

Fig. S1: Latitude–pressure cross section of mean ozone concentration (in ppb) obtained from HIPPO aircraft measurements (first row), control run (second row), and reanalysis (third row). The relative difference (in %) between the control run and the observation (fourth row) and between the reanalysis and the observation (fifth row) is also shown. Results are shown for all HIPPO campaigns (from left to right: HIPPO I, 8–30 January 2009; HIPPO II, 31 October to 22 November 2009; HIPPO III, 24 March to 16 April 2010; HIPPO IV, 14 June to 11 July 2011; and HIPPO V, 9 August to 9 September 2011).

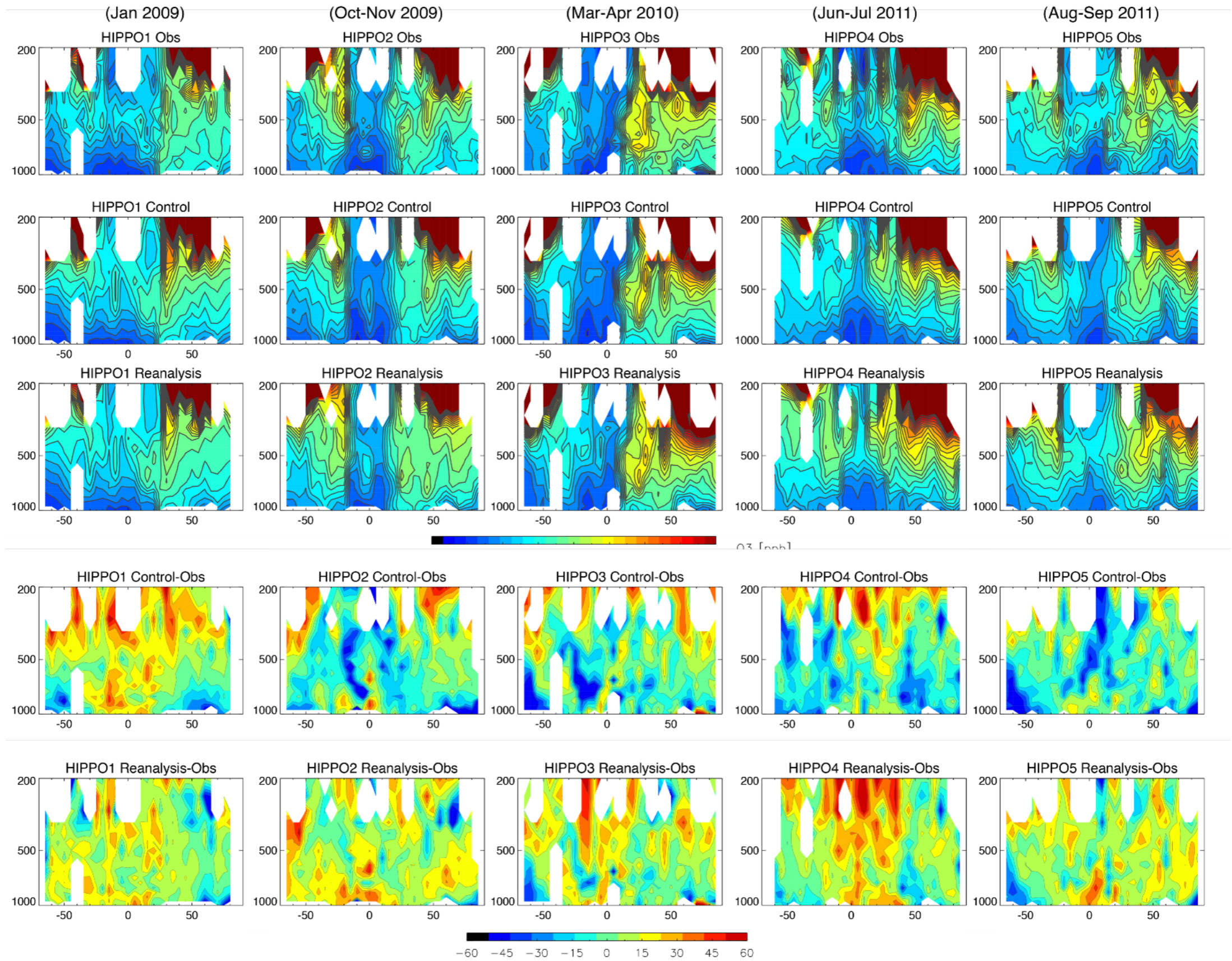


Fig. S2: Same as in Fig. S1, but for CO concentration (in ppb, from first to third row) and its absolute difference (in ppb, from fourth to fifth row) obtained from HIPPO aircraft measurements.

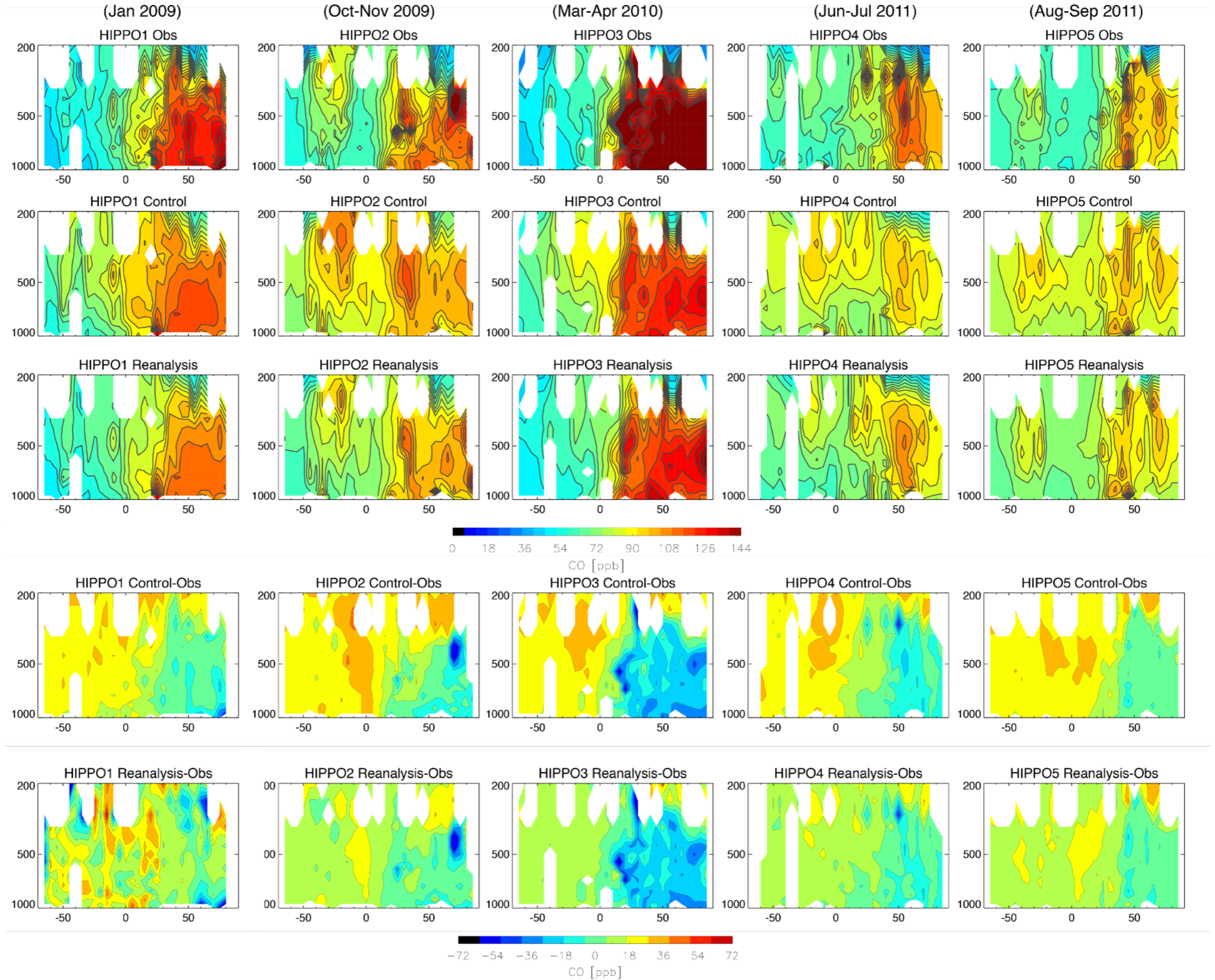


Fig. S3: Anomaly of annual mean tropospheric OH concentration from the 14-year mean (2005-2018) in TCR-2.

