Influence of the Bermuda High on interannual variability of summertime ozone in the Houston-Galveston-Brazoria region

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ABSTRACT

The Bermuda High (BH) quasi-permanent pressure system is the key large-scale circulation pattern influencing summertime weather over the eastern and southern US. Here we developed a multiple linear regression (MLR) model to characterize the effect of the BH on year-to-year changes of monthly-mean maximum daily 8-hour average (MDA8) ozone in the Houston-Galveston-Brazoria (HGB) metropolitan region during June, July and August (JJA). The BH indicators include the longitude of the BH western edge (BH-Lon), and the BH intensity index (BHI) defined as the pressure gradient along its western edge. Both BH-Lon and BHI are selected by MLR as significant predictors ($p < 0.05$) of the interannual (1990-2015) variability of the HGB-mean ozone throughout JJA, while local-scale meridional wind speed is selected as an additional predictor for August only. Local-scale temperature and zonal wind

24   speed are not identified as important factors for any summer month. The best-fit

25   MLR model can explain 61%-72% of the interannual variability of the HGB-mean

26   summertime ozone over 1990-2015 and shows good performance in cross-validation

27   (R^2 higher than 0.48). The BH-Lon is the most important factor, which alone explains

28   38%-48% of such variability. The location and strength of the Bermuda High appears

29   to control whether or not low-ozone maritime air from the Gulf of Mexico can enter

30   southeastern Texas and affect air quality. This mechanism also applies to other

31   coastal urban regions along the Gulf Coast (e.g. New Orleans, LA; Mobile, AL; and

32   Pensacola, FL), suggesting that the BH circulation pattern can affect surface ozone

33   variability through a large portion of the Gulf Coast.

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35   **Keyword**: Bermuda High, ozone, Houston, Gulf Coast, multiple linear regression

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37   **1. Introduction**

38   Surface ozone, as an important air pollutant, has significant adverse impacts on

39   both public health and agriculture (NRC, 1991). Ozone is produced in the

40   troposphere by photochemical oxidation of carbon monoxide (CO) and volatile

41   organic carbon (VOCs), initiated by reaction with hydroxyl radicals (OH) in the

42   presence of nitrogen oxides (NO_x). Surface ozone is influenced not only by emissions

43   of its precursors but also by meteorological conditions (e.g. Jacob and Winner 2009).

44   Large-scale circulation patterns can lead to local meteorological conditions that are

45   favorable for ozone episodes, such as high temperatures, low wind speeds, clear

46   skies, and stagnation (Nielsen-Gammon et al., 2005; Ngan and Byun, 2011; Pearce et

47   al., 2011; Psiglogloue et al., 2013; Pugliese et al., 2014). Previous studies have

48   demonstrated certain associations between large-scale circulations and surface

49   ozone concentrations over the US (e.g. Darby, 2005; Rappenglück et al., 2008; Lin et

50   al., 2012; Zhu and Liang, 2013; Shen et al., 2015; Lin et al., 2015). For example,

51   surface ozone in the western US is affected by mid-latitude cyclones that transport

52   Asian pollutions eastward across the Pacific (Lin et al., 2012) and late-spring

53   stratospheric intrusions occurring more frequently following strong La Nina winters

54   (Lin et al., 2015). In the Midwest and Northeast US, polar jet frequency is a good

55   indicator for the interannual variability of surface ozone in summer (Shen et al.,

56   2015).

57   The Bermuda High (BH), a quasi-permanent system located over the North

58   Atlantic Ocean (Davis et al., 1997), is the key large-scale circulation pattern that

59   influences the regional climate over the eastern and southern US in summer (Li et al.,

60   2012; Zhu and Liang, 2013; Hegarty et al., 2007; Hogrefe et al., 2004). The BH

61   circulation pattern influences ozone air quality in the US through two mechanisms.

62   First, as the BH shifts westward from late spring to summer, it places the eastern US

63   under high pressures, resulting in meteorological conditions favorable for local

64   production of ozone, such as high temperatures, clear skies, and stagnation. A

65   number of studies have shown that high ozone concentrations easily occur over

66   large parts of the Northeast under the BH pressure pattern (Eder et al., 1993; Fiore

67   et al., 2003; Hogrefe et al., 2004). Second, the southerly flows at the western edge of

68 the BH bring clean marine air from the Gulf of Mexico to the southern Great Plains

69 (Higgins et al. 1997). This maritime inflow has lower concentrations of ozone

70 compared with the continental air it replaces, thus responsible for low ozone

71 background over much of the Gulf States in summer (Langford et al., 2009; Ngan and

72 Byun et al., 2011). These two mechanisms have been illustrated by Zhu and Liang

73 (2013). Using observational data, they found positive correlations over the Northeast

74 between maximum daily 8h average (MDA8) surface ozone in summer and the

75 intensity of the BH on the interannual time scale due to the first mechanism, but

76 negative correlations over the South-Central US due to the second mechanism. Shen

77 et al. (2015) further suggested that the location of the BH western edge has an

78 influence on the summer mean MDA8 ozone in the Southeast.

79   The Houston-Galveston-Brazoria (HGB) area is a major metropolitan area on the

80 Gulf coast located near the western edge of the BH in summer (Figure 1). The HGB

81 region was classified as a "marginal" nonattainment zone for ozone by the U.S.

82 Environmental Protection Agency (EPA) under the 2008 standard (TCEQ, 2012),

83 although mean and peak ozone of the HGB area has decreased significantly during

84 the past decades due to control of anthropogenic emissions (Berlin et al. 2013). A

85 number of studies have demonstrated the importance of meteorology and

86 circulation patterns on ozone over the HGB (Ngan et al., 2011; Rappenglück et al.,

87 2008; Pakalapati et al., 2009; Haman et al., 2014), focusing predominantly on a

88 typical year or episodic high ozone cases, such as those observed during the Texas

89 Air Quality Study-II (Rappenglück et al., 2008; Bridget et al., 2009). Doppler lidar

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measurements of the strength and direction of the nocturnal low level jet (LLJ) during the TexAQS 2006 field campaign (Tucker et al., 2009) found a relationship between the nocturnal LLJ and ozone concentrations on the next day. A strong southerly nocturnal LLJ was linked to strong BH conditions due to the superposition of the sea breeze cycle on a strong synoptic-scale southerly flow. Given the evidence established by prior investigations on the overall influence of the BH on summertime ozone over the southern US (e.g. Zhu and Liang 2013; Shen et al., 2015), we hypothesize that the large-scale circulation patterns associated with the BH play a key role in driving the year-to-year change in surface ozone over the HGB. In this study we will test this hypothesis by examining the statistical relationship between the variability of the BH and MDA8 ozone over the HGB during June, July, and August (JJA), a time period when the BH is located closer to North America and exerts large influences to circulation patterns over the HGB.

