

Table S1. Number of data points averaged per altitude bin for the HOOVER experimental values.

altitude [m]	NO	O ₃	CO	HO ₂	OH	CH ₄	CH ₃ O ₂	j(NO ₂)	j(O ¹ D)	P(O ₃)
0 - 1000	5	10	10	5	4	8	4	10	10	3
1000 - 2000	11	20	15	18	17	15	14	19	19	10
2000 - 3000	3	8	4	7	6	4	3	8	7	2
3000 - 4000	5	7	5	6	5	4	3	7	7	2
4000 - 5000	4	8	5	6	6	5	4	8	8	3
5000 - 6000	7	10	9	9	8	9	8	9	9	6
6000 - 7000	10	18	12	16	14	11	10	18	18	7
7000 - 8000	20	31	27	29	28	27	26	31	31	19
8000 - 9000	48	62	47	61	46	47	45	62	62	37
9000 - 10000	21	40	30	24	31	30	15	33	33	12
10000 - 11000	7	17	7	18	18	7	7	18	18	7
11000 - 12000	3	6	3	6	6	3	3	6	6	3

Table S2. Number of data points averaged per altitude bin for the model values. The number is identical for all species and calculations.

altitude [m]	HOOVER	UTOPIHAN	BLUESKY
0 - 1000	10	19	71
1000 - 2000	20	36	50
2000 - 3000	8	11	35
3000 - 4000	7	16	24
4000 - 5000	8	9	53
5000 - 6000	10	16	32
6000 - 7000	18	16	14
7000 - 8000	31	32	21
8000 - 9000	63	68	29
9000 - 10000	40	78	52
10000 - 11000	18	32	64
11000 - 12000	6	10	42
12000 - 13000	1	9	24

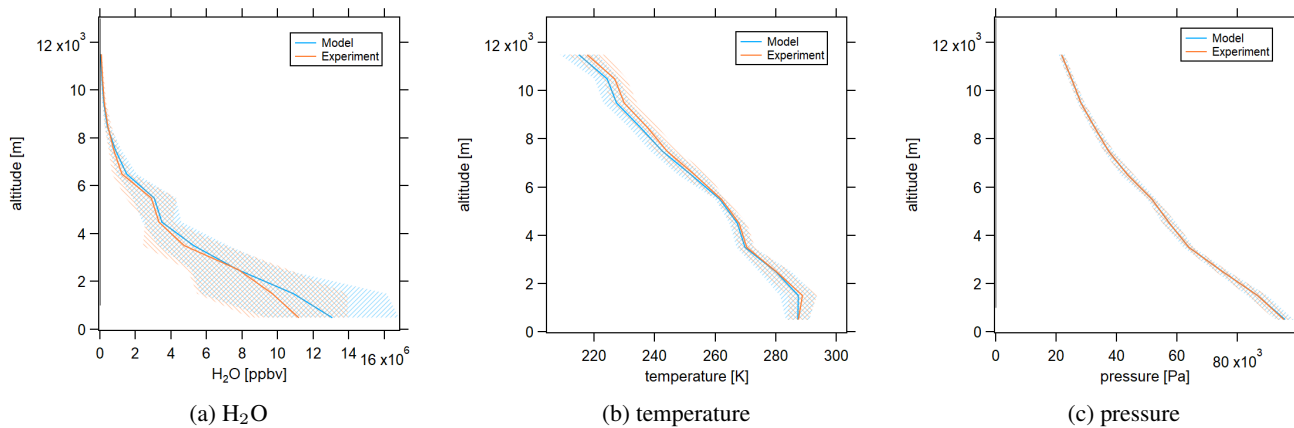


Figure S1. Vertical profiles of water, temperature and pressure for observations (orange) and model simulations (blue) during the HOOVER campaign. All three parameters show close agreement for model and experiment.

