

Rapid O₃ assimilations – Part 1: background and local contributions to tropospheric O₃ changes in China in 2015-2020

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Abstract

A single ozone (O₃) tracer mode was developed in this work to build the capability of the GEOS-Chem model for rapid O₃ simulation. The single O₃ tracer simulation demonstrates consistency with the GEOS-Chem full chemistry simulation, with dramatic reductions in computational costs of approximately 91-94%. The single O₃ tracer simulation was combined with surface and Ozone Monitoring Instrument (OMI) O₃ observations to investigate the changes in tropospheric O₃ over eastern (E.) China in 2015-2020. The assimilated O₃ concentrations demonstrate good agreement with O₃ observations: surface O₃ concentrations are 43.2, 41.8 and 42.1 ppb, and tropospheric O₃ columns are 37.1, 37.9 and 38.0 DU in the simulations, assimilations and observations, respectively. The assimilations indicate rapid rises in surface O₃ concentrations by 1.60 (spring), 1.16 (summer), 1.47 (autumn) and 0.80 (winter) ppb yr⁻¹ over E. China in 2015-2020, and the increasing trends are underestimated by the a priori simulations. More attention is suggested to the rapid increases in O₃ pollution in spring and autumn. We find stronger rises in tropospheric O₃ columns over highly polluted areas due to larger local contributions, for example, 0.12 DU yr⁻¹ (North China Plain) in contrast to -0.29 (Sichuan Basin) -0.25 DU yr⁻¹ (Southern China). Furthermore, our analysis demonstrated noticeable contributions of the interannual variability in background O₃ to the trends in surface O₃ (particularly in the summer) and tropospheric O₃ columns over E. China in 2015-2020. This work highlights the importance of rapid simulations and assimilations to extend and interpret

32 atmospheric O₃ observations.

33

34 **1. Introduction**

35 Tropospheric ozone (O₃) is produced when volatile organic compounds (VOCs) and
36 carbon monoxide (CO) are photochemically oxidized in the presence of nitrogen oxides (NO_x).
37 Tropospheric O₃ has important influences on the climate (Mickley, 2004; Iglesias-Suarez et al.,
38 2018), atmospheric oxidation capacity (Thompson, 1992; Prinn, 2003), human health and crop
39 growth (Zhang et al., 2021; Li et al., 2022). The important role of O₃ in the atmosphere has led
40 to many efforts focusing on O₃ observations that have improved our understanding of
41 atmospheric O₃ (Logan et al., 2012; Oetjen et al., 2016; Parrish et al., 2021). The limited spatial
42 coverage of O₃ observations promotes the efforts of spatial extensions of O₃ observations
43 (Chang et al., 2015; Peng et al., 2016). Recent advances in machine learning techniques further
44 provide a new method to extend O₃ observations by fusing satellite and surface observations
45 (Li et al., 2020; Liu et al., 2022; Wei et al., 2022).

46 Chemical transport models (CTMs), as powerful tools, have been widely used to simulate
47 and interpret observed O₃ variabilities (Parrington et al., 2012; Jiang et al., 2016; Li, Ke et al.,
48 2019). Despite the advances in CTMs, an accurate simulation of observed O₃ is still challenging
49 because of uncertainties in physical and chemical processes (Peng et al., 2021; Chen et al.,
50 2022), emission inventories (Elguindi et al., 2020; Jiang et al., 2022) and coarse model
51 resolutions (Schaap et al., 2015; Benavides et al., 2021). Furthermore, the high computational
52 cost is a bottleneck for rapid simulations, which poses a possible barrier to better understanding
53 tropospheric O₃. Alternatively, researchers may consider simulations of atmospheric O₃ with
54 the archived O₃ product and loss rates. For example, the tagged-O_x mode of the GEOS-Chem
55 model has been used to analyze the sources and transport of tropospheric O₃ (Zhang et al., 2008;
56 Zhu et al., 2017; Han et al., 2018). However, it may not be an ideal choice to perform O₃

57 simulations based on the tagged- O_x mode because O_x is the combination of multiple species
58 ($O_x=O_3+NO_2+2NO_3+3N_2O_5+HNO_3+HNO_4+PANs$) and thus cannot be accurately compared
59 with O_3 observations.

60 In this study, we developed the single O_3 tracer mode (tagged- O_3) of the GEOS-Chem
61 model, driven by archived O_3 product and loss rates provided by GEOS-Chem full chemistry
62 simulations, to build the capability of the GEOS-Chem model for rapid simulations of
63 tropospheric O_3 (rather than O_x). Data assimilations, by combining modeled and observed O_3
64 concentrations, can take advantage of both simulations and observations to produce more
65 accurate O_3 concentrations (Parrington et al., 2008; Ma et al., 2019; Huijnen et al., 2020). The
66 single O_3 tracer simulations were thus further combined with the Ozone Monitoring Instrument
67 (OMI) and China Ministry of Ecology and Environment (MEE) monitoring network O_3
68 observations (in this paper) and United States (US) Air Quality System (AQS) and European
69 AirBase network O_3 observations (in the companion paper, (Zhu et al., 2023)) via a sequential
70 Kalman Filter (KF) assimilation system (Tang et al., 2022; Han et al., 2022) to perform a
71 comparative analysis to investigate the changes in tropospheric O_3 in eastern (E.) China in
72 2015-2020 (in this paper) and the US and Europe in 2005-2020 (Zhu et al., 2023).

73 Satellite instruments provide globally covered O_3 observations that are sensitive to O_3
74 concentrations in the free troposphere. The OMI-based assimilations can thus reflect the
75 optimized adjustments in both global background and local O_3 concentrations. On the other
76 hand, surface observations are sensitive to local O_3 concentrations. Surface observation-based
77 assimilations can reflect the optimized adjustments in local contributions, and the information
78 of local contributions can be transported into the free troposphere via vertical convection in the
79 assimilation processes, which is different from the fusion of satellite and surface observations
80 (Li et al., 2020; Liu et al., 2022; Wei et al., 2022). Consequently, a comparative analysis by
81 assimilating satellite and surface O_3 observations is useful for better characterization of O_3

82 changes in the surface and free troposphere. Furthermore, the low computational costs of the
83 single O₃ tracer simulations allow us to design and perform different experiments much more
84 efficiently. Multiple simulation and assimilation experiments (see details in Table 1) were thus
85 conducted in this work to analyze the impacts of background O₃ (particularly, the interannual
86 and seasonal variabilities in the background O₃ as well as optimization in the background O₃)
87 and local O₃ formation on the changes in surface and free tropospheric O₃ over E. China.

88 This paper is organized as follows: in Section 2, we provide descriptions of the MEE and
89 OMI O₃ observations, the GEOS-Chem model and the single O₃ tracer simulation and
90 assimilation system used in this work. Tropospheric O₃ changes in E. China in 2015-2020 are
91 then demonstrated in Section 3 by assimilating MEE and OMI O₃ observations. As shown in
92 Fig. 1, five regions (i.e., North China Plain (#1), Yangtze River Delta (#2), Central China (#3),
93 Sichuan Basin (#4) and Southern China (#5)) are defined within the E. China domain. Regions
94 #1 and #2 are defined as highly polluted regions by excluding grids with low and medium
95 anthropogenic NO_x emissions. Tropospheric O₃ changes over these regions are discussed to
96 investigate the possible regional discrepancies in surface and free tropospheric O₃ associated
97 with different local pollution levels. Our conclusions follow in Section 4.

98

99 **2. Data and Methods**

100 **2.1 Surface O₃ measurements**

101 We use MEE surface in situ O₃ concentration data (<https://quotsoft.net/air/>) for the period
102 2015-2020. These real-time monitoring stations report hourly concentrations of criteria
103 pollutants from 1691 sites in 2020. All stations (1441 urban sites and 250 urban background
104 sites) are assimilated in our analysis. Concentrations were reported by the MEE in μg m⁻³ under
105 standard temperature (273 K) until 31 August 2018. This reference state was changed on 1
106 September 2018 to 298 K. We converted the O₃ concentrations to ppb and rescaled the post-

107 August 2018 concentrations to the standard temperature (273 K) to maintain consistency in the
108 trend analysis. It should be noted that the assimilation of O₃ observations from urban and urban
109 background sites may result in possible overestimation of surface O₃ concentrations over rural
110 areas.

111 **2.2 OMI PROFOZ product**

112 The OMI instrument was launched in July 2004 on the Aura spacecraft with a spatial
113 resolution of 13 × 24 km (nadir view). It provides globally covered measurements with
114 backscattered sunlight in the ultraviolet–visible range from 270 to 500 nm (UV1: 270–310 nm;
115 UV2: 310–365 nm; visible: 350–500 nm). In this study, we use the OMI O₃ profile retrieval
116 product (PROFOZ v0.9.3, level 2, Liu et al. (2010); Huang et al. (2017)) from the Smithsonian
117 Astrophysical Observatory (SAO). The retrieval uses the vector linearized discrete ordinate
118 radiative transfer model (VLIDORT) (Spurr, 2006) and Bayesian optimal estimation. Profiles
119 of partial O₃ columns (unit: DU) are retrieved in the spectral region of 270–330 nm with 24
120 vertical layers: approximately 2.5 km for each layer from the surface to approximately 60 km.
121 The following filters are applied in our analysis following Huang et al. (2017): 1) nearly clear-
122 sky scenes with effective cloud fraction < 0.3; 2) solar zenith angles (SZA) < 75°; and 3) fitting
123 root mean square (RMS, ratio of fitting residuals to assumed measurement error) < 2.0.

124 Starting in 2009, anomalies were found in the OMI data and diagnosed as attenuated
125 measured radiances in certain cross-track positions. This instrument degradation has been
126 referred to as the “row anomaly”. To enhance the quality and stability of the data, only across-
127 track positions between 4-11 (within 30 positions in the UV1 channels) are assimilated in our
128 main assimilation experiment (Exp. #8). This treatment is similar to the production of row-
129 isolated data by using across-track positions between 3-18 (within 60 positions in the UV2
130 channels) in the OMI/MLS O₃ data (Ziemke et al., 2019; Wang, X. et al., 2022). The effects of
131 the usage of row-isolated data will be evaluated by comparing the main assimilation experiment

132 with the sensitivity assimilation experiment (Exp. #10) by assimilating OMI O₃ observations
133 at across-track positions 4-27.

134 The modeled tropospheric O₃ profiles in the assimilation processes and subsequent
135 analyses are convolved by using the OMI retrieval averaging kernels and a priori O₃ profile
136 based on the following equation (Liu et al., 2010; Huang et al., 2017):

$$137 \quad \hat{\mathbf{x}} = \mathbf{x}_a + \mathbf{A}(\mathbf{x} - \mathbf{x}_a) \quad (\text{Eq. 1})$$

138 where $\hat{\mathbf{x}}$ is the modeled O₃ profile convolved by the retrieval averaging kernels, \mathbf{x}_a is the
139 OMI a priori O₃ profile, \mathbf{x} is the modeled O₃ profile, and \mathbf{A} is the OMI averaging kernel
140 matrix. Here $A(i, j) = \frac{\partial \hat{x}_j}{\partial x_i}$, representing the sensitivity of the retrieved partial O₃ column (DU)
141 at layer j to the change in O₃ (DU) at layer i. The unit for averaging kernels in this OMI product
142 is DU/DU because the conversion from DU to ppb varies with altitude.

143 **2.3 GEOS-Chem model configuration**

144 The GEOS-Chem chemical transport model (<http://www.geos-chem.org>, version 12-8-1)
145 is driven by assimilated meteorological data from MERRA-2. The GEOS-Chem full chemistry
146 simulation includes fully coupled O₃-NO_x-VOC-halogen-aerosol chemistry. Our analysis is
147 conducted at a horizontal resolution of nested 0.5°×0.625° over E. China with chemical
148 boundary conditions archived every 3 hours from global simulations with 4°×5° resolution.
149 Emissions are computed by the Harvard-NASA Emission Component (HEMCO). Global
150 default anthropogenic emissions are from the CEDS (Community Emissions Data System)
151 (Hoesly et al., 2018). Regional emissions are replaced by MEIC (Multiresolution Emission
152 Inventory for China) in China and MIX in other regions of Asia (Li et al., 2017). The reference
153 year for the CEDS inventory is 2010 with annual scaling factors in 2005-2014, and the
154 reference year for the MEIC/MIX inventory is 2010 with annual scaling factors in 2008-2010
155 in the GEOS-Chem model. Open fire emissions are from the Global Fire Emissions Database
156 (GFED4) (van der Werf et al., 2010).

