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Improved estimate of the policy-relevant background ozone in the United States using the GEOS-Chem global model with $1/2^\circ \times 2/3^\circ$ horizontal resolution over North America

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1 **Improved estimate of the policy-relevant background ozone in the United States**
2 **using the GEOS-Chem global model with $1/2^\circ \times 2/3^\circ$ horizontal resolution over**
3 **North America**

4
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23
24

24 **Abstract**

25

26 The policy-relevant background (PRB) ozone is defined by the US Environmental
27 Protection Agency (EPA) as the surface ozone concentration that would be present over
28 the US in the absence of North American anthropogenic emissions. It is intended to
29 provide a baseline for risk and exposure assessments used in setting the National
30 Ambient Air Quality Standard (NAAQS). We present here three-year statistics (2006-
31 2008) of PRB ozone over the US calculated using the GEOS-Chem global 3-D model of
32 atmospheric composition with $1/2^\circ \times 2/3^\circ$ horizontal resolution over North America and
33 adjacent oceans ($2^\circ \times 2.5^\circ$ for the rest of the world). We also provide estimates of the US
34 background (no anthropogenic US emissions) and natural background (no anthropogenic
35 emissions worldwide and preindustrial methane). PRB is particularly high in the
36 intermountain West due to high elevation, arid terrain, and large-scale subsidence. We
37 present for this region a detailed model evaluation showing that the model is successful in
38 reproducing ozone exceedances up to 70 ppbv. However, the model cannot reproduce
39 PRB-relevant exceptional events associated with wildfires or stratospheric intrusions. The
40 mean PRB estimates for spring-summer are 27 ± 8 ppbv at low-altitude sites and 40 ± 7
41 ppbv at high-altitude sites. These include a mean enhancement from intercontinental
42 pollution and anthropogenic methane of 9 ppbv at low-altitude sites and 13 ppbv at high-
43 altitude sites. The PRB is higher than average when ozone exceeds 60 ppbv, particularly
44 in the intermountain West. The annual 4th-highest PRB values in the intermountain West
45 are typically 50-60 ppbv, as compared to 35-45 ppbv in the East or on the West Coast.
46 Our PRB estimates are on average 4 ppbv higher than in previous GEOS-Chem studies
47 and this may reflect higher lightning, increasing Asian emissions, and improved model
48 resolution.

49

50 **Keywords:** Ozone; background ozone; policy relevant background; air quality standard

51

52

53 **1. Introduction**

54

55 The US Environmental Protection Agency (US EPA, 2006) defines the policy-relevant-
56 background (PRB) for ozone air quality as the surface ozone concentration that would be
57 present in the US in the absence of anthropogenic emissions from North America
58 (defined as the ensemble of the US, Canada, and Mexico). The PRB is used in the setting
59 of the National Ambient Air Quality Standard (NAAQS) to estimate the maximum ozone
60 reduction that could be achieved through North American emission controls. It provides
61 a baseline for risk and exposure assessments. The present US NAAQS is 75 ppbv (annual
62 4th-highest daily maximum 8-h average concentration), but the EPA is considering
63 decreasing it to a value in the 60-70 ppbv range. As the standard becomes more stringent
64 and approaches the PRB, accurate specification of the PRB becomes increasingly
65 important

66

67 Ozone is produced in the troposphere by photochemical oxidation of CO and volatile
68 organic compounds (VOCs) in the presence of nitrogen oxides ($\text{NO}_x \equiv \text{NO} + \text{NO}_2$). It is
69 also transported to the troposphere from the stratosphere. The NO_x , CO, and VOC

70 precursors of ozone have major anthropogenic sources from fuel combustion as well as
71 natural sources including lightning, wildfires, and the biosphere. Ozone has a lifetime of
72 only a few days in the continental boundary layer but weeks in the free troposphere (Y.
73 Wang et al., 1998; Fiore et al., 2002). Ozonesonde, aircraft, and satellite observations
74 show typical ozone concentrations of 50-70 ppbv in the free troposphere over North
75 America (Thompson et al., 2007; L. Zhang et al., 2010), with frequent occurrence over 80
76 ppbv in plumes from intercontinental pollution, fires, and stratospheric intrusions (Heald
77 et al., 2003; Nowak et al., 2004; Bertsch and Jaffe, 2005; Liang et al., 2007; Thompson
78 et al., 2007; Oltmans et al., 2010). Subsidence of this high-ozone air to the surface could
79 result in PRB values approaching or exceeding the NAAQS (Jaffe, 2011). However,
80 ozone decreases during entrainment into the boundary layer because of dilution,
81 deposition, and chemical loss (Fiore et al., 2002; Hudman et al., 2004; L. Zhang et al.,
82 2009; Cooper et al., 2011).

83
84 A region of particular interest from a PRB perspective is the intermountain West,
85 extending between the Sierra Nevada/Cascades Mountains to the west and the Rocky
86 Mountains to the east. This region features elevated plateaus and mountains with surface
87 elevations typically in excess of 1.5 km, arid terrain, and large-scale subsidence. As a
88 result, background ozone there is higher than in the eastern US (Lefohn et al., 2001; Fiore
89 et al., 2002; Jaffe, 2011). Exceptionally high ozone events have been observed in
90 association with stratospheric intrusions (Langford et al., 2009). Positive correlations
91 have been observed between ozone and regional wildfires (Jaffe et al., 2008; Jaffe, 2011).
92 There is evidence that ozone inflow from the Pacific to the western US has been
93 increasing over the past decades (Lin et al., 2000; Jaffe and Ray, 2007; Parrish et al.,
94 2009; Cooper et al., 2010). This could reflect increasing Siberian wildfires (Jaffe et al.,
95 2004) and Asian pollution (L. Zhang et al., 2008; Cooper et al., 2010).

96
97 The PRB ozone is not an observable quantity, if only because of the contribution of North
98 American anthropogenic sources to the northern mid-latitudes ozone background. It
99 needs to be calculated with a global model of atmospheric composition that is evaluated
100 with observations at remote sites where the PRB drives much of the variability. Fiore et
101 al. (2003) previously used the GEOS-Chem CTM with $2^\circ \times 2.5^\circ$ global horizontal
102 resolution to estimate PRB ozone over the US. H. Wang et al. (2009) updated those
103 estimates by using a $1^\circ \times 1^\circ$ nested continental-scale version of GEOS-Chem, and also
104 estimated the US background ozone (defined by zeroing anthropogenic US emissions).
105 Here we present a further update of PRB, US background, and natural background ozone
106 estimates with a 3-year (2006-2008) GEOS-Chem simulation at $1/2^\circ \times 2/3^\circ$ resolution
107 featuring a number of improvements over previous versions. Our motivation for this
108 work is to assist the EPA in its revision of the ozone NAAQS, scheduled to be released in
109 2014. We include a detailed model evaluation in the intermountain West where elevated
110 PRB is of particular relevance to the NAAQS.

111 112 **2. Model description**

113
114 We use the GEOS-Chem 3-D global model of atmospheric composition (v8-02-03;
115 <http://acmg.seas.harvard.edu/geos/>). The model is driven by GEOS-5 assimilated

116 meteorological fields from the NASA Global Modeling and Assimilation Office
117 (GMAO). Meteorological fields in the GEOS-5 data have a temporal resolution of 6
118 hours (3 hours for surface variables and mixing depths) and a horizontal resolution of
119 $1/2^\circ$ latitude by $2/3^\circ$ longitude. GEOS-Chem includes a detailed simulation of
120 tropospheric ozone- NO_x -VOC-aerosol chemistry. The ozone simulation over the US and
121 adjacent oceans has been previously evaluated with measurements from surface sites
122 (Fiore et al., 2002, 2003; Goldstein et al., 2004; H. Wang et al., 2009), aircraft (Hudman
123 et al., 2007; L. Zhang et al., 2008; Walker et al., 2010), ozonesondes (Li et al., 2002,
124 2005), and satellites (Parrington et al., 2008; L. Zhang et al., 2010). L. Zhang et al.
125 (2010) found in particular that the GEOS-Chem simulation for 2006 is unbiased in the
126 middle troposphere at northern mid-latitudes compared with ozonesondes and satellite
127 measurements.

