Atmospheric Environment 43 (2009) 1310–1319

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Surface ozone background in the United States: Canadian and Mexican pollution influences

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ARTICLE INFO

Article history:
Received 28 May 2008
Received in revised form
20 November 2008
Accepted 30 November 2008

Keywords:
Background ozone
Surface ozone
Air quality standard
Chemical transport model
Global model

ABSTRACT

We use a global chemical transport model (GEOS-Chem) with $1^\circ \times 1^\circ$ horizontal resolution to quantify the effects of anthropogenic emissions from Canada, Mexico, and outside North America on daily maximum 8-hour average ozone concentrations in US surface air. Simulations for summer 2001 indicate mean North American and US background concentrations of 26 ± 8 ppb and 30 ± 8 ppb, as obtained by eliminating anthropogenic emissions in North America vs. in the US only. The US background never exceeds 60 ppb in the model. The Canadian and Mexican pollution enhancement averages 3 ± 4 ppb in the US in summer but can be occasionally much higher in downwind regions of the northeast and southwest, peaking at 33 ppb in upstate New York (on a day with 75 ppb total ozone) and 18 ppb in southern California (on a day with 68 ppb total ozone). The model is successful in reproducing the observed variability of ozone in these regions, including the occurrence and magnitude of high-ozone episodes influenced by transboundary pollution. We find that exceedances of the 75 ppb US air quality standard in eastern Michigan, western New York, New Jersey, and southern California are often associated with Canadian and Mexican pollution enhancements in excess of 10 ppb. Sensitivity simulations with 2020 emission projections suggest that Canadian pollution influence in the Northeast US will become comparable in magnitude to that from domestic power plants.

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1. Introduction

Air quality standards to protect public health from the harmful effects of surface ozone are becoming increasingly stringent. Present standards for ozone daily maximum 8-h average concentrations are 75 ppb in the United States (US EPA, 2008), 65 ppb in Canada, and 55 ppb in the European Union. The European Union also has a dose standard to protect vegetation of 3000 ppb h^{-1} above 40 ppb in daytime during the growing season, corresponding to a mean concentration of about 43 ppb.

Tougher ozone standards increase the importance of long-range transport in determining compliance. Observed ozone concentrations at remote sites in northern mid-latitudes are in the range of 15–50 ppb (Altshuller and Lefohn, 1996; Fiore et al., 2002a; Vingarzan, 2004), representing a significant increment toward the standards. These remote concentrations have increased over the past decades (Lin et al., 2000; Jaffe and Ray, 2007), due at least in part to hemispheric-scale pollution (Fiore et al., 2002a). A number of model studies have examined the effects on US ozone air quality of emissions in Asia (Jacob et al., 1999; Hudman et al., 2004), Europe (Li et al., 2002), and global methane (Fiore et al., 2002b), but there has been little investigation of the nearer-scale international influences from Canada and Mexico. We examine these influences here and the implications for meeting US ozone air quality standards, for both present and projected future (2020) conditions.

Ozone is produced in the troposphere by photochemical oxidation of volatile organic compounds (VOCs) and CO in the presence of nitrogen oxides ($NO_x \equiv NO + NO_2$). Rapid ozone production takes place in polluted regions in summer, at a rate generally determined by the supply of NO_x from fossil fuel combustion. Ozone has a lifetime of only a few days in the continental boundary layer in summer but several weeks in the free troposphere (Wang et al., 1998; Fiore et al., 2002a), so that ozone pollution ventilated from the continental boundary layer can be

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doi:10.1016/j.atmosenv.2008.11.036

Table 1
Anthropogenic NO_x emissions (Tg N a^{-1})[a].

Simulation year	US	Canada	Mexico	Rest of the world
2001	6.7	0.76	0.93	16.0
2020	3.3	0.39	0.61	24.8

[a] July mean emissions scaled to the whole year. Seasonal variation is slight.

Table 2
Model simulations.

Name	Description
1. Standard (2001)	Anthropogenic emissions as described in the text
2. NA background (2001)	Same as 1 but without North American anthropogenic emissions
3. US background (2001)	Same as 1 but without US anthropogenic emissions
4. NA background (2006)	Same as 2 but with 2006 East Asian anthropogenic emissions
5. Standard (2020)	Same as 1 but with 2020 anthropogenic emissions
6. NA background (2020)	Same as 5 but without 2020 North American anthropogenic emissions
7. US background (2020)	Same as 5 but without 2020 US anthropogenic emissions
8. US power plant background (2020)	Same as 5 but without 2020 US power plant emissions

transported on intercontinental scales. Ozone is also produced in the remote troposphere from methane and NO_x, both of which have large anthropogenic sources (Wang et al., 1998; Fiore et al., 2002b; Hudman et al., 2004).

The US Environmental Protection Agency (EPA) (2006) uses the Policy-Relevant-Background (PRB) in its standard-setting process as the surface ozone concentration that would be present in the US in the absence of anthropogenic emissions from North America (defined as the ensemble of the US, Canada, and Mexico). The PRB represents the ozone concentration that is not amenable to reduction under current policy frameworks. It is important for regulatory decisions as it sets the maximum ozone reduction and relative health benefits that can be achieved through North American emission controls. Lefohn et al. (2001) pointed out that ozone concentrations at remote US sites in winter–spring are often in the 50–60 ppb range, so that air quality standards close to those values would be difficult to achieve. However, Fiore et al. (2002a) pointed out that these values are not representative of surface air in summer, Using a global chemical transport model (CTM), they estimated a PRB of 15–35 ppbv in the US during summer with little variability, and demonstrated consistency with observations at remote sites (Fiore et al., 2002a; Goldstein et al., 2004).

An issue in the definition of the PRB is the role of Canadian and Mexican emissions. The US EPA (2006) assumes that these emissions are amenable to control to achieve US air quality objectives, but it is not clear to what degree they actually are. Unlike intercontinental transport that manifests itself mainly through enhancement of the hemispheric ozone background, pollution plumes from Canada and Mexico can be transported in the continental boundary layer to affect US areas immediately downwind. There is to our knowledge no peer-reviewed literature on the spatial extent and the magnitude of this near-field international pollution influence.

