Effects of 2000–2050 changes in climate and emissions on global tropospheric ozone and the policy-relevant background surface ozone in the United States

Shiliang Wu,[1] Loretta J. Mickley,[1] Daniel J. Jacob,[1] David Rind,[2] and David G. Streets[3]

Received 26 November 2007; revised 15 April 2008; accepted 26 June 2008; published 27 September 2008.

[1] We use a global chemical transport model (GEOS-Chem) driven by a general circulation model (NASA Goddard Institute for Space Studies GCM) to investigate the effects of 2000–2050 global change in climate and emissions (the Intergovernmental Panel on Climate Change A1B scenario) on the global tropospheric ozone budget and on the policy-relevant background (PRB) ozone in the United States. The PRB ozone, defined as the ozone that would be present in U.S. surface air in the absence of North American anthropogenic emissions, has important implications for setting national air quality standards. We examine separately and then together the effects of changes in climate and anthropogenic emissions of ozone precursors. We find that the 2000–2050 change in global anthropogenic emissions of ozone precursors increases the global tropospheric ozone burden by 17%. The 2000–2050 climate change increases the tropospheric ozone burden by 1.6%, due mostly to lightning in the upper troposphere, and also increases global tropospheric OH by 12%. In the lower troposphere, by contrast, climate change generally decreases the background ozone. The 2000–2050 increase in global anthropogenic emissions of ozone precursors increases PRB ozone by 2–6 ppb in summer; the maximum effect is found in April (3–7 ppb). The summertime PRB ozone decreases by up to 2 ppb with 2000–2050 climate change, except over the Great Plains, where it increases slightly as a result of increasing soil NO_x emission. Climate change cancels out the effect of rising global anthropogenic emissions on the summertime PRB ozone in the eastern United States, but there is still a 2–5 ppb increase in the west.

Citation: Wu, S., L. J. Mickley, D. J. Jacob, D. Rind, and D. G. Streets (2008), Effects of 2000–2050 changes in climate and emissions on global tropospheric ozone and the policy-relevant background surface ozone in the United States, *J. Geophys. Res.*, *113*, D18312, doi:10.1029/2007JD009639.

1. Introduction

[2] Surface ozone is a toxic air pollutant. As of 2003–2005, 157 million people in the United States lived in areas exceeding the ozone National Ambient Air Quality Standards (NAAQS) of 0.08 ppm (8-h average, not to be exceeded more than 3 times per year) [*U.S. Environmental Protection Agency* (*EPA*), 2007]. Ozone in the troposphere is produced by photochemical oxidation of volatile organic compounds (VOCs) and carbon monoxide (CO) in the presence of nitrogen oxides ($NO_x \equiv NO + NO_2$). The lifetime of tropospheric ozone varies from days to months, and the lifetimes of its precursors span an even wider range, making ozone air quality not only a regional problem, but also subject to global change in anthropogenic emissions [*Jacob et al.*, 1999; *Wild and Akimoto*, 2001; *Prather et al.*, 2003].

[3] The EPA defines a "policy-relevant background" (PRB) ozone for the United States as the surface ozone concentration that would be present in the absence of anthropogenic emissions from North America (United States, Canada, and Mexico). The PRB is an important factor in the NAAQS-setting process, as it represents a baseline of risk not amenable to domestic regulation [*U.S. EPA*, 2006]. Model studies indicate that the PRB background has a large intercontinental anthropogenic component [*Fiore et al.*, 2002a, 2003a] and observations show that it has been rising over the past three decades [*Lin et al.*, 2000; *Jaffe et al.*, 2003].

[4] Future global socioeconomic scenarios (A1B, A1T, A1FI, A2, B1, B2) by the Intergovernmental Panel on Climate Change (IPCC) project major changes in anthropogenic emissions of greenhouse gases and ozone precursors over the 21st century [*Intergovernmental Panel on Climate Change* (*IPCC*), 2000]. All scenarios project a global increase of anthropogenic emissions of ozone precursors for 2000–2050, largely driven by economic growth in developing countries, but three of them (A1B, A1T, B1)

[1]School of Engineering and Applied Sciences and Department of Earth and Planetary Sciences, Harvard University, Cambridge, Massachusetts, USA.
[2]NASA Goddard Institute for Space Studies, New York, New York, USA.
[3]Argonne National Laboratory, Argonne, Illinois, USA.

Copyright 2008 by the American Geophysical Union.
0148-0227/08/2007JD009639

project decreasing emissions in North America and Europe. Such a geographical shift in emissions could significantly increase the role of intercontinental transport of pollution as a factor affecting U.S. air quality. *Prather et al.* [2003] found that all IPCC scenarios for anthropogenic emissions of ozone precursors imply a 2000–2030 increase in surface ozone by 2–7 ppb over much of the Northern Hemisphere. *Unger et al.* [2006] calculated an increase of annual mean surface ozone by 2–5 ppb over the United States by 2030 with the A1B scenario. *Fiore et al.* [2002a] projected a longer and more intense U.S. ozone pollution season under the A1B emission scenario for 2030 because of a higher background and despite domestic emission reductions.

[5] Another factor affecting background ozone could be the changing climate. A number of studies (summarized in Table 1) have used general circulation models (GCMs) to examine the effect of 21st -century climate change on the global tropospheric ozone budget, independently from change in anthropogenic emissions of ozone precursors. Most studies project global ozone decreases due to increasing water vapor shortening the ozone chemical lifetime [*Johnson et al.*, 1999], but *Zeng and Pyle* [2003] and *Hauglustaine et al.* [2005] project a 3% increase by 2100 due to increased stratosphere-troposphere exchange (STE) and natural emissions of ozone precursors.

[6] Fewer studies have examined the effect of climate change on the surface ozone background. From a multi-model ensemble simulation, *Dentener et al.* [2006] projected that 2000–2030 climate change will reduce annual mean surface ozone by 0.5–1 ppb over most continental areas including the United States. *Murazaki and Hess* [2006] calculated a decrease of background ozone by 0–2 ppb in the United States in summer due to 1990–2090 climate change (A1B scenario). With the 1996–2095 climate change, *Lin et al.* [2008] found increases in summer surface ozone over the United States by 1–15 and 0–7 ppb under the A1FI and B1 scenarios respectively, largely driven by changes in biogenic emissions.

[7] In previous work we used a global chemical transport model (GEOS-Chem CTM) driven by meteorological fields from a general circulation model (NASA/GISS GCM 3) to investigate the effects of 2000–2050 global changes in climate and anthropogenic ozone precursor emissions (A1B scenario) on ozone air quality in the United States [*Wu et al.*, 2008]. We apply this model here to study the effects on global tropospheric ozone, natural background ozone, and intercontinental transport of ozone pollution to the United States. The A1B scenario is of particular interest as it represents a world where the United States is aggressively reducing its domestic ozone precursor emissions to meet air quality standards, but where the results of these efforts could be compromised by increases in emissions abroad [e.g., *Lelieveld and Dentener*, 2000; *Fiore et al.*, 2002a]. As we will see, climate change further complicates the picture.

2. Model Description

[8] The model and emission scenario used in this work are the same as that used by *Wu et al.* [2008]. We apply the NASA/GISS GCM 3 [*Rind et al.*, 2007] to simulate 2000–2050 climate change following the IPCC A1B scenario [*IPCC*, 2001]. The GCM output is used to drive the GEOS-Chem chemical transport model (CTM) for global simulation of ozone-NO_x-VOC-aerosol chemistry. The interface between the GEOS-Chem CTM and the GISS GCM is described by *Wu et al.* [2007].

[9] We use the "qflux" version of the GISS GCM 3, with a horizontal resolution of 4° latitude by 5° longitude and 23 vertical layers in a sigma coordinate system extending from the surface to 0.002 hPa [*Rind et al.*, 2007]. Comparison with higher-resolution models and observations shows that although the coarse 4° × 5° resolution compromises the ability to capture local ozone maxima, it does not induce significant mean bias and can still capture the major factor of ozone variability on synoptic scales [*Fiore et al.*, 2003b; *Wu et al.*, 2008].

[10] We perform a 1950–2000 spin-up starting from climate equilibrium to adequately initialize the ocean, using observed trends of the well-mixed greenhouse gases (including CO_2, CH_4, N_2O, and halocarbons), ozone, and aerosols [*Hansen et al.*, 2002]. For 2001 to 2050 we follow the IPCC A1B scenario for the well-mixed greenhouse gases, with CO_2 as calculated in the Bern-CC model [*IPCC*, 2001]. CO_2 levels reach 522 ppm by 2050. The GISS GCM yields a global mean surface temperature increase of 1.6 K for 2000–2050. Summertime temperatures increase by as much as 2–3 K over the United States. The equilibrium climate sensitivity (global mean surface temperature increase following a doubling of atmospheric CO_2 concentration) of the GISS GCM is 2.8 K, in the range of 2.1–4.4 K for the current generation of GCMs [*Randall et al.*, 2007].

[11] The 2000–2050 trends in anthropogenic emissions of ozone precursors are based on the IPCC A1B scenario. Fossil fuel NO_x emissions decrease by 40% in the United States but rise by 90% globally (Table 2a). Following *Fiore et al.* [2002a], we first derived 2000–2050 growth factors for different categories of anthropogenic emissions (fossil fuel, biofuel, and biomass burning emissions) and for different countries, on the basis of data from the IMAGE socioeconomic model [*IMAGE Team*, 2001]. These national growth factors were mapped onto the GEOS-Chem model grid and applied to the present-day GEOS-Chem emission inventories to generate the emission inventories for 2050.

