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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Global and regional trends in aerosol optical depth based on remote sensing products and pollutant emission estimates between 2000 and 2009" *by* A. de Meij et al.

A. de Meij et al.

a.demeij@cyi.ac.cy

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Reviewer 4 This paper starts by presenting a global, decadal trend analysis of AOD retrieved from the MODIS MISR aboard Terra, as well as from a selection of AERONET stations around the globe. Different resolutions (Level 3 and then Level 2) are used for the satellite data products. Statistically significant negative trends are seen over eastern North America and Europe, with some less robust, but positive trends over South and East Asia. The paper then relates the AOD trends to changes in emissions of various precursor species, such as decreasing emissions of SO2 and NO2 over Eu-

rope and North America, and increasing pollutant emissions over Asia. I find this paper ambitious. Combining MODIS + MISR + AERONET + emissions is a step forward in the community, and work is timely and very important as the world discusses future environmental impacts. The attempt to quantify trends both at Level 3 and Level 2 is a very good idea. Unfortunately, I think the paper falls far short of the mark. It is poorly organized, and has a tendency to lead the reader off on tangents. Global and regional single scattering albedo, while given one sentence in the abstract, is not really discussed in the paper (no figures). There are obvious omissions to the cited literature discussing aerosol trends based on satellite data (e.g. many papers by Mishchenko and AVHRR teams). Some of the cited literature (e.g. Zhang and Reid) seems to be cited based on abstracts, without actually reading the papers. While tests for statistical significance are one way to determine robustness of trends, the paper makes no attempt to determine if trends are due to increased "background" AOD values, or increased number of high AOD "events". Trends are also less "believable" due to the use of incomplete annual cycle (Feb 2000-Nov 2009). Waiting until end of Jan 2010 at least look at entire years would have been better idea. Finally, while it is important to trace AOD trends to emission trends, the chemistry and physics of these processes is complicated and nonlinear. Detailed analyses of chemical transport models are probably necessary. The captions on the figures tend to be incomplete. Also, this is kind of strange, but it took me until the middle of the paper, to realize that the comparisons were being made over land only! I cannot know from reading the paper, but it seems to me that data from MODIS and MISR were used in a carefree way, without much interaction with the satellite science teams. For example, both MODIS and MISR data were made "operational" in Feb of 2000, however it was not until March or April of 2000 that the data were considered stable. The Zhang and Reid (ACP, 2010) paper (cited here) shows that at least part of the MODIS AOD trend can be traced to changes in the MODIS instrument itself (calibration drifts?). Levy et al., (ACP, 2010) show that MODIS AOD comparison with AERONET has changed over time. Although the general corroboration between MODIS, MISR and AERNOET helps this paper's case, it should be

acknowledged that instrument artifacts will impact a global or regional trend.

To help overcome that the paper is poorly organized, we inserted in the Introduction an overview of the structure of the manuscript in order to provide a clear overview of how the manuscript is constructed. We added to the text in section 1: “The paper is organized as follows. In chapter 2 we present the methodology and a description of the remote sensing data. In section 3.1 we evaluate the significance of the MODIS and MISR global AOD trends, followed in section 3.2 by a more detailed statistical evaluation of the AOD trends over land by MODIS, MISR and AERONET between 2000 and 2009. Then in section 3.3, we compare AOD MODIS Level 3 with Level 2 data and we link the derived trends with the changes in the anthropogenic emissions for Europe, North America and Asia. We present the conclusions in section 4.” The single scattering albedo by MISR is a product which has not been fully validated (personal communication Ralph Kahn, 2010) and therefore one should be careful in drawing conclusions. For this reason we think that showing plots, which describe SSA trends, is not appropriate. We have added the following references to the Introduction and we give a short description of the instruments they used and their findings: Mishchenko et al. (2007), Mishchenko and Geogdzhayev (2007), Zhao et al. (2008) and Thomas et al. (2010). We added to our evaluation the error of the slope in order to understand better the uncertainty of the slope for MODIS, MISR and AERONET and to determine if the trend is significant (Table 2 and Tables of the Supplement). The error of the slope is calculated according to Numerical Recipes in Fortran 77, Second Edition, Press, W. H. (1992).

