Dear Editor,

First, we would like to thank the reviewer for all the remarks. We made important modifications in the new version, mainly by removing part 3.3.3, focused on the satellite observations (see details bellow). We also re-worked the AERONET part following the different points highlighted by the reviewer.

1. General Comment

This paper presents an overview of the temporal and spatial characteristics of aerosol absorption in the Mediterranean utilizing AERONET ground based observations and available satellite measurements. The ground-based component of the analysis is methodologically sound, and constitutes an important contribution in the regional and temporal analysis of AERONET aerosol absorption observations.

The satellite based analysis of three aerosol single scattering albedo data sets, however, is not well carried out. It lacks a consistent methodology in the description of the products, their expected accuracies, and associated validation/assessment analysis. The authors seem particularly unfamiliar with the OMI and MODIS Deep Blue SSA products. In their analysis and conclusions they tend to exaggerate the quantitative significance of the temporal and spatial variability of MISR SSA, a product admittedly meant to be used categorically rather than quantitatively. If the authors are indeed interested in assessing the strengths and limitations of SSA retrieval techniques from space observations they should carry out a direct quantitative evaluation of these products using AERONET, SKYNET and other data bases as reference. Such an evaluation effort will benefit from the participation of scientists associated with the OMI and MODIS SSA products as well.

As mentioned in the first version of the article, the analysis of satellite data included in this article was a first exploitation of SSA satellite observations over the Mediterranean basin. The aim of this work was to present the regional SSA pattern obtained over this specific region from satellite observations, which is complementary to local AERONET data, and to present possible geographical gradient depending on the aerosol types. Anyway and as mentioned by the reviewer, this part is not robust enough in its present form. We already attempted to evaluate SSA in the MISR, MODIS and OMI products over this region using AERONET data (MPASS website) but Level 2 data conjoined with satellite over passing was not sufficient for producing relevant statistical estimations. This is why we did not present AERONET versus satellite SSA comparisons in this work.

To our knowledge, a rigorous evaluation of SSA MODIS-DB, OMI and MISR satellite products does not exist at the present time (except the work of Jethva and Torres, ACP 2011, for OMI) and clearly requires an enlarged number of AERONET sites (certainly at the global scale), including AERONET sites associated with large aerosol concentrations (AOD higher than 0.4 in the visible bands) and affected by different aerosol types (smoke, dust and pollution). In that respect, the amount of data over Mediterranean basin seems to be not sufficient. Therefore, we have decided to remove the last part of the article focused on satellite analyses (3.3.3).

We plan to reanalyze these results and possibly submit a second article focused on satellite data analysis. A specific study (evaluation of SSA from MISR, MODIS Deep Blue and OMI) is now ongoing using a large number of AERONET sites as mentioned below but is beyond the scope of the present study. Hence, we have re-written the introduction and conclusion, and changed the title of

the article which is now: «<u>Absorption properties of Mediterranean aerosols obtained from multi-year ground-based remote sensing observations</u>».

Below, we tried to answer to reviewer questions including those on the satellite part, which will be helpful for the on-going work.

<u>2. Specific Comments</u>

<u>Page 9274</u>
Line 10. The authors characterization of AERONET SSA retrieval accuracy is inaccurate. Dubovik's 2000 paper states a SSA accuracy of 0.03 for AOD > 0.2 only for water soluble aerosols. The same paper states the accuracy of desert dust and biomass burning SSA as 0.03 for AOD > 0.5 and solar zenith angle larger than 50 degrees. The Dubovik et al papers (2000, 2002) describes the SSA accuracy for AOD (440 nm) < 0.2 as 0.05 ~ 0.07. The authors should present a more detailed discussion of version 2.0 assurance criteria based on Holben et al, 2006 (AERONET's version 2.0 quality assurance criteria, SPIE, Volume 6408, SPIE paper number 6408-27).

This is effectively right and this point is now presented in more detail in the new version (Part 2.1). We've added the following sentence: "…whereas SSA uncertainty is estimated to be ±0.03 for AOD (440 nm) >0.2 for water soluble aerosols and for AOD (440 nm) >0.5 (zenith angle larger than 50 degrees) for desert dust and biomass burning particles. Finally, for AOD (440 nm) < 0.2, the SSA accuracy is ±0.05-0.07 (Dubovik et al., 2000, 2002). In the present study, the analysis is mostly focused on AAOD and SSA for AERONET level 2.0, cloud-screened and quality-assured AOD (Smirnov et al. 2000) and level 2.0 inversion products (Dubovik et al. 2002). The AERONET's Version 2.0 quality assured aerosol products were used. The criteria used for Version 2.0 quality assurance are described in details by Holben et al. (2006)."

<u>Page 9276</u>
Line 4. Mention the spatial resolution of the monthly MISR SSA products. What is the typical number of days per grid-point per month when MISR SSA is retrieved? How representative of a monthly average are these measurements?

