Sensitivity of the WRF-Chem v4.4 ozone, formaldehyde, and precursor simulations to multiple bottom-up emission inventories over East Asia during the KORUS-AQ 2016 field campaign

Supporting Information

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Table S1. The physics and chemistry schemes that are used in this study

Physics	**Scheme**	**Reference**
Planetary Boundary Layer (PBL)	Yonsei University Scheme (YSU)	Hong and Noh, 2006
Land surface	Unified Noah Land Surface Model	Tewari et al., 2004
Microphysics	Purdue Lin Scheme	Chen and Sun, 2002
Cumulus parameterization	Grell 3D Ensemble	Grell, 1993 Grell and Devenyi, 2002
Chemistry	**Scheme**	**Reference**
Photolysis	Madronich	Madronich, 1987
Gas-phase chemistry	NOAA/ESRL RACM	Stokwell et al., 1997
Aerosols	MADE/VBS	Ackermann et al., 1998 Ahmadov et al., 2012

Table S2. The list of RACM species.

Species	Definition
ISO	Isoprene
SO2	Sulfur dioxide
NO	Nitric oxide
NO2	Nitrogen dioxide
CO	Carbon monoxide
ETH	Ethane
HC3	Alkanes, alcohols, esters, and alkynes with HO rate constant(298 K, 1 atm) less than 3.4 x 10^{-12} cm^3 s^{-1}
HC5	Alkanes, alcohols, esters, and alkynes with HO rate constant(298K , 1 atm) between 3.4 x 10^{-12} and 6.8 x 10^{-12} cm^3 s^{-1}
HC8	Alkanes, alcohols, esters, and alkynes with HO rate constant(298 K, 1 atm) greater than 6.8 x 10^{-12} cm^3 s^{-1}
XYL	Xylene and more reactive aromatics
OL2	Ethene
OLT	Terminal alkenes
OLI	Internal alkenes
TOL	Toluene and less reactive aromatics
CSL	Cresol and other hydroxy substituted aromatics
HCHO	Formaldehyde
ALD	Acetaldehyde and higher aldehydes
KET	Ketones
ORA2	Acetic acid and higher acids
NH3	Ammonia
SULF	Sulfuric acid
PM2.5	Particulate matter under 2.5 μm diameter
PM10	Particulate matter under 10 μm diameter
OC	Organic carbon
BC	Black carbon

Table S3. The area total anthropogenic 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) for each emission data set in May.

Region	Eastern China			South Korea			SMA		
Species	**EDV2**	**EDV3**	**KOV5**	**EDV2**	**EDV3**	**KOV5**	**EDV2**	**EDV3**	**KOV5**
				unit = mol/s					
ISO*	0.0	0.0	31.3	0.0	0.0	2.5	0.0	0.0	0.1
SO2	3627	1991	1648	183	349	165	18	92	10
NO	10063	9034	5482	990	1191	886	196	214	191
NO2	0	0	0	0	0	0	0	0	0
CO	52304	53183	48489	921	3004	2113	268	240	388
ETH	519	715	579	18	16	30	5	5	6
HC3	406	542	545	60	58	45	16	18	10
HC5	508	695	507	66	65	36	18	20	8
HC8	317	435	534	53	54	41	13	16	9
XYL	176	246	270	15	16	41	4	4	9
OL2	1144	1599	1043	62	59	73	16	17	14
OLT	410	573	352	30	29	26	7	8	4
OLI	118	165	312	14	13	27	3	4	6
TOL	294	410	810	27	27	98	6	8	26
CSL	176	246	0	15	16	0	4	4	0
HCHO	96	134	47	15	16	9	4	5	2
ALD	430	599	41	34	34	6	8	10	1
KET	106	144	43	7	6	5	3	2	1
ORA2	0	0	0	0	0	0	0	0	0
NH3	4065	6056	4594	80	395	510	9	30	43
SULF	0	0	0	0	0	0	0	0	0
				unit = kg/s					
PM2.5	98.8	95.2	42.1	2.5	4.3	1.0	0.3	0.8	0.1
PM10	142.3	133.2	96.7	3.6	6.6	7.3	0.4	1.3	1.1
OC	18.0	16.8	13.7	0.2	0.4	1.7	0.0	0.1	0.1
BC	12.5	11.6	8.5	0.7	0.5	0.6	0.1	0.1	0.1

* Note ISO in Table S3 is only from anthropogenic sources. ISO is mainly emitted from biogenic sources using MEGAN.

Table S4. The chemical species of anthropogenic emissions used in RACM chemistry option and their mapping formulas from MOZART chemistry option that is the input format of *anthro_emiss* program

WRF-Chem	EDGAR-HTAP v2 (v3)	KORUS v5
RACM	**MOZART to RACM**	**SAPRC-99 to RACM**
ISO	0	ISOP
SO2	SO2	SO2
NO	NOx	NO + NO2
NO2	0	0
CO	CO	CO
ETH	C2H6[1)]	ALK1[2)]
HC3	0.5*C2H5OH[1)] + CH3OH[1)] + C3H8[1)]	ALK2[2)] + 1.11 x ALK3[2)] + 0.4 x MEOH[2)]
HC5	0.5 x BIGALK[1)] + 0.5 x C2H5OH[1)]	0.97*ALK4[2)]
HC8	0.5 x BIGALK[1)]	ALK5[2)]
XYL	0.2 x TOLUENE[1)]	ARO2[2)]
OL2	C2H4[1)]	ETHE[2)]
OLT	0.3 x BIGENE[1)] + C3H6[1)]	OLE1 + 0.5 x MACR + 0.5 x MVK
OLI	0.4 x BIGENE[1)]	OLE2[2)]
TOL	0.1 x BIGENE[1)] + 0.3 x TOLUENE[1)]	ARO1[2)]
CSL	0.2 x TOLUENE[1)]	PHEN[2)] + CRES[2)]
HCHO	CH2O	HCHO
ALD	0.2 x BIGENE[1)] + CH3CHO[1)] + 0.3 x TOLUENE[*]	CCHO[2)] + RCHO[2)] + BALD[2)] + GLY[2)] + MGLY[2)] + BACL[2)] + 0.5 x MACR[2)]
KET	CH3COCH3[1)] + MEK[1)]	0.3 x ACET[2)] + 1.61 x MEK[2)] + 1.61 x PRD2[2)] + 0.5 x MVK[2)] + IPRD[2)]
ORA2	0	0
NH3	NH3	NH3
PM2.5	PM2.5	PM2.5+PMFINE
PM10	PM10	PM10
OC	OC	POA
SULF	0	SULF
BC	BC	PEC

[1)] Note that those are MOZRT VOC species (Emmons et al., 2010).