We apply here the GEOS-Chem CTM (Bey et al., 2001; Fiore et al., 2002a, 2003) to quantify these different international influences on US ozone concentrations for present-day and for projected 2020 conditions. We distinguish between the 'North American background' used by US EPA (2006) as the PRB, and the 'US background' defined as the ozone concentration that would be present in US surface air in the absence of US anthropogenic emissions (but allowing for Canadian and Mexican emissions). GEOS-Chem results for the North American background (Fiore et al., 2002a) have been used previously by the US EPA (2006) as best estimates of the PRB. For the present application, we use a high-resolution version of GEOS-Chem (1° latitude × 1° longitude) to resolve transport from Canada and Mexico. This version was previously used by Park et al. (2006) to quantify transboundary pollution influences on aerosol concentrations in the US in the context of the Regional Haze Rule (US EPA, 1999).

2. Model description

We use GEOS-Chem v7-02-01 (http://www-as.harvard.edu/chemisry/trop/geos) in a global simulation of tropospheric ozone-NO_x–VOC–aerosol chemistry for year 2001. Meteorological input is from NASA/GEOS-3 assimilated data with a horizontal resolution of 1° latitude × 1° longitude, 48 vertical levels, and a temporal resolution of 6 h (3 h for surface variables and mixing depths). The

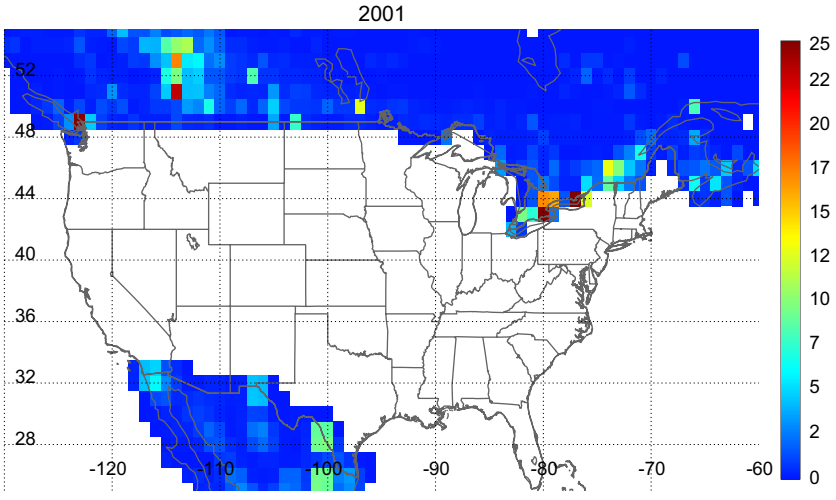

Fig. 1. Canadian and Mexican anthropogenic NO_x emissions (Gg N a^{-1} per 1°x1° grid square) near the US border in the 2001 simulations. Emissions are for July and are scaled to the whole year (per annum units). Canadian emissions are from the CAC inventory for 2002 and Mexican emissions are from the BRAVO inventory for 1999. US emissions in border grid squares are excluded (these come from the NEI 99 inventory in the standard simulation). The BRAVO 1° × 1° inventory grid is offset by 0.5° relative to the GEOS-Chem grid and the required re-mapping introduces some smearing.

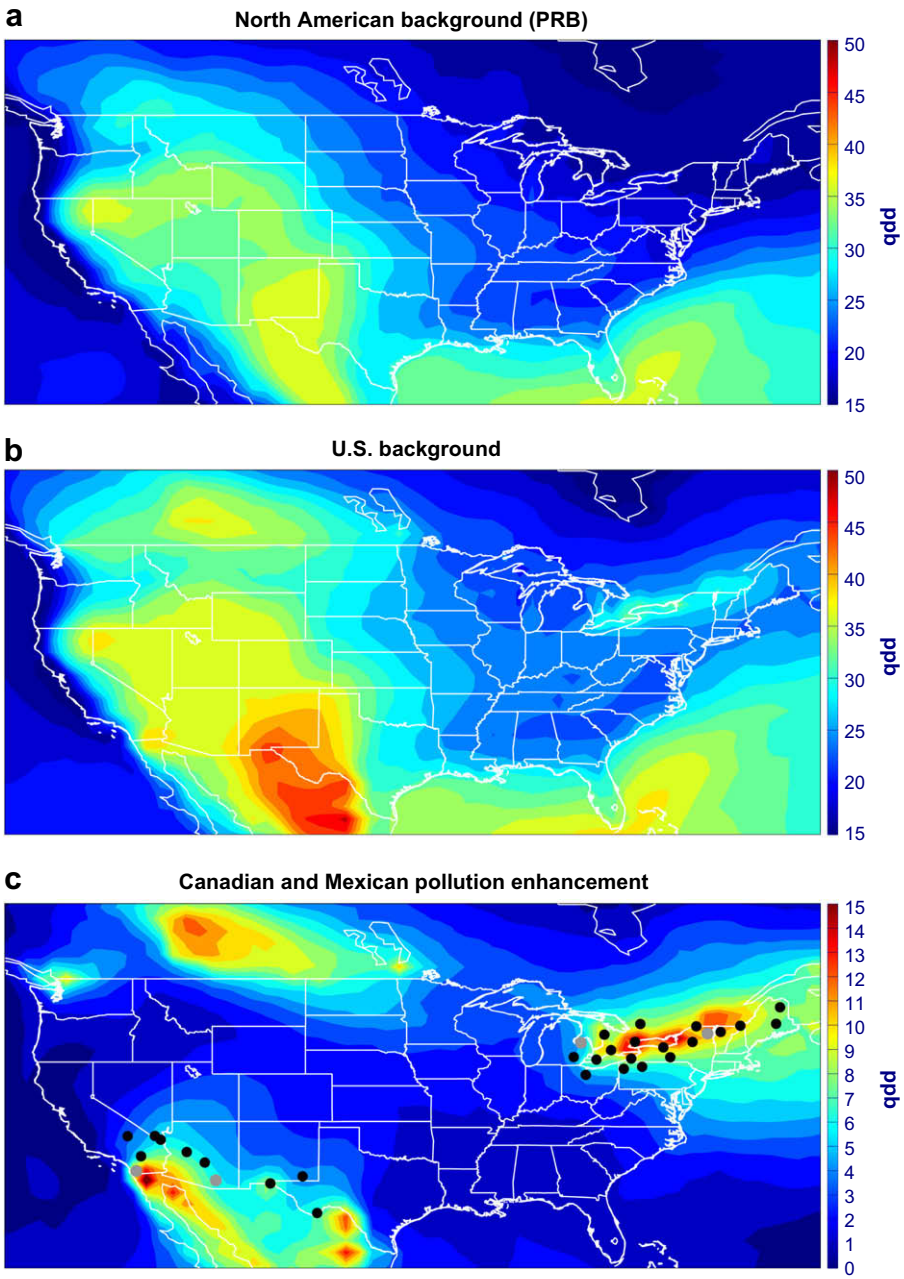

Fig. 2. Jun–Aug mean daily-8 h-max ozone concentrations in surface air for (a) North American background, (b) US background, (c) Canadian and Mexican pollution enhancement (determined as the difference between the US and North American backgrounds). The circles in (c) show the observation sites used for model evaluation. The grey circles identify the sites used in the time series plots of Fig. 4 (time series plots for other sites are included in supplementary materials).

