Review of "Causes of growing middle-upper tropospheric ozone over the North West Pacific region" by Xiaodan Ma et al.

Overall Assessment

This study leverages ozonesonde records over a relatively long (>20 year) period for four locations bordering the North-West Pacific Ocean, in comparing a state-of-the-art chemistry climate model (EMAC) in simulating ozone across different latitudes spanning the near-tropics to mid-latitudes, as well as from the surface to the upper-troposphere-lower stratosphere region. The authors confirm good overall agreement in terms of EMAC's ability to capture the seasonality in tropospheric ozone, as well as in many cases relatively large, robust trends/changes that have occurred at each location from the 1990s to 2010s, according to the ozonesonde records. They do this by separating out upper, middle and lower troposphere, abbreviated UT, MT and LT respectively, and computing monthly/seasonal means. By exploiting the stratosphere-tagged ozone tracer (O_3S) in the EMAC model, the authors then proceed to quantify the attribution from the stratosphere versus that formed in the troposphere according to the model climatology. A detailed assessment of the stratospheric contribution to recent trends/changes is then performed. Calculation of the residual amount of ozone formed in the troposphere (O_3T) helps to then attribute the role of tropospheric production.

The work is of clear importance as ozone is a non-well mixed greenhouse gas, with a typical lifetime of ~3 weeks in the free troposphere, and is sensitive to multiple factors, including in situ photochemical production, long-range transport and large-scale dynamics. Uncertainty in what drives/influences tropospheric ozone is therefore large and there is an obvious need to understand this better as there is an obvious radiative forcing and air quality implication to the results, as recognised by the wider literature. Dynamical influence mainly entails changes in the downward transport of ozone from the stratosphere, which is strongly determined by the strength of the Brewer-Dobson Circulation (the residual meridional stratospheric overturning circulation forced by wave-mean flow interactions). Facilitation of such downward transport arises from the action of tropopause folding, which preferentially occurs in association with the subtropical/eddy-driven jets and transient synoptic-scale systems (e.g., cut-off lows). Filamentation of ozone-rich air near the tropopause can subsequently be entrained into the troposphere (so-called 'stratospheric intrusions').

This regional-focussed study is therefore highly valuable and is complementary to many earlier studies which have a more general focus (e.g., global). The study is logically presented and well executed, fitting well within the scope of Atmospheric, Chemistry and Physics journal. I would recommend publication of the study, following a minor round of revisions as per my general and minor comments which I detail below:

General Comments

L56: "…Brewer-Dobson circulation promotes stratospheric intrusions…" → I wouldn't say this is true. An enhanced BDC may facilitate an enrichment of ozone into the extratropical lowermost stratosphere that is readily available to be transported down into the troposphere. The development of stratospheric intrusions is instead governed by processes operating on much finer spatial scales, typically synoptic-scale or less, such as associated with the rear-flank of extratropical cyclones/cut-off lows. The authors detail this later (L63-65).

The authors I think need to change the wording to reflect that an increase in BDC strength can promote greater seasonal build-up of ozone into the extratropical lowermost stratosphere over winter (Ray et al., 1999; Konopka et al., 2015; Ploeger & Birner, 2016). Subsequent stratospheric intrusions can them facilitate greater stratosphere-troposphere exchange of ozone as a result of this enrichment, particularly around March time when the lowermost stratospheric reservoir of ozone reaches an annual maximum and is seasonally “flushed thereafter” (Hegglin and Shepherd, 2007; Bönisch et al., 2009). This however does depend on changes to each of the deep and shallow branches of the BDC; a strengthening of the deep branch indeed serves to increase lowermost stratospheric ozone. Strengthening of the shallow branch on the other hand favours enhanced transport/mixing of low-ozone air from the tropical upper troposphere (e.g., Fig. 4 in Bönisch et al., 2009).

