1    Impacts of the East Asian summer monsoon on interannual variations of

2    summertime surface-layer ozone concentrations over China

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25 **Abstract**

26     We apply a global three-dimensional Goddard Earth Observing System

27 (GEOS) chemical transport model (GEOS-Chem) driven by the NASA/GEOS-4

28 assimilated meteorological fields to quantify the impacts of the East Asian

29 summer monsoon (EASM) on interannual variations of June-July-August (JJA)

30 surface-layer $O_3$ concentrations over China. With anthropogenic emissions fixed

31 at year 2005 levels, model simulation for years 1986–2006 shows that the

32 changes in meteorological parameters alone lead to interannual variations in JJA

33 surface-layer $O_3$ concentrations by 2–5% over central eastern China, 1–3% in

34 northwestern China, and 5–10% over the Tibetan Plateau as well as the border

35 and coastal areas of South China, as the interannual variations are relative to the

36 average $O_3$ concentrations over the 21-yr period. Over 1986–2006, $O_3$

37 concentration averaged over the whole China is found to correlate positively with

38 the EASM index with a large correlation coefficient of +0.75, indicating that JJA $O_3$

39 concentrations are lower (or higher) in weaker (or stronger) EASM years. Relative

40 to JJA surface-layer $O_3$ concentrations in the strongest EASM years (1990, 1994,

41 1997, 2002, and 2006), $O_3$ levels in the weakest EASM years (1988, 1989, 1996,

42 1998, and 2003) are lower over almost whole China with a national mean lower

43 $O_3$ concentration by 2.0 ppbv (or 4%). Regionally, the largest percentage

44 differences in $O_3$ concentration between the weakest and strongest EASM years

45 are found to exceed 6% in northeastern China, southwestern China, and over the

46  Tibetan Plateau. Sensitivity studies show that the difference in transboundary

47  transport of $O_3$ is the most dominant factor that leads to lower $O_3$ concentrations

48  in the weakest EASM years than in the strongest EASM years, which, together

49  with the enhanced vertical convections in the weakest EASM years, explain about

50  80% of the differences in surface-layer $O_3$ concentrations between the weakest

51  and strongest EASM years. We also find that the impacts the EASM strength on

52  JJA surface-layer $O_3$ can be particularly strong (comparable in magnitude to the

53  impacts on $O_3$ by changes in anthropogenic emissions over 1986–2006) for

54  certain years. The largest increases in $O_3$ by anthropogenic emissions are

55  simulated over southeastern China, whereas the largest impacts of the EASM on

56  $O_3$ are found over central and western China.

## 1 Introduction

Tropospheric $O_3$ is an air pollutant harmful to human health and ecosystems (Shindell et al., 2012). It is also (after $CO_2$ and $CH_4$) the third most important anthropogenic greenhouse gas (Intergovernmental Panel on Climate Change (IPCC), 2007). High $O_3$ concentrations have been observed in many Chinese sites, with seasonal mean concentrations of 20–60 ppbv (Tang et al., 1995; Yan et al., 1997, 2003; Cheung and Wang, 2001; Zhu et al., 2004; Gao et al., 2005; Shao et al., 2006; H. Wang et al., 2005; Chou et al., 2006; Ding and Wang, 2006; T. Wang et al., 2006a, 2009; Takami et al., 2006; Tu et al., 2007; Ding et al., 2008; Lin et al., 2008; Xu et al., 2008; Yang et al., 2008; Zhang et al., 2008; Y. Wang et al., 2011) and episodic concentrations of exceeding 100 ppbv (T. Wang et al., 2006b; Z. Wang et al., 2006; Duan et al., 2008). Concentrations of $O_3$ are driven by a combination of precursor emissions and the regional meteorological conditions.

Meteorological parameters in summer in eastern China vary with the East Asian summer monsoon (EASM). The EASM prevails in May–September every year, with strong southerlies bringing clean, warm, and moist air from the oceans to eastern China and rain belts that stretch for thousands of kilometers in the west–east direction in eastern China (Tao and Chen, 1987; Wang and Ding, 2008). Previous observational and modeling studies have shown that such patterns of winds and precipitation of the EASM influence the seasonal variations

78  of $O_3$ in China (Chan et al., 1998; Li et al., 2007; He et al., 2008; Wang et al., 2011)

79  and in the west Pacific region (Pochanart et al., 2002; Tanimoto et al., 2005;

80  Yamaji et al., 2006). He et al. (2008) analyzed the seasonal variations of $O_3$

81  concentrations measured over 2004–2006 and found that $O_3$ concentrations peak

82  in spring and autumn with a summer trough in central eastern China and the west

83  Pacific, the areas that are influenced by clean air from the southern oceans during

84  the summer monsoon. Studies by Wang et al. (2008), Lin et al. (2009), and Zhao

85  et al. (2010) reported that the increasing clouds associated with the EASM rainfall

86  suppress photochemical production of $O_3$ by altering solar radiation, which also

87  contribute to the minimum $O_3$ concentrations in summer.

88      The strength of the EASM exhibits large interannual variations as a result of

89  the interactions between the atmosphere and oceans (Webster et al., 1998). No

90  previous studies, to our knowledge, have systematically examined the impacts of

91  the EASM on interannual variations of summertime $O_3$ in China. Recently, Zhou et

92  al. (2013) analyzed 2000–2010 ozonesonde data from Hong Kong and found a

93  close link between lower tropospheric $O_3$ and the East Asian monsoon on

94  interannual scales, but their analyses were focused on $O_3$ in Hong Kong in spring

95  and autumn. We present here a study to examine the impacts of the EASM on

96  interannual variations of summertime surface-layer $O_3$ concentrations over China,

97  based on 1986–2006 simulations of $O_3$ concentrations using the global chemical

98  transport model GEOS-Chem driven by the assimilated meteorological fields. This

99    work is a companion study to the work of Zhu et al. (2012), which investigated the

100   impacts of the EASM on interannual to decadal variations of summertime

101   aerosols in China.

