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 ≤ 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.

→ 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.

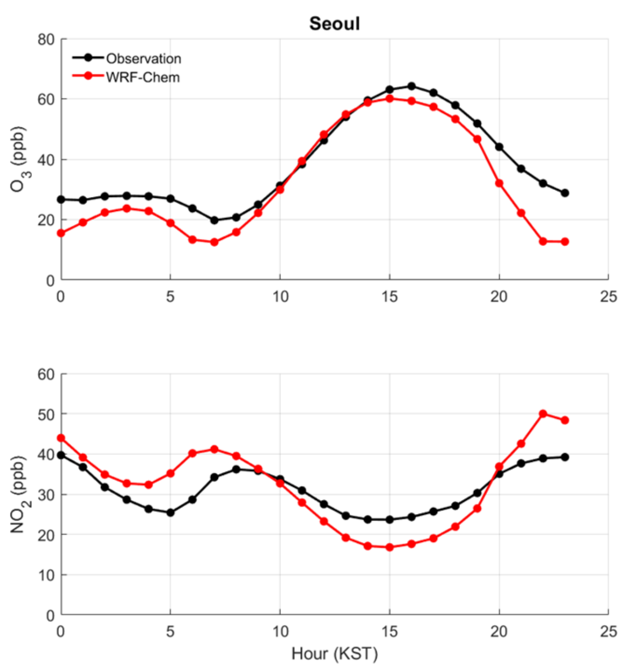

Figure R9. The diurnal variations of observed and simulated O_3 and NO_2 averaged for the simulation period.

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

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.

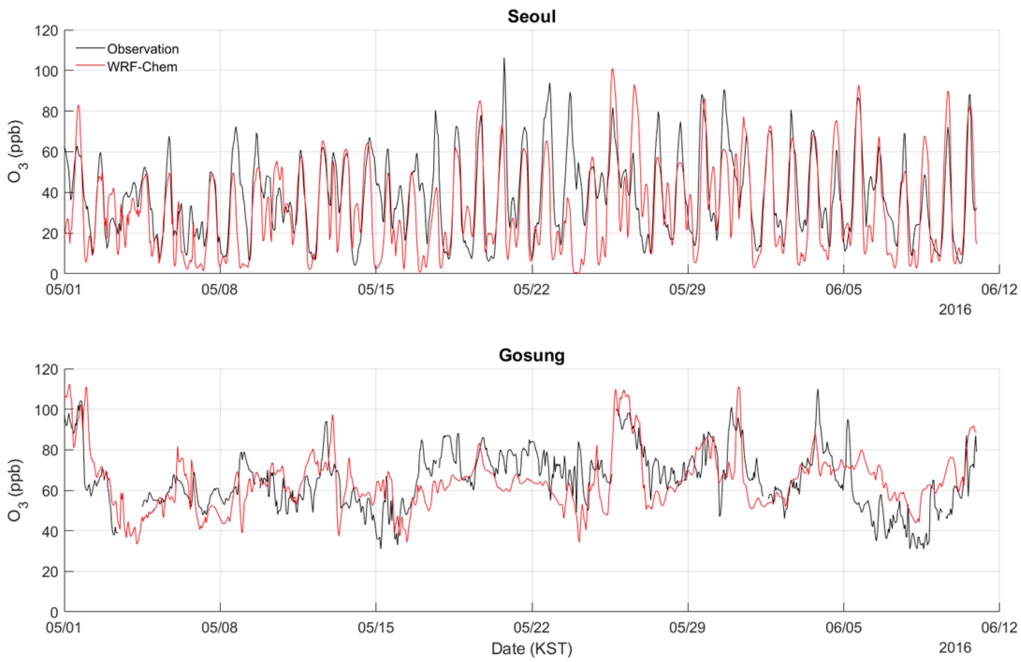

Figure R10. The time series of observed and simulated hourly ozone in (top) Seoul and (bottom) Gosung. Basic statistics are shown as follows. Mean bias (MB): Seoul -6.2 ppb /Gosung -0.9 ppb, Root Mean Square Errors (RMSE): Seoul 18.2 ppb/Gosung 13.7 ppb, Correlation Coefficient(R): Seoul 0.68/Gosung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

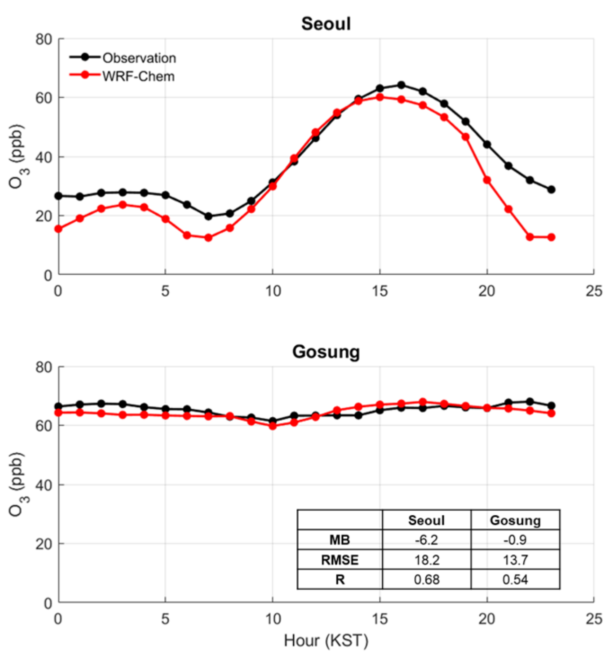

Figure R11. Diurnal variations of observed and simulated ozone concentrations averaged for the entire simulation period: (top) Seoul and (bottom) Gosung. Basic statistics are shown in the plot.

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.