

Response to the Editor

The authors would like to thank the Editor for his comments. Below, please find attached our response to each one of the Editor's minor comments:

1. In the first paragraph of page 4 there is a long list of publications dealing with Mediterranean and satellite use. This collection of papers is based simply on satellite use. I would suggest since you are not commenting on what anyone of these authors really did and since this is not an overview paper, to limit the publications on this section based on the importance of their results.

We thank the Editor for this comment. We agree with the Editor that this is not a review paper as it discusses new findings. On the other hand our goal was to submit a kind of comprehensive reference study for the area which could be used not only for scientific but also for educational purposes. The main idea behind giving all these studies is that 1. the studies are separated according to the satellite instruments they use and 2. the statistics given in Fig. 4 are produced using these references. If we removed some of them, then Fig. 4 which shows the importance of the region and the increasing interest about it from the scientific community would not be valid any more.

Therefore, we kindly ask from the Editor to allow us keeping these references in the text.

2. There are several papers comparing aeronet stations with MODIS AOD's. Based on figure 3 and table 2 and the discussion included, I do not see any comments on the impact of the differences exhibited, to the final results of your study. Comments are limited to: "The results discussed in this paragraph are comparable to the ones appearing in previous studies focusing on the Mediterranean region (see Papadimas et al., 2009; Koukouli et al., 2010). In general, it is shown here that the MODIS Terra Collection 051 data exhibit a better agreement with the ground-based observations from AERONET than MODIS Aqua data do."

We thank the Editor for giving us the opportunity to clarify this in the paper. We have added the following lines in the revised manuscript.

Page 18: "...Therefore, the statistics appearing for MODIS Terra throughout the paper could be considered more robust..."

Page 19: "...Only areas in Northern Africa are expected to be affected by the use of DB data due the extended lack of DT data there..."

3. So in the conclusions it would be important to include some discussion on this issue. For example figure 3c (DB) statistics are quite random in my opinion. So the importance of the results based on these data have to be discussed.

We thank the Editor for giving us the opportunity to clarify this. We have added the following lines in the first paragraph of the Summary and Conclusions section of the revised manuscript.

"...According to the validation, the statistics appearing for MODIS Terra throughout the paper could be considered more robust while areas in Northern Africa are expected to be affected by the extended use of DB data which do not exhibit a very good matching with the ground-based observations..."

4. Personally I do not like the function and function statistics of the articles vs years in figure 4. I think you can simply mention it or leave the line there for demonstrating what you want to say.

The function has been removed from the Fig. 4 according to the Editor's recommendation.

5. Figure 6. It was strange for me not to see any AOD hints in and around the area of Istanbul at all seasons. There are studies showing the importance of the aerosol flow from this area over various directions but in this study it is completely invisible e.g. compared to Ankara, Turkey area, or even Thessaloniki and Athens area.

We thank the Editor for his comment. We have double-checked our calculations following the Editor's recommendation. The same appears also in a very recent paper by Georgoulias et al. (2016)* where we compare the Collection 6 and 5.1 MODIS data. So, this has definitely not to do with the Collection used here. On the other hand the AODs are pretty high over the greater Istanbul region. This is more prominent in Fig. 11 (first column) where the anthropogenic aerosols are presented.

**Georgoulias, A. K., Alexandri, G., Kourtidis, K. A., Lelieveld, J., Zanis, P., and Amiridis, V.: Differences between the MODIS Collection 6 and 5.1 aerosol datasets over the greater Mediterranean region, Atmos. Environ., 147, 310-319, doi:10.1016/j.atmosenv.2016.10.014, 2016.*

6. Figure 7. FMF fraction use over land as the authors report is quite uncertain. Could you quantify the effect of its use on the land related percentages of this figure ?

As discussed in detail in the methodology we do not use FMF over land. We use the MACC-GOCART dust/total AOD ratios instead. Therefore our results do not suffer from the use of FMF.

7. Figure 11 dust cases/column. We expect a gradient of AOD based on the dust sources of Sahara and middle East. However e.g. figure 11j there are significant contributions in the order of 0.1 to 0.15 of AOD to specific isolated areas e.g. in Greece. Could it be due to uncertainties related with the way of characterizing dust here ?

Working with regional climate models in the region we have seen that generally local dust sources are not taken into account. Industrial, construction and agricultural activities are expected to produce significant amount of dust. For example in Greece, high dust concentrations appear over such regions (e.g. the industrial region in Thessaloniki, the Thessaly plain in central Greece, etc.).

Main document changes and comments

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Therefore, the statistics appearing for MODIS Terra throughout the paper could be considered more robust.

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Only areas in Northern Africa are expected to be affected by the use of DB data due the extended lack of DT data there.

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; Georgoulas et al., 2016

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; Georgoulas et al., 2016

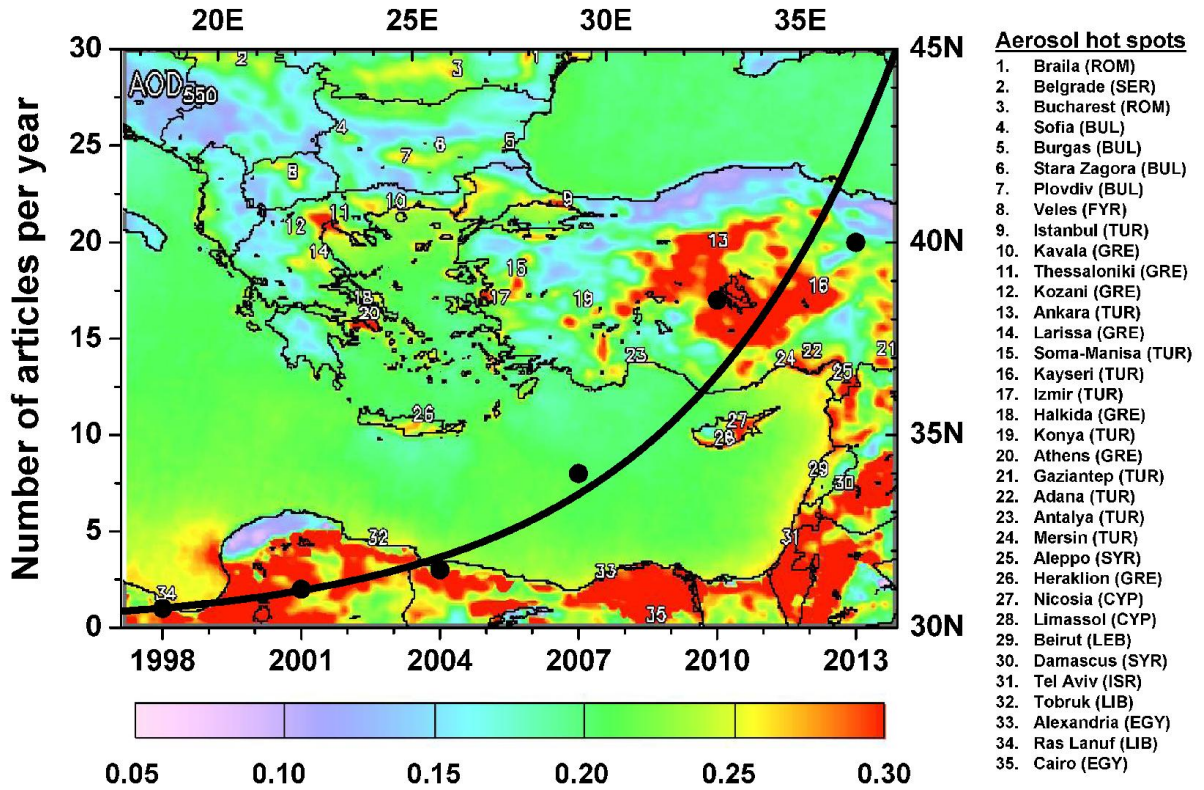
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According to the validation, the statistics appearing for MODIS Terra throughout the paper could be considered more robust while areas in Northern Africa are expected to be affected by the extended use of DB data which do not exhibit a very good matching with the ground-based observations.

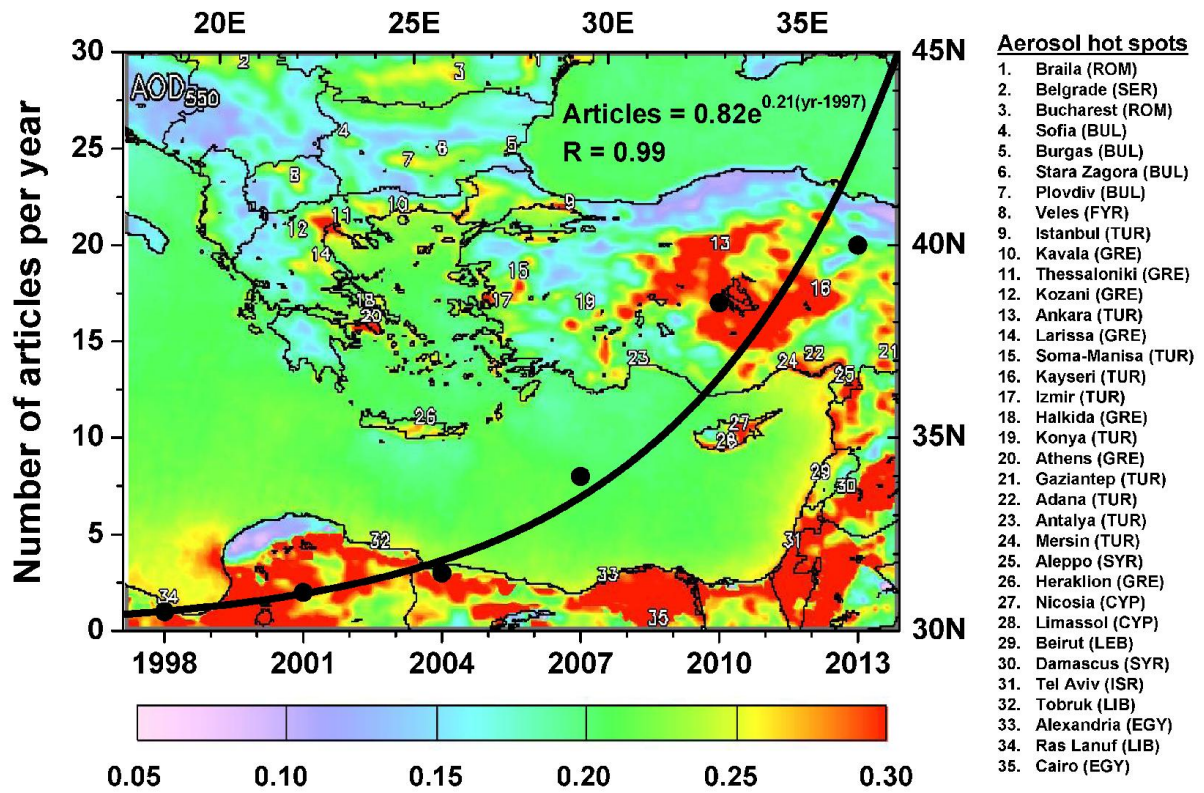
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English (U.K.)



