Supplement of Geosci. Model Dev., 15, 4657–4687, 2022
https://doi.org/10.5194/gmd-15-4657-2022-supplement

Supplement of

Regional evaluation of the performance of the global CAMS chemical modeling system over the United States (IFS cycle 47r1)

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Table S1: Updates to the C_5H_8 oxidation scheme in CBA as compared to Huijnen et al. (2019). [1] Stavrakou et al. (2010), [2] Lamarque et al. (2012), [3] Myriokefalitakis et al. (2020), [4] spectral absorption data from http://iupac.pole-ether.fr (last access: 21 Sept 2021), [5] Quantum Yields as for 298K.

Reaction	Rate expression	Reference
OH + ISOP → 0.65 $ISOPBO_2$ + 0.35 $ISOPDO_2$	2.7 x 10^{-12} x exp(390/T)	[1]
$ISOPBO_2$ → HPALD1 + HO_2	4.1 x10^{8} x exp(-7700/T)	[1],[3]
$ISOPBO_2$ → ISPD + CH_2O + HO_2	2.08 x 10^{11} x exp(-8993/T)	[1]
$ISOPDO_2$ → HPALD2 + HO_2	4.1x10^{8} x exp(-7700/T)	[1],[3]
$ISOPDO_2$ → ISPD + CH_2O + HO_2	2.08 x 10^{11} x exp(-8993/T)	[1]
$ISOPBO_2$ + HO_2 → ISOPOOH	2.05 x 10^{-13} x exp(1300/T)	[2]
$ISOPBO_2$ + NO → 0.08 ORGNTR + 0.92 NO_2 + HO_2 + 0.51 CH_2O + 0.55 ISPD + 0.37 HPALD1	4.4 x 10^{-12} x exp(180/T)	[2]
$ISOPDO_2$ + HO_2 → ISOPOOH	2.05 x 10^{-13} x exp(1300/T)	[2]
$ISOPDO_2$ + NO → 0.08 ORGNTR + 0.92 NO_2 + HO_2 + 0.51 HCHO + 0.55 ISPD + 0.37 HPALD2	4.4 x 10^{-12} x exp(180/T)	[2]
OH + ISOPOOH → 0.1 XO2 + 0.4 CH_3COCHO + 0.3 CHOCHO + 0.12 ISOPBO2 + 0.08 ISOPDO2	1.52 x 10^{-11} x exp(200/T)	[1]
ISOPOOH + hν → 0.69 ISPD + 0.69 HCHO + HO_2	Explicit	[4]
OH + HPALD1 → 0.65 XO_2+ 0.25 CHOCHO + 0.1 CH_3COCHO	1.86 x 10^{-11} x exp(175/T)	[2]
HPALD1 + hν → OH + HO_2 + 0.5 HYAC + 0.5 CH_3COCHO + 0.5 GLYALD + HCHO	Explicit	[4]
OH + HPALD2 → 0.65 XO_2+0.25 CHOCHO + 0.1 CH_3COCHO	1.86 x 10^{-11} x exp(175/T)	[1]
HPALD2 + hν → HO_2 + OH + 0.5 HYAC + 0.5 CHOCHO +0.5 GLYALD + HCHO	Explicit	[4]
OH + CHOCHO → 0.63 HO_2 + 1.26 CO + C_2O_3	3.1 x 10^{-12} x exp(340/T)	[1]
CHOCHO + hν → 2 CO + 2 HO_2	Explicit	[4],[5]
CHOCHO + hν → HCHO + CO	Explicit	[4],[5]
OH + GLYALD → 0.25 OH + 0.75 HO_2 + 0.17 CHOCHO + 0.17 HCOOH + 0.67 HCHO + 0.5 CO	8.0 x 10^{-12}	[1]
GLYALD + hν → 2HO_2 + CO + HCHO	Explicit	[4]
OH + HYAC → 0.1 OH + 0.825 HO_2 + 0.75 CH_3COCHO + 0.125 HCOOH + 0.125 CH_3O_2 + 0.125 CH_3COOH + 0.05 CO	2.0 x 10^{-12} x exp(320/T)	[1]
HYAC + hν → C_2O_3 + HO_2 + HCHO	As J(CH_3COCH_3)	[1]

Table S2: Updates to the oxidation of TOL and XYL as implemented in the IFS(CBA) chemistry as compared to Huijnen et al. (2019). The reaction scheme is adapted from Karl et al. (2009), with modification to the product distribution for loss of $AROO_2$ following Myriokefalitakis et al. (2020). (*) This indicates the final rate applied accounts for the ortho-, meta- and para-isomers of the cyclic aromatics.