2. Data and Methods

2.1. Ozone observations

The HGB region is delineated by longitude from -94.5°W to -96.0°W, and by latitude from 28.5°N to 30.5°N (blue box in Figure 1a). Surface ozone concentrations over the HGB have been routinely monitored at continuous ambient monitoring stations (CAMSs) maintained by the Texas Commission on Environmental Quality (TCEQ), the City of Houston, and Harris County. Observational records of surface

111   ozone in JJA from 1990 to 2015 were obtained from the EPA AirData website

112   (http://www3.epa.gov/airquality/airdata/ad_data_daily.html). Ozone observations

113   from 28 ozone CAMS sites in the HGB region that have data records longer than 10

114   years were used for analysis. Figure 1b displays the site distributions and long-term

115   mean ozone in JJA at each site. The site locations and operation time periods are

116   provided in the supplementary material (Table S1). The number of CAMS sites

117   increases to 16 in 1998 and 21 in 2004. There are 8 sites with continuous

118   observations from 1990 to present. The overall data coverage at each selected site is

119   99% during its operation period, except the Houston East site that has ozone

120   observations since 1990 but no data during 1993-1995. In what follows, all the ozone

121   data and related discussions are MDA8 ozone and the correlation coefficient, r,

122   reported in this study is the Pearson correlation coefficient unless stated otherwise.

123   As shown in Figure 1b, the sites in Galveston and Brazoria counties have lower

124   ozone concentrations than the sites in the Houston region, due in part to lower local

125   emissions. Since the scope is on synoptic scale circulation patterns, we focus on the

126   HGB-mean ozone rather than ozone at individual sites. To calculate the HGB-mean

127   ozone for a given month, we averaged ozone observations from all eligible sites

128   (all-site average) and compared that with the average of only those sites with

129   continuous observations since 1990 (continuous-site average). The difference in the

130   HGB-mean ozone calculated from the two averaging approaches is within 5 ppbv and

131   diminishes in the 2000s as the sites number increases (Fig 2a). To remove the

132   influence of the decreasing trend in anthropogenic emissions on the interannual

133   variability of ozone, the HGB-mean ozone of each month (JJA) was de-trended by

134   subtracting the 7-year moving average of the corresponding month. The de-trending

135   process further decreases the difference in the HGB-mean ozone calculated from the

136   two averaging approaches (Fig 2b). Therefore we use the all-site average to present

137   the HGB-mean ozone. The time series of the de-trended HGB-mean ozone is from

138   1993 to 2012.

139   **2.2. Meteorological data**

140   The meteorological data consists of the geopotential height at 850hPa and sea

141   level pressure (SLP) from the National Centers for Environmental Prediction (NCEP)

142   Reanalysis 1 with a spatial resolution of $2.5^{o} \times 2.5^{o}$ (Kalnay et al., 1996), which were

143   used to derive the BH indices described below. We adopted 2-meter temperature (T),

144   zonal (U) and meridional (V) component of wind on 850hPa from the European

145   Centre for Medium-Range Weather Forecasts (ECMWF) Interim reanalysis with a

146   finer spatial resolution of $0.5^{o} \times 0.5^{o}$. These data were used to calculate the local-scale

147   meteorological parameters, including the HGB-mean temperature and 850 hPa

148   winds (U and V).

149   **2.3. BH indices**

150   Figure 3 illustrates the mean BH circulation patterns from June to August. The

151   HGB region is located to the west of the BH, thus under strong southerly winds from

152   June to August. The intensity of the BH peaks in July. As the BH intensifies from June

153   to July, its west edge moves westward toward the North America continent; when it

154 weakens from July to Aug, its west edge retreats eastward away from the continent.

155 Separate indices have been used to define the intensity and location of the BH

156 (Stahle and Cleaveland, 1992; Ortegren et al. 2011, Li et al. 2011; Zhu and Liang,

157 2013). For the BH location, we adopted the BH longitudinal index (BH-Lon) from Li et

158 al. (2011) as a measure of the westward extension of the BH. They defined the

159 BH-Lon as the longitude of the cross-point of the 1560 geopotential meter (gpm)

160 isoline and the 850hPa wind ridgeline. The wind ridgeline refers to the roughly zonal

161 line that separates the easterly trade winds in the south from the westerly winds in

162 the north, and it can be written mathematically as u=0 and $\frac{\partial u}{\partial y} \geq 0$, where u is the

163 zonal wind component and y is meridional coordinate. The location of the

164 1560-gpm isoline is linearly interpolated to the precision of 0.1^{o} from the gridded

165 reanalysis data. The BH-Lon is always negative in longitude, thus more negative

166 values meaning closer proximity to Houston.

167   The intra-seasonal variability of the BH (c.f. Fig 3) will affect the value of BH-Lon

168 when defined with reference to a fixed isoline. For instance, the 1560 gpm isoline is

169 located further away from the HGB in August when the BH is weaker than in July,

170 even when the center location of the BH from the HGB does not change between the

171 two months. To reduce such effect, we tried different isolines with an interval of 4

172 gpm from 1560 to 1536 gpm in the calculation of the BH-Lon by month and

173 evaluated which definition captures the most variability of the HGB-mean ozone. For

174 June and July, the BH-Lon is best defined on the basis of the 1560 gpm isoline, the

175 same as in Li et al. (2011), and this definition results in the interannual (1993-2012)

176   correlation (r) of 0.69 between detrended BH-Lon and HGB-mean ozone. The BH-Lon

177   for August is defined using the 1556 gpm and the corresponding r is 0.62.

178   The time series of monthly-mean BH-Lon is shown in Figure 4. In

179   correspondence with the intra-seasonal movement of the BH shown in Fig 3, the

180   mean value of BH-Lon from 1990-2015 was 80.3°W in June, 93.1°W in July, and

181   91.6°W in August. There were no significant trends in BH-Lon for the months of June

182   and July. In August, however, BH-Lon exhibited a significant increasing trend (i.e., an

183   eastward shift) of 0.51° a^{-1} (p<0.1) over 1990-2015, but this trend becomes not

184   significant if the first four years (1990-1993) were excluded. Shen et al. (2015) found

185   an increasing trend of 0.35° a^{-1} in the JJA-mean BH-Lon over 1980 - 2010 using the

186   definition of Li et al. (2011). Rather than a change in the BH circulation patterns, they

187   attributed this trend to a spatially uniform decrease of SLP over the US and adjacent

188   Atlantic Ocean in the reanalysis data. Given their work as well as the lack of a

189   consistent trend in the monthly-mean BH-Lon over 1990-2015, we do not consider

190   the trend of BH-Lon in the present work. To be consistent with the ozone data, the

191   BH-Lon time series were processed by removing the 7-year moving averages.

192   Another type of BH index is defined on the basis of pressure differences

193   between two representative locations, with their exact locations varying among

194   studies. Zhu and Liang (2013) defined a pressure-based BH index (BHI) as the mean

195   SLP difference between a location in the Gulf of Mexico and the other in the

196   southern Great Plains where the SLP has the largest positive and negative correlation

197   with LLJ respectively. As a result, their BHI exhibits a significant positive association

198 between the strength of LLJ, which determines the transport of clean marine air

199 from the Gulf of Mexico. Similar to that study, we defined a pressure-based BHI as

200 the mean SLP difference along the west edge of the BH, between the same location

201 in southern Great Plains (35°-39°N, 105.5°-100°W) as selected by Zhu and Liang

202 (2013) (box 2 in Figure 1a) and the other in the Gulf of Mexico (25.3°-29.3°N,

203 92.5°-87.5°W) (box 1 in Figure 1a) where the SLP exhibits the largest correlation with

204 the HGB-mean ozone. Our Gulf of Mexico domain is located 2.5° east of that defined

205 by Zhu and Liang (2013). This BHI shows only weak to moderate correlations with

206 BH-Lon, with *r* being -0.57, -0.26 and -0.39 for JJA respectively, suggesting the

207 position of the BH west edge may not vary coherently with the pressure gradient

208 over the west edge. Therefore, both BH-Lon and BHI are used as predictors in the

209 regression model described below.

