General comment

We would again like to thank the reviewers for their critical and constructive comments. They led us to reconsider the use of CALIPSO aerosol subtype information, to include additional AOD (MISR) and emission (CEDS) datasets in the analysis, and to more clearly present the very large changes in aerosol and cloud properties observed in late autumn/early winter, including a more elaborate statistical evaluation.

Following concerns expressed by both reviewers, we decided to omit the usage of CALIPSO level 3 aerosol subtypes from our analysis. As stated in our previous replies, the primary purpose of using CALIPSO data was the inclusion of information on the aerosol vertical distribution, which is still possible based on the CALIPSO data on total aerosol. We understand, however, that it would be highly likely for the reader to misunderstand the usage of aerosol subtypes as an attempt to attribute corresponding sources. As we explain in specific comments below, this omission led to further adjustments in the description of our results. Most importantly, the question of how absorbing the aerosols are, is now answered based mainly on the GFED analysis.

Furthermore, we included in our analysis results based on two-month aggregations. This was triggered by concerns expressed by the reviewers regarding the validity of the monthly changes. We recognized that pairs of months showed similar changes in cloud and aerosol properties and a bimonthly analysis added further robustness to the results.

In the following, the referee comments and our first replies are provided for clarity and completeness purposes, with our final responses highlighted in red. Page and line numbers in our final responses refer to the marked-up version of the manuscript, attached here after our responses.

Final response to Anonymous Referee #1

General comments:

There are many (admitted) observational limitations, especially with respect the interpretation of aerosol type thus I recommend also to look at the fine-mode AOD of the MISR retrieval, which I placed on ftp ftp://ftp-projects.zmaw.de/aerocom/satellite/mis_v23/

We thank the referee for this suggestion and for providing the data. Using fine mode AOD as a better CCN indicator (compared to total AOD) will add robustness to our findings. We intend to include this analysis in a revised version of the manuscript.

Final response: MISR data of total, fine and dust mode AOD were included in the analysis.

It seems very promising to examine cloud property changes in regions where large changes in aerosol loading have occurred. However for that region an opportunity is missed by just exploring periods of a recent AOD decline (especially since 2012), whereas it was contrasted by a strong AOD increase before

2008. Thus opposing cloud property changes should have been observed, if there was an aerosol impact on clouds. Unfortunately even with these AOD changes the aerosol loading was quite high so that CCN concentrations may been already saturated with respect to droplet formation which in part explain the largely 'missing' first indirect effect. Overall, I like the paper but I am sometimes dismayed at the recognition of shortfalls without going into further detail. The data-consistency (e.g. CLARA) also is often a major handicap so that despite of significant data-analysis often there remain relatively little useful information to work.

We acknowledge the value of performing the same analysis for larger periods of time, in view of the known alternating signs of aerosol changes over this region. However, reliable satellite-based aerosol observations are not available before the 2000s. Additionally, to minimize the effects of interannual variability on the detected changes, the latter should be computed for relatively long periods (here we required at least 10 years). These factors render the combined examination of aerosol and cloud changes over larger periods practically unfeasible. The strong AOD decrease before 2008 that the referee mentions is not apparent in our analysis (Fig. 2), probably because the latter includes only two years prior to this switching point. However, we took the opportunity to examine clouds changes over a larger time period, offered by CLARA-A2. As the referee mentions, issues of long-term consistency in this data set reduce the availability of useful information. The remaining information, however, shows that the changes reported during the more recent years are considerably different (and in some cases opposing) than those based on earlier time periods (as shown in Fig. 6), suggesting a connection with the switch in the sign of aerosol changes in recent years.

The referee's remark on possible saturation in CCN concentrations is an important one that we plan to include in the relevant discussion.

Final response: The referee's remark on possible saturation in CCN concentrations was also included (page 9, lines 18-20).

The use of reanalysis data is an interesting aspect, and I just wonder if they the MODIS data assimilation in MACC actual changes in cloud-properties in the examined China regions are simulated. I suggest also not to look just at biofuel but also fossil fuel emission (trends) as alternate aerosol change indicator although working with actual AODf is probably best.

Comparisons of satellite-based aerosol and cloud data with corresponding CAMS data show consistent changes. However, in the case of CAMS this is due to data assimilation, and does not prove a successful model reproduction of aerosol-cloud interactions that occurred in reality.

Working with fine mode AOD is a constructive suggestion that we intend to incorporate.

Final response: Fine mode AOD data were incorporated based on MISR.

Minor comments:

figures

Figure 1 biomass burning emissions are at a maximum in spring but AOD/ AODf reductions (of MISR see below) are largest in fall. I would also add seasonalities of MISR data (AOD, AODfine and AODnonsphere[dust])

From MODIS and CALIPSO we also get largest AOD reductions in fall and early winter (see Fig. 3). We plan to include MISR data in the revised manuscript.

Final response: The suggested seasonalities were included.

Figure 2 MODIS data over land have limited accuracy and MISR data are there usually more accurate … although the reduced AOD in that region is not questioned. The component AOD type assignments of CALIPSO involve many assumptions and should not be overinterpreted.

We intend to include MISR data in the analysis. CALIPSO assumptions and issues are acknowledged, and they will be further emphasized in the framework of a more conservative interpretation.

Final response: Figure 2 was restructured to include MISR data, while CALIPSO aerosol subtypes were omitted.

Figure 3 the AOD reductions by MODIS and CALIPSO are max in October, for which GEFD changes can be hardly used to explain. Also consider that the CALIPSO 'smoke' in fact could be 'pollution'

As we explain in the manuscript, GFED does not provide a full picture of aerosol emissions (page 4, lines 37-38). Hence it should not be expected to explain all changes in AOD found from MODIS or CALIPSO. CALIPSO uncertainties will also be further highlighted, although possible misclassifications of the "smoke" subtype are already mentioned (page 5, lines 29-33).

Final response: Figure 3 was updated and CALIPSO aerosol subtypes were omitted.

Figure 4 cloud fraction is relatively high (ca 80%) so that the number of successful pixels in MODIS retrievals maybe very small and that CALIPSO will miss cases with strong pollution below clouds. Why is the water cloud phase dominant in the coldest seasons (that is not very intuitive). Also I would assume the largest cloud-fraction, COT and LWP during the monsoon season in summer, which the seasonality does not show.

Because of the high cloud fraction, there are indeed relatively few successful MODIS AOD retrievals. Therefore, we required only 10 daily AOD values to be present (compared to 18 for cloud properties) to accept a MODIS monthly-mean AOD for further analysis. On the relatively coarse grid scale of the MODIS products (1 x 1 degree) this is usually fulfilled. Similarly, the even much coarser (2 x 5 degrees) spatial resolution of the CALIPSO level-3 products also yields sufficient retrievals for the monthly means.

The monsoon season in summer is characterized by a larger fraction of high clouds with ice near the top, in particular convective clouds. In winter, low stratus/stratocumulus clouds prevail. In both seasons the cloud fraction is rather high, but one can imagine that persistent stratus will give a larger mean cloud fraction than intermittent convective systems. The seasonal cycle of cloud properties is retrieved very

consistently from two independent satellite records, CLARA-A2 and MODIS, so we think it can be trusted.

Final response: Part of our reply regarding the seasonality of cloud fraction was added in the revision (page 6, lines 36-37).

Figure 5 the increase to LWP and cloud cover change have a strong local signature so I wonder if working with large regional changes is too simple. Even these annual averages display strong variations over time, so that I wonder if the identical 'trends' are significant. Also COT an REF retrievals are only possible for overcast conditions – so in that context tendencies in overcast cloud conditions would be of interest as well.

The LWP and cloud cover changes do show spatial variations, in particular with larger (positive) changes over land, and even slightly negative changes over sea. These patterns are consistent between CLARA-A2 and MODIS. We do not want to 'cherry-pick' exactly the grid boxes where we see the largest changes, and therefore we analyze time series of monthly-mean cloud properties averaged over the full region from both satellites, even if this might give somewhat weaker effects, because some parts with weaker trends are included. COT and REF retrievals are performed for cloudy pixels under the assumption that these are overcast. This will not always be the case, but we do not have sub-pixel information to determine that.

Final response: no additional changes.

Figure 6 AVHRR over the years have different overpass time (trends may reflect in part daily cycles) and different sensors, so CLARA may not have the homogeneity needed for long-term trends. On the other hand the long time series is of advantage as (industrial) fine-mode AOD likely increased until 2008 and likely decreased since 2008 over China. So if there is a cloud property response I would expect a clear bi-polar response, which I do not see.

Orbital drift in NOAA satellites is important especially in the 80s and 90s. For the 10-year period examined in this study, data from NOAA-18 and NOAA-19 were used. Specifically, only the "primary" satellite was used in each month, meaning that when NOAA-19 data became available, NOAA-18 was not used any more. In this way any possible drift would be minor. The very good agreement with MODIS ensures that drift is not an issue for the 10-year period examined. For longer periods, depicted in Fig. 6, the effects of drifts are identified and acknowledged.

Final response: The orbital drift issue is discussed further in Sect. 2.2 (page 3, lines 27-31).

Figure 7 the largest cloud property changes (increases to all examined cloud properties!) are in December – not the month with the largest AOD changes (October). Also the largest AOD and AODf values of MISR in that region are in spring (consistent with your Figure 1)

Not all cloud properties have the largest change in December: liquid CFC has the largest increase in November. Apart from that, we agree that the largest aerosol and cloud changes don't coincide, but we also claim that such a coincidence is not a prerequisite for possible interactions. Aerosol type and height

relative to clouds are also important factors, and constituted the main reason for CALIPSO data to be included in our analysis. We also don't see why the largest changes in AOD should coincide with the largest AOD concentrations. Large changes in autumn and large amounts in spring are not inconsistent.

Final response: no additional changes.

Figure 8 the changes in October are likely pollution rather than smoke and the category of dust mixtures is too imprecise for me to be useful.

The most relevant part of aerosol type in this case is the optical properties and how these might affect interactions with clouds. We agree that attributing sources of aerosols based on their types is uncertain, but it is also of secondary importance in our case. The two main important points are (i) that the aerosols are absorbing and (ii) that they reside at a certain height, which is different in these two months. We will try to clarify this even more carefully in the revised version of the manuscript.

Final response: Figure 8 was updated by omitting CALIPSO aerosol subtypes and aggregating results in September-October and November-December. The relevant discussion was also adjusted, since there is no strong evidence that aerosols in October are indeed absorbing. In November, however, the correlation between decreasing CALIPSO AOD and GFED emissions does strongly suggest that the aerosols are absorbing.

Figure 9 Wouldn't it be better to show the histogram just for Oct and Nov when you focus on Oct and Nov changes. Actually I am bit uncomfortable just to focus on two individual months rather than on an entire season – in the context of meteorological variability

Yes, this suggestion is reasonable and we will incorporate it. The focus on two separate months is due to the different behavior in CALIPSO aerosol subtypes (Fig. 3) and corresponding differences in vertical changes in these months (Fig. 8). Our results suggest that these changes affected clouds, based on the same explanatory mechanism which manifested in two different ways depending on the relative positions of aerosols and clouds. These effects would be indiscernible, had we averaged these months or examined the season as a whole.

Final response: Based on the revised results, the lack of strong evidence on absorbing aerosols in October renders the explanation proposed for this month rather hypothetical. Additionally, Fig. 9c does not provide substantially additional information for the November case. For these reasons, Fig. 9 was omitted.

text

page 2 / 20 there are some issues with the total AOD, since the it is contaminated by larger dust particles, which do not contribute as smaller particles from wildfire and pollution to particle concentrations and potential CCNs. MODIS only offers to total AOD and there are issues with the accuracy over land (for which the data are applied here) and the opportunity to retrieve aerosol maybe be limited by the relatively high low cloud cover. And the aerosol typing of CALIPSO is only quantitative – especially for 'mixtures' and it is extremely difficult to distinguish between pollution and biomass aerosol

types. I think the use of MISR data also should be included in order to include AOD trends, especially since MISR also addressed the AODf.

We acknowledge the issues in using AOD as proxy to CCN. Possible reduced retrievals due to high low cloud cover are treated by applying all the requirement thresholds in the estimation of monthly means, described in Section 2.4. Regarding the quantitative aspects of AOD from CALIPSO, our focus is primarily on their optical properties and how they can affect interactions with clouds. Inferring sources of aerosols based on their types is secondary and we will try to clarify this in the revised version.

Final response: MISR AOD data were included in the analysis and CALIPSO aerosol subtypes were excluded to avoid misinterpretations.

page 2 / 40 the mentioning of the GFEDv4 data is an interesting concept to justify aerosol changes due to biomass burning, but I would argue that industrial emission reductions (talk to Steve Smith to provide his industrial emission data for that region) are much more likely to explain satellite observed AODf reductions in recent years.

Reductions in aerosols from biomass burning activities over this area in recent years is well documented in the literature, which we include in our study, and the GFED analysis confirms these previous findings in terms of both seasonal distribution and changes. We claim that the change in these activities explain only part of the reduction in total AOD, as is also obvious e.g. from Fig. 3 and the relevant discussion (page 5). We agree that reductions in industrial emissions may also play a role in explaining the overall AOD reduction, and it is a constructive idea to include some relevant analysis as the referee suggests. It should be noted, however, and will be emphasized in the revised manuscript version, that the main goal of this study is to examine possible aerosol-cloud-radiation interaction mechanisms manifested due to the AOD decrease, rather than explain in detail every aspect in this decrease over the region. These interactions should not be expected to correlate with the overall aerosol change reported, but depend on specific aerosol types, relative positions and overall conditions.

Final response: A figure showing changes in various emissions from the suggested data set was included in the supplement.

page 3 / 10 the MODIS data for (liquid) cloud cover is the most reliable property (though possibly biased by the overpass time and also dependent on the non-obscurance by higher altitude clouds). Details (LWC, Reff and COT) only refer to an overcast cloud subsample – so I wonder if also changes for overcast low cloud pixels can be addressed as well

It is true that MODIS COT, REFF and consequently LWP are retrieved for a subsample of cloud-detected pixels (e.g. cloud edges are excluded). In that sense, analyzing the fraction of "overcast"-only cloudy pixels is a useful addition.

Final response: Our first reply did not address the question appropriately. The MODIS analysis is already based on overcast pixels, i.e. excluding cloud edges, where retrievals are more ambiguous. Hence no further action was required.

page 3 / 15 I wonder how stable the CLARA data-set is, especially involving AVHRR data with drifting overpass time and different sensors.

The issues that the referee mentions are more pronounced during the first two decades of CLARA-A2 data. In fact it has been shown that CLARA-A2 data are stable from the 2000s onwards (see also Karlsson et al., 2017). We use data from the 10 most recent years of the data set, from afternoon satellites NOAA-18 and NOAA-19. For every month, the most recent satellite available was used. Hence, the rapid succession of satellites ensures that when their orbital drift could start compromising the stability of the time series, their data were already not being used in the analysis. This point will be included in the revised manuscript to clarify the issue.

Final response: The relevant information was included in Sect. 2.2 of the revised manuscript (page 3, lines 27-31).

page 3 / 30 consistent changes of different data is certainly a help but for clouds I wonder how independent the cloud retrievals (other than cloud cover) are.

We understand the referee's point. Since both CLARA-A2 and MODIS use the Nakajima-King principle to retrieve cloud optical thickness and effective radius, their retrievals are not completely independent. We will rephrase accordingly.

Final response: This part of Sect. 2.3 was rephrased.

page 4 / 12 what is a pixel? (I assume a 1x1 lat-lon region), thus there are 50 pixels in the regions investigated region

Correct, it will be clarified.

Final response: The term "pixel" was replaced by "grid cell" throughout the manuscript, when level 3 data are mentioned.

page 4 / 20 domestic burning may be largest in winter … but what about industrial emissions as I also expect that mitigations (e.g. to electric vehicles) have lowered emissions?

Yes, industrial emissions may also have been decreased, and their seasonality pattern will be different. A relevant description will be included in the revision for completeness. However, these changes are not depicted in the GFED analysis, since they originate in a different aerosol source.

Final response: industrial emissions from CEDS were included in the analysis. According to this inventory, industrial emissions have not decreased but rather increased over the decade studied (Figure S5).

page 4 / 35 the aerosol typing of calipso should be taken with care. What does 'polluted dust' really tell you on fractions of dust and pollution? without a good distinction between pollution and smoke how to argue via biofuel (GFED) rather than fossil fuel (IPCC6) changes?

As we mention in other parts, the primary goal of the study is not to attribute sources on different aerosol types, especially given the uncertainty that characterizes the CALIPSO aerosol subtypes. We argue, however, that consistency in seasonal characteristics and changes in an aerosol subtype with GFED constraints the possibilities of the origin of this subtype. In this sense, we claim that the "polluted dust" subtype is probably of local origin. For its possible effects on interactions with clouds, however, fractions of dust and pollution are of less importance.

Final response: As explained above, CALIPSO aerosol subtypes were excluded from the analysis, to avoid misinterpretations on possible source attributions.

page 4 / 38 given the potential of advection it would be useful also to look at trends including surrounding regions (at least for the aerosol data)

While this would support conclusions on the specifics of the origins of these changes, this analysis would exceed the scope of the study, since attributing specific reasons of changes in aerosol concentrations is not the primary goal.

Final response: We tried to provide a more complete view of possible reasons for the reported aerosol decrease by including industrial emissions along with biomass burning emissions from GFED.

page 5 / 2 MODIS pixels for AOD are usually provided at 10x10km so the use of 'pixel' here for 1x1 deg is a bit misleading

The term here refers to level 3 "grid cells". We will use this term consistently for more clarity.

Final response: The term "pixel" was replaced by "grid cell" throughout the manuscript, when level 3 data are mentioned.

page 5 / 16 please also add industrial emissions in Figure 3 (contact S.Smith, who provides industrial emissions over time – also for China – for IPCC6 simulations)

This is a constructive idea adding to the completeness of the analysis regarding sources of aerosol change and will be considered in the revised manuscript.

Final response: Figures showing the overall seasonal cycle and changes in emissions over the area and period examined were added in the supplement.

page 5 / 42 it does not seem too intuitive why there should be so much less liquid water clouds the monsoon season: is this an artifact since there are more ice clouds so that the lower altitude clouds cannot be seen?

Our results are verified by two independent studies cited in this part of the analysis (Pan et al., 2015; Cai et al., 2017). Following the referee's question, the same analysis for ice clouds will be included. Please refer also to our reply on the referee's previous comment on Figure 4.