157 Following Jiang et al. (2022), the total anthropogenic NO_x and VOC emissions in the
158 GEOS-Chem model are scaled based on Zheng et al. (2018) and Li, M. et al. (2019) so that the
159 modeled surface nitrogen dioxide (NO₂) and O₃ concentrations in the a priori simulations are
160 identical to Jiang et al. (2022) in 2005-2018. The total anthropogenic NO_x and VOC emissions
161 in 2019-2020 are further scaled based on linear projections. The total anthropogenic NO_x
162 emissions in the a priori simulations declined by 19% in China in 2015-2020. The total
163 anthropogenic VOC emissions in the a priori simulations increased by 1% in China in 2015-
164 2020. We refer the reader to Jiang et al. (2022) for the details of the model configuration and
165 performance, particularly the modeled trends of surface and tropospheric column NO₂ in 2005-
166 2018.

167 **2.4 Single O₃ tracer simulation**

168 A new chemical mechanism was developed in this work to allow the running of the single
169 O₃ tracer mode (tagged-O₃). As shown in Fig. S1 (see the SI), the package of the Kinetic
170 PreProcessor (KPP) module was modified to define the production (PO₃) and loss (LO₃) of O₃.
171 The GEOS-Chem full chemistry simulations with the updated KPP module were then
172 performed to produce PO₃ (unit kg cm⁻³ s⁻¹) and relative LO₃ (i.e., LO₃/[O₃] with unit cm⁻³ s⁻¹)
173 every 20 minutes. Here the 20 minutes are selected to be the same as the chemical time step in
174 the GEOS-Chem full chemistry mode to ensure consistency between the single O₃ tracer and
175 full chemistry simulations. The single O₃ tracer simulation (tagged_o3_mod.F90) was then
176 performed by reading the archived PO₃ and relative LO₃. Because we are interested in
177 tropospheric chemistry, we archived O₃ concentrations instead of O₃ production and loss rates
178 in the stratosphere in the full chemistry simulations. The archived stratospheric O₃
179 concentrations were read in the single O₃ tracer simulation process as boundary conditions to
180 ensure a reasonable stratospheric-tropospheric O₃ exchange.

181 The major advantage of the single O₃ tracer simulation is dramatic reductions in

182 computational costs by approximately 91%-94%; for example, the computational costs (hours
183 of wall time for one year simulation) are 57.5 and 5.2 hours at the global scale ($4^\circ \times 5^\circ$) and 80.2
184 and 4.5 hours within the nested China domain ($0.5^\circ \times 0.625^\circ$) by full chemistry and single O₃
185 tracer simulations, respectively. Consequently, once PO₃ and LO₃ are produced, the
186 computational costs of performing additional single O₃ tracer simulations are almost negligible.
187 The low computational costs of the single O₃ tracer simulation allow us to design and perform
188 different simulation and assimilation experiments much more efficiently. As shown in Table
189 1, there are 10 different simulation and assimilation experiments performed in this work, which
190 requires 4812 hours (wall time) with the full chemistry simulation but only 270 hours (wall
191 time) with the single O₃ tracer simulation.

192 Here we evaluate the consistency in modeled O₃ concentrations between single O₃ tracer
193 and full chemistry simulations. Fig. 2A1-A5 show the annual and seasonal averages of the
194 surface maximum daily 8-hour average (MDA8) O₃ over E. China in 2015-2020 from the full
195 chemistry simulation. The modeled surface MDA8 O₃ concentrations are as high as 60-70 ppb
196 in the summer and as low as 10-20 ppb in the winter over northern China. The simulation with
197 the single O₃ tracer mode (Fig. 2B1-B5) demonstrates spatial consistency with the full
198 chemistry simulation (Fig. 2A1-A5) and temporal consistency at both the daily (Fig. 3A) and
199 monthly (Fig. 3B) scales in 2015-2020. In contrast, the tagged-O_x mode of the GEOS-Chem
200 model is driven by the archived production and loss of O_x, which is the combination of multiple
201 species including O₃. There are large discrepancies between full chemistry (Fig. 2A1-A5) and
202 tagged-O_x (Fig. 2C1-C5) simulations. As shown in Fig. 3, the O_x concentrations are higher than
203 the O₃ concentrations by approximately 6 ppb, and the relative difference can reach 40% in the
204 winter. Our analysis thus indicates the reliability of the single O₃ tracer simulations developed
205 in this work.

206 **2.5 Data assimilation method**

207 We employ the sequential KF to assimilate O₃ observations, which has been used in
 208 recent studies to optimize tropospheric CO concentrations (Tang et al., 2022; Han et al., 2022).
 209 As a brief description of the assimilation algorithm, the forward model (**M**) predicts the O₃
 210 concentration (\mathbf{x}_{at}) at time t:

$$211 \quad \mathbf{x}_{at} = \mathbf{M}_t \mathbf{x}_{t-1} \quad (\text{Eq. 2})$$

212 The optimized O₃ concentrations can be expressed as:

$$213 \quad \mathbf{x}_t = \mathbf{x}_{at} + \mathbf{G}_t (\mathbf{y}_t - \mathbf{K}_t \mathbf{x}_{at}) \quad (\text{Eq. 3})$$

214 where \mathbf{y}_t is the observation and \mathbf{K}_t represents the operation operator that projects O₃
 215 concentrations from the model space to the observation space. \mathbf{G}_t is the KF gain matrix, which
 216 can be described as:

$$217 \quad \mathbf{G}_t = \mathbf{S}_{at} \mathbf{K}_t^T (\mathbf{K}_t \mathbf{S}_{at} \mathbf{K}_t^T + \mathbf{S}_\epsilon)^{-1} \quad (\text{Eq. 4})$$

218 where \mathbf{S}_{at} and \mathbf{S}_ϵ are the model and observation covariances, respectively. The optimized O₃
 219 concentrations provided by Eq. 3 are then forwarded (hourly) to Eq. 2. The model errors are
 220 assumed to be 50% because the objective of our assimilations is to provide dynamic extensions
 221 of atmospheric O₃ observations. The a posteriori O₃ concentrations with the assumption of 50%
 222 model errors are expected to match better with atmospheric O₃ observations. The measurement
 223 errors are calculated as $\epsilon_0 = ermax + 0.0075 * \Pi_0$, where *ermax* is the base error (1.5 $\mu\text{g m}^{-3}$)
 224 and Π_0 represents the observed O₃ concentrations (unit: $\mu\text{g m}^{-3}$). The representation errors
 225 are calculated as $\epsilon_r = \gamma \epsilon_0 \sqrt{\Delta l / L}$, where γ is a scaling factor (0.5), Δl is the model resolution
 226 (~56 km in this study), and L represents the range that the observation can reflect, which
 227 depends on the station type (2 km for urban, 4 km for suburban). The total observation error is
 228 then defined as $\epsilon_t = \sqrt{\epsilon_0^2 + \epsilon_r^2}$. Furthermore, the "superobservation" method was applied in
 229 this work to further reduce the influence of representative error (Miyazaki et al., 2017; Tang et
 230 al., 2022):

$$231 \quad \omega_j = 1 / \epsilon_j^2 \quad (\text{Eq. 5})$$

232
$$y_s = \sum_{j=1}^k \omega_j y_j / \sum_{j=1}^k \omega_j \quad (\text{Eq. 6})$$

233
$$1/\varepsilon_s^2 = \sum_{j=1}^k 1/\varepsilon_j^2 \quad (\text{Eq. 7})$$

234 where y_j is O₃ observation of the j th station, ω_j represents the weighting factor of the j th
 235 station, y_s and ε_s are the grid-based O₃ observations and errors (superobservation),
 236 respectively.

237

238 **3. Results and Discussion**

239 **3.1 Surface O₃ by assimilating MEE O₃ observations**

240 We first investigate the effects of surface O₃ observations on single O₃ tracer
 241 assimilations. O₃ at the surface level is formed by precursors mixed in the planetary boundary
 242 layer (PBL). Thus, it may not be accurate to assume that the differences between simulated and
 243 observed surface O₃ concentrations are completely caused by biased O₃ production and loss at
 244 the surface level. Here we adjust O₃ concentrations above the surface level within the PBL
 245 when assimilating surface O₃ observations:

246
$$\Delta O_3^n = \Delta O_3^1 \times \gamma^{n-1} \quad (\text{Eq. 8})$$

247 where ΔO_3^1 is the adjustment at the surface level calculated with Eq. 3; ΔO_3^n is the adjustment
 248 at model level n , which is based on ΔO_3^1 but decays exponentially with the increase in model
 249 level, and the decay speed is adjusted by the γ parameter. As shown in Table 1, three
 250 assimilation experiments (Exp. #5-#7) were conducted to evaluate the effects of the decay
 251 speed: 1) $\gamma = 0$ by assuming that the biased surface O₃ concentrations are completely caused
 252 by biased O₃ production and loss at the surface level; 2) $\gamma = 1$ by assuming full mixing of O₃
 253 biases within the PBL; and 3) $\gamma = 0.8$ by assuming partial mixing of O₃ biases within the
 254 PBL, i.e., the adjustment at the 4th model level is approximately 50% of ΔO_3^1 , and the
 255 adjustment at the 10th model level (close to the top of PBL) is approximately 10% of ΔO_3^1 .

256 As shown in Fig. S2A (see the SI), the assimilated surface MDA8 O₃ concentrations
257 show good agreement by using different γ parameters: 42.3, 41.8 and 42.0 ppb ($\gamma = 0, 0.8$ and
258 1.0) in 2015-2020; there are noticeable discrepancies in the trends of assimilated surface O₃
259 concentrations: 0.80, 1.24 and 1.50 ppb yr⁻¹ ($\gamma = 0, 0.8$ and 1.0) in 2015-2020 (Fig. S2B), and
260 the trends obtained by considering the mixing of O₃ biases ($\gamma = 0.8$ and 1.0) match better with
261 MEE O₃ observations (1.77 ppb yr⁻¹). Fig. S3 (see the SI) further demonstrates tropospheric O₃
262 columns by assimilating MEE O₃ observations in 2015-2020. We find good agreement in the
263 assimilated tropospheric O₃ columns by using different γ parameters, i.e., the mean
264 tropospheric O₃ columns are 38.1, 37.9 and 37.9 DU, and the trends of tropospheric O₃ columns
265 are 0.11, 0.17 and 0.21 ppb yr⁻¹ ($\gamma = 0, 0.8$ and 1.0). Considering the better agreement in the
266 trends of assimilated surface O₃ concentrations ($\gamma = 0.8$ and 1.0) with observations, we finally
267 decide to set $\gamma = 0.8$ as our main assimilation setting by assuming partial mixing of O₃ biases
268 within the PBL.

269 Fig. 4A1-A5 show the annual and seasonal averages of surface MDA8 O₃ observations
270 from MEE stations in 2015-2020. Fig. 4C1-C5 show the annual and seasonal averages of the a
271 posteriori O₃ concentrations by assimilating the MEE O₃ observations. As shown in Fig. 5, the
272 assimilated O₃ concentrations (blue lines) show good agreement with MEE O₃ observations
273 (red lines): the mean surface MDA8 O₃ in 2015-2020 are 43.2, 41.8 and 42.1 ppb (E. China),
274 42.4, 45.6 and 47.6 ppb (North China Plain), 44.6, 45.0 and 44.9 ppb (Yangtze River Delta),
275 45.1, 43.1 and 43.5 ppb (Central China), 45.7, 37.5 and 36.9 ppb (Sichuan Basin), and 43.2,
276 39.2 and 38.3 ppb (Southern China) in the a priori simulations, a posteriori simulations and
277 MEE observations, respectively. It should be noted that Fig. 5A exhibits broadly good
278 agreement between the a priori and a posteriori O₃ concentrations over E. China except for a
279 larger difference in the summer. However, as shown in Fig. 4D1-D5, the good agreements
280 between the a priori and a posteriori O₃ concentrations are caused by the counterbalance of

281 positive biases (i.e., overestimated surface O₃ in the a priori simulations over southern China)
282 and negative biases (i.e., underestimated surface O₃ in the a priori simulations over northern
283 China). The good agreements in Fig. 5A thus cannot represent good performance in the
284 simulations of surface O₃ concentrations.