[12] For simulations with present-day anthropogenic emissions, the methane mixing ratios are specified with a global mean of 1750 ppb and a 5% interhemispheric gradient based on observations. Methane concentrations have leveled off over the past decade [*Forster et al.*, 2007], which could reflect stabilization of emissions [*Dlugokencky et al.*, 2003], but could also be only a temporary reprieve [*Wuebbles and Hayhoe*, 2002]. *IPCC* [2001] projects methane mixing ratio to rise to 2400 ppb by 2050 in the A1B scenario, on the basis of increasing emissions and a methane-OH positive feedback factor [*Prather*, 1996; *Prather et al.*, 2001], but not including the effect of climate change on OH concentrations. 2000–2050 climate change increases OH because of increasing water vapor (Table 1), thus shortening the methane lifetime.

[13] In our simulations with 2050 climate, we adjust the methane concentrations to ensure consistency in the implied methane emissions with the corresponding simulations with 2000 climate (Table 2a). For example, in the sensitivity simulations with 2050 climate but present-day anthropogenic emissions, we first set the methane concentrations to

Table 1. Projected Perturbations to Global Tropospheric Ozone and OH From 21st-Century Climate Change[a]

Reference	Model	Scenario	Time Horizon	STE and Natural Emissions[b]	Ozone Burden	$P(O_x)$[c]	$L(O_x)$[c]	Ozone Lifetime[d]	OH
Johnson et al. [1999]	STOCHEM	IS92a[e]	1990–2075		−10%	+4%	+9%	−18%	+13%[f]
Stevenson et al. [2000][g]	STOCHEM	IS92a	1990–2030	STE +6%	−3%	+1%	+2%	−5%	
Stevenson et al. [2000][g]	STOCHEM	IS92a	1990–2060	STE +15%	−4%	+2%	+3%	−7%	+7%[h]
Stevenson et al. [2000][g]	STOCHEM	IS92a	1990–2100	STE +15%	−10%	+3%	+6%	−14%	
Zeng and Pyle [2003][g]	UM_CAM	A2[i]	2000–2100	STE +80%	+3%	+11%	+21%	−10%	
Hauglustaine et al. [2005]	LMDz-INCA	A2	2000–2100	lightning +50%, soil +22% isoprene +59% STE +35%	+3%	+14%	+19%	−14%	+9%
Stevenson et al. [2005]	STOCHEM/HadAM3	IS92a	1990–2020	isoprene +9%	−4%	+2%	+2%	−4%	
Brasseur et al. [2006]	MOZART-2/MPI-OM-1	$2 \times CO_2$	2000–2100	lightning +20%	+5%				+12%[h]
Liao et al. [2006]	CACTUS	A2	2000–2100	lightning +21% soil +24% isoprene +55%	−12%	+9%	+12%	−18%	
Racherla and Adams [2006]	CACTUS	A2	1990–2050		−6%	+2%	+7%	−9%	
Stevenson et al. [2006][j]	10 models	Various	2000–2030	various changes	−0.4% ± 1.2%	+0.6% ± 1.2%	+1.7% ±1.4%	−1.5% ±1.7%	+4%[h]
Unger et al. [2006]	GISS 3	B1[k]	1990–2030	lightning +5%	−2%	[l]		+4%[h]	
This study	GEOS-Chem/GISS	A1B	2000–2050	lightning +18% soil +8% isoprene +25%	+2%	+8%	+9%	−5%	+12%

[a]From global modeling studies using constant anthropogenic emissions to isolate the effect of climate change. The studies differ in their time horizons, climate change scenarios, and treatments of changes in natural emissions and stratosphere-troposphere exchange (STE), as described in the table. Unless noted otherwise, percentage perturbations to the tropospheric ozone and OH budgets are mass-weighted annual means for the global atmospheric domain extending from the surface to tropopause.

[b]No changes in STE and natural emissions of ozone precursors (lightning NO_x, soil NO_x, and vegetative isoprene) are reported in the reference unless noted otherwise in this column.

[c]Chemical production rate ($P(O_x)$) and loss rate ($L(O_x)$) of tropospheric ozone, expressed in terms of the odd oxygen family (O_x) to account for rapid cycling between ozone and other O_x components. In our work, the odd oxygen family is defined as $O_x = O_3 + O + NO_2 + 2 \times NO_3 + 3 \times N_2O_5 + PANs + HNO_3 + HNO_4$; other studies might use slightly different definitions, but the effect is minor [*Wu et al.*, 2007].

[d]Calculated as the tropospheric O_x burden divided by the O_x chemical and deposition loss rates.

[e]Older scenario described by *IPCC* [1995].

[f]Average of effects in January (11%) and July (14%).

[g]Derived by difference of simulations including changes of anthropogenic emissions with versus without climate change.

[h]Derived from the change in methane lifetime.

[i]One of the most pessimistic scenarios projected by *IPCC* [2001].

[j]Intercomparison of 10 models using different climate change scenarios and treatments of impacts on STE and natural emissions. Results are given as the means and standard deviations for all models.

[k]With CO_2 abundance change from 360 ppm in 1995 to 429 ppm in 2030 and hence close to the B1 scenario.

[l]Only net ozone production ($P(O_x) - L(O_x)$) is reported (−4%).

Table 2a. The 2000–2050 Trends in Anthropogenic Emissions of Ozone Precursors[a]

Species	Global			North America[b]			Contiguous United States		
	2000[c]	2050	Change (%)	2000[c]	2050	Change (%)	2000[c]	2050	Change (%)
NO_x (Tg N a^{-1})									
Fossil fuel	24.6	47.3	+90%	6.7	4.8	−28%	5.9	3.6	−40%
Biomass burning	6.5	8.1	+25%	0.17	0.19	+12%	0.03	0.06	+100%
Biofuel	2.2	2.1	−5%	0.05	0.06	+11%	0.01	0.01	0%
Fertilizer	0.5	0.9	+80%	0.07	0.08	+22%	0.05	0.06	+8%
CO (Tg CO a^{-1})									
Fossil fuel	381	363	−5%	94	45	−50%	81	35	−57%
Biomass burning	459	750	+63%	19.2	27.0	+40%	3.4	8.4	+150%
Biofuel	176	169	−4%	6.6	7.0	+5%	3	2.6	−13%
NMVOCs[d] (Tg C a^{-1})									
Anthropogenic	43	98	+130%	10.6	5.5	−48%	9.3	4.4	−52%
Biomass burning	10	17	+66%	0.47	0.63	34%	0.09	0.21	+133%
Methane (ppb)	1750[e]	2400[e]	+37%						

[a]From the *IPCC* [2001] A1B scenario.
[b]Including the United States, Canada, and Mexico.
[c]The base year used for present-day anthropogenic emission inventories is 1999 for the United States and 1995 elsewhere.
[d]Nonmethane volatile organic compounds including alkanes, alkenes, and acetone.
[e]For simulations with 2000 climate. When 2000–2050 climate change is taken into account, the methane concentration is reduced to 1520 ppb (for 2000 anthropogenic emissions) and 2090 ppb (for 2050 anthropogenic emissions) because of OH increase, as discussed in the text.

be the same as present-day levels, and find that increasing OH increases methane oxidation by 9%. We then apply the methane-OH feedback factor of 1.4 [*Prather et al.*, 2001] and repeat the simulation with a 13% decrease of methane concentration. We verified in all cases that the resulting methane oxidation rate matches that in the corresponding simulation with present-day climate, so that the implied methane emission is indeed the same in both simulations.

[14] Natural emissions of ozone precursors (Table 2b), including nonmethane VOCs (NMVOCs) from vegetation, and NO_x from lightning and soil, are computed locally within the model on the basis of meteorological variables and hence allowed to change in response to climate change. Biogenic emissions of NMVOCs are influenced solely by temperature and solar radiation [*Guenther et al.*, 1995; *Wang et al.*, 1998]; we do not account for the effects of changes in atmospheric CO_2 concentrations [*Constable et al.*, 1999; *Rosenstiel et al.*, 2003; *Centritto et al.*, 2004] or land cover [*Sanderson et al.*, 2003]. The global total isoprene emission rate calculated in our model for the 2000 climate is 430 Tg C a^{-1} (Table 2b), which is 14% less than the global emission rate reported by *Guenther et al.* [1995], reflecting difference in the driving meteorology. Lightning NO_x emissions are parameterized as a function of deep convective cloud top [*Price and Rind*, 1992; *Wang et al.*, 1998] and are distributed vertically following *Pickering et al.* [1998]. Soil NO_x emissions are computed as a function of vegetation type, temperature, precipitation, fertilizer usage, and leaf area index [*Yienger and Levy*, 1995; *Wang et al.*, 1998]. Further details on the emission inventories are given by *Wang et al.* [1998] and *Bey et al.* [2001].

[15] We do not account for possible 2000–2050 changes in stratosphere-troposphere exchange (STE) of ozone. In all simulations, STE for ozone is represented by the Synoz flux boundary condition [*McLinden et al.*, 2000] with an imposed global annual mean STE flux of 470–530 Tg O_3 a^{-1} depending on the meteorological year.