Final response: The same analysis was repeated for ice clouds, showing that they indeed peak during summer. While the studies mentioned here verify the prevalence of liquid clouds in winter, the possibility of the liquid CFC being artificially decreased in summer due to the presence of ice clouds, as the referee suggests, cannot be excluded. Hence it was added in the relevant discussion (page 6, lines 39-41).

page 6 / 5 the 1x1 region based changes to water cloud cover and water content show a lot of sub-regional signals … so maybe also aerosol data (e.g. AODf data) should be examined on that basis…?

Following the referee's suggestion also in other comments regarding analysis of fine mode AOD, corresponding data from MISR will be included in the revised study.

Final response: Results from the suggested analysis are shown in Fig. S2 of the supplement.

page 6 / 20 in figure 6 changes in the y-axis direction are relevant, so that the Reff results are rather meaningless – other than could drops were relatively large in 2003 … which however was identified in the text as an artifact. Although Figure 6 is a nice analysis the results may not be robust enough to make good cases for cloud property changes.

In Fig. 6 the x-axis position is also important since it determines the start year of the period examined each time. Hence in the R_{eff} case, apart from start and end years around 2000, which are known to be artifacts, the plot shows that there was a relatively constant decrease in the period before 2000 and a relatively constant increase afterwards. We consider that even after identification of the artifacts, this analysis still contains useful information, and it is a relatively robust way to exploit the long time series available from CLARA-A2 and highlight the importance of changes during the past 10 years compared to earlier periods.

Final response: No additional changes implemented.

page 6 / 20 the changes to aerosol (earlier) and cloud (later) do not have the quite same seasonality … and the admittance of potential meteorological causes is good though limiting to the goals of the study.

The seasonality in changes is indeed different. However, it would be oversimplified for ACI to manifest through common seasonality in changes of total aerosol and cloud properties. But in cases where strong changes in clouds and specific aerosols occur simultaneously, possible interactions playing a role are worth investigating. We consider however a prerequisite in such a study to "admit" potential meteorological causes and examine if these can explain the changes reported or if they can be excluded as explanatory mechanisms. Our analysis indicates that the latter is true.

Final response: No additional changes implemented.

page 7 / 5 unfortunately no renalaysis data are shown (in that context it would be interesting if reanalysis sees the same 'trends' for aerosol and cloud properties as the observations. The reanalysis also could address if there were changes to the monsoon (strength and location) … e.g. were there trends in precipitation and are there temporal shifts (e.g. associated potentially with temporal delay with more

fine-mode aerosol)? Also do the reanalysis data see shifts in fine-mode AOD altitude as indicated by CALIPSO?

All the analyses suggested by the referee would be interesting and possibly strengthen some of our conclusions, although there is always a question when reanalysis agrees with observations: does this agreement verify an underlying mechanism or is it just due to assimilation? In the case of CAMS data, probably the latter holds, since the aerosol-cloud-radiation interactions mechanisms are not included in the model (Flemming et al. 2017). Such an analysis would extend considerably the present study, of which the purpose is to examine what can be possibly inferred using observations only, minimizing the need for analyzing reanalysis data.

Final response: No additional changes implemented.

page 7 / 25 to exclude monsoon impacts the focus is on changes to aerosol and clouds in the dry season … so maybe this can be even part of the title.

The season of largest aerosol and cloud changes is prominently mentioned in the abstract and elsewhere. We think that including this in the title would make it excessively long.

Final response: No additional changes implemented.

page 7 / 37 add 'rapid' after 'more'

Ok.

Final response: Added (page 9, line 21).

page 8 / 1 also aerosol from pollution is absorbing (ssa ~ 0.92) – although usually not so strong as from fresh biomass aerosol (ssa ~ 0.85).

This shows that even if part of the aerosol change is from pollution rather than biomass burning, there is still considerable absorption. We will include this detail in the revised version.

Final response: The possibility of absorbing aerosols from pollution was added (page 9, line 28).

page 8 / 13 if there I so much low altitude cloud cover in Oct and Nov, how reliable is the CALIPSO statistics (other than the strong trend changes within 1 month seem highly suspicious)

In October and November the cloud fraction is around 70% (Fig. 4). Therefore, there are still considerable cloud-free periods, allowing to retrieve aerosol information from CALIPSO for the full atmospheric column. The aggregation of CALIPSO data over rather coarse (2 x 5 degree) grid boxes further increases the number of valid aerosol profiles. In addition, when low clouds are present, potential aerosols above these clouds can still be identified with CALIPSO.

Final response: No additional changes implemented.

page 8 / 35 interesting but speculative, although possible at least to some degree

This mechanism is indeed mentioned as a possibility, with no claim of proof based on the present analysis.

Final response: This statement was largely rephrased, and now focuses on the November-December case, since after the omission of CALIPSO aerosol subtypes the October case is not adequately supported (page 10, lines 29-34).

page 9 / 10 to make the stated case on absorbing aerosol decrease in that region (other than aerosol in that region is absorbing to start with) it would be useful to demonstrate that these aerosol reductions are associated with the fine-mode AOD (at this point there is a reliance on Calipso typing which is quite general and its association with biomass burning is not very convincing as also urban/industrial (fossil fuel) emissions were reduced and may be even more relevant.

The association with biomass burning was based on the good agreement with the GFED, rather than reliance on CALIPSO aerosol subtypes only. However, fine mode AOD analysis will be included to add robustness to our conclusions.

Final response: MISR fine mode AOD and industrial emissions were included in the analysis and the CALIPSO aerosol subtypes were omitted. GFED is still used as an indicator of association with biomass burning.

page 9 / 20 the disclaimer is warranted (though disappointing)

We agree with the referee's comment. However, one should acknowledge these limitations, which are inherent to observations, and recommend the most promising way forward, which includes modelling simulations in order to overcome these limitations.

Final response: No additional changes implemented.

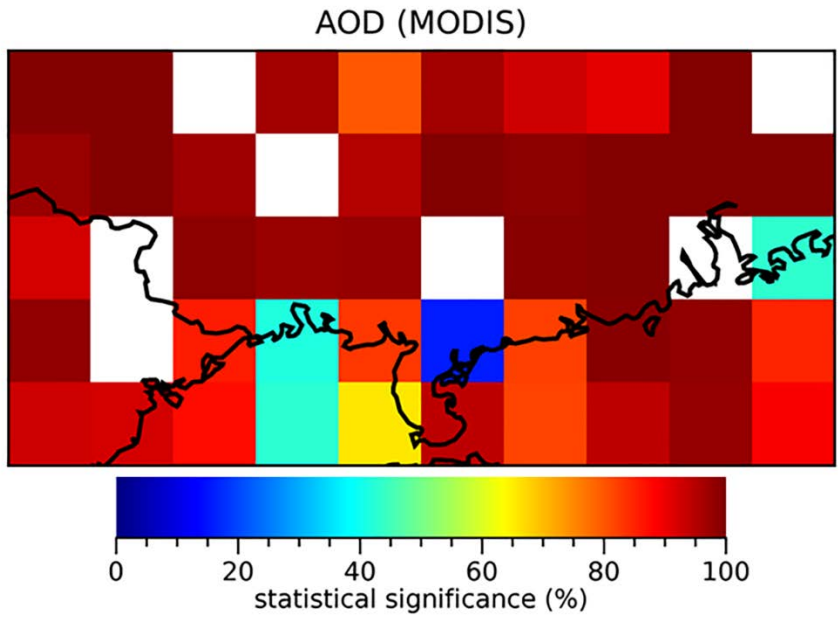
Final response to Anonymous Referee #2

Pg. 1, Lines 10-12: This statement in the abstract follows from Fig. 2 and Fig. 5 (panels above). It is important to see predictive statistics associated with these trend lines rather than just the percent changes. There's quite a bit of scatter in the data, some of which may be seasonal variability, some of which may be interannual variability, and then there's the uncertainty of the Level 3 data product itself. What are the slopes of the dotted lines (yr^{-1})? What are the p-values for the statistical test that one can reject the null hypothesis that the slope of the dotted line is zero?

Figures 2 and 5 provide information on the statistical significance of the plotted lines in the 95% confidence intervals, with corresponding percent changes highlighted in bold. This is explained in the caption of Fig. 2. It was omitted from Fig. 5 caption, but it should indeed be included for clarity. Regarding Fig. 5, it is also clearly stated (page 6, line 10) that in most of the cases the statistical significance level is below the 95% confidence interval. This is the reason why the analysis proceeds further to monthly changes, where the seasonal variability is removed. Reporting percent changes instead of (absolute) slopes was selected as a more intuitive measure of change. Regarding p-values, a table could also be added for completeness. The following table provides the information requested by the referee:

Parameter	Unit	CALIPSO	MODIS	CLARA-A2
		Change (%)/slope (<unit> yr^{-1})/p-value	Change (%)/slope (<unit> yr^{-1})/p-values	change (%)/slope (<unit> yr^{-1})/p-value
Total AOD	1	-23.3/-0.013/0.013	-17.6/-0.010/0.002	
Dust AOD	1	+8.4/0.0003/0.797		
Smoke AOD	1	-22.5/-0.006/0.071		
Polluted Dust AOD	1	-33.5/-0.008/0.003		
All-sky LWP	g m^{-2}		+12.4/0.837/0.204	+14.2/0.913/0.242
Liquid CFC	1		+6.8/0.003/0.219	+3.4/0.002/0.465
Liquid COT	1		+5.5/0.089/0.399	+3.6/0.058/0.607
Liquid REFF	μm		+1.6/0.018/0.239	+5.2/0.034/0.0003

The following figure depicts, on a pixel basis, the level of statistical significance for MODIS AOD changes (corresponding to Fig. 2a). For similar maps corresponding to the changes shown in Fig. 5a and 5b, the reviewer is referred to one of our later replies (page 11 of this document).



Final response: The requested statistical measures are provided in Table S3 of the supplement. The map provided here is also provided in the supplement (Fig. S1), updated to highlight grid cells with levels of statistical significance over 95%.

There's also quite a bit of day-to-day and sub-pixel variability that is not reflected in the Level 3 gridded monthly mean product as well as different numbers of measurements (i.e., samples) in each pixel that need to be considered and this is not really discussed in the manuscript. Some mention of area weighting of pixels is given on Pg. 3, Line 38, but is not described; how was this done? The monthly-averaged CALIPSO observations also have different numbers of observations that are averaged to yield the reported gridded mean and standard deviation – in addition to the area weighting, were these differences in number of samples accounted for when averaging across the region or across different months/years?

The area weighting mentioned by the referee concerns the differences in surface areas of grid boxes due to different latitudes. Because of the small size of the domain these differences are minor. This could be clarified in the statement of page 3, line 38. The different number of observations was accounted for by applying a threshold on the minimum number of days used in the monthly mean calculation (on a pixel basis) before estimating the spatial average (see also Section 2.4). In the case of CALIPSO, averages were weighted by the number of samples used, which is available in the level 3 data. Data sets from different sources will of course have different numbers of observations being averaged, as the referee mentions. The same concern led us to apply the thresholds described in Section 2.4, in order to minimize ensuing discrepancies. While we agree that sub-pixel variability is not reflected in the gridded monthly mean products, we consider that some rephrasing could answer the referee points previously mentioned.

Final response: The statement of page 3, line 38 was clarified in page 4, lines 12-14 of the revised manuscript, and the clarification regarding the CALIPSO spatial averaging was also included (page 4, lines 16-17).

It would helpful for the reader to see the Level 3 standard deviations on these trendline graphs as error bars or as a shaded region. How is this additional sub-month, sub-grid-cell variability being captured in the statistical tests to assess whether or not there is a trend? Assuming there are indeed, statistically-significant trends (which I don't think hasn't been discussed very extensively at all) is the trend in AOD is related to the trend in CFC or LWP or are they coincidental? The italics statement above from the abstract implies that there is a non-coincidental relationship.

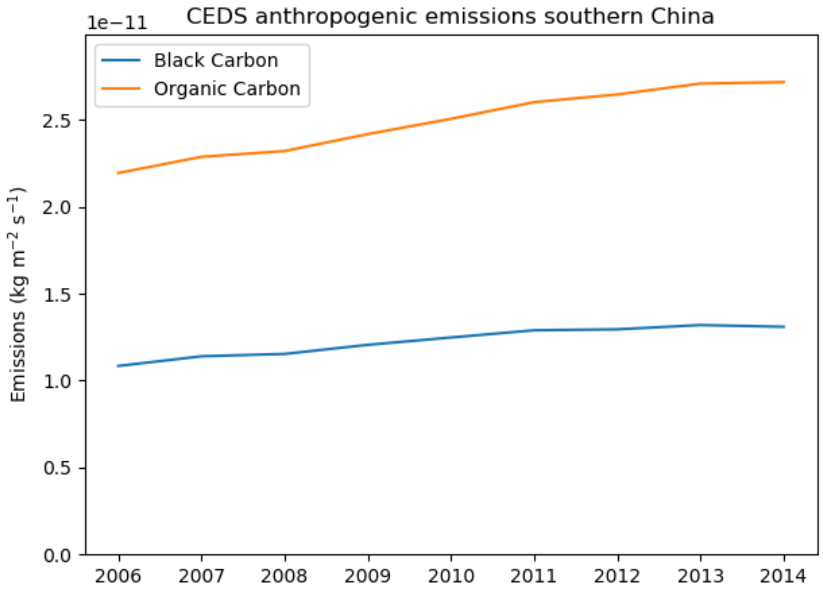
The requested information could be added in the graphs. While we considered that the statistical significance of trends was adequately discussed, this discussion could also be extended, with an addition of a relevant table of p-values, as shown before. However, based on the present discussion in the paper, it is explicitly mentioned that some changes are statistically significant and some are not, as was also explained in our first reply. Hence, we don't understand why the referee would still "assume" that "there are indeed, statistically significant trends". Regarding the question on the relation or coincidence of changes in aerosols and clouds, it is one of the main science questions of this study, as described in the Introduction (page 2, lines 11-12), and we attempt to address it based on the analysis described in Section 3.3. Our results imply that there is indeed a non-coincidental relationship.

Final response: Standard deviations were included in the updated Fig. 2 as shaded regions. Corresponding statistical measures, including p-values, are given in supplementary table S3.

Pg. 1, Line 12-13: The fundamental flaw with this conclusion is that it is not clear that all aerosol types have been captured, so one cannot say that the "main driver" of AOD trends over the last decade is biomass burning or continental pollution or marine aerosol or other types, because only the three aerosol types are included in the Level 3 CALIPSO product (dust, smoke, and polluted dust), and critical information about the trends of these other aerosol types is lacking. One must also ask the question, are the CALIPSO aerosol types sufficient to answer the question that's being posed, or does one need more specificity with regard to aerosol composition (e.g., sulfate, organics, dust, black carbon) that must be obtained from a model? Therefore, I would characterize this conclusion as unsupported by the underlying data and highly speculative. One way to address this criticism would be to not use the Level 3 data, but rather to use the Level 2 data that has more aerosol type classifications. Another approach would be to use model data products to explore this research question. Of course there would be uncertainties associated with any aerosol type classification scheme that would make it difficult to compare across different data sets – for example, the CALIPSO smoke aerosol probably is not only associated with biomass burning and also includes the contribution of other anthropogenic combustion sources. Another advantage of using the Level 2 data products is that they are not gridded and temporally averaged, so they capture a truer range of measured variability. This helps avoid biases, because the mean of the means is not always the same as the mean of the population if sample sizes are not constant and this unequal weighting is not accounted for properly.

It is true that the lack of the full set of aerosol subtypes is prohibitive for the attribution of their overall decrease to a "main driver". This expression should be corrected accordingly. Our results, however, show a statistically significant decrease in total AOD and in an aerosol subtype. This may indeed not be the main driver, but the lacking information on how some other subtypes change is not "critical" for further analyzing this specific subtype. Regarding the sufficiency of the CALIPSO data used "to answer the question that's being posed", and based on the referee's suggestions (level 2 data, model data) and some later comments, it seems that there is a misunderstanding on the reason of using CALIPSO in this study, which should be clarified also in the manuscript: it is neither to attribute aerosol sources, nor to unveil ACIs in their process level. It is to include information on the vertical distribution of aerosols, and its possible changes, which is critical for their position relative to clouds. This is why, while the CALIPSO subtypes are analyzed in terms of changes and compared to other data sets (e.g. GFED), the ambiguity regarding their origin is repeatedly stressed throughout the manuscript.

Following a suggestion by referee #1 we have collected CEDS anthropogenic emission estimates compiled for CMIP6. As shown in the figure below, these estimates suggest that both the organic carbon and black carbon emissions cannot explain the decrease in AOT in southern China, because both show slight increases in the 2006-2014 period.



Regarding the referee's remark on the unequal weighting in averaging, as explained in our previous reply, the unequal number of observations is actually accounted for.

Final response: As explained in our general comment, CALIPSO aerosol subtypes were omitted from the revised study. Information on other emissions is now included in the supplement based on the CEDS data set.

Putting aside the major flaw of the missing aerosol types, it is also hard for me to see the trends in the data that the authors are using as a basis for saying that AOD changes occur in late Autumn and early Spring. From Fig. 1a and Fig.3 (shown at right), the peak in biomass burning in GFED is apparent

between Nov.-Mar., while the ΔAOD traces vary quite a bit but don't really peak in this period. There is some decrease (the traces are below zero), but there is also a good bit of scatter in the data.

The combination of plots provided here by the referee to support the above criticism actually compares the seasonal variation in biomass burning emissions from GFED with the *changes* in the seasonal variation of AOD and biomass burning emissions. It is not obvious why the maximum change in a parameter should also coincide with its maximum average value, as the referee seems to expect. It is also worth noting that the original statement, cited by the referee (page 1, lines 12-13) reads "changes occurred mainly in late autumn and early winter months*",* not "*early Spring*". In fact, nowhere in the manuscript is said that "*AOD changes occur in late Autumn and early Spring"*. The sentence cited by the referee mentions "early winter", and actually refers to AOD from MODIS and CALIPSO, and their changes in October, November and December (Figs 3a and 3b). We would be happy to rephrase this part in order to clarify it. However, the referee has drawn here a red box ranging from November to March, to show that (indeed) ΔAOD does not peak in this period. The same mistake is repeated in a later comment (page 9 in this document). Hence, this criticism is rather superficial and obviously unsupported.

Final response: The misunderstood term, along with large part of the abstract, has been rewritten, based on the updated analysis.

