At first we want to thank the reviewer for the very helpful comments to improve the manuscript.

<u>A general remark by the authors:</u>

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.

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.
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 (in particular of the related trajectory analysis). 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 major objective of our work, was to identify pollution events using MOPITT data and link them to source regions and 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*

Abstract: the latitude range of the study region should be specified in the abstract.

There are several instances where a sentence is started with "Though…" Without a following clause, these are not complete sentences and are confusing in terms of meaning. Either connect these to the previous sentence or use a different starting word for example:

L22, "Though these studies…" should be "However, these studies…"

L144: "Though it can be expected…" Do you mean "Nonetheless, it can be expected…"?

L179: "Though, this region…" I think you can make this one sentence with: "…events occur, even though this region…"

L39 and throughout – I would capitalize the WCB acronym – wcb looks a bit like web and could be confusing to the reader.

L107 "As a reference for CO emissions…" This is confusing wording – could imply that MPTRAC uses an emissions inventory, which I don't think is the case. Maybe say explicitly: "We use the IPCC AR5/RCP8.5 emission inventory (Lamarque et al., 2010) to determine emission regions above the threshold.

L124 "extension" should be "extent"

L173 "extraordinary high pollution" => "significant high pollution"

L175 sentence starting with "Thus we can assume.." is confusing. Please re-word

L180 "level" can be confused with vertical levels – maybe use "High CO events"

→ *We rephrased these sections of the manuscript!*

L90 By using the grid cells with the highest 2% mixing ratios, you still have a seasonal dependence on the contribution of the a priori to the denominator CO amount due to differences in MOPITT sensitivity over seasons, as well as the global contributions from seasonal SH biomass burning (as you acknowledge). The contributions of these effects should be determined, including the trends in biomass burning (e.g. Andela et al., 2017) and how they affect the selection of pollution outflow events should be quantified.
Reference:

Andela, D.C. Morton, L. Giglio, Y. Chen, G.R. van der Werf, P.S. Kasibhatla, R.S. DeFries, G.J. Collatz,S. Hantson, S. Kloster, D. Bachelet, M. Forrest, G.Lasslop, F. Li, S. Mangeon, J.R. Melton, C. Yue, J.T. Randerson, A human-driven decline in global burned area, Science, 356 (6345) (2017), pp. 1356-1362, 10.1126/science.aal4108

1) Please refer to our general remark at the beginning of our reply.

2) It would be rather difficult to determine quantitatively the effect of biomass burning on our selection of pollution events as we cannot distinguish between CO sources by solely using MOPITT data. We can only assume that most of the CO which is observed over the southern hemisphere and over central Africa is emitted by fires.
Finally, it was not our objective to investigate the impact of biomass burning on observed CO level.

3) 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 dataset.

We considered it as 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.
This gives the motivation for this manuscript: Quantification of the source regions and long-range transport pathways underlying these high levels of CO in the upper troposphere far away from strong CO sources.

It is a strength of our 2%-method, that CO 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%.

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 source region and uplift 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).
The increase of the number of selected grid points over the Pacific during NH-spring is however, striking. Therefore, we discuss hypothetical reasons for this increase (one is a change in biomass

burning). A more detailed analysis of the MOPITT data was however, beyond the scope of this manuscript and as mentioned above, would not change our main results (the trajectory analysis).

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.

At the end of this reply we add a figure showing fig. 4 of the manuscript for Central Africa, South Africa and South America. This gives an indication about the seasonal and regional distribution of the 2% grid points.

L101 "number of trajectories is defined by delLat x delLon x 100" What is the range and typical value for this number?

The size of the domain covering a cluster varies strongly. It ranges from rather small areas having a size of 2°x2° degrees up to areas having a size of 5°x6°. Larger cluster are detected rarely (roughly once per month).

The cluster shown in fig. 1 (lower most row) are therefore of average size. The squares covering the five cluster being discussed as case studies have a size of (6°x3°)[a], (2°x4°)[b], (3°x6°)[c], (5°x5°)[d] and (4°x5°)[e] .

