

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.

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Reviewer 1 The paper is poorly organised, with a lot of repetition and waffle. It also has the feel of a rushed job, with many typographical and grammatical errors. We respectfully disagree and assure the reviewer that it was not a rushed job. However, we accept that there is room for improvement, merged sections 3.2 and 3.3, carefully checked the manuscript for errors and inserted in the Introduction an overview of the structure of the manuscript in order to provide a clearer overview. We added the following text in section 1: “The paper is organized as follows. In chapter 2 we present the

C13893

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 figures are not well designed and there are not enough of them (for instance, the authors discuss trends in single scatter albedo derived from MISR measurements repeatedly, but nowhere are these trends presented in a figure). The single scattering albedo by MISR is a product which has not been fully validated (personal communication Ralph Kahn, 2010) and therefore conclusions cannot be quantitative. For this reason we think that showing plots, which describe SSA trends, would not be appropriate.

Furthermore, many of the results presented by the authors are of dubious relevance. Many of the trends, particularly for individual sites, are tiny. There is no indication of an uncertainty or confidence interval on these values, so they are essentially useless. This is especially so when the two or three independent measurements available appear to disagree.

We respectfully disagree that many of the trends at individual sites in Fig. 3 and Fig. 5 are tiny and of dubious relevance. From Fig. 2 (significant AOD trends with confidence level 95% or higher) we see that for Europe, North America and Asia the trends are significant (positive or negative). To determine whether a positive or negative trend is significant we applied the statistical method of Weatherhead et al. (1998). Even if the trends for individual sites are sometimes tiny positive/negative we must consider them as a scientific result. Not having a significant positive or negative trend does not mean that the result is insignificant. It indicates that the AOD trend is small, even though air pollution reduction measures may have been implemented. For example,

C13894

over eastern China emission factors have been reduced (i.e. improved technology), which partly balances the concurrent increase in fossil fuel use. If the trends between the two instruments disagree many measurement and algorithm issues come into play (cloud screening, multi-angle view versus single-angle view, the number of successful overpasses, etc.). This is explained in the text. We agree that an estimation of the error of the slope is required to better understand the uncertainty of the slope and to determine if the trend is significant. We included in Table 2 and in the Tables of the Supplement the error of the slope for MODIS, MISR and AERONET. The error of the slope was calculated according to Numerical Recipes in Fortran 77, Second Edition, Press, W. H. (1992).

Finally, the authors make no attempt to physically relate the changes in emissions (discussed at the end of the paper) to the observed trends in AOD. The paper's conclusions amount to little more than a statement that in regions where emissions of aerosol precursors have decreased over the past decade, a decreasing trend in AOD tends to be seen, and vice versa. Thus the authors fail to present anything new to the literature.

Several studies (Mishchenko and Geogdzhayev, 2007; Mishchenko, 2007; Zhao et al 2008, Streets et al., 2006 and Streets et al., 2009) suggest that the dimming and the brightening of the atmosphere is related to the change in cloud cover and the increase/decrease of the anthropogenic pollution in the atmosphere depending on the related economical activities and legislation. To our knowledge only Streets et al., 2006, 2009 quantify the changes in anthropogenic emissions (1980 – 2006) and link these changes with the trends in the AODs. Analyzing the emissions of the different inventories (EMEP, IPCC and REAS) up to 2010 and directly link their trends with AOD trends makes our study relevant and new, because (1) we use the latest emission estimates up to 2010 for North USA, Europe and Asia, (2) we try to link the trends in the AODs by MODIS and MISR and AERONET with the emission inventories, which has not been done before and (3) our study confirms or rebuts earlier findings. We believe that our approach contributes to the understanding of the regional changes in

C13895

AOD, even though it is not possible to make a direct physical link with anthropogenic emissions but rather their tendencies between 2000 and 2009. In section 3.4 we have strengthened the discussion of the emissions in relation to AOD changes.

General comments and suggestions Ensure that ALL acronyms are defined once, on their first use.

Done.

The authors make extensive use of the terms level 1,2,3 when referring to satellite and AERONET data. It should be made clear in each instance, exactly what these terms mean. (i.e. level 1 is generally calibrated radiances on a grid determined by the instrument measurement system, when referring to satellite data, etc.).

Done. We added to the text in section 1, also suggested by Reviewer 3: "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."

