Reply to the review of "Changes in surface ozone in South Korea on diurnal to decadal time scale for the period of 2001-2021"

We provide our replies below. The review is written in blue and our replies in black.

This manuscript addressed an issue of observed surface ozone increases in South Korea by analyzing a long-term dataset and 3-d air quality model simulations for divulging its attribution. The surface ozone increase in South Korea and China is a compelling issue for which previous literature extensively attempted to investigate its causes. Compared to them, I find it quite challenging that this work shows a new contribution to the scientific understanding of the issue or a new idea that needs to be investigated in the future. In addition, the manuscript should be reshaped to highlight its main findings by adding descriptions of how the authors reached conclusions, which were mostly based on immature analyses. I will elaborate on them below.

Thank you for constructive criticism and introducing recent publications about surface ozone over South Korea that probed the sources of its abundance. We appreciate these studies and will include them for discussions in the revised manuscript as elaborated in the responses below.

Your comments are greatly appreciated. But we respectively disagree with the reviewer to the point that previous literature extensively attempted to investigate its causes and there are hardly any new contributions and ideas in our study. We hope that our responses below help better identify the values of this study and bring up many ideas to be studied/tested in the future. Past and recent publications (several publications) pointed out the possibility of long-range transport of ozone from China to South Korea and high background ozone value external to East Asia or South Korea for a certain period. However, the atmospheric/environmental science community is far from understanding the causes for the long-term trends of surface ozone over South Korea that were summarized in our study. Colombi et al. (2022) nicely demonstrated one possible cause for ozone increase over South Korea from 2015 to 2019. There are good agreements between our results and Colombi et al. (2022). And there are differences too. It is good that the two different approaches reach the similar conclusions, an importance of large background ozone in spring and existence of long-range transport from China to South Korea. Our study is different from Colombi et al. (2022) in terms of investigation of vertical sensitivity of ozone to surface emission changes and the period of the data including the COVID-19 pandemic. We found a large reduction of ozone exceedances over most of the sites over South Korea in spring during the COVID-19 pandemic, which were not reported and were not extensively studied. We believe our study motivates more detailed modeling research encompassing the long-term period or the period including the COVID-19 pandemic for better understanding of ozone over South Korea and China.

In the responses below, we explain how we reached the conclusions and will include the discussed contents to the revised manuscript. We were preparing several manuscripts regarding the WRF-Chem and CAM-Chem performances and did not include details and evaluation results to the current manuscript. This is the reason why we omitted the model evaluations. The authors have full pictures, but the reviewer and reader would not have them. Therefore, it is helpful to provide more information about model performances as the reviewer asked. In the revised manuscript, we will include evaluations of the model ozone simulations to Supporting Information and refer to the manuscripts submitted or to be submitted.

- Papers submitted and in preparation

Jeong, YuJoo, et al., 2023, Influence of ENSO on tropospheric ozone variability in Asia, submitted. (evaluations of CAM-Chem ozone simulations)

Kim, Kyoung-Min, et al., 2023, Sensitivity of the WRF-Chem v4.4 ozone, formaldehyde, and their precursor simulations to multiple bottom-up emission inventories over East Asia during the KORUS-AQ 2016 field campaign, *in preparation*.

P2,L2 - “Increasing trends of tropospheric ozone in South Korea” is a bit misleading because ozone in surface air does not always reflect tropospheric ozone. Needs to be revised to surface ozone.

→ Gaudel et al. (2020) found that tropospheric ozone in China and South Korea increased from 1996 to 2016. Both surface and tropospheric ozone in South Korea increased during the last decades. However, for the abstract of this manuscript, we changed “Increasing trends of tropospheric ozone” to “Increasing trends of surface ozone” as the reviewer suggested.

P4,L11 - Here and elsewhere, references at not in the reference section. Please check all the citations and include other previous studies on the same issue (e.g., Colombi et al., ACPD, 2022, and the references are therein).

→ Thank you for introducing Colombi et al and references therein. We originally included the references that focused on the analysis of surface ozone measurements in South Korea. Now in the revised manuscript, we include more references including modeling or analysis studies (see the reference section in this reply).

P4,L11 - "Ozone in South Korea …" this sentence requires a citation.

→ We will cite the papers, Oh et al. (2010) and Lee and Park (2022) (see the reference section in this reply).

P8,L11 - Stratospheric ozone appears to have a significant effect on ozone in the troposphere and even in surface air in this study. However, I cannot find out how the effect of stratospheric ozone on tropospheric and surface ozone was quantified in the manuscript. I think that it should be elaborated on here.

→ 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 of chemical reactions and variables, please refer to Emmons et al. (2020). We will include explanations about O_{3S} in the revised manuscript. The representation of O_{3S} has uncertainties, but it can be used as a parameter that indicates the contribution of stratospheric ozone to tropospheric ozone at each altitude at least qualitatively. We will explain how O_{3S} is calculated and mention uncertainty of using O_{3S} in the revised manuscript.

Sections 2.4, 2.5. – This study used model simulations to understand the observed characteristics of surface ozone in South Korea. Therefore, an extensive model evaluation should be conducted and discussed somewhere in the manuscript by focusing on how good the model is to reproduce the observations and their variability.