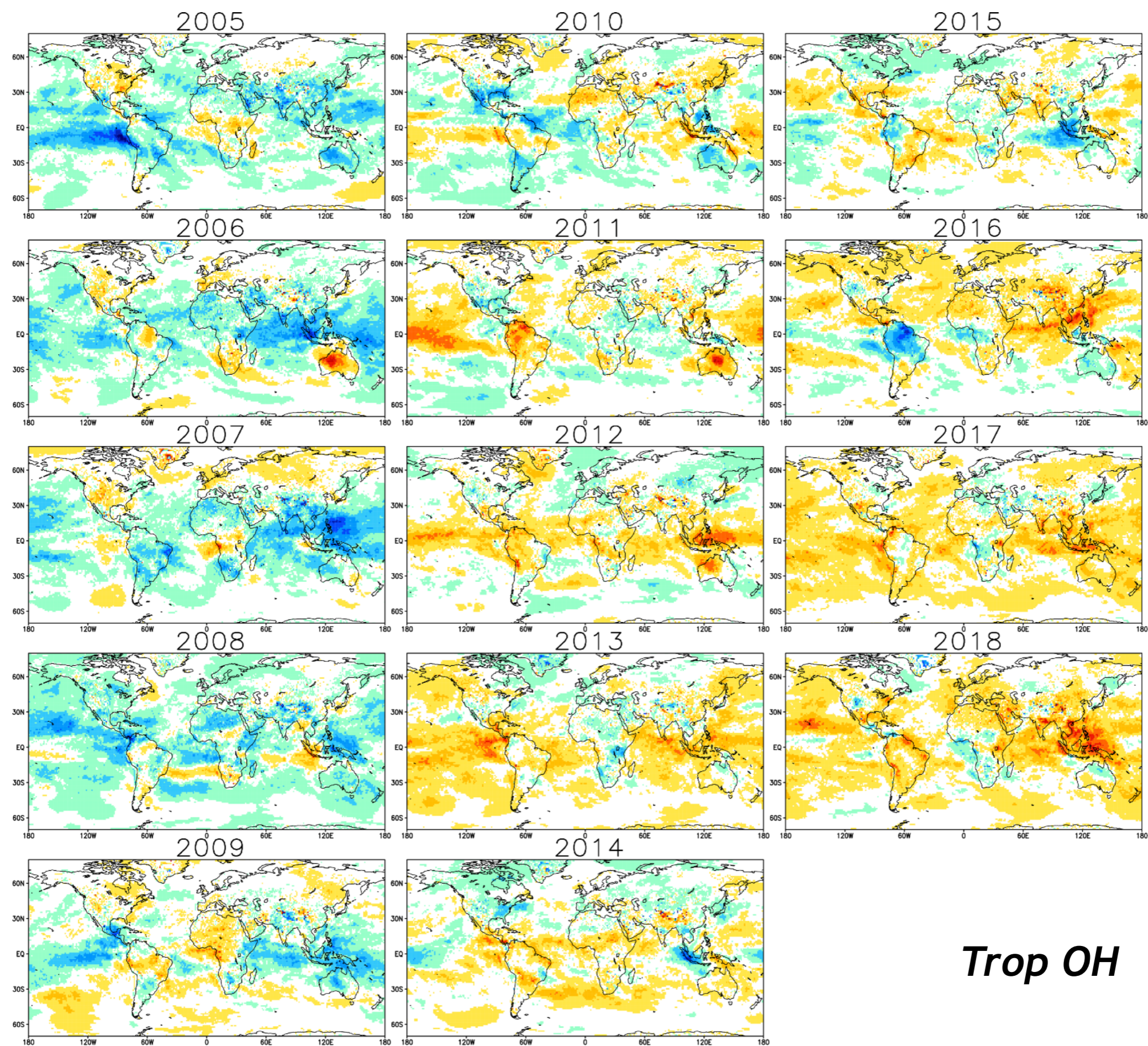


Fig. S4: Same as in Fig S3, but for the noTES reanalysis.

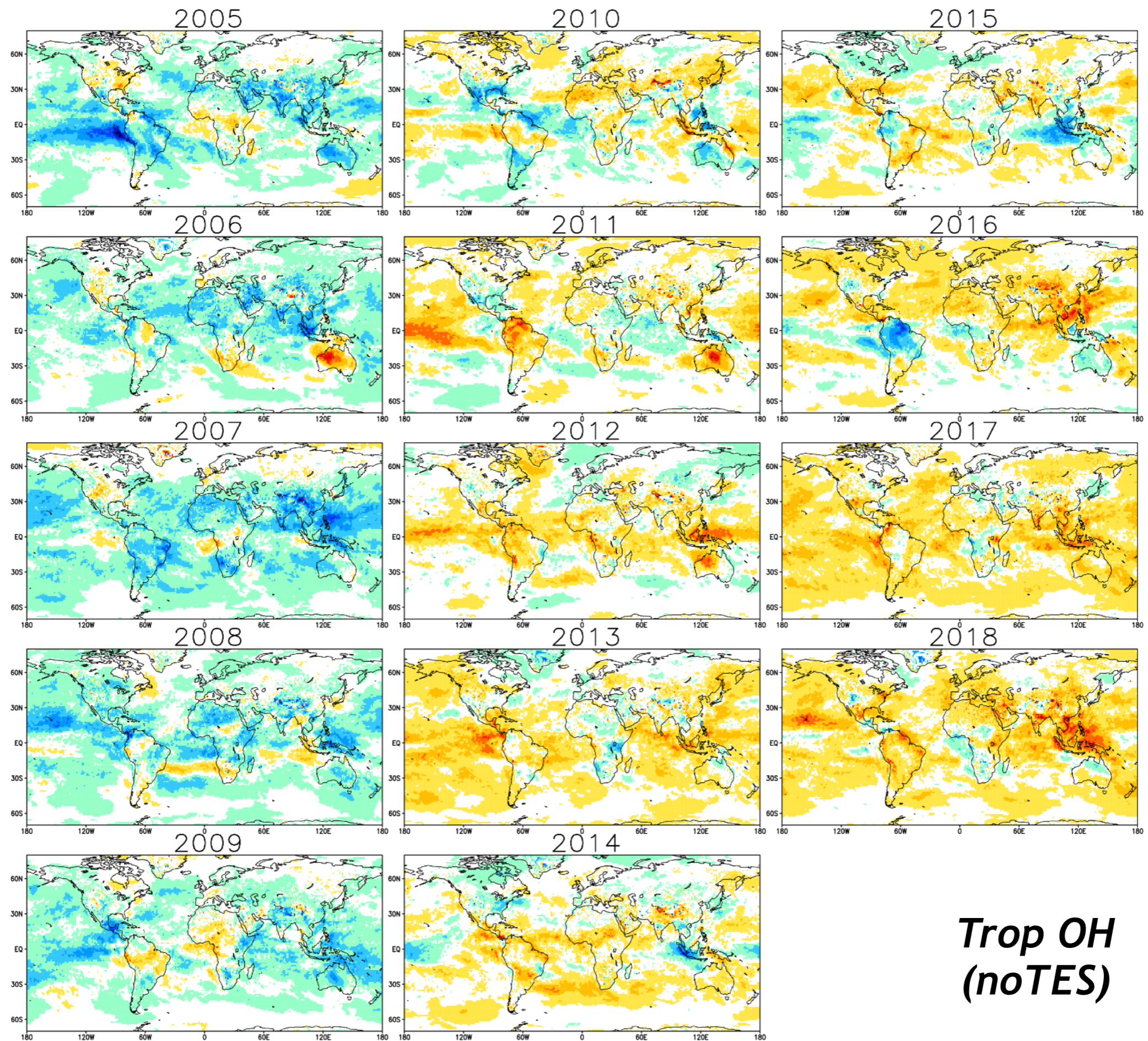


Fig. S5: Vertical profiles of OH concentrations (pptv) from ATom measurements (black), control run (blue), and chemical reanalysis from ATom-1 in August 2016 and ATom-2 in February 2017.