For these five cluster we calculated 1800[a] , 800[b] , 1800[c] , 2500[d] and 2000[e] trajectories at four different start times. Thus the final number of trajectories which we calculated for each cluster is: 7200[a] , 3200[b] , 7200[c] , 10000[d] and 8000[e] .

We performed sensitivity simulations calculating e.g. twice the number of trajectories for each cluster. As the statistics regarding the source region contribution did not change significantly we decided to keep the number of trajectories as small as possible (but still sufficient!) for computational reasons.

L105-106: What is the sensitivity of the results to these threshhold values?

We have performed indeed sensitivity tests regarding these threshold values. The sensitivity of the results was very small regarding the CO emission flux as almost all CO source regions (apart from Siberia) have rather high CO emissions throughout the year.

The results (trajectory statistics) are also not very sensitive regarding the exact criterion for the descend into the boundary layer. The total number of trajectories following this criterion is very large and almost all of these trajectories reach the surface level (in the ERA-INTERIM data set used to drive MPTRAC) quickly after descend below 850 hPa within one source region. As we defined each source region as a rather large area, it happens only rarely that a trajectory crosses a 'source-region border' while being in the boundary layer within 24 hours after descend.

Figure 2: Grey crosses are difficult to distinguish- maybe use these to indicate areas without statistical significance. Also, please state pressure level in the figure caption.

We added "400 hpa" to the figure caption.

L201 "At the same time, the total number of valid MOPITT grid points…" Does this analysis consider the number of MOPITT retrievals per grid cell? This could also be indicative of sampling changes over time due to clouds.

Yes, indeed. We replaced fig. 3 (showing so far the total number of CO-maxima events which we detect) with a figure showing the number of CO-maxima events weighted with the number of MOPITT retrievals for each grid cell to exclude sampling changes.

L263 – Sec. 4.2 How do these results agree or disagree quantitatively with previous results using different approaches? A broader discussion and/or table would be useful.