128
129 We use a nested version of GEOS-Chem (Y.X. Wang et al., 2004; Chen et al., 2009; Y.X.
130 Wang et al., 2011) with the native $1/2^\circ \times 2/3^\circ$ horizontal resolution over North America
131 and adjacent oceans (140° - 40° W, 10° - 70° N), and $2^\circ \times 2.5^\circ$ horizontal resolution for the
132 rest of the world. We first conduct the global GEOS-Chem simulation at $2^\circ \times 2.5^\circ$
133 resolution, and then use the output archived at 3-hour temporal resolution as dynamic
134 boundary conditions for the nested model.

135
136 Global anthropogenic emissions are from the Emission Database for Global Atmospheric
137 Research (EDGAR) inventory (Olivier and Berdowski, 2001) for 2000, superseded by
138 regional emission inventories from the EPA 2005 National Emission Inventory (NEI-05)
139 for the US, Q. Zhang et al. (2009) for Asia in 2006, the European Monitoring and
140 Evaluation Program (EMEP) for Europe (Vestreng and Klein, 2002), the Canada Criteria
141 Air Contaminants (CAC) emission inventory for Canada
142 (http://www.ec.gc.ca/pdb/cac/cac_home_e.cfm), and the Big Bend Regional Aerosol and
143 Visibility Observational (BRAVO) emission inventory for Mexico (Kuhns et al., 2005).
144 The EMEP, CAC, and BRAVO emissions are scaled on the basis of energy statistics to
145 2006 as described by van Donkelaar et al. (2008). We doubled the Japanese and Korean
146 NO_x anthropogenic emissions from Q. Zhang et al. (2009) as constrained by OMI
147 tropospheric NO_2 column measurements (L. Zhang et al. 2008). Anthropogenic NO_x
148 emissions from fertilizer application are from Yienger and Levy (1995).

149
150 Natural sources of ozone include open fires, lightning, the biosphere, and transport from
151 the stratosphere. We use monthly biomass burning emissions from the Global Fire
152 Emission Database version 2 (GFED-v2) (van der Werf et al., 2006). Lightning NO_x
153 emissions are linked to deep convection following the parameterization of Price and Rind
154 (1992) with vertical profiles from Pickering et al. (1998). The global spatial distribution
155 of lightning flashes is rescaled to match the 10-year climatology of OTD/LIS satellite
156 observations (Sauvage et al., 2007) with higher NO_x yield per flash at northern mid-
157 latitudes than in the tropics (Hudman et al., 2007). The global lightning source is imposed
158 to be 6 Tg N a^{-1} (Martin et al., 2007). Soil NO_x emissions are computed using a modified
159 version of the Yienger and Levy (1995) algorithm with canopy reduction factors as
160 described in Y. Wang et al. (1998). Stratospheric ozone is simulated with a linearized

161 ozone (Linoz) parameterization (McLinden et al., 2000) that provides a mechanistic
162 representation of stratospheric influences on tropospheric ozone.

163
164 We conducted three-year GEOS-Chem simulations for 2006-2008. The standard
165 simulation includes all sources and is used for evaluation with observations. We also
166 conducted simulations with: (1) zero North American anthropogenic emissions (North
167 American background or PRB) for 2006-2008, (2) zero US anthropogenic emissions (US
168 background) for 2006, and (3) zero anthropogenic emissions worldwide and methane set
169 to its 700 ppbv pre-industrial value (natural background) for 2006. All ozone
170 concentrations presented in this paper are daily 8-h average maxima (daily 8-h max), the
171 metric used for the US NAAQS. We find that the US background is on average 1-3 ppbv
172 higher than the North American background, reflecting anthropogenic sources in Canada
173 and Mexico, with little variability except in border regions. Our results for the US
174 background are similar to those reported in the focused GEOS-Chem analysis of H.
175 Wang et al. (2009) and hence we do not discuss them further.

176 177 **3. Evaluation with observations in the intermountain West**

178
179 We evaluated our GEOS-Chem simulation with the nationwide ensemble of surface
180 ozone observations from the Clean Air Status and Trends Network (CASTNet;
181 <http://www.epa.gov/castnet>), which monitors air quality in rural areas. The CASTNet
182 sites are shown in Figure 1. Comparison results are generally consistent with the previous
183 GEOS-Chem evaluations presented by Fiore et al. (2003) and H. Wang et al. (2009); time
184 series for individual sites and summary statistics for each region are given in the
185 Supplementary Materials. We focus here on the 12 sites in the intermountain west US,
186 identified in Figure 1 and listed in Table 1, for the year 2006. Interannual variability for
187 seasonal mean concentrations at individual sites is weak during 2006-2008 in both model
188 and observations, generally less than 2 ppbv. However, interannual variation in the
189 number of exceedance days for thresholds of 65-75 ppbv is larger as discussed in Jaffe
190 (2011).

191
192 Figure 2 shows the time series of observed vs. simulated daily 8-h max ozone
193 concentrations in spring-summer 2006 at four representative sites in the intermountain
194 West, and Figure 3 shows scatterplots for the ensemble of sites. Spring-summer is when
195 concentrations are highest. Also shown in the figures are the North American
196 background (PRB) and natural background values. Mean values and correlation
197 coefficients for the simulated vs. observed ozone time series at all 12 sites are
198 summarized in Table 1. We find that seasonal mean ozone concentrations in the model
199 are generally within ± 2 ppbv of the observations in Table 1. The correlation coefficients
200 between model and observations are only 0.2-0.5 for the individual sites, which may
201 reflect the small dynamic range of variability in the observations. The correlation
202 coefficient is 0.6 in spring and 0.3 in summer for the ensemble of sites in Figure 3.

203
204 The North American background (PRB) averages 39-44 ppbv (spring) and 35-45 ppbv
205 (summer) for the ensemble of sites in the intermountain West and drives most of the day-
206 to-day variability. The North American anthropogenic enhancement (difference between

207 standard and PRB simulations) averages only 10-22 ppbv depending on the site. As
208 shown in Figure 3, PRB increases with increasing ozone concentration in the
209 intermountain West, whereas for surface sites in the East there is little correlation of PRB
210 with ozone (see Supplementary Materials). The natural background is on average 25-28
211 ppbv (spring) and 25-33 ppbv (summer) and is strongly correlated with the PRB. The
212 model difference between the PRB and natural ozone reflects intercontinental pollution
213 influences plus anthropogenic methane. It averages 13-16 ppbv in spring and 11-13 ppbv
214 in summer. Intercontinental pollution influence on ozone is larger in spring because of
215 stronger winds and slower chemical loss (Jacob et al., 1999). Annual 4th-highest ozone
216 values at the intermountain West sites are 51-59 ppbv for PRB and 34-45 ppbv for the
217 natural background.

218
219 It is of particular interest to evaluate the ability of the model to reproduce the frequencies
220 of exceedance of proposed air quality standards. Figure 4 shows the simulated vs.
221 observed number of days at individual sites when daily 8-h max ozone concentrations
222 exceed thresholds of 60, 65, and 70 ppbv in spring and summer 2006. The model captures
223 most of the ozone exceedances except for the 65 and 70 ppbv thresholds in spring and for
224 the Mesa Verde site in summer. There is observational evidence that stratospheric
225 intrusions cause high-ozone events in the intermountain West in spring (Langford et al.,
226 2009) and these may not be properly represented in the model. The Mesa Verde model
227 overestimate may reflect excessive summer lightning NO_x emissions over Mexico and
228 the US Southwest.

