Supplementary

Recent decreasing trends in surface $PM_{2.5}$ over East Asia in the winter-spring season: Different responses to emissions and meteorology between upwind and downwind regions

Young-Hee Ryu[1], Seung-Ki Min[1*], Alma Hodzic[2]

[1]Division of Environmental Science and Engineering, Pohang University of Science and Technology (POSTECH), Pohang, South Korea
[2]National Center for Atmospheric Research (NCAR), Boulder, USA

Text S1. Estimation of emissions of CO, NO_X, SO_2, NMVOC, and NH_3

The reduction amounts of other gases are determined in a similar manner as those for PM emissions. The trends from REASv3.1 and our assumptions are similar to those estimated by Zheng et al. (2018) except for CO. It if found from our simulations that CO is largely underestimated both over South Korea and China (not shown), and thus CO emissions are increased by a factor of 3 over South Korea during 2013–2018 and CO emissions during 2016–2018 remain the same as in 2015 over China. A future study regarding CO would be required, but it is out of scope of this study. Similarly, NO_X emissions are found to be too low over South Korea, and hence they are increased by 60% from the original NO_X emissions in REASv3.1 during 2013–2015 and for the later period the NO_X emissions in 2015 are increased by 50% in 2016, 40% in 2017, and 30% in 2018. The adjustment of NO_X emissions is based on a study by (Oak et al., 2019); they demonstrated that the increase in NO_X emissions by 50% during the 2016 KORUS-AQ field campaign best reproduces O_3 production characteristics. The ratios of NO_X emissions applied for 2017 and 2018 are roughly estimated based on surface observation data of NO_2. A recent study of Ryu and Min (2021) has constructed gridded pollutant concentration data at horizontal resolutions of 0.25°×0.25° using surface observations over South Korea. The observed NO_2 concentration averaged over Seoul is 27.78 ppb in 2016, 26.11 ppb in 2017, and 24.41 ppb in 2018. Given that 1.5 factor (corresponding to 50% increase) is applied to NO_X emissions in 2016 relative to the NO_X emissions in 2015, approximately 1.4 factor in 2017 (26.11 ppb/27.78 ppb × 1.5 = 1.43) and 1.3 factor in 2018 (24.41 ppb/27.78 ppb × 1.5 = 1.34) are applied. NH_3 emissions over South Korea are increased by a factor of 3 to reproduce NH_3 concentrations observed in Seoul of ~9 ppb in winter and 12–16 ppb in spring by (Phan et al., 2013). The monthly NH_3 concentration averaged over the SMA in our simulations ranges from 9 to 11 ppb in February and from 12 to 15 ppb in May. Similar approach that utilizes surface observations was also used to estimate SO_2 emissions over East Asia by Bae et al. (2020).

Text S2. PM speciation

The $PM_{2.5}$ allocation values are obtained by using SPECIATE version 5.1 developed by EPA (https://www.epa.gov/air-emissions-modeling/speciate). We choose simplified PM profiles in SPECIATE under all sectors except for transport sector. For example, for industry sector, we choose “Draft Cast Iron Cupola” (profile code = 92012), “Draft Ferromanganese Furnace” (92027), “Draft Pulp & Paper” (92061), and many more. Each profile provides weight percentages among which are elemental carbon (EC), organic carbon (OC), nitrate, sulfate, other PM (non-speciated PM). We average those weight percentages over all profiles. Because the REASv3.1 inventory explicitly provides EC (BC) and OC, the weight percentages of EC and OC obtained from SPECIATE profiles are excluded and then the weight percentages of remaining species (sulfate, nitrate, ammonium, and PM others) are normalized again.

For transport sector, only two profile codes (92122: Onroad Gasoline Exhaust–Simplified and 8993VBS: Light-duty Gasoline Vehicles Exhaust–Stabilized Running) are employed assuming that those are representatives of transport sector profiles. We take EC and OC weight percentages from 92122 code, which are 19.0% and 54.9%, respectively. The simplified PM profiles of SPECIATE (e.g., 92122 code) does not provide weight percentages for ammonium. Therefore, the weight percentages from 8993 VBS code are used for sulfate (7.2%), ammonium (2.78%), and nitrate (0.29%). The remaining percentage (15.83%) is assigned to other PM. As done for residential, industry, and power sectors, EC and OC percentages for transport sector are excluded because the REASv3.1 directly provides EC (BC) and OC. Then, the weight percentages for species other than EC and OC are re-normalized.