[16] Following *Fiore et al.* [2003a], we quantify the ozone background over the United States with three groups of simulations: (1) a standard simulation as described above, (2) a PRB ozone simulation excluding anthropogenic emissions of NO_x, CO, and nonmethane VOCs (NMVOCs) from North America, and (3) a natural ozone simulation excluding global anthropogenic emissions of NO_x, CO, and NMVOCs, and setting the methane mixing ratio to its observed preindustrial value of 700 ppb. We define as anthropogenic the emissions of NO_x, CO, and NMVOCs from fuel use, industry, and fertilizer application but not those from open fires (i.e., wildfires, prescribed fires, and agricultural waste burning). Although humans are responsible for most of fire ignition, they are also responsible for most of fire suppression and so the net effect of human activity on fire emissions is highly uncertain.

[17] The standard and PRB ozone simulations were done for four cases to separate the effects of change in climate and change in anthropogenic emissions of ozone precursors: (1) present-day climate and emissions, (2) 2050 climate and

Table 2b. The 2000–2050 Trends in Natural Emissions of Ozone Precursors[a]

Species	Global			North America			Contiguous United States		
	2000	2050	Change (%)	2000	2050	Change (%)	2000	2050	Change (%)
NO_x (Tg N a^{-1})									
Lightning	4.9	5.8	+18%	0.22	0.26	+24%	0.14	0.17	+21%
Soil	6.1	6.6	+8%	0.58	0.64	+10%	0.35	0.39	+11%
Isoprene (Tg C a^{-1})	430	540	+25%	37	45	+23%	28	35	+25%
Other biogenic NMVOCs[b] (Tg C a^{-1})	180	210	+20%	19	22	+18%	12	14	+20%

[a]Results are based on 3-year general circulation model averages (1999–2001 and 2049–2051).
[b]Nonmethane volatile organic compounds, including alkenes, monoterpenes, and acetone but not methanol.

Table 3. The 2000–2050 Changes in Global Tropospheric Ozone and OH[a]

Ozone	2000 Conditions[b]	2050 Climate[c]	2050 Emissions[d]	2050 Conditions[e]
$P(O_x)$ (Tg a^{-1})	4530	4880 (+8%)	5900 (+30%)	6290 (+39%)
$L(O_x)$ (Tg a^{-1})	4090	4460 (+9%)	5200 (+27%)	5620 (+38%)
Ozone deposition (Tg a^{-1})	960	950 (−1.5%)	1230 (+28%)	1210 (+26%)
Ozone burden (Tg)	313	318 (+1.6%)	367 (+17%)	370 (+18%)
Ozone lifetime[f] (days)	22.6	21.5 (−5%)	20.8 (−8%)	19.8 (−13%)
OH[g] (10^6 molecule cm^{-3})	1.10	1.23 (+12%)	1.10 (−0.1%)	1.23 (+12%)

[a]Global annual mean budgets. Numbers in parentheses show the percentage changes relative to the 2000 conditions. The tropospheric domain is defined using 2000 tropopause heights in all cases. Transport from the stratosphere provides an ozone source to the troposphere of about 500 Tg a^{-1} in all simulations (with slight interannual variation of up to 30 Tg a^{-1} reflecting the year-to-year differences in the model circulation). The ozone budget is defined as that of odd oxygen (O_x); see footnote a in Table 1.

[b]The 2000 anthropogenic emissions of ozone precursors and 2000 climate.

[c]The 2000 anthropogenic emissions of ozone precursors and 2050 climate.

[d]The 2050 anthropogenic emissions of ozone precursors and 2000 climate.

[e]The 2050 anthropogenic emissions of ozone precursors and 2050 climate.

[f]Lifetime against chemical loss and deposition.

[g]Mean tropospheric concentration weighted by air mass. The changes take into account the methane-OH positive feedback factor, including the effect of changing climate (see section 2).

present-day anthropogenic emissions of ozone precursors, (3) present-day climate and 2050 anthropogenic emissions of ozone precursors, and (4) 2050 climate and emissions. The natural ozone simulations were done for two cases, 2000 and 2050 climate. To account for interannual variability, each simulation was conducted for three GCM years (1999–2001 or 2049–2051) following 1 year of model spin-up [*Wu et al.*, 2008]. Unless noted otherwise, all results in this paper are averages over those 3 years. Interannual variability of ozone in the model is small and examination of the 2000–2050 time series of GISS meteorological data fields indicates that 3-year samples are sufficient to characterize changes in the 2050 versus 2000 climate [*Wu et al.*, 2007, 2008].

3. Global Tropospheric Ozone

[18] Table 3 gives our global budget of tropospheric ozone for the present-day atmosphere and for the different 2000–2050 perturbations. 2000–2050 global change in anthropogenic emissions of ozone precursors (not including climate change) increases the global chemical production rate of tropospheric ozone by 30% and the tropospheric ozone burden by 17%. This is mainly driven by the projected increases of anthropogenic NO_x and methane emissions (Table 2a), which are the principal limiting precursors for ozone formation on a global scale [*Wang and Jacob*, 1998]. The ozone lifetime decreases by 8% because much of the added production is in the continental boundary layer where ozone has a short lifetime [*Fiore et al.*, 2002a]. There is no significant change in mean tropospheric OH due to compensating effects from increasing ozone and NO_x versus increases in CO and VOCs; regionally, OH generally decreases over remote areas but increases over regions with increasing NO_x emissions.

[19] When only climate change is considered we find that the lifetime of tropospheric ozone decreases by 5% (Table 3), consistent with previous studies (Table 1) and due to higher water vapor driving ozone chemical loss [*Johnson et al.*, 1999]. However, we also find an 8% increase in global ozone production, so that the tropospheric ozone burden increases by 1.6%. These changes are for the troposphere as defined by tropopause heights for the 2000 climate; tropopause heights generally increase by 0.2–0.6 km in our 2050 climate, which would further increase tropospheric ozone production and burden.

[20] Most previous studies except *Zeng and Pyle* [2003] and *Hauglustaine et al.* [2005] found by contrast that the tropospheric ozone burden decreases in the future climate. As shown in Table 1, this appears to reflect differences in the accounting of the effect of climate change on natural ozone sources. When increases in lightning, soil NO_x, biogenic VOCs, and STE are all taken into account, tropospheric ozone is more likely to show an increase in response to 21st -century climate change. We find in our simulation that the principal factor increasing the tropospheric ozone burden in the future climate is the 18% increase of lightning NO_x (Table 2b) due to more frequent deep convection [*Del Genio et al.*, 2007], which enhances ozone production in the upper troposphere where its lifetime is long [*Martin et al.*, 2002; *Labrador et al.*, 2004, 2005; *Sauvage et al.*, 2007; *Wu et al.*, 2007]. Previous studies compiled in Table 1 similarly find 5–50% increases in the lightning NO_x source from the 21st -century climate change.

[21] Our results show a 12% increase in tropospheric OH from 2000 to 2050 climate change. This is on the high end of the range from previous studies (Table 1), particularly considering that these tended to cover a longer time horizon or stronger climate change. Our result could reflect our correct treatment of the methane-OH feedback factor, which to our knowledge was not considered in previous studies. Tropospheric OH in our simulation would only increase by 8% if we ignored this feedback factor, as discussed in section 2.

[22] We find that the effects of changes in climate and anthropogenic emissions on tropospheric ozone and OH are nearly additive (Table 3). The combined effect is to increase the chemical production rate of tropospheric ozone by 39% and the ozone burden by 18%; these increases are dominated by emission changes. On the other hand, the combined effect on tropospheric OH (increase by 12%) is dominated by climate change.

[23] Figure 1a shows our simulated 2000–2050 changes in the global distribution of tropospheric ozone. The increase due to changes in anthropogenic emissions of