Given the BDC is pertinent to this study, I suggest the authors also introduce this key distinction between the shallow and deep branches and their significance in regard to stratosphere-troposphere exchange of ozone.

L208-209: “The tropopause folding on the south part of the STJ could lead to more stratospheric intrusion contributions to the ozone tongue.” → I think it would help if the authors provided an overlay in Figure 2 of the standard deviation or similar (to highlight interannual variability). Not only because this could provide more useful information, but it would be expected I think that higher standard deviation would support this speculative point. I wonder if the authors could also go a step further and attempt to diagnose the STJ from one year to the next, which would bolster this assertion?

Sect 3.2.1 (L337-346): Although EMAC generally overestimates ozone, there is clearly a tendency towards higher overestimation for lower ozone mixing ratios and lower overestimation at higher ozone mixing ratios (which changes in sign for most distributions, such that EMAC typically underestimates ozone at the highest ozone mixing ratios). The authors neglect to mention this so should consider adding description/quantification of this.

I also think the authors need to be a little bit careful with the interpretation for instance that EMAC better represents the upper and lower troposphere versus the middle troposphere for Hong Kong/Naha. The RMSE and MAE imply otherwise at face value, though this is because the differences are less systematic and thus compensated for. The total error summed for all soundings versus model values would maybe be larger for the MT but hard to tell as the axes’ ranges are smaller for MT and LT compared with UT. Perhaps the authors would like to compute this, add and compare, which would help support their claim?

Minor Comments

L46: "Tropospheric O_3 increases of 7% (measured as a partial column between 3-9 km)…" → The authors may wish to specify this increase is of "Free tropospheric O_3", thereby excluding potentially different trends in the lower troposphere/ABL.

L51-52: "…, the counterpart in the middle-upper troposphere…" → "…, the evolution in the middle-upper troposphere…"?

L58: "a stratospheric chemistry-climate model" → Is this correct or should this be a chemistry-climate model which has a well-resolved stratosphere?

L118-120: "The research from the cross-evaluation of OMI data and the ozonesonde observation in Japan sites shows that CI ozonesonde measurements are negatively biased relative to ECC measurements by 2–4 DU compared with the OMI data (Bak et al., 2019)". → Unclear if the bias of 2-4 DU is for total column, or partial column only in troposphere.

L199: "Figure 2 depicts the climatologically vertically resolved tropospheric O3 distribution with respect to months" → Suggested sentence revision: "Figure 2 depicts the monthly climatological vertically resolved tropospheric O_3 distribution throughout the year".

Figure 2: Yellow colour shading missing for Tsukuba. This needs checking.

Figure S1: Same as issue as above for panels c1 (Tsukuba) and d1-d2 (Sapporo).

L208: "Figure S2" → Change to Figure S1 and reverse order of existing Figures S1 and S2 so that they are in order.

Figure 3 Caption: I think it would be helpful to remind the reader of what is used to represent upper, middle and lower troposphere. I'm not entirely convinced that each should be best represented by a single pressure level though, particularly for the lower troposphere where ABL and free tropospheric ozone amounts and variability (including its trends) can differ substantially. Also, are the time periods the same as in Figure 2?

Figure 4c1 and 4d1-2: Same issue as before with contour shading, certain colours missing.

Figure 4 Caption: "Dashed lines in the i-l represent…" → Typo? I assume this refers to the c panels for each site?

L280: "Figure S1" → See L208 comment.

L287: "20 to 40 ppb." → Difficult to tell as colour scale saturates above 20 ppb? Should this be extended?

L288: "…Hong Kong shows more significant O3 changes in the lower troposphere" → This seems true in the ABL (~<1.6 km) and above what is likely 850 hPa (which I think the authors still use to represent the lower troposphere) for April and August (~2-4 km). However, there is a region in between which is statistically insignificant throughout the calendar year. I don't feel the text gives full justice to the nature of the results shown in Figure 4a3.

