

Reply to A 4-D climatology (1979-2009) of the monthly aerosol optical depth distribution over the Mediterranean region from a comparative evaluation and blending of remote sensing and model products.

Anonymous referee #1

Reply : We would like first to thank the reviewer for the evaluation of our work and his positive comments. We have addressed all the comments and questions in detail, and clarified the mentioned points. Please find below our point-by-point replies highlighted in bold. Corrections in the text are indicated in italics.

Positives

- comprehensive comparison of available AOD retrieval data from satellite remote sensing
- separate assessment of spatial and temporal AOD correlations (to AERONET reference data)
- use (MODIS aqua) satellite data to describe local monthly AOD variability for 2003-2009 period

Concerns

- focus only on AOD data at a solar single wavelength
- composition data rely on interpretations (by two different) models
- indirect path to optical properties (assumed SSA and g for components - rather than AERONET)
- less useful (completely model based) for the 1979-2003 period

Reply : The first three points of concerns are answered further. Concerning the 1979-2003 period, we know that very few measurements are available over this period. However, it is an essential period for aerosol-climate studies in the context of the dimming/brightening phenomenon (Wild, 2009), and this is why we extended the 2003-2009 aerosol climatology over this period. The final product indeed includes only a sulfate trend from the LMDz-OR-INCA model before 2003, but it seems to be the dominant trend in the passed period (no significant trend over the Euro-Mediterranean region has been found for the other aerosols).

In order to reinforce the assessment of aerosols over the 1979-2003 period, we have added in the paper a comparison of the trends given by the models taking part in the ACCMIP exercise (Lamarque et al., 2013). With regards to observations, the TOMS data series has been completed between 1997 and 2000, and a new SeaWiFS product (Hsu et al., 2012) has also been added.

General comments

The paper explains multi-data source monthly average composites for the mid-visible aerosol optical depth (AOD) for the Mediterranean region (in the troposphere). A comprehensive inter-comparison among different satellite retrieval products and among model output of different simulations and assimilations is conducted. AOD data are assessed in comparisons to ground-based remote sensing statistics sampled at AERONET sites, to justify selected AOD data choices. Vertical stratification is scaled by active remote sensing from space, component sub-divisions are scaled by preferences in modeling and the temporal extension for the last 30 years is mainly tied to simulated sulfate AOD multi-annual trends by a single model according to prescribed changes in sulfur emission input to that model.

The AOD data-sets comparisons are the major part of the paper while the description of the different aspects of the climatology is relatively brief. There is a strong reliance on MODIS AOD data, also maintaining their regional variations of monthly AOD. Thus, if MODIS retrievals are correct (and there are larger uncertainties over land and there are sampling biases), then this climatology is well suited to address the reduction of the visible solar radiative energy that reaches the surface (for the 2003-2009 period in the Mediterranean). Still, to perform flux calculations, spectral variations must be addressed, not only for AOD but also for the properties describing the aerosol composition (single scattering albedo and asymmetry-factor) – especially in the context of

climate assessments (at TOA).

Reply : In order to address spectral variations in AOD, we have added an estimation of the Angström exponent from MODIS (figure below, section 4.3 in the paper). This parameter is derived from variation of AOD between 470 and 660nm over land (including bright surfaces with the deep blue algorithm) and between 550 and 865 nm over the sea. AOD can then be calculated at different wavelengths using equation 1 of the paper.

As far as the other optical properties (SSA, g) are concerned, Table 5 of the paper has been clarified further in the revised version and includes values for both visible and infrared spectral bands.

However it is worth mentioning that not all climate models can take into account this information. It depends on the spectral bands of their radiative schemes (Randles et al., 2013).