Header and footer changes

Text Box changes

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Footnote changes

Endnote changes

1 **Spatiotemporal variability and contribution of different aerosol**
2 **types to the Aerosol Optical Depth over the Eastern Mediterranean**

3
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28
29 **Abstract**

30 This study characterizes the spatiotemporal variability and relative contribution of different
31 types of aerosols to the Aerosol Optical Depth (AOD) over the Eastern Mediterranean as
32 derived from MODIS Terra (3/2000-12/2012) and Aqua (7/2002-12/2012) satellite
33 instruments. For this purpose, a 0.1° x 0.1° gridded MODIS dataset was compiled and

1 validated against sunphotometric observations from the AERosol RObotic NETwork
2 (AERONET). The high spatial resolution and long temporal coverage of the dataset allows for
3 the determination of local hot spots like megacities, medium sized cities, industrial zones, and
4 power plant complexes, seasonal variabilities, and decadal averages. The average AOD at 550
5 nm (AOD₅₅₀) for the entire region is $\sim 0.22 \pm 0.19$ with maximum values in summer and
6 seasonal variabilities that can be attributed to precipitation, photochemical production of
7 secondary organic aerosols, transport of pollution and smoke from biomass burning in Central
8 and Eastern Europe, and transport of dust from the Sahara Desert and the Middle East. The
9 MODIS data were analyzed together with data from other satellite sensors, reanalysis projects
10 and a chemistry-aerosol-transport model using an optimized algorithm tailored for the region
11 and capable of estimating the contribution of different aerosol types to the total AOD₅₅₀. The
12 spatial and temporal variability of anthropogenic, dust and fine mode natural aerosols over
13 land and anthropogenic, dust and marine aerosols over the sea is examined. The relative
14 contribution of the different aerosol types to the total AOD₅₅₀ exhibits a low/high seasonal
15 variability over land/sea areas, respectively. Overall, anthropogenic aerosols, dust and fine
16 mode natural aerosols account for $\sim 51\%$, $\sim 34\%$ and $\sim 15\%$ of the total AOD₅₅₀ over land,
17 while, anthropogenic aerosols, dust and marine aerosols account $\sim 40\%$, $\sim 34\%$ and $\sim 26\%$
18 of the total AOD₅₅₀ over the sea, based on MODIS Terra and Aqua observations.

19

20 **1 Introduction**

21 For more than fifteen years, two MODIS (Moderate Resolution Imaging Spectroradiometer)
22 satellite sensors monitor tropospheric aerosols at a global scale on a daily basis. The retrieved
23 aerosol optical properties have been used in numerous air quality studies as well as studies
24 related to the effect of airborne particles on various climatic parameters (e.g. radiation, clouds,
25 precipitation, etc.). The $1^\circ \times 1^\circ$ daily gridded level-3 dataset is primarily used for global as well
26 as regional studies while the single pixel level-2 data with a 10 km resolution (at nadir) are
27 mostly used for regional and local scale studies. Nevertheless, the use of the coarse resolution
28 MODIS data has predominated even in regional studies. The reasons for this could be the
29 smaller file size which makes their processing and storage easier or the fact that they are easily
30 accessible through user-friendly data bases which also allow for a very basic analysis like e.g.
31 NASA's GIOVANNI website (<http://giovanni.gsfc.nasa.gov/giovanni/>) (Acker and Leptoukh,
32 2007).

1 The same holds for studies focusing on the Mediterranean Basin, an area which is considered of
2 particular sensitivity as far as air pollution and climate change is concerned (Lelieveld et al.,
3 2002; Giorgi, 2006). The Mediterranean basin is one of the regions with highest aerosol optical
4 depths (AODs) in the world (Husar et al., 1997; Ichocku et al., 2005; Papadimas et al., 2008),
5 causing significant climate forcing especially in summer, which is characterized by low
6 cloudiness and high incoming solar radiation levels (Papadimas et al., 2012; Alexandri et al.,
7 2015). The Mediterranean is also recognized as a crossroads between three continents where
8 aerosols of various types accumulate (Lelieveld et al., 2002). Marine aerosols from the
9 Mediterranean Sea and even the Atlantic Ocean combine with aerosols from continental Europe
10 (urban and rural), dust particles transported from the Sahara Desert and Middle East as well as
11 biomass burning aerosols from occasional wild fires and agricultural burning (Lelieveld et al.,
12 2002). Specifically, as discussed in Hatzianastassiou et al. (2009), Eastern Mediterranean, the
13 region under investigation here, is located at a "key" point of this crossroads. There is a
14 significant number of ground and satellite-based studies on the abundance and optical properties
15 of tropospheric aerosols in the area; however, these studies are either focused on specific spots
16 or used a coarse spatial and temporal resolution.

17 The ground-based instrumentation used in studies focusing on the aerosol load and optical
18 properties over the Eastern Mediterranean includes active and passive sensors such as Lidars
19 (e.g. Papayannis and Balis, 1998; Balis et al., 2004; Papayannis et al., 2005, 2009; Amiridis et
20 al., 2005, 2009; Mamouri et al., 2013; Kokkalis et al., 2013; Nisantzi et al., 2015), Cimel
21 sunphotometers (e.g. Israelevich et al., 2003; Kubilay et al., 2003; Derimian et al., 2006;
22 Kalivitis et al., 2007; Kelektsoğlu and Rapsomanikis, 2011; Nikitidou and Kazantzidis, 2013),
23 Brewer spectrophotometers (e.g. Kazadzis et al., 2007; Koukouli et al., 2010), Multi-Filter
24 Radiometers (e.g. Gerasopoulos et al., 2009, 2011; Kazadzis et al., 2014), ceilometers (e.g.
25 Tsaknakis et al., 2011), Microtops sunphotometers (e.g. El-Metwally and Alfaro, 2013), etc.
26 However, these and other studies not referenced here either refer to specific spots with the
27 majority of the ground stations being situated in large urban centers (e.g. Athens, Thessaloniki,
28 Cairo) or to specific events (e.g. Sahara dust intrusions, biomass burning events, etc.).

29 On the other hand, AOD and other aerosol optical properties have been studied over the greater
30 Eastern Mediterranean region based on data from Meteosat (Moulin et al., 1998), SeaWiFS
31 (Koren et al., 2003; Antoine and Nobileau, 2006; Mélin et al., 2007; Nabat et al., 2013), TOMS
32 (Alpert and Ganor, 2001; Israelevich et al., 2002; Koukouli et al. 2006; Hatzianastassiou et al.,
33 2009; Koukouli et al., 2010, Israelevich et al., 2012, Kaskaoutis et al., 2012a; Nabat et al., 2013;

1 Gkikas et al., 2013, 2014; Varga et al., 2014), MODIS Terra and Aqua (Barnaba and Gobbi,
2 2004; Papayannis et al., 2005; Kaskaoutis et al., 2007, 2008, 2010, 2011, 2012a,b,c,d;
3 Kosmopoulos et al., 2008; Papadimas et al., 2008, 2009; Rudich et al., 2008; Carmona and
4 Alpert, 2009; Karnieli et al., 2009; Gkikas et al., 2009, 2013; Hatzianastassiou et al., 2009; El-
5 Metwally et al., 2010; Koukouli et al., 2010; Kanakidou et al., 2011; Gerasopoulos et al., 2011;
6 de Meij and Lelieveld, 2011; Marey et al., 2011; de Meij et al., 2012; Nabat et al., 2012, 2013;
7 Nikitidou and Kazantzidis, 2013; Athanasiou et al., 2013; Benas et al., 2011, 2013; Sorek-
8 Hamer et al., 2013; Kabatas et al., 2014; Kourtidis et al., 2014; Mishra et al., 2014; Flaounas et
9 al., 2015; Kloog et al., 2015), OMI/AURA (Kaskaoutis et al., 2010; El-Metwally et al., 2010;
10 Marey et al., 2011; Kaskaoutis et al., 2012b,c; Gkikas et al., 2013, 2014; Sorek-Hamer et al.,
11 2013; Varga et al., 2014; Flaounas et al., 2015), CALIOP/CALIPSO (Amiridis et al., 2009,
12 2013, Mamouri et al., 2009; Marey et al., 2011; Kaskaoutis et al., 2012c; de Meij et al., 2012;
13 Nabat et al., 2012, 2013; Mamouri and Ansmann, 2015), MISR/Terra (Kanakidou et al., 2011;
14 Marey et al., 2011; de Meij and Lelieveld, 2011; de Meij et al., 2012; Nabat et al., 2013;
15 Kabatas et al., 2014; Abdelkader et al., 2015) as well as NOAA/AVHRR, MERIS/ENVISAT,
16 AATSR/ENVISAT, PARASOL/POLDER, MSG/SEVIRI, and Landsat satellite data (see
17 Retalis and Sifakis, 2010; Nabat et al., 2013; Benas et al., 2013; Sifakis et al., 2014). To our
18 knowledge, these studies comprise the majority of work focusing on tropospheric aerosols over
19 the Eastern Mediterranean by means of satellite remote sensing, published in peer reviewed
20 journals the last ~ 15 years. As shown in Fig. 4 of this work, the publication rate of satellite-
21 based studies focusing on the Eastern Mediterranean aerosols nearly doubled every three years
22 during the period 1997-2014 which is indicative of the increasing scientific interest in the area.
23 In a very large fraction of the satellite-based studies referenced above, the used data are either of
24 coarse mode (usually 1° which is ~ 100 km for the mid-latitudes) or focus on specific spots for
25 validation purposes. In a few cases, high resolution data were used in spatiotemporal studies;
26 however, either these studies are restricted over surfaces covered by water or examine a short
27 period only. For example, Moulin et al. (1998) investigated the dust AOD patterns over the
28 oceanic areas of the Mediterranean Basin at a resolution of $35 \times 35 \text{ km}^2$ for a period of 11 years
29 (1984-1994) using Meteosat observations. A 7-year climatology (1998-2004) of total and dust
30 AOD for the same regions at a resolution of $0.16^\circ \times 0.16^\circ$ was compiled by Antoine and
31 Nobileau (2006) using observations from SeaWiFS. Mélin et al. (2007) merged SeaWiFS and
32 MODIS data and presented high resolution AOD patterns ($2 \times 2 \text{ km}^2$) for May 2003. As far as
33 MODIS is concerned, only Barnaba and Gobbi (2004) presented a high resolution ($0.1^\circ \times 0.1^\circ$)

