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A nitrate photolysis source of tropospheric HONO is incompatible with current understanding of atmospheric chemistry

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Supplementary information

Table S1. Summary of key terms for atmospheric nitrogen inthe three simulations.

	No pNO_3^- + hv	Shah et al. 2023	This study
Emissions (TgN yr^{-1})			
Total NOx	53.98	53.80	53.46
Anthropogenic NO_x	30.86	30.86	30.86
Soil NO_x	7.52	7.52	7.52
Biomass Burning NO_x	6.67	6.67	6.67
Lightning NO_x	6.13	6.13	6.13
Shipping NO_x	2.80	2.62	2.32
Shipping HNO_3	3.09	3.26	2.21
Deposition (TgN yr^{-1})			
HNO_3	35.20	37.62	36.09
pNO_3	15.37	12.53	12.71
PAN	1.26	1.38	2.32
NO_2	1.73	1.76	1.67
XNO2 / XNO3	0.29	0.33	0.38
HONO	0.07	0.14	0.20
Photolysis Reactions (s^{-1})			
pNO_3 + hv → HONO	0.00	7.96	32.89
pNO_3 + hv → NO_2	0.00	3.98	16.44
HONO + hv → OH + NO	16.90	27.89	68.05
HNO_3 + hv → OH + NO_2	2.07	2.14	1.81
Gas-Phase Reactions (TgN yr^{-1})			
OH + NO → HONO	17.05	20.32	36.20
OH + NO_2 → HNO3	27.13	34.49	59.73
NO_2 + MCO_3 → PAN	117.57	133.84	150.17
PAN → NO_2 + MCO_3	114.69	130.67	145.21
HNO_3 + OH → NO_3	3.44	3.85	3.84
HNO_2 + OH → H_2O + NO_2	0.19	0.37	0.98
O_3 + NO_2 → NO_3 + O_2	41.53	49.07	67.31
Heterogeneous Reactions (TgN yr^{-1})			
N_2O_5 (+ H_2O) → HNO_3	10.08	11.52	16.17
NO_3 → HNO_3	2.59	3.08	3.59
NO_2 → HNO_3	0.98.	1.06	1.07
Tropospheric Burdens (Tg X)			
O_3	309	332	393
NO_x	0.36	0.43	0.57
HNO_3	1.21		1.23
pNO_3^-	0.43	0.38	0.30
HONO (Gg)	1.64		6.21
CO	326	306	271

Figure S1. Location and value in ppt of all available HONO measurements, from aircraft and ship campaigns (coloured by HONO concentration in pptv) and in-situ measurements (red markers).