102       The GEOS-Chem model and numerical experiments are described in Sect. 2.

103   Section 3 presents simulated interannual variations of summertime $O_3$ in China.

104   Section 4 shows simulated impacts of the EASM on interannual variations of

105   summertime $O_3$, and Sect. 5 examines the mechanisms through which the EASM

106   influences the interannual variations. Section 6 compares the impacts of changing

107   monsoon strength with those of changing anthropogenic emissions on $O_3$

108   concentrations in China.

109

110   **2    Model description and experimental design**

111   **2.1    GEOS-Chem model**

112   We simulate tropospheric $O_3$ using the global chemical transport model

113   GEOS-Chem (version 8.2.1, http://acmg.seas.harvard.edu/geos) driven by the

114   assimilated meteorological fields from the Goddard Earth Observing System

115   (GEOS) of the NASA Global Modeling and Assimilation Office (GMAO). The

116   version of the model used here has a horizontal resolution of 2° latitude by 2.5°

117   longitude and 30 vertical layers from the surface to 0.01 hPa. The GEOS-Chem

118   model includes a fully coupled treatment of tropospheric $O_3$-NOx-VOC chemistry

119   and aerosol components. Tropospheric $O_3$ is simulated with about 80 species and

120   over 300 chemical reactions (Bey et al., 2001). Photolysis rates are computed

121 using the Fast-J algorithm (Wild et al., 2000). The cross-tropopause $O_3$ flux in this

122 version of GEOS-Chem is specified with the synthetic ozone ("Synoz") method

123 (McLinden et al., 2000) as implemented by Bey et al. (2001), which includes a

124 passive, ozone-like tracer released into the stratosphere at a constant rate

125 equivalent to that of the prescribed cross-tropopause ozone flux of 499 Tg $O_3$ $yr^{-1}$.

126 **2.2    Emissions**

127 Global emissions of $O_3$ precursors, aerosol precursors, and aerosols in the

128 GEOS-Chem model follow Park et al. (2003, 2004), but anthropogenic emissions

129 of $NO_x$, CO, $SO_2$, and $NH_3$ over East Asia are overwritten by the emissions

130 inventory of Streets et al. (2003). Global anthropogenic emissions of nonmethane

131 hydrocarbons are from the GEIA inventory (Piccot et al., 1992). Biomass burning

132 emissions are taken from the GFED-2 inventory (van der Werf et al., 2006). These

133 inventories are then scaled for 2005 on the basis of economic data and energy

134 statistics as described by van Donkelaar et al. (2008). The biogenic emissions in

135 the GEOS-Chem model are simulated using the MEGAN module (Guenther et al.,

136 2006; Wiedinmyer et al., 2007). Soil $NO_x$ emissions are computed using a

137 modified version of the algorithm proposed by Yienger and Levy (1995). Lightning

138 emissions follow Price and Rind (1992), with the $NO_x$ vertical profile proposed by

139 Pickering et al. (1998).

140 The simulations of tropospheric $O_3$ by the GEOS-Chem model have been

141 evaluated in previous studies for the United States (Fiore et al., 2005; Wu et al.,

142 2008; H. Wang et al., 2009) and China (Y. Wang et al., 2008, 2011; Jeong and

143 Park, 2013; Lou et al., 2014). The model was found to be able to capture the

144 magnitude and spatial distribution of $O_3$ in China.

145 **2.3    Experiments**

146 In this study concentrations of $O_3$ in China for years 1986–2006 are simulated

147 using the GEOS-4 meteorological fields. To identify the key processes that

148 influence $O_3$ concentrations in different monsoon years, we perform the following

149 simulations:

150 (1)  O3_TOT: The standard simulation of $O_3$ concentrations for years 1986–2006.

151      Global anthropogenic and biomass burning emissions of $NO_x$, CO,

152      nonmethane hydrocarbons, aerosols and aerosol precursors are fixed at year

153      2005 levels. Meteorological fields are allowed to vary over 1986–2006. The

154      cross-tropopause $O_3$ flux is set to 499 Tg $yr^{-1}$ using the "Synoz" scheme.

155 (2)  O3_TB: The sensitivity simulation of $O_3$ concentrations for years 1986–2006

156      to quantify the role of transboundary transport of $O_3$ in different monsoon

157      years. The model setups are the same as those in O3_TOT except that all

158      natural and anthropogenic emissions in China are turned off.

159 (3)  O3_ST: The sensitivity simulation of $O_3$ concentrations for years 1986–2006

160      to quantify the impact of cross-tropopause $O_3$ flux on surface-layer $O_3$

161      concentrations. The model setups are the same as those in O3_TOT except

162      that natural and anthropogenic emissions are turned off globally.

163 (4)  O3_EMIS: The sensitivity simulations to compare the impacts of changing

164    anthropogenic emissions with those of changing monsoon strength. Two

165    sensitivity simulations are conducted with 1986 and 2006 anthropogenic

166    emissions, respectively. The emissions in year 1986 are simulated using the

167    default scaling factors in the model (van Donkelaar et al., 2008). Year 2006

168    meteorological parameters are used to drive both simulations. The

169    cross-tropopause $O_3$ flux is set to 499 Tg yr$^{-1}$ using the "Synoz" scheme.

170    **2.4    East Asian summer monsoon index**

171    The interannual variations in strength of the EASM are commonly represented by

172    the EASM index (EASMI). The EASMI introduced by Li and Zeng (2002) is used in

173    this study. The formulation for calculating EASMI based on the GEOS-4

174    meteorological parameters was given in Zhu et al. (2012). Positive values of

175    EASMI indicate strong monsoon years whereas negative values indicate weak

176    monsoon years. Physically, a strong summer monsoon in China is characterized

177    by strong southerlies extending from southern China to northern China, a deficit of

178    rainfall in the middle and lower reaches of the Yangtze River, and large rainfall in

179    northern China. On the contrary, in a weak summer monsoon year, China

180    experiences weak southerlies, large rainfall in southern China, and a deficit of

181    rainfall in northern China. The movement of the rain belts is associated with the

182    strength of the southerlies.