229
230 When the ensemble of sites is considered as in Figure 3, we find that the model can
231 provide an unbiased ozone simulation up to about 75 ppbv but fails to reproduce
232 exceptional events of higher concentrations. Such events are illustrated in Figure 2 for
233 Pinedale (80 ppbv) and Rocky Mountain NP (91 ppbv). This may reflect a general
234 difficulty in Eulerian models to preserve the structure of plumes of dimensions
235 comparable to the grid resolution (Rastigeyev et al., 2010).

236 237 **4. Distribution of background ozone and contribution to pollution episodes.**

238
239 Figure 5 shows the simulated and observed frequency distributions of ozone for the
240 ensemble of CASTNet sites in the US in March-August 2006, separately for low-altitude
241 (< 1.5 km) and high-altitude sites. Also shown are the model frequency distributions for
242 North American (PRB) and natural ozone backgrounds. The model is unbiased in its
243 simulation of the overall distribution. The PRB averages 27 ± 8 ppbv at the low-altitude
244 sites and 40 ± 7 ppbv at the high-altitude sites. The natural background averages 18 ± 6
245 ppbv at the low-altitude sites and 27 ± 6 ppbv at the high-altitude sites. The difference
246 between PRB and natural background reflects intercontinental pollution influence
247 including anthropogenic methane; it averages 9 ppbv at the low-altitude sites and 13 ppbv
248 at the high-altitude sites. The mean 2006 value of the annual 4th-highest daily 8-h max
249 ozone is 54 ppbv for PRB and 39 ppbv for the natural background at the ensemble of
250 high-altitude sites, compared with 42 ppbv for PRB and 29 ppbv for the natural
251 background at the low-altitude sites.

252

253 Our general PRB statistics for the US can be compared to the previous GEOS-Chem
254 studies of Fiore et al. (2003) and H. Wang et al. (2009). H. Wang et al. (2009) found a
255 mean PRB of 26 ± 8 ppbv for summer 2001, whereas we find 30 ± 10 ppbv for summers
256 2006-2008. Fiore et al. (2003) reported a typical PRB range of 15-35 ppbv for March-
257 October 2001 using a slightly different metric (mean afternoon concentrations). Our
258 results are overall about 4 ppbv higher than these previous estimates. A contributing
259 factor is our higher lightning NO_x source, 6 Tg a^{-1} as compared to 4.5 Tg N a^{-1} in H.
260 Wang et al. (2009). Another factor is the 2001-2006 increase in Asian anthropogenic NO_x
261 emissions, which we previously estimated to have increased PRB ozone by up to 3 ppbv
262 in the West in spring (L. Zhang et al., 2008). We also find some dependence on the
263 model resolution, as our outer nest with $2^\circ \times 2.5^\circ$ horizontal resolution yields mean PRB
264 values that are 1-2 ppbv lower than the nested simulation. Our results are consistent with
265 those of Parrington et al. (2009), who found a 5 ppbv increase in background ozone in the
266 western US compared to Fiore et al. (2002) after assimilation of TES satellite ozone data
267 into the GEOS-Chem model.

268
269 Figure 6 shows the spatial distribution of the seasonal mean PRB ozone concentrations
270 for spring and summer 2006. The PRB is highest in the intermountain West because of
271 the combination of high elevation, deep boundary layer mixing, large-scale subsidence,
272 slow ozone deposition to the arid terrain, and slow ozone chemical loss due to dry
273 conditions (Fiore et al., 2002). The PRB ozone generally decreases from spring to
274 summer, reflecting faster chemical ozone loss; this seasonal decrease is particularly
275 pronounced in the Northeast and on the West Coast. An increase in PRB from spring to
276 summer is found in the Southwest due to summer lightning. The maximum PRB value
277 over New Mexico in summer reflects intense lightning and deep boundary layer mixing.

278
279 Also shown in Figure 6 is the mean PRB ozone on the days when simulated daily 8-h
280 max ozone exceeds 60 ppbv. We find that the mean PRB on these high-ozone days is
281 higher than the seasonal mean almost everywhere. The difference is particularly
282 pronounced in the West, where the PRB is on average 7 ppbv higher than the seasonal
283 mean for both spring and summer. It is also large in the Great Lakes region in spring
284 where high ozone values are associated with model lightning. Fiore et al. (2002)
285 previously found PRB values to be maximum for ozone concentrations in the 50-70 ppbv
286 range, and this is consistent with our result. If the NAAQS is lowered in the 60-70 ppbv
287 range, areas of the intermountain West will have little or no ability to reach compliance
288 through North American regulatory controls.

289
290 Finally, we show in Figure 7 the simulated annual 4th-highest North American
291 background (PRB) ozone in surface air averaged over 2006-2008, representing the lowest
292 air quality standard that can be achieved by North American emission controls. Values
293 are typically 35-45 ppbv in the East and on the West Coast but 50-60 ppbv in the
294 intermountain West, with a maximum of 64 ppbv over New Mexico and a secondary
295 maximum of 59 ppbv over Idaho due to large wildfires in 2007. A recent study with the
296 CMAQ regional model found much larger contributions from wildfires on surface ozone
297 in the western US (Mueller and Mallard, 2011). Aircraft observations of California fire

298 plumes indicate however no significant ozone enhancements unless mixed with urban
299 pollution (Singh et al., 2010).

300

301 **5. Conclusions**

302

303 We have used the GEOS-Chem global 3-D model of atmospheric composition with $1/2^\circ$
304 $\times 2/3^\circ$ nested horizontal resolution over North America to provide updated estimates of
305 the PRB ozone for the US in 2006-2008. Our work is intended to assist the US EPA in its
306 current risk and exposure assessments as part of the NAAQS-setting process.

307

308 We evaluated the GEOS-Chem simulation with the ensemble of ozone observations from
309 CASTNet sites across the US. Comparisons show in general similar results to previous
310 GEOS-Chem PRB studies (Fiore et al., 2003; H. Wang et al., 2009) and are documented
311 in the Supplementary Materials. We focused our attention on the intermountain West,
312 where the PRB is particularly high and may interfere with the achievability of ozone air
313 quality standards. We showed that the model gives an unbiased representation of ozone in
314 that region and that the PRB drives most of the ozone variability. The model captures the
315 frequency of high-ozone events up to about 70 ppbv but fails to reproduce events of
316 exceptionally high ozone that may be due to stratospheric or wildfire influences. We
317 expect following Rastigeyev et al. (2009) that Eulerian models in general would have
318 difficulty in capturing exceptional events.

319

320 We obtained mean PRB values for the US in spring-summer of 27 ± 8 ppbv at low-
321 altitude sites (< 1.5 km) and 40 ± 7 ppbv at high-altitude sites. These values are 9-13
322 ppbv higher than the natural background due to intercontinental pollution including
323 anthropogenic methane. Our PRB estimates are on average 4 ppbv higher than in
324 previous GEOS-Chem studies (Fiore et al., 2003; H. Wang et al., 2009) and we attribute
325 this to a combination of increasing Asian emissions, higher model lightning, and higher
326 model resolution. We find that the PRB generally decreases from spring to summer
327 except in regions strongly affected by summer lightning. We also find that the PRB is
328 higher than average when ozone exceeds 60 ppbv, particularly in the intermountain West.
329 The annual 4th-highest PRB value in the model (representing the minimum standard
330 achievable through suppression of North American anthropogenic emissions) is typically
331 in the 35-45 ppbv range over the East and the West Coast but 50-60 ppbv in the
332 intermountain West. Such high PRB values in the intermountain West compared to the
333 proposed revisions of the ozone NAAQS (60-70 ppbv) suggest that special consideration
334 of that region may be needed in the NAAQS-setting process.