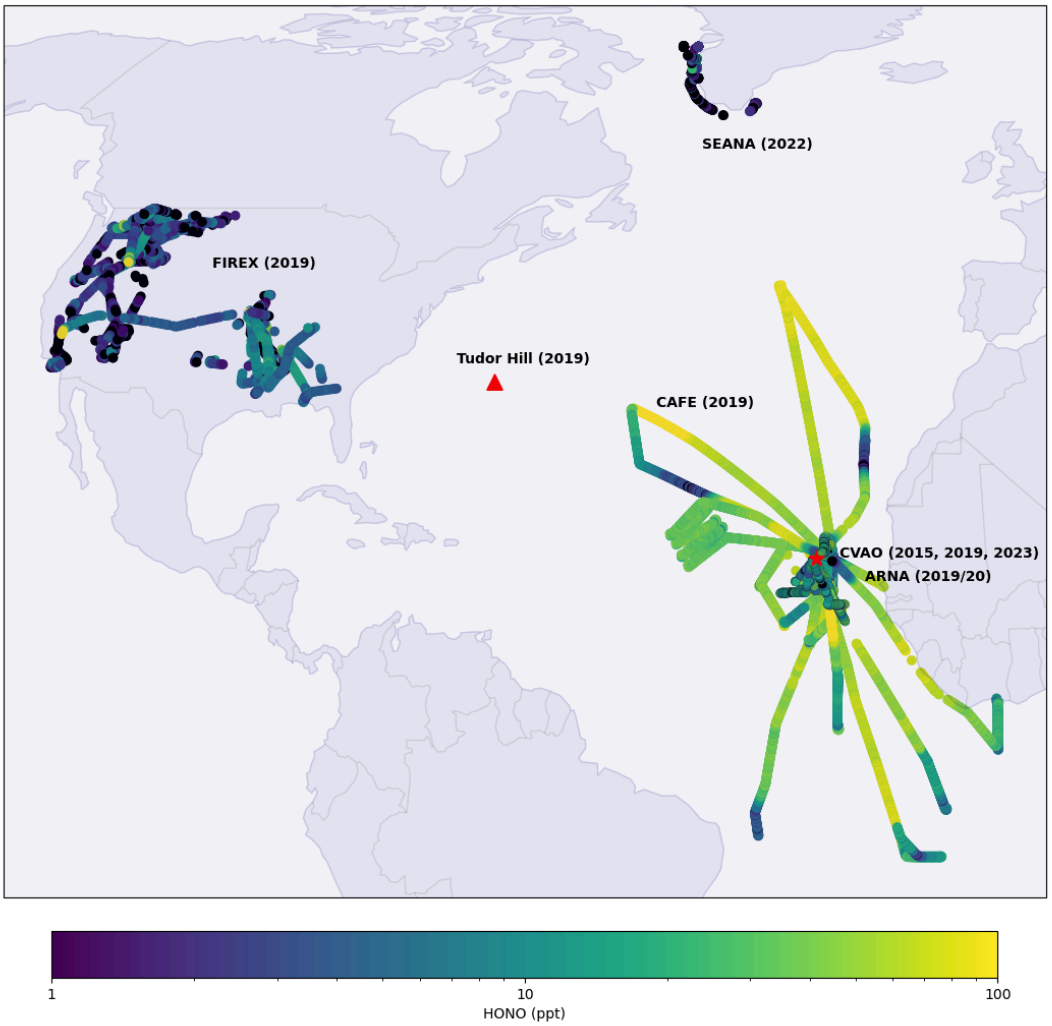


Figure S2. Nitrate aerosol photolysis enhancement factor necessary to balance the observed HONO against its photolytic loss as a function of the nitrate aerosol concentration, including parameterisations generated with additional dependencies on RH and Cl (see Methods).

