This supplement includes Table S1-S12 and Figure S1-S15 as listed below.

Table S1. Information about the surface monitor stations used in this study

Table S2. Comparison of the ground-based hourly O_3, NO_2, and CO observations with the simulations utilizing EDGAR-HTAP v2 (EDV2) and v3 (EDV3) and KORUS v5 (KOV5) emission inventories in each regional box (unit = ppb). N denotes number of samples. R is correlation coefficient.

Table S3. Comparison of aircraft-based 1-minuite-interval O_3, NO_2, CO, and HCHO observations with EDV2, EDV3, and KOV5 in each case distinguished by China contribution to O_3 concentration under 2 km height (unit = ppb). N denotes number of samples. σ is standard deviation. R is correlation coefficient.

Table S4. Comparison of surface meteorological observations and WRF-Chem for the KORUS-AQ campaign period. N is number of samples. R (RMSE) denotes correlation coefficient (root-mean-square-error).

Table S5. The area sum emissions in Eastern China (27.7-40˚N, 115-123˚E), South Korea (34.5-38˚N, 126-130˚E), and Seoul Metropolitan Area (SMA: 37.2-37.8˚N, 126.5-127.3˚E) in May 2016 for NOx.

Table S6. Spring and summer NO_2 concentrations in Korean metropolitan cities and provinces. Both daytime (10-20 LT) and nighttime (01-06 LT) averages are shown. Differences in concentrations between spring and summer ($NO_{2\ spring} - NO_{2\ summer}$) are in the parenthesis. The data from 2002 to 2019 are utilized.

Table S7. Spring and summer O_x ($=O_3+NO_2$) concentrations in Korean metropolitan cities and provinces. Both daytime (10-20 LT) and nighttime (01-06 LT) averages are shown. Differences in concentrations between spring and summer ($O_{X\ spring} - O_{X\ summer}$) are in the parenthesis. The data from 2002 to 2019 are utilized.

Table S8. Spring and summer mean temperatures, mean maximum temperatures, and mean wind velocities in Korean metropolitan cities and provinces. Differences in values between spring and summer are in the parenthesis. The cities and provinces listed in the table are in counterclockwise order in regards to the South Korean map. The data from 2001 to 2021 are utilized.

Table S9. Altitudes (m) of monitoring sites in Gangwon-do. Ganseong-eup represents Gosung.

Table S10. Surface and stratospheric O_3 concentrations (O_{3S}) and their ratio in Korea simulated by CESM. The concentrations and ratios for the altitude of 1 km are shown in parenthesis.

Table S11. The trends of NO_x and CO emissions from linear fits of the data covering 2001-2020.

Table S12. The observed trends of NO_2 and CO concentrations from linear fits of the data covering 2001-2021.

Figure S1. The examples of CAM-Chem U, V wind components for spring, 2003. Without nudging, the model simulated U, V do not closely agree with the MERRA2 data.

Figure S2. Monthly variations of CAM-Chem simulated ozone concentration (red line) and observed ozone concentration (black solid line) at each pressure level near Pohang, between 14-17 KST. (Left panel) ~1 degree horizontal resolution simulation, and (right panel) ~2 degree horizontal resolution simulation.

Figure S3. The diurnal variations of observed and simulated O_3 and NO_2 averaged for the simulation period.

Figure S4. The time series of observed and simulated hourly ozone in (top) Seoul and (bottom) Gosung. Basic statistics are shown as follows. Mean bias (MB): Seoul -6.2 ppb /Gosung -0.9 ppb, Root Mean Square Errors (RMSE): Seoul 18.2 ppb/Gosung 13.7 ppb, Correlation Coefficient(R): Seoul 0.68/Gosung 0.54.

Figure S5. Diurnal variations of observed and simulated ozone concentrations averaged for the entire simulation period: (top) Seoul and (bottom) Gosung. Basic statistics are shown in the plot. MB is mean bias. RMSE is root-mean-square-error. R is correlation coefficient.

Figure S6. Averaged O_3 from the ground-based observations and model results for regional boxes that distinguish urban (red box) and non-urban (green box) region (central plot). Box averaged diurnal cycle (solid lines) of O_3 and 1/4 of standard deviations (filled area) from observations (black), the WRF-Chem simulations using EDGAR-HTAP version 2 (EDV2, green), EDGAR-HTAP version 3 (EDV3, blue), and KORUS-AQ version 5 (KOV5, red) are shown. The diurnal cycle plots represent Northern China (NOC, 38-42˚N/106-110˚E), Sichuan-Chongqing-Guizhou (SCG, 27-33˚N/103-109˚E), Pearl River Delta (PRD, 21.5-24˚N/112-115.5˚E), Southeastern China (SEC, 24-28˚N/116-120˚E), Yangtze River Delta (YRD, 30-33˚N/119-122˚E), South Korea (KOR, 34.5-38˚N/126-130˚E), North China Plain (NCP, 34-41˚N/113-119˚E), and Northeastern China (NEC, 43-47˚N/124-130˚E).

Figure S7. Averaged model and airborne observations of (a) O_3, (b) NO_2, (c) CO, and (d) HCHO (bars) and 1/4 of standard deviations (whiskers) (unit: ppbv) under 2 km height for the Local, Transport, and Chungnam cases from DC-8 (grey), EDV2 (green), EDV3 (blue), and KOV5 (red). The Chungnam (Chungcheongnam-do) region has large point sources like coal-burning power plants and petrochemical facilities that are not well-represented in the bottom-up emission inventories. The local case (May/4, May/20, June/2, June/3) and transport case (May/25, May/26, May/31) represent the dates with the smallest and largest influence from Chinese emissions, respectively. The Chungnam case represents the dates when DC-8 had survey flights targeting the urban and point sources in Chungcheongnam-do and downwind.

Figure S8. Vertically averaged O_3 from DC-8 (black), EDV2 (green), EDV3 (blue), and KOV5 (red) for the Local and Transport cases under 2 km height above ground level. The 1/2 of standard deviations are represented with black whiskers in each 200m layer. Sensitivity tests are conducted with doubled anthropogenic CO and VOC emissions in China (EDV3_Ch2, blue triangle dots and dashed lines) and in both China and South Korea (EDV3_ChKo2, blue open square and dotted lines). The model results co-located with the observations are sampled and compared with each other. The sampling numbers in the layers are represented with magenta color. (a) and (b) include the data from all flights while (c) and (d) select the data over SMA (Seoul Metropolitan Area).

Figure S9. Diurnal variations of the model ozone concentrations at surface from various emission scenarios. The model results were averaged for the full simulation period.

Figure S10. (Top) topography map of South Korea and (bottom) West-East vertical cross-section of terrain connecting Seoul and Gosung. Seoul and Gosung in Gangwon-do are

highlighted with color circles. Color bar denotes elevations above sea level (m). A simplified potential ozone transport path is depicted in the bottom plot. Here the ozone layer is colored orange and the terrain is colored gray.

Figure S11. Diurnal O_3 exceedances for the sites that have more exceedances during summer than spring

Figure S12. Diurnal O_3 exceedances for the locations that have similar exceedances for spring and summer

Figure S13. Diurnal O_3 exceedances for the locations that have more exceedances during spring than summer

Figure S14. (Left panel) the number of cars passing highway tolls near the Seoul Metropolitan Area (SMA) from January to October in 2019 and 2020 (top) and 2019 and 2021 (bottom) and (Right panel) reductions (%) in mobility based on transit station mobility metrics and driving mobility index in SMA from Google and Apple mobility report, respectively, covering spring and summer in 2020 (top) and 2021 (bottom) relative to 2019.

Figure S15. Difference (%) in the toll numbers, NO_2, SO_2, CO, PM_{10}, and $PM_{2.5}$ concentrations in SMA during spring and summer: (top) 2020-2019 and (bottom) 2021-2019.

Table S1. Information about the surface monitor stations used in this study

Index	Station code	City·Province	Site name	Latitude	Longitude
1	111121	Seoul	Jung-gu	37.56	126.97
2	111123		Jongno-gu	37.57	127.00
3	111131		Yongsan-gu	37.54	127.00
4	111141		Gwangjin-gu	37.55	127.09
5	111142		Seongdong-gu	37.54	127.04
6	111151		Jungnang-gu	37.58	127.09
7	111152		Dongdaemun-gu	37.58	127.03
8	111161		Seongbuk-gu	37.61	127.03
9	111171		Dobong-gu	37.65	127.03
10	111181		Eunpyeong-gu	37.61	126.93
11	111191		Seodaemun-gu	37.59	126.95
12	111201		Mapo-gu	37.56	126.91
13	111212		Gangseo-gu	37.54	126.84
14	111221		Guro-gu	37.50	126.89
15	111231		Yeongdeungpo-gu	37.53	126.90
16	111241		Dongjak-gu	37.48	126.97
17	111251		Gwanak-gu	37.49	126.93
18	111261		Gangnam-gu	37.52	127.05
19	111262		Seocho-gu	37.50	126.99
20	111273		Songpa-gu	37.50	127.09
21	111274		Gangdong-gu	37.55	127.14
22	111281		Geumcheon-gu	37.45	126.91
23	111291		Gangbuk-gu	37.65	127.01
24	111301		Yangcheon-gu	37.53	126.86
25	111311		Nowon-gu	37.66	127.07
26	831492	Incheon	Baengnyeong island	37.96	124.63
27	823611		Sinheung	37.47	126.63
28	823621		Songnim	37.47	126.64
29	823631		Guwol	37.45	126.72
30	823632		Sungui	37.46	126.65
31	823641		Bupyeong	37.50	126.72
32	823651		Yeonhui	37.54	126.68
33	823671		Gojan	37.40	126.70
34	823681		Seongnam	37.50	126.67
35	823691		Songhae	37.76	126.46
36	823633		Seokbawi	37.46	126.69
37	525111	Daejeon	Eumnae-dong	36.37	127.42
38	525112		Munpyeong-dong	36.45	127.40
39	525121		Munchang-dong	36.32	127.44
40	525141		Guseong-dong	36.37	127.37
41	525151		Daeheungdong2	36.32	127.42
42	525173		Wolpyeong-dong	36.35	127.35
43	324115	Gwangju	Seoseok-dong	35.15	126.92
44	324121		Nongseong-dong	35.15	126.89
45	324133		Duam-dong	35.17	126.93
46	324141		Songjeong1-dong	35.14	126.80
47	221112	Busan	Gwangbok-dong	35.10	129.03
48	221172		Daeyeon-dong	35.13	129.09