285 The assimilations exhibit noticeable declines in surface O₃ concentrations over regions
286 #2-5 in June-July, and the declines are underestimated by the a priori simulations (Fig. 5C-F).
287 The inaccurate simulation in June-July thus results in overestimated surface O₃ concentrations
288 in the summer. There is dramatic seasonality in surface O₃ concentrations (Fig. 5): maximum
289 in June in the North China Plain, May and August in the Yangtze River Delta, Central China
290 and Sichuan Basin, and September-October in Southern China. Fig. 4E1-E5 exhibits the effects
291 of seasonal variabilities in background O₃ (Exp. #3) by fixing background O₃ in the spring in
292 the simulations. The fixed background O₃ has limited influences on surface O₃ concentrations,
293 and consequently, the seasonality in surface O₃ concentrations is dominated by local
294 contributions. As we expected, MDA8 O₃ concentrations are higher over areas with higher
295 anthropogenic NO_x emissions, for example, 45.6 and 45.0 ppb in the North China Plain and
296 Yangtze River Delta, respectively, in contrast to 43.1, 37.5 and 39.2 ppb in Central China,
297 Sichuan Basin and Southern China. The influences of regional transport on surface O₃
298 concentrations are limited; for example, O₃ generated within the North China Plain PBL by
299 setting O₃ formation rates within the North China Plain PBL to zero (Exp. #4) are mainly
300 contained within the North China Plain (Fig. 4F1-F5).

301 **3.2 Rapid increasing trends in surface O₃ concentrations**

302 Here we investigate the changes in surface O₃ concentrations from observations and
303 assimilations. As shown in Fig. 6B1-B5, the a priori simulation suggests slightly increasing
304 trends of MDA8 O₃ in 2015-2020: 0.31 (spring), -0.12 (summer), 0.45 (autumn) and 0.40
305 (winter) ppb yr⁻¹, and the relative increasing trends are 0.7 (spring), -0.2 (summer), 1.1

306 (autumn) and 1.4 (winter) % yr⁻¹. The a priori simulation suggests increasing trends of surface
307 O₃ concentrations in the summer over areas with higher local pollution levels, for example,
308 0.68 and 0.63 ppb yr⁻¹ over the North China Plain and Yangtze River Delta, respectively, and
309 decreasing trends of surface O₃ concentrations in the summer over areas with lower local
310 pollution levels, for example, -0.83 and -1.01 ppb yr⁻¹ over the Sichuan Basin and Southern
311 China, respectively. The decreasing trends over areas with lower local pollution levels in the
312 simulations are not surprising, given the decreases in anthropogenic NO_x emissions (Zheng et
313 al., 2018; Jiang et al., 2022) and the reported NO_x-limited O₃ nonlinear chemical regimes in
314 model simulations (Chen et al., 2021; Liu et al., 2021). Furthermore, as shown in Fig. 6D1-D5,
315 the interannual variabilities in background O₃ (Exp. #2) are suggested to result in increases in
316 surface O₃ concentrations in the a priori simulations in 2015-2020 by 0.02 (spring), 0.05
317 (summer), 0.02 (autumn) and 0.00 (winter) ppb yr⁻¹, and the relative contribution is particularly
318 pronounced in the summer.

319 In contrast, the increasing trends in surface O₃ are much stronger in the assimilations. As
320 shown in Table 2.1, our assimilation suggests 1.60 (spring), 1.16 (summer), 1.47 (autumn) and
321 0.80 (winter) ppb yr⁻¹ increases in surface O₃ over E. China in 2015-2020, and the relative
322 increasing trends are 3.4 (spring), 2.2 (summer), 3.7 (autumn) and 2.7 (winter) % yr⁻¹. The
323 annual increasing trend (1.24 ppb yr⁻¹) in the assimilated surface O₃ concentrations is more
324 consistent with the MEE O₃ observations (1.77 ppb yr⁻¹) which are comparable with the
325 reported recent trends in surface O₃ concentrations in China of 1.25-2.0 ppb yr⁻¹
326 (Mousavinezhad et al., 2021; Wei et al., 2022; Wang, W. et al., 2022). The increasing trends
327 are weaker when the modeled surface O₃ concentrations are averaged over E. China (Table
328 2.2) instead of sampling at the locations and times of MEE observations: 0.71 (spring), 0.36
329 (summer), 0.69 (autumn) and 0.54 (winter) ppb yr⁻¹ because most MEE stations are urban sites.
330 Our analysis thus indicates a noticeable underestimation in the increasing trends of surface O₃

331 concentrations in China in the a priori simulations, particularly in the summer, despite the
332 anthropogenic NO_x and VOC emissions having been scaled in the simulations following Jiang
333 et al. (2022).

334 The changes in surface O₃ concentrations have significant regional and seasonal
335 discrepancies. As shown in Tables S1-S5 (see the SI), our assimilations demonstrate strong
336 increasing trends in surface O₃ concentrations in 2015-2020 in spring (1.94 ppb yr⁻¹ or 3.8%
337 yr⁻¹) and summer (2.52 ppb yr⁻¹ or 4.0% yr⁻¹) over the North China Plain; in spring (2.21 ppb
338 yr⁻¹ or 4.4% yr⁻¹) and autumn (1.84 ppb yr⁻¹ or 4.1% yr⁻¹) over the Yangtze River Delta; in
339 spring (2.07 ppb yr⁻¹ or 4.3% yr⁻¹) and autumn (2.09 ppb yr⁻¹ or 4.7% yr⁻¹) over Central China;
340 in spring (1.69 ppb yr⁻¹ or 3.8% yr⁻¹) over the Sichuan Basin; and in autumn (2.21 ppb yr⁻¹ or
341 4.9% yr⁻¹) over Southern China. While surface O₃ concentrations are higher over areas with
342 higher anthropogenic NO_x emissions, the increasing trends in surface O₃ concentrations over
343 Central China and Southern China are comparable with those in the North China Plain and
344 Yangtze River Delta. Our analysis advises more attention to O₃ pollution in spring and autumn
345 over areas with lower anthropogenic NO_x emissions because of the rapid increases in surface
346 O₃ concentrations.

347 **3.3 Tropospheric O₃ columns by assimilating OMI O₃ observations**

348 Fig. 7A1-A5 show the annual and seasonal averages of tropospheric OMI O₃ columns in
349 2015-2020. OMI is sensitive to O₃ at different vertical levels (Huang et al., 2017; Fu et al.,
350 2018), and thus, the standard KF algorithm (Eq. 3) was employed to adjust tropospheric O₃
351 vertical profiles with the application of OMI O₃ averaging kernels. Fig. 7C1-C5 show the
352 annual and seasonal averages of the a posteriori tropospheric O₃ columns by assimilating OMI
353 O₃ observations. The assimilated tropospheric O₃ columns show good agreement with OMI O₃
354 observations: the mean tropospheric O₃ columns in 2015-2020 (Table 2.3) are 37.1 DU in the
355 a priori simulations and 37.9 and 38.0 DU in the a posteriori simulation and OMI observations,

356 respectively. The discrepancies between the a priori and a posteriori simulations in tropospheric
357 O₃ columns (Fig. 7) are smaller than those in surface O₃ concentrations (Fig. 4). A better
358 simulation capability in tropospheric column O₃ is expected because model simulation with
359 0.5°×0.625° horizontal resolution may not be enough to accurately resolve O₃ nonlinear
360 chemical regimes over urban surface stations.

361 The above assimilated tropospheric O₃ columns (Exp. #8) are driven by optimized O₃
362 background conditions provided by global assimilations of OMI O₃ as well as row-isolated
363 OMI data by using across-track positions between 4-11. Fig. 7E1-E5 exhibits the effects of
364 optimization on regional O₃ background conditions. The mean assimilated tropospheric O₃
365 column driven by the original O₃ background conditions is 37.6 DU (Exp. #9), which is slightly
366 lower than the 37.9 DU in the main assimilation (Exp. #8). The usage of original O₃ background
367 conditions can result in overestimations over southern China in the spring and winter, and
368 underestimations over northern China in the spring and summer (Fig. 7E1-E5). Fig. 7F1-F5
369 further exhibits the effects of the usage of row-isolated data. The mean assimilated tropospheric
370 O₃ column by assimilating OMI O₃ observations at across-track positions 4-27 is 37.7 DU (Exp.
371 #10), which is slightly lower than the 37.9 DU in the main assimilation (Exp. #8). The
372 underestimations in the assimilated tropospheric O₃ columns are particularly significant in the
373 spring and summer (Fig. 7F2-F3).

374 As shown in Fig. 8, the trends of tropospheric O₃ columns in 2015-2020 (Table 2.3) are
375 0.02 DU yr⁻¹ in the a priori simulations and -0.17 and -0.30 DU yr⁻¹ in the a posteriori
376 simulation and OMI observations, respectively. In contrast to the wide distributions of
377 increasing trends of O₃ at the surface level (Fig. 6), both OMI O₃ observations (-0.30 DU yr⁻¹)
378 and the OMI-based assimilations (-0.17 DU yr⁻¹) suggest decreasing trends in tropospheric O₃
379 columns over E. Asia in 2015-2020 (Fig. 8). The decreasing trends are stronger in the summer
380 and weaker in the spring. Furthermore, the usage of original O₃ background conditions can

381 result in overestimated trend by approximately 0.08 DU yr^{-1} (Fig. 8D1); and the assimilation
382 of OMI O_3 observations at across-track positions 4-27 can result in a similar overestimated
383 trend, by approximately 0.08 DU yr^{-1} (Fig. 8E1). These discrepancies demonstrate the
384 importance of optimized usages of regional O_3 background conditions and OMI O_3
385 observations in the assimilations.

386 **3.4 Changes in tropospheric O_3 columns**

387 The trends shown in Fig. 8 may not represent the actual tropospheric O_3 changes well
388 because the convolution of OMI O_3 averaging kernels on the output O_3 profiles can affect the
389 weights of the derived tropospheric columns to O_3 at different vertical levels. Consequently,
390 Fig. 9 shows the annual and seasonal averages of tropospheric O_3 columns in which the output
391 O_3 profiles are not convolved with OMI retrieval averaging kernels so that they can better
392 represent the actual atmospheric O_3 state. The assimilated tropospheric O_3 columns are 37.9
393 and 38.8 DU (E. China), 42.9 and 43.7 DU (North China Plain), 47.5 and 48.1 DU (Yangtze
394 River Delta), 47.4 and 48.1 DU (Central China), 43.8 and 44.6 DU (Sichuan Basin), and 39.6
395 and 40.6 DU (Southern China) in 2015-2020 by assimilating MEE and OMI O_3 observations,
396 respectively.

397 In contrast to the higher surface MDA8 O_3 concentrations over areas with higher
398 anthropogenic NO_x emissions, tropospheric O_3 columns over Central China and the Sichuan
399 Basin are even higher than those over the highly polluted North China Plain. In addition,
400 tropospheric O_3 columns obtained by assimilating MEE surface O_3 observations are lower than
401 those obtained by assimilating OMI O_3 observations, and their difference is larger in the
402 summer and smaller in the winter. As shown in Fig. S4 (see the SI), the impacts of different
403 surface and satellite O_3 observations on the assimilated O_3 vertical profiles are limited. The
404 assimilation of MEE surface O_3 observations leads to decreases in O_3 concentrations in the
405 lower troposphere from the surface to 600 hPa levels over the Sichuan Basin and Southern

406 China; the assimilation of OMI O₃ observations leads to enhancement in O₃ concentrations in
407 the middle and upper troposphere over the highly polluted North China Plain.