simulation is as described by Park et al. (2006). It uses a global horizontal resolution of $4^\circ \times 5^\circ$, with a $1^\circ \times 1^\circ$ nested domain for North America and the adjacent oceans (10°N–60°N, 40°W–140°W). The nested domain encompasses essentially all emissions in the US, Canada, and Mexico. The one-way nesting capability in GEOS-Chem is described by Wang et al. (2004) and application of the nested model to North American ozone simulations has been presented previously by Fiore et al. (2005) and Li et al. (2005).

Emissions used for the 2001 simulations are as described by Park et al. (2006) unless stated otherwise. Global anthropogenic emissions are from the Global Emission Inventory Activity (GEIA), scaled to 1995 as described by Bey et al. (2001). This is overridden in the US with the EPA National Emission Inventory (NEI) for 1999, in northern Mexico with the Big Bend Regional Aerosol and Visibility Observational (BRAVO) inventory for 1999 (Kuhns et al., 2005) and in Canada with the Environment Canada Criteria Air Contaminants (CAC) inventory for 2002 (http://www.ec.gc.ca/pdb/cac/cac_home_e.cfm). Emissions in US–Canada and US–Mexico border grid squares are the sums of the two corresponding national inventories.

The scaled GEIA emissions for East Asia are consistent with TRACE-P aircraft observations in Asian outflow in 2001 (Heald et al., 2003), but NO_x emissions in East Asia have grown rapidly since then (Zhang et al., 2007, Zhang et al., 2008). We also conducted a simulation using an Asian emission inventory for 2006 featuring a 50% increase in Chinese anthropogenic NO_x emissions relative to 2001

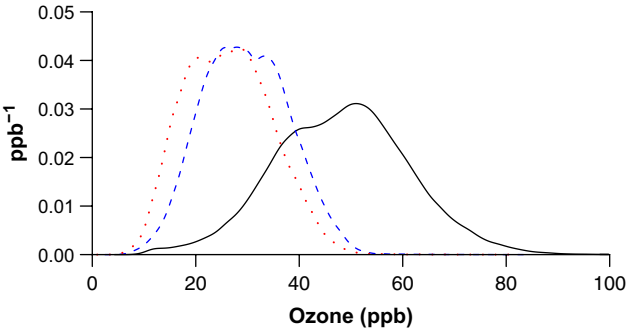

Fig. 3. Frequency distribution of the simulated daily-8 h-max surface ozone (black solid line), North American background (red dotted line) and US background (blue dashed line), for the contiguous US during Jun–Aug 2001. The frequency distributions are constructed from the $1^\circ \times 1^\circ$ daily model output.

(Zhang et al., 2007). We find that the effect of this increase on the mean North America background is less than 1 ppb anywhere in the US in summer. The effect is greater in spring, as discussed by Zhang et al. (2008).

Table 1 lists the anthropogenic NO_x emissions used in our simulations. Here and elsewhere, anthropogenic emissions include contributions from fuel combustion, industry, and fertilizer use, but not from open fires. Non-anthropogenic NO_x sources for both 2001 and 2020 include climatological (monthly) biomass burning from Duncan et al. (2003) (8.0 Tg N year^{-1} globally), soil (5.8 Tg N year^{-1} globally) and lightning (4.5 Tg N year^{-1} globally). North America (10°N–60°N, 40°W–140°W) accounts for 34% of global anthropogenic emissions in our 2001 simulation. The US accounts for 80% of North American anthropogenic emissions, Canada for 9%, and Mexico for 11%.

Our global projections of anthropogenic emissions for 2020 are based on the Current LEgislation (CLE) gridded inventory of Dentener et al. (2005). The CLE inventory includes consideration of air pollution control legislation as of 2002 and assumes full compliance. We override the CLE projections in the US with the more recent gridded projections based on the implementation of the Clean Air Interstate Rule (CAIR) (http://www.epa.gov/cair/rule.html). We also override the CLE projections for China with our own sector-specific growth factors, based on calculations by Tsinghua University for the China State Environmental Protection Administration (D.G. Streets, unpublished). The 2020 anthropogenic NO_x emissions in the US, Canada and Mexico decrease respectively by 51%, 49% and 34% relative to our 2001 inventory, while emissions in the rest of the world increase by 54%.

Methane concentrations for 2001 are specified as a global mean of 1750 ppb with a 5% interhemispheric gradient based on observations. For 2020 we assume a global mean concentration of 1845 ppb based on the CLE inventory, consistent with the B1 scenario of Watson et al. (2001) which assumes a convergent world with rapid introduction of clean technologies.

Table 3
Background ozone statistics in the contiguous US for 2001 and 2020[a].

Year	Standard	US background	North American background	Canadian/Mexican enhancement[c]
2001	48 ± 13	30 ± 8	26 ± 8	3 ± 4
2020	45 ± 10 (43 ± 9)[b]	30 ± 9	28 ± 9	2 ± 3[d]

[a] Values are means ± standard deviations of the Jun–Aug daily-8 h-max ozone.
[b] Values for a simulation with US power plant emissions eliminated.
[c] Pollution contribution from Canada and Mexico as determined by difference between US and North American backgrounds.
[d] Lower limit estimate since Canadian/Mexican emissions in border grid squares are eliminated.