→ We have extensively evaluated our model results with the airborne and surface observations acquired during the KORUS-AQ campaign and the routine surface monitors in China and South Korea. The results are summarized and will be submitted as a separate manuscript to a relevant journal:

Kim, Kyoung-Min, et al., 2023, Sensitivity of the WRF-Chem v4.4 ozone, formaldehyde, and their precursor simulations to multiple bottom-up emission inventories over East Asia during the KORUS-AQ 2016 field campaign, *in preparation*.

For example, the diurnal variations of the model and observed surface ozone concentrations in China and South Korea are compared below (Figure R1 and Table R1). We found decent model performances in the surface ozone concentrations with the bottom-up emission inventories EDGAR-HTAPv2(EDV2), EDGAR-HTAPv3(EDV3), and KORUS-AQv5(KOV5). EDV3 and KOV5 performed a little better.

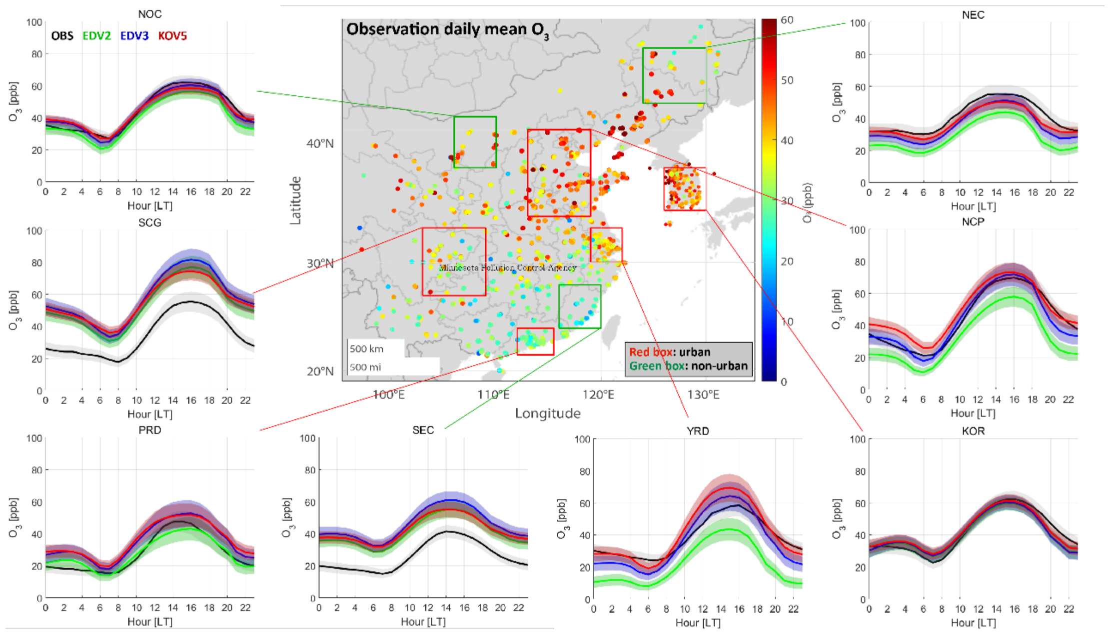

Figure R1. Averaged O_3 from the ground-based observations and model results for regional boxes that distinguish urban (red box) and non-urban (green box) region (central plot). Box averaged diurnal cycle (solid lines) of O_3 and 1/4 of standard deviations (filled area) from observations (black), the 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).

Table R1. 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) in each regional box (unit = ppb).

	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

Evaluation of the model results with the aircraft data acquired during the KORUS-AQ campaign are shown below (Figure R2 and Table R2).

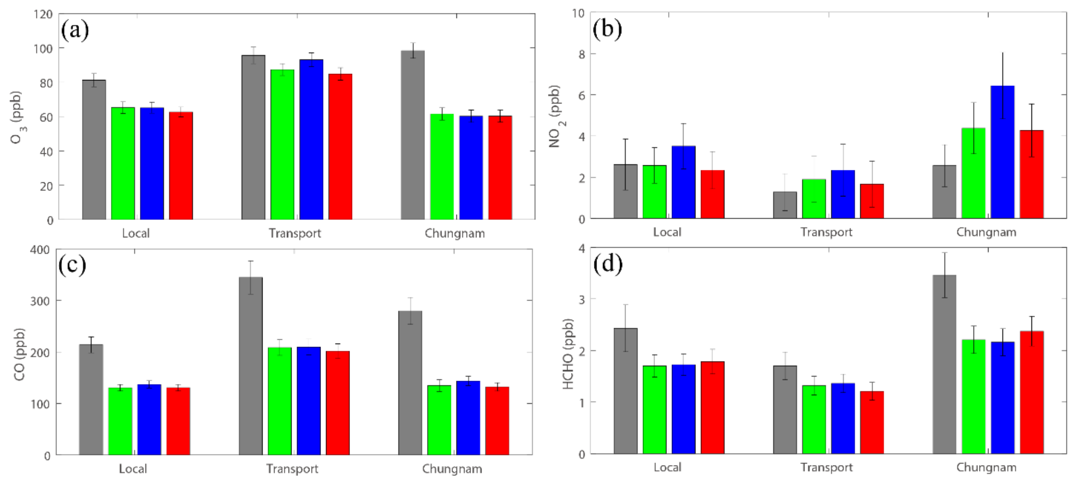

Figure R2. Averaged model and airborne observations of (a) O_3, (b) NO_2, (c) CO, and (d) HCHO (bars) and 1/4 of standard deviations (whiskers) (unit: ppbv) under 2 km height for the Local, Transport, and Chungnam cases from DC-8 (grey), EDV2 (green), EDV3 (blue), and KOV5 (red). The Chungnam (Chungcheongnam-do) 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, June/1) 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.