[2)] Note that those are SAPRC99 VOC species (Carter, 2000).

Table S5. Comparison of surface meteorological variables from SYNOP and WRF-Chem for the KORUS-AQ campaign period. N is the number of samples. R is correlation coefficient. RMSE is 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 S6. The mapping table from WAS to RACM VOC based on Lu et al., 2013.

RACM	WAS
TOL	**Toluene**, Benzene, Ethylbenzene, i-Propylbenzene, n-Propylbenzene
XYL	**m/p-Xylene**, **o-Xylene**, 1-3-5-Trimethylbenzene, 1-2-4-Trimethylbenzene, 1-2-3-Trimethylbenzene, 4-Ethyltoluene
ETE	Ethene
ISO	Isoprene

Table S7. Comparison of the aircraft-based 1-minuite-interval O_3, NO_2, CO, HCHO, TOL, XYL, ETE, and ISO observations with EDV2, EDV3, and KOV5 in SMA under 2 km height (unit = ppb). N is the number of samples. R is correlation coefficient. RMSE is root-mean-square-error.

Species	Type	N	Mean	Bias	σ	R
O_3	**OBS**	1081	80.9		21.6	
	EDV2		64.0	-16.9	16.3	0.61
	EDV3		66.6	-14.2	16.3	0.63
	KOV5		62.7	-18.1	15.4	0.70
NO_2	**OBS**	1068	4.89		7.53	
	EDV2		5.33	0.44	7.28	0.81
	EDV3		5.55	0.66	7.44	0.80
	KOV5		5.34	0.46	7.56	0.82
CO	**OBS**	1150	247		98	
	EDV2		148	-98	53	0.65
	EDV3		151	-96	49	0.67
	KOV5		145	-102	49	0.71
HCHO	**OBS**	1126	2.65		1.75	
	EDV2		1.89	-0.76	1.24	0.84
	EDV3		1.99	-0.66	1.32	0.82
	KOV5		1.86	-0.78	1.26	0.85
TOL	**OBS**	328	3.12		1.71	
	EDV2		0.63	-2.49	0.43	0.38
	EDV3		0.78	-2.34	0.57	0.36
	KOV5		2.24	-0.88	1.47	0.40
XYL	**OBS**	182	0.76		0.60	
	EDV2		0.31	-0.46	0.25	0.41
	EDV3		0.40	-0.37	0.34	0.41
	KOV5		0.64	-0.12	0.51	0.45
ETE	**OBS**	870	0.33		0.44	
	EDV2		0.79	0.46	0.90	0.71
	EDV3		0.89	0.56	1.07	0.73
	KOV5		0.69	0.35	0.71	0.71
ISO	**OBS**	555	0.10		0.11	
	EDV2		0.28	0.17	0.27	0.40
	EDV3		0.27	0.16	0.26	0.40
	KOV5		0.26	0.16	0.26	0.41

Table S8. Comparison of the aircraft-based 1-minuite-interval O_3, NO_2, CO, HCHO, TOL, XYL, ETE, and ISO observations with EDV2, EDV3, and KOV5 in the Chungnam region under 2 km height (unit = ppb). N is the number of samples. R is correlation coefficient. RMSE is root-mean-square-error.

Species	Type	N	Mean	Bias	σ	R
O_3	**OBS**	560	101.6		17.1	
	EDV2		63.5	-38.1	14.3	0.10
	EDV3		60.2	-41.3	13.7	0.06
	KOV5		62.4	-39.2	14.1	0.11
NO_2	**OBS**	557	3.46		5.07	
	EDV2		4.88	1.43	4.75	0.26
	EDV3		8.09	4.63	6.34	0.30
	KOV5		4.79	1.34	4.79	0.28
CO	**OBS**	578	302		102	
	EDV2		148	-153	49	0.51
	EDV3		157	-145	37	0.59
	KOV5		142	-159	34	0.61
HCHO	**OBS**	579	4.04		2.48	
	EDV2		2.25	-1.79	1.06	0.41
	EDV3		2.18	-1.86	1.02	0.39
	KOV5		2.49	-1.55	1.19	0.44
TOL	**OBS**	130	2.65		2.36	
	EDV2		0.47	-2.18	0.35	0.13
	EDV3		0.50	-2.15	0.35	0.09
	KOV5		1.17	-1.48	0.81	-0.06
XYL	**OBS**	30	1.20		1.01	
	EDV2		0.11	-1.10	0.09	0.01
	EDV3		0.14	-1.06	0.10	0.03
	KOV5		0.17	-1.03	0.21	-0.42
ETE	**OBS**	255	2.08		4.64	
	EDV2		0.53	-1.55	0.50	0.05
	EDV3		0.59	-1.49	0.48	0.06
	KOV5		0.75	-1.33	0.64	0.10
ISO	**OBS**	101	0.06		0.06	
	EDV2		0.06	0.00	0.06	-0.17
	EDV3		0.09	0.04	0.09	-0.10
	KOV5		0.06	0.00	0.06	-0.13

Table S9. Comparison of the aircraft-based 1-minuite-interval O_3, NO_2, CO, and HCHO observations with EDV2, EDV3, and KOV5 in each case distinguished by Chinese contribution to O_3 concentration under 2 km height (unit = ppb). N is the number of samples. R is correlation coefficient. RMSE is root-mean-square-error.

