

SUPPLEMENTAL MATERIAL

Net radiative forcing and air quality responses to regional CO emission reductions

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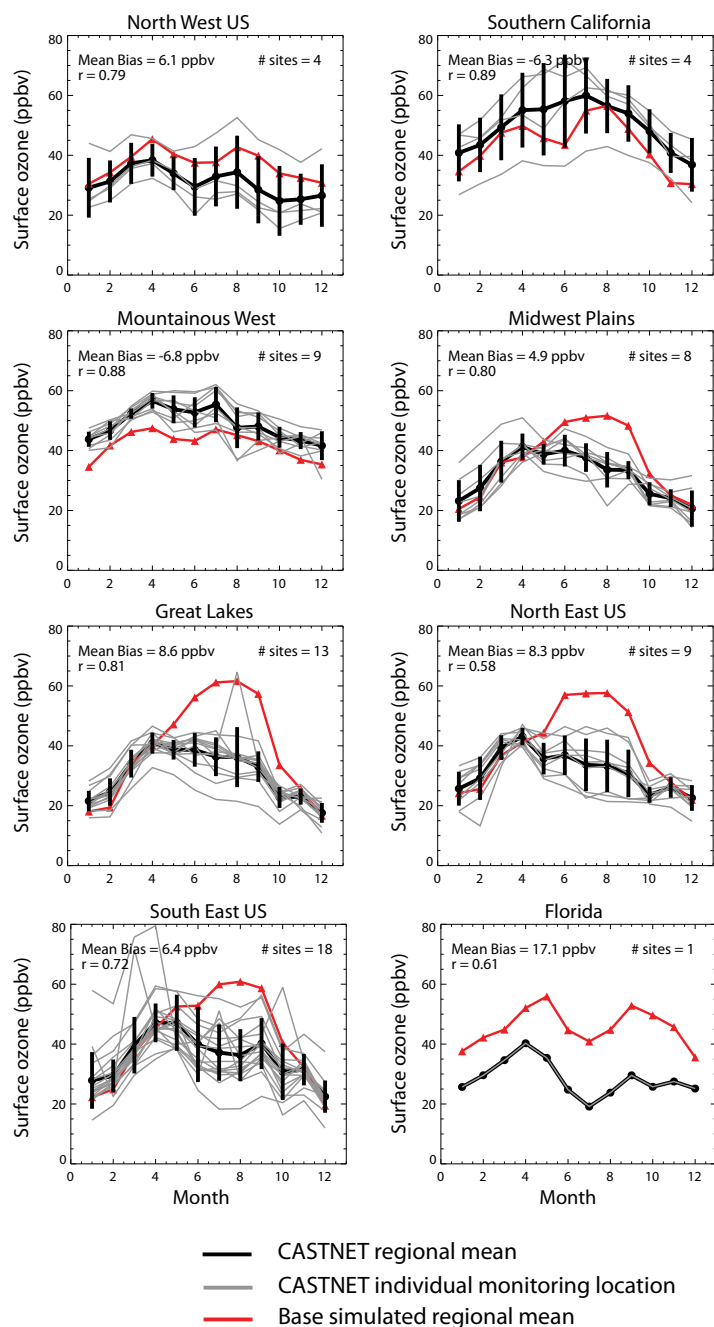


Figure S1. Comparison of the base simulated regional monthly mean short-term surface O_3 concentrations (red) for the year 2005 with 2005 observations from the CASTNET monitoring network in the U.S., showing CASTNET regional mean (black) and individual monitoring locations (grey). An overall model bias of 4.5 ppbv is calculated across all stations.

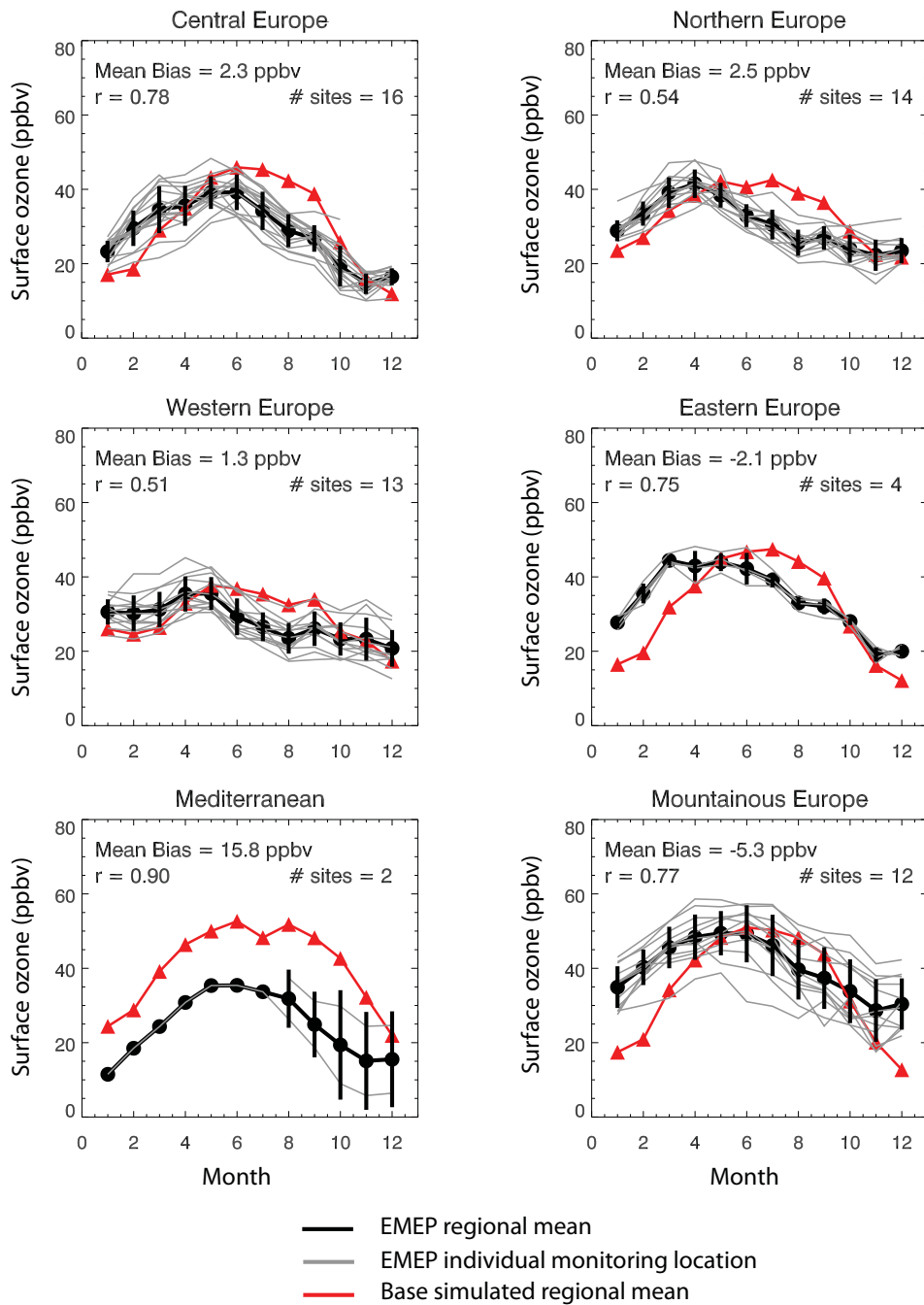
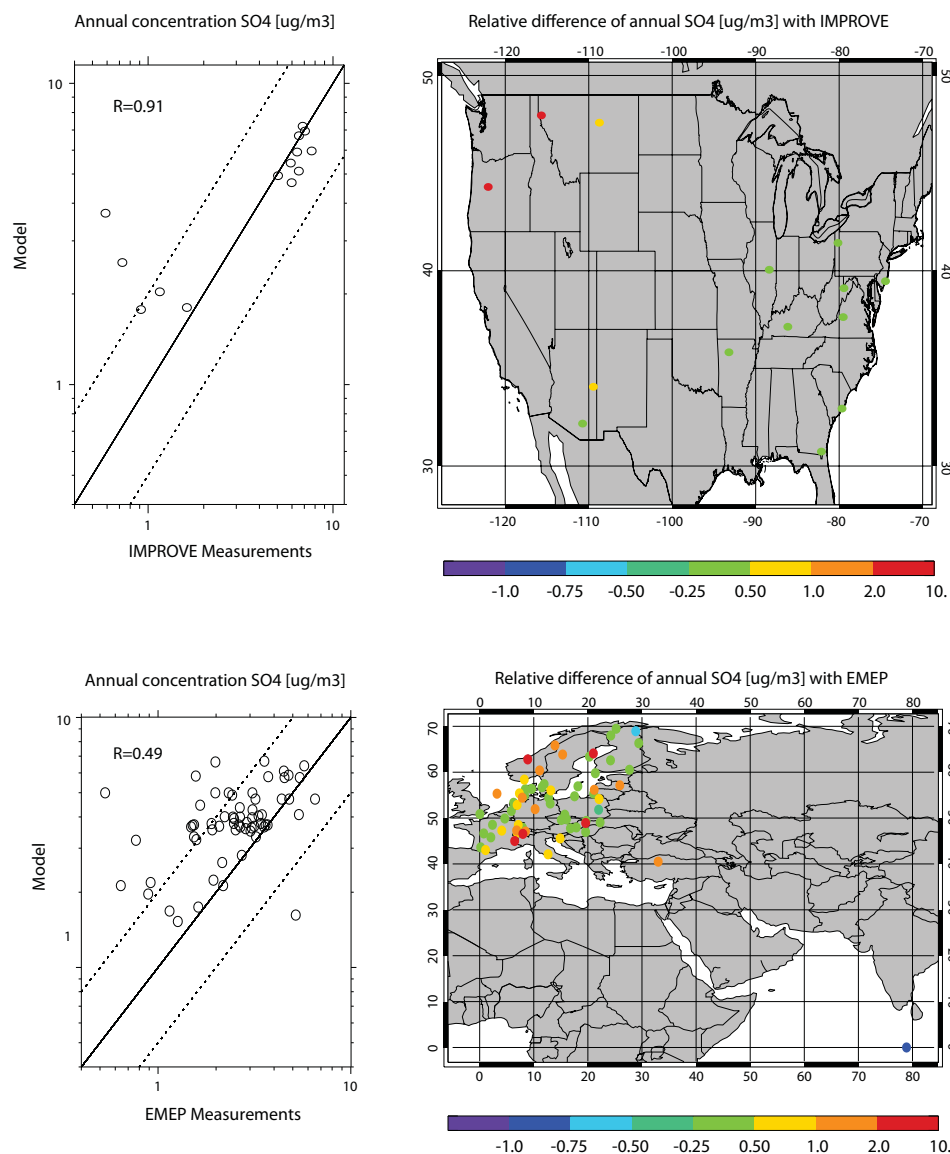