Table R2. 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).

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

In summary, the model reasonably simulated ozone concentrations (particularly for the Transport Case), but they are overall underestimated compared to the observations. Potential causes for the discrepancy are underestimated CO and volatile organic compound emissions/concentrations in China and South Korea and/or uncertainties in the background ozone external to East Asia. Details about the model performances of precursor emissions are discussed in the manuscript by Kim, Kyoung-Min et al. (2023) and are beyond the scope of this study. We included some of the model results for discussions for our manuscript and will add some evaluation results to Supporting Information.

P9,L4 – Years for the WRF-Chem simulations were missing. Did you conduct simulations for all years or for a particular year?

→ The WRF-Chem model was conducted for 2016. We will specify the model year in the revised manuscript.

P9,L7 – It appears that the authors used different meteorology to drive CAM-Chem simulations and WRF-Chem simulations. Have you ever thought about using identical meteorology for both models?

→ The WRF-Chem and CAM-Chem model results were shown for different purposes. The WRF-Chem runs were used to analyze the sensitivity of ozone over South Korea to the emissions over China and South Korea for a limited time window (May-June 2016). The CAM-Chem runs inform the seasonal changes in the background ozone including the contribution of stratospheric ozone to the troposphere for the long-term period. Thorough comparisons of the two model results are beyond the scope of this study. Meanwhile, both WRF-Chem and CAM-Chem accurately simulated meteorology (Table R3 and Figure R3).

Table R3. Comparison of surface meteorological observations and WRF-Chem for the KORUS-AQ campaign period. 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

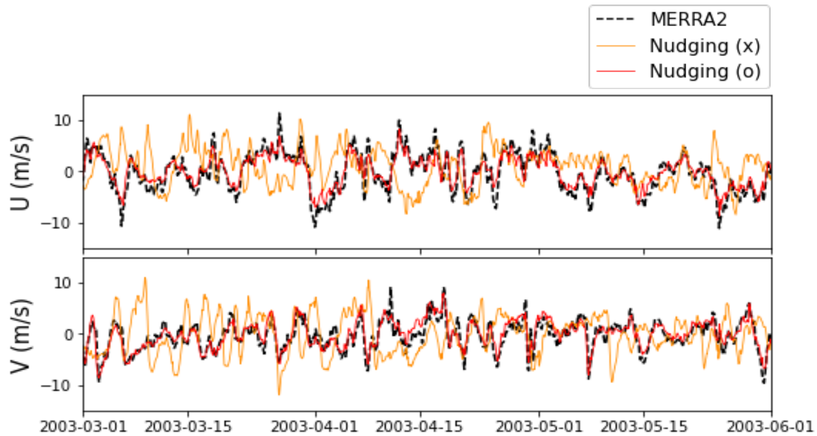

Figure R3. The examples of CAM-Chem U, V wind components for spring, 2003. Without nudging, the model simulated U, V do not closely agree with the MERRA2 data.

P9,L14 – The time information of emissions inventory used in the model is missing. Did you also consider biomass burning emissions in the model?

→ EDGAR-HTAPv2 represents the year 2010. Since there are 6 years difference in EDGAR-HTAPv2 from 2016, we also utilized EDGAR-HTAPv3 representing the year 2016 in the revised manuscript. Park et al (2021) informed that biomass burning was not an important factor affecting air quality in South Korea during KORUS-AQ. Therefore, we did not include the biomass burning.

P10,L9, - You analyzed the 4th highest MDA8 O3. I wonder how this metric well represents ozone air quality because these could be rather extreme events, which rarely happen. In other words, how frequently people in South Korea were exposed to this metric?

→ The published works on the trend of surface ozone in South Korea presented the ozone metrics such as annual mean of hourly ozone, annual mean of MDA8 ozone, annual mean of daily maximum hourly ozone, and frequency of hourly concentrations greater than 120 ppb. The trends based on those metrics have already been published (e.g., Yeo and Kim, 2021). Since the US EPA National Ambient Air Quality Standard (NAAQS) for ozone is 70 ppb, as the fourth-highest MDA8 ozone concentration, averaged across three consecutive years, and the recent study by Wang et al. (2022) adopted the 4th highest MDA8 ozone concentrations as one of the metrics for study of Chinese ozone pollution, it would be nice to have analyses adopting the 4th highest MDA8 ozone for a global comparison. The EPA standard is also designed for public health protection. Exceedances presented in our study are similar to the frequency exposed to MDA8 ozone > 70 ppb (relevant to EPA standard).

P10,L14 – The trend in Jeollanam-do differs from other provinces. This is explained by "MDA8O3 in Jeollanam-do is high before 2010". I do not understand why this is the case. Here and elsewhere, please check out the proper usage of provinces and city names.

→ The monitoring sites in Jeollanam-do include the Yeosu-Kwangyang region in which many petrochemical industry (e.g., GS Caltex, LG Chem), and iron steel complexes (e.g., POSCO) are located, similar to Houston, Texas. This region experienced severe ozone problems in the 1990's to early 2000's (Ghim, Y. S. 2000). We will mention large unique sources in this area in the revised manuscript. And we will double-check the consistency of names for provinces and cities.