408 The assimilated tropospheric O₃ columns are maximum in June-July over the highly
409 polluted North China Plain and March-May over other lower polluted regions (Fig. S5, see the
410 SI). Fig. 9E1-E5 exhibit the effects of seasonal variabilities in background O₃ (Exp. #3). The
411 fixed background O₃ in the spring can result in dramatic increases in tropospheric O₃ columns
412 by 14.3 (summer), 15.1 (autumn) and 4.8 (winter) DU over E. China. Fig. 9F1-F5 further
413 exhibit the effects of O₃ formation within the North China Plain PBL (Exp. #4) on tropospheric
414 O₃ columns, which are 5.4 (spring), 8.1 (summer), 3.6 (autumn) and 1.3 (winter) DU over the
415 North China Plain. In addition, as shown in Fig. S6 (see the SI), there is a larger enhancement
416 in O₃ production rates in the free troposphere (600-300 hPa) over the North China Plain in the
417 summer than in other lower polluted regions. Consequently, the spring maximum in
418 tropospheric O₃ columns over lower polluted regions is caused by the enhanced background O₃
419 (Fig. 9E1-E5), and the summer maximum in tropospheric O₃ columns over the highly polluted
420 North China Plain is caused by the local contributions from enhanced O₃ formation within the
421 North China Plain PBL (Fig. 9F1-F5) and free troposphere (Fig. S6).

422 As shown in Fig. 10A1-A5, the trends of tropospheric O₃ columns in the a priori
423 simulations in 2015-2020 are -0.02 (spring), 0.02 (summer), 0.29 (autumn) and 0.09 (winter)
424 DU yr⁻¹ over E. China. The interannual variability in background O₃ (Fig. 10D1-D5, Exp. #2)
425 is suggested to have important contributions to the trends of tropospheric O₃ columns by 0.09
426 (spring), -0.11 (summer), -0.10 (autumn) and -0.08 (winter) DU yr⁻¹. The trends of assimilated
427 tropospheric O₃ columns are 0.17 and -0.10 DU yr⁻¹ (E. China), which are comparable with the
428 reported recent trend in free tropospheric O₃ concentrations over China by -0.14 DU yr⁻¹
429 (Dufour et al., 2021), and are 0.47 and 0.12 DU yr⁻¹ (North China Plain), 0.45 and 0.13 DU yr⁻¹
430 (Yangtze River Delta), 0.32 and -0.06 DU yr⁻¹ (Central China), 0.03 and -0.29 DU yr⁻¹

431 (Sichuan Basin), and 0.06 and -0.25 DU yr⁻¹ (Southern China) by assimilating MEE and OMI
432 O₃ observations, respectively.

433 The stronger increasing trends in tropospheric O₃ columns over the highly polluted North
434 China Plain (Fig. 10A1) are suggested to be caused by larger local contributions because of
435 relatively uniform influences from interannual variability in background O₃ (Fig. 10D1).
436 Higher positive trends by assimilating MEE observations are expected, given the increasing
437 trends in surface O₃ concentrations (1.77 ppb yr⁻¹) and decreasing trends in OMI O₃
438 concentrations (-0.30 DU yr⁻¹) over E. China. Furthermore, it should be noted that while the
439 Yangtze River Delta is defined as a highly polluted region in our analysis, its area is much
440 smaller than that of the North China Plain (Fig. 1); thus, the impact of local contributions on
441 tropospheric O₃ columns over the Yangtze River Delta is not as strong as that over the North
442 China Plain.

443 **4. Conclusion**

444 A single O₃ tracer (tagged-O₃) mode was developed in this work to build the capability
445 of the GEOS-Chem model for rapid simulations of tropospheric O₃. The single O₃ tracer
446 simulation demonstrates consistency with the GEOS-Chem full chemistry simulation. In
447 contrast, the O_x concentrations provided by the tagged-O_x mode are higher than the O₃
448 concentrations by approximately 6 ppb, and the relative difference can reach 40% in the winter,
449 which is thus not suitable for direct comparison with observed O₃. The computational costs of
450 the single O₃ tracer mode are reduced by approximately 91-94% with respect to the full
451 chemistry mode. For example, the computational costs (hours of wall time per simulation year)
452 are 57.5 and 5.2 hours at the global scale (4°×5°) and 80.2 and 4.5 hours within the nested
453 China domain (0.5°×0.625°) by full chemistry and single O₃ tracer simulations, respectively.
454 The low computational costs allow us to design and perform different experiments much more
455 efficiently. As shown in Table 1, 10 different simulation and assimilation experiments are

456 performed in this work to analyze the impacts of background and local contributions to surface
457 and free tropospheric O₃ changes over E. China in 2015-2020, which requires 4812 hours (wall
458 time) with the full chemistry simulation but only 270 hours (wall time) with the single O₃ tracer
459 simulation.

460 As an application of the single O₃ tracer mode, the assimilated surface O₃ concentrations
461 demonstrate good agreement with surface O₃ observations: 43.2, 41.8 and 42.1 ppb over E.
462 China in a priori and a posteriori simulations and observations, respectively. We find noticeable
463 biases in modeled surface O₃ concentrations, for example, overestimated surface O₃ over
464 southern China and underestimated surface O₃ over northern China. The assimilations indicate
465 rapidly increasing trends in surface O₃ concentrations by 1.60 (spring), 1.16 (summer), 1.47
466 (autumn) and 0.80 (winter) ppb yr⁻¹ over E. China in 2015-2020, and the increasing trends are
467 underestimated by the a priori simulations. While surface O₃ concentrations are higher over
468 areas with higher anthropogenic NO_x emissions, the increasing trends in surface O₃
469 concentrations over Central China and Southern China are comparable with those in the North
470 China Plain and Yangtze River Delta. Our analysis thus advises more attention to O₃ pollution
471 in spring and autumn over areas with lower anthropogenic NO_x emissions in China because of
472 the rapid increases in surface O₃ concentrations.

473 Similarly, the assimilated tropospheric O₃ columns demonstrate good agreement with
474 OMI observations: 37.1, 37.9 and 38.0 DU over E. China in a priori and a posteriori simulations
475 (convolved with OMI retrieval averaging kernels) and OMI observations, respectively. The
476 trends of assimilated tropospheric O₃ columns in 2015-2020 over E. China are 0.09 and -0.17
477 (spring), 0.17 and -0.22 (summer), 0.38 and 0.04 (autumn), and 0.12 and -0.02 (winter) by
478 assimilating MEE and OMI O₃ observations, respectively. We find stronger increasing trends
479 in tropospheric O₃ columns over highly polluted areas due to the larger local contributions, for
480 example, 0.47 and 0.12 DU yr⁻¹ (North China Plain) in contrast to 0.03 and -0.29 DU yr⁻¹

481 (Sichuan Basin) and 0.06 and -0.25 DU yr⁻¹ (Southern China) by assimilating MEE and OMI
482 O₃ observations, respectively. The large discrepancy by assimilating surface and satellite
483 observations indicates the possible uncertainties in the derived free tropospheric O₃ changes.
484 The usage of optimized O₃ background conditions and row-isolated OMI data is important to
485 produce more reliable results, for example, the usage of original O₃ background conditions can
486 result in an overestimated trend by approximately 0.08 DU yr⁻¹ in 2015-2020.

487 Our analysis demonstrates noticeable contributions of the interannual variability in
488 background O₃ to the trends in tropospheric O₃ over E. China. The seasonality in surface O₃
489 concentrations is dominated by local contributions; however, the interannual variabilities in
490 background O₃ have noticeable contributions to the increasing trends in surface O₃ particularly
491 in the summer in the a priori simulations. Moreover, the spring maximum in tropospheric O₃
492 columns over lower polluted regions is caused by the enhanced background O₃, and the summer
493 maximum in tropospheric O₃ columns over the highly polluted North China Plain is caused by
494 enhanced local O₃ formation. The interannual variabilities in background O₃ have important
495 contributions to the trends in tropospheric O₃ columns; for example, the trends of tropospheric
496 O₃ columns in 2015-2020 are -0.02 (spring), 0.02 (summer), 0.29 (autumn) and 0.09 (winter)
497 DU yr⁻¹ over E. China, and the contributions from interannual variability in background O₃ are
498 0.09 (spring), -0.11 (summer), -0.10 (autumn) and -0.08 (winter) DU yr⁻¹ in the a priori
499 simulations. Our analysis thus suggests more attention to the impact of background O₃ to
500 tropospheric O₃ changes in China, particularly in the free troposphere.

501 The capability of rapid O₃ simulation developed in this work is a useful tool for
502 interpreting atmospheric O₃ observations. Assimilations of surface and satellite observations,
503 as shown in this work, can provide useful information to better describe the changes in surface
504 and free tropospheric O₃. Despite these advantages, it should be noted that the linear chemistry
505 assumption by reading the archived PO₃ and LO₃ implies single O₃ tracer mode is good for

506 representing near-current O₃ chemical conditions, particularly, for scientific issues associated
507 with the sources and transport of tropospheric O₃ as well as assimilations in this work and the
508 companion paper (Zhu et al., 2023). More cautious applications are suggested under
509 substantially different O₃ chemical conditions as the linear chemistry assumption could not be
510 satisfied.

511

512 **Code and data availability:** The MEE O₃ data can be downloaded from

513 <https://quotsoft.net/air/>. The AQS and AirBase surface O₃ data can be downloaded from

514 <https://www.eea.europa.eu/data-and-maps/data/aqereporting-8> and

515 https://aqs.epa.gov/aqsweb/airdata/download_files.html#Row. The OMI PROFOZ product

516 can be acquired at

517 <https://avdc.gsfc.nasa.gov/pub/data/satellite/Aura/OMI/V03/L2/OMPROFOZ/>. The GEOS-

518 Chem model (version 12.8.1) can be downloaded from [http://wiki.seas.harvard.edu/geos-](http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_12#12.8.1)

519 [chem/index.php/GEOS-Chem_12#12.8.1](http://wiki.seas.harvard.edu/geos-chem/index.php/GEOS-Chem_12#12.8.1). The KPP module for tagged-O₃ simulations can be

520 downloaded from <https://doi.org/10.5281/zenodo.7545944>.

521

522 **Author Contributions:** Z.J. designed the research. R.Z. developed the model code and

523 performed the research. Z.J. and R.Z. wrote the manuscript. X.L. provided instruction for the

524 usage of OMI data. All authors contributed to discussions and editing the manuscript.

525

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528

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536 **Table and Figures**

537 **Table 1.** Single O₃ tracer simulation and assimilation experiments (Exp.) conducted in this
538 work. Exp. #1: the main a priori simulation; Exp. #2: O₃ boundary conditions and stratospheric
539 O₃ concentrations are fixed in 2015; Exp. #3: O₃ boundary conditions and stratospheric O₃
540 concentrations are fixed in the spring; Exp. #4: O₃ formation rates within the North China Plain
541 PBL are set to zero; Exp. #5: the main assimilation by assimilating MEE surface O₃
542 observations with $\gamma = 0.8$; Exp. #6: only surface O₃ concentrations are adjusted ($\gamma = 0$); Exp.
543 #7: full mixing of O₃ biases within the PBL ($\gamma = 1.0$); Exp. #8: the main assimilation by
544 assimilating OMI O₃ observations; Exp. #9: O₃ boundary conditions are not optimized; Exp.
545 #10: assimilating OMI O₃ observations at across-track positions 4-27.