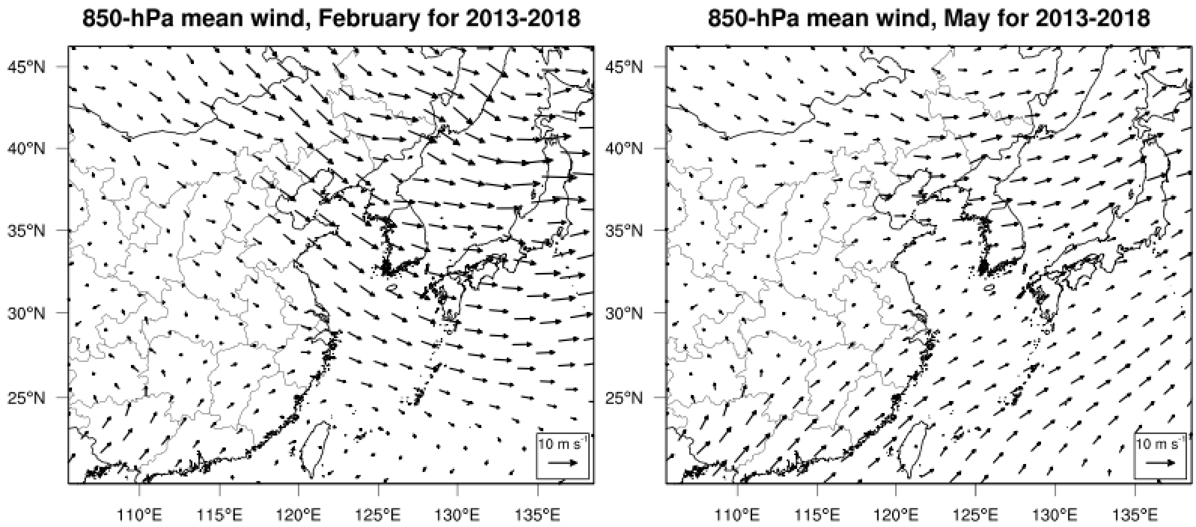


Figure S1. Six-year (2013–2018) mean 850-hPa wind fields obtained from the ERA-5 reanalysis in (left) February and (right) May.

