

Supplements

Table S 1: Lists of EANET stations grouped by their countries with ID number as in Error! Reference source not found..

Country	Station (ID number)
Cambodia	PhnomPenh (31)
China	Jinyunshan (48), Hongwen (14)
Indonesia	Jakarta (18), Serpong (36), Bandung (1)
Japan	Rishiri (33), Ochiishi (26), Tappi (38), Sado-seki (34), Happo (10), Ijira (15), Oki (28), Banryu (3), Yusuhara (45), Hedo (11), Ogasawara (27), Tokyo (40)
Lao	Vientiane (42)
Malaysia	Petaling Jaya (30), Tanah Rata (37), Danum Valley (7)
Mongolia	Ulaanbaatar (41), Terelj (39)
Myanmar	Yangon (43), Mandalay (50)
Philippines	Metro Manila (22), Mt. Sto. Tomas (24)
Republic of Korea	Kanghwa (19), Cheju (Kosan) (5), Imsil (16)
Russia	Mondy (23), Listvyanka (21), Irkutsk (17), Primorskaya (32)
Thailand	Bangkok (2), Samutprakarn (47), Pathumthani (29), Khanchanaburi (20), Chiang Mai (6), Sakaerat (35), Nai Mueang (25), Chang Phueak (46), Si Phum (49)
Vietnam	Hanoi (8), Hanoi (Relocated) (9), Hoa Binh (13), Can Tho (4), Ho Chi Minh (12), Yen Bai (44)

5 Table S 2: Model comparison with ATom1 flights, calculated for all flight, and for North Pacific (NP) region: no outlier detection is applied. N is number of available data for each calculation, R is the correlation coefficients. R and bias of the STD run are shown as bold if better than that of the OLD run. Unit of bias is ppt for NO₂, OH, ppb for O₃, CO.

	NO ₂	NO ₂ (NP)	O ₃	O ₃ (NP)	OH	OH (NP)	CO	CO (NP)
<i>N</i>	29,509	2,283	29,204	2,246	7,601	608	27,467	2,172
<i>R</i> (STD)	0.730	0.621	0.751	0.609	0.579	0.407	0.659	0.596
<i>R</i> (OLD)	0.697	0.306	0.752	0.598	0.584	0.374	0.643	0.596
bias (STD)	-11.277	0.588	11.637	8.471	-0.038	-0.003	1.698	-1.713
bias (OLD)	-6.940	4.450	15.025	13.050	-0.015	0.015	-7.521	-12.393

10 **Table S 3: Tables of correlation coefficient (*R*) and model biases against EMeRGe measurements for HONO.**

“Alt.” columns show altitude ranges (± 500 m). “*N*” column show the numbers of hourly-averaged values calculated for each altitude range. Left table: darker colours represent higher absolute values of *R* (closer to ± 1). Right table: lighter colours show smaller model biases (closer to 0). The darkness of blues (negative values) and reds (positive values) are scaled to ± 1 for *R* and \pm maximum values of each row for biases. Unit of biases is ppt for HONO and NO₂, ppb for O₃ and CO.

<i>R</i>(HONO)									Bias(HONO)							
Alt.	<i>N</i>	STD	GRx8	EMx8	AIRC	maxST	ratR4	ratR4+CLD	Alt.	STD	GRx8	EMx8	AIRC	maxST	ratR4	ratR4+CLD
0	970	-0.23	-0.39	-0.29	-0.27	-0.17	-0.22	-0.21	0	-112.46	-94.09	-102.65	-112.18	-70.32	-106.14	-102.87
1000	1714	0.49	0.36	0.51	0.44	0.56	0.24	0.24	1000	-105.31	-95.51	-94.24	-105.61	-71.70	-99.76	-96.14
2000	1538	0.31	0.47	0.38	0.36	0.47	0.12	0.07	2000	-64.10	-62.86	-64.11	-64.36	-61.76	-63.33	-62.79
3000	2296	0.16	0.05	0.11	0.11	-0.03	0.13	0.05	3000	-44.17	-42.81	-44.09	-44.22	-43.21	-43.89	-43.71
4000	192	-0.17	-0.24	-0.08	-0.11	0.28	-0.11	-0.04	4000	-25.95	-24.25	-25.84	-25.99	-25.61	-25.67	-25.44
5000	836	0.04	0.03	0.14	0.21	0.53	0.19	0.75	5000	-18.94	-17.31	-18.80	-18.97	-18.49	-18.77	-18.34
6000	506	-0.01	0.02	-0.03	0.03	0.11	-0.03	0.05	6000	-4.99	-2.87	-4.64	-5.07	-4.92	-4.78	-4.76
7000	76	-0.31	-0.33	-0.31	-0.33	-0.30	-0.30	-0.30	7000	-4.07	0.70	-3.45	-4.26	-4.12	-3.77	-3.65
8000	44	-0.67	-0.64	-0.64	-0.64	-0.64	-0.68	-0.67	8000	-2.83	2.90	-1.93	-2.72	-2.72	-2.51	-2.50

