Effects of intercontinental transport on surface ozone over the United States: Present and future assessment with a global model

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[1] Future changes in intercontinental transport (ICT) of European and Asian (EURA) pollutants can have substantial consequences on U.S. pollution. This is investigated with 27 simulations using a newly improved version of the Model for OZone And Related chemical Tracers to simulate ICT contributions to summertime U.S. surface ozone for 1999, 2049 and 2099, with future climate and emissions perturbations following the Intergovernmental Panel on Climate Change A1fi and B1 scenarios. We find that projected ICT changes are primarily affected by changes in EURA anthropogenic emissions, whose increases under A1fi lead to 3–8 ppb, 2–6 ppb due to Asian emissions increases alone, more ozone from 1999 to both 2049 and 2099 over the western U.S.; whereas EURA emissions changes under B1 lead to 0–0.7 ppb and 1–2 ppb less ozone by 2049 and 2099, respectively. Therefore global, especially Asian, emission control is important for U.S. pollution mitigation. **Citation:** Lin, J.-T., D. J. Wuebbles, and X.-Z. Liang (2008), Effects of intercontinental transport on surface ozone over the United States: Present and future assessment with a global model, *Geophys. Res. Lett.*, *35*, L02805, doi:10.1029/2007GL031415.

1. Introduction

[2] Surface air quality over the contiguous United States is being affected by the intercontinental transport (ICT), through transpacific and transatlantic pathways, of European and Asian (EURA) pollutants as a result of anthropogenic/natural emissions [*Wilkening et al.*, 2000; *Biscaye et al.*, 2000; *Akimoto*, 2003; *Zhao et al.*, 2006; *Wuebbles et al.*, 2007]. In particular, the transpacific transport (TPT) is the primary process to bring EURA pollutants to the U.S. *Jaffe et al.* [1999] found that the observed ground-level chemical concentrations over the northwest U.S. were greatly affected by episodic pollutant plumes from Asia. A global modeling simulation by *Jacob et al.* [1999] indicated an increasing TPT effects on U.S. ozone from 1985–2010 with tripling Asian anthropogenic emissions. Since then, various ground-level observation analyses, field campaigns, and model simulations have been conducted to investigate the current pathways and characteristics of TPT, as well as their influence on U.S. air quality [e.g., *Yienger et al.*, 2000; *Jaeglé et al.*, 2003; *Hudman et al.*, 2004; *Parrish et al.*, 2004; *Bertschi and Jaffe*, 2005; *Heald et al.*, 2006]. These studies confirmed an important source of U.S. pollutants from Europe and Asia, especially the latter, in recent years. As compared to the TPT, transatlantic transport of EURA pollutants has minor effects on U.S. air quality [*Li et al.*, 2002] due to the prevailing westerly at the northern mid-latitudes.

[3] One critical issue related to the ICT is its changes and resulting impacts on downwind regions over the coming century [*Wuebbles et al.*, 2007]. In future years, the changing EURA anthropogenic emissions are likely to affect U.S. air quality. Specifically, if EURA anthropogenic emissions increase (decrease) in the future, there will likely be more (less) pollutants over the U.S. as a result of the enhancing (reducing) ICT. In addition, climate changes (e.g., air temperature, water vapor) will also influence the ICT by affecting biogenic emissions over source regions, pollutant lifetimes [*Fiore et al.*, 2002; *Murazaki and Hess*, 2006] and transport pathways. While many modeling studies have investigated the overall effects of projected global climate and emissions perturbations on U.S. pollution in future years [e.g., *Intergovernmental Panel on Climate Change* (*IPCC*), 2001, and references therein; *Murazaki and Hess*, 2006; *Lin et al.*, 2008], changes in ICT and their specific effects on U.S. air quality are seldom examined [*Wuebbles et al.*, 2007].