The methodology section is written essentially as a list of steps. This is fine, but it would be a lot clearer if presented as either a bulleted or numbered list.

Done.

Note that there are two MODIS instruments, one on Terra and one on Aqua. Although the authors mention that MODIS is on Terra, they should explicitly state that the Aqua instrument is not used.

We added to the Introduction the following: "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 (around 10.30am), which could lead to discrepancies between the AOD retrievals. Aerosol

C13896

loads and cloud cover can change within this time difference affecting the AOD retrieval.”

The authors reference the global trend in oceanic AOD derived from AVHRR data (Mishchenko et al. 2007) – they may be interested in Thomas et al., *Atmos. Chem. Phys.*, 10, 4849-4866, 2010, which includes further analysis of this result.

Done. Thank you for the reference. Reviewer 2 and 4 suggested to add more references to the Introduction. We added the following: “Mishchenko et al. (2007) and Mishchenko and Geogdzhayev (2007) use the AOD from the Advanced Very High Resolution Radiometer (AVHRR) satellite to study the dimming and brightening tendencies. They found a decrease of 0.033 ($\sim 0.0024/\text{year}$) of the global tropospheric AOD. Zhao et al. (2008) studied the long-term (nearly 25 years) AOD trends over the global oceans using the AVHRR Pathfinder Atmosphere extended (PATMOS-x) data set and their results are consistent with those of Mishchenko and Geogdzhayev (2007). Thomas et al. (2010) evaluated the Global Retrieval of Along Track Scanning Radiometer (ATSR-2) Cloud Parameters Evaluation (GRAPE) AOD product over the ocean by comparing against measurements of the AErosol RObotic NETwork (AERONET) and AOD of the Advanced Very High Resolution Radiometer (AVHRR) of the Global Aerosol Climatology Project (GACP). They concluded that the GRAPE AODs corroborate the AERONET observations, but some discrepancies are apparent with the GACP data. “

On a related point to the previous one, have the authors contacted the MODIS aerosol team regarding this work? If my memory serves me correctly, Lorraine Remer suggested that MODIS Terra might not have sufficiently good calibration stability for use in monitoring aerosol trends, at the 2010 EGU General Assembly (I suggest the authors check on this).

We have contacted Lorraine Remer and read her review on Zhang and Reid (2010). She suggested to include in the discussion that ‘there has been artificial drift in the AOD product due to drift in the radiometric calibration’. We added to the text in section

C13897

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 ~ 0.005 prior to 2004 and underestimation of the same magnitude afterwards.”

AOD has a strong seasonal cycle in many regions, which the authors acknowledge several times. Care must be taken when attempting to fit 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. The authors don’t provide details of how they have calculated their trends; have they accounted for the seasonal cycle in their calculations, or investigated its potential impact?

We agree with the reviewer and special attention has been devoted 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. 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.”

Regarding the impact of not deseasonalizing, we inserted this to the text in section 3.1. “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

C13898

locations with no statistical significance 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.”

The authors need to be more quantitative in their analyses. Statements like “the trends are similar” are not sufficient. When comparing two measurements or two trends the authors need to provide a rigorous measure of whether they agree or not.

Corrected. 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). Secondly we added to 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 we added to Table S4 a column which shows the difference in AOD of the regression line for AERONET Monthly Level 2 products. This helps 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.

Throughout the paper, figures are referred to as Fig. 2a, Fig. 5b etc. However, none of the figures are correspondingly labelled.

Done. We deleted Fig. 5, see answer next comment.

The authors have extensively used the absolute and percentage change in the AOD trend from 2000 to 2009 (in preference to the slope of their fitted trend line) to quantify the observed trends. This is fine in principle, but repeatedly referring to these values as a change in observed AOD (for example the first sentence of section 3.4.1, “In Fig. 5a, the change in MODIS Level 2 AOD between 2000 and 2009 is presented...”) is incorrect. The difference between observed AOD between two years would be the difference between some average AOD value in those two years, not a trend calculated from a time series over the whole period.

C13899

We have deleted Figure 5, because we inserted in Table 2 the slope and error of the slope for MODIS Level 2, which allows a direct comparison with the trends of MODIS Level 3.