183

184    **3    Simulated interannul variations of summertime $O_3$ in China**

185    Figure 1a shows the simulated spatial distribution of June-July-August (JJA)

186    surface-layer $O_3$ concentrations averaged over 1986–2006 from the $O_3\_TOT$

187    simulation. Over eastern China, simulated $O_3$ concentrations are 50–65 ppbv in

188    northern China and 25–50 ppbv in southern China. Such pattern of higher $O_3$

189    concentrations in northern China than in southern China results from that the

190    strong southerlies bring clean air from the oceans to southern China during the

191    summer monsoon season (Chan et al., 1998; Yamaji et al., 2006; Li et al., 2007;

192    He et al., 2008; Wang et al., 2011). In western China, simulated $O_3$ exhibits

193    maximum concentrations of 65 ppbv. Our simulated surface-layer concentrations

194    of $O_3$ are high in both western and eastern China, which agree with the modeling

195    studies of Wang et al. (2011), Jeong and Park (2013), and Lou et al. (2014). The

196    high $O_3$ concentrations in western China are associated with the downward

197    transport of $O_3$ from the stratosphere to troposphere (Wild and Akimoto, 2001; T.

198    Wang et al. 2006a; Ding and Wang, 2006), and the high $O_3$ concentrations in

199    eastern China result from anthropogenic emissions (Wang et al., 2011).

200        The interannual variations in simulated JJA $O_3$ concentrations can be

201    quantified by mean absolute deviation (MAD) and absolute percent departure

202    from the mean (APDM) defined as $\mathrm{MAD} = \frac{1}{n}\sum_{i=1}^{n}\left|P_i - \frac{1}{n}\sum_{i=1}^{n}P_i\right|$ and $\mathrm{APDM} =$

203    $100\% \times \mathrm{MAD}/\left(\frac{1}{n}\sum_{i=1}^{n}P_i\right)$, where $P_i$ is the simulated JJA mean $O_3$ concentration of

204    year $i$, and $n$ is the number of years examined ($n$=21 for years 1986–2006).

205    Therefore MAD represents the absolute interannual variation and APDM

206  represents the interannual variation relative to the average of $O_3$ concentration

207  over the $n$ years. The MAD values of JJA surface-layer $O_3$ concentrations (Fig.

208  1b) are 1.0–4.0 ppbv in China, with the largest values exceeding 2 ppbv found

209  over northeastern China, coastal areas of South China, and the Tibetan Plateau.

210  As shown in Fig. 1c, the APDM values are in the range of 2–5% over central

211  eastern China where summer monsoon prevails, 1–3% in northwestern China,

212  and 5–10% over the Tibetan Plateau as well as the border and coastal areas of

213  South China. These interannual variations in $O_3$ are significant as compared to

214  the impacts of reductions in emissions of $O_3$ precursors. Han et al. (2005) showed

215  by modeling studies that, in eastern China, reductions in $NO_x$ or total VOCs

216  (anthropogenic plus biogenic VOCs) by 50% lead to changes in JJA $O_3$

217  concentrations by 10–20%.

218    The GEOS-Chem simulations of present-day surface-layer $O_3$ concentrations

219  in China have been evaluated in Wang et al. (2011) and Lou et al. (2014). Wang

220  et al. (2011) demonstrated that the model captured well the magnitude and

221  seasonal variation of surface-layer concentrations and column burdens of $O_3$ in

222  China. Lou et al. (2014) reported that the simulated $O_3$ in China agreed fairly well

223  with measurements collected from the literature with an average high bias of 9%.

224  Because of the lack of long-term $O_3$ measurements in China, we evaluate the

225  simulated interannual variations of JJA surface-layer $O_3$ concentrations at two

226  sites (Fig. 2): Hok Tsui in Hong Kong (22°13' N, 114°15' E) and Ryori in Japan

227  (39°03'N, 144°82'E). The measurements at Hok Tsui are taken from T. Wang et al.

228  (2009), and those at Ryori site are from the WMO World Data Center for

229  Greenhouse Gases (WDCGG, http:// http://ds.data.jma.go.jp/gmd/wdcgg/). At Hok

230  Tsui, simulated $O_3$ concentrations are higher than the observations in JJA. This

231  discrepancy may due to the model's overestimate of $O_3$ in marine boundary layers

232  in summer (Liu et al., 2006). Interannually, the model captures well the peaks and

233  troughs of the observed JJA $O_3$ concentrations, with a high correlation coefficient

234  of +0.87. The model underestimates JJA $O_3$ concentrations at Ryori, probably due

235  to the uncertainties with local emissions, but captures mostly the years with

236  maximum or minimum $O_3$ levels with a correlation coefficient of +0.47. The

237  simulated APDM values at Hok Tsui and Ryori are both 7%, smaller than the

238  observed interannual variations of 22% and 8% at these two sites, respectively,

239  which can be attributed to the fixed anthropogenic emissions of $O_3$ precursors in

240  our O3_TOT simulation.

241

242  **4  Impacts of the EASM on interannual variations of summertime $O_3$ in**

243     **China**

244  Simulated JJA surface-layer $O_3$ concentration averaged over the whole China

245  (defined by the national borders of China) is shown in Fig. 3a for years 1986–2006

246  based on the O3_TOT simulation. Summertime $O_3$ concentrations show large

247  interannual variations, with high (or low) $O_3$ concentrations in strong (or weak)

248     monsoon years. Concentrations of $O_3$ are found to correlate positively with the

249     EASMI; the correlation coefficient is +0.75 and is statistically significant at the

250     95th percentile. The spatial distribution of the correlation coefficients between the

251     EASMI and $O_3$ concentrations from the $O_3$_TOT simulation is presented in Fig. 3b.

252     Positive correlations are found in almost the whole China, and large positive

253     correlation coefficients that exceed +0.5 are found over the region between 90°E

254     and 110°E. Since anthropogenic emissions of $O_3$ precursors are kept unchanged

255     in the simulation for 1986–2006, these strong positive correlations demonstrate

256     that the EASM strength has large impacts on JJA $O_3$ concentrations over China.