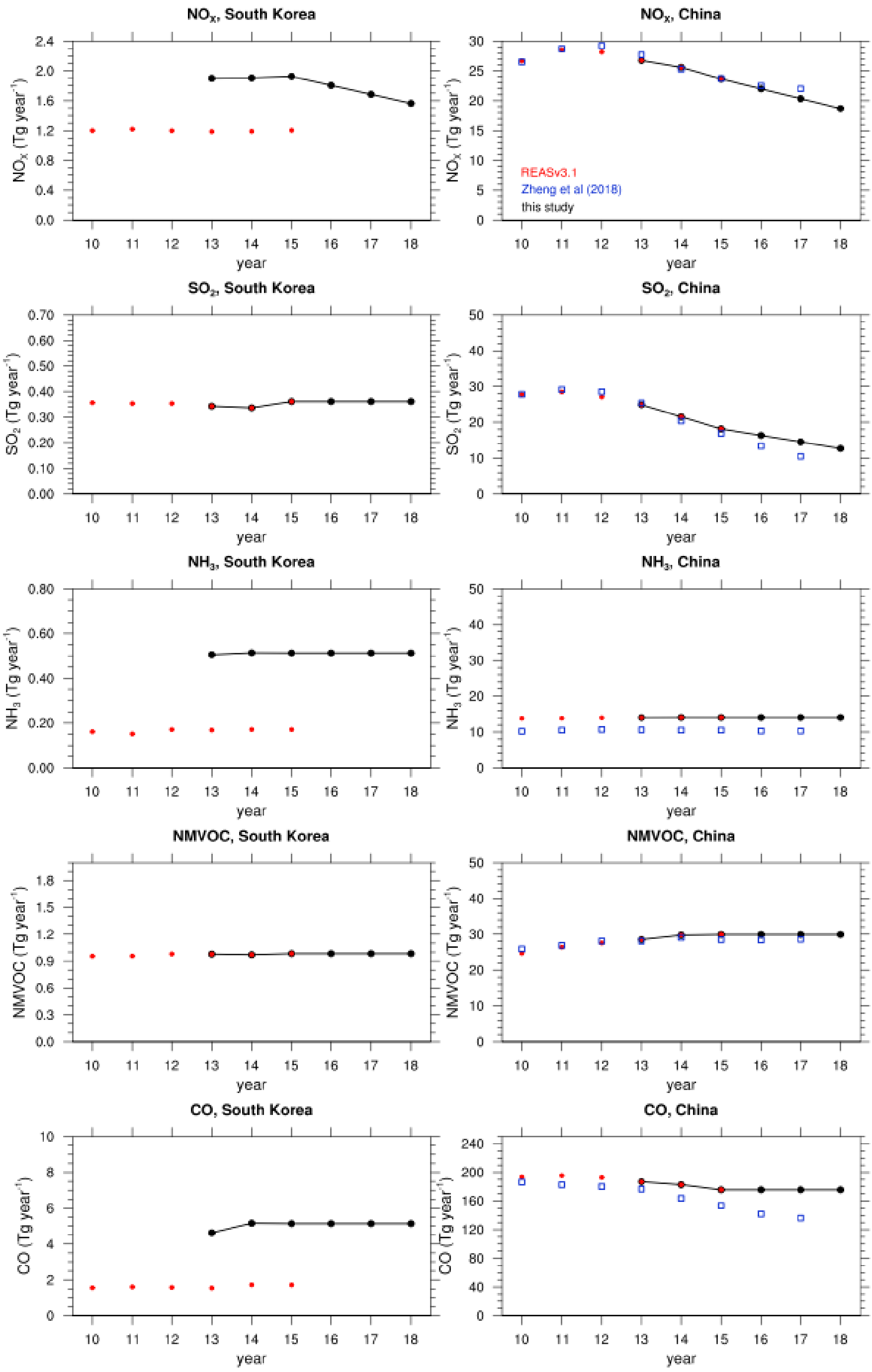
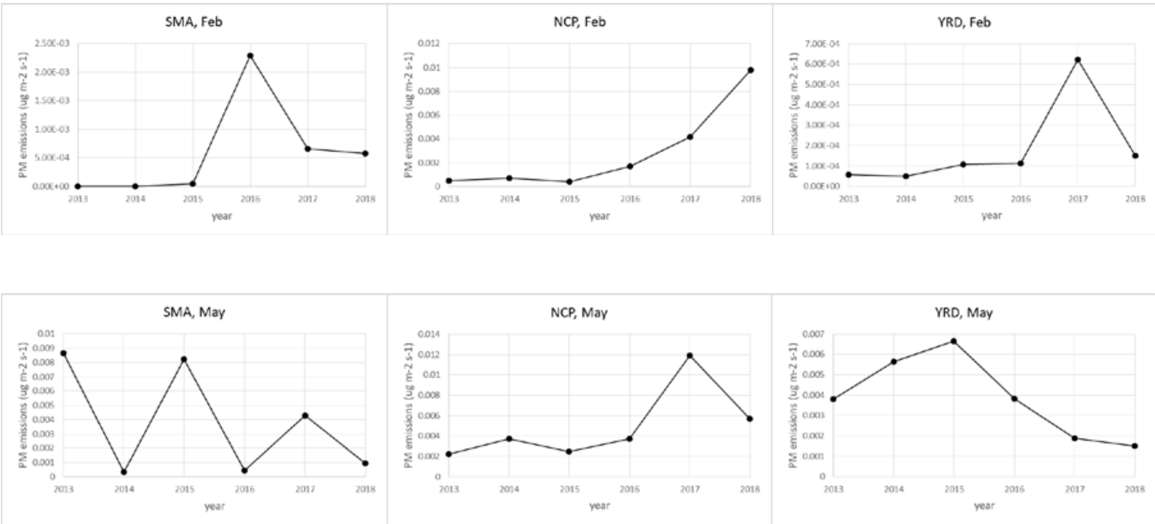


Figure S2. Trends of year total gaseous emissions over South Korea (left) and over China (right) for 2000–2015 from REASv3.1 inventory. The black dots indicate the year total emissions used (and assumed for 2016–2018) in this study. The blue squares indicate the estimated emissions by Zheng et al. (2018).