Reaction	Rate expression
$OH + TOL \rightarrow 5PAR + AROO_2$	5.96×10^{-12}
$O_3 + TOL \rightarrow 5PAR + AROO_2$	$2.34 \times 10^{-17} \times \exp(-6694/T)$
$NO_3 + TOL \rightarrow ORGNTR + PAR$	6.8×10^{-17}
$OH + XYL \rightarrow 5PAR + AROO_2$	avg of (1.3×10^{-11}, 2.36×10^{-11}, 1.43×10^{-11})*
$O_3 + XYL \rightarrow 5PAR + AROO_2$	avg of ($5.37 \times 10^{-13} \times \exp(-6039/T)$, $1.91 \times 10^{-13} \times \exp(-5586/T)$, $2.4 \times 10^{-13} \times \exp(-5586/T)$)
$NO3 + XYL \rightarrow CH_3COCHO + PAR$	avg of (3.6×10^{-16}, 2.33×10^{-16}, 4.5×10^{-16})*
$NO + AROO_2 \rightarrow NO_2 + CHOCHO + 0.33\ CH_3COCHO$	$4.2 \times 10^{-12} \times \exp(180/T)$
$XO_2 + AROO_2$	$1.7 \times 10^{-14} \times \exp(1300/T)$
$HO_2 + AROO_2 \rightarrow ROOH + CHOCHO$	$3.5 \times 10^{-13} \times \exp(1000/T)$
$AROO_2 + AROO_2$	$1.7 \times 10^{-14} \times \exp(1300/T)$

Table S3: Details related to the inclusion of HCN and CH_3CN in IFS(CBA), with rate expressions coming from Atkinson et al. (2004).

Reaction	Rate expression	Comments
$OH + HCN \rightarrow$	$1.2 \times 10^{-13} \times \exp(-400/T)$	No products defined
$OH + CH_3CN \rightarrow 0.3HCN$	$8.1 \times 10^{-13} \times \exp(-1080/T)$	Products not completely defined

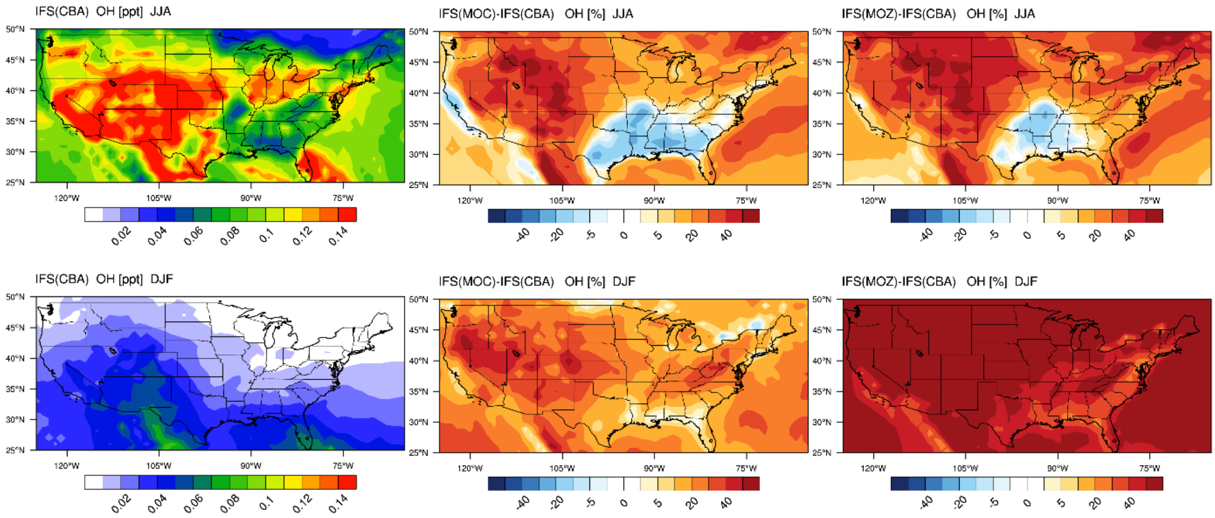

Figure S1. The horizontal seasonal mean for tropospheric OH below 1km over the US domain for JJA 2014 (top) and DJF (2014/2015).

