To: Dr. Toshihiko Takemura, Editor, *Atmospheric Chemistry and Physics*

From: Anonymous Referee # 2

Date: 10 December 2018

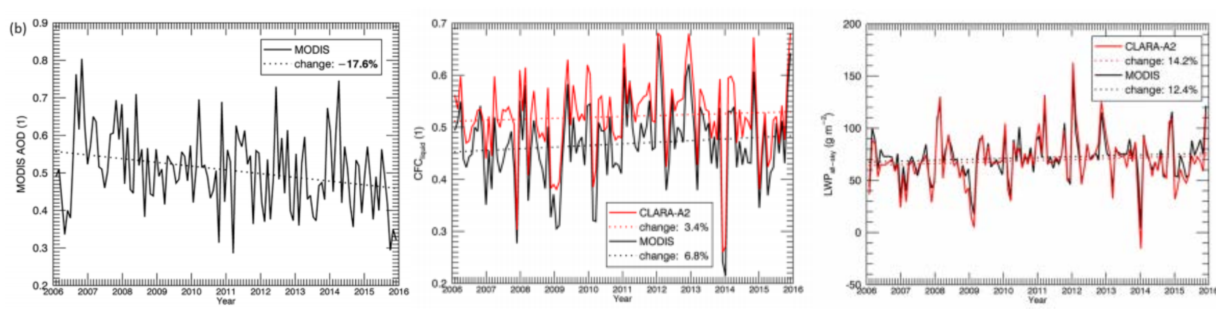
Subject: Further explanation of recommendation to reject “Satellite Observations of Aerosols and Clouds over Southern China From 2006 to 2015: Analysis of Changes and Possible Interaction Mechanisms” for publication in *Atmospheric Chemistry and Physics*.

In your email dated 04 December 2018, you advised that the authors’ reply to my initial review of their paper did not mesh with the content of my review. You asked me to comment on the manuscript again and to more concretely indicate relevant sentences and figures supporting my initial concerns with the paper and to reply to the authors’ comments. I have re-read the manuscript, my first review, and my notes from the first review. Based on this re-review, my recommendation to reject the manuscript is unchanged. In this memo, I provide the specific details that underpin this recommendation.

The fundamental flaw that I find with this paper is that the data and analyses presented in the manuscript do not support the conclusions. It is also implied that some variables must be related because they trend together, but no causal relationship is established. Statistical tests are mentioned as having been performed, but no statistics such as p-values establishing significance or correlation coefficients establishing correlation (except for the minor case discussion on Pg. 9, Line 2) are presented. The authors are good about owning up to the limitations of the data, which is appreciated. However, acknowledging the limitations doesn’t make them go away or make the dataset better suited for addressing the chosen science questions. Additional depth of analysis is needed to overcome these limitations in order to draw meaningful conclusions.

Here, I highlight my concerns with specific manuscript statements (italicized and quoted). Because of the extensive nature of these concerns, I do not think that the paper should be published without new data and analyses and extensive re-writing. Since new data and a considerable increase in the depth of analysis is required beyond that typically encountered for a major revision, I recommended the paper be rejected. I did not reach this recommendation lightly and only after a thorough reading, and now re-review, of the manuscript.

Pg. 1, Lines 10-12: “The results show a 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.”



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?