1 spatiotemporal analysis for a period of 1 year (2001) over sea only. In a recent paper,
2 Athanasiou et al. (2013) presented in detail a method for compiling a 0.5-degree resolution
3 AOD gridded dataset using level-2 MODIS Terra data for the greater region of Greece (2000-
4 2008). However, the spatial resolution they used (~ 50 km) is not high enough to reveal local
5 sources (e.g. cities, islands, river banks, etc.). Overall, there has not been so far any detailed
6 high resolution spatiotemporal study of the AOD over the Eastern Mediterranean.
7 In this paper, the AOD₅₅₀ spatiotemporal variability over the Eastern Mediterranean (30°N-
8 45°N, 17.5°E-37.5°E) is presented at a high spatial resolution (0.1° x 0.1°) based on MODIS
9 Terra and Aqua observations. Level-2 MODIS data are used for the compilation of a 0.1-degree
10 gridded dataset which is validated against ground-based observations. In order to calculate the
11 contribution of different aerosol types to the total AOD, the MODIS data were analyzed
12 together with other satellite data, ERA-Interim reanalysis data and the Goddard Chemistry
13 Aerosol Radiation and Transport (GOCART) model using an algorithm optimized for the
14 surface properties of the Eastern Mediterranean region. The different datasets used in this
15 research are presented in detail in Sect. 2 while a detailed description of the method is given in
16 Sect. 3. Sect. 4 includes the results from the MODIS validation procedure, the annual and
17 seasonal variability of AOD₅₅₀ over the region with a discussion on the local aerosol sources
18 and the differences between Terra and Aqua, and the annual and seasonal contribution of
19 different aerosol types to the total AOD₅₅₀. Finally, in Sect. 5, the main conclusions of the paper
20 are presented along with a short discussion on how these results could contribute to future
21 studies in the area.

22

23 **2 Observations, reanalysis data and model simulations**

24 **2.1 MODIS Terra and Aqua satellite observations**

25 The main data used in this work come from the level-2 MODIS Terra (MOD04_L2) and
26 MODIS Aqua (MYD04_L2) Collection 051 dataset and have been acquired through NASA's
27 Level 1 and Atmosphere Archive and Distribution System (LAADS)
28 (<http://ladsweb.nascom.nasa.gov>). The fact that MODIS Terra and Aqua have a daytime
29 equator crossing time at 10:30 LT (morning) and 13:30 LT (noon), respectively. MODIS
30 instruments with a viewing swath of 2330 km measure backscattered radiation at 36 spectral
31 bands between 0.415 and 14.235 μm with a spatial resolution of 250, 500 and 1000 m,
32 providing a nearly global coverage on a daily basis. Aerosol optical properties for the
33 standard MODIS aerosol product are retrieved using two different "Dark Target" (DT)

1 algorithms. The one is used over land surfaces (Kaufman et al., 1997; Levy et al., 2007a, b;
2 Remer et al., 2005; Levy et al., 2010) and the other over oceanic regions (Tanré et al., 1997;
3 Levy et al., 2003; Remer et al., 2005). The "Deep Blue" algorithm (DB) (Hsu et al., 2004;
4 Hsu et al., 2006) has been used for retrievals over bright land surfaces (e.g. deserts) where the
5 DT algorithm fails. Only recently, updates to the algorithm allowed for extending the spatial
6 coverage of the DB aerosol product over all land areas (Hsu et al., 2013; Sayer et al., 2013;
7 2014). AERONET Cimel sunphotometric measurements have been extensively used for the
8 validation of the MODIS over-land and over-ocean products (e.g. Chu et al., 2002; Remer et
9 al., 2002; Remer et al., 2005; Levy et al., 2010; Shi et al., 2013).

10 In this work, AOD₅₅₀ over both land and sea and the Fine Mode Ratio (FMR₅₅₀) over sea from
11 Collection 051 were used at a spatial resolution of 10 km (at nadir). The uncertainty of the
12 MODIS aerosol optical depth has been estimated at $\pm(0.05+0.15AOD)$ over land (Chu et al.,
13 2002; Levy et al., 2010) and $\pm(0.03+0.05AOD)$ over ocean (Remer et al., 2002) relative to the
14 AERONET AOD. Specifically, for the DT data used in this work only high quality retrievals
15 are used over land. This means that the data have a Quality Assurance Confidence (QAC) flag
16 equal to 3 (high confidence). For retrievals over sea we use data with a QAC flag of 1
17 (marginally good), 2 (good) and 3 (see Levy et al., 2009 for details). The pre-launch
18 uncertainty of FMR₅₅₀ is $\pm 30\%$ over ocean (Remer et al., 2005) while over land this
19 parameter is by no means trustworthy and should only be used in qualitative studies (e.g. see
20 Georgoulias and Kourtidis, 2011). In cases where DT algorithm does not provide products
21 over land, especially over bright arid and semi-arid regions of North Africa, AOD₅₅₀ values
22 from the DB algorithm are used in our work. The expected uncertainty of the DB product
23 used here is $\pm(0.05+0.2AOD)$ relative to the AERONET AOD (Hsu et al., 2006). The
24 analyzed datasets cover the period from 3/2000 to 12/2012 for Terra and from 7/2002 to
25 12/2012 for Aqua MODIS covering the region of the Eastern Mediterranean. The Collection
26 051 DB data for Terra are available only until 12/2007 due to calibration issues; nevertheless,
27 these data are carefully used within our analysis to get a complete image of the aerosol load
28 over the region.

29

30 **2.2 AERONET ground-based observations**

31 For the evaluation of the MODIS AOD₅₅₀, data from 13 AERONET Cimel network ground
32 stations in the region of the Eastern Mediterranean have been acquired
33 (<http://aeronet.gsfc.nasa.gov>). The stations were selected such that their operation period

1 covers at least 2 years and there are at least 100 common days of co-localized AERONET and
2 MODIS observations. AERONET Cimel sunphotometers measure solar radiation every 15
3 minutes within the spectral range from 340 to 1020 nm (Holben et al., 2001). The spectral
4 measurements allow for the retrieval of columnar aerosol properties (see Holben et al., 1998;
5 Dubovik and King, 2000; Dubovik et al., 2000, 2002). The AERONET AOD uncertainty is in
6 the order of 0.01-0.02 (Eck et al., 1999), being larger at shorter wavelengths. Here, we use
7 quadratic fits on a log-log scale to interpolate the AERONET data (AODs at 440, 500, 675
8 and 870 nm) to the MODIS band-effective wavelength of 550 nm (Eck et al., 1999; Levy et
9 al., 2010). So, we can directly compare the MODIS AOD₅₅₀ retrievals against AERONET
10 observations. Simultaneous measurements of the Ångström Exponent (AE) for the spectral
11 range 440-870 nm (AE₄₄₀₋₈₇₀) from the 13 AERONET stations mentioned above were also
12 utilized in this work in order to account for days with dust dominance. The uncertainty of the
13 AE is significantly higher than the AOD uncertainty, especially under low-AOD conditions.
14 Li et al. (2014) found that the uncertainty for a typical Northern Hemispheric AERONET
15 station (GSFC) is ~ 0.6 during winter when AODs are significantly lower compared to
16 summer (~ 0.15).

17

18 **2.3 LIVAS CALIOP/CALIPSO dust climatology**

19 Dust aerosol optical depths at 532 nm (AOD₅₃₂) from CALIOP/CALIPSO (Cloud-Aerosol
20 Lidar with Orthogonal Polarization instrument aboard Cloud-Aerosol Lidar and Infrared
21 Pathfinder Satellite Observations satellite) at a resolution of $1^\circ \times 1^\circ$ are also used here for the
22 period 2007-2012. CALIPSO measures cloud and aerosol properties flying at a 705 km sun
23 synchronous polar orbit with a 16 day repeat cycle and an equator-crossing time close to that
24 of the Aqua satellite (13:30 LT). The dust product used here comes from a Saharan-dust-
25 optimized retrieval scheme that was developed within the framework of the LIVAS (Lidar
26 Climatology of Vertical Aerosol Structure for Space-Based LIDAR Simulation Studies)
27 project (Amiridis et al. 2015) and has been presented in detail in Amiridis et al. (2013). In
28 brief, the LIVAS dust product is optimized for Europe by applying a lidar ratio of 58 sr
29 instead of 40 sr to Level 2 dust related backscatter products. This correction results to an
30 improvement of the AOD₅₃₂ product. Comparison against spatially and temporally co-located
31 AERONET observations (Amiridis et al., 2013) returned an absolute bias of ~ -0.03 . The
32 corresponding reported biases for the original CALIPSO data are significantly higher ($\sim -$
33 0.10). The bias is even lower when compared against MODIS satellite-based observations.