In this study, we have directly used the seasonal MISR-SSA product available on the https://eosweb.larc.nasa.gov/PRODOCS/misr/level3/download_data.html website. The spatial resolution for the aerosol product is 0.5 x 0.5 degree grid (indicated in the Table 3).

Following this remark, we have now downloaded the "Single_scatter_albedo_count" MISR product, i.e, number of observations (not exactly the number of days) per grid-point for the two seasons (MAM & JJA) averaged for the 2000 to 2010 period (Figure 1 below). We can clearly see that the number of MISR observations is higher during summer (right) compared to spring (left), as might be expected. During summer, the number of observations is generally higher than 80 over a large part of the Mediterranean basin, except over the Tyrrhenian Sea and Gulf of Lion (~ 50 observations). The number of observations decreases significantly over the European continent, with values lower than 50. Over Northern Africa, the number of observations is very high and reaches ~150 for certain regions during summer. In contrast, more caution should be used during spring, as the number of observations decreases over the Western basin, with values around 20-50. Over the eastern part, the new figure displays numbers of observations ranging from 50 to 70. Over Northern Africa, we obtain an acceptable number of observations (larger than 50 over a large part of the domain) except over

Morocco, Algeria and Northern Tunisia, where caution should be used when analyzing MISR observations.

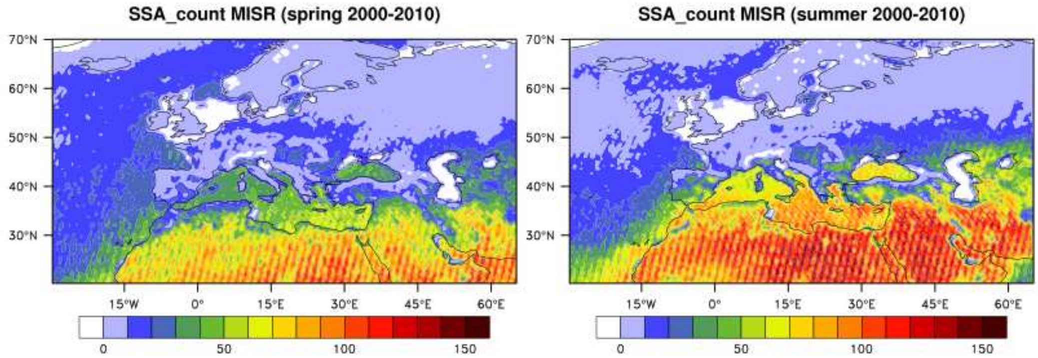

Figure 1. Seasonal SSA count for MISR sensor, for spring and summer (mean of 2000 to 2010 period).

Concerning the representativeness of such averages, it should be noted that the retrieval of detailed aerosol properties from remote-sensing observations is much more sensitive to scene conditions than the AOD retrieval. Among such aerosol properties as particle size, shape, and SSA, SSA is by far the most difficult to constrain, depending on the scene, provided there are good retrieval conditions. SSA retrievals are obtained under a more limited set of conditions than AOD, which bears on the "representativeness" of the product on the global scale. An actual work is made for providing a "quality flag" for aerosol type. Specifically situations with low sensitivity of retrievals to SSA were outlined as the situations with AOD < 0.2, when the surface is very bright and when the AOD is very variable within a retrieval region.

Line 8. In spite of the stated 'categorical' nature of the MISR SSA product, later in the paper it is used quantitatively with an implicitly assumed unprecedented accuracy. If the MISR SSA is to be used quantitatively, a reference-supported statement of the expected accuracy should be provided, indicating and what the possible sources of error are. Provide references of correlative analysis of MISR SSA with AERONET or other independent measurements. Also provide references on MISR assumptions on real and imaginary refractive index.

For the time and to our knowledge, there is no complete SSA evaluation with AERONET Level 2 derivation. Initially, we tried to perform SSA scatter plots (not shown) using the MAPSS database, but the number of points was too low for making rigorous statistical calculations. A similar attempt was made by Kahn et al. (JGR 2005; 2010), but even the Level 1.5 AERONET data are much too sparse for statistically meaningful validation of SSA. Anyway, as mentioned above, the additional work is going on to evaluate the different SSA satellite products at the global scale using more AERONET sites, including those characterized by large aerosol load (AOD larger than 0.4 in the visible wavelengths).

