

# 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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## ABSTRACT

The policy-relevant background (PRB) ozone is defined by the US Environmental Protection Agency (EPA) as the surface ozone concentration that would be present over the US in the absence of North American anthropogenic emissions. It is intended to provide a baseline for risk and exposure assessments used in setting the National Ambient Air Quality Standard (NAAQS). We present here three-year statistics (2006–2008) of PRB ozone over the US calculated using the GEOS-Chem global 3-D model of atmospheric composition with  $1/2^\circ \times 2/3^\circ$  horizontal resolution over North America and adjacent oceans ( $2^\circ \times 2.5^\circ$  for the rest of the world). We also provide estimates of the US background (no anthropogenic US emissions) and natural background (no anthropogenic emissions worldwide and pre-industrial methane). Our work improves on previous GEOS-Chem PRB estimates through the use of higher model resolution, 3-year statistics, better representation of stratospheric influence, and updated emissions. PRB is particularly high in the intermountain West due to high elevation, arid terrain, and large-scale subsidence. We present for this region a detailed model evaluation showing that the model is successful in reproducing ozone exceedances up to 70 ppbv. However, the model cannot reproduce PRB-relevant exceptional events associated with wildfires or stratospheric intrusions. The mean PRB estimates for spring–summer are  $27 \pm 8$  ppbv at low-altitude sites and  $40 \pm 7$  ppbv at high-altitude sites. Differences between the PRB simulation and the natural simulation indicate a mean enhancement from intercontinental pollution and anthropogenic methane of 9 ppbv at low-altitude sites and 13 ppbv at high-altitude sites. The PRB is higher than average when ozone exceeds 60 ppbv, particularly in the intermountain West. Our PRB estimates are on average 4 ppbv higher than previous GEOS-Chem studies and we attribute this to higher lightning, increasing Asian emissions, and improved model resolution. Whereas previous studies found no occurrences of PRB exceeding 60 ppbv, we find here some occurrences in the intermountain West. The annual 4th-highest PRB values in the intermountain West are typically 50–60 ppbv, as compared to 35–45 ppbv in the East or on the West Coast. Such high PRB values in the intermountain West suggest that special consideration of this region may be needed if the ozone NAAQS is decreased to a value in the 60–70 ppbv range.

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## 1. Introduction

The US Environmental Protection Agency (US EPA, 2006) defines the policy-relevant-background (PRB) for ozone air quality as the surface ozone concentration that would be present in the US in the

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absence of anthropogenic emissions from North America (defined as the ensemble of the US, Canada, and Mexico). The PRB is used in the setting of the National Ambient Air Quality Standard (NAAQS) to estimate the maximum ozone reduction that could be achieved through North American emission controls. It provides a baseline for assessing risk and exposure from ozone pollution. The present US NAAQS is 75 ppbv as the annual 4th-highest daily maximum 8-h average concentration averaged over three years, but the EPA is considering decreasing it to a value in the 60–70 ppbv range. As the standard becomes more stringent and approaches the PRB, accurate specification of the PRB becomes increasingly important.

Ozone is produced in the troposphere by photochemical oxidation of CO and volatile organic compounds (VOCs) in the presence of nitrogen oxides ( $\text{NO}_x \equiv \text{NO} + \text{NO}_2$ ). It is also transported to the troposphere from the stratosphere. The  $\text{NO}_x$ , CO, and VOC precursors of ozone have major anthropogenic sources from fuel combustion as well as natural sources including lightning, wildfires, and the biosphere. Ozone has a lifetime of only a few days in the continental boundary layer but weeks in the free troposphere (Y. Wang et al., 1998; Fiore et al., 2002). Ozone sonde, aircraft, and satellite observations show typical ozone concentrations of 50–70 ppbv in the free troposphere over North America (Thompson et al., 2007; Zhang et al., 2010), with frequent occurrences above 80 ppbv in plumes from intercontinental pollution, fires, and stratospheric intrusions (Heald et al., 2003; Nowak et al., 2004; Bertschi and Jaffe, 2005; Liang et al., 2007; Thompson et al., 2007; Oltmans et al., 2010). Subsidence of this high-ozone air from the troposphere to the surface could result in PRB values approaching or exceeding the NAAQS (Jaffe, 2011). However, ozone decreases during entrainment into the boundary layer because of dilution, deposition, and chemical loss (Fiore et al., 2002; Hudman et al., 2004; Zhang et al., 2009a; Cooper et al., submitted for publication).