49	221181		Hakjang-dong	35.15	128.98
50	221182		Deokcheon-dong	35.21	129.01
51	221202		Jangnim-dong	35.08	128.97
52	221152		Jeonpo-dong	35.16	129.06
53	221221		Yeonsan-dong	35.18	129.07
54	221141		Taejongdae	35.06	129.08
55	221191		Cheongnyong-dong	35.28	129.09
56	221192		Jwa-dong	35.17	129.17
57	221211		Daejeo-dong	35.21	128.97
58	221212		Noksan-dong	35.09	128.86
59	221231		Gijang-eup	35.24	129.22
60	221251		Bugok-dong	35.23	129.09
61	221271		Gwangan-dong	35.16	129.12
62	221163		Myeongjang-dong	35.20	129.10
63	221233		Yongsu-ri	35.33	129.18
64	221241		Sujeong-dong	35.13	129.05
65	221281		Daesin-dong	35.12	129.02
66	221282		Deokpo-dong	35.17	128.99
67	221283		Gaegeum-dong	35.16	129.02
68	221284		Dangni-dong	35.11	128.97
69	221193		Jaesong-dong	35.18	129.12
70	221142		Cheonghak-dong	35.09	129.06
71	221183		Hwamyeong-dong	35.23	129.01
72	238120		Daesong-dong	35.50	129.42
73	238121		Seongnam-dong	35.56	129.32
74	238122		Bugok-dong	35.50	129.34
75	238124		Yaeum-dong	35.53	129.33
76	238125		Samsan-dong	35.54	129.33
77	238127	Ulsan	Sinjeong-dong	35.53	129.31
78	238128		Deoksil-ri	35.43	129.31
79	238129		Mugeo-dong	35.56	129.26
80	238130		Hyomun-dong	35.56	129.37
81	238371		Hwasal-ri	35.44	129.34
82	238373		Sangnam-ri	35.49	129.31
83	238126		Sinjeongno	35.53	129.30
84	422114		Suchang-dong	35.87	128.58
85	422115		Jisan-dong	35.83	128.63
86	422121		Seoho-dong	35.87	128.71
87	422132		Ihyeon-dong	35.87	128.55
88	422141		Daemyeong-dong	35.85	128.57
89	422153	Daegu	Nowon-dong	35.89	128.56
90	422154		Sinam-dong	35.89	128.63
91	422161		Manchon-dong	35.87	128.64
92	422171		Horim-dong	35.84	128.49
93	422113		Namsan-dong	35.87	128.59
94	422133		Pyeongni-dong	35.88	128.56
95	339111		Ido-dong	33.50	126.53
96	339121	Jeju	Donghong-dong	33.25	126.57
97	339312		Gosal-ri(Gosan)	33.29	126.16
98	533112	Chungcheongbuk	Songjeong-dong	36.64	127.44
99	533113		Sacheon-dong	36.67	127.47

100	533114		Yongdam-dong	36.64	127.50
101	633122		Hoam-dong	36.96	127.93
102	633131		Jangnak-dong	37.15	128.22
103	534111		Seonghwang-dong	36.81	127.15
104	534421	Chungcheongnam	Dokgot-ri	36.99	126.39
105	534422		Dongmun-dong	36.78	126.46
106	132112		Jungangno	37.88	127.72
107	132113		Seoksa-dong	37.86	127.75
108	632132		Okcheon-dong	37.76	128.90
109	632121		Jungang-dong	37.35	127.95
110	632122	Gangwon	Bangok-dong	37.34	127.98
111	632421		Ganseong-eup(Gosung)	38.29	128.39
112	132401		Bangsan-myeon	38.22	127.96
113	632371		Bukpyeong-myeon	37.43	128.66
114	632431		Chiaksan	37.36	128.13
115	131111		Sinpung-dong	37.28	127.01
116	131112		Ingye-dong	37.26	127.03
117	131113		Gwanggyo-dong	37.29	127.07
118	131114		Yeongtong-dong	37.25	127.06
119	131141		Anyang6-dong	37.39	126.93
120	131142		Burim-dong	37.39	126.96
121	131144		Hogye-dong	37.38	126.95
122	131145		Anyang2-dong	37.41	126.92
123	131121		Dandae-dong	37.45	127.16
124	131123		Jeongja-dong	37.36	127.11
125	131131		Uijeongbu-dong	37.74	127.04
126	131161		Cheolsan-dong	37.48	126.87
127	131191		Gojan-dong	37.32	126.83
128	131192		Wonsi-dong	37.33	126.81
129	131193		Bono-dong	37.30	126.87
130	131194		Wongok-dong	37.33	126.80
131	131195		Bugok-dong	37.33	126.86
132	131196	Gyeonggi	Daebu-dong	37.24	126.59
133	131341		Bijeon-dong	36.99	127.11
134	131201		Byeollyang-dong	37.42	126.99
135	131202		Gwacheon-dong	37.45	127.00
136	131211		Gyomun-dong	37.59	127.13
137	131212		Donggu-dong	37.62	127.14
138	131222		Bugok3-dong	37.32	126.95
139	131381		Haengsin-dong	37.62	126.84
140	131382		Siksa-dong	37.69	126.81
141	131501		Dang-dong	37.35	126.95
142	131502		Sanbon-dong	37.36	126.93
143	131231		Jeongwang-dong	37.35	126.74
144	131232		Sihwasandan	37.34	126.72
145	131233		Daeya-dong	37.44	126.79
146	831151		Sosabon-dong	37.48	126.80
147	831152		Nae-dong	37.52	126.77
148	831153		Jung2-dong	37.49	126.77
149	831154		Ojeong-dong	37.53	126.80
150	131241		Geumgok-dong	37.64	127.22

151	131411		Gimnyangjang-dong	37.23	127.20
152	131471		Sau-dong	37.62	126.72
153	131472		Gochon-eup	37.61	126.76
154	131473		Wolgot-myeon	37.72	126.55
155	131531		Osan-dong	37.16	127.08
156	131541		Sinjang-dong	37.54	127.21
157	437112	Gyeongsangbuk	Jangheung-dong	35.98	129.37
158	437113		Jangnyang-dong	36.07	129.38
159	437151		Gongdan-dong	36.10	128.39
160	437152		Wonpyeong-dong	36.13	128.33
161	437153		Hyeonggok-dong	36.11	128.34
162	437131		Mundang-dong	36.14	128.09
163	437122		Seonggeon-dong	35.85	129.21
164	437141		Myeongnyun-dong	36.57	128.73
165	437161		Gaheung-dong	36.82	128.61
166	437402		Yeongcheon-si	35.97	128.94
167	437541		Taeha-ri	37.52	130.80
168	238111	Gyeongsangnam	Hoewon-dong	35.22	128.57
169	238112		Bongam-dong	35.22	128.60
170	238141		Myeongseo-dong	35.24	128.64
171	238142		Ungnam-dong	35.21	128.66
172	238143		Seongju-dong	35.20	128.71
173	238151		Gyeonghwa-dong	35.15	128.69
174	238131		Sangbong-dong	35.20	128.08
175	238181		Dongsang-dong	35.24	128.88
176	238361		Bukbu-dong	35.35	129.04
177	335115	Jeollabuk	Nosong-dong	35.83	127.15
178	735111		Samcheon-dong	35.80	127.12
179	735114		Palbok-dong	35.85	127.09
180	735121		Sinpung-dong	35.97	126.70
181	735122		Soryong-dong	35.97	126.65
182	735123		Gaejeong-dong	35.96	126.75
183	735132		Namjung-dong	35.95	126.96
184	735151		Jukang-dong	35.41	127.38
185	336121	Jeollanam	Seogang-dong	34.74	127.73
186	336124		Wollae-dong	34.85	127.72
187	336352		Jung-dong	34.94	127.70
188	336353		Taein-dong	34.94	127.76
189	336354		Jinsang-myeon	35.02	127.72
190	336131		Jangcheon-dong	34.95	127.49
191	336111		Yongdang-dong	34.81	126.39
192	336441		Nabul-ri	34.77	126.44

Table S2. Comparison of the ground-based hourly O_3, NO_2, and CO observations with the simulations utilizing EDGAR-HTAP v2 (EDV2) and v3 (EDV3) and KORUS v5 (KOV5) emission inventories in each regional box (unit = ppb). N denotes number of samples. R is correlation coefficient.