→ Conclusions: We extended and rephrased this section

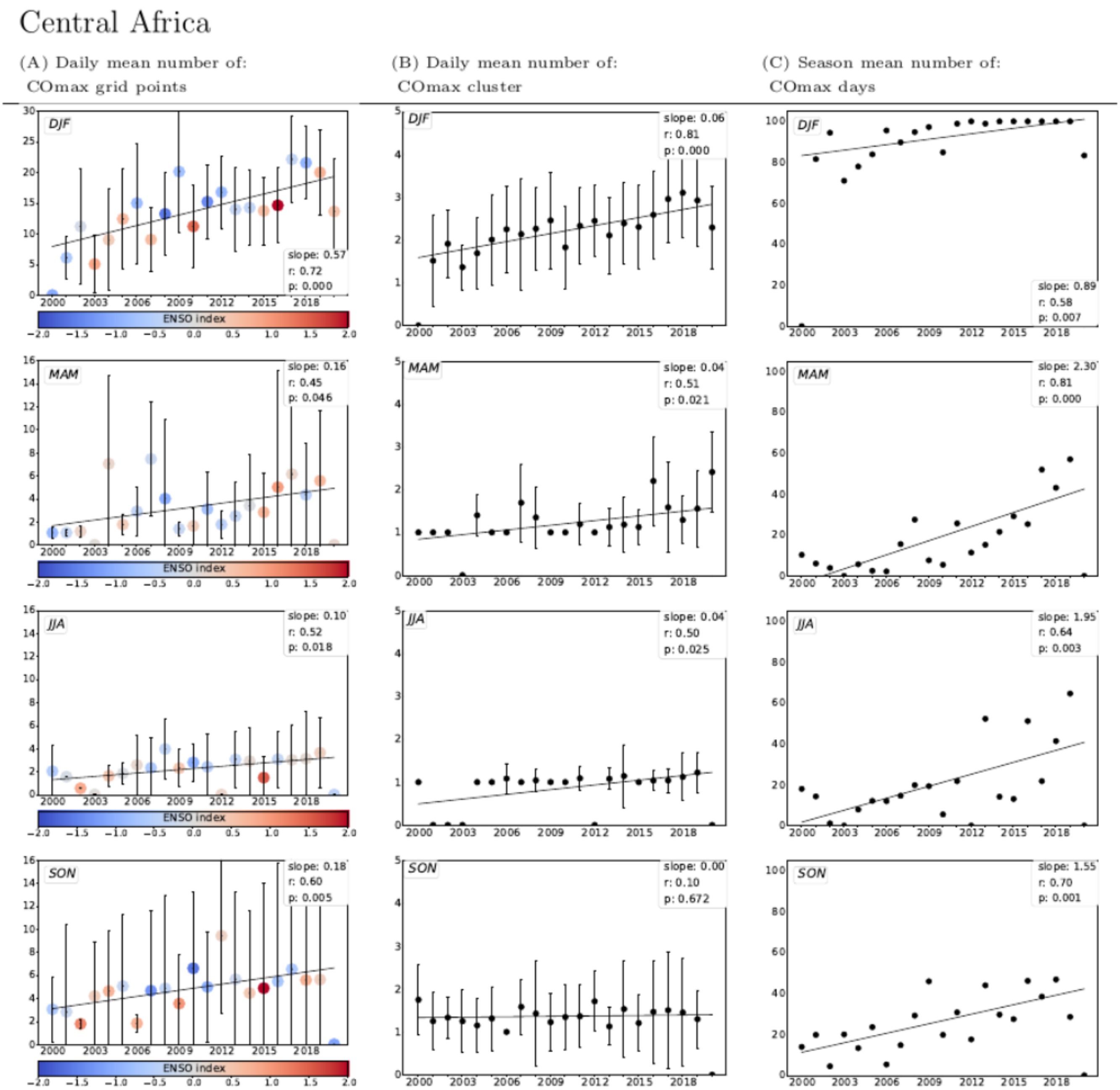
Comparable studies from e.g. Luan and Jaeglé (2013), Liang et al., 2005, 2004, 2007 use numerical models and/or satellite data to filter long range transport (LRT) events across the Pacific. They come to similar conclusions like us regarding the seasonal distribution of LRT events and the processes leading to uplift of pollution into the free troposphere. Some of these studies use however, a composite approach. This is a justified and well documented approach to investigate LRT events but, the analysis refers to the mean properties of the composite. As we analyze each pollution event individually, our results regarding transport pathways and transport times are more accurate. Furthermore, we analyze a much larger time period than comparable studies. This gives us the opportunity to assess the contribution of different CO source regions and their seasonal variability to CO observed over the Pacific.
In addition we are not aware that any other study so far has not pointed out that Asian emissions impact severely and regularly CO level in the upper troposphere (400hpa) far away from CO source regions.

L307-310: I had a hard time following this logic and implications. Does this mean these cases are not included?

All these cases are included in our statistics.

It would be desirable to find a clear correlation between the occurence of high levels of CO in the upper troposphere over the remote Pacific (e.g. over the Aleutian peninsula/north-eastern NH-Pacific during winter) and the prevailing synoptic situation.
This is very generally the case as we find common uplift mechanisms for trajectories having a common source region and a common start region (thus the region where CO maxima are found).
Though, we find strong differences when comparing in detail individual pollution events with each other (regarding: location of CO maxima, location, strength and development of the Aleutian low, transport time from a CO-source region over the continent to the observation location of elevated CO over the Pacific, location of uplift from the boundary layer to the upper troposphere, position and strength of cyclones leading to uplift, residence time in the boundary layer/free troposphere during transport).
Therefore, we conclude that it is very difficult to further group (= create composites of) trajectories with similarities especially regarding a statistical correlation to meteorological parameter.

To better quantify the contribution of Asian emissions to north American pollution level however, it might be important to link the occurence of long range transport events and transport pathways in more detail with the prevailing synoptic situation over the Pacific.