[4] To investigate the effects of future ICT changes on U.S. pollution, this study uses the global chemical transport model (CTM), Model for OZone And Related chemical Tracers version 2.4 (MOZART-2.4), with additional improvements in model chemistry/physics, to simulate the effects of ICT on U.S. ozone levels for the year of 1999 and project their changes from 1999 to 2049 and to 2099 due to changes in global climate and EURA anthropogenic and biogenic emissions. However, future changes in climate and emissions are determined by both natural variability and human activities, projections of which contain large uncertainties. Therefore two distinct climate/emission pathways from the Intergovernmental Panel on Climate Change (IPCC) Special Report on Emissions Scenarios (SRES, http://sres.ciesin.org/final_data.html) are considered, including the A1fi and B1 scenarios representing upper and lower bounds of climate warming over the 21st century, respectively. The latest estimate of global temperature change from 1980–1999 to 2080–2099 is 4.0°C under the A1fi scenario and 1.8°C under the B1 scenario [*IPCC*, 2007]. The corresponding changes in ozone precursor emissions are more complicated and dependent on individual species, as shown in Table 1. These two scenarios have been used by *Lin et al.* [2008] to study the overall surface ozone changes over the U.S. and China. The analyses focus on summer months (June–August), when the U.S. ozone concentrations typically maximize and when any increases in ICT

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Table 1. Projected Summer Emission Budgets of Ozone Precursor Over Europe and Asia Together for 1999 and Their Fractional Changes by 2049 and 2099

Species	Year	Anthropogenic	Biogenic	Total
NO_x	1999	4.5 TgN	1.6 TgN	6.2 TgN
NO_x	2049	294% / 23%[a]	0% / 0%	217% / 17%
NO_x	2099	314% / −45%	0% / 0%	232% / −33%
CO	1999	109.7 Tg	26.7 Tg	136.4 Tg
CO	2049	296% / −30%	2% / 1%	239% / −24%
CO	2099	332% / −42%	4% / 2%	267% / −34%
Isoprene	1999	—	47.9 TgC	47.9 TgC
Isoprene	2049	—	41% / 20%	41% / 20%
Isoprene	2099	—	74% / 35%	74% / 35%

[a]The numbers before and after '/' denote projected fractional emissions changes under the A1fi and B1 scenarios, respectively, relative to the 1999 budgets.

would have significant consequences, e.g., increasing non-attainment of the national ozone standard.

2. Experiment Design

[5] MOZART-2.4 (described and evaluated in detail by *Horowitz et al.* [2003]) is a state-of-the-art CTM in studying distributions and changes of ozone and related tracer gases in the troposphere [e.g., *Wuebbles et al.*, 2001; *Wei et al.*, 2002; *Horowitz et al.*, 2003; *Lamarque et al.*, 2005; *Tie et al.*, 2005; *Murazaki and Hess*, 2006; *Lin et al.*, 2008]. This study incorporates in MOZART-2.4 three improvements in model chemistry and physical parameterizations, following *Lin et al.* [2008]. First, there is a likely underprediction of the removal of nitrogen oxides (NO_x) through isoprene nitrate chemistry in the original MOZART-2.4 (i.e., the model prior to the improvements incorporated here) because it likely underestimates the yield of isoprene nitrates and the conversion from isoprene nitrates to nitric acid [see *Fiore et al.*, 2005, and references therein]. In the improved model, the yield of isoprene nitrate is determined as 12% instead of the original 8%, which is then converted immediately to nitric acid as the permanent sink of NO_x, following *Fiore et al.* [2005]. Second, the original MOZART-2.4 may underestimate, by over 30%, the summertime ozone dry deposition over the deciduous forest prevailing in the eastern U.S. [*Padro*, 1996; *Zhang et al.*, 1996; *Wesely and Hicks*, 2000]. Here the summertime ozone dry deposition is increased by 30% over the eastern U.S. (east of 100°W) as a conservative improvement. Third, the critical Richardson number is increased from the original 0.3 to 1.0, which is suggested to be more realistic by more recent research [*Stull*, 1997; *Cheng et al.*, 2002, and references therein]. *Lin et al.* [2008] found that the simulated summer average ozone concentrations by the improved version of MOZART-2.4 are within 20 ppb, often within 10 ppb, of the ground-level measurements over most of the U.S.; overall, the model improvements reduced the simulation biases by 10–20 ppb over the eastern U.S. and have minor effects over the western U.S. Hereafter the model is referred to as the improved version if not otherwise described.

[6] This study conducted 27 experiments for the years of 1999, 2049 and 2099. The detailed experiment design is described in Table 2. These experiments were conducted with the improved version of MOZART-2.4 driven by meteorological data derived from the Parallel Climate Model (PCM [*Washington et al.*, 2000]) outputs. The MOZART resolution is T42 (∼2.8°) horizontally with 18 vertical layers. Following *Lin et al.* [2008], the modeled ozone concentrations were outputted at 3-hour instead of 1-hour intervals to save storage. However, *Lin et al.* [2008] found that the calculation of daily (24 hours) mean ozone was insignificantly affected by this temporal resolution.