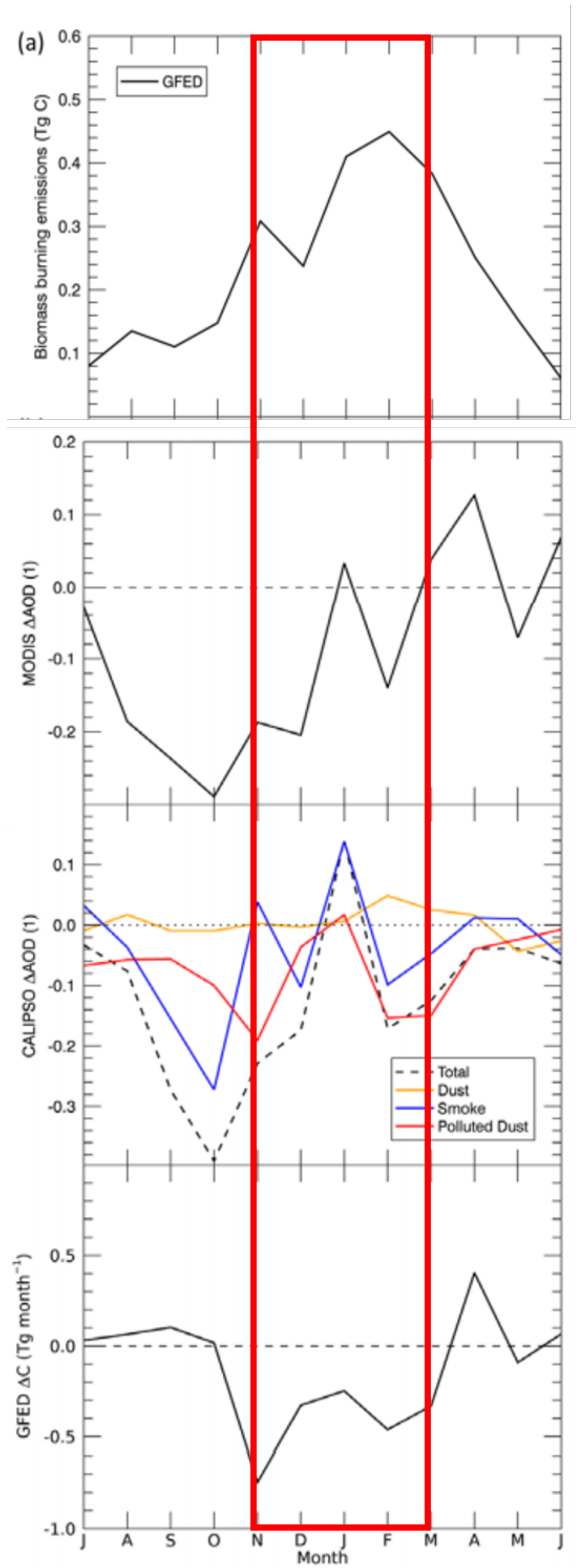
There's also quite a bit of day-to-day and sub-pixel variability that is not reflected in the Level 3 gridded monthly mean product as well as different numbers of measurements (i.e., samples) in each pixel that need to be considered and this is not really discussed in the manuscript. Some mention of 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?

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.

Pg. 1, Line 12-13: "Analysis of aerosol types and emissions suggests that the main driver for their reduction is a decrease in biomass burning aerosols. These changes occurred mainly in late autumn and early winter months..."

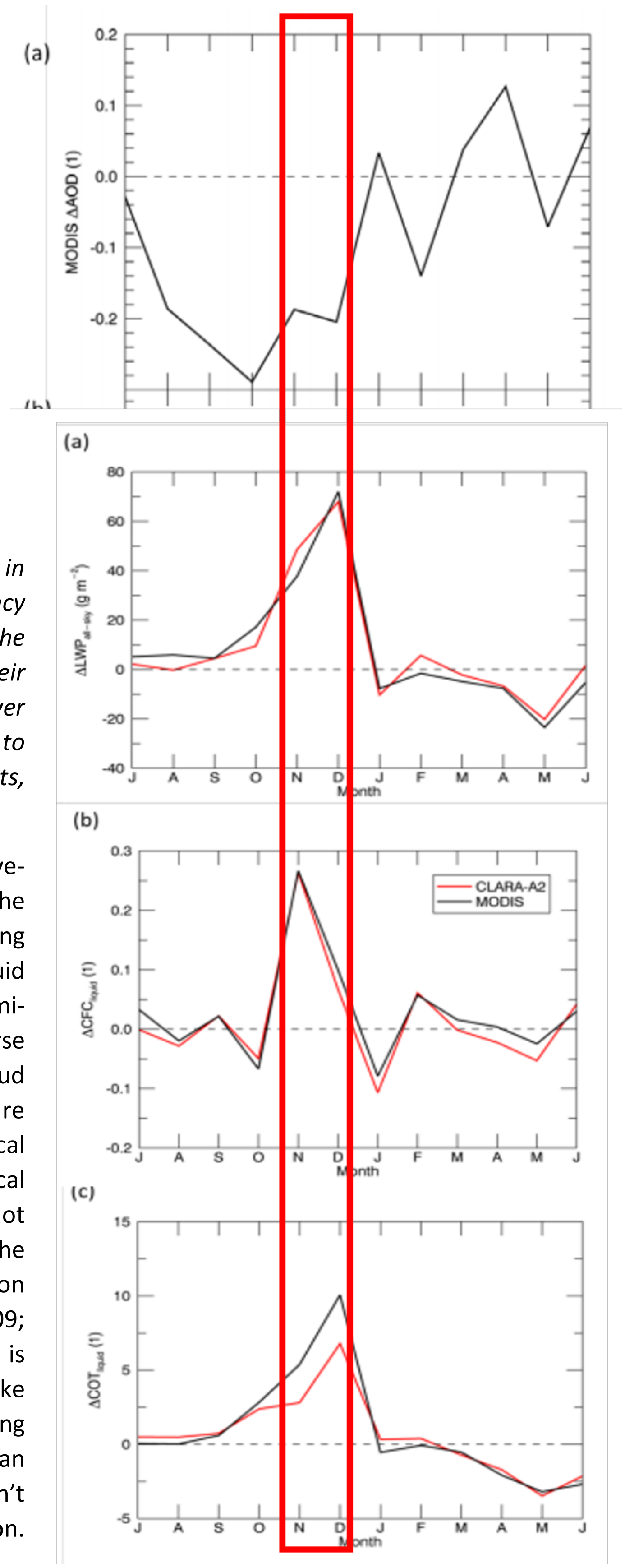
The fundamental flaw with this conclusion is that it is not clear that all aerosol types have been captured, so one cannot say that the "main driver" of AOD trends over the last decade is biomass burning or continental pollution or marine aerosol or other types, because only the three aerosol types are 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.