1 Other improvements of this product are related to the use of a new methodology for the
2 calculation of pure dust extinction from dust mixtures and the application of an averaging
3 scheme that includes zero extinction values for the non-dust aerosol types detected. Overall,
4 this product (hereafter denoted as LIVAS dust product) exhibits better agreement with
5 observations from MODIS and AERONET and simulations from the BSC-DREAM8b dust
6 model over North Africa and Europe than the standard CALIPSO data hence being an ideal
7 tool for the evaluation of other satellite-based products.

9 **2.4 Earth Probe TOMS and OMI satellite observations**

10 For this work, UV Aerosol Index (AI) data (Herman et al., 1997) from the Earth Probe TOMS
11 (Total Ozone Mapping) spectrometer aboard Earth Probe for the period 1/2000-9/2004 at a
12 resolution of 1° (latitude) x 1.25° (longitude) and the OMI (Ozone Monitoring Instrument)
13 sensor aboard EOS AURA for the period 10/2004-12/2012 at a resolution of 1° x 1° were
14 acquired through the GIOVANNI web database (<http://giovanni.gsfc.nasa.gov/giovanni/>).
15 Earth Probe TOMS continued the record of the first three TOMS instruments aboard Nimbus-
16 7, Meteor-3 and ADEOS flying in a sun synchronous orbit at an altitude of 740 km with an
17 instantaneous field of view size of $39 \times 39 \text{ km}^2$ at nadir. The instrument had an ascending
18 node equator crossing time at 12:00 LT covering 85 % of the globe on a daily basis from
19 7/1996 until 12/2005. The satellite was originally set to a 500 km sun synchronous orbit but
20 was set to its final orbit after the failure of ADEOS satellite in 6/1997. OMI is a UV/VIS
21 nadir solar backscatter spectrometer (Levelt et al., 2006) that continues the long TOMS
22 record. OMI flies in a sun synchronous polar orbit at an altitude of 705 km with an ascending
23 node equator crossing time at 13:45 LT. Its 2600 km viewing swath allows for almost daily
24 global coverage while the spatial resolution of the instrument is $13 \times 24 \text{ km}^2$ at nadir. The AI
25 (also known as Absorbing Aerosol index) which is calculated by the two instruments
26 constitutes a qualitative indicator of the presence of UV absorbing aerosols in the atmosphere
27 such as biomass burning and dust (Torres et al., 1998). Positive AI values generally represent
28 absorbing aerosols while small or negative values represent non-absorbing aerosols. The
29 Version 8 algorithm is applied to spectral measurements from both TOMS and OMI sensor to
30 produce a consistent long-term AI timeseries (Li et al., 2009). AI is calculated from the
31 difference in surface reflectivity derived from the 331.2 and 360 nm measurements exhibiting
32 an uncertainty of $\pm 30 \%$ (Torres et al., 2007).

2.5 ERA-Interim reanalysis data

Wind speed (ws) data at 10 m above surface from the ERA-Interim reanalysis (Dee et al., 2011) are used for 9:00 and 12:00 UTC on a daily basis for the period 2000-2012. We use 9:00 and 12:00 UTC data in order to be closer to the Terra and Aqua overpass time in the area, respectively. The various ERA-Interim reanalysis fields are produced by ECMWF's Integrated Forecast System (IFS) assimilating satellite and ground-based observations. The system includes a 4-D variational analysis with a 12-hour analysis window. The spatial resolution of the ERA-Interim data is ~ 79 km with 60 vertical levels from the surface up to 0.1 hPa while the data can be acquired at various resolutions (in this work $1^\circ \times 1^\circ$) through ECMWF's website (<http://apps.ecmwf.int/datasets/data/interim-full-daily/>). Over ocean, the 10 m ERA-interim wind speed exhibits a bias of less than -0.5 m/s compared to quality-controlled in situ observations on a global scale (Dee et al., 2011). Specifically, for the region of the Eastern Mediterranean examined here, the 10 m ERA-interim wind speed exhibits a bias of -0.96 m/s (-16 %) compared to satellite-based observations from QuikSCAT (Hermann et al., 2011).

2.6 MACC reanalysis data

The daily MACC total and dust AOD₅₅₀ data for the period 2003-2012 come from the aerosol analysis and forecast system of ECMWF which consists of a forward model (Morcrette et al., 2009) and a data-assimilation module (Benedetti et al., 2009). AOD₅₅₀ measurements from the two MODIS instruments aboard Terra and Aqua are assimilated by the MACC forecasting system through a 4D-Var assimilation algorithm to produce the aerosol analysis, leading to an improved AOD representation compared to observations (see Benedetti et al., 2009; Mangold et al., 2011). Five aerosol species are included within MACC, namely, mineral dust, sea salt, sulfates, black carbon and organic matter. Three different size bins are used for mineral dust and sea salt particles while the black carbon and organic material are distributed to a hydrophilic and a hydrophobic mode. Dust and sea salt emissions are given as a function of surface wind speed, while the emissions of the other species are taken from inventories. The spatial resolution of the MACC reanalysis data is ~ 79 km with 60 vertical levels from the surface up to 0.1 hPa and can be acquired through: <http://apps.ecmwf.int/datasets/data/macc-reanalysis/> for the period 2003-2012. The MACC total and dust AODs have been evaluated against ground and satellite-based observations (see Elguindi et al., 2010; Bellouin et al., 2013; Inness et al., 2013; Cesnulyte et al., 2014; Cuevas et al., 2015) showing that the MACC

1 aerosol products generally capture well the daily, seasonal and interannual variability of
2 aerosols. As discussed in Bellouin et al. (2013) the uncertainties of MACC total AOD₅₅₀ (~
3 0.03) and dust AOD₅₅₀ (~ 0.014) arise from uncertainties in the MODIS retrievals which are
4 assimilated into the model and errors in the forward modeling of total and component AODs.
5

6 **2.7 GOCART data**

7 Daily total and dust AOD₅₅₀ data from the GOCART chemistry-aerosol-transport model
8 simulations (version 006) are used in this study for the period 2000-2007. The GOCART
9 model (see Chin et al., 2000, 2002, 2004, 2007; Ginoux et al., 2001, 2004) uses the
10 assimilated meteorological fields of the Goddard Earth Observing System Data Assimilation
11 System (GEOS DAS) which are generated by the Goddard Global Modeling and Assimilation
12 Office (GMAO). The data which are used were acquired from an older version of NASA's
13 GIOVANNI web database (<http://disc.sci.gsfc.nasa.gov/giovanni/>) and come from a
14 simulation implemented at a spatial resolution of 2° (latitude) x 2.5° (longitude) with 30
15 vertical sigma layers (Chin et al., 2009). The model includes physicochemical processes of
16 major tropospheric aerosol components (sulfates, dust, black carbon, organic carbon, sea salt)
17 and precursor gases (SO₂ and dimethylsulfide) incorporating various atmospheric processes.
18 The total AOD₅₅₀ from GOCART compared to ground-based observations from the
19 AERONET exhibits a relative mean bias [$\text{mean(GOCART)/mean(AERONET)}$] of 1.120,
20 1.135 and 0.959 over Europe, North Africa and for the whole globe, respectively.
21

22 **2.8 Ancillary data**

23 Apart from the primary datasets presented above, three additional datasets were used in order
24 to support our findings. OMI/AURA daily gridded (Bucsela et al., 2013) tropospheric NO₂
25 columnar data (OMNO2d version 2.1) at a spatial resolution of 0.25° x 0.25° were acquired
26 from NASA's GIOVANNI web database (<http://giovanni.gsfc.nasa.gov/giovanni/>) for the
27 period 2005-2012. The quality checked data used in this work correspond to sky conditions
28 where cloud fraction is less than 30 %. Planetary boundary layer (PBL) SO₂ daily gridded
29 columnar data (OMSO2e version 1.1.7) were also acquired from GIOVANNI for the same
30 period. The OMSO2e gridded data (0.25° x 0.25°) used in this work are produced from best
31 level-2 pixel data, screened for OMI row anomaly and other data quality flags. The PBL SO₂
32 column retrievals are produced with an algorithm based on principal component analysis
33 (PCA) of the OMI radiance data (Li et al., 2013). Finally, monthly precipitation data from the

1 3B43 TRMM and Other Sources Monthly Rainfall Product (version 7) at a spatial resolution
2 of $0.25^\circ \times 0.25^\circ$ for the period 2000-2012 were obtained from GIOVANNI. This dataset is
3 derived from 3-hourly precipitation retrievals from the Precipitation Radar (PR), the TRMM
4 Microwave Imager (TMI) and the Visible and Infrared Scanner (VIRS) aboard the TRMM
5 (Tropical Rainfall Monitoring Mission) satellite merged with other satellite-based
6 precipitation data and the Global Precipitation Climatology Centre (GPCC) rain gauge
7 analysis (Huffman et al., 2007).

8

9 **3 Methodology**

10 **3.1 Compiling a MODIS $0.1^\circ \times 0.1^\circ$ gridded dataset**

11 To investigate the spatial and temporal variability of aerosols over the Eastern Mediterranean
12 we first created a $0.1^\circ \times 0.1^\circ$ daily gridded aerosol dataset using single pixel level-2 AOD_{550} and
13 FMR_{550} data from MODIS Collection 051. The same resolution has been utilized in previous
14 studies (e.g. Barnaba and Gobbi, 2004) in the region; however, without reporting on the
15 gridding methodology followed. In this work we present a gridding methodology that could be
16 used as a reference for future regional studies. The methodology has been successfully applied
17 in the past on level-2 MODIS Terra data in different cases studies, e.g. in order to examine the
18 weekly cycle patterns of AOD_{550} over the region of Central Europe and the aerosol load
19 changes observed over a cement plant in Greece due to changes in the deposition practices of
20 the primary materials (see Georgoulas and Kourtidis, 2012; Georgoulas et al., 2012; Kourtidis
21 et al., 2014). In the following lines we proceed to a detailed description of the method
22 underlining the potential of being used in detailed quantitative studies like this one.