P10,L15-17 – This sentence includes several factors, contributing to ozone increases in South Korea. Proper citations are required.

→ "Widely increasing long-term ozone trends in South Korea indicate a regional nature of this pollutant" is the statement we made from our analysis. However, we will include some references that support this statement with modeling (e.g., Lee and Park, 2022, Colombi et al., 2022).

P11,L2 – "Investigating seasonal differences in ozone in South Korea" has been examined by Lee and Park (2022). Any consistency or dissimilarity from the previous study is worth being mentioned.

→ Lee and Park (2022) reported the April mean ozone concentration of 39.3 ppb, which is slightly higher than the July counterpart (38.3 ppb) from their model simulations for the year 2016 and the selected surface monitor sites for 4 main regions (Seoul, Chungbuk, Gwangju, and Pusan). Our study summarizes the differences between spring (March, April, May) and summer (June, July, August) for 21 years including 192 monitoring sites covering the whole of South Korea focusing on the analysis of long-term surface ozone observations. On overage, the observed spring mean ozone is 34.3 ppb and the summer mean ozone is 29.0 ppb over South Korea in our study. Lee and Park (2022) indicated that ozone air quality in South Korea is determined mainly by year-round regional background contributions (peak in spring). With some differences in details, the results from the two studies are qualitatively similar arguing high springtime background ozone value. In the revised manuscript, we will add discussions above. One unique aspect of our modeling study is demonstrations of the impact of emission in Seoul on Gangwon-do, causing slight ozone decrease in Gangwon-do with zero-Seoul emissions from surface to 2 km in May

2016. Our study highlights the diverse impacts of surface emission changes (over China or Seoul) on downwind ozone at different altitudes (Figure 11 in our original manuscript). In the future, more detailed analysis of ozone in Gangwon-do will be helpful since the Gangwon-do region is highly elevated (Figure R4), potentially receiving upwind ozone at high altitude. In the original manuscript, "Gangwon-do" meant "Gosung, Gangwon-do" in Figure 11. In the revised manuscript, we will correct this title and typing error in the x-axis (Ozobe → Ozone). We will also include the map of South Korea to Supporting Information and explain potential paths of ozone transport from China to Seoul to Gosung with a simplified diagram.

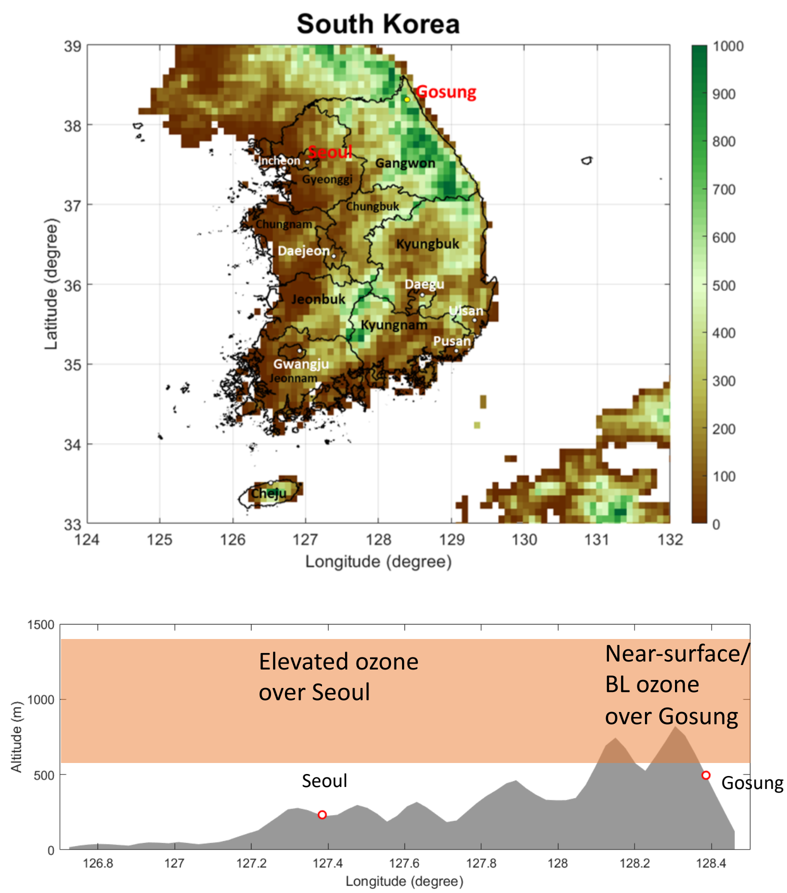

Figure R4. (Top) topography map of South Korea and (bottom) West-East vertical cross-section of terrain connecting Seoul and Gosung. Seoul and Gosung in Gangwon-do are highlighted with color circles. Color bar denotes elevations above sea level (m). A simplified potential ozone transport path is depicted in the bottom plot. Here the ozone layer is colored orange and the terrain is colored gray.

P13,L1-13 – Stratospheric influences are quite large, which are still debatable. As I mentioned above, how did you obtain the stratospheric ozone influences on low tropospheric and surface ozone concentrations in South Korea? Does the model reproduce observations well? You have to elaborate a lot on this part.