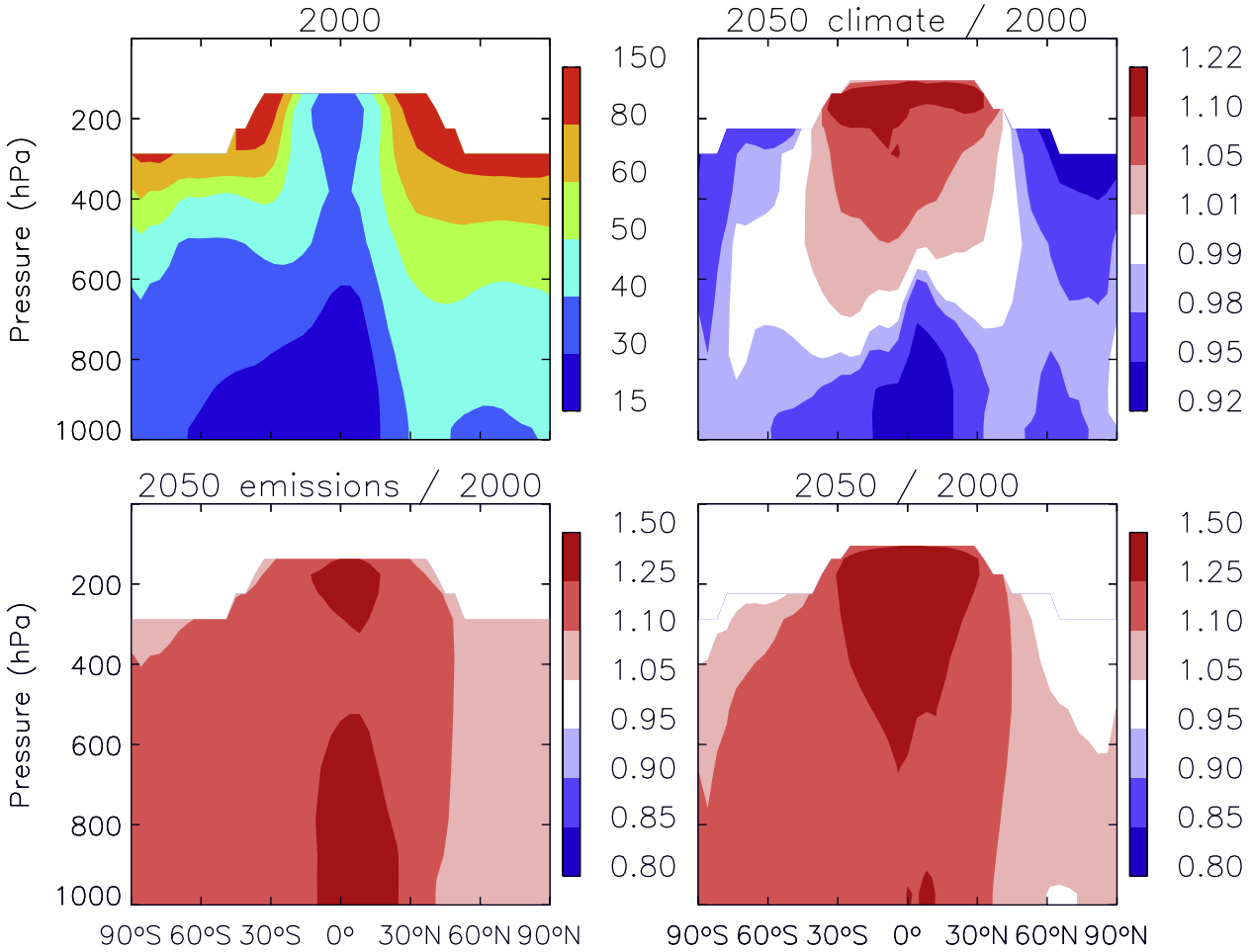

Figure 1a. (top left) Simulated present-day (2000 climate) annual zonal mean tropospheric ozone concentrations (ppb). Perturbations shown as ratios from (top right) changes in climate (2050 climate–2000), (bottom left) changes in anthropogenic emissions (2050 emissions–2000), and (bottom right) changes in both (2050–2000). Note differences in scales.

ozone precursors extends throughout the troposphere, with the largest effect in the tropics. The largest increase in ozone due to climate change is in the tropical upper troposphere, reflecting the increase in lightning. In contrast, we find that climate change decreases mean ozone levels in the lower troposphere (below 800 hPa) by up to 8%, especially in the tropics and Northern Hemisphere. This decrease reflects the shorter lifetimes of ozone and of peroxyacetylnitrate (PAN), a low-temperature NO_x reservoir produced from NO_x-NMVOC chemistry that provides a major source of NO_x to the remote atmosphere [*Wang et al.*, 1998]. The higher temperature in the future climate reduces the potential for long-range transport of PAN and hence the corresponding source of ozone in the remote troposphere. This effect is most important in the Northern Hemisphere where the continental NO_x and NMVOC sources are concentrated and thus where most of the PAN formation takes place. Conversely, less efficient PAN formation increases NO_x and hence ozone in polluted source regions [*Jacob et al.*, 1993; *Sillman and Samson*, 1995]. The large decrease of ozone due to climate change in the upper troposphere at high latitudes also reflects the increase of tropopause heights.

[24] Figure 1b examines more specifically the change in the global distribution of surface ozone (annual mean afternoon concentrations). Changes in anthropogenic emissions increase surface ozone in most of the world, except in the eastern United States where there is a decrease of up to 7 ppb reflecting emission reductions (Table 2a). Other regions with similar reductions in emissions in the A1B scenario (western United States, western Europe) do not show such decreases because of the compensating effect of the rising background [*Lelieveld and Dentener*, 2000; *Fiore et al.*, 2002a].

[25] Changes in climate affect surface ozone in a complicated way, with increases over most polluted continental regions (including the United States) and decreases over the oceans by as much as 5 ppb in the Northern Hemisphere. The continental increases reflect a combination of meteorological and natural emission factors including higher temperature, reduced convective ventilation, decrease in midlatitude cyclone frequency, as well as enhanced biogenic emissions for NMVOCs and NO_x, as discussed in detail for the United States by *Wu et al.* [2008]. The decreases over the oceans reflect the strong influence of increasing chemical loss driven by water vapor [e.g., *Johnson et al.*, 1999],

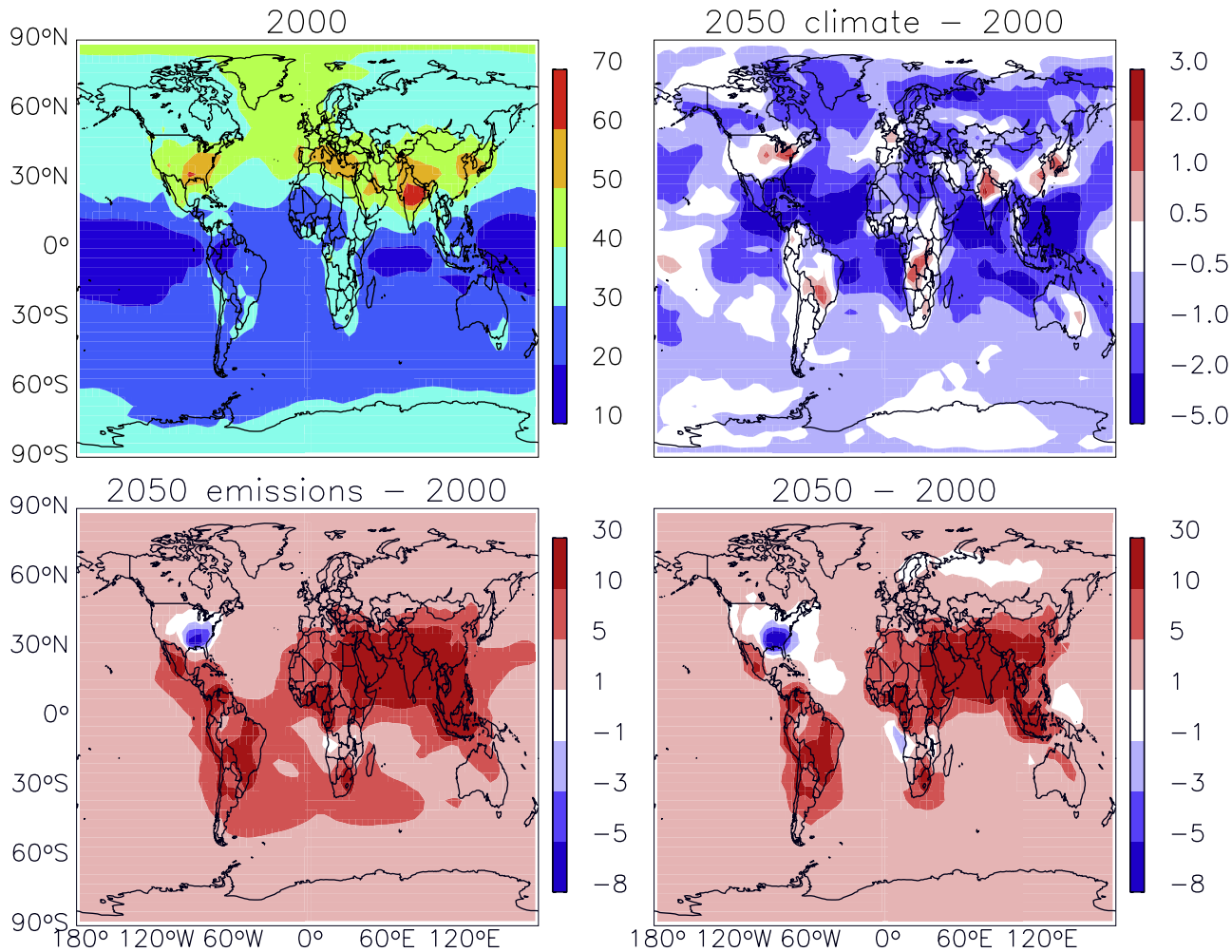

Figure 1b. Same as Figure 1a, but for annual mean afternoon (1300–1700 local time (LT)) surface ozone in ppb and with perturbations shown as absolute differences. Note differences in scales.

and also (and most strongly in the Northern Hemisphere) the reduced long-range transport of PAN to serve as a NO_x reservoir [*Wu et al.*, 2008].