We conducted a number of model simulations to quantify North American and US background ozone concentrations for present-day and 2020 conditions (Table 2). Anthropogenic emissions of NO_x, non-methane VOCs, and CO were shut off for either North America (140°W–40°W, 10°N–60°N) or the contiguous US (including shipping emissions within 400 km of the US coastline) in order to obtain the corresponding backgrounds. In the 2001 US background simulation, the border grid squares contain Canadian/Mexican emissions, but no US emissions. In the 2020 US background simulation, the border grid squares contain zero emissions due to lack of information to separate contributions from different countries. The simulation without North American anthropogenic emissions defines the North American background (i.e., the PRB as used by EPA). The simulation without US anthropogenic emissions defines the US background. The difference between the US and North American backgrounds defines the ozone enhancement from Canadian and Mexican anthropogenic emissions. Fig. 1 shows emissions near the US borders in 2001. We also conducted an additional simulation for 2020, eliminating US NO_x emissions from power plants only, as reductions in this sector are a particular target of CAIR. Power plants account for 24% of anthropogenic US NO_x emissions in the 2001 simulation (NEI 99 inventory) and 18% in 2020 (CAIR inventory).

All simulations were conducted for Apr–Sep, preceded by a 6-month spin-up with the $4^\circ \times 5^\circ$ global simulation and a 1-month spin-up with the $1^\circ \times 1^\circ$ simulation to remove the effect of initial conditions. Hourly surface ozone concentrations were archived for all $1^\circ \times 1^\circ$ grid squares in the North America domain. All ozone concentrations presented in this paper are daily 8-h average maxima (daily-8 h-max).

3. Mean background concentrations

Fig. 2 shows the Jun–Aug mean North American background (PRB) and US background ozone concentrations for the 2001 conditions. Also shown is the difference between the two, representing the Canadian/Mexican pollution enhancement. Jun–Aug is the peak of the ozone pollution season in the US and is also found in the model to be the peak of Canadian/Mexican pollution influence (by contrast, Asian pollution influence peaks in Apr–May; Jacob et al. (1999)). The North American ozone background in Fig. 2 is 13–38 ppb, consistent with the results previously reported by Fiore et al. (2003). It is higher in the west than in the east mostly because of a longer ozone lifetime (Fiore et al., 2002a). The US background is 1–3 ppb higher than the North American background over most of the US as a result of Canadian/Mexican pollution. Larger Canadian/Mexican pollution enhancements are found in the northeast (up to 15 ppb on a summer mean basis) and southwest (up to 13 ppb).

Fig. 3 shows the model frequency distributions for Jun–Aug 2001 of surface ozone concentrations, North American background, and US background, sampled daily for all grid squares in the contiguous US. The distributions are near-normal. Means and standard deviations are 48.2 ± 12.9 ppb for surface ozone, 26.3 ± 8.3 ppb for the North American background, and 29.6 ± 8.3 ppb for the US background (Table 3). The North American background values are consistent with the mean value of 26 ± 7 ppb previously reported by Fiore et al. (2003) for a slightly different metric (mean afternoon concentrations in Mar–Oct). Canadian and Mexican enhancements in the contiguous US are 3.4 ± 3.6 ppb.

4. Evaluation with observations at US sites downwind of Canada and Mexico

Previous evaluations of GEOS-Chem with US ozone air quality data have been presented by Fiore et al. (2002a, 2003), Goldstein

Table 4
Monitoring sites used for evaluation of Canadian and Mexican influence.

Site	Daily-8 h-max ozone, ppb[a]				
	r[b]	Observed	Model	US background[c]	Canadian/Mexican enhancement[d]
US sites under Canadian influence					
Whiteface Base, New York (44.4N, 73.8W)	0.77	49.7	46.6	28.6	10.8
Lake Frances Dam, New Hampshire (45.1N, 71.4W)	0.78	42.3	44.2	28.3	10.6
Williamson, New York (43.2N, 77.2W)	0.65	50.5	50.7	28.4	10.4
Westfield, New York (42.3N. 79.6W)	0.62	55.8	53.9	28.9	8.8
Howland, Maine (45.2N, 68.7W)	0.67	40.3	42.2	26.5	8.5
Kane Exp Forest, Pennsylvania (41.6N, 78,8W)	0.73	56.4	53.9	27.1	8.2
Ashland, Maine (46.6N, 68.4W)	0.74	37.2	36.3	25.3	7.9
Connecticut Hill, New York (42.4N, 76.7W)	0.72	54.8	51.2	25.7	7.5
Unionville, Michigan (43.6N, 83.3W)	0.84	52.8	48.3	23.1	5.8
M.K. Goddard, Pennsylvania (41.4N, 80.1W)	0.79	57.9	62.3	25.2	5.7
Lykens, Ohio (40.9N, 83.0W)	0.65	59.7	55.0	25.0	5.0
Ann Arbor, Michigan (42.4N, 83.9W)	0.71	54.7	52.1	24.6	4.6
US Sites under Mexican influence					
Alpine, California (32.8N, 116.8W)	0.67	64.3	66.1	30.6	10.3
Big Bend National Park, Texas (29.3N, 103.2W)	0.54	43.0	50.3	42.7	9.1
US–Mexico border, New Mexico (31.8N, 106.7W)	0.47	56.5	55.9	45.0	8.3
Joshua Tree, California (34.1N, 116.4W)	0.44	55.7	63.4	35.7	7.7
Hillside, Arizona (34.4N, 113.0W)	0.68	58.5	53.8	37.1	7.5
Tucson, Arizona (32.0N, 110.8W)	0.66	51.5	52.9	37.5	6.8
Searchlight, Nevada (35.5N. 114.9W)	0.53	59.4	57.7	37.2	6.6
Blue Point-Sheriff, Arizona (33.5N, 111.6W)	0.44	64.1	61.1	36.6	6.4
Jean, Nevada (35.8N, 115.4W)	0.53	63.7	59.5	37.4	5.7
Carlsbad, New Mexico (32.4N, 104.3W)	0.44	50.7	53.1	42.2	4.7
Mojave Desert, California (35.8N, 117.4W)	0.55	57.5	59.9	35.7	3.8
Canadian sites					
Cornwall, Ontario (45.0N, 74.7W)	0.73	51.4	42.0		
Dorset, Ontario (45.2N, 78.9W)	0.82	45.9	42.7		
London, Ontario (43.0N, 81.1W)	0.79	49.8	50.6		
Merlin, Ontario (42.2N, 82.2W)	0.69	53.6	58.5		
Tiverton, Ontario (44.3N, 81.6W)	0.83	51.5	50.8		
Toronto East, Ontario (43.7N, 79.3W)	0.75	47.3	47.3		

[a] Daily-8 h-max mean values for Jun–Aug 2001.
[b] Pearson's correlation coefficient between model and observations.
[c] As derived in the model from a sensitivity simulation with US anthropogenic emissions eliminated.
[d] As derived in the model by difference between the US background and North American background simulations.