The assumptions on real and imaginary refractive index used for the different aerosol types (reported in the Table 2 of Kahn et al. (2010) Journal of Geophysical Research paper) in the MISR algorithm are the following: for dust particles, the real refractive indices are 1.45 for all spherical particles (components No.1- 6 in Table 2 of Kahn et al. (2010)), and for the "medium_grains" (No. 19) and "coarse_spheroids" (No. 21), 1.51, at all wavelengths. The imaginary part for the "medium_grains" and "coarse_spheroids" are 0.021 at 0.55 microns, 0.0011 at 0.672 microns, 0.000721 at 0.866 microns. In addition, the spectral imaginary parts used for particles No. 8 and No. 14, named

"spherical_absorbing_0.12_SSA_0.9" and "spherical_absorbing_0.12_SSA_0.8", are 0.0147 for all wavelengths and 0.0325 for all wavelengths, respectively.

Line 13. At what temporal resolution and to what SSA accuracy can MSIR separate adjacent air masses in terms of SSA? Are MISR statistics suitable for an analysis of regional patterns as suggested as the aim of this work?

SSA temporal resolution depends on retrieval conditions at each location. In general, the values would be "seasonal," except for plumes, where SSA can be retrieved with greater confidence. For example, a recent study based on a field validation case, shows the ability to separate aerosol air masses based on MISR-retrieved properties (see Figures 6 and 7 of Kahn, R., A. et al. (2009). Desert Dust Aerosol Air Mass Mapping in the Western Sahara, Using particle properties derived from space-based multi-angle imaging, Tellus 61B, 239-251). In general, these authors used the combination of size, shape, and SSA constraints from MISR to assess aerosol type. In addition and due to the presence of marked different aerosol regimes, the Mediterranean region appears quite appropriate for testing/using SSA products and inferring regional gradients, between North and South, for example between pollution and mineral dust aerosols.

Line 25: Provide reference for stated accuracies of OMI retrieved products.

To our best knowledge, the Jethva and Torres (ACP, 2011) article presents the best-documented uncertainties on OMI-derived AOD and SSA, based on comparisons with AERONET retrievals for the two parameters.

Line 26: The OMI near UV algorithm does not retrieve SSA at 500 nm. The retrieval is actually carried out at 388 nm, and converted to 500 nm [Torres et al., 2007]. What is the spatial resolution of the monthly OMI SSA product. What is the typical number of days per grid-point per month when OMI SSA is retrieved? How representative are these monthly statistics? Provide references of correlative analysis of near UV retrieved SSA with AERONET [JGR, Torres et al., 2005; JGR, Torres et al, 2007]

Thank you for this remark on the estimation of SSA at 500 nm, which will be helpful for the subsequent work on SSA satellite evaluation. Concerning the spatial resolution, the OMI SSA monthly product is about 1*1°. As for MODIS, the number of days per grid-point is unfortunately not provided in the monthly product, which requires downloading directly the daily product to calculate it.

<u>Page 9277</u>
Line 1. Provide a reference to support the statement on the higher reliability of OMI retrievals over land.

OMI should have about the same quality over land and water. Aerosol type, vertical distribution, and AOD should matter more than surface type for the OMI retrieval.

Line 7. OMAERUV assumes real refractive index and particle size distribution. No assumption on imaginary refractive index is made.

Thank you for this remark. This point will be mentioned in subsequent work on SSA satellite evaluation.

Line 8. What is the typical number of days per grid-point per month when MODIS SSA is retrieved? How representative are these monthly statistics? Provide references of correlative analysis of MODIS retrieved SSA with AERONET observations.

The number (for Deep Blue observations) of days per grid-point and per month is not a product provided in the monthly MODIS data. In that sense, its estimation requires downloading daily MODIS data, which is difficult due to the size of folders. Anyway, we will certainly use this technique for estimating the number of days per month in subsequent work.

As mentioned below, unlike OMI, there is no inter-evaluation between SSA-MODIS DB and AERONET at this time. A recent study (Shi et al., AMT, 2013) investigated the MODIS-DB AOD derivation but not the SSA. This motivates the second work we propose on SSA evaluation.

<u>Page 9280</u>
Line 8. In addition to retrieval accuracy, an important difference between versions 1.5 and 2.0 is instrument calibration. Version 2 data is produced after the instrument is recalibrated. Aerosol inverted products are particularly dependent on calibration. Therefore, version 1.5 and 2.0 sky-radiance-derived data should not be combined. Please comment.

This is effectively right and this specific point is now included in the new version. In this study, we did not combine level 2 and level 1.5. Level 2 was used to derive AAOD and AAE. In parallel and just for the part 3.2.2, we have used level 1.5 for deriving information about the possible influence of other absorbing particles than mineral dust over the Mediterranean basin.

Line 6. The presence of OC in biomass burning plumes has been confirmed by OMI satellite observations [Jethva and Torres, ACP, 2011]

This specific point and the associated reference are now added in the new version (part 3.2.2). "In addition, the work of Kirchstetter et al. (2004) and Jethva and Torres (2011) confirm the high AAE values of smoke aerosols when taking into account wavelengths shorter than 440 nm."