Section 3.1 is sloppy and requires a significant overhaul. Figure 2 essentially plots the same information in 3 slightly different ways, and the authors descriptions of the patterns seen in each of these plots is overly long and repetitive. Furthermore, the authors seem to be slightly confused as to what is plotted in the last two panels of Fig. 2, as they state that these panels show “the significance” of the trends, whereas the plots show the trends themselves. I suggest that the number of panels in this plot is reduced to four, showing the decade trends for MODIS and MISR followed by maps of the significance of these trends (or, alternatively, their uncertainty). A rewrite of section 3.1 based on this simplified figure should produce a more coherent and succinct description.

To a large extent we implemented the suggestions of reviewer 1 in section 3.1. We removed Figures 2a and 2b and we changed the text accordingly. As recommended (see comment earlier) we included in the manuscript the errors of the slopes (Tables in the supplement); see earlier for the method. Before submitting the manuscript to ACPD we had valuable discussions with Ralph Kahn about the scientific content of the manuscript. We were very pleased that Ralph Kahn took the time to read the manuscript and provided suggestions/comments. We believe that his suggestions and comments improved the manuscript, especially section 3.1, because most of the suggestions were related to the significance of the trends.

I don't see the point of detailed analysis of MODIS and MISR AOD trends at specific AERONET sites when there is insufficient AERONET data to provide a comparison. Essentially, this is just rehashing the results presented in section 3.1, but for a small subset of single pixels scattered around the globe. I thus feel Section 3.2 is superfluous.

C13900

Reviewer 3 has a similar comment, therefore we removed section 3.2. We merged the results of the previous section 3.2 with the section "Global AOD trends by MODIS, MISR and AERONET" (now 3.2). We removed Table 1a and moved Figure 3 to the Electronic Supplement and we made the appropriate changes and references to the Tables and Figures in the text.

I find Fig. 3 (and Fig. 5) virtually unreadable. If the authors want to show the trends seen by MODIS and MISR (and surely AERONET should be included as well) at these sites, I would suggest a miniature bar chart at the location of each station, with one bar each representing the MODIS, MISR and AERONET trends. Again, this should only be done where an AERONET trend is available. It is also very difficult to relate the plots to the names of the individual stations used elsewhere in the paper.

Here we show in more detail the trends by MODIS and MISR between 2000 and 2009, now moved to the Electronic Supplement. Showing the trends by MODIS, MISR and AERONET, based on the number of months available by AERONET, will (1) change the trends with respect to the period 2000-2009, trends will be shown for stations with different time periods of observations in one figure (this will be confusing) and comparison with MODIS Level 2 will not be possible. We show the MODIS and MISR trends for the full period (2000 -2009) to enable a comparison with MODIS Level 2 data. This is an important part of the paper, as drawing firm conclusions based on Level 3 products is not recommended (personal communication R. Kahn). We believe that miniature bar charts will not improve the readability of the plots.

In section 3.3 the authors are comparing the AOD trends seen by the satellites over specific AERONET stations with those derived from AERONET measurements themselves. This is a sensible thing to do, provided the AERONET measurements provide a complete enough time series (and the authors have filtered the data for this). Why do they then present the data as averages for each region in Fig. 4? What is the point this plot trying to make? By averaging over the AERONET stations in each region the details of the comparison are lost; but the sampling across the region doesn't appear

C13901

sufficient for these plots to be considered representative of the regions overall.

Figure 4 (now Fig.3) presents a qualitative analysis of the regional assessment of the trends by averaging all the trends for each region (Europe, North America, South America, Africa and Asia). Details for each of the stations in the regions are given in the appendix (S2, now S1). This qualitative comparison has three advantages, i.e. (1) it removes outliers when the trends are presented for a region, (2) it provides insight about how large-scale aerosol optical properties change between 2000 and 2009 for each of the region in relation to air pollution legislation and (3) it can be linked with emissions inventories.

The comparisons presented in section 3.3 are not informative without uncertainties on the trends being compared. How is one to know if a changes in mean AOD of -30

Done, we included in the tables in the supplement the errors of the slopes.

The authors also compute trends using both AERONET daily mean and monthly mean data in section 3.3. I approve of this, but the manuscript goes no further than saying the trends are mostly similar. How similar? Do they actually agree?

Corrected. We added to Table S4 (now S3) a column which shows the error of the regression line for AERONET Daily Level 2 products, and we present the results of the comparison between AERONET Monthly and Daily products in the text.