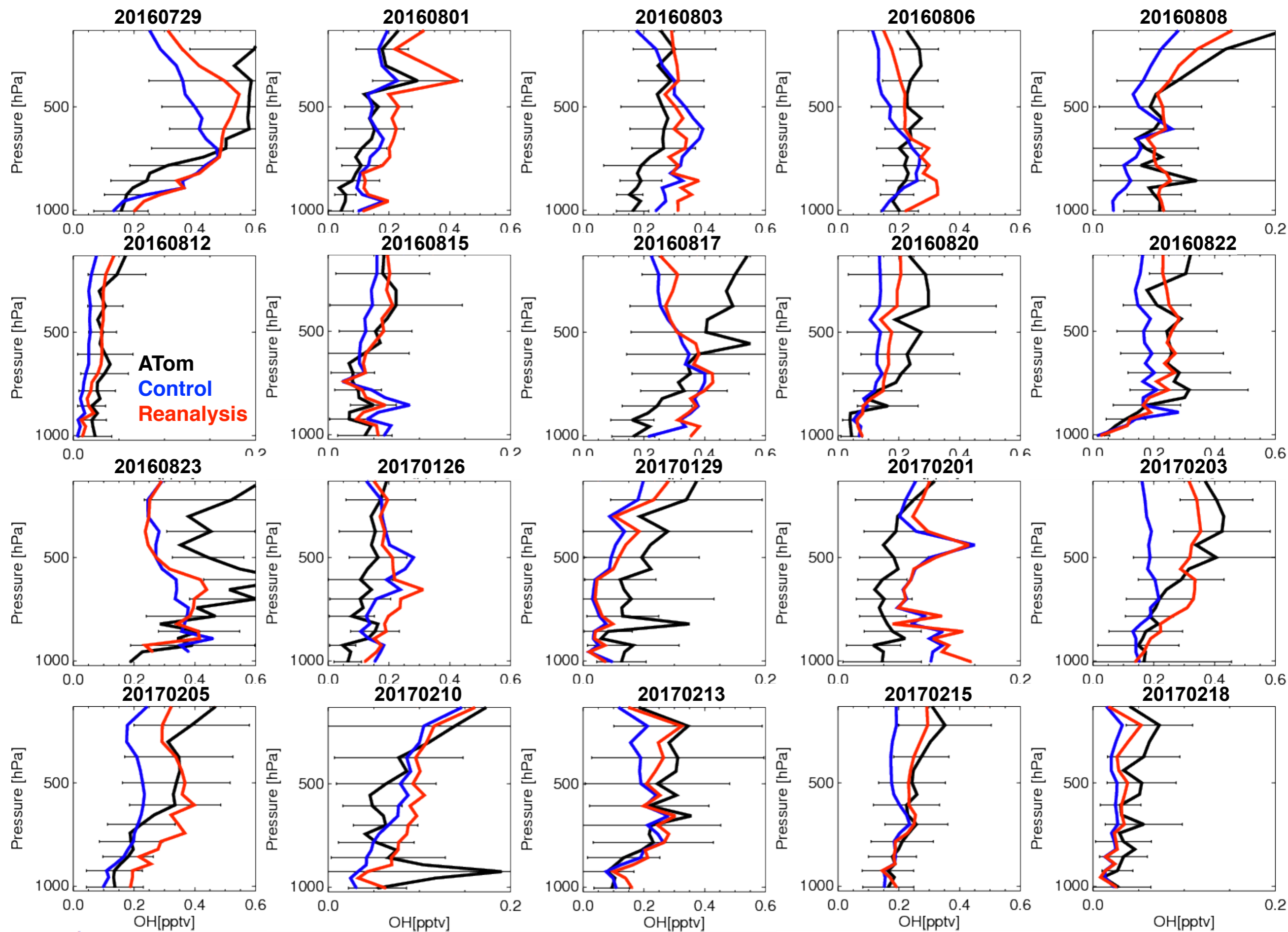


Fig. S6: Anomaly of annual mean surface NO_x emissions (in 10^{-11} kg N m⁻² s⁻¹) from the 14-year mean (2005-2018) in TCR-2.

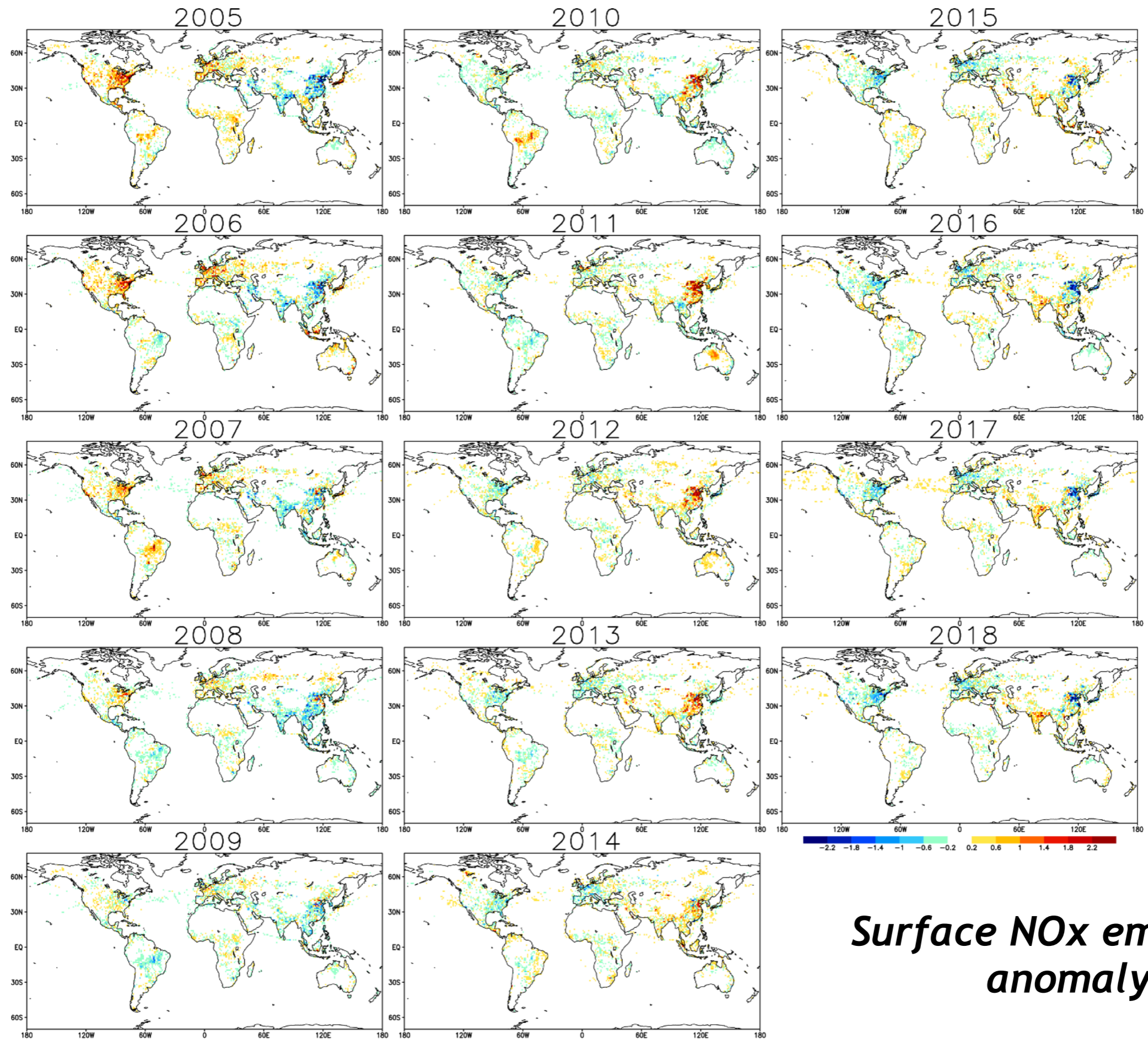
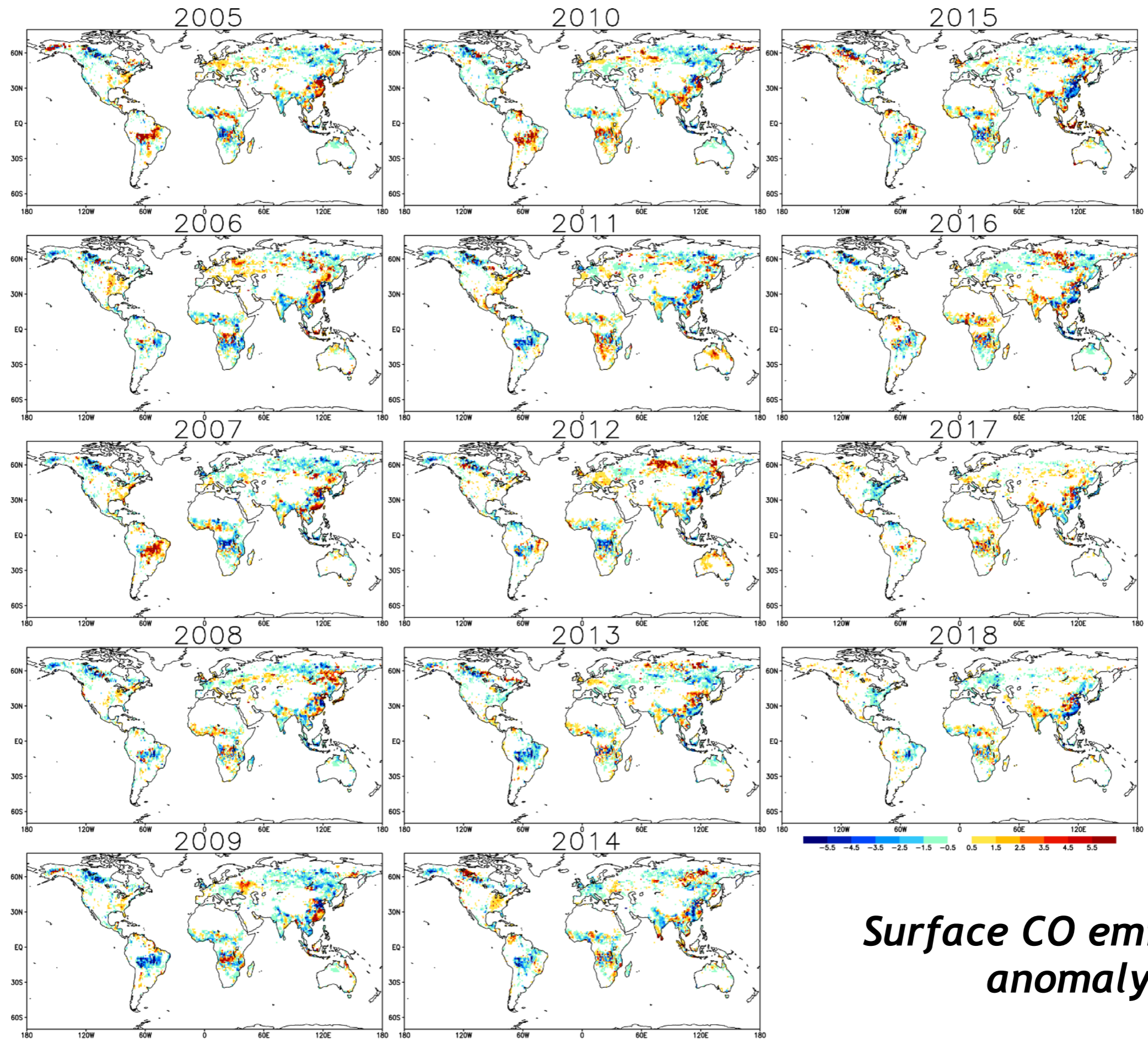