546
547 **Table 2.** Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface
548 and tropospheric column O₃ concentrations in 2015-2020 over E. China from observations
549 (MEE and OMI) and a priori (Exp. #1) and a posteriori (KF) simulations (Exp. #5 and #8). The
550 domain definition of E. China is shown by Fig. 1A. T2.1): the modeled surface O₃ is sampled
551 at the locations and times of MEE surface O₃ observations; T2.2): the modeled surface O₃ is
552 averaged over E. China (land only); T2.3): the output O₃ profiles from the a priori and a
553 posteriori simulations are convolved with OMI O₃ averaging kernels; T2.4): the output O₃
554 profiles are NOT convolved with OMI O₃ averaging kernels. The uncertainties in the averages
555 are calculated using the bootstrapping method. The trends and uncertainties in the trends are
556 calculated using the linear fitting of averages by using the least squares method (see details in
557 the SI).

558
559 **Fig. 1.** (A) Anthropogenic NO_x emissions over E. China in 2015; (B) Region definitions for
560 the North China Plain (#1), Yangtze River Delta (#2), Central China (#3), Sichuan Basin (#4)
561 and Southern China (#5). The different colors (red, gray and green) represent grids with high
562 (highest 15%), medium (15-50%) and low (lowest 50%) anthropogenic NO_x emissions.

563 Regions #1 and #2 are defined as highly polluted (HP) regions by excluding grids with low and
564 medium anthropogenic NO_x emissions.

565

566 **Fig. 2.** Surface MDA8 O₃ in 2015-2020 (annual and seasonal averages) simulated by GEOS-
567 Chem model with (A1-A5) full chemistry mode; (B1-B5) single O₃ tracer (tagged-O₃) mode;
568 and (C1-C5) tagged-O_x mode. The 8-hour range of surface O_x is selected according to the time
569 range of MDA8 O₃.

570

571 **Fig. 3.** (A) Daily averages of surface MDA8 O₃ over E. China in 2015-2020 from GEOS-Chem
572 full chemistry (black), single O₃ tracer (tagged-O₃) (blue) and tagged-O_x (red) simulations; (B)
573 Monthly averages of MDA8 O₃. The dashed lines in panel B are annual averages.

574

575 **Fig. 4.** Surface MDA8 O₃ in 2015-2020 (annual and seasonal averages) from (A1-A5) MEE
576 stations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem a
577 posteriori simulation by assimilating MEE O₃ observations (Exp. #5); (D1-D5) Bias in the a
578 priori simulations (Exp. #1 minus #5). (E1-E5) Effects of seasonal variabilities in background
579 O₃ (Exp. #3 minus #1); (F1-F5) Effects of O₃ formation within the North China Plain PBL
580 (Exp. #1 minus #4).

581

582 **Fig. 5.** (A-F) Daily averages of surface MDA8 O₃ in 2015-2020 from MEE stations (red) and
583 GEOS-Chem a priori (black, Exp. #1) and a posteriori (blue, Exp. #5) simulations by
584 assimilating MEE O₃ observations. (G-L) Monthly averages of MDA8 O₃. The dashed lines in
585 panels G-L are annual averages. The domain definition of E. China is shown by Fig. 1A.

586

587 **Fig. 6.** Trends of surface MDA8 O₃ in 2015-2020 (annual and seasonal averages) from (A1-
588 A5) MEE stations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem
589 a posteriori simulation by assimilating MEE O₃ observations (Exp. #5). (D1-D5) Effects of
590 interannual variabilities in background O₃ (Exp. #1 minus #2).

591

592 **Fig. 7.** Tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from (A1-A5)
593 OMI observations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem
594 a posteriori simulation by assimilating OMI O₃ observations (Exp. #8). (D1-D5) Bias in the a
595 priori simulations (Exp. #1 minus #8). (E1-E5) Effects of optimization on regional O₃

596 background conditions (Exp. #9 minus #8); (F1-F5) Effects of the usage of row-isolated data
597 (Exp. #10 minus #8). The output O₃ profiles are convolved with OMI averaging kernels.

598

599 **Fig. 8.** Trends of tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from
600 (A1-A5) OMI observations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5)
601 GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations (Exp. #8). (D1-D5)
602 Effects of optimization on regional O₃ background conditions (Exp. #9 minus #8); (E1-E5)
603 Effects of the usage of row-isolated data (Exp. #10 minus #8). The output O₃ profiles are
604 convolved with OMI averaging kernels.

605

606 **Fig. 9.** Tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from (A1-A5)
607 GEOS-Chem a priori simulation (Exp. #1); (B1-B5) Assimilations of MEE surface O₃
608 observations (Exp. #5); (C1-C5) Assimilations of OMI O₃ observations (Exp. #8). (D1-D5)
609 Difference in tropospheric O₃ columns calculated by OMI-based assimilations minus MEE-
610 based assimilations (Exp. #8 minus #5). (E1-E5) Effects of seasonal variabilities in background
611 O₃ (Exp. #3 minus #1); (F1-F5) Effects of O₃ formation within the North China Plain PBL
612 (Exp. #1 minus #4). The output O₃ profiles are NOT convolved with OMI averaging kernels.

613

614 **Fig. 10.** Trends of tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from
615 (A1-A5) GEOS-Chem a priori simulation (Exp. #1); (B1-B5) Assimilations of MEE surface
616 O₃ observations (Exp. #5); (C1-C5) Assimilations of OMI O₃ observations (Exp. #8). (D1-D5)
617 Effects of interannual variabilities in background O₃ (Exp. #1 minus #2). The output O₃ profiles
618 are NOT convolved with OMI averaging kernels.

619

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815

	Experiments	Observations	O3 Boundary Conditions	Other Settings
A priori Simulations	#1 (Main)	N/A	Original (2015-2020)	
	#2	N/A	Original (2015, fixed)	
	#3	N/A	Original (2015-2020, fixed in spring)	
	#4	N/A	Original (2015-2020)	PO3 = 0 (NCP)
Kalman Filter Assimilations	#5 (Main)	MEE	Original (2015-2020)	$\gamma = 0.8$
	#6	MEE	Original (2015-2020)	$\gamma = 0.0$
	#7	MEE	Original (2015-2020)	$\gamma = 1.0$
	#8 (Main)	OMI	Optimized (2015-2020)	positions: 4-11
	#9	OMI	Original (2015-2020)	positions: 4-11
	#10	OMI	Optimized (2015-2020)	positions: 4-27

Table 1. Single O₃ tracer simulation and assimilation experiments (Exp.) conducted in this work. Exp. #1: the main a priori simulation; Exp. #2: O₃ boundary conditions and stratospheric O₃ concentrations are fixed in 2015; Exp. #3: O₃ boundary conditions and stratospheric O₃ concentrations are fixed in the spring; Exp. #4: O₃ formation rates within the North China Plain PBL are set to zero; Exp. #5: the main assimilation by assimilating MEE surface O₃ observations with $\gamma = 0.8$; Exp. #6: only surface O₃ concentrations are adjusted ($\gamma = 0$); Exp. #7: full mixing of O₃ biases within the PBL ($\gamma = 1.0$); Exp. #8: the main assimilation by assimilating OMI O₃ observations; Exp. #9: O₃ boundary conditions are not optimized; Exp. #10: assimilating OMI O₃ observations at across-track positions 4-27.

E. China (2015-2020)		Annual		Spring		Summer		Autumn		Winter	
		Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend	Mean	Trend
T2.1 surface (sampled)	MEE	42.1±0.3	1.77±0.38	48.4±0.4	2.25±0.46	51.7±0.6	1.70±0.64	39.8±0.4	2.01±0.60	29.6±0.2	1.14±0.49
	a priori	43.2±0.2	0.21±0.13	48.0±0.2	0.31±0.15	56.3±0.5	-0.12±0.38	40.1±0.3	0.45±0.19	28.5±0.3	0.40±0.17
	KF-MEE	41.8±0.2	1.24±0.28	47.2±0.3	1.60±0.34	51.7±0.5	1.16±0.55	39.5±0.3	1.47±0.47	29.5±0.2	0.80±0.37
T2.2 surface	a priori	42.6±0.1	0.10±0.11	47.7±0.1	0.16±0.11	53.1±0.2	-0.19±0.29	39.1±0.1	0.25±0.19	30.8±0.2	0.35±0.13
	KF-MEE	41.3±0.1	0.55±0.17	46.7±0.1	0.71±0.17	49.8±0.2	0.36±0.36	38.0±0.1	0.69±0.31	31.0±0.2	0.54±0.19
T2.3 trop. column (convolved)	OMI	38.0±0.2	-0.30±0.19	40.9±0.2	0.12±0.20	45.9±0.2	-0.66±0.44	34.6±0.2	-0.41±0.30	30.4±0.2	-0.48±0.40
	a priori	37.1±0.1	0.02±0.14	41.0±0.2	0.17±0.24	43.2±0.2	-0.19±0.16	32.6±0.1	0.15±0.19	31.3±0.2	-0.06±0.18
	KF-OMI	37.9±0.1	-0.17±0.15	41.1±0.2	0.08±0.07	45.5±0.2	-0.51±0.37	34.2±0.1	-0.17±0.24	30.7±0.1	-0.17±0.23
T2.4 trop. Column	a priori	38.3±0.1	0.07±0.14	42.8±0.2	-0.02±0.46	42.5±0.2	0.02±0.16	33.3±0.1	0.29±0.11	34.8±0.2	0.09±0.32
	KF-MEE	37.9±0.1	0.17±0.16	42.6±0.2	0.09±0.47	41.8±0.2	0.17±0.15	33.0±0.1	0.38±0.12	34.7±0.2	0.12±0.32
	KF-OMI	38.8±0.1	-0.10±0.25	42.9±0.2	-0.17±0.57	44.1±0.2	-0.22±0.26	34.4±0.1	0.04±0.12	34.2±0.2	-0.02±0.30

Table 2. Averages (with units ppb or DU) and trends (with units ppb yr⁻¹ or DU yr⁻¹) of surface and tropospheric column O₃ concentrations in 2015-2020 over E. China from observations (MEE and OMI) and a priori (Exp. #1) and a posteriori (KF) simulations (Exp. #5 and #8). The domain definition of E. China is shown by Fig. 1A. T2.1): the modeled surface O₃ is sampled at the locations and times of MEE surface O₃ observations; T2.2): the modeled surface O₃ is averaged over E. China (land only); T2.3): the output O₃ profiles from the a priori and a posteriori simulations are convolved with OMI O₃ averaging kernels; T2.4): the output O₃ profiles are NOT convolved with OMI O₃ averaging kernels. The uncertainties in the averages are calculated using the bootstrapping method. The trends and uncertainties in the trends are calculated using the linear fitting of averages by using the least squares method (see details in the SI).

