At first, we want to thank the reviewer for his helpful suggestions to improve the manuscript.

A general remark by the authors:

Apparently, the objectives of our work and the filtering method of the MOPITT data and its purpose did not become clear enough in our manuscript.
Therefore, we give an explanation at the beginning of this reply, hopefully clarifying misunderstandings.

(1)

We should have stated more clearly, that it was NOT our objective, to find ALL severe pollution events over the Pacific during the last 20 years.

In fact we wanted to identify long-term pollution in the UT on the basis of daily observations. For this purpose we used CO from MOPITT and assumed the highest 2% of the daily global observations to represent pollution.
We decided to chose a filter (among many possible filtering methods using either numerical models, satellite data or other observational data), which ensures that really all events which we select represent episodes of very high CO level in the upper troposphere (i.e. the globally highest 2% mixing ratios).

Therefore, the statistics we present in section 3.1, refers solely to our specific selection of pollution events.
Overall we analyze 17232 individual pollution cluster. We consider this number as sufficient to create a statistics. The number of individual events which we analyze is much larger than in comparable studies from e.g. Luan and Jaeglé (2013), Liang et al., 2005, 2004, 2007.

The objective of our work, was to identify pollution events in the upper most troposphere (other studies do not focus on this altitude range!) using MOPITT CO data and link them to (i) source regions and (ii) specific uplift mechanisms.

→ We rephrased the last paragraph of the introduction (‘objectives’) and the abstract to emphasize the motivation and objectives of our work.

→ We also rephrased section 3.1

(2) *Idea behind the 2% filter:*

The choice of the 2%-filter is originally based on an analysis of the frequency distribution of measured CO mixing ratios (we will add a figure showing this frequency distribution to the manuscript). These have a gaussian like distribution at 400 hPa. Literally spoken, we ‘cut off’ the right tail of the distribution and analyze only this part of the MOPITT data set.

We considered it as rather surprising that many of these 2% grid points are found very regularly over the remote Pacific – also during winter and spring when biomass burning is known to lead to high CO emissions over Central Africa and South America.

High levels of CO over the remote Pacific must be related to long range transport events as the locations where we detect these pollution cluster are far away from CO source regions.

(3) *Selection of pollution events/long range transport (LRT) events:*

As mentioned above, many selection criteria for LRT events exist.
The idea behind the approach of Luan and Jaeglé (2013) is very similar to our method: Based on modelled AOD data they create a frequency distribution of AOD values in a certain latitude range.

This distribution is log-normal (similar like the CO mixing ratio frequency distribution). They choose the top 20% days in the frequency distribution as LRT events.

To compose a robust statistics about the CO source regions and CO transport mechanisms (our major objective!), it is necessary to include a large number of pollution events in the analysis which is given by the 2%-filtering method. The average number of pollution events for which we calculated trajectories is: 195 for DJF per year, 330 for MAM per year, 239 for JJA per year, 98 for SON per year.
As the total number of events in our trajectory analysis is rather large (17232 individual pollution cluster) and events are distributed over 18 years, our trajectory statistics is not impacted by the fact that the number of events (selected with the 2% criterion) varies slightly during time or that we miss some events (e.g. due to clouds over pollution cluster).

→ *As we mentioned above, we rephrased section 3.1 to point out that the statistics (especially fig. 4) solely refer to our individual selection of pollution events (which however, are not chosen randomly but follow a mathematical criterion) and have to be considered as additional information to interpret the trajectory data.*

General Comments

The manuscript describes the use of satellite CO data for the 2000-2019 period to investigate trends over the Pacific region at 400 hPa. Trajectory calculations are used to determine the source regions of CO enhancements observed over the same area. MOPITT thermal infrared (TIR) L3 CO data from version 8 are used. This is an interesting topic and the MOPITT dataset is well suited to the goals stated.

Some key points need to be clarified in order to demonstrate if the data selection method is appropriate to reach the goals stated, though. According to the manuscript, "to capture only severe pollution outflow events from Asia" the highest 2 % MOPITT mixing ratio values (or CO-maxima) are selected. Regions with 3 or more neighbouring CO-maxima (a CO-maxima cluster) are then analyzed.