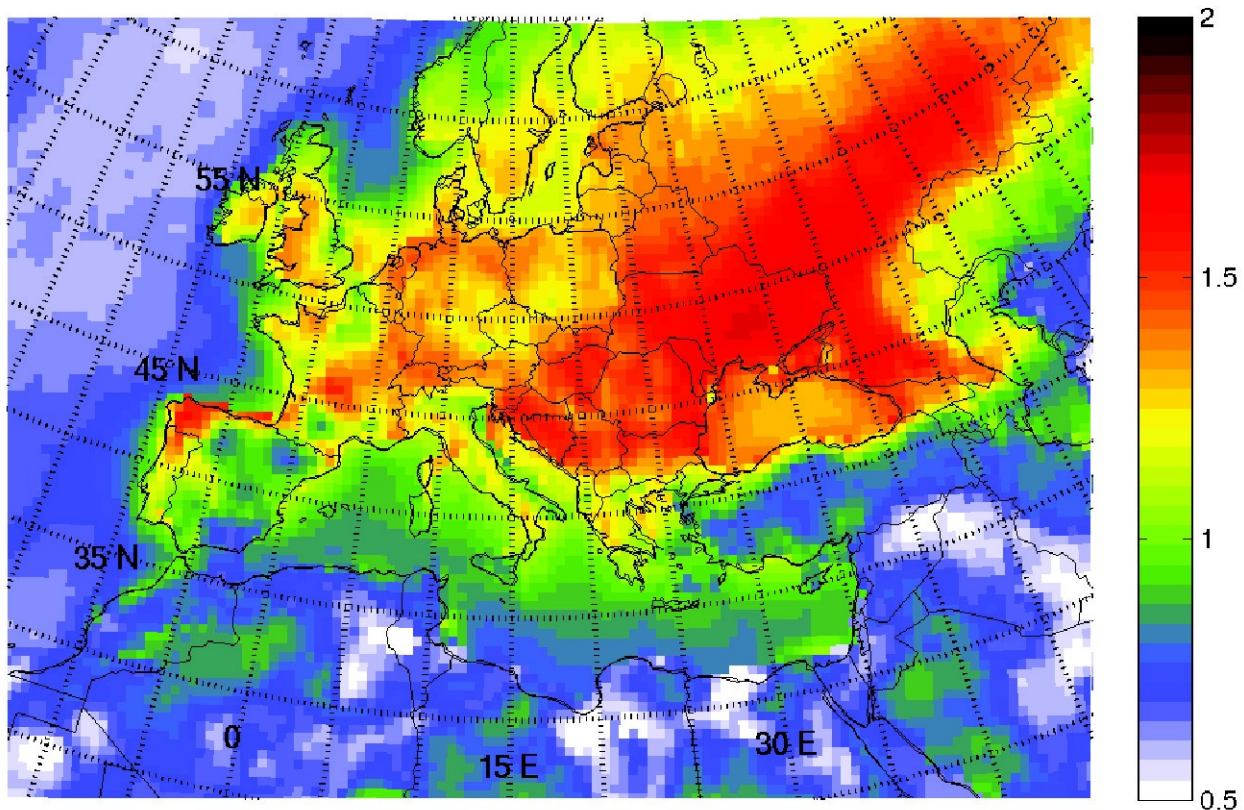


Figure 1 : Angstrom exponent (AQUA/MODIS) over the Mediterranean region for the 2003-2009 period.

The aerosol composition approach takes a rather indirect path, hereby introducing additional uncertainties. Rather than linking directly to AERONET optical properties, model simulations are applied to split the MODIS AOD data into components of SU, SS, DU, OC and BC. Now, these models usually include model size schemes, so there is an automatic split between AOD by coarse mode sizes (DU, SS) and by fine-mode sizes (mainly SU, OC, BC). Thus, the quality of the assumed split could be demonstrated with respect to the fine mode AOD and coarse mode AOD and in conjunction with assumed component SSA of Table 5 (which seem to be high in terms of absorption for OC and high on absorption or size for DU) via AAOD data to monthly statistics at AERONET sites. It should also be demonstrated that this climatology is superior to data by any single model (e.g. LMD-INCA) without the use of data from satellite remote sensing. In that sense, why not including the final climatology as additional ‘participant’ in AOD assessments of Figures 5 to 7?

Reply : This new climatology can be seen as a first attempt to have a realistic aerosol forcing available for climate models. Thanks to our intercomparison between each data set, including also AERONET ground-based measurements, we have shown several limitations for some

products (e.g., the underestimation of sulfate aerosols in RegCM-4, the problem in dust aerosols over Sahara in MACC, ...). Our reconstruction aims at selecting and combining the best product for each aerosol in order to get rid of the major drawbacks of the existing products. We are aware it is not a perfect product, but we believe that it is better than already existing products used for aerosol-climate studies.