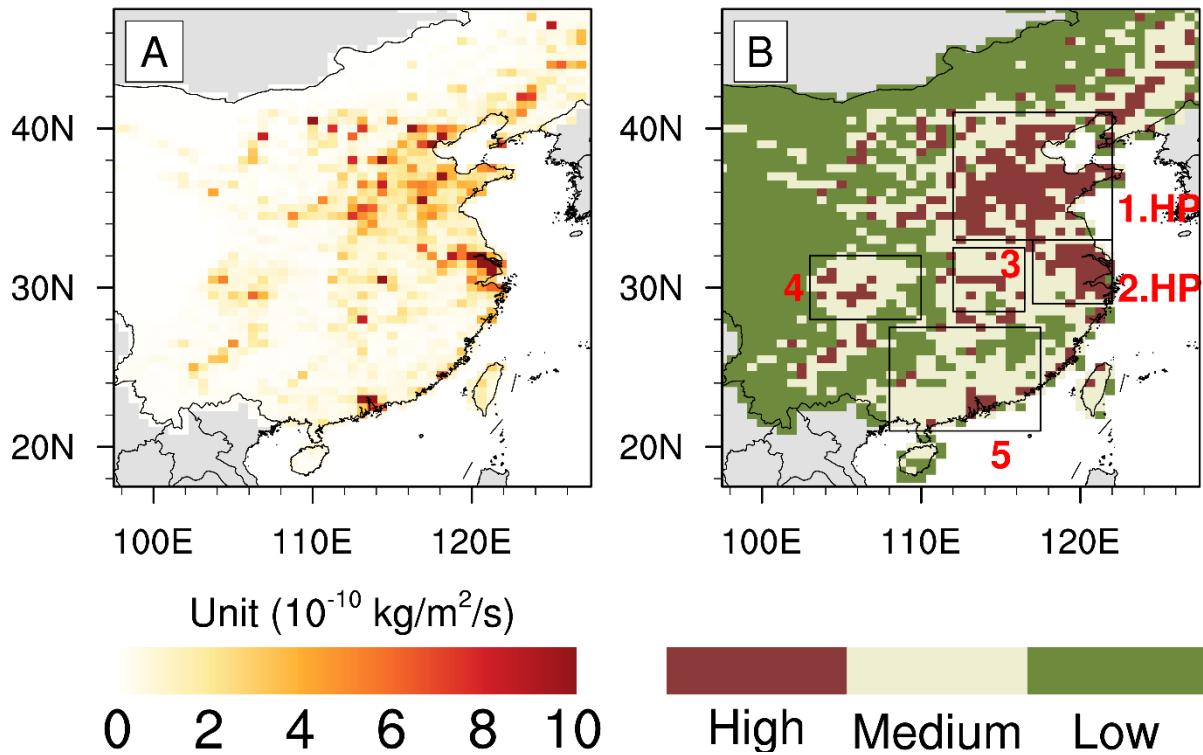


Fig. 1. (A) Anthropogenic NO_x emissions over E. China in 2015; (B) Region definitions for the North China Plain (#1), Yangtze River Delta (#2), Central China (#3), Sichuan Basin (#4) and Southern China (#5). The different colors (red, gray and green) represent grids with high (highest 15%), medium (15-50%) and low (lowest 50%) anthropogenic NO_x emissions. Regions #1 and #2 are defined as highly polluted (HP) regions by excluding grids with low and medium anthropogenic NO_x emissions.

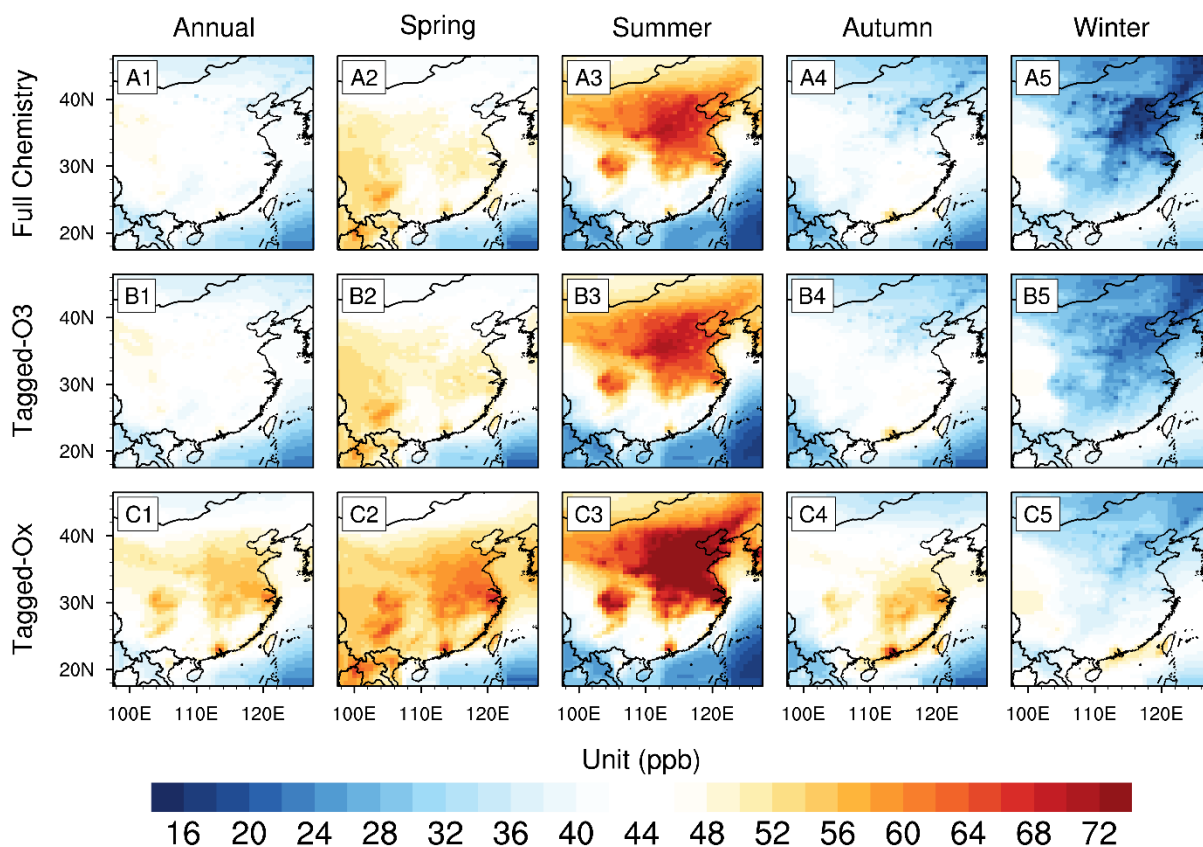


Fig. 2. Surface MDA8 O₃ in 2015-2020 (annual and seasonal averages) simulated by GEOS-Chem model with (A1-A5) full chemistry mode; (B1-B5) single O₃ tracer (tagged-O₃) mode; and (C1-C5) tagged-O_x mode. The 8-hour range of surface O_x is selected according to the time range of MDA8 O₃.

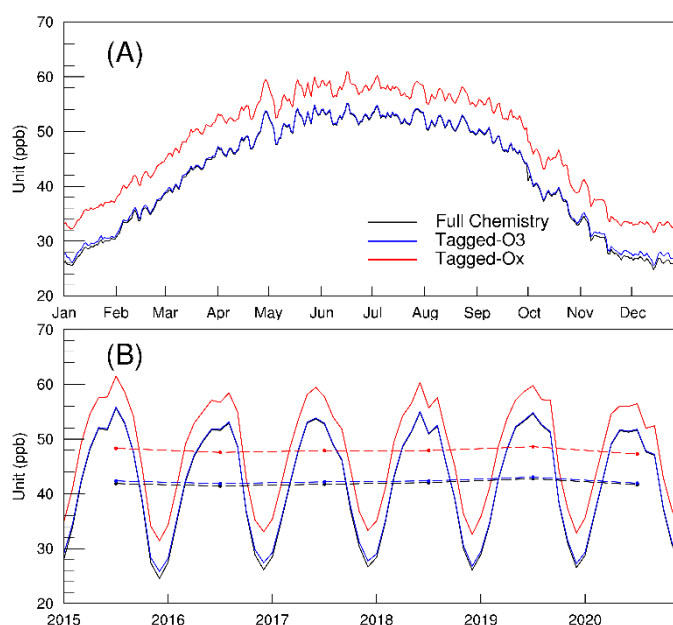


Fig. 3. (A) Daily averages of surface MDA8 O₃ over E. China in 2015-2020 from GEOS-Chem full chemistry (black), single O₃ tracer (tagged-O₃) (blue) and tagged-O_x (red) simulations; (B) Monthly averages of MDA8 O₃. The dashed lines in panel B are annual averages.

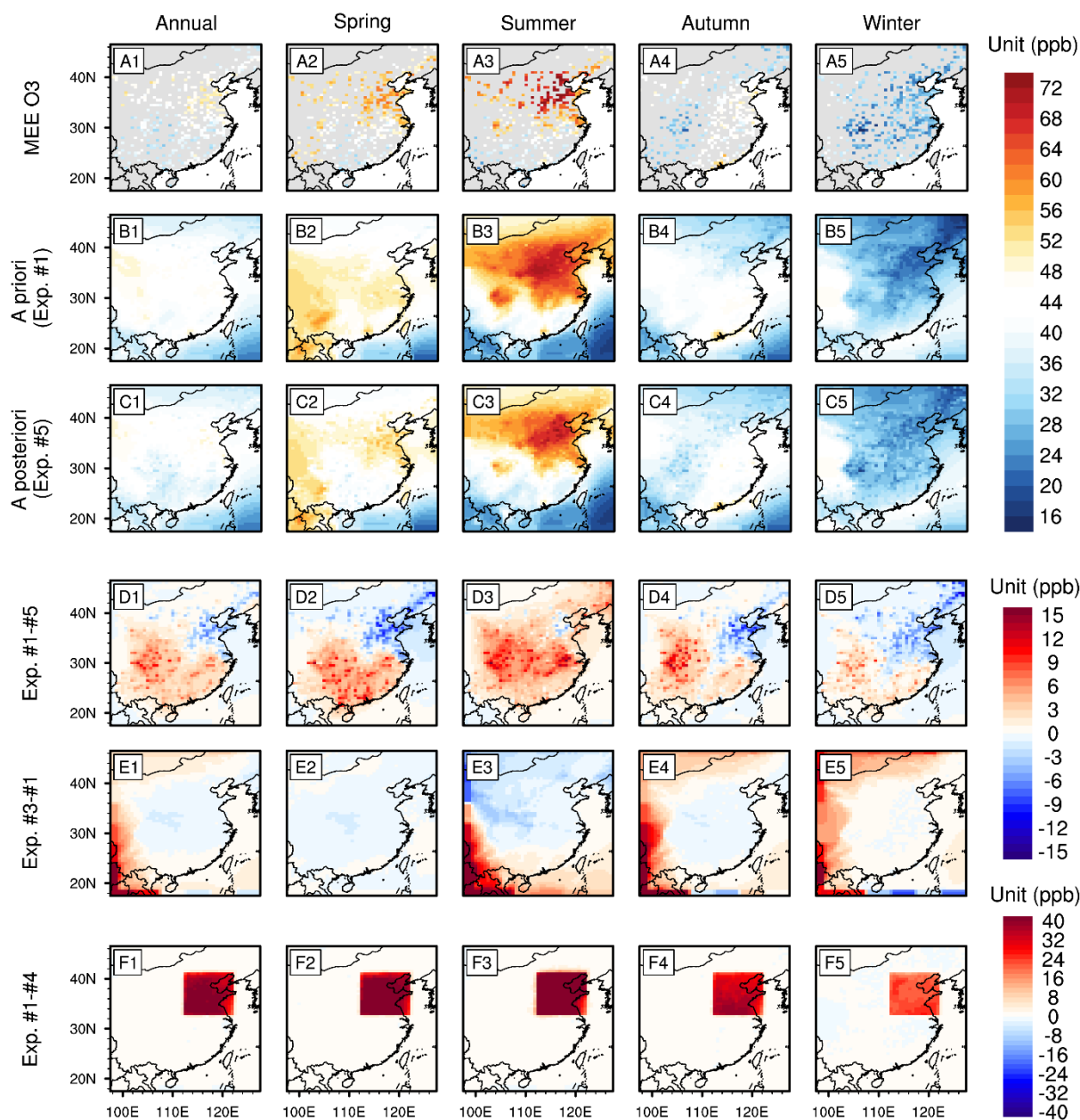


Fig. 4. Surface MDA8 O₃ in 2015-2020 (annual and seasonal averages) from (A1-A5) MEE stations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem a posteriori simulation by assimilating MEE O₃ observations (Exp. #5); (D1-D5) Bias in the a priori simulations (Exp. #1 minus #5). (E1-E5) Effects of seasonal variabilities in background O₃ (Exp. #3 minus #1); (F1-F5) Effects of O₃ formation within the North China Plain PBL (Exp. #1 minus #4).