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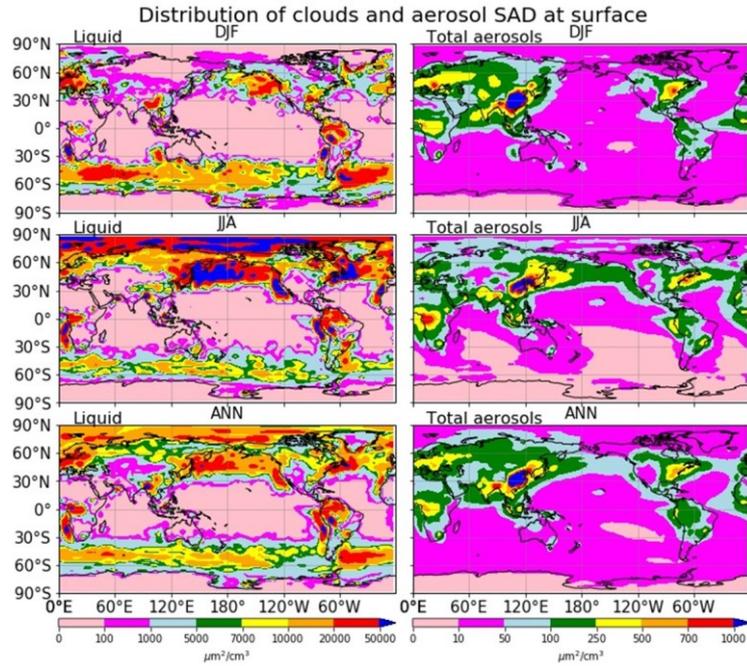


Figure S 1: Seasonal and annual mean distributions of cloud droplet (left) and total aerosols SAD (right).

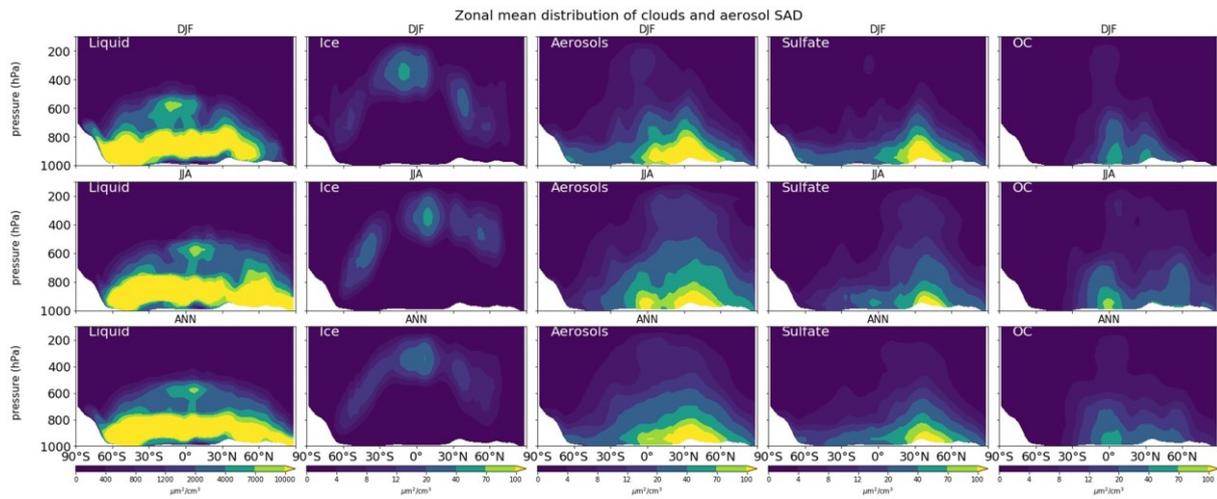
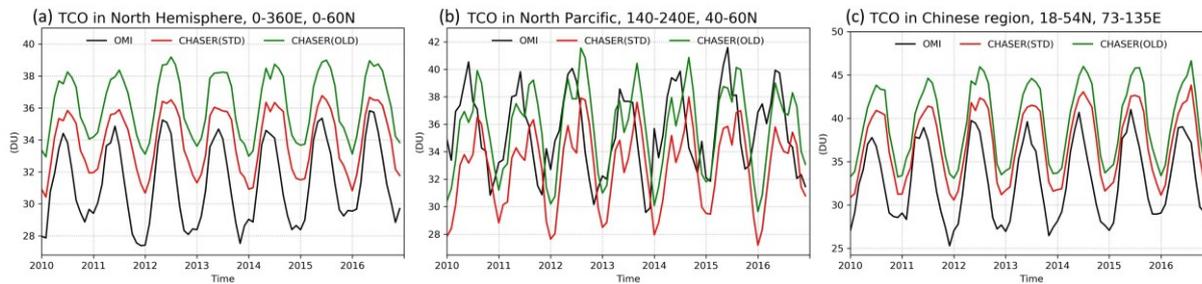
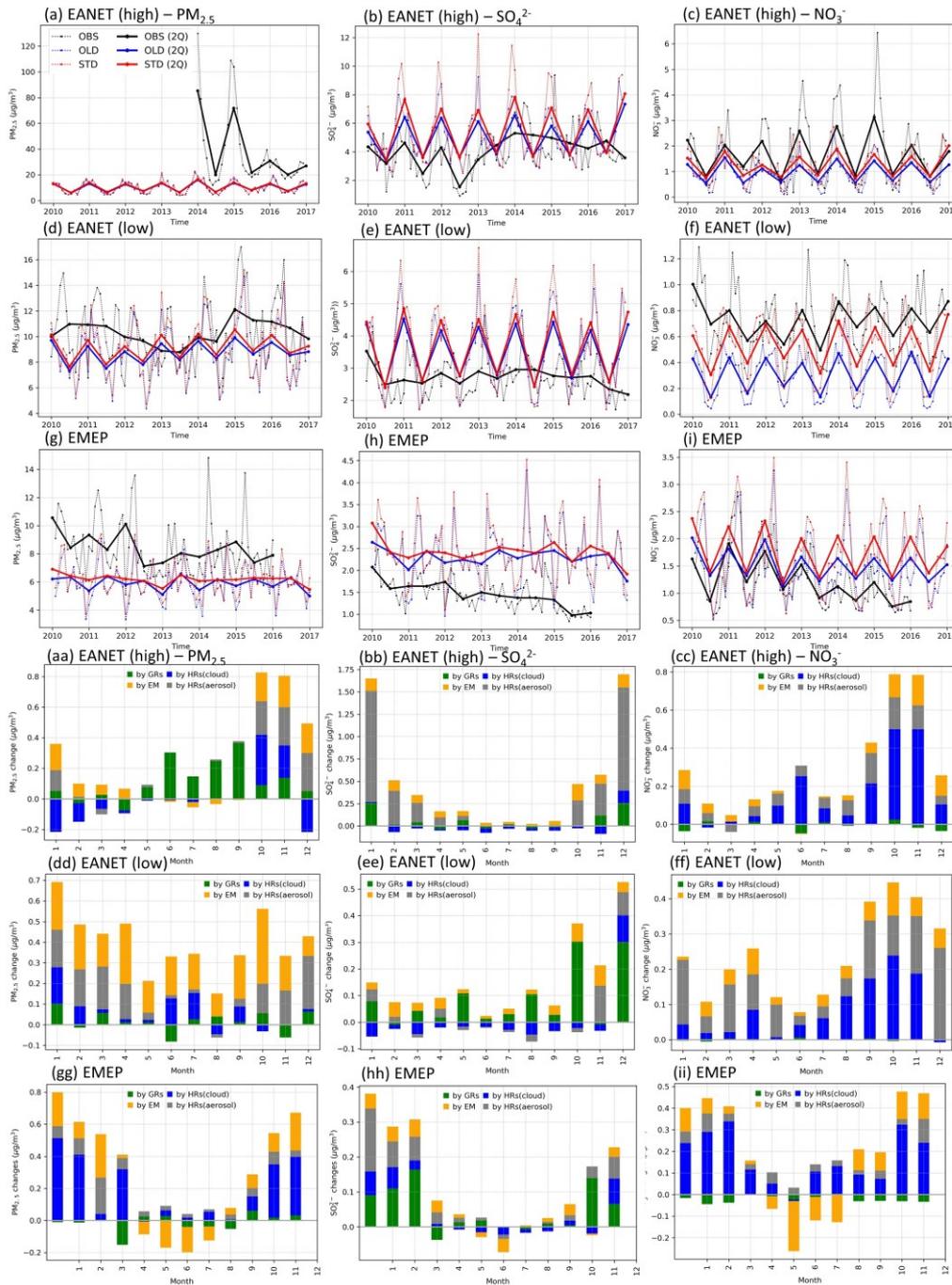