210 **2.4. Statistical Method**

211 We applied a multiple linear regression (MLR) model, which has been

212 commonly used in air quality and meteorological studies (e.g. Kutner et al., 2004; Tai

213 et al., 2010), to construct the statistical relationship between the detrended

214 HGB-mean ozone and the five meteorological predictors described above, including

215 BH-Lon, BHI, T, U, and V. Since our focus is on variability, all the meteorological

216 predictors were detrended using the same approach as the ozone data. For ease of

217 comparison, all the meteorological predictors in the regression analysis were

218 normalized. The regression was conducted on the monthly scale from 1993 to 2012.

219 The model is of the form：

$$y = \beta_0 + \sum_{k=1}^{n} \beta_k x_k \tag{1}$$

where y is the detrended monthly HGB-mean MDA8 O_3, n is the number of predictors, x_k represents the kth predictor which is detrended and normalized, β_k is the corresponding regression coefficient for x_k, and β_0 is the intercept. The predictors are separated into two groups. We applied a stepwise regression using the BH indices (BH-Lon, BHI) first, which represent the large-scale effects. The HGB-mean T, U, and V that characterize local meteorological conditions were added subsequently only if they result in significant improvements in model performance. The predictor selection is based on the Akaike Information Criterion (AIC) statistics to obtain the best model fit (Venables and Ripley, 2003).

3. RESULTS

3.1. Ozone and the BH relationship

The HGB-mean ozone shows a large intra-seasonal variation during JJA (Figure 5), with a minimum of monthly-mean ozone in July. We first examined if this feature can be explained by near-surface temperature, which has been suggested as an important meteorological factor affecting surface ozone in many regions (Fu et al., 2015; Rasmussen et al., 2012; Camalier et al., 2007). As shown in Figure 5, the HGB-mean temperature is the highest in July when the monthly mean ozone is lowest, precluding temperature as the driver of the intra-summer variability of ozone over the HGB region. This can be explained by the fact that summertime

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241 temperatures over the HGB region are always high, thus the temperature variation is

242 relatively less significant and the ozone formation might not be limited by

243 temperature over this region in summer. To further support this argument, on the

244 interannual time scale (1993-2012) the HGB-mean ozone shows no correlation with

245 temperature in June (r = -0.14), although the correlation coefficient between the two

246 increases to 0.29 and 0.41 in July and August, respectively.

247   The BH-Lon reaches its westernmost location in July (Figure 3), coincident with

248 the ozone minimum (monthly mean) in the same month. This coincidence supports

249 the mechanism that the more westward shift of the BH (the lower BH-Lon) brings

250 the stronger inflow of cleaner maritime air into the HGB leading to lower surface

251 ozone. The decrease of background ozone over the HGB region from spring to

252 summer is reported by a number of observational and modeling studies

253 (Nielsen-Gammon et al., 2005a; Nielsen-Gammon et al., 2005b; Li et al., 2002;

254 Reidmiller et al., 2009) and can be attributed to strengthening of this maritime

255 inflow in summer. Indeed, ~~as shown in Figure 6a,~~ the BH-Lon shows a significantly

256 stronger correlation with the HGB-mean ozone during JJA (*r* = 0.64~0.74) than

257 temperature does. However, the BH-Lon does not correlate well with the HGB-mean

258 temperature, with r being -0.04, 0.44, and 0.33 for JJA respectively. The BH-Lon has a

259 higher correlation with the HGB-mean meridional wind (V) (r = -0.4~-0.7) but does

260 not correlate with the zonal wind (U). This is expected because the BH determines

261 the strength of the meridional flows that bring maritime air masses from the Gulf of

262 Mexico to southeastern Texas.

263   Considering the large intra-summer variation in ozone, BH-Lon and their

264 association, the effects of the BH on the HGB-mean ozone are analyzed month by

265 month in the following sections. For comparison, Zhu and Liang (2013) combined the

266 intra-seasonal and interannual variations of ozone and Shen et al. (2015) focused on

267 the variability of the JJA-mean ozone. Both studies investigated the southern US as a

268 whole. We note here that HGB ozone exhibits a bimodal seasonality (c.f. Figure 5),

269 with 41% of exceedance days occurring in JJA, and the rest in the spring and early fall.

270 The meteorological features identified here are not expected to predict peak ozone

271 outside of JJA, which is a limitation of the study.

272 **3.2. Statistical model**

273   Using the stepwise regression (Equation 1), we obtained the best-fit MLR

274 equations for the interannual variability of the HGB-mean ozone by month. Table 1

275 summarizes the regression results, including the predictors selected for each month

276 and their regression coefficients. Both BH-Lon and BHI are selected as significant

277 predictors ($p < 0.05$) for each month, while V is selected as an additional predictor

278 for August only. Temperature is not selected as a predictor by the MLR for any

279 month, which is consistent with insignificant correlation between ozone and

280 temperature presented above. The HGB-mean zonal wind (U) is not selected for any

281 month either. The coefficients of determination (R^2) from the MLR are 0.61 for Jun,

282 0.72 for Jul, and 0.70 for Aug.

283   We verified that the MLR model results are not sensitive to the data

284 de-trending methods (Table S2 in the supplementary material). The use of raw ozone

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Figure 7 displays the time series of the observed and MLR-regressed

monthly-mean ozone (both detrended) from 1993 to 2012. Many of the extremely

high and low ozone months are well reproduced by the MLR model, for example, the

low ozone month of June 2004 and high ozone month of August 2011. The BH-Lon is

the most important predictor throughout JJA and has the highest regression

coefficient (absolute values) that is positive throughout JJA (Table 1). Given the

BH-Lon being negative, this means that as the Bermuda High extends more

westward, ozone over the HGB is lowered, which supports the mechanism that the

BH brings maritime inflow of low ozone background into the HGB region. The MLR

relationship indicates that monthly-mean MDA8 ozone over the HGB area will

decrease by 4.52, 3.44, and 3.34 ppbv, respectively, for every degree of westward

extension of the BH-Lon in June, July, and August.

While the MLR model developed here captures 61%-72% (based on R^2) of the

interannual variance of the HGB-mean ozone from June to August, the

307   meteorological predictors may be correlated with each other, leading to overfitting

308   of the data. To examine the multi-collinearity between the meteorological predictors,

309   the variance inflation factor (VIF), a widely used index of the collinearity in the

310   regression analysis, was calculated for each variable. Table 2 summarizes the VIF of

311   each predictor by month. Most of the VIFs are smaller than 3, much lower than the

312   commonly-used VIF threshold of 10 in determining significant collinearity (Kutner et

313   al., 2004), indicating the problem of multi-collinearity among the predictors is

314   generally unimportant.