The adoption of the 2 % method seems to be based on "potential undetected slow drifts of the data over the 20 years of available data (Yoon et al., 2013)";

This is a misunderstanding. The potential drift is only a minor criterion for our filter. Please see our explanation on top of the reply regarding the choice of the filtering method.

the Yoon reference discusses version 5 of the MOPITT dataset. For completeness, the manuscript should consider other relevant studies. For example, Deeter et al. (2019) demonstrated that bias drift at 400 hPa (i.e., the pressure level analyzed and discussed in the manuscript) is 0.0 % / year for the MOPITT version 8 TIR dataset, which is the same version of the dataset analyzed in this manuscript. Since the premise of "potential undetected slow drifts" has not been sufficiently demonstrated, the use of the 2 % method needs to be better justified.

→ We included the discussion of Deeter et al, 2019.

Was the number of MOPITT L2 observations that went into each L3 data point taken into consideration? Some L3 data points are based on a few L2 observations, sometimes as few as 2. L3 data points based on a few L2 observations may not be representative.

An analysis of L2 data was beyond the scope of this manuscript. We rely on the V8-MOPITT user guide and the related publication (Deeter et al., 2019) that L3 data which are publicly available (and has been used in other peer reviewed publications) have undergone a thorough quality control.

As we can link the locations above the Pacific having CO mixing ratios belonging to the globally highest 2% with CO source regions by our trajectory analysis (for individual events but also on a statistical basis) it appears to us that the observed signal in the MOPITT data is robust.

It is not clear if the possible effects of missing data (due to clouds, calibration events, etc.) have been taken into consideration when calculating statistics and interpreting results. If they have, then the manuscript should clarify how. Missing data may follow seasonal/annual patterns and could affect the results and interpretations, if not properly accounted for.

1) To account for missing data points, all quantities shown in Fig. 4 are weighted with the total number of valid data points (see caption of Fig. 4 and text p. 10, l.201/202).

2) Periods with data gaps are not considered for calculating the regression line. We forgot to mention this information in text and the label of Fig. 4 and thank the reviewer for this hint.

3) We exchanged Figure 3 showing the total number of CO maxima at a given grid point with a plot showing the total number of CO maxima grid points weighted with the total number of valid MOPITT data points (the same quantity plotted in Fig. 4, left column)

The two lower panels in Fig. 1 seem to indicate that nighttime MOPITT CO values (half of the daily measurements) were not included in this analysis. This point should be clarified and, if true, justified.

The two plots in the lower row of Fig. 1 show by purpose only daytime observations to demonstrate our filtering methods for which each overpath is considered separately.

In general, global CO maps from MOPITT and other instruments show that pollution plumes emitted in China, other Asian regions, Siberia, etc. are transported across the Pacific and often reach North America and beyond. The manuscript mentions fires as the source of the CO; that would be the case for emissions from Siberia during the northern summer months and in some cases for emissions from SE Asia. However, most emissions from China are due to fossil fuel combustion and they occur during all seasons. This should be discussed in the manuscript.

We do not state at any point in the manuscript, that we assign all CO to fire emissions. In Section 3.1, we discuss the (potential!) impact of biomass burning (in other regions on earth which are known to have rather low industrial emissions of CO but seasonally high CO emissions by biomass burning) on our selection of the CO maximum grid points.

Using solely MOPITT data, it is not possible to draw any further conclusions about specific CO sources (such as fires, traffic, and many others). Indeed our analysis shows (Fig. 7b) that the densely populated areas potentially contribute to upper tropospheric CO.

The manuscript does not discuss the seasonal effects on CO lifetime and overall CO background values which would result in more frequent/persistent CO enhancements during the winter months.

The seasonal variation of the CO lifetime is rather short compared to the total tropospheric lifetime of CO. The trajectory analysis reveals, that air masses need less than 10 days to be transported from CO source regions into the upper troposphere over the Pacific. Therefore, transport time is always shorter than the tropospheric CO lifetime.

In addition, it is a strengths of our 2%-method, that lifetime effects or seasonalities would not strongly affect the relative contributions of our daily frequency distribution. The 2% tail of the frequency distribution is affected in the same way by ambient conditions as the remaining 98%.

The descriptions of results and their interpretations are hard to follow.

Following the suggestions of the 2nd reviewer, we rephrased the ‘conclusions’ section.

Specific Comments

An explicit definition of “elevated CO event”, “severe pollution events” should be provided.

Do CO-maxima clusters represent elevated CO events/severe pollution events? Do CO values/statistics support the idea that CO-maxima cluster represent only severe pollution events as intended? CO-maxima clusters may or may not represent actual pollution events, since they are relative. It would be more appropriate to call those “relative daily CO maxima” or similar. Using an absolute CO threshold value would have resulted in the selection of absolute CO-maxima, most likely corresponding to severe pollution events only.