→ O_{3s} was explained in the response above. In this reply, we show how the CAM-Chem simulations are compared with the ozonesonde data acquired over Pohang, South Korea from 1996 to 2020 (Figure R5). The model results and observations reasonably agree in terms of seasonal variability and absolute values. The model run with ~1 degree horizontal resolution reduces positive model surface ozone biases in summer compared to ~2 degree horizontal resolution run, but increases biases in late autumn to early spring at 500-850 hPa. At the 200 hPa level (close to tropopause), the CAM-Chem with both resolutions agree well with the observations. We presented the results with ~1 degree resolution in the original manuscript. We will explain the performance of CAM-Chem against the ozonesonde data in the revised manuscript.

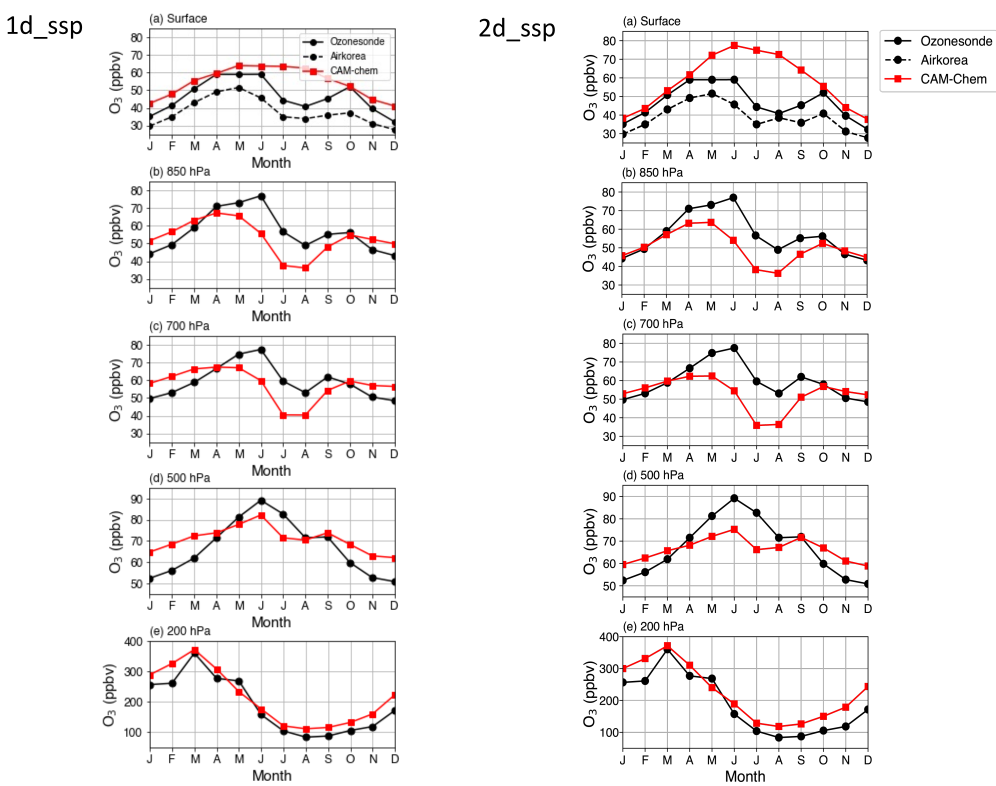

Figure R5. Monthly variations of CAM-Chem simulated ozone concentration (red line) and observed ozone concentration (black solid line) at each pressure level near Pohang, between 14-17 KST. (Left panel) ~1 degree horizontal resolution simulation, and (right panel) ~2 degree horizontal resolution simulation.

P13,L14-20 – Colombi et al. (2022) already performed a nice analysis on the effect of precursor changes on observed surface ozone increases in South Korea. You have to compare your work with theirs.

→ We think it is very nice to have several publications about the topic of ozone in South Korea, which agree in general, but have a different emphasis and details. A main difference between Colombi et al. (2022) and our study is the period of the study and whether it focuses on the surface ozone or vertical sensitivity explaining ozone variability at different locations in South Korea. Our study investigated surface ozone and ozone at various altitudes to consider the transport within and above the boundary layer between China and South Korea. Colombi et al. (2022) analyzed the surface ozone and NO_2 concentrations mainly over the Seoul Metropolitan Area from 2015 to 2019. The increase of ozone was mostly attributed to decrease in NO_2 for the studied period in their study. It is nice to identify one possible cause for the increase of surface ozone in the SMA from 2015 to 2019 as in Colombi et al. (2022). We will mention this study in the revised manuscript. One question that remains is the existence of long-term increasing trends of surface ozone over South Korea (SMA and other regions) when NO_x concentrations were steady before 2015-2019. What is the cause for this increase? Further research would be necessary to understand the long-term trends of ozone over South Korea.

Both Colombi et al. (2022) and Lee and Park (2022) indicated high background ozone concentration external to East Asia (or South Korea), suggesting difficulty of achieving ozone standards. Our study agrees to this point. Probably one different message is that reducing emissions of NOx and VOC here and there all together have positive impacts on reducing ozone downwind. For example, emission reductions associated with the COVID-19 would lead to decrease of ozone at most sites over South Korea in spring. Global efforts associated with greenhouse mitigation (use of cleaner fuel) eventually help to alleviate ozone pollution.

P14,L6-20 – Previous studies published the observed increase in ozone in China and South Korea during the pandemic due to less titration of NOx. This result is contrary to previous studies and please compare the differences between this and previous work.

