Changes in atmospheric aerosol loading retrieved from space based measurements during the past decade

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Dear Anonymous Referee #1,

We thank you for the constructive comments, which replies are listed on the supplement.

General Comments

(i) A good number of papers have been published in the last few years on aerosol changes and trends. What is missing in this paper is a comparison of its findings to the existing ones from other studies.

-> Yes, there have been many papers for aerosol changes and trends. As your comment, we have improved the manuscript by citing the papers and comparing their corresponding results with our ones.

(ii) I also have some reservations regarding the methodology, which are discussed below.

-> We have answered the reservations below.

(iii) Since the study period is relatively short, it is better to use the term "tendencies" instead of "trends".

-> We agree that the time lengths of the satellite data used in this study are relatively short to discuss "trend", especially only 6 years for MODIS-Aqua AOT data. Nonetheless, they are the longest available and obtainable when we started this work. In the papers comparable to this work (e.g. Li et al., 2009; de Meij et al., 2012; Hsu et al., 2012; Karnieli et al., 2009; Thomas et al., 2010; Xie and Xia, 2008; Yoon et al., 2011, 2012; Zhang and Reid, 2010; Zhao et al., 2008), the term "trend" has been used. Therefore, we would keep the term "trend" and use it together with the "tendency" in this work.

(iv) In the manuscript, explanations of obtained changes of AOT are quite often based on speculations reaching the limit of hypotheses. Often, they are based on discussions of the regime of aerosol sources and transport, for each area. However, I am afraid that this is not enough. More support is necessary and authors may search to get some in other way. For example, seasonal analyses of trends could provide some evidence and explanations, shedding more light.

-> Yes, we totally agree your comment and thank you for the constructive suggestion. However, unfortunately, the weighted trend model introduced in this study is designed to minimize the uncertainty caused by the cloud disturbance (i.e. no AOT retrieval due to cloud occurrence). The cloud occurrence is strongly correlated with the number of observations or retrievals as shown in Figure S1 (Yoon et al., 2012). Figure S1 shows the removal ratio of cloud and the number of observations per month. Because the square roots of the numbers in same season can be similar, the weighted model is not enough to minimize the cloud disturbance for seasonal trend estimate and the seasonal weighted trend can be more or less the same with the simple linear trend estimate. This is reason why we didn't discuss the seasonal trends in Yoon et al. (2012). Therefore, since the main goals of this study are to improve and to estimate the more convincing trend of atmospheric aerosol by minimizing the uncertainty caused by the unrepresentative sampling, we would more focus on showing how the weighted trend model works and how much

the different/limited temporal sampling can bring the uncertainty in the trend estimate. To identify and explain the major cause leading to the trend, actually further study is needed. Instead, we would cite other relevant publications, which have discussed the cause.

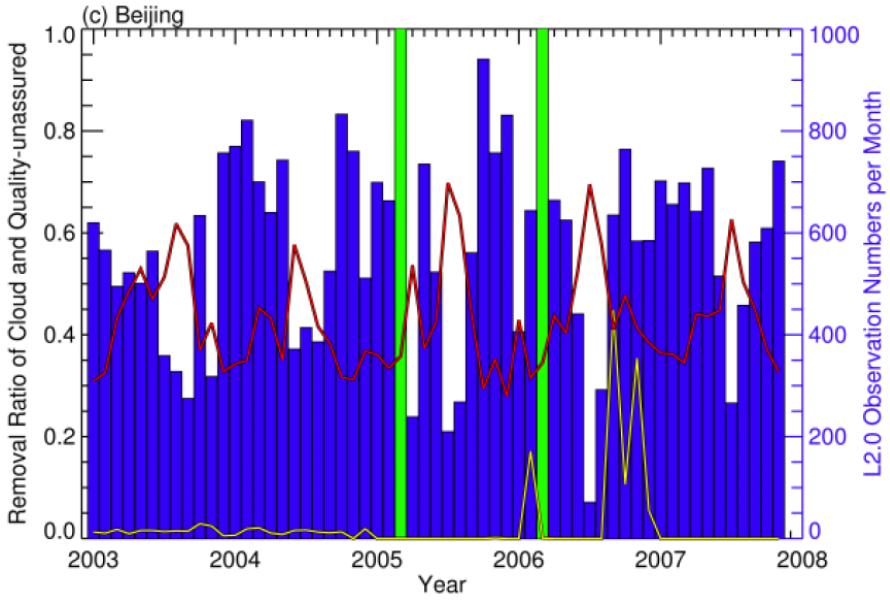

Figure S1. The removal ratio of cloud (red line) and quality-unassured (yellow line) cases to AERONET level 2.0 data (blue bar) within each of research period at the Beijing AERONET stations. Green bars mean that the observation numbers per month are over 1000 times (Yoon et al., 2012).

