Supplements

Table S1. Information on seven sub-air masses in the CHN case during E-AS-08 and E-AS-09. Bold and plain numbers in the fifth – ninth columns indicate data from observations and simulations.

Segments (CHN case)	**Flight altitudes**	**Periods**	*N*	**BC/CO Obs. Mod.**	**CO/CO_2 Obs.**	**BC/CO_2 Obs.**	BC Mean(Max.) **Obs.** Mod.	Mean(Max.)-baseline-ΔCO **Obs.** **M**od.
Units	*km*	*UTC (s)*		*ng m^{-3} ppb^{-1}*	*ppb ppm^{-1}*	*ng m^{-3} ppm^{-1}*	*μg m^{-3}*	*ppbv*
E-AS-08 S1	0.3	022715-024615	77	**4.7** 8.3	**16.5**	**77**	**0.76 (1.01)** 1.79 (1.87)	**320 (360)- 159- 161** 250 (261)--
E-AS-08 S2	1	024746-030946	89	**3.9** 13.9	**22.6**	**82**	**0.45 (0.73)** 1.22 (1.29)	**268 (303)- 153- 115** 191 (198)- 104- 87
E-AS-08 S3	1	042501-044516	82	**3.9** 12.6	**18.0**	**48**	**0.62 (0.94)** 2.00 (2.12)	**314 (387)- 152- 162** 263 (274)- 104- 159
E-AS-08 S4	0.3	044716-053746	203	**2.6** 11.5	**21.6**	**60**	**1.05 (1.55)** 2.05 (2.60)	**367 (537)--** 278 (320)- 101- 178
E-AS-08 S5	1	053746-060701	113	**2.4** 12.4	**19.2**	**24**	**1.11 (1.66)** 1.76 (2.10)	**433 (628)--** 239 (264)- 97- 142
E-AS-09 S1	1	025800-032415	99	**2.7** 10.7	**16.0**	**39**	**0.64 (0.88)** 1.45 (1.53)	**309 (349)- 75- 234** 219 (226)- 84- 135
E-AS-09 S2	0.3	032645-041945	213	**3.8** 12.6	**24.3**	**98**	**0.89 (1.24)** 1.77 (1.98)	**372 (415)- 142- 230** 246 (262)- 105- 140
NS-CEC: E-AS-08 S4–5, E-AS-09 S1			415	**2.9** 10.4	**19.5**	**59**	**0.85 (1.66)** 1.89 (2.60)	**371 (628)** 253 (320)- 78- 175
S-CEC: E-AS-08 S1–3, E-AS-09 S2			563	**3.5** 11.2	**22.0**	**76**	**0.64 (1.24)** 1.59 (2.12)	**305 (415)- 122- 183** 229 (274)- 88- 141
All			872	**3.5** 10.5	**21.1**	**77**	**0.84 (1.66)** 1.77 (2.60)	**351 (628)-106- 245** 246 (320)- 78- 168

Table S2. Emissions of BC, CO, and CO_2 from China in 2018 (or the most recent year stated in the first column) were prescribed in bottom-up inventories or other references (A) and this study (B). In part A, the numbers in brackets indicate the relative biases ("+" for positive, "–" for negative; unit %) of the emission in 2018 to the values estimated by E(BC)-based method (first number) and E(CO)-based method (second number). In B part, the first row shows emissions in CMAQ_HTAPv2.2z; the numbers in brackets show the percentage bias needs to be reduced in HTAPv2.2z to meet the values estimated by E(BC) or E(CO) methods, respectively. The last two rows show estimated emissions by E(BC) and E(CO) in this study, and the numbers in brackets show uncertainty ranges in Tg yr^{-1}.

