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.

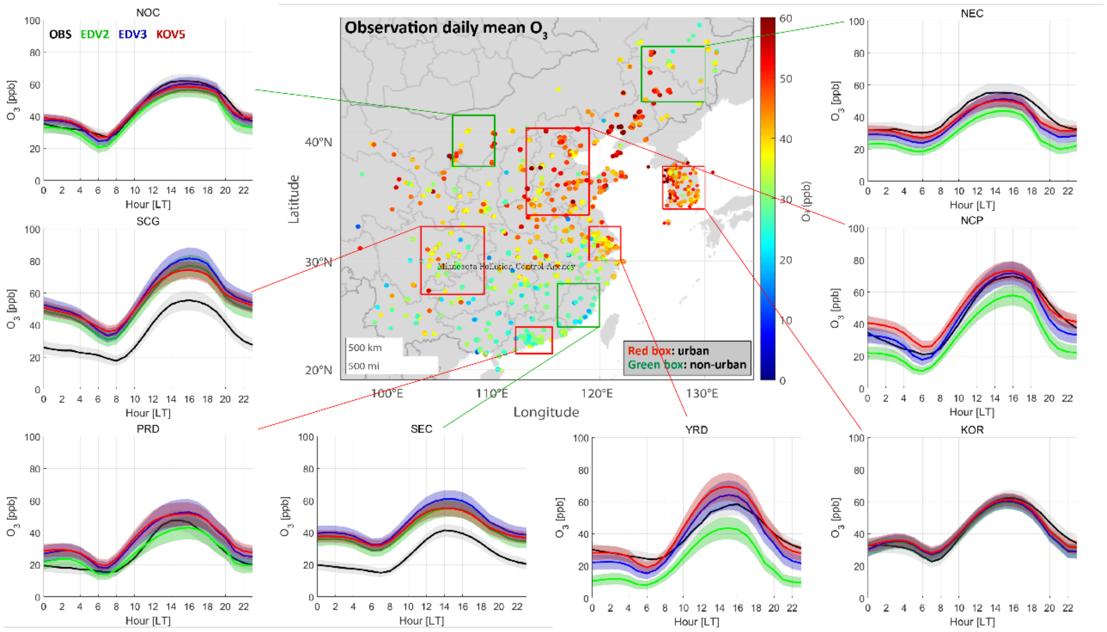

Figure R1. Averaged O_3 from the ground-based observations and model results for regional boxes that distinguish urban (red box) and non-urban (green box) region (central plot). Box averaged diurnal cycle (solid lines) of O_3 and 1/4 of standard deviations (filled area) from observations (black), the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

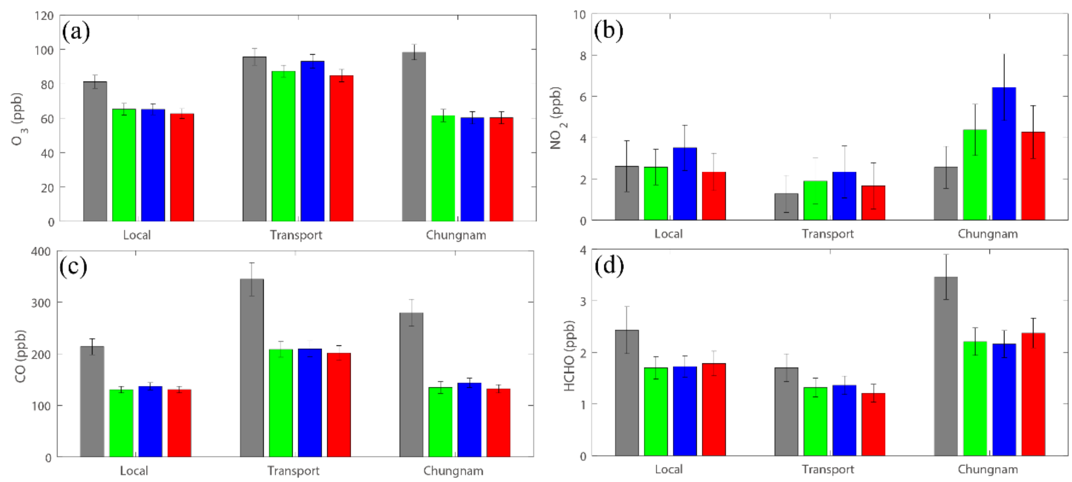

Figure R2. Averaged model and airborne observations of (a) O_3, (b) NO_2, (c) CO, and (d) HCHO (bars) and 1/4 of standard deviations (whiskers) (unit: ppbv) under 2 km height for the Local, Transport, and Chungnam cases from DC-8 (grey), EDV2 (green), EDV3 (blue), and KOV5 (red). The Chungnam (Chungcheongnam-do)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

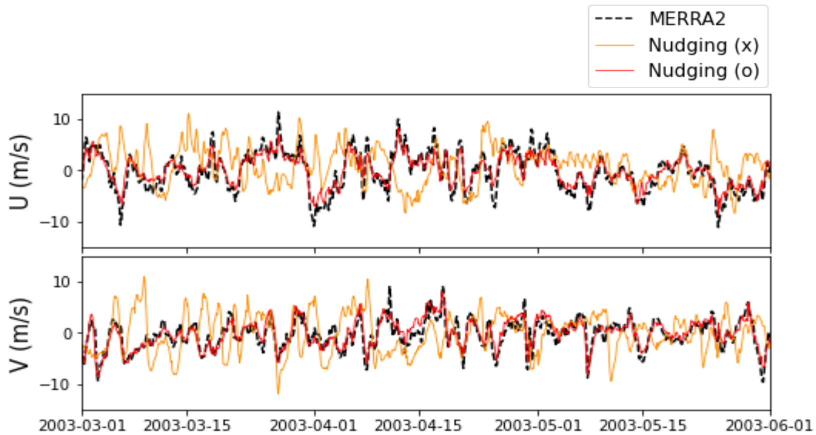

Figure R3. The examples of CAM-Chem U, V wind components for spring, 2003. Without nudging, the model simulated U, V do not closely agree with the MERRA2 data.

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

→ P12 L6-L9.

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?

→ Refer to our reply to reviewer1's comments

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.

→ P13 L16-L19.

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

→ P13 L19-P14 L1.

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.

→ P29 Section 3.4.3 Comparisons with recent modeling research

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.

→ P10 L13-P11 L4 and SI1 Figure S2.

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.

→ P29 Section 3.4.3 Comparisons with recent modeling research

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.

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.

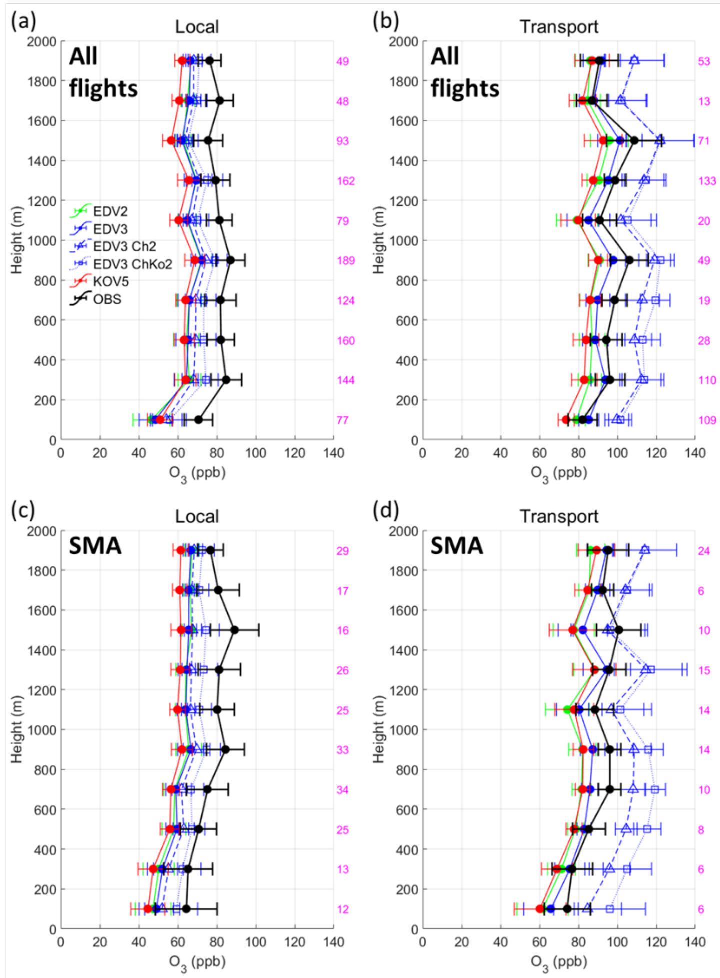

Figure R8. Vertically averaged O_3 from DC-8 (black), EDV2 (green), EDV3 (blue), and KOV5 (red) for the Local and Transport cases under 2 km height above ground level. The 1/2 of standard deviations are represented with black whiskers in each 200m layer. Sensitivity tests are conducted with doubled anthropogenic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

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Reply to the review 2 of “Changes in surface ozone in South Korea on diurnal to decadal time scale for the period of 2001-2021”

Thank you for your comments that improve our manuscript. Our replies to the specific comments are written below (the reviewer’s comments in blue and our replies in black).