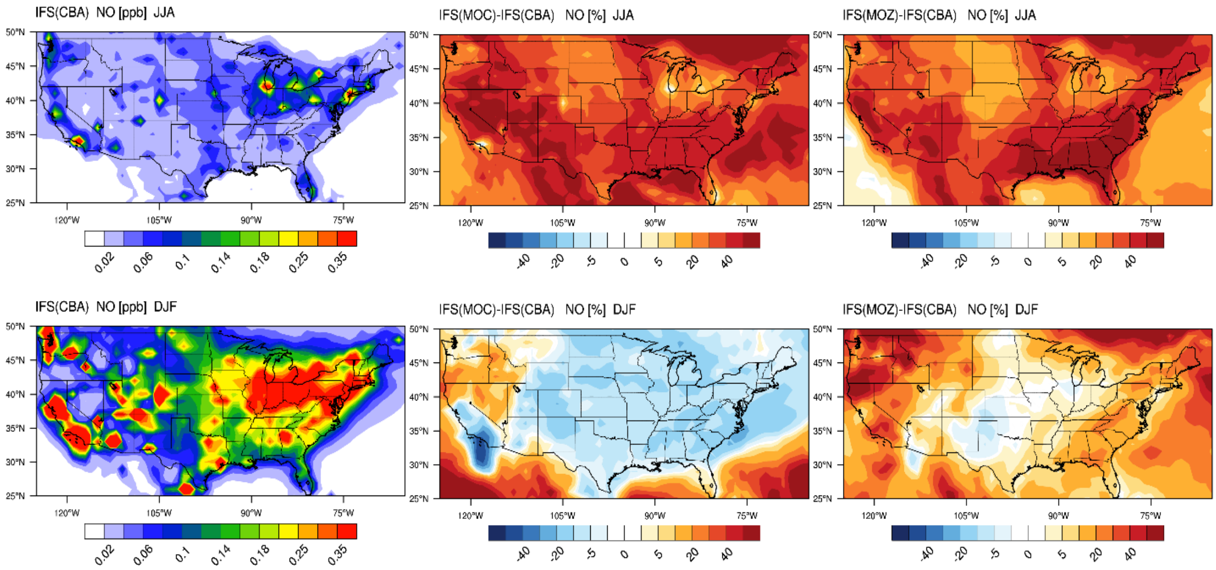

Figure S2. The horizontal seasonal mean for tropospheric NO below 1km over the US domain for JJA 2014 (top) and DJF (2014/2015).

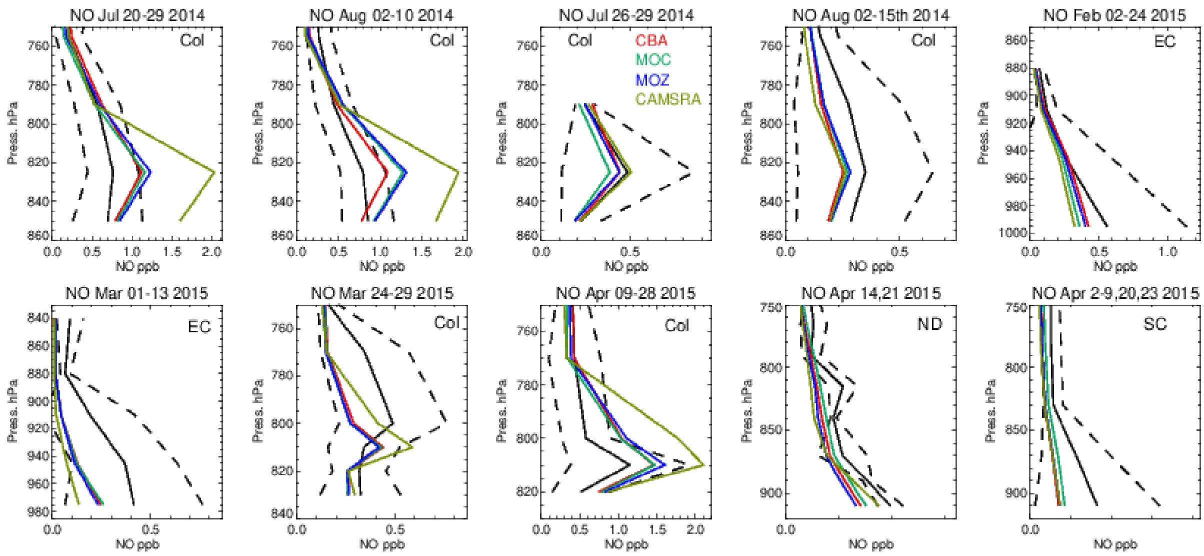

Figure S3. Comparisons of lower tropospheric NO profiles for 2014/2015 against composites of aircraft measurements for the regional domains shown in Figure 1. Campaigns shown (top left to bottom right) are DISCOVER-AQ, FRAPPE, WINTER and SONGNEX.