Main comments

Abstract

1. Page 26002, lines 1-14: text is not appropriate here since it is pure theoretical and does not refer to the findings of this study

2. Based on the previous comment, the remained Abstract must be enhanced to stand up alone.

3. The studied period has to be explicitly indicated in the Abstract and not be referred as "during the past decade".

Introduction

4. A considerable number of studies dealing with trends of AOD have been studied so far, either at local or regional and global scale. Therefore, it should be clearly stated here what new the present study brings and adds to scientific knowledge with respect to the previous and existing studies.

-> We have improved and modified our manuscript as your comments.

Section 2.4 (AERONET)

5. It should be reported how many AERONET stations are used in the study, making reference to Table 2. It would be also useful to show them on a map.

-> We have added a new figure (i.e. Figure 2 in the manuscript) showing the AERONET stations used in this study.

Section 3

6. Has any care been taken with regards the continuity of AERONET data? It is stated that their minimum temporal coverage is 5 years but it is not clarified whether they cover continuous periods or they do have breaks on it.

-> The AERONET stations were selected using the series of criteria as defined in Yoon et al. (2012) to check the temporal continuity and coverage of AERONET data as following:

i) The significant monthly mean reflecting the population mean is calculated with the number of observation larger than 300 per month.

ii) The complete yearly data set is composed out of more than seven qualified monthly means to minimize a bias from data missing in trend estimate.

iii) Minimum temporal coverage for estimating a convincing trend is a continuous five-year set of the complete yearly data.

They have been described in the revised manuscript.

7. It is also important to provide the number of compared satellite-AERONET AOD data pairs, for each AERONET station.

8. Page 26007, line 21, "… are caused only by the different and limited sampling": it should be clarified whether the term "sampling" refers to time or space. The entire sentence has to be re-written in a much cleared manner.

-> We have improved and modified our manuscript as your comments.

Section 4

9. Page 26009, lines 2-4, "A relatively large … cloud-free AOT retrieval (Yoon et al., 2011).": yes, but large standard deviations can also be attributed to strong temporal variability of AOT itself induced by aerosol- and not cloud- related changes. How can the authors isolate the contribution of this to their computed overall standard deviation of daily AOTs within a month? This should be important in areas, for example, undergoing aerosol transport or biomass burning.

-> Yes, as you pointed out, relatively large standard deviation of AOT can be attributed to cloud contamination in in cloud-free AOT retrieval as well as high variability itself of transported and biomass burning aerosols. In this study, we didn't apply any method to distinguish between them. Nonetheless, since in statistics and probability theory, a large standard deviation indicates statistical unrepresentativeness of the mean value, the mean value can bring a significant bias in the trend estimate. In other words, even though a large deviation really comes from high variability of transported and biomass burning aerosols, it is a kind of irregular event that should be removed in the trend estimate. This has been explained in the revised manuscript.

10. sub-section 4.1: when applying the Grubbs and Gaussian tests to remove outliers in weighting factors, it is "assumed that that an approximately normal distribution is the most probable one.". This, as shown in Figure 4, indeed effectively removes outliers. Nevertheless, what happens if these outliers are meaningful? For instance, if values correspond to AOT, how/why can/should very low or high values be excluded? Authors should discuss this.

-> The outlier of the weighting factors is a crucial influence in the trend estimate based on the weighted trend model. It happens when the standard deviation is very small or large compared to the mean value. The small standard deviation is generally attributed to the small number of observations and the large one is caused by the high variability of aerosol itself.

Section 5

11. Last sentence is meaningless; you should rather state whether trends are nicely correlated or not.

-> We have improved and modified our manuscript as your comments.

Section 6

12. second paragraph. The discussion of AOT trends over western and eastern Europe and especially their attribution to causes must be further supported. For example, the decreases of emissions in eastern Europe are not given any reference while hypothetical explanations and assumptions should not be presented as facts unless supported by references. Also, for both regions, the role of natural aerosols should be also assessed given its contribution to total AOD.

-> Since there has been no study for the significant uncertainties, which are different/limited temporal sampling of polar-orbiting satellites and cloud disturbance in the trend estimate of cloud-free AOT, this study presents a new approach to minimize the uncertainties by use of weighted least squares regression and multiple satellite-derived AOTs from the space-born instruments. This study has unambiguously and first shown how significant the uncertainty from unrepresentative sampling is in trend estimate. We agree that this study is not enough to identify and explain directly the major cause leading to the trend and further study based on modelling and in-situ observation is needed. Instead, to support our results, we would cite and compare the results from other relevant publications, which have analysed the cause.

13. Page 26014, lines 9-11: there are available references for the megacity of Cairo (e.g. Kanakidou et al., 2011).

14. Page 26015, lines 3-4, "… with nearly about 1 billion people living in and around the Ganges valley, are …": nevertheless, the increase of AOD is not in that region.

15. The statements concerning the increase of AOT over Korea and Japan should be supported by some evidence, literature included.

-> We have improved and modified our manuscript as your comments.

