

## *Interactive comment on* "AOD trends during 2001–2010 from observations and model simulations" *by* A. Pozzer et al.

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We thank the reviewer for her/his comments. Following the suggestions, a deeper analysis of the meteorologically driven trends has been conducted which will be included in the revised manuscript. Here, however, only the comments of the referee will be addressed.

1. It is necessary to have some verification on the credibility of water uptake by the soluble aerosols. The model study of Pringle et al. (2010b), which used the same water uptake coefficients, has shown that there is both qualitatively and quantitatively good agreement of aerosol hygroscopicity (and hence water uptake) with measurement campaigns all over the world. Pringle et al. (2010b) C12118

included comparison of the modeled values with surface concentrations and vertical profiles (available from several campaigns). This agreement provides us with confidence that the representation of water uptake of the aerosol particles in the model is sufficiently credible to draw conclusions from temporal trends. Concerning the optical properties of the assumed internal mixture of aerosol particles and therefore the water contribution to the AOD uncertainties, these are discussed in Klingmüller et al. (2014). A definite conclusion on which approach is best suited to determine the overall aerosol optical properties in a computing cost efficient way is still to be investigated.

2. Show the time series of AOD in each region to see if linear fitting is appropriate. We have produced the figures as suggested by the referee (see Fig.1-3), adding also the regions of Central Africa and South America (see also answer #3 below). One important note here is that a large interannual variability would increase the term  $N_t$  (Page 26629, line 21) and this would make the trends non significant. Therefore it is well possible that a linear fit is not appropriate for some regions, but this would also imply a non-significance of the trends. For example, in Fig.4 we enlarge the Fig.3 for the South East Asia (SEA) region, which was referred by the referee many times. The application of a linear fit can be debatable (as the referee correctly mentioned); nevertheless the large noise caused by biomass burning in the years 2002 and 2006 cause the trend to be **non significant**, despite the fact that a decrease can be observed and a linear fitting could be even appropriate. Therefore we are confident with our results and their significance.

## 3. Comparison with satellite data on regional trends:

(a) Some regions present large discrepancies between observations and **model results.** This question was somehow raised also by referee # 1 as well. The regions mentioned by the referee are Central Africa and Southern America. As shown by Yoon et al. (2012) the strong cloud coverage on these regions

severely affects the satellite retrievals. This is indicated by the non significance of the observation estimated trends over South America for all the satellite instruments. In Central/South Africa MISR and SeaWiFS do also present low data coverage and/or non significance. MODIS nstead shows significant negative trends, but we are not convinced of such trends as this is not corroborated by the other instruments. Again, as discussed in the previous point, we are confident that significant trends have been estimated only where the noise did not cover the possible increasing/decreasing signal.

(b) I wonder how much difference is there between the model-calculated regional averages with and without considering the matching time. The model and satellite are not matched spatially as this was not necessary in this study. The global maps have been plotted on the original resolution (either  $1 \times 1$  degree,  $0.5 \times 0.5$  degree for the satellite or  $1.1 \times 1.1$  degree for the model). The model was sampled at the correct local time of satellite overpass and these values were used to calculate the monthly values used in the comparison. It must be underlined that the satellite products do present information about number of observations used to construct the monthly values, although no real information is given about the time location of the missing observations. The calculation of the modeled monthly values included all available data points.

For the special case of missing values in the monthly satellite product, we produced Fig.5 for the SEAWiFS dataset (which had the highest number of gaps), where the model was also masked in the same temporal location. The differences in the modeled calculated trends (see Fig.3 and Fig.5) are very low and this masking does not alter the trends and their significance.

(c) Remove fitting line from Fig.5. We will do it in the revised version of the manuscript.

4. Role of meteorology in influencing trends. We would like to thank the reviewer for the suggestion to include the role of meteorology in influencing the trends. C12120

A deeper analysis has been performed and the manuscript will be extended. Trends in precipitation, wind speed and relative humidity have been calculated for clarifying the reason of meteorologically driven trends (see reply to specfic comments).