Besides, the final total averaged AOD is not really worth to be added as « participant » in AOD climatology assessments of figures 5 to 7 since the result will be exactly the same as MODIS AOD, on which the final product is based.

Page 8499 Line 27 : As a result, the method used in the present work has been to evaluate and compare the total AOD data from each model, highlighting the limitations for some components (e.g. Sulfates in RegCM-4, dust aerosols in MACC). The final reconstruction is a first attempt to have an AOD climatology over the Mediterranean basin, that has none of the mentioned limitations.

Personally, I am biased that the detail and accuracy of AERONET should not be wasted only on evaluations, but should be an integral part of an aerosol climatology at least for the 1996-2012 period.

Reply : It is not possible to build a complete AOD gridded product from AERONET measurements. AERONET data are indeed only provided at a limited number of localized stations. Even if some methods to build a gridded product from localized measurements exist (e.g. the sea level, Calafat and Gomis, 2009), the AERONET network is limited by its spatial and temporal coverage of the basin. Very few measurements are indeed available over the sea and northern Africa, and most of the data last much less than 10 years. Among the 22 Mediterranean AERONET stations with more than 2,5 yr of monitoring compiled by Mallet et al. (2013), the total number of days with level-2 AOD at a given station averages at 1075, i.e. less than 3 yrs. Only 3 stations provide time series starting before 2000 with the oldest time series of Sede Boker in Israel dating back to early 1996 and the 2 other station of Erdemli in Turkey and Avignon in France starting in late 1999. Our reconstruction based on MODIS data and several model products (MACC, RegCM-4 and LMDz-OR-INCA) should be seen as a first attempt to have a realistic AOD climatology over the Mediterranean basin. Other reconstructions are obviously welcome.

While I am less enthusiastic about the climatology, the comparison of all these different satellite AOD retrievals alone warrants a publication, but here it would also help to be more clear, which particular versions are used or have been downloaded.

Reply : Precisions have been added for the versions which have been used and downloaded.

minor comments

when introducing SeaWiFS data you may mention that there is now a better product available, as described in Hsu et al. 2012 (the assessed version is very discouraging)

Reply : This new version (Hsu et al., 2012) has been added to the comparison.

when introducing CALIPSO data make sure to mention, which version is used (I assume version 2 and for difference between version 2 and 3 it could be referred to the Koffi et al 2012 paper)

Reply : We have used the version 3, this reference has been added.

Page 8480 line 1 : We use here the level-2 LIDAR product (CAL_LID_L2, version 3.01) for the "Atmospheric_Volume_Description" variable

replace at several places (e.g. 556, 594) 'inferior' with 'lower' or 'smaller', since inferior gives the impression of lower in quality

Reply : Corrected.

It should not surprise that GEMS and MACC ‘reproduce the AOD seasonality’ of MODIS (and MISR) since MODIS data are used in these assimilations.

Reply : Indeed this was an expected result. Added in the text.

Page 8487 line 9 : This was expected for GEMS and MACC which include MODIS AOD assimilation.

The pick of Aqua over Terra, may also be related to recently discovered issues with Terra (since 2005 increasingly lower AOD), it is not clear if these problematic data or updated data are here assessed - so from that perspective in the Aqua (/deep blue) data are certainly the safer choice

Reply : We agree with the reviewer, this point has indeed been mentioned in Levy et al. (2010), and has been added in the text.

Page 8489 line 27 : Besides, Levy et al. (2010) have recently underlined discovered issues with TERRA (notably an increasingly lower AOD since 2005). They have also shown at the global scale for dark-target products over land that Terra AOD presented a negative trend compared to AERONET measurements while comparisons between AERONET and Aqua AOD have shown no trend.

The pick for the best models in order to extract info on composition seems tied to the best scores with respect to total AOD to AERONET but not with respect to AOD components. Another aspect is that the regional choices are picked based on MODIS data (which are not really a reference). Note, that for the fine/coarse mode split, AERONET are available as reference. The RegCM4 is chosen to represent dust, but there is some concern about the rather poor correlations of these data to AERONET is the Taylor plots. Good scores for coarse mode AOD with respect to AERONET would be more convincing.