Figure S2. Comparison of the base simulated regional monthly mean short-term surface O_3 concentrations (red) for the year 2005 with 2005 observations from the EMEP monitoring network in Europe, showing EMEP regional mean (black) and individual monitoring locations (grey). An overall model bias of 0.8 ppbv is calculated across all stations.



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3 Figure S3. Comparison of the base simulated annual average (2005) surface SO₄²⁻
 4 concentrations (μg m⁻³) with annual average (2005) observations from (a) the IMPROVE
 5 surface monitoring network for remote locations in the U.S., and (b) the EMEP surface
 6 monitoring network for Europe. The left panels show modeled versus observed
 7 concentrations (μg m⁻³) with the dashed 1:2 and 2:1 lines representing agreement within a
 8 factor of 2. The right panels show a map of [(modeled-observed)/observed] values.

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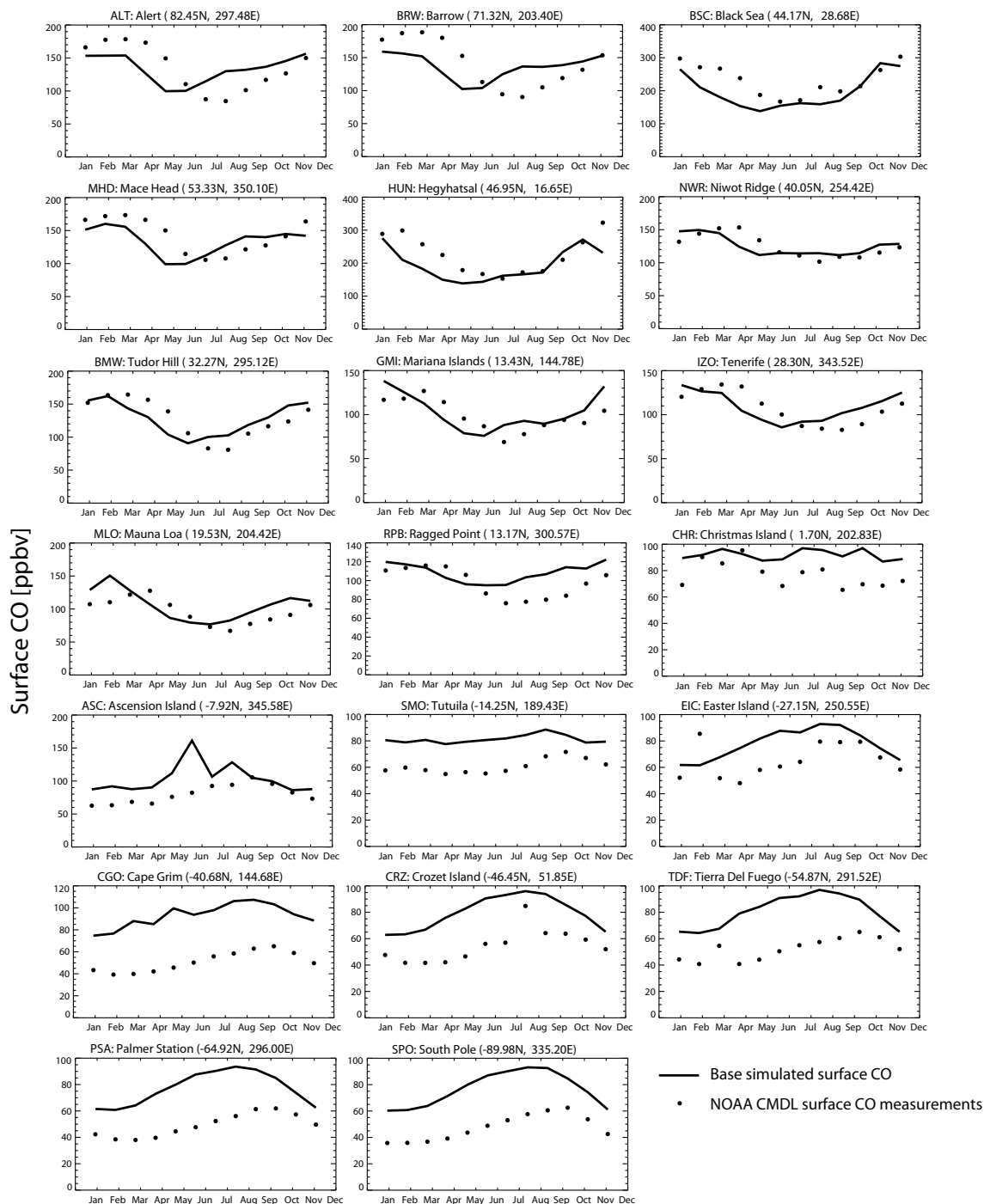


Figure S4. Comparison of the base simulated monthly mean (2005) surface CO concentrations with monthly mean NOAA CMDL surface CO measurements (in ppbv).