There are no metrics of statistical variability included in this graph (Fig. 3) – only the means – so it is hard for me to assess the statistical significance of the data. The authors say they did t-tests, but on what? Where are these statistical results presented? Statements are made in multiple places that trends are statistically significant but no p-values are provided. Where variables are thought to be correlated (as in the case of biomass burning emissions and AOD), there are no correlation coefficients provided. I see this lack of scientific rigor as a major flaw in this study. It also led me to comment that most of the correlations suggested are determined by whether one or more variables trend up/down together over time, which I guess is determined visually. Having some numbers here related to the statistics, I think, is very important.

We appreciate the referee's request for more details on the metrics of statistical variability. There are indeed statements in multiple places that changes are statistically significant, with corresponding p-values not provided, and a concentrated report of corresponding metrics could help. However, describing the method used for the assessment of statistical significance, and then reporting results individually, is a rather common practice in similar cases and does not constitute "lack of scientific rigor". In fact, it is a matter of a simple revision for these metrics to be provided. However, the "visual determination" of correlations, mentioned here by the referee, does not qualify as a valid scientific approach. Hence it is surprising that this is the referee's "guess" regarding our methodology, and unfortunately it is not even accompanied by the "benefit of the doubt". In our opinion, such a serious statement in a review process should at least give to the authors the opportunity to disprove it, instead of leading directly to such a negative judgement. Nevertheless, we take the opportunity here and provide the requested metrics:

- Page 5, line 6: p-value=0.002 for MODIS, 0.013 for CALIPSO.
- Page 6, line 4: please refer to the maps provided here in page 11.
- Page 6, line 10: please refer to the table provided here in page 1.
- Page 6, lines 33-34: please refer to the table provided here in pages 11-12.
- Page 8, line 9: the referred decrease is not "significant" in a statistical sense (95% confidence interval). The term should be replaced to avoid misunderstandings.
- Page 9, line 9: p-value=0.03. This decrease refers to polluted dust aerosols, should be replaced for clarification.

The following table shows the Pearson's correlation coefficients, on a monthly basis, of each CALIPSO aerosol subtype with GFED emissions. Please note that nowhere in the text is the total AOD correlated with GFED, as mentioned by the referee. Please also note the correlation between GFED and polluted dust AOD in November, which led to the discussion in page 5, lines 24-29 of the manuscript.

	Dust AOD	Smoke AOD	Polluted dust AOD
January	-0.27	-0.06	-0.10
February	-0.28	0.37	0.18
March	0.14	0.59	0.16
April	-0.38	-0.50	0.30
May	0.42	-0.07	0.27
June	-0.37	0.24	-0.07
July	-0.31	0.16	-0.23
August	0.28	0.00	0.06
September	-0.44	-0.37	-0.37
October	-0.09	-0.07	0.37
November	-0.43	-0.05	0.74
December	-0.30	0.62	0.25

Final response: Apart from those referring to CALIPSO aerosol subtypes, all the metrics provided in our reply above are now given either in the main text or in the supplement.

Pg. 1, Line 13: The panels from Figs. 3 and 7 shown at right on this page indicate the seasonal variation in changes of AOD (top) and cloud properties (bottom three panels). There is a very clear and distinct change in cloud properties in Nov.-Dec. that does not appear to be related to the changes in AOD during this period. I don't understand the basis for the italicized statement made in the abstract that changes in AOD "coincided with changes in cloud properties".

The term "late autumn and early winter months" should probably be replaced by "November and December" to clarify the issue. It should be obvious also from the red box that the referee has drawn that in these months there are changes in AOD and cloud properties that coincide. It is not clear what the referee means by "*does not appear to be related to the changes in AOD during this period*". The term "related" was not used in the statement that the referee cites, and no "relation" was established based on the plots that the referee has compiled. The fact that in other months (e.g. October) AOD and

cloud property changes do not coincide, does not negate our statement. It is actually the main finding of our paper that the AOD changes in October occur at a higher level and thus have different effects on clouds.

Final response: The cited term, along with large part of the abstract, have been rephrased.

Pg. 1, Line 15-17: The semi-direct posits that solar heating of above-cloud absorbing aerosol layers changes the temperature profile of the atmosphere, reducing buoyancy, and ultimately cloud cover and liquid water path. To be consistent, then, with the semi-direct effect, I would expect to see an inverse correlation between absorbing aerosols above cloud and these cloud properties. What is shown in Figure 8 are monthly-averaged differences in the vertical profile of aerosol extinction as well as the vertical profile of cloud extinction. First, extinction is not absorption. Even relatively close to fires, the scattering-to-extinction ratio is > 0.8 (e.g., Yokelson et al., Atmos. Chem. Phys., 2009; https://doi.org/10.5194/acp-9-5785-2009), and it is known that the ratio is much higher as the smoke plumes age. No data is being presented regarding smoke age, whether or not the smoke is from urban pollution or biomass burning, or that there is or isn't any trend in absorbing aerosols over this region.

Second, the CALIPSO Level 3 data typing algorithm identifies smoke only when the layer is elevated – by definition! Therefore, it is not appropriate to use the positioning of this smoke product to suggest that there is some sort of vertical relationship with cloud. The smoke classification type shares many similar features to the polluted continental classification type, except that the latter is at the surface and not elevated. The polluted continental classification type has not been considered in the present analysis, which is a major gap in the analysis. Finally, I don't understand the relevance of the ISCCP classification types to this discussion – this classification scheme seems much too coarse to be meaningful. In sum, I see no conclusive evidence that aerosol changes are altering the temperature profile of the atmosphere to effect changes in clouds. Consequently, I don't think that it's appropriate for the authors to suggest that the semi-direct effect is a causal mechanism for the observed, 5-13% increase in LWP and cloud fraction from 2006-2015.

The referee's statement that the semi-direct "posits that solar heating of above-cloud absorbing aerosol layers changes the temperature profile of the atmosphere, reducing buoyancy, and ultimately cloud cover and liquid water path" contradicts the widely described semi-direct effect mechanism for absorbing aerosols above stratocumulus clouds (e.g., Koch and Del Genio, 2010). The buoyancy is indeed reduced but this will lead to less entrainment at the cloud top and consequently an **increase** in cloud cover / liquid water path. Hence, a decrease in absorbing aerosols above clouds would be consistent with a corresponding decrease in stratocumulus clouds below. This is exactly what is shown in Figs. 8b and 9b.

The purpose of Fig. 8b and c is to indicate at which height the changes in aerosol occur. The CALIPSO extinction profile suits that purpose. The fact that "extinction is not absorption" is irrelevant for the conclusions regarding the vertical location of the aerosol changes. We acknowledge in the manuscript that the CALISPO smoke classification is accompanied with some uncertainty. As a result we do not know for sure how absorbing these elevated aerosols are. However, for the polluted dust aerosols,

showing largest decreases in November, we have solid indications that they are strongly absorbing because their decrease goes together with a decrease in GFED biomass burning emissions (Fig. 3c) while anthropogenic emissions did not show a decrease (see figure on page 4 of this reply).

The ISCCP classification was included to highlight the changes in low clouds for October and November. We don't see why this classification, which has been used widely in the past, does not serve this purpose.

Final response: Figure 8 was updated to omit aerosol subtypes and aggregate months. Figure 9 was omitted, since, following the revised analysis and the ambiguity of aerosol absorption in the October case, it does not provide any additional information.

Pg. 4, Line 32-33: Why was it not possible to explore this discrepancy? How would further investigation be carried out? This is a very shallow approach to analyzing the data.

Our statement reads "*…it was not possible to pinpoint specific reasons for the March-April differences based on the data sets used here".* Contrary to the referee's understanding, this statement denotes that this discrepancy was actually explored, based on the data sets used here, but no explanation was found. Hence, further investigation would require further analysis of additional data sets, focusing on these months. This would extend beyond the scope of this study, which focuses on the October and November months.

Final response: No additional changes implemented.

Pg. 4, Line 35-36: Biomass burning aerosols do contribute to smoke layers, but so do other sources of combustion. Similarly, biomass burning, urban pollution, and fossil fuel combustion aerosols contribute to the polluted continental aerosol type (which is not accounted for in this study). A key difference between the CALIPSO smoke and polluted continental aerosol types is whether or not the layer is at the surface or elevated. Since the aerosol classification types are based on aerosol intensive and extensive parameters, there can be misclassification and some ambiguity across aerosol types, particularly for categories dominated by smoke and urban pollution because both types of aerosol are dominated by relatively small, non-depolarizing aerosols. The polluted dust category isn't necessarily a mix of biomass burning and dust – it represents the middle part of the continuum between smoke/continental-pollution (small and weakly depolarizing) and dust (large and strongly depolarizing). The satellite aerosol-typing products are very useful, but they are not unambiguous. This statement is too strong and not supported by the data.

We thank the referee for these clarifications, which could be added in the relevant discussion along with the appropriate references. We also agree that the aerosol-typing products are useful, but they are not unambiguous. In fact, the ambiguity regarding especially the smoke aerosol type is explicitly stated in page 5, lines 31-32. This statement could also be rephrased in accordance with these ambiguities.

Final response: This statement was removed, since CALIPSO aerosol subtypes analysis was omitted.

Pg. 4, Lines 36-37: I agree with the authors' statement here, and yet, Figure 1 and Figure 3 attempt to make precisely this comparison.

This statement, along with the next sentence (page 4, lines 37-39), explain why biomass burning emissions and satellite-based AOD are expected to differ (i.e. not being directly comparable). However, including them as subplots in Figs. 1 and 3 is useful, in our opinion, since the former can help explain the latter.

Final response: This statement was expanded for clarification (page 5, lines 19-20).

Pg. 5, Lines 7-8: It is true that fitted lines to both polluted dust and smoke aerosols trend down during this period along with the overall AOD. However, it is unclear what the trend in continental pollution or marine aerosols are for this period because they have not been considered by this study. Certainly, decreases in polluted dust and smoke contribute to the decrease in AOD, but I don't think that the authors can "attribute" the change to only these two aerosol types when there are other types that are not being considered.

We understand that the term "can be attributed" may be misunderstood as rather definitive, when other possibilities are not excluded. However, the total AOD from the three CALIPSO categories matches quite closely with the total AOD from MODIS, so other categories appear to play a minor role. Moreover, anthropogenic emission estimates (including continental pollution) do not show a decrease over the investigated time period (see page 4 of this reply).

Final response: This statement was removed, since CALIPSO aerosol subtypes analysis was omitted.

Pg. 5, Lines 14-15: What evidence is there that the smoke and polluted dust aerosol types are dominated by biomass burning aerosol versus other sources of combustion or pollution aerosols? The CALIPSO aerosol type is not specific to biomass burning. Consequently, for the authors to make this conclusion, they need to provide some other evidence. Since no such evidence is apparent in this manuscript, this seems highly speculative.

One piece of evidence that biomass burning emissions play a major role, already provided in the paper, is GFED. Another piece of evidence are the CMIP emission inventories, mentioned here in page 4, which could be added to the manuscript.

Final response: This statement was removed, since CALIPSO aerosol subtypes analysis was omitted.

Pg. 5, Line 15-18: No data on residential energy sources are provided or discussed in this manuscript, so this statement is entirely speculative, and, frankly irrelevant to the present study. The previous studies cited in the next sentence are also not sufficient to support this statement, as they are not recent enough be cover the 2006-2015 time period in this study. Even if there was a decrease in residential biomass burning emissions starting in the 1990s, such a decrease does not necessarily extend to present day. This conclusion is unfounded.

Information on the seasonal peak in residential energy sources is provided in He et al., (2011). While this reference should be added here, it should also be clear that this is not a conclusion of the present study, since no relevant data are provided here. The way the referee connects this sentence with the next ones is rather arbitrary. We hope it is clear now that these previous studies are not provided here to support the referred statement, but in a more general discussion on how previous findings relate to ours, which is a rather common and necessary practice.

Final response: This statement was rephrased for clarification (page 6, lines 5-6).

Pg. 5, Line 20-: Again, showing the same figure as before at right, it can be seen that there is no agreement between the change in AOD and the change in C emissions (delta-AOD even becomes positive in January, while delta-C is fairly constant). I'm not sure I understand what is being meant by the term, "partially agrees". It appears that during the seasons where delta-C reaches a local minimum and is fairly stable that both MODIS and CALIPSO delta-AOD are quite variable and not at a local minimum or maximum. Finally, is it even appropriate to be trying to establish this comparison, as it was already stated on Pg. 4, Lines 36-37: "Biomass burning emissions and satellite-based AOD are not directly comparable"?

Biomass burning emissions constitute part of the total aerosol load. Hence, they are expected to agree better with the total aerosol load concentration (or changes) when they dominate compared to other sources, rather than when other sources (or their corresponding changes) dominate the total aerosol load. This is the intended meaning of the term "partial agreement". ΔC actually reaches a minimum in November, when MODIS and CALIPSO ΔAOD also exhibit large decreases, but not necessarily their minimum values (since they are not expected to always agree). It is obvious that the term "not directly comparable" causes a misunderstanding, and could be replaced by "not expected to always agree", which is probably more appropriate for the intended meaning. Regarding the appropriateness of the comparison between biomass burning emissions and satellite-based AOD, the referee is referred to our previous reply (page 8 of this document). In short: yes, comparing a total with its part gives insights on the relative contribution of the latter to the former.

Final response: The misunderstood term was rephrased and expanded (page 5, lines 19-20).

Pg. 5, Lines 23-26: What evidence is there to assert that the aerosols or smoke observed over this region is transported from neighboring regions such as Indochina (versus long-range transport or local emissions)? No data on fire activity in neighboring regions is presented, nor is any information on air mass back trajectories. What about the confounding influences of local, urban pollution and non-biomass combustion aerosols on the CALIPSO types? This italicized statement seems highly speculative.

This statement is not an "assertion" based on "evidence". It is a "suggestion", a possible explanation for the reported results, not excluding other possibilities. We agree that there are alternative possibilities, and we thank the referee for suggesting methods that could lead to an assertion based on evidence.

Final response: This statement was removed, following the omission of CALIPSO aerosol subtypes from the analysis.

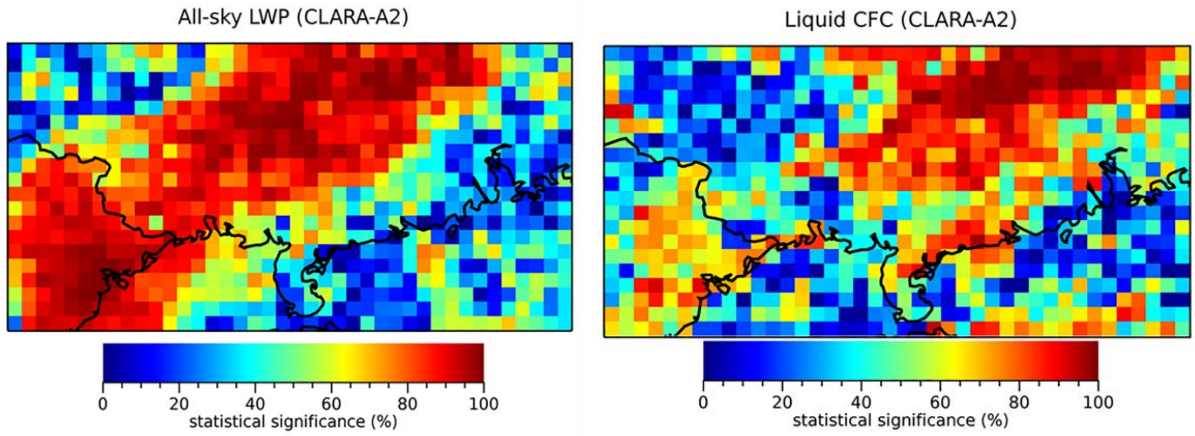
Pg. 5, Lines 29-33: I agree 100% with this statement. The problem is that this ambiguity undercuts many of the conclusions put forward in this manuscript. This is made even more problematic in that the continental pollution aerosol type is not included in this analysis. Given these uncertainties, a fundamental question that must be asked is, is this satellite-based AOD aerosol type data set appropriate to address source attribution? Given that some major aerosol types are missing, I think the answer is that it is not appropriate.

We agree that there are serious limitations in using this data set to address source attribution (see page 8, lines 40-42). As stated in page 2, lines 26-27, this data set was not used here for this purpose. It could also be (further) clarified that possible relations of CALIPSO aerosol types with aerosol sources were based only on previous studies (page 4, lines 35-36, page 5, lines 28-29), and do not constitute "strong" conclusions of the present study. This should be obvious from page 8, line 41, but it could be further emphasized.

Final response: This statement was removed, following the omission of CALIPSO aerosol subtypes from the analysis.

Pg. 6, Lines 8-10: I'm struggling to interpret the meaning of this statement. Spatial distributions of the change in LWP and cloud fraction are presented that demonstrate large increases in LWP and cloud fraction over land and decreases over water. No indication is given in Figure 5 of the areas where these changes are or are not statistically significant (often I have seen this done with speckling overlaid on the statistically significant portions). It also sounds like from this statement that most of the cases in Fig. 5 are not statistically significant. Yet, these are the numbers that are being quoted in the abstract for changes in liquid cloud cover and liquid water path of "5% and 13%, respectively" (Pg. 1, Line 12) and for drawing other conclusions later on. Are these numbers statistically significant?

The information requested by the referee can be easily provided. In fact, the following two maps show the level of statistical significance, on a pixel basis, of all-sky LWP (left) and liquid CFC (right):



In the paper, these results were summarized in the statement that the referee mentions, but they could also be included, to avoid struggles with interpretation. We think, however, that the statement "*reduces this significance to levels below 95% in most cases of Fig. 5."* is clear. The referee is also referred to the

table in the first page of this reply. It is not clear, however, what the referee means by the term "drawing other conclusions later on". After stating that the statistical significance is reduced to levels below 95% in most cases of Fig. 5 due to averaging (page 6, lines 9-10), the 34-year CLARA-A2 time series is analyzed (page 6, lines 13-26), and then changes are examined on a seasonal basis. In page 8, lines 33-34 it is stated which of the cloud properties exhibit statistically significant changes in which months. To clarify any further misunderstandings, we include here a detailed table with corresponding levels of statistical significance (in %) for every cloud property examined and every month:

	All-sky LWP		Liquid CFC		In-cloud LWP		Liquid COT		Liquid REFF	
	CLARA	MODIS	CLARA	MODIS	CLARA	MODIS	CLARA	MODIS	CLARA	MODIS
Jan	14.13	13.49	54.18	39.26	6.15	5.13	3.59	6.41	14.13	56.52
Feb	10.79	3.68	44.67	39.57	10.60	15.01	5.28	1.13	14.10	52.25
Mar	5.52	15.19	1.47	12.82	12.85	7.65	12.19	11.08	47.77	45.10
Apr	33.05	42.88	28.30	4.58	58.39	61.04	50.12	54.49	73.79	72.08
May	79.19	94.20	61.31	30.53	96.01	99.22	85.28	95.12	86.93	48.38
Jun	13.69	26.27	85.18	49.95	39.64	62.92	36.34	55.18	61.09	59.25
Jul	22.71	39.33	2.57	56.52	26.96	26.94	23.06	2.27	90.11	93.16
Aug	4.06	77.65	57.83	42.97	74.50	29.39	34.42	1.00	86.12	97.35
Sep	36.62	27.02	55.97	37.12	30.88	33.54	21.79	22.90	92.01	46.53
Oct	46.82	69.47	74.85	81.51	72.35	70.81	59.18	66.37	79.20	50.22
Nov	90.07	90.99	99.26	99.55	65.37	86.26	48.39	78.23	89.59	88.76
Dec	97.05	99.33	35.75	54.15	99.43	99.91	96.45	98.57	97.46	67.40

In fact, contrary to the referee's statement, no conclusion is reported based on the changes in the entire time series of Fig. 5. The reason is that "most of the cases in Fig. 5 are not statistically significant". We hope this is clearer now.