315   **3.3.   Model cross-validation**

316   The MLR model described above shows good regression performance in

317   explaining the interannual variations of the summertime HGB-mean ozone on the

318   monthly scale. To evaluate the predictability of this model, a cross-validation (CV)

319   method was implemented. We first isolated one year at a time, performed model

320   fitting with the remaining years, and then applied the model to predict the

321   monthly-mean ozone on the isolated year. Figure S1 shows the CV results by month.

322   The R^2 between the observed and CV-predicted ozone is 0.48~0.59, indicating the

323   MLR model is capable of predicting about 50% interannual (1993-2012) variability of

324   monthly mean ozone over the HGB area in summer. However, some of the extreme

325   ozone values are not well predicted, suggesting that other factors are responsible for

326   those high ozone events, e.g. emissions and stagnation conditions, which are not

327   considered as predictors in the MLR model.

328

4. DISCUSSION

4.1. Comparison with other studies

To the best of our knowledge, the MLR-regression correlation coefficients (r = 0.78~0.84) are significantly higher than those from previously published studies on the regression relationship between interannual variability of meteorological factors and surface ozone over a metropolitan region or the southern US as a whole. Zhu and Liang (2013) reported a negative correlation of -0.5 ~ -0.7 between the BHI and summer-mean MDA8 ozone over the southern Great Plains (including Houston) during 1993-2010. Shen et al. (2015) identified the polar jet, the Great Plains low level jet, and the BH as major synoptic-scale patterns influencing surface O_3 variability in the eastern US in summer, which in combination explain 53% (r = 0.73) of the interannual variance of summer-mean MDA8 ozone in South Central US during 1980-2010. These studies averaged surface ozone over a large geographical region and onto a seasonal mean and thus smoothed out some variability. By comparison, the present study investigates the interannual variability of monthly mean ozone over a smaller region (HGB) and extends to more recent time periods (1990-2015). With just three meteorological predictors (BH-Lon, BHI, and V), the MLR model developed here captures 61% - 72% of the interannual variance of the HGB-mean ozone during JJA. The MLR model developed here shows a good prediction skill with the CV R^2 higher than 0.48 for each month of JJA.

4.2. Mechanism

350   Among the meteorological predictors examined here, indicators of the BH

351   location (BH-Lon) and strength (BHI) explain more interannual variability of the

352   summertime HGB-mean ozone than local meteorological factors (i.e. winds and

353   temperature), indicating the dominant role of large-scale circulation patterns

354   controlling ozone variability over this region in summer. The BH-Lon is the most

355   important predictor throughout JJA, which alone explains 38%-48% of the observed

356   interannual variability of monthly mean ozone over the HGB region. As discussed

357   above, the BH circulation patterns in summer are responsible for the inflow of low

358   ozone air masses from the Gulf of Mexico into the HGB region. The MLR results

359   suggest that the BH-Lon is a better predictor than local-scale meridional wind to

360   indicate the variability of this maritime inflow and consequently surface ozone.

361   As a further illustration of this mechanism, Figure 8 compares circulation

362   patterns and associated changes in the HGB-mean ozone between two

363   representative months, Jul 2000 and Jul 2001. The BH was not only stronger in Jul

364   2001 but also extended closer to the southeastern US during that month than Jul

365   2000. The BH-Lon was 88.7^{o}W in Jul 2001 as compared to that of 78.7 ^{o}W in Jul 2000,

366   indicating stronger maritime influence to HGB during Jul 2001. Correspondingly,

367   surface ozone concentrations were significantly lower at all the CAMS monitors

368   during Jul 2001 than Jul 2000 (Fig 8). The HGB-mean ozone was 38.9 ppbv in Jul 2001,

369   compared with 49.0 ppbv in Jul 2000. The number of ozone nonattainment days was

370   also lower in Jul 2001 (2 days) than Jul 2000 (6 days). Despite different BH-Lon, the

371   HGB-mean V happened to be the same during the two months, both -2.1 m/s. This

372   indicates that local-scale wind alone may not be sufficient to indicate the origin of air

373   masses because winds are affected by both large-scale and local-scale factors.

374   Given the large-scale influence of the BH, the mechanism underlying the BH and

375   O_3 association over the HGB region, if correct, should apply for other urban regions

376   along the Gulf Coast which experience similar onshore maritime flow from the Gulf

377   of the Mexico in summer. To test this, we chose New Orleans in Louisiana

378   (29.8°-30.6°N, 89.9°-90.7°W), Mobile in Alabama (30.4°-30.8°N, 88.0°-88.2°W), and

379   Pensacola in Florida (30.3°-30.5°N, 87.2°-87.3°W), all of which are at about 30°

380   latitude, similar to HGB. Figure 6b-d presents the time series of surface ozone

381   (detrended and averaged over all the CAMS monitors) at these urban areas during

382   JJA along with BH-Lon. On interannual time scales (1993-2013), monthly mean ozone

383   concentrations at the three urban regions all exhibit significantly positive

384   correlations with the BH-Lon throughout JJA (r = 0.38~0.64), which is consistent with

385   the BH-O_3 association over HGB and thus confirms the large-scale impact of the BH

386   circulation patterns on surface ozone variability along the Gulf Coast. It is interesting

387   to note that the BH-O_3 correlation coefficients at HGB are highest among the four

388   urban regions along the Gulf Coast. This can be partly explained by the fact that HGB

389   has the largest quantity of ozone-precursor emissions among the four urban areas,

390   so that the intrusion of clean maritime air masses would cause a larger relative

391   reduction of ozone at HGB, and therefore the BH location and strength could cause

392   more ozone variability.

393   **4.3.   Threshold value of BH-Lon**

394   Shen et al. (2015) identified a threshold value of 85.4°W for the JJA-mean

395 BH-Lon that marks two different associations of the BH-Lon with surface ozone in the

396 eastern US. When the BH-Lon is east of 85.4°W, westward movement of the BH-Lon

397 leads to a decrease of surface ozone; when it is west of that threshold, its westward

398 movement leads to an increase of ozone. We did not find such a threshold value of

399 BH-Lon for the monthly scale analysis presented above; the monthly-mean

400 relationship between BH-Lon and HGB-mean ozone is consistently linear throughout

401 the study period for each summer month (c.f. Fig 6 and Fig S1).