335

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339

340 **References**

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549 **Figure captions**

550 **Figure 1.** CASTNet ozone monitoring sites in the continental United States for 2006.
551 Sites in the intermountain West (Table 1) are indicated in red. Pluses denote sites above
552 1.5 km altitude.

553
554 **Figure 2.** March-August 2006 time series of daily 8-h max ozone concentrations at four
555 representative sites in the US intermountain West. Model results (red line) are compared
556 with observations (black line). Also shown is the North American background or PRB
557 (blue line) and the natural background (green line). The mean concentrations for the time
558 period and annual 4th-highest ozone values (in parentheses) in ppbv are shown inset.
559

560 **Figure 3.** Simulated vs. observed daily 8-h max ozone concentrations for spring (March-
561 May) and summer (June-August) 2006 at the 12 intermountain West CASTNet sites of
562 Table 1. Also shown is the 1:1 line. The box-and-whisker plots (minimum, 25th, 50th, 75th
563 percentile, and maximum) give statistics of the North American background (PRB) and
564 natural background for 10-ppbv bins of observed ozone concentrations.
565

566 **Figure 4.** Simulated (GEOS-Chem) vs. observed number of days with daily 8-h max
567 ozone concentrations exceeding thresholds of 60, 65, and 70 ppbv in spring and summer
568 2006 at the 12 CASTNet sites in the intermountain West (Table 1). Symbols identify the
569 individual sites.

570
571 **Figure 5.** Frequency distributions of daily 8-h max ozone concentrations in March-
572 August 2006 for the ensemble of low-altitude (<1.5 km) and high-altitude CASTNet sites
573 in the US (Figure 1). Model results (red) are compared to observations (black). Also
574 shown are frequency distributions for the North American background (solid blue) and
575 natural background (dashed green).
576

577 **Figure 6.** North American background (PRB) ozone concentration in surface air for
578 spring and summer 2006. The top panels show seasonal means while the bottom panels
579 show the means for days with ozone > 60 ppbv. Gray areas in the bottom panels had no
580 days with ozone > 60 ppbv.

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582 **Figure 7.** Annual 4th-highest value of North American background ozone (PRB)
583 calculated in GEOS-Chem as daily 8-h max and averaged for 2006-2008.
584

1 **Table 1.** Ozone concentrations at CASTNet monitoring sites in the US intermountain West ^a

Sites ^b	<i>r</i>	Spring		Summer		Annual 4 th highest	
		Observed	GEOS-Chem (PRB) ^c	Observed	GEOS-Chem (PRB)	Observed	GEOS-Chem (PRB)
Yellowstone N.P., WY (44.6N, 110.4W, 2.4 km)	0.40	56.7	52.7 (40.8)	56.0	47.2 (35.3)	69.6	61.4 (51.3)
Pinedale, WY (42.9N, 109.8W, 2.4 km)	0.48	56.5	54.8 (41.9)	57.3	54.1 (38.6)	68.4	65.7 (53.5)
Centennial, WY (41.4N, 106.2W, 3.2 km)	0.20	59.5	54.9 (42.4)	56.1	56.1 (40.1)	70.4	66.6 (52.9)
Rocky Mountain NP, CO (40.3N, 105.6W, 2.8 km)	0.33	56.5	57.9 (44.6)	59.8	61.7 (39.4)	76.1	77.4 (55.3)
Gothic, CO (38.9N, 107.0W, 2.9 km)	0.21	58.8	56.0 (44.4)	53.9	55.4 (40.0)	70.0	65.3 (55.5)
Mesa Verde N.P., CO (37.2N, 108.5W, 2.2 km)	0.31	58.5	57.9 (44.4)	61.2	68.3 (45.6)	74.4	79.6 (58.1)
Great Basin N.P., NV (39.0N, 114.2W, 2.1 km)	0.52	54.5	52.8 (41.2)	58.9	59.2 (40.6)	72.2	72.6 (52.6)
Canyonlands N.P., UT (38.5N, 109.8W, 1.8 km)	0.36	56.6	56.3 (43.4)	59.7	60.3 (42.1)	70.6	70.9 (56.1)
Grand Canyon N.P., AZ (36.1N, 112.2W, 2.1 km)	0.38	58.8	56.6 (43.8)	58.8	60.6 (42.7)	70.8	69.9 (56.4)
Petrified Forest, AZ (34.8N, 109.9W, 1.7 km)	0.57	56.7	55.4 (42.5)	61.5	61.7 (43.3)	71.5	75.2 (57.9)
Chiricahua NM, AZ (32.0N, 109.4W, 1.6 km)	0.41	54.7	53.8 (43.1)	56.5	61.3 (45.1)	74.0	72.3 (58.9)
Big Bend NP, TX (29.3N, 103.2W, 1.1 km)	0.49	52.4	51.3 (39.3)	48.6	54.6 (40.8)	65.3	65.0 (52.3)

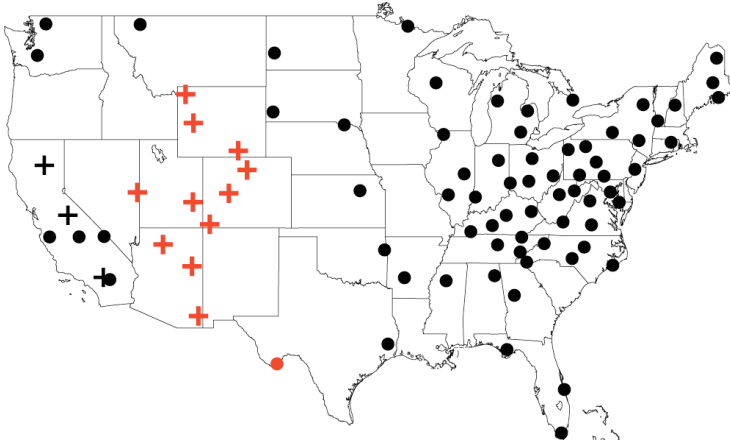
2 ^a Seasonal mean and annual 4th-highest daily 8-h max ozone values in ppbv for 2006 and correlation coefficients (*r*) between model and
3 observations for the daily data. Spring is March-April and summer is June-August.

4 ^b NP = National Park, NM = National Monument, WY = Wyoming, CO = Colorado, NV = Nevada, UT = Utah, AZ = Arizona, TX = Texas.