The data that we are using is until December 2009. With respect to the seasonal cycles special care has been taken to avoid having incomplete cycles. First, an integer number of cycles has been taken to avoid constant biases in the trends calculation. This implied the removal of the first year (2000), where the data are not complete. In our calculation of the trend, the seasonal cycle has been removed from the observations. We did not apply a sinusoidal fit for the seasonal cycle. Instead, each monthly component

has been calculated separately based on the multi-year average of the observations in that month. This approach removes the seasonal cycle, independently from the yearly shape and it is useful when the seasonal cycle is not defined as sinusoidal function, which is sometimes the case for many locations. We inserted to the text: “In our trend calculations we removed the seasonal cycle from the observations by subtracting for each month separately the multi-year average of the observations in that month.”

Before submitting the manuscript to ACPD, R. Kahn of the MISR science team commented the manuscript and provided suggestions, which we were keen to include in the manuscript. Regarding the drift in the AOD due to calibration of the MODIS product, we contacted L. Remer of the MODIS science team. We added to the text in section 4: “There has been an artificial tendency in the MODIS C005 AOD product due to drift in the radiometric calibration (Levy et al., 2010; Zhang and Reid 2010; Remer personal communication, 2011). Levy et al. (2010) validated MODIS C005 AOD by comparing with AERONET AODs and estimated an overestimation of $\sim$0.005 prior to 2004 and underestimation of the same magnitude afterwards.”

Specific comments: 1. Abstract: In general the abstract is qualitative, not quantitative. I think the readers want some specific numbers. The SSA analyses seems unrelated to rest of paper (AOD). I would even suggest removing SSA analysis throughout the paper, because of likely huge uncertainties and lack of validation. The linkage of AOD and emissions is really a modeling effort, and there is too much speculation (“it appears that”, “emissions may become”, “may have weakened”..)

The single scattering albedo gives information about the composition of the studied column. We believe that the analysis of SSA trends is a worthwhile exercise, knowing that indeed the SSA products are not validated (personal communication R. Kahn), which is also clearly stated in the manuscript. The increase / decrease in the SSA can be linked with the black carbon emissions, a finding that, according to us, is new and meaningful with respect to the impact of the implementation of soot filters on diesel engine vehicles and the improvement of combustion technology. Therefore the analysis

of SSA is not unrelated to the content of the manuscript in section 3.4 and contributes to the scientific discussion, even though the results are qualitative rather than quantitative, which has been emphasized in the manuscript.

2. Introduction: Needs a more thorough literature search. Also, is this a study over land only? Or over both ocean and land? We have added the following references to the Introduction and we give a short description of the instruments they used and their findings: Mishchenko et al. (2007), Mishchenko and Geogdzhayev (2007), Zhao et al. (2008), Thomas et al. (2010), see comment earlier. Then we mention in the Introduction that we first compare in section 3.1 (and Fig. 2) the AOD trends globally (land and ocean) followed by a more detailed analysis of the trends over land.

3. The paper should clearly explain what Level 3, Level 2, etc is. It should be done ONCE! Also, why was MODIS-Terra used and not MODIS-Aqua? We received a similar comment of reviewer 1 and 3, therefore we added in the Introduction the following: "MODIS and MISR data are available at different processing Levels. Level 1 data contain calibrated radiances. Level 2 products are produced at a spatial resolution of 10x10km for MODIS and 17.6x17.6km for MISR. Level 3 products contain parameters from the Level 2 products on a 1°x1° for MODIS and 0.5°x0.5° for MISR. We did not use products of the MODIS-Aqua instrument, because Aqua was launched in May 2002 a comparison with MISR data based on 10 years is therefore not possible. Secondly, MODIS Aqua has a different equatorial overpass time (around 13.30) than the Terra platform (around 10.30am), which could lead to discrepancies between the AOD retrievals. Aerosol loads and cloud cover can change within this time difference affecting the successful AOD retrieval."