257         In order to quantify the impacts of the EASM on $O_3$ concentrations over China,

258     we show in Figs. 4a and 4b, respectively, the absolute and percentage differences

259     between $O_3$ concentrations averaged over five weakest EASM years (1988, 1989,

260     1996, 1998, and 2003) and those averaged over five strongest EASM years (1990,

261     1994, 1997, 2002, and 2006). These weakest (or strongest) monsoon years are

262     selected within 1986–2006 based on the five largest negative (or positive) values

263     of the normalized EASMI as shown in Fig. 3a. Relative to the concentrations in the

264     strongest monsoon years, $O_3$ levels in the weakest monsoon years are lower over

265     almost the whole China, with the largest reductions of exceeding 3 ppbv (or 6%) in

266     northeastern and southwestern China and over or near the Tibetan Plateau.

267     Concentrations of $O_3$ in the weakest monsoon years are simulated to be higher

268     than those in the strongest monsoon years by 3–5 ppbv (or 6–15%) over the East

269 China Sea. The pattern of the differences in $O_3$ concentrations agrees with the

270 spatial distribution of the correlation coefficients (Fig. 3b). Averaged over China,

271 $O_3$ level in the weakest monsoon years is lower than that in the strongest

272 monsoon years by 2.0 ppbv (or 4%). Note that the monsoon region covers almost

273 the whole China except for the northwestern China (Gao, 1962; An et al., 2000).

274 Our simulated monsoon-induced changes in $O_3$ in China (Fig. 4) are mostly within

275 the monsoon region. The changes in $O_3$ over the Siberia are large, which can be

276 explained by the anomalous northerlies over the north border of China between

277 the five weakest and strongest EASM years (weakest–strongest) (Fig. 5) that

278 transport $O_3$ to China.

279     Figures 4c and 4d are pressure-longitude plots of the differences in $O_3$

280 averaged over the latitude range of 20°–46°N. From the surface to 250 hPa

281 altitude, $O_3$ levels in the weakest monsoon years are lower by up to 5 ppbv (or 8%)

282 over 80°–115°E and are higher by 1–3 ppbv (or 3–6%) over 120°–135°E relative

283 to the concentrations in the strongest monsoon years. Concentrations of $O_3$ at

284 130–250 hPa altitudes exhibit increases in the weakest monsoon years, with

285 maximum increases of 4–7 ppbv (3–6%) over 80°E and 130°E, as a result of the

286 anomalous horizontal convergence at these layers.

287

288 **5   Mechanisms of the impacts of the EASM on summertime $O_3$**

289 **5.1   Impacts of the EASM on transboundary transport of $O_3$**

290   Considering that $O_3$ concentrations in almost whole China are lower in the

291   weakest monsoon years than in the strongest monsoon years and that

292   tropospheric $O_3$ has a relatively long lifetime of 3–4 weeks (Seinfeld and Pandis,

293   2006), we examine firstly the impacts of the EASM on transboundary transport of

294   $O_3$. Figure 5 shows JJA horizontal winds at 850 hPa and 500 hPa averaged over

295   1986–2006 as well as the composite differences in JJA horizontal winds between

296   the five weakest and five strongest EASM years at these two layers. The typical

297   features in winds during the EASM can be seen in Figs. 5a and 5b. The

298   southerlies prevail in southeastern China in the lower and middle troposphere in

299   JJA. Figs. 5c and 5d also present the differences in winds between the weakest

300   and strongest EASM years (weakest minus strongest). In JJA, anomalous

301   southerlies are found in southern China and anomalous westerlies are found in

302   southeastern China, as the winds in the five weakest monsoon years are

303   compared with those in the five strongest monsoon years. Such differences in

304   winds between weak and strong EASM years were also reported in Li and Zeng

305   (2002) and Huang (2004).

306   The differences in winds in different monsoon years lead to differences in

307   transboundary transport of $O_3$. We show in Fig. 6 the differences in simulated

308   horizontal mass fluxes of $O_3$ at the four lateral boundaries of the selected box of

309   (85°–120°E, 20°–46°N, the surface to 250 hPa) and in Table 1 the summary of the

310   composite analysis on fluxes of $O_3$ in and out of this box, based on simulation

311    O3_TOT. This box is selected to capture the features of transboundary $O_3$

312    transport that influence $O_3$ concentrations in China. The location of the box is

313    shown in Fig. 5d. Relative to the strongest monsoon years, the weakest monsoon

314    years have less inflow by 0.1 Tg season$^{-1}$ at the west boundary, larger inflow

315    fluxes of $O_3$ by 4.2 Tg season$^{-1}$ at the south boundary and by 5.5 Tg season$^{-1}$ at

316    the north boundary, and larger outflow by 12.9 Tg season$^{-1}$ at the east boundary

317    (Table 1 and Fig. 6), as mass fluxes are summed over JJA. The net effect is a

318    larger transboundary outflow of $O_3$ by 3.3 Tg season$^{-1}$ in the weakest monsoon

319    years than in the strongest monsoon years. The anomalous westerlies in

320    southeastern China are especially important, which bring polluted air with high $O_3$

321    concentrations to the coastal areas and the East China Sea (Fig. 6d), leading to

322    reductions in $O_3$ concentration in China.