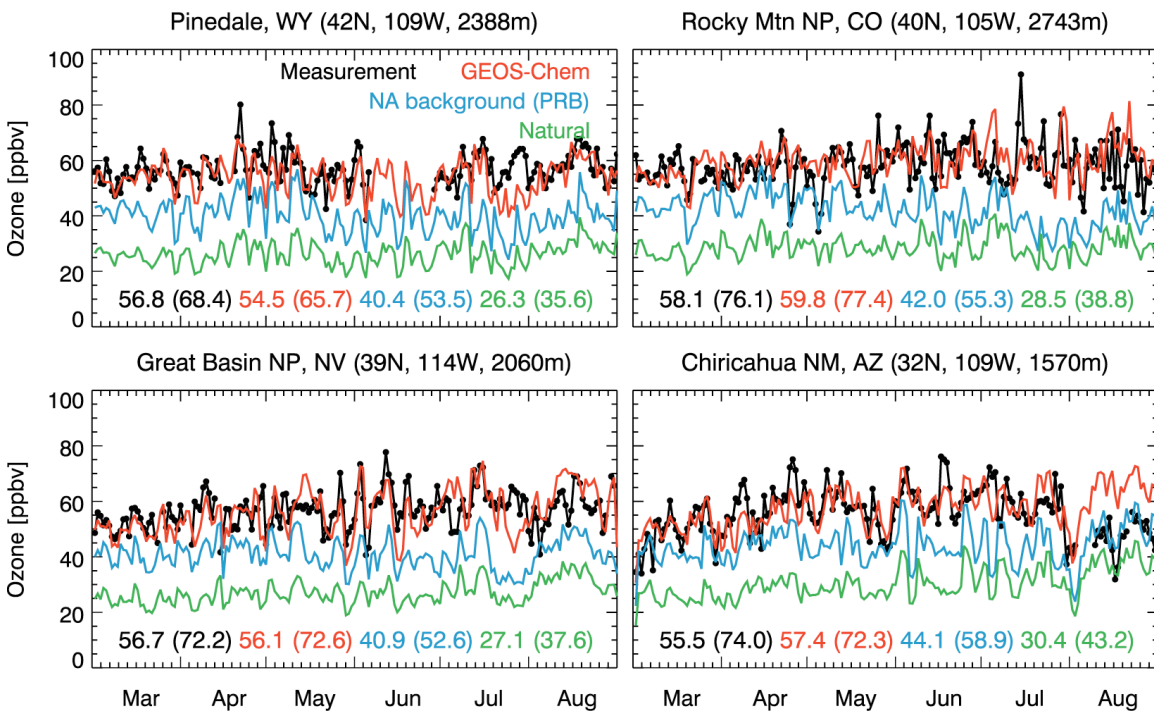
5 ^c GEOS-Chem values in parentheses are the policy-relevant background ozone (PRB) as determined by a simulation with zero North
6 American anthropogenic emissions.

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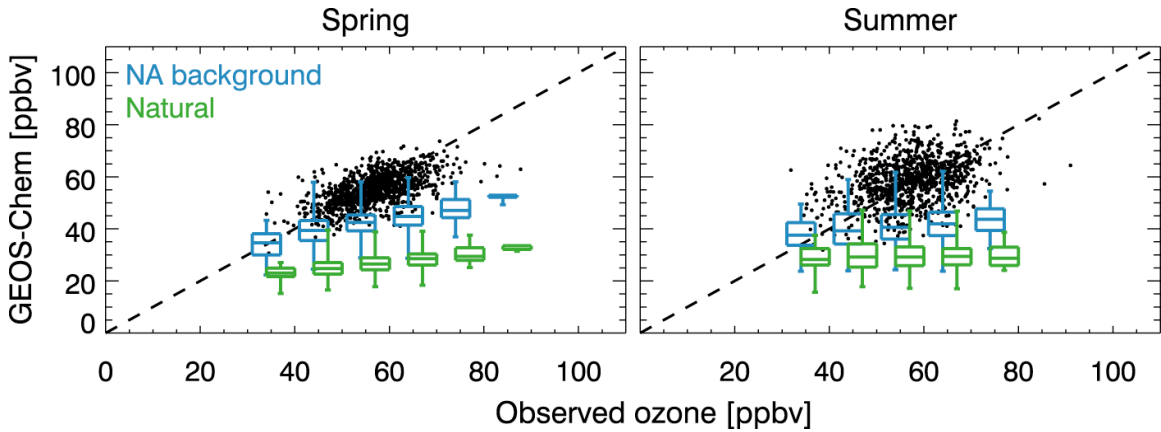
1 **Figures**
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 5 **Figure 1.** CASTNet ozone monitoring sites in the continental United States for 2006.
 6 Sites in the intermountain West (Table 1) are indicated in red. Pluses denote sites above
 7 1.5 km altitude.
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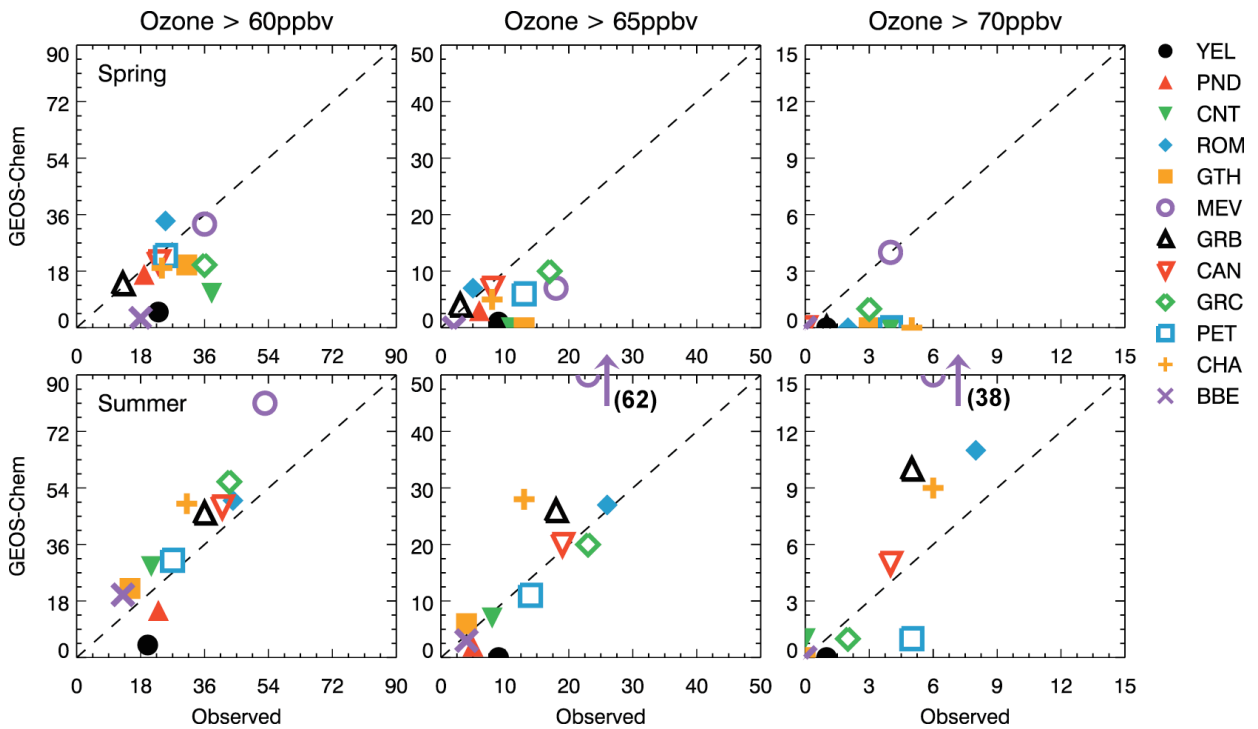


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 13 **Figure 2.** March-August 2006 time series of daily 8-h max ozone concentrations at four
 14 representative sites in the US intermountain West. Model results (red line) are compared
 15 with observations (black line). Also shown is the North American background or PRB
 16 (blue line) and the natural background (green line). The mean concentrations for the time
 17 period and annual 4th-highest ozone values (in parentheses) in ppbv are shown inset.