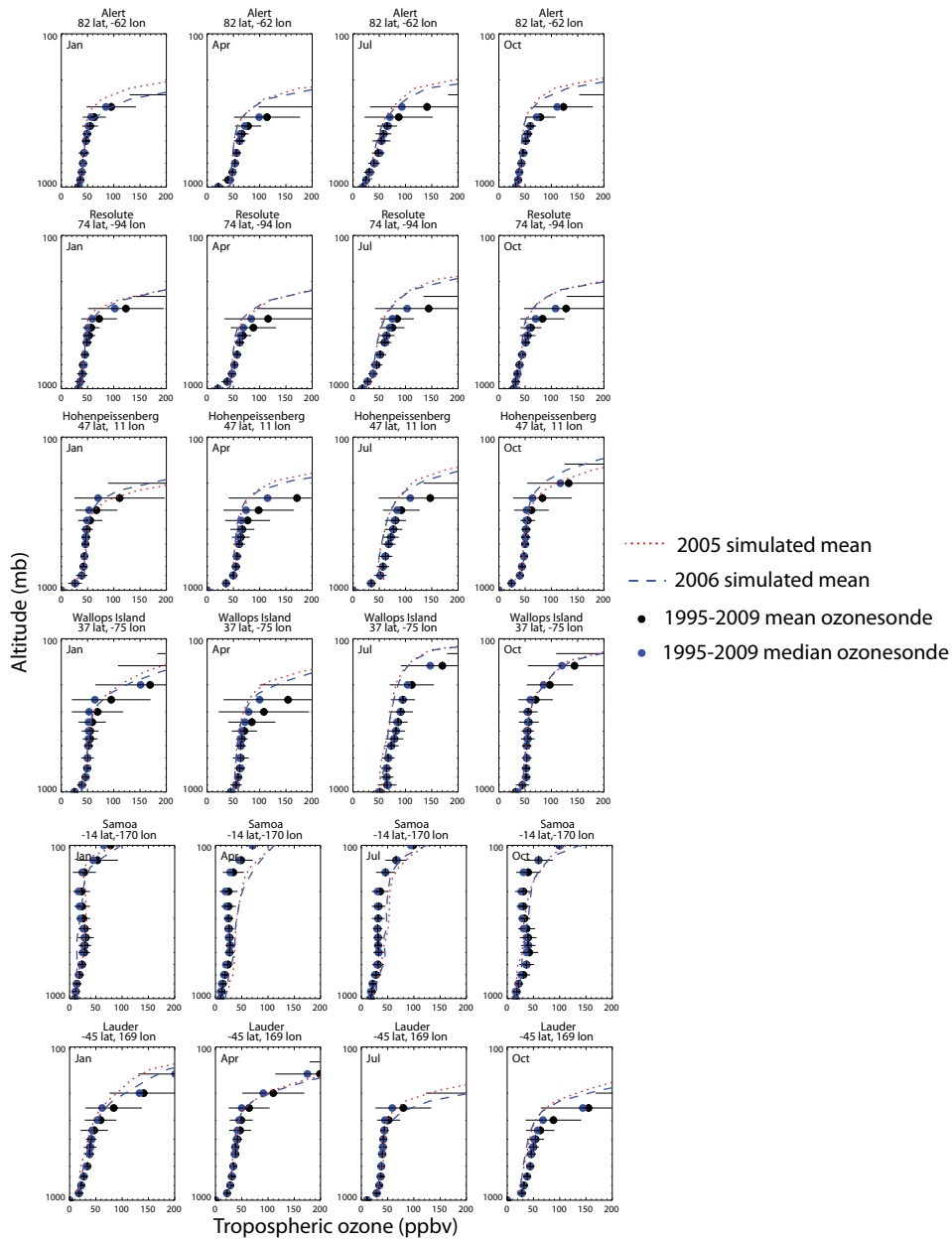


Figure S5. Vertical profile comparisons of the base simulated monthly mean tropospheric O_3 concentrations (ppbv) for the years 2005 (red line) and 2006 (blue line) with the monthly mean (black dot) and median (blue dot) ozonesonde climatology (average of 1995 through 2009) (Tilmes et al., 2011) for six selected ozonesonde stations that are latitudinally representative across the northern and southern hemispheres (Alert, Resolute, Hohenpeissenberg, Wallops Island, Samoa, and Lauder) in the months of January, April, July, and October.

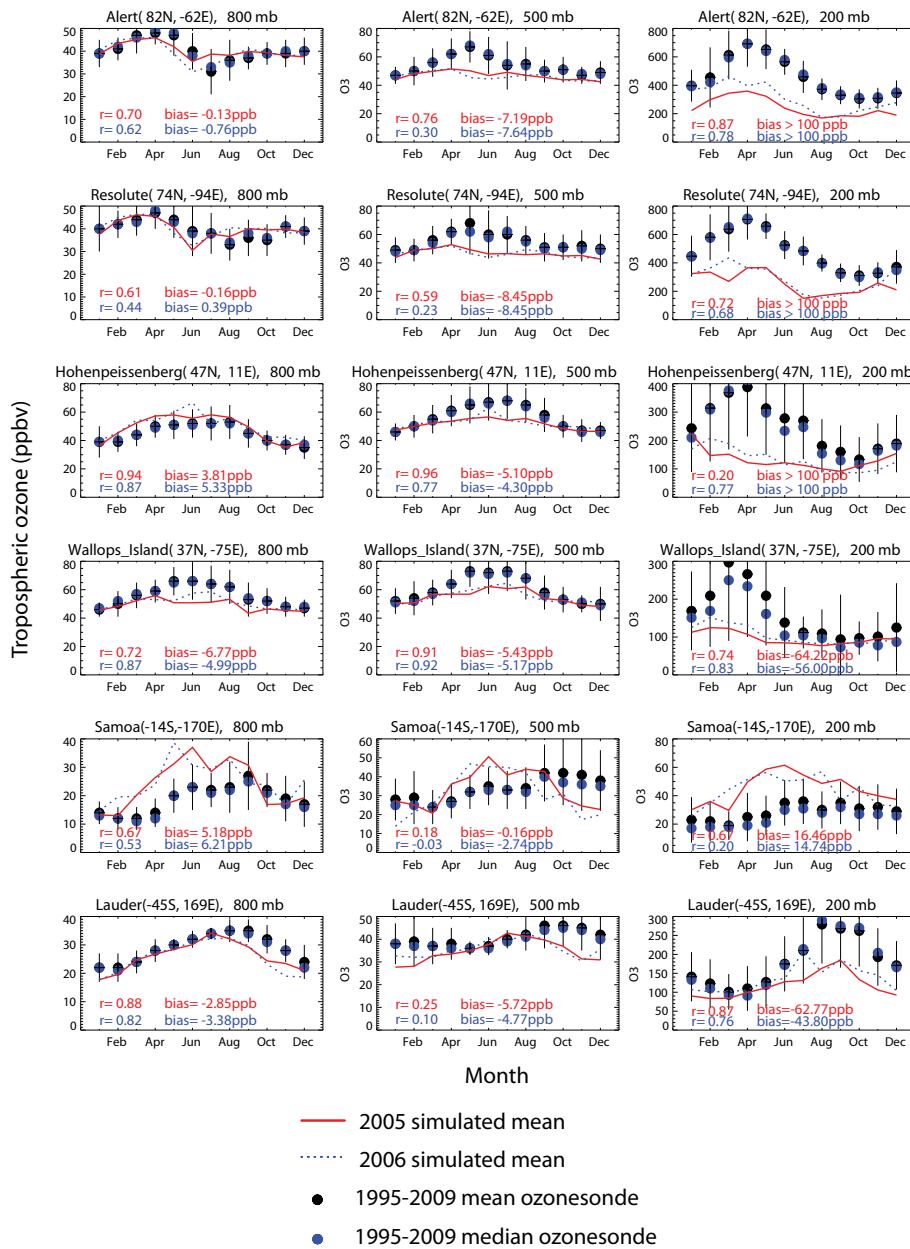
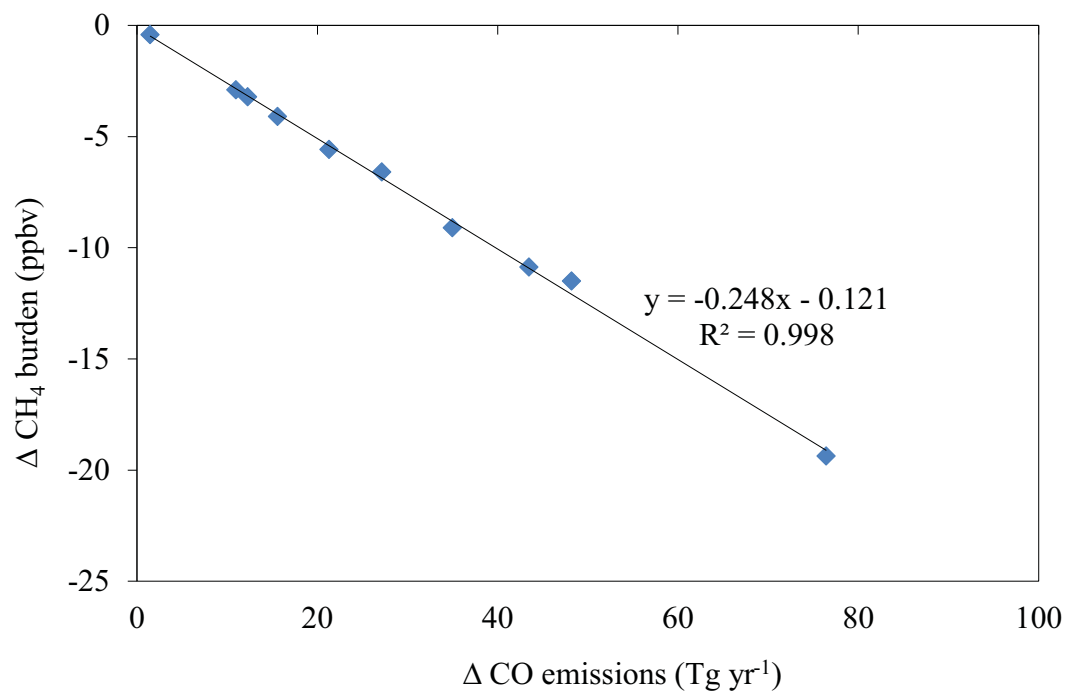


Figure S6. Time series comparisons of the base simulated monthly mean tropospheric O₃ concentrations (ppbv) for the years 2005 (red line) and 2006 (blue line) with the monthly mean (black dot) and median (blue dot) ozonesonde climatology (averaged over 1995 through 2009) (Tilmes et al., 2011) for six selected ozonesonde stations that are latitudinally representative across the northern and southern hemispheres (Alert, Resolute, Hohenpeissenberg, Wallops Island, Samoa, and Lauder) at altitudes of 800, 500, and 200 millibars (mb).

