Note: Author responses are in blue, italicized text.

This is a well written paper with reasonable conclusions. In particular, the SOM analysis seems to do a good job of selecting more strongly convectively influenced versus subsidence profiles. And the discussion around the importance of diagnosing the seasonal variation in the ozone trend is very useful to say. My main comment is that the paper seems longer than necessary due to using so many convective proxies, rather than simply just using rain itself (more in comment below). The technical standard of the paper was quite high, and I don't have any significant comments to make on these aspects of the paper.

We thank the Reviewer for the thoughtful comments. They inspired us to generate more analyses in the form of Figures for Reviewers that bolster our results indicating the tropospheric ozone/convection trends relationship even further. As the Reviewer noted that they thought the paper was a bit long anyway, we decided not to include these Reviewer Figures in the manuscript.

The SHADOZ sondes also contain RH, and it would have been interesting to do a trend analysis of that quantity also, since it is also linked to convection. Could be issues with the data of course, especially in the upper troposphere.

This is a good suggestion. We converted the SHADOZ radiosonde RH measurements into water vapor mixing ratio and calculated monthly 5-15 km MLR trends in the exact same way as with ozone. This is shown in Figure R1. There has been a significant moistening of the lower troposphere in most months after April, but the only significant decrease in water vapor mixing ratio is in fact found in March. This corresponds with the PWAT analysis (Figure 10) contained in the manuscript. We agree that the radiosonde RH (and the derived water vapor mixing ratio) above ~12 or 13 km should always be taken with a grain of salt.

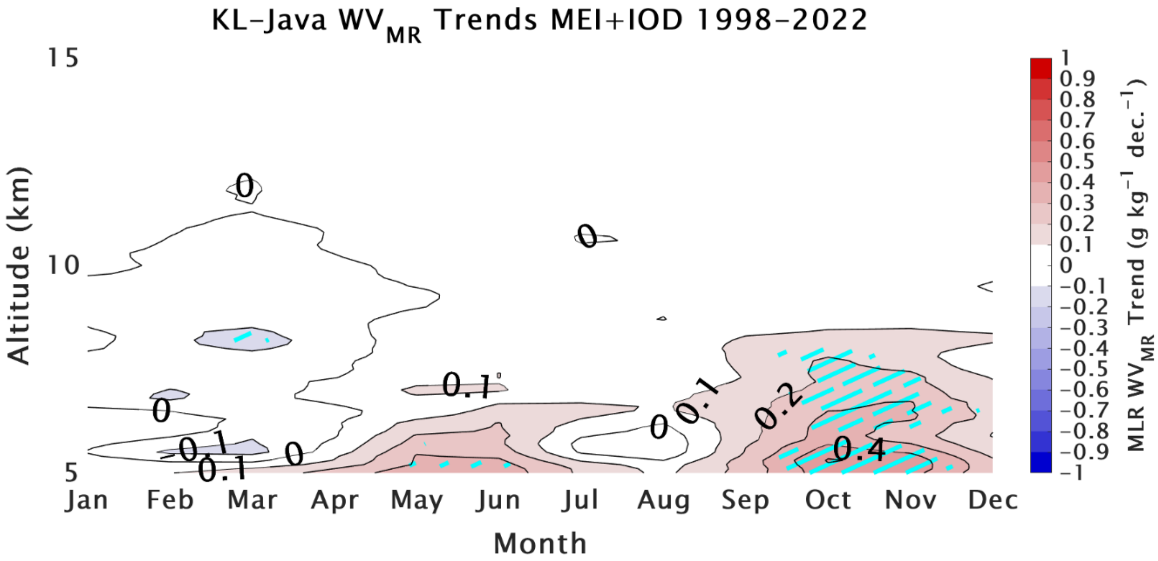


Figure R1. 1998-2022 Monthly MLR trends of water vapor mixing ratio (in grams per kilogram per decade) derived from SHADOZ radiosonde (coupled to the

ozonesondes) profiles. As in the manuscript, cyan hatching denotes trends that exceed the 95% confidence interval.