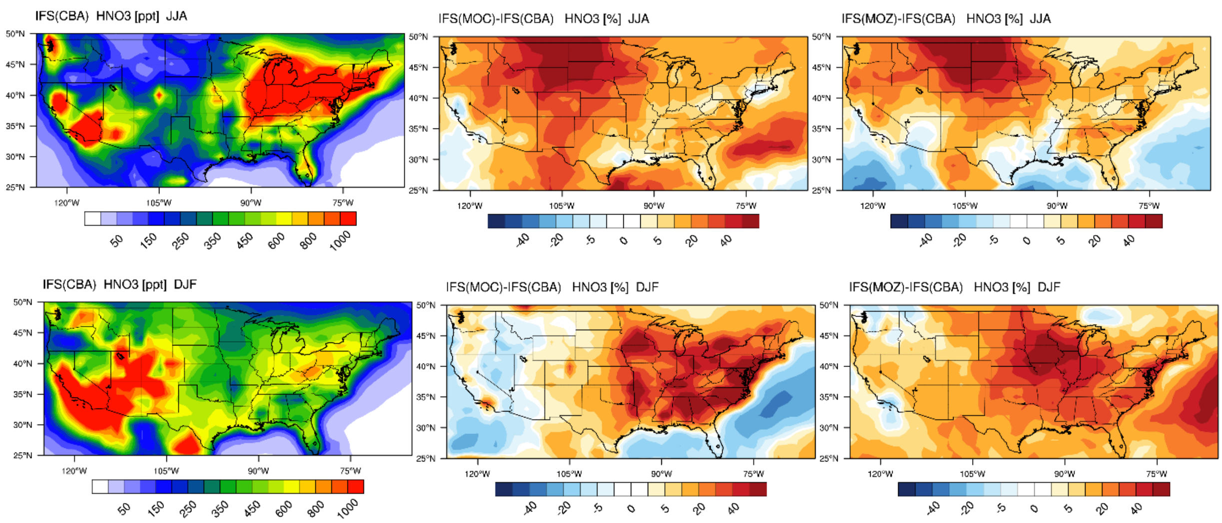

Figure S4. The horizontal seasonal mean for tropospheric HNO_3 below 1km over the US domain for JJA 2014 (top) and DJF (2014/2015).

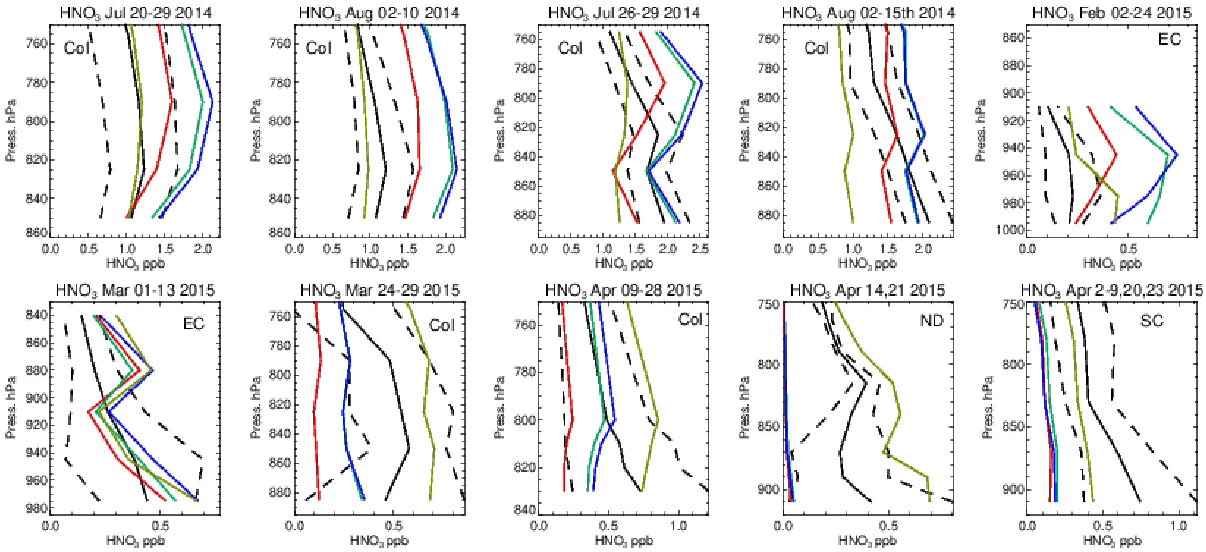

Figure S5. Comparisons of lower tropospheric HNO_3 profiles for 2014/2015 against composites of aircraft measurements for the regional domains shown in Figure 1. Campaigns shown (top left to bottom right) are DISCOVER-AQ, FRAPPE, WINTER and SONGNEX.