	Tg BC yr^{-1} (%, %)	Tg CO yr^{-1} (%, %)	Tg CO_2yr^{-1} (%, %)	References / Notes
A. Other references				
MEICv1.0 (2010)	1.76	171	10,124	Li et al., 2017
Zheng (interpolated for BC, CO; 2017 for CO_2)	1.17 (+80, +53)	132 (–20, –32)	10,434 (–16, –28)	Zheng et al., 2018, 2021
REASv2.1 (2008)	1.59	202	8,155	Kurokawa et al., 2013
REASv3.2 (2015)	1.64	165	11,941	Kurokawa and Ohara, 2020
HTAPv3	1.29 (+98, +68)	129 (–22, –34)	/	Crippa et al., 2023
CEDS (CMIP6) (2014)	2.54	/	/	Hoesly et al., 2018
CEDS v_2021_02_05	1.22 (+87, +59)	150 (–10, –23)	10,200 (–17, –30)	O'Rourke et al., 2021
ECLIPSEv6b (interpolated)	0.96 (+47, +25)	137 (–18, –30)	10,210 (–17, –30)	IIASA 2019; Klimont et al. 2017
EDGARv6.1	1.11 (+71, +45)	114 (–32, –42)	11,499 (–7, –21)	https://edgar.jrc.ec.europa.eu/index.php/dataset_ap61
EDGAR_v8.0_GHG	/	/	11,554 (–6, –20)	Crippa et al., 2023
GCB	/	/	9,964 (–19, –31)	Friedlingstein et al., 2020
CO_TCR2 (2019-2020)	/	153 (–8, –22)	/	Miyazaki et al., 2020
Fukue (Estimated)	1.06 (+62, +38)	/	/	Kanaya et al., 2020
B. This study				
CMAQ_HTAPv2.2z	1.36 (↓52, ↓44)	134 (↑24, ↑46)	/	Model
E(BC)$_{HTAPv2.2z}$ - based estimated	0.65 (0.40–0.90)	166 (102–231)	12,355 (7,542–17,168)	Estimated and uncertainty range
E(CO)$_{HTAPv2.2z}$ - based estimated	0.77 (0.54–1.00)	195 (137–254)	14,521 (10,163–18,880)	Estimated and uncertainty range

Table S3. Emissions of BC from hard coal, grade 3 (HC3) grouped by abatement measures and sectors prescribed in ECLIPv6b inventory (Klimont, personal communications, 2021)

Abatement Measures and Sectors	Max of Level of activity [PJ]	Max of Unabated emission factor [kt/unit of activity]	Max of Removal efficiency [%]	Max of Abated emission factor [kt/unit of activity]	Average of Capacities controlled [%]	Sum of Emissions [kt BC]
Hard coal, grade 3 (HC3)	8395.3161	0.2200	99.02	0.2200	49.96	410.2067
Cyclone (MB_CYC)	1312.8621	0.0040	11.00	0.0036	89.63	3.9924
Medium boilers (<50MW) - automatic (DOM_MB_A)	1191.7341	0.0040	11.00	0.0036	83.12	3.5801
Medium boilers (<1MW) - manual (DOM_MB_M)	121.1281	0.0040	11.00	0.0036	96.36	0.4123
No control (NOC)	4163.1457	0.2200	0.00	0.2200	58.85	405.5393
Medium boilers (<50MW) - automatic (DOM_MB_A)	1124.8112	0.0040	0.00	0.0040	4.20	0.1884
Medium boilers (<1MW) - manual (DOM_MB_M)	119.0261	0.0040	0.00	0.0040	4.20	0.0199
Single house boilers (<50 kW) - manual (DOM_SHB_M)	23.3138	0.2150	0.00	0.2150	90.00	4.0752
Cooking stoves (DOM_STOVE_C)	2526.4034	0.1350	0.00	0.1350	95.00	324.0112
Heating stoves (DOM_STOVE_H)	369.5911	0.2200	0.00	0.2200	95.00	77.2445
Coal single house boiler new (SHB_NEW_C)	23.3138	0.2150	20.00	0.1720	10.00	0.3622
Medium boilers (<1MW) - manual (DOM_SHB_M)	23.3138	0.2150	20.00	0.1720	10.00	0.3622
Briquette stove (STV_BRIQ)	2895.9945	0.2200	99.02	0.0022	5.00	0.3128
Cooking stoves (DOM_STOVE_C)	2526.4034	0.1350	98.40	0.0022	5.00	0.2729
Heating stoves (DOM_STOVE_H)	369.5911	0.2200	99.02	0.0022	5.00	0.0399