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

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

intermountain West where elevated PRB is of particular relevance to the NAAQS.

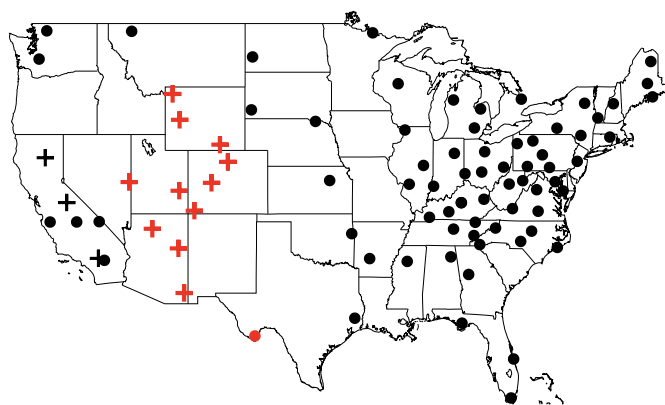
## 2. Model description

We use the GEOS-Chem 3-D global model of atmospheric composition (v8-02-03; <http://acmg.seas.harvard.edu/geos/>) driven by GEOS-5 assimilated meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO) for 2006–2008. The GEOS-5 data have a temporal resolution of 6 h (3 h for surface variables and mixing depths) and a horizontal resolution of  $1/2^\circ$  latitude by  $2/3^\circ$  longitude. GEOS-Chem includes a detailed simulation of tropospheric ozone– $\text{NO}_x$ –VOC–aerosol chemistry. The ozone simulation over the US and adjacent oceans has been previously evaluated with measurements from surface sites (Fiore et al., 2002, 2003; Goldstein et al., 2004; Wang et al., 2009), aircraft (Hudman et al., 2007; Zhang et al., 2008; Walker et al., 2010), ozonesondes (Li et al., 2002, 2005), and satellites (Parrington et al., 2008; Zhang et al., 2010). Zhang et al. (2010) found in particular that the GEOS-Chem simulation for 2006 is unbiased in the middle troposphere at northern mid-latitudes compared with ozonesondes and satellite measurements.

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

Global anthropogenic emissions are from the Emission Database for Global Atmospheric Research (EDGAR) inventory (Olivier and Berdowski, 2001), superseded by regional emission inventories from the EPA 2005 National Emission Inventory (NEI-05) for the US, Zhang et al. (2009b) for Asia in 2006, the European Monitoring and Evaluation Program (EMEP) for Europe (Vestreng and Klein, 2002), the Canada Criteria Air Contaminants (CAC) emission inventory for Canada ([http://www.ec.gc.ca/pdb/cac/cac\\_home\\_e.cfm](http://www.ec.gc.ca/pdb/cac/cac_home_e.cfm)), and the Big Bend Regional Aerosol and Visibility Observational (BRAVO) emission inventory for Mexico (Kuhns et al., 2005). The EDGAR, EMEP, CAC, and BRAVO emissions are scaled on the basis of energy statistics to 2006 as described by van Donkelaar et al. (2008). We doubled the Japanese and Korean  $\text{NO}_x$  anthropogenic emissions from Zhang et al. (2009b) as constrained by OMI tropospheric  $\text{NO}_2$  column measurements (L. Zhang et al., 2008). Anthropogenic  $\text{NO}_x$  emissions from fertilizer application are from Yienger and Levy (1995).

Natural sources of ozone include open fires, lightning, the biosphere, and transport from the stratosphere. We use monthly biomass burning emissions from the Global Fire Emission Database version 2 (GFED-v2) (van der Werf et al., 2006). Lightning  $\text{NO}_x$  emissions are linked to deep convection following the parameterization of Price and Rind (1992) with vertical profiles from Pickering et al. (1998). The global spatial distribution of lightning flashes is rescaled to match the 10-year climatology of OTD/LIS satellite observations (Sauvage et al., 2007) with higher  $\text{NO}_x$  yield per flash at northern mid-latitudes than in the tropics (Hudman et al., 2007). The global lightning source is imposed to be  $6 \text{ Tg N a}^{-1}$  (Martin et al., 2007). Soil  $\text{NO}_x$  emissions are computed using a modified version of the Yienger and Levy (1995) algorithm with canopy reduction factors as described in Y. Wang et al. (1998). Stratospheric ozone is simulated with a linearized ozone (Linoz) parameterization (McLinden et al., 2000) that provides a mechanistic representation of stratospheric influences on tropospheric ozone.