Figures S3. Timeseries of monthly mean biomass burning PM emissions averaged over (top, left) the SMA, (top, middle) the NCP, and (top, right) the YRD in February. The bottom subfigures are the same as in top subfigures, but for May.

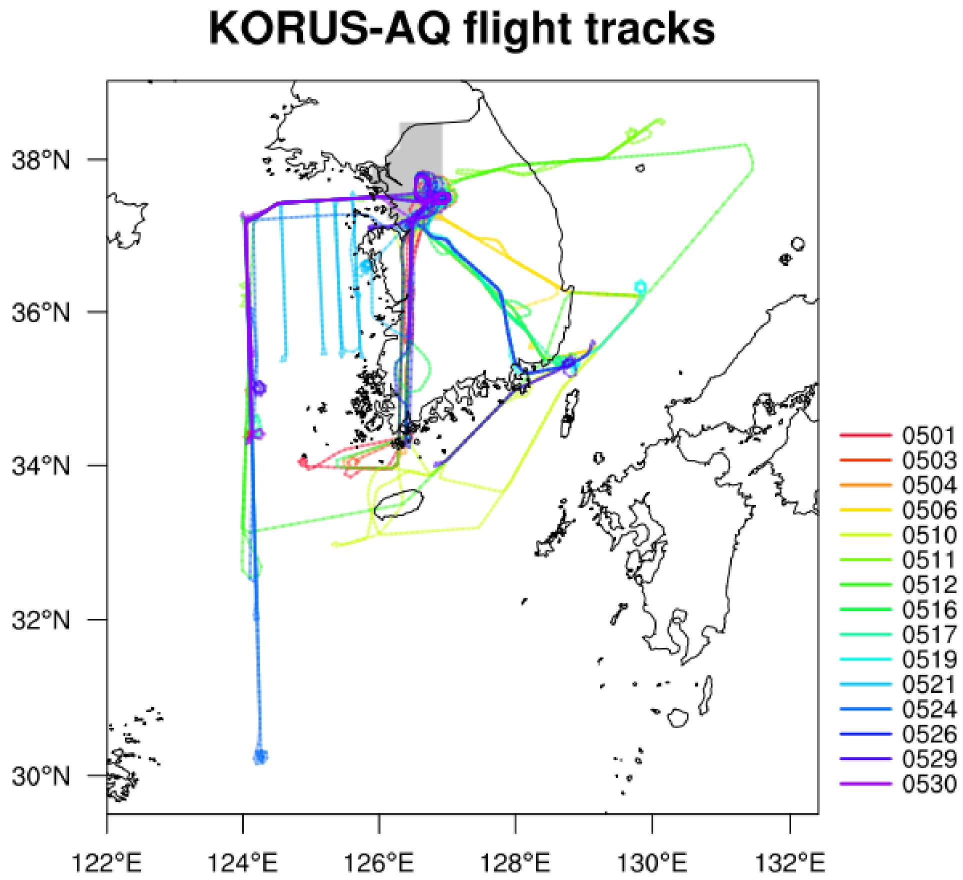


Figure S4. Flight tracks during the KORUS-AQ campaign in May 2016. The Seoul Metropolitan Area (SMA) is indicated by the gray shaded area.