<u>Page 9282</u>
Line 10. What it is even more important AERONET does not make any absorption measurements at wavelengths shorter than 440 nm where the influence of brown carbon absorption is more prevalent.

This statement is correct and now reported in the text (part 3.2.2). "Furthermore, most AERONET sites do not provide observations in near UV where the influence of brown carbon is most prevalent".

Line 15. The work of kirchstetter et al [JGR, 2004] and Jethva and Torres [ACP, 2011] confirm the high AAE values of smoke aerosols when taking into account wavelengths shorter than 440 nm.

This specific point (and the associated references) is now added in the new version (part 3.2.2) of the new version." In addition, the work of Kirchstetter et al. (2004) and Jethva and Torres (2011) confirm the high AAE values of smoke aerosols when taking into account wavelengths shorter than 440 nm."

<u>Page 9284</u>
Lines 2-4. This is a highly speculative statement that adds nothing to the discussion, taking into account the documented uncertainty of AERONET SSA retrieval (0.03).

We agree with this remark and we have now removed this sentence in the new version.

Page 9286
Line 9. It should be mentioned that the North-South gradient observed by OMI is consistent with AERONET AAOD observations in Figure 3 while MISR shows an opposite trend.

Thank you for this remark, this point will be discussed in the subsequent study.

Page 9287
The author's interpretation of the MISR SSA data in this paper is ambiguous and confusing at best. On page 9275, the authors clearly state that the MISR SSA data is to be treated as' categorical' rather than quantitative. They seemed to stick to that position when discussing the N-S gradient absorption clearly present in both AERONET (Figure 3) and OMI observations but missed by MISR (Figures 13 and 14) which actually shows a reversed gradient. However, in the analysis of data over the Mediterranean the authors suddenly shift to a detailed numerical interpretation of the MISR data assigning to it unprecedented accuracy in satellite derived SSA. They report an alleged east (0.96~0.98) - west (0.97--0.98) on the basis of a seasonal difference of ~ 0.01 in single scattering albedo difference between the two regions over different time periods. That regional difference would be statistically insignificant even for AERONET measurements whose accuracy is +/- 0.03. How can then any significance be given to such small number based on a parameter not meant to be used quantitatively, without regard to the poor temporal and spatial sampling of the MISR sensor and undocumented accuracy of the SSA product?

We agree with this and now we have removed the entire section on satellite analyses in the new version of the abstract, text and conclusion. Indeed, based on SSA uncertainty linked to MISR retrieval, the West-East gradient is too small to argue about a realistic geographical gradient. This could be observed for a specific event as shown in the following study for a single volcanic plume (Kahn, R.A., and J.A. Limbacher, 2012. Eyjafjalljökull Volcano Plume Particle-Type Characterization from Space-Based Multi-angle Imaging. Atmosph. Chem. Phys. 12, 9459–9477, doi:10.5194/acp-12-9459-2012) but not in this case for a seasonal mean.

The existence of the alleged east-west gradient in the AERONET data can't be supported either based on the data presented in the paper. There are no AERONET measurements of SSA in the Mediterranean geographically representative of the satellite measurements. An analysis of AERONET SSA data in the Mediterranean coastal and island stations in Table 2 shows statistically insignificant differences (~0.92) for both basins.

We agree with this remark and we used the term "moderate" in the first version of the article, which is maybe not fully accurate. We've now re-written this specific point in the abstract and conclusion using the following sentence:" AERONET level-2 sun-photometer data indicate a possible East-West gradient, with higher values over the eastern basin ($AAE_{East.} = 1.39$ / $AAE_{West.} = 1.33$)".

Page 9288
Lines 10-12. Can any conclusion really be derived about the differences in absorbing properties between the Eastern and Western basins within the undisclosed uncertainty of the MISR data?

As mentioned above, due to uncertainties associated with MISR SSA derivation, we have now removed this conclusion in the new version of the manuscript.

3. Minor Comments/corrections

Page 9278
Line 5 What is the meaning of the quotes in 'pure'

We'd just like to identify those AERONET sites as representative of "urban-industrialized" atmosphere. "Pure" is not necessary the good term. We have now just indicated "Over urban-industrialized regions…"

Lines 9 and 13. Replace tendency with trend.

This is now corrected in the new version.

line 23 What is the meaning of the quotes in 'dusty'?

This term was used to identify the AERONET sites mostly affected by mineral dust aerosols.

Page 9279
Line 9. Is this correlation scientifically relevant?

A linear correlation between AAOD and latitude (Fig. 3) is found significant (at the 0.01 level). The increasing trend of AE and decreasing trend of AAOD at 440 nm from South to North indicates that mineral dust controls these parameters.

Line 9285
Line 2. Replace 'explains' with 'would explain'.

This is now corrected in the new version.