**Fig. 1.** CASTNet ozone monitoring sites in the continental United States for 2006. Sites in the intermountain West (Table 1) are indicated in red. Pluses denote sites above 1.5 km altitude. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

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

### 3. Evaluation with observations in the intermountain West

We evaluated our GEOS-Chem simulation with the nationwide ensemble of surface ozone observations from the Clean Air Status and Trends Network (CASTNet; <http://www.epa.gov/castnet>), which monitors air quality in rural areas. The CASTNet sites are shown in Fig. 1. Comparison results are generally consistent with

the previous GEOS-Chem evaluations presented by Fiore et al. (2003) and Wang et al. (2009); time series for individual sites and summary statistics for each region are given in the Supplementary Materials. We focus here on the 12 sites in the intermountain west US, identified in Fig. 1 and listed in Table 1, for the year 2006. Interannual variability for seasonal mean concentrations at individual sites is weak during 2006–2008 in both model and observations, generally less than 2 ppbv. There is larger interannual variability in the occurrence of high-ozone events as discussed by Jaffe (2011).

Fig. 2 shows the time series of observed vs. simulated daily 8-h max ozone concentrations in spring–summer 2006 at four representative sites in the intermountain West, and Fig. 3 shows scatterplots for the ensemble of 12 sites. Spring–summer is when concentrations are highest. Also shown in the figures are the North American background (PRB) and natural background values. Mean values and correlation coefficients for the simulated vs. observed ozone time series at all 12 sites are summarized in Table 1. We find that seasonal mean ozone concentrations in the model are generally within  $\pm 2$  ppbv of the observations in Table 1. The correlation coefficients between model and observations are only 0.2–0.5 for the individual sites. Much of the variability in the observations is on a high-frequency (day-to-day) basis (Fig. 2), and the 6-h temporal resolution of the GEOS-5 meteorological data may limit the ability of the model to reproduce this variability. The correlation coefficient is 0.6 in spring and 0.3 in summer for the ensemble of sites in Fig. 3.

The North American background (PRB) averages 39–44 ppbv (spring) and 35–45 ppbv (summer) for the ensemble of sites in the intermountain West and drives 20–54% of the day-to-day variability in the model. The North American anthropogenic enhancement (difference between standard and PRB simulations) averages only 10–22 ppbv depending on the site. As shown in Fig. 3, PRB increases with increasing ozone concentration in the intermountain West, whereas for surface sites in the East there is little correlation of PRB with ozone (see Supplementary Materials). The natural background is on average 25–28 ppbv (spring) and 25–33 ppbv (summer) and is strongly correlated with the PRB. The model difference between the PRB and natural ozone reflects intercontinental pollution influences plus anthropogenic methane. It averages 13–16 ppbv in spring and 11–13 ppbv in summer. Intercontinental pollution influence on ozone is larger in spring because of stronger winds and slower chemical loss (Jacob et al., 1999). Annual 4th-highest ozone values at the intermountain

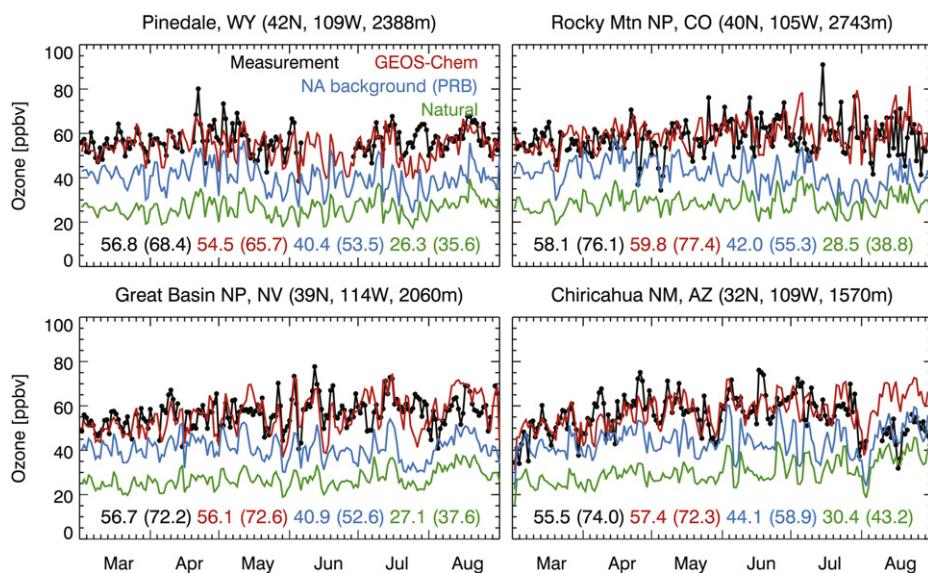
**Table 1**  
Ozone concentrations at CASTNet monitoring sites in the US intermountain West.<sup>a</sup>