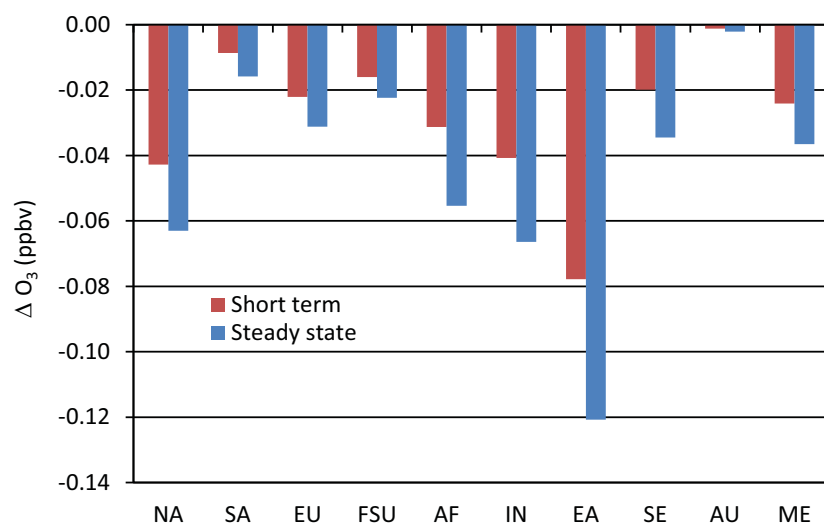


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3 Figure S7. Changes in global steady-state tropospheric CH₄ burden as a function of CO
 4 emissions change for each of the regional reductions relative to the base.

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3 Figure S8. Global annual average surface O₃ concentration changes (ppbv) for each of the
 4 regional reduction simulations, in the short term and at steady state.

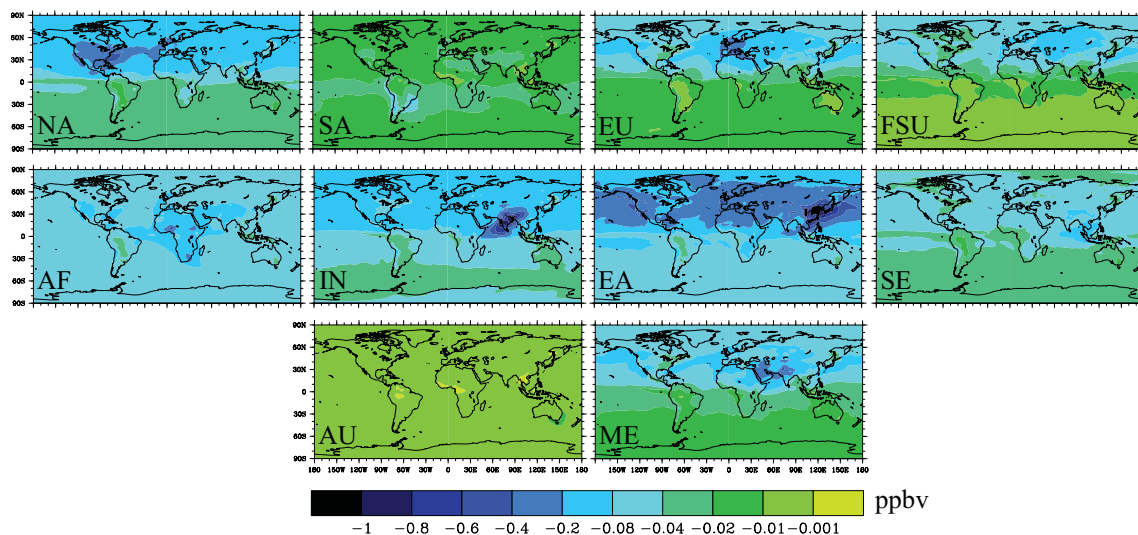
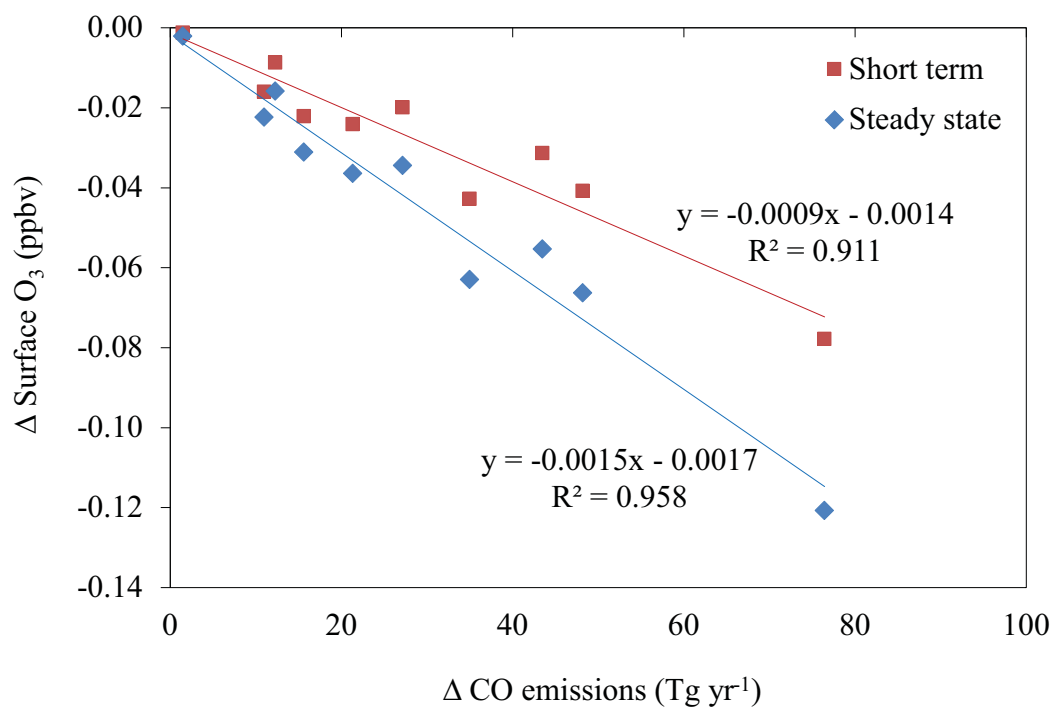


Figure S9. Changes in the three-month average of daily 8-hour maximum surface O_3 concentration changes (ppbv) relative to the base, for each of the regional reduction simulations, for the consecutive three-month period where daily 8-hour maximum surface O_3 changes are highest in each grid cell.



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3 Figure S10. Short-term and steady-state surface ozone changes as a function of CO emissions
 4 change for each of the regional reductions relative to the base.

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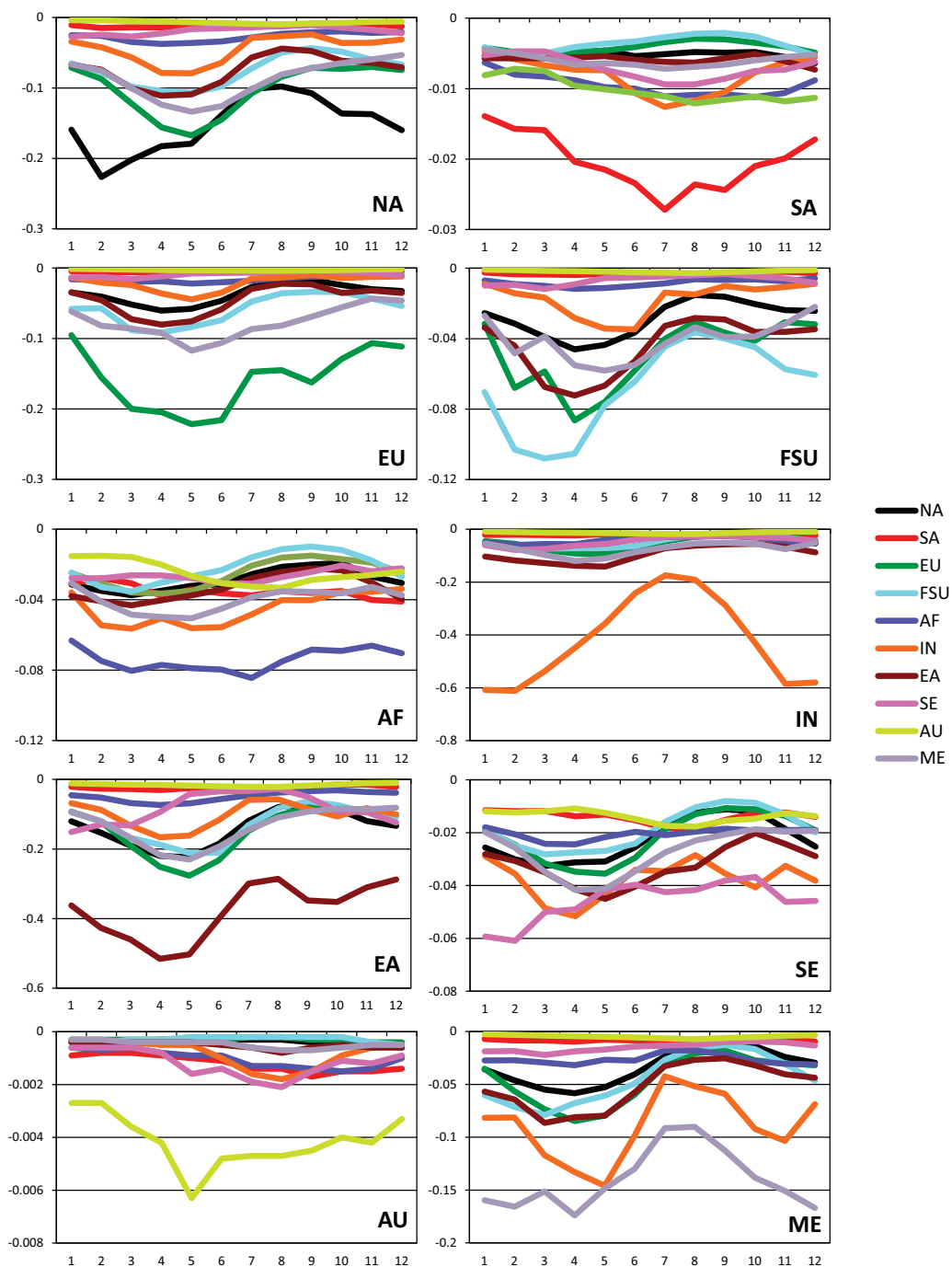


Figure S11. Changes in monthly average short-term surface O_3 (ppbv) for each of the regional reduction simulations, where the influence of each reduction region (noted in bottom right of each plot) on all 10 receptor regions (colored lines) is shown.