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 ,31)	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
NO_2	Local (5/4,20 , 6/2,3)	OBS	1066	2.62		4.92	
		EDV2		2.57	-0.05	3.45	0.63
		EDV3		3.51	0.89	4.39	0.67
		KOV5		2.34	-0.28	3.62	0.67
	Transport (5/25,26 ,31)	OBS	591	1.28		3.60	
		EDV2		1.89	0.61	4.45	0.85
		EDV3		2.34	1.06	5.02	0.83
		KOV5		1.67	0.39	4.47	0.86
CO	Local (5/4,20 , 6/2,3)	OBS	1225	214		61.7	
		EDV2		130	-83	24.9	0.64
		EDV3		137	-77	27.7	0.64
		KOV5		131	-83	25.0	0.67
	Transport (5/25,26 ,31)	OBS	651	345		128.5	
		EDV2		209	-136	60.7	0.59
		EDV3		209	-135	61.6	0.58
		KOV5		201	-143	57.7	0.58
HCHO	Local (5/4,20 , 6/2,3)	OBS	1177	2.43		1.82	
		EDV2		1.70	-0.73	0.86	0.49
		EDV3		1.72	-0.71	0.84	0.48
		KOV5		1.78	-0.65	0.96	0.54
	Transport (5/25,26 ,31)	OBS	605	1.70		1.08	
		EDV2		1.32	-0.38	0.72	0.74
		EDV3		1.36	-0.34	0.71	0.73
		KOV5		1.21	-0.49	0.69	0.72

Table S10. Comparison of the aircraft-based 1-minuite-interval TOL, XYL, ETE, and ISO observations with EDV2, EDV3, and KOV5 in each case distinguished by Chinese contribution to O_3 concentration under 2 km height. TOL, XYL, ETE, and ISO are defined in regional atmospheric chemical model (RACM) and compared with WAS based on Table S5 (unit = ppb). N is the number of samples. R is correlation coefficient. RMSE is root-mean-square-error.

Species	Case	Type	N	Mean	Bias	σ	R
TOL	**Local** (5/4,20 , 6/2,3)	**OBS**	170	1.67		1.40	
		EDV2		0.35	-1.32	0.25	0.22
		EDV3		0.42	-1.25	0.35	0.19
		KOV5		1.14	-0.53	1.00	0.03
	Transport (5/25,26 ,31)	**OBS**	72	1.99		1.48	
		EDV2		0.49	-1.50	0.37	0.54
		EDV3		0.59	-1.41	0.50	0.55
		KOV5		1.64	-0.35	1.42	0.57
XYL	**Local** (5/4,20 , 6/2,3)	**OBS**	77	0.76		0.85	
		EDV2		0.13	-0.63	0.17	0.21
		EDV3		0.18	-0.58	0.24	0.23
		KOV5		0.26	-0.50	0.40	0.15
	Transport (5/25,26 ,31)	**OBS**	30	0.70		0.42	
		EDV2		0.28	-0.42	0.22	0.23
		EDV3		0.37	-0.33	0.30	0.24
		KOV5		0.60	-0.10	0.48	0.19
ETE	**Local** (5/4,20 , 6/2,3)	**OBS**	535	0.75		2.78	
		EDV2		0.44	-0.30	0.51	-0.01
		EDV3		0.52	-0.23	0.66	0.02
		KOV5		0.49	-0.26	0.49	0.09
	Transport (5/25,26 ,31)	**OBS**	290	0.24		0.38	
		EDV2		0.37	0.13	0.60	0.65
		EDV3		0.39	0.15	0.70	0.65
		KOV5		0.31	0.06	0.51	0.65
ISO	**Local** (5/4,20 , 6/2,3)	**OBS**	352	0.08		0.09	
		EDV2		0.16	0.07	0.20	0.32
		EDV3		0.17	0.09	0.20	0.31
		KOV5		0.16	0.07	0.20	0.34
	Transport (5/25,26 ,31)	**OBS**	76	0.10		0.10	
		EDV2		0.18	0.08	0.19	0.56
		EDV3		0.19	0.08	0.18	0.58
		KOV5		0.17	0.06	0.17	0.55

Table S11. Comparison of the aircraft-based 1-minuite-interval O_3, NO_2, CO, HCHO, TOL, XYL, ETE, and ISO observations with EDV3_Ch2 and EDV3_ChKo2 in each case. The sampling number (N), mean, mean bias compared to DC-8 observations, standard deviations (σ), and correlation coefficient (R) with observations are presented (unit = ppb).