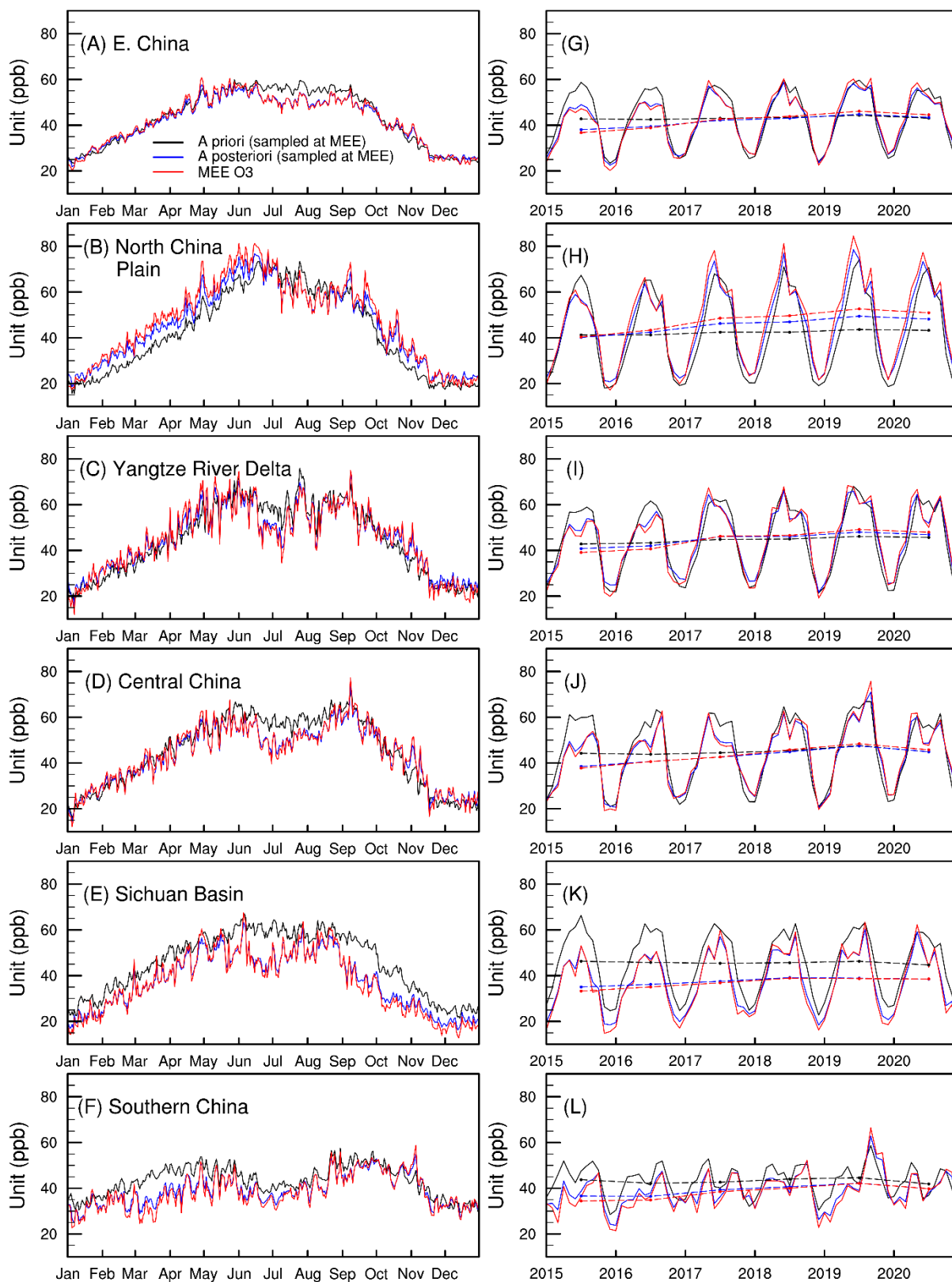


Fig. 5. (A-F) Daily averages of surface MDA8 O₃ in 2015-2020 from MEE stations (red) and GEOS-Chem a priori (black, Exp. #1) and a posteriori (blue, Exp. #5) simulations by assimilating MEE O₃ observations. (G-L) Monthly averages of MDA8 O₃. The dashed lines in panels G-L are annual averages. The domain definition of E. China is shown by Fig. 1A.

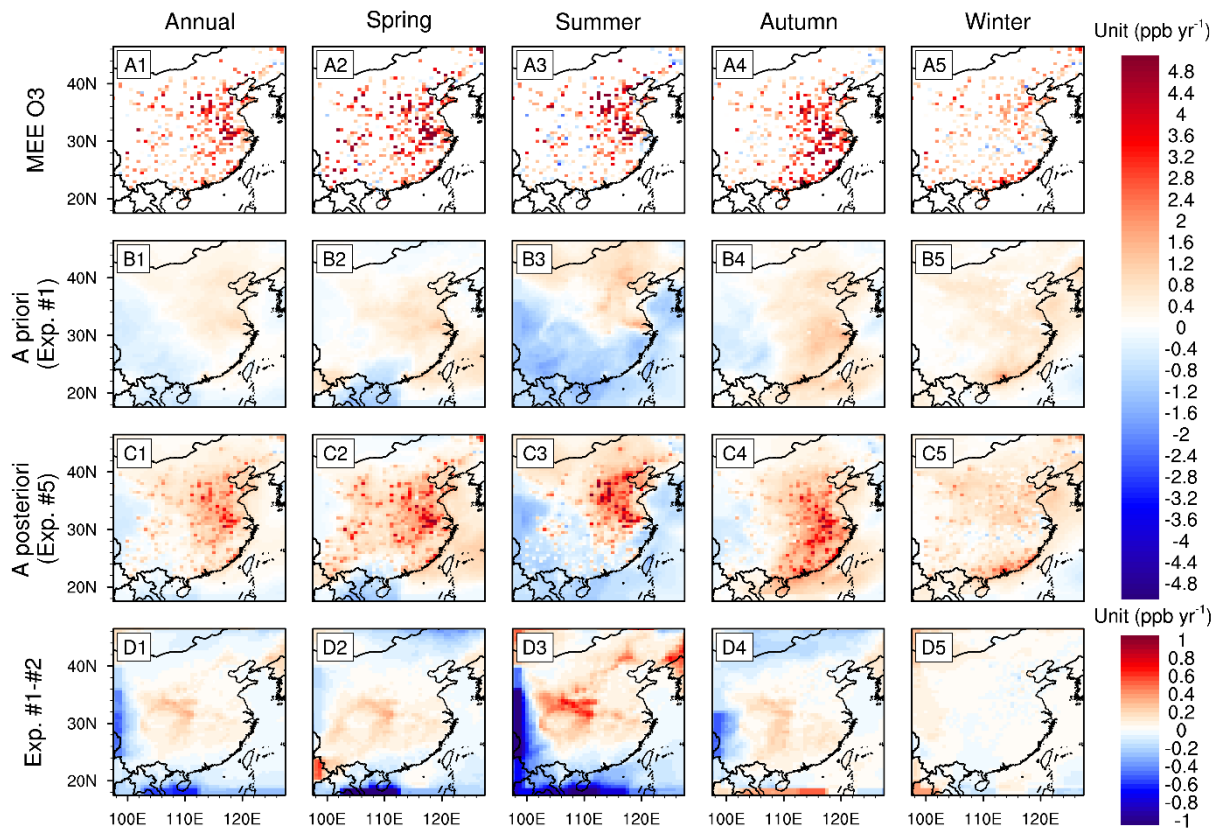


Fig. 6. Trends of surface MDA8 O₃ in 2015-2020 (annual and seasonal averages) from (A1-A5) MEE stations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem a posteriori simulation by assimilating MEE O₃ observations (Exp. #5). (D1-D5) Effects of interannual variabilities in background O₃ (Exp. #1 minus #2).

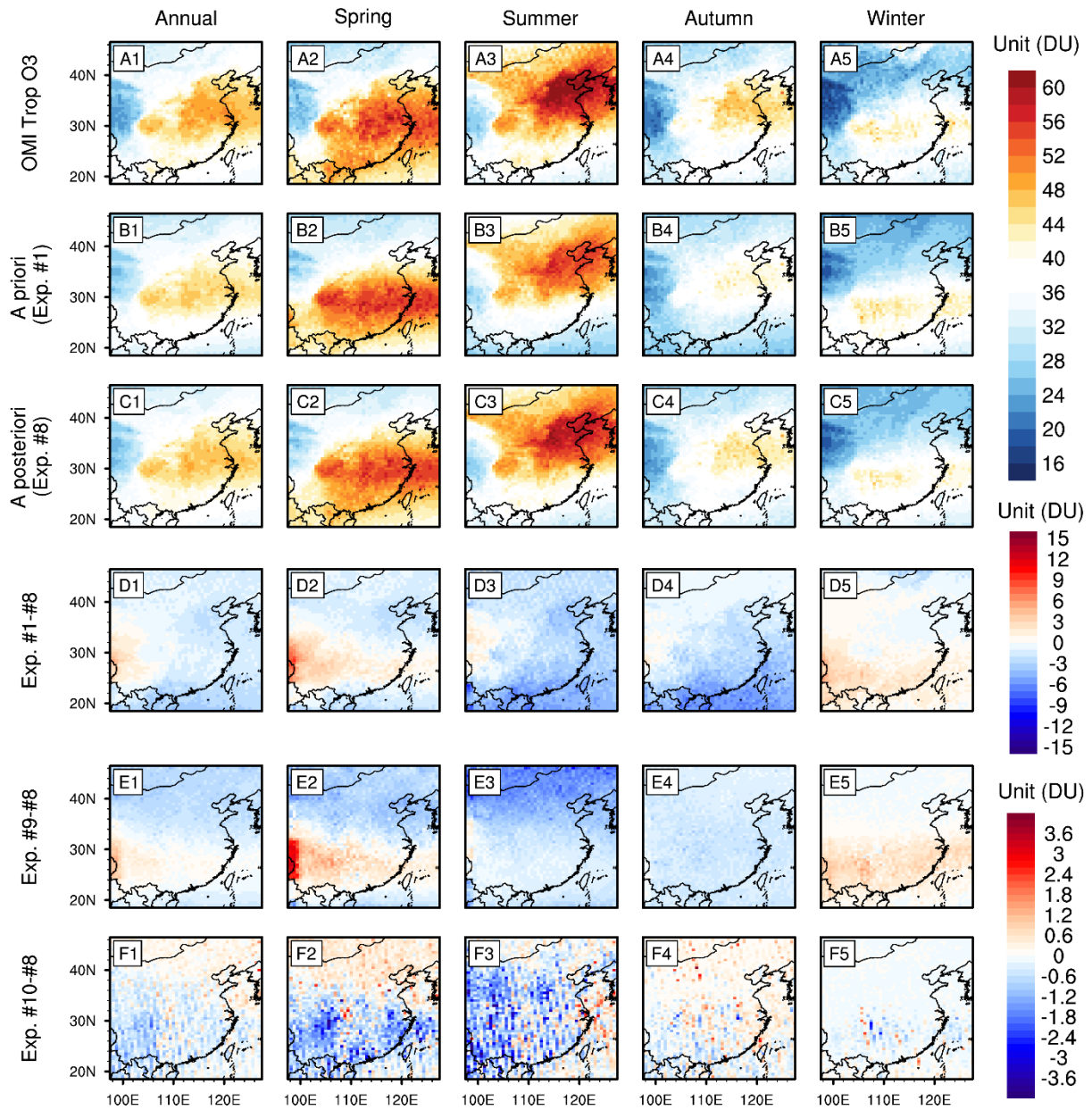


Fig. 7. Tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from (A1-A5) OMI observations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations (Exp. #8). (D1-D5) Bias in the a priori simulations (Exp. #1 minus #8). (E1-E5) Effects of optimization on regional O₃ background conditions (Exp. #9 minus #8); (F1-F5) Effects of the usage of row-isolated data (Exp. #10 minus #8). The output O₃ profiles are convolved with OMI averaging kernels.