Specific Comments

L2 P2: I believe there is a typo: Change “Increasing trends of tropospheric ozone in South Korea in the last decades have reported in several studies” to “Increasing trends of tropospheric ozone in South Korea in the last decades have been reported in several studies”.

→ We corrected this typo in the revised manuscript.

L4 P5: Could you give some details on the impact of the COVID-19 pandemic on the atmospheric composition in spring versus in summer? Has South Korea experienced several lock-downs in spring and summer 2020, or only in spring, with reduction of human activities/emissions of the precursors of ozone?

→ Nationwide social distancing protocol enforced by Korean government started February 25 of 2020 and lasted until April 18 of 2022, although levels of protocol differ. During spring in 2020 (until May 6, 2020), facilities for public use (libraries, swimming pools, museums, and national parks) and religious, indoor sports, entertainment facilities were forced to close, and people were refrain from going out except for buying necessities, visiting a doctor, and commuting to/from work. Since May 6 of 2020, as number of new confirmed COVID-19 cases remain relatively steady, the guidelines have shifted from social distancing to distancing in daily life, no restrictions on people going out. Because a cluster of new COVID-19 cases emerged in mid-August, social distancing protocol (since August 16 until early October) was again forced by the government, people were strongly recommended to stay indoors. After August 16 of 2020, there were well-defined government protocols as Level1, 2, and 3: Level1 is no restricted personal gathering and daily life, Level 2 allows personal gathering up to 8 people and discourage unnecessary and unurgent travel, and Level 3 allows personal gathering up to 3 people, requires remote work and online classes, and discourage travels. Most days in spring and summer in 2021 were the period under the Level2 protocol. In summary, most distinct changes in social-distancing protocols and traffic/mobile activities occurred between spring and summer in 2020. This discussion is now included in the revised manuscript.

L12 P5: I believe there is a typo: Change “as following” to “as follow”.

→ We corrected this typo in the revised manuscript.

L7 P6: Could you be more specific? Could you give the starting year? Are all the 500 stations still working now? Maybe add a column “time period” in Table S1.

→ More specific information on the time period of the observations and missing period is given in a excel file as Supporting Information.

L9 P7: Could you be more specific on the stricter recommendations: quality assurance and cloud fraction?

→ The stricter recommended filter is selecting pixels passing quality assurance > 0.75 and cloud radiance fraction < 0.5.

L10 P7: Have you conducted or are you aware of any sensitivity test to see how much the compromise sampling statistics/quality may change the results?

→ We conducted the sensitivity test by applying different sampling conditions and found consistent results irrespective of quality control parameters: larger tropospheric NO_2 column reduction during spring than during summer between 2019 and 2020-2021 (COVID-19 periods). Differences between KNMI and NASA retrievals are large when the original filter was applied (quality assurance > 0.5 and cloud radiance fraction < 0.4). When the stricter filter was applied, differences between KNMI and NASA retrievals are small. Therefore, in the revised manuscript, the stricter filter (quality assurance > 0.75 and cloud radiance fraction < 0.5) is used. Since the NASA product released in November, 2022 were generated in a consistent manner for May 2018-December 2021, we presented the NASA MINDS product in the revised manuscript instead of the KNMI product. We summarized the sensitivity tests in the Supporting Information. The distribution of absolute tropospheric NO_2 columns for different years are also shown in the Supporting Information.

L4 P9: Typo: Change "11st" to "11th" (eleventh). Could you add the year?

→ Yes. We added year "2016". We corrected this part to 11th June 12 UTC in 2016.

L11 P10: Could you add the uncertainties on the trend estimate?

→ We included the uncertainties on the trend as the reviewer suggested.

L14 P10: "Insignificant" is not used anymore (Wasserstein et al., 2019). Trend reliability can be expressed with p-value (Wasserstein et al., 2019) and/or signal-to-noise (SNR) ratio (Chang et al., 2021). Then you can apply the trend reliability scale (see table below from the guidance note on best statistical practices for tropospheric ozone assessment report -TOAR-analyses by Kai-Lan Chang, Martin Schultz, Gerbrand Koren and co-authors pending their approval, February 2023; the document will be posted on the TOAR website by end of April 2023 upon the TOAR steering committee approval, https://igacproject.org/activities/TOAR/TOAR-II) to report the trend and its uncertainty.

Table 3. Trend reliability scale

p-value	SNR (signal-to-noise) value	Term
$p \leq 0.01$	SNR ≥ 3	very high certainty
$0.05 \geq p > 0.01$	$2 \leq$ SNR < 3	high certainty
$0.10 \geq p > 0.05$	$1.65 \leq$ SNR < 2	medium certainty
0.33[1] $\geq p > 0.10$	$1 \leq$ SNR < 1.65	low certainty
$p > 0.33$[1]	SNR < 1	very low certainty or no evidence

[1]This boundary is meant to be fuzzy around 1/3 (Mastrandrea et al., 2010).

Table taken from the guidance note on best statistical practices for tropospheric ozone assessment report -TOAR-analyses by Kai-Lan Chang, Martin Schultz, Gerbrand Koren and co-authors pending their approval, February 2023; the document will be posted on the TOAR website upon the TOAR steering committee approval, https://igacproject.org/activities/TOAR/TOAR-II.

→ We added p-value and SNR in a separate Table in the main text. The table is displayed below.

Table R4. Trends estimates based on the 4th highest MDA8 O_3 values

Location		Slope (ppb yr^{-1})	2-Sigma (ppb yr^{-1})	P value	SNR
City	Seoul (SUL)	1.19	0.38	< 0.01	6.23
	Incheon (INC)	1.07	0.37	< 0.01	5.72
	Daejeon (DJN)	1.22	0.49	< 0.01	4.96
	Gwangju (GWJ)	0.98	0.46	< 0.01	4.30
	Busan (BSN)	0.98	0.36	< 0.01	5.47
	Ulsan (ULS)	1.40	0.34	< 0.01	8.14
	Daegu (DGU)	1.12	0.46	< 0.01	4.89
Province	Gyeonggi-do (GGI)	1.26	0.27	< 0.01	9.33
	Chungcheongbuk-do (CCB)	0.79	0.51	< 0.01	3.09
	Chungcheongnam-do (CCN)	1.45	0.47	< 0.01	6.12
	Jeollabuk-do (JLB)	1.83	0.32	< 0.01	11.30
	Jeollanam-do (JLN)	0.08	0.39	0.67	0.41
	Jeju Island (JEJ)	0.66	0.46	< 0.01	2.89
	Gyeongsangnam-do (GSN)	0.83	0.52	< 0.01	3.18
	Gyeongsangbuk-do (GSB)	1.10	0.35	< 0.01	6.32
	Gangwon-do (GWO)	0.67	0.48	< 0.01	2.79

L5 P11: Spell out LT = Local Time, at least the first time it is used.

→ Corrected.

L12 P11: It would be worth adding a discussion with references on summer/spring differences: meteorology condition in Seoul and Gyeonggi-do compared with other sites/regions. That would probably fit in the "Discussions" section.

→ The mean temperature, mean maximum temperature, and mean wind velocity values during spring and summer, 2001 – 2021 are listed in Table R5. Unlike opposite patterns of spring/summer peak time ozone in Seoul and Gyeonggi-do, the meteorological factors show similar differences in the area of interests. Thus, the meteorological factors are not main drivers of high summertime exceedances in Seoul and Gyeonggi-do region. The data are obtained from the Korea Meteorological Administration (KMA) website (https://data.kma.go.kr/).