[7] Emissions of ozone precursors for 1999 are based on the present-day inventory as described by *Horowitz et al.* [2003]. Emissions projections from 1999 to 2049 and 2099 were described in detail by *Lin et al.* [2008]. Briefly, projected changes in anthropogenic (fossil fuel, industrial, transportation, biofuel, etc.) emissions were derived from the economic models used by *IPCC* [2001], miniCAM for A1fi and IMAGE for B1, respectively. Emissions from biomass burning sources are fixed at the present-day level for all experiments. Projections of biogenic emissions of non-methane volatile organic compounds (VOC) take into account the PCM calculated changes in temperature [after

Table 2. Experiment Design of Climate and Ozone Precursor Emissions

Experiment	Climate[a]	Anthropogenic Emissions[b]
(1) CURRemis	1999	C / C / C[c]
(2) noEURAant	1999	0 / 0 / C
(3) noASIAant	1999	C / 0 / C
(4) A_CURRemis_M	A1fi 2049	C / C / C
(5) A_noEURAant_M	A1fi 2049	0 / 0 / C
(6) A_EURAchgANT_M	A1fi 2049	M_A / M_A / C
(7) A_ASIAchgANT_M	A1fi 2049	C / M_A / C
(8) A_ASIA_B_EUROPE_M	A1fi 2049	M_B / M_A / C
(9) A_EURAchgALL_M[d]	A1fi 2049	M_A / M_A / C
(10) A_EURAchgANT_nox_M[e]	A1fi 2049	M_A / M_A / C
(11) A_EURAchgANT_voc_M[f]	A1fi 2049	M_A / M_A / C
(12) A_CURRemis_L	A1fi 2099	C / C / C
(13) A_noEURAant_L	A1fi 2099	0 / 0 / C
(14) A_EURAchgANT_L	A1fi 2099	L_A / L_A / C
(15) A_ASIAchgANT_L	A1fi 2099	C / L_A / C
(16) A_ASIA_B_EUROPE_L	A1fi 2099	L_B / L_A / C
(17) A_EURAchgALL_L[d]	A1fi 2099	L_A / L_A / C
(18) B_CURRemis_M	B1 2049	C / C / C
(19) B_noEURAant_M	B1 2049	0 / 0 / C
(20) B_EURAchgANT_M	B1 2049	M_B / M_B / C
(21) B_EURAchgALL_M[d]	B1 2049	M_B / M_B / C
(22) B_EURAchgANT_nox_M[e]	B1 2049	M_B / M_B / C
(23) B_EURAchgANT_voc_M[f]	B1 2049	M_B / M_B / C
(24) B_CURRemis_L	B1 2099	C / C / C
(25) B_noEURAant_L	B1 2099	0 / 0 / C
(26) B_EURAchgANT_L	B1 2099	L_B / L_B / C
(27) B_EURAchgALL_L[d]	B1 2099	L_B / L_B / C

[a]Physical environment of the troposphere, including temperature, water vapor, etc.

[b]Including emissions from fossil fuel, industry, transportation, biofuel, etc.

[c]The three symbols separated by '/' denote the emissions over Europe, Asia and the rest of the world, respectively (from left to right), designed for each experiment. Symbol 'C' denotes current emissions; 'M_A' and 'M_B' denote future emissions for the mid 21st century (i.e., 2049) under the A1fi and B1 scenarios, respectively; 'L_A' and 'L_B' denote future emissions for the late 21st century (i.e., 2099) under A1fi and B1 scenarios, respectively; '0' denotes zero emissions. Europe and Asia are separated by 60°E in this study.

[d]Changes in biogenic and soil emissions are incorporated, corresponding to the specified climate scenarios (A1fi or B1) and future years (2049 or 2099) for each experiment.

[e]Changes in anthropogenic emissions are for NO_x only.

[f]Changes in anthropogenic emissions are for CO and VOC only.