Fig. S7: Anomaly of annual mean surface CO emissions (in 10^{-10} kg CO m⁻² s⁻¹) from the 14-year mean (2005-2018) in TCR-2.



***Surface CO emissions
anomaly***

Fig. S8: Anomaly of annual mean surface SO₂ emissions (in 10⁻¹² kg S m⁻² s⁻¹) from the 14-year mean (2005-2018) in TCR-2.

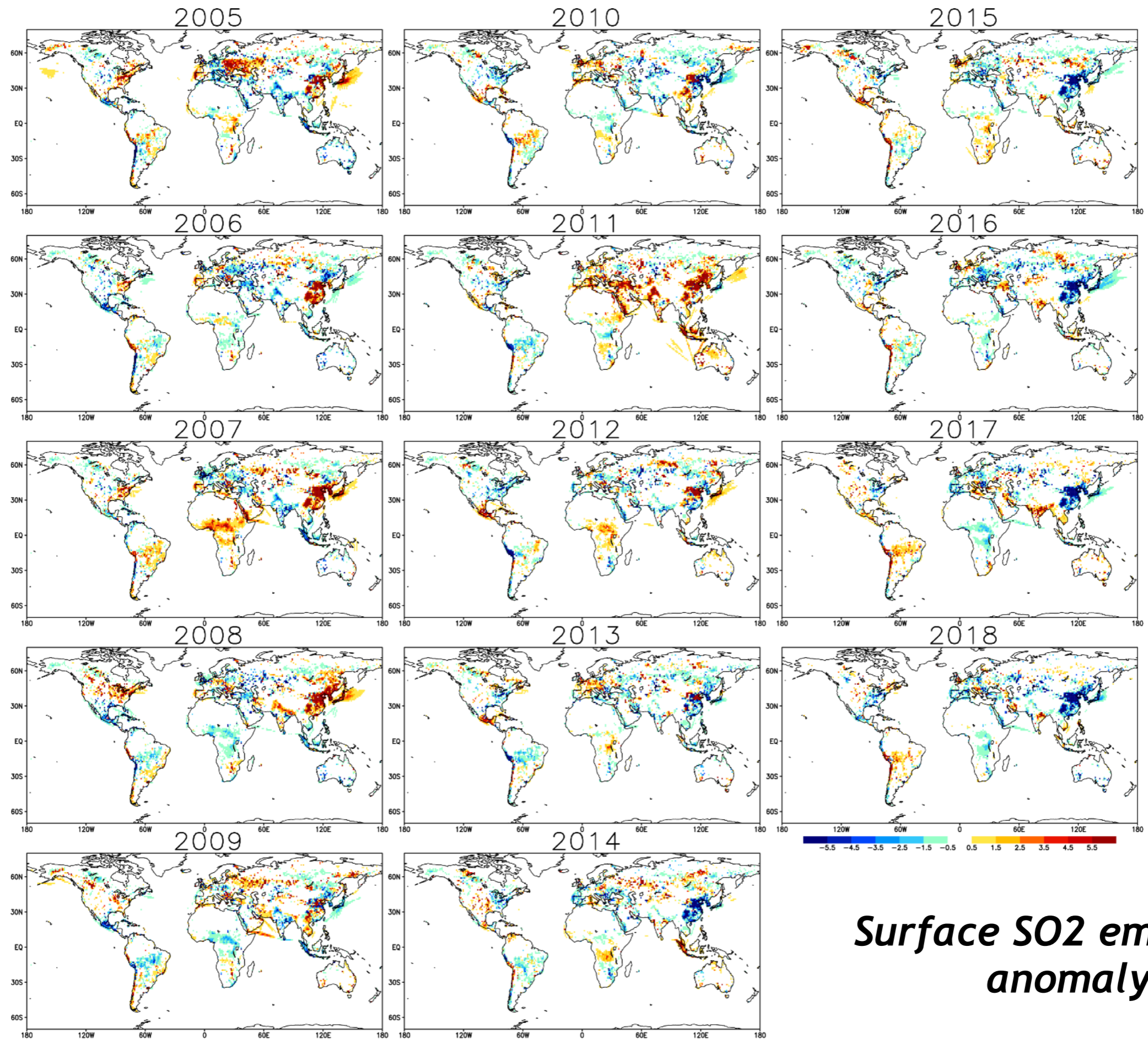


Fig. S9: Anomaly of annual mean lightning NO_x sources (in 10^{-12} kg N m⁻² s⁻¹) from the 14-year mean (2005-2018) in TCR-2.