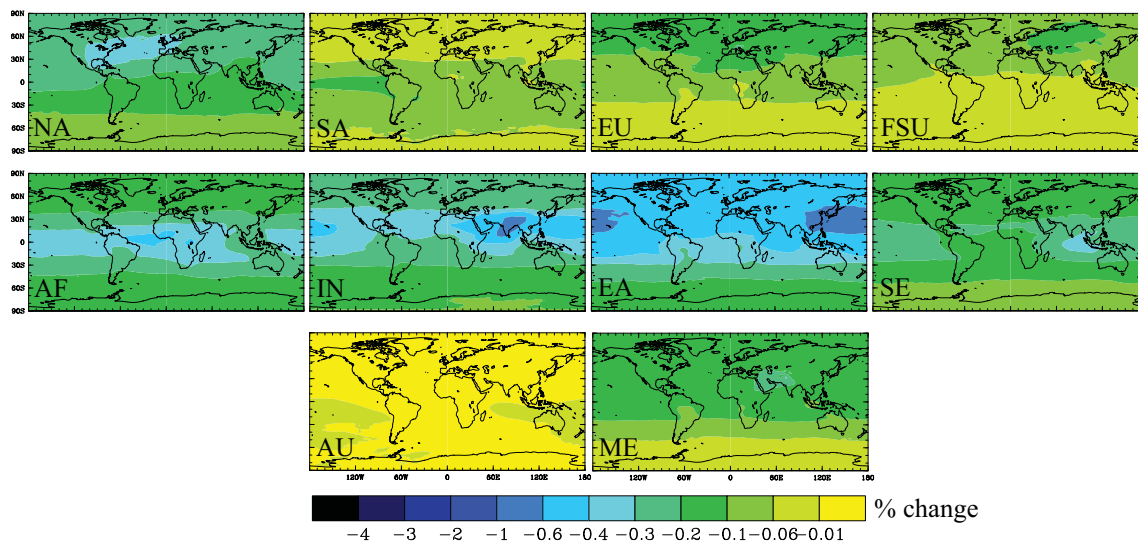
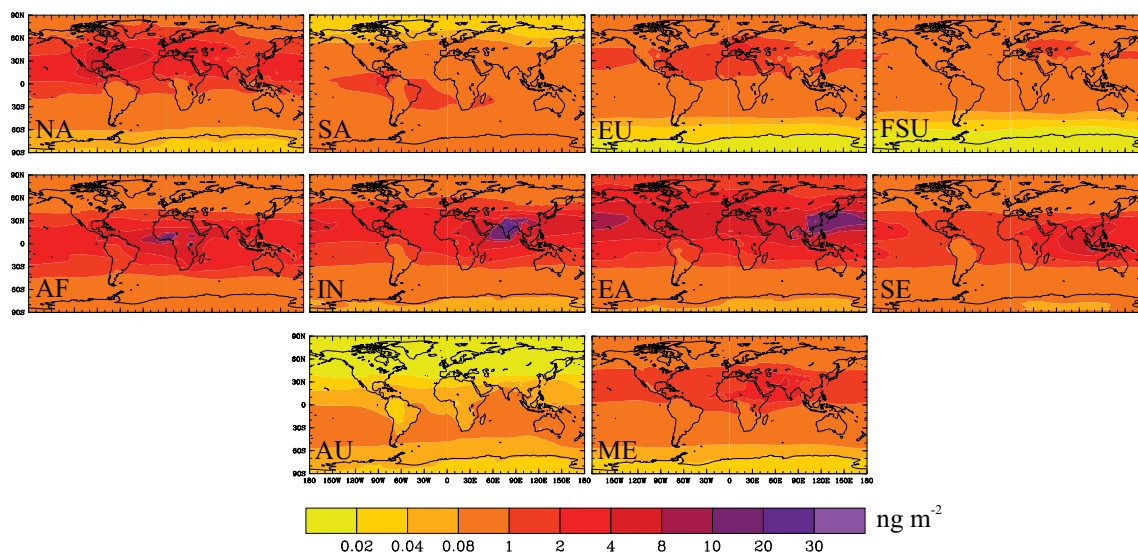


Figure S12. Global distribution of annual average percentage changes in tropospheric total column O_3 at steady state for each of the regional reduction simulations relative to the base.

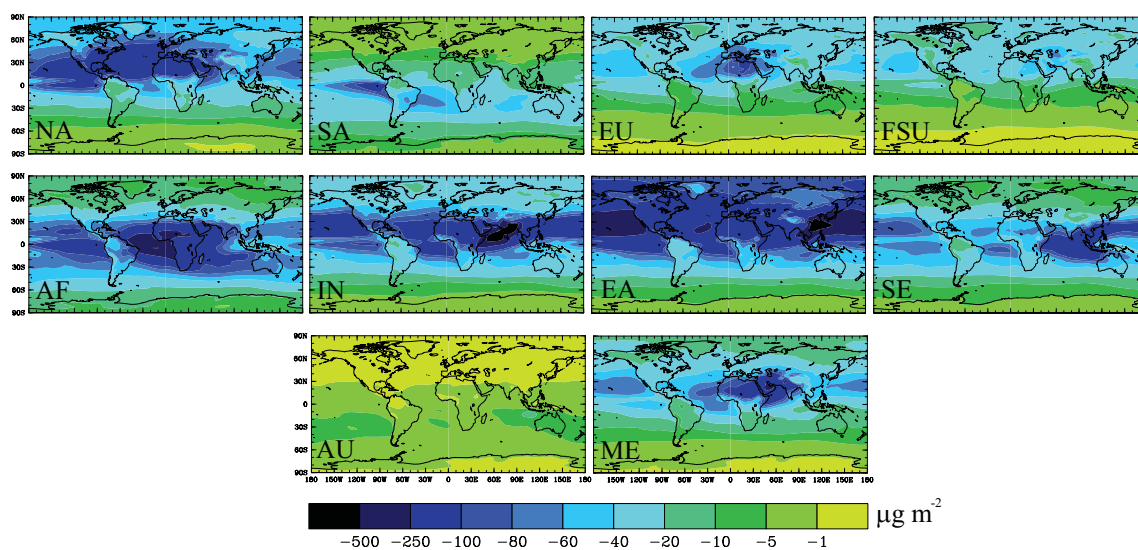


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3 Figure S13. Global distribution of annual average changes in tropospheric total column OH

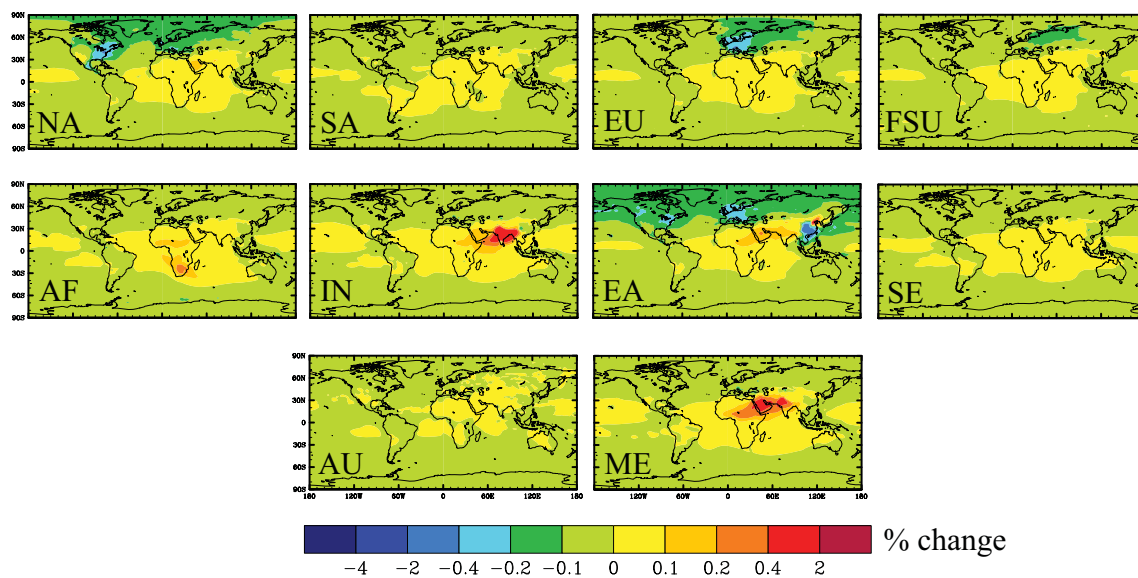
4 burden (ng m⁻²) for each of the regional reduction simulations relative to the base.