Sites <sup>b</sup>	r	Spring		Summer		Annual 4th-highest	
		Observed	GEOS-Chem (PRB) <sup>c</sup>	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)

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

<sup>b</sup> NP = National Park, NM = National Monument, WY = Wyoming, CO = Colorado, NV = Nevada, UT = Utah, AZ = Arizona, TX = Texas.

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



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

West sites are 51–59 ppbv for PRB and 34–45 ppbv for the natural background.

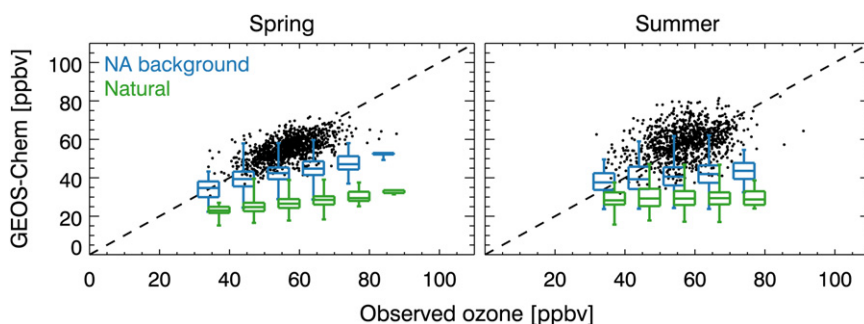
It is of particular interest to evaluate the ability of the model to reproduce the frequencies of exceedance of proposed air quality standards. Fig. 4 shows the simulated vs. observed number of days at individual sites when daily 8-h max ozone concentrations exceed thresholds of 60, 65, and 70 ppbv in spring and summer 2006. The model is successful in capturing the frequencies of exceedance at the individual sites except for the 65 and 70 ppbv thresholds in spring and for the Mesa Verde site in summer. There is observational evidence that stratospheric intrusions cause high-ozone events in the intermountain West in spring (Langford et al., 2009) and these may not be properly represented in the model. The Mesa Verde model overestimate may reflect excessive summer lightning  $\text{NO}_x$  emissions over Mexico and the US Southwest.

When the ensemble of sites is considered as in Fig. 3, we find that the nested model with  $1/2^\circ \times 2/3^\circ$  horizontal resolution has more success in reproducing high-ozone events up to 75 ppbv than the  $2^\circ \times 2.5^\circ$  simulation (outer nest) or previous GEOS-Chem work (Fiore et al., 2003). But it still fails to reproduce exceptional events of higher concentrations. Such events are illustrated in Fig. 2 for Pinedale (80 ppbv) and Rocky Mountain NP (91 ppbv).