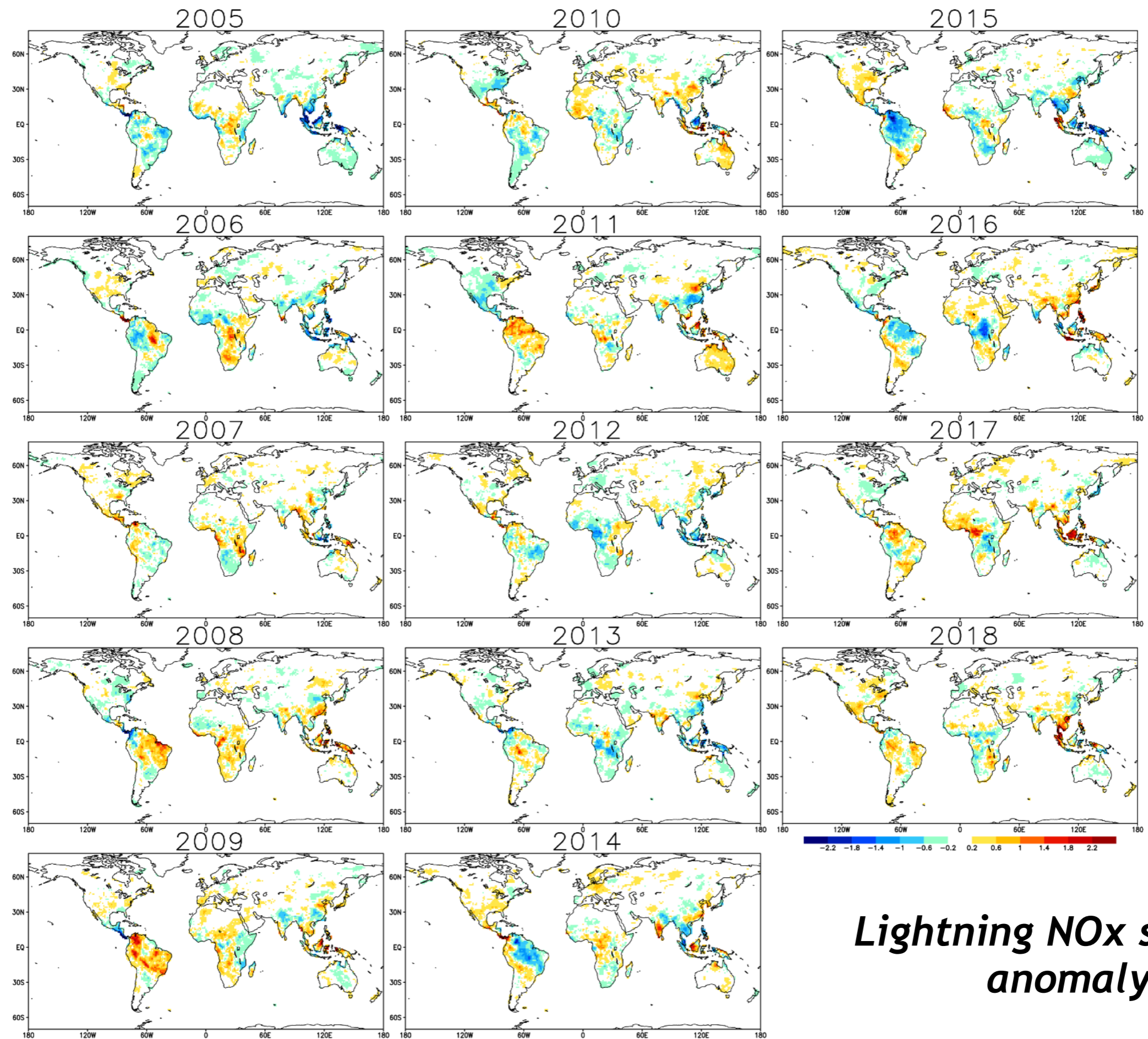
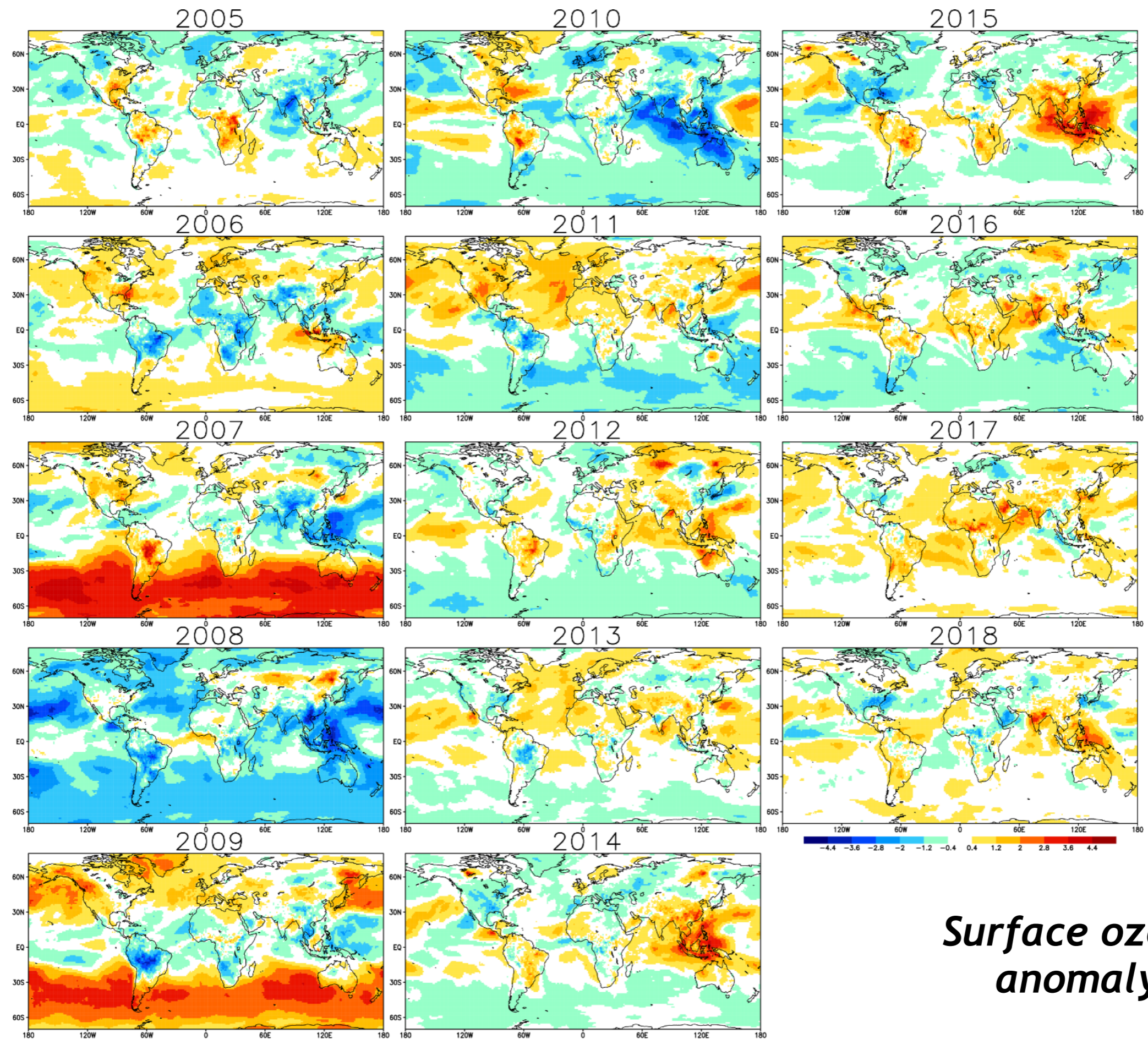


Fig. S10: Anomaly of annual mean surface ozone concentrations (in ppb) from the 14-year mean (2005-2018) in TCR-2.



***Surface ozone
anomaly***

Fig. S11: Same as in Fig S10, but for the noTES reanalysis.

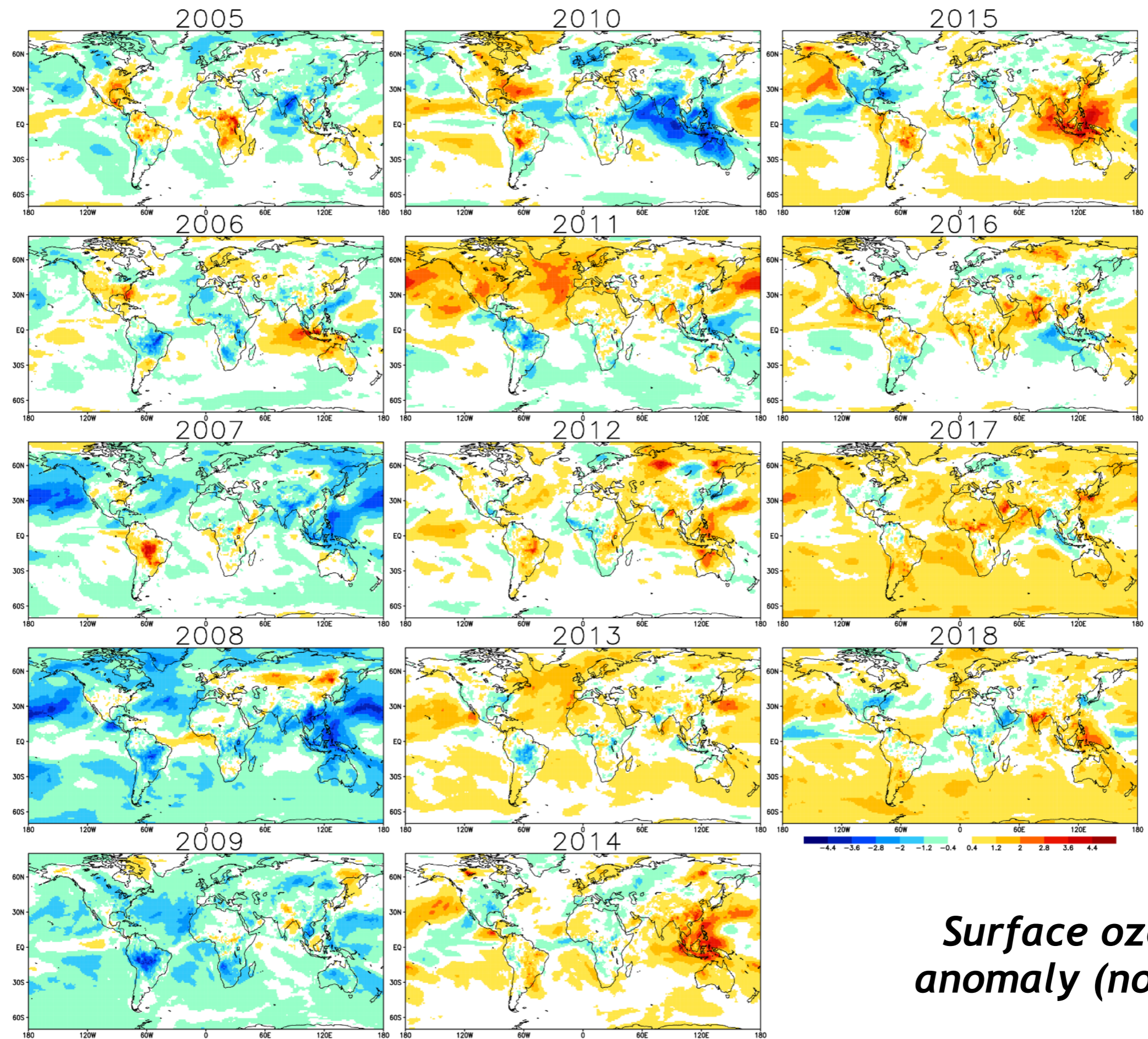


Fig. S12: Anomaly of annual mean 500 hPa ozone concentrations (in ppb) from the 14-year mean (2005-2018) in TCR-2.