4. Background Surface Ozone in the United States

[26] Figure 2 shows the natural summer afternoon ozone levels in surface air for the 2000 climate (anthropogenic emissions shut off worldwide). Values range from 10 to 15 ppb in the eastern United States to 15–25 ppb in the west. The higher values in the west reflect the higher elevation as well as the deeper boundary layer mixing and longer O_3 lifetimes in the arid climate [*Fiore et al.*, 2002b, 2003a]. Our natural ozone values are consistent with previous preindustrial ozone simulations [*Wang and Jacob*, 1998; *Mickley et al.*, 1999; *Hauglustaine and Brasseur*, 2001; *Fiore et al.*, 2003a]. *Lelieveld and Dentener* [2000] calculated slightly higher preindustrial ozone mixing ratios of 15–25 ppb over the United States, which could reflect their higher methane abundance (800 versus 700 ppb) and STE (600 versus 500 Tg a^{-1}).

[27] Also shown in Figure 2 is the effect of 2000–2050 climate change on this natural ozone background. Mixing ratios decrease by up to 1.5 ppb along the coasts, reflecting the general decrease of lower tropospheric ozone over the oceans discussed in the previous section. This effect is compounded in the southeast by increasing biogenic isoprene emissions (Table 2b), which leads to net ozone destruction under low-NO_x conditions [*Sillman et al.*, 1990; *Fiore et al.*, 2005; *Wu et al.*, 2008]. By contrast, we find that the natural background ozone increases by 1–2 ppb in the southwest and the Great Plains, largely driven by increasing soil NO_x emissions due to higher temperatures (Table 2b).

[28] Figure 3 shows the summer afternoon surface mixing ratios of PRB ozone (defined by shutting off anthropogenic emissions in North America) for 2000 conditions, along with the perturbations due to 2000–2050 global change. The 2000–2050 change in anthropogenic emissions increases the mean summer PRB ozone by 3–6 ppb in the west and 2–3 ppb in the east. The 2000–2050 climate change decreases the mean summertime PRB ozone in the United States by up to 3 ppb, except in the southwest and Great Plains where it increases by up to 1 ppb because of increase in soil NO_x emissions. *Murazaki and Hess* [2006] previously calculated a 0–2 ppb decrease of summer background ozone throughout the United States in response

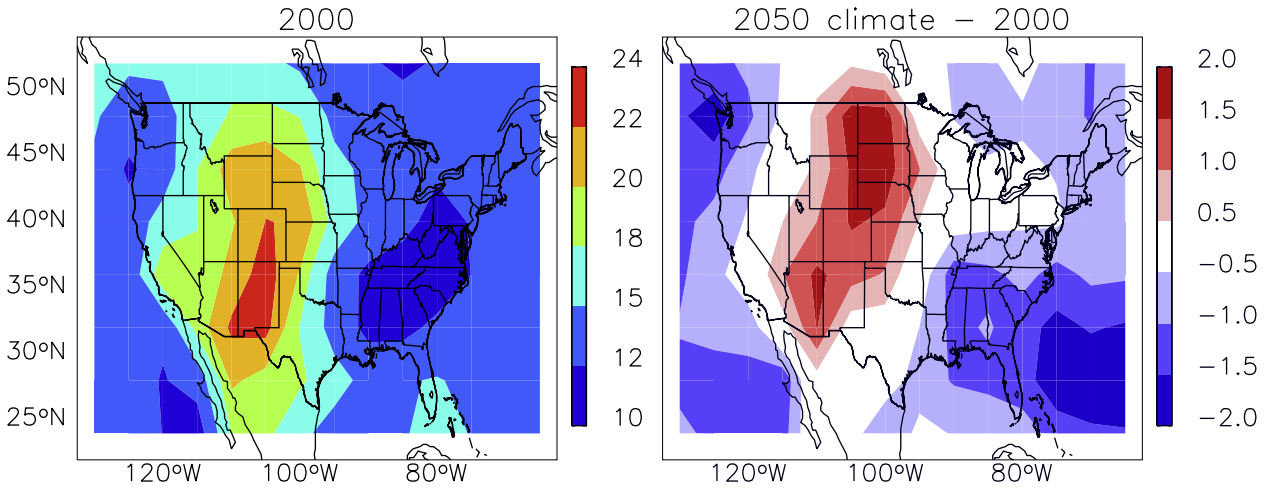

Figure 2. Mean natural ozone concentrations (ppb) in the United States in surface air for summer (June–August) afternoon (1300–1700 LT): model values for 2000 climate (left) and perturbation from 2000 to 2050 climate change (right).

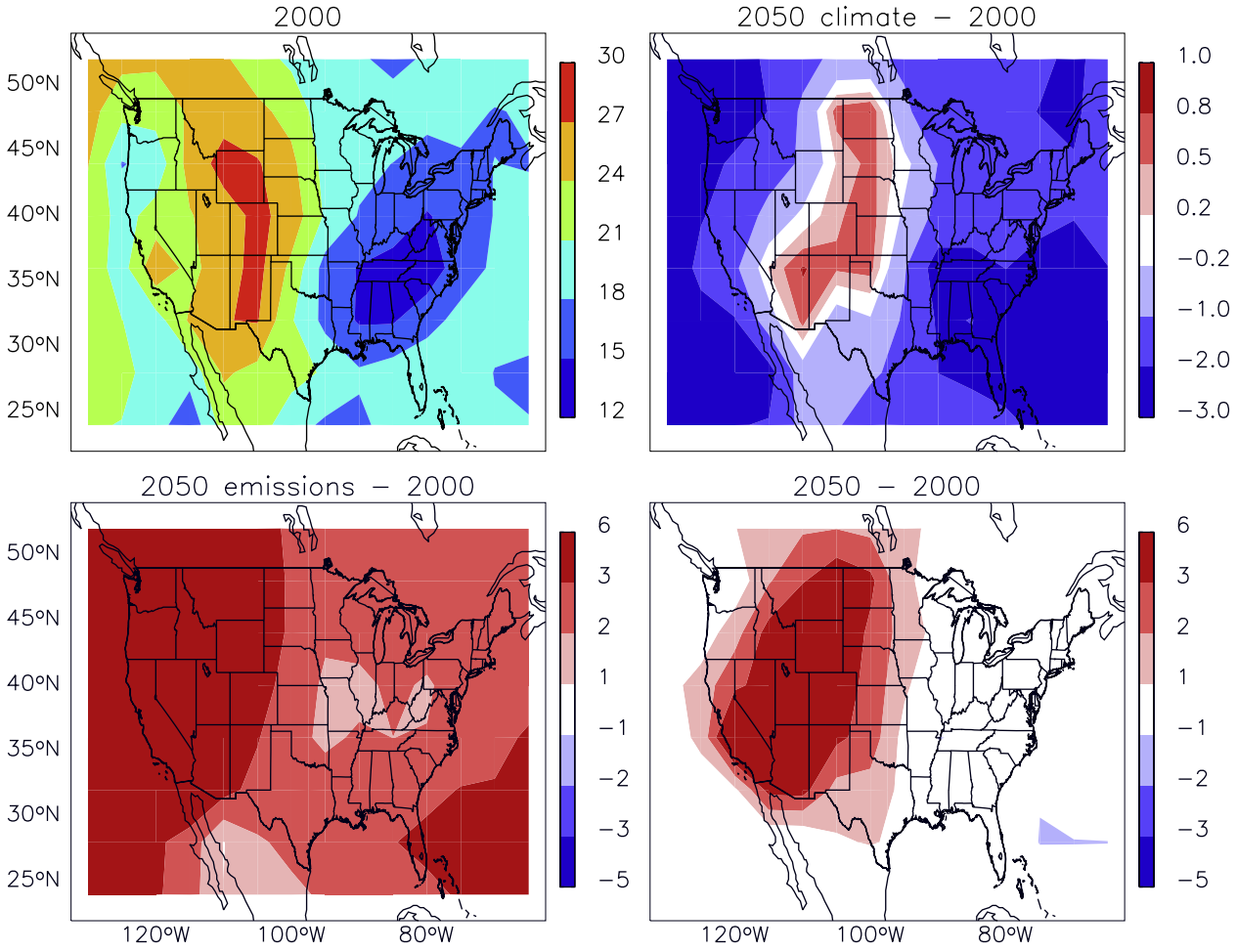

Figure 3. (top left) Mean simulated policy-relevant background (PRB) ozone in U.S. surface air in summer afternoon (June–August, 1300–1700 LT) for present-day conditions. PRB ozone is defined by a simulation with North American anthropogenic emissions shut off. Perturbations to PRB ozone from (top right) changes in climate (2050 climate–2000), (bottom left) changes in anthropogenic emissions (2050 emissions–2000), and (bottom right) changes in both (2050–2000). Units are in ppb. Note differences in scales.