Species	Case	Type	N	Mean	Bias	σ	R
O_3	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	1125	68.5	-12.7	12.3	0.6
		EDV3_ChKo2		72.3	-8.9	14.3	0.68
	Transport (5/25,26 ,31)	**EDV3_Ch2**	605	111.0	15.4	23.4	0.65
		EDV3_ChKo2		112.6	17.0	23.1	0.66
NO_2	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	1066	3.30	0.68	4.31	0.7
		EDV3_ChKo2		3.05	0.44	3.95	0.67
	Transport (5/25,26 ,31)	**EDV3_Ch2**	591	2.09	0.81	4.75	0.83
		EDV3_ChKo2		1.92	0.64	4.17	0.84
CO	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	1225	158	-56	43	0.7
		EDV3_ChKo2		176	-38	53	0.65
	Transport (5/25,26 ,31)	**EDV3_Ch2**	651	331	-13	122	0.56
		EDV3_ChKo2		339	-6	122	0.56
HCHO	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	1177	1.82	-0.61	0.87	0.5
		EDV3_ChKo2		2.13	-0.30	1.17	0.51
	Transport (5/25,26 ,31)	**EDV3_Ch2**	605	1.78	0.08	0.98	0.69
		EDV3_ChKo2		1.94	0.24	1.33	0.72
TOL	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	170	0.46	-2.33	0.40	0.1
		EDV3_ChKo2		0.79	-2.00	0.65	0.18
	Transport (5/25,26 ,31)	**EDV3_Ch2**	72	0.68	-2.20	0.46	0.50
		EDV3_ChKo2		1.14	-1.75	0.90	0.55
XYL	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	77	0.19	-0.64	0.25	0.2
		EDV3_ChKo2		0.31	-0.52	0.42	0.23
	Transport (5/25,26 ,31)	**EDV3_Ch2**	30	0.35	-0.43	0.30	0.22
		EDV3_ChKo2		0.61	-0.17	0.53	0.23
ETE	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	535	0.62	-0.13	0.92	0.0
		EDV3_ChKo2		0.87	0.12	1.17	0.00
	Transport (5/25,26 ,31)	**EDV3_Ch2**	290	0.51	0.26	0.69	0.65
		EDV3_ChKo2		0.74	0.50	1.22	0.67
ISO	**Local** (5/4,20 , 6/2,3)	**EDV3_Ch2**	352	0.17	0.08	0.20	0.3
		EDV3_ChKo2		0.15	0.06	0.17	0.33
	Transport (5/25,26 ,31)	**EDV3_Ch2**	76	0.16	0.06	0.16	0.57
		EDV3_ChKo2		0.14	0.03	0.14	0.55

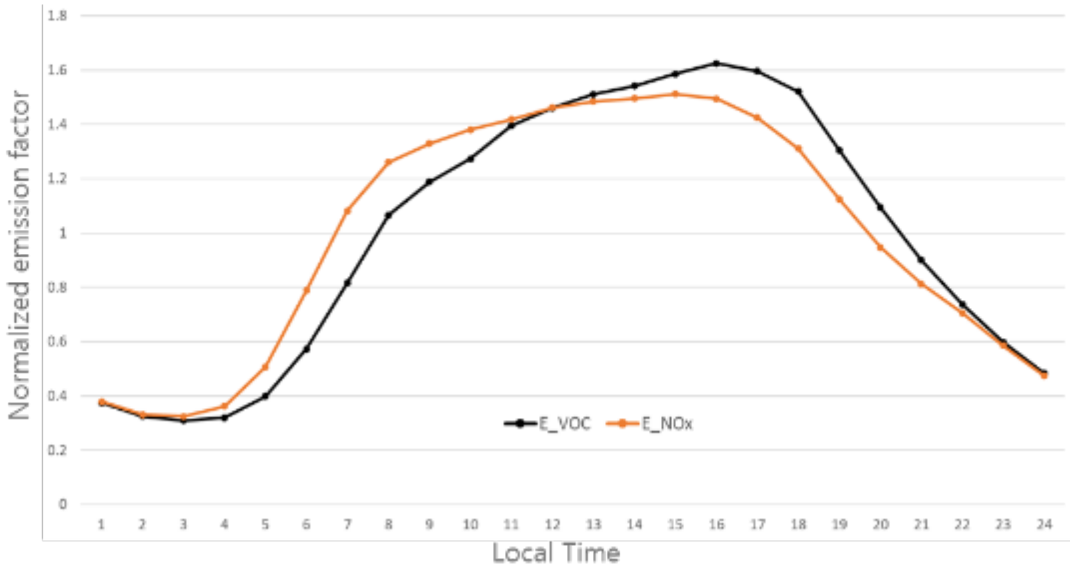

Figure S1. Diurnal emission factors of VOC and NO_x.