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. There are no metrics of statistical variability included in this graph – 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.



Pg. 1, Line 13: "These changes occurred mainly in late autumn and early winter months and coincided with changes in cloud properties."

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".



Pg. 1, Line 15-17: "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."

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.

Pg. 4, Line 32-33: "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."

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.

Pg. 4, Line 35-36: "According to the CALIPSO classification, smoke aerosols originate in biomass burning activities…"

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.

Pg. 4, Lines 36-37: "Biomass burning emissions and satellite-based AOD are not directly comparable."

I agree with the authors' statement here, and yet, Figure 1 and Figure 3 attempt to make precisely this comparison.

Pg. 5, Lines 7-8: "Based on the CALIPSO aerosol types classification, this decrease can be attributed to corresponding reductions in polluted dust and smoke aerosols"

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.

Pg. 5, Lines 14-15: "Based on CALIPSO, this decrease [in AOD] is driven by biomass burning aerosols: as for the full time series (Fig. 2c), dust aerosols show no significant change."

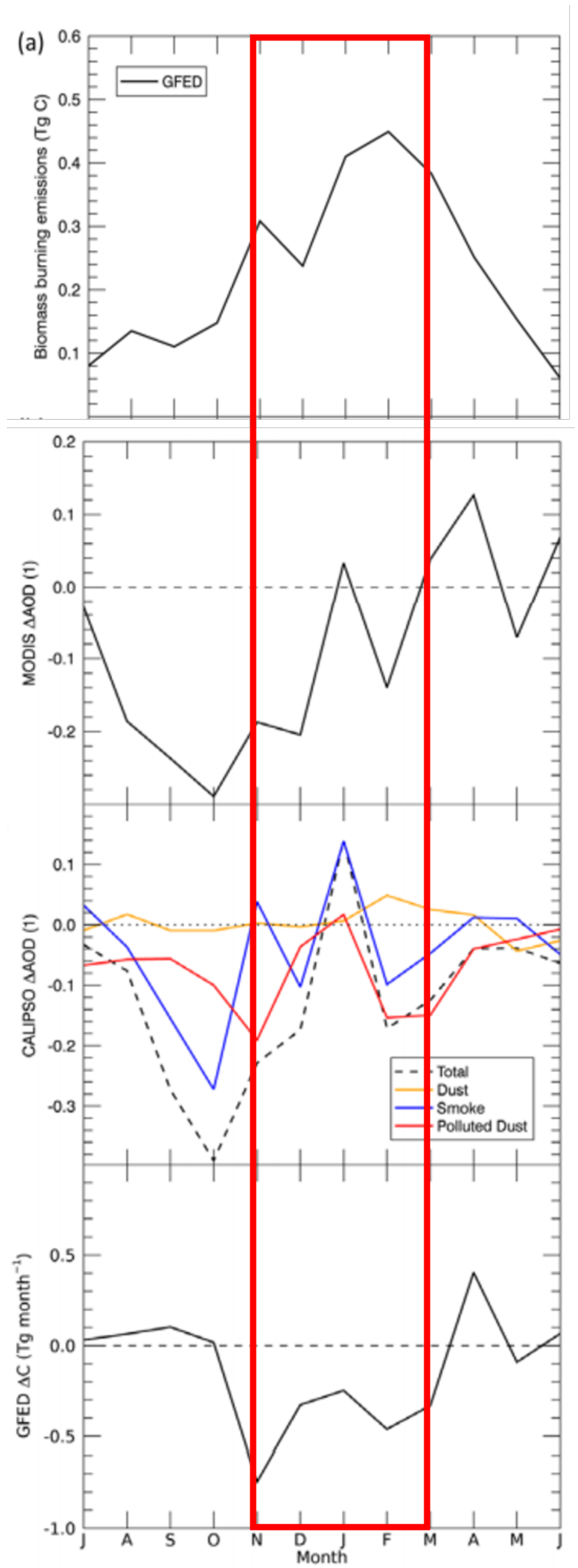
What evidence is there that the smoke and polluted dust aerosol types are dominated by biomass burning aerosol versus other sources of combustion or pollution aerosols? The CALIPSO aerosol type is not specific to biomass burning. Consequently, for the authors to make this conclusion, they need to provide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