	Region		[1]NCP	[1),a)]SCG	[1)]YRD	[1)]PRD	[1),b)]KOR (SMA)	[2),c)]NEC	[2),d)]NOC	[2),e)]SEC
	N		190	104	93	68	358 (125)	45	28	43
O_3	**OBS**	**Mean**	44.5	34.6	38.2	27.9	41.5 (36.6)	40.9	44.3	26.1
	EDV2	**Mean**	32.2	53.5	21.6	27.6	40.5 (31.1)	28.6	39.4	40.8
		Bias	-12.3	18.9	-16.6	-0.3	-1.0 (-5.5)	-12.3	-4.9	14.7
		R	0.65	0.53	0.62	0.61	0.59 (0.60)	0.48	0.63	0.52
	EDV3	**Mean**	43.4	57.5	35.7	34.7	41.0 (32.6)	35.2	43.7	45.5
		Bias	-1.1	23.0	-2.5	6.8	-0.5 (-4.0)	-5.7	-0.6	19.4
		R	0.68	0.55	0.66	0.65	0.56 (0.57)	0.63	0.67	0.55
	KOV5	**Mean**	49.0	55.3	41.1	35.7	42.2 (33.1)	37.1	43.8	42.4
		Bias	4.5	20.7	2.8	7.8	0.7 (-3.5)	-3.8	-0.5	16.3
		R	0.71	0.53	0.65	0.70	0.62 (0.64)	0.62	0.67	0.54

1) Urban area, 2) Non-urban area

a) Sichuan-Chongqing-Guizhou, b) South Korea (SMA-Seoul Metropolitan Area), c) Northeastern China, d) Northern China, e) Southeastern China

Table S3. Comparison of aircraft-based 1-minuite-interval O_3, NO_2, CO, and HCHO observations with EDV2, EDV3, and KOV5 in each case distinguished by China contribution to O_3 concentration under 2 km height (unit = ppb). N denotes number of samples. σ is standard deviation. R is correlation coefficient.

Species	Case	Type	N	Mean	Bias	σ	R
O_3	**Local** (5/4,20 , 6/2,3)	**OBS**	1125	81.2		15.3	
		EDV2		65.2	-15.9	13.4	0.66
		EDV3		65.2	-16.0	12.8	0.59
		KOV5		62.6	-18.5	11.5	0.70
	Transport (5/25,26 , 6/1)	**OBS**	605	95.6		19.1	
		EDV2		87.3	-8.3	13.8	0.64
		EDV3		93.1	-2.5	16.0	0.67
		KOV5		84.8	-10.8	14.3	0.69
	Chungnam (5/22 , 6/5)	**OBS**	812	98.4		17.8	
		EDV2		61.6	-36.8	14.3	0.14
		EDV3		60.2	-38.2	14.2	0.07
		KOV5		60.3	-38.1	14.0	0.17

Table S4. Comparison of surface meteorological observations and WRF-Chem for the KORUS-AQ campaign period. N is number of samples. R (RMSE) denotes correlation coefficient (root-mean-square-error).

Nation		Eastern China (sites = 271)			South Korea (sites = 48)		
Variable		Temperature (˚C)	Relative humidity (%)	Wind speed (m/s)	Temperature (˚C)	Relative humidity (%)	Wind speed (m/s)
N		83698	83696	79595	14948	14946	14103
Mean	Obervation	20.13	65.02	2.87	18.94	65.81	2.56
	WRF-Chem	19.22	65.35	4.12	17.23	71.35	3.84
R		0.90	0.85	0.55	0.88	0.76	0.62
Mean bias		-0.91	0.32	1.25	-1.71	5.54	1.27
RMSE		3.20	13.94	2.45	2.84	15.88	2.31

Table S5. The area sum emissions in Eastern China (27.7-40˚N, 115-123˚E), South Korea (34.5-38˚N, 126-130˚E), and Seoul Metropolitan Area (SMA: 37.2-37.8˚N, 126.5-127.3˚E) in May 2016 for NOx. NO_x emissions from SSP5-8.5 (CESM/CAM-Chem) and EDGAR-HTAP v3, v2, and KORUS v5 (WRF-Chem) are compared. Over China, SSP5-8.5 NOx emissions are slightly larger than those in KORUS v5 and are lower than those in EDGAR-HTAP v3. SSP5-8.5 has much lower NOx emissions over South Korea and SMA, compared to EDGAR-HTAP v3. "No SMA" simulations with WRF-Chem may help estimate the uncertainty in the simulated O_3 originated from the emission discrepancy. "No SMA" increases O_3 concentrations over South Korea (SMA) by 1.87 (22.1) ppb. CMIP6 (CESM/CAM-Chem) (not shown in the table) is based on EDGAR v4.2 or v4.3.2 described in Feng et al. (2020) (Feng, L., Smith, S. J., Braun, C., Crippa, M., Gidden, M. J., Hoesly, R., Klimont, Z., van Marle, M., van den Berg, M., and van der Werf, G. (2020). The generation of gridded emissions data for CMIP6. Geoscientific Model Development, 13, 461-482, doi.org/10.5194/gmd-13-461-2020).

NOx emission (unit = mols/s)	**SSP5-8.5**	**EDGAR-HATP v3**	**EDGAR-HATP v2**	**KORUS v5**
Eastern China	6638	9034	10063	5482
South Korea	303	1097	990	886
SMA	26	214	196	191

Table S6. Spring and summer NO_2 concentrations in Korean metropolitan cities and provinces. Both daytime (10-20 LT) and nighttime (01-06 LT) averages are shown. Differences in concentrations between spring and summer ($NO_{2\ spring} - NO_{2\ summer}$) are in the parenthesis. The data from 2002 to 2019 are utilized.

Location		Daytime period	Nighttime period
		Spring / Summer (difference)	Spring / Summer (difference)
City	Seoul (SUL)	36.1 / 29.5 (6.6)	36.9 / 24.6 (12.3)
	Incheon (INC)	30.5 / 24.3 (6.3)	28.5 / 19.7 (8.8)
	Daejeon (DJN)	16.1 / 12.0 (4.1)	20.5 / 13.6 (6.9)
	Gwangju (GWJ)	19.8 / 13.4 (6.4)	19.0 / 10.4 (8.6)
	Busan (BSN)	20.9 / 13.9 (7.0)	24.3 / 14.5 (9.8)
	Ulsan (ULS)	23.9 / 20.1 (3.8)	21.2 / 15.3 (5.9)
	Daegu (DGU)	24.3 / 16.6 (7.7)	26.0 / 15.4 (10.6)
Province	Gyeonggi-do (GGI)	27.3 / 20.8 (6.5)	31.2 / 20.7 (10.5)
	Chungcheongbuk-do (CCB)	18.9 / 13.6 (5.3)	20.8 / 13.7 (7.1)
	Chungcheongnam-do (CCN)	17.4 / 12.9 (4.5)	18.0 / 11.9 (6.1)
	Jeollabuk-do (JLB)	15.1 / 11.2 (3.9)	14.1 / 9.2 (4.9)
	Jeollanam-do (JLN)	17.4 / 14.0 (3.4)	14.1 / 10.5 (3.6)
	Jeju Island (JEJ)	12.0 / 8.5 (3.5)	8.5 / 6.0 (2.5)
	Gyeongsangnam-do (GSN)	18.6 / 15.0 (3.6)	18.9 / 13.3 (5.6)
	Gyeongsangbuk-do (GSB)	17.9 / 13.4 (4.5)	20.5 / 13.6 (6.9)
	Gangwon-do (GWO)	14.0 / 9.9 (4.1)	13.4 / 8.1 (5.3)
Background	Ulleung Island (ULL)	3.4 / 2.7 (0.7)	3.3 / 2.8 (0.5)
	Gosung (GSU)	4.5 / 2.6 (1.9)	4.6 / 2.8 (1.8)

Table S7. Spring and summer O_x ($=O_3+NO_2$) concentrations in Korean metropolitan cities and provinces. Both daytime (10-20 LT) and nighttime (01-06 LT) averages are shown. Differences in concentrations between spring and summer ($O_{X\ spring} - O_{X\ summer}$) are in the parenthesis. The data from 2002 to 2019 are utilized.

Location		Daytime period	Nighttime period
		Spring / Summer (difference)	Spring / Summer (difference)
City	Seoul (SUL)	70.8 / 65.0 (5.8)	57.4 / 41.6 (15.8)
	Incheon (INC)	65.2 / 57.0 (8.2)	53.6 / 39.4 (14.2)
	Daejeon (DJN)	57.8 / 48.7 (9.1)	43.4 / 32.3 (11.1)
	Gwangju (GWJ)	59.9 / 48.6 (11.3)	47.5 / 34.0 (13.5)
	Busan (BSN)	61.4 / 47.6 (13.8)	54.6 / 36.4 (18.2)
	Ulsan (ULS)	62.8 / 53.2 (9.6)	46.9 / 33.5 (13.4)
	Daegu (DGU)	64.3 / 53.9 (10.4)	50.0 / 34.5 (15.5)
Province	Gyeonggi-do (GGI)	65.1 / 58.9 (6.2)	51.9 / 38.1 (13.8)
	Chungcheongbuk-do (CCB)	61.5 / 53.2 (8.3)	45.7 / 34.2 (11.5)
	Chungcheongnam-do (CCN)	58.8 / 49.9 (8.9)	47.3 / 34.2 (13.1)
	Jeollabuk-do (JLB)	53.4 / 45.5 (7.9)	40.7 / 32.0 (8.7)
	Jeollanam-do (JLN)	60.5 / 48.9 (11.6)	47.5 / 34.4 (13.1)
	Jeju Island (JEJ)	61.3 / 42.7 (18.6)	52.5 / 34.6 (17.9)
	Gyeongsangnam-do (GSN)	63.3 / 54.8 (8.5)	47.8 / 34.9 (12.9)
	Gyeongsangbuk-do (GSB)	63.4 / 50.9 (12.5)	49.0 / 33.7 (15.3)
	Gangwon-do (GWO)	58.5 / 49.0 (9.5)	41.3 / 28.6 (12.7)
Background	Ulleung Island (ULL)	60.0 / 46.6 (13.4)	59.2 / 45.9 (13.3)
	Gosung (GSU)	62.8 / 45.5 (17.3)	62.7 / 47.9 (15.0)

Table S8. Spring and summer mean temperatures, mean maximum temperatures, and mean wind velocities in Korean metropolitan cities and provinces. Differences in values between spring and summer are in the parenthesis. The cities and provinces listed in the table are in counterclockwise order in regards to the South Korean map. The data from 2001 to 2021 are utilized.