23 First, a $0.1^\circ \times 0.1^\circ$ resolution grid covering the Eastern Mediterranean (30°N - 45°N , 17.5°E -
24 37.5°E) is defined which corresponds to 30000 grid cells. As already mentioned in Sect. 2.1,
25 only level-2 single pixel AOD_{550} measurements with a QAC flag of 3 and a QAC flag greater
26 than 0 were used over land and over sea, respectively, to ensure the high quality of the data.
27 Pixels are attributed to a specific grid cell if their center falls within a $25 \times 25 \text{ km}^2$ square
28 window around the grid cell (see Fig. S1 in the Supplement). These pixels are then used for the
29 calculation of daily averages. As shown in Figure S1, a grid cell of 0.1° ($\sim 10 \text{ km}$) is as big as
30 the centre of a large Mediterranean city like Thessaloniki, Northern Greece (~ 1 million
31 inhabitants). The procedure was followed separately for MODIS Terra and Aqua data. In cases
32 of grid cells with no DT MODIS observations, data from the DB algorithm were used (over

1 bright arid and semi-arid regions of North Africa) constituting only a small part of the gridded
2 dataset.

3 The size of the gridding window was selected following Koukouli et al. (2007). They used both
4 10 and 25-km windows showing that the latter allows for the inclusion of more data points
5 without undermining the ability of monitoring accurately the aerosol load over a specific spot.
6 In addition, in cases of urban sites, a window of 25 km allows for the inclusion of pixels from
7 the surrounding non-urban surfaces where the MODIS surface reflectance parameterization is
8 better (Levy et al., 2010). The size of each MODIS pixel is 10 km at nadir, but at the swath
9 edges, it may become 2-3 times larger. Hence, ideally the maximum number of pixels that could
10 be used in the daily averaging is nine. The overlap between the windows of neighbouring grid
11 cells does not affect the representativeness of the dataset over each grid cell. Aerosols are
12 transported by air masses throughout the day and thus the aerosol load in neighbouring grid
13 cells is not expected to be completely independent.

14 In order to make sure that the use of a 25-km gridding window is optimal for capturing local
15 pollution sources we repeated the same procedure for bigger gridding windows (50-km, 75km
16 and 100-km) using MODIS Terra AOD₅₅₀ data for the year 2004. Numerous aerosol hot spots
17 cannot be seen as the gridding window becomes bigger and there is a significant smoothing of
18 the aerosol patterns mainly over land (Fig. S2). The use of the MODIS gridded dataset in the
19 detection of local aerosol hot spots is discussed in more detail in Sect. 4. In addition, we
20 conducted a detailed validation of the MODIS data against sunphotometric data from a total of
21 13 AERONET stations in the region (see Fig. 1). The validation procedure was repeated several
22 times for different spatial collocation windows which were equal to the windows used for the
23 gridding procedure (i.e. 25, 50, 75 and 100-km) and for different data quality criteria. The
24 results of the validation procedure are presented in Sect. 4.1 while part of them is given in the
25 Supplement of this manuscript (see Table S2). Overall, it is shown that the gridding
26 methodology followed here offers the best compromise for studying the spatial variability of
27 aerosols on a regional or local scale, preserving at the same time the representativeness of the
28 real aerosol load over each specific spot.

29 In order to generalize our results, nine different sub-regions (Fig. 1) were selected apart from
30 the three basic regions of interest, namely, the whole Eastern Mediterranean (EMT) and the land
31 (EML) and oceanic (EMO) areas of the region. The selection was done mainly taking into
32 account geographical but also land type and land use criteria. The four sub-regions that
33 correspond to the land regions of the Eastern Mediterranean are the Northern Balkans Land

1 (NBL), the Southern Balkans Land (SBL), the Anatolia Land (ANL) and the Northern Africa
2 Land (NAL) region while the five sub-regions that correspond to the oceanic regions are the
3 Black Sea Oceanic (BSO), the North-Western Oceanic (NWO), the South-Western Oceanic
4 (SWO), the North-Eastern Oceanic (NEO) and the South-Eastern Oceanic (SEO) region. Mean
5 values of the total AOD₅₅₀ from the Terra and Aqua MODIS are reported for each one of the
6 three basic regions of interest and their nine sub-regions in Sect. 4.

7

8 **3.2 Contribution of different aerosol types to AOD₅₅₀**

9 **3.2.1 Sea**

10 In order to quantify the contribution of different types of aerosols to the total AOD₅₅₀ we
11 followed a different approach for sea and land. This is due to the lack of reliable FMR₅₅₀
12 retrievals over land (e.g. see Levy et al., 2010; Georgoulias and Kourtidis, 2011) which are
13 crucial for the algorithms used in this work. Over the sea we utilize wind speed data at 10 m
14 above surface from the ERA-Interim reanalysis, AI data from TOMS and OMI along with
15 AOD₅₅₀ and FMR₅₅₀ from the MODIS Terra and Aqua gridded datasets presented above. All the
16 datasets were brought to the same 0.1 degree spatial resolution as MODIS by using bilinear
17 interpolation. In the case of TOMS and OMI we used monthly mean AI data following Bellouin
18 et al. (2008) in order to avoid gaps especially during the TOMS period.

19 In general, the algorithm used over the oceanic regions (see Fig. 2) is similar with the one
20 presented in Bellouin et al. (2008). First, the marine AOD₅₅₀ (τ_m) is calculated from near surface
21 wind speed using a linear relation which has been obtained from ground-based studies over
22 pollution free oceanic regions. Bellouin et al. (2008) use the linear relation of Smirnov (2003).
23 Then, if τ_m is greater or equal than AOD₅₅₀ it is assumed that there are marine particles only
24 over this region. If τ_m is smaller than AOD₅₅₀ a decision tree is followed which is first based on
25 FMR₅₅₀ and then on AI in order to reach a conclusion about the type of aerosols that account for
26 AOD₅₅₀. If FMR₅₅₀ is smaller than the critical value of 0.35 and AI is greater than or equal to a
27 critical value it is assumed that there are both marine aerosols (τ_m) and dust ($\tau_d=AOD_{550}-\tau_m$)
28 while if AI is smaller than this critical value it is assumed that there are marine aerosols only.
29 The AI critical value is equal to 1 in Bellouin et al. (2008). If FMR₅₅₀ is greater than or equal to
30 0.83 it is assumed that there are both anthropogenic ($\tau_a=AOD_{550}-\tau_m$) and marine aerosols (τ_m).
31 In the occasion of having a FMR₅₅₀ equal to 0.35 or greater than 0.35 but smaller than 0.83 one
32 has to take again AI into consideration. If AI is less than the critical value it is assumed that
33 there are marine aerosols (τ_m) only while in the opposite occasion it is assumed that all the three

1 types of aerosols that can be defined over oceanic regions by this algorithm, namely, dust
2 [$\tau_d=(1-FMR_{550})(AOD_{550}-\tau_m)$], anthropogenic [$\tau_a=FMR_{550}(AOD_{550}-\tau_m)$] and marine aerosols (τ_m)
3 are present. One should keep in mind that all the biomass burning aerosols are classified as
4 anthropogenic by this method.

5 In this work, we proceeded to a "fine-tuning" of the algorithm for the region of the Eastern
6 Mediterranean. First, we applied the algorithm on MODIS Terra data using the same equations
7 and critical values as in Bellouin et al. (2008). The results showed that the original Bellouin et
8 al. (2008) method might be valid for global studies but for a "closed" sea like the Mediterranean
9 the method leads to a large overestimation of sea salt AODs and therefore underestimation of
10 dust and anthropogenic aerosol AODs. Indicative of this situation is Fig. S3 in the Supplement
11 where we present the relative contribution of dust, marine and anthropogenic aerosols per
12 month over the oceanic regions of the Eastern Mediterranean as calculated using the original
13 Bellouin et al. (2008) method. It is shown that the marine contribution is several times higher
14 than the values reported for the Mediterranean Basin in previous studies (e.g. see Nabat et al.,
15 2012). Evaluation of the algorithm was done using dust AOD₅₃₂ data from the LIVAS
16 CALIOP/CALIPSO product. From LIVAS we only use the high quality Sahara dust product as
17 a reference and not other aerosol type retrievals (e.g. marine aerosols) since the dust retrievals
18 from CALIOP/CALIPSO are by far the most reliable (e.g. Burton et al., 2013). We performed
19 several tests by changing the linear relation that connects τ_m with near surface wind speed and
20 the AI critical values and compared each time the dust AOD₅₅₀ seasonal variability with the
21 LIVAS AOD₅₃₂ seasonal variability for the sea covered sub-regions of the Eastern
22 Mediterranean. Results from this algorithm-tuning procedure can be found in Figs. S4e-i of the
23 Supplement where one can also see the underestimation of dust AOD₅₅₀ from the original
24 Bellouin et al. (2008) algorithm.

25 The linear relation given in Kaufman et al. (2005) was finally selected ($\tau_m=0.007ws+0.02$). The
26 2000-2012 average wind speed over the sea for the region of the Eastern Mediterranean is ~ 5.3
27 m/s. Kaufman et al. (2005) reduced the offset in the linear relation of Smirnov (2003) from 0.06
28 to 0.02 to fit the average baseline AOD of 0.06 for the typical wind speed of 6 m/s. In addition,
29 our tests showed that an AI critical value of 1 performs well over the region of the Eastern
30 Mediterranean. The results did not change significantly when using other AI thresholds (e.g. 0.5
31 which is suggested in Jones and Christopher, 2011) and therefore we decided to adopt 1 as the
32 AI critical value. Another test, following the example of other studies (see Lehahn et al., 2010),
33 was to assume that for wind speed less than 5 m/s there is very little or no sea-spray particle

1 production (limited bursting of entrained air bubbles associated with whitecap formation). In
2 this case, τ_m is stable, equal to the offset of the linear relation between τ_m and wind speed which
3 is indicative of the background sea salt AOD₅₅₀. However, this test reveals that the effect of
4 assuming stable τ_m for wind speed less than 5 m/s is insignificant and therefore we selected to
5 follow the Kaufman et al. (2005) linear relationship for the whole wind speed range. As shown
6 in Figs. S4e-i, the seasonal variability when applying our modified algorithm over oceanic
7 regions is very close to the LIVAS dust AOD₅₃₂ especially for the months with lower dust load
8 (June-January). It is also shown that dust AODs from this algorithm are closer to the LIVAS
9 dust product than dust AODs from MACC reanalysis do. The slight overestimation of dust
10 AOD or the shift of the maximum dust load we observe for the period of high dust loads in the
11 region (February-May) is probably connected to the narrow swath and the 16-day time of
12 CALIPSO which means that several dust events might be not observed by the CALIOP
13 instrument contrary to MODIS which has a daily coverage.