5. Model evaluation. As mentioned in the manuscript, AOD simulated by the model was already evaluated in numerous publications (Pringle et al., 2010a; de Meij et al., 2012b; Pozzer et al., 2012; Astitha et al., 2012). Pozzer et al. (2012) in particular analysed the biases and correlations between model simulated AODs and observed AODs from both MODIS and MISR instruments. Therefore we do not think it would be reasonable to repeat such analysis in this work, but we would prefer to leave the references to these studies.

Specific comments:

- Page 26620, line 3: we will add the name of the EMAC model (ECHAM5/MESSy Atmospheric Chemistry).
- Page 26625, line 18-24: yes, only one year for dust and sea-salt emissions were considered in the simulation. We will rephrase the discussion about dust, underlining that any trends involving dust is purely due to changes in its transport/deposition, while the emissions remain constant. Additionally we will change the abstract, underlining that only biomass burning and anthropogenic emissions were time dependent.
- Page 26626, line 10 and 13: we will change the name using the suggested names (BASE and FIXEMI), as the names could create some misunderstanding.
- Page 26628: We agree with the reviewer. This work was also performed in (de Meij et al., 2012b). We will reformulate the sentence.

- Page 26629, line 3: we thank the referee for noticing this error. Indeed AERONET is a "ground based remote sensing instrument". We will correct this error in the revised version.
- Page 26629, line 4: we fully agree with the referee that "correctly" is wrong here. We will reformulate the sentence as : "the model is generally able to reproduce the AOD observed by AERONET within a factor of two for most of the observations."
- Page 26630, line 4: SD stands for standard deviation.
- Page 26630, line 9: "Small significant decrease" was a bad formulation. Small was referring to the trends estimated from MISR data compared to the one estimated from MODIS data. We will rephrase the sentence as "More specifically, both MISR and SeaWIFS show a significant decrease over North America and partially Europe, being smaller than what observed by MODIS. Strong significant increase is instead visible over Saudi Arabia for both MISR and SeaWIFS."
- Page 26630, line 15-16: We agree with the reviewer's comment. Nevertheless, as mentioned also in our reply to Dr. Sayer, the MODIS Deep Blue algorithm does not cover years after the 2007. Therefore we will change the manuscript mentioning the Deep Blue Algorithm (see reply to Dr. Sayer) but we won't include the data in the analysis.
- Page 26630, line 20: The submodel SORBIT is explained in detail in Jöckel et al. (2010). Fig.3 of the manuscript is created using the correct (local) overpass time of the platform. Having the TERRA and OrbView-2 two different overpass time, two figures are present for each simulation, as the model was sampled differently via SORBIT.
- Page 26630, line 22-25 : We agree with the referee and we will reformulate the sentence, removing the claim that a general agreement is present between model results and observations. However, as mentioned above (major comments), some C12122

of the regions where the agreement is missing (South America and Southern Africa) is based only on the MODIS comparison, as the MISR and SeaWiFS do not show any clear trends is these regions. We will also remove the "anthropogenically driven" remark, which is not coherent with our conclusion.

- Page 26632, line 15-16: The referee is correct. We will reformulate the sentence as "This means that the variation in the **anthropogenic and biomass burning** emission did not affect the AOD trend in this region"
- Page 26632, line 21-22: In Fig.6 precipitation trends in the model simulations RCP00 and RCP85 (same meteorology) for the decade 2001-2010 have been estimated. As written in the text, a clear decrease of precipitation in the Middle East can be observed, confirmed also by other studies.
- Page 26632, line 26: With this region we mean "Middle East". We will correct the paragraph.
- Page 26633: WASO compounds are not fully responsible for the increasing trends, as explained in Sect.6, Page 26634 line 20 to Page 26635, line 7. In Fig.6 of this reply we can notice that the decrease in precipitation does not only influence the Middle East but also the Sahara Desert. Therefore the changes in precipitation is one of the causes of increase in the dust aerosol lifetime.
- Page 26633, line 21-22: We cannot be more precise as only the Level 3 data are used. Therefore every dataset used its own processing algorithm, and these products are subject of different thresholds and decision processes. It is therefore natural that such observational datasets produce different results for critical locations where either the trends are very low or the noise is very high. In this case the high noise of the data caused by frequent presence of clouds (or haze) does not allow a clear estimation of the trends in the region (Yoon et al., 2014a). A study

on the reasons for diverging results between MODIS and MISR datasets is the one of de Meij et al. (2012a).