Table S4. BC and CO information from the Chinese air mass was detected in EMeRGe flights (CHN case) and at Fukue Island. Simulated and observed BC mean concentrations, enhanced CO mean concentrations (ΔCO) in observations and simulations, correction factors (E(BC), E(CO)), and observed BC/CO ratios are displayed. The 2nd column shows data in the CHN case in order of all eight segments / NS-CEC air mass / S-CEC air mass. The 3rd column shows data at Fukue Island in the following order: spring peaks mean during 24th - 28th March 2018 (SP18) from N-CEC air mass / S-CEC air mass. The 4th column shows data at Fukue Island in order of spring 2018 mean (S18) from all Chinese sources (WCN) / N-CEC air mass / S-CEC air mass. E(BC) for the CHN case regards HTAPv2.2z, while data at Fukue regards REASv2.1(2008) (Kurokawa et al., 2013). The number of data used for the CHN case is 15-second intervals, while data at Fukue Island is hourly. E(BC) and E(CO) for data at Fukue Island are from Kanaya et al. (2020).

	All CHN case / NS-CEC / S-CEC	**Fukue: SP18_N-CEC / S-CEC**	**Fukue: S18_WCN/ N-CEC / S-CEC**
Number of recorded data	978 / 415 / 562 (15-s)	38 / 37 (1-h)	210 / 93 / 101 (1-h)
Mean [BC] observation (μg m^{-3})	0.78 / 0.85 / 0.64	0.92 / 0.67	0.51 / 0.50 / 0.56
Mean [BC] simulation (μg m^{-3})	1.69 / 1.89 / 1.59	/	/ 0.79 / 0.89
Mean [ΔCO] observation (ppb)	226 / 338 / 183	254 / 190	130 / 142 / 131
Mean [ΔCO] simulation (ppb)	158 / 175 / 141	/	/
E(BC)	0.46 / 0.45 / 0.40	/	/ 0.6 / 0.48
E(CO)	1.43 / 1.93 / 1.29	/	/ 1.02 / 0.87
Observed BC/CO (ng m^{-3} ppb^{-1})	3.5 / 2.9 / 3.5	3.6 / 3.5	4.9 / 4.4 / 5.2

Table S5. BC/CO and CO/CO_2 ratios from biomass burning in the THL case and recorded in other references

References	BC/CO (ng m^{-3} ppb^{-1})	CO/CO_2 (%)	Characteristics
This study (THL case)	7.1	4	
Akagi et al., 2011	7.0 ± 4.1	8.9 ± 2.6	Emission inventory for tropical forests,
	7.3 ± 4.3	5.9 ± 1.6	savanna,
	16 – 21	4.1 – 6	and garbage burning and open cooking
Lee et al., 2018	6.98		Simulated by WRF-Chem (FINNv1.5) for fire biomass burning in Southeast Asia 2002 – 2008
Warneke et al., 2009	7 ± 4 10 ± 5	4.2 ± 1.9 5.0 ± 2.5	Biomass burning under plumes sampled by flights over Alaska in April 2008: - Lake Baikal. - Agricultural fires in Kazakhstan
Kondo et al., 2011	8.5 ± 5.4 1.7±0.8 3.4±1.6	1.5 ± 0.5 22.2 ± 11.8 2.6 ± 1.0	Flaming-phase fires from Asian biomass burning. Summer mix fires in North America and Canada: - Smoldering - Flaming
Zhu et al., 2019	> 7		Biomass burning air mass observed at Rishiri Island (Japan)
Chi et al., 2013, Cristofanelli et al., 2013	21.8 – 29.8		Agricultural fires
Chi et al., 2013	9.3		Winter air masses affected by anthropogenic emissions.
Paris et al., 2009	4.1; 6.8	4.6 ± 2.0	One-day fresh flaming plumes
Vasileva et al. 2017	6.1–6.3	10.0 ± 0.6	One-day fresh flaming plumes
		15.2 ± 0.7	Dominantly smouldering fires
Pirjola et al., 2015		3.2	Dominantly smouldering fires
Cofer et al., 1989, 1998; Goode et al., 2000; Laursen et al., 1992; McRae et al., 2006; Simpson et al., 2011; Urbanski et al., 2009		6–16	Aircraft measurements of forest fire plumes in the northern US, Canada, Alaska, and Siberia