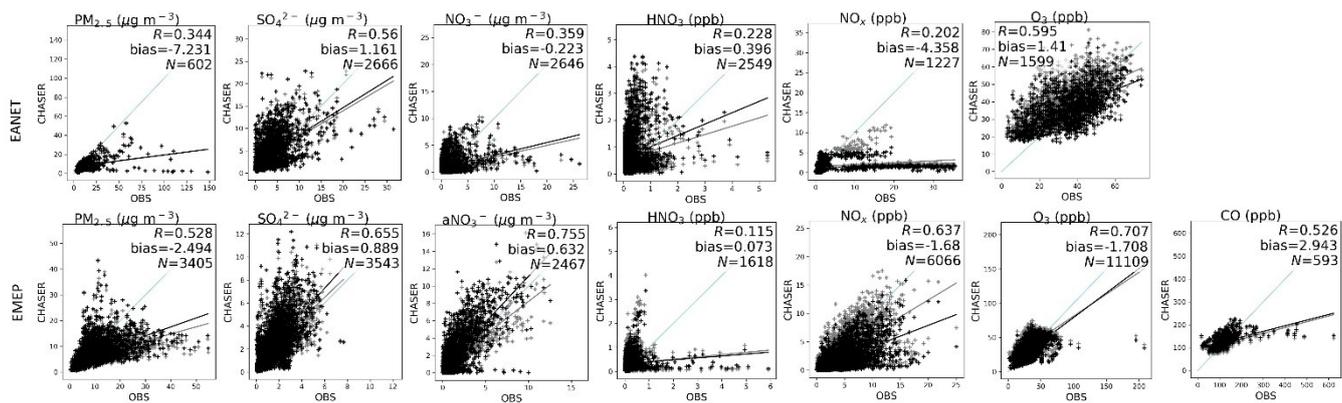
Figure S 2: Zonal, seasonal mean (upper and middle panels), and annual mean (lower panels) distribution of cloud droplet surface aerosol density (SAD), cloud ice SAD, total aerosol SAD, sulfate aerosol SAD, and organic carbon SAD (from left to right).