16. Page 26015, line 28 through to page 26016, line 2: is this enough to explain the increasing tendency of MODIS-Aqua afternoon AOT? For example, wildfires usually do not last for half a day but are more persisting sometimes lasting for a few days.

-> Yes, generally the fire activity lasts for several days. Nonetheless there is a significant difference between MODIS-Terra and -Aqua AOTs from 2003 to 2008 as shown in Figure S2.

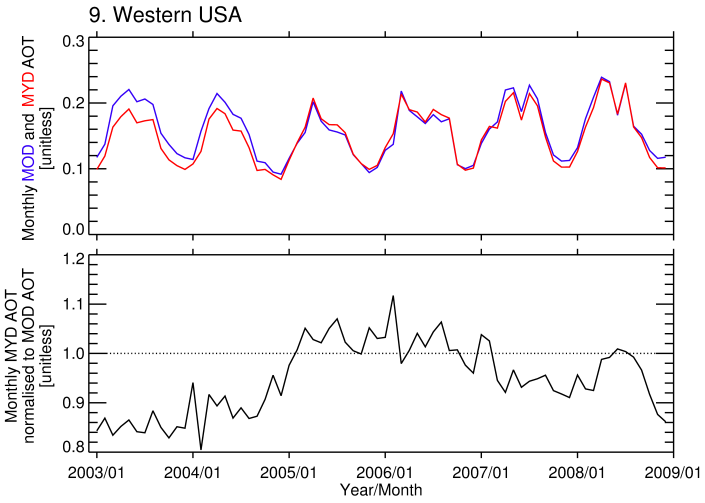

Figure S2. Plots of the time series of MODIS-Terra (MOD) and -Aqua (MYD) AOTs, and MYD AOT normalized to MOD AOT from 2003 to 2008.

As discussed in the manuscript, since there is no difference in retrieval accuracy, cloud filtering method, and spatial resolution between MODIS-Terra and -Aqua, therefore it is attributed to different sampling. The region where

significant trend is observed in Figure 9 (d) is overlapped with where wildfires frequently break out and the wildfires typically ignite in the afternoon. Therefore we supposed that the fire activity enhanced during 2003 to 2008 could be a cause for the trend of MODIS-Aqua AOT. Since it is difficult to identify what is the main cause to this difference in MODIS-Terra and -Aqua AOT without further studies, we would mention this possible cause in the manuscript and cite a paper reporting the significant positive trend of the aerosol scattering coefficient.

17. As to the discussion of AOT tendencies over China and the relevant Fig. 10, what are the changes suggested by each dataset (and applied liner regression fit)? Also, why there are more spikes in red curve, i.e. the one of AERONET?

-> As discussed before in the manuscript, although there is good chance of deriving different trends from the different/limited samplings over such a large urban agglomeration, we wanted to show the similar seasonal variation of the satellite-retrieved AOTs from 2003 to 2008 over East China and some relation to NO_2 and SO_2. The AERONET AOT at Beijing seems to be different to other satellite-retrieved AOTs and its spikes is due to its small spatial coverage and small number of observation shown in Figure S3 (Yoon et al., 2012).

18. Page 26016, line 29 through page 26017, line 2: a R value equal to 0.6 certainly cannot justify statements like "strong correlation".

-> We have modified our manuscript as your comments.

Other details

1. Page 26002, line 17: replace "Whilst the aerosol optical thickness, AOT, over Western Europe decreases …" by "While the aerosol optical thickness, AOT, decreased over Western Europe …"

-> We have improved and modified our manuscript as your comments.

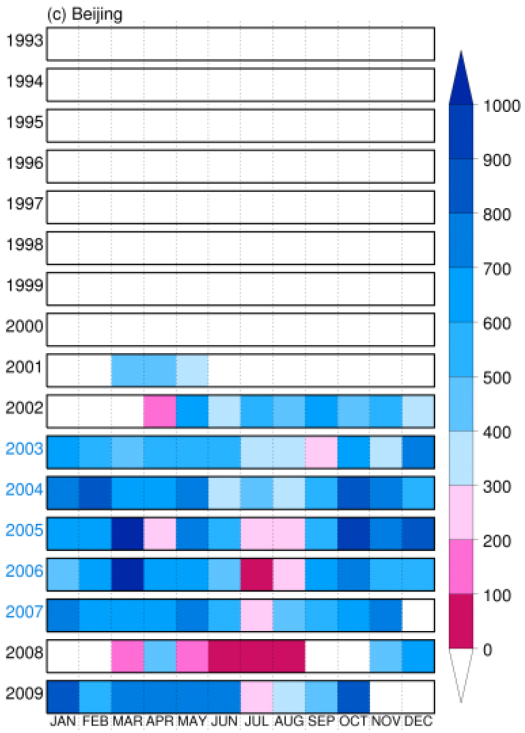

Figure S3. The monthly observation numbers of level 2.0 AOT (440 nm) at the Beijing station since 1993. The research period for each station is shown by the blue years at vertical axis (Yoon et al., 2012).

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