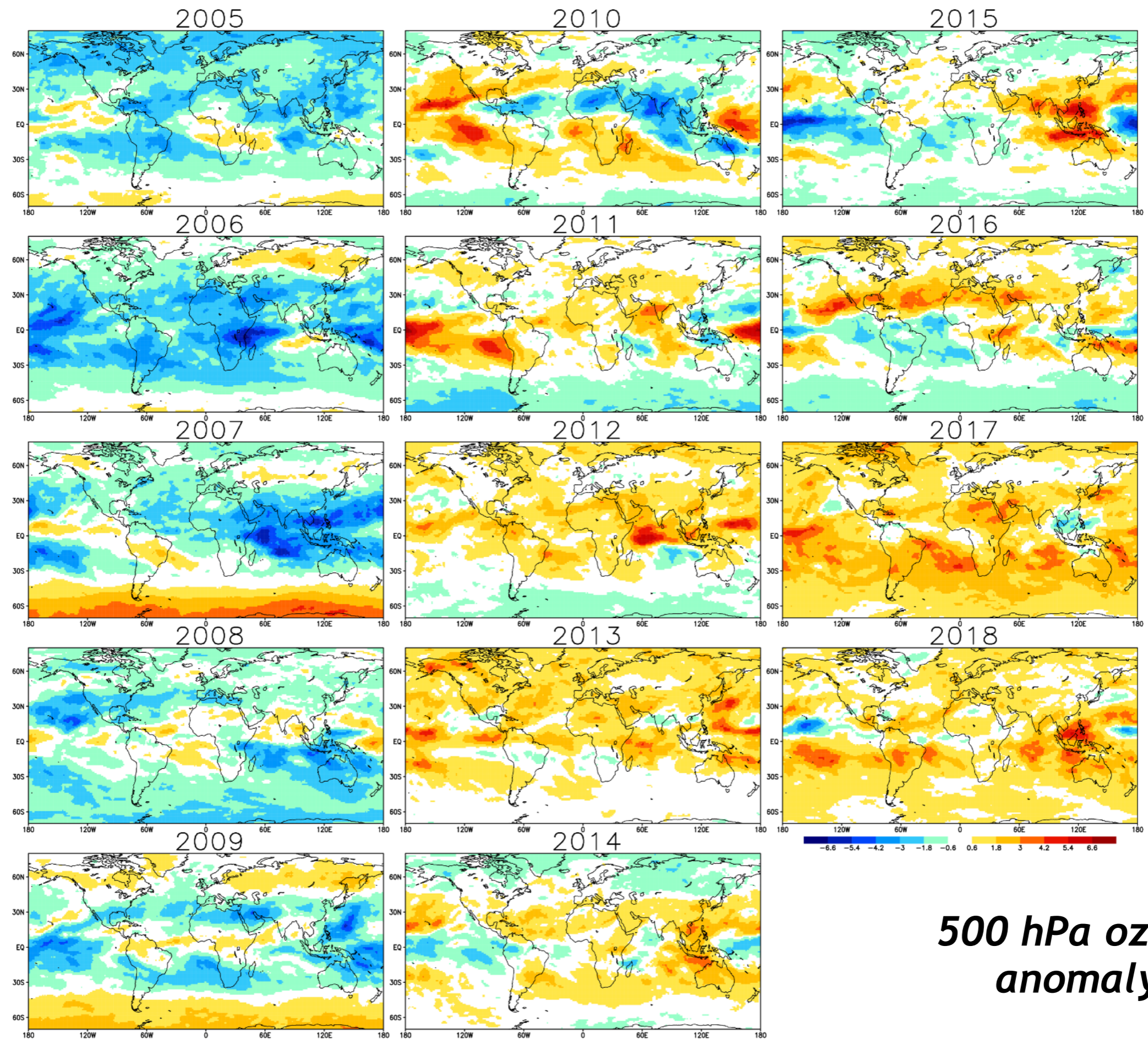


Fig. S13: Same as in Fig S12, but for the noTES reanalysis.

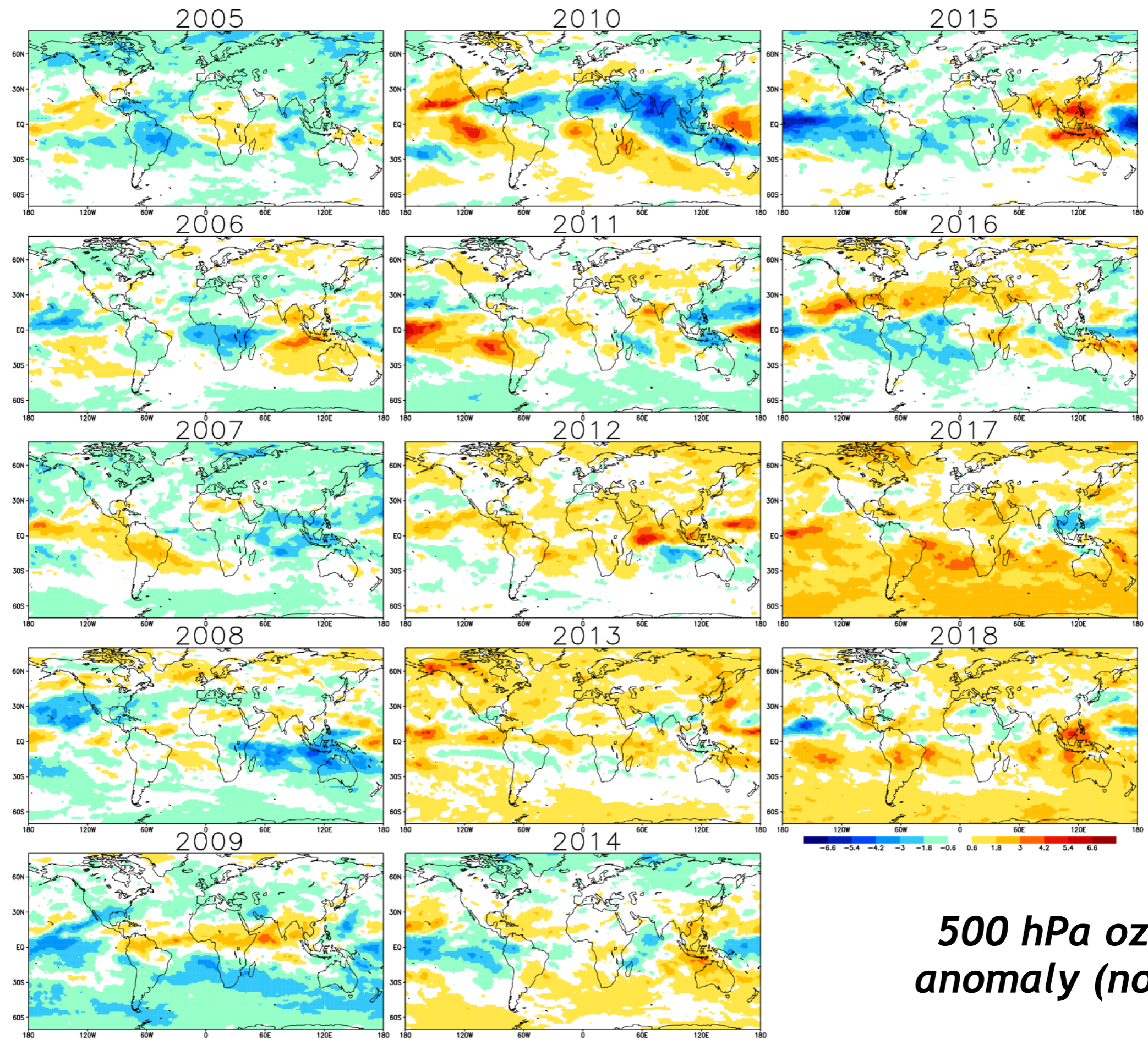
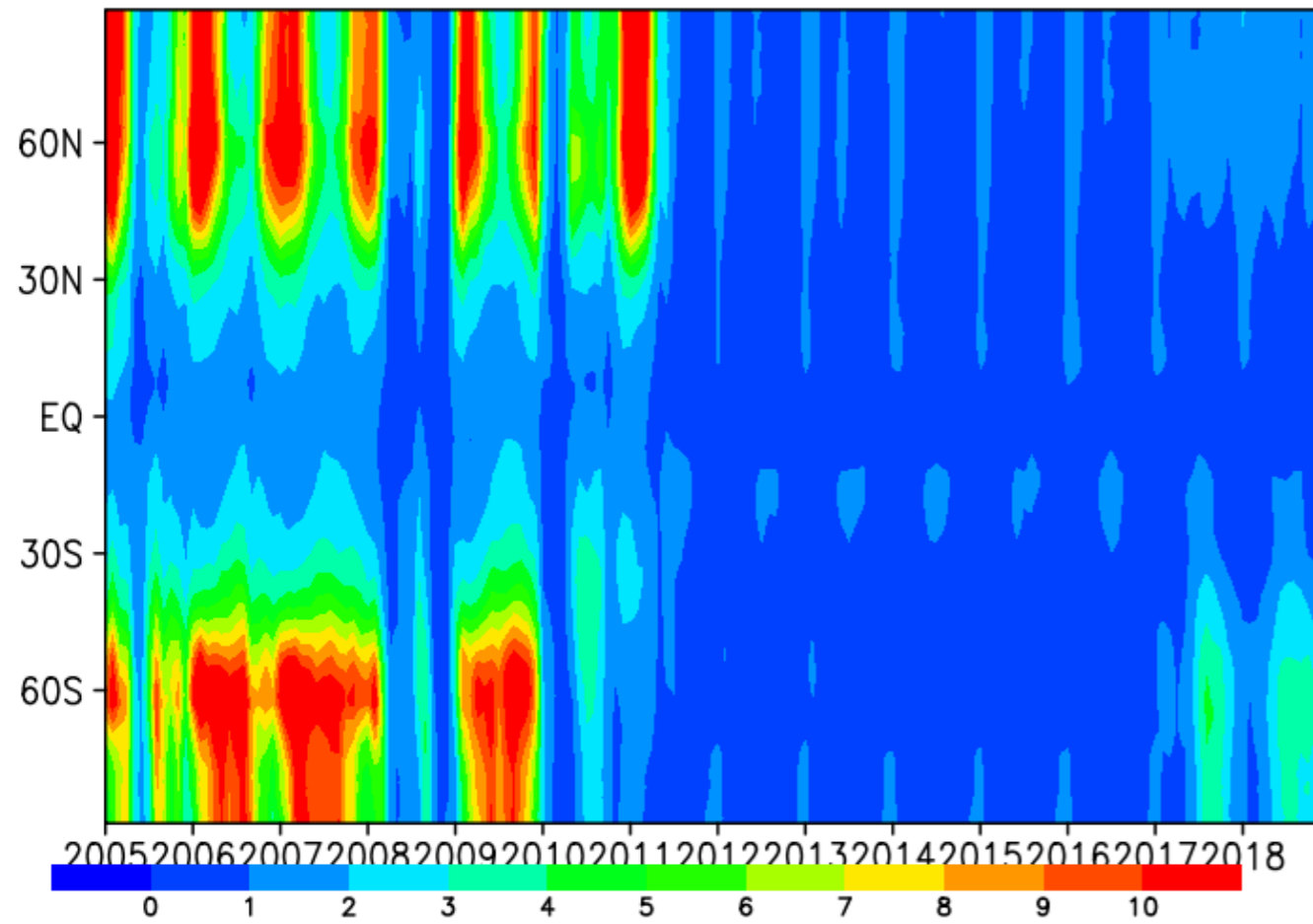