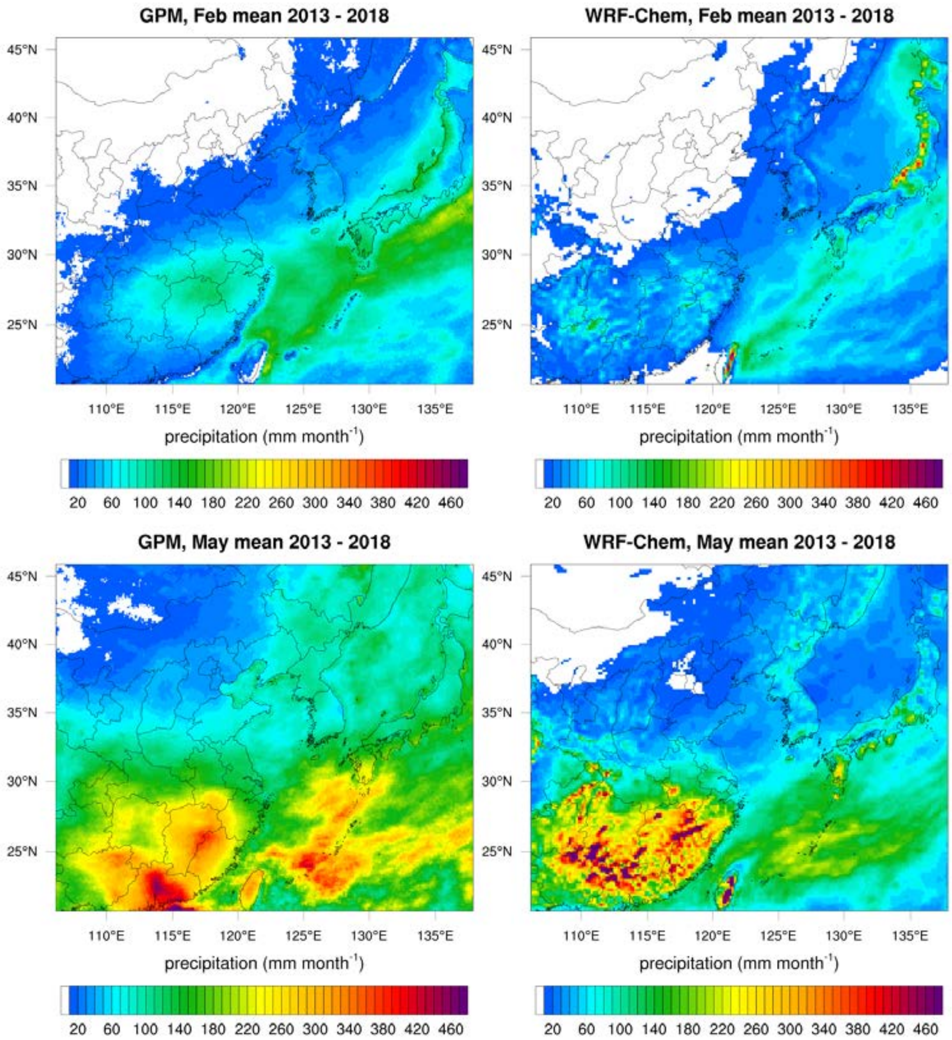


Figure S5. 6-year (2013–2018) averaged monthly precipitation from the (top left/bottom left) Global Precipitation Measurement (GPM) by NASA and (top right/bottom right) WRF-Chem simulations in February/May.

Table S1. Mapping CAMS species to WRF-Chem species. The size ranges in diameter for the MOSAIC 4 bins are 0.039–0.156 μm for a01, 0.156–0.625 μm for a02, 0.625–2.5 μm for a03, 2.5–10 μm for a04.

WRFChem	CAMS	WRFChem	CAMS	WRFChem	CAMS	WRFChem	CAMS	WRFChem	CAMS
CO	1.3CO	C_2H_4	C_2H_4	CH_3OOH	CH_3OOH	DMS	DMS	so4_a03	0.3SU
NO	NO	C_2H_6	C_2H_6	HCHO	CH_2O	O_3	O_3	so4_a04	0.1SU
NO_2	NO_2	C_2H_8	C_2H_8	ALD	ALD2	bc_a02	0.2BC[a]	oin_a02	0.6DUST1[c]
HNO_3	HNO_3	BIGALK	PAR	ISOPR	C_5H_8	bc_a03	0.8BC	oin_a03	0.6DUST2
N_2O_5	N_2O_5	BIGENE	OLE	MACR	0.5ISPD	oc_a02	0.05OM[b]	oin_a04	0.6DUST3
PAN	PAN	CH_3OH	CH_3OH	MVK	0.5ISPD	oc_a03	0.1OM		
SO_2	SO_2	C_2H_5OH	C_2H_5OH	MGLY	MGLY	oc_a04	0.6OM		
NH_3	NH_3	ACET	CH_3COCH_3	ONIT	ONIT	so4_a02	0.2SU		