Location		Mean temperature (℃)	Mean maximum temperature (℃)	Mean wind velocity (m/s)
		Spring / Summer (difference)		
City	Seoul	12.4 / 24.9 (-12.5)	17.7 / 29.0 (-11.3)	2.6 / 2.2 (0.4)
	Incheon	11.6 / 23.9 (-12.3)	16.1 / 27.5 (-11.4)	3.2 / 2.5 (0.7)
	Daejeon	12.9 / 24.9 (-12.0)	19.1 / 29.3 (-10.2)	2.0 / 1.8 (0.2)
	Gwangju	13.5 / 25.2 (-11.7)	19.7 / 29.8 (-10.1)	2.0 / 2.0 (0.0)
	Busan	13.7 / 24.0 (-10.3)	18.1 / 27.4 (-9.3)	3.5 / 3.2 (0.3)
	Ulsan	13.6 / 24.4 (-10.8)	19.1 / 28.7 (-9.6)	2.3 / 2.0 (0.3)
	Daegu	14.3 / 25.5 (-11.2)	20.3 / 30.3 (-10.0)	2.4 / 2.2 (0.2)
Province	Gyeonggi-do	11.5 / 24.0 (-12.5)	17.1 / 28.4 (-11.3)	2.3 / 2.0 (0.3)
	Chungcheongbuk-do	11.6 / 23.7 (-12.1)	18.4 / 28.8 (-10.4)	2.1 / 1.5 (0.6)
	Chungcheongnam-do	11.3 / 24.0 (-12.7)	17.8 / 28.8 (-11.0)	2.0 / 1.6 (0.4)
	Jeollabuk-do	12.3 / 24.7 (-12.4)	18.7 / 29.6 (-10.9)	1.9 / 1.6 (0.3)
	Jeollanam-do	12.6 / 24.2 (-11.6)	18.0 / 28.2 (-10.2)	3.0 / 2.5 (0.5)
	Jeju Island	14.7 / 25.1 (-10.4)	18.4 / 28.1 (-9.7)	3.1 / 2.8 (0.3)
	Gyeongsangnam-do	13.0 / 24.4 (-11.4)	19.6 / 29.4 (-9.8)	1.8 / 1.5 (0.3)
	Gyeongsangbuk-do	12.4 / 23.7 (-11.3)	18.8 / 28.7 (-9.9)	2.3 / 1.7 (0.6)
	Gangwon-do	11.5 / 23.4 (-11.9)	17.6 / 28.2 (-10.6)	2.0 / 1.6 (0.4)

Table S9. Altitudes (m) of monitoring sites in Gangwon-do. Ganseong-eup represents Gosung.

	Name	Latitude	Longitude	Altitude(m)
Gangwon	Jungangno	37.87564	127.72048	110.1613
	Seoksa-dong	37.85707	127.7495	195.0629
	Okcheon-dong	37.76003	128.90297	81.9188
	Jungang-dong	37.35279	127.94746	194.5183
	Bangok-dong	37.3356	127.9771	274.9333
	Ganseong-eup	38.28744	128.38521	586.4231
	Bangsan-myeon	38.22439	127.95856	456.5462
	Bukpyeong-myeon	37.43023	128.66476	631.8139
	Chiaksan	37.36014	128.12509	587.2285

Table S10. Surface and stratospheric O_3 concentrations (O_{3S}*) and their ratio in Korea simulated by CESM. The concentrations and ratios for the altitude of 1 km are shown in parenthesis.

Season	Surface O_3	Stratospheric O_3 (O_{3S})	Ratio (O_{3S}/O_3) (%)
DJF	30.4 (46.2)	23.2 (26.4)	76.2 (57.2)
MAM	45.3 (64.2)	16.7 (21.4)	36.9 (33.3)
JJA	45.9 (53.9)	2.0 (3.0)	4.3 (5.6)
SON	36.3 (51.1)	9.8 (12.1)	26.9(23.8)

*CESM2.2 calculates O_{3S} as a 3-D variable in space. Originally, O_{3S} is O_3 above tropopause. The O_{3S} is transported and undergoes chemical losses below tropopause as

$$O_{3S} = O_{3S} * \exp(-O_{3S}\_Loss).$$

The O_{3S}_Loss rate by chemical reactions in the troposphere is calculated:

O_{3S}_Loss = 2.0*O_O_3 + O1D_H_2O + HO_2_O_3 + OH_O_3 + H_O_3 + 2.0*NO_2_O + 2.0*jno_3_b + 2.0*CLO_O + 2.0*jcl_2o_2 + 2.0*CLO_CLOa + 2.0*CLO_CLOb + 2.0*BRO_CLOb + 2.0*BRO_CLOc + 2.0*BRO_BRO +2.0*BRO_O + CLO_HO_2 + BRO_HO_2 + S_O_3 + SO_O_3 + C_2H_4_O_3 + C_3H_6_O_3 + ISOP_O_3 + MVK_O_3 + MACR_O_3 + MTERP_O_3 + BCARY_O_3.

ISOP=isoprene

MVK= methyl vinyl ketone

MACR=methacrolein

MTERP= pinene_a + carene_3 + thujene_a + 2met_styrene + cymene_p + cymene_o + terpinolene + bornene + fenchene_a + ocimene_al + pinene_b + sabinene + camphene + limonene + phellandrene_a + terpinene_g + terpinene_a + phellandrene_b + myrcene + ocimene_t_b + ocimene_c_b

BCARY= caryophyllene_b + bergamotene_a + bisabolene_b + farnescene_b + humulene_a.

For details in chemical mechanism, refer to Emmons, L. K. et al., 2020 [The chemistry mechanism in the Community Earth System Model Version 2 (CESM2), Journal of Advances in Modeling Earth Systems, https://doi.org/10.1029/2019MS001882].

Table S11. The trends of NOx and CO emissions from linear fits of the data covering 2001-2020.

Stations		NOx (kton/yr) Slope (Correlation Coefficient)	CO (kton/yr) Slope (Correlation Coefficient)
City	Seoul	-2.35 (-0.72)	-8.02 (-0.97)
	Incheon	-1.14 (-0.60)	-0.74 (-0.73)
	Daejeon	-0.56 (-0.84)	-0.84 (-0.88)
	Gwangju	-0.29 (-0.63)	-0.72 (-0.94)
	Busan	-1.23 (-0.77)	-2.01 (-0.94)
	Ulsan	-1.27 (-0.90)	-0.12 (-0.37)
	Daegu	-0.85 (-0.74)	-1.37 (-0.87)
Province	Gyeonggi-do	-1.30 (-0.47)	-1.51 (-0.67)
	Chungcheongbuk-do	0.52 (0.46)	0.40 (0.40)
	Chungcheongnam-do	-5.32 (-0.74)	1.49 (0.93)
	Jeollabuk-do	-0.66 (-0.82)	0.61 (0.53)
	Jeollanam-do	0.74 (0.57)	1.63 (0.75)
	Jeju Island	0.27 (0.64)	0.31 (0.58)
	Gyeongsangnam-do	-5.47 (-0.83)	-0.09 (-0.14)
	Gyeongsangbuk-do	0.78 (0.52)	2.19 (0.76)
	Gangwon-do	0.25 (0.17)	0.95 (0.67)

Table S12. The observed trends of NO_2 and CO concentrations from linear fits of the data covering 2001-2021.

Stations		NO_2 Spring / Summer Slope (Correlation Coefficient)	CO Spring / Summer Slope (Correlation Coefficient)
City	Seoul	-0.77 (-0.85)/-0.72(-0.91)	-7.56(-0.77)/-5.34(-0.83)
	Incheon	-0.37(-0.62)/-0.50(-0.62)	-7.65(-0.71)/-4.64(-0.66)
	Daejeon	-0.10(-0.29)/-0.12(-0.50)	-15.53(-0.79)/-9.71(-0.64)
	Gwangju	-0.51(-0.85)/-0.35(-0.88)	-10.64(-0.81)/-8.00(-0.69)
	Busan	-0.64(-0.89)/-0.49(-0.90)	-12.32(-0.83)/-11.05(-0.81)
	Ulsan	-0.04(-0.08)/-0.06(-0.16)	-4.80(-0.39)/-0.75(0.07)
	Daegu	-0.65(-0.87)/-0.51(-0.89)	-23.49(-0.90)/-19.87(-0.87)
Province	Gyeonggi	-0.41(-0.66)/-0.44(-0.79)	-14.50(-0.95)/-8.82(-0.94)
	Chungcheongbuk	-0.18(0.39)/-0.16(-0.45)	-17.68(-0.78)/-6.49(-0.61)
	Chungcheongnam	-0.10(-0.30)/-0.12(-0.41)	-20.95(-0.76)/-9.33(-0.69)
	Jeollabuk	-0.17(-0.42)/-0.25(-0.65)	-21.33(-0.87)/-15.07(-0.85)
	Jeollanam	-0.21(-0.51)/-0.21(-0.58)	-5.86(-0.53)/-5.32(-0.48)
	Jeju Island	-0.18(-0.38)/-0.16(-0.46)	-10.74(-0.71)/-6.95(-0.50)
	Gyeongsangnam	-0.12(-0.31)/-0.10(-0.40)	-6.76(-0.58)/-3.92(-0.46)
	Gyeongsangbuk	-0.76(-0.89)/-0.49(-0.88)	-27.54(-0.82)/-17.48(-0.78)
	Gangwon	-0.16(-0.50)/-0.20(-0.69)	-15.31(-0.86)/-9.03(-0.71)

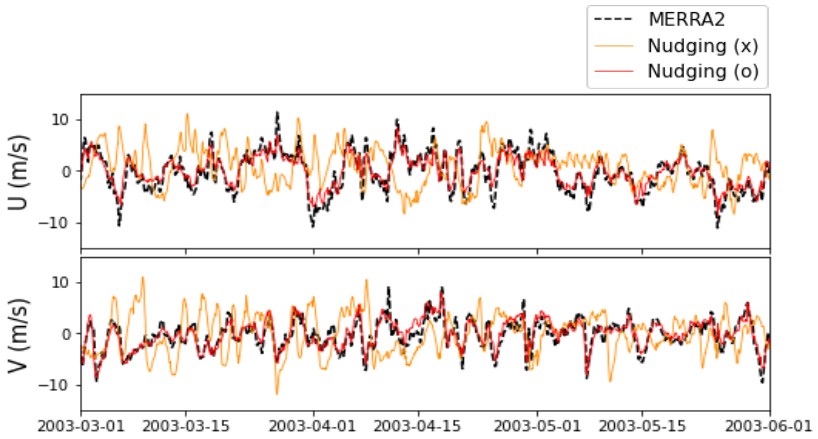

Figure S1. The examples of CAM-Chem U, V wind components for spring, 2003. Without nudging, the model simulated U, V do not closely agree with the MERRA2 data.