Reply : We are aware of the difficulties in evaluating the different aerosol contributions to total AOD. We could indeed use the separation between fine and coarse mode (available in MODIS and AERONET), considering fine aerosols are essentially sulfates, BC and organic aerosols. However, in this case we would ignore the fine fraction of dust aerosols, as shown in Shindell et al., 2013. It is the same problem for the absorption aerosol optical depth (AAOD), which could be estimated for the black carbon and dust aerosols. However dust aerosols are more or less absorbing depending notably on their size and hematite content, and some organic aerosols are not exclusively scattering (Mallet et al., 2013).

As a result, our approach is limited to the evaluation and comparison of the total AOD data from each model. MACC has got the best scores against AERONET ground-based measurements, but since its AOD seems to be underestimated over the Sahara compared to all other satellite retrievals, we decided not to take dust aerosols from this model product. We have preferred using dust aerosols from the RegCM-4 model, forced by the ERA-INTERIM reanalysis and including a complete dust interactive scheme validated in several studies (e.g. Solmon et al., 2008, 2012 ; Nabat et al., 2012). The poor correlations of RegCM-4 to AERONET are linked to the underestimation of sulfate aerosols by this model.

Besides, we hope the coming ChArMEx campaign in summer 2013 could help us bringing more validation data for our models, notably through case studies.

Section 4.3 (discussion) has been modified to add these explanations.

Sulfate aerosol seems to dominate among the fine-mode (not necessarily ‘anthropogenic’ [line 812 ... ‘anthropogenic dominated’ as in line 834 sounds better]) sizes, but organics are not completely zero and larger not just in TEG97 [line 831] but also in GEMS.

Reply : Corrected.

Page 8491 line 27 : Organic carbon (OR) have lower AOD, ranging from 0.02 (MACC and ACCMIP) to 0.07 (TEG97) on annual average over Europe. All models agree on the very low AOD for black carbon (BC) aerosols, estimated between 0.01 and 0.02 over Europe.

At the stage in the middle of the paper, it is unclear, how the MODIS Aqua defined total AOD is split into subcomponents, because the sum of DU and SS of RegCM4 and SU, BC and OC of MACC will locally differ from the Aqua total AOD. It would help to show the assumed AOD maps for all five components in an extra figure since the compositional separation is also used to define the AOD vertical distribution, based on the CALIPSO type identifiers. Please refer already here to the later description and definition.

Reply : The AOD maps for all five components have been presented in Figure 15, which has been modified. Details about the split into subcomponents in the final product are given in Section 4.1, a reference has been added at the end of the Section 3.3.

The relative vertical distributions of CALIPSO are determined for each of the 16 regions. Are there now sharp altitude gradients at the layer boundaries? Some explanations are needed.

Reply : Even if CALIOP could have more difficulties to retrieve aerosols near surface, such a sharp gradient in the bottom layer is also observed by airborne lidar profiles (Dulac and Chazette, 2003).

Page 8493 line 13 : These sharp gradients near surface are in agreement with airborne lidar profiles (Dulac and Chazette, 2003).

For the pre-2000 data, the LMD simulations seem quite agreeable to the NOAA Patmos data. I would hesitate even to show (broadband sensor data related) MVIRI data, which are not only biased low and have apparent discontinuities with sensor changes, but also seem to drift to lower values with space deployment time (these data are certainly less mature than Patmos, as also AVHRR had to deal and correct for sensor changes and overpass-time drifts).

Reply : We believe that Meteosat/MVIRI data are worth in this study as they represent 14 years of AOD data over the Mediterranean basin, and have been used in several former studies (e.g. Moulin et al. 1997, Chiapello et al. 2002). These discontinuities that we found have never been shown before, which is for us a message to give to data users. Text has been clarified.

Page 8496 line 2 : Figure 14 illustrates the issue for MVIRI data, already used in several former studies (e.g. Moulin et al. 1997, Chiapello et al. 2002).

Since stratospheric AOD enhancements in the years after the ElChichon and Mt.Pinatubo volcanic eruptions were removed, maybe the title and text should refer to a 'tropospheric' AOD data.