- Page 26633, line 24: As "another reason" we meant "another reason for differences between the trends estimated from satellite observational datasets".
- Page 26634, line 2-3: The emissions (biomass burning) have been decreased in the regions, although not significantly (Yoon et al., 2014b). We will clarify this in the revised version.
- Page 26634: as listed at Page 26624 line 26, as WASO we meant water-soluble compounds, i.e. all water soluble inorganic ions (e.g.:  $NH_4^+$ ,  $SO_4^2$ -,  $HSO_4$  and  $NO_3$ -) which do not include sea salt. We will clarify the sentence.
- Page 26634, line 11: The contribution of water strongly depends on the location and the relative humidity of the location. As mentioned in the reply to referee #1, this was studied by de Meij et al. (2012b), who found that over Europe and North America the associated aerosol water contributes around 40-45% to the total AOD. Nevertheless this contribution is highly variable, with low values in regions where AOD is mainly controlled by dust (i.e. arid locations). We will reformulate the sentence mentioning that "Aerosol water content has the largest contribution to the total AOD, with the notable exception of desert area where dust dominates, [...]"
- Page 26634, line 16-17: We did not claim that aerosol amount and water uptake are connected, but rather we claimed that a decrease of both is present in some regions (Eastern part of US and Western part of Europe). We will reformulate the sentence.
- Page 26634, line 21: The increase of Dust over the Middle East (ME) and Sahara Desert (SD) are mainly due to two reasons: (i) decrease of the rain events over ME

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and SD, (ii) decrease of coating effect (and therefore lower deposition) due to the decreased emissions in Europe of WASO compounds. (i) can be seen from Fig.6, while (ii) can be seen in Fig.6 of the manuscript, where the WASO compounds have a decreasing trend in the SD region.

- Page 26635, line 4: We will clarify this, mentioning that the increasing trends are due to the decreased precipitation events in the regions.
- Page 26635, line 4-5: In the model we can track both the amount of hydrophilic material as well as the species contribution to the overall extinction by scaling the total extinction in that grid box with the volume fraction of the hydrophilic material. Consequently, we can approximate the contribution of each species used in the internal mixture of the aerosol particles either by mass and by volume fraction, with the latter providing the contribution to the extinction. We will clarify the sentence in the revised version of the manuscript.
- Page 26635, line 13-16: Following the referee's suggestions, we calculated the average relative humidity in the two regions. As suggested in the manuscript, the RH averaged in the period 2001-2010 is  $\sim 73\%$  and  $\sim 59\%$  over East China and South Asia, respectively. Therefore we can conclude that the higher RH in the China region causes a more effective water uptake due to the exponential relationship between water uptake and relative humidity. As mentioned to referee #1, there is a positive trend present in the Indian region; nevertheless, despite such trends, RH values at the surface over East China are higher than the one over South Asia (India), being the difference between 6 and 30%. We will add this information in the revised manuscript.
- Page 26635, line 19: It is still unclear to us what is really causing the trends in South East Asia. Following simulation RCP00, this trends must be meteorologically driven, although we were not able to clearly identify what caused it. The only identifiable meteorological trend in the region is a decrease in the wind speed

(see Fig.7), which possibly causes a decrease of SS concentration over this region (as SS present a significant decreasing trends in this region).

- Page 26635, paragraph about SEA. Indeed Biomass Burning does not have linear trends in all regions. Nevertheless, the linear trends in this region for biomass burning is negative, as shown by Yoon et al. (2014a). Following Tab.1 and Fig.6 in the manuscript, we can see that SS and Dust AOD trends contribute directly only to  $\sim 5\%$  of the total trends.
- Page 26635, line 23: As direct we mean the direct decrease of AOD due to the decreases of the aerosol burden. As indirectly we mean a decrease of AOD due to the decrease in the aerosol water uptake by decreasing the hydrophilic mass. This sentence will be reformulated.

Page 26636, line 8-9: We will reformulate the sentence.

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 26619, 2014.