323        The role of changes in transboundary transport of $O_3$ can be further quantified

324    by the simulation O3_TB with natural and anthropogenic emissions of $O_3$

325    precursors in China turned off. The spatial distribution of $O_3$ from simulation

326    O3_TB (referred to as $TBO_3$ hereafter) is presented in Fig. 7a. $TBO_3$

327    concentrations show a distinct spatial gradient over China, decreasing from about

328    55 ppbv in northwestern China to about 10 ppbv in southeastern China. The JJA

329    $TBO_3$ concentration averaged over the whole China is 30 ppbv, which is about 62 %

330    of the average concentration simulated in O3_TOT, indicating that a significant

331    fraction of surface-layer $O_3$ over China is from transboundary transport. The

332  differences in TBO$_3$ concentrations between the weakest and strongest monsoon

333  years (Fig. 7d) are, to a large extent, similar to those from simulation O3_TOT (Fig.

334  4a), in terms of both distributions and magnitudes. Averaged over China, the

335  difference in TBO$_3$ between the weakest and strongest monsoon years is −1.6

336  ppbv, which accounts for 80% of the corresponding difference obtained in

337  O3_TOT. The JJA mass fluxes of TBO3 for the selected box of (85°–120°E, 20°–

338  46°N, the surface to 250hPa) are also similar to those simulated in O3_TOT

339  (Table 1), with a net horizontal outflow of 2.8 Tg season$^{-1}$. These model results

340  indicate that the differences in transboundary transport of O$_3$ is a dominant

341  mechanism through which the EASM influences interannual variations of JJA O$_3$

342  concentrations in China.

343  **5.2    Impacts of the EASM on vertical transport of O$_3$**

344  Vertical circulations in China have some unique features during the EASM. In

345  summer, two major ascending branches of winds (or strong convections) are

346  observed throughout the entire troposphere (Fig. 8a). One branch is located over

347  the Yangtze River valley, associated with the Mei-yu front (rain belt) of the EASM

348  (Ding and Chan, 2005). The other branch is over the Tibetan Plateau, because

349  the Tibetan Plateau during summer serves as a large heat source, which uplifts

350  heated air to the upper troposphere and even to the stratosphere (Ye, 1981). Fig.

351  8a shows JJA vertical velocity and simulated vertical mass flux of O$_3$ at 500 hPa

352  averaged over 1986–2006. Since vertical velocity is not available from the

17

353 reanalyzed GEOS-4 meteorological fields, vertical winds presented here are from

354 the NCEP/NACR reanalysis data. The simulated mass fluxes are obtained in

355 O3_TOT. The largest vertical mass fluxes of $O_3$ are simulated to occur over

356 northern China and western China where surface-layer $O_3$ concentrations are

357 high, and to occur over the Tibetan Plateau and its surrounding areas as a result

358 of the strong convections here.

359     Figure 8b shows the differences in JJA vertical wind velocity and simulated

360 vertical mass flux of $O_3$ at 500 hPa between the weakest and strongest monsoon

361 years. Relative to the strongest monsoon years, anomalous convections are

362 found over central and western China in the weakest monsoon years, leading to

363 enhanced upward transport of $O_3$ from the surface to upper troposphere at these

364 locations. The differences in vertical winds shown in Fig. 8b agree with those

365 reported in Huang (2004).

366     Table 1 also summarizes the composite analysis on vertical fluxes of $O_3$

367 through the top side of the selected box (85°–120°E, 20°–46°N, the surface to

368 250hPa). In simulation O3_TOT, the upward flux of $O_3$ through the top plane of

369 the box in the weakest monsoon years is larger than that in the strongest

370 monsoon years by 1.4 Tg season$^{-1}$. This anomalous vertical outflow of 1.4 Tg

371 season$^{-1}$ is smaller than the anomalous horizontal transboundary outflow of 3.3

372 Tg season$^{-1}$ (Section 5.1), indicating that the differences in vertical transport of $O_3$

373 also contribute to lower JJA $O_3$ concentrations in China in the weakest monsoon

374  years than in the strongest monsoon years, but the impact of the differences in

375  vertical transport is smaller than that of the differences in transboundary transport

376  of $O_3$.

377  **5.3   Impact   of   cross-tropopause   transport   on   surface-layer   $O_3$**

378  **concentrations**

379  The cross-tropopause transport of $O_3$ from the stratosphere is an important

380  source of tropospheric $O_3$. Simulation $O_3\_ST$ is performed to quantify the impact

381  of cross-tropopause transport on JJA surface-layer $O_3$ concentrations. The

382  simulated JJA concentrations of $O_3$ in simulation $O_3\_ST$ (referred to as $STO_3$) at

383  the surface-layer averaged over 1986–2006 are presented in Fig. 7b. $STO_3$

384  shows maximum concentrations of 15–20 ppbv over northwestern China,

385  contributing to the high $O_3$ concentrations in western China simulated in O3_TOT.

386  Averaged over China, the surface-layer concentration of JJA $STO_3$ is 8 ppbv,

387  which is about 17% of that of JJA $O_3$ simulated in O3_TOT. Concentrations of

388  $STO_3$ increase with altitude; the ratio of $STO_3$ to $O_3$ simulated in O3_TOT is about

389  50% at 200 hPa and about 80% at 100 hPa. Figure 7e shows the differences in

390  surface-layer $STO_3$ concentrations between the weakest and strongest monsoon

391  years.   Relative   to   the   strongest   monsoon   years,   surface-layer   $STO_3$

392  concentrations are lower by 1–2 ppbv over central China and the Tibetan Plateau

393  in the weakest monsoon years. Averaged over China, the difference in $STO_3$

394  between the weakest and strongest monsoon years is −0.45 ppbv, which only

395    accounts for 23% of the corresponding difference in TOTO$_3$ and hence indicates

396    that the variations in O$_3$ transported from the stratosphere in different monsoon

397    years are not the major factors that drive the interannual variations of JJA

398    surface-layer O$_3$ concentrations in China. Note that the cross-tropopause O$_3$ flux