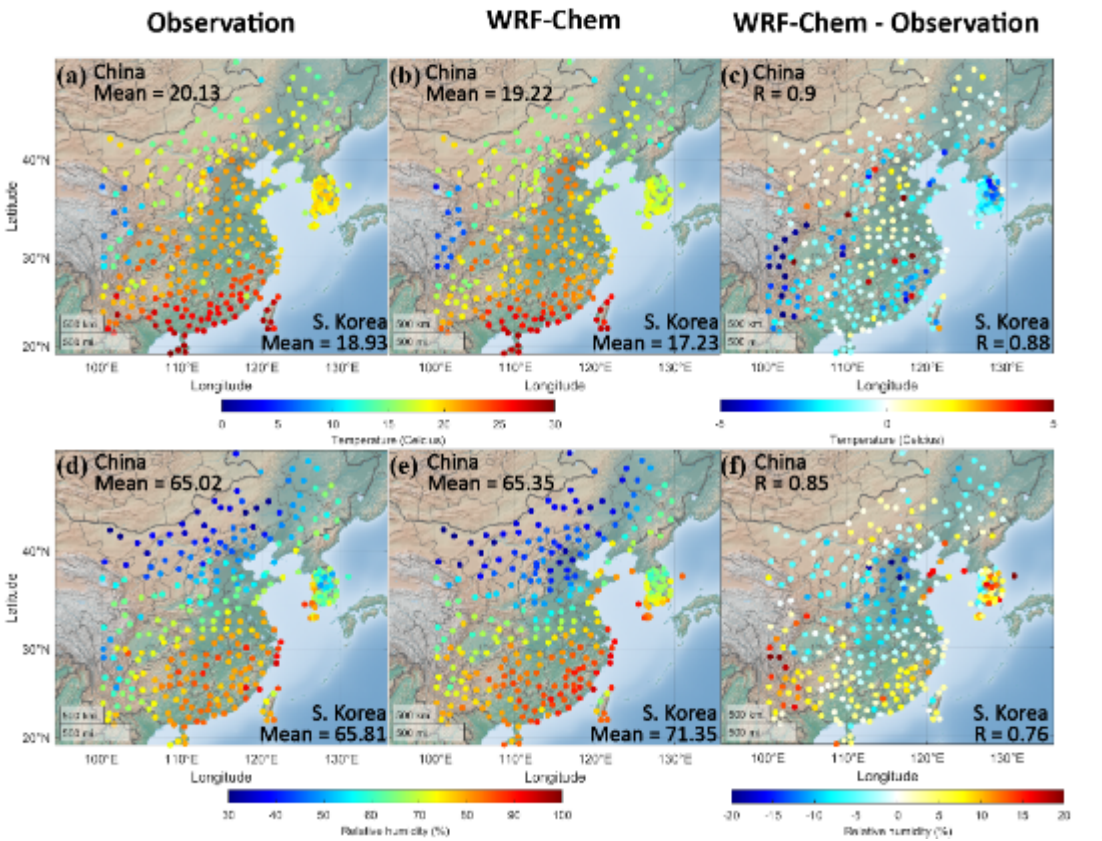

Figure S2. Averaged surface temperature (unit: ˚C) and relative humidity (unit: %) from (a, d) ground-based observations (SYNOP) and (b, e) the weather research and forecast (WRF) model coupled with chemistry (WRF-Chem) from 1st May to 10th June for each station and countries. The differences and correlation coefficients between averaged observations and WRF-Chem are shown (c, f).

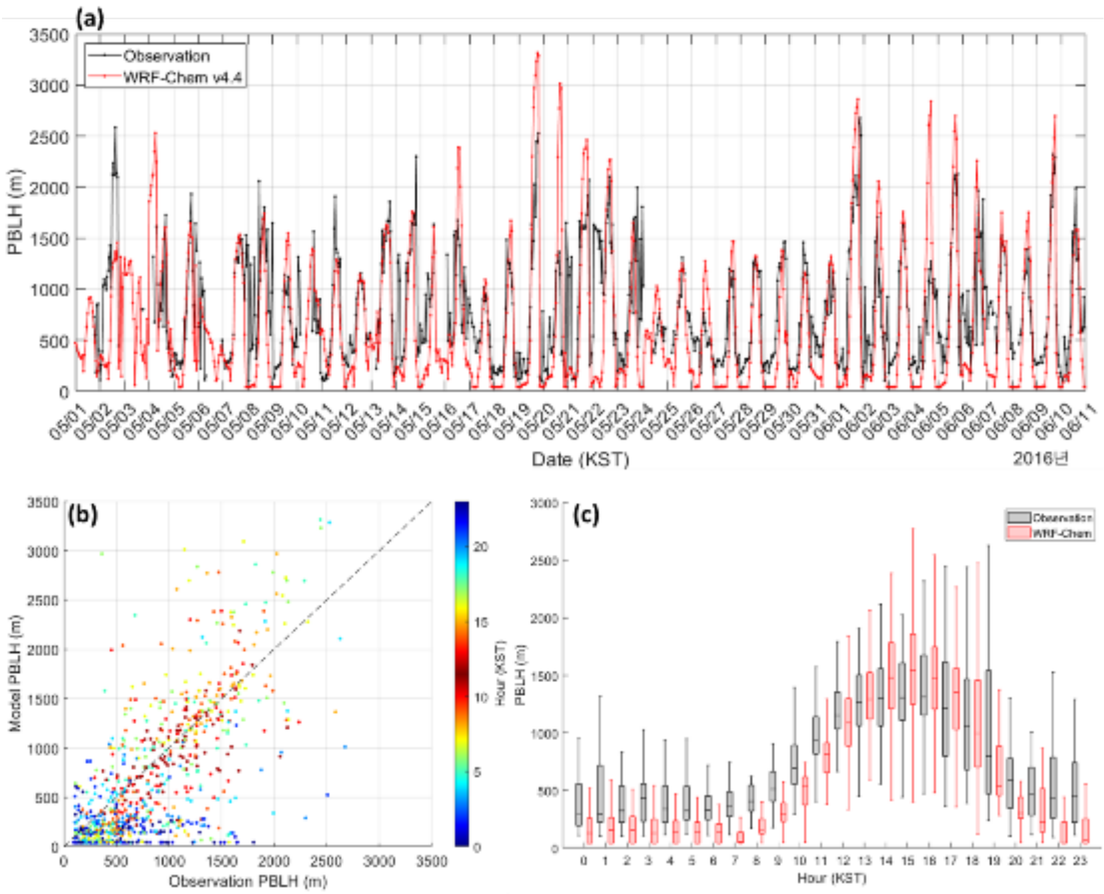

Figure S3. Comparison of PBL heights derived from the ceilometer at Yonsei University (37.564˚N, 126.935˚E) with the WRF-Chem results during the KORUS-AQ campaign period: (a) time series of planetary boundary layer height from ceilometer and WRF-Chem (unit = m), (b) scatter plot of which x axis is ceilometer and y axis is WRF-Chem PBL height, (c) comparison of diurnal variations of PBL heights from ceilometer (grey) and WRF-Chem (red) with box whisker plot.