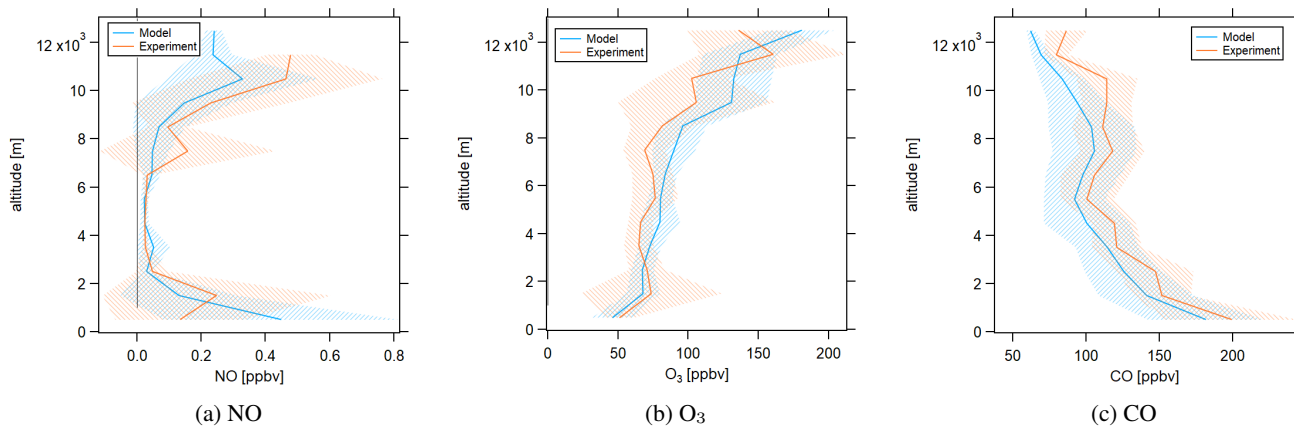


Figure S2. Vertical profiles of NO, O_3 and CO for observations (orange) and model simulations (blue) during the UTOPIHAN campaign. All three trace gases show close agreement for model and experiment.

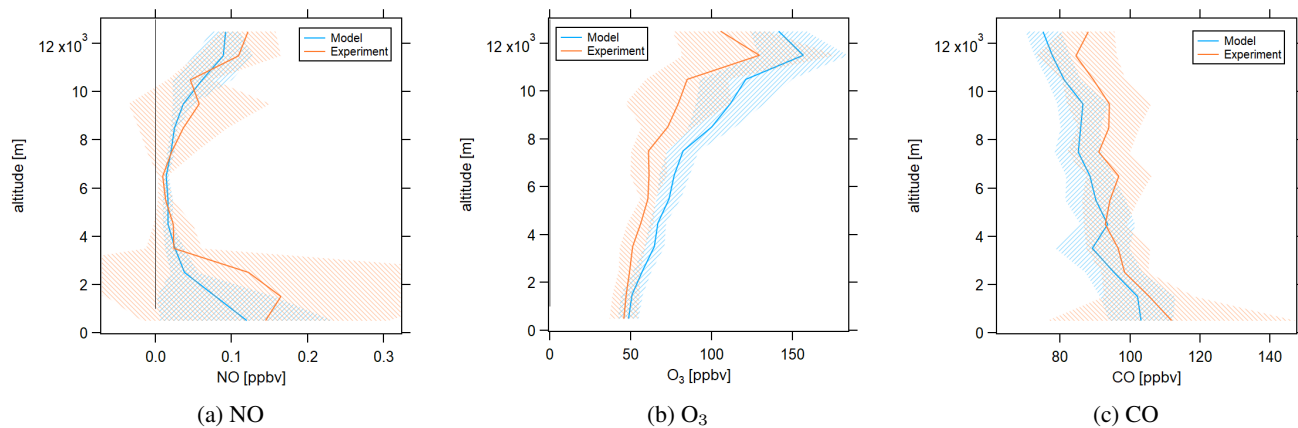


Figure S3. Vertical profiles of NO, O₃ and CO for observations (orange) and model simulations (blue) during the BLUESKY campaign. All three trace gases show close agreement for model and experiment.

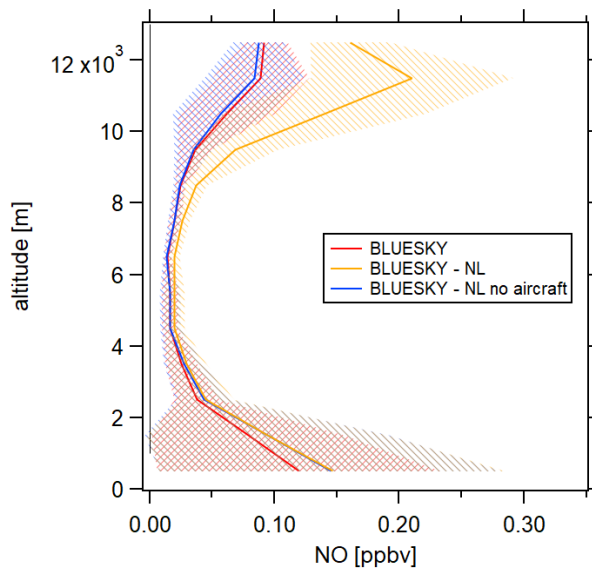


Figure S4. NO vertical profiles for BLUESKY (red), for the BLUESKY no-lockdown scenario (yellow) and for the BLUESKY no-lockdown scenario without aircraft emissions (blue) (model data). Upper tropospheric NO reductions observed for BLUESKY can be attributed to reduced air traffic during the COVID-19 lockdowns.