Regarding the use of only MODIS-Terra we added the following in the Introduction: We did not use products of the MODIS-Aqua instrument, because Aqua was launched in May 2002 and a comparison with MISR data based on 10 years is therefore not possible. Secondly, MODIS Aqua has a different equatorial overpass time (around 13.30) than the Terra platform, which could lead to discrepancies between the AOD re-

trievals from the Terra platform which has an equatorial overpass time around 10.30am. Aerosol loads and cloud cover can change within this time difference affecting the successful AOD retrieval.

4. Methodology section: might be easier to read if some of it was bulleted.

Done.

5. For collocation why was geographical range of $\pm$15-30 km chosen? (Reference?) What is the temporal collocation protocol?

There is not really a reference regarding the selection of the sampling of MODIS Level 2 based on $\pm$15-30 km. The MODIS Level 2 data was selected for the period 2000-2009, which resulted in $\sim$40000 files for each of the region. Knowing longitude and latitude for the location, we extracted data from those pixels which are within the range of $\pm$15-30 km for each file (i.e. overpass).

6. AERONET section: Probably the errors will not be too large, but AERONET team (e.g. Eck et al., 1999) suggests using quadratic interpolation to retrieve AOD at 550 nm.

MODIS and MISR AODs are given on 550nm and 558 nm respectively. We interpolate AERONET AOD to 550nm using AOD from 440nm and 870nm (S. Kinne, personal communication, and Kinne 2003 JGR).

7. MISR section: Does small sampling of MISR Level 2 lead to artifacts in Level 3 data?

It does not lead to artifacts in the Level 3 data, but to a monthly average based on less successful AOD retrievals. The more successful AOD retrievals one has to construct a monthly mean value the better the representation of the monthly average. MODIS has a daily global coverage. It is therefore more likely that more successful AOD retrievals are available to construct a monthly average than for MISR.

8. MODIS section: MODIS isn't really a "camera", it is an imager. The point is that it measures only on a single geometry for each ground target.

Corrected. Thank you.

9. MODIS section: There is a "deep blue" algorithm (Hsu et al., 2004) that retrieves AOD over bright desert surfaces. At least mention it, and say it is not used here.

Done. We added to following to the text in section 2.1.3: "In addition to the Terra MODIS AOD products, MODIS Aqua AOD products are processed with the Deep Blue algorithm in order to retrieve aerosol optical properties over bright surfaces (Hsu et al., 2004). The Deep Blue algorithm employs radiances from the blue channels (412nm) of the MODIS Aqua instrument, where surface reflectance is low enough to make such retrievals possible."

10. Emissions inventories: Are there any uncertainty estimates? It is difficult and also beyond the scope of this manuscript to quantify the uncertainties of the emissions in the EMEP, IPCC and REAS inventories for the different species. EMEP provides some information regarding the uncertainties of SO2 and NOx emissions in their inventories ($\pm$ 10% and $\pm$ 30%, respectively), see: http://www.eea.europa.eu/data-and-maps/indicators/emissions-of-primary-particles-and#uncertainties.

11. Results: Here is where I believe that the Zhang and Reid paper (ACP, 2010) was cited, but never read thoroughly. Zhang and Reid clearly stated that they had to do a lot of cloud screening and product screening in order to believe the MODIS AOD product. Are any similar screening/massaging techniques performed here?

See previous comment regarding the drift in the AOD due to calibration issues.

12. Results: Entire section! AOD has a strong seasonal cycle in many regions. Care must be taken when attempting to ïnËĞAËŻt a linear trend to a time series that is dominated by a cyclic variation, especially if the time series doesn't span an integer number of cycles (Feb 2000 to Nov 2009). The seasonal cycle must be accounted for,

or at least some statements made as to potential impacts.

We agree with the reviewer and special attention has been taken to avoid this. First, an integer number of cycles has been taken to avoid constant biases in the trends calculation. This implied the removal of the first year (2000), where the data are not complete. In our calculation of the trend, the seasonal cycle has been removed from the observations. We did not apply a sinusoidal fit for the seasonal cycle. Instead, each monthly component has been calculated separately based on the multi-year average of the observations in that month. This approach removes the seasonal cycle, independently from the yearly shape and it is useful when the seasonal cycle is not defined as sinusoidal function, which is sometimes the case for many locations, see comment earlier. For MISR we have data until December 2009. We corrected this in section 2.