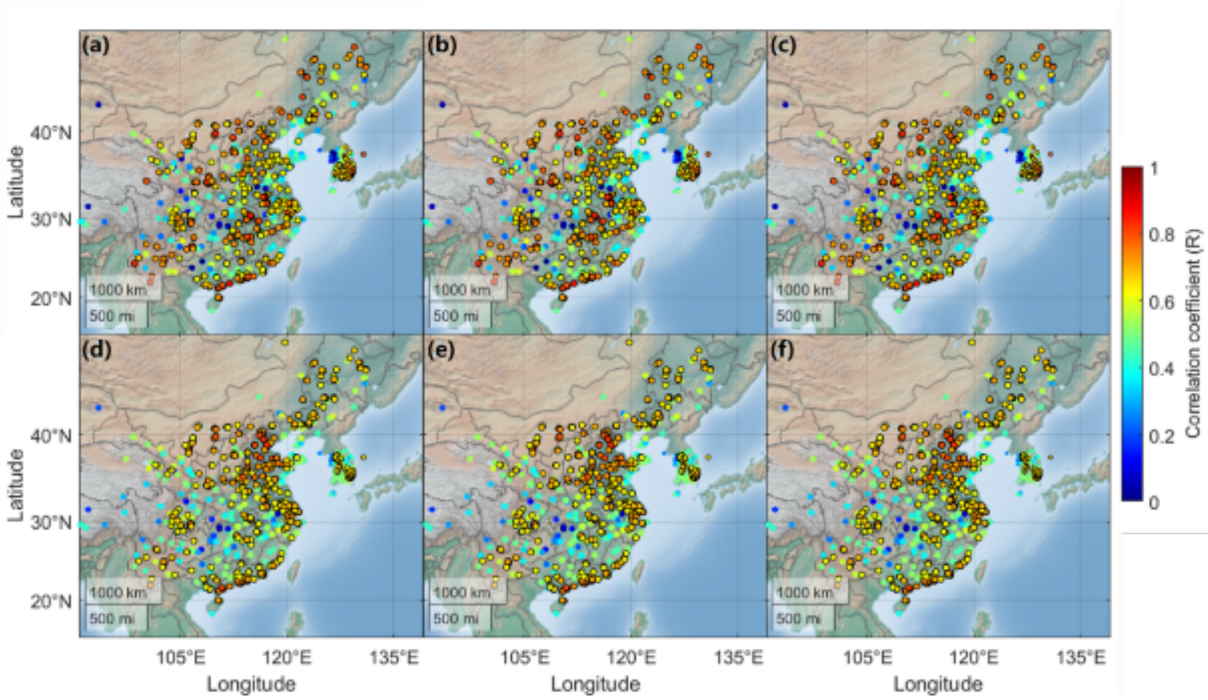

Figure S4. Correlation coefficient (R) between observed and simulated (a-c) MDA8 O_3 and (d-f) hourly O_3 with (a, d) EDV2, (b, e) EDV3, and (c, f) KOV5 emissions. The observation sites with R greater than 0.6 are indicated by a black circle.

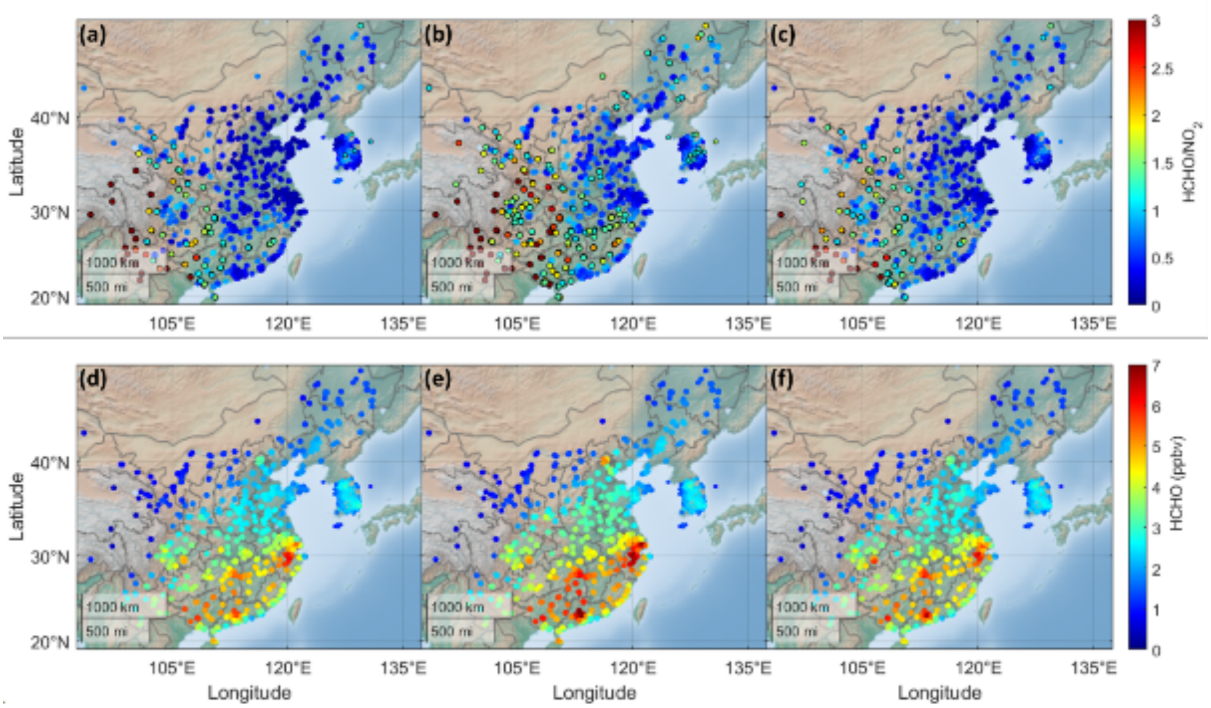

Figure S5. (a-c) Simulated surface HCHO to NO_2 ratio (FNR) and (d-f) HCHO concentrations with (a, d) EDV2, (b, e) EDV3, and (c, f) KOV5 emissions for 14-16 LST. FNR greater than 1 is marked with black circles. The simulated FNR and HCHO are linearly interpolated to ground-based observation sites.