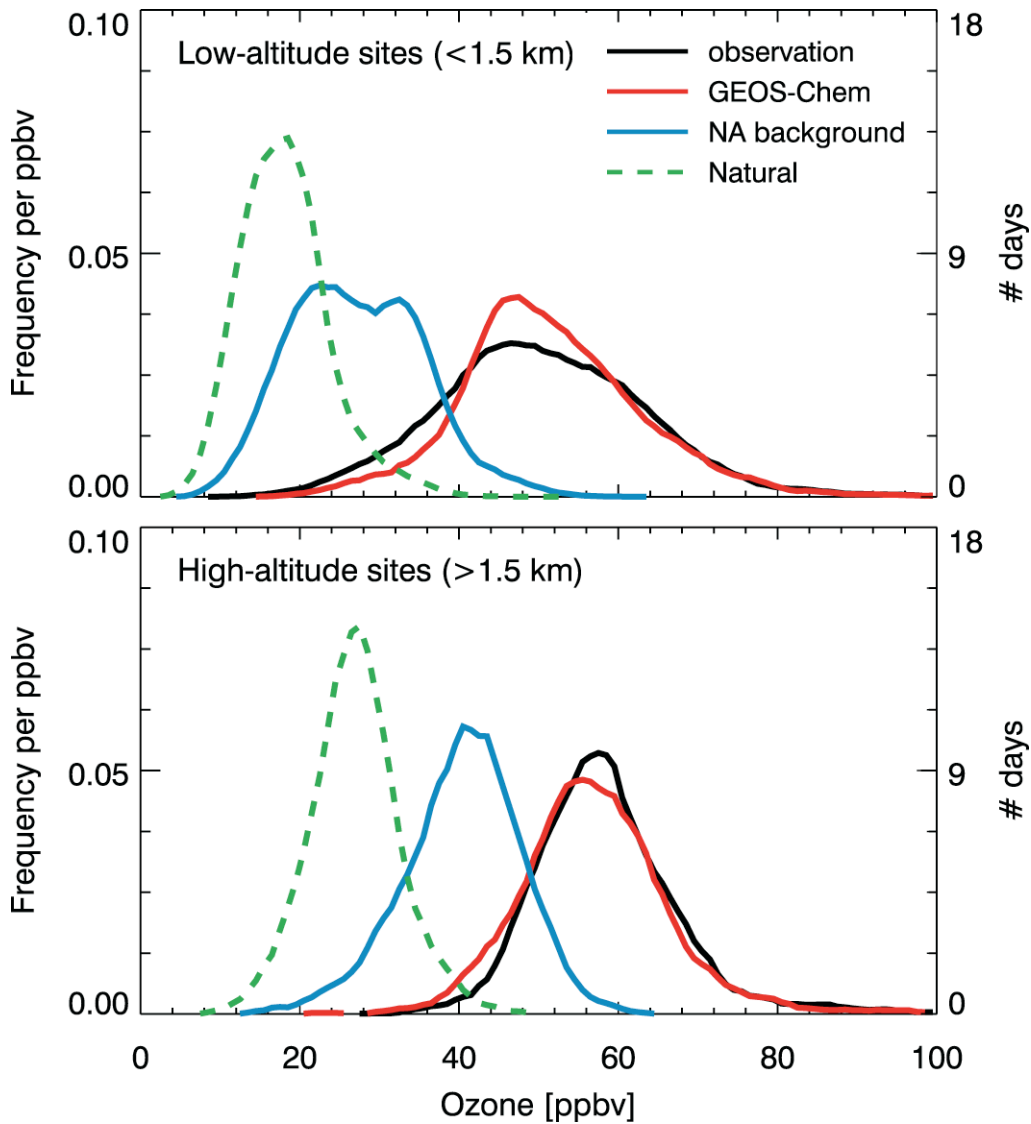
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Figure 3. Simulated vs. observed daily 8-h max ozone concentrations for spring (March-May) and summer (June-August) 2006 at the 12 intermountain West CASTNet sites of Table 1. Also shown is the 1:1 line. The box-and-whisker plots (minimum, 25th, 50th, 75th percentile, and maximum) give statistics of the North American background (PRB) and natural background for 10-ppbv bins of observed ozone concentrations.



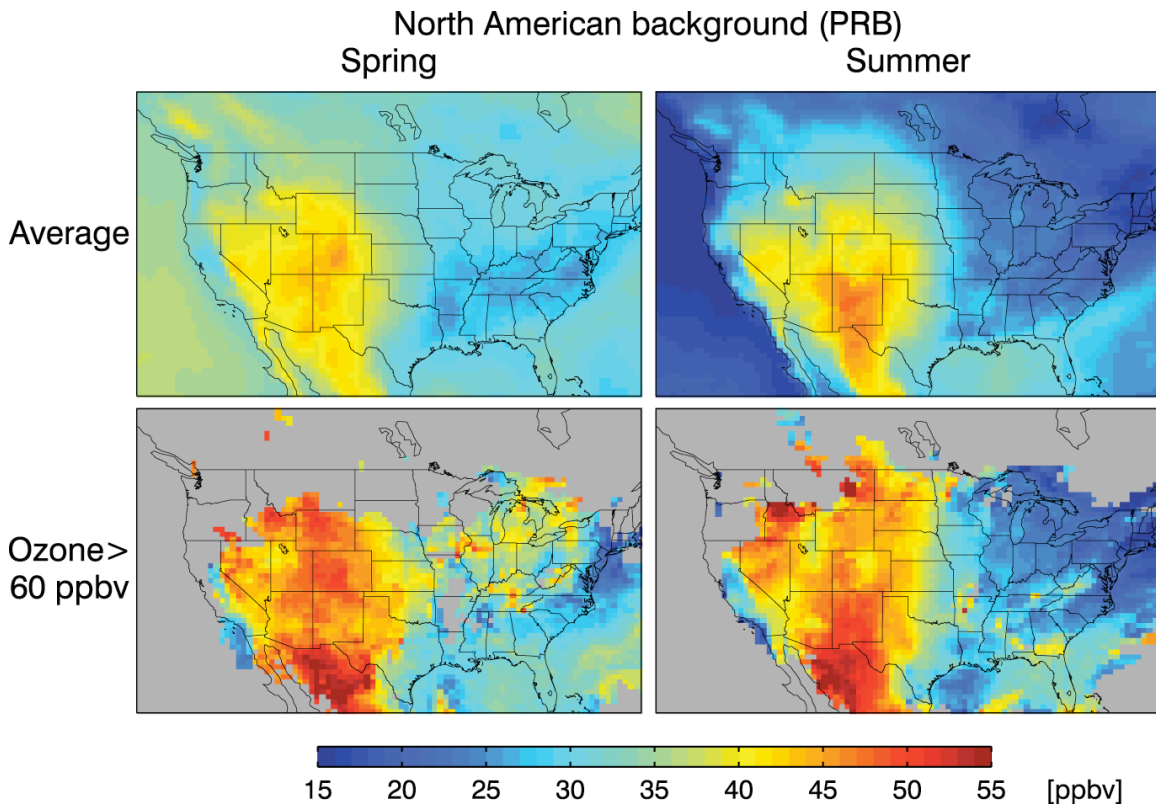
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Figure 4. Simulated (GEOS-Chem) vs. observed number of days with daily 8-h max ozone concentrations exceeding thresholds of 60, 65, and 70 ppbv in spring and summer 2006 at the 12 CASTNet sites in the intermountain West (Table 1). Symbols identify the individual sites.