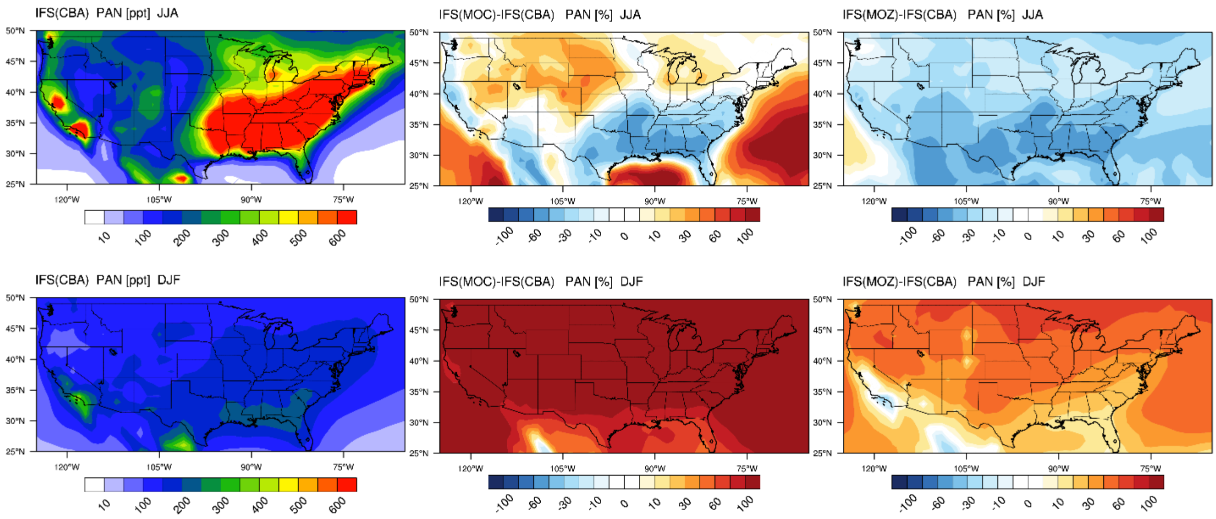

Figure S6. The horizontal seasonal mean for tropospheric PAN below 850 hPa over the US domain for JJA 2014 (top) and DJF (2014/2015).

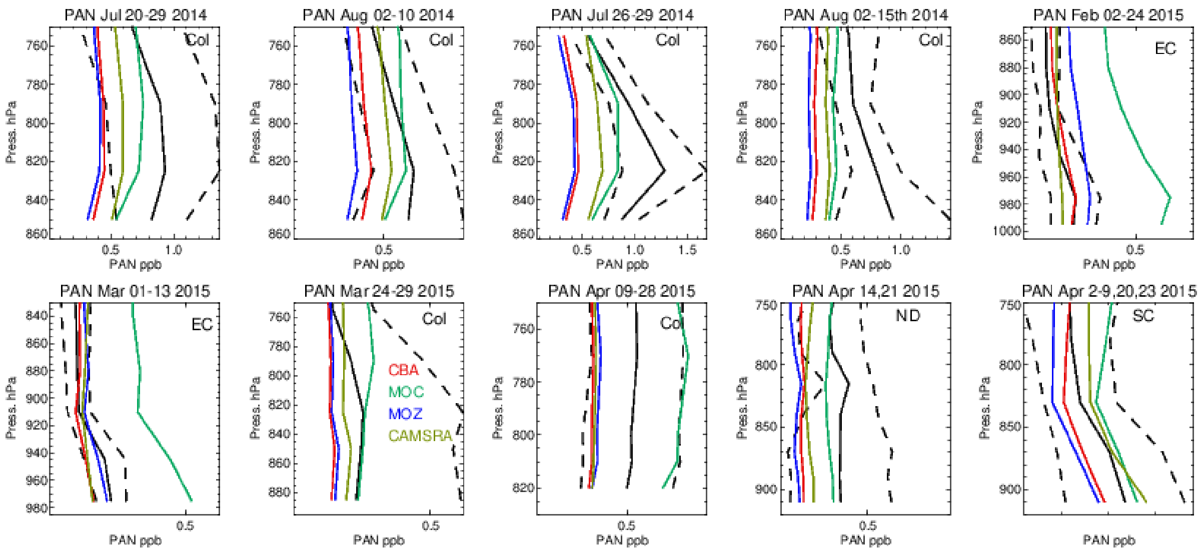

Figure S7. Comparisons of lower tropospheric PAN profiles for 2014/2015 against composites of aircraft measurements for the regional domains shown in Figure 1. Campaigns shown (top left to bottom right) are DISCOVER-AQ, FRAPPE, WINTER and SONGNEX.