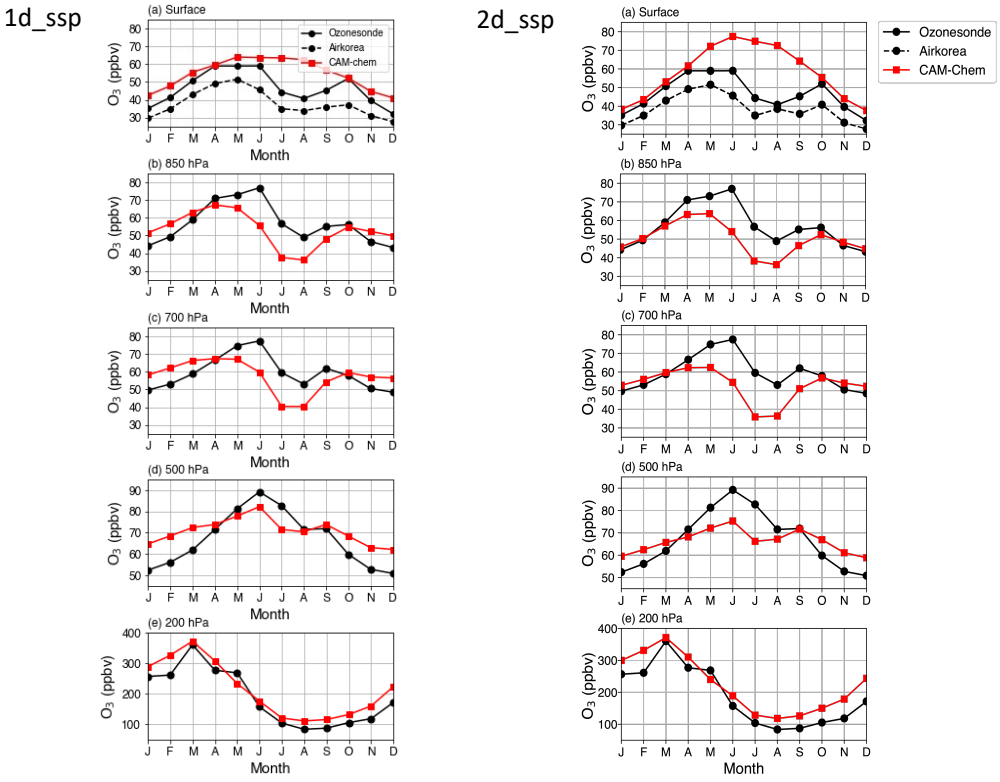

Figure S2. Monthly variations of CAM-Chem simulated ozone concentration (red line) and observed ozone concentration (black solid line) at each pressure level near Pohang, between 14-17 KST. (Left panel) ~1 degree horizontal resolution simulation, and (right panel) ~2 degree horizontal resolution simulation.

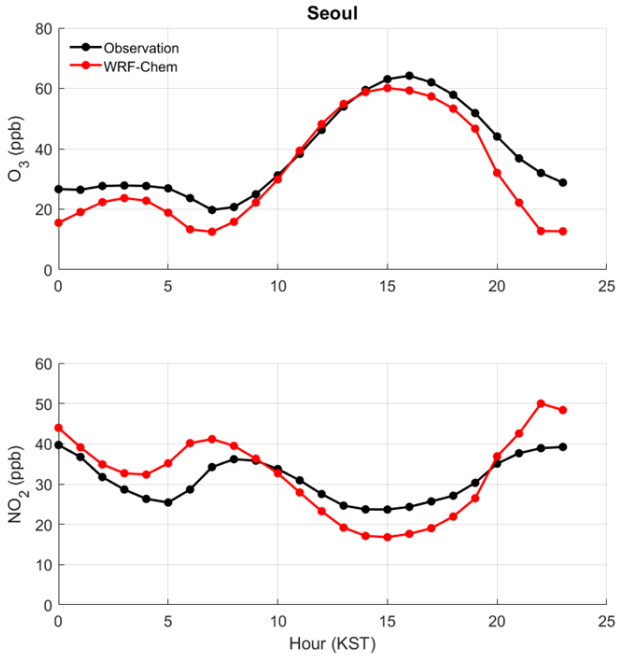

Figure S3. The diurnal variations of observed and simulated O_3 and NO_2 averaged for the simulation period.

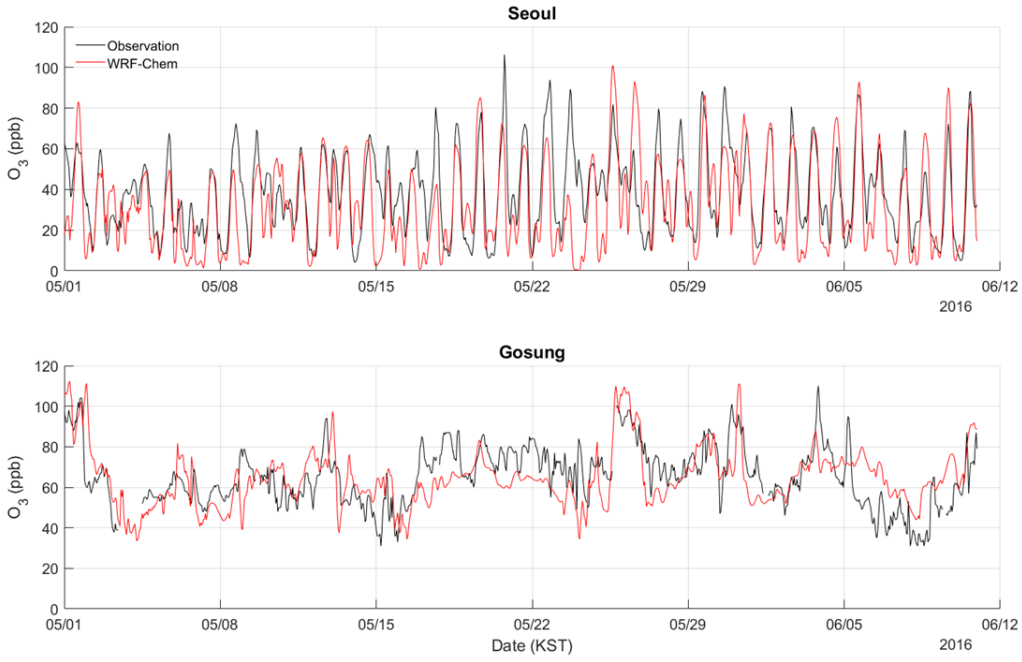

Figure S4. The time series of observed and simulated hourly ozone in (top) Seoul and (bottom) Gosung. Basic statistics are shown as follows. Mean bias (MB): Seoul -6.2 ppb /Gosung -0.9 ppb, Root Mean Square Errors (RMSE): Seoul 18.2 ppb/Gosung 13.7 ppb, Correlation Coefficient(R): Seoul 0.68/Gosung 0.54.

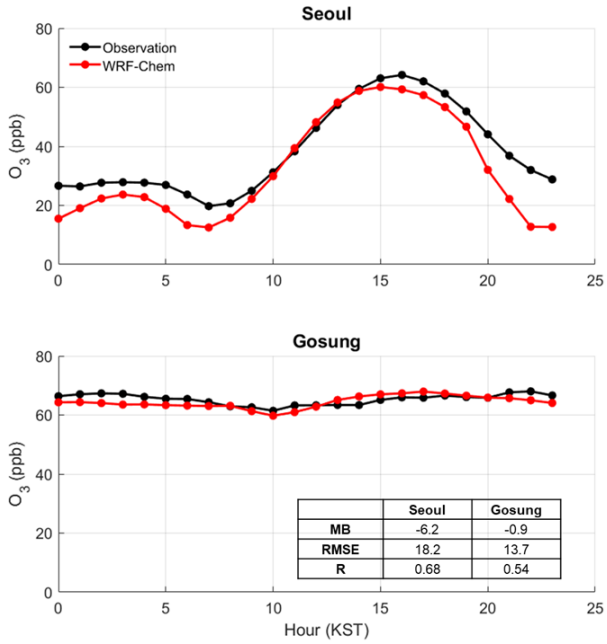

Figure S5. Diurnal variations of observed and simulated ozone concentrations averaged for the entire simulation period: (top) Seoul and (bottom) Gosung. Basic statistics are shown in the plot. MB is mean bias. RMSE is root-mean-square-error. R is correlation coefficient.