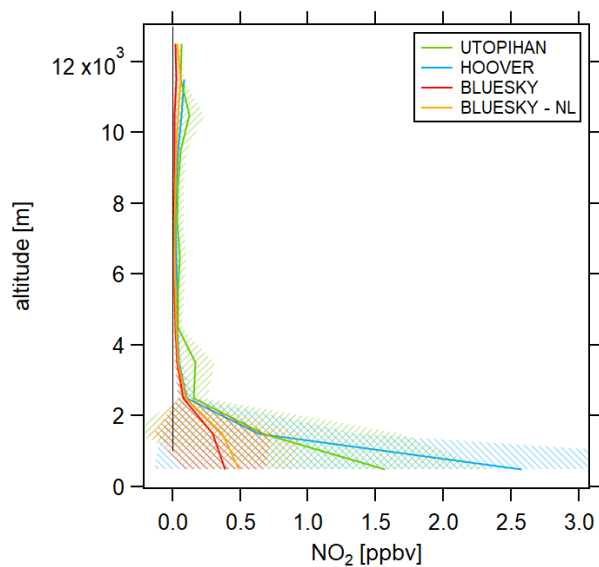


Figure S5. NO_2 vertical profiles during UTOPIHAN, HOOVER, BLUESKY with and without lockdown scenario.

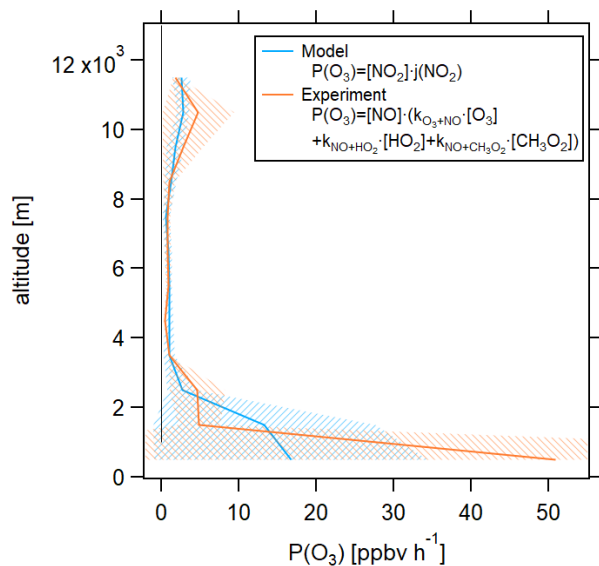


Figure S6. Modeled and experimental vertical profiles of $\text{P}(\text{O}_3)$ for HOOVER. Modeled $\text{P}(\text{O}_3)$ was calculated via NO_2 photolysis and experimental $\text{P}(\text{O}_3)$ was calculated via the extended Leighton ratio as shown in Equation (2).