25 **Figure S 3: Verifications with OMI satellite data for tropospheric column ozone (TCO). TCO (DU) by OMI (black) and CHASER (red: STD case; green: OLD case) in the Northern Hemisphere (a), NP (b), and Chinese (c) regions are plotted.**



30 **Figure S 4: Observed and simulated mass concentrations (a-i) and monthly-mean changes (aa-ii) for $PM_{2.5}$, SO_4^{2-} , and NO_3^- for EANET and EMEP stations, grouped as high- NO_x EANET, low- NO_x EANET, and all EMEP stations. In (a-j), black lines: observation; red: STD case; blue: OLD case. Dotted lines are all stations' median from monthly-mean for each station in that group. Thick solid lines represent two quarters averaged from dotted lines. In (aa-jj), green bars: monthly changes by GRs; blue: by HRs on clouds; grey: by HRs on aerosols; orange: by EM.**



35 **Figure S 5: Correlations of STD and OLD runs with EANET (upper) and EMEP (lower) stations for $PM_{2.5}$, SO_4^{2-} , NO_3^- , HNO_3 , NO_x , O_3 , and CO (CO for EMEP only). Fitting lines for STD (black) and OLD (grey) with observations are also plotted. N (no unit) is the number of available data after outlier filtering, which is similar as described in Error! Reference source not found.**

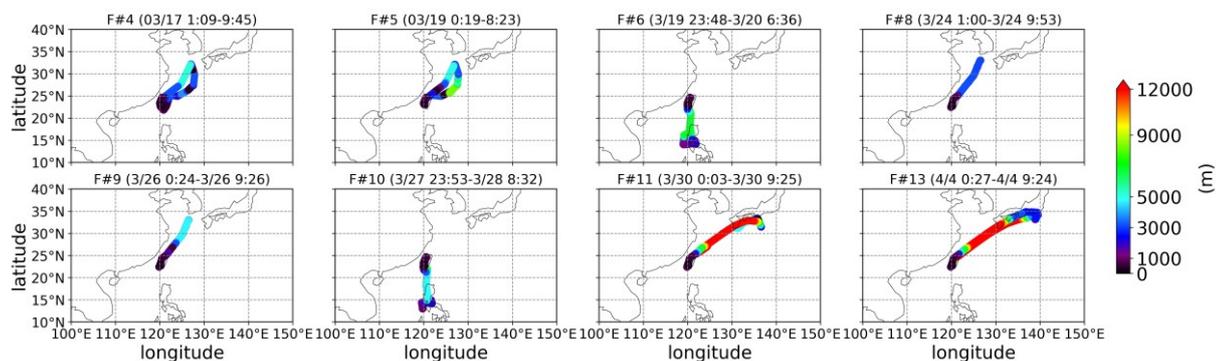


Figure S 6: Cruising altitudes in EMERGe-Asia 2018 campaign.