→ This is the novel aspect of our manuscript. There are several studies reporting the increase of near-surface ozone after COVID lockdowns in the urban areas (e.g., Shi & Brasseur, 2020) because of expected non-linear relationship between ozone and NO_x in the highly polluted regions. However, there are also studies reporting reductions of ozone concentrations from 1 to 8 km altitude in the northern extratropics during COVID

(Steinbrecht et al., 2021). Our study shows both increases and decreases of ozone with COVID-like NO_x emission changes: near-surface ozone concentrations over the polluted regions increase, but there are reductions of ozone concentrations in the elevated layer (Figure R6 and R7). Novel findings in our study are **the decrease of downwind ozone near surface to upper layer with reductions of NO_x/VOC emission in upwind pollution hot spots** (see Figure R6 and R7 for several sensitivity runs). For example, 50%-75% of Chinese NO_x emission reductions decrease ozone concentrations in Korea and surrounding seas and the Pacific Ocean from the surface to upper layers although near-surface ozone in Northeast China increases due to these emission changes. Therefore, our study does not fully support the findings in Lee et al. (2021) that stated "These NO_x-saturated conditions in megacities contribute to the increased O_3 due to NO_x reduction, which could also affect the enhanced O_3 concentrations throughout the Asia–Pacific region via long-range transport". Chinese VOC reductions cause reduced ozone concentrations from surface to upper layer and from hot spots to downwind areas. Our study suggests potential changes in photochemical regimes with altitudes over the pollution hot spots (NO_x-saturated near surface versus NOx-limited in the elevated layer). Thus, combined effects of vertical and horizontal ozone transport and local production dependent on altitude would determine the ultimate changes in ozone concentrations at certain locations and altitudes. We will add the discussions in the revised manuscript with Figure R6. One thing to note is that the assessment also depends on the accuracy of VOC emissions estimations. This part is vastly uncertain and is the matter of further study.