13. Results: More quantification is necessary. Statements like "the trends are similar" are not sufficient. A fundamental problem is that the trends from level 2 and 3 MODIS data are plotted for different resolutions. For level 3 the closest 1x1 degree pixel to each AERONET station is used, while for level 2 it is data within a 15-30 km radius. Are differences due to averaging/sampling/aggregation methods?

To overcome the issue of statements like "the trends are similar" we added to our evaluation the estimation of the error of the slope for MODIS, MISR and AERONET in order to understand better the uncertainty of the slope and to determine if the trend is significant (Tables of the supplement). The error of the slope is calculated according to Numerical Recipes in Fortran 77, Second Edition, Press, W. H. (1992). The formulas we used to calculate the error of the slope are given in the reply to Reviewer's 1 comments. Secondly we added Table 2 the slope and the error of the slope of MODIS Level 2. This allows us to estimate the trend with a certain confidence level and compare that with MODIS Level 3 and AERONET and to Table S4 (now S3) a column which shows the difference in AOD of the regression line for AERONET Monthly Level 2 products. This helps us to quantify the difference in the trend line between AERONET Daily and AERONET Monthly products and the satellite products. The results of this comparison

are included in the text of section 3.3 (now 3.2).

The reviewer has a good point and this is why we compare MODIS L3 with L2 in order to see if there are differences in the AOD due to grid cell resolution and/or aggregation.

14. MODIS L2 and emissions: Why are chemical equations listed here? Why not the introduction, if at all? Also, gas to particle conversions are not really as linear as a few chemical equations suggest. There are many more, plus interaction with clouds and hydrologic cycles. Chemical transport models should be used or at least acknowledged.

We deleted the reactions, but we kept some information on how SO4=, NO3- and NH4+ are formed. We agree that modelling studies could be more appropriate, but this is beyond the scope of this manuscript.

15. Figures: a. Fig 1. Really makes no sense for the difference between red dots and blue dots Corrected, see earlier comment.

b. Fig 2. Why plot the same information 2 or 3 different ways? Bottom row of panels is really confusing. The x-axis suggests confidence > 95% but the plot shows values from -1 to 1.

Reviewer 1 and 3 had a similar comment regarding Fig. 2. Therefore we removed Figures 2a and 2b and we changed the text accordingly.

c. Fig 3: What are the locations? Are these AEROENT sites? These are plots of satellite trends, what about the trends from the AERONET sites? Do they agree?

These are indeed corresponding AERONET sites, this is also mentioned in the manuscript. Detailed analysis of the trends and the slopes by MODIS and MISR is given in section 3.2 (now deleted) and in Table S1 (now S4). The comparison between MODIS and MISR with AERONET is presented in section 3.3 (now 3.2). As we have now also mentioned the error of the slopes a more robust statistical comparison is made.

d. Fig 4: Incomplete year cycles are a problem. The fonts are too small to be read clearly. Maybe by “deasonalizing” (normalizing with respect to average seasonal cycle), the trends would be much clearer. We received a similar comment of reviewer 1. We added the following to the text: “We performed a sensitivity analysis in order to estimate the impact of not deseasonalizing the AOD trends by MODIS and MISR for the period 2001 – 2009. We found small differences in the slope of the trends ($\sim$ 5%) over Europe, North America and Asia, which are densely populated regions with strong anthropogenic emissions. Only for locations for which the error of the slope is of the same order of magnitude as the slope of the trend (i.e. below 60% significance level), the trend can sometimes change. However, such changes are not relevant considering their small magnitude and level of significance.” We improved the fonts and we added the trend lines of the three instruments to make the trend clearer.

e. Fig 5: Same questions as Fig 3, and the caption is even less descriptive. We have deleted Fig. 5 because we inserted in Table 2 the slope and error of the slope for MODIS Level 2 products.

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