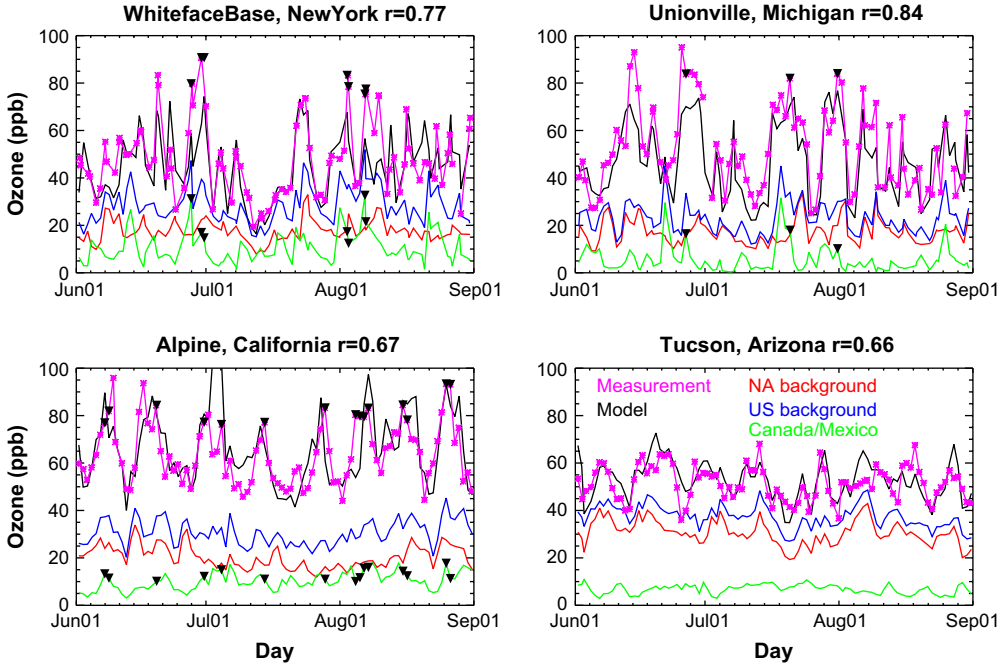

Fig. 4. Jun–Aug 2001 time series of daily-8 h-max ozone concentrations at two US sites in the northeast (top) and southwest (bottom) where Canadian and Mexican influences are particularly strong. Model results (black line) are compared to observations (magenta line with stars). Also shown are the North American background (blue line), the US background (red line), and the Canadian and Mexican pollution enhancement determined by difference of the US and North American backgrounds (green line). Black triangles highlight days when observed ozone exceeds 75 ppb and Canadian/Mexican enhancement exceeds 10 ppb. Site locations and statistics are given in Table 4. Time series plots for additional sites are included in supplementary materials.

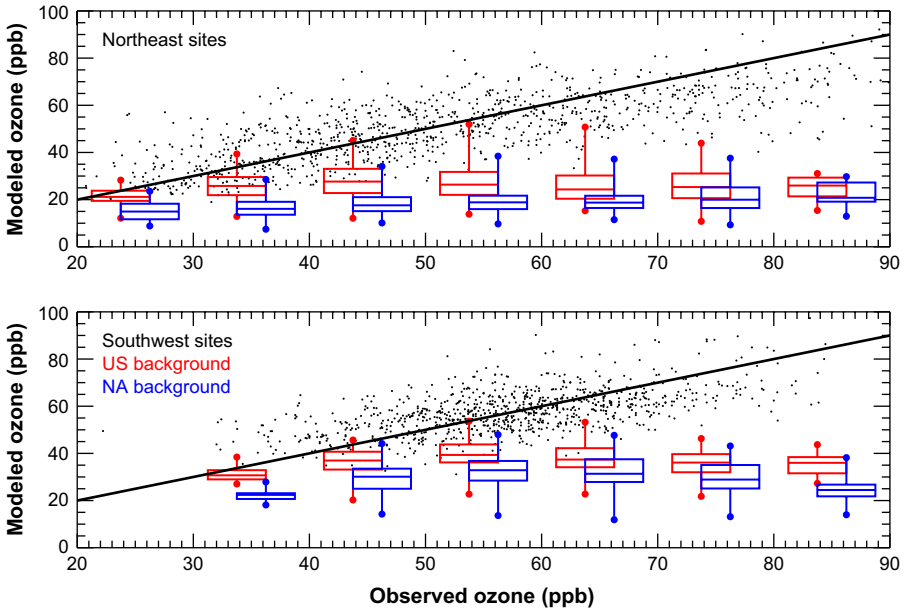

Fig. 5. North American and US background statistics as a function of observed ozone concentration for Jun–Aug 2001 at the northeast and southwest US sites of Table 4. Also shown is the scatterplot of simulated vs. observed daily-8 h-max concentrations for the ensembles of sites. The black line is the $y = x$ relationship. The box-and-whisker plots show the minimum, 25th, 50th, 75th percentile, and maximum North American background (blue) and US background (red) in 10-ppb increments of observed ozone concentrations.

et al. (2004), and Hudman et al. (2004). These have included comparisons with observations at remote sites to test the ability of the model to simulate the North American background and its variability (Li et al., 2002; Fiore et al., 2003; Goldstein et al., 2004). They found that the model had no significant bias and captures much of the variability in the observations. We focus our evaluation on summer 2001 ozone observations at northeast and southwest US sites where Canadian and Mexican influences are the highest. We also evaluate the model at a few near-border Canadian sites. For US sites, we use the ensemble from the Clean Air Status and Trends Network (CASTNet, http://www.epa.gov/castnet) and the Air Quality System (AQS, http://www.epa.gov/ttn/airs/airsaqs) where the summer mean Canadian and Mexican pollution enhancements in Fig. 2 exceed 3 ppb. We exclude urban/suburban, mountain top, and high-traffic sites (>2000 counts per year) as identified by the network database. We thus selected 12 sites in the northeast and 11 in the southwest, labeled as black/gray circles in Fig. 2c and listed in Table 4. All have near-continuous records for Jun–Aug 2001 except for Big Bend National Park which starts in late July. For Canada, we used an ensemble of non-urban sites from the Ontario Ministry of the Environment (http://www.airqualityontario.com/reports/historical_data.cfm); these are also listed in Table 4. We could not find suitable non-urban sites for model evaluation on the Mexican side of the US–Mexico border.