Fig. S14: Time-latitude cross section of zonal mean analysis spread of ozone (in ppb) at 600 hPa and 200 hPa.

Ozone analysis spread 600 hPa



Ozone analysis spread 200 hPa

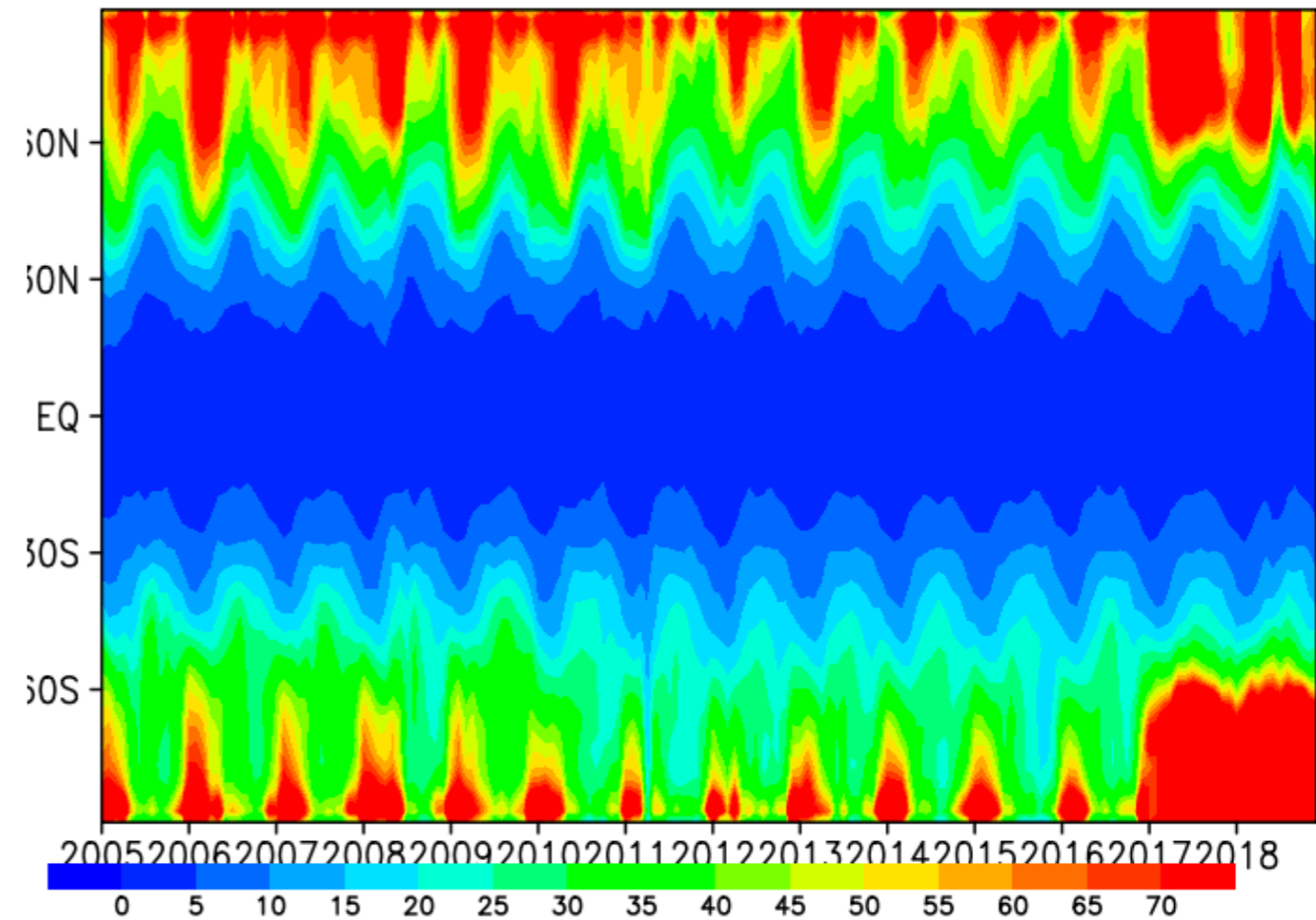


Table S1: Model minus observation comparisons of the mean ozone concentrations(in ppb) between the analysis or control run (in brackets) and the observations for HIPPO aircraft measurements during 2009–2011.

Ozone (ppb)		90-55S		55-15S		15S-15N		15N-55N		55N-90N	
		Bias	RMSE	Bias	RMSE	Bias	RMSE	Bias	RMSE	Bias	RMSE
850-500 hPa	Assim	-0.9	6.1	2.0	6.6	2.4	7.4	1.6	8.7	2.0	7.7
	Free	-4.5	6.2	-4.1	7.2	-3.6	7.5	-4.6	9.5	-4.0	7.3
500-200 hPa	Assim	4.6	1.9	4.6	14.3	5.0	10.0	4.5	19.4	2.5	36.2
	Free	42.6	58.7	4.3	19.6	-2.6	19.6	8.1	24.1	31.9	56.6

Table S2: Same as in Table S1, but for CO (in ppb).

CO (ppb)		90-55S		55-15S		15S-15N		15N-55N		55N-90N	
		Bias	RMSE	Bias	RMSE	Bias	RMSE	Bias	RMSE	Bias	RMSE
850-500 hPa	Assim	6.5	6.6	6.2	7.5	3.3	9.5	-9.7	16.5	-15.1	17.4
	Free	17.8	17.8	18.6	18.9	15.7	18.7	-5.9	16.4	-15.5	16.9
	Assim	8.7	8.9	7.9	9.4	7.1	10.1	-2.4	16.5	-4.0	6.5
	Free	19.9	20.0	21.6	22.1	21.7	22.4	5.0	20.0	-1.2	21.4

Table S3: Yearly surface NOx emissions in TgNyr⁻¹.