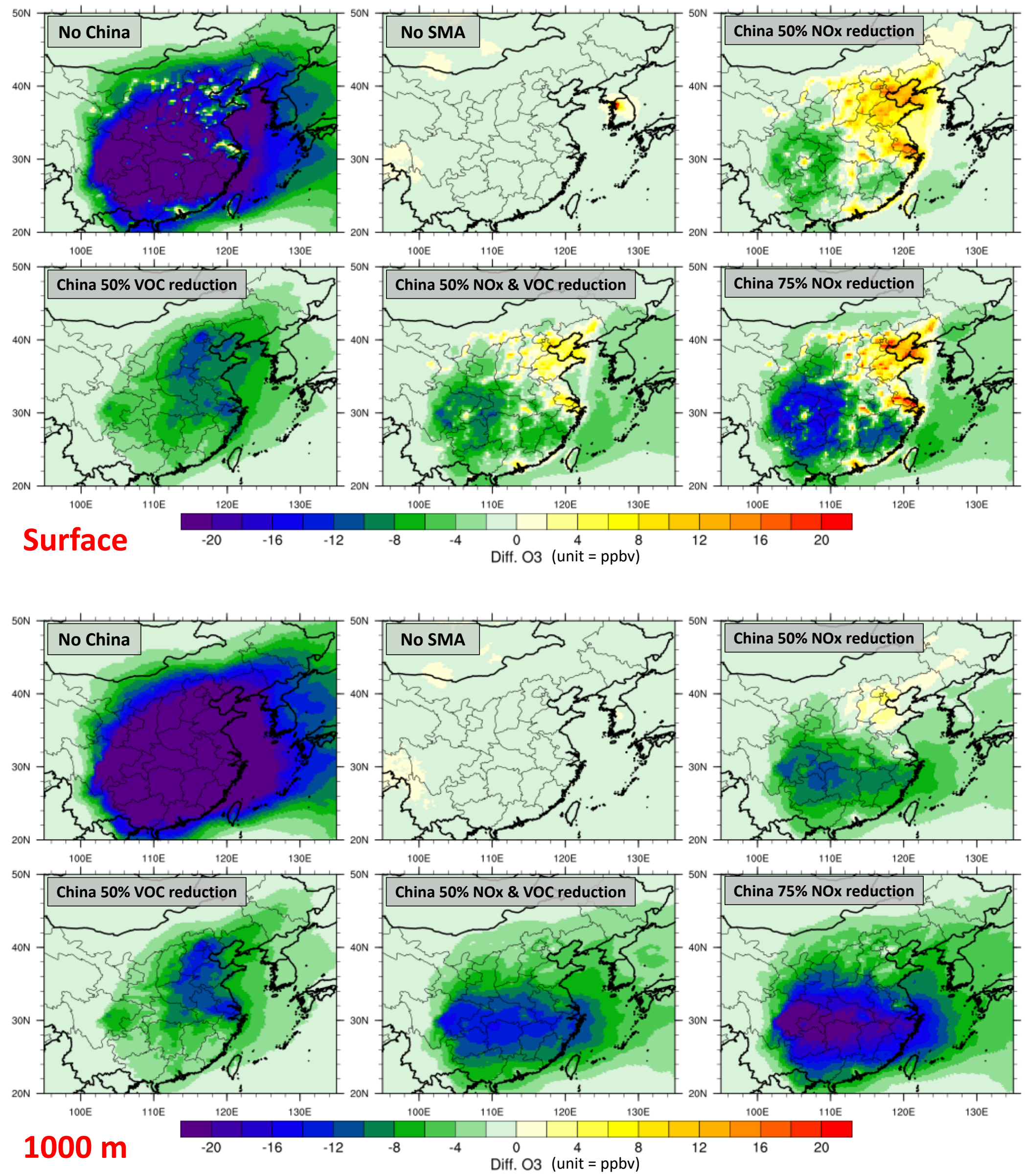

Figure R6. Differences in the WRF-Chem simulated ozone concentrations (ΔO_3 = O_3_emission reduction case-O_3_control case) at (top) surface and (bottom) 1000 m above ground level. Green to blue colors (yellow to red colors) denotes reduced (increased) ozone concentration due to the emission changes.

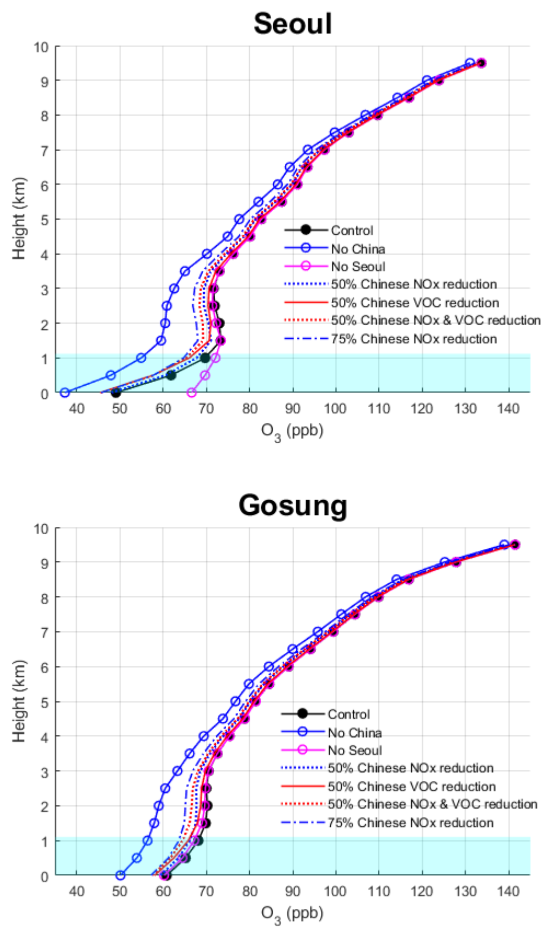

Figure R7. Vertical profiles of ozone from the WRF-Chem model simulations based on various emission scenarios: (top) Seoul, and (bottom) Gosung, Gangwon-do. The model ozone is averaged from 10 LT to 20 LT. Averaged boundary layer height is shaded as cyan color.

Section 4. You presented simulated vertical profiles in Seoul and Gangwondo during the KORUS-AQ. Could you include aircraft observations in Figure 11? I also wonder how the model simulates surface ozone concentrations.

→ The vertical profiles of ozone from the DC-8 observations and co-located the WRF-Chem results in our study are shown below (Figure R8). The model generally follows the vertical distributions measured by the DC-8 aircraft. The model ozone has a low bias of 16-19 ppb for the cases influenced by the local emissions (Local case: May/4, May/20, June/2, June/3). The model performed better for the cases strongly influenced by the Chinese emissions (Transport case: May/25, May/26, June/1) with a low bias of 3-11 ppb. The EDGAR-HTAP v3 emissions led to the smallest bias for the Transport case. The emission sensitivity runs with doubling Chinese CO and VOC emissions and with doubling both

Chinese and South Korean CO and VOC emissions improve ozone simulations for the Local case, but overestimate ozone concentration for the Transport case. This indicates that more efforts need to be put into the evaluation and improvement of the local CO and VOC emissions estimations. It is still important to improve the emission estimations for China for better ozone simulations of South Korea and beyond. Both surface and boundary layer ozone in the model runs were evaluated and discussed in the responses above. We include this discussion in the Supporting Information. In the revised manuscript, we replace the WRF-Chem model results using EDGAR-HTAPv2 by those using EDGAR-HTAPv3.