Correlation coefficients for the simulated vs. observed Jun–Aug time series of daily-8 h-max concentrations are shown in Table 4 (individual time series are discussed below). For US sites, correlation coefficients range from 0.62 to 0.84 in the northeast and from 0.44 to 0.67 in the southwest. The correlation coefficients for Canadian sites range from 0.73 to 0.83. The weaker correlation in the southwest likely reflects model difficulties in dealing with complex topography as well as the smaller range of variability in concentrations. At most of the sites, the modeled summer mean surface ozone is within 3 ppb of the observations (although there are a few cases with larger discrepancies). The summer mean Canadian influence is 4.6–10.8 ppb at the northeast sites and the summer mean US background at those sites is in the range of 23.1–28.9 ppb. The summer mean Mexican influence is 3.8–10.3 ppb at the southwest sites and the summer mean US background at those sites is in the range of 30.6–45.0 ppb.

Fig. 4 shows Jun–Aug time series of simulated vs. observed daily-8 h-max concentrations for two sites each in the northeast and southwest where Canadian/Mexican influences are particularly strong. Also shown in the figure are the corresponding time series of the simulated North American and US backgrounds, as well as the Canadian/Mexican pollution enhancements. Similar plots for other sites listed in Table 4 are included in the supplementary materials.

Whiteface Base (New York) has both the largest summer mean (11 ppb) and the largest episode (33 ppb) of Canadian enhancement among the selected sites in Table 4. The model accounts for most of the observed variability at the site ($r = 0.77$) with no significant bias in the mean. Unlike the North American background, the Canadian pollution enhancement shows considerable structure and manifests itself mostly in events. It ranges from 1 to 33 ppb over the course of the summer and averages 11 ± 7 ppb. The largest episode of 33 ppb occurs on a day with 75 ppb of surface ozone. There are 7 episodes in summer 2001 when Canadian pollution enhancement exceeds 10 ppb and surface ozone exceeds 75 ppb. The North American background at this site varies between 8 and 33 ppb with an average of 18 ± 5 ppb, and the US background varies between 15 and 52 ppb with an average of 29 ± 8 ppb.

Unionville (Michigan) is occasionally downwind of industrial southern Ontario. The summer mean Canadian influence at Unionville is 6 ± 6 ppb, only about half of that at Whiteface Base (New York). However, episodic Canadian enhancement at Unionville can reach 32 ppb (on a day with 75 ppb of surface ozone). The model captures the observed variability well ($r = 0.84$), including most of the high ozone episodes. The summer mean North American background and US background are 17 ± 5 ppb and 23 ± 7 ppb. There are 3 cases when Canadian enhancement exceeds 10 ppb and surface ozone exceeds 75 ppb.

Alpine (California) is the site in Table 4 most affected by Mexican pollution, both in terms of mean and variance. Although the correlation coefficient between model and observations is only moderately high ($r = 0.67$), Fig. 4 shows that the model captures most of the relevant structure in the observations. The North