In section 3.4, trends computed from MODIS level 2 and MODIS level 3 data are compared. Again, this is most certainly a worthwhile exercise, however I don't believe the approach taken by the authors is valid. Once again, the comparisons are done at the locations of AERONET stations. Although the trends from the AERONET stations themselves are mentioned, comparison is no more than stating whether the sign of the three data sets agree. A more quantitative analysis should be presented.

We inserted in Table 2 the slope and the error of the slope of MODIS Level 2. This allows estimating the trend with a certain confidence level and comparing with MODIS

C13902

Level 3 and AERONET. We adapted the text in section 3.4 (now 3.3) accordingly.

A more fundamental problem is that the trends from level 2 and 3 MODIS data are not calculated for the same regions: for level 3 the closest 1x1 degree pixel to each AERONET station is used, while for level 2 it is data within a 15 km radius of the station. Thus it is not possible to determine whether the differences in the trends seen are due to the averaging used to generate level 3 data, or due to the differences in the area used in the trend analysis. The authors need to decide which of these potential sources of differences they want to investigate, and design their experiment accordingly.

We selected from the MODIS and MISR data sets the pixels in which the AERONET station is located, not the nearest 1x1 degree pixel. MODIS Level 2 data is within a radius of $\pm 15 \times 30$ km of the AERONET station, so this falls within the MODIS Level 3 pixel. Averaging the nearby Level 3 pixels (1x1 degree) around the AERONET station will worsen the statistical approach of comparing this average with a point measurement. Indeed the large grid scale of the Level 3 data (1x1 degree for MODIS and 0.5x0.5 degree for MISR) introduces difficulties in terms of representativity of the AERONET observations, which are point observations. This problem occurs often in air quality transport modelling using regional and global models (ranging between 10x10 km to 1°x1° or coarser) when ground based measurements (gas and aerosol concentrations) and satellite observations, which are typically provided on resolution of 10-20 km, are used for evaluation and validation purposes. We agree that a higher resolution of the satellite products will increase the quality of the comparison with ground based observations from different networks.

I am puzzled as to why the authors have included descriptions in the change in emissions inventories in section 3.4. The data in this section are samples from MODIS and AERONET taken at specific locations, whereas the discussions of the emissions relate to overall regions.

The emissions between 2000 and 2010 for the coincident grid cell with the AERONET

C13903

station do not directly show the impact of the sources on the AOD trends for that specific location. Not only local emissions contribute to the AOD for a specific location, but also the primary and secondary aerosols from surrounding areas. Synoptic-scale transport contributes importantly to air pollution. Therefore showing the emissions for a whole region and analyzing the AODs for point locations in the domain is according to us, a worthwhile exercise.

The inclusion of chemical schemes detailing the conversion of gaseous precursors to aerosol constituents given in section 3.4.1 should appear in an introductory section, if at all. These reactions are well known and the references to Seinfeld and Pandis etc would be sufficient.

One of the above comments of reviewer 1 states that no physical attempt is made to relate the changes in the emission inventories with the observed trends in AOD. As mentioned earlier and in the manuscript, Wild et al. (2010) and Streets et al. (2006, 2009), Mishchenko and Geogdzhayev (2007) suggest that the dimming and the brightening of the atmosphere are caused by the change in anthropogenic emissions. We removed the reactions, but kept some information on how SO₄⁼, NO₃⁻ and NH₄⁺ are formed.

On page 30757 the authors state that they don't know the sampling time of MODIS level 3 data. This statement is nonsensical; MODIS is in a sun synchronous orbit, and level 3 data is calculated from the same level 2 data the authors are using in their analysis.

It is mentioned in the manuscript that we don't know the sampling time of the MODIS level 3 data. We mean that we do not know which days of successful AOD retrieval enter into the Monthly mean Level 3 product, e.g. is the average based on the first few days of the month, a few days at the beginning and at the end of the month or all the days of the month? Only counting of the number of successful AOD retrievals is available. We changed the text to explain this more clearly.

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The final paragraph is, frankly, nonsense. The paper hasn't shown that consistent AOD trends have been derived from multiple datasets. Nor has it linked these trends to changes in emissions in any meaningful way. Finally, linking emissions to AOD using chemical transport models is a well established practice, not a "next step".

We removed the paragraph.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 30731, 2010.

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