The authors might have considered rain itself as a more direct proxy for convection than water column, cloud brightness temperature, OLR, velocity potential, etc. There may have been a bit of "over analysis" here in using all these proxies instead of going straight to rain. I think the paper would be more useful if a direct connection between ozone and rain could be established, since rainfall is more directly relevant. Presumably, some version of the TRMM dataset (3 hourly) would be appropriate. I realize rain is a more "noisy" variable, but it is also more interesting.

In the paper we analyzed multiple observational and assimilated datasets to show that several independent physical phenomena arrived at the same conclusion of waning convection in ~Feb-Apr from 1998-2022, leading to the observed positive free-tropospheric (5-15km) ozone trends. Because convective precipitation is a mesoscale feature often influenced by land and topography, while VP200/MJO, ENSO etc., act on synoptic or even larger scales, it's better to look at those regional metrics rather than more localized (rainfall) conditions to diagnose FT ozone variability and trends. Nonetheless, here we show an analysis similar to Figures in the manuscript for Global Precipitation Climatology Project (GPCP; https://psl.noaa.gov/data/gridded/data.gpcp.html; Figure R2) and MERRA-2 convective precipitation (Figure R3). As with the GridSat-B1 product, GPCP is Climate Data Record (CDR) quality. Indeed, the GPCP data are a bit noisier than the proxies that we analyzed, but both datasets also arrive at the same conclusion. Note that in both Figure R2 and R3 that the precipitation trend spatial patterns, especially in March, correspond to those in Figures 7-10 in the manuscript. We have chosen to retain our analyses and Figures in the paper, but hope you find these Figures for Reviewers insightful.