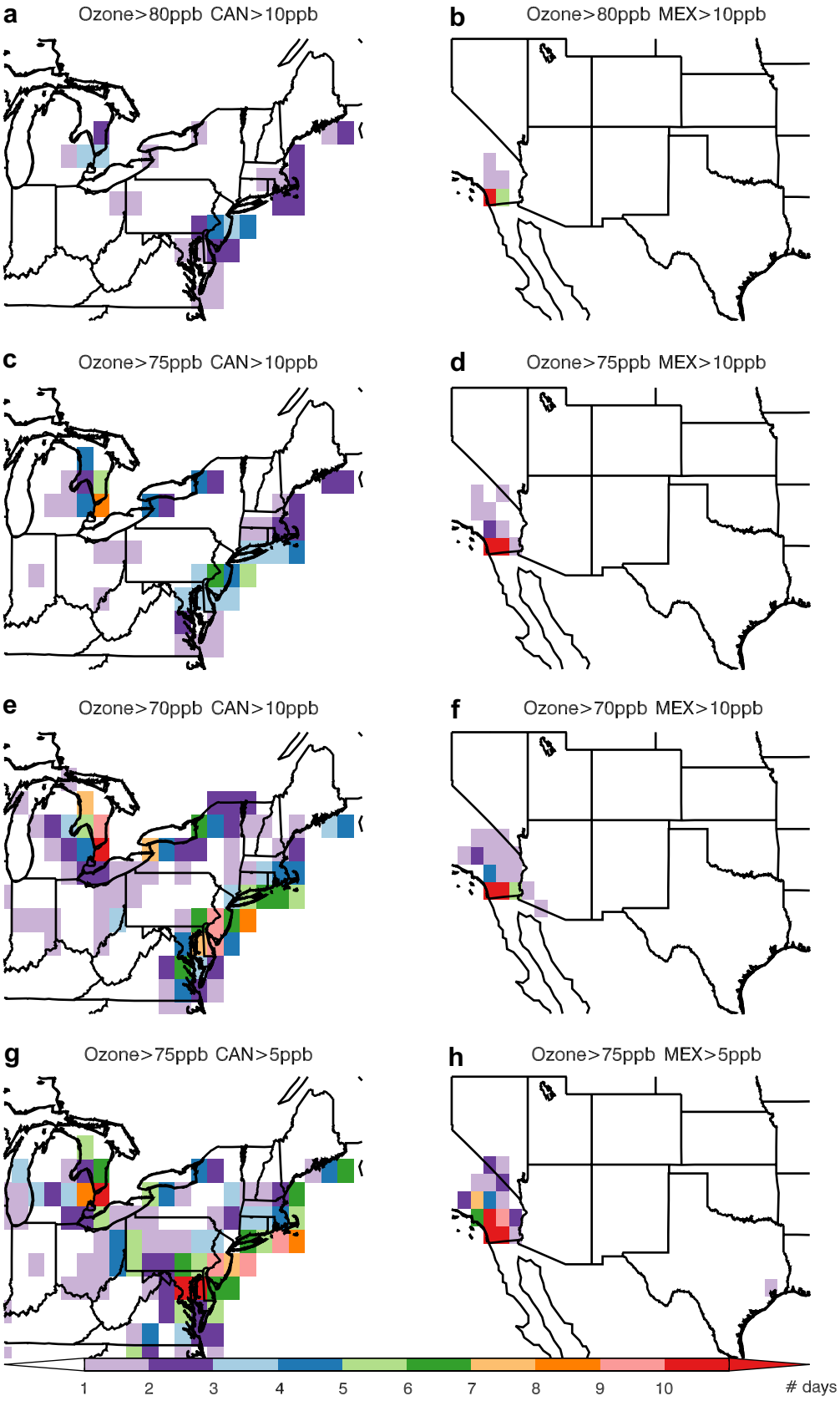

Fig. 6. Frequency of occurrence of high ozone levels in the northeast and southwest US associated with large Canadian/Mexican pollution enhancements. Shown is the number of days in Jun–Aug 2001 when the simulated ozone exceeds 80, 75, or 70 ppb and Canadian/Mexican pollution enhancements exceed 10 ppb. Also shown (bottom panels) is the number of days when the simulated ozone exceeds 75 ppb and the Canadian/Mexican pollution enhancement exceeds 5 ppb. Additional plots for other combinations of thresholds are included in the supplementary materials. Canadian/Mexican pollution enhancements are determined by the difference between model sensitivity simulations with US vs. North American anthropogenic emissions eliminated.

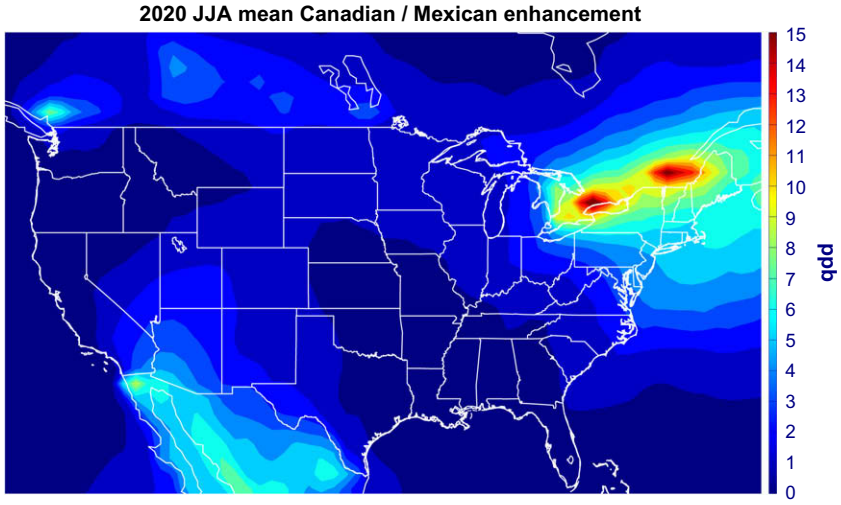

Fig. 7. Mean Canadian/Mexican pollution enhancement of the Jun–Aug daily-8 h-max ozone (ppb) in 2020. The Canadian/Mexican enhancement is determined by the difference between a simulation with 2020 US anthropogenic emissions eliminated and a simulation with 2020 North American anthropogenic emissions eliminated. Values can be compared to the present-day simulation results shown in the bottom panel of Fig. 2.

American background averages 20 ± 5 ppb with a maximum of 34 ppb, while the US background averages 31 ± 5 ppb with a maximum of 45 ppb. Mexican pollution influence averages 10 ± 4 ppb with a maximum of 18 ppb (on a day with 68 ppb of ozone); it is markedly less variable than Canadian influence in the northeast. The largest US background (45 ppb) occurs on Aug 25 when the observed ozone concentration is 93 ppb. Also shown in Fig. 4 is the time series at a rural site near Tucson (Arizona, $r = 0.66$). The North American background (31 ± 5 ppb on average) is higher than that at Alpine, but the Mexican influence is smaller and even less variable (7 ± 2 ppb on average). The largest US background occurrence at that site is 49 ppb (August 7) with 6 ppb enhancement from Mexican pollution.

The higher variability of Canadian influence in the northeast vs. Mexican influence in the southwest reflects higher wind variability in the northeast. Prevailing surface winds in the northeast in summer are from the southwest, unfavorable for Canadian pollution influence; Canadian influence manifests itself mostly through episodic frontal passages. Winds in the southwest are less variable and the mean flow can carry pollution from northern Mexico (Fiore et al., 2002a).

5. Correlation of background concentrations with pollution episodes

It is of particular interest to derive background concentration statistics for conditions when ozone concentrations approach or exceed the air quality standard, as this addresses the issue of achievability of the standard. Fig. 5 shows the North American and US background statistics as a function of the observed ozone concentration (binned in 10-ppb increments) for the ensemble of northeast and southwest US sites of Table 4. Also shown in the figure are the scatterplots of simulated vs. observed concentrations and the 1:1 lines; the high correlation and lack of bias support the approach of sorting model background concentration statistics against observed ozone concentrations. We see that ozone concentrations up to 60 ppb can be largely determined by background influence, but this influence diminishes rapidly in a relative sense when concentrations exceed 60 ppb. The maximum US background concentration in Fig. 5 is 52 ppb for the northeast sites and 54 ppb for the southwest sites. Statistics for all $1^\circ \times 1^\circ$ US grid squares in the model indicate maximum US background concentrations of 57 ppb in the northeast and 55 ppb in the southwest. In the model at least, eliminating US anthropogenic emissions would maintain ozone concentrations below 60 ppb at all times.

Fiore et al. (2002a) previously found that the North American background contribution is generally highest when ozone is in the 50–70 ppb range, and decreases when ozone is higher because of restricted import in stagnant air masses. They noted an exception in the northeast, where the background does not decrease under polluted conditions due to higher local emissions of natural ozone precursors (isoprene from vegetation, NO_x from soil) at the high temperatures associated with stagnation. Fig. 5 shows the same general patterns of correlation of background with ozone concentration as in Fiore et al. (2002a, 2003). The US background behaves similarly to the North American background.

Fig. 6 shows the spatial distribution of occurrences of peak ozone days in summer 2001 (simulated ozone >80, 75, or 70 ppb) that include a Canadian/Mexican pollution enhancement in excess of 10 ppb. We find that much of the US area around the Great Lakes bordering southern Ontario experiences such occurrences. So does the northeast corridor, where occurrences of ozone >75 ppb are frequent. From the standpoint of exceedance of the 75 ppb air quality standard, the areas most sensitive to Canadian pollution are eastern Michigan, western New York, and New Jersey.