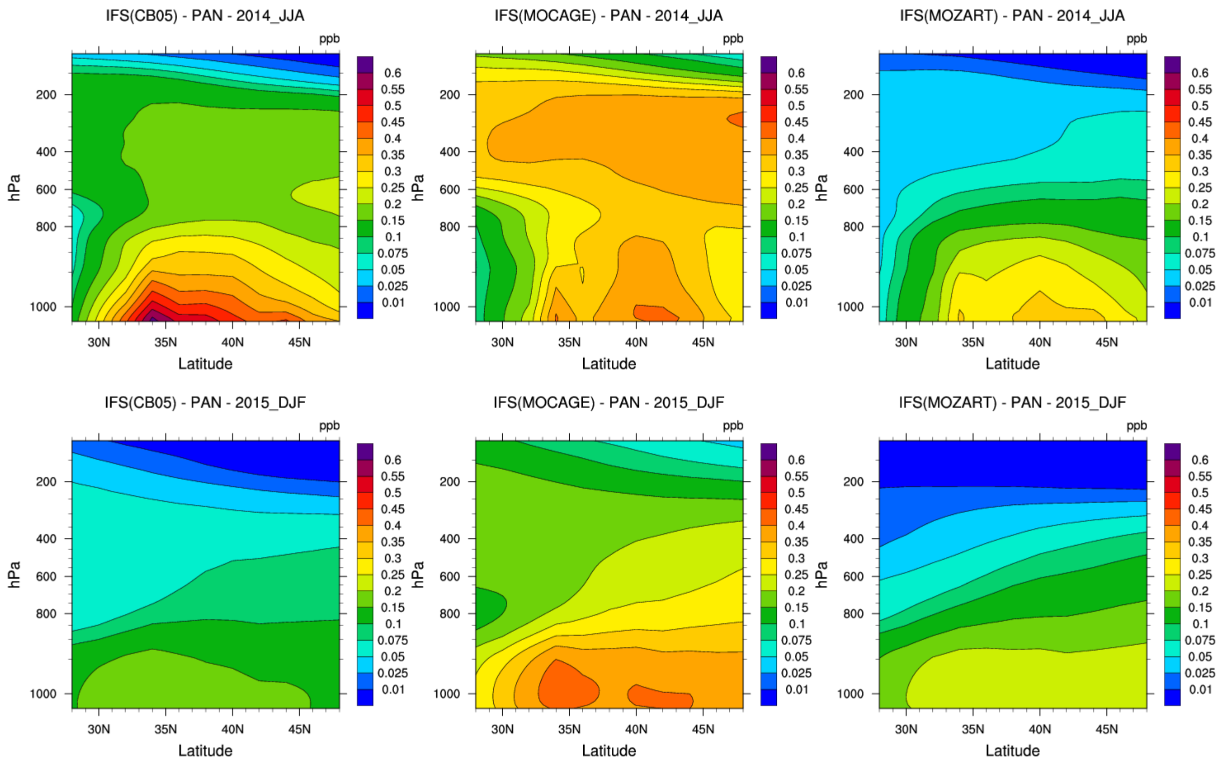

Figure S8. Seasonal zonal mean distributions of PAN profiles for 2014/2015 over the US domain.

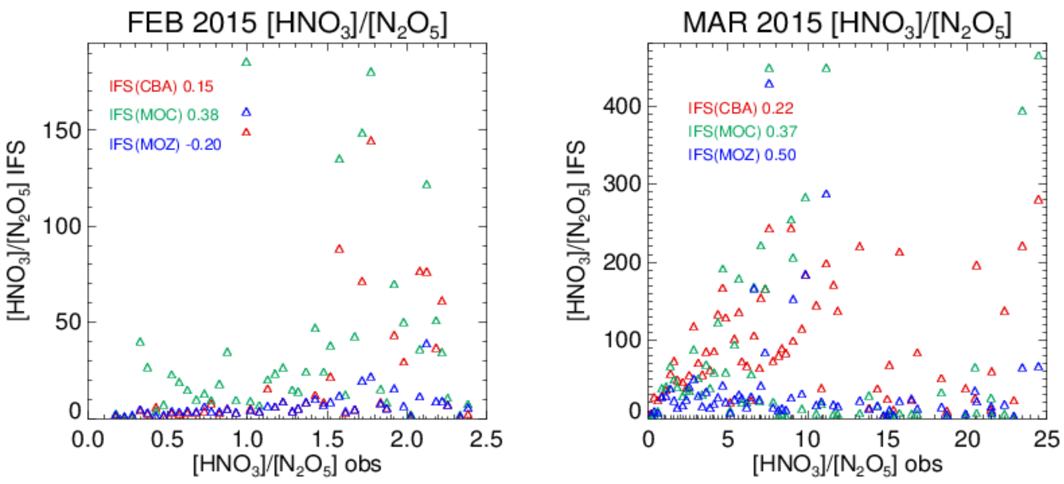

Figure S9. Comparisons of the $[HNO_3]/[N_2O_5]$ ratio between nighttime observations and those simulated in the IFS, with the associated correlation coefficients being given in each panel.