402   On the daily scale, however, we found evidence for a threshold of BH-Lon that

403 tends to create conducive or non-conducive conditions for high ozone days over the

404 HGB region. Figure 9a shows the variation of HGB-mean daily ozone anomaly as a

405 function of daily BH-Lon during July 2011, a high ozone month. The daily ozone

406 anomaly is calculated as the difference between daily and 30-day moving average of

407 ozone. The daily data shown in Fig 9a clearly display two populations, warranting a

408 threshold of BH-Lon to separate them. As a simplified attempt, a linear piecewise

409 fitting was applied to the daily data, using different segmentation points with varying

410 BH-Lon from 100° W to 80° W at an interval of 0.5°. The least regression error (sum

411 of the squared errors) was obtained when the segmentation point was set to 90° W,

412 which represents the threshold of BH-Lon for July 2011. The resulting regression

413 indicates that when the BH-Lon is east of 90°W, the westward extension of BH-Lon

414 tends to cause a decrease of HGB ozone; the opposite relationship holds when the

415 BH-Lon is west of that threshold (Fig 9).

416   To understand causes of the two regimes, Figure 9b-c compares circulation

417   patterns between two representative days, July 3 and July 10 2011, when the BH-Lon

418   is located to the west and east of its threshold respectively. The circulation patterns

419   on July 10 (9c), when the BH-Lon was located to the east of 90°W, are similar to the

420   typical monthly-mean patterns described before: clean maritime air masses flow

421   southeasterly to southeast Texas along the west edge of the BH, leading to lower

422   surface ozone in HGB (26.4 ppbv on July 10 2011). On July 3 (9b), there was a

423   high-pressure system over southeast US, which appears to be separate from the BH

424   and in this case the choice of 1560 gpm isoline may not be appropriate to define the

425   BH west edge on a daily scale. According to the wind fields, however, this

426   high-pressure system over land is likely resulted from the westward extension of the

427   BH to the continental US. The high-pressure system typically brings stagnant weather,

428   high temperatures, and clear skies, which are all favorable meteorological conditions

429   leading to higher ozone on July 3 (42.2 ppbv).

430   To predict daily ozone on the basis of its statistical relationships with

431   meteorology alone is a challenging task and beyond the scope of the present study.

432   The simple daily analysis presented above for the month of July 2011 was intended

433   to demonstrate that the linear relationship between the BH-Lon and HGB-mean

434   ozone on a monthly scale is useful to explain some portion of HGB ozone variability

435   on a daily scale, but there are certain degrees of nonlinearity in the daily relationship

436   associated with a threshold value of the BH-Lon that separates different circulation

437   regimes and that threshold value may not be constant for every month. When the

438 BH-Lon is used to predict the monthly-mean ozone over the HGB area, however, it is

439 not necessary to consider the threshold of BH-Lon since the mean variations of the

440 BH-Lon is a lot smaller on the monthly scale than that on the daily time scale.

441

442 **5. Conclusion**

443 More than two decades (1990 - 2015) long observational record of MDA8 ozone

444 and meteorology were analyzed to characterize the effects of the BH circulation

445 patterns on interannual variations of surface ozone in the HGB region during the

446 summer months (June to August). The BH indicators are the longitude of the BH

447 western edge (BH-Lon) and the pressure-based BH intensity index (BHI) along its

448 western edge. Statistical relationships between the HGB-mean ozone variability and

449 meteorological predictors, including both large-scale (BH-Lon, BHI) and local-scale

450 ones (T, U, V), were tested and developed through multiple linear regression (MLR).

451 The best-fit MLR equations select both BH-Lon and BHI as significant predictors ($p <$

452 0.05) of interannual variability of the HGB-mean ozone for each month and

453 meridional wind speed (V) as a significant predictor for August only. Temperature or

454 zonal wind speed (U) is not selected as a predictor by the MLR for any of the summer

455 month. The exclusion of temperature in the MLR model is supported by the lack of

456 significant correlations between ozone and temperature over the HGB region on

457 both intra-seasonal and interannual time scales. This suggests temperature is not a

458 key driver of summertime ozone variability in the HGB region despite its importance

459 for other regions.

460   With only three meteorological predictors (BH-Lon, BHI, and V), the MLR model

461   developed here captures 61%-72% (based on R^2) of the interannual variance of the

462   HGB-mean ozone from June to August. The MLR model developed here also show a

463   good prediction skill with the CV R^2 higher than 0.45. The BH-Lon alone explains

464   38%-48% (r = 0.62~ 0.69) of the year-to-year variability in monthly mean ozone over

465   HGB during JJA for the period 1990-2015, indicating the dominant role of large-scale

466   circulation patterns controlling ozone variability over this region in summer. Such a

467   high correlation is explained by the mechanism that the western extension of the BH

468   determines the strength of the inflow of maritime air masses with lower ozone

469   background from the Gulf of Mexico to the HGB during summer. This mechanism

470   also applies to other coastal urban regions, such as New Orleans LA, Mobile AL and

471   Pensacola FL, confirming the large-scale impact of the BH circulation patterns on

472   surface ozone variability along the Gulf Coast. The linear relationship between the

473   BH-Lon and HGB-mean ozone on a monthly scale is useful to explain some portion of

474   HGB ozone variability on a daily scale, but there are certain degrees of nonlinearity

475   in the daily relationship associated with a threshold value of the BH-Lon that

476   separates different circulation regimes and that threshold value may not be constant

477   for every month. The statistical relationship between surface ozone and large-scale

478   circulation patterns on derived herein will be useful to distinguish the role of

479   meteorology versus anthropogenic emissions in controlling the interannual

480   variability of ozone along the Gulf Coast as well as to serve a benchmark to test the

481   performance of air quality models in representing such distinctions, which will be

482 investigated in future studies.

483

489

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622

Tables

624   Table 1. Regression coefficients and coefficients of determination (R^2) of the best-fit

625   MLR models for June, July, and August.   The model cross-validation R^2 is shown in

626   the last column.

	β_0 (intercept)	BH-Lon	BHI	V	R^2	Adjusted R^2	Cross-validation R^2
June	0.16	4.52	-3.84	-	0.61	0.56	0.48
July	-0.26	3.44	-2.98	-	0.72	0.69	0.59
August	-0.14	3.34	3.21	-5.21	0.70	0.64	0.51

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629   Table 2. Variance inflation factor (VIF) of the predictors selected in the MLR mode for

630   June, July and August.

	BH-Lon	BHI	V
June	1.47	1.47	-
July	1.07	1.07	-
August	1.21	2.21	2.22

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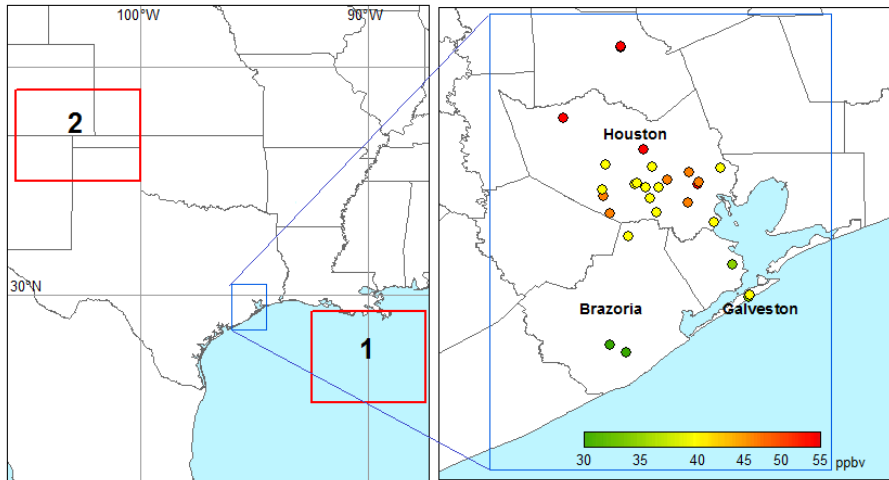
Figures

636   Figure 1. (a) Locations of the HGB region (blue box). The red boxes show the regions

637   used to define the BH intensity indices BHI; (b) Locations of the CAMS sites within