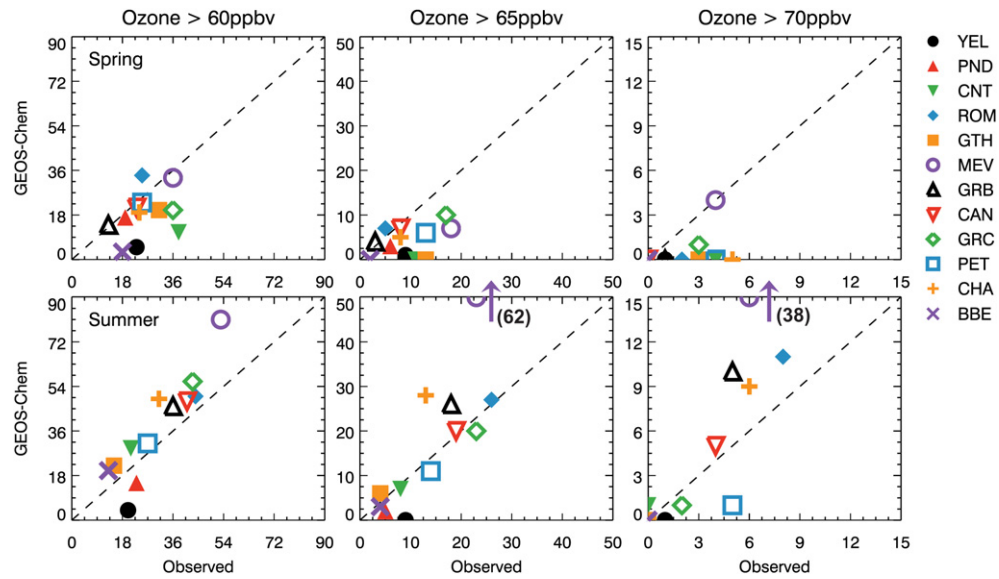
#### 4. Distribution of background ozone and contribution to pollution episodes

Fig. 5 shows the simulated and observed frequency distributions of ozone for the ensemble of CASTNet sites in the US in March–August 2006, separately for low-altitude (<1.5 km) and high-altitude sites. Also shown are the model frequency distributions for North American (PRB) and natural ozone backgrounds. The model is unbiased in its simulation of the overall distribution. The PRB averages  $27 \pm 8$  ppbv at the low-altitude sites and  $40 \pm 7$  ppbv at the high-altitude sites. The natural background averages  $18 \pm 6$  ppbv at the low-altitude sites and  $27 \pm 6$  ppbv at the high-altitude sites. The difference between PRB and natural background reflects intercontinental pollution influence plus anthropogenic methane; it averages 9 ppbv at the low-altitude sites and 13 ppbv at the high-altitude sites. The mean 2006 value of the annual 4th-highest daily 8-h max ozone is 54 ppbv for PRB and 39 ppbv for the natural background at the ensemble of high-altitude sites, compared with 42 ppbv for PRB and 29 ppbv for the natural background at the low-altitude sites.

Our general PRB statistics for the US can be compared to the previous GEOS-Chem studies of Fiore et al. (2003) and Wang et al.



**Fig. 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.



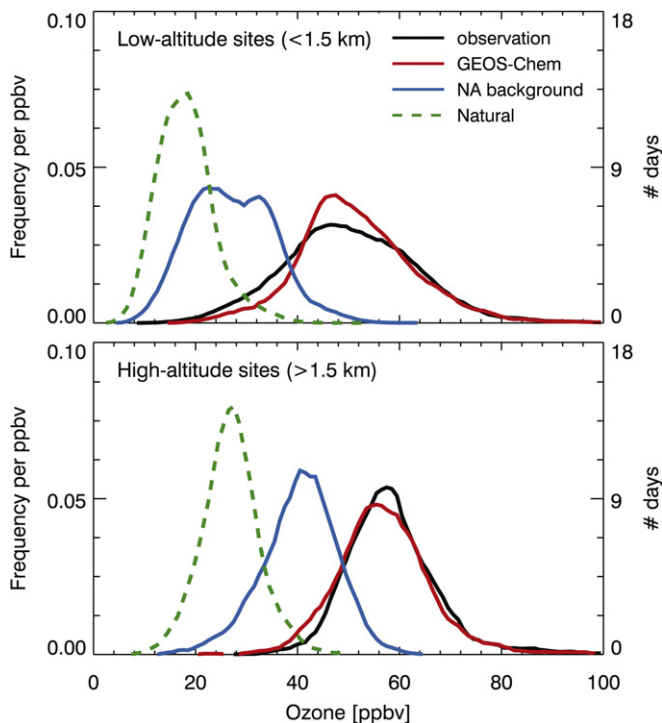
**Fig. 4.** Simulated (GEOS-Chem) vs. observed number of days with daily 8-h max ozone concentrations exceeding thresholds of 60, 65, and 70 ppbv in spring and summer 2006 at the 12 CASTNet sites in the intermountain West (Table 1). Symbols identify the individual sites.