Table R5. 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.

Location		Mean temperature (°C)	Mean maximum temperature (°C)	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-do	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)

L15 P11: I found 7 sites showing more exceedances in summer than in springs according to Figure 4. Why do you report only 3 of them? I also found 10 sites showing more exceedances in spring than in summer, why do you report only 3 of them?

→ We just exemplified the diurnal cycles for representative cases since Figure 4 also have this information. In the revised manuscript, we included the diurnal variations at all locations in the Supporting Information.

L7 P12: "than Incheon" is not clear. I believe there is a typo in the sentence. Could you rephrase?

→ We changed to "compared to the time of exceedance in Incheon".

L13-14 P12: Is it a statement from previous studies or from this current study? Could you give a reference or cite a figure to support this statement?

→ During nighttime, NO reacts with ozone forming NO_2 and oxygen molecule, which is the main loss of ozone (Jacob, D. J., 1999; Seinfeld and Pandis, 2016). In Figure R9, both model and observations exhibit high NO_2 concentrations and low ozone concentrations during night.

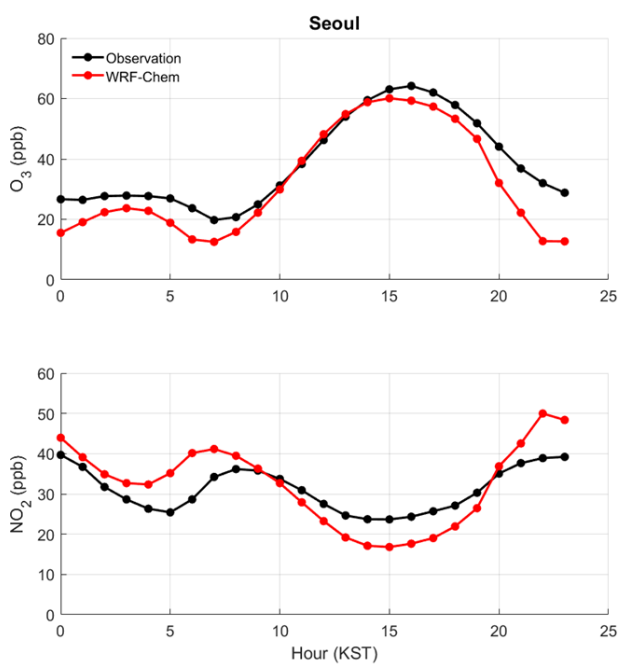

Figure R9. The diurnal variations of observed and simulated O_3 and NO_2 averaged for the simulation period.

L20 P14: Does "large reduction of ozone" refer to the difference between the time periods P2 and P3? It would be helpful to clarify.

→ Yes. It means the time periods P2 and P3. We clarified it.

L14 P15: Does "likely to be VOC-limited" mean that VOCs did not decrease between P2 and P3 in South Korea? Any reference?

→ It meant that "VOC-limited" is a dominant photochemical regime in the cities over South Korea (e.g., Kim et al., 2020). We clarified in the manuscript.

L20 P15: Do we know why there are more NO_2 in MAM 2019 than JJA 2019? Specific human activities, Meteorological conditions? It would be interesting to see the maps of MAM 2020 and JJA 2020.

→ Meteorological condition such as sunlight is the main driver. NO_2 concentrations at surface or vertically integrated column concentrations are lower during summer than during spring because of enhanced OH radical concentrations due to increased sunlight during summer increase loss of NO_2 via a reaction of NO_2 with OH (Martin et al., 2003; Lamsal et al., 2010). The reduced chemical lifetime of NO_x leads to decreased NO_2 columns in JJA 2019 compared to those in MAM 2019. We also included the maps of TROPOMI NO_2 columns for MAM 2020 and JJA 2020 in the Supporting Information.

L20 P16: Why did you choose Seoul and Gangwon-do over other sites?

→ In the reply to the Reviewer1, we explained the reason to investigate Gangwon-do, in particular Gosung. The elevations of monitoring sites in Gangwon-do are high as in Table R6. Gosung (Ganseong-eup in Table R6) is elevated to ~600 m, is located to leeside of mountain, and is close to the East Coast of South Korea. Therefore, this remote site is ideally located to investigate the impacts of long-range transport of ozone at high elevations.

Table R6. Altitudes (m) of monitoring sites in Gangwon-do. Ganseong-eup represents Gosung.

	Name	Latitude	Longitude	Altitude
	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
Gangwon	**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

L1 P17: An evaluation of WRF-Chem above Seoul and Gangwon-do would be helpful. How does the control run compare with the observations? Any sondes launched during KORUS-AQ that can be used for this evaluation? Was this model study done with annual means or did you perform it for a specific season? Showing summer and spring would be useful to echo the seasonal results on trends estimate.

→ The model results from the WRF-Chem control run were compared with the observations from the surface monitor over Seoul and Gosung in Figure R10 and R11. The model decently simulated the observations in an hourly basis (Figure R10) and on average (Figure R11). The model was conducted for the KORUS-AQ field campaign (May 1 – June 10 in 2016) and was averaged for the period. The model simulation period covers mainly springtime. Longer simulations will be required to contrast spring and summer. This is an interesting modeling topic for future study. In reply to the Reviewer 1, we showed the evaluations of vertical profiles of simulated ozone with the DC-8 aircraft observations.

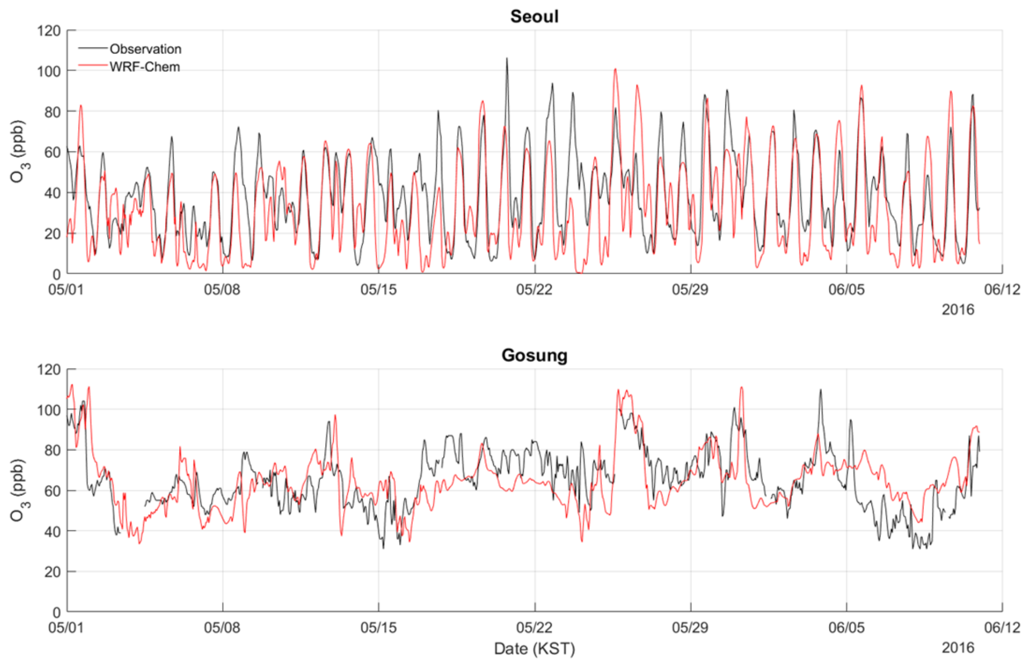

Figure R10. The time series of observed and simulated hourly ozone in (top) Seoul and (bottom) Gosung. Basic statistics are shown as follows. Mean bias (MB): Seoul -6.2 ppb /Gosung -0.9 ppb, Root Mean Square Errors (RMSE): Seoul 18.2 ppb/Gosung 13.7 ppb, Correlation Coefficient(R): Seoul 0.68/Gosung 0.54.