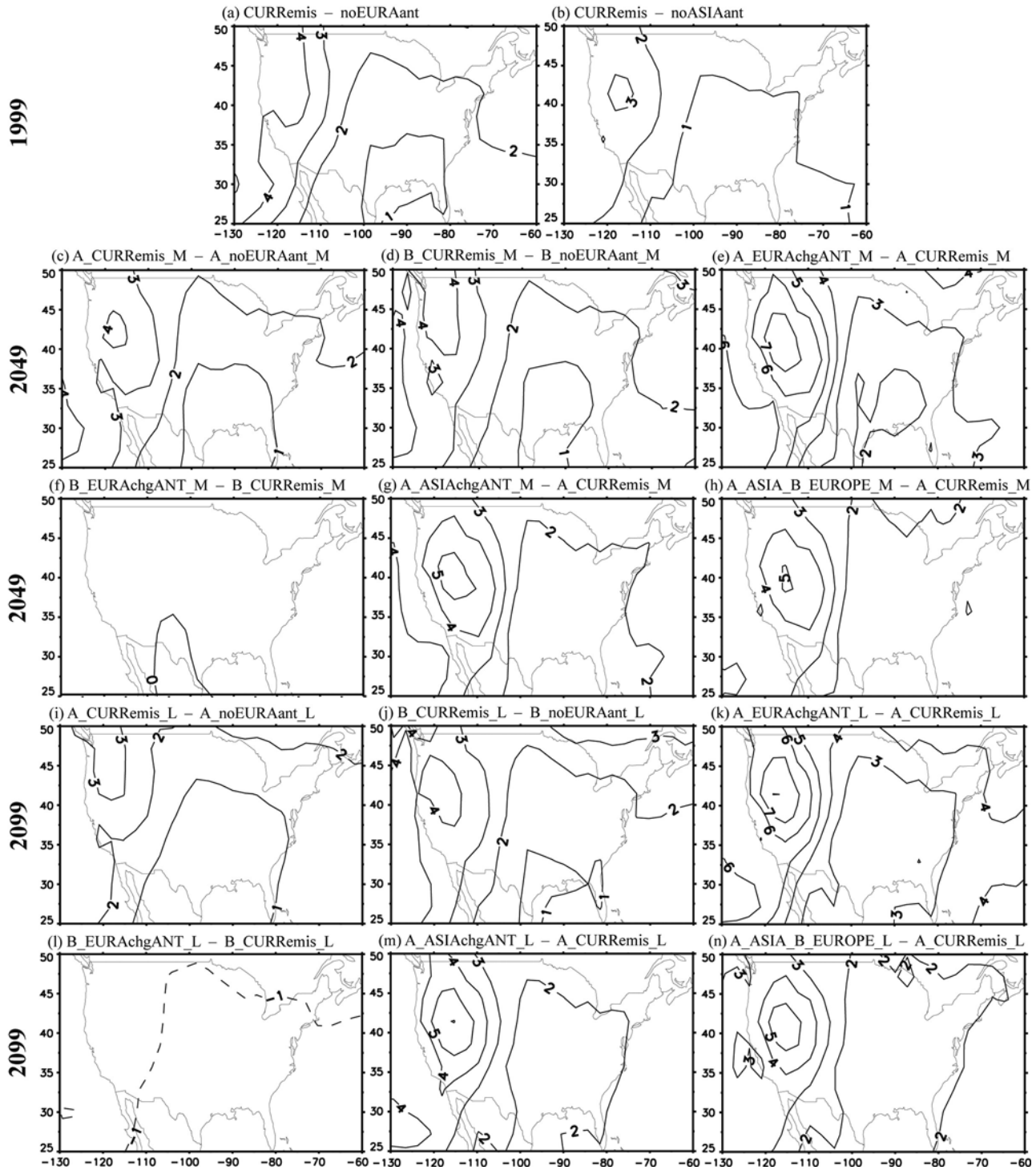

Figure 1. Contributions (in ppb) of intercontinental transport of European and Asian pollutants to summertime average daily mean surface ozone concentrations over the United States for the year of (a and b) 1999, (c–h) 2049, and (i–n) 2099. The contour interval is 1 ppb. The daily means are calculated from the MOZART outputs at 3-hour intervals. See Table 2 for the detailed experiment design.

Guenther, 1997] and carbon dioxide (CO_2) level [after *Potosnak*, 2002]. Changes in soil emissions of carbon monoxide (CO) due to the temperature dependence of net primary production (NPP) were also considered [*Lin et al.*, 2008]. However, soil emissions of NO_x and oceanic emissions of all ozone precursors were kept at the same present-day levels in all experiments because of the large uncertainties in emissions estimations and the difficulty in future projections. The fractional changes in emissions of NO_x,

CO and isoprene from 1999 to 2099 under both the A1fi and B1 scenarios are presented in Table 1.