Final response: The statistical information provided in our reply above is now included in the supplement. The maps provided here are also provided in the supplement (Fig. S6), updated to highlight grid cells with levels of statistical significance over 95%.

Pg. 6, Lines 33-34: Do I understand this statement correctly to say that there is only one month where each of the cloud property changes is statistically different from zero, and that only cloud fraction is statistically different from zero in November? Why would there be a trend in cloud properties in only a single month? Reading between the lines here, does this mean that the overall change in cloud property changes is not statistically significant, or is the effect size of this single month enough to drive the entire trend?

Yes, this is exactly what this sentence states. Regarding the referee's second question, a possible reason could be "differences in the seasonal (or monthly) characteristics and changes in factors affecting cloud properties". Such a factor is aerosols, and a large part of the present study investigates exactly this question. This single month is not enough to drive the entire trend, hence most of the cases in Fig. 5 are not statistically significant, as stated in page 6, lines 9-10. Our previous reply also provides further clarifications.

Final response: No additional changes implemented. Relevant statistical significance measures are now given in the supplement.

Pg. 6, Lines 37-38: I don't think this statement is correct. Liquid clouds increased only in November on a cloud fraction basis and only in December on a LWP or COT basis (based on the statistical significance discussion in the previous comment). The seasonal pattern of the AOD changes and the cloud properties changes are not similar (either correlated or anti-correlated). This statement suggests that they are anti-correlated, which is not true.

This statement was phrased carefully, to avoid misinterpretations. The referee correctly states that liquid clouds increased only in November on a cloud fraction basis and only in December on a LWP or COT basis, based on the statistical significance results (see also the relevant table in pages 11-12 of this document). We summarized these changes using the phrase "liquid clouds increased mainly in late autumn and early winter", but this could be rephrased, based on the referee's statement, to be more clear. We also agree that the seasonal pattern of the AOD changes and the cloud properties changes are not similar. In fact, we don't see why they should be similar (either correlated or anti-correlated) throughout the year. However, it is one of our findings that some statistically significant changes in properties of clouds and aerosols occurred in the same months, and they are indeed anti-correlated (see also the table in page 16 of this document).

Final response: A new subsection was included (Sect. 3.3) summarizing the results on aerosol and cloud seasonal changes, their statistical significance and correlations.

Pg. 7, Line 10-11: First, statistical significance of the cloud changes is unclear (discussed previously). There does appear to be a decreasing trend in AOD, which the authors assert is statistically significant. That surface pressure and geopotential height do not show a statistically significant trend is insufficient to rule out meteorological drivers. The atmospheric temperature profile, moisture, and lower tropospheric stability are also important variables that do not appear to have been considered. Even if these variables fail to demonstrate a statistically significant trend that does not in and of itself rule out the existence of such a trend. All it means is that the available data are insufficient to reject the null hypothesis, but there may indeed be a trend that might be uncovered by additional data and/or a longer timeseries. The italicized statement is not demonstrated conclusively by the data presented, which are rather superficial.

We hope the statistical significance of cloud and aerosol changes in now clearer, based on our previous replies. Apart from surface pressure and geopotential height, other parameters could indeed be considered, as the referee suggests. It is not clear, however, what the referee means by the following: "*Even if these variables fail to demonstrate a statistically significant trend that does not in and of itself rule out the existence of such a trend. All it means is that the available data are insufficient to reject the null hypothesis, but there may indeed be a trend that might be uncovered by additional data and/or a longer timeseries*". This statement seems to suggest that there is no way of excluding meteorological variability as a factor of cloud and aerosol changes, since there might always be a trend waiting to be

uncovered by additional data and/or longer time series. This contradicts common practices followed in similar studies.

Final response: No additional changes were implemented.

Pgs. 7-8, Section 3.3.2: I think that this paragraph is not at all supported by the underlying data, which until this point has focused on trends and changes over time. In this paragraph, process-level explanations are invoked, but are done at a highly-averaged level spanning months and 5 x 10 degree area. These are not the scales at which aerosol-cloud interactions would be expected to be evident (e.g., McComiskey and Feingold, ACP, 2012, https://www.atmos-chem-phys.net/12/1031/2012/), so the failure to see ACI effects in the trend data is not surprising. Saying that the authors' findings are "inconsistent" with the first and second indirect effects is too strong a statement. These effects may very be visible in this region if a more appropriate data set is used (e.g., aircraft, balloon, surface remote sensor scales measuring clouds over minutes to hours or a model with better space and time resolution). One cannot know. The same is true for the discussion on the semi-direct effect, with the additional comments that have been described above in this review that the attribution of the particles to biomass burning, as absorbing particles, and that these particles are at cloud level are all not established by this data set. In fact, the CALIPSO smoke type is not unambiguous as a marker for biomass burning. That the smoke type often occurs near cloud level is unsurprising given that the layer must be elevated by definition of that aerosol type in the CALIPSO scheme. Similarly, the continental pollution aerosol type is very similar to the smoke type, but it is not in an elevated layer. Since the data set used in this manuscript and its analyses is so highly averaged in space and time, it is of little utility for discussing ACI effects. Consequently, the conclusions as stated are not definitive and this entire paragraph should be removed.

This paragraph examines what can (and cannot) be deduced from the previous analysis regarding ACI and clearly states what can (and cannot) be supported by the data. It is not true that results of ACI cannot be evident at scales larger than their process scale, as the referee suggests. The problem with the temporal and spatial scales used here, is one of quantification of ACI, as clearly stated in the study cited by the referee, and acknowledged also in our study (page 1, lines 33-35). However, no ACI quantification was attempted in the present study, so the referee's criticism is rather unsupported. In the same sense, if a particular mechanism dominated over a large area and period, one would expect to see the consequences in a data set covering this area and period. If the analyzed data sets show changes in a direction opposite to the one expected, then they are "inconsistent" with the previous assumption. We don't see why such a term would be "strong" or "unsupported" by the data.

We agree with the referee that ACI may be visible based on the suggested data and resolutions. However, the purpose of this study is not to provide evidence of ACI on their process level. It is (among others) to examine if their consequences can explain observed changes in a larger scale.

We acknowledge the referee's concerns on the limitations of the CALIPSO aerosol types based on their definitions. In fact, their limitations are acknowledged in several parts of the manuscript (page 5, lines 29-33, page 8, lines 18-20, page 8, lines 40-42). However, this section examines *changes* in their profiles, not concentrations. The referee finds "unsurprising" that "the smoke type often occurs near cloud level". This is a rather confusing statement, since it is nowhere made in the manuscript.

Final response: A part of Section 3.3.2 (now Section 4.2) was revised to reflect the new analysis, namely focusing on the November-December changes, since the degree of absorption of aerosols in October is more ambiguous after the omission of CALIPSO aerosol subtypes (page 9, lines 28-29).

Pg. 8, Line 10: Are the aerosol and cloud profiles shown in Figure 8 an average profile or an individual, typical profile for each? What is meant by "autumn" or "Fall" for the cloud extinction profile – both October and November? If they are averaged profiles, how was that averaging carried out (e.g., was a weighted average of the sample numbers in each pixel used)? Are there meaningful differences in the profiles across the spatial area? A single set of averaged profiles over the entire spatial domain seem difficult to meaningfully interpret to me, as I would expect these profile changes to be very different over land and over water. How should the reader interpret these profiles with regard to representativeness? Are the changes in Fig. 8b and 8c statistically significant at all height levels? The commentary on Pg. 8, Line 21-23 suggests that only certain layers are statistically significant and in different months (e.g., 1-1.5 km altitude for smoke in October and 0.7-1.2km for polluted dust in November). It would appear that the CALIPSO smoke aerosol change is not statistically significant in November when the cloud fraction change is statistically significant. Conversely, the smoke change is statistically significant in October when the cloud fraction change is not statistically significant. What about the December profiles where the other cloud property changes are statistically significant? It is very difficult to unravel what is being presented here, but it certainly does not seem to be suggestive of an aerosol-cloud semi-direct effect (as is stated on Pg. 1, Lines 15-18).

The cloud profile shown in Figure 8 is spatially averaged over the study area and autumn (fall) months (September, October, November), based on measurements from 2007 to 2011 (see also Amiridis et al. 2015, for details on the LIVAS data set). The aerosol profiles of Figure 8 actually show changes, calculated, for each profile level, based on the method described in Section 2.4. The averaging was indeed carried out using the numbers of averaged samples, also provided in the data set, as weights. It is not clear what the referee means by "meaningful differences". It is true that differences should be expected, especially over land and sea, and selecting two of the four pixels covering the study region would probably be more representative of the land profiles. A separate analysis could be performed to answer the question on representativeness.

The referee interprets correctly the statement in page 8, lines 21-23. To clarify further: in October, smoke changes are statistically significant between 1-1.5 km, liquid cloud changes are not; in November, polluted dust changes are statistically significant between 0.7-1.2km and liquid CFC change is also statistically significant; in December, all liquid cloud properties changes examined, except for CFC, are statistically significant, and aerosol changes are not. While some rephrasing might help, we consider reporting the results on statistical significance along with corresponding changes really crucial, hence some difficulty in unravelling the findings should be expected.

Final response: Figure 8 has been updated, omitting the profiles of CALIPSO aerosol subtypes and aggregating data from different months. Due to the coarse resolution of CALIPSO level 3 data, no meaningful discrimination between land and sea was possible.

Figure 9 and related discussion: Why is necessary to break out the cloud optical thickness data by cloud type? It has already been established from Figure 7 and associated discussion that delta-COT is not

statistically significant in October or November. This is mentioned in passing on Pg. 8, Line 28. Yet, there is then extensive discussion on the coincidence between decreased biomass burning, increased liquid cloud fraction and water content in November and a decrease in smoke aerosols in October (Pg. 8, Lines 28-32). I find this discussion very confusing, but much of it appears to be based on source attribution that has already been discussed in this review as being speculative. The ISCCP cloud type classifications does not bring any additional clarity or information to the major flaws in the prior conclusions.

It is not clear what the referee means here. Establishing a non-significant change in a parameter of liquid clouds, does not necessarily exclude the same parameter from changing significantly in a cloud sub-type. Similarly, establishing a non-significant change in a time series does not exclude significance on a monthly basis, as was shown in this study. As the referee mentions in a previous comment, establishing non-significance does not rule out the existence of a significant change that might be uncovered by additional data and/or longer time series.

The part of the discussion mentioned by the referee seems indeed confusing and should be rephrased, since it is not based on source attribution, as the referee claims, but rather on the position of the aerosols relative to clouds. The notion that a non-significant change in COT should prevent an analysis and discussion of changes in biomass burning and smoke aerosols, coinciding with changes in liquid cloud fraction and water content, is also unsupported.

Final response: Based on the revised results, the lack of strong evidence on absorbing aerosols in October renders the explanation proposed for this month rather hypothetical. Additionally, Fig. 9c does not provide substantially additional information for the November case. For these reasons, Fig. 9 was omitted.

Pg. 8, Lines 40 – Pg. 9, Line 1: I fundamentally disagree with second part of this statement. The manuscript is saying that smoke aerosols are from biomass burning and are absorbing, and therefore, an association between smoke aerosol height and cloud height in Figure 8 is somehow related to the semi-direct effect. If the aerosols are misclassified or smoke dominated urban aerosols that are weakly absorbing then this would indeed call the conclusions of the manuscript into question. The missing CALIPSO aerosol types and the compositional ambiguity provided by this typing method make this particular dataset less capable for addressing the types of science questions and drawing the types of conclusions that are sought in this manuscript. This limitation is a significant one that cannot be overcome without new data and new analyses.

The referee keeps repeating the same argument. We acknowledge that CALIPSO does not give unambiguous information about certain aerosol types, in particular biomass burning smoke and urban pollution. However, as stated before, the GFED dataset demonstrates that (absorbing) biomass burning aerosol emissions have markedly decreased over the decade studied. At the same time, anthropogenic emissions have not decreased (see Figure on page 4 of this reply). These pieces of information give strong additional indications that the aerosols are not largely misclassified.

Final response: CALIPSO aerosol subtypes were omitted from the analysis. Hence this statement was also omitted.

Pg. 9, Lines 1-4: Here are the monthly timeseries from Fig. 1 and Fig. 4. I think that it's hard to make the case as is done here that there is a meaningful anti-correlation between polluted dust AOD and liquid CFC only in November and December (circled regions) and that such an anti-correlation can be used to draw a conclusion. There is just too much variability (and that's before the additional requested information of standard deviation error bars or shaded regions are added to the graph). The decreasing pattern in the polluted dust aerosol is present for the smoke aerosol and does occur during other months. Given the scale, it's difficult to discern the trend for the dust trace.

The following table shows the Pearson's coefficients of monthly liquid CFC from CLARA-A2 and MODIS, and AOD from CALIPSO. Results are shown separately for total, dust, smoke and polluted dust AOD.

	Total		Dust		Smoke		Polluted dust	
	CLARA-A2	MODIS	CLARA-A2	MODIS	CLARA-A2	MODIS	CLARA-A2	MODIS
January	-0.21	-0.27	0.26	0.18	-0.23	-0.26	-0.11	-0.17
February	-0.32	-0.23	-0.10	0.05	-0.45	-0.44	-0.15	-0.07
March	-0.42	-0.29	0.01	<0.01	-0.60	-0.53	-0.09	0.07
April	0.02	0.05	0.30	0.35	0.35	0.43	-0.48	-0.51
May	0.49	0.55	-0.13	-0.34	0.39	0.53	0.34	0.36
June	0.15	0.42	-0.06	0.17	-0.07	-0.17	0.11	0.38
July	0.36	0.26	-0.24	-0.53	0.21	0.41	0.47	0.12
August	0.79	0.74	-0.04	-0.15	0.69	0.75	0.41	0.26
September	0.02	0.18	0.18	0.37	-0.31	-0.18	0.08	0.26
October	<0.01	-0.29	0.42	0.01	0.02	-0.32	0.05	-0.15
November	-0.53	-0.50	0.21	0.21	0.08	0.10	-0.73	-0.69
December	-0.83	-0.81	0.22	0.14	-0.69	-0.66	-0.80	-0.79

A quick inspection of the table makes our statement obvious: *the liquid CFC and the polluted dust AOD are anti-correlated in November and December, with correlation coefficients around -0.7 to -0.8.* This statement is true for both CLARA-A2 and MODIS liquid CFC. Inclusion of this table would clarify this issue. However, the referee inadequately uses the plots showing the monthly averages of these variables and falsely calls these plots "monthly time series", to disprove a statement that was not based on those plots. This is an unsettling level of misunderstanding.

Final response: This sentence was omitted due to the updated analysis. Updated results on correlations (and their significance) between aerosol and cloud properties are provided in Section 3.3 of the revised manuscript.

Pg. 9, Line 8-10: No data on aerosol absorption are presented in this study. Some CALIPSO aerosol types are missing, and those that are included cannot be unambiguously attributed to biomass burning emissions. This conclusion is not true.

This same argument has been made many times, and we refer to our reply on page 16.

Final response: This sentence was updated following the updated analysis (page 11, lines 6-9).

Pg. 9, Line 10-11: Technically this statement just says that cloud fraction and thickness both change across months. The changes are neither correlated nor anti-correlated (Figure 4a and 4c). At best, this sentence doesn't reach a meaningful conclusion, and at worst it misleads the reader into thinking that concurrent changes somehow track each other. I recommend that this sentence be removed.

This sentence should indeed be rephrased. It is apparent that the referee is confused and makes an inadequate judgement, since Figure 4a and 4c do not show changes in liquid cloud fraction and optical thickness, but monthly averages of these parameters during the period examined. The sentence refers to changes concurrent with aerosol changes, in different months.

Final response: The statement was rephrased based on the updated analysis (page 11, lines 9-10).

Pg. 9, Line 11-13: No data actually relating the position of aerosols to clouds is presented. Instead, the vertical distribution of cloud extinction and the vertical distribution of aerosol temporal change are presented in Figure 8. These are not the same thing. It is not shown that the sign of cloud changes is determined by the position of aerosol relative to clouds. This statement is not true.

We thank the referee for this remark. Indeed, the data provided actually relates the position of aerosol *changes* relative to clouds, and this statement should read "*Further analysis of vertical profiles of aerosol changes and clouds showed that the signs of cloud changes depended on the position of aerosol changes relative to clouds…"* in order to be true.

Final response: The statement was rephrased according to the referee's correction and the updated results (page 11, lines 10-13).

Pg. 9, Lines 17-18: It is very difficult to relate the trend analysis changes for the aerosol AOD and cloud changes to a process-level causal mechanism. Even comparing the trend changes of aerosol and cloud in this manuscript are difficult because they appear to vary differently across monthly and with different (and poorly explained) levels of statistical significance. It is not true to suggest that the data show a "high level of consistency" with the semi-direct or any ACI effect. This is because the data used here are highly averaged in both space and time, and are, therefore, less ideal for tackling these sorts of science questions.

Acknowledging the limitations of the data and concerns similar to the ones of the referee, we did not attempt to establish any process-level causal mechanism, as was emphatically noted in the statement right after the one cited here by the referee. We also tried to establish consistency in space and time (see Section 2.4) before attempting any comparison of changes. We hope that the levels of statistical significance are better explained now based on our previous replies. However, we consider that suggesting "*a high level of consistency with predictions"* of the semi-direct effect is a modest statement, which takes into account the limits of our analysis. The reason invoked by the referee to dismiss this statement, namely that the data used are highly averaged in both space and time, is indeed prohibitive for quantifying any ACI effect. However, this is not what we do here. And there is no physical reason limiting the *consequences* of any ACI on their process-level space and time scales only.