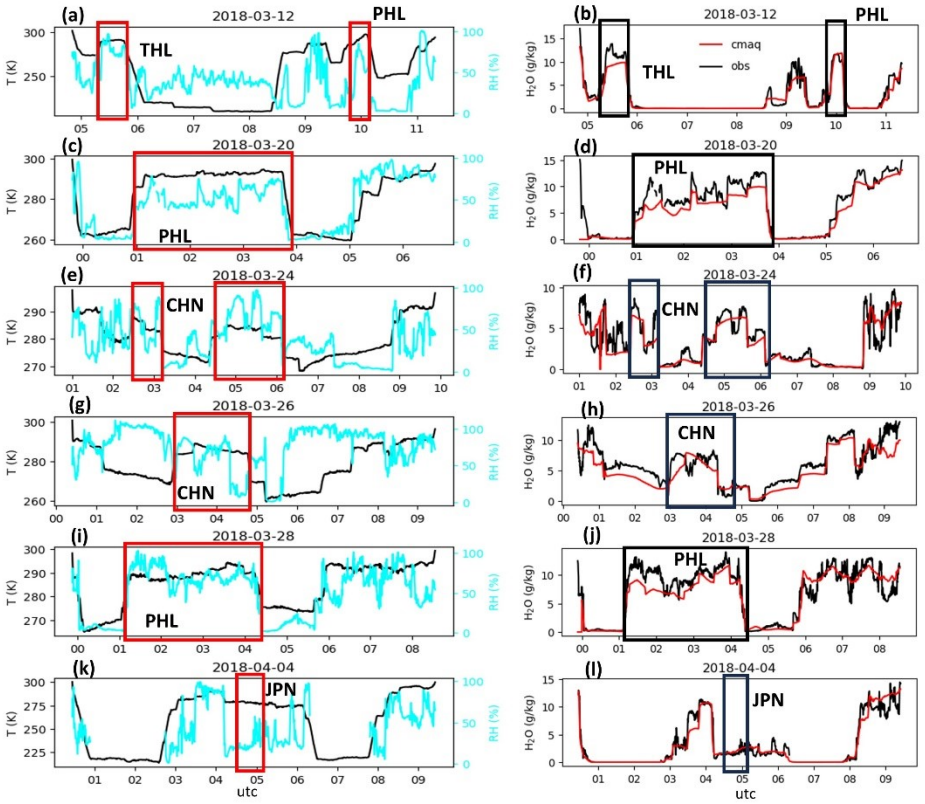

Figure S1. The left column shows temperature (black) and relative humidity (cyan) during the flights; the right column shows the H_2O mass mixing ratio by observation (black) and simulation (red). Red and black boxes show investigated flight segments (similar to Figs. 2 and 3); corresponding cases are noted in black abbreviations.