3. Results

3.1. Current ICT Effects

[8] Figure 1a shows that, during 1999, EURA anthropogenic emissions contribute 4–5 ppb to the summer average daily mean (as calculated from the 3-hour MOZART outputs) surface ozone levels over the northwest U.S. and 3–4 ppb over California. Over the eastern U.S., the ICT effect is less than 2 ppb. The interannual variability of ICT effects is very small during 1996–2000, with the 5-year standard deviation less than 0.5 ppb over the U.S. (see discussions in section 4). Anthropogenic emissions over Asia (east of 60°E) alone contribute 2–4 ppb over the western U.S. and less than 1 ppb over the eastern U.S. (Figure 1b). Our results are similar to those of *Jacob et al.* [1999], who found that tripling Asian anthropogenic emissions upon the 1985 levels, with 12 TgN/yr increases in NO_x emissions, a magnitude of increase similar to this study (10 TgN/yr), would lead to an increase of 1–4 ppb in U.S. surface ozone averaged over the summer months.

[9] Using the global model, GEOS-Chem, *Fiore et al.* [2002] found that 4–7 ppb out of the 1995 summer average afternoon (1:00 pm–5:00 pm) mean U.S. background ozone levels were contributed by EURA anthropogenic emissions. The differences between the results here and those given by *Fiore et al.* [2002] are due to many factors such as meteorology, emissions, resolution, model physics/chemistry and evaluation metric (daily mean ozone vs. afternoon mean ozone). As one factor, for the vertical mixing in the planetary boundary layer (PBL), MOZART incorporates the non-local scheme by *Holtslag and Boville* [1993] capable of simulating various PBL states from unstable in the afternoon to stable in the nighttime; while GEOS-Chem assumes a fully mixed PBL throughout the day, resulting in stronger interaction between the surface and higher altitudes in favor of the ICT. In addition, the horizontal resolution here is much finer than that of $4° \times 5°$ used by *Fiore et al.* [2002], and thus should generally lead to better simulation of the dynamical, physical and chemical processes important to ozone formation and transport.

3.2. Projected Future ICT Effects

[10] Projected future climate change, especially increasing tropospheric water vapor content, is likely to result in a shorter atmospheric lifetime for ozone and thus weaker ICT, if global precursor emissions are fixed at the present-day levels [see also *Fiore et al.*, 2002; *Murazaki and Hess*, 2006]. It is found that, from 1999 to 2049, the northern hemisphere tropospheric (below 220 hPa) water vapor content derived by PCM is increased by about 9% under A1fi and 2% under B1; the derived increases from 1999 to 2099 are 26% under A1fi and 5% under B1. As a result, from 1999 to 2049, projected contributions of ICT to U.S. ozone are slightly reduced, i.e., by less than 0.5 ppb over most of the country, under both scenarios (comparing Figures 1a, 1c, and 1d); the ICT reductions from 1999 to 2099 are more significant, especially under the A1fi scenario where the reduction exceeds 1 ppb over the western U.S. (comparing Figures 1a, 1i, and 1j).

[11] Projected changes in EURA anthropogenic emissions can have significant impacts on the ICT. Over the coming century, anthropogenic emissions of ozone precursors over Europe and Asia are projected to change greatly and distinctively under the A1fi and B1 scenarios, relative to the present-day levels (Table 1). As a result, from 1999 to 2049, increases in EURA anthropogenic emissions under A1fi lead to 3–8 ppb more surface ozone over the western U.S. and 1–3 ppb more ozone over the eastern U.S. (Figure 1e), about one third of which are contributed by the increased NO_x emissions and two thirds by the increased emissions of CO and VOC together (not shown). Under B1, by comparison, the ICT effect is slightly reduced, i.e., by not more than 0.7 ppb (Figure 1f). The magnitudes of ICT changes from 1999 to 2099 are about 0.5 ppb larger than those from 1999 to 2049 under both scenarios due to the greater emissions changes (Figures 1k and 1l). Overall, under A1fi, total EURA anthropogenic emissions contribute to 3–11 ppb of surface ozone over the western U.S. and 2–4 ppb over the eastern U.S. in both 2049 and 2099; whereas under B1, contributions of total EURA anthropogenic emissions reduce to less than 4 ppb in 2049 and less than 3 ppb in 2099 over the western U.S.