399    in O3_TOT and O3_ST is specified with the synthetic ozone ("Synoz") method

400    (McLinden et al., 2000) with a constant cross-tropopause ozone flux of 499 Tg O$_3$

401    yr$^{-1}$. The latest version of the GEOS-Chem model also has an option of using the

402    linearized ozone ("Linoz") parameterization scheme of McLinden et al. (2000) to

403    represent O$_3$ in the stratosphere, in which the ozone vertical profiles across the

404    tropopause are relaxed back toward climatological profiles and hence

405    cross-tropopause O$_3$ flux varies with time step and location. We have tested using

406    "Linoz" instead of "Synoz" and found that these two schemes obtain same

407    conclusion about the impact of cross-tropopause O$_3$ on JJA surface-layer O$_3$ in

408    China.

409    **5.4    Impacts of the EASM on local chemical production of O$_3$**

410    The differences in O$_3$ concentrations between O$_3$_TOT and O$_3$_TB simulations

411    are attributed to the enhancement of O$_3$ due to Chinese emissions, referred to as

412    Chinese local O$_3$ (LOCO$_3$). Figure 7c shows that high concentrations of LOCO$_3$

413    are located in eastern China where the emissions of O$_3$ precursors are large. The

414    maximum LOCO$_3$ concentrations reach 40–45 ppbv. The LOCO$_3$ concentration

415    averaged over the whole China is 18 ppbv, which is about 38% of the value

416    simulated in O3_TOT. Figure 7f presents the differences in simulated $LOCO_3$

417    between the weakest and strongest EASM years. Relative to the strongest

418    monsoon years, $LOCO_3$ concentrations in the weakest monsoon years are lower

419    by 2–5 ppbv over southern China and slightly higher by up to 1 ppbv over central

420    China. Averaged over China, the difference in $LOCO_3$ between the weakest and

421    strongest monsoon years is −0.4 ppbv, which accounts for 20% of the

422    corresponding difference in $TOTO_3$ and hence reflects the small impacts of

423    monsoon strength on local chemical production of $O_3$.

424        The small impacts of monsoon strength on local chemical production of $O_3$

425    can be further justified by examining the net chemical production of $O_3$ within the

426    selected box (85°–120°E, 20°–46°N, the surface to 250hPa). Sum over the

427    selected box, the net chemical production (chemical production − chemical loss)

428    averaged over the weakest monsoon years is 39.0 Tg $season^{-1}$ in JJA, which is

429    smaller than that averaged over the strongest monsoon years by 0.5%.

430

431    **6    Comparison of the impact of monsoon strength with the role of**

432        **changing emissions**

433    We perform simulations $O_3$_EMIS to compare the impact of changing monsoon

434    strength (Section 4) with that of changing anthropogenic emissions on JJA $O_3$

435    concentrations. Two sensitivity simulations are conducted with 1986 and 2006

436    anthropogenic emissions, respectively, and year 2006 meteorological parameters

437    are used to drive both simulations. Relative to 1986, year 2006 emissions of $NO_x$,

438  CO, and NMVOCs in China increase by 111%, 56%, and 9%, respectively,

439  leading to increases in JJA surface-layer $O_3$ by 9–15 ppbv in southeastern China

440  (Fig. 9). Note the locations of large increases in $O_3$ here are different from those of

441  large differences in $O_3$ between the weakest and strongest monsoon years (Fig.

442  4b). Averaged over China, the change in JJA surface-layer $O_3$ concentration

443  owing to changes in emissions over 1986–2006 is +5.3 ppbv, which is larger than

444  the difference in JJA surface-layer $O_3$ of 2.0 ppbv between the selected weakest

445  and strongest monsoon years. However, the difference in surface-layer $O_3$

446  between the weak monsoon year 1998 and the strong monsoon year 1997 is −4.2

447  ppbv, indicating that the impacts of the EASM on JJA $O_3$ can be particularly strong

448  for certain years.

449

450  **7  Conclusions**

451  We examine the impacts of the East Asian summer monsoon (EASM) on

452  interannual variations of summertime surface-layer $O_3$ concentrations over China

453  using the GEOS-Chem model driven by the assimilated GEOS-4 meteorological

454  data. The interannual variations of $O_3$ concentrations are quantified by values of

455  mean absolute deviation (MAD) and absolute percent departure from the mean

456  (APDM), based on the simulation of $O_3$ for years 1986–2006 with changes in

457  meteorological parameters but fixed anthropogenic emissions at year 2005 levels.

458  The MAD values of JJA surface-layer $O_3$ concentrations in China are in the range

459  of 1.0–4.0 ppbv, with the largest values exceeding 2 ppbv found over

460  northeastern China, coastal areas of South China, and the Tibetan Plateau. The

APDM values of JJA surface-layer $O_3$ concentrations are 2–5% over central eastern China, 1–3% in northwestern China, and 5–10% over the Tibetan Plateau as well as the border and coastal areas of South China.

With fixed anthropogenic emissions, simulated JJA $O_3$ concentrations averaged over China exhibit strong positive correlation (with a correlation coefficient of +0.75) with the East Asian summer monsoon index (EASMI) in the time period of 1986–2006, indicating that JJA $O_3$ concentrations are lower (or higher) in weaker (or stronger) EASM years. Relative to JJA surface-layer $O_3$ concentrations in the strongest EASM years (1990, 1994, 1997, 2002, and 2006), $O_3$ levels in the weakest EASM years (1988, 1989, 1996, 1998, and 2003) are lower over almost whole China with the national mean $O_3$ concentration lower by 2.0 ppbv (or 4%).