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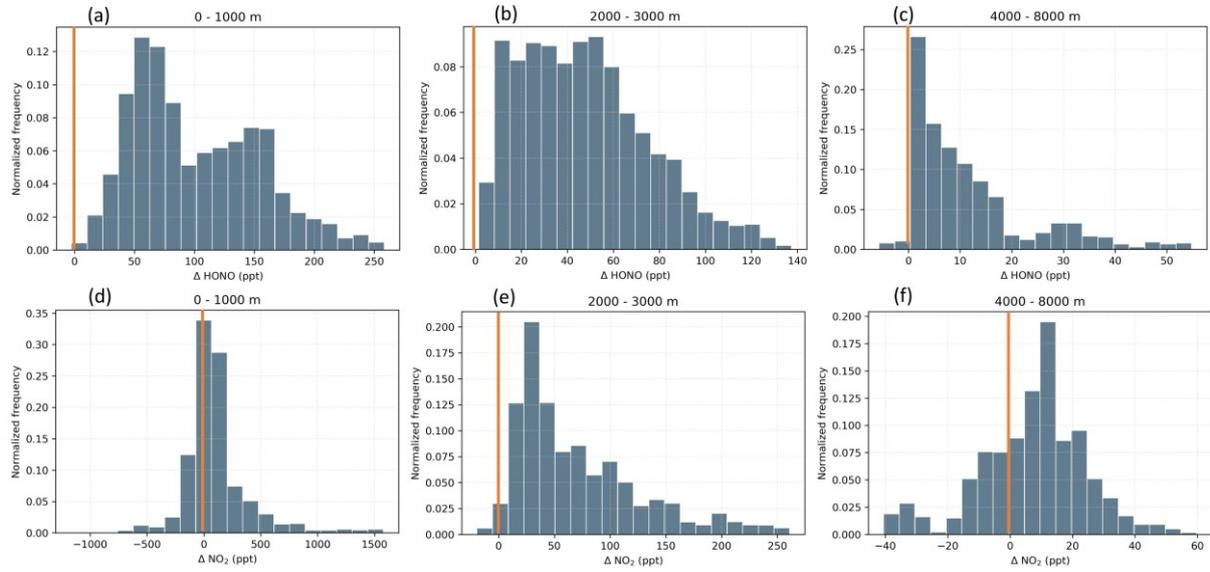
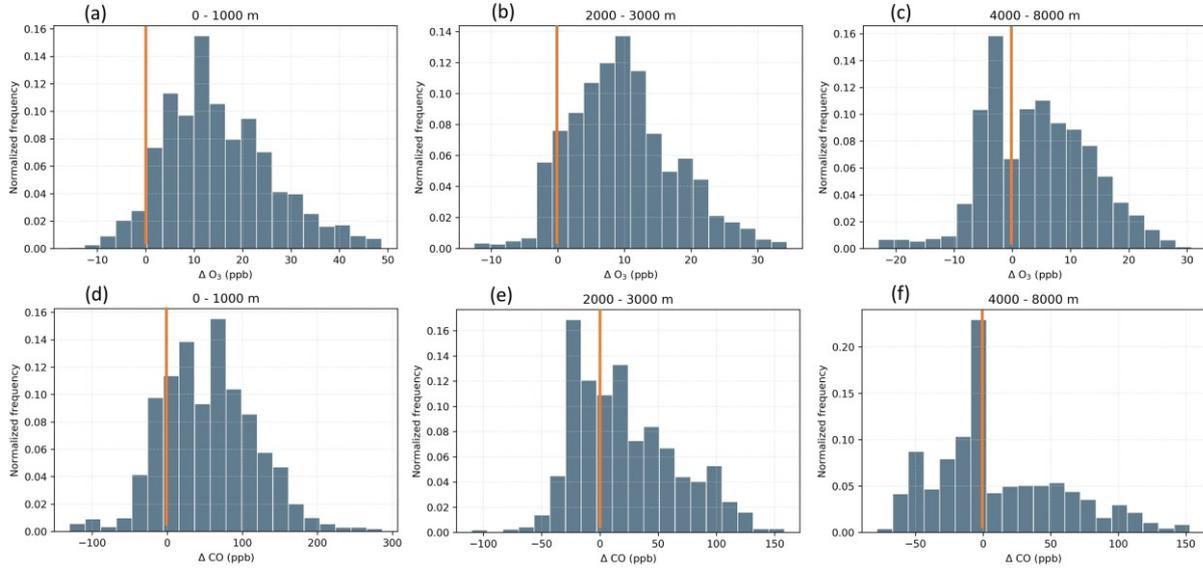
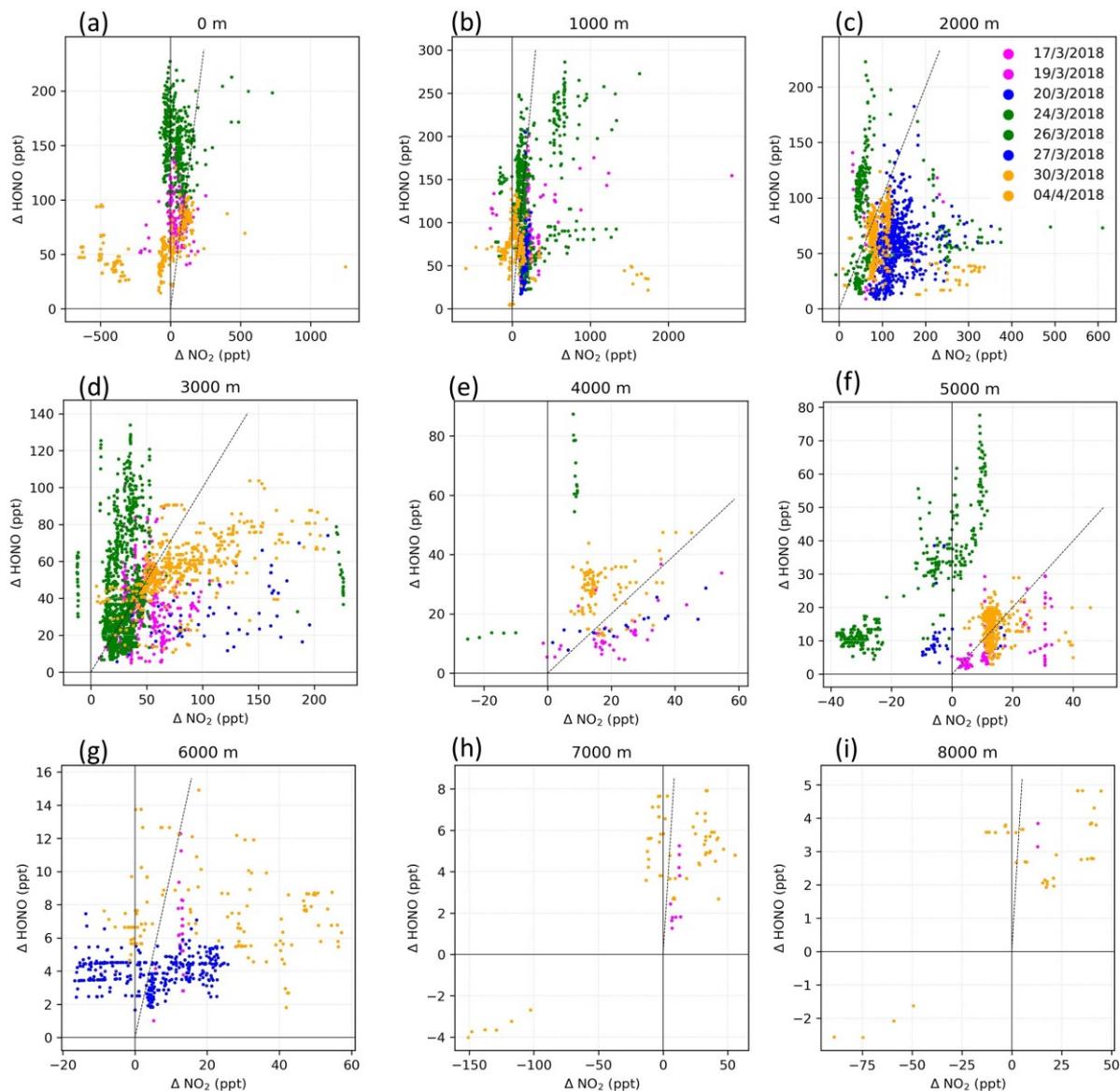