Reply : The title has been modified and now includes the word « tropospheric ».

Aha, now the component normalization is explained. Apparently, multi-annual (7 year) monthly averages are used to the effect that the AOD of the climatology for a month of particular year (e.g. 2006) will not be the same as that for MODIS Aqua (... only similar over the 2003-2009 period).

Reply : In fact, each month of the 2003-2009 period in the final product is the same as in MODIS for total AOD. The normalization is applied to each month individually. This has been clarified in the new version.

Page 8497 Line 9 : For each month of the period 2003-2009, the raw AOD coming from the model is normalized by the total AOD from MODIS. This normalization is applied to each month individually so that total AOD of the reconstruction is identical to AQUA/MODIS for every month.

I wonder if a 60m vertical resolution is actually necessary or overkill (there are significant uncertainties with the CALIPSO data). I rather would focus on a finer spatial resolution than just the 16 areas.

Reply : We agree that a 60m vertical resolution is actually not necessary, and could be misleading given uncertainties with the CALIPSO data. Consequently, we have interpolated the CALIPSO data to the 60 MACC vertical levels. Having more than 16 areas will reduce the

number of available data for each zone, so that profiles would be more uncertain. 16 areas seems to be a compromise between the number of data and the size of the region.

Page 8494 Line 19 : Given the uncertainties in CALIOP data, the vertical resolution for our final product is based on the 60 MACC vertical levels.

The MACC resolution is 1.125 degree. I assumed the 0.5deg resolution is based on (linear?) interpolations?

Reply : Linear interpolation as now indicated in the manuscript (page 8497 line 3).

Is it correct that the coarse mode monthly AOD varies from year to year between 2003 and 2009 based on the RegCM-4 model? What happen with the coarse mode AOD for the years before the year 2000 - just the 2003-2009 multi-annual monthly data? Please clarify.

Reply : Each aerosol type varies from year to year between 2003 and 2009. Before 2003, we apply the 2003-2009 multi-annual monthly data, with the addition of the trend for sulfate aerosols. Text has been clarified.

Page 8498 Line 4 : As a result, contrary to the 2003-2009 period, there is no year to year variability in the reconstruction between 1979 and 2003, but only a decreasing trend for sulfate aerosols.

The temporal trend validation is largely a LMD INCA model evaluation, indicating that the (sulfate) emission changes are relatively well represented in that model... which begs the question, why this climatology is needed (a demonstration of a better performance over the model would go a long way).

Reply : In order to assess this sulfate trend between 1979 and 2009, we have added in the comparison the data coming from the ACCMIP exercise (Lamarque et al., 2013), whose simulations include compulsory time-slices (1980 and 2000). The following table (added in the new version) presents the sulfate AOD differences between the timeslices 2000 and 1980 for each ACCMIP model. The decrease in sulfate AOD ranges from -0.05 to -0.19 over Europe, and from -0.03 to -0.08 over the Mediterranean Sea. LMDz-OR-INCA is a median model, showing sulfate trends close to the ACCMIP mean. It should also be noted that no significant trend has been noticed for BC and OR aerosols over Europe and the Mediterranean Sea (the ACCMIP mean trend is -0.00 for BC and OR).

Page 8496 line 10 : It should also be noted that no significant 1140 trend has been noticed for BC and OR aerosols over Europe and the Mediterranean Sea (the ACCMIP mean trend is -0.00 for BC and OR).

Region	CICERO	GFDL	GISS	GISS-E2-R-TOMAS	HadGEM2	LMDzORI NCA	NCAR-CAM3.5	NCAR-CAM5.1	Mean
Europe	-0.14	-0.14	-0.19	-0.05	-0.10	-0.12	-0.10	-0.05	-0.11
Med Sea	-0.06	-0.08	-0.06	-0.03	-0.05	-0.05	-0.05	-0.03	-0.05

Table 1 : AOD differences between ~2000 and ~1980 in the ACCMIP models.

In the conclusion it is claimed that there ‘is no means to evaluate properly the separation of components’. The argument that dust (or sea-salt) may also contribute to the fine-mode is rather weak, since most of the the DU AOD and SS AOD will contribute to the coarse mode.