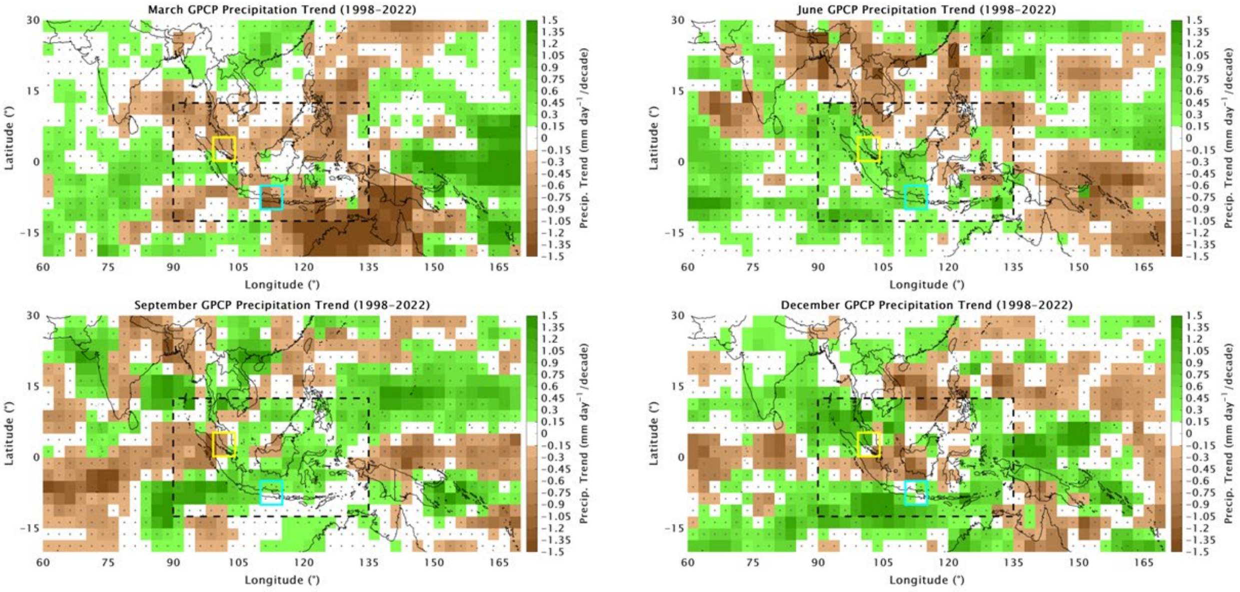
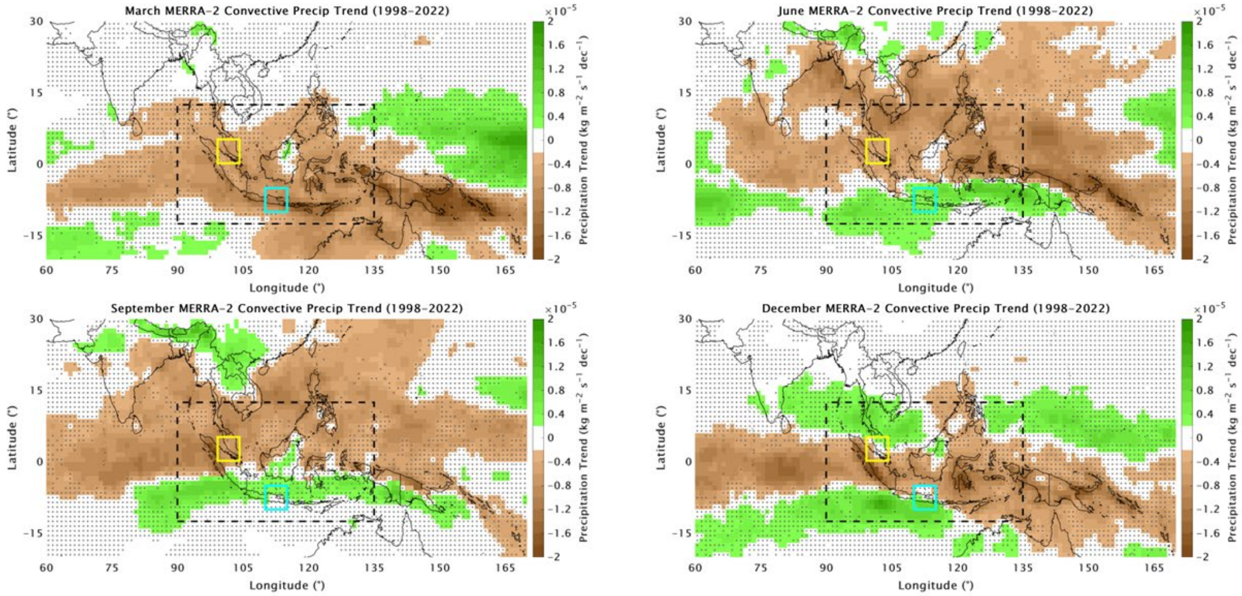


Figure R2. 1998-2022 MLR GPCP (2.5°x2.5°) total precipitation trends in mm per day per decade for March, June, September, and December. As in the manuscript, stippling shows trends <u>within</u> the 95% confidence interval bounds (i.e., "insignificant").



March MERRA-2 Convective Precip Trend (1998-2022)

June MERRA-2 Convective Precip Trend (1998-2022)

September MERRA-2 Convective Precip Trend (1998-2022)

December MERRA-2 Convective Precip Trend (1998-2022)

Figure R3. 1998-2022 MLR MERRA-2 (1.0°x1.0°) convective precipitation trends in kg per square meter per second per decade for March, June, September, and December. As in the manuscript, stippling shows trends <u>within</u> the 95% confidence interval bounds (i.e., "insignificant").

It might be borderline appropriate to call the ozone trend using 22 years of data a trend instead of some form of longer term variance. In this context, it also might be worth mentioning, or even showing, the absolute ozone trends (with MJO, ENSO variability included), since there could be changes in the fraction of the total rain/ozone variance that are part of these oscillations, so that there may not be an absolute trend in these quantities.