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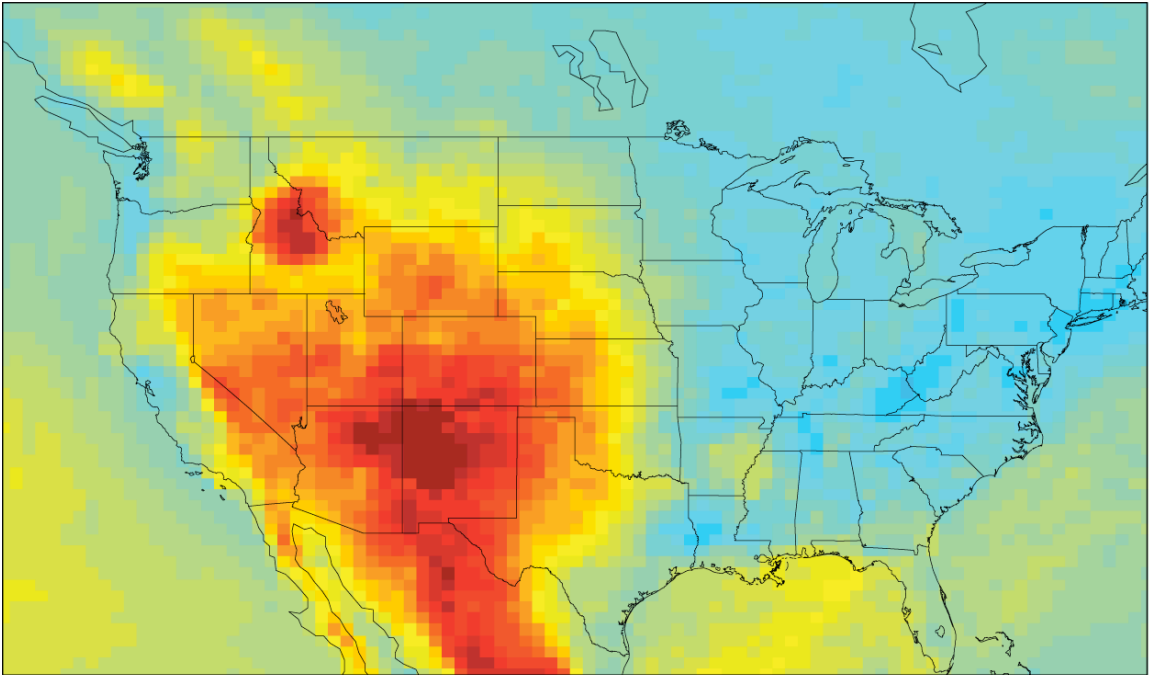
Figure 5. Frequency distributions of daily 8-h max ozone concentrations in March-August 2006 for the ensemble of low-altitude (<1.5 km) and high-altitude CASTNet sites in the US (Figure 1). Model results (red) are compared to observations (black). Also shown are frequency distributions for the North American background (solid blue) and natural background (dashed green).



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Figure 6. North American background (PRB) ozone concentration in surface air for spring and summer 2006. The top panels show seasonal means while the bottom panels show the means for days with ozone > 60 ppbv. Gray areas in the bottom panels had no days with ozone > 60 ppbv.

Annual 4th highest PRB ozone for 2006-2008



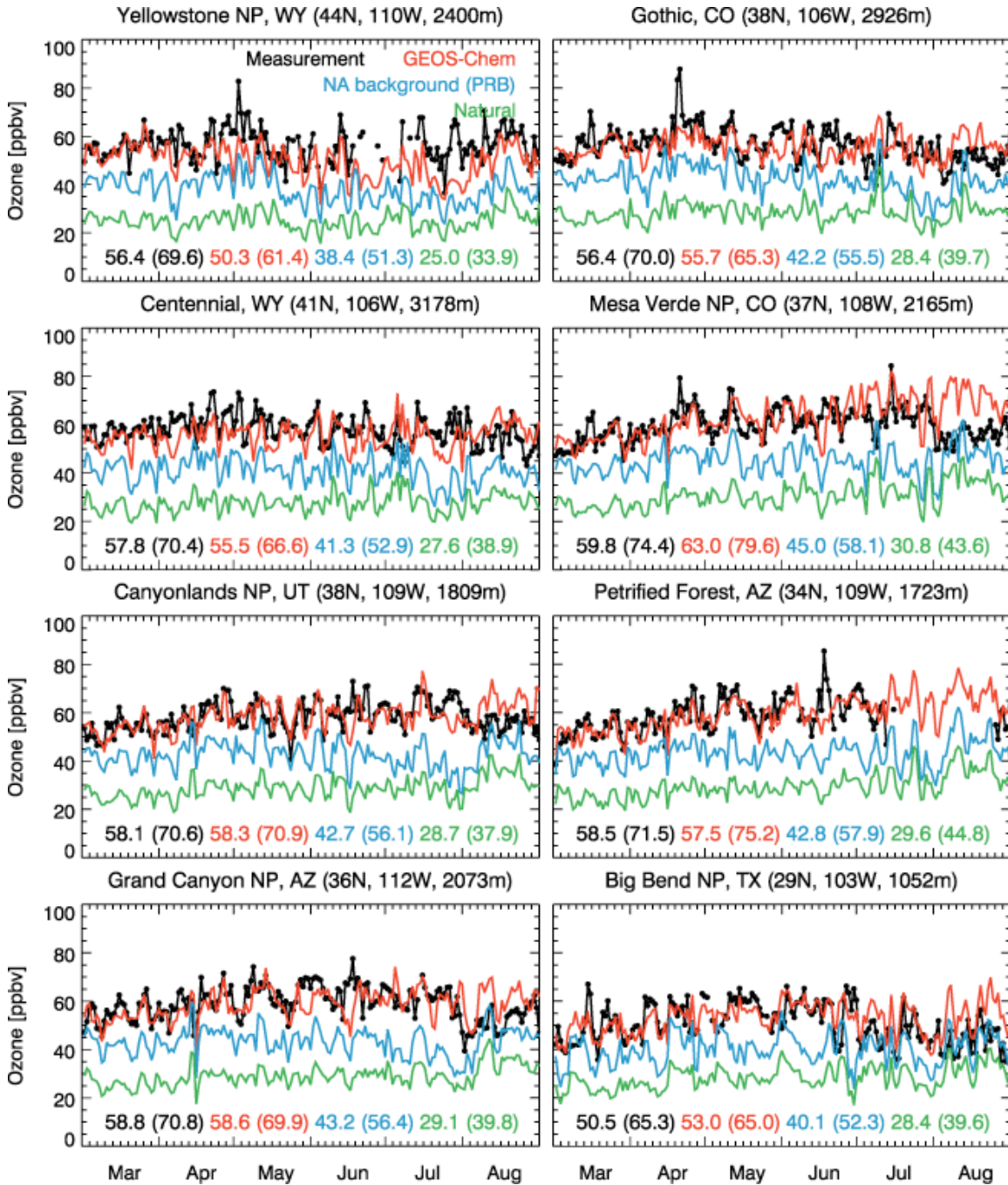
25 30 35 40 45 50 55 60 [ppbv]

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Figure 7. Annual 4th-highest value of North American background ozone (PRB) calculated in GEOS-Chem as daily 8-h max and averaged for 2006-2008.