TgNyr ⁻¹	Globe	NH	Tropics	SH	Europe	USA	C Africa	E China	S Africa	S America	N Africa	SE Asia	Australia	India
2005	50.59	29.51	17.59	3.48	5.05	6.74	3.06	5.45	0.69	1.40	3.49	0.65	1.58	2.91
2006	50.60	30.24	16.74	3.62	5.21	6.46	2.89	5.64	0.68	1.15	3.00	0.57	1.85	3.01
2007	50.39	30.04	16.65	3.70	4.98	6.07	2.80	5.83	0.71	1.46	3.26	0.62	1.57	2.92
2008	48.50	30.04	15.17	3.29	4.78	5.65	2.75	5.83	0.67	0.99	3.12	0.49	1.36	2.96
2009	47.14	28.23	15.56	3.34	4.54	5.36	2.87	5.87	0.64	0.88	3.13	0.52	1.47	3.16
2010	49.83	30.23	16.29	3.31	4.57	5.08	2.87	6.77	0.67	1.56	2.91	0.61	1.25	3.11
2011	49.41	29.82	16.05	3.55	4.69	5.18	2.78	7.06	0.66	1.01	3.20	0.54	1.78	3.43
2012	49.87	28.92	17.16	3.78	4.63	5.05	2.88	6.86	0.68	1.15	3.13	0.59	1.78	3.45
2013	49.87	29.82	16.67	3.39	4.42	5.05	2.83	6.84	0.67	1.00	3.10	0.59	1.52	3.50
2014	50.30	29.19	17.50	3.61	4.40	4.98	2.77	6.43	0.72	1.08	3.22	0.65	1.62	3.54
2015	49.17	27.57	17.96	3.65	4.33	4.85	2.93	5.84	0.70	1.17	3.23	0.65	1.62	3.64
2016	47.93	27.39	17.19	3.35	4.47	4.84	2.84	5.63	0.68	1.05	3.30	0.67	1.45	3.66
2017	47.41	26.46	17.34	3.61	4.47	4.91	2.85	5.75	0.68	1.12	3.30	0.60	1.57	3.75
2018	48.27	27.87	16.91	3.48	4.41	4.66	2.83	5.76	0.68	1.13	3.03	0.61	1.58	3.78

Table S4: Yearly surface CO emissions in TgCOyr⁻¹.

TgCOyr ⁻¹	Globe	NH	Tropics	SH	Europe	USA	C Africa	E China	S Africa	S America	N Africa	SE Asia	Australia	India
2005	1159.57	616.55	503.78	39.26	47.91	67.80	156.43	195.03	7.53	79.30	107.30	14.75	11.79	73.85
2006	1111.34	612.92	460.56	37.88	48.94	71.27	159.31	183.60	7.10	35.76	92.31	11.28	16.99	68.19
2007	1108.76	589.86	477.70	41.22	38.30	71.19	142.79	177.46	7.68	68.18	104.14	16.68	14.27	76.75
2008	1058.13	591.94	429.61	36.59	40.65	69.87	164.48	174.37	8.29	30.49	112.71	10.29	9.77	74.54
2009	1022.17	549.56	433.24	39.38	38.55	63.43	172.43	176.35	6.70	10.06	95.46	10.91	15.52	70.61
2010	1152.88	577.29	530.65	44.96	41.97	56.93	186.01	179.26	8.40	86.27	102.68	17.59	8.66	79.78
2011	1088.59	583.90	457.03	47.68	41.65	73.54	174.98	176.92	8.64	23.87	110.75	10.53	25.58	73.81
2012	1130.63	638.17	448.83	43.64	49.94	67.46	150.93	179.53	7.94	32.26	106.95	13.31	26.85	80.96
2013	1065.01	591.91	430.82	42.29	43.38	57.67	168.20	183.18	8.71	12.67	105.12	14.83	12.34	76.58
2014	1102.37	597.61	467.14	37.63	34.96	69.74	164.90	168.54	9.03	19.09	102.23	11.70	14.94	73.35
2015	1137.98	577.93	524.99	35.08	41.53	66.87	160.97	157.55	7.22	32.48	108.25	13.93	16.73	75.38
2016	1122.68	591.68	494.34	36.67	44.12	60.55	166.87	173.70	6.65	33.90	127.14	18.31	11.17	88.99
2017	1130.49	597.30	495.79	37.41	44.85	52.35	175.64	177.02	8.80	42.29	114.21	12.69	14.16	93.13
2018	1062.53	564.73	457.88	39.94	37.45	55.31	164.94	174.46	7.34	32.13	110.05	14.44	16.90	86.58

Table S5: Yearly surface SO2 emissions in TgNyr⁻¹.

TgS yr ⁻¹	Globe	NH	Tropics	SH	Europe	USA	C Africa	E China	S Africa	S America	N Africa	SE Asia	Australia	India
2005	36.33	25.91	7.24	3.18	2.87	2.86	0.69	7.00	0.50	0.42	0.66	0.13	1.01	1.51
2006	35.54	25.17	7.11	3.26	2.94	2.52	0.59	7.54	0.49	0.37	0.58	0.13	1.13	1.63
2007	39.04	27.65	7.99	3.40	2.87	2.73	0.75	8.25	0.48	0.46	1.13	0.13	1.17	1.54
2008	37.12	27.00	6.89	3.24	2.90	2.88	0.60	7.70	0.39	0.30	0.45	0.13	1.13	1.84
2009	35.50	24.87	7.09	3.54	2.95	2.64	0.66	6.09	0.50	0.28	0.47	0.15	1.35	1.66
2010	34.65	23.76	7.48	3.40	3.03	2.38	0.71	5.85	0.56	0.47	0.53	0.13	1.21	1.74
2011	39.13	27.74	7.89	3.51	3.21	2.48	0.69	6.89	0.43	0.29	0.69	0.13	1.54	2.19
2012	35.43	24.32	7.72	3.40	2.79	2.33	0.74	6.45	0.52	0.34	0.77	0.12	1.28	1.74
2013	34.28	23.62	7.20	3.46	2.89	2.42	0.70	5.74	0.50	0.29	0.61	0.11	1.27	1.79
2014	33.76	22.70	7.65	3.41	2.67	2.54	0.74	4.61	0.50	0.34	0.61	0.12	1.28	1.86
2015	33.25	21.88	7.76	3.61	2.70	2.42	0.74	4.11	0.57	0.40	0.60	0.13	1.31	1.88
2016	32.09	21.26	7.30	3.53	2.67	2.39	0.60	3.79	0.58	0.40	0.57	0.14	1.24	2.01
2017	33.05	21.80	7.66	3.59	2.70	2.51	0.56	3.62	0.60	0.52	0.48	0.15	1.31	2.25
2018	31.58	20.65	7.49	3.44	2.68	2.46	0.58	3.46	0.58	0.49	0.56	0.12	1.18	1.93

Table S6: Yearly lightning NOx sources in TgNyr⁻¹.

TgN yr ⁻¹	Globe	NH	Tropics	SH	N America	S America	S Africa	N Africa	Siberia	India	SE Asia	Pacific	Atlantic	Australia
2005	6.80	2.32	3.69	0.79	0.48	0.98	0.80	0.74	0.51	0.06	0.77	0.19	0.02	0.21
2006	7.08	2.37	3.87	0.84	0.50	1.07	0.90	0.66	0.55	0.05	0.86	0.17	0.01	0.26
2007	7.61	2.55	4.19	0.87	0.52	1.11	0.83	0.73	0.58	0.06	0.98	0.21	0.03	0.26
2008	7.72	2.44	4.39	0.89	0.43	1.25	0.90	0.73	0.53	0.06	1.06	0.22	0.02	0.28
2009	7.88	2.54	4.38	0.96	0.50	1.34	0.82	0.75	0.58	0.06	1.04	0.23	0.02	0.25
2010	7.45	2.49	4.07	0.89	0.40	1.04	0.75	0.72	0.57	0.06	0.98	0.24	0.03	0.32
2011	7.70	2.35	4.36	0.99	0.39	1.28	0.81	0.70	0.58	0.07	0.98	0.26	0.02	0.36
2012	7.38	2.53	3.89	0.96	0.50	1.07	0.74	0.66	0.56	0.07	0.88	0.28	0.02	0.27
2013	7.33	2.43	3.97	0.93	0.46	1.14	0.69	0.67	0.57	0.07	0.87	0.28	0.03	0.26
2014	7.63	2.58	4.09	0.97	0.54	0.99	0.81	0.74	0.57	0.07	0.93	0.27	0.03	0.28
2015	7.13	2.44	3.75	0.94	0.55	0.94	0.77	0.69	0.53	0.07	0.88	0.28	0.03	0.22
2016	7.67	2.62	4.05	1.01	0.44	1.05	0.75	0.76	0.58	0.07	1.06	0.27	0.02	0.24
2017	7.76	2.47	4.33	0.97	0.45	1.20	0.76	0.82	0.54	0.05	1.08	0.23	0.03	0.25
2018	7.86	2.59	4.31	0.96	0.55	1.20	0.79	0.71	0.58	0.06	1.08	0.26	0.02	0.26