Pg. 5, Line 15-18: "analysis of the total mass of carbon particles (C) from local 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 should be attributed to changes in residential energy sources, which peak during the same period."

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.

Pg. 5, Line 20- : "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."

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"?



Pg. 5, Lines 23-26: "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 differences in seasonality patterns (Fig. 1). In such cases, AOD and local emissions do not agree well (e.g., smoke aerosols in October). Forest fires and biomass burnig activities in Indochina could be such sources."

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.

Pg. 5, Lines 29-33: "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)…Hence, some ambiguity probably exists regarding the origin of the aerosols, especially for the smoke aerosol type."

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.

Pg. 5, Lines 38-40: A minor comment is that COT and REFF should be defined.

Pg. 6, Lines 8-10: "Cloud changes appear statistically significant at the 95% level over large areas of the study region, especially over land, when studied on a pixel basis. Analysis of spatially averaged values, however, over the entire (5x10-degree) study region, reduces this significance to levels below 95% in most cases of Fig. 5."

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?

Pg. 6, Lines 33-34: "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."

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?

Pg. 6, Lines 37-38: "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."

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.

Pg. 7, Line 10-11: "These results suggest that meteorological variability is not among the major factors contributing to the aerosol and cloud changes reported."

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.

Pgs. 7-8, Section 3.3.2 "Possible effects of ACIs and ARIs:"; "our results appear to be inconsistent with the standard definition of the first and second indirect effects, although the possibility of multiple mechanisms occurring simultaneously cannot be excluded."; "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."; "It is important noting that this mechanism holds primarily for absorbing aerosols, such as biomass burning particles, which is the case in this study. 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".

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.

Pg. 8, Line 10: "Figure 8a shows the typical profile of cloud extinction in autumn over southern China…"

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).

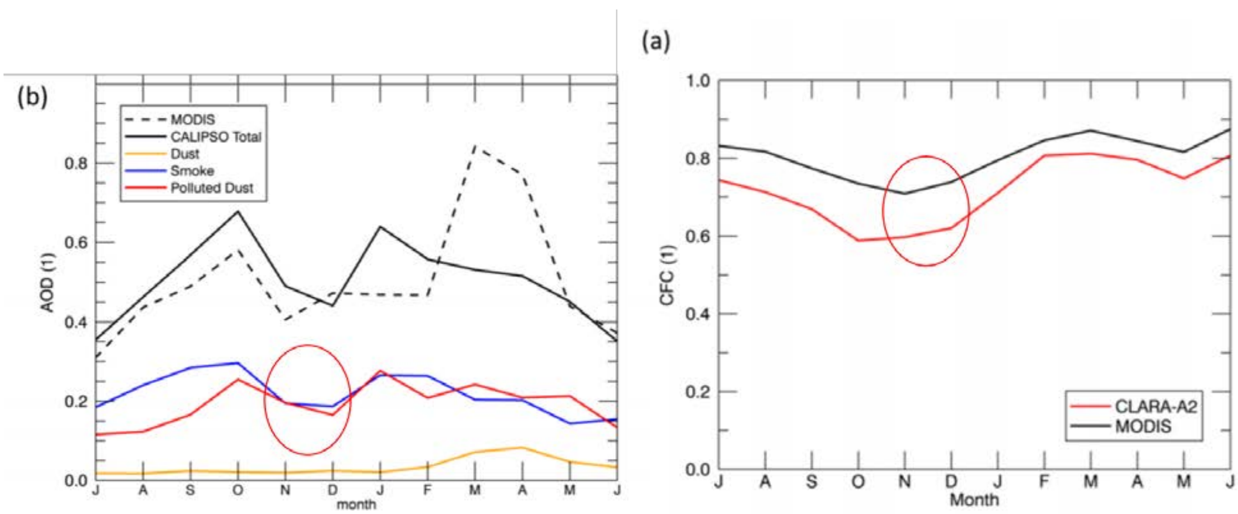
Figure 9 and related discussion.