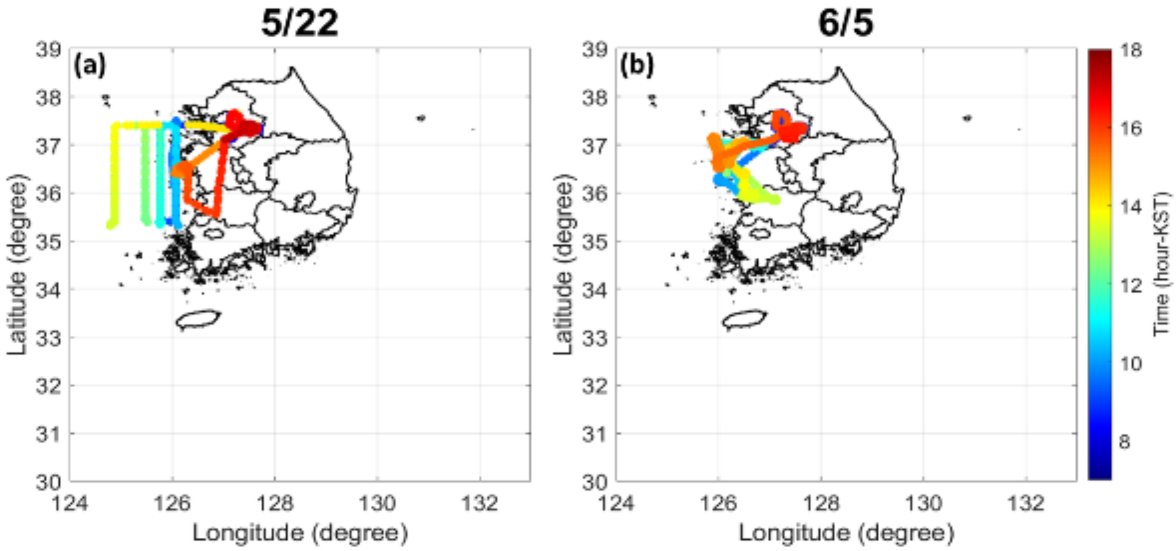

Figure S6. The DC-8 flight tracks on the 22nd May and 5th June.

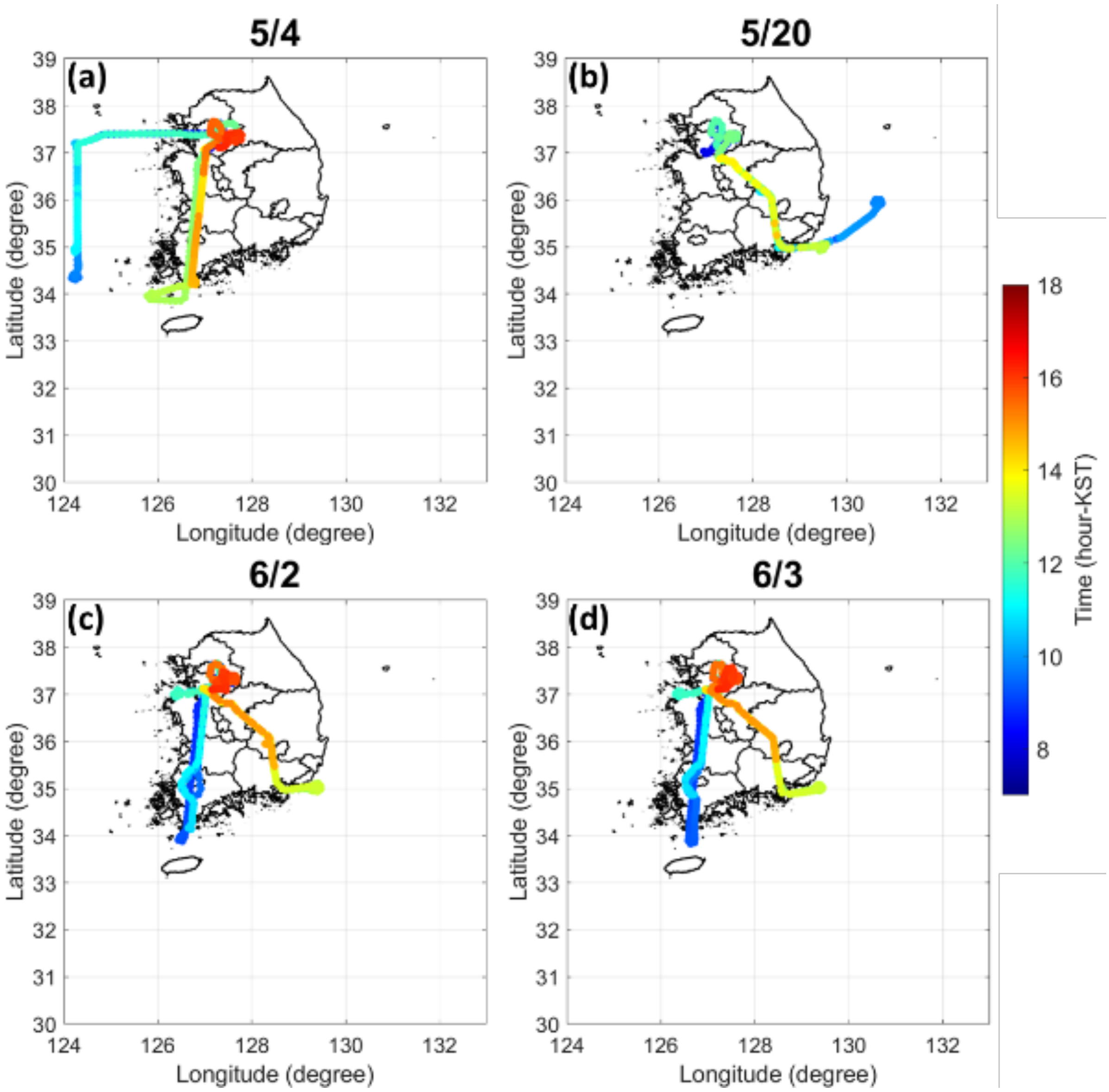

Figure S7. The DC-8 flight tracks on the 4th, 20th May and 2nd, 3rd June (Local case).

