Second 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:
The authors have made marked improvements in the paper. However, the major revisions that I identified in my first review as necessary before this paper is can be published have not been adequately addressed. Until they are addressed, I cannot support publication of this paper. Those major revisions are addressed in further detail below. For the most part the minor issues identified have been addressed, but a few remaining are also listed below.

Major issues:

1) In my judgement, the authors must begin their observation-based analysis with a consideration of the ozone distribution that would be present in South Korea if there were no local continental influences, i.e., if observed concentrations were due to transported baseline ozone alone. In my first review I suggested how this might be approached. This consideration would then provide a basis for understanding the continental influences, both from local South Korean emissions and from the Asian mainland emissions.

   However, the authors have not attempted this approach. Instead they argue that "It would not be straighforward to delineate background O3 (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." This argument is not adequate. In fact the background ozone is quite readily approximated to the degree of accuracy required. As I noted in my first review, 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 6 of that paper shows that there is a small seasonal cycle (~ ± 5 ppb) in the background O3 outside of the marine boundary layer. Thus, the surface concentration that would be expected in South Korea in the absence of continental emissions is ~35-45 ppb at the spring-summer seasonal maximum; this expectation is in close accord with the peak time ozone concentrations at city and province sites throughout South Korea. To my mind, this discussion must be the starting point for the discussion of O3 concentrations throughout South Korea.

   The authors also respond: "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." However, Figure R12 shows only relatively small differences in mean annual ozone at the diurnal peak times, even in Seoul, the largest city in South Korea. The Control simulation gives a maximum of ~60 ppb. No Seoul emission simulation gives ~ 70 ppb and No China emission simulation gives ~47 ppb. This clearly emphasizes my point that the ~35-45 ppb expected for background only is an excellent starting point for the discussion of the local and regional South Korean influences.

2) In my first review I objected to the author's attempt to use the lower ozone concentrations in the 01-06 period to characterize transport of background ozone, because loss of ozone beneath the nocturnal inversion, both due to reaction with fresh local NOx emissions, but

also due to surface deposition, especially to vegetated surfaces. Thus, nighttime ozone concentrations do not provide direct information regarding transported baseline ozone.

However, if the authors insist upon inclusion of analysis of the data in the 01-06 period, it is essential to base that analysis of Ox = O3 + NO2 concentrations. The figure below is derived from the Seoul data that the authors included in their Figure R9. The Ox concentrations are not affected by the reaction of O3 with fresh local NOx emissions (but is affected by loss to surface deposition), providing further reactions to form NO3, N2O5 and nitrate are not important. Ox recorded during the 01-06 period is a much more accurate indicator of transported baseline ozone than is O3 itself.

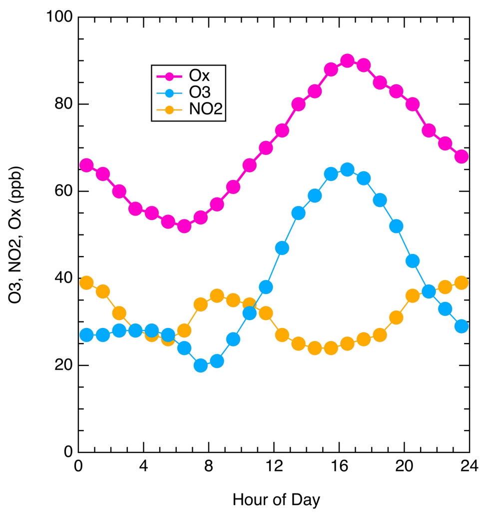

Figure 1. The diurnal variations of observed O_3, NO_2 and Ox averaged for the simulation period, based on reading data from the authors' Figure R9.