14

15 **3.2.2 Land**

16 As already mentioned in the previous paragraph a different approach is followed over the land
17 regions of the Eastern Mediterranean due the low confidence on the MODIS FMR₅₅₀ and
18 Ångström exponent retrievals over land compared to that over ocean (e.g. see Levy et al., 2010;
19 Georgoulias and Kourtidis, 2011). This limitation does not allow us to distinguish the
20 contribution of fine and coarse mode aerosols in terms of AOD₅₅₀. In this case, we choose to use
21 daily model fields of the dust contribution to the total AOD (here MACC reanalysis and
22 GOCART). We follow a method similar with the one presented in Bellouin et al. (2013).
23 Specifically, we calculate the dust AOD₅₅₀ by scaling the MODIS AOD₅₅₀ data with the MACC
24 or GOCART dust/total AOD₅₅₀ ratios [$f_d = \tau_{d(model)} / \tau_{(model)}$] on a daily basis.

25 Since the MACC data are available only from 2003 to 2012, in order to take advantage of the
26 full MODIS dataset (3/2000-12/2012), data from the GOCART model were used for the period
27 2000-2002. The GOCART data were normalized in order to be consistent with the MACC data.
28 Daily dust/total AOD₅₅₀ ratios (f_d) from the common GOCART-MACC period 2003-2007 were
29 first brought to a common 1° x 1° spatial resolution using bilinear interpolation and then we
30 calculated the regression line for each grid cell on a seasonal basis. The linear relations were
31 afterwards used in order to normalize the 2000-2002 GOCART ratios to have a homogeneous
32 dataset. The slopes and offsets of these regression lines and the corresponding correlation
33 coefficients (R) can be seen in Figs. S5, S6 and S7 of the Supplement, respectively. Overall, for

1 the whole time period, the MACC reanalysis f_d ratios are lower by $\sim 26\%$ from the GOCART f_d
2 ratios and the linear relation connecting the two products is $f_{dMACC}=0.4964f_{dGOCART}+0.0952$
3 with a correlation coefficient R of 0.74. The f_d values of the merged GOCART-MACC (2000-
4 2012) timeseries were checked using the Standard Normalized Homogeneity Test (SNHT) as
5 described in Alexandersson (1986). The statistical significance was checked following Khaliq
6 and Ouarda (2007) and the f_d timeseries were found to be homogeneous (see Fig. S8 of the
7 Supplement). Hence, this test verifies that the use of the merged GOCART-MACC f_d dataset
8 will not insert any artifacts (e.g. trends or breaks) in the algorithm. Finally, the f_d data were
9 brought to the same spatial resolution with MODIS data ($0.1^\circ \times 0.1^\circ$) using bilinear interpolation
10 again.

11 After the calculation of τ_d with the use of f_d values ($\tau_d=f_dAOD_{550}$), we proceed to the calculation
12 of the anthropogenic contribution to the total AOD_{550} (τ_a) by multiplying the non-dust part of
13 AOD_{550} with the anthropogenic fraction f_a for the region of Eurasia (0.77 ± 0.20) given in
14 Bellouin et al. (2013) [$\tau_a=f_a(1-f_d)AOD_{550}$]. The rest of the total AOD_{550} is attributed to the fine
15 mode natural aerosols [$\tau_n=(1-f_a)(1-f_d)AOD_{550}$] (see Fig. 2). As discussed in Bellouin et al.
16 (2013), the fine mode natural aerosols consist of sea salt, dimethyl sulfide from land and
17 oceanic sources, SO_2 from degassing volcanoes and secondary organic aerosols from biogenic
18 emissions. It has to be highlighted that like in the case of oceanic regions the biomass burning
19 aerosols are classified as anthropogenic by this algorithm. As shown in Figs. S4a-d, the seasonal
20 variability of τ_d over land covered regions is very close to the LIVAS dust AOD_{532} which is
21 used as a reference.

22 Overall, the algorithm described above performs well as far as dust is concerned. This is further
23 shown when comparing MODIS Terra and Aqua τ_d values with collocated AERONET
24 observations for dust dominated days (see Fig. S9 in the Supplement). The method followed for
25 the collocation of the data is similar to the one presented in Sect. 4.1 while dust dominated days
26 were days with an AERONET AE smaller than 1 (see Mateos et al., 2014) and a MODIS based
27 τ_d greater than τ_a and τ_n or τ_m . The uncertainties of the calculated τ_a , τ_d , τ_n and τ_m values which
28 are inserted by the input data and the assumptions of the algorithm are expected to be similar
29 with the ones presented in Bellouin et al. (2013). Bellouin et al. (2013) using a Monte-Carlo
30 analysis indicated that τ_a can be specified with an uncertainty of $\sim 23\%$ over land and $\sim 16\%$
31 over the ocean, τ_d can be specified with an uncertainty of $\sim 19\%$ over land and $\sim 33\%$ over the
32 ocean, τ_n can be specified with an uncertainty of $\sim 41\%$ and τ_m with an uncertainty of $\sim 28\%$.
33 The results of the application of the algorithm described in the paragraphs above are presented

1 in the following section (Sect. 4) by means of maps, pie charts, plots and tables for each one of
2 the three basic regions of interest and their nine sub-regions.

3

4 **4 Results and discussion**

5 **4.1 Validation of MODIS gridded data using ground-based observations**

6 As discussed in Sects. 2 and 3, the high quality (QAC: 3) DT level-2 Collection 051 MODIS
7 data used in this work were validated in detail against data from 13 AERONET stations (see
8 Fig. 1). The stations were selected to make sure that their version 2.0 level 2.0 high quality
9 cloud screened Cimel sunphotometric observations were covering at least 2 years and there
10 were at least 100 common days of AERONET and MODIS observations. The exact
11 geolocation of the AERONET stations is given in Table 1 (see also Fig.1) along with the
12 period of available data, the hosting country, the type of the station (e.g. urban/rural,
13 coastal/continental, etc.) and the corresponding mean overpass time of Terra and Aqua
14 MODIS. First, we collocated spatially and temporally the MODIS and AERONET
15 observations by temporally averaging AERONET measurements within ± 30 min from the
16 MODIS overpass time (see Levy et al., 2010) and spatially averaging MODIS measurements
17 centered within a $25 \times 25 \text{ km}^2$ window around each station (see Koukouli et al., 2010). The
18 use of a collocation window equal to the one used for the gridding procedure, practically,
19 allows us to validate at the same time the $0.1^\circ \times 0.1^\circ$ MODIS gridded product.

20 The regression lines between MODIS and AERONET AODs are shown in Fig. 3 while
21 details about the validation results can be found in Table 2. Overall, the MODIS Terra DT
22 Collection 051 data overestimate AOD_{550} by 11.59 % (Normalized Mean Bias - NMB) with
23 63.28 % of the data falling within the expected error (EE) envelope and 67.78% within the
24 pre-launch expected error (pLEE) envelope. The expected error envelope is define as: $\text{AOD} -$
25 $|\text{EE}| \leq \text{AOD}_{\text{MODIS}} \leq \text{AOD} + |\text{EE}|$ with EE being $\pm(0.05 \pm 0.15\text{AOD})$ (Levy et al., 2010) and
26 pLEE being $\pm(0.05 \pm 0.20\text{AOD})$ (Kaufman et al., 1997). On the other hand, the MODIS Aqua
27 DT Collection 051 data overestimate AOD_{550} by 25.18 % (NMB) with 57.14 % of the data
28 falling within the EE envelope and 61.87 % within the pLEE envelope. The percentage of the
29 MODIS Terra and Aqua data falling within the EE envelope are close to the 57 % given in
30 Remer et al. (2005) for the Eastern Mediterranean. The validation results for each station
31 separately can be found in Table S1 of the Supplement. The results discussed in this
32 paragraph are comparable to the ones appearing in previous studies focusing on the
33 Mediterranean region (see Papadimas et al., 2009; Koukouli et al., 2010). In general, it is