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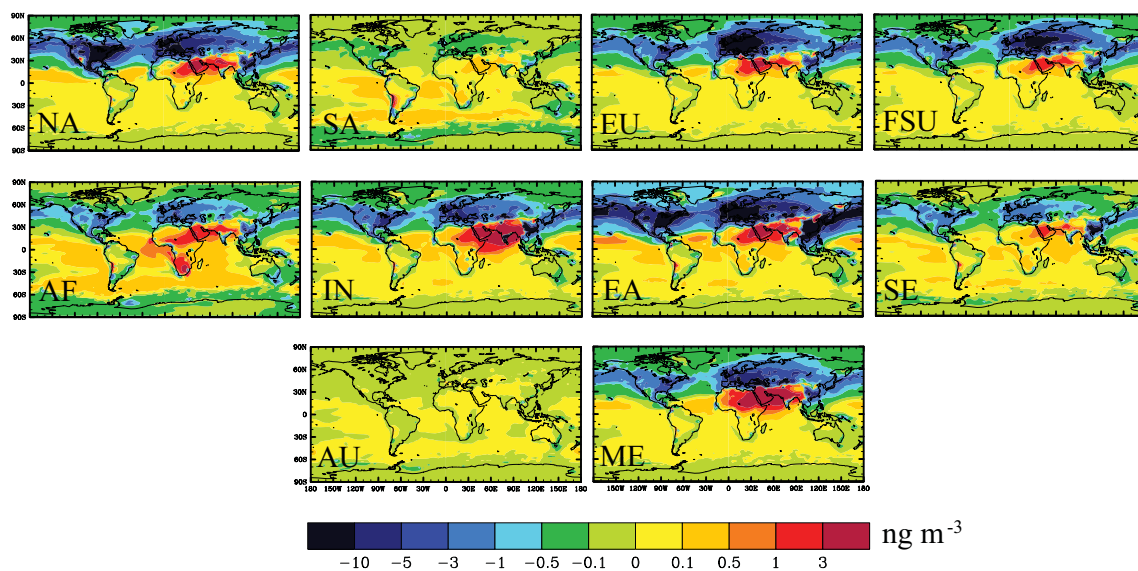
3 Figure S14. Global distribution of annual average changes in tropospheric total column H_2O_2
 4 burden ($\mu\text{g m}^{-2}$) for each of the regional reduction simulations relative to the base.



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3 Figure S15. Global distribution of annual average percentage changes in tropospheric total
 4 column SO_4^{2-} for each of the regional reduction simulations relative to the base.

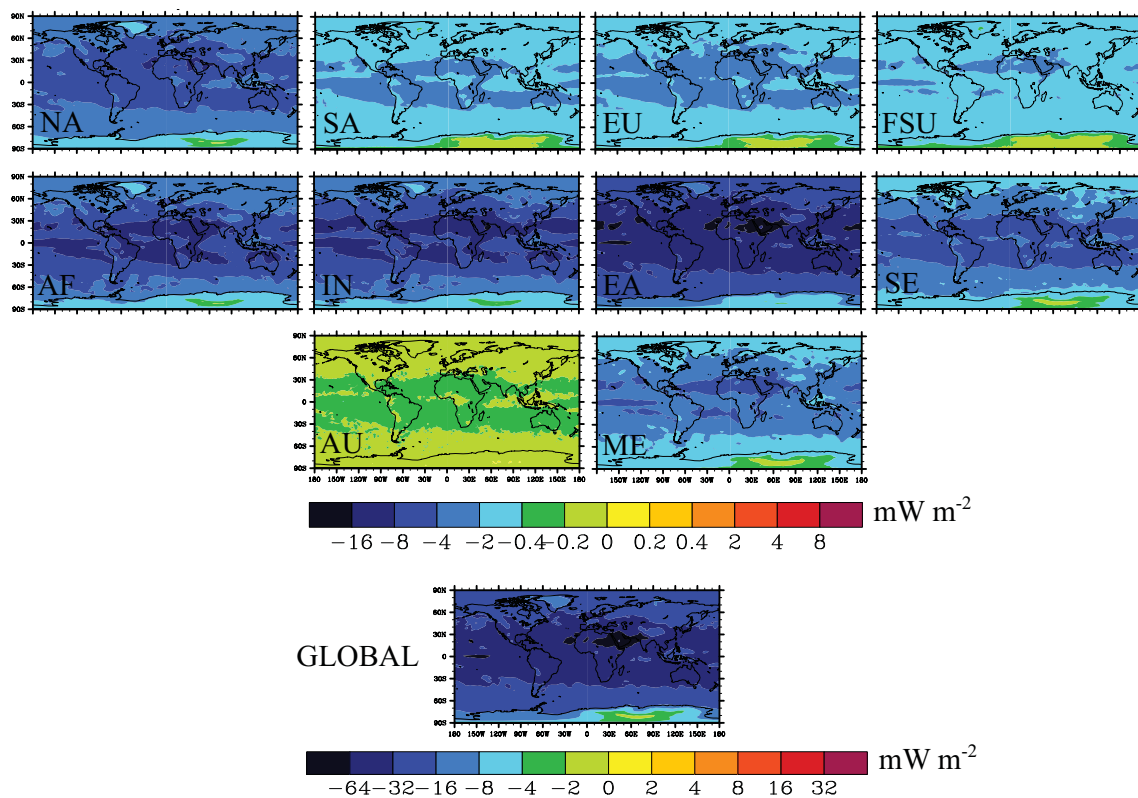


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3 Figure S16. Global distribution of annual average surface SO_4^{2-} concentration changes (ng m^{-3}) for each of the regional reduction simulations relative to the base.

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3 Figure S17. Annual average longwave (infrared) radiation distributions (mW m⁻²) due to
 4 changes in tropospheric steady-state O₃, CH₄, and SO₄²⁻ for the regional and global CO
 5 reduction simulations minus the base simulation.

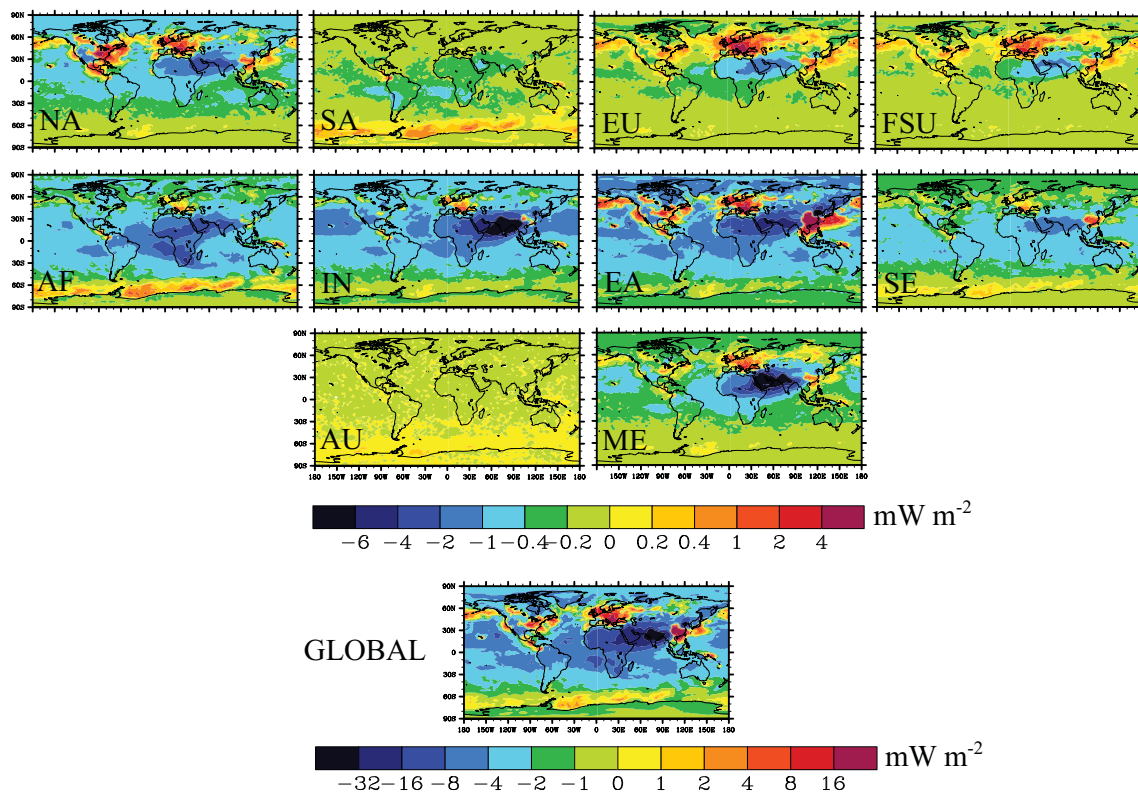
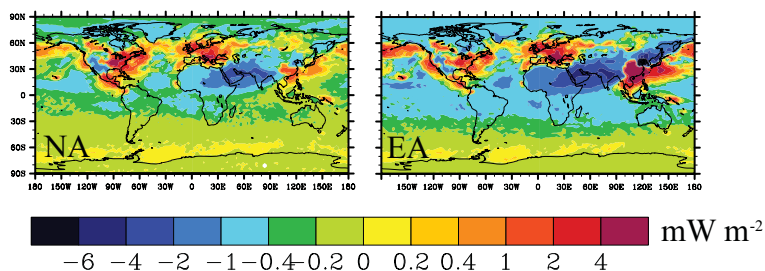
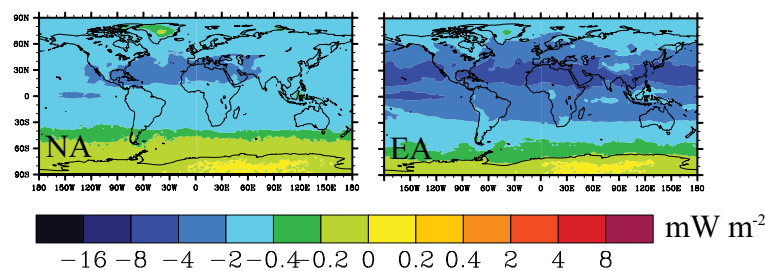