1 Supplemental Material

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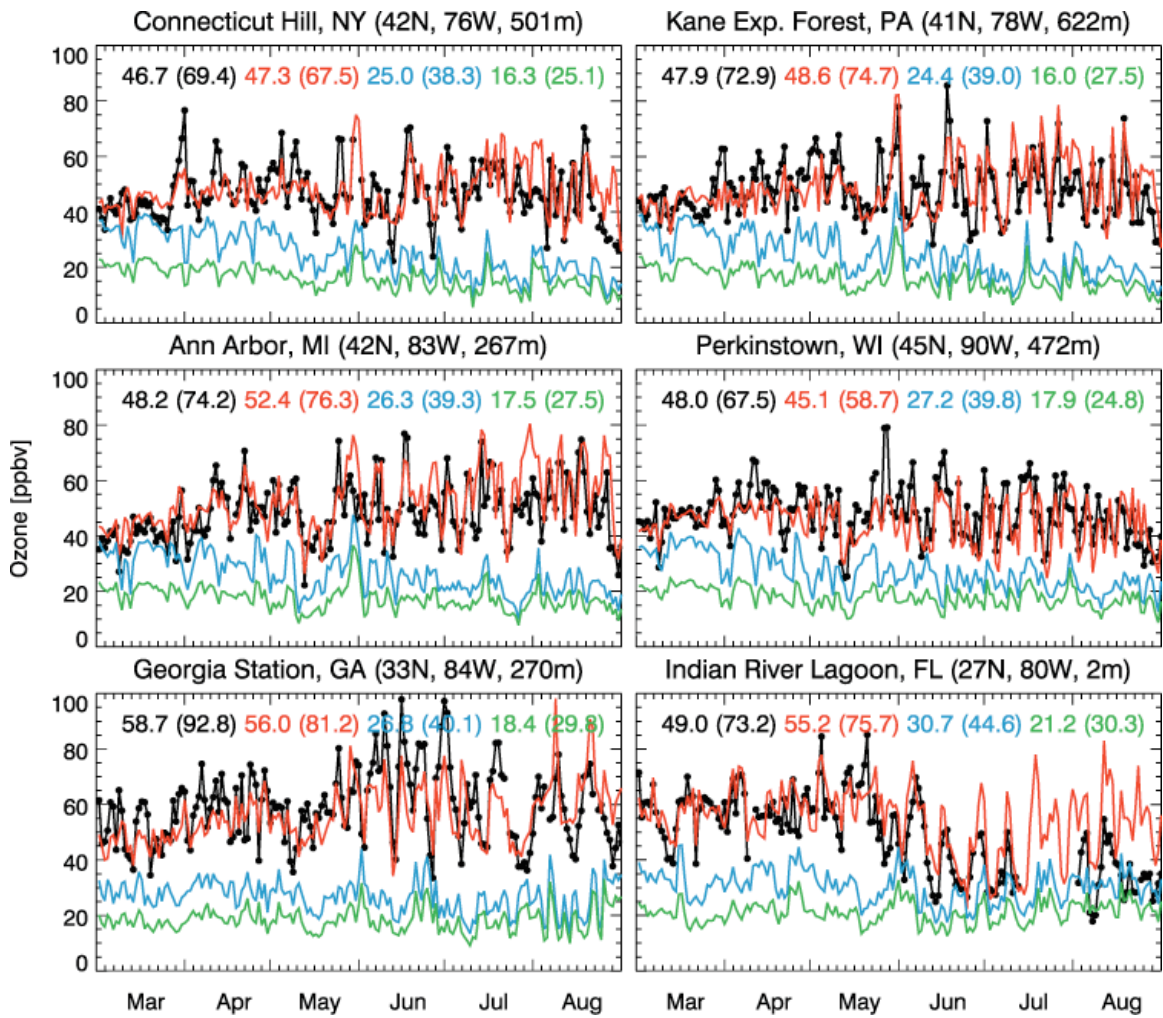


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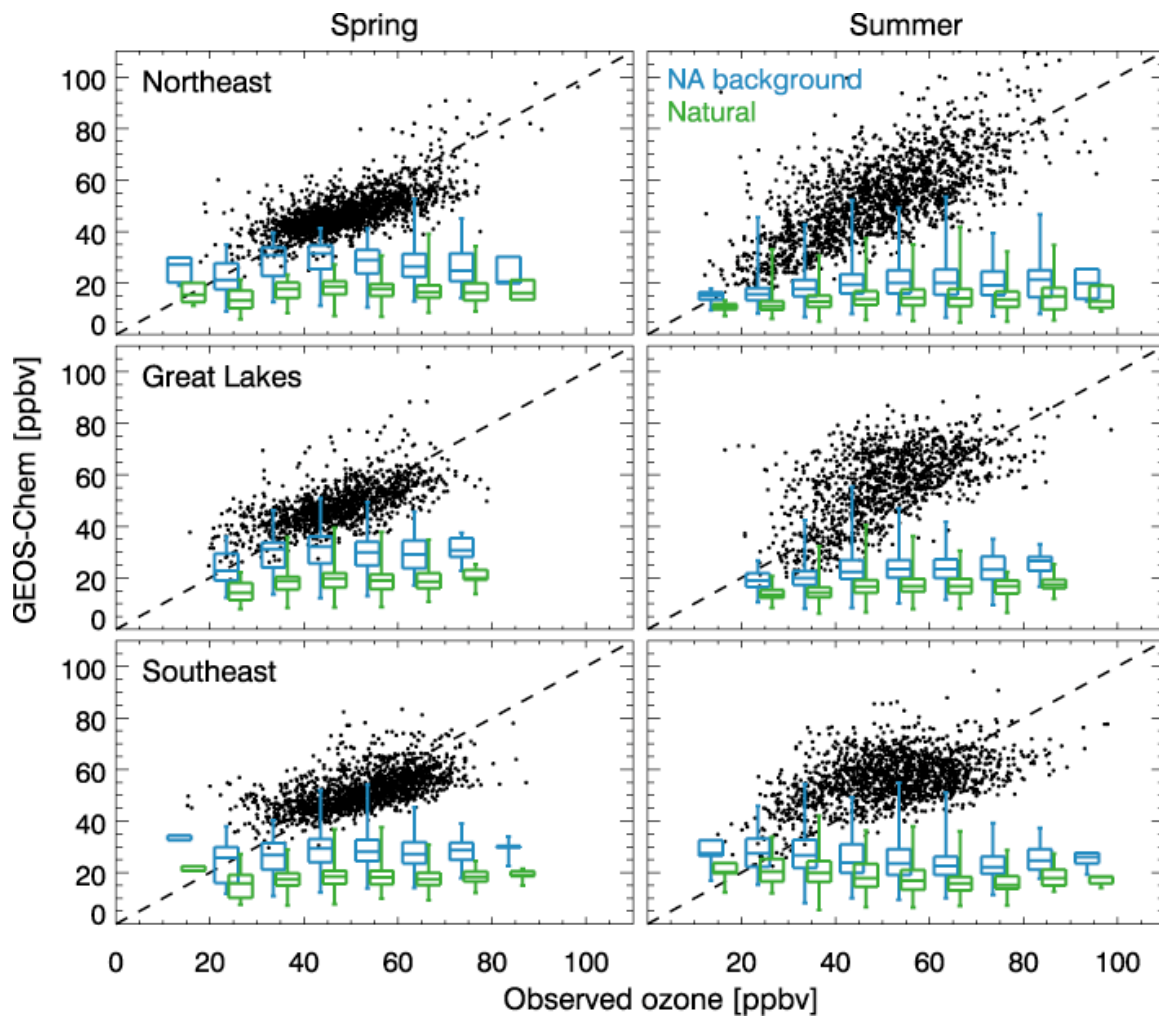
5 **Figure S1.** March-August 2006 time series of daily 8-h max ozone concentrations at the
 6 additional intermountain West sites of Table 1. Model results (red line) are compared
 7 with observations (black line). Also shown is the North American background or PRB
 8 (blue line) and the natural background (green line). The mean concentrations for the time
 9 period and annual 4th-highest ozone values (in parentheses) in ppbv are shown inset.

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Figure S2. The same as Figure S1, but for representative CASTNet sites in the Northeast US (top), Great Lakes (middle), and the Southeast US (bottom).



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Figure S3. Simulated vs. observed daily 8-h max ozone concentrations for spring (March-May) and summer (June-August) 2006 for the ensembles of CASTNet sites in the Northeast US, Great Lakes, and the Southeast US. Also shown is the 1:1 line and North American background (PRB) and natural background model statistics for 10-ppbv bins of observed ozone concentrations: the minimum, 25th, 50th, 75th percentile, and maximum.