638   the HGB region and the long-term (1990-2015) mean MDA8 ozone from June to

639   August.

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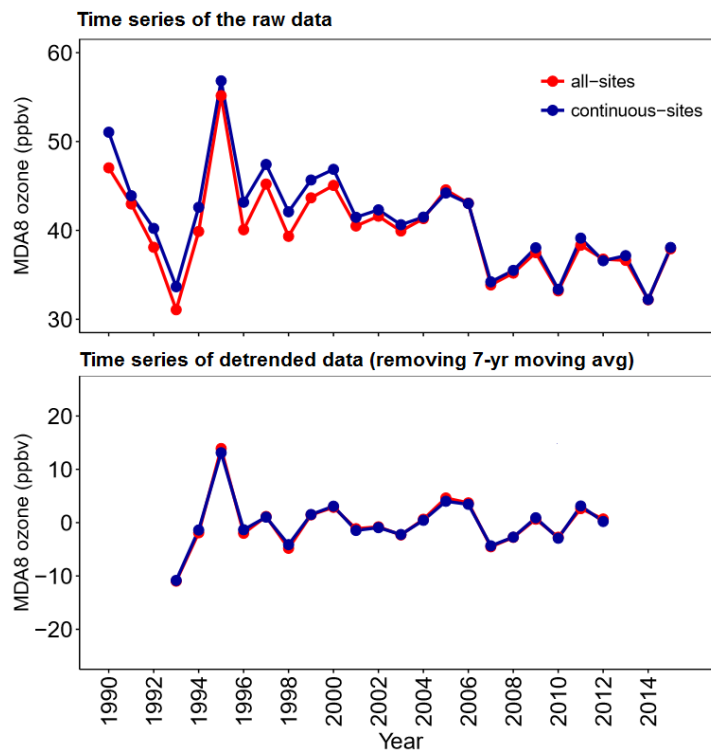
642   Figure 2. Summer (JJA)-mean ozone over the HGB area during the period from 1990

643 to 2015. The upper panel is the time series of the raw data and the lower panel is the

644 detrended time series after subtracting the 7-year moving averages. The red line is

645 the average from all available sites for each year and the blue line is the average of

646 those sites with continuous coverage from 1990 to 2015.

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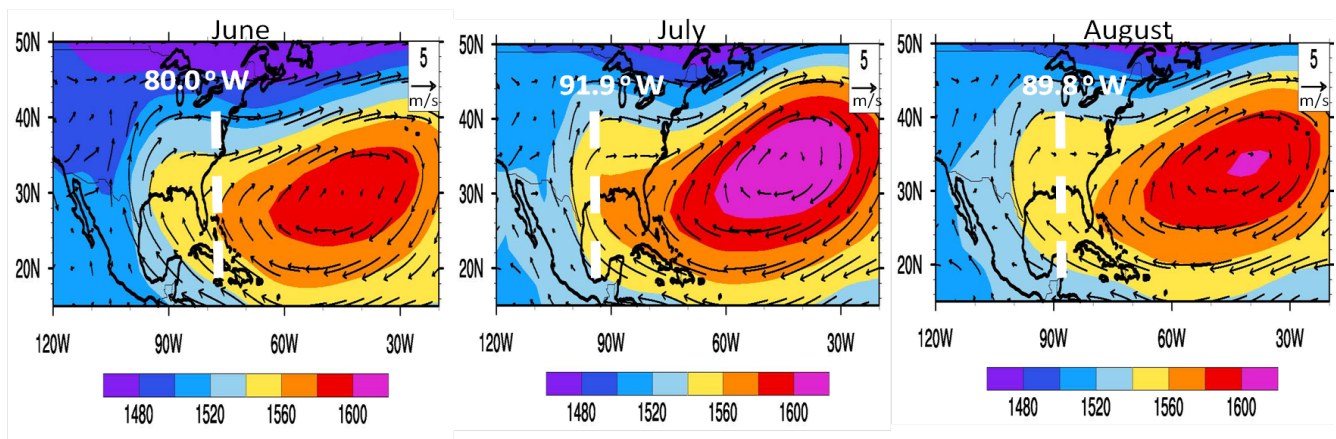
649 Figure 3. Distributions of the 1998-2013 mean 850hPa geopotential height (color

650 contour) and wind fields (arrows; m/s) in June (left), July (center), and August (right).

651 The thick white dashed line shows the longitude of the BH-Lon (values shown in

652 white).

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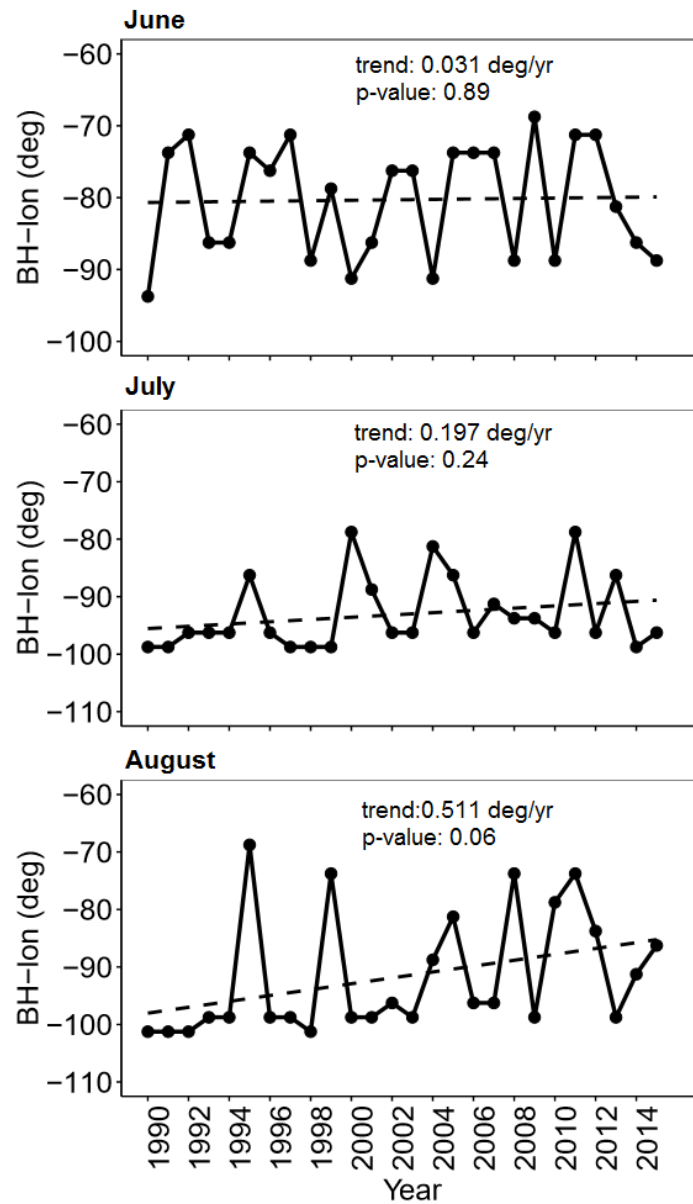
Figure 4. Time series of the BH-Lon (solid line) in June, July and August from 1990 to