Figure S18. Annual average shortwave (solar) radiation distributions (mW m⁻²) due to changes in tropospheric steady-state O₃, CH₄, and SO₄²⁻ for the regional and global CO reduction simulations minus the base simulation.

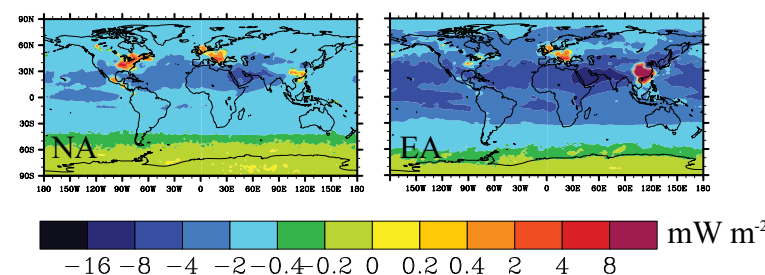
Net Solar



Net Infrared



Net RF



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3 Figure S19. Short-term annual average shortwave (solar) radiation, longwave (infrared)
 4 radiation, and net RF distributions (mW m^{-2}) due to changes in tropospheric short-term O_3
 5 and SO_4^{2-} for the NA and EA reduction simulations minus the base simulation.

6

1 Table S1. Base simulated tropospheric OH burdens (labeled OH) compared to tropospheric
2 OH burdens of Spivakovsky et al. (2000) (labeled OH-S), Lawrence et al. (1996) (labeled
3 OH-L1), Lawrence et al. (1999) (labeled OH-L2), von Kuhlmann (2001) (labeled OH-V), and
4 Emmons et al. (2010) (labeled OH-E) in $1\text{e6 OH molecules cm}^{-3}$.

Region	OH	OH-S	OH-L1	OH-L2	OH-V	OH-E
Surface-750 hPa						
90°S - 30°S	0.43	0.47	0.44	0.35	0.51	0.42
30°S - 0°	1.11	1.44	1.56	1.30	1.51	1.25
0° - 30°N	1.35	1.52	1.86	1.52	1.76	1.37
30°N - 90°N	0.81	0.76	0.86	0.79	0.86	0.77
750 - 500 hPa						
90°S - 30°S	0.46	0.72	0.56	0.36	0.46	0.53
30°S - 0°	1.15	2.00	1.65	1.20	1.48	1.39
0° - 30°N	1.41	1.99	1.94	1.37	1.61	1.59
30°N - 90°N	0.67	0.88	0.91	0.65	0.72	0.72
500 - 250 hPa						
90°S - 30°S	0.40	0.64	0.65	0.37	0.36	0.54
30°S - 0°	0.80	1.43	1.55	1.04	0.82	1.05
0° - 30°N	1.05	1.36	1.76	1.15	0.96	1.26
30°N - 90°N	0.59	0.64	0.95	0.59	0.52	0.72

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1 Table S2. Base simulated tropospheric CH₄ loss by reaction with OH (%) compared to Fiore
 2 et al. (2008).

Region	CH ₄ loss by	
	Base CH ₄ loss by OH (%)	OH (%) (Fiore et al., 2008)
Surface-750 hPa		
90°S - 30°S	4.7%	4.4%
30°S - 0°	15.5%	18.9%
0° - 30°N	19.6%	23.6%
30°N - 90°N	9.7%	9.8%
Total	49.4%	56.7%
750 - 500 hPa		
90°S - 30°S	3.7%	3.2%
30°S - 0°	12.0%	11.3%
0° - 30°N	14.6%	13.1%
30°N - 90°N	5.8%	5.3%
Total	36.1%	32.9%
500 - 250 hPa		
90°S - 30°S	1.5%	1.2%
30°S - 0°	4.5%	3.4%
0° - 30°N	6.0%	4.0%
30°N - 90°N	2.5%	1.8%
Total	14.5%	10.4%

1 Table S3. Source-receptor matrix of annual average surface CO concentration changes per
2 unit change in CO emissions (pptv CO (Tg CO yr⁻¹)⁻¹), for the regional reduction simulations,
3 with the US also defined as a receptor in addition to the 10 regions. The largest changes for
4 each source reduction region are in bold.

Source	Receptor										
	NA	SA	EU	FSU	AF	IN	EA	SE	AU	ME	US
NA	-358	-19.8	-130	-116	-38.3	-50.0	-93.6	-33.4	-9.1	-96.0	-552
SA	-15.7	-310	-10.4	-10.4	-45.4	-25.7	-14.1	-37.0	-67.9	-14.6	-12.6
EU	-149	-20.0	-1174	-318	-63.0	-54.8	-164	-40.1	-8.3	-250	-139
FSU	-187	-16.8	-335	-886	-43.1	-62.8	-270	-45.1	-7.7	-185	-172
AF	-24.3	-56.9	-20.0	-20.0	-235	-42.4	-25.5	-33.9	-36.2	-32.6	-21.8
IN	-40.0	-21.3	-37.8	-41.4	-45.9	-1567	-87.8	-72.7	-11.9	-57.3	-40.9
EA	-92.6	-16.8	-85.4	-100	-30.8	-62.6	-654	-109	-9.5	-66.4	-98.3
SE	-30.1	-26.4	-27.3	-26.9	-33.2	-69.9	-54.4	-330	-27.0	-30.5	-30.6
AU	-8.3	-74.2	-6.3	-6.3	-47.4	-24.6	-10.2	-70.1	-551	-10.3	-7.4
ME	-77.6	-23.0	-109	-139	-79.3	-277	-118	-44.0	-10.3	-642	-77.9

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1 Table S4. Source-receptor matrix of annual average steady-state changes in surface O₃
2 concentrations per unit change in CO emissions (pptv O₃ (Tg CO yr⁻¹)⁻¹), for the regional
3 reduction simulations, with the United States (US) also defined as a receptor in addition to the
4 10 regions. The largest changes for each source reduction region are in bold.

Source	Receptor										
	NA	SA	EU	FSU	AF	IN	EA	SE	AU	ME	US
NA	-5.09	-0.74	-3.75	-2.78	-1.34	-2.04	-2.93	-1.00	-0.59	-3.42	-7.24
SA	-1.17	-2.06	-1.16	-0.97	-1.34	-1.43	-1.27	-1.05	-1.24	-1.39	-1.26
EU	-3.10	-0.72	-11.0	-4.42	-1.58	-2.11	-3.71	-1.05	-0.58	-5.87	-3.59
FSU	-3.37	-0.69	-5.32	-6.86	-1.32	-2.36	-4.87	-1.10	-0.58	-4.65	-3.97
AF	-1.38	-1.20	-1.37	-1.15	-2.24	-1.78	-1.53	-1.06	-0.95	-1.79	-1.47
IN	-1.76	-0.78	-1.85	-1.59	-1.39	-9.43	-2.68	-1.39	-0.64	-2.38	-1.99
EA	-2.58	-0.70	-2.74	-2.33	-1.18	-2.07	-5.73	-1.56	-0.60	-2.64	-3.08
SE	-1.53	-0.89	-1.57	-1.31	-1.27	-2.10	-1.93	-2.13	-0.88	-1.84	-1.71
AU	-1.09	-1.30	-1.09	-0.96	-1.30	-1.43	-1.23	-1.36	-3.28	-1.36	-1.16
ME	-2.33	-0.80	-2.98	-2.77	-1.80	-4.97	-3.24	-1.17	-0.63	-7.48	-2.70

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1 Table S5. For each regional reduction, changes in global annual average, steady-state full and
 2 upper tropospheric (UT) O₃ burden (B_{O₃}) per unit change in CO emissions (E_{CO}) (Tg O₃/Tg
 3 CO).