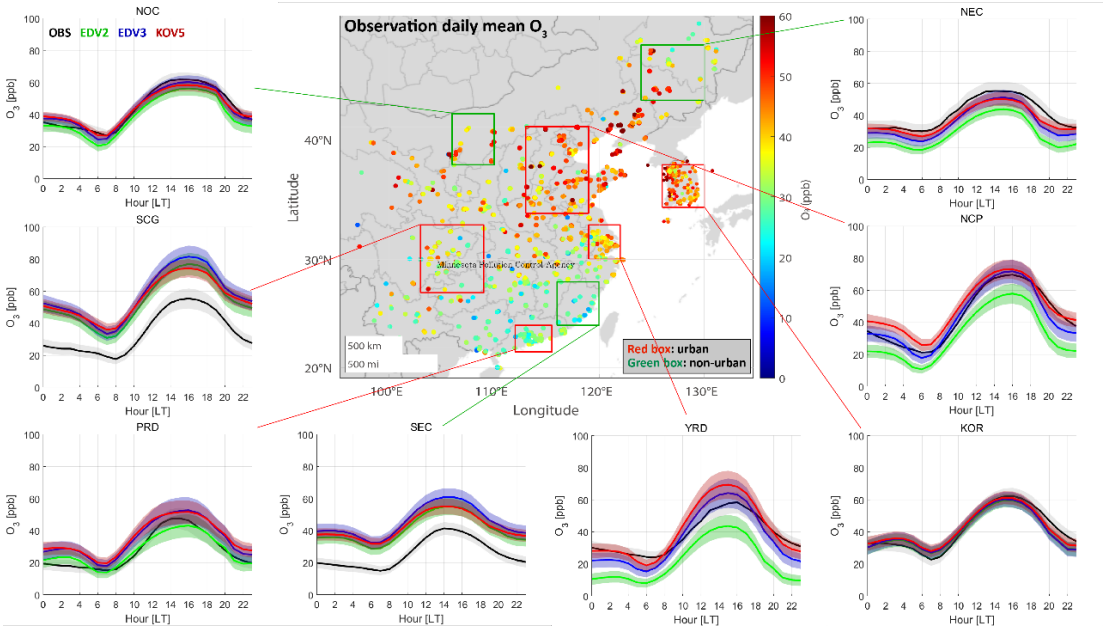

Figure S6. Averaged O_3 from the ground-based observations and model results for regional boxes that distinguish urban (red box) and non-urban (green box) region (central plot). Box averaged diurnal cycle (solid lines) of O_3 and 1/4 of standard deviations (filled area) from observations (black), the WRF-Chem simulations using EDGAR-HTAP version 2 (EDV2, green), EDGAR-HTAP version 3 (EDV3, blue), and KORUS-AQ version 5 (KOV5, red) are shown. The diurnal cycle plots represent Northern China (NOC, 38-42˚N/106-110˚E), Sichuan-Chongqing-Guizhou (SCG, 27-33˚N/103-109˚E), Pearl River Delta (PRD, 21.5-24˚N/112-115.5˚E), Southeastern China (SEC, 24-28˚N/116-120˚E), Yangtze River Delta (YRD, 30-33˚N/119-122˚E), South Korea (KOR, 34.5-38˚N/126-130˚E), North China Plain (NCP, 34-41˚N/113-119˚E), and Northeastern China (NEC, 43-47˚N/124-130˚E).

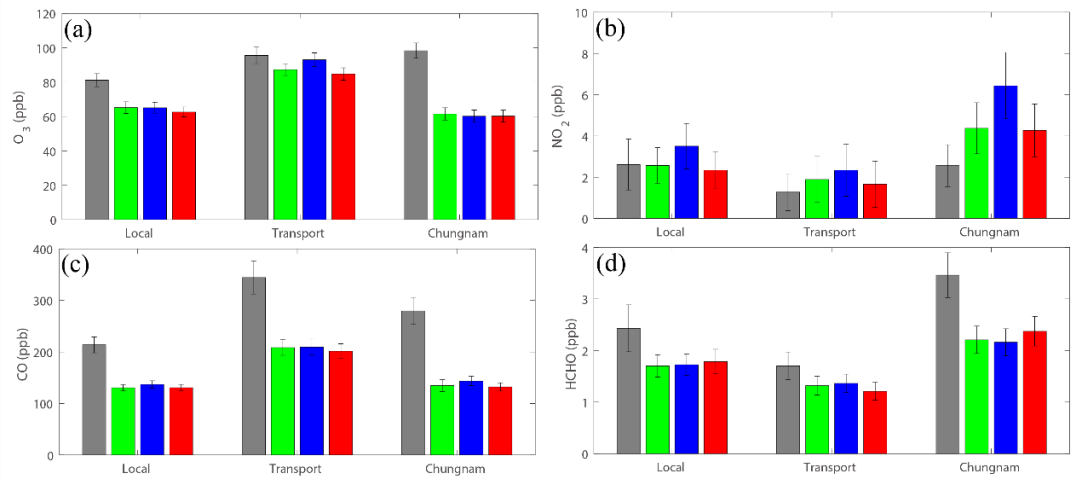

Figure S7. Averaged model and airborne observations of (a) O_3, (b) NO_2, (c) CO, and (d) HCHO (bars) and 1/4 of standard deviations (whiskers) (unit: ppbv) under 2 km height for the Local, Transport, and Chungnam cases from DC-8 (grey), EDV2 (green), EDV3 (blue), and KOV5 (red). The Chungnam (Chungcheongnam-do) region has large point sources like coal-burning power plants and petrochemical facilities that are not well-represented in the bottom-up emission inventories. The local case (May/4, May/20, June/2, June/3) and transport case (May/25, May/26, May/31) represent the dates with the smallest and largest influence from Chinese emissions, respectively. The Chungnam case represents the dates when DC-8 had survey flights targeting the urban and point sources in Chungcheongnam-do and downwind.

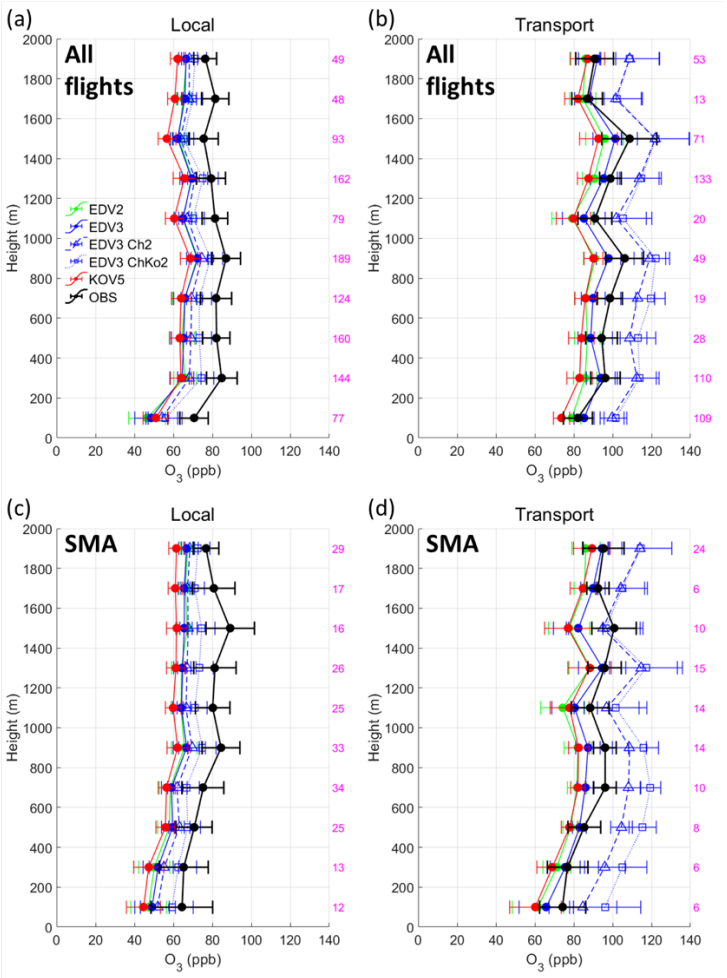

Figure S8. Vertically averaged O_3 from DC-8 (black), EDV2 (green), EDV3 (blue), and KOV5 (red) for the Local and Transport cases under 2 km height above ground level. The 1/2 of standard deviations are represented with black whiskers in each 200m layer. Sensitivity tests are conducted with doubled anthropogenic CO and VOC emissions in China (EDV3_Ch2, blue triangle dots and dashed lines) and in both China and South Korea (EDV3_ChKo2, blue open square and dotted lines). The model results co-located with the observations are sampled and compared with each other. The sampling numbers in the layers are represented with magenta color. (a) and (b) include the data from all flights while (c) and (d) select the data over SMA (Seoul Metropolitan Area).

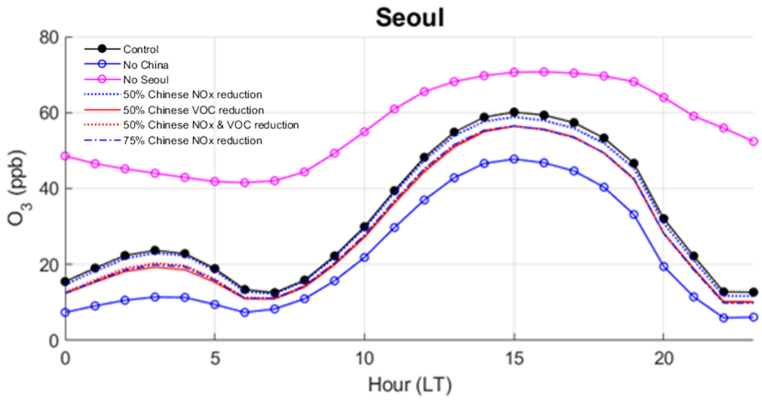

Figure S9. Diurnal variations of the model ozone concentrations at surface from various emission scenarios. The model results were averaged for the full simulation period.