1) Please see our general comment at the beginning of the reply.

2) “Elevated CO” is defined by the choice of the LRT events themselves (i.e. we only include grid points in our analysis with mixing ratios belonging to the globally highest 2% mixing ratios).

3) As CO mixing ratios differ regionally and seasonally strongly it would be rather difficult to chose a threshold mixing ratio to select pollution events. Therefore, we decided to chose a filter which is independent of absolute mixing ratios. As the difference between mean CO mixing ratios including all CO data and CO mixing ratios including the selected maxima is of statistical significance we can conclude, that we do select events of very high levels of CO.

What are “neighbouring CO-maxima”? Do CO-maxima need to be adjacent to each other? Within some fixed distance?
Why is 3 the minimum number of neighbouring CO-maxima to form a cluster?

On a quadratic grid, each grid point has 8 neighbouring grid points. We require, that at least 2 out of these 8 grid points are also selected as a CO-maximum (given then in total at least 3 grid points being neighbours). This is done to ensure that we only include larger regions of elevated CO in our trajectory analysis and not single grid points.

It is unclear if the 2 % method does “capture only severe pollution outflow events from Asia”. Elevated CO at 400 hPa (the only pressure level discussed in the manuscript) could potentially come from other regions, including Europe or even North America.

Therefore, we performed the trajectory analysis to determine the source regions of the observed CO at 400hPa. Due to the long tropospheric lifetime of CO, emissions from other regions than Asia contribute to background CO level. Though, the CO-maxima cluster analyzed by us, are predominantly fed with CO from Asia. In addition to the source region, we can also identify distinct transport patterns.

We explain on p. 5, l. 147-153 why we analyze the 400hPa level.

We have restricted the analysis to 400hPa as we focus by purpose on the upper troposphere. We applied the filtering method also to other pressure level giving us also cluster of very high CO mixing ratios over the Pacific. Though, the 400hPa level is chosen as it is closest to the tropopause but reliably in the troposphere throughout the year. The 300hPa level is presumably (based on low CO mixing ratios) often stratospheric.

Fig. 2. Blue (low) values in the upper half of the DJF map may indicate that the number of data points is very low. If the CO-maxima data points are basically the only data points available, then difference values will be close to 0. Season and geographical location are consistent with clouds resulting in a low number of data points.

According to Fig.3a, we detected the highest number of events during DJF over the northern Pacific. If the number of valid data points is small (but for sure not too small!) over the northern Pacific during winter but almost all valid data points belong to our selection of CO maxima grid points (which would lead to a small difference in CO in Fig. 2, DJF) our conclusion would not change: Based on considering all available data points, episodes of highly elevated CO mixing ratios determine the overall mean over the northern most Pacific during winter.

Fig. 3, MAM panel. Could the region with high number of CO-maxima be a region with few clouds and, thus, more MOPITT observations which could result in more CO-maxima cases? VIIRS true color images show that this may be the case. ISCCP maps of seasonal mean cloud amount (%) support this point. If the statistical analysis did not account for the effect of clouds in the number of observations, then some of the manuscript results and conclusions may be invalid.

→ Please see our general comment at the beginning of the reply.

By analysing the global distribution of the daily 2% highest CO mixing ratios we found these pollution cluster surprisingly often over the NH-Pacific. This gives the motivation of this manuscript: We want to quantify the source regions and transport pathways explaining the finding of high levels of CO in the upper troposphere far away from strong CO sources.
In deed, we cannot detect pollution cluster in cloudy regions by using MOPITT CO data.

Therefore, Fig.3 gives the reader *solely* information about the locations of the CO cluster which are later on investigated in more detail by the trajectory analysis.
The reader can e.g. see that in DJF more trajectories are initiated over the northern NH-Pacific than in MAM. To account for seasonal differences in the location and number of pollution cluster, the

trajectory statistics is given for the total Pacific as well as northern and southern NH-Pacific separately.

Fig. 4. Right panels. The very low value of the JJA 2001 data point coincides with MOPITT not acquiring data between May and August 2001. Similarly suspect points: DJF 2016 (no data acquired some days in the Feb-Mar period), DJF 2009 (no data acquired some days in the Jan-Feb period). Were periods with missing data accounted for? If not, then some results and conclusions may be invalid. Unclear what other results/conclusions could be affected by this issue.