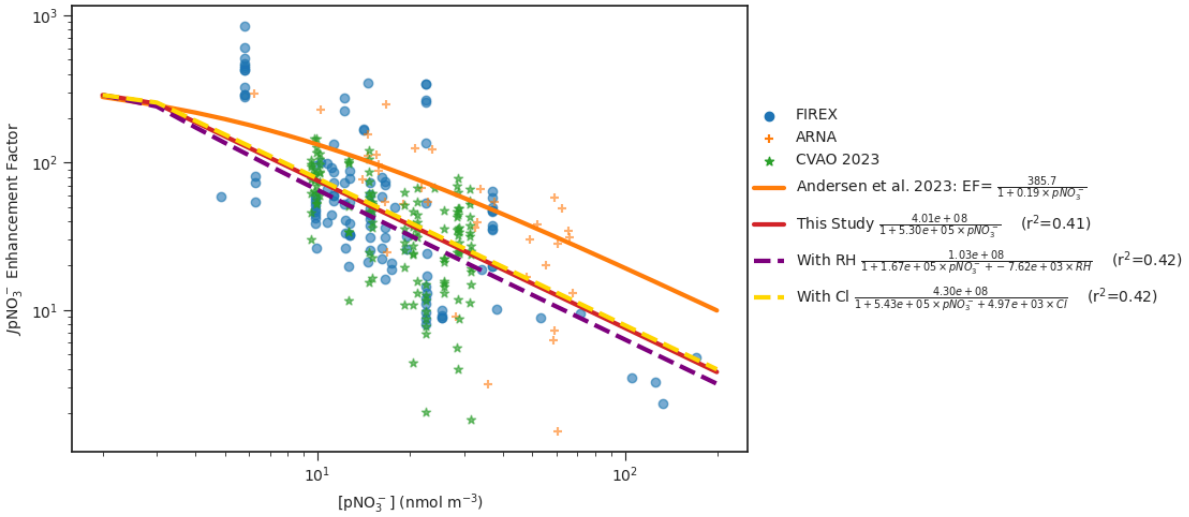


Figure S3. Comparison of FIREX-AQ AMS fine-mode nitrate (black) with simulated fine-mode nitrate from the base model version without nitrate photolysis (blue). Data is split into Western and Eastern USA (at -115°W), and averaged over 500m altitude bins. Horizontal grey lines represent 1 standard deviation on the measurements.