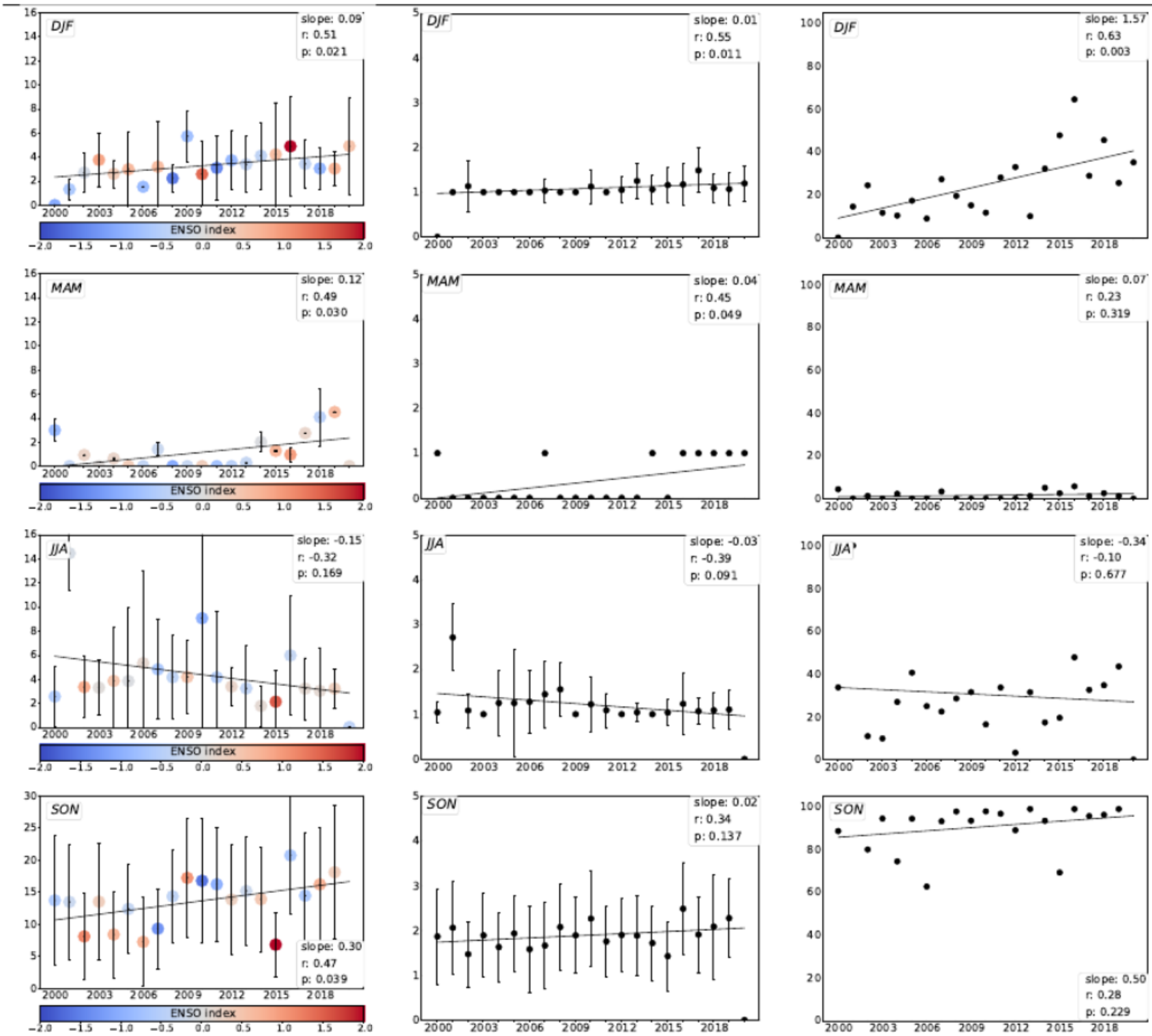


South Africa

(A) Daily mean number of: COmax grid points

(B) Daily mean number of: COmax cluster

(C) Season mean number of: COmax days



South America

(A) Daily mean number of: COmax grid points

(B) Daily mean number of: COmax cluster

(C) Season mean number of: COmax days