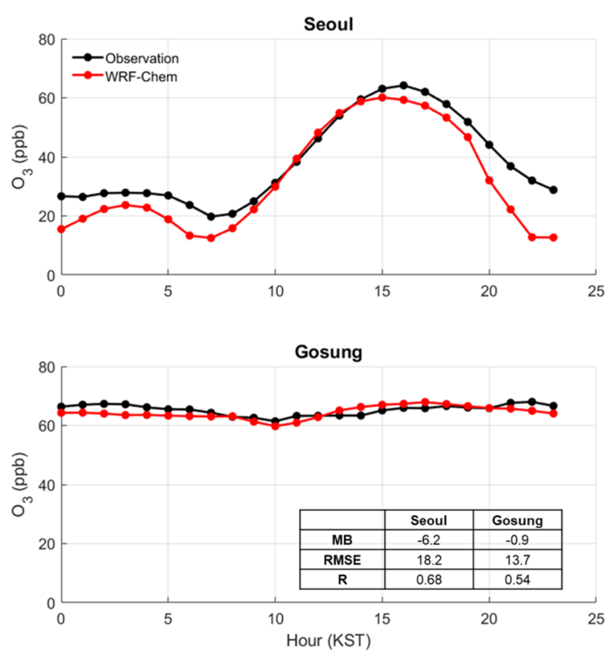

Figure R11. Diurnal variations of observed and simulated ozone concentrations averaged for the entire simulation period: (top) Seoul and (bottom) Gosung. Basic statistics are shown in the plot.

L7 P17: It seems to be very small changes (almost none). Could you be more quantitative?

→The reduction is -1 ppb (-2%). In the revised manuscript, we used EDGAR-HTAPv3 emission inventory. This statement was omitted.

L3-5 P18: You probably should inform on the altitude of both Gosung and Gangwon-do sites because it is a little confusing as it is written.

→ The altitudes are informed in Table R6. We include this information in Supporting Information.

L1-2 P35: Are NO_2 and CO values from CAM-Chem? It is worth clarifying in the caption.

→ The trends are calculated from the surface monitor observations (www.airkorea.or.kr). We clarified it.

L2 P36: Can you have colors or signs to differentiate cities, provinces and background sites, as well as the definitions of these three categories. Is it according to ozone diurnal/seasonal variability? Could you add a legend?

→ We used the colors to differentiate the three categories. We added it to the Figure caption.

L2 P37: Could you add the uncertainties (2-sigma values), or p-value or signal-to-noise ratio associated with the slope values S? (see my previous comment on how to report trend and its uncertainty)

→ We added p-value and SNR in the newly added Table in the revised manuscript.

L4 P41: Is the extraction over the entire country? It should be specified in the caption and section 2.4.

→ Yes. It was extracted over the entire country. Now we include this information in the Figure caption in the revised manuscript.

L4 P44: Typo in the legend of Figure 11: change “Contorl” to “Control”

→ The typo is corrected in the revised manuscript. Thank you for paying attention to detail.

References:

Chang, K.-L., Schultz, M. G., Lan, X. et al. (2021). Trend detection of atmospheric time series: Incorporating appropriate uncertainty estimates and handling extreme events. Elementa: Science of the Anthropocene, 9.

Wasserstein, R.L., Schirm, A. L. & Lazar, N. A. (2019). Moving to a world beyond “p < 0.05". The American Statistician.

References

Jacob, D. J. (1999), Introduction to Atmospheric Chemistry, *Princeton University Press*, 260pp.

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.44 4.

Lamsal, L. N., et al., 2010, Indirect validation of tropospheric nitrogen dioxide retrieved from the OMI satellite instrument: Insight into the seasonal variation of nitrogen oxides at northern midlatitudes, 115, D5, https://doi.org/10.1029/2009JD013351.

Randall, V. M., et al., 2003, Global inventory of nitrogen oxide emissions constrained by space-based observations of NO2 columns, 108, D17, https://doi.org/10.1029/2003JD003453.

Seinfeld, J. H. and Pandis, S. N. (2016), Atmospheric Chemistry and Physics: From Air Pollution to Climate Change (3rd ed.), Wiley, 1152pp.

Reply to the review 3 of “Changes in surface ozone in South Korea on diurnal to decadal time scale for the period of 2001-2021”

Thank you very much for your insights about trend and seasonality of background ozone values at northern midlatitude. The background ozone beyond Asia should have been discussed in the manuscript. In the revised manuscript, we included the references and mentioned this point. We also thank you for recognizing the strengths of our study. Our replies to the major concerns and specific comments are written below (the reviewer’s comments in blue and our replies in black).

The reviewer’s main concern was the use of surface O_3 data from the base time (01-06 LT) to gain information about background value because O_3 loss reacting with NO is dominant at this time over the highly polluted area. It is typical to ignore the data at this time when analyzing trends over polluted regions. However, in this study, we would like to utilize O_3 data at this time to find information about background O_3 because ozone is transported throughout a day and this process is very important in the region of study. The Figure R12 shows the WRF-Chem simulated surface O_3 in Seoul from various emission scenarios. Blue line in the plot denotes the model results only with local emissions (zero-out Chinese emissions, labeled as “No China”) and black line represents the results from Control run with all emissions. The local emissions case (blue line) shows much reduced O_3 compared to the Control case throughout a day (including 01-06 LT). The difference between the Control case (black line) and local emissions case (blue line) at 01-06 LT indicates increase of ozone from transport from upwind sources at this time.

High NO_x condition in Seoul tends to suppress the photochemical production of O_3 during daytime and enhance O_3 destruction during nighttime as exhibited in differences between black (Control case) and magenta lines (zero-out Seoul emission case, labeled as “No Seoul”). This indicates that chemistry plays a critical role in determining O_3 value in Seoul. Therefore, similarity of mean O_3 values in the Control case to clean background tropospheric O_3 value (climatological value) may be just a coincidence. These modeling exercises demonstrate that O_3 at the base time can be analyzed to derive information about background ozone even over the highly polluted (high NO_x) sites. The other point the reviewer commented is the impact of different NO_x concentrations during spring and summer on background ozone at 01-06 LT. Because it is not daytime, differences in boundary layer height between the two seasons should be small. Lower stable boundary layer height during summer than during spring is not well theoretically supported.

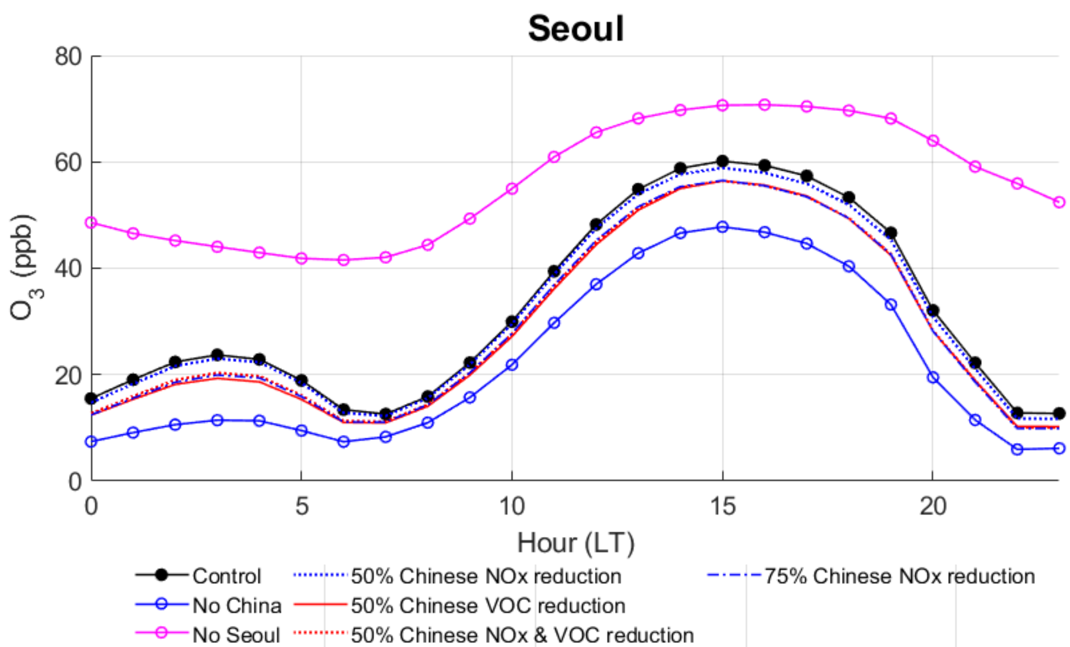

Figure R12. Diurnal variations of the model ozone concentrations at surface from various emission scenarios. The model results were averaged for the full simulation period.