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.

Pg. 8, Lines 40 – Pg. 9, Line 1: "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."

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.

Pg. 9, Lines 1-4: "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.



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.

Pg. 9, Line 8-10: "it was found that absorbing aerosol loads over the region decreased significantly, and this decrease was attributed mainly to changes in biomass burning activities."

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.

Pg. 9, Line 10-11: "Concurrent changes in liquid cloud fraction and thickness were observed, with notable increases and decreases in different months."

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.

Pg. 9, Line 11-13: "Further analysis of vertical profiles of both aerosols and clouds showed that the signs of cloud changes depended on the position of aerosols relative to clouds, being in agreement with the predictions of the aerosol semi-direct effect, under different aerosol and cloud configurations."

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.

Pg. 9, Lines 17-18: "Here, the combined analysis of different aerosol and cloud data sets showed a high level of consistency with predictions of [the semi-direct effect]."

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.

Pg. 9, Lines 18-20: "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 will focus on model simulations of the conditions described here, in order to provide more insights regarding the underlying physical mechanism."

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.

Authors' Comments (in italics) and reviewer's reply (in plain text):

1) *Multiple pieces of information are provided that are suggestive of the authors' conclusion that AOD changes are related to carbon emissions from burning, and the authors' conclusion is supported by a number of previous studies.*

   While the data may be suggestive, there are some significant gaps (e.g., missing aerosol types, lack of unambiguity in aerosol typing) that prevent direct attribution of AOD changes to specific aerosol emissions sources. If previous studies have shown a link between biomass burning and AOD changes, then it is not clear to me how this paper meaningfully contributes to that body of work.

2) *The manuscript acknowledges the limitations associated with the CALIPSO Level 3 data. Because this data has been validated and used by other papers, the limitations do not necessarily invalidate their conclusions. It just limits the scope of conclusions that can be drawn regarding the sources of the aerosol load over the region.*

   My concerns are not with the underlying data, which are useful for many different purposes. I do not think that these data are sufficient, however, to draw the conclusions that are presented in this paper. I appreciate that the authors have been upfront in acknowledging the data limitations; perhaps a future study employing a model or additional datasets/tools could move beyond just identifying them to overcoming them.

3) *The reviewer read the paper incorrectly when commenting that the seasonal trend in AOD does not track with the cloud properties during the November-December time period. There is a strong decrease in AOD during these months.*

   Yes, but the seasonality of delta-AOD and delta-Cloud Properties do not look particularly similar. I also don't know what is a strong vs. weak decrease in AOD. Some additional discussion on what these changes are (I assume the decadal change) and what the annual trend slopes are would be helpful for elucidating strong vs. weak changes.

4) *The averaged data are standard and have been used by many studies. This paper finds strong changes that cannot be caused by data averaging.*

   Again, the strength of the changes and their statistical significance is not clear to me from the manuscript. The Level 3 data has more than just the mean values, and includes standard deviations and numbers of observations. These need to be accounted for in the statistics. My concerns are not with the data themselves, but rather with the conclusions that are being drawn using them.

5) *A two-sided t-test was used to calculate the statistical significance of all calculated changes. Statements in the text that the results being discussed are statistically significant are made at multiple places throughout the manuscript.*

   It would be good to include p-values and correlation coefficients (and their p-values) to support the qualitative statements that are being made in the text. Is the t-test being applied to the 2006 data and the 2015 data to evaluate a difference or is it being applied to the entire timeseries? Some additional detail on the trend analysis and statistical methodology would be welcome here so that these details are not easily overlooked.

6) *Characterizing the conclusion regarding the attribution of aerosol sources to biomass burning in the manuscript as "highly speculative" and the study more broadly as drawing "strong, unsupported conclusions" is unfair. The authors have been careful in indicating limitations.*

   I appreciate that the authors have been careful in indicating limitations. I do think that the limitations limit the ability to draw conclusions based on this dataset. Consequently, strong conclusions like those detailed above are going to be at least somewhat speculative without better data or tools for source attribution and unambiguous aerosol typing.

7) *No concrete or specific flaws are mentioned. A path toward publication is not articulated.*

   I hope that this re-review provides an appropriate level of specificity. While I do not see a path toward publication for this particular manuscript, I think that a study employing preferably both a model and Level 2 satellite data (with the complete set of aerosol types) to look at aerosol trends and the source attribution of those trends to specific emissions sources would be a very worthwhile paper. However, using that type of data to try to establish a connection to long-term trends in cloud properties and trying to relate cloud trends to ACI effects seems like a step too far to me given the known scale issues in identifying and quantifying ACIs.