3) In my first review I suggested that the background sites be included in Figure 3 and Table 1 in order to emphasize the similarity of the ozone concentrations throughout the country, and the predominant role played by transported baseline ozone. The authors have not made that inclusion; they argue that missing data require that exclusion. However, the data are missing only for periods of only 1 to 4 months out of 21 years. Such minor periods of missing data do not significantly compromise trend analyses. The great value of the background sites for comparison with other south Korean sites is shown in the authors' Figure R11 which clearly demonstrates that peak, mid-day mean ozone concentrations are very similar (in both the observations and model simulation) at the largest South Korean urban area (Seoul) and one of the background sites (Gosung). In my view, it is imperative that all tables, figures and discussion clearly address the 7 cities, 9 provinces, and 2 background sites in a consistent manner to the fullest extent possible. I do understand that measurements of precursor species may not be available from background sites, and thus cannot be included. However, on lines 12-15 of page 30 in the Conclusions Section the authors state: "The 4th highest maximum daily 8-hour average (MDA8) ozone concentrations showed an increasing trend in all cities, most provinces, and background sites during this period, with a yearly increase of 1-2 ppb." Certainly the data from the background sites must be shown in the paper to support that conclusion. The increasing trend at background sites should also be included in the similar sentence in the Abstract (lines 8-9, page 2).

4) In my first review I mentioned that 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. The authors disagree, and respond that they "don't think that there are clear mechanisms driving differences in nocturnal inversion between spring and summer." I am not a meteorologist, but is has been my understanding that atmospheric stability is at a minimum in spring and significantly greater in summer – hence tighter inversion layers in summer. I believe that this may be clearly shown in the authors' Table R5, which shows that mean wind velocity is generally higher in spring than in summer

at all of the stations considered. The nocturnal inversion is much more sharply defined in calm than in windy conditions. Regardless of my meager meteorological expertise, if the authors really wish to compare nighttime ozone concentrations between spring and summer, it is their responsibility to demonstrate that the nocturnal boundary layer dynamics do not change between seasons to a degree large enough to affect that comparison – they have not provide that demonstration. Again, given the very local processes that determine the 01-06 LT ozone, including the unaccounted for effects of surface deposition, and for the reasons discussed in my point 2) above, I recommend that the authors eliminate the discussion of this nighttime ozone; this discussion does not seem to be central to the main points of the paper. Notably, the analysis discussed in Section 3.2.1 is not mentioned either in the Abstract or in Section 4 Conclusions.

5) The Conclusions section requires some modifications. Specifically:
   - On page 31, lines 4-7 has the sentence: "The majority of ozone exceedances occurred between 16-19 LT (4-7 PM). Interestingly, exceedances also occurred frequently at night in background sites such as Gosung and Ulleung Island, indicating a strong influence of long-range transport on surface ozone levels in these locations." I suggest that the final phrase "in these locations." be changed to "over South Korea". The only reason that nighttime exceedances are not seen at most sites in South Korea is that loss of ozone to fresh NOx emissions at night reduce the ozone concentrations below the concentration of transported background ozone.
   - Page 31, lines 13-14 has the sentence: "Therefore, it is not clear what drove increase of ozone exceedances over South Korea from P1 to P2." I disagree; I believe that it is abundantly clear that the increases in ozone exceedances in South Korea can be attributed to increased anthropogenic emissions in China. This certainly follows from the following sentence which states: "We observed significant reductions in ozone exceedances across all monitoring sites in South Korea during the spring of the COVID-19 pandemic (period P3, 2020-2021), which was attributed to decreased anthropogenic activities and subsequent lower emissions in both China and South Korea." This discussion should be clarified.

Minor issues:

1) Table 1: I presume that the tabulated ozone concentrations are means over all days in each season. This should be stated in the Table caption.

2) The description of the two models indicates that different anthropogenic emission inventories are used in the two models. The authors have now given a brief discussion regarding how well these inventories compare (their Table R9), but they include that discussion only in their response to the reviewers' comments. This is very important discussion; it should also be included in the paper itself, possible in the Supplementary Material.

3) Pg. 14, line 1 contains the term "Asian emissions". I think more specificity is required here. Perhaps "Chinese emissions" or East Asian emissions".