1) Please see our general comment at the beginning of the reply.

2) We decided to include the points in the figure, as the quantities are weighted, thus the values given in Fig. 4 are actually independent of the number of days/events considered. Though, apparently there are extraordinary few CO maxima events on the (rather few) days considered in JJA in 2001.

Fig. 4 (left panels) What is the relationship between ENSO and the data points plotted? It is unclear from the text why ENSO is discussed.

We included the ENSO index in Fig. 4 to get an idea if years with a strong deviation from the mean (or better from the trend shown by the regression line) are correlated with El Nino episodes as these are known to change transport patterns over the Pacific and therefore also pollution export from Asia.
→ We slightly rephrased the corresponding part of the text (p-10).

Fig. 4: Could those trends be caused by CO emissions elsewhere? Do we see less clusters and/or less CO-maxima days through time in the region studied because other regions in the planet are "dominating" that (relative) top 2 %? Consider a hypothetical scenario with increasing summer CO fire emissions over N America during the 2000-2019 period. Under such scenario: 1) the number of daily observations at the top 2 % would, increasingly, be found over/near N America and 2) conversely, the number of daily observations at the top 2 % over the N Pacific region would decrease during the same period.

As we discuss in section 3.1 (p.10, l.205-210) this could indeed be the case.

We composed the same statistics as shown if fig. 4 also for other regions where we detect the 2%-grid points most often (i.e. Central Africa, southern Africa, South America). We included these plots at the end of this reply.

Fig. 8b seems to indicate that a very large proportion of the trajectories (most of the trajectories in some cases; e.g., Russia DJF) initiate over the ocean. Does that mean that the CO source is at the ocean? If so, please explain. What's the relevance/significance of trajectories initiating over land versus over ocean?

Fig. 8b refers to the ***uplift*** region. Not the ***source*** region.

Fig 1, top left panel. The sharp contrast in average CO between land and ocean is suspect.

After rephrasing section 3 we removed Fig. 1 from the manuscript.

The color code of the figure might have been misleading. This figure and the shown quantity is not used for any analysis and we had only included it into the manuscript to give a reader unfamiliar with the global CO distribution a vague orientation.

Figures need to be arranged in the order in which they are mentioned in the text. For example, Fig. 3 is mentioned in the text after Fig. 1 but before Fig. 2.

We mention Fig.3 once before Fig.2 but not to discuss the science shown (which is done after Fig.2 is discussed) but to show generally the region which is included in our analysis – which can best be seen in Fig.3. Therefore we think it is justified to deviate from the standard to mention the figures in the order of their numbering.

- The legends of panels 8.d, 8.e, and 8.f include a "NE-Asia" class in white. Please show its boundaries in panel 8.a.

As we only consider continental source regions, the land/sea boundary marks the boundary of the NE-Asia source region (the same for all other source regions).

- Revise standard deviation representation in Fig. 8, for consistency. For clarity, consider plotting +- 1 standard deviation lines (not just -1 st. dev. lines) in different colors and/or with an horizontal offset to avoid overlaps.

We mention in the figure caption that the standard deviation is only plotted in the negative direction. This was done by purpose as the standard deviation is the same in positive and negative direction and the bar charts already contain a lot of information. We considered the plot as confusing if more lines were included.

→ We revised the manuscript carefully following the suggestions below.

- Fig. 1. Please label panels. Same comment applies to other figures.

- Maps lack latitude and longitude labels (Fig. 1, 2, 3, 5, 7, 8.a).

- Fig. 8 caption: please explain yellow area in panel 8.a, not represented by color or name in any other panel. The explanation is in the text (lines 275-276); please include in caption too, for clarity.

- Fig 8.c: white bars. According to the text (lines 133-134) those represent the "rest" class. Please add label to panel 8.c for clarity. Rename "rest" to "other" or similar.

- Fig. 8.a. caption: For clarity and to avoid language issues, please consider rewording to, for example: "Figure 8. Summary of trajectory analysis results. (a) Source regions: China (green), NE-Asia (white), India (red), SE-Asia (blue), Russia (gray), and ??? (yellow). (b) Source surface type (land, ocean) per region and season. (c) Uplift type (warm conveyor belt, frontal system, other) per region and season. (d) Source region per season. (e) Same for the NE-Pacific region only. (f) Same for the Southern Pacific region only.”

- Expressions such as “surprisingly high” (line 7), “extraordinary [sic] high” (165, 172), “extraordinary [sic] large” (212) should be avoided. Objective, quantitative statements should be used instead.