The reviewer's suggestion to construct frames to analyze O_3 in South Korea by using the observations and chemical transport model results sounds interesting, but we are not sure if that can/should be conducted in this study. It would not be straightforward to delineate background O_3 (without continental influences) and to assess the impacts of local South Korean emissions and Asian mainland emissions by mainly analyzing observations for the complex atmospheric environment of South Korea. We agree with the reviewer to the point that the models like CESM constrain many important parameters to develop the model reproducing O_3 seasonality and trends. To rely on the models, however, the uncertainties of the models should be well accounted for. This alone is a quite challenging work. It would be interesting to conduct research the reviewer suggested. But it would require considerable times and that work would be beyond the scope of this study. In this study, we used models to help interpret observations as shown in the discussion section in the original manuscript, which is moved to the result section in the revised manuscript.

Summary:
This a very useful and informative paper. It has two major strengths: 1) Investigation of ozone in a region with strong local anthropogenic emissions, that also receives marine inflow with a highly polluted continent lying directly upwind of the marine area. 2) An effective incorporation of both observation and modelling based analysis. However, I believe that a major revision of the paper is required before it is ready for publication. One major need is for the authors to begin their observation-based analysis with a consideration of the ozone distribution that would be present in South Korea if there were no continental influences, i.e., if observed concentrations were due to transported baseline ozone alone. That consideration can rely on both the CESMv2.2 model calculations of these ozone concentrations (evidently shown in Figure 6), where results at ~1 km likely represent baseline ozone, and on analysis of observations as suggested below in the first major issue. This consideration would then provide a basis for understanding the continental influences, both from local South Korean emissions and from the Asian mainland emissions. Also much of the discussion is difficult to follow and requires substantial improvement; suggestions in this regard are given in the major and minor issues described below.
→ Please see the replies above.

Major issues:

1) I believe that the discussion based on Table 1 requires reconsideration. I assume that these are mean ozone concentrations for the peak and base time periods in spring and summer. First, I think the period names are misleading. The 10-20 LT period has higher ozone concentrations than does the 01-06 period. However, those higher (10-20 LT) ozone concentrations are similar to that expected for northern mid-latitude baseline ozone concentrations. For example, Figure 5 of Parrish et al., (2020) shows that annual mean ozone is 30 to 40 ppb in the lower 1 km of the troposphere. Figure S14 of that paper shows that ozone at Mt. Walinguan (upwind of South Korea, but at higher elevation) has mean ozone of about 45 to 60 ppb in spring and summer. To my mind, the mean ozone in Table 1 in the 10-20 LT period predominately reflects baseline ozone transported into the country; this is the reason that these mean concentrations are similar throughout the country.

→ We agree with the reviewer about the possibility of baseline ozone transported into the country, judging from similar mean values throughout the country. However, Figure R12 also illustrates various responses of surface ozone to emission scenarios in Seoul. It demonstrates that chemistry is an important factor to determine mean annual ozone in Seoul and other regions in South Korea. Therefore, we would like to avoid oversimplification of factors to determine the ozone in South Korea.

2) If the interpretation above is correct, then the lower ozone concentrations in the 01-06 period are caused by loss of ozone due to surface deposition and reaction with fresh NO emissions under a shallow nocturnal inversion. Such a diurnal cycle (low at night, higher during the day) is a ubiquitous feature of urban ozone.

→ Agreed. See the replies above (including Figure R12). Ozone in the 01-06 LT period is lower than that in the 10-20 LT period because of different chemical and physical processes involved. But there are still influence of transport in the 01-06 LT period as shown in Figure R12. Therefore, we would like to use the data in the 01-06 period.

3) To emphasize the similarity of the ozone concentrations throughout the country, and the predominant role of transported baseline ozone, I suggest that the background sites be included in Figure 3 and Table 1.

→ It was difficult to derive the trends for the background sites because some of ozone season data are missing for the sites. The data from March 31, 2011 to August 31, 2011 are missing in Gosung, Gangwon-do. The data from Mary 1, 2012 to June 8, 2012 are missing in Gosan, Jeju. The data from March 30, 2011 to June 30, 2011 are missing in Ulleung Island (in Gyeongsangbuk-do). Therefore, we limit the trend analysis for the region with multiple monitoring sites covering the full period of analysis.

4) More generally, I suggest that all tables, figures and discussion clearly address the 7 cities, 9 provinces, and 3 background sites in a consistent manner to the fullest extent possible. The discussion is often difficult to follow when varying lists of cities, provinces and sites are mentioned.

→ We presented the results for the 7 cities, 9 provinces, and 2 background sites consistently in the revised manuscript whenever possible. We omitted Gosan, Jeju Island because NO_2 and CO data need quality assurance from mid of 2010 to current date. The names of the sites were updated consistently throughout the manuscript. For the trend study (Figure 2 and 3), we did not include the background sites because of some missing data during ozone season.

5) The primary reason that mean ozone is generally higher in spring than in summer is that the lower troposphere baseline ozone is higher in spring than in summer, particularly in marine influenced air; e.g., see Figures 4 and 6 of Parrish et al., (2020).

→ Thank you for the reference. We explained the seasonal difference including marine influenced air in the revised manuscript and referred to Parrish et al (2020).

6) Pg. 11, lines 5-8: One reason the 01-06 LT ozone is higher in the spring is that the nocturnal inversion is tighter in the summer, so ozone loss at night is more pronounced in summer than in spring. Given the very local processes that determine the 01-06 LT ozone, for simplicity, the authors may wish to eliminate the discussion of this nighttime ozone.

→ We don't think that there are clear mechanisms driving differences in nocturnal inversion between spring and summer. See the replied above for the reason why we keep the discussions about the ozone concentrations in the 01-06 LT period.

7) A discussion of local CO and NOx trends begins near the bottom of pg. 13. These observation-based trends should be compared and discussed in relation to the trends of these species derived from the model emission inventories. This may be more relevant to NOx, since it does have more local influence than CO.

→ We listed the trends of NOx and CO emissions from linear fits of the data covering 2001-2020, obtained from Clean Air Policy Support System (CAPSS) emission inventory (https://www.air.go.kr/) (Table R7 and R8 for emission inventories and ambient concentrations, respectively). Overall, signs of slopes agree between emission inventory and ambient concentrations at least for the cities, but site-to-site variations do not agree even for the cities. And there are disagreements of signs of slopes between emission inventory and ambient concentrations for the provinces. This can be attributed to the uncertainties in long-term emission inventories of NOx and CO.

Table R7. 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-do	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 R8. 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)

8) The discussion of the COVID-19 influence on ozone (pg. 14-15) is interesting, particularly the “large reduction of ozone in the background sites”. There are other studies of the influence of COVID-19 emission reduction on background ozone at northern mid-latitudes. The findings in these other studies should be quantitatively compared to the present results.

→ Thank you for introducing publications. In the revised manuscript, we refer the study by Steinbrecht et al. (2021) that reported about 7% reductions of mid-latitude free atmosphere ozone concentrations in 2020 from the climatology value covering 2000-2020. Our study focused on the analysis surface ozone over South Korea that substantially increased for the period of 2000-2020. Thus, it is not straightforward to quantitatively compare the anomaly in 2020 from climatology in this study with that in Steinbrecht et al. (2021). It is still worthwhile to mention agreement in declining ozone concentration/exceedances during COVID in our study and Steinbrecht et al (2021).

9) I find the discussion beginning on line 14, page 13 and continuing to the end of the Results section on page 16 to be very confusing, with many topics discussed in a disjointed manner. Please revise and clarify this discussion. Any topic that cannot be clearly and concisely explained without speculation should be eliminated.

→ Agreed. There are indeed many topics. Following your suggestions, to clarify the contents, we made several subsections with appropriate titles within the results section. The discussion section is also incorporated into the result section. The titles for the subsections in the results section in the revised manuscript are written below.

3.1 Surface ozone trends

3.2 Difference between spring and summer: background value, exceedance, stratospheric influence, and precursor concentrations

3.2.1 Background values at the base and peak times

3.2.2 Ozone exceedances

3.2.3 Influence of stratospheric ozone

3.2.4 Long-term trends of surface NO_2 and CO concentrations

3.3 Changes detected during the COVID-19 pandemic (2020-2021) compared to 2002-2019

3.3.1 Changes in ozone exceedances and local precursors during springtime

3.3.2 Changes in ozone exceedances and local precursors during summertime

3.3.3 Changes in precursor concentrations at a regional scale during spring and summer: TROPOMI tropospheric NO_2 columns

3.4. Impacts of changes in East Asian emissions on surface/boundary layer ozone in South Korea: a modeling analysis

3.4.1. Changes in surface/boundary layer ozone due to emissions reductions: East Asian region

3.4.2. Vertical sensitivity of ozone changes in South Korea to East Asian emission Reductions

3.4.3. Comparisons with recent modeling research

10) Similarly, the Section 4 discussion section is difficult to follow. The authors should aim to convey the main points of the modeling results as clearly and concisely as possible. The last two sentences of the section appear to be the main points; they should be clearly and concisely supported by the preceding discussion.