2015. A linear trend (dash line) is used to fit each time series.

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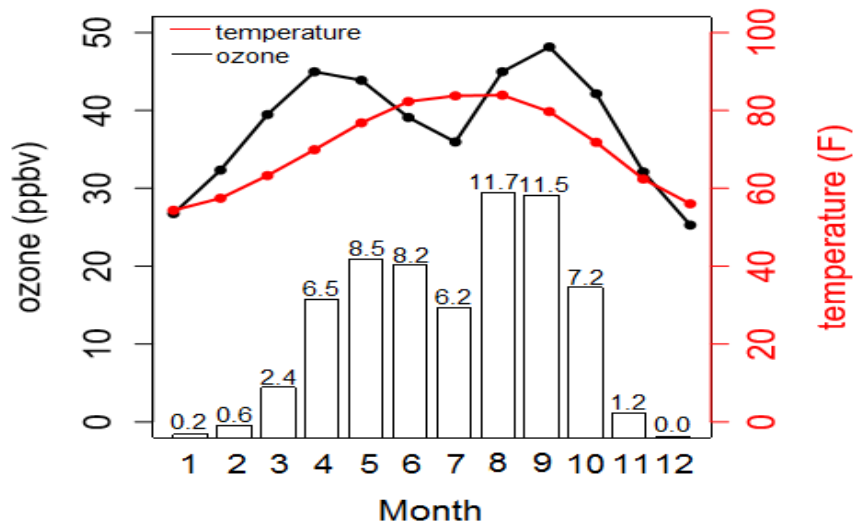
659 Figure 5. The 1990-2015 mean monthly MDA8 ozone over the HGB area (black line),

660 overlaid with the average number of exceedance days by month (bars) and the

661 HGB-mean temperature (red line). An exceedance day is defined when MDA8 ozone
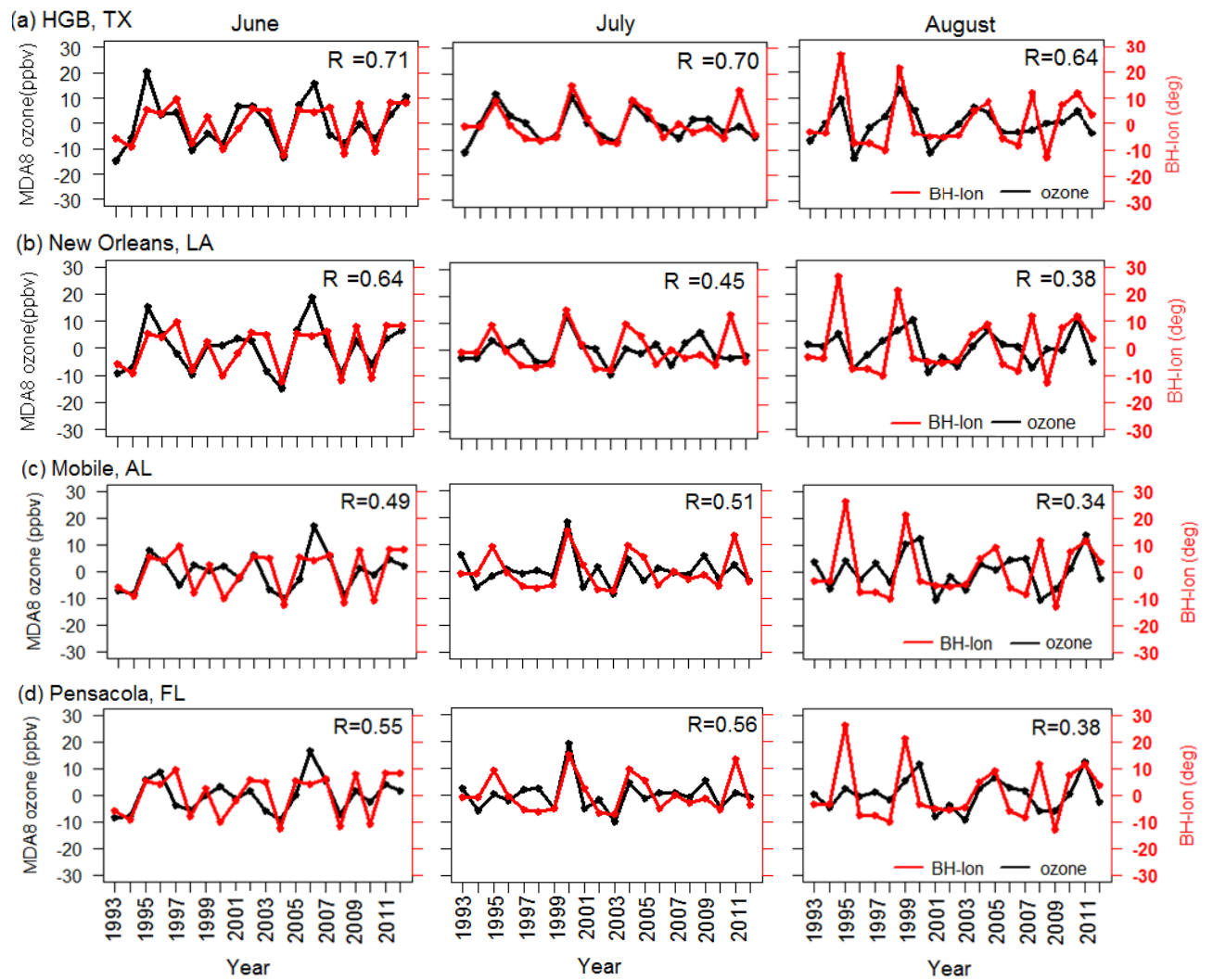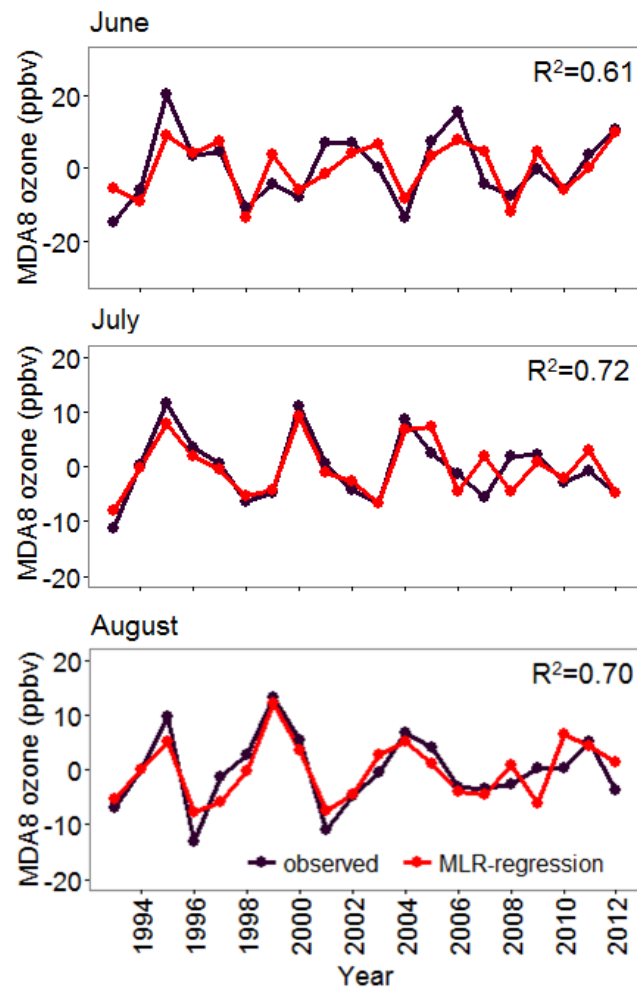
662 at one or more monitors in HGB is higher than 70 ppbv.