1 shown here that the MODIS Terra Collection 051 data exhibit a better agreement with the
2 ground-based observations from AERONET than MODIS Aqua data do. Therefore, the
3 statistics appearing for MODIS Terra throughout the paper could be considered more robust.
4 To be in line with the global validation of the DT Collection 051 product by Levy et al.
5 (2010) we also performed a validation with the specifications used in their work. We used a
6 50 x 50 km² window for the spatial collocation of the MODIS and AERONET data while
7 only days with at least 5 MODIS retrievals and 2 AERONET measurements were taken into
8 account. The increased size of the collocation window improves the results of the validation.
9 As shown in Table 2, MODIS Terra DT Collection 051 data overestimate AOD₅₅₀ by 5.10 %
10 (NMB) with 70.17 % of the data falling within the EE envelope and 74.64 % within the pLEE
11 envelope. For MODIS Aqua, the NMB is 15.34%, while the percentage of the measurements
12 falling within the EE and pLEE envelope is 66.76 % and 70.45 %, respectively. These results
13 for the Eastern Mediterranean are close to the global ones presented in Levy et al. (2010).
14 As discussed in Sect. 3.1, data from the DB algorithm were used over bright arid and semi-
15 arid regions of North Africa for the production of the 0.1° x 0.1° MODIS gridded dataset for
16 grid cells with no DT data. Therefore, in this work we also perform a validation of the DB
17 Collection 051 product over the region of the Eastern Mediterranean. In the case of DB data,
18 we first make use of all the available DB observations without any quality filtering over the
19 13 AERONET stations. A spatial window of 25-30 km has been typically used in the past for
20 the collocation of MODIS DB data with the AERONET observations (see Shi et al., 2011;
21 Ginoux et al.; 2012; Sayer et al., 2013; 2014) which is in line with the 25 x 25 km² window
22 used here. The MODIS Terra DB data overestimate AOD₅₅₀ by 21.38 % (NMB) with 51.90 %
23 of the data falling within the expected uncertainty (EU) envelope assuming a DB expected
24 uncertainty of $\pm 0.05 \pm 20\% \text{AOD}_{\text{AERONET}}$ (Hsu et al., 2006). The MODIS Aqua DB Collection
25 051 data overestimate AOD₅₅₀ by 33.03 % (NMB) with 55.30 % of the data falling within the
26 expected uncertainty envelope. We repeated the validation procedure for DB data taking into
27 account the highest quality data only. The sample of available measurements was diminished
28 by a factor of 5 in the case of MODIS Terra and 6 in the case of MODIS Aqua but the results
29 were pretty similar with the ones for the unfiltered data. Therefore, the use of unfiltered DB
30 data during the gridding procedure does not insert any significant uncertainty. The DB results
31 for the 13 AERONET stations examined here are not of the same agreement than the DT
32 results and the ones presented in previous studies utilizing DB Collection 051 data for other
33 stations and larger regions (see Shi et al., 2011; Ginoux et al., 2012). However, it has been

1 reported that stations in the region (e.g. Sede Boqer in Israel) are among the ones with the
2 greatest discrepancies between MODIS DB and AERONET measurements (Ginoux et al.,
3 2012). Nevertheless, as commented in Sect. 3.1, the DB data constitute only a small fraction
4 of the data used for the production of the MODIS gridded dataset (~ 1 % only of the 30000
5 grid cells covering the Eastern Mediterranean has only DB retrievals) and therefore they do
6 not affect significantly its quality. Only areas in Northern Africa are expected to be affected
7 by the use of DB data due the extended lack of DT data there.

8 As discussed in Sect. 3.1 the gridding procedure was repeated four times using a gridding
9 window of 25, 50, 75 and 100-km using MODIS Terra AOD₅₅₀ data for the year 2004 showing
10 that the 25-km window is optimal for capturing local pollution sources. In order to see how the
11 size of the gridding window affects the agreement between MODIS and AERONET data we
12 also proceeded to a validation of MODIS DT data against AERONET measurements using
13 different spatial collocation windows (25, 50, 75 and 100-km) and two quality criteria, a "strict"
14 one: at least 2 AERONET measurements for each MODIS-AERONET pair and "stricter" one:
15 at least 5 MODIS retrievals and 2 AERONET measurements for each MODIS-AERONET pair
16 as in Levy et al. (2010). The results for the DT MODIS Terra and Aqua data are presented in
17 Table S2 of the Supplement. In general, it is shown that the increased size of the spatial
18 collocation window leads to an improvement of the bias between satellite and ground-based
19 observations. This is probably due to the inclusion of more observations into the calculations
20 which diminishes the noise of the MODIS observations. In addition, as expected, the stricter
21 quality criteria lead to a better agreement between MODIS DT and AERONET data. Taking
22 into account not only the NMB but also the regression lines and the other metrics appearing in
23 Table 2S, it is concluded that the 50-km window is the best choice for the validation procedure
24 in line with Ichoku et al. (2002). On the other hand, the 25-km validation results are close to the
25 50-km ones (see Table S2) and at the same time the 25-km gridding window allows for a more
26 efficient detection of local aerosol sources as shown in Sect. 3.1. Taking this into account, we
27 suggest that the 25-km window used for the production of the 0.1° x 0.1° gridded MODIS
28 dataset is the optimal selection for studying the spatial variability of aerosols, preserving at the
29 same time the representativeness of the real aerosol load over each specific spot.

30

31 **4.2 Aerosol spatial variability and hot spots**

32 The AOD₅₅₀ spatial variability over the greater Eastern Mediterranean region for the period
33 2000-2012 as seen from the Terra MODIS 0.1° x 0.1° dataset is presented in Fig. 4. Several

1 aerosol hot spots that coincide with megacities (e.g. Cairo, Istanbul), large cities (e.g. Athens,
2 Ankara, Alexandria, Izmir, Thessaloniki) or even medium sized cities (e.g. Larissa,
3 Limassol), industrial zones (e.g. OSTIM Industrial Zone in Ankara, Turkey), power plant
4 complexes (e.g. Maritsa Iztok complex at the Stara Zagora Province in Bulgaria, Ptolemaida-
5 Kozani power plants in Western Macedonia, Greece), river basins (e.g. Evros river Basin at
6 the borders between Greece and Turkey), etc, can be detected on the map. Indicatively, in Fig
7 4 we give a list of 35 local particle pollution sources in the region; however, careful
8 inspection of this map and the seasonal maps presented in Fig. 6 allows for the detection of
9 many more aerosol sources. The results from the analysis of Aqua MODIS data are pretty
10 similar as shown in Fig. S10 of the Supplement. A significant number of the local aerosol
11 sources can also be detected on the OMI 2004-2012 tropospheric NO₂ and PBL SO₂ maps
12 given in Figs. 5a and b which reveals the origin of aerosols over these regions (e.g. traffic,
13 industrial activities, etc). However, there are regions of high aerosol load which cannot be
14 seen in Fig. 5a and b and vice versa which is indicative of the significant role of other
15 anthropogenic or natural processes that contribute to the local aerosol load (e.g. fires, soil dust
16 from agricultural activities or arid regions, Sahara dust transport).

17 The topography (Fig. 5c) and precipitation (see Fig. 5d for annual precipitation levels for the
18 period 2000-2012 from TRMM) are also major determinants of the local AOD₅₅₀ levels. For
19 example, regions with mountain ranges on the Balkan Peninsula (e.g. Pindus mountain range
20 in Greece, Dinaric Alps that run through Albania and the former Yugoslav republics, the
21 Balkan mountain range in Central Bulgaria) are characterized by low AODs (see Fig. 4). On
22 the contrary, regions of low altitude are generally characterized by higher AODs because the
23 majority of anthropogenic activities is usually concentrated there. Also, low altitude regions
24 surrounded by high mountains are characterized by higher AODs as aerosols cannot be easily
25 transported by the wind (e.g. the industrialized regions in Central Bulgaria which are confined
26 between the high Balkan and Rodopi mountain ranges). As precipitation is the major removal
27 mechanism of pollutants in the atmosphere, regions with high AOD₅₅₀ are in many cases
28 connected to low precipitation levels and vice versa (see Figs. 4 and 5d). It has to be
29 highlighted that the AOD₅₅₀ over these regions is high primarily due to the emissions and the
30 atmospheric processes forming aerosol particles. The low removal rates from precipitation
31 just preserve the AOD₅₅₀ levels high. A striking example is the region of Anatolia in Central
32 Turkey which is characterized by lower precipitation levels and higher aerosol loads

1 compared to the surrounding regions. Also, the low precipitation levels are partly responsible
2 for the high aerosol loads appearing over Northern Africa.

3 Overall, the mean AOD₅₅₀ for the whole period of interest is estimated at 0.215 ± 0.187 for
4 Terra and 0.217 ± 0.199 for Aqua MODIS for the Eastern Mediterranean region which is ~ 45
5 % higher than the global average appearing in recent studies (e.g. Kourtidis et al., 2015). Over
6 land higher mean AODs are generally recorded (0.219 ± 0.165 for Terra and 0.239 ± 0.189
7 for Aqua MODIS) than over the sea (0.213 ± 0.201 for Terra and 0.202 ± 0.205 for Aqua
8 MODIS). All these values along with the mean AODs for the 9 sub-regions of interest
9 covering the Eastern Mediterranean can be found in Table 3.

10 The AOD₅₅₀ spatial variability on a seasonal basis from MODIS Terra and Aqua is presented
11 in Fig. 6 along with the difference between the two products. The majority of the local aerosol
12 sources over land are more prominent in summer. The limited washout by precipitation (see
13 also Papadimas et al., 2008) and also the enhanced photochemical production of secondary
14 organic aerosols (Kanakidou et al., 2011 and references therein) contribute to the high AODs
15 appearing over local sources. In addition, during summer, over the region, there is typically a
16 significant transport of aerosols (e.g. see Kanakidou et al., 2011 and references therein) and
17 gaseous pollutants like SO₂ and NO₂ (see Georgoulas et al., 2009; Zyrichidou et al., 2009)
18 and biomass burning aerosols from Central-Eastern Europe. Over the sea, a profound
19 maximum is observed in spring extending across the North African coast and the neighboring
20 oceanic areas which is due to the well documented transport of significant amounts of dust
21 from the Sahara Desert (see Barnaba and Gobbi., 2004 and the list of references given in the
22 introduction). The seasonal variability of aerosols and the relative role of different aerosol
23 types and various processes is discussed in more details in Sect 4.4.