Final response: no further changes implemented.

Pg. 9, Lines 18-20: This statement succinctly highlights the lack of depth of the analyses in this manuscript. There are a large number of critical limitations associated with the use of these data to try to draw these sorts of conclusions. The limitations are acknowledged in the manuscript, but there is no real attempt to overcome them. The model analyses discussed in this last sentence hold promise for being able to attribute aerosol to sources as well as to link those aerosol to clouds. I recommend that those tools be brought to bear on the questions being tackled here, perhaps with some context being provided by more complete set of aerosol types in the Level 2 version of the satellite data that is discussed here.

It is unfortunate that the acknowledgement of limitations in a data set and/or method of analysis is characterized as "lack of depth", especially when the referee refers to another kind of study and conclusions. Specifically, in the previous comment, the referee reasons that the data used here are "*less ideal for tackling these sorts of science questions"*, because they are "*highly averaged in both space and time"*. Indeed, these data are inadequate to establish an ACI cause and effect mechanism. This is clearly acknowledged as a limitation, but it was never described as a science question to be tackled in this study. Similarly, using level 2 data which are closer to the ACI process-level in terms of both spatial and temporal resolution, and/or model analyses (especially the latter), could probably lead to robust conclusions on aerosol sources and links with clouds, again, in the process level. In a previous comment, the referee invokes the study by McComiskey and Feingold (2012) to justify the inadequacy of the scales used here and a predefined failure to "*see ACI effects*". That study, however, tackles the question of quantifying the ACIs. This was never a goal in the present study, exactly because this limitation was acknowledged. The same holds for the suggestion, by the referee, of a "*more appropriate data set … (e.g., aircraft, balloon, surface remote sensor scales measuring clouds over minutes to hours or a model with better space and time resolution)*".

Final response: no further changes implemented. Model-based emission data were included in the study to provide a more broad overview of possible sources of aerosols.

Satellite observations of aerosols and clouds over southern China from 2006 to 2015: analysis of changes and possible interaction mechanisms

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Abstract. Aerosol and cloud properties over southern China during the 10-year period 2006-2015 are analysed based on observations from passive and active satellite sensors and emission data. The results show a strong decrease in aerosol optical depth over the study area ~~by about 20% on average~~, accompanied by an increase in liquid cloud cover and cloud liquid water path (LWP) ~~by 5% and 13%, respectively~~. Analysis of ~~aerosol types and~~ emissions suggests that ~~the main driver for their reduction is~~ a decrease in biomass burning aerosols played an important role in the overall aerosol reduction. These changes in biomass burning emissions occurred mainly in late autumn and early winter months, leading to a decrease in AOD by about 40% and coincid~~ed~~ing with ~~changes~~ an increase in liquid cloud ~~properties~~fraction by 40% and a near-doubling of LWP in November and December. ~~For the latter, p~~Possible explanatory mechanisms for these changes were examined, including changes in circulation patterns and aerosol-cloud interactions. Further analysis of changes in aerosol vertical profiles demonstrates a consistency of the observed aerosol and cloud changes with the aerosol semi-direct effect, which depends on their relative heights. Based on this mechanism, fewer absorbing aerosols in the cloud layer would lead to an overall decrease in evaporation of cloud droplets, thus increasing cloud LWP and cover.

1 Introduction

The role of atmospheric aerosols in climate change has been studied widely in the past. Their various effects are broadly defined based on their interactions with atmospheric radiation and clouds. The direct effect is described through scattering and absorption of radiation whereas indirect effects describe interactions with clouds, which can lead to changes in both cloud albedo (Twomey, 1977) and cloud lifetime (Albrecht, 1989). The semi-direct effect is a third category that describes aerosol-induced changes in clouds through interaction with radiation. According to the latest terminology (Boucher et al., 2013), the semi-direct effect is described as a "rapid adjustment" induced by aerosol radiative effects, and along with the direct effect it is grouped into the "Aerosol-Radiation Interactions" (ARI) category, whereas the indirect effects are termed "Aerosol-Cloud Interactions" (ACI).

Observations of these mechanisms and their effects on climate have been elusive, and the uncertainties associated with them remain high (Boucher et al., 2013). The main reasons for this lack of substantial progress originate in the high complexity of these phenomena, with multiple possible feedback mechanisms and dependences on various parameters in different regimes (Stevens and Feingold, 2009, Bony et al., 2015). Although there are continuous improvements, the mechanisms related to aerosol and cloud interactions and feedbacks are still inadequately represented in models (Feingold et al., 2016), and poorly captured by remote sensing measurements (Seinfeld et al., 2016). Regarding the latter approach, many studies have highlighted the difficulties and limitations of remote sensing methods, which usually include limitations in spatial and

temporal samplings (Grandey & Stier, 2010; McComiskey & Feingold, 2012). On the other hand, progress is steadily being made, as data sets of aerosols and clouds based on remote sensing retrievals gradually improve. Additionally, independent data sets with complementary characteristics and properties become constantly available, allowing more in-depth analyses of the aerosol and cloud conditions and opening new possibilities for synergistic usage, towards further constraining the effects of aerosols on clouds.

The present study builds on these developments by providing an analysis of aerosol and cloud characteristics and changes in recent years over a climatically important and sensitive area in southern China. This region (20°-25° N, 105°-115° E) was selected, being a densely populated area with intense human activities, ranging from urban and industrial to agricultural, which also constitute different sources of aerosol emissions. Furthermore, significant changes in aerosol loads during the past years over the wider surroundings have previously been reported (e.g. Zhao et al., 2017; Sogacheva et al., 2018), providing the opportunity for an analysis of possible effects on clouds. Hence, the purpose of this study is dual. The primary aim is to analyse aerosol and cloud characteristics and changes during the previous years over southern China. Using multiple data sets, created based on different retrieval approaches, adds robustness to the results. The secondary purpose of this study is to investigate the possibilities and limitations of the synergistic use of this multitude of aerosol and cloud data sets for the assessment of possible aerosol and cloud interaction mechanisms. For this purpose, data sets are analysed in combination, to either help exclude possible explanatory mechanisms, or provide indications of their manifestation.

The study is structured as follows: Section 2 provides a description of the aerosol and cloud data sets used, and the methodology for analysing their changes. Results of this analysis are described in Sections 3 and 4, including time series and seasonal changes in aerosols and clouds, possible effects of large-scale meteorological variability, and indications of possible effects of aerosol changes on corresponding cloud changes. Our findings are summarized in Section 5~~4~~.

2 Data and methodology

2.1 Aerosol and emissions data

Analysis of aerosol changes was based on MODerate resolution Imaging Spectroradiometer (MODIS), Multi-angle Imaging SpectroRadiometer (MISR) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) data. MODIS is a sensor on board NASA's Terra and Aqua polar orbiters, providing aerosol and cloud data products since 2000 and 2002 from Terra and Aqua, respectively. The Aqua MODIS level 3 Collection 6 daily Aerosol Optical Depth (AOD) was used here, available over both land and ocean at 1° × 1° spatial resolution (Levy et al., 2013).

AOD data from MISR were also analysed. MISR flies on board NASA's Terra satellite and acquires measurements at nine viewing angles, providing information on specific aerosol types along with the total aerosol load (Khan & Gaitley, 2015). Here, MISR products of total AOD, along with fine mode AOD and dust (non-spherical) particles AOD were analysed on a monthly basis and at 1° × 1° spatial resolution, available at level 3 of version V23.

The CALIPSO level 3 monthly aerosol profile product was also used ~~along with MODIS~~, to include information on ~~different~~ the aerosol ~~types and their~~ vertical distribution in the analysis. CALIPSO level 3 parameters ~~were~~ are derived from the corresponding instantaneous level 2 version 3 aerosol product (Winker et al., 2009; Omar et al., 2009; Tackett et al., 2018) and include column AOD of total aerosol, ~~dust, smoke and polluted dust,~~ available globally at 2° × 5° latitude/longitude resolution, along with the~~ir~~ extinction profiles at 60 m vertical resolution, up to 12 km altitude. The standard quality filters implemented to ensure the quality of the level 3 product, described in Tackett et al. (2018), were also adopted here. ~~It is worth noting that, while the CALIPSO aerosol models consist of six aerosol types, only three of them are included in the level 3 product. The reason is given in the discussion of the relevant paper (Tackett et al., 2018), where the authors mention that they chose to include a subset of aerosol types for better management of the level 3 file sizes. Nevertheless, this product has been evaluated and used in many studies: for example, it has been compared against AERONET data (Mielonen et al.,~~

~~2009), with agreement on 70% of the daily mean aerosol types; it has also been used for the assessment of aerosol impacts e.g. on solar radiation (Yang et al., 2016), on cloud phase changes (Zhang et al., 2015), and on pollution patterns (Kar et al., 2015).~~

Apart from the ~~characterization~~ analysis of aerosol loads and vertical distributions over the region with MODIS, MISR and CALIPSO data, aerosol sources were investigated using the Global Fire Emissions Database (GFED), which provides information ~~about~~ on trace gas and aerosol emissions from different fire sources on a global scale. Here, version 4 of the data set was used (GFED4s), available at 0.25° × 0.25° spatial resolution and on a monthly basis. GFED emission estimates are based on data of burned areas and active fires, land cover characteristics and plant productivity, and the use of a global biogeochemical model (Van der Werf et al., 2017). Additionally, a recent inventory of anthropogenic emissions of aerosols and precursor gases from the Community Emissions Data System (CEDS, Hoesly et al., 2018) was included in the analysis, to provide a more complete overview of possible origins of AOD changes. It should be noted that, due to the long-range transport of aerosols, local aerosol emissions are not expected to fully explain corresponding properties and characteristics of aerosol types and loads in the atmosphere of the same region. Emission data were rather used here for partially explaining the origin of aerosol types and distributions detected from space. They were also useful as an indicator of local aerosol-producing human activities, with biomass burning being a major source.

2.2 Cloud data

Two independently derived, satellite-based cloud data sets, were used for the analysis of cloud properties and changes over southern China. The Aqua MODIS level 3 Collection 6 daily 1° × 1° product was used (Platnick et al., 2017), as in the case of AOD, for the estimation of monthly averages and corresponding changes in cloud properties, including total and liquid Cloud Fractional Coverage (CFC), in-cloud and all-sky Liquid Water Path (LWP), as well as liquid Cloud Optical Thickness (COT) and Effective Radius (REFF).

The same cloud properties were analyzed using the second edition of the Satellite Application Facility on Climate Monitoring (CM SAF) cLoud, Albedo and surface RAdiation data set from AVHRR data (CLARA-A2), a recently released cloud property data record, created based on Advanced Very High Resolution Radiometer (AVHRR) measurements from NOAA and MetOp satellites (Karlsson et al., 2017). It covers the period from 1982 to 2015 and includes, among other parameters, CFC and cloud phase (liquid/ice), cloud top properties and cloud optical properties, namely COT, REFF and water path, separately for liquid and ice clouds. Orbital drift in NOAA satellites is an important issue regarding the stability of the CLARA-A2 time series, especially in the 80s and 90s. For the 10-year period examined in this study, CLARA-A2 level 3 data, available at 0.25° × 0.25° spatial resolution from AVHRR on NOAA-18 and NOAA-19 were used. Specifically, only the "primary" satellite was used in each month, meaning that when NOAA-19 data became available, NOAA-18 was not used any more. As a result, orbital drifts are minor.~~CLARA-A2 level 3 data, available at 0.25° × 0.25° spatial resolution, from the afternoon satellites NOAA-18 and NOAA-19, were analyzed in the present study.~~

2.3 Uncertainties in aerosol and cloud products

Uncertainties in pixel-based (level 2) data can in many cases be estimated by propagation of error sources through the retrieval algorithms and through validation with collocated independent reference observations. For example, Levy et al. (2013) showed by comparison with Aerosol Robotic Network (AERONET) observations that the MODIS AOD has a 1-sigma uncertainty of about ±(0.05+0.15AOD) over land. However, the propagation of pixel-based error estimates to monthly aggregates is difficult because it needs to separate contributions from systematic and random errors. Similarly, validation at monthly scales is cumbersome, and no level-3 validation results have been reported for the aerosol and cloud data sets used in this study.

Therefore, the use of three independent aerosol data sets and two ~~independent~~ cloud data sets, derived from different sensors is an important element of this study, which suggests that the detected changes reflect actual changes, rather than possible sensor degradations or retrieval artifacts. ~~and~~ This is especially true in the case of aerosol data, which were obtained by ~~with~~ different retrieval approaches. ~~is an important element of this study. The similarity between results from different data sets strongly suggests that the detected changes reflect actual changes in aerosols and clouds, rather than possible sensor degradations or retrieval artifacts.~~

2.4 Analysis of time series and changes

The analysis of all data sets and their changes was based on monthly average values. This temporal resolution is appropriate for studying both long-term interannual as well as seasonal changes. Furthermore, data from afternoon satellites ~~only~~ were mainly used (MODIS Aqua, AVHRR on NOAA-18 and -19 and the daytime product of CALIPSO), to minimize differences due to different temporal samplings. Additionally, due to the different ~~pixel~~ grid cell sizes of the products used, the analysis was based only on area-weighted averaged values over the entire study region, rather than individual ~~pixels~~grid cells. Area-weighted averages were computed based on the cosines of the latitudes of the grid cells covering the study region. However, due to the small size of the domain the ensuing differences were minor. It should be noted that, in the case of GFED, monthly values of emissions over the study area were calculated by summing the corresponding ~~pixel~~ grid cell values, instead of averaging. Additionally, in the case of CALIPSO, spatial averages were weighted by the number of samples used, which is available in the level 3 data.

The quantification of changes during the study period was based on linear regression fits to the spatially averaged deseasonalized monthly time series. Deseasonalization was performed by subtracting from each month the corresponding time series average of this month and then adding the average of all months in the time series. For every aerosol and cloud variable X studied, the change ΔX was calculated as $\Delta X = X_f - X_i$, where X_i and X_f are the initial and final monthly values of the regression line. The corresponding percent change was estimated as $\Delta X = 100(X_f - X_i)/ X_i$.

Spatial and temporal representativeness of the study area and time period in the change analysis were ensured by applying thresholds to both the area covered with valid data and the number of months used in the calculations. Specifically, the following thresholds were applied: a) on a ~~pixel~~ grid cell basis, a monthly average value was used only if it was computed from at least 18 daily values (10 daily values for AOD, due to sparsity of data). Application of this threshold requires the number of days used in the calculation of the monthly average. This information ~~is~~ was available in all data sets used, except for MISR; b) a spatially averaged value was used if it was computed from at least 50% of the ~~pixels~~ grid cells in the study area; c) it was required that at least 80% of monthly averages are present in the time series, for the corresponding 10-year changes to be estimated. Further analysis included a per month estimation of changes, in order to assess their seasonal variation. In this case, no deseasonalization was applied. Statistical significance of all calculated changes was estimated using the two-sided t-test.

3 Results

3.1 Aerosol characteristics and changes

Aerosol sources in southern China include biomass burning activities, such as residential biofuel consumption, crop residues burning, firewood consumption and agricultural waste open burnings (Chen et al., 2017). These sources exhibit different seasonal characteristics and relative contributions to the total aerosol load. Higher emissions of domestic biomass burning occur in autumn and winter, specifically November to March (He et al., 2011), while agricultural field fires are mostly observed after harvesting seasons, when rice and wheat straw field burning takes place, typically in late May and October

(Zha, 2013; Chen et al., 2017). Domestic burning is the major contributor, reaching over 60% of the total biomass burning emissions (He et al., 2011).

Figure 1 shows the seasonal variation of emissions from GFED and AOD from MODIS, MISR and CALIPSO over southern China, based on data during 2006-2015. The seasonal variation of carbon emitted from biomass burning over the region shows that the highest emissions occur between November and April (Fig. 1a). This seasonal pattern in biomass burning carbon emissions is in good agreement with the seasonal variation of biomass burning activities described before, verifying the high contribution of domestic fuelwood burning during the same months. MODIS, MISR and CALIPSO total AOD (Fig. 1b) are in relatively good agreement in most months, with the largest differences occurring in March and April, when CALIPSO deviates from the other two data sets. While the present analysis was designed to minimize discrepancies due to differences in spatial and temporal resolutions, as described in Section 2.3, some disagreement between ~~MODIS and~~ CALIPSO and the passive sensors should be expected, considering their differences in areas sampled, overpass times and retrieval methodologies. While it was not possible to pinpoint specific reasons for the March-April differences based on the data sets used here, this feature deserves further investigation. Based on ~~CALIPSO~~MISR, which offers additional information on aerosol types, ~~smoke and polluted dust have similar~~the fine mode AOD follows a seasonal pattern similar to the total AOD, and appears to constitute a large part of the latter. This highlights the important role that anthropogenic emissions (including biomass burning) play in the overall aerosol load over the region.~~s,~~ ~~while~~ On the other hand, the contribution of dust is minimal, with a small peak in spring. ~~According to the CALIPSO classification, smoke aerosols originate in biomass burning activities, and polluted dust aerosols are a mixture of dust with biomass burning smoke (Omar et al., 2009).~~ Biomass burning emissions (Fig. 1a) and satellite-based AOD (Fig. 1b) are not ~~directly comparable~~expected to always agree, since the former contributes to only part of the latter. ~~Apart from a~~Additional aerosol sources that contribute to the total AOD and are not represented in GFED include mostly scattering aerosols from anthropogenic sources such as industry and transportation. ~~,~~ Furthermore, transportation of aerosols from neighbouring regions can also cause large differences.