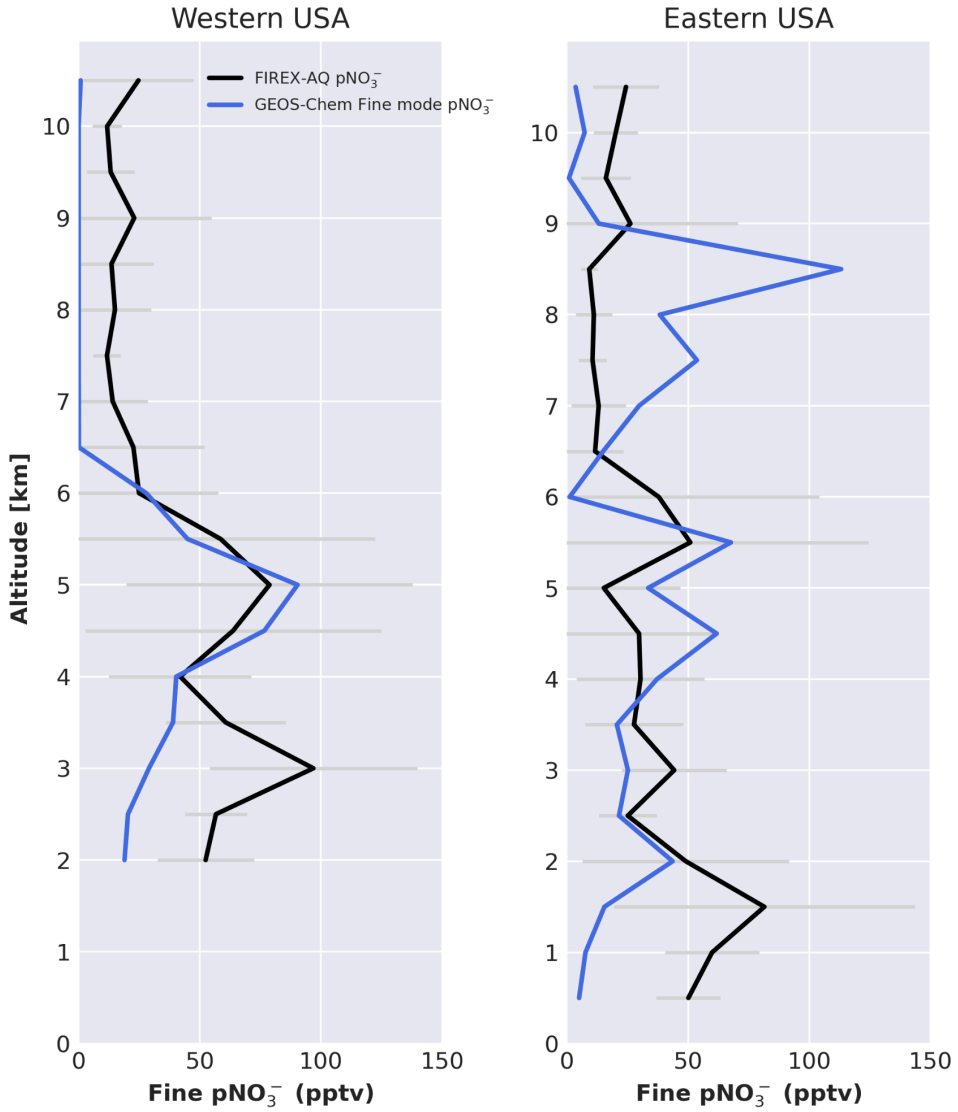
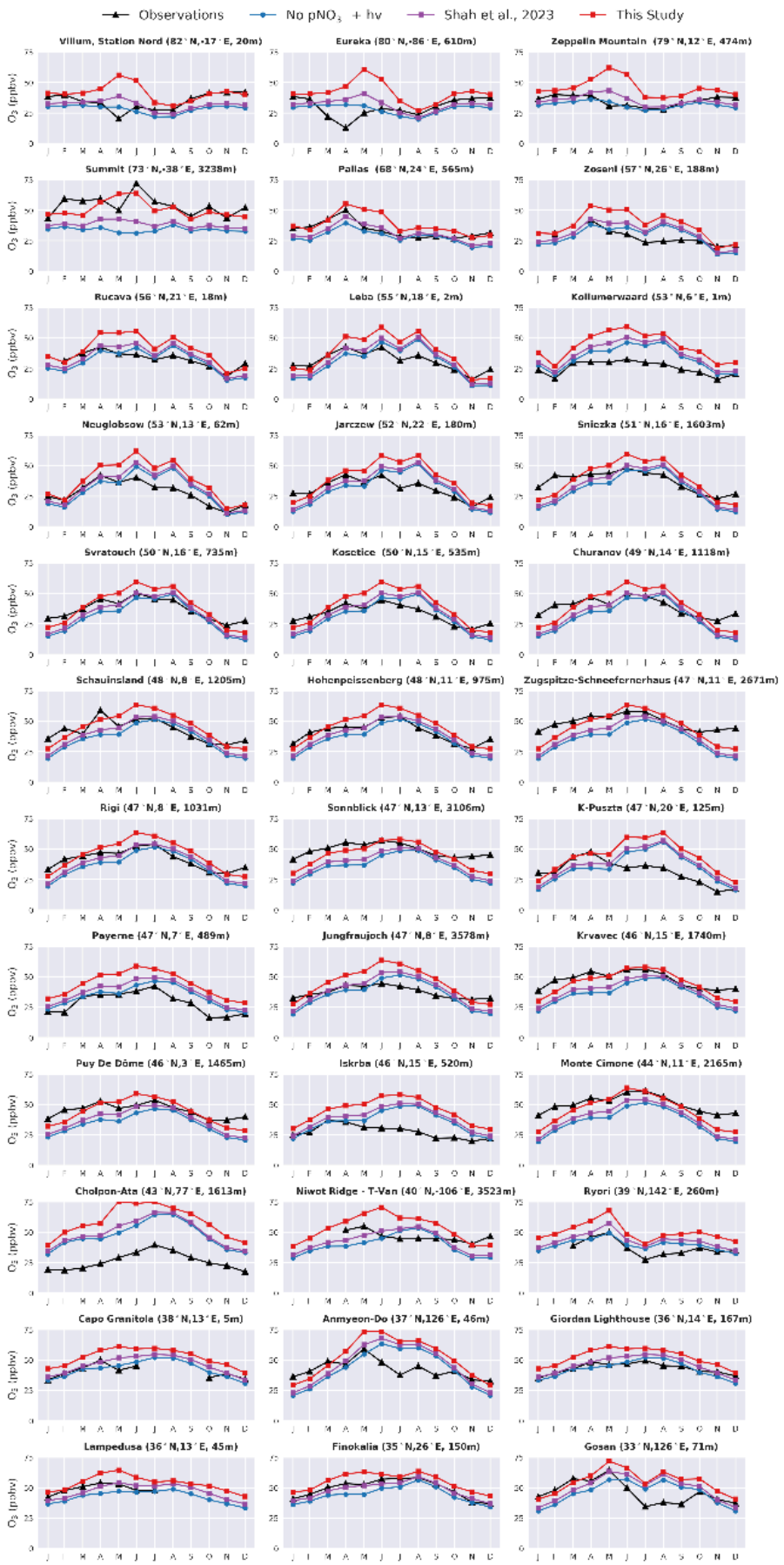
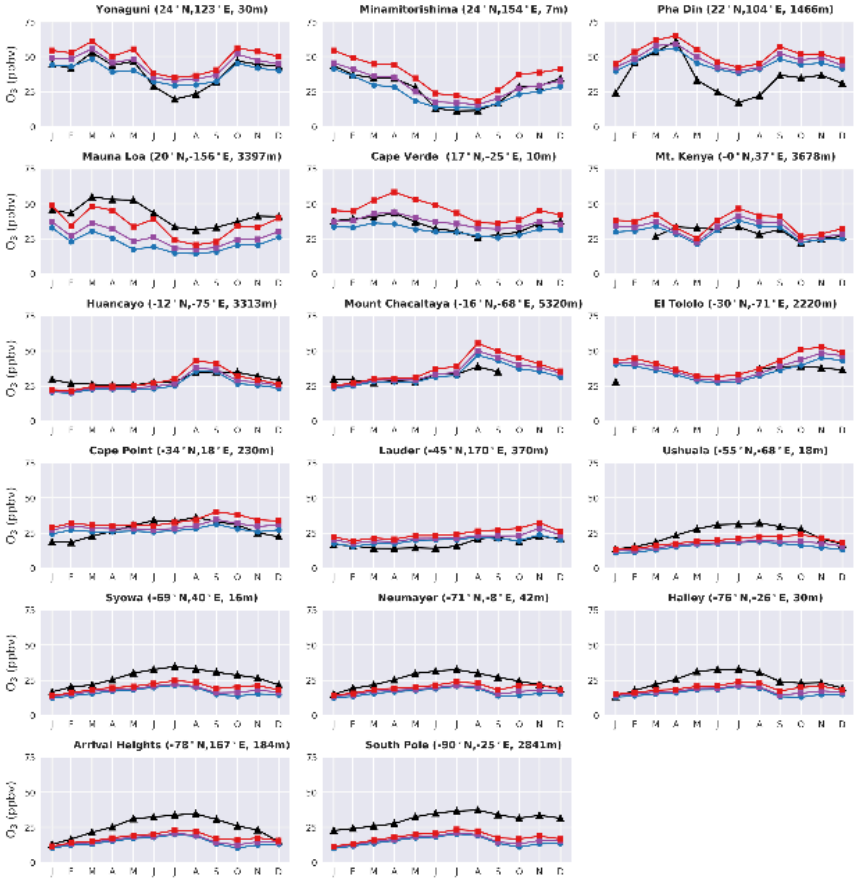


Figure S4. Monthly mean O_3 from Global Atmospheric Watch (GAW) sites (black) compared with simulated O_3 from the 3 simulations.




Yonaguni (24°N,123°E, 30m)
Minamitorishima (24°N,154°E, 7m)
Pha Din (22°N,104°E, 1466m)
Mauna Loa (20°N,-156°E, 3397m)
Cape Verde (17°N,-25°E, 10m)
Mt. Kenya (-0°N,37°E, 3678m)
Huancayo (-12°N,-75°E, 3313m)
Mount Chacaltaya (-16°N,-68°E, 5320m)
El Tololo (-30°N,-71°E, 2220m)
Cape Point (-34°N,18°E, 230m)
Lauder (-45°N,170°E, 370m)
Ushuaia (-55°N,-68°E, 18m)
Syowa (-69°N,40°E, 16m)
Neumayer (-71°N,-8°E, 42m)
Halley (-76°N,-26°E, 30m)
Arrival Heights (-78°N,167°E, 184m)
South Pole (-90°N,-25°E, 2841m)
O3 (ppbv)
J F M A M J J A S O N D