[a] BC is the sum of hydrophobic and hydrophilic black carbon

[b] OM is the sum of hydrophobic and hydrophilic organic matter

[c] DUST1, DUST2, and DUST3 are dust aerosols with radius of 0.03–0.55, 0.55–0.9, and 0.9–20 μm, respectively.

Table S2. Mapping of REASv3.1 VOC to emitted VOC species of MOZART-4

MOZART-4	REAS	MOZART-4	REAS	MOZART-4	REAS
e_c2h2	Acetylene	e_benzene	Benzene+ 0.05*Other_Aromatics	e_macr	0.08*Other_Aldehyde
e_c2h4	Ethylene	e_toluene	Toluene+ 0.8*Other_Aromatics	e_ch3coch3	0.63*Ketones
e_c2h6	Ethane	e_xylene	Xylenes+ 0.15*Other_Aromatics	e_mek	0.36*Ketones
e_c3h6	Propene	e_ch2o	Formaldehyde	e_mvk	0.01*Ketones
e_c3h8	Propane	e_ch3cho	0.38*Other_Aldehyde	e_ch3oh	0.03*Others
e_bigene	Terminal_Alkenes +Internal_Alkenes	e_gly	0.35*Other_Aldehyde	e_isop	0.03*Others
e_bigalk	Butanes+Pentanes +Other_Alkanes	e_mgly	0.18*Other_Aldehyde	e_c10h16	0.05*Others

Table S3. Allocation of $PM_{2.5}$ emissions to sulfate (SO_4), nitrate (NO_3), ammonium (NH_4), and inorganic aerosols based on emission sectors

	SO_4	NO_3	NH_4	Other Inorganic
Residential	0.230	0.0142	0.00177	0.754
Industry	0.264	0.0218	0.00136	0.713
Energy	0.188	0.00269	0.00117	0.808
Transport	0.276	0.0111	0.107	0.607

Table S4. Performance statistics in the sensitivity simulations using the fixed emissions with base year of 2014 for 2013–2018. The unit of normalized mean bias (NMB) is %, and the unit of systematic root-mean-square-error (RMSEs) and unsystematic root-mean-square-error (RMSEu) is μg m^{-3}.

	SMA February			NCP February			YRD February			SMA May			NCP May			YRD May		
year	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu	NMB	RMSEs	RMSEu
2013	–7.4	2.2	6.7	–11.0	19.9	24.0	–6.3	8.1	16.0	1.7	2.1	10.1	–20.5	15.6	9.0	–18.4	9.9	12.3
2014	5.4	10.4	17.8	–3.4	11.5	16.8	–4.5	2.8	14.3	2.4	2.8	6.6	–1.4	0.9	13.2	–11.1	6.6	14.2
2015	4.7	3.3	10.6	12.0	10.8	18.2	23.6	16.5	16.4	21.8	6.0	10.1	0.3	2.9	9.9	–12.7	8.6	12.0
2016	4.3	1.2	6.6	39.6	26.5	19.1	33.9	19.0	19.9	–0.4	5.0	7.5	17.6	8.0	8.5	13.0	5.4	12.9
2017	–13.5	5.7	6.0	18.7	15.5	17.5	23.8	14.2	20.4	20.3	5.8	12.5	28.3	11.6	9.8	27.0	9.5	11.7
2018	–15.0	5.5	7.0	42.8	28.8	20.8	37.3	21.6	17.9	7.2	1.7	6.4	44.4	17.8	7.4	15.7	6.7	10.5