Month/region	nobs	mean observations	IFS(CBA)	IFS(MOC)	IFS(MOZ)	CAMSRA
05.2014 /ND	2558	59.4 ± 32.4	-15.3 ± 16.9	-14.3 ± 16.9	-13.2 ± 17.0	-9.5 ± 17.2
06.2014 /NM	478	60.5 ± 3.5	11.4 ± 7.8	10.8 ± 7.8	8.8 ± 7.8	6.8 ± 8.1
*07.2014/ Col	450	62.7 ± 6.3	-7.3 ± 7.0	-5.7 ± 7.9	-5.2 ± 7.2	-16.6 ± 7.4
07.2014/ Col	2838	61.8 ± 9.9	-5.3 ± 2.2	0.5 ± 2.6	-4.5 ± 2.4	-11.1 ± 2.3
*08.2014/ Col	354	62.0 ± 8.4	-4.6 ± 6.7	0.0 ± 7.2	-5.0 ± 6.8	-16.2 ± 7.6
08.2014/ Col	1024	63.2 ± 9.5	2.2 ± 4.1	6.0 ± 4.0	0.1 ± 4.0	-7.9 ± 3.9
02.2015/EC	1054	37.7 ± 2.7	1.2 ± 2.9	4.9 ± 2.9	11.8 ± 3.0	0.7 ± 3.4
03.2015/EC	477	45.5 ± 4.3	-1.7 ± 3.8	2.5 ± 3.8	9.8 ± 3.9	0.5 ± 4.2
03.2015/Col	3434	49.4 ± 3.7	0.6 ± 1.3	3.9 ± 1.6	4.7 ± 1.6	2.5 ± 1.3
04.2015/ND	4018	51.2 ± 7.7	0.5 ± 0.6	2.0 ± 0.6	5.8 ± 0.6	6.7 ± 0.8
04.2015/Col	2560	53.3 ± 3.6	-0.9 ± 1.8	6.8 ± 1.5	6.6 ± 1.7	-0.8 ± 1.6
04.2015/Texas	1869	44.0 ± 11.9	5.5 ± 2.6	5.3 ± 2.6	5.1 ± 2.6	5.3 ± 2.6

Table S4. Mean biases and standard deviations from the mean of tropospheric O_3 for the lower troposphere using all valid points aggregated over the selected days for each of the aircraft campaigns presented in Table 9. Only points below 815 hPa (Colorado) or 900 hPa (EC,ND,Texas) are included thus limiting the sample size but minimizing transport effects. Each mean bias is calculated as the difference between the IFS mini-ensemble member minus observational value from the respective aircraft campaign. In addition, the number of observations ('n. obs') and observational mean, and its standard deviation, are given.

Month/region	nobs	mean observations	IFS(CBA)	IFS(MOC)	IFS(MOZ)	CAMSRA
05.2014 /ND	2558	125.5 ± 6.4	-3.6 ± 3.9	-11.1 ± 3.9	-31.9 ± 3.8	2.4 ± 4.1
06.2014 /NM	478	145.2 ± 34.3	-41.6 ± 13.6	-50.8 ± 13.5	-62.1 ± 13.6	-34.9 ± 13.6
*07.2014/ Col	437	131.3 ± 6.6	22.2 ± 27.2	5.5 ± 26.6	6.1 ± 26.6	15.8 ± 32.7
*08.2014/ Col	366	125.3 ± 12.0	15.2 ± 33.9	1.5 ± 40.0	-0.4 ± 34.1	15.4 ± 42.2
08.2014/Col	1081	118.1 ± 26.9	4.1 ± 7.8	-9.2 ± 7.6	-11.1 ± 7.6	-9.3 ± 7.5
02.2015/EC	1047	158.4 ± 10.0	-12.0 ± 13.9	-21.0 ± 13.9	-36.5 ± 13.8	-11.7 ± 14.6
03.2015/EC	487	147.5 ± 11.5	-10.8 ± 11.7	-20.1 ± 11.7	-38.0 ± 11.8	-8.5 ± 12.1
03.2015/Col	3291	135.4 ± 20.6	-10.9 ± 7.3	-21.8 ± 7.2	-38.1 ± 7.2	-7.5 ± 8.2
04.2015/ND	3742	127.9 ± 17.5	-5.1 ± 0.7	-16.8 ± 0.4	-36.2 ± 0.1	-2.1 ± 0.8
04.2015/Col	2459	173.1 ± 47.3	-10.9 ± 21.2	-22.8 ± 20.7	-39.9 ± 20.1	-10.6 ± 15.1
04.2015/Texas	1793	133.0 ± 11.0	-4.8 ± 1.8	-18.0 ± 1.7	-29.2 ± 1.5	1.5 ± 2.8

Table S5: As for Table S4 except for tropospheric CO

Month/region	nobs	mean observations	IFS(CBA)	IFS(MOC)	IFS(MOZ)	CAMSRA
*07.2014/ Col	452	2462 ± 572	-359 ± 829	-564 ± 849	-345 ± 802	324 ± 1096
07.2014/ Col	2894	1764 ± 109	-33 ± 291	-350 ± 299	8 ± 306	91 ± 314
*08.2014/Col	367	1971 ± 464	-249 ± 592	-380 ± 594	-209 ± 558	254 ± 799
08.2014/Col	1038	2125 ± 1041	-816 ± 303	-931 ± 302	-752 ± 304	-857 ±296
02.2015/EC	837	458 ± 239	-199 ± 157	-128 ± 159	-158 ± -156	-318 ± 169
03.2015/EC	638	506 ± 186	-211 ± 170	-127 ± 186	-126 ± 167	-320 ± 172
03.2015/Col	3345	800 ± 140	-306 ± 150	-243 ± 149	-163 ± 149	-371 ± 145
04.2015/Col	2579	1055 ± 384	-313 ± 362	-220 ± 355	-150 ± 361	-578 ± 363
04.2015/Texas	1659	874 ± 530	136 ± 77	-2 ± 63	182 ± 65	-35 ± 65

Table S6. As for Table S4 except for tropospheric CH_2O.