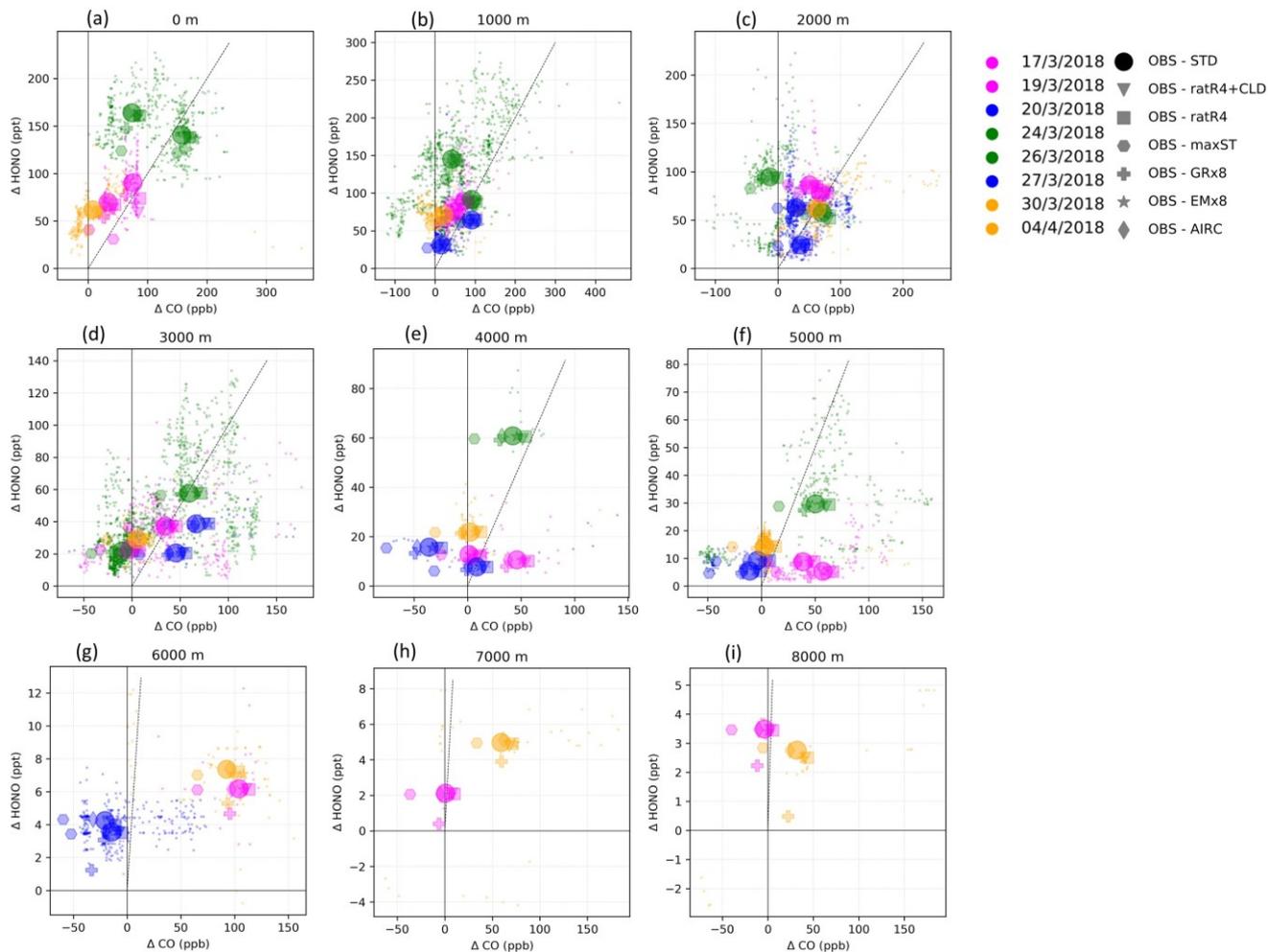
Figure S 7: Normalized distributions of the differences between measured and simulated data (OBS – STD) for HONO (upper panel) and NO₂ (lower panel). Data are separated into three categories (± 500 m): 0 – 1000 m (a-d), 2000 – 3000 m (b-e), 4000 – 8000 m (c-f). Three-sigma-rule outlier detection is applied for each altitude range before grouped.



45 Figure S 8: Normalized distributions of the differences between measured and simulated data (OBS – STD) for O₃ (upper panels) and CO (lower panels). Data are separated into three categories (± 500 m): 0 – 1000 m (a-d), 2000 – 3000 m (b-e), 4000 – 8000 m (c-f). Outliers of each altitude group are filtered by 3-order rule.