0.2	(a) EUS (Eastern USA) RCP85 Trend=-0.00539±0.00097 yr <sup>-1</sup>
0.0 -0.2	MOD Trend=-0.00492±0.00207 yr <sup>-1</sup>
0.4 0.0 - -0.4	(b) SAM (South America) RCP85 Trend=+0.00158±0.00425 yr
	MOD Trend=-0.00420±0.00519 yr <sup>-1</sup>
Se 0.2	(c) WE (Western Europe) RCP85 Trend=-0.00462±0.00120 yr <sup>-1</sup>
H-0.2	MOD Trend=-0.00426±0.00170 yr <sup>-1</sup>
0.4	(d) SD (Sahara Desert) RCP85 Trend=+0.00133±0.00123 yr <sup>-1</sup>
₹-0.4	MOD Trend=-0.00076±0.00208 yr <sup>-1</sup>
10.4	(e) ME (Middle East) RCP85 Trend=+0.00060±0.00099 yr <sup>-1</sup>
L- 0.0	MOD Trend=+0.00491±0.00332 yr <sup>-1</sup>
0.4 ·	(f) CA (Central Africa) RCP85 Trend=-0.00191±0.00140 yr
2-0.4	MOD Trend=-0.00224±0.00327 yr <sup>-1</sup>
ເສັ0.5 ເດັດດູ	(g) SA (South Asia) RCP85 Trend=+0.00157±0.00175 yr 1
0.0	MOD Trend=+0.00199±0.00311 yr <sup>-1</sup>
≥ 0.4 ⊽ ∩ ∩.	(h) EC (East China) RCP85 Trend=+0.00569±0.00357 yr 1
0.0 ·	MOD Trend=-0.00084±0.00449 yr <sup>-1</sup>
0.3	(i) SEA (SouthEast Asia) RCP85 Trend=-0.00267±0.00290 yr1
eg-0.3	MOD Trend=-0.00262±0.00425 yr <sup>-1</sup>
ö 0.2 <sup>∙</sup>	(j) NH (Northern Hemishere) RCP85 Trend=-0.00086±0.00052 yr 1
-0.2 .	MOD Trend=-0.00070±0.00127 yr <sup>-1</sup>
0.1	(k) SH (Southern Hemishere) RCP85 Trend=-0.00018±0.00030 yr <sup>-1</sup>
-0.1	MOD Trend=-0.00011±0.00069 yr <sup>-1</sup>
0.1	(I) GL (GLobe) RCP85 Trend=-0.00052±0.00030 yr1
-0.1	MOD Trend=-0.00043±0.00081 yr <sup>-1</sup>
2	001 2002 2003 2004 2005 2006 2007 2008 2009 2010 2011 Year

Fig. 1. Time series of deseasonalized MODIS and RCP85 AODs.

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0.2	(a) EUS (Eastern USA)	RCP85 Trend=-0.00539±0.00097 yr <sup>-1</sup>
-0.2		MIS Trend=-0.00268±0.00141 yr <sup>-1</sup>
0.4	(b) SAM (South America)	RCP85 Trend=+0.00158±0.00425 y
-0.4		MIS Trend=-0.00140±0.00388 yr <sup>-1</sup>
[s] 0.2	(c) WE (Western Europe)	RCP85 Trend=-0.00462±0.00120 yr1
- 0.0 -	And the second s	MIS Trend=-0.00215±0.00116 yr <sup>-1</sup>
크0.4	(d) SD (Sahara Desert)	RCP85 Trend=+0.00133±0.00123 yr1
O 0.0 -	and the second s	MIS Trend=+0.00020±0.00242 yr <sup>-1</sup>
0.4	(e) ME (Middle East)	RCP85 Trend=+0.00060±0.00099 yr <sup>-1</sup>
μ <u>0.0</u>		MIS Trend=+0.00677±0.00230 yr <sup>-1</sup>
0.4	(f) CA (Central Africa)	RCP85 Trend=-0.00191±0.00140 yr <sup>-1</sup>
C 0.0 -	anona and have	MIS Trend=+0.00076±0.00194 yr <sup>-1</sup>
UB 0.5	(g) SA (South Asia)	RCP85 Trend=+0.00157±0.00175 yr <sup>-1</sup>
<u><u> </u></u>		MIS Trend=+0.00210±0.00223 yr <sup>-1</sup>
≥ 0.4	(h) EC (East China)	RCP85 Trend=+0.00569±0.00357 yr1
0.0 -	source and the source	MIS Trend=+0.00208±0.00327 yr <sup>-1</sup>
0.3	(i) SEA (SouthEast Asia)	RCP85,Trend=-0.00267±0.00290 yr1
- 0.0 -		MIS Trend=-0.00179±0.00310 yr <sup>-1</sup>
0.2	(j) NH (Northern Hemishere)	RCP85 Trend=-0.00086±0.00052 yr1
0.0-		MIS Trend=+0.00012±0.00090 vr <sup>-1</sup>
0.1	(k) SH (Southern Hemishere)	RCP85 Trend=-0.00018±0.00030 yr <sup>-1</sup>
0.0 -		MIS Trend=-0.00045+0.00051 vr <sup>-1</sup>
0.1	(I) GL (GLobe)	RCP85 Trend=-0.00052±0.00030 yr <sup>-1</sup>
0.0 -		MIS Trend=-0.00016+0.00050 vr <sup>-1</sup>
-0.1 _ 20	001 2002 2003 2004 2005	2006 2007 2008 2009 2010 2011
		Year