L333: "3.2 Quantification of stratospheric intrusion versus tropospheric production using EMAC simulations" → It seems only later subsection 3.2.3 directly relates to this heading (the later sub-heading is essentially worded the same).

May I suggest to the authors re-writing the section 3.2 header to encompass the model-measurement evaluation of tropospheric ozone first in 3.2.1, something along the lines of: "3.2 Comparison with observations and stratospheric versus tropospheric attribution using EMAC simulations"

L344: "Figure 7c2" → I think this refers to Figure 6c2.

L345: "Figure 7 b1" → I think this refers to Figure 6b1.

L356: "Figure .7" → Figure 7

L375-376: "Bold indicates the agreement with the observations for significance and the sign of the trend" → Not all bold entries have asterisks next to both the model and observed trend. Is there a mistake or did I misinterpret the description?

L388: "The EMAC simulations of O3 at different portions of the troposphere…" → Revised sentence suggestion: "The EMAC simulations of O3 for different altitude ranges in the troposphere…"

L389: "Dots in the i-l represent…" → Again, not clear what i-l stands for. Is it a typo?

Figure 8d1-d2 → Check colour scale. Seems that yellow filled contour is missing in Figure 8d1 and possibly cyan filled contour missing in both.

L406-407: "Furthermore, together with dynamical processes such as tropopause folding in the vicinity of the subtropical jet (Baray et al., 2000), stratospheric O_3 is transported downward into the troposphere." → The authors should also add mention of the seasonal lifting of the which will naturally contribute to entrainment of ozone-rich air from the stratosphere into the troposphere (e.g., Monks, 2000).

Figure 9d2 → Yellow colour missing.

L434: "3.2.3 Quantification of stratospheric intrusion vs. tropospheric production using EMAC" → This is almost word for word identical to the 3.2 heading on L333. It is more fitting here, but I would simplify by heading it as follows: "3.2.3 Attribution of EMAC tropospheric O3 changes: O_3S vs. O_3T"

L442-443: "The contribution of O3S to observed O3 increases by up to 96% and 40% in the middle-upper troposphere during winter and summer." → I'm a little unclear where these numbers are from. Are they directly shown in Table 3? 96 % is shown for Sapporo in JJA and 40 % for Tsukuba in

JJA (UT), could that be what the authors refer to? It needs to be clearer exactly what season, site and region of troposphere this is (the authors state middle-upper troposphere so reading this implies the quantified contribution is a conflation of the two regions).

Table 3: It seems to me there is a missing set of numbers and that is the total change in O_3 between the 2010s and 1990s for the EMAC model (noting that similar such values in Table 2 are linear trend changes and not the difference between the later and earlier period). These numbers would be good to have so the reader can fully appreciate attribution from the stratosphere versus troposphere without having to do the math.

There appears to be a few inconsistencies in the Table. Presumably the absolute and percentage change values should be equivalent in sign for each season, location and each of UT, MT and LT, which is not always the case. This need checking.

L469: "110°N to150°N" → "110°E to 150°E"

Figure 11: I wonder if the authors could add an equivalent Figure but expressed as a percentage (this could go in the supplementary information at the end). This would be extremely handy to compare and should be minimal effort for the authors to produce and include.

L488-490: "On the other hand, ozone originating from the stratosphere dominates the large portion of middle-upper tropospheric O3 enhancement by up to 96% and 40% in the mid-latitude during winter and spring. → Again, these numbers are very specific and I'm not sure if they are plucked directly from Table 3 or not. If the latter, it is surely better to give a range that encompasses both MT and UT, including both Tsukuba and Sapporo as I think the authors are using to represent mid-latitudes.

L518: "With increasing greenhouse gasses…" → "With increasing greenhouse gases…"

L521: "…facilitating downward transport to the troposphere u under the influence of the Pacific jet…" → "…facilitating downward transport to the troposphere under the influence of the Pacific jet…"

References

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