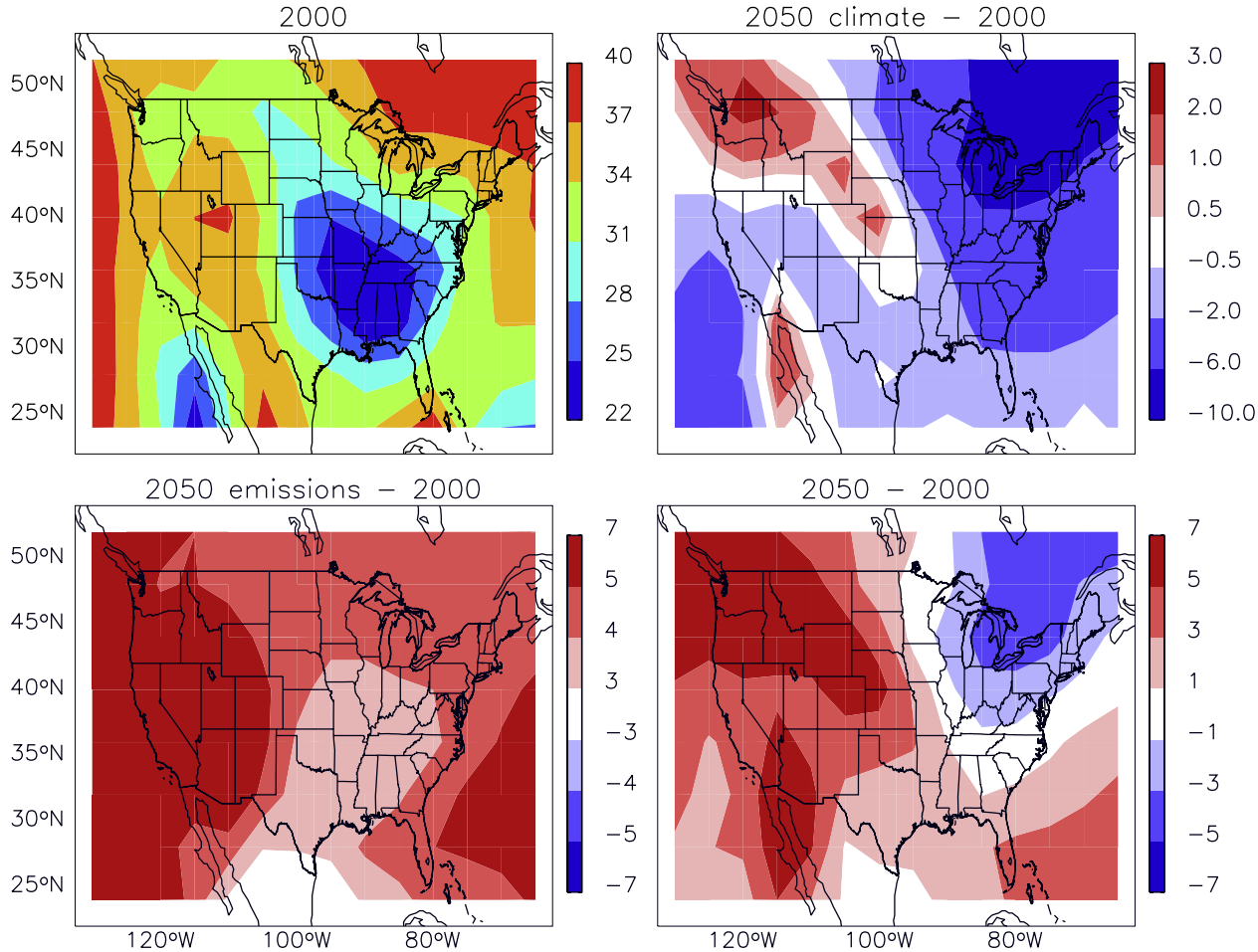

Figure 4. Same as Figure 3, but for April.

to 2000–2100 climate change with the A1B scenario, but they did not account for the effect of changing soil NO_x emissions.

[29] Combination of the 2000–2050 changes in climate and anthropogenic emissions outside North America leads to compensating effects on PRB ozone in the eastern United States, where the two effects cancel. In the western United States the effect of changing external emissions is larger so that surface ozone is projected to increase by 1–5 ppb because of the two combined effects. We find that the effects of changes in climate and anthropogenic emissions on PRB ozone are not additive because of the nonlinearity in ozone chemistry.

[30] The increase of PRB ozone in the United States with 2050 anthropogenic emissions is larger during spring, when the ozone lifetime is longer and the intercontinental transport from Asia is stronger than in summer [*Liu et al.*, 2003]. We find that the largest effect is in April, when 2050 emissions increase the PRB ozone by 4–7 ppb in the western United States and 3–5 ppb in the east (Figure 4). This implies that the 2000–2050 projected changes in anthropogenic emissions outside North America will not only exacerbate the ozone pollution in the summer but also increase the length of the ozone pollution season, as previously pointed out by *Fiore et al.* [2002a].

[31] The effect of 2000–2050 climate change on PRB ozone is also much larger in April than in summer (Figure 4 versus Figure 3) and shows different patterns. There are increases in the northwest (up to 3 ppb) and large decreases in the northeast (up to 10 ppb). These changes appear largely driven by changes in the atmospheric circulation, particularly the northward shifts of the midlatitude westerly flow and the Aleutian Low as results of the greenhouse warming [*IPCC*, 2007]. The poleward displacement of the midlatitude westerlies reduces the transport of ozone from high latitudes to the northeast while the poleward shift of the Aleutian Low allows more Asian pollution to reach northwest. Changes in boundary layer mixing depth, which expands in the model by up to 200 m in the northwest and shrinks by up to 300 m in the northeast, can also contribute to the changes in PRB ozone.

5. Conclusions

[32] We investigated the effects of 2000–2050 global change in climate and emissions on global tropospheric ozone and background surface ozone concentrations in the United States by using a global chemical transport model (GEOS-Chem CTM) with meteorological input from a general circulation model (GISS GCM 3). We considered separately and then together the effects of changes in

climate and changes in global anthropogenic emissions of ozone precursors based on the *IPCC* [2001] A1B scenario. The A1B scenario describes a future world with rapid economic growth and introduction of new technologies, reduction in regional differences of per capita income, and balanced energy generation from fossil and alternative fuels. The NO_x emissions from fossil fuel in that scenario are projected to decrease by 40% in the United States by 2050 but increase by 90% globally.

[33] We began by examining the effects of changing climate and anthropogenic emissions on the global budget of tropospheric ozone. We define as anthropogenic the emissions of NO_x, CO, and NMVOCs from fuel use, industry, and fertilizer application but not those from open biomass burning. Our results indicate that the projected 2000–2050 changes in anthropogenic emissions of ozone precursors would increase the global burden of tropospheric ozone by 17%, reflecting the increase of NO_x and methane emissions. In contrast to most previous studies that found the tropospheric ozone burden to decrease with climate change, we find that climate change increases the tropospheric ozone burden by 1.6%. This is largely driven in our model by an 18% increase of NO_x emissions from lightning, reflecting increased deep convection, and compensating for the shorter lifetime of ozone due to increased water vapor. Increasing stratosphere-troposphere exchange (STE) due to climate change [*Collins et al.*, 2003; *Zeng and Pyle*, 2003; *Sudo et al.*, 2003] is not included in our model and would further increase the tropospheric ozone burden. We find that the largest increase in ozone due to climate change is in the tropical upper troposphere (up to 20%), where ozone is an important greenhouse gas. On the other hand, ozone in the remote lower troposphere is calculated to decrease because of the increase in water vapor. As a result, the background surface ozone imported into the United States is expected to decrease.

[34] We examined the effect of 2000–2050 climate change on the natural ozone background in the United States (no anthropogenic emissions worldwide). The simulated present-day summer afternoon average mixing ratios of natural ozone in surface air are 10–15 ppb in the eastern United States and 15–25 ppb in the west. The 2000–2050 climate change decreases natural ozone background in the east by up to 1.5 ppb, reflecting enhanced ozone destruction from increased water vapor as well as increased biogenic isoprene. In contrast, natural ozone background in the southwest and the Great Plains increase by up to 2 ppb, largely driven by soil NO_x emissions.

[35] We then examined the effect of 2000–2050 changes in climate and anthropogenic emissions on the policy-relevant background (PRB) ozone, defined as the ozone that would be present in surface air over the United States in the absence of anthropogenic emissions in North America. The 2000–2050 climate change decreases the summertime PRB ozone in the United States by up to 1–3 ppb except over the Great Plains where soil NO_x emissions cause a small increase. The decrease of PRB ozone as a result of climate change is mostly driven by the decrease of intercontinental pollution enhancement associated with the shorter lifetimes of ozone and PAN in the warmer wetter climate.

[36] The 2000–2050 changes in anthropogenic emissions of ozone precursors outside of North America increase the mean summer PRB ozone by 3–6 ppb in the western United States and by 2–3 ppb in the east. The effects of 2000–2050 global changes in climate and anthropogenic emissions on background ozone cancel out for the eastern United States resulting in unchanged PRB ozone in 2050 compared to 2000. For the west, the combined effects are dominated by the effects from changing anthropogenic emissions and the PRB ozone increases by 2–5 ppb.

[37] The increase of PRB ozone in the United States due to 2000–2050 changes in anthropogenic emissions outside of North America is larger in spring than in summer. We find that the April PRB ozone increases by 4–7 ppb in the west and 3–5 ppb in the east, which implies that the ozone pollution season in the United States could become longer. Climate change is also found to have large regional effects on the April PRB ozone in our model through perturbation to atmospheric circulation and mixing depths. The poleward shifts of the midlatitude westerly flow and the Aleutian Low as well as the changes in mixing depth (decreases in the northeast but increases in the northwest) lead to decreases of PRB ozone by up to 10 ppb in the northeast and increases by up to 3 ppb in the northwest.

[38] **Acknowledgments.** This work was supported by the U.S. EPA STAR Program. We thank Arlene Fiore (NOAA GFDL) for help in processing the emission growth factors.