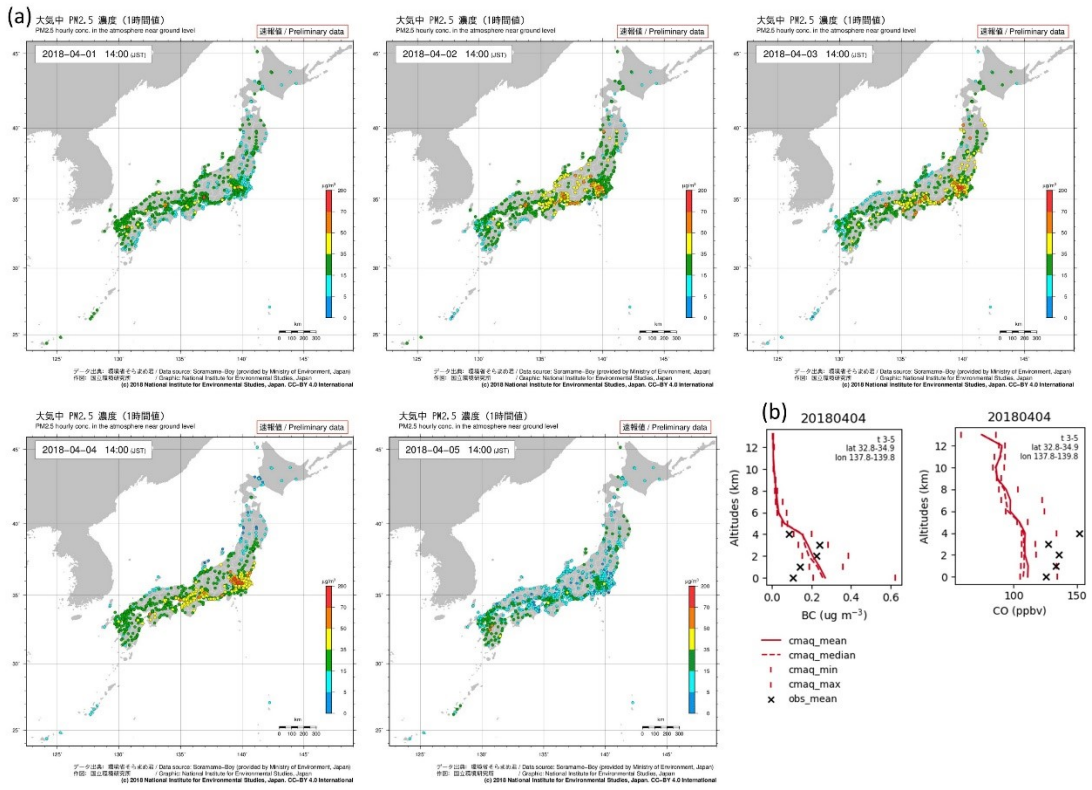

Figure S2. (a) Japan's $PM_{2.5}$ levels at 14:00 JST from 1st to 5th April 2018, according to Ministry of the Environment Air Pollutant Wide-Area Monitoring System (https://soramame.env.go.jp/) and (b) Vertical profiles of BC (left) and CO (right) in observations (black: batches for mean) and simulations (red: solid lines for mean, dashed lines for median, vertical bars for minimum and maximum values). Map graphics created by the National Institute for Environmental Studies, Japan.