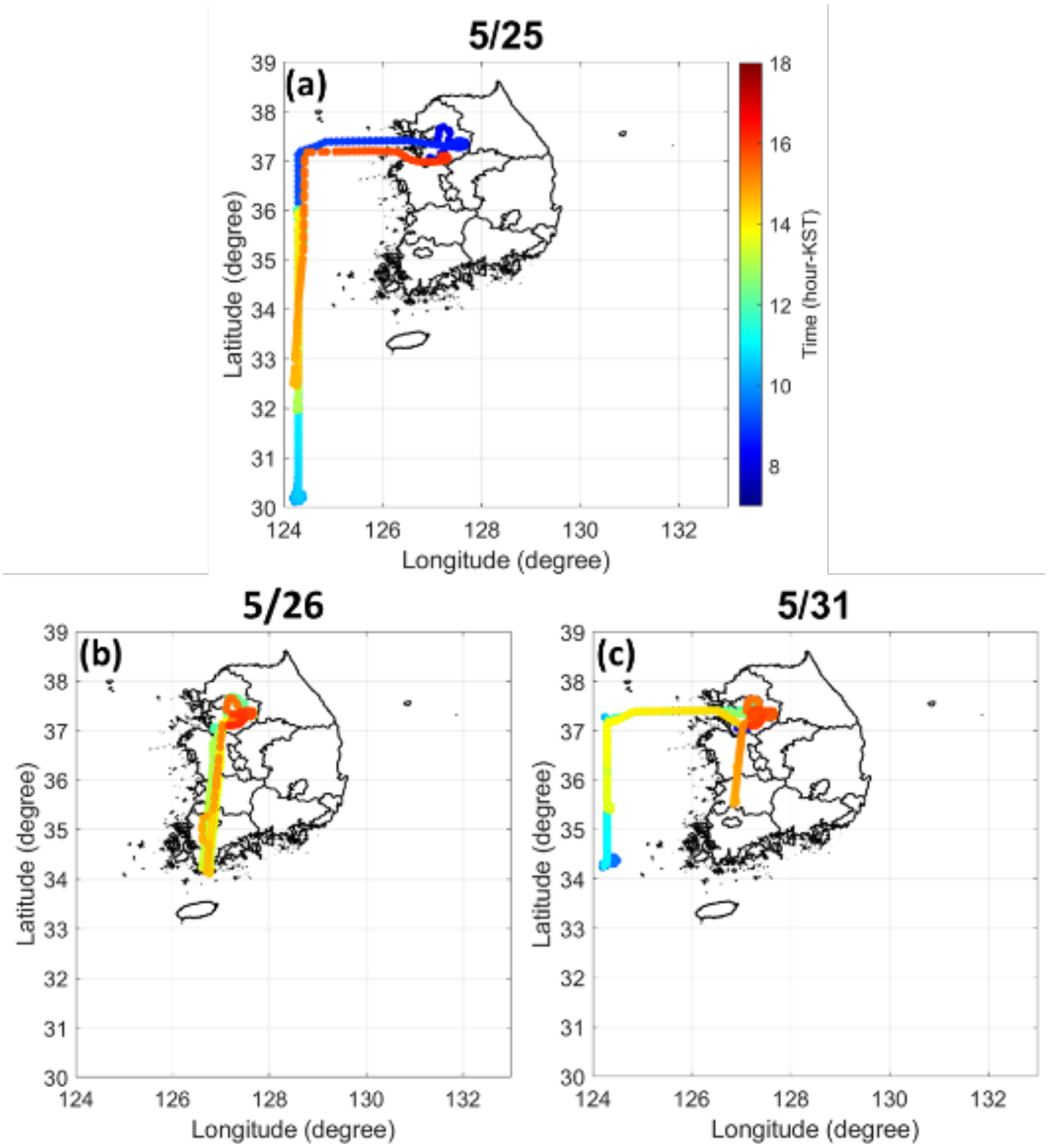

Figure S8. The DC-8 flight tracks on the 22nd, 27th, 31st May (Transport case).

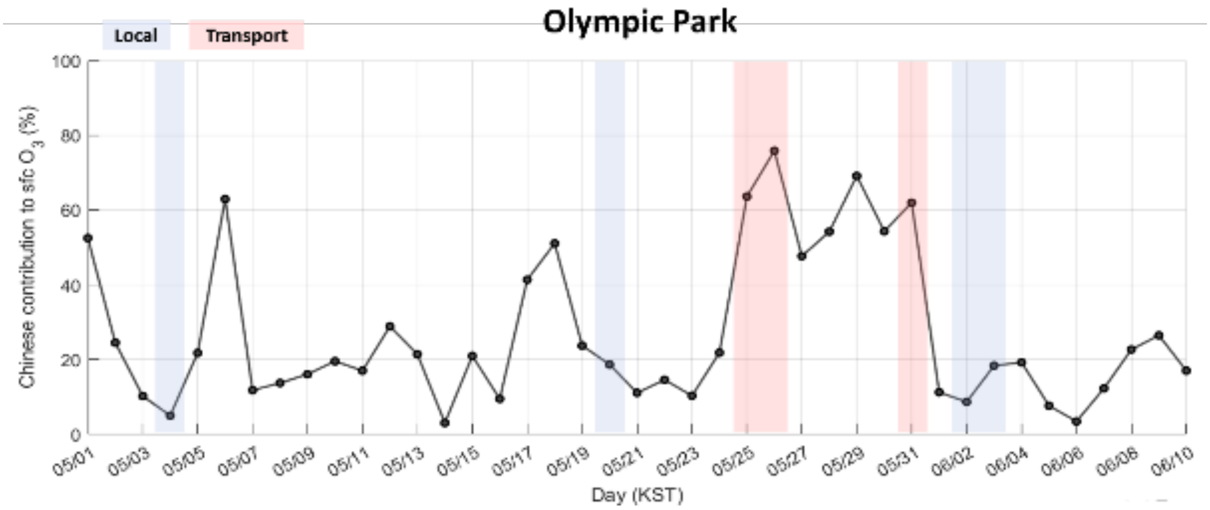

Figure S9. The contribution of Chinese emissions (%) to daily surface O_3 concentrations at Olympic Park obtained from the EDV3 simulations with/without Chinese anthropogenic emissions.