Month/region	nobs	mean observations	IFS(CBA)	IFS(MOC)	IFS(MOZ)	CAMSRA
*07.2014/ Col	469	2284 ± 955	488 ± 1913	-64 ± 1838	-6 ± 1808	865 ± 2216
07.2014/ Col	2329	915 ± 776	-218 ± 642	-358 ± 641	-383 ± 641	-330 ± 666
*08.2014/Col	359	2071 ± 603	141 ± 2086	33 ± 2120	-22 ± 2108	793 ± 2496
08.2014/Col	907	1732 ± 1766	-965 ± 661	-1048 ± 655	-1097 ± 652	-1216 ± 659
02.2015/EC	973	2535 ± 812	-276 ± 1128	-374 ± 1139	-299 ± 1120	-896 ± 1108
03.2015/EC	470	1446 ± 879	-300 ± 1170	-240 ± 1125	-406 ± 1089	-717 ± 1045
03.2015/Col	3345	788 ± 236	-164 ± 501	-230 ± 507	-254 ± 502	-129 ± 501
04.2015/ND	3962	163 ± 167	-70 ± 194	-77 ± 194	-69 ± 194	-61 ± 196
04.2015/Col	2297	2293 ± 834	1408 ± 834	404 ± 651	125 ± 650	2293 ± 640
04.2015/Texas	1799	389 ± 287	195 ± 271	202 ± 269	158 ± 267	161 ± 266

Table S7. As for Table S4 except for tropospheric NO_2 with mean biases and associated standard deviations are given in ppt.

Month/region	nobs	mean observations	IFS(CBA)	IFS(MOC)	IFS(MOZ)	CAMSRA
*07.2014/ Col	448	741 ± 406	162 ± 749	235 ± 839	273 ± 836	1194 ± 1651
07.2014/ Col	2330	241 ± 199	-57 ± 233	-67 ± 232	-67 ± 233	-73 ± 238
*08.2014/Col	344	846 ± 325	-26 ± 1018	75 ± 1087	141 ± 1145	962 ± 1625
08.2014/Col	953	510 ± 515	-316 ± 358	-301 ± 358	-303 ± 359	-326 ± 360
02.2015/EC	799	368 ± 299	58 ± 372	-7 ± 372	37 ± 375	-50 ± 403
03.2015/EC	408	356 ± 178	-192 ± 291	-174 ± 285	-199 ± 291	-254 ± 312
03.2015/Col	3356	345 ± 198	-95 ± 233	-99 ± 235	-102 ± 235	-90 ± 211
04.2015/ND	4002	152 ± 25	-94 ± 130	97 ± 130	-96 ± 130	-98 ± 131
04.2015/Col	2481	455 ± 352	96 ± 237	143 ± 234	174 ± 235	155 ± 234
04.2015/Texas	1787	117 ± 86	-18 ± 190	0.0 ± 189	14 ± 189	-1 ± 193

Table S8. As for Table S4 except for tropospheric NO with mean biases and associated standard deviations are given in ppt.

Month/region	nobs	mean observations	IFS(CBA)	IFS(MOC)	IFS(MOZ)	CAMSRA
*07.2014/ Col	251	1058 ± 414	75 ± 373	428 ± 450	513 ± 474	-21 ± 302
07.2014/ Col	11962	1991 ± 466	-745 ± 1083	-208 ± 1229	-180 ± 1252	-793 ± 986
*08.2014/Col	262	1122 ± 425	482 ± 403	878 ± 493	960 ± 484	-156 ± 295
08.2014/Col	12541	2061 ± 361	-561 ± 756	-182 ± 758	-175 ± 745	-1072 ± 765
02.2015/EC	752	174 ± 95	97 ± 150	441 ± 275	328 ± 264	89 ± 164
03.2015/EC	487	509 ± 246	55 ± 564	64 ± 544	272 ± 673	309 ± 518
03.2015/Col	3356	618 ± 542	-559 ± 202	-493 ± 189	-494 ± 191	-64 ± 201
04.2015/ND	3230	88 ± 67	-47 ± 33	-49 ± 34	-38 ± 33	-3 ± 34
04.2015/Col	2481	657 ± 460	-562 ± 179	-474 ± 184	-450 ± 186	56 ± 251
04.2015/Texas	1746	320 ± 211	-279 ± 48	-279 ± 50	-261 ± 51	-70 ± 57

Table S9. As for Table S1 except for tropospheric HNO_3 with mean biases and associated standard deviations are given in ppt