In the southwest, we find that peak ozone days (>80, 75, or 70 ppb) associated with Mexican pollution influence in excess of 10 ppb are mostly restricted to southern California, where these peak ozone days are frequent. The most frequent occurrences are in San Diego County, where the Alpine site is located (Fig. 4). Alpine experienced a total of 21 days with more than 75 ppb of ozone in summer 2001, and the Mexican pollution enhancement exceeded 10 ppb on 15 of those days.

Also shown in Fig. 6 is the frequency of occurrence of ozone greater than 75 ppb and Canadian/Mexican influence greater than 5 ppb. Much of the northeast is affected since the summer mean Canadian influence is of similar magnitude (see Fig. 2), with the greatest impacts in eastern Michigan and in Maryland/DC area. In the southwest, most of southern California is affected. Additional plots for other combinations of thresholds are included in the supplementary materials.

6. Projection for 2020

Table 3 gives the Jun–Aug mean statistics for US surface ozone, US background, North American background, and Canadian/

Mexican influence in the contiguous US in the 2020 simulation. The mean US ozone concentration decreases by 3 ppb relative to 2001, reflecting domestic emission reductions. Despite the large increase in Asian emissions, the mean North American background in summer increases by only 2 ppb, consistent with Zhang et al. (2008). The summer mean Canadian/Mexican pollution influence is in the range of 0–11 ppb (Fig. 7). This is a lower limit estimate since (unlike for the 2001 simulation) we do not account for Canadian/Mexican emissions in border grid squares. The CLE 2020 inventory projects a future environment where Canadian pollution influence in the northeast US would emerge as a major concern. At Buffalo (New York), Canadian pollution influence in the 2020 simulation averages 10 ppb over the course of the summer, and three out of the five >70 ppb events at the site have Canadian influences exceeding 10 ppb. In comparison, the summer mean Canadian influence at Buffalo in 2001 is also 10 ppb, but none of the eight >70 ppb events have Canadian influences exceeding 10 ppb. The maximum Canadian pollution event in the 2020 simulation is in Michigan with an episode of 37 ppb.

The Canadian pollution influence in the US for the 2020 simulation can be placed in the perspective of the influence from domestic power plants, which are particularly targeted in the CAIR emission control strategy. We conducted a sensitivity simulation for 2020 with the US power plant emissions eliminated. We found a 2 ppb decrease of Jun–Aug mean surface ozone concentrations in the contiguous US relative to the standard 2020 simulation, with larger reductions of 3–7 ppb in the east. Canadian pollution influence over the US in the 2020 simulation is comparable in magnitude to that from domestic power plants in some areas of the northeast.

7. Conclusions

We used the GEOS-Chem chemical transport model with $1^\circ \times 1^\circ$ horizontal resolution to quantify the effect of Canadian and Mexican anthropogenic emissions on surface ozone in the contiguous US for both present-day (2001) and projected future (2020) conditions. The US EPA (2006) defines a policy-relevant background (PRB) in its standard-setting process as the ozone concentration that would be present in US surface air in the absence of North American anthropogenic emissions. We compared this North American background to the US background allowing for anthropogenic emissions in Canada and Mexico, in order to assess the importance of the latter for emission control strategies to meet US ozone standards. All background concentrations were expressed as maximum daily 8-h average (daily-8 h-max) concentrations, corresponding to the metric used for the air quality standard.

For 2001 conditions and the ensemble of the contiguous US, we find mean concentrations of 26 ± 8 ppb for the North American background, 30 ± 8 ppb for the US background, and 3 ± 4 ppb for the Canadian/Mexican pollution enhancement obtained by difference. Much larger Canadian and Mexican influences are found in downwind regions of the northeast and southwest US, with Jun–Aug mean values reaching 15 ppb in the northeast and 13 ppb in the southwest.

We evaluated the model by comparison to observed ozone concentrations for Jun–Aug 2001 at selected northeast US, southwest US, and Ontario sites near the US border. The model shows strong correlations with observations and no significant bias. It is able to reproduce the occurrence of peak ozone episodes exceeding 75 ppb (the new US air quality standard) with a high degree of fidelity at most sites, implying that the $1^\circ \times 1^\circ$ resolution is adequate for simulating these episodes. The North American background and the Mexican pollution influence show little temporal variability, but the Canadian pollution influence shows large temporal variability. The highest Canadian influence of 33 ppb occurs in New York State on a day with 75 ppb of surface ozone. The highest Mexican influence of 18 ppb occurs in southern California on a day with 68 ppb of surface ozone.

Statistics for the ensemble of sites show that both the North American and US backgrounds peak when ozone is in the range 50–70 ppb. The highest US background concentrations in the model simulations are 57 ppb in the northeast and 55 ppb in the southwest. We thus find that eliminating US anthropogenic emissions would maintain surface ozone concentrations in the US below 60 ppb at all times.

We examined the frequency of occurrence of Canadian/Mexican pollution influences greater than 10 ppb under model conditions when surface ozone exceeds the US air quality standard (75 ppb). We found such occurrences in large areas of the northeast US and in southern California. Most affected are eastern Michigan, western New York, New Jersey, and San Diego County. More stringent air quality standards will increase the relative importance of Canadian/Mexican pollution influence in these areas and thus the importance of coordinated transboundary emission control strategies.

Our 2020 simulation used emission projections from the Clean Air Interstate Rule (CAIR) for the US, an independent estimate for China, and the Current LEgislation (CLE) inventory for the rest of the world. The summer mean surface ozone averaged over the contiguous US decreases by 3 ppb relative to the 2001 standard simulation, the North American background increases by 2 ppb, and the US background remains the same. Our results indicate that Canadian pollution influence could emerge as an important concern for US air quality in the northeast in the future, of comparable importance to the influence from domestic power plant emissions in some areas.

Acknowledgments

This work was sponsored by US Department of Energy, Office of Fossil Energy, through a contract with Argonne National Laboratory. We thank Naresh Kumar at EPRI and Thomas Grahame at DOE for their constructive input to this work.

Appendix A. Supplemental material

Supplementary information for this manuscript can be downloaded at doi: 10.1016/j.atmosenv.2008.11.036.

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