References

Bey, I., D. J. Jacob, R. M. Yantosca, J. A. Logan, B. D. Field, A. M. Fiore, Q. Li, H. Y. Liu, L. J. Mickley, and M. G. Schultz (2001), Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation, *J. Geophys. Res.*, *106*, 23,073–23,095, doi:10.1029/2001JD000807.

Boersma, K. F., H. J. Eskes, E. W. Meijer, and H. M. Kelder (2005), Estimates of lightning NO_x production from GOME satellite observations, *Atmos. Chem. Phys.*, *5*, 2311–2331.

Brasseur, G. P., M. Schultz, C. Granier, M. Saunois, T. Diehl, M. Botzet, E. Roeckner, and S. Walters (2006), Impact of climate change on the future chemical composition of the global troposphere, *J. Clim.*, *19*, 3932–3951, doi:10.1175/JCLI3832.1.

Butchart, N., et al. (2006), Simulations of anthropogenic change in the strength of the Brewer-Dobson circulation, *Clim. Dyn.*, *27*, 727–741, doi:10.1007/s00382-006-0162-4.

Centritto, M., P. Nascetti, L. Petrilli, A. Raschi, and F. Loreto (2004), Profiles of isoprene emission and photosynthetic parameters in hybrid poplars exposed to free-air CO_2 enrichment, *Plant Cell Environ.*, *27*, 403–412, doi:10.1111/j.1365-3040.2003.01156.x.

Collins, W. J., R. G. Derwent, B. Garnier, C. E. Johnson, M. G. Sanderson, and D. S. Stevenson (2003), Effect of stratosphere-troposphere exchange on the future tropospheric ozone trend, *J. Geophys. Res.*, *108*(D12), 8528, doi:10.1029/2002JD002617.

Constable, J. V. H., A. B. Guenther, D. S. Schimel, and R. K. Monson (1999), Modelling changes in VOC emission in response to climate change in the continental United States, *Global Change Biol.*, *5*, 791–806.

Del Genio, A. D., M.-S. Yao, and J. Jonas (2007), Will moist convection be stronger in a warmer climate?, *Geophys. Res. Lett.*, *34*, L16703, doi:10.1029/2007GL030525.

Dentener, F., et al. (2006), The global atmospheric environment for the next generation, *Environ. Sci. Technol.*, *40*, 3586–3594, doi:10.1021/es0523845.

Dlugokencky, E. J., et al. (2003), Atmospheric methane levels off: Temporary pause or a new steady state, *Geophys. Res. Lett.*, *30*(19), 1992, doi:10.1029/2003GL018126.

Fiore, A. M., D. J. Jacob, B. D. Field, D. G. Streets, S. D. Fernandes, and C. Jang (2002a), Linking ozone pollution and climate change: The case for controlling methane, *Geophys. Res. Lett.*, *29*(19), 1919, doi:10.1029/2002GL015601.

Fiore, A. M., D. J. Jacob, I. Bey, R. M. Yantosca, B. D. Field, A. C. Fusco, and J. G. Wilkinson (2002b), Background ozone over the United States in

summer: Origin, trend, and contribution to pollution episodes, *J. Geophys. Res.*, *107*(D15), 4275, doi:10.1029/2001JD000982.

Fiore, A., D. J. Jacob, H. Liu, R. M. Yantosca, T. D. Fairlie, and Q. Li (2003a), Variability in surface ozone background over the United States: Implications for air quality policy, *J. Geophys. Res.*, *108*(D24), 4787, doi:10.1029/2003JD003855.

Fiore, A. M., D. J. Jacob, R. Mathur, and R. V. Martin (2003b), Application of empirical orthogonal functions to evaluate ozone simulations with regional and global models, *J. Geophys. Res.*, *108*(D14), 4431, doi:10.1029/2002JD003151.

Fiore, A. M., L. W. Horowitz, D. W. Purves, H. Levy II, M. J. Evans, Y. Wang, Q. Li, and R. M. Yantosca (2005), Evaluating the contribution of changes in isoprene emissions to surface ozone trends over the eastern United States, *J. Geophys. Res.*, *110*, D12303, doi:10.1029/2004JD005485.

Forster, P., et al. (2007), Changes in atmospheric constituents and in radiative forcing, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 130–234, Cambridge Univ. Press, New York.

Guenther, A., et al. (1995), A global model of natural volatile organic compound emissions, *J. Geophys. Res.*, *100*, 8873–8892.

Hansen, J., et al. (2002), Climate forcings in Goddard Institute for Space Studies SI2000 simulations, *J. Geophys. Res.*, *107*(D18), 4347, doi:10.1029/2001JD001143.

Hauglustaine, D. A., and G. P. Brasseur (2001), Evolution of tropospheric ozone under anthropogenic activities and associated radiative forcing of climate, *J. Geophys. Res.*, *106*, 32,337–32,360, doi:10.1029/2001JD900175.

Hauglustaine, D. A., J. Lathière, S. Szopa, and G. A. Folberth (2005), Future tropospheric ozone simulated with a climate-chemistry-biosphere model, *Geophys. Res. Lett.*, *32*, L24807, doi:10.1029/2005GL024031.

Hudman, R. C., et al. (2004), Ozone production in transpacific Asian pollution plumes and implications for ozone air quality in California, *J. Geophys. Res.*, *109*, D23S10, doi:10.1029/2004JD004974.

IMAGE Team (2001), The Image 2.2 implementation of the SRES scenarios, *Publ. 481508018*, [CD-ROM], Natl. Inst. for Public Heath and the Environ., Bilthoven, Netherlands, July.

Intergovernmental Panel on Climate Change (IPCC) (1995), *Climate Change 1995: The Science of Climate Change. Contribution of Working Group I to the Second Assessment of the Intergovernmental Panel on Climate Change*, edited by J. T. Houghton et al., 572 pp., Cambridge Univ. Press, Cambridge, New York.

Intergovernmental Panel on Climate Change (IPCC) (2000), *IPCC Special Report on Emissions Scenarios*, edited by N. Nakicenovic et al., Cambridge Univ. Press, Cambridge, U. K.

Intergovernmental Panel on Climate Change (IPCC) (2001), *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, edited by J. T. Houghton et al., Cambridge Univ. Press, New York.

Intergovernmental Panel on Climate Change (IPCC) (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., 996 pp., Cambridge Univ. Press, New York.

Jacob, D. J., J. A. Logan, G. M. Gardner, R. M. Yevich, C. M. Spivakovsky, S. C. Wofsy, S. Sillman, and M. J. Prather (1993), Factors regulating ozone over the United States and its export to the global atmosphere, *J. Geophys. Res.*, *98*, 14,817–14,826, doi:10.1029/98JD01224.

Jacob, D. J., J. A. Logan, and P. P. Murti (1999), Effect of rising Asian emissions on surface ozone in the United States, *Geophys. Res. Lett.*, *26*, 2175–2178, doi:10.1029/1999GL900450.

Jaffe, D., and J. Ray (2007), Increase in surface ozone at rural sites in the western U.S., *Atmos. Environ.*, *41*, 5452–5463, doi:10.1016/j.atmosenv.2007.02.034.

Jaffe, D., I. McKendry, T. Anderson, and H. Price (2003), Six "new" episodes of trans-Pacific transport of air pollutants, *Atmos. Environ.*, *37*, 391–404, doi:10.1016/S1352-2310(02)00862-2.

Johnson, C. E., W. J. Collins, D. S. Stevenson, and R. G. Derwent (1999), The relative roles of climate and emissions changes on future oxidant concentrations, *J. Geophys. Res.*, *104*(D15), 18,631–18,645, doi:10.1029/1999JD900204.

Labrador, L. J., R. von Kuhlmann, and M. G. Lawrence (2004), Strong sensitivity of the global mean OH concentration and the tropospheric oxidizing efficiency to the source of NO_x from lightning, *Geophys. Res. Lett.*, *31*, L06102, doi:10.1029/2003GL019229.

Labrador, L. J., R. von Kuhlmann, and M. G. Lawrence (2005), The effects of lightning-produced NO_x and its vertical distribution on atmospheric chemistry: Sensitivity simulations with MATCH-MPIC, *Atmos. Chem. Phys.*, *5*, 1815–1834.

Lambert, S. J., and J. C. Fyfe (2006), Changes in winter cyclone frequencies and strengths simulated in enhanced greenhouse warming experiments: Results from the models participating in the IPCC diagnostic exercise, *Clim. Dyn.*, *26*, 713–728, doi:10.1007/s00382-006-0110-3.

Langner, J., R. Bergstrom, and V. Foltescu (2005), Impact of climate change on surface ozone and deposition of sulphur and nitrogen in Europe, *Atmos. Environ.*, *39*, 1129–1141, doi:10.1016/j.atmosenv.2004.09.082.

Lelieveld, J., and F. J. Dentener (2000), What controls tropospheric ozone?, *J. Geophys. Res.*, *105*, 3531–3551, doi:10.1029/1999JD901011.

Liao, H., W.-T. Chen, and J. H. Seinfeld (2006), Role of climate change in global predictions of future tropospheric ozone and aerosols, *J. Geophys. Res.*, *111*, D12304, doi:10.1029/2005JD006852.