We assume you mean 25 years (1998-2022) of data, not 22. We include an ENSO proxy (in the form of MEIv2) in the MLR trend model. When ENSO is accounted for in the model, and despite the last three years of our analysis ending in a "triple dip" La Nina which lowers tropospheric ozone in this region, we still find strongly positive ~Feb-Apr free-tropospheric (5-15 km) ozone trends. You bring up a good point about whether these trends will continue in the future. Clearly there has been a change in MJO (as given by the VP200 proxy) and thus FT ozone in the early part of the year in this region over the last 25 years. Whether the VP200/OLR/T_B trends calculated here are the result of changes or shifts in the spatial patterns, seasonality, strength, etc. of MJO, ENSO, and their interactions with each other is beyond the scope of our study, but they are interesting questions nonetheless and should be examined in future efforts.

We show here (Figure R4) the relationships between MEIv2 and ozone, and VP200 anomalies (computed for the black dash boxed region on previous Figures) and ozone, further indicating that any trend or change in VP200 anomalies in particular will result in tropospheric ozone changes. The relationship between these quantities and ozone is stronger for the 5-15 km layer (top of Figure R4) than the surface-5 km layer (bottom of Figure R4). The fact that there is still a weak relationship between surface-5 km ozone and VP200 anomalies may result in the correspondence between near-surface and FT ozone trends in Feb-Apr (Figure 6, and as you note below).

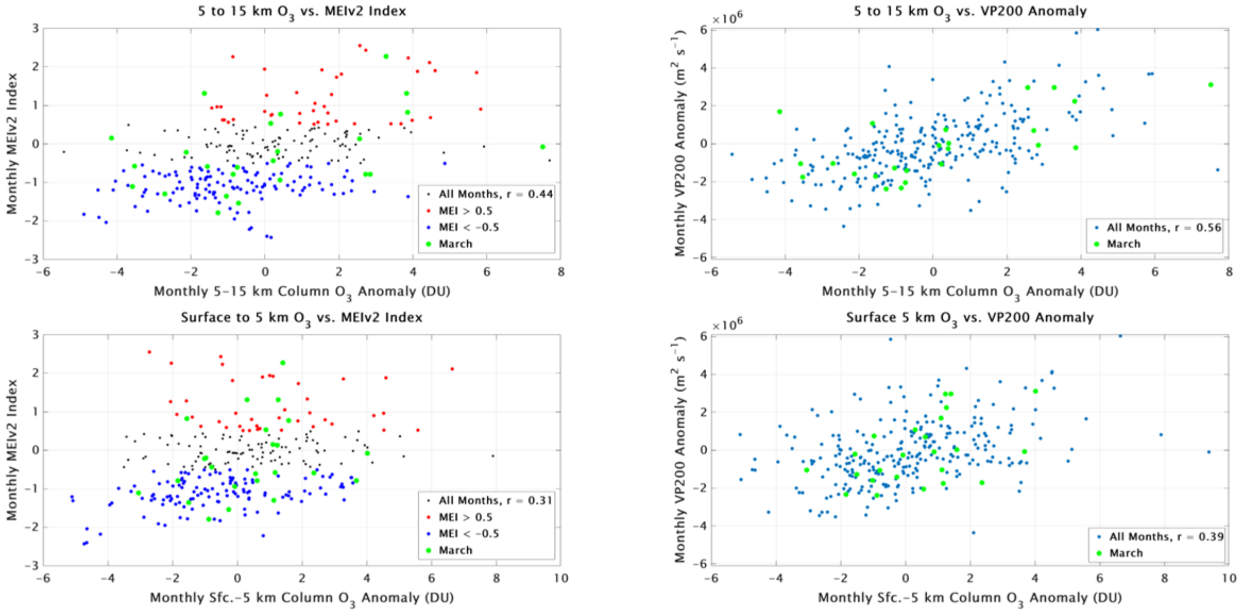


Figure R4. Scatterplots of 5 to 15 km (top row) and surface to 5 km (bottom row) SHADOZ monthly partial column ozone anomalies corresponding to MEIv2 (left column) and VP200 anomaly values (right column). VP200 anomalies are computed for the black dash boxed region shown on Figures in the manuscript and above.