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Figure 6. The time series of detrended BH-Lon (red line) and MDA8 ozone (black line) during June, July and August over the HGB region, TX (a), New Orleans, LA (b), Mobile, AL (c) and Pensacola, FL (d). The correlation coefficients between BH-Lon and ozone are shown in each figure.

678   Figure 7. Time series of observed HGB-mean MDA8 ozone (black line) and

679   MLR-regressed ozone (red line) in June (top), July (middle) and August (bottom). The

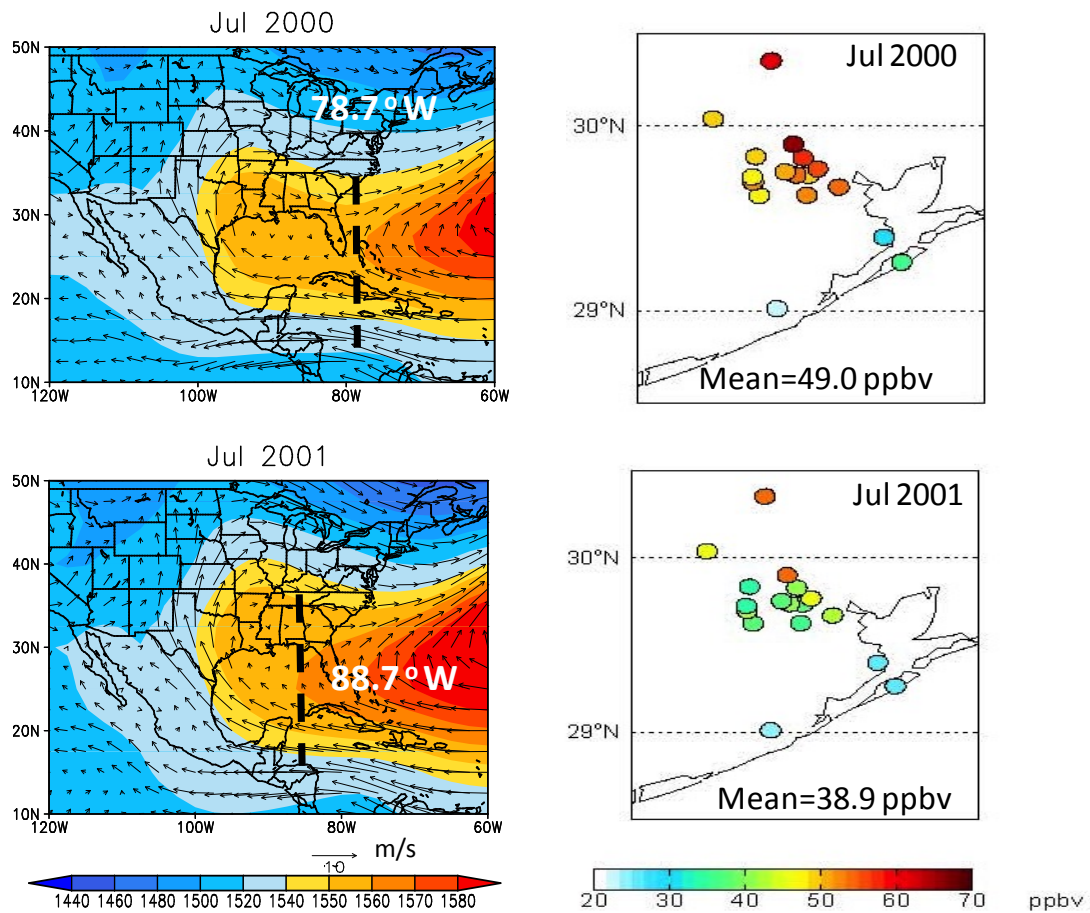680   data presented are detrended.

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Figure 8. (Left) 850 hPa geopotential height (gpm) and wind field (m/s) for the month

July 2000 and July 2001. The black dashed line shows the longitude of the BH-Lon

(values shown in white). (Right) MDA8 ozone concentrations at the CAMS sites in
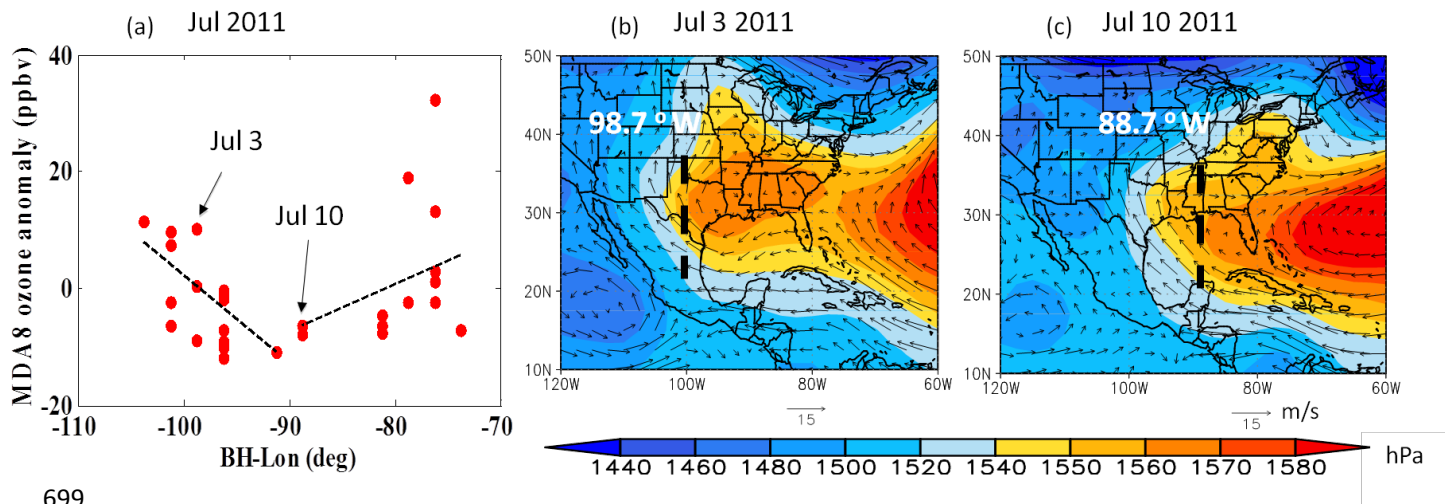
HGB for July 2000 and July 2001.

Figure 9. (a) Relationship between daily ozone anomaly (y-axis) and daily BH-Lon (y-axis) for July 2011. (b-c) Distribution of SLP (color contours) and 850hPa wind fields (arrows) on July 3rd (b) and July 10th 2011 (c). The black dashed line shows the longitude of the BH-Lon (values shown in white).