~~In fact, high smoke AOD values combined with low emissions (e.g. in September and October), suggest that these aerosols were transported to the study region from different areas.~~

Figure 2 shows the changes in AOD over the southern China region during the 10-year period examined, both on a ~~pixel~~ grid cell basis from MODIS (Fig. 2a) and as spatially averaged time series from MODIS, CALIPSO and MISR (Figs. 2b, 2c and 2~~d~~e). The ~~pixel~~grid cell-based changes in AOD (Fig. 2a)~~, deduced from MODIS level 3 data,~~ reveal an almost uniform reduction throughout the area, with stronger decreases over land. The time series of the deseasonalized spatially averaged monthly values of the AOD, separately from MODIS, ~~and~~ CALIPSO and MISR, are shown in Figs. 2b, 2c and 2~~d~~e, along with their linear regression fits and corresponding changes (in percent). The reduction in total AOD during the 10-year period is apparent and statistically significant in the 95% confidence interval in ~~both MODIS and CALIPSO~~all three data sets. The levels of statistical significance on a grid cell basis, corresponding to Fig. 2a, are shown for MODIS AOD in supplementary Fig. S1. Similar results in terms of both spatial distribution and statistical significance of changes were acquired from the analysis of MISR total and fine mode AOD (Fig. S2). Table S3 provides additional information on the time series analysis, i.e. slopes and p-values. ~~Based on the CALIPSO aerosol types classification, this decrease can be attributed to corresponding reductions in polluted dust and smoke aerosols.~~ The reduction in AOD reported here is in agreement with changes over the same region or wider Chinese regions during recent years, reported based on different satellite sensors, e.g. MODIS (He et al., 2016), MODIS and AATSR (Sogacheva et al., 2018) and MODIS and MISR (Zhao et al., 2017).

The seasonality variability of aerosols over the study region (Fig. 1) suggests that their changes could also exhibit seasonal variations. Hence, the time series changes were further analyzed in terms of their seasonal variability. Results for both AOD and emissions are shown in Fig. 3. For AOD (Fig. 3a), the main decrease occurs in autumn and early winter. ~~MODIS (Fig.~~

3a) and CALIPSO All three data sets (Fig. 3b) agree well in this seasonal pattern. Based on CALIPSOMISR, this decrease is driven by biomass burningfine mode aerosols, while: as for the full time series (Fig. 2c), dust aerosols show no significant change. The same analysis of the total mass of carbon particles (C) from local fire emissions (Fig. 3c) shows that the largest decrease in emitted particles occurs during late autumn to early spring, with a minimum in November, suggesting that this decrease cshould be attributed to changes in residential energy sources. This stems from the finding by He et al. (2011), that this activity dominates biomass burning emissions during this period., which peak during the same period. This explanation is also consistent with previous studies, which report a diminishing contribution of residential biomass burning, starting already in the 1990s (Qin and Xie, 2011; 2012; Streets et al., 2008), mainly through a replacement of fuelwood by electricity (Yevich and Logan, 2003). Furthermore, a direct comparison of changes in satellite-based AOD and surface emissions offers additional insights into the origins of these changes: the seasonal variation of changes in C emissions partially agrees with the total AOD change pattern, while this agreement improves in the case of polluted dust. These results suggest that part of the aerosol load over the study area (especially smoke aerosols) is transported from neighboring regions, as was also inferred from the differences in seasonality patterns (Fig. 1)., e.g. from November to January, when both decrease. In such cases, AOD and local emissions do not agree well (e.g. smoke aerosols in October). Forest fires and biomass burning activities in Indochina could be such sources. In November, on the other hand,

This coincidence suggests that large part of the AOD aerosol load during these months probably originates mainly from local biomass burning sources, leading to a coincidence in AOD and fire emission reductions. Further support to this hypothesis is given by the CEDS data, which indicate that other anthropogenic emissions of black and organic carbon as well as nitrate and sulfate precursor gases may be of comparable magnitude but are not expected to have strong intra-annual variations (Fig. S4). Furthermore, these emissions showed increases rather that decreases over southern China in the period 2006-2014 (Fig. S5), and thus cannot explain the observed decrease in AOD. This agreement between emissions and the polluted dust aerosol type from CALIPSO (Figs. 3b and 3c) also suggests that aerosols of biomass burning origin, which contribute to this type (Omar et al., 2009), may play a leading role in its overall characteristics. It should be noted here that misclassification of aerosol type in the CALIPSO data set does occur. In particular, smoke aerosols may be confused with other small aerosol types such as urban pollution (Burton et al., 2013). The best agreement in classification comparisons with AERONET was found for polluted dust and desert dust aerosols (Mielonen et al., 2009). Hence, some ambiguity probably exists regarding the origin of the aerosols, especially for the smoke aerosol type.

3.2 Cloud characteristics and changes

The seasonality of main cloud properties over the study region, comprising total and liquid cloud cover, and optical thickness and effective radius for liquid clouds, is shown in Fig. 4. While the total cloud cover does not exhibit strong seasonal characteristics (Fig. 4a), varying between 0.7 and 0.8 throughout the year (based on CLARA-A2 and MODIS, respectively), liquid clouds appear to prevail from late autumn to early spring (Fig. 4b). A similar seasonal pattern appears in liquid COT, which is not necessarily related to the variation in the extent of liquid clouds. Liquid REFF ranges between 10 μm and 14 μm throughout the year. The LWP, which is proportional to the product of liquid COT and REFF, also varies seasonally, with higher values in winter (not shown here). The main driving factor for the seasonality in total and liquid cloud cover is the Asian Monsoon (AM). The monsoon season in summer is characterized by a larger fraction of high clouds with ice near the top, in particular convective clouds. In winter, low stratus/stratocumulus clouds prevail., which Overall, there areleads to more clouds in summer compared to winter, but more liquid clouds in winter (Pan et al., 2015). The prevalence of low, liquid clouds in winter, which are mostly single-layer clouds, is also verified based on CALIPSO data (Cai et al., 2017). On the other hand, in summer higher ice clouds, constituting about half of the CFC, probably shield a considerable amount of low liquid clouds.

Figure 5 shows ~~pixel~~ grid cell based and spatially averaged changes in cloud properties over southern China during the period examined. The all-sky LWP and liquid CFC have increased over most parts of the land and significantly in most cases (Figs. 5a and 5b, with corresponding maps of statistical significance levels given in Fig. S6). In fact, Fig. 5 shows increases in all liquid cloud properties, with the largest increase found for the total liquid water content present in clouds (12%-14%). Liquid COT changes appear similar to those of LWP, with very good agreement between the two data sets (CLARA-A2 and MODIS), while liquid REFF changes are also positive but more ambiguous. Cloud changes appear statistically significant at the 95% level over large areas of the study region, especially over land, when studied on a ~~pixel~~ grid cell basis. Analysis of spatially averaged values, however, over the entire (5° × 10°) study region, reduces this significance to levels below 95% in most cases of Fig. 5 (see also Table S3). Overall, MODIS and CLARA-A2 are in good agreement and consistent in terms of the changes reported, with biases of around 10% appearing for liquid CFC (Fig. 5d) and REFF (Fig. 5f).

The long time range available from CLARA-A2 data (34 years, starting in 1982) offers the opportunity for further evaluation of the cloud properties changes reported before, especially with respect to changes during the past three decades. For this purpose, changes from all possible time ranges, at least 10 years long and starting from 1982 onward, were estimated for the study region. Results, shown in Fig. 6, suggest that the ranges of changes reported in Fig. 5 are not typical of the entire 34-year CLARA-A2 period. Specifically, for LWP, liquid CFC and liquid COT, the largest increases occur when the time range examined ends within the last five years of the CLARA-A2 period (2011-2015), indicating that corresponding values reached maxima during these years. Furthermore, for liquid REFF, a switch in the sign of change appears in the last years: while liquid REFF is mainly decreasing for most start and end year combinations, only positive changes appear after 2003, indicating a consistent increase during the last years. It should be noted that abrupt changes appearing in the plots of Fig. 6 should be attributed to artifacts especially in the early years of the CLARA-A2 data record. Specifically, negative changes in liquid CFC occurring for starting years between 1988 and 1994 coincide with the period when AVHRR on NOAA-11 was operational, which caused a small discontinuity in the time series. Additionally, the switch from channel 3b (at 3.7 μm) to channel 3a (at 1.6 μm) on NOAA-16 AVHRR during 2001-2003 caused a discontinuity in the cloud property time series, most prominently visible for REFF. A similar, long time range analysis of aerosols was not possible, due to the lack of available aerosol data.

As for aerosols, the seasonality of cloud property changes was also analyzed. Figure 7 shows that the overall increase in liquid clouds during the 10-year period examined can be attributed to changes occurring mainly in November and December. In fact, the patterns of seasonal changes show that CLARA-A2 and MODIS agree very well, with an increase in LWP occurring primarily in December and secondarily in November (Fig. 7a), and liquid CFC increases prevailing also in November and December (Fig. 7b). Corresponding results for liquid COT and liquid REFF (Figs. 7c and 7d) indicate the similarity in change patterns between COT and LWP, and the ambiguity in the REFF change between CLARA-A2 and MODIS, especially in November. The liquid CFC change is statistically significant in the November case, while all other cloud property changes shown in Fig. 7 are significant in December. Corresponding levels of significance for all cloud properties and months examined, for both CLARA-A2 and MODIS, are provided in Table S7.

3.3 Summary of aerosol and cloud seasonal changes

The results presented in the previous section show that during the 10-year study period, monthly changes in cloud properties and GFED emissions occurred almost exclusively in November and December (Figs. 3b and 7), while AOD changes also occurred in earlier autumn months (Fig. 3a). To add robustness to our findings, and realizing that averaging over full seasons will dilute the results too much, we have further aggregated the aerosol and cloud parameters to two-month periods. Table 1 summarizes the changes in GFED emissions, AOD and liquid clouds on a bimonthly basis, with statistically significant changes highlighted in bold. This analysis makes clear that the period September-December drove the AOD changes found in the 10-year period examined, with significant decreases by about 40%, while GFED emissions only changed significantly

in November-December. As mentioned before, liquid cloud changes occurred mainly in November and December, with liquid CFC increasing by around 40% and LWP almost doubling. Hence, there is a concurrence of substantial aerosol and cloud variations in late autumn and early winter.

Further statistical analysis showed that there is indeed a strong, statistically significant anti-correlation between GFED emissions and AOD, on one side, and liquid cloud CFC and LWP, on the other. Results for all possible combinations examined are shown in Table 2, with statistically significant correlation coefficients in the 95% confidence interval highlighted in bold. These results reveal a persistent anti-correlation, independently from the aerosol or cloud data sets used. The same analysis was performed for the entire seasonal cycle, showing that, apart from some spurious cases, significant correlations occur consistently only in November-December (Table S8).

An important question is which mechanisms could explain the concurrent variation of aerosol and cloud properties. A first possibility is that large-scale meteorological variability affects both aerosols and clouds simultaneously. Secondly, local-scale ACI and/or ARI mechanisms would lead to cloud changes as a result of aerosol changes. A combination of these factors should not be excluded either. A second question arising from the previous results, is why significant cloud changes occur in November-December only, while aerosols change significantly also in September-October (Table 1). We attempt to address these questions in the following section.

~~3.~~4~~3~~ Discussion

~~3.~~4~~3~~.1 Possible effects of meteorological variability and large-scale phenomena

~~The results presented in the previous section show that during the study period, aerosols decreased over southern China particularly in autumn and early winter, while liquid clouds increased mainly in late autumn and early winter. Hence, there is a concurrence of substantial aerosol and cloud changes during the same months, namely in late autumn and early winter. There are two major mechanisms that could lead to this concurrence: large-scale meteorological variability could affect both aerosols and clouds simultaneously, while local-scale ACI and/or ARI mechanisms, would lead to cloud changes due to corresponding aerosol changes. A combination of these two factors should also not be excluded.~~

In order to analyse meteorological variability, namely changes in atmospheric circulation patterns and their possible role in the changes reported before, we used surface pressure and 500 hPa geopotential height fields from the Copernicus Atmospheric Monitoring Service (CAMS) reanalysis data record (Flemming et al., 2015; 2017). Similarly to the aerosol and cloud properties, the analysis was based on deseasonalized linear regressions of the entire time series of monthly averages, as well as changes on a monthly basis, focusing especially on months when aerosol and cloud changes maximize (i.e. November-December). For this analysis, however, the study area was extended by 10° in every direction, to include large-scale patterns that could be affecting the southern China region.

The analysis showed 500 hPa geopotential height changes at the ~~pixel~~ grid cell level in the order of several meters and surface pressure changes up to a few hPa, none of which were statistically significant, when either the entire time series or specific months were examined. These results suggest that meteorological variability is not among the major factors contributing to the aerosol and cloud changes reported.

Changes in atmospheric circulation could also be related to larger scale phenomena affecting the wider South-East Asia region, namely the El Nino Southern Oscillation (ENSO) and Asian Monsoon (AM) cycles. Regarding possible effects of ENSO over southern China, the Oceanic Nino Index (ONI) was used to examine possible correlations between ENSO and the aerosol and cloud properties analysed here. ONI is the National Oceanic and Atmospheric Administration (NOAA) primary indicator for measuring ENSO; it is defined as the 3-month running Sea Surface Temperature (SST) anomaly in the Nino 3.4 region, based on a set of improved homogeneous SST analyses (Huang et al., 2017). This analysis showed no particular correlation between ONI and cloud or aerosol properties; Correlation coefficients were around -0.2 for the entire

time series and slightly larger for specific months. A very similar, not significant, anti-correlation between ENSO and low cloud amount was found by Liu et al. (2016), examining the entire China and the period 1951-2014.

The overall effects of AM on the area are most pronounced in summer. Although AM is known to affect aerosol concentrations (through wet deposition during the raining season) and cloud cover, this seasonality pattern does not coincide temporally with the seasonal aerosol and cloud changes reported here. Furthermore, it is known that AM and ENSO are strongly correlated (Li et al. 2016), hence the effects of the former on these changes are expected to be similarly insignificant with those of the latter.

~~3.~~4~~3~~.2 Possible effects of ACIs and ARIs

Although cause and effect mechanisms cannot be proven based on observations only, possible underlying ACI and ARI mechanisms are worth investigating, since the combination of aerosol and cloud changes can also be used to exclude some of them.

Following this approach, our results appear inconsistent with the standard definitions of the first and second aerosol indirect effects, although the possibility of multiple mechanisms occurring simultaneously cannot be excluded. Specifically, according to the first aerosol indirect effect, a decrease in aerosols would lead to an increase in cloud droplet size, under constant liquid water content. In our case, while both CLARA-A2 and MODIS indicate an overall increase in liquid REFF (Fig. 5f), these changes do not coincide seasonally with any significant aerosol change~~, which occurred mainly in autumn~~ (Fig. 3). In fact, mixed signs in liquid REFF change were observed in November (Fig. 7d). Additionally, the LWP increases considerably, suggesting that the first indirect effect mechanism does not play a major role. Furthermore, the already high aerosol loads over the region in the recent past may have led to a saturation in the role of cloud condensation nuclei (CCN) to droplet formation. According to the second aerosol indirect effect, a decrease in aerosols implies reduced cloud life time through more rapid precipitation. However, the increase in observed cloud fraction suggests increased cloud life time, which is contrary to this mechanism.

Contrary to the first and second aerosol indirect effects, the semi-direct effect cannot be excluded as an explanatory process, since the signs of changes of all aerosol and cloud variables presented here are consistent with what would be expected based on this mechanism. Specifically, this effect predicts that decreasing absorbing aerosols inside the cloud layers would lead to reduced evaporation of cloud droplets and hence increased cloudiness and cloud water content. It is important noting that this mechanism holds primarily for absorbing aerosols, such as biomass burning particles, ~~which is the case in this study~~while aerosols from air pollution can also be absorbing. Based on the GFED emissions analysed here, there are strong indications that at least in the November-December case, aerosol changes refer mainly to absorbing aerosols. It is also important noting that the position of the aerosols relative to the cloud layer determines the sign of the semi-direct effect: a decrease in aerosols will lead to increased cloudiness only if the aerosols are at the same level with clouds. If the aerosols are above clouds, the effect will be the opposite (Koch and Del Genio, 2010).

~~3.~~4~~3~~.3 Profiles of aerosol and cloud changes

In order to further examine the possibility of the semi-direct effect as an underlying mechanism, an analysis of the vertically resolved changes in aerosol extinction profiles was conducted, based on CALIPSO data, combined with typical values of cloud extinction profiles for this region. September-October and November-December were selected~~ as the most characteristic months in terms of aerosol changes~~, since ~~both~~ they exhibit a significant decrease in aerosols, with the main difference being that in November-December GFED changes suggest that a decrease in biomass burning emissions contributed to the corresponding decrease in aerosols~~which however is attributed to different aerosol types, namely smoke aerosols for October and polluted dust for November~~ (Table 1 and Fig. 3b). Additionally, November and December are the months when cloud changes were prominent. Figure 8a shows the typical profile of cloud extinction in autumn over southern

China, available from the LIVAS data set (Lidar climatology of Vertical Aerosol Structure for space-based lidar simulation studies; Amiridis et al., 2015) based on measurements from 2007 to 2011. It is apparent that low clouds prevail during this season. Figures 8b and 8c show, for the same height range, changes in the aerosol extinction profiles in September-October and November-December during 20076-2015~~, separately for total, dust, smoke and polluted dust aerosols~~. In September-October, changes ~~in smoke prevail, in agreement with Fig. 3b~~occurred mainly at an elevated altitude. When compared with the cloud extinction profile, it appears that the decrease in ~~smoke~~ aerosols tended to occur mostly above clouds. In November-December, however, ~~polluted dust is the leading decreasing aerosol type (see also Fig. 3b) and this~~the decrease was more pronounced towards the surface. In fact, the shape of the profile change suggests that most of the November-December decrease occurred near or within clouds. The aerosol profile change in November-December is also consistent with our previous conclusion on the local origin of aerosols, based on Fig. 3b. A decrease in aerosols from local sources is expected to be proportional to their typical profile (higher concentrations at lower atmospheric levels). It should be noted here, that the uncertainty in aerosol extinction profiles retrieval from CALIPSO increases in lower atmospheric layers (Young et al., 2013), thus decreasing the confidence in the results towards the surface. The vertically resolved analysis of aerosol ~~type~~ changes showed that the significance level ~~for smoke~~ in September-October (Fig. 8b) exceeds ~~90% and~~ 95% between 1.3 km and 2~~1~~.5 km altitude, while ~~polluted dust~~ changes in November-December are significant between 0.6~~7~~ km-1.0 km and 2~~1~~.0-2.5~~2~~ km.