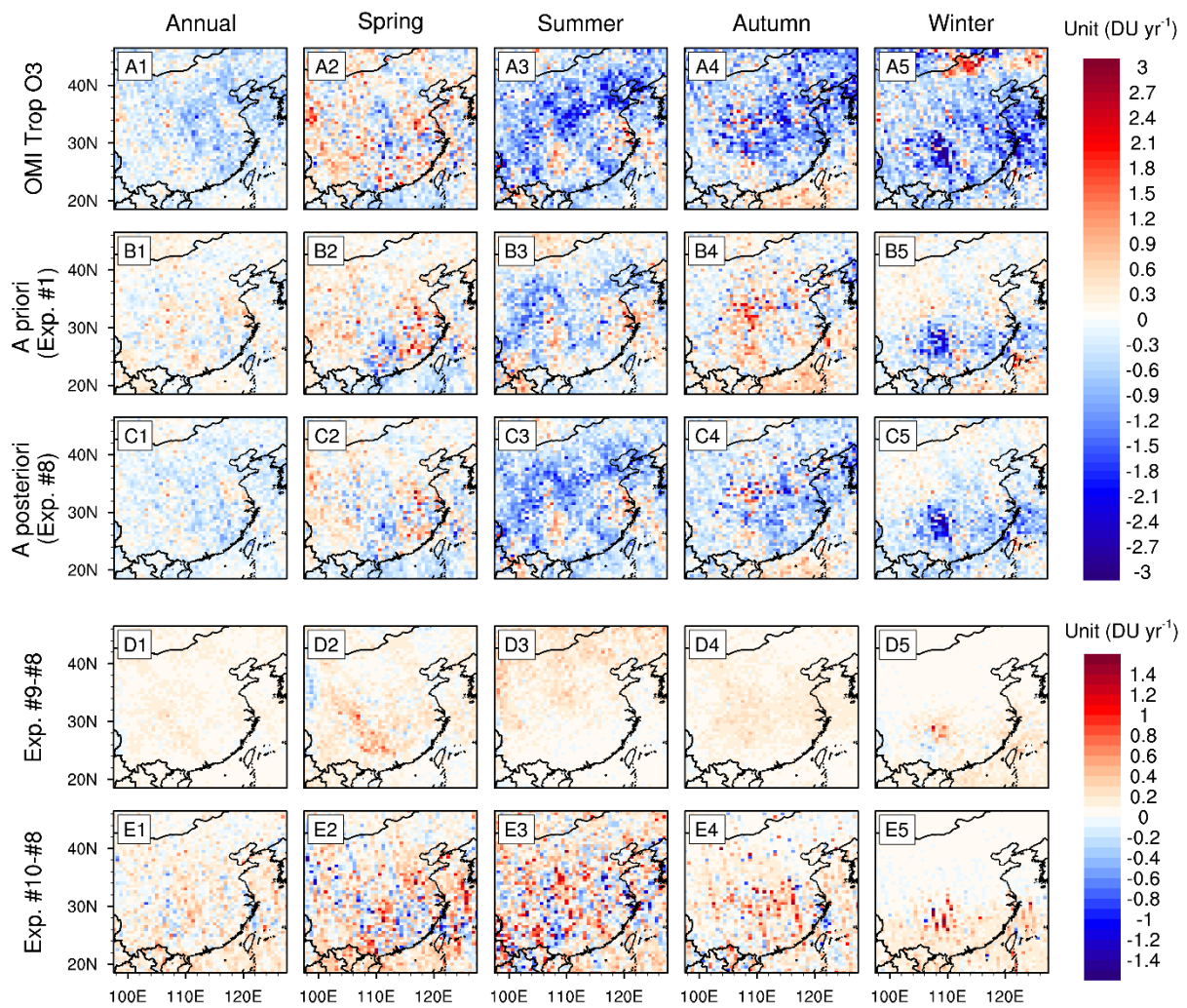


Fig. 8. Trends of tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from (A1-A5) OMI observations; (B1-B5) GEOS-Chem a priori simulation (Exp. #1); (C1-C5) GEOS-Chem a posteriori simulation by assimilating OMI O₃ observations (Exp. #8). (D1-D5) Effects of optimization on regional O₃ background conditions (Exp. #9 minus #8); (E1-E5) Effects of the usage of row-isolated data (Exp. #10 minus #8). The output O₃ profiles are convolved with OMI averaging kernels.

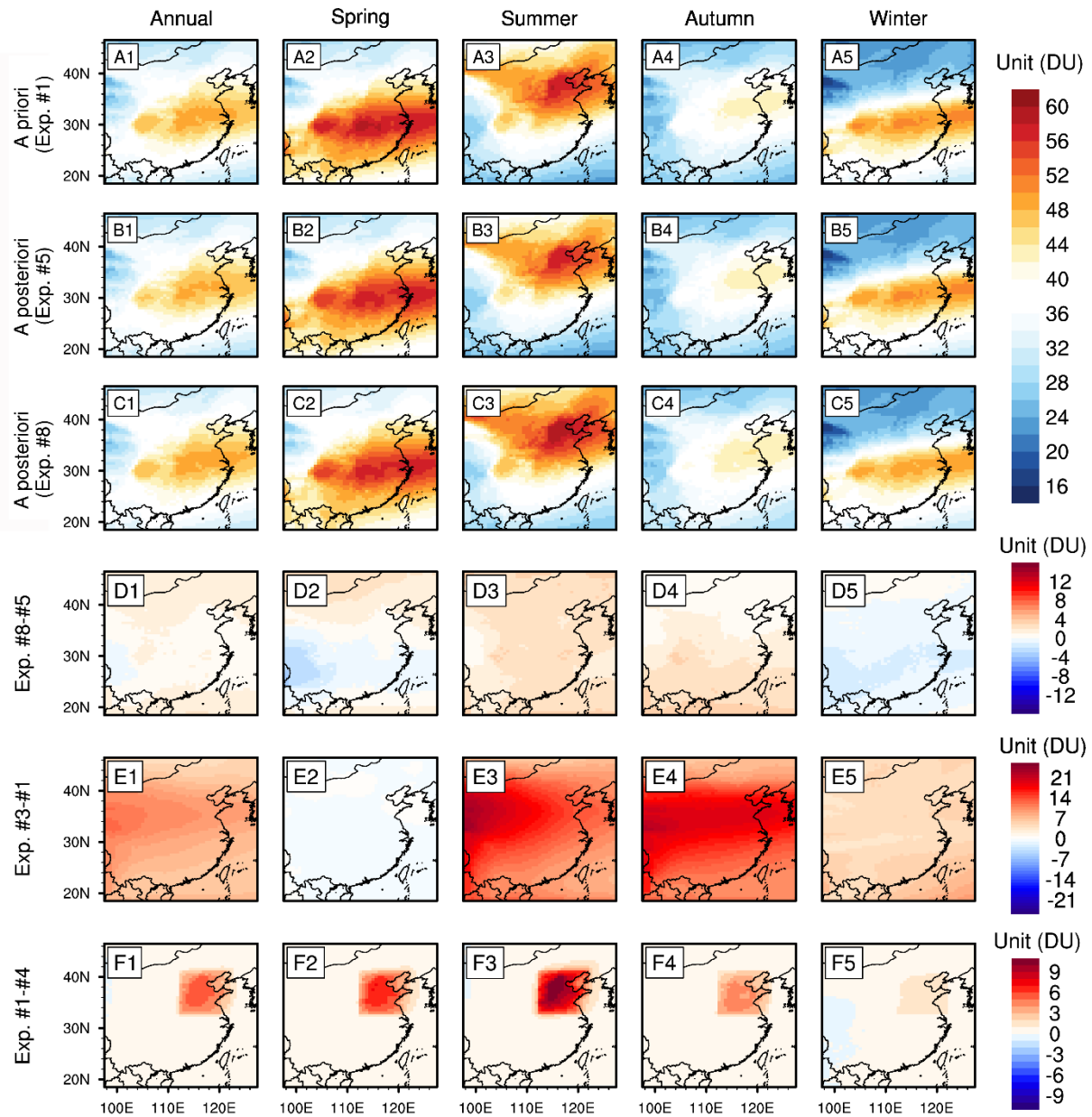


Fig. 9. Tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from (A1-A5) GEOS-Chem a priori simulation (Exp. #1); (B1-B5) Assimilations of MEE surface O₃ observations (Exp. #5); (C1-C5) Assimilations of OMI O₃ observations (Exp. #8). (D1-D5) Difference in tropospheric O₃ columns calculated by OMI-based assimilations minus MEE-based assimilations (Exp. #8 minus #5). (E1-E5) Effects of seasonal variabilities in background O₃ (Exp. #3 minus #1); (F1-F5) Effects of O₃ formation within the North China Plain PBL (Exp. #1 minus #4). The output O₃ profiles are NOT convolved with OMI averaging kernels.

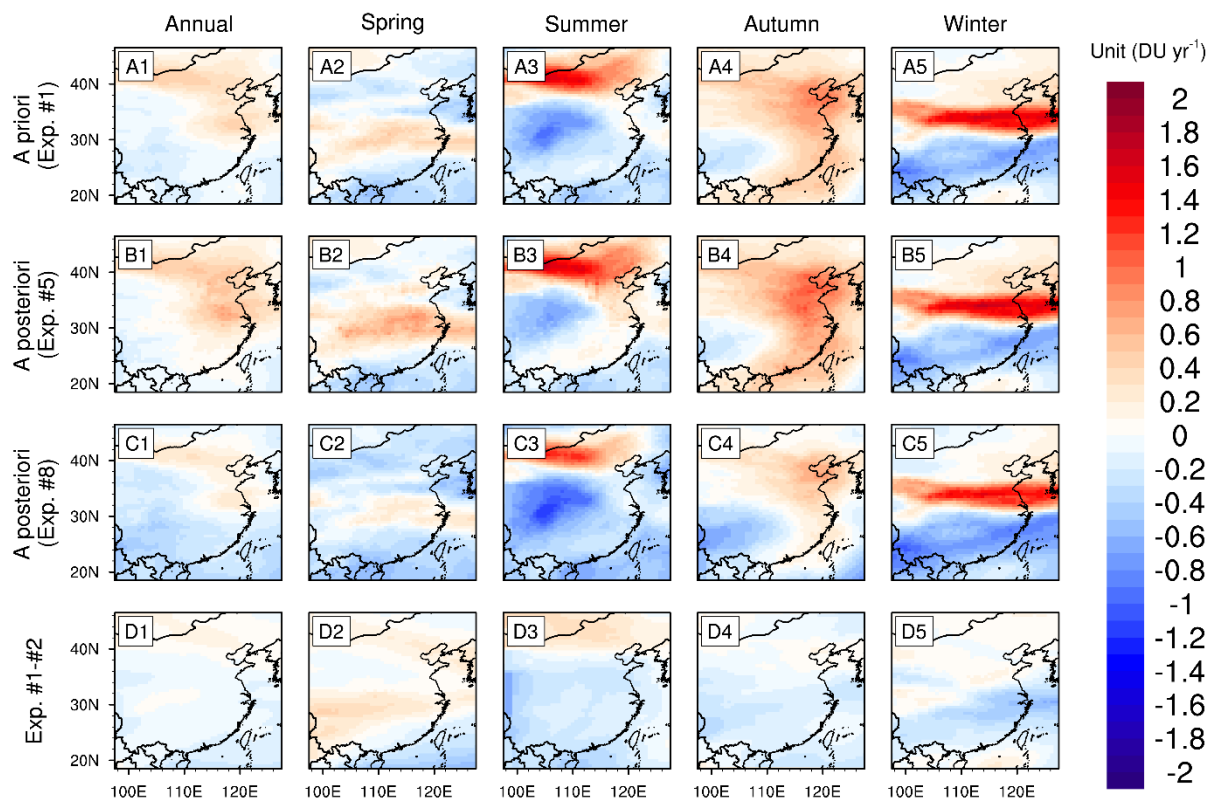


Fig. 10. Trends of tropospheric O₃ columns in 2015-2020 (annual and seasonal averages) from (A1-A5) GEOS-Chem a priori simulation (Exp. #1); (B1-B5) Assimilations of MEE surface O₃ observations (Exp. #5); (C1-C5) Assimilations of OMI O₃ observations (Exp. #8). (D1-D5) Effects of interannual variabilities in background O₃ (Exp. #1 minus #2). The output O₃ profiles are NOT convolved with OMI averaging kernels.