Sensitivity studies are performed to identify the key processes through which the variations in EASM strength influence interannual variations of JJA $O_3$ in China. The difference in transboundary transport of $O_3$ is found to be the most dominant factor that leads to lower $O_3$ concentrations in the weakest EASM years than in the strongest EASM years. Relative to the strongest EASM years, the weakest EASM years have less inflow by 0.11 Tg season$^{-1}$ at the west boundary, larger inflow fluxes of $O_3$ by 4.2 Tg season$^{-1}$ at the south boundary and by 5.5 Tg season$^{-1}$ at the north boundary, and larger outflow by 12.9 Tg season$^{-1}$ at the east boundary, as horizontal mass fluxes of $O_3$ at the four lateral boundaries of the selected box (85°–120°E, 20°–46°N, the surface to 250 hPa) are calculated. As a

23

483 result, the weakest EASM years have larger outflow of $O_3$ than the strongest

484 EASM years, which, together with the enhanced vertical convections in the

485 weakest EASM years, explain about 80% of the differences in surface-layer $O_3$

486 concentrations between the weakest and strongest EASM years.

487     We also perform a sensitivity simulation $O_3\_EMIS$ to compare the impact of

488 changing monsoon strength with that of changing anthropogenic emissions on

489 JJA $O_3$ concentrations. Averaged over China, the change in JJA surface-layer $O_3$

490 concentration owing to changes in emissions over 1986–2006 is +5.3 ppbv, which

491 is larger than the difference in JJA surface-layer $O_3$ of 2.0 ppbv between the

492 selected weakest and strongest monsoon years. However, the difference in

493 surface-layer $O_3$ between the weak monsoon year 1998 and the strong monsoon

494 year 1997 is −4.2 ppbv, indicating that the impacts of the EASM on JJA $O_3$ can be

495 particularly strong for certain years. Note that while the largest increases in $O_3$ by

496 anthropogenic emissions are located over southeastern China, the largest

497 impacts of EASM on $O_3$ are found over central and western China.

498

504　　Administration special funding in atmospheric science GYHY200906020.

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763 **Table 1.** The composite analyses of horizontal and vertical fluxes of $O_3$ (Tg
764 season$^{-1}$) for the selected box of (85°–120°E, 20°–46°N, the surface to 250 hPa)
765 based on simulations O3_TOT and O3_TB. The values are averaged over the five
766 weakest (1988, 1989, 1996, 1998, and 2003) and five strongest monsoon years
767 (1990, 1994, 1997, 2002, and 2006), and the differences are calculated as
768 (weakest-strongest). For horizontal fluxes, positive values indicate eastward or
769 northward transport and negative values indicate westward or southward
770 transport. For vertical fluxes, positive values indicate upward transport.

| Side of the selected box | O3_TOT | | | O3_TB | | |
|---|---|---|---|---|---|---|
| | Weakest | Strongest | Difference | Weakest | Strongest | Difference |
| Horizontal mass fluxes | | | | | | |
| West | +83.7 | +83.8 | −0.1 | +78.8 | +78.9 | −0.1 |
| East | +106.5 | +93.5 | +12.9 | +84.1 | +73.1 | +11.0 |
| South | +13.4 | +9.2 | +4.2 | +13.8 | +10.5 | +3.3 |
| North | −19.3 | −13.7 | −5.5 | −20.0 | −15.0 | −5.0 |
| Vertical mass fluxes | | | | | | |
| Top | +22.1 | +20.7 | +1.4 | +16.5 | +15.5 | +1.0 |

771

**Figure Captions**

**Fig. 1.** (a) Simulated JJA surface-layer $O_3$ concentrations (ppbv) that are averaged over years 1986–2006 of simulation O3_TOT. (b) Mean absolute deviation (MAD, ppbv) and (c) absolute percent departure from the mean (APDM, %) calculated with 1986–2006 simulation of $O_3$ in O3_TOT.

**Fig. 2.** Comparisons of observed and simulated JJA mean surface-layer $O_3$ concentrations at Hok Tsui (22°13' N, 114°15' E) in Hong Kong (left) and Ryori (39°03'N, 144°82'E) in Japan (right). Correlation coefficient between simulations and observations is shown at top right corner of each panel, which is calculated over the time period with observations available.

**Fig. 3.** (a) The normalized time series of EASMI (blue bars, left y-axis) and the simulated JJA surface-layer $O_3$ concentrations (black line, right y-axis, ppbv) averaged over China for years of 1986–2006. (b) Spatial distribution of the correlation coefficients between the EASMI and the JJA surface-layer $O_3$ concentrations. The dotted areas indicate statistical significance with 95% confidence from a two-tailed Student's t-test. The EASMI are calculated using the GEOS-4 assimilated meteorological data.

**Fig. 4.** Horizontal distributions of (a) absolute and (b) percentage differences in JJA surface-layer $O_3$ concentrations between the five weakest and strongest EASM years (weakest–strongest). Pressure-longitude cross sections averaged over 20°–46°N for (c) absolute and (d) percentage differences in JJA $O_3$ concentrations between the five weakest and strongest EASM years (weakest–strongest). Results are from simulation O3_TOT and unit is shown on top of each panel.

**Fig. 5.** The JJA mean horizontal winds at (a) 850 hPa and (b) 500 hPa averaged over 1986–2006. The composite differences in horizontal winds between the five weakest and strongest EASM years (weakest–strongest) at (c) 850 hPa and (d) 500 hPa. Horizontal winds are from the GEOS-4 assimilated meteorological data. Unit is shown on top of each panel. The location of box (85°–120°E, 20°–46°N, the surface to 250 hPa) selected to capture the features of transboundary $O_3$ transport is also shown in (d).