~~Analysis of MODIS level 3 data for these same months, classified according to the International Satellite Cloud Climatology Project (ISCCP) cloud type classification scheme, shows that stratocumulus clouds prevail over the region during this season. While this is shown in Fig. 9a on an autumn average basis, the pattern is similar if October and November are examined separately. Stratocumulus clouds also exhibit the largest changes, with a considerable decrease in October (Fig. 9b) and increase in November (Fig. 9c), although not in a statistically significant sense. Nevertheless,~~ These results show consistency with an aerosol semi-direct effect mechanism acting under decreasing aerosol loads in the November-December case. Specifically, the decrease in biomass burning aerosols within clouds ~~(November case)~~in these months coincides with an increase in liquid cloud fraction and water content in low liquid clouds (Figs. 7a, 7b), with a significant anti-correlation (Table 2). ~~, attributed primarily to stratocumulus clouds (Fig. 9c), while t~~The decrease in ~~smoke~~ aerosols above clouds (September-October case), on the other hand, ~~coincides with a considerable decrease in stratocumulus CFC (Fig. 79b)~~has no coincidence with any significant cloud change. A possible explanation for this difference between the two periods examined is that in September and October aerosols are not strongly absorbing, compared to the November-December case. The lack of any significant change in GFED emissions during these two months supports this conclusion. In ~~both~~ these ~~months~~ November-December case, however, the positions of aerosols and clouds and their signs of changes agree well with the ~~different~~ semi-direct effect mechanism prediction:~~s~~ ~~reported for different aerosol-cloud configurations: fewer absorbing aerosols above stratocumulus clouds would lead to a decrease in CFC due to a weakened inversion and enhanced cloud-top entrainment (as in October), while~~ fewer absorbing aerosols within clouds would lead to more and thicker clouds, by reducing cloud evaporation. ~~(as in November).~~

~~The results on aerosol profile changes also agree with our previous conclusions on the origins of aerosols, based on Figs. 3b and 3c. A decrease in smoke aerosols that are transported from remote areas (as in the October case) would probably occur at higher altitudes, whereas a decrease in aerosols from local sources (as in polluted dust in November) is expected to be proportional to their typical profile (higher concentrations at lower atmospheric levels). It should be noted here, as was also mentioned in Section 3.1, that possible misclassifications in CALIPSO aerosol types add ambiguity to our conclusions regarding the origin of these aerosols loads, especially smoke aerosols. They would not affect, however, our findings regarding possible interaction mechanisms. Further analysis of the monthly time series showed that the liquid CFC and the polluted dust AOD are anti-correlated in November and December, with correlation coefficients around -0.7 to -0.8. This~~

~~anti-correlation is not apparent in other months, and the decreasing pattern in the polluted dust profile in November is not present for the other aerosol types or months.~~

5~~4~~ Summary

In the present study, aerosol and cloud characteristics and changes were analysed based on a synergistic use of multiple independent remote sensing data sets. The study focused on the southern China region, which is characterised by intense aerosol-producing human activities, while a significant decrease in aerosol loads has previously been reported. In agreement to these previous reports, it was found that ~~absorbing~~ aerosol loads over the region decreased significantly in autumn and early winter months, and this decrease ~~was attributed mainly to~~coincided with large ~~changes~~ decreases in biomass burning emissions in November and December~~ activities~~. Concurrent changes in liquid cloud fraction and ~~thickness~~ water path were observed in these two months, with notable increases ~~and decreases~~ in ~~different month~~boths. Further analysis of vertical profile observations showed that~~ the signs of cloud changes depended on~~ the decrease in aerosol loads occurred at low elevations, where the liquid clouds are typically position~~ed. It was concluded that the observed aerosol and cloud changes are~~, ~~being~~ in agreement with the predictions of the aerosol semi-direct effect, ~~under different aerosol and cloud configurations~~ by which less absorbing aerosols residing in liquid clouds lead to a reduction in cloud evaporation and a corresponding increase in cloud cover and LWP. In the months September and October the decrease in AOD occurred at higher elevations and could not be related to a decrease in local biomass burning emissions. In line with this, a similar cloud response like in November and December was not observed.

The aerosol semi-direct effect has been studied in the past through both model simulations (e.g. Allen and Sherwood, 2010; Ghan et al., 2012) and analysis of observations (e.g. Wilcox, 2012; Amiri-Farahani et al., 2017). While its magnitude on a global average scale appears less pronounced compared to indirect aerosol effects, it has been shown that on local scales and in specific aerosol-cloud regimes its consequences can be significant. Here, the combined analysis of different aerosol and cloud data sets showed a high level of consistency with predictions of this mechanism. It should be stressed however, that apart from strong indications, these results do not constitute evidence of any cause and effect mechanism, which cannot be proved based on observations only. They rather represent a contribution to the observational approaches in aerosol-cloud-radiation interaction studies, highlighting both the possibilities and limitations of these approaches. To overcome some of these limitations, further research should focus on model simulations of the conditions described here, in order to provide more insights regarding the underlying physical mechanism.

Author contributions

N.B. and J.F.M. developed the methodology and performed the analysis. All authors contributed in interpreting the results, writing, editing and finalizing the manuscript.

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Table 1. Relative change (in %) of two-monthly emission, aerosol and cloud parameters over southern China during the period 2006-2015 (2007-2015 for CALIPSO AOD). Significant changes are indicated with boldface.

parameter	Jan+Feb	Mar+Apr	May+Jun	Jul+Aug	Sep+Oct	Nov+Dec
GFED carbon emissions	-58	12	-10	56	60	**-99**
CALIPSO total AOD	-2	-14	-11	-12	**-42**	-34
MODIS total AOD	-10	10	0	-24	**-38**	**-35**
MISR total AOD	-8	7	3	-20	**-39**	**-35**
MISR fine mode AOD	-11	2	3	-19	**-40**	**-41**
CLARA liquid CFC	-3	-1	-1	-3	-3	35
MODIS liquid CFC	-1	1	0	2	-5	**42**
CLARA all-sky LWP	-1	-4	-20	3	17	**92**
MODIS all-sky LWP	-4	-7	-23	18	22	**80**

Table 2. Linear correlation coefficients of November-December-mean emission and AOD time series with cloud property time series over southern China during the period 2006-2015 (2007-2015 for CALIPSO AOD). Significant correlations are indicated with boldface.

parameter	CLARA liquid CFC	MODIS liquid CFC	CLARA all-sky LWP	MODIS all-sky LWP
GFED carbon emissions	-0.51	-0.51	**-0.69**	**-0.75**
CALIPSO total AOD	**-0.77**	**-0.75**	**-0.69**	**-0.71**
MODIS total AOD	**-0.76**	**-0.81**	**-0.75**	**-0.84**
MISR total AOD	**-0.66**	**-0.74**	**-0.66**	**-0.81**
MISR fine mode AOD	**-0.66**	**-0.74**	**-0.70**	**-0.84**

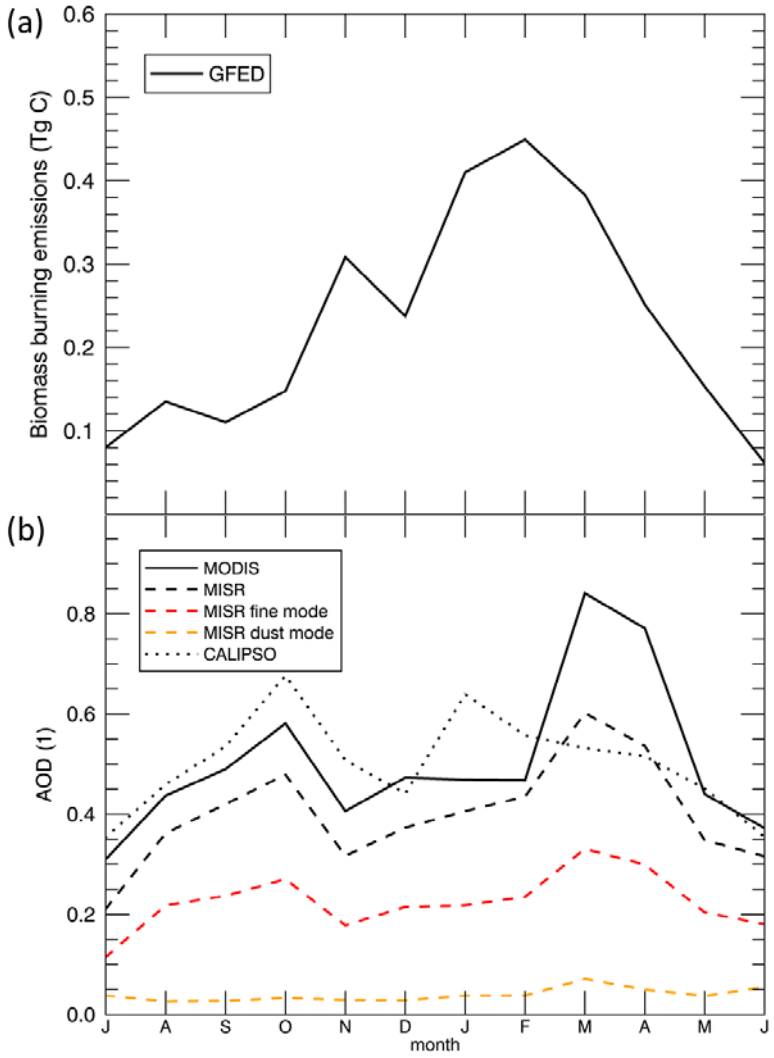

Figure 1. Seasonal variations in biomass burning emissions and aerosols over southern China, based on the period 2006-2015. (a) GFED biomass burning emissions (Tg C), (b) AOD from MODIS, MISR and CALIPSO, including ~~CALIPSO~~ MISR ~~components of dust, smoke and polluted~~fine and dust mode AOD. Note that the horizontal axis starts in July and ends in June.

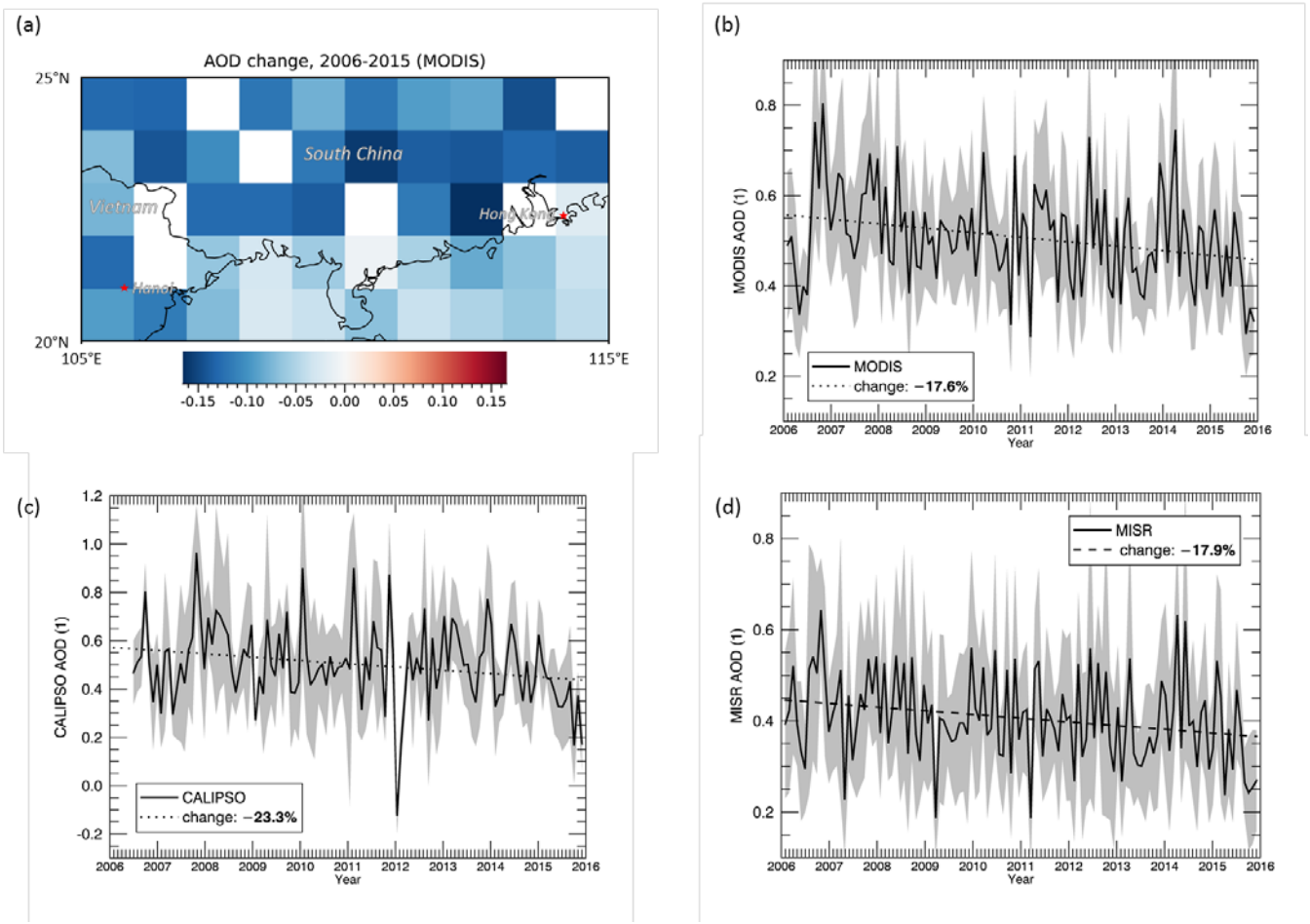

Figure 2. Changes in AOD over southern China during 2006-2015. (a) Spatial distribution of AOD change over the study region deduced from MODIS data. Spatially averaged monthly deseasonalized values of AOD from MODIS (b), ~~and~~ CALIPSO (c), and MISR (d) ~~including components of dust, smoke and polluted dust AOD (c)~~. Shaded areas correspond to one standard deviation of the grid-scale monthly averages. Dotted lines correspond to linear regression fits. Percent changes during the period examined are also shown, with the statistically significant ones indicated in bold.

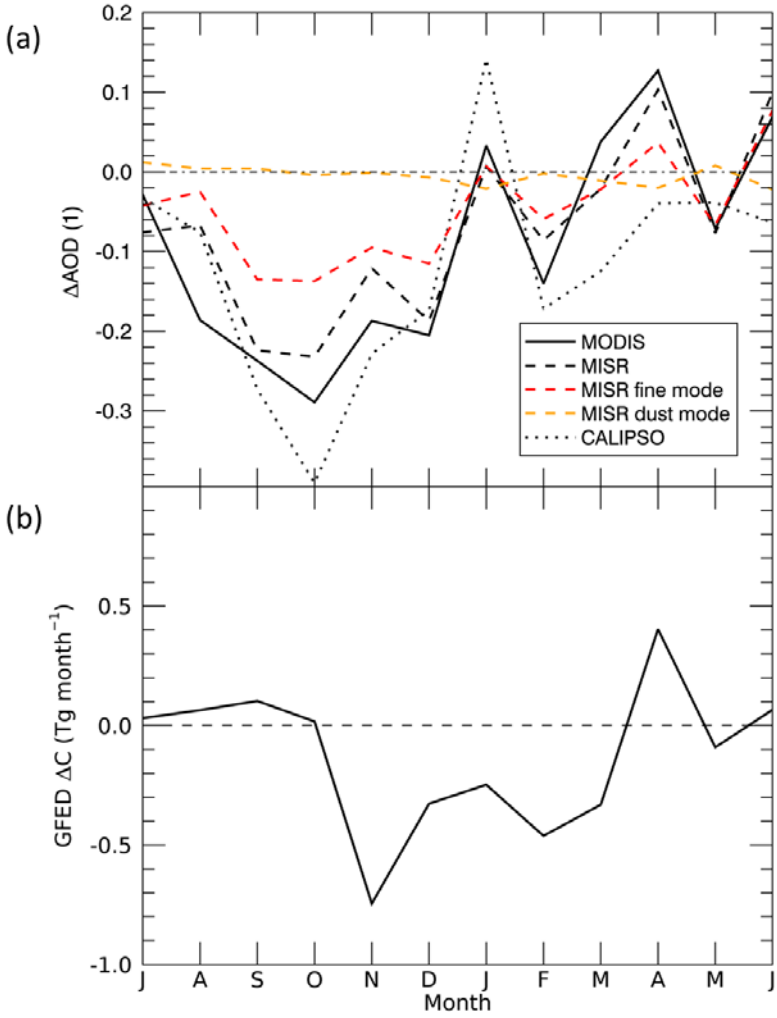

Figure 3. Seasonal variation of changes in aerosols and emissions over southern China. (a) AOD changes from 2006 to 2015 deduced from MODIS, MISR and CALIPSO data. (b) ~~AOD changes from 2007 to 2015 deduced from CALIPSO data, in total and per aerosol type. (c)~~ Biomass burning aerosol emission changes from 2006 to 2015 based on GFED data.

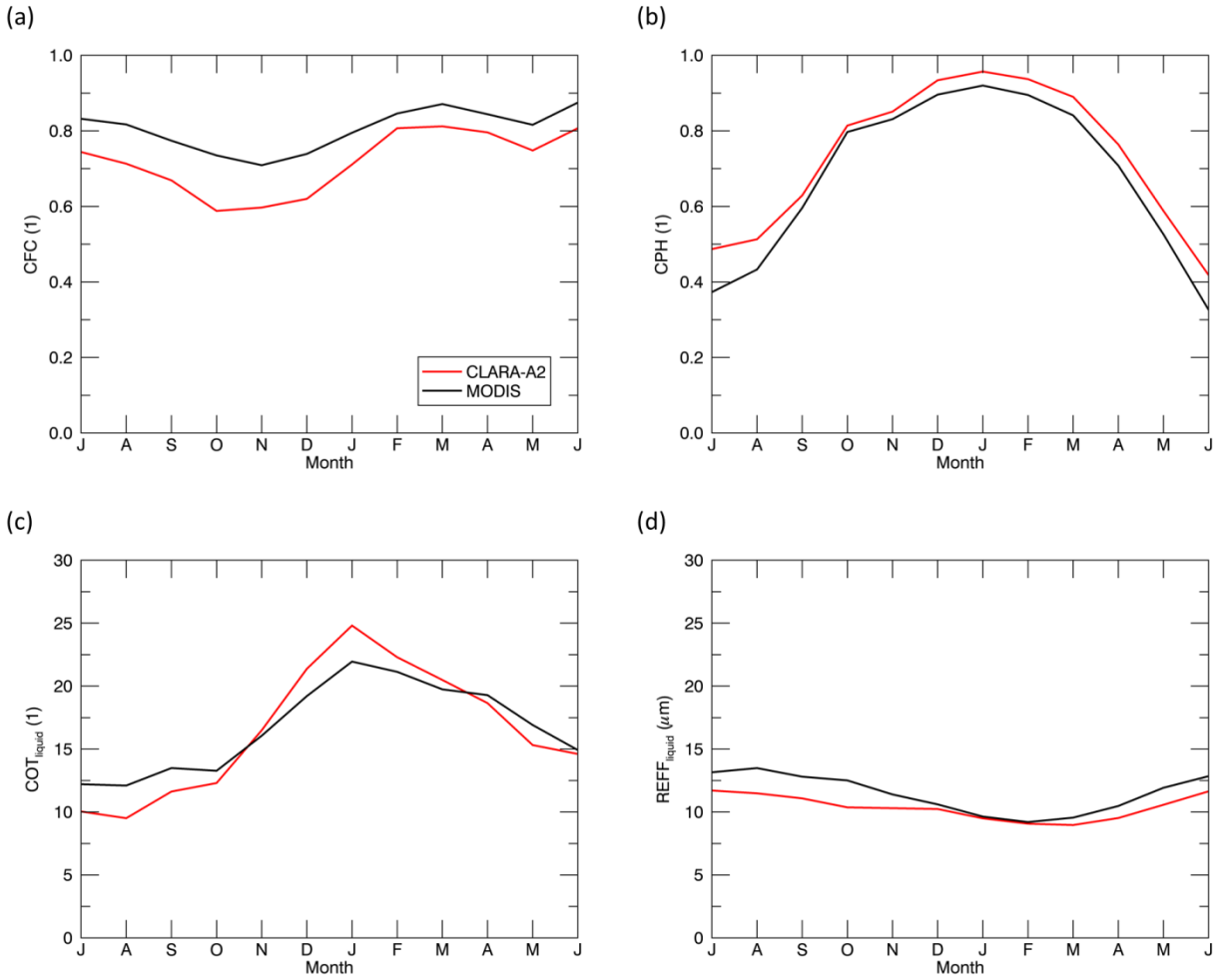

Figure 4. Seasonal variations in cloud properties over southern China, based on CLARA-A2 and MODIS data, during the period 2006-2015. (a) Total CFC, (b) cloud phase (CPH; fraction of liquid clouds relative to total CFC), (c) COT for liquid clouds and (d) REFF.