→ The discussion section is now incorporated into the results section for better support of the content and a smooth connection. We emphasized the last two sentences by reorganizing the results section and adding more explanations.

11) The Summary and Conclusions section will need to be rewritten when the issues identified here are addressed. Specifically:

- The ozone in the 01-06 LT period is so affected by local conditions that it should not be included in the 2nd paragraph of this Section.

→ Please see our replies above. We kept using data at 01-06 LT to get information about background/transport.

- Page 19, discussion beginning on line 17 should be improved. If there is strong influence of long-range transport on the surface ozone at the background sites, then that influence must also be present at all sites throughout South Korea. That influence is not apparent at night at most sites due to rapid nighttime loss of ozone at most sites.

→ Please see our replies above. Even with rapid nighttime loss of ozone at most sites, there is still information about long-range transport. We would like to maximize the use of the data at 01-06 LT.

- An explanation should be given as to why there is such large regional differences in overall percentage decline in NO_2. Perhaps this can be related to the model emission inventory?

→ There are many sources of NO_2 besides mobile sources in South Korea, such as power plant and industries. Thus, decline of NO_2 varies at the monitoring sites that have different source profiles. As mentioned, uncertainty in the emission inventory is generally large and was not extensively estimated.

Minor issues:

1) Pg. 4, Line 11: Four references are given for papers that have previously reported increasing ozone trends in South Korea. Two of those are missing from the reference list. The introduction should briefly summarize what these papers found, and discuss the advances that the authors' make in this paper beyond what is known from those earlier papers.

→ The missing references were added. We clarify the contribution of our study compared to the previous study. In reply to the Reviewer 1, we wrote "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)". This state some of our contributions to ozone analysis over South Korea, compared to the previous studies. This study reveals characteristics of newly defined exceedances (hourly O3 concentration > 70 ppb) that captured large changes of ozone during COVID and emphasizes long-range transport of ozone over eastern part of South Korea such as Gangwon-do and Ulleung-Island.

2) There are minor problems with the English usage, which should be corrected by editing by a native English speaker.

→ We improved English for the revised manuscript with the aid of a native speaker without changing contents.

3) Page 5, line 9 mentions that 8 provinces are studied; however Table 1 lists 9 provinces. Please develop a list of cities, provinces, and background sites, and consistently use that list throughout the paper.

→ We kept listing 9 provinces in the main text, tables, and figures in the revised manuscript.

4) In the Figure 1 caption, the different colors used for the city province and site names should be described. Also it is not clear exactly what is being plotted here: Is each symbol the mean 4th highest (MDA8) over all sites in the city or province? Confidence limits should be given for all derived slopes.

→ In the revised manuscript, we explained the meaning of different colors in Figure 1. In Table 1 in the revised manuscript, we showed slope, standard deviation, P-value, and signal-to-noise value. The information about all sites in the city or province is shown in the Supporting Information.

5) In the description of the two models evidently different anthropogenic emission inventories are used in the two models (CMIP6 for 2000-2014 and SSP5-8-5 for 2015-2020 in CAM-Chem and WRF-Chem and EDGAR-HTAPv2). There should be a brief discussion regarding how well these inventories compare, and if any problems arise

from using perhaps incompatible emissions in the two models. Also mention should be made regarding whether these inventories correctly simulate the emissions reductions during the COVID-19 period.

→ CMIP6 is based on EDGAR v4.2 or v4.3.2 described in Feng et al. (2020). SSP5-8.5 and EDGAR-HTAP v3 can be compared for the KORUS-AQ campaign period in 2016, as the WRF-Chem simulations were conducted during the period. In this reply, we compared NO_x emissions of SSP5-8.5, EDGAR-HTAP v3, v2, and KORUS v5. In Table R9, 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.

We acknowledge the emission differences for the two models. However, we are conducting research utilizing CAM-Chem and WRF-Chem separately for different purposes. Separate papers for different models are in review and in preparation. In this study, we utilized the results from CAM-Chem to analyze the contribution of stratospheric ozone to tropospheric ozone and use WRF-Chem model to investigate the impacts of anthropogenic emission changes on local and regional air quality. Thus, one-to-one comparison of the two models are beyond the scope of this study.

Table R9. The area sum emissions in Eastern China (27.7-40̊N, 115-123̊E), South Korea (34.5-38̊N, 126-130̊E), and Seoul Metropolitan Area (SMA: 37.2-37.8̊N, 126.5-127.3̊E) in May 2016 for NOx.

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

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

6) Page 10, line 11-12: For greater accuracy, I suggest changing "... increases by 1-2 ppb yr-1 for most of cities and provinces across South Korea ..." to "... increases by 1.0-1.5 ppb yr-1 for most cities and provinces across South Korea ..."

→ We changed it to 1.0-1.5 ppb yr^{-1}.

7) Page 10, line 12-13: For greater accuracy, I suggest changing "The most of cities and provinces have the 4th highest MDA8 O3 higher than 70 ppb after 2010." to "In nearly all cities and provinces, the 4th highest MDA8 O3 has been higher than 70 ppb since 2010 or earlier."

→ Thank you for your suggestion. We replaced the original sentence by the one the reviewer suggested.

8) I suggest vertical lines be added to Figure 4 to separate the cities, provinces, and background sites from each other. Similarly for Figures 7 and 8. Also simplify the figure captions.

→ We noted cities, provinces, and background sites with labels and lines in Figure 4, 7, and 9. The names of the location were redefined and were used consistently throughout the manuscript.

9) The discussion illustrated in Figures 4, 5 and 7 is based on "exceedances"; however, I cannot find where "exceedance" is defined in the paper. Please define. (I assume it is a day when MDA8 ozone exceeds 70 ppb).

→ Agreed. The mistakes in the abstract were corrected. The definition of exceedances is clarified in the abstract. In this study, exceedance is defined as hourly O_3 > 70 ppb.

10) Figure 5 needs to be clearly explained. If an exceedance is based on MDA8, how can there be a diurnal cycle, since there is only one MDA8 per day? Is this percent of days with ozone above 70 ppb in a given hour? I suggest using the same ordinate scale in all 3 graphs, so that the comparison is made easy for the reader. Also the general description of the sites included in the 3 graphs should be given; i.e., top = Seoul area, middle = secondary cities, bottom = remote sites.

→ Please see the reply above for minor point (9). We also used the same ordinate scale for Figure 5. The general description of the sites is included in the figure caption as suggested by the reviewer.

11) It seems that the information included in Table 2 and Figure 6 are identical; I suggest that Table 2 be eliminated.

→ Agreed. We deleted Table 2 in the original manuscript and moved to the Supporting Information for the readers who may want to obtain the details.

12) Please give units for the slopes in Table 3; confidence limits should be given for the derived slopes. Also please give the slopes for the background sites for comparison, if those data are available.

→ The units are shown in the table caption. The results from statistical analysis are included in the revised manuscript. Because of discontinuous record of the data, the slopes for the background sites are not shown.

13) Figure 11 – x-axis labels have typo.

→ Corrected.

14) Page 20 – Please define SMA

→ SMA (Seoul Metropolitan Area) was defined in Page 2 in the original manuscript.

References:

Parrish, D.D., et al. (2020), Zonal similarity of long-term changes and seasonal cycles of baseline ozone at northern mid-latitudes. J. Geophys. Res.: Atmos., doi: 10.1029/2019JD031908.

References

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

Summary of changes in the revised manuscript in response to reviews

Here we inform the reviewers and the editor of the updates in the revised manuscript corresponding to reviewer's comments. We improved English in the manuscript, but kept the contents almost the same.

The review is written in blue and our updates in black.

- **Reviewer 1**

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.
→ P2, L2 in the revised manuscript.

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).
→ Added missing or new references in the Reference section. Refer to P4, L 9-13.