- line 58: “North America”

- line 61: Please consider rewording to “However, in that particular case study” or similar. There are several other cases where “Though” is used at the beginning of a sentence (line 22, 144, 151, 163, 179, 200, 204, 219, 254, 269, 272, 299, 304). Please reword.

- line 125: Please reword “tends to rather underestimate than overestimate” to “ tends to underestimate rather than overestimate”.

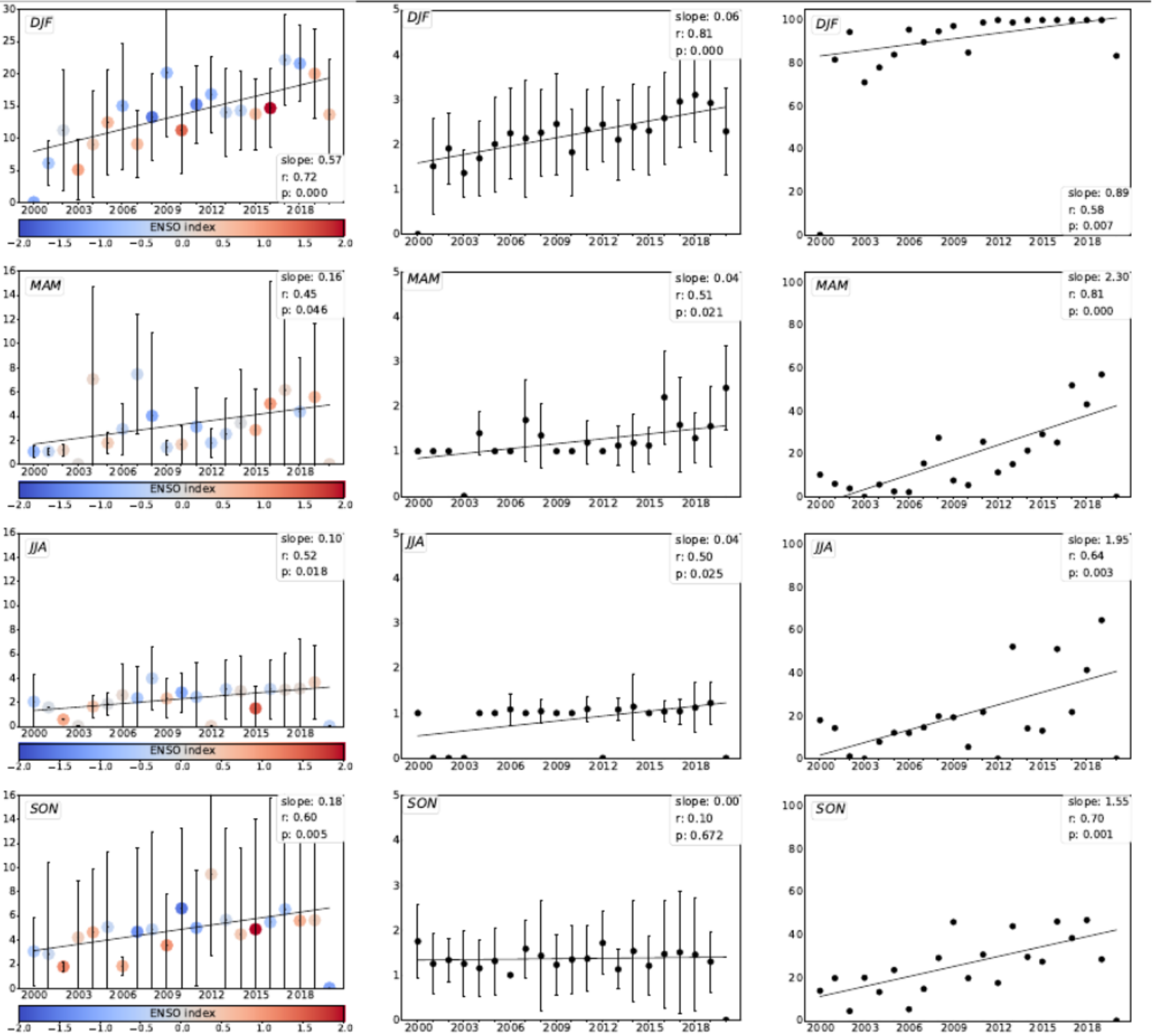
- line 154: “top and centre rows”.
- line 234: “both clusters”.
- line 277: “The two case studies indicate that pollution”.

