reduction of VOCs is
789 much higher than that of NO_x, the O₃ concentration can be reduced, otherwise if the regional reduction
790 of VOCs is much less than that of NO_x, the inhibitory effect of NO_x on O₃ generation will be
791 weakened, and the O₃ concentration will increase remarkably (Wang et al. 2015). According to the
792 studies of Su et al. (2017), Zheng et al. (2019), and Wu et al. (2019), it can be deduced that NO_x has
793 been significantly reduced by about 60%, at least two times of the reduction of VOCs in Hangzhou.
794 The influence of stringent emission control measures on VOCs is not as immediate and effect as that
795 on NO_x, which is associated with the fact that there was a large amount of biogenic VOC emission in
796 Hangzhou and surrounding regions (Liu et al. 2018; Wu et al. 2020). In fact, the average temperature
797 during the study period is as high as around 31°C (Fig. 4c), which facilitates the biogenic VOC
798 emissions and photochemical reactions. As a result, the photochemical generation of O₃ was not under
799 control and high concentration of ozone appeared. However, it is worth noting that after the emergent
800 VOCs control measures had been implemented in the area during the third stage, the net generation
801 rate of O₃ gradually reduces since September 2, 2016, leading to a period of relatively low ozone
802 concentration together with other meteorological effects. These discussions implicate that to alleviate
803 ozone pollution, the ratio of reduction of VOCs to that of NO_x is the key parameter based on the O₃-
804 NO_x-VOCs sensitivity analysis. As the biogenic VOCs are important sources of the total VOCs in the

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805 YRD region, it is necessary to balance the reduction of NO_x to make the ratio within effective regime
806 in the future.

807 ~~2019). Based on our results, we inferred that a considerably high background O₃ concentration~~
808 ~~in the upper air markedly contributed to surface O₃ pollution; this inference agreed (hemispheric~~
809 ~~background) with the findings of studies conducted in Europe (Wilson et al., 2012) and the United~~
810 ~~States (Lin et al., 2012, 2015).~~

811 We demonstrated that local chemical generation in Hangzhou was enhanced during episodes of
812 high O₃ concentrations before and after the tropical cyclone that occurred during the study period.

813 ~~Chemical generation of O₃ is the net effect of photochemical generation and titration consumption.~~

814 ~~VOC oxidation (Jenkin et al., 1997; Sillman, 1999) in photochemical reactions provides critical~~
815 ~~oxidants (i.e., RO₂) that efficiently convert NO to NO₂, resulting in further accumulation of O₃ (Wang~~

816 ~~et al., 2017). In the present study, downward shortwave flux at the ground level (Fig. S3) was more~~
817 ~~intense on days with high O₃ concentrations than on those with low O₃ concentrations. This strong~~

818 ~~solar radiation strengthened O₃ photochemical generation. In addition to the stagnant weather~~
819 ~~conditions, air subsidence in peripheral circulations of tropical cyclones helps to trap heat and~~

820 ~~pollutants at the surface (Jiang et al., 2015; Shu et al., 2016). Furthermore, a tropical system with~~
821 ~~calm, hot dry weather favors the development of an urban heat island, which causes thermal~~

822 ~~circulations as well as the convergence of the surrounding O₃ and its precursors (Lai and Cheng,~~
823 ~~2009). The increased temperature also accelerates the photochemical reactions (Narumi et al., 2009;~~

824 ~~Waleek et al., 1995). In the present study, these enhanced photochemistry processes dominated O₃~~
825 ~~chemical generation, resulting in high O₃ concentrations. This result was consistent with the results~~

826 ~~of a previous field study (Su et al., 2017). Low-level O₃ episodes (i.e., 06 September) in Hangzhou~~
827 ~~were accompanied by a rain band in the YRD region. Rain band related cumulus clouds blocked~~

828 ~~solar radiation, thereby weakening O₃ photochemical generation. Consequently, titration~~
829 ~~consumption dominated the chemical generation process, resulting in low or negative O₃ chemical~~

830 ~~production.~~

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831 **5. Conclusions**

832 ~~Changes in O₃ concentrations in Hangzhou~~ To understand the unique response of ozone to short-
833 ~~term emission control measures~~ during the G20 summit ~~were well represented in Hangzhou, the~~
834 ~~spatial-temporal characteristics and process analysis of O₃ pollution~~ are investigated by using the
835 WRF-Chem model. Statistical evaluations of meteorological and chemical parameters
836 ~~suggested~~ suggest that the model system ~~results satisfactorily matched~~ is able to reasonably predict the
837 observed data for both the ground and upper-air levels in MICS-ASIA Asia III. ~~The model results~~
838 ~~revealed that the O₃ High ozone concentrations~~ are observed, temporarily during most day time of the
839 ~~emission control period of G20 summit, and spatially in Hangzhou were highly related to a tropical~~
840 ~~eyclone over the East China Sea. Throughout the simulation period, large-scale air mass and even the~~
841 ~~whole YRD region, from the surface to the top of the planetary boundary layer. Horizontal and~~
842 ~~vertical advection, circulations and energy transport~~ are captured in Hangzhou, with horizontal
843 ~~advection the source and vertical advection the sink of the surface O₃ in Hangzhou. Consequently,~~
844 ~~the serious ozone pollution is mainly resultant from the local photochemical reactions which are not~~
845 ~~under good control~~ by the ~~tropical eyclone probably caused the high~~ emission reduction measures. As
846 ~~the surface O₃ formation in Hangzhou is dominant by the VOCs-limited regime, the significant~~
847 ~~reduction of NO_x compared to that of VOCs is unfavorable to chemical generation of O₃. The ratio~~
848 ~~of reduction of VOCs to that of NO_x based on the O₃-NO_x-VOCs sensitivity analysis is a critical~~
849 ~~parameter for reduction of ozone formation from photochemical reactions. In addition, it is found that~~
850 ~~the vertical diffusion from the upper-air O₃-rich mass in the horizontal and vertical scales of the YRD~~
851 ~~region; this phenomenon engendered a negative~~ notable background O₃ also plays an important role
852 ~~in shaping the surface ozone concentration. As the tropical eyclone approached, bringing with it a~~
853 ~~prevailing north wind component, Hangzhou was affected by pollutant transport from North China.~~
854 ~~After or before the tropical eyclone, peripheral downdraft or air subsidence produced stable and calm~~
855 ~~weather, with high pressure and temperature and~~ when the photochemical reactions are weak wind,
856 ~~and the urban heat island effect was aggravated. The combination of these conditions enhanced the~~

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857 chemical-generation process, resulting in a marked increase in surface O₃ concentrations. Our study
858 provides scientific insight into urban O₃ formation and dispersion under conditions where short-term
859 emission reduction measures had been applied in East China during a tropical cyclone event. _

860 Author contribution

861 Zhizhen Ni: Data curation, Investigation, Writing - original draft. Kun Luo: Methodology, Resources,
862 Writing - review & editing, Supervision. Yang Gao: Formal analysis, Methodology, Writing - review
863 & editing. Xiang Gao: Data curation, Resources. Fei Jiang: Methodology, Writing - review &
864 editing. Cheng Huang: Data curation, Formal analysis. Jianren Fan: Resources,
865 Supervision. Joshua Fu: Writing - review & editing. Changhong Chen: Formal analysis.

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873 the official documents of emission control policies were obtained from the Hangzhou Environmental
874 Monitoring Center.

875 Reference

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876 Competing interests

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

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