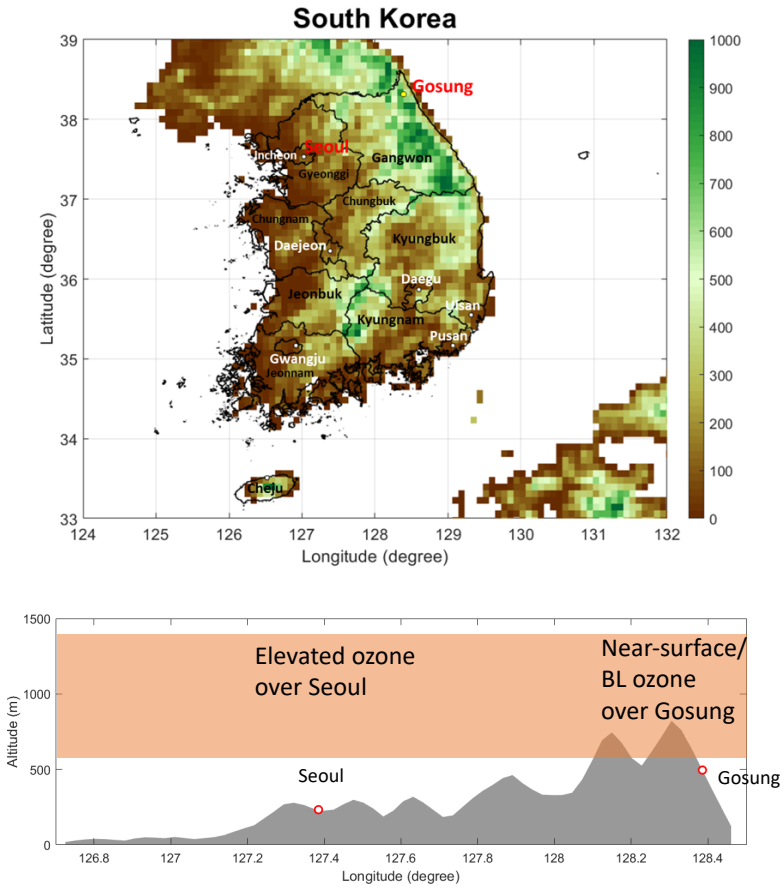

Figure S10. (Top) topography map of South Korea and (bottom) West-East vertical cross-section of terrain connecting Seoul and Gosung. Seoul and Gosung in Gangwon-do are highlighted with color circles. Color bar denotes elevations above sea level (m). A simplified potential ozone transport path is depicted in the bottom plot. Here the ozone layer is colored orange and the terrain is colored gray.

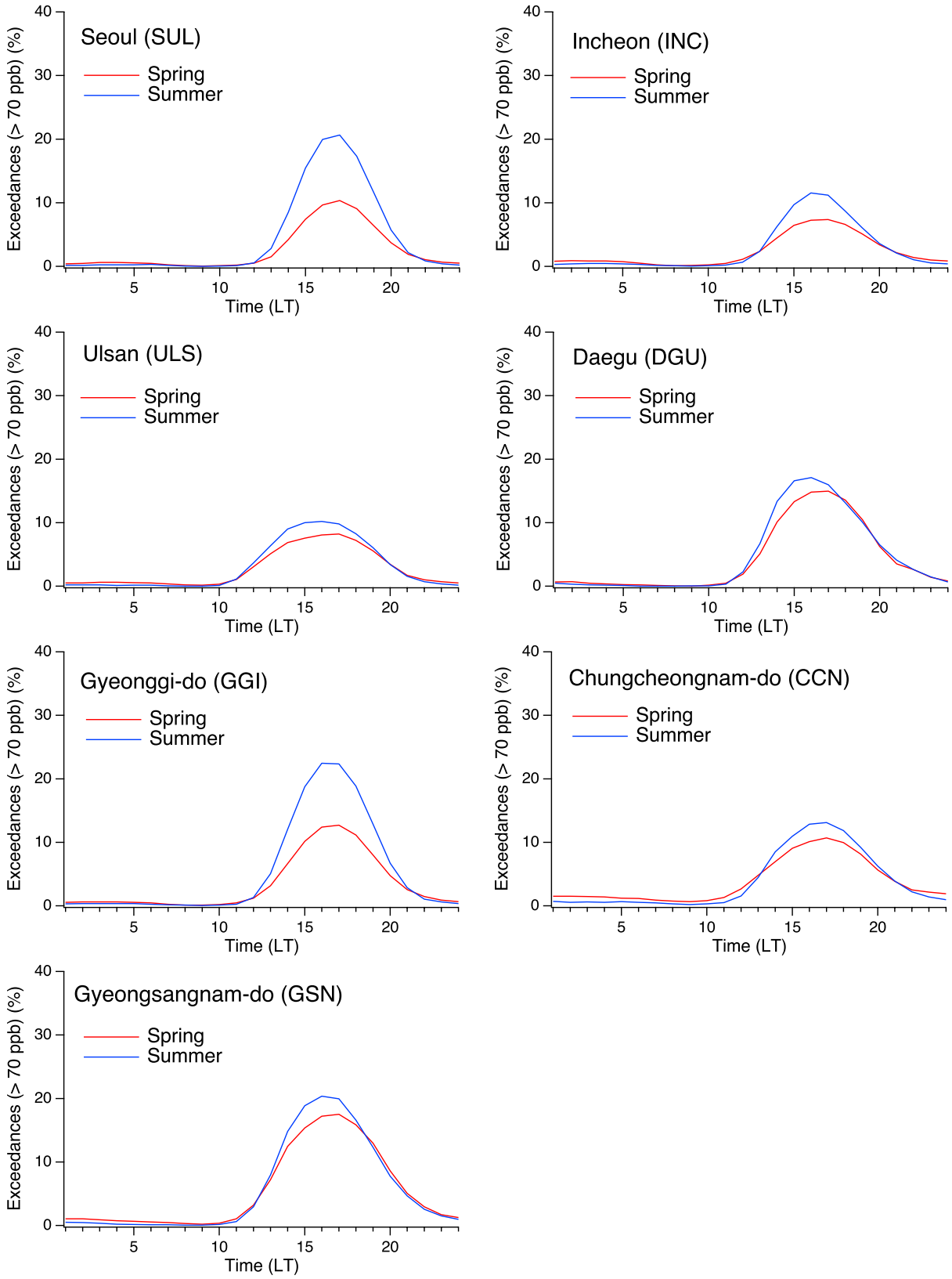

Figure S11. Diurnal O_3 exceedances for the sites that have more exceedances during summer than spring

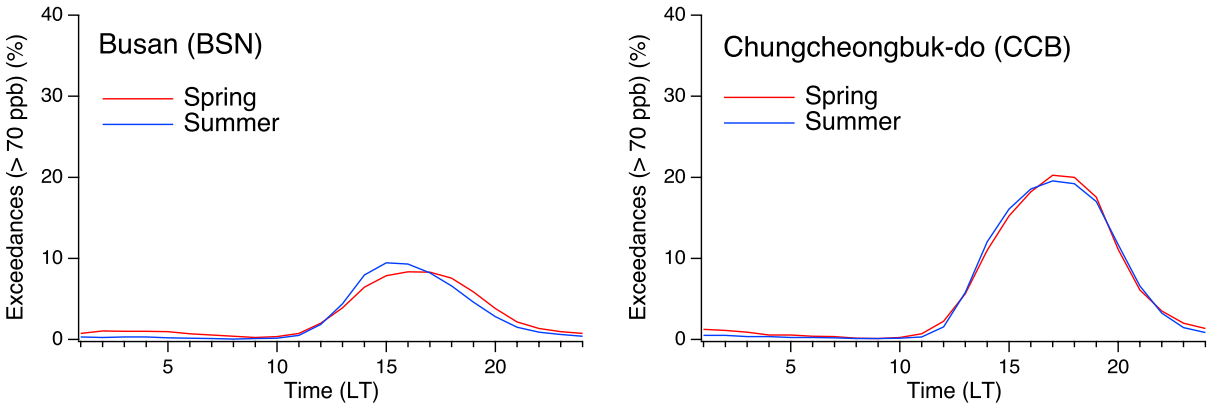

Figure S12. Diurnal O_3 exceedances for the locations that have similar exceedances for spring and summer