P4,L11 - "Ozone in South Korea ..." this sentence requires a citation.
→ Refer to P4, L 14-15.

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.
→ P10, L13-P11 L4

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.
→ P12, L15-18. Also refer to Supporting Information 1 (SI1) Table S2-S4 and Figures S3-S8.

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

→ P11, L14

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?

→ Refer to our reply to Reviewer 1's comment. Evaluations of meteorology can be found in P10 L10-11 and SI1 Figure S1. P12 L16-18 and SI1 Table S4.

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

→ P12, L 7-9.

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?

→ Please refer to our reply to reviewer1's comment.

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.

→ P13, L17-L20.

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

→ P13, L20-P14 L2.

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.

→ P28-p29, Section 3.4.3 Comparisons with recent modeling research

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.

→ P10, L13-P11, L4 and SI1 Figure S2. Also refer to P18, L5-8.

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.

→ P28-p29, Section 3.4.3 Comparisons with recent modeling research

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.

→ P21, L20-P22, L9.

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.

→ P25, L10-L16. Also refer to SI1 Table S3 and Figure S8.

- **Reviewer 2**

Specific Comments

L2 P2: I believe there is a typo: Change “Increasing trends of tropospheric ozone in South Korea in the last decades have reported in several studies” to “Increasing trends of tropospheric ozone in South Korea in the last decades have been reported in several studies”.

→ P2, L2 in the revised manuscript.

L4 P5: Could you give some details on the impact of the COVID-19 pandemic on the atmospheric composition in spring versus in summer? Has South Korea experienced several lock-downs in spring and summer 2020, or only in spring, with reduction of human activities/emissions of the precursors of ozone?

→ P19, L19-P20 L15 and SI1 Figure S14-15.

L12 P5: I believe there is a typo: Change “as following” to “as follow”.

→ P6, L5.

L7 P6: Could you be more specific? Could you give the starting year? Are all the 500 stations still working now? Maybe add a column “time period” in Table S1.

→ Updated Table S1 in Supporting Information 2 (SI2, an excel file) including data availability.

L9 P7: Could you be more specific on the stricter recommendations: quality assurance and cloud fraction?

→ P8 L6-P9 L17 and Supporting Information 3 (SI3) summarizing TROPOMI data quality assurance tests.

L10 P7: Have you conducted or are you aware of any sensitivity test to see how much the compromise sampling statistics/quality may change the results?

→ P8 L6-P9 L17 and Supporting Information 3 (SI3) summarizing TROPOMI data quality assurance tests.

L4 P9: Typo: Change "11st" to "11th" (eleventh). Could you add the year?

→ P11, L13-L14.

L11 P10: Could you add the uncertainties on the trend estimate?

→ Table 1 in the revised manuscript.

L14 P10: "Insignificant" is not used anymore (Wasserstein et al., 2019). Trend reliability can be expressed with p-value (Wasserstein et al., 2019) and/or signal-to-noise (SNR) ratio (Chang et al., 2021). Then you can apply the trend reliability scale (see table below from the guidance note on best statistical practices for tropospheric ozone assessment report -TOAR-analyses by Kai-Lan Chang, Martin Schultz, Gerbrand Koren and co-authors pending their approval, February 2023; the document will be posted on the TOAR website by end of April 2023 upon the TOAR steering committee approval, https://igacproject.org/activities/TOAR/TOAR-II) to report the trend and its uncertainty.

Table 3. Trend reliability scale

p-value	SNR (signal-to-noise) value	Term
$p \leq 0.01$	SNR ≥ 3	very high certainty
$0.05 \geq p > 0.01$	2 ≤ SNR < 3	high certainty
$0.10 \geq p > 0.05$	1.65 ≤ SNR < 2	medium certainty
0.33[1] $\geq p > 0.10$	1 ≤ SNR < 1.65	low certainty
$p > 0.33$[1]	SNR < 1	very low certainty or no evidence

[1]This boundary is meant to be fuzzy around 1/3 (Mastrandrea et al., 2010).

Table taken from the guidance note on best statistical practices for tropospheric ozone assessment report -TOAR-analyses by Kai-Lan Chang, Martin Schultz, Gerbrand Koren and co-authors pending their approval, February 2023; the document will be posted on the TOAR website upon the TOAR steering committee approval, https://igacproject.org/activities/TOAR/TOAR-II.

→ Table 1, 3, and 4 including uncertainties in trend estimates.

L5 P11: Spell out LT = Local Time, at least the first time it is used.

→ P14, L15.

L12 P11: It would be worth adding a discussion with references on summer/spring differences: meteorology condition in Seoul and Gyeonggi-do compared with other sites/regions. That would probably fit in the "Discussions" section.

→ P15, L18-P16, L1 and SI1 Table S5.

L15 P11: I found 7 sites showing more exceedances in summer than in springs according to Figure 4. Why do you report only 3 of them? I also found 10 sites showing more exceedances in spring than in summer, why do you report only 3 of them?

→ P17, L4-L6 and SI1, Figure S11-S13.

L7 P12: "than Incheon" is not clear. I believe there is a typo in the sentence. Could you rephrase?

→ P16, L9 "compared to the time of exceedance in Incheon".

L13-14 P12: Is it a statement from previous studies or from this current study? Could you give a reference or cite a figure to support this statement?

→ Please refer to our reply to reviewer 2.

L20 P14: Does "large reduction of ozone" refer to the difference between the time periods P2 and P3? It would be helpful to clarify.

→ P21, L17, L20 and P22, L6.

L14 P15: Does "likely to be VOC-limited" mean that VOCs did not decrease between P2 and P3 in South Korea? Any reference?

→ P23, L3, Kim et al. (2020) in the reference section.

L20 P15: Do we know why there are more NO_2 in MAM 2019 than JJA 2019? Specific human activities, Meteorological conditions? It would be interesting to see the maps of MAM 2020 and JJA 2020.

→ Please refer to our reply to reviewer 2. For TROPOMI map, see SI3.

L20 P16: Why did you choose Seoul and Gangwon-do over other sites?

→ SI1 Table S6 and Figure S10. Also refer to P28 L2-L14.

L1 P17: An evaluation of WRF-Chem above Seoul and Gangwon-do would be helpful. How does the control run compare with the observations? Any sondes launched during KORUS-AQ that can be used for this evaluation? Was this model study done with annual means or did you perform it for a specific season? Showing summer and spring would be useful to echo the seasonal results on trends estimate.

→ SI1 Figure S4 and S5.

L7 P17: It seems to be very small changes (almost none). Could you be more quantitative?

→ In the revised manuscript, WRF-Chem model results utilizing EDGAR-HTAPv3 are presented. This part was removed.

L3-5 P18: You probably should inform on the altitude of both Gosung and Gangwon-do sites because it is a little confusing as it is written.

→ SI1, Table S6.

L1-2 P35: Are NO_2 and CO values from CAM-Chem? It is worth clarifying in the caption.

→ Captions in Table 3 and 4.

L2 P36: Can you have colors or signs to differentiate cities, provinces and background sites, as well as the definitions of these three categories. Is it according to ozone diurnal/seasonal variability? Could you add a legend?

→ P53 Figure 1 and caption.

L2 P37: Could you add the uncertainties (2-sigma values), or p-value or signal-to-noise ratio associated with the slope values S? (see my previous comment on how to report trend and its uncertainty)

→ Table 1, 3, and 4 in the revised manuscript (P49, P51, P52).

L4 P41: Is the extraction over the entire country? It should be specified in the caption and section 2.4.

→ P58, Figure 6 caption.

L4 P44: Typo in the legend of Figure 11: change “Contorl” to “Control”

→ P63, Figure 11.

- **Reviewer 3**

Major issues:

1) I believe that the discussion based on Table 1 requires reconsideration. I assume that these are mean ozone concentrations for the peak and base time periods in spring and summer. First, I think the period names are misleading. The 10-20 LT period has higher ozone concentrations than does the 01-06 period. However, those higher (10-20 LT) ozone concentrations are similar to that expected for northern mid-latitude baseline ozone concentrations. For example, Figure 5 of Parrish et al., (2020) shows that annual mean ozone is 30 to 40 ppb in the lower 1 km of the troposphere. Figure S14 of that paper shows that ozone at Mt. Walinguan (upwind of South Korea, but at higher elevation) has mean ozone of about 45 to 60 ppb in spring and summer. To my mind, the mean ozone in Table 1 in the 10-20 LT period predominately reflects baseline ozone transported into the country; this is the reason that these mean concentrations are similar throughout the country.