(2009). Wang et al. (2009) found a mean PRB of  $26 \pm 8$  ppbv for summer 2001, whereas we find  $30 \pm 10$  ppbv for summers 2006–2008. Fiore et al. (2003) reported a typical PRB range of 15–35 ppbv for March–October 2001 using a slightly different metric (mean afternoon concentrations). Our results are overall

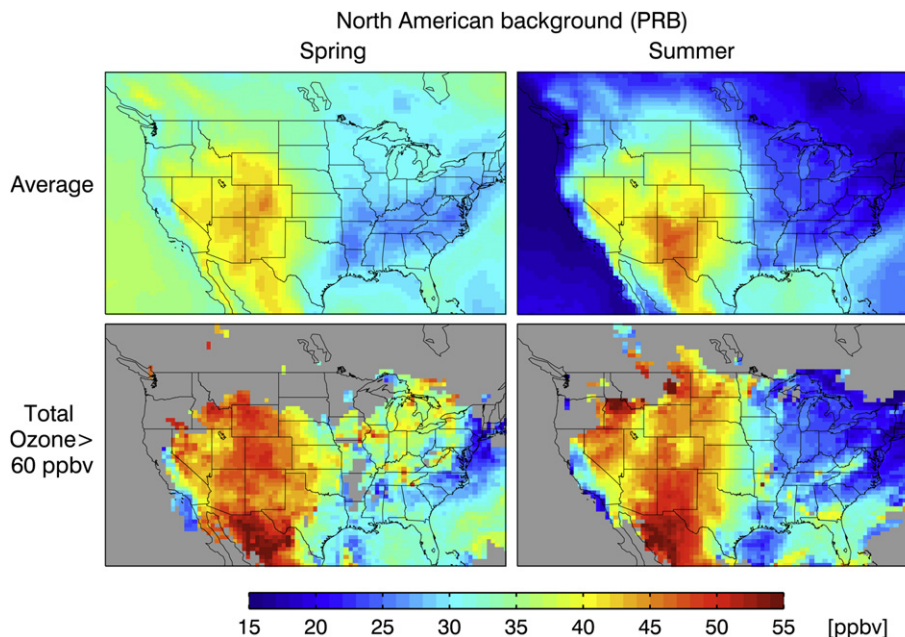
about 4 ppbv higher than these previous estimates. A contributing factor is our higher lightning  $\text{NO}_x$  source,  $6 \text{ Tg a}^{-1}$  as compared to  $4.5 \text{ Tg N a}^{-1}$  in Wang et al. (2009). Another factor is the 2001–2006 increase in Asian anthropogenic  $\text{NO}_x$  emissions, which we previously estimated to have increased PRB ozone by up to 3 ppbv in the West in spring (Zhang et al., 2008). We also find some dependence on the model resolution, as our outer nest with  $2^\circ \times 2.5^\circ$  horizontal resolution yields mean PRB values that are 1–2 ppbv lower than the nested simulation. Wang et al. (2009) found that the PRB never exceeded 60 ppbv, but in our work the PRB occasionally exceeds 60 ppbv in the intermountain West (Fig. 3). Our results are consistent with those of Parrington et al. (2009), who found a 5 ppbv increase in background ozone in the western US compared to Fiore et al. (2002) after assimilation of TES satellite ozone data into the GEOS-Chem model.

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

Also shown in Fig. 6 is the mean PRB ozone on the days when simulated daily 8-h max ozone exceeds 60 ppbv. We find that the mean PRB on these high-ozone days is higher than the seasonal mean almost everywhere. The difference is particularly pronounced in the West, where the PRB is on average 5 ppbv higher than the seasonal mean for both spring and summer. There is also a large difference in the Great Lakes region in spring where high-ozone values are associated with model lightning. Fiore et al. (2002) previously found PRB values to be maximum for ozone concentrations in the 50–70 ppbv range, and this is consistent with our result. If the NAAQS is lowered in