50 **Figure S 9: Differences of measured data from simulated data (OBS – STD) for HONO and NO₂ in ranges of altitudes (0 – 8000 m ±500 m). Colours represent each flight cruise, i.e. Taiwan – South Korea – Taiwan on 17/3 and 19/3 (magenta), Taiwan – South Korea on 24/3 and 26/3 (green), Taiwan – Philippines on 20/3 and 27/3 (blue), and Taiwan – Japan on 30/3 and 4/4 (orange). Vertical, horizontal, and diagonal lines indicate $\Delta\text{NO}_2 = 0$, $\Delta\text{HONO} = 0$, $\Delta\text{NO}_2 = \Delta\text{HONO}$, respectively.**



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Figure S 10: Differences of measured and simulated data for HONO and CO at different altitudes (± 500 m). Colours represent each flight cruise, similar to Figure S 9. All values of (OBS – STD) are illustrated as small dots. Median values of differences are plotted for (OBS – STD) as circles, (OBS – ratR4+CLD) as triangles, (OBS – ratR4) as squares, (OBS – maxST) as hexagones, (OBS – GRx8) plus, (OBS – EMx8) as stars, (OBS – AIRC) as diamonds. Vertical, horizontal, and diagonal lines indicate $\Delta \text{NO}_2 = 0$, $\Delta \text{HONO} = 0$, $\Delta \text{NO}_2 = 10^3 \Delta \text{HONO}$, respectively.