**Fig. 6.** The composite differences in horizontal $O_3$ fluxes (shades, kg s$^{-1}$) and winds (contours, m s$^{-1}$) at (a) south, (b) north, (c) west, and (d) east boundaries of the selected box of (85°–120°E, 20°–46°N, the surface to 250 hPa) between the five weakest and strongest EASM years (weakest–strongest). Fluxes and winds

813 that are eastward or northward are positive, and those that are westward and
814 southward are negative. Mass fluxes are from simulation O3_TOT. Longitudinal
815 and meridional winds are from the GEOS-4 assimilated meteorological data.
816
817 **Fig. 7.** The JJA surface-layer $O_3$ concentrations (ppbv) from (a) simulation
818 O3_TB (referred to as $TBO_3$), (b) simulation O3_ST (referred to as $STO_3$), and (c)
819 simulation O3_TOT minus simulation O3_TB (referred to as $LOCO_3$). Panels (a),
820 (b), and (c) are the averages over 1986–2006. Panels (d), (e), and (f) are the
821 composite differences (ppbv) in $TBO_3$, $STO_3$, and $LOCO_3$, respectively, between
822 the five weakest and strongest EASM years (weakest–strongest).
823
824 **Fig. 8.** (a) The spatial distributions of JJA mean vertical wind velocity (contours,
825 Pa $s^{-1}$*1000) and simulated upward mass flux of $O_3$ (shades, kg $s^{-1}$) averaged
826 over 1986–2006. (b) The composite differences in vertical wind (contours, Pa
827 $s^{-1}$*1000) and simulated upward mass flux of $O_3$ (shades, kg $s^{-1}$) between the five
828 weakest and strongest EASM years (weakest–strongest). Vertical winds are from
829 the NCEP/NACR reanalysis data.
830
831 **Fig. 9.** The changes in simulated JJA surface-layer $O_3$ concentrations (ppbv)
832 owing to the changes in anthropogenic emissions of $O_3$ precursors over 1986–
833 2006, based on simulations of O3_EMIS.
834
835

(a) O₃ Conc. (ppbv)

(b) MAD (ppbv)

(c) APDM (%)

**Fig. 1.** (a) Simulated JJA surface-layer $O_3$ concentrations (ppbv) that are averaged over years 1986–2006 of simulation O3_TOT. (b) Mean absolute deviation (MAD, ppbv) and (c) absolute percent departure from the mean (APDM, %) calculated with 1986–2006 simulation of $O_3$ in O3_TOT.

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(a) EASMI & Surface O$_3$ Conc.

(b) Corr. EASMI & Surface O$_3$ Conc.

**Fig. 3.** (a) The normalized time series of EASMI (blue bars, left y-axis) and the simulated JJA surface-layer O$_3$ concentrations (black line, right y-axis, ppbv) averaged over China for years of 1986–2006. (b) Spatial distribution of the correlation coefficients between the EASMI and the JJA surface-layer O$_3$ concentrations. The dotted areas indicate statistical significance with 95% confidence from a two-tailed Student's t-test. The EASMI are calculated using the GEOS-4 assimilated meteorological data.

**Fig. 4.** Horizontal distributions of (a) absolute and (b) percentage differences in JJA surface-layer $O_3$ concentrations between the five weakest and strongest EASM years (weakest–strongest). Pressure-longitude cross sections averaged over 20°–46°N for (c) absolute and (d) percentage differences in JJA $O_3$ concentrations between the five weakest and strongest EASM years (weakest–strongest). Results are from simulation O3_TOT and unit is shown on top of each panel.

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881

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winds (contours, m s$^{-1}$) at (a) south, (b) north, (c) west, and (d) east boundaries of
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that are eastward or northward are positive, and those that are westward and
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and meridional winds are from the GEOS-4 assimilated meteorological data.

889

**Fig. 7.** The JJA surface-layer $O_3$ concentrations (ppbv) from (a) simulation O3_TB (referred to as $TBO_3$), (b) simulation O3_ST (referred to as $STO_3$), and (c) simulation O3_TOT minus simulation O3_TB (referred to as $LOCO_3$). Panels (a), (b), and (c) are the averages over 1986–2006. Panels (d), (e), and (f) are the composite differences (ppbv) in $TBO_3$, $STO_3$, and $LOCO_3$, respectively, between the five weakest and strongest EASM years (weakest–strongest).

(a) OMEGA & O$_3$ vertical mass flux at 500 hPa
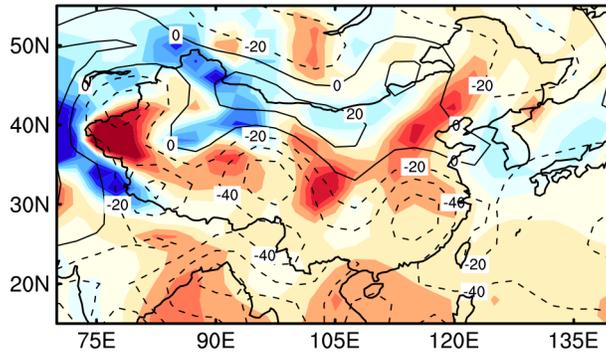
(b) Diff. in OMEGA & O$_3$ vertical mass flux at 500 hPa
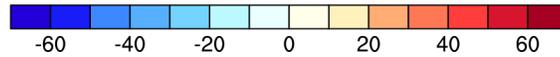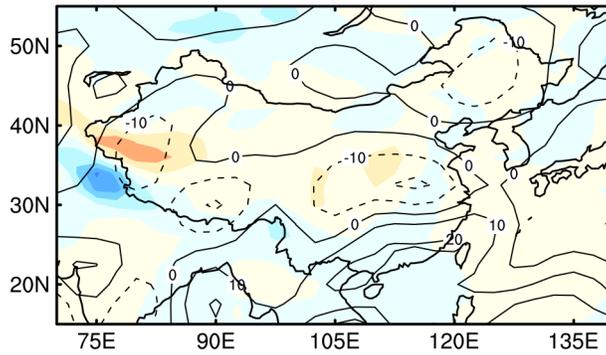
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Diff. in O$_3$ Conc. (ppbv)



**Fig. 9.** The changes in simulated JJA surface-layer O$_3$ concentrations (ppbv) owing to the changes in anthropogenic emissions of O$_3$ precursors over 1986–2006, based on simulations of O3_EMIS.