[12] Over the coming decades, it is more likely that anthropogenic emissions over Europe will be reduced as a result of the pollution control policies while Asian emissions will likely grow, at least for the next few decades as a result of the fast on-going industrialization and economic/population growth, although in the longer term, these emissions may also be reduced under stronger policy controls. Therefore it is of interest to investigate the effects of this likely emission trend on the ICT. To this regard, we find that, from 1999 to 2049, the summer average U.S. ozone concentrations increase 2–6 ppb over the western region and 1–2 ppb over the eastern region due to the increasing Asian anthropogenic emissions following the A1fi scenario, whether European anthropogenic emissions are fixed at present-day levels (Figure 1g) or even allowed to change following the B1 scenario (Figure 1h). The corresponding ICT changes from 1999 to 2099 are about 0.5 ppb higher (Figures 1m and 1n). This suggests that the projected Asian emissions increases dominate the future ICT enhancement and, if indeed they occurred, would greatly impact the U.S. pollution levels. Projected changes in European emissions, by comparison, have a much smaller effect on U.S. ozone, i.e., for both 2049 and 2099, calculated surface ozone concentrations increase 1–3 ppb over the western U.S. when European emissions increase from the B1 to A1fi case.

[13] Projected future changes in EURA biogenic emissions impact the ICT as well. From 1999 to 2049 and 2099, projected EURA biogenic emissions of isoprene greatly increase, more under the A1fi than B1 scenario (Table 1). In response, relative to 1999, the U.S. surface ozone concentrations are increased by less than 0.9 ppb under A1fi and less than 0.4 ppb under B1 by 2049; the ozone increases from 1999 to 2099 are less than 1.4 ppb under A1fi and less than 0.6 ppb under B1 (not shown). Therefore projected future changes in ICT due to potential changes in

EURA biogenic emissions are not as significant as those due to potential changes in EURA anthropogenic emissions.

4. Conclusions and Discussions

[14] Through 27 model experiments conducted with a newly improved version of MOZART-2.4, this study finds that future changes in intercontinental transport may have significant consequences on summertime U.S. ozone mitigation.

[15] Projected changes in EURA anthropogenic emissions dominate the summertime ICT changes when considering IPCC SRES scenarios. From 1999 to both 2049 and 2099, projected increases in EURA anthropogenic emissions under A1fi lead to 3–8 ppb more ozone over the western U.S., with the ICT effect being about 0.5 ppb larger in 2099 than in 2049. In particular, a 2–6 ppb increase of ozone over the western U.S. is due to the Asian emissions enhancements alone, suggesting the importance of Asian emissions changes to U.S. pollution. On the other hand, relative to 1999, projected changes in EURA anthropogenic emissions under B1 lead to up to 0.7 ppb less ozone by 2049 and 1–2 ppb less ozone by 2099 over the western U.S. This reveals that the success of U.S. ozone mitigation depends on not only the country-wide emission reductions but also on what happens to emissions in Asia and Europe, especially in Asia. *Holloway et al.* [2003] similarly concluded that emissions controls in Asia may be crucial to the future of ozone mitigation in the U.S.

[16] Projected changes in EURA biogenic emissions have a secondary effect on the summertime ICT as compared to changes in EURA anthropogenic emissions. They lead to a summertime ozone increase of less than 0.9 ppb by 2049 and less than 1.4 ppb by 2099 under the A1fi scenario throughout the U.S.; the corresponding ozone increases under B1 are even smaller. Nevertheless, this amount of ozone increase is a result of the climate warming and not affected by local emission reductions. Thus, global climate change could also impact regional pollution outside of the direct effects of the local temperature changes.

[17] It is noted that the present-day ICT calculated here is (1) affected insignificantly by current model improvements, and (2) largely independent on the year of choice, i.e., 1999. Previously we have used the original MOZART-2.4 (without improvements) to conduct two experiments for 1996–2000 where EURA anthropogenic emissions are included and excluded, respectively, to simulate the climatological level of the present-day ICT. We find that the simulated summertime ICT effects on U.S. ozone for 1999 with and without the model improvements, respectively, and the ICT effects averaged over 1996–2000 are very similar, i.e., the differences among the three cases are less than 0.5 ppb in most regions (not shown). Moreover, the 5-year standard deviation of ICT effects calculated without the model improvements is also less than 0.5 ppb (less than 17% of the 5-year mean) over the U.S. Therefore our findings here are largely climatological, at least for the present-day ICT, despite of the 1-year rather than multi-year simulations. Future research should focus on better understanding the dependence of soil emissions of NO_x on climate and resulting impacts on the ICT, given their significant contribution to NO_x budget and thus ozone production.

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