→ SI1, Figure S9. Please refer to our reply to reviewer3's comment.

2) If the interpretation above is correct, then the lower ozone concentrations in the 01-06 period are caused by loss of ozone due to surface deposition and reaction with fresh NO emissions under a shallow nocturnal inversion. Such a diurnal cycle (low at night, higher during the day) is a ubiquitous feature of urban ozone.

→ SI1, Figure S9. Please refer to our reply to reviewer3's comment.

3) To emphasize the similarity of the ozone concentrations throughout the country, and the predominant role of transported baseline ozone, I suggest that the background sites be included in Figure 3 and Table 1.

→ Please refer to our reply to reviewer3's comment.

4) More generally, I suggest that all tables, figures and discussion clearly address the 7 cities, 9 provinces, and 3 background sites in a consistent manner to the fullest extent possible. The discussion is often difficult to follow when varying lists of cities, provinces and sites are mentioned.

→ Please refer to our reply to reviewer3's comment. Because of potential issues in NO_2 and CO measurements in Gosan, Jesu Island, this site was omitted among the background sites.

5) The primary reason that mean ozone is generally higher in spring than in summer is that the lower troposphere baseline ozone is higher in spring than in summer, particularly in marine influenced air; e.g., see Figures 4 and 6 of Parrish et al., (2020).

→ Parrish et al (2020) in the reference section.

6) Pg. 11, lines 5-8: One reason the 01-06 LT ozone is higher in the spring is that the nocturnal inversion is tighter in the summer, so ozone loss at night is more pronounced in summer than in spring. Given the very local processes that determine the 01-06 LT ozone, for simplicity, the authors may wish to eliminate the discussion of this nighttime ozone.

→ Please refer to our reply to reviewer3's comment.

7) A discussion of local CO and NOx trends begins near the bottom of pg. 13. These observation-based trends should be compared and discussed in relation to the trends of these species derived from the model emission inventories. This may be more relevant to NOx, since it does have more local influence than CO.

→ SI1, Table S8 and S9. P19 L2-L7.

8) The discussion of the COVID-19 influence on ozone (pg. 14-15) is interesting, particularly the "large reduction of ozone in the background sites". There are other studies of the influence of COVID-19 emission reduction on background ozone at northern mid-latitudes. The findings in these other studies should be quantitatively compared to the present results.

→ P21,L16-P22, L9.

9) I find the discussion beginning on line 14, page 13 and continuing to the end of the Results section on page 16 to be very confusing, with many topics discussed in a disjointed manner. Please revise and clarify this discussion. Any topic that cannot be clearly and concisely explained without speculation should be eliminated.

→ Following Reviewer3, the section 3 was partitioned into several subsections.

3.1 Surface ozone trends

3.2 Difference between spring and summer: background value, exceedance, stratospheric influence, and precursor concentrations

3.2.1 Background values at the base and peak times

3.2.2 Ozone exceedances

3.2.3 Influence of stratospheric ozone

3.2.4 Long-term trends of surface NO_2 and CO concentrations

3.3 Changes detected during the COVID-19 pandemic (2020-2021) compared to 2002-2019

3.3.1 Changes in ozone exceedances and local precursors during springtime

3.3.2 Changes in ozone exceedances and local precursors during summertime

3.3.3 Changes in precursor concentrations at a regional scale during spring and summer: TROPOMI tropospheric NO_2 columns

3.4. Impacts of changes in East Asian emissions on surface/boundary layer ozone in South Korea: a modeling analysis

3.4.1. Changes in surface/boundary layer ozone due to emissions reductions: East Asian region

3.4.2. Vertical sensitivity of ozone changes in South Korea to East Asian emission Reductions

3.4.3. Comparisons with recent modeling research

10) Similarly, the Section 4 discussion section is difficult to follow. The authors should aim to convey the main points of the modeling results as clearly and concisely as possible. The last two sentences of the section appear to be the main points; they should be clearly and concisely supported by the preceding discussion.

→ The discussion section is incorporated into Section 3.

11) The Summary and Conclusions section will need to be rewritten when the issues identified here are addressed. Specifically:

- The ozone in the 01-06 LT period is so affected by local conditions that it should not be included in the 2nd paragraph of this Section.

→ This part was kept in the revised manuscript.

- Page 19, discussion beginning on line 17 should be improved. If there is strong influence of long-range transport on the surface ozone at the background sites, then that influence must also be present at all sites throughout South Korea. That influence is not apparent at night at most sites due to rapid nighttime loss of ozone at most sites.

→ Please refer to our reply to reviewer3's comment.

- An explanation should be given as to why there is such large regional differences in overall percentage decline in NO_2. Perhaps this can be related to the model emission inventory?

→ P19, L2-7.

Minor issues:

1) Pg. 4, Line 11: Four references are given for papers that have previously reported increasing ozone trends in South Korea. Two of those are missing from the reference list. The introduction should briefly summarize what these papers found, and discuss the advances that the authors' make in this paper beyond what is known from those earlier papers.

→ P4 L9-13. P5 L18-P6 L2.

2) There are minor problems with the English usage, which should be corrected by editing by a native English speaker.

→ We improved English for the revised manuscript with the aid of a native speaker. It did not affect the contents of the manuscript.

3) Page 5, line 9 mentions that 8 provinces are studied; however Table 1 lists 9 provinces. Please develop a list of cities, provinces, and background sites, and consistently use that list throughout the paper.

→ We kept listing 9 provinces in the main text, tables, and figures in the revised manuscript.

4) In the Figure 1 caption, the different colors used for the city province and site names should be described. Also it is not clear exactly what is being plotted here: Is each symbol the mean 4th highest (MDA8) over all sites in the city or province? Confidence limits should be given for all derived slopes.

→ Figure 1 and Table 1.

5) In the description of the two models evidently different anthropogenic emission inventories are used in the two models (CMIP6 for 2000-2014 and SSP5-8-5 for 2015-2020 in CAM-Chem and WRF-Chem and EDGAR-HTAPv2). There should be a brief discussion regarding how well these inventories compare, and if any problems arise from using perhaps incompatible emissions in the two models. Also mention should be made regarding whether these inventories correctly simulate the emissions reductions during the COVID-19 period.

→ Refer to our reply to reviewer3's comment.

6) Page 10, line 11-12: For greater accuracy, I suggest changing "… increases by 1-2 ppb yr-1 for most of cities and provinces across South Korea ..." to "… increases by 1.0-1.5 ppb yr-1 for most cities and provinces across South Korea ..."

→ P13, L13.

7) Page 10, line 12-13: For greater accuracy, I suggest changing "The most of cities and provinces have the 4th highest MDA8 O3 higher than 70 ppb after 2010." to "In

nearly all cities and provinces, the 4th highest MDA8 O3 has been higher than 70 ppb since 2010 or earlier."

→ P13, L14-15.

8) I suggest vertical lines be added to Figure 4 to separate the cities, provinces, and background sites from each other. Similarly for Figures 7 and 8. Also simplify the figure captions.

→ Figure 4, 7, and 8 separating the cities, provinces, and background sites.

9) The discussion illustrated in Figures 4, 5 and 7 is based on "exceedances"; however, I cannot find where "exceedance" is defined in the paper. Please define. (I assume it is a day when MDA8 ozone exceeds 70 ppb).

→ P2, L11-13. P30, L18-19.

10) Figure 5 needs to be clearly explained. If an exceedance is based on MDA8, how can there be a diurnal cycle, since there is only one MDA8 per day? Is this percent of days with ozone above 70 ppb in a given hour? I suggest using the same ordinate scale in all 3 graphs, so that the comparison is made easy for the reader. Also the general description of the sites included in the 3 graphs should be given; i.e., top = Seoul area, middle = secondary cities, bottom = remote sites.

→ Figure 5. P2, L11-13. P30, L18-19.

11) It seems that the information included in Table 2 and Figure 6 are identical; I suggest that Table 2 be eliminated.

→ SI1 Table S7.

12) Please give units for the slopes in Table 3; confidence limits should be given for the derived slopes. Also please give the slopes for the background sites for comparison, if those data are available.

→ Table 3 and 4.

13) Figure 11 – x-axis labels have typo.

→ Figure 11.

14) Page 20 – Please define SMA

→ P2, Line 13.