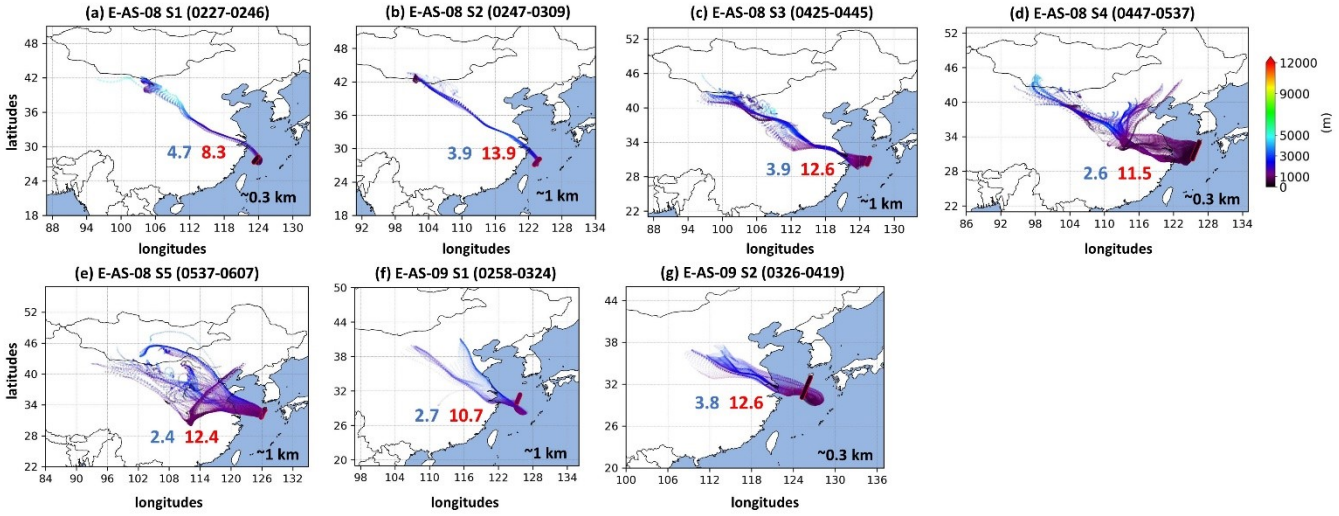

Figure S3. HYSPLIT backward trajectories for seven segments in the CHN case. Numbers in blue and red indicate observed and simulated BC/CO ratios, respectively.

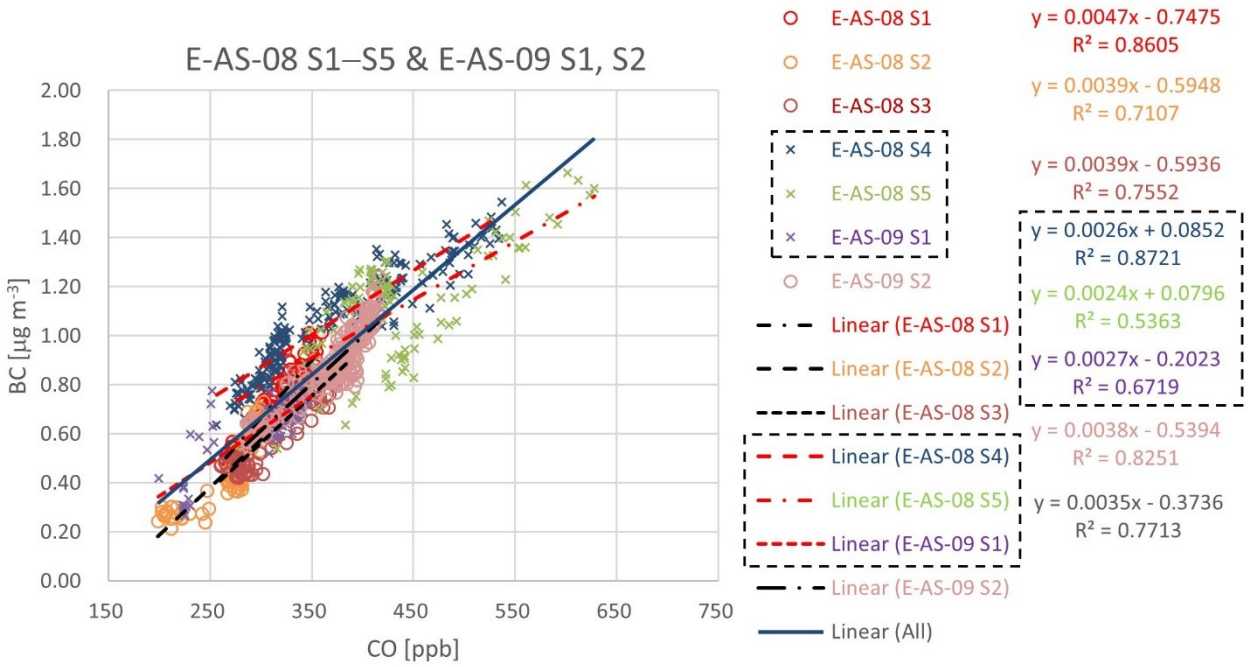

Figure S4. Observed BC/CO concentration ratios calculated for seven segments in CHN case. The dashed boxes in the legend indicate the functions of NS-CEC sub-air masses (lower BC/CO ratios than S-CEC sub-air masses). The air masses from NS-CEC include segments E-AS-08 S4–5 and E-AS-09 S1 with the data in batches and regression lines in red. The air masses from S-CEC include segments E-AS-08 S1–3 and E-AS-09 S2 with the data in open circles and regression lines in black. The total linear regression line for all data is shown in blue. Regression functions are shown in the same colour as the dataset.