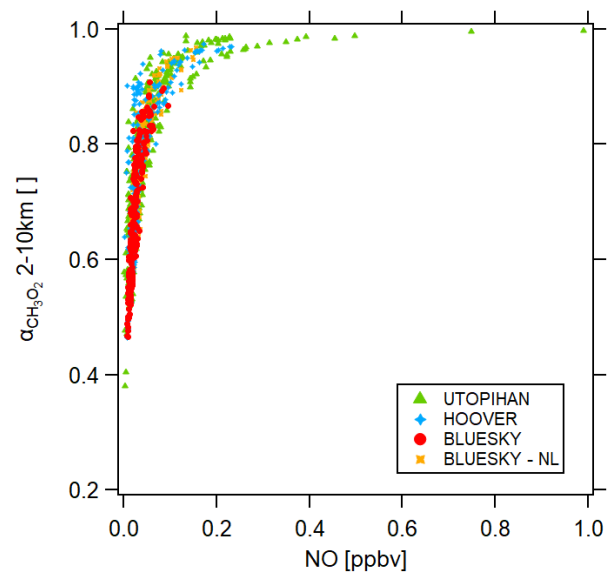
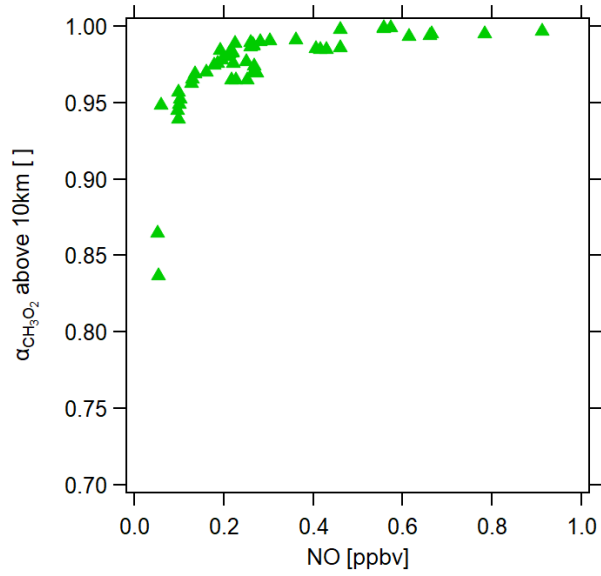
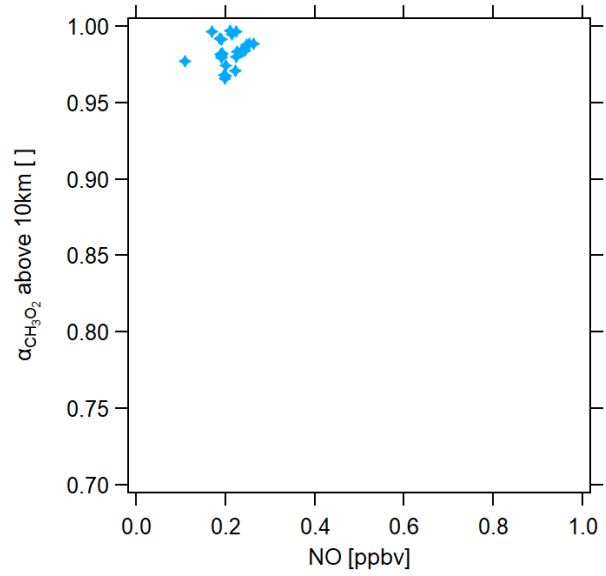


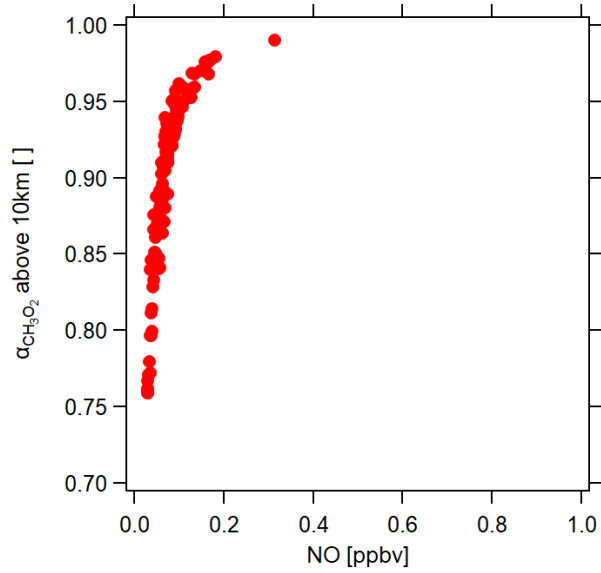
Figure S7. $\alpha_{CH_3O_2}$ for the campaigns UTOPIHAN (green), HOOVER (blue), BLUESKY (red) and the no-lockdown (NL) scenario (yellow) in correlation with NO between 2 and 10 km altitude.



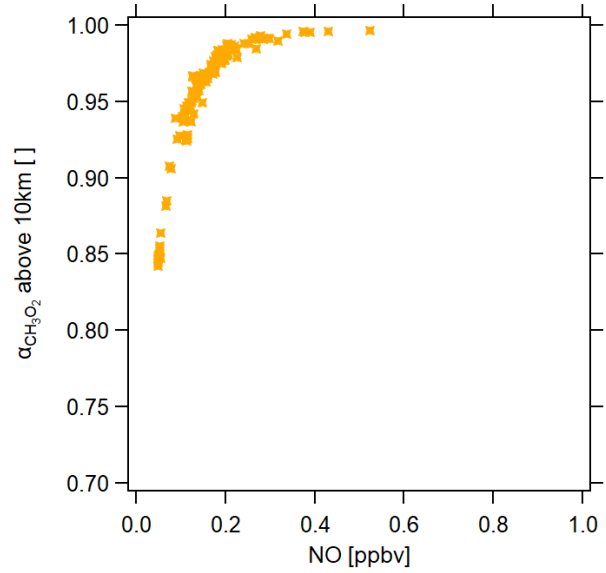
(a) UTOPIHAN



(b) HOOVER



(c) BLUESKY



(d) BLUESKY - NL

Figure S8. $\alpha_{CH_3O_2}$ correlated with NO mixing ratios for all campaigns as shown in Figure 5c of the manuscript in individual panels.