Reply : Dust aerosols have a submicronic fraction that contributes significantly to AOD, as shown by model studies (e.g. Nabat et al., 2012). This fine dust fraction is consequently a problem to compare fine MODIS AOD with the anthropogenic aerosols of the model AOD in the Mediterranean region where dust is relatively frequent, or even dominant. Moreover, uncertainties are still important in this kind of product (Yu et al., 2009), showing no consensus on an observationally-constrained anthropogenic AOD.

Page 8499 line 23 : Previous studies (Lee and Adams, 2010, Shindell et al., 2013) have

determined the dominant mass type in different locations around the world, which is more difficult in this regional study (except for dust over the Sahara desert) as the Mediterranean area is affected by mixtures of different aerosols. The separation between fine and coarse mode available for example in the MODIS data set (Remer et al., 2005) could be used considering fine aerosols are essentially sulfates, BC and organic aerosols. However, the fine fraction of dust aerosols would not be taken into account in this case. Moreover, uncertainties are still important in this kind of product (Yu et al., 2009) showing no consensus on an observationally-constrained anthropogenic AOD.

The same problem is raised for the absorption aerosol optical depth (AAOD), which could be estimated with the black carbon and dust aerosols. Dust aerosols are however more or less absorbing depending notably on their size and hematite content. Some organic aerosols are not exclusively scattering (Mallet et al., 2009).

The exclusion of the volcanic data should have been mentioned right from the start and day-to-day can certainly not be addressed with monthly averages.

Reply : Added in the introduction.

Page 8475 line 17 : It should be noted that the present work focuses on tropospheric aerosols, and that consequently the consideration of volcanic aerosols will only be raised in the discussion part.

The attempt to address the optical properties by components with averages for components in Table 5 is rather simple as they ignore assume fixed size, shape and refractive indices and ignore their variations. This is disappointing, since (via AERONET) monthly reference data for these properties actually exist and could be tested: ftp://ftp-projects.zmaw.de/aerocom/aeronet/STATISTICS/grd_1203 ‘osunal’ (direct attenuation data AOD and ANG) and ‘oskyl’ (inversion data: AOD, ANG, AAOD, fineAODfraction)

I do not always agree with SSA and g values for the component data, listed in Table 5. In the table below I attach values I would use (based on single scattering simulations). I am concerned about the relatively low SSA values for DU and OC. The low SSA values for DU seem plausible only for very large dust sizes (radii of ca 4um!) and I wonder if these are realistic for transported dust over the Med. Regions. Also the properties for BC, as demonstrated, are strongly size-dependent.

Reply : The table 5 (below) has been precised : refractive index and effective radius have been added. These calculations have been carried out with a Mie code from effective radius observed in the ESCOMPTE campaign (Mallet et al., 2003) and refractive indices published by Krekov (1993). We have also separated the fine and accumulation modes for dust and organic aerosols, which could explain that reviewer has found low SSA values for these types. Uncertainties have been taken into account using an error range of 10 % for the effective radius. Associated intervals are indicated between square brackets.

These optical parameters are provided as advised values for aerosol-climate studies over the Mediterranean region. We know we would need a further comparison with all these optical parameters but this is beyond the scope of the paper. Another work concerning the absorption issue has just been submitted to ACP (Mallet et al., 2013) in the same special issue as the present work.

Type	Reff (μm)	Std (μm)	Refractive index 550nm 1 μm	SSA 550nm	SSA 1μm	g 550nm	g 1μm
SS	0.35 [0.32 0.38]	1.75	1.45-0.000031i 1.45-0.00006i	0.99 [0.99 0.99]	0.99 [0.99 0.99]	0.72 [0.71 0.72]	0.73 [0.73 0.73]