Central Africa

(A) Daily mean number of: COmax grid points

(B) Daily mean number of: COmax cluster

(C) Season mean number of: COmax days

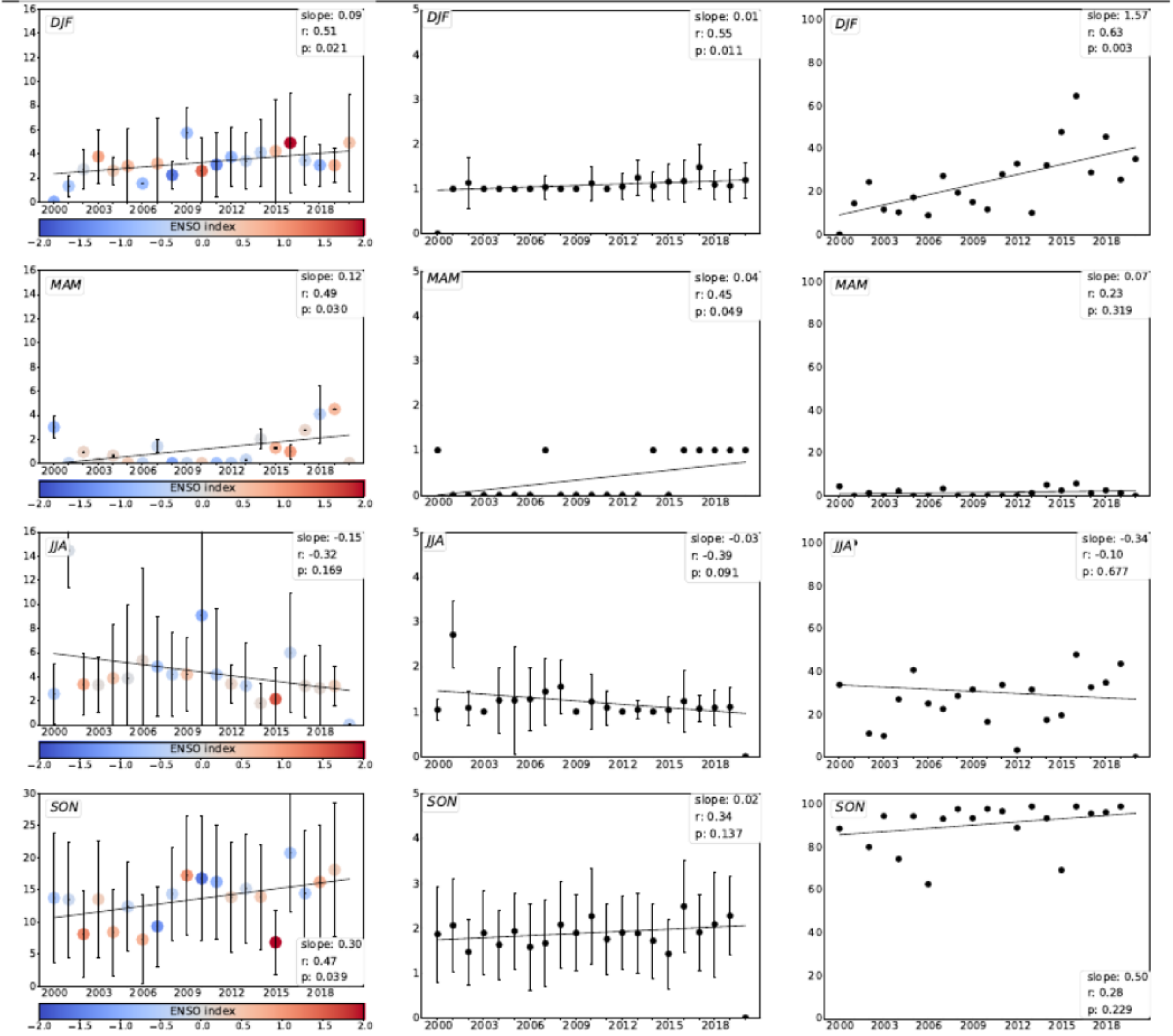


South Africa

(A) Daily mean number of: COmax grid points

(B) Daily mean number of: COmax cluster

(C) Season mean number of: COmax days



Central Africa

(A) Daily mean number of:
COmax grid points

(B) Daily mean number of:
COmax cluster

(C) Season mean number of:
COmax days