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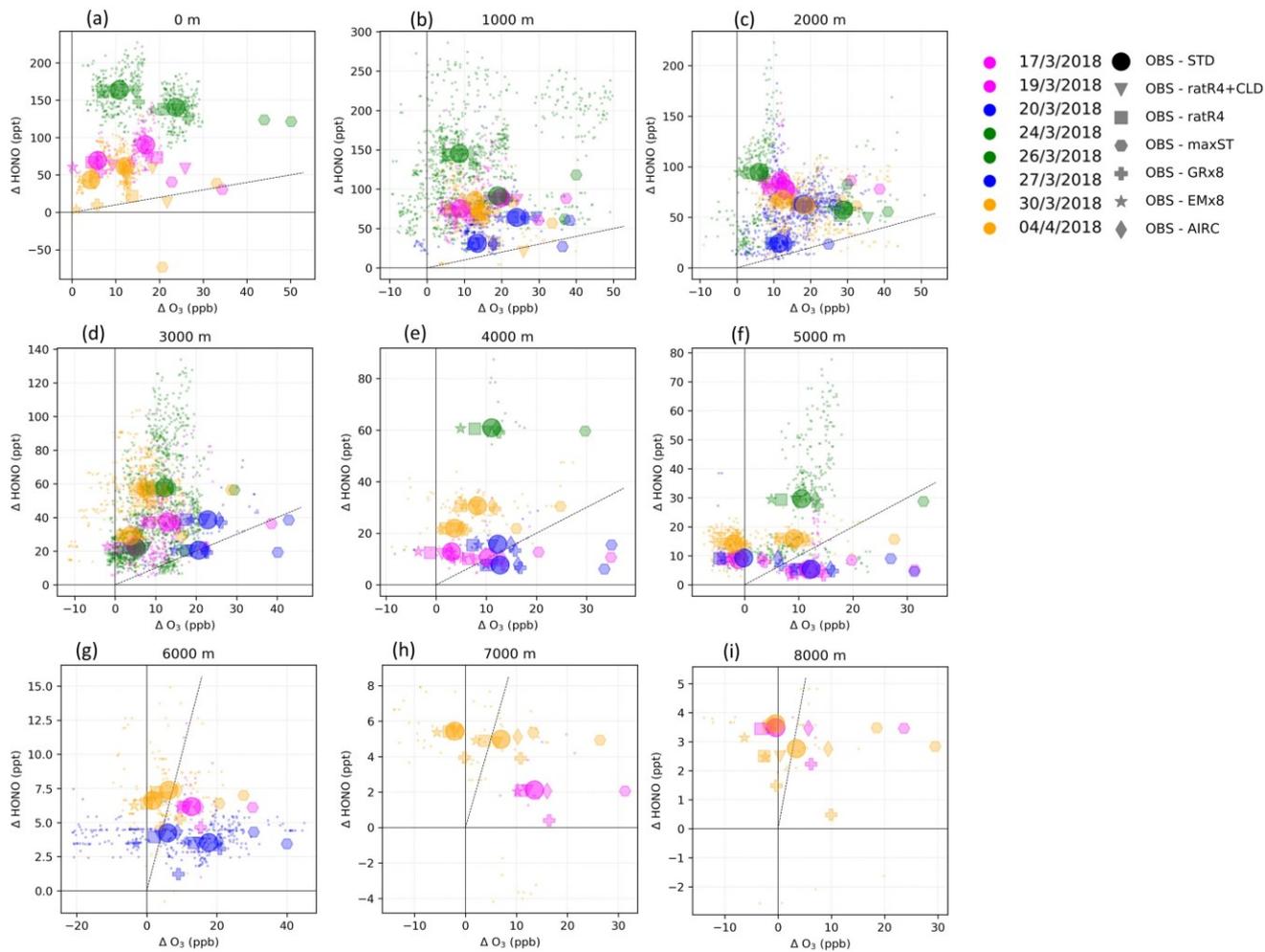
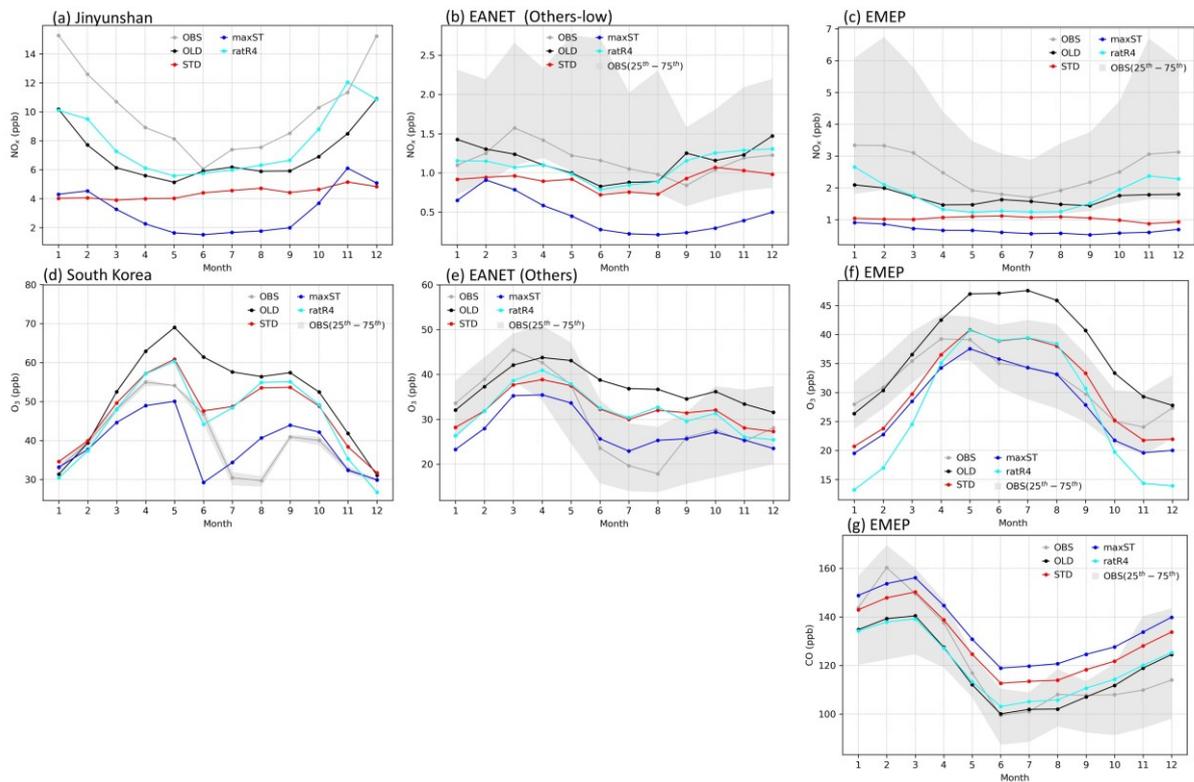
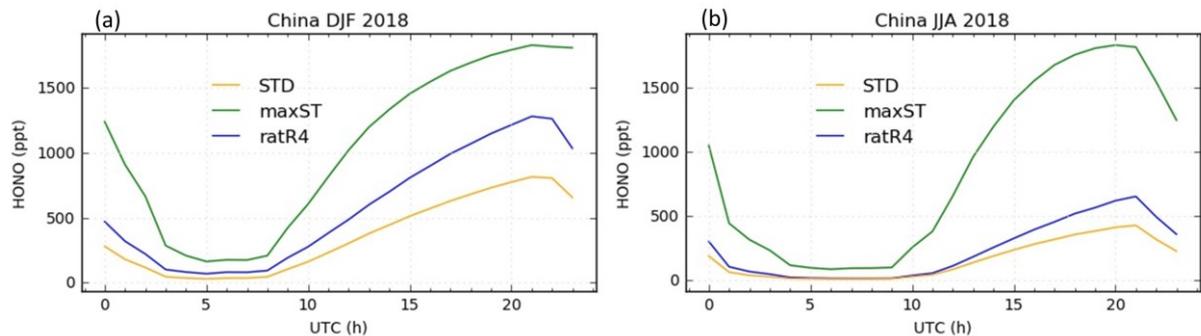


Figure S 11: Similar to Figure S 10 but for HONO and O₃.



65 **Figure S 12: Monthly variance for NO_x (first row), O_3 (second row), and CO (last row) for EANET and EMEP stations: (a) Jinyunshan (China), (b) other EANET stations except some high- NO_x stations (see text), (d) South-Korean stations (Kanghwa, Imsil, Cheju), (e) other EANET stations, (c, f, g) all EMEP stations. Grey lines with shades show observation and 25th – 75th range; black lines show OLD case; red lines show STD; blue lines show maxST; cyan lines show ratR4.**



70 **Figure S 13: Diurnal variance of HONO for the Chinese region in December, January, February (a), and June, July, August (b). Orange lines show STD run; green lines show maxST run; blue lines show ratR4 run.**