Lin, C. Y. C., D. J. Jacob, J. W. Munger, and A. M. Fiore (2000), Increasing background ozone in surface air over the United States, *Geophys. Res. Lett.*, *27*, 3465–3468, doi:10.1029/2000GL011762.

Lin, J. T., K. O. Patten, K. Hayhoe, X. Z. Liang, and D. J. Wuebbles (2008), Effects of future climate and biogenic emissions changes on surface ozone over the United States and China, *J. Appl. Meteorol. Climatol.*, *47*(7), 1888–1909.

Liu, H. Y., D. J. Jacob, I. Bey, R. M. Yantosca, B. N. Duncan, and G. W. Sachse (2003), Transport pathways for Asian combustion outflow over the Pacific: Interannual and seasonal variations, *J. Geophys. Res.*, *108*(D20), 8786, doi:10.1029/2002JD003102.

Martin, R. V., et al. (2002), Interpretation of TOMS observations of tropical tropospheric ozone with a global model and in situ observations, *J. Geophys. Res.*, *107*(D18), 4351, doi:10.1029/2001JD001480.

McLinden, C. A., et al. (2000), Stratospheric ozone in 3-D models: A simple chemistry and the cross-tropopause flux, *J. Geophys. Res.*, *105*, 14,653–14,665.

Mickley, L. J., P. P. Murti, D. J. Jacob, and J. A. Logan (1999), Radiative forcing from tropospheric ozone calculated with a unified chemistry climate model, *J. Geophys. Res.*, *104*, 30,153–30,172, doi:10.1029/1999JD900439.

Murazaki, K., and P. Hess (2006), How does climate change contribute to surface ozone change over the United States?, *J. Geophys. Res.*, *111*, D05301, doi:10.1029/2005JD005873.

Pickering, K. E., Y. Wang, W. Tao, C. Price, and J. Muller (1998), Vertical distributions of lightning NO_x for use in regional and global chemical transport models, *J. Geophys. Res.*, *103*, 31,203–31,216.

Prather, M. J. (1996), Time scales in atmospheric chemistry: Theory, GWPs for CH_4 and CO, and runaway growth, *Geophys. Res. Lett.*, *23*, 2597–2600, doi:10.1029/96GL02371.

Prather, M., et al. (2001), Atmospheric chemistry and greenhouse gases, in *Climate Change 2001: The Scientific Basis. Contribution of Working Group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change*, edited by J. T. Houghton et al., pp. 239–288, Cambridge Univ. Press, New York.

Prather, M., et al. (2003), Fresh air in the 21st century?, *Geophys. Res. Lett.*, *30*(2), 1100, doi:10.1029/2002GL016285.

Price, C., and D. Rind (1992), A simple lightning parameterization for calculating global lightning distributions, *J. Geophys. Res.*, *97*, 19,919–19,933, doi:10.1029/92JA01459.

Price, C., and D. Rind (1994), Possible implications of global climate change on global lightning distributions and frequencies, *J. Geophys. Res.*, *99*, 10,823–10,831, doi:10.1029/94JD00019.

Price, C., J. Penner, and M. Prather (1997), NO_x from lightning: 1. Global distribution based on lightning physics, *J. Geophys. Res.*, *102*, 5929–5941, doi:10.1029/96JD03504.

Racherla, P. N., and P. J. Adams (2006), Sensitivity of global tropospheric ozone and fine particulate matter concentrations to climate change, *J. Geophys. Res.*, *111*, D24103, doi:10.1029/2005JD006939.

Randall, D. A. (2007), Climate models and their evaluation, in *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., pp. 590–662, Cambridge Univ. Press, Cambridge, U. K.

Rind, D., J. Lerner, J. Jonas, and C. McLinden (2007), Effects of resolution and model physics on tracer transports in the NASA Goddard Institute for Space Studies general circulation models, *J. Geophys. Res.*, *112*, D09315, doi:10.1029/2006JD007476.

Rosenstiel, T. N., M. J. Potosnak, K. L. Griffin, R. Fall, and R. K. Monson (2003), Increased CO_2 uncouples growth from isoprene emission in an agriforest ecosystem, *Nature*, *421*, 256–259, doi:10.1038/nature01312.

Sanderson, M. G., C. D. Jones, W. J. Collins, C. E. Johnson, and R. G. Derwent (2003), Effect of climate change on isoprene emissions and surface ozone levels, *Geophys. Res. Lett.*, *30*(18), 1936, doi:10.1029/2003GL017642.

Sauvage, B., et al. (2007), Remote sensed and in situ constraints on processes affecting tropical tropospheric ozone, *Atmos. Chem. Phys.*, *7*, 815–838.

Sillman, S., and P. J. Samson (1995), The impact of temperature on oxidant formation in urban, polluted rural, and remote environments, *J. Geophys. Res.*, *100*, 11,497–11,508, doi:10.1029/94JD02146.

Sillman, S., J. A. Logan, and S. C. Wofsy (1990), The sensitivity of ozone to nitrogen oxides and hydrocarbons in regional ozone episodes, *J. Geophys. Res.*, *95*, 1837–1851, doi:10.1029/JD095iD02p01837.

Stevenson, D. S., et al. (2000), Future tropospheric ozone radiative forcing and methane turnover: The impact of climate change, *Geophys. Res. Lett.*, *27*, 2073–2076, doi:10.1029/1999GL010887.

Stevenson, D. S., R. M. Doherty, M. G. Sanderson, C. E. Johnson, W. J. Collins, and R. G. Derwent (2005), Impacts of climate change and variability on tropospheric ozone and its precursors, *Faraday Discuss.*, *130*, doi:10.1039/b417412g.

Stevenson, D. S., et al. (2006), Multimodel ensemble simulations of present-day and near-future tropospheric ozone, *J. Geophys. Res.*, *111*, D08301, doi:10.1029/2005JD006338.

Stocks, B. J., et al. (1998), Climate change and forest fire potential in Russian and Canadian boreal forests, *Clim. Change*, *38*, 1–13, doi:10.1023/A:1005306001055.

Sudo, K., M. Takahashi, and H. Akimoto (2003), Future changes in stratosphere-troposphere exchange and their impacts on future tropospheric ozone simulations, *Geophys. Res. Lett.*, *30*(24), 2256, doi:10.1029/2003GL018526.

Unger, N., D. T. Shindell, D. M. Koch, M. Amann, J. Cofala, and D. G. Streets (2006), Influences of man-made emissions and climate changes on tropospheric ozone, methane, and sulfate at 2030 from a broad range of possible futures, *J. Geophys. Res.*, *111*, D12313, doi:10.1029/2005JD006518.

U.S. Environmental Protection Agency (EPA) (2006), Air quality criteria for ozone and related photochemical oxidants, *Rep. EPA/600/R-05/004aF-cF,* Washington, D. C.

U.S. Environmental Protection Agency (EPA) (2007), Report on air quality in nonattainment areas for 2003–2005 covering ozone, particulate matter, carbon monoxide, sulfur dioxide, nitrogen dioxide, and lead, Washington, D. C. (Available at http://www.epa.gov/air/airtrends/pdfs/20070214_aq_na_2003-2005.pdf)

Wang, Y., and D. J. Jacob (1998), Anthropogenic forcing on tropospheric ozone and OH since preindustrial times, *J. Geophys. Res.*, *103*, 31,123–31,135, doi:10.1029/1998JD100004.

Wang, Y., D. J. Jacob, and J. A. Logan (1998), Global simulation of tropospheric O_3-NO_x-hydrocarbon chemistry: 3. Origin of tropospheric ozone and effects of nonmethane hydrocarbons, *J. Geophys. Res.*, *103*, 10,757–10,767, doi:10.1029/98JD00156.

Wild, O., and H. Akimoto (2001), Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, *J. Geophys. Res.*, *106*, 27,729–27,744, doi:10.1029/2000JD000123.

Wu, S., L. J. Mickley, D. J. Jacob, J. A. Logan, R. M. Yantosca, and D. Rind (2007), Why are there large differences between models in global budgets of tropospheric ozone?, *J. Geophys. Res.*, *112*, D05302, doi:10.1029/2006JD007801.

Wu, S., L. J. Mickley, E. M. Leibensperger, D. J. Jacob, D. Rind, and D. G. Streets (2008), Effects of 2000–2050 global change on ozone air quality in the United States, *J. Geophys. Res.*, *113*, D06302, doi:10.1029/2007JD008917.

Wuebbles, D. J., and K. Hayhoe (2002), Atmospheric methane and global change, *Earth Sci. Rev.*, *57*, 177–210, doi:10.1016/S0012-8252(01)00062-9.

Yienger, J. J., and H. Levy II (1995), Empirical model of global soil-biogenic NO_x emissions, *J. Geophys. Res.*, *100*(D6), 11,447–11,464, doi:10.1029/95JD00370.

Zeng, G., and J. A. Pyle (2003), Changes in tropospheric ozone between 2000 and 2100 modeled in a chemistry-climate model, *Geophys. Res. Lett.*, *30*(7), 1392, doi:10.1029/2002GL016708.

D. J. Jacob, L. J. Mickley, and S. Wu, Department of Earth and Planetary Sciences, Harvard University, Cambridge, MA 02138, USA. (s.wu@post.harvard.edu)

D. Rind, NASA Goddard Institute for Space Studies, 2880 Broadway, New York, NY 10025, USA.

D. G. Streets, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439, USA.