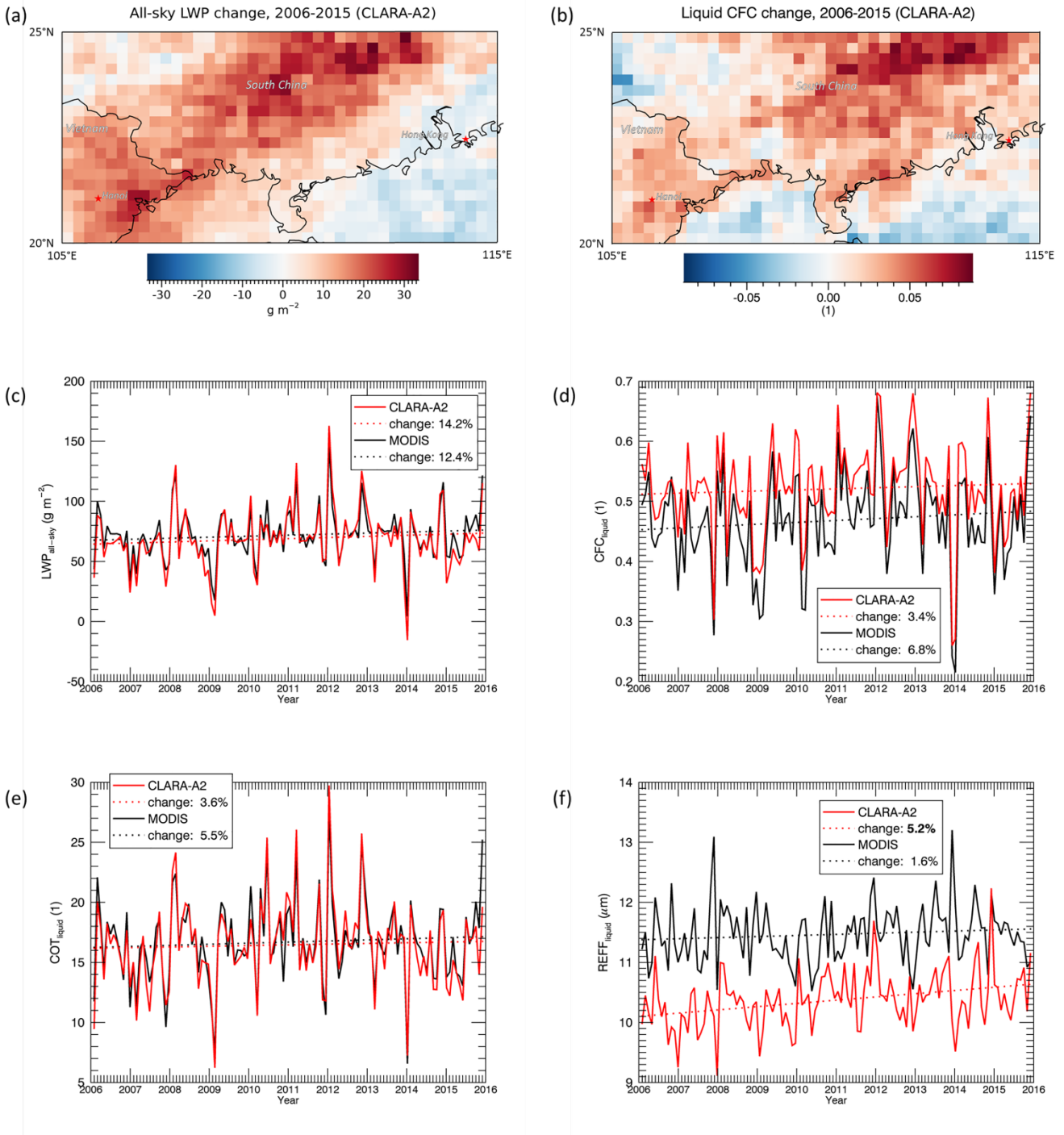

Figure 5. Changes in cloud properties over southern China during 2006-2015, based on CLARA-A2 and MODIS data. (a), (b) Spatial distributions of changes in all-sky LWP and liquid CFC based on CLARA-A2 data. Spatially averaged monthly deseasonalized values of all-sky LWP (c), liquid CFC (d), liquid COT (e) and REFF (f). Percent changes during the period examined are also shown, with the statistically significant ones (only CLARA-A2 liquid REFF) indicated in bold.

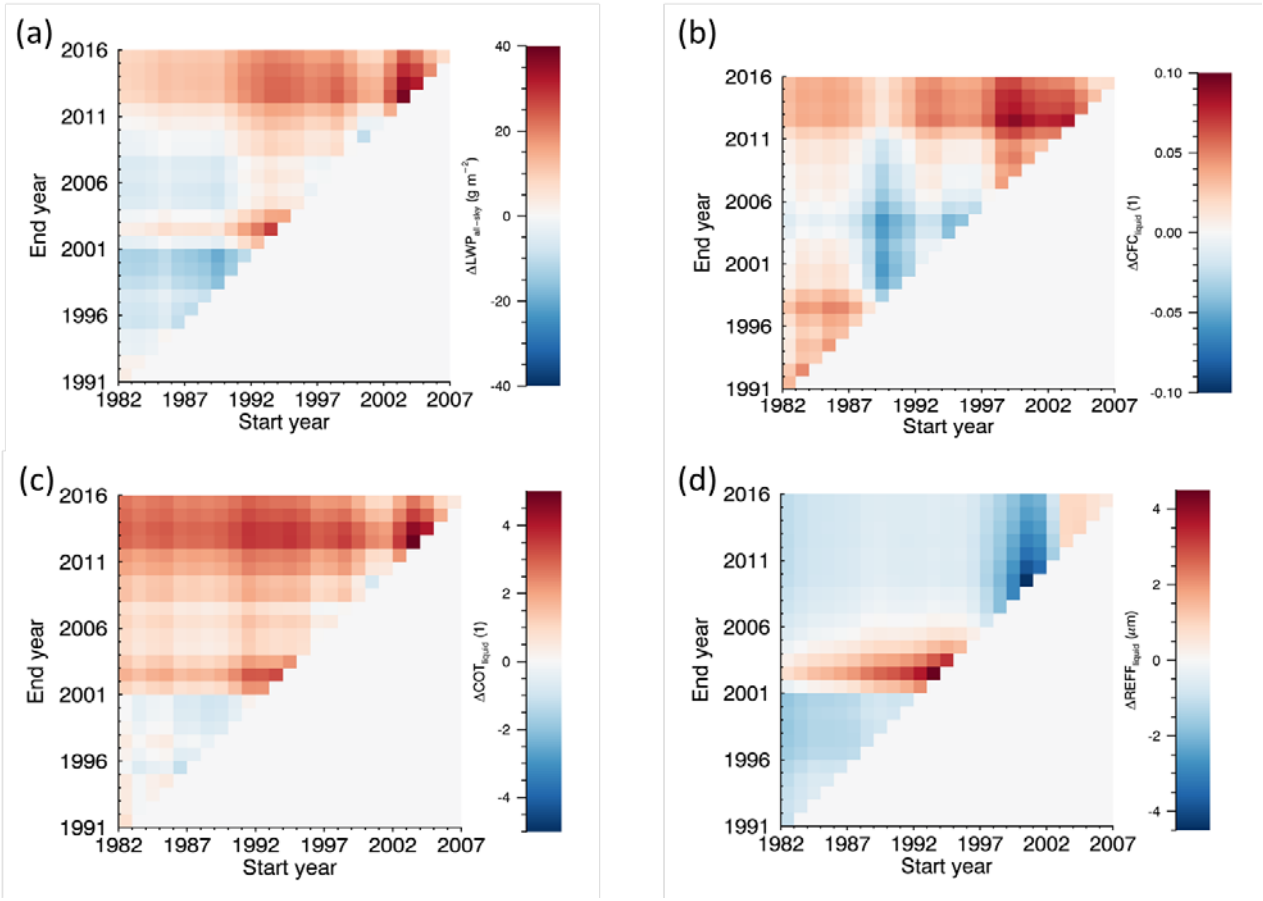

Figure 6. Changes in liquid cloud properties over southern China, based on 34 years of CLARA-A2 data (1982-2015) and estimated for all possible combinations of start and end years, with a minimum time range of 10 years. The four plots show corresponding changes in (a) all-sky LWP, (b) liquid CFC, (c) liquid COT and (d) liquid REFF.

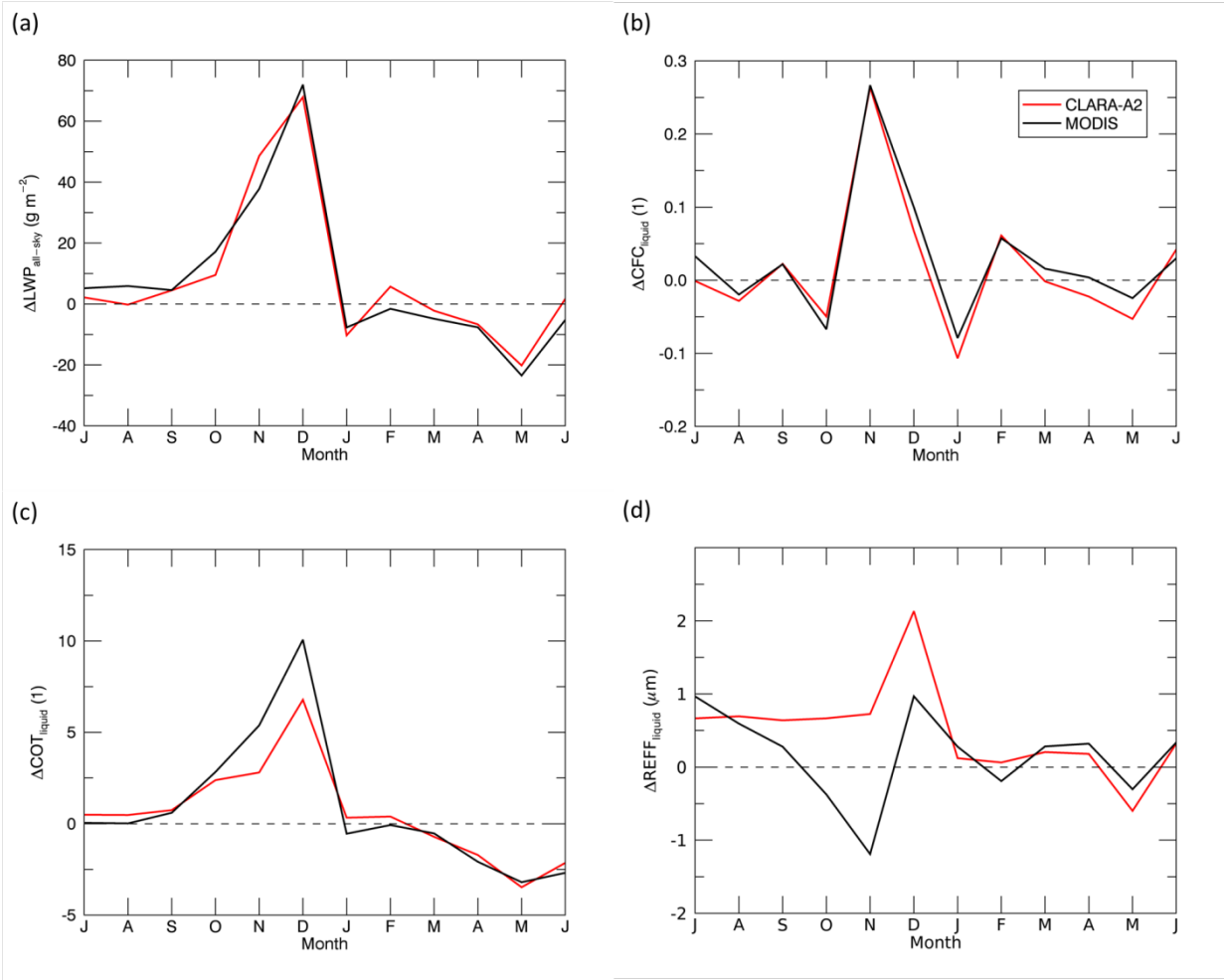

Figure 7. Seasonal variation of changes in liquid cloud properties over southern China. (a) all-sky LWP, (b) liquid CFC, (c) liquid COT and (d) liquid REFF changes from 2006 to 2015 based on CLARA-A2 and MODIS data.

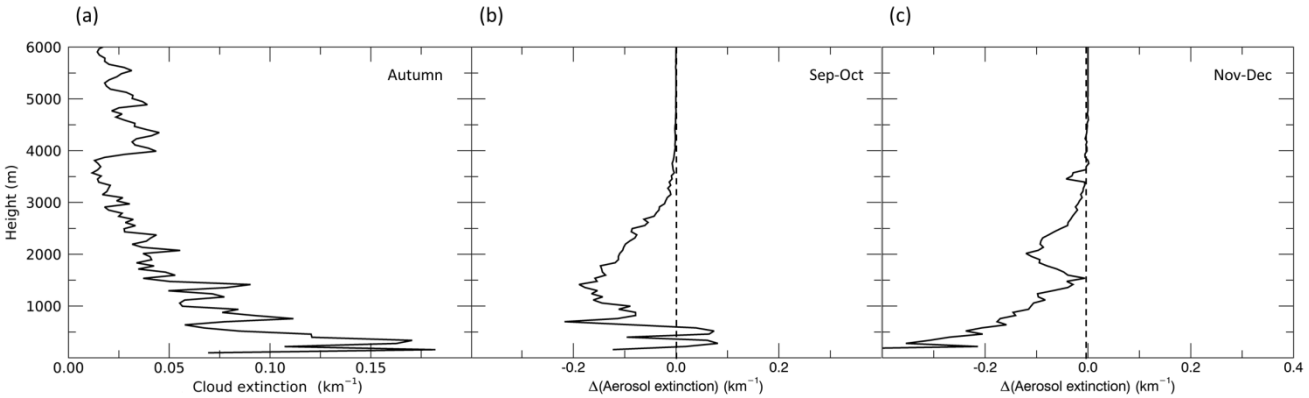

Figure 8. Profiles of cloud and aerosol changes over southern China. (a) Cloud extinction in autumn (September-November), estimated based on LIVAS CALIPSO data from 2007-2011. Aerosol extinction change for September-October (b) and November-December (c) based on CALIPSO level 3 data from 2007-2015. ~~**Changes are estimated for total aerosol and separately for dust, smoke and polluted dust aerosol types.**~~

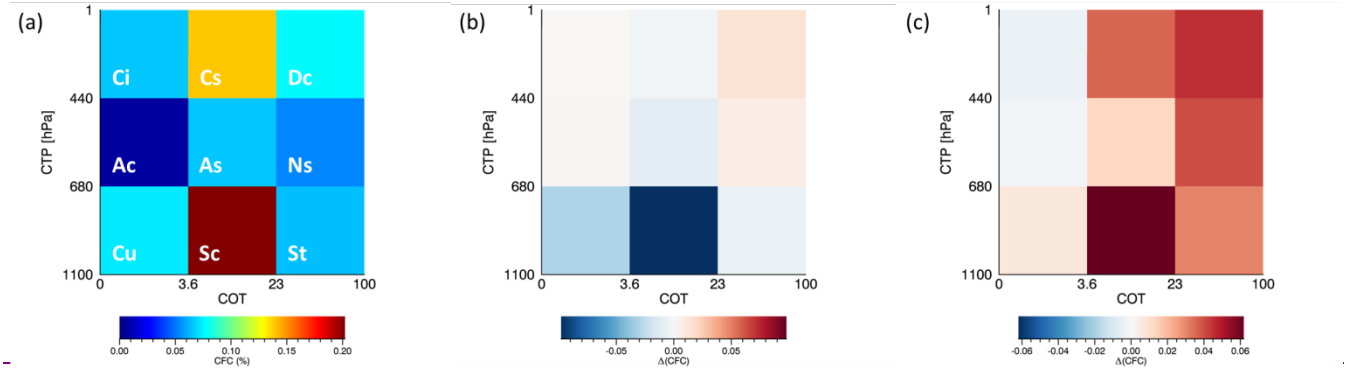

Figure 9. (a) Average autumn CFC for different cloud types, from MODIS data and the ISCCP cloud classification scheme based on joint COT – CTP (cloud top pressure) histograms. Cloud type abbreviations are as follows: Ci: Cirrus, Cs: Cirrostratus, Dc: Deep convection, Ac: Altocumulus, As: Altostratus, Ns: Nimbostratus, Cu: Cumulus, Sc: Stratocumulus, St: Stratus. Changes in CFC for each cloud type in October and November are shown in (b) and (c), respectively.