	$\Delta \text{Full B}_{\text{O}_3} /$ $\Delta \text{E}_{\text{CO}}$	$\Delta \text{UT B}_{\text{O}_3} /$ $\Delta \text{E}_{\text{CO}}$
NA	0.0182	1.05E-02
SA	0.0174	1.10E-02
EU	0.0172	9.43E-03
FSU	0.0174	9.68E-03
AF	0.0184	1.18E-02
IN	0.0194	1.27E-02
EA	0.0180	1.10E-02
SE	0.0205	1.38E-02
AU	0.0171	9.55E-03
ME	0.0180	1.07E-02

1 Table S6. Source-receptor matrix of annual average surface SO_4^{2-} concentration changes (ng m^{-3}), for the regional reduction simulations, with
 2 the United States (US) also defined as a receptor in addition to the 10 regions.

Source	Receptor										
	NA	SA	EU	FSU	AF	IN	EA	SE	AU	ME	US
NA	-8.82	-0.11	-9.10	-2.83	0.16	1.01	-3.10	-0.37	-0.04	-0.18	-15.59
SA	-0.16	-0.06	-0.25	-0.09	0.01	-0.05	-0.19	-0.11	-0.14	0.06	-0.20
EU	-1.61	-0.04	-19.21	-4.26	0.17	0.53	-2.22	-0.22	-0.02	-0.43	-2.58
FSU	-1.29	-0.02	-4.70	-4.67	0.10	0.45	-2.07	-0.18	-0.01	-0.28	-2.08
AF	-0.96	-0.15	-2.15	-0.77	0.71	0.36	-1.18	-0.32	-0.24	0.32	-1.47
IN	-1.92	-0.11	-4.34	-1.88	0.39	21.75	-3.90	0.55	-0.07	0.82	-3.30
EA	-6.65	-0.10	-13.42	-4.96	0.28	1.17	-41.29	-3.70	-0.08	-0.15	-11.23
SE	-0.85	-0.05	-1.60	-0.56	0.08	0.01	-2.98	-0.81	-0.07	0.11	-1.44
AU	-0.01	-0.01	-0.01	-0.01	0.00	-0.01	-0.02	-0.02	-0.02	0.01	-0.01
ME	-1.40	-0.05	-4.91	-3.05	1.10	6.73	-2.39	-0.18	-0.03	5.98	-2.36

3

- 1 Table S7. Source-receptor matrix of percentage changes in surface CO concentrations, for each of the regional reduction simulations relative
2 to the base. The largest percentage changes for each source reduction region are in bold.

Source	Receptor									3
	NA	SA	EU	FSU	AF	IN	EA	SE	AU	ME ⁴
NA	-8.29%	-0.47%	-2.75%	-2.45%	-0.77%	-0.63%	-1.39%	-0.67%	-0.31%	-2.28%
SA	-0.13%	-2.58%	-0.08%	-0.08%	-0.32%	-0.11%	-0.07%	-0.26%	-0.82%	-0.12%
EU	-1.53%	-0.21%	11.03%	-2.98%	-0.57%	-0.31%	-1.09%	-0.36%	-0.13%	-2.64%
FSU	-1.36%	-0.12%	-2.21%	-5.83%	-0.27%	-0.25%	-1.26%	-0.28%	-0.08%	-1.38%
AF	-0.70%	-1.68%	-0.52%	-0.52%	-5.91%	-0.66%	-0.47%	-0.84%	-1.55%	-0.96%
IN	-1.28%	-0.70%	-1.10%	-1.20%	-1.28%	27.02%	-1.80%	-2.01%	-0.56%	-1.87%
EA	-4.69%	-0.87%	-3.93%	-4.62%	-1.36%	-1.71%	21.26%	-4.79%	-0.72%	-3.44%
SE	-0.54%	-0.49%	-0.45%	-0.44%	-0.52%	-0.68%	-0.63%	-5.14%	-0.72%	-0.56%
AU	-0.01%	-0.07%	-0.01%	-0.01%	-0.04%	-0.01%	-0.01%	-0.06%	-0.79%	-0.01%
ME	-1.10%	-0.33%	-1.40%	-1.78%	-0.98%	-2.11%	-1.07%	-0.54%	-0.21%	-9.28%

- 1 Table S8. Source-receptor matrix of percentage changes in steady-state surface O₃ concentrations, for each of the regional reduction
- 2 simulations relative to the base. The largest percentage changes for each source reduction region are in bold.

Source	Receptor									
	NA	SA	EU	FSU	AF	IN	EA	SE	AU	ME
NA	-0.50%	-0.10%	-0.36%	-0.29%	-0.16%	-0.17%	-0.24%	-0.12%	-0.09%	-0.30%
SA	-0.04%	-0.10%	-0.04%	-0.04%	-0.05%	-0.04%	-0.04%	-0.05%	-0.07%	-0.04%
EU	-0.14%	-0.05%	-0.47%	-0.21%	-0.08%	-0.08%	-0.14%	-0.06%	-0.04%	-0.23%
FSU	-0.10%	-0.03%	-0.16%	-0.23%	-0.05%	-0.06%	-0.13%	-0.04%	-0.03%	-0.13%
AF	-0.17%	-0.21%	-0.16%	-0.15%	-0.32%	-0.19%	-0.16%	-0.16%	-0.18%	-0.19%
IN	-0.24%	-0.15%	-0.24%	-0.23%	-0.22%	-1.10%	-0.30%	-0.24%	-0.13%	-0.28%
EA	-0.56%	-0.22%	-0.57%	-0.54%	-0.30%	-0.38%	-1.03%	-0.42%	-0.20%	-0.50%
SE	-0.12%	-0.10%	-0.12%	-0.11%	-0.11%	-0.14%	-0.12%	-0.21%	-0.11%	-0.12%
AU	0.00%	-0.01%	0.00%	0.00%	-0.01%	-0.01%	0.00%	-0.01%	-0.02%	0.00%
ME	-0.14%	-0.07%	-0.17%	-0.18%	-0.13%	-0.26%	-0.16%	-0.09%	-0.06%	-0.39%

1 Table S9. For each regional reduction, changes in global annual average steady-state tropospheric CO production (P_{CO}), loss (L_{CO}), burden
2 (B_{CO}), and B_{CO} per unit change in CO emissions (E_{CO}). Global annual average changes in steady-state tropospheric O₃ burden (B_{O_3}), B_{O_3} per
3 unit change in E_{CO} , and steady-state tropospheric CH₄ per unit change in E_{CO} are also shown. Steady-state CO and O₃ changes represent the
4 sum of short and long-term changes, where long-term changes are calculated by scaling ΔP_{CO} , ΔL_{CO} , ΔB_{CO} , and ΔB_{O_3} from the CH₄ control
5 simulation to the ratio of global CH₄ burden changes from each scenario divided by that from the CH₄ control simulation.

Reduction region	ΔP_{CO} (Tg CO)	ΔL_{CO} (Tg CO)	ΔB_{CO} (Tg CO)	$\Delta B_{CO} / \Delta E_{CO}$ (days)	ΔB_{O_3} (Tg O ₃)	$\Delta B_{O_3} / \Delta E_{CO}$ (days)	$\Delta CH_4 / \Delta E_{CO}$ (pptv (Tg CO yr ⁻¹) ⁻¹)
NA	1.12	-27.1	-7.56	78.9	-0.637	6.64	260
SA	0.39	-9.13	-2.78	82.6	-0.214	6.37	262
EU	0.49	-12.0	-3.76	88.0	-0.269	6.29	263
FSU	0.34	-8.45	-2.77	92.4	-0.191	6.37	265
AF	1.42	-33.1	-8.86	74.4	-0.800	6.72	250
IN	1.48	-35.9	-9.02	68.3	-0.934	7.08	239
EA	2.41	-57.7	-16.4	78.4	-1.38	6.58	253
SE	0.85	-20.4	-5.53	74.4	-0.557	7.49	243
AU	0.04	-1.09	-0.38	93.7	-0.024	6.08	289

ME	0.67	-16.6	-4.48	76.8	-0.384	6.58	262
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1 Table S10. Comparison of GWP₂₀ and GWP₁₀₀ estimates, due to regional changes in CO
2 emissions, to those in Fry et al. (2012) and Berntsen et al. (2005), where the region definitions
3 differ slightly in the three studies.

Source Region	Current study		Fry et al. (2012)		Berntsen et al. (2005)	
	GWP ₂₀	GWP ₁₀₀	GWP ₂₀	GWP ₁₀₀	GWP ₂₀	GWP ₁₀₀
North America	3.94	1.30	5.21	1.70	--	--
Europe	3.67	1.23	4.60	1.50	10.5	3.8
East Asia	3.87	1.27	5.07	1.64	--	--
South Asia	4.31	1.38	5.35	1.70	13	4.4

4