The positive ozone trends near the surface in the top panel of Fig. 6 from June onward look like they are confined to the boundary layer. Comparison with the stability could confirm. This would make more physical sense than saying "below 700 hPa". Could simply then say that, for whatever reason, BL increases in ozone are not being communicated to the free troposphere (maybe since BL increases are a local response to emissions increases).

Another good suggestion. We computed 1998-2022 monthly MLR trends (Figure R5) for the vertical gradient of potential temperature from the SHADOZ radiosondes to investigate changes in boundary layer stability and possible links to the ozone trends shown on Figure 6a. Significant negative trends in dθ/dz (less stability) below 2 km are found in ~Mar-Jul. This does not perfectly align with the trends in Figure 6a, but may help somewhat explain why positive ozone trends between about 2 and 5 km are found only in ~Feb-Apr. We also point out the increasing trend in stability at 14-15 km in March in

Figure R5, another indication of decreasing favorability for convection. The opposite is true in ~Jul-Oct, when negative ozone trends are found at these altitudes in Figure 6.

Thank you for the suggestion on wording. We have added this sentence to Section 3.2 the paper: "However, these near-surface trends are not being communicated to the FT for most of the year. This indicates that surface ozone trends may be primarily driven by emissions changes, while FT trends, as we will show, are the result of changes to convective activity. The trends appear to be somewhat independent of each other in the two different segments of the profile."

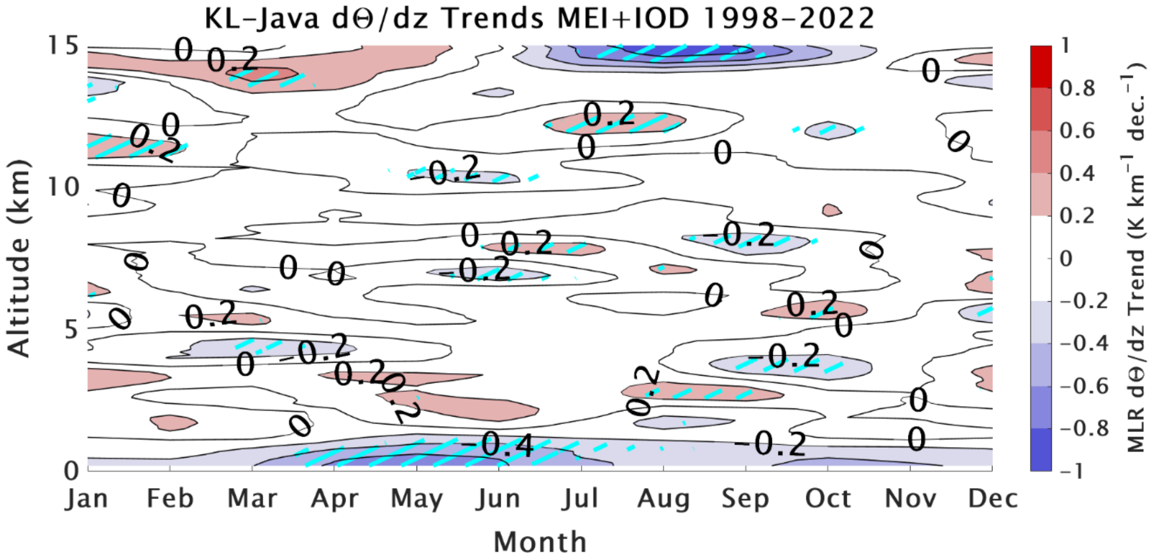


Figure R5. 1998-2022 Monthly MLR trends of the vertical potential temperature gradient (in Kelvin per kilometer per decade) derived from SHADOZ radiosonde (coupled to the ozonesondes) profiles. As in the manuscript, cyan hatching denotes trends that exceed the 95% confidence interval.