Fig. 2. Time series of deseasonalized MISR and RCP85 AODs.

0.2	(a) EUS (Eastern USA) RCP85 Trend=-0.00539±0.00096 yr <sup>-1</sup>
-0.2	SEA Trend=-0.00299±0.00128 yr <sup>-1</sup>
0.4	(b) SAM (South America) RCP85 Trend=+0.00162±0.00438 yg
S-04	SEA Trend=+0.00080±0.00387 yr <sup>-1</sup>
10.2	(c) WE (Western Europe) RCP85 Trend=-0.00470±0.00123 yr <sup>-1</sup>
<u> </u>	SEA Trend=-0.00234±0.00147 vr <sup>-1</sup>
Q 0.4	(d) SD (Sahara Desert) RCP85 Trend=+0.00134±0.00126 yr <sup>-1</sup>
∩ 0.0 √	SEA Trand- 10 00039+0 00203 vr
.0.4 .5 0.4	(e) ME (Middle East) RCP85 Trend=+0.00061±0.00100 vr <sup>-1</sup>
Q 0.0	CEA Trend _ 0.0077010.00005.vrl
0-0.4 68 0.4	SEA Trend=+0.00770±0.00285 yr
0.0	for a contraction of the contrac
20.5	SEA Trend=+0.00182±0.00354 yr
0.0 g	
₩-0.5	SEA Trend=+0.00077±0.00246 yr <sup>-1</sup>
0.0 ·	Children China) RCP85 Trend=+0.00590±0.00362 yr 1
0.4	SEA Trend=+0.00146±0.00355 yr <sup>-1</sup>
0.0 alize	(i) SEA (SouthEast Asia) RCP85 Trend=-0.00276±0.00301 yr <sup>-1</sup>
Б-0.3.	SEA Trend=-0.00133±0.00234 yr <sup>-1</sup>
80.2 900.2	(j) NH (Northern Hemishere) RCP85 Trend=-0.00088±0.00052 yr <sup>-1</sup>
Ö-0.2	SEA Trend=+0.00097±0.00114 yr <sup>-1</sup>
0.1	(k) SH (Southern Hemishere) RCP85 Trend=-0.00018±0.00031 yr <sup>-1</sup>
-0.1	SEA Trend=+0.00115±0.00038 yr <sup>-1</sup>
0.1	(I) GL (GLobe) RCP85 Trend=-0.00053±0.00030 yr <sup>-1</sup>
0.0 -0 1	SEA Trend=+0.00098±0.00061 vr <sup>-1</sup>
2	001 2002 2003 2004 2005 2006 2007 2008 2009 2010 2011 Year

Fig. 3. Time series of deseasonalized SeaWiFS and RCP85 AODs.

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Fig. 4. Time series of modeled AOD from RCP85 for OrbView-2 overpass over South East Asia. Black: original data, Blue: deseasonalized data, Red: linear trend.



Fig. 5. Time series of deseasonalized SeaWiFS and RCP85 AODs. Model was masked for observational missing data.

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Fig. 6. Linear trend in %/year for surface precipitation in simulations RCP85 and RCP00.



Fig. 7. Linear trend in %/year for surface wind in simulations RCP85 and RCP00.

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