

Review of "Changes in surface ozone in South Korea on diurnal to decadal time scale for the period of 2001-2021" by Si-Wan Kim et al.

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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.

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.
- 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.
- 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.
- 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.
- 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).

- 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.
- 7) A discussion of local CO and NO_x 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 NO_x, since it does have more local influence than CO.
- 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.
- 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.
- 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.
- 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.
 - 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.
 - An explanation should be given as to why there is such large regional differences in overall percentage decline in NO₂. Perhaps this can be related to the model emission inventory?

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

- 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.
- 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.
- 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 ..."
- 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."
- 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.
- 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).
- 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.
- 11) It seems that the information included in Table 2 and Figure 6 are identical; I suggest that Table 2 be eliminated.
- 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.
- 13) Figure 11 – x-axis labels have typo.
- 14) Page 20 – Please define SMA

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.