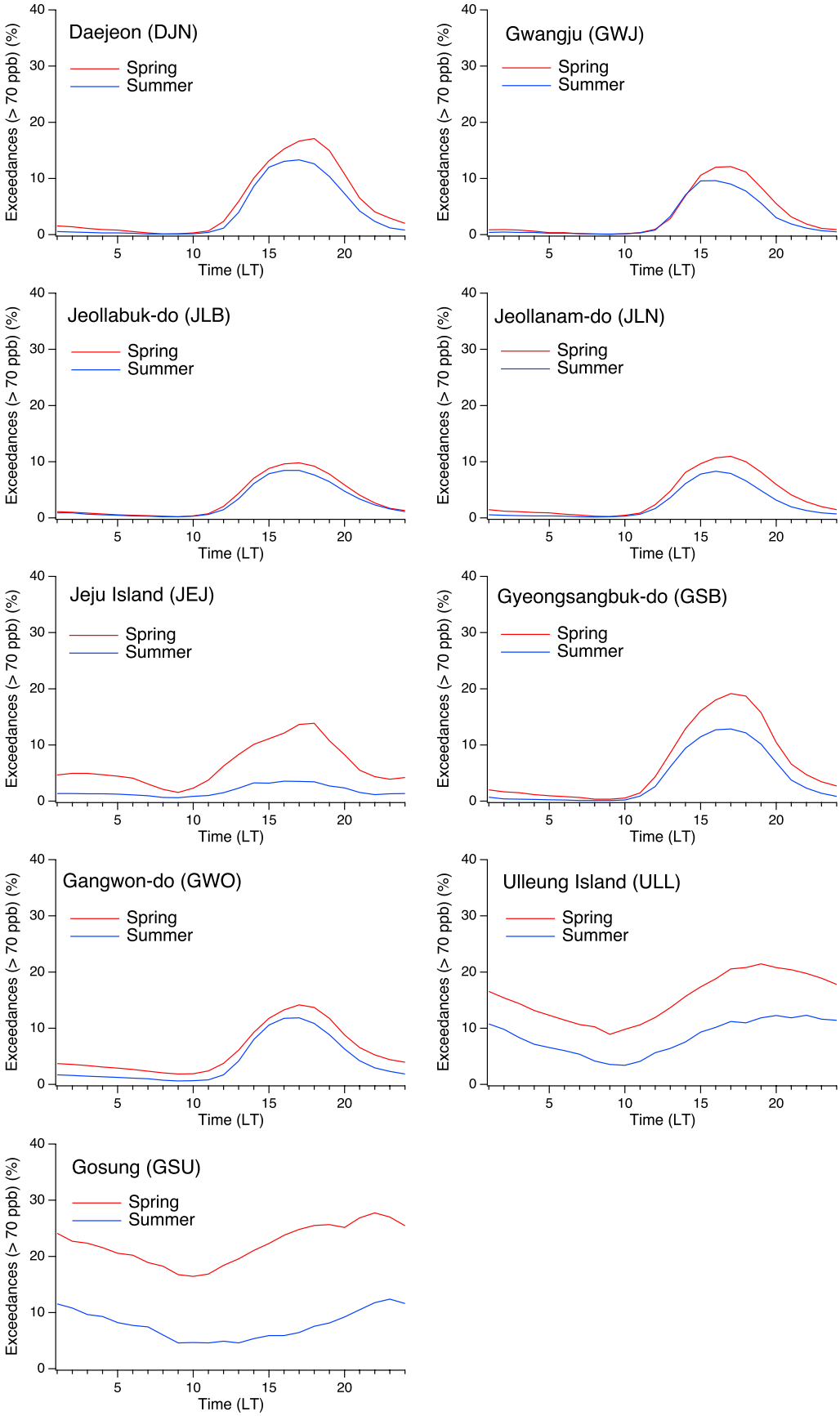

Figure S13. Diurnal O_3 exceedances for the locations that have more exceedances during spring than summer

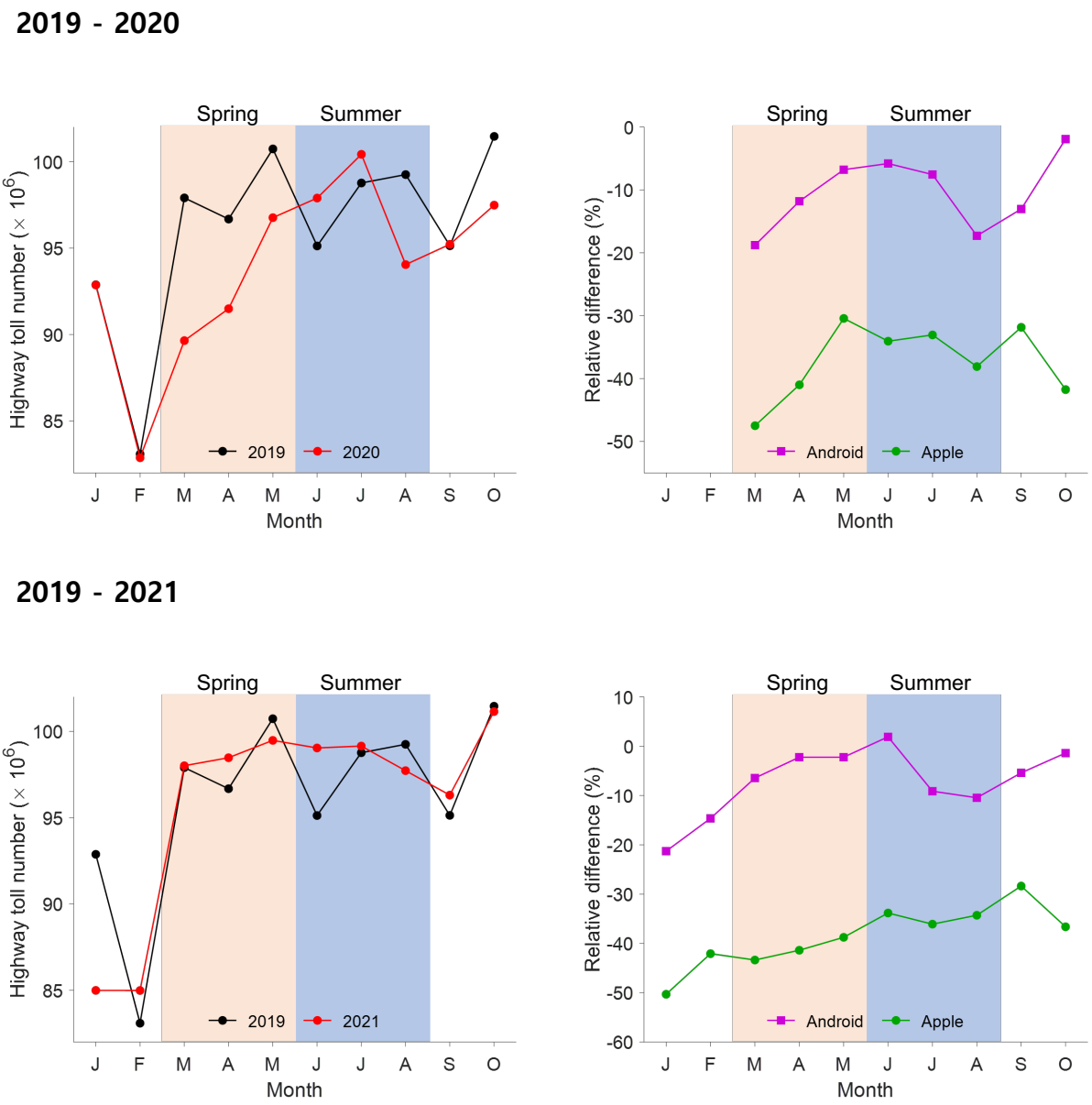

Figure S14. (Left panel) the number of cars passing highway tolls near the Seoul Metropolitan Area (SMA) from January to October in 2019 and 2020 (top) and 2019 and 2021 (bottom) and (Right panel) reductions (%) in mobility based on transit station mobility metrics and driving mobility index in SMA from Google and Apple mobility report, respectively, covering spring and summer in 2020 (top) and 2021 (bottom) relative to 2019.

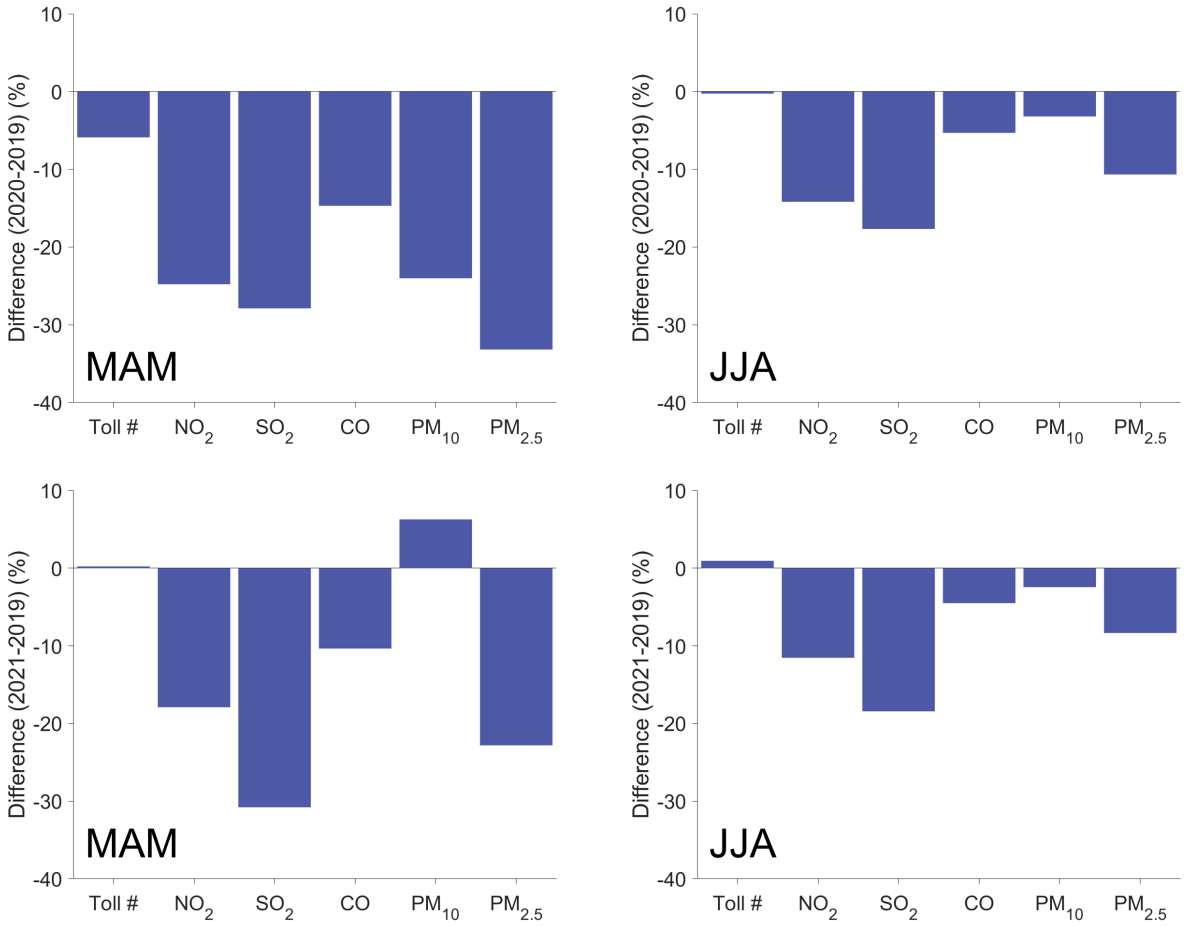

Figure S15. Difference (%) in the toll numbers, NO_2, SO_2, CO, PM_{10}, and $PM_{2.5}$ concentrations in SMA during spring and summer: (top) 2020-2019 and (bottom) 2021-2019.