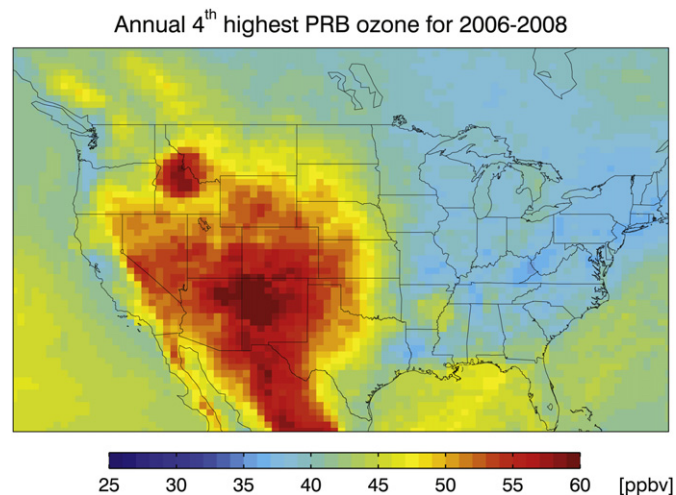
**Fig. 5.** Frequency distributions of daily 8-h max ozone concentrations in March–August 2006 for the ensemble of low-altitude (<1.5 km) and high-altitude CASTNet sites in the US (Fig. 1). Model results (red) are compared to observations (black). Also shown are frequency distributions for the North American background (solid blue) and natural background (dashed green). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)



**Fig. 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 total ozone > 60 ppbv. Gray areas in the bottom panels had no days with total ozone > 60 ppbv.

the 60–70 ppbv range, areas of the intermountain West will have little or no ability to reach compliance through North American regulatory controls.

Finally, we show in Fig. 7 the simulated annual 4th-highest North American background (PRB) ozone in surface air averaged over 2006–2008, representing the lowest air quality standard that can be achieved by North American emission controls. Values are typically 35–45 ppbv in the East and on the West Coast but 50–60 ppbv in the intermountain West, with a maximum of 64 ppbv over New Mexico and a secondary maximum of 59 ppbv over Idaho due to large wildfires. A recent study with the CMAQ regional model found much larger contributions from wildfires on surface ozone in the western US (Mueller and Mallard, 2011). Aircraft observations of California fire plumes indicate however no significant ozone enhancements unless mixed with urban pollution (Singh et al., 2010).



**Fig. 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.

## 5. Conclusions

We have used the GEOS-Chem global 3-D model of atmospheric composition with  $1/2^\circ \times 2/3^\circ$  nested horizontal resolution over North America to provide updated estimates of the PRB ozone for the US in 2006–2008. Our work is intended to assist the US EPA in its current risk and exposure assessments as part of the NAAQS-setting process. It improves on previous GEOS-Chem PRB estimates through the use of higher model resolution, 3-year statistics, better representation of stratospheric influence, and updated emissions.

We evaluated the GEOS-Chem simulation with the ensemble of ozone observations from CASTNet sites across the US. Comparisons in the eastern US show in general similar results to previous GEOS-Chem PRB studies (Fiore et al., 2003; Wang et al., 2009) and are documented in the Supplementary Materials. We focused our discussion on the intermountain West, where the PRB is particularly high and may interfere with the achievability of ozone air quality standards. We showed that the model gives an unbiased representation of ozone in that region and that the PRB drives 20–54% of the ozone variability. The model captures the frequency of high-ozone events up to about 70 ppbv but fails to reproduce events of exceptionally high ozone that may be due to stratospheric or wildfire influences.

We obtained mean PRB values for the US in spring-summer of  $27 \pm 8$  ppbv at low-altitude sites (<1.5 km) and  $40 \pm 7$  ppbv at high-altitude sites. These values are 9–13 ppbv higher than the natural background due to intercontinental pollution plus anthropogenic methane. Our PRB estimates are on average 4 ppbv higher than in previous GEOS-Chem studies (Fiore et al., 2003; Wang et al., 2009) and we attribute this to a combination of increasing Asian emissions, higher model lightning, and higher model resolution. We find that the PRB generally decreases from spring to summer except in regions strongly affected by summer lightning. We also find that the PRB is higher than average when ozone exceeds 60 ppbv, particularly in the intermountain West. The annual 4th-highest PRB value in the model (representing the minimum standard achievable through suppression of North American anthropogenic emissions)

is typically in the 35–45 ppbv range over the East and the West Coast but 50–60 ppbv in the intermountain West. Whereas previous GEOS-Chem studies found no occurrences of PRB ozone exceeding 60 ppbv, we find here some occurrences in the intermountain West. The high PRB values in that region compared to the proposed revisions of the ozone NAAQS (60–70 ppbv) suggest that special consideration may be needed in the NAAQS-setting process.

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## Appendix. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.atmosenv.2011.07.054.

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