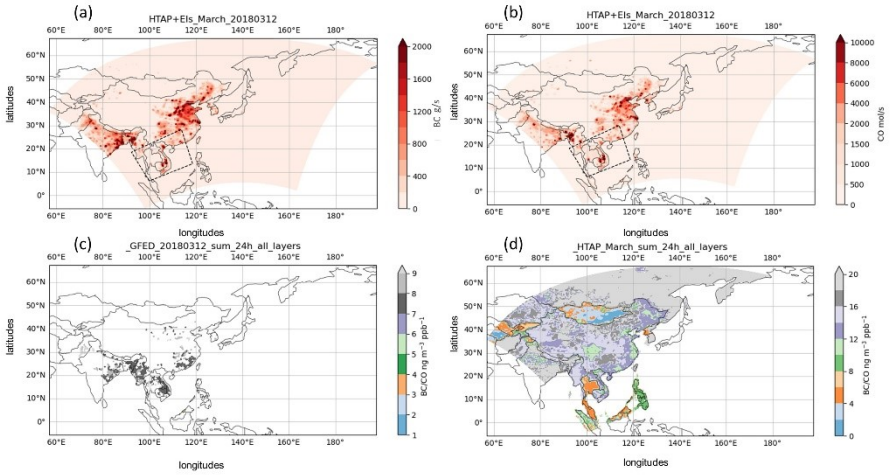

Figure S5. (a) BC and (b) CO emissions in CMAQ's emissions inventories, including HTAPv2.2z. Loose dashed boxes for the THL case. (c) BC/CO emission ratio from GFED inventory and (d) HTAPv2.2z inventory (without JEI-DB data for Japan).

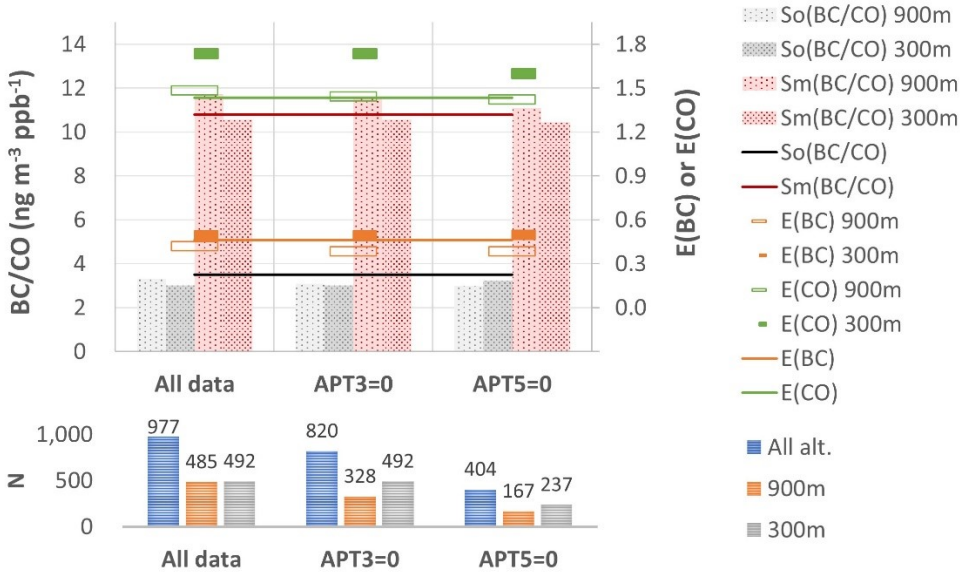

Figure S6. Influences of Accumulated Precipitation along Trajectories (APT) and altitudes to observed (So) and simulated (Sm) BC/CO, E(BC), and E(CO). Columns show BC/CO values scaled to the left axis; grey columns and black line for aircraft data; red columns and line for CMAQ simulation; first and second columns in each set show data at 900 m and 300 m, respectively, while lines show values from all data, all altitudes. Oranges and greens represent E(BC) and E(CO), respectively, scaled to the right axis; long and short boxes show data at 900 m and 300 m, respectively, while lines show values from all data, all altitudes. The lower panel shows the amount of data extracted for each case.