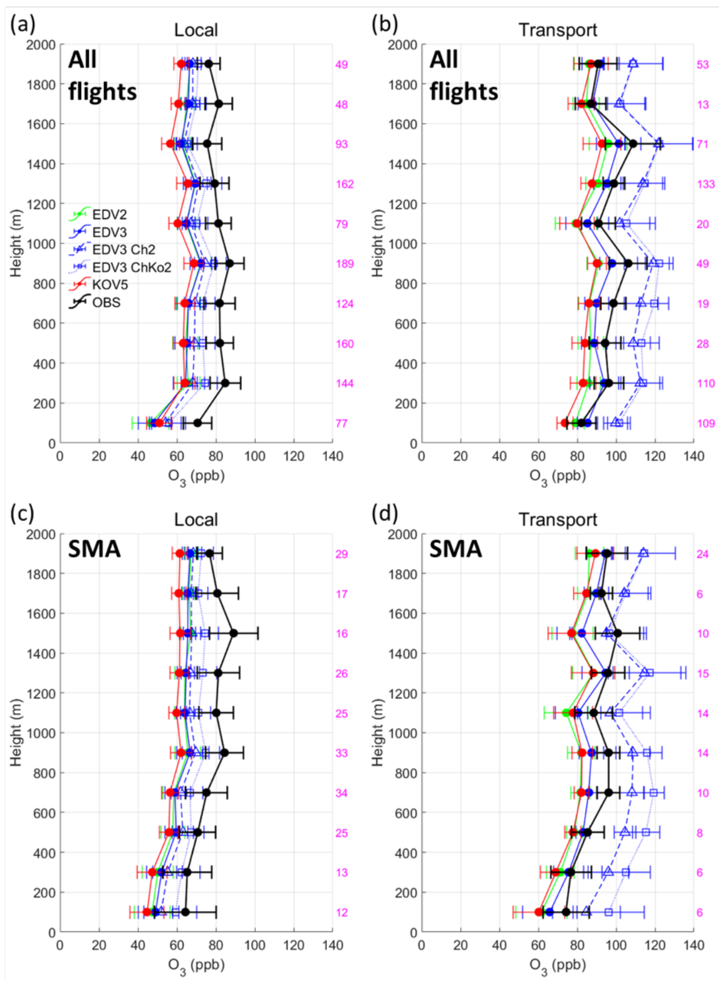

Figure R8. Vertically averaged O_3 from DC-8 (black), EDV2 (green), EDV3 (blue), and KOV5 (red) for the Local and Transport cases under 2 km height above ground level. The 1/2 of standard deviations are represented with black whiskers in each 200m layer. Sensitivity tests are conducted with doubled anthropogenic CO and VOC emissions in China (EDV3_Ch2, blue triangle dots and dashed lines) and 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).

References:

Colombi, N. K., et al., 2022, Why is ozone in South Korea and the Seoul Metropolitan Area so high and increasing? ACP Discuss., https://doi.org/10.5194/egusphere-2022-1366.

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.

Gaudel, A., et al., 2020, Aircraft observations since the 1990s reveal increases of tropospheric ozone at multiple locations across the Northern Hemisphere, *Sci. Adv.*, 6, eaba8272.

Ghim, Y. S., 2000, Trends and factors of ozone concentration variations in Korea, Journal of Korean Society for Atmospheric Environment, Vol. 16, No. 6, pp 607-623.

Hwang, S.-H., et al., 2007, Observation of secondary ozone peaks near the tropopause over the Korean Peninsula associated with stratosphere-troposphere exchange, *Journal of Geophysical Research: Atmosphere* 112(D16). https://doi.org/10.1029/2006JD007978.

Jeong, YuJoo, et al., 2023, Influence of ENSO on tropospheric ozone variability in Asia, submitted.

Kim, H., et al., 2020, Factors controlling surface ozone in the Seoul Metropolitan Area during the KORUS-AQ Campaign, *Elementa* 8(46): 10.1525/elementa.444. https://doi.org/10.1525/elementa.444.

Kim, Kyoung-Min, et al., 2023, Sensitivity of the WRF-Chem v4.4 ozone, formaldehyde, and their precursor simulations to multiple bottom-up emission inventories over East Asia during the KORUS-AQ 2016 field campaign, *in preparation*.

Lee, H.-J., et al., 2021, Ozone continues to increase in East Asia despite decreasing NO_2: Causes and Abatements. *Remote Sensing* 13(11):2177. https://doi.org/10.3390/rs13112177.

Lee, H.-M. and R. J. Park, 2022, Factors determining the seasonal variation of ozone air quality in South Korea: Regional Background versus Domestic Emission Contributions, *Environmental Pollution* 308:119645, https://doi.org/10.1016/j.envpo.2022.119645.

Oh, I.-B., et al., 2010, Elevated ozone layers over the Seoul Metropolitan Region in Korea: Evidence for long-range ozone transport from Eastern China and its contribution to surface concentrations, *Journal of Applied Meteorology and Climatology* 49: 203-20. Httos://doi.org/10.1175/2009JAMC2213.1.

Park et al. (2021). Multi-model intercomparisons of air quality simulations for the KORUS-AQ campaign. ELEMENTA: Science of the Anthropocene, 9(1), 00139, doi: https://doi.org/10.1525/elementa.2021.00139

Shi, X., & Brasseur, G. P. (2020). The response in air quality to the reduction of Chinese economic activities during the COVID-19 outbreak. *Geophysical Research Letters*, 47, e2020GL088070. https://doi.org/10.1029/2020GL088070.

Steinbrecht, W., Kubistin, D., Plass-Dülmer, C., Davies, J., Tarasick, D. W., von der Gathen, P., et al. (2021). COVID-19 crisis reduces free tropospheric ozone across the Northern Hemisphere. *Geophysical Research Letters*, 48, e2020GL091987. https://doi.org/10.1029/2020GL091987.

Wang, W., Parrish, D. D., Wang, S., Bao, F., Ni, R., Li, X., Yang, S., Wang, H., Cheng, Y., and Su, H.: Long-term trend of ozone pollution in China during 2014–2020: distinct seasonal and spatial characteristics and ozone sensitivity, Atmos. Chem. Phys., 22, 8935–8949, https://doi.org/10.5194/acp-22-8935-2022, 2022.

Yeo, M. J., and Y. P. Kim (2021), Long-term trends of surface ozone in Korea, Journal of Cleaner Production, 294, https://doi.org/10.1016/j.jclepro.2020.125352.