SD	0.036 [0.032 0.040]	1.97	1.51-0.008i	0.95 [0.95 0.95]	0.93 [0.92 0.93]	0.62 [0.60 0.63]	0.52 [0.49 0.54]
	0.36 [0.32 0.40]	1.98	1.50-0.008i	0.85 [0.84 0.87]	0.91 [0.90 0.92]	0.74 [0.73 0.75]	0.72 [0.71 0.72]
OR	0.027 [0.024 0.030]	1.86	1.45-0.001i	0.99 [0.99 0.99]	0.98 [0.97 0.98]	0.52 [0.49 0.54]	0.35 [0.32 0.38]
	0.29 [0.26 0.32]	1.95	1.47-0.001i	0.98 [0.98 0.98]	0.99 [0.99 0.99]	0.73 [0.73 0.73]	0.71 [0.71 0.71]
BC	0.028 [0.025 0.031]	1.94	1.83-0.74i	0.32 [0.30 0.33]	0.24 [0.22 0.26]	0.43 [0.40 0.45]	0.31 [0.29 0.33]
			1.91-0.68i				
SU	0.040 [0.036 0.044]	1.74	1.52-0.0005i	0.99 [0.99 0.99]	0.99 [0.99 0.99]	0.53 [0.50 0.55]	0.35 [0.32 0.38]
			1.51-0.0005i				

Table 2 : Aerosol optical properties obtained from Mie calculations for different particle types. Values between square brackets correspond to an uncertainty range of 10% on the effective radius. SSA=Single Scattering Albedo, g=asymmetry factor. These properties are provided in dry state. Changes on optical properties should be made depending on the relative humidity (Mallet et al., 2003).

Page 8500 Line 19 : For that reason, we propose in Table 6 values for single scattering albedo (SSA) and asymmetry parameter (g). These values come from calculations carried out with a Mie code from effective radius observed in the ESCOMPTE campaign (Mallet et al., 2003) and refractive indexes published by Krekov (1993). An uncertainty range of 10% has been taken into account for the effective radius: the associated values for SSA and g are indicated between square brackets. All these parameters are provided in dry state. Fine and accumulation modes have been separated for dust and organic aerosols.

Table 1 the spatial resolutions are confusing as the higher resolutions are not actually applied in the (coarse) 1 degree assessments. In addition, also MODIS (10km) and MISR (17.6km) official aerosol products are available at higher resolutions than indicated in the Table. I would the original resolution and the resolution used.

Reply : Monthly level-3 products used in this study have a 1°x1° resolution. Original MODIS products are indeed available at 10 km resolution, but they are level-2 products. We have preferred using level-3 products to have a better robustness in data. The same idea has been applied for MISR, whose level-3 products used in this study have a 0.5°x0.5° resolution.

Figure 5 from the seasonal difference in Figure 4 I would expect larger deviations in Figure 5. The box whisker Figure 5 needs definitions as the boundary PDF values (20%/80% for the box?, 95%/5% for the dashed line?, central line for the median (50%)?, what is the meaning of the individual small circles?)

Reply : The thick black line represents the median while the central box is limited by the first and third quartile. The whisker limits correspond to the inner quartile range multiplied by 1.5. All the points out of this range (outliers) are indicated with small circles. These explanations have been added in the methodology section (3.1), and the legend of Figure 5 has been detailed.

Figure 6 and 7 both figures apparently indicate that at some sites there are even ANTI-correlations at least spatially ... so that even the 'better' retrievals of MODIS and MISR are far from perfect.

Reply : There are indeed AERONET stations which show anti-correlations for some products

(e.g. MERIS) but not for MODIS and MISR.

Figure 8 the label in the figure display 'OR' while 'OC' is mentioned in the caption. Similarly, in the text both OR and OC are both mentioned although I think the same is meant (is confusing)

Reply : Corrected : all 'OC' have been replaced by 'OR'.

Figure 14 this is a sorry figure. Since the data are so poor, why even show them, especially since they are irrelevant for the climatology.

Reply : As mentioned previously in this reply, we think it is worth mentioning the discontinuities of Meteosat data with this figure, in order to warn future data users.

Figure 15 I am struggling with the figure and I strongly suggest to remove it unless it is strongly improved. The text is quite clear about the normalization and the temporal extension - but not this figure. Some diamonds indicate a component, so I assume the lower right refers to dust. I have no idea what should be in the pink box and using a black background for a box with black letters is not too smart.

Reply : This figure has been modified : the scheme has been removed given that the explanation in the text with equations was clear enough. Only the maps with the contributions of different aerosols have been kept.

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