24 The difference between MODIS Terra and Aqua Collection 051 AOD₅₅₀ over the Eastern
25 Mediterranean is -0.002 (-1.40 %) for winter, -0.009 (-3.27 %) for spring, -0.011 (-4.46 %)
26 for summer and 0.008 (4.40 %) for autumn. AOD₅₅₀ levels from Terra MODIS are lower than
27 that from Aqua MODIS over land for all seasons. Over the sea, Terra MODIS AOD₅₅₀ levels
28 are lower than that of Aqua MODIS only in winter. The fact that Terra MODIS measurements
29 are systematically higher than that from Aqua over the sea by ~ 0.01 on an annual basis is in
30 line with the findings of previous global studies for Collection 5 (e.g. Remer et al., 2006;
31 2008). Locally, one can see regions with positive and negative differences between Terra and
32 Aqua MODIS AOD₅₅₀. The patterns of the Terra-Aqua difference per season are presented in
33 Figs. 6c, f, i and l while the patterns of the percent difference are given in Fig. S11 of the

1 Supplement. The largest part of the Terra-Aqua MODIS differences over land and sea which
2 are observed here may be attributed to the known calibration and sensor degradation issues of
3 MODIS (for details see Levy et al., 2010; 2013; Lyapustin et al., 2014; [Georgoulas et al.,](#)
4 [2016](#)). A significant effort has been undertaken to address these issues in the new (Collection
5 6) MODIS product (e.g. Levy et al., 2013; Lyapustin et al., 2014; [Georgoulas et al., 2016](#))
6 and a repetition of a similar analysis with Collection 6 data in the future would be a valuable
7 contribution. Taking into account the aforementioned issues and the retrieval uncertainty of
8 MODIS it becomes more than obvious that the attribution of observed differences between
9 Terra and Aqua to the diurnal variability of aerosol load (e.g. over biomass burning regions)
10 in the region is a difficult task. It is shown in Fig. S12 of the Supplement that the diurnal
11 variability of AOD₅₅₀ from AERONET ranges significantly from station to station. The
12 average hourly departure from the daily mean for the total of the 13 stations ranges from ~ -5
13 % to ~ 5 %. Specifically, for the MODIS Terra and Aqua overpass times, the AERONET
14 AOD₅₅₀ difference ranges from ~ -10 % to ~ 10 % (see Fig. S12b). The Terra-Aqua AOD₅₅₀
15 difference is negative for the total of the 13 stations ranging from ~ -25 % to ~ -5 %. It is
16 shown in Fig. S12b that the two differences exhibit a similar variability from station to station
17 which indicates that part of the observed Terra-Aqua difference is indeed due to the diurnal
18 variability of aerosols. However, as mentioned above, the diurnal variability of aerosols is a
19 very delicate issue and should be comprehensively addressed in a future study. The same
20 stands for other kind of variabilities which could be connected to local and regional
21 anthropogenic activities like e.g. the weekly cycle of aerosols (see Georgoulas and Kourtidis,
22 2011; Georgoulas et al., 2015).

23

24 **4.3 Contribution of different aerosol types to the total AOD₅₅₀**

25 **4.3.1 Annual contribution**

26 As mentioned above, we attempt to estimate in our work the contribution of different aerosol
27 types to the total AOD₅₅₀ over the region of the Eastern Mediterranean was calculated
28 following the methodology presented in Sect. 3.2. For the land covered areas, based on
29 MODIS Terra observations, we estimate that 52 % (0.112±0.087) of the total AOD₅₅₀ is due
30 to anthropogenic aerosols, 32 % (0.074±0.080) due to dust and 16 % (0.034±0.026) due to
31 fine mode natural aerosols (see Fig. 7). For the oceanic areas, 41 % (0.086±0.085) of the total
32 AOD₅₅₀ is due to anthropogenic aerosols, 34 % (0.076±0.185) due to dust and 25 %
33 (0.054±0.018) due to marine aerosols (see Fig. 7). The results based on observations from

1 MODIS Aqua are similar. Over land, 50 % (0.117 ± 0.093) of the total AOD_{550} is
2 anthropogenic, 35 % (0.090 ± 0.102) is due to dust and 15 % (0.035 ± 0.028) due to fine mode
3 natural aerosols, while, over the sea, 40 % (0.079 ± 0.080) of the total AOD_{550} is of
4 anthropogenic origin, 33 % (0.070 ± 0.181) is due to dust and 27 % (0.054 ± 0.018) due to
5 marine aerosols (see Fig. 7). These results along with the relative contributions and the annual
6 τ_a , τ_d , τ_n and τ_m levels for each one of the nine sub-regions of interest (see Fig. 1) are given in
7 Table 4.

8 For anthropogenic aerosols, the region with the highest relative contribution is NBL (59 % for
9 both Terra and Aqua MODIS) while the region with the lowest relative contribution is SWO
10 (32 % for both Terra and Aqua MODIS) (see also Table 4). The spatial variability of τ_a is
11 presented in Fig. 8a for Terra MODIS and Fig. S13a of the Supplement for Aqua MODIS, the
12 patterns being similar in both cases. Over land, the annual τ_a patterns are similar to the
13 AOD_{550} patterns, the highest values appearing over local particle pollution sources (cities,
14 industrial zones, etc.). Over the sea, τ_a is higher along the coasts, while it drops significantly
15 towards other directions. An interesting feature here is that the oceanic region of Black Sea
16 (BSO) presents higher relative anthropogenic contributions than the rest of the oceanic sub-
17 regions but also than land areas with significant anthropogenic sources (e.g. ANL and NAL).
18 This is indicative of the transport of atmospheric particles from Central Europe and biomass
19 burning aerosols during the biomass burning seasons in April-May from Russia (across the
20 latitudinal zone 45°N - 55°N) and July-August from South-Western Russia and Eastern Europe
21 (Amiridis et al., 2010). These aerosols are transported at much lower latitudes as shown in
22 previous studies (e.g. Vrekoussis et al., 2005; Karnieli et al., 2009) reaching the Sahara Desert
23 and the Middle East regions (Pozzer et al., 2015). The fact that τ_a drops gradually from the
24 coasts is also seen in Fig. 9 where the latitudinal variability of the optical depths of the
25 different aerosol types (τ_a , τ_d , τ_n and τ_m) is presented for four bands that cover the whole
26 Eastern Mediterranean. An interesting feature is that τ_a increases nearby the shoreline
27 (particularly along the North African coastal zone) before it gradually decreases. Over land
28 aerosols are located within the atmospheric boundary layer, close to the emission sources, and
29 hence, their deposition and removal from the atmosphere is more efficient than over the sea.
30 The particles which are transported over the sea on the other hand usually reach greater
31 heights which prolongs their lifetime.

32 As shown in Fig. 9, the same feature is observed for dust. Indicatively, τ_d and the relative
33 contribution of dust to the total AOD_{550} on an annual basis over the oceanic regions of SWO

1 and SEO are in general higher or comparable to the ones over NAL (see Table 4 for more
2 details). In Fig. 9, the MODIS-based τ_d latitudinal variability is presented along with the
3 latitudinal variability of dust AOD₅₃₂ and extinction coefficients of dust at 532 nm from
4 LIVAS. As expected, in all cases τ_d decreases with distance from the large dust sources in the
5 South and South-East (Sahara Desert, Middle East deserts) with local maxima over the
6 latitudinal zone from 35°N to 40°N (especially for band 2 and band 3). The latitudinal
7 variability of τ_d is similar to the latitudinal variability of dust AOD₅₃₂ for all the four bands
8 despite the fact that the MODIS-based data have a resolution 100 times higher (0.1° vs 1°) and
9 therefore are more sensitive to local characteristics. Dust reaches heights up to ~ 4-5 km in the
10 area; however, the largest fraction of dust mass is confined within the first 2-3 km of the
11 troposphere (see Fig. 9). The annual τ_d patterns are shown in Fig. 8b for Terra MODIS (Fig.
12 S13b of the Supplement for Aqua MODIS). The main dust transport pathways over the
13 oceanic areas of the Eastern Mediterranean can be seen along with various local maxima over
14 land. The highest τ_d values over land appear over the regions of NAL and ANL (see Table 4)
15 and along the coasts. The high dust concentrations appearing over these regions are not only
16 due to the transport of dust from the nearby deserts but also due to local dust sources. A
17 recent study by Liora et al. (2015) reports various local sources of wind blown dust along the
18 coastal regions of Greece and Turkey, over the region of Anatolia in Turkey, over the Greek
19 islands, Crete, Cyprus and regions close to the coastal zone of Middle East. Their results are
20 in good agreement with the τ_d patterns presented in this work.

21 As shown in Fig. 7, fine mode natural aerosols exhibit the lowest contribution to the total
22 AOD₅₅₀ compared to the other aerosol types over land. The spatial variability of τ_n is very low
23 compared to τ_a and τ_d as shown in Figs. 8c and 9. It is inferred from the values appearing in
24 Table 4 that τ_n increases slightly as one moves from North to South; however, the relative
25 contribution of fine mode natural aerosols to the total AOD₅₅₀ slightly decreases (i.e. 17.67 %
26 over NBL and 14.97 % over NAL according to Terra MODIS observations). The latitudinal
27 variability and the percentages appearing in Table 4 are in accordance to the relative
28 contributions of biogenic aerosols to the total AOD₅₅₀ appearing over the Eastern
29 Mediterranean in a recent modeling study (Rea et., 2015).

30 Similar to fine mode natural aerosols over land, marine aerosols generally have the lowest
31 contribution to the total AOD₅₅₀ compared to the other aerosol types over the sea (see Fig. 7
32 and Table 4) except for BSO. The variability of τ_m is very low compared to τ_a and τ_d . On an
33 annual basis, high τ_m values appear over the Aegean Sea and the oceanic area between Crete

1 and the North African coast while slightly lower values appear along the coasts of the Eastern
2 Mediterranean (see Figs. 8d and 9). The τ_m patterns follow the near surface wind speed
3 patterns in the region (see Fig. S14 of the Supplement) being in accordance to the τ_m , marine
4 particulate matter concentration or sea salt emission patterns appearing in other studies (Im et
5 al., 2012; Nabat et al., 2013; Rea et al., 2015; Liora et al., 2015).

6

7 **4.3.2 Seasonal contribution**

8 The contribution of different aerosol types to the total AOD₅₅₀ over the Eastern Mediterranean
9 varies from season to season. The relative contribution of each aerosol type over EML and
10 EMO for each season is shown in Fig. 10. Over land, the relative contribution of τ_a , τ_d and τ_n
11 to the total AOD₅₅₀ exhibits a low seasonal variability. The relative contribution of
12 anthropogenic aerosols to the total AOD₅₅₀ ranges from 49 % in SON to 55 % in DJF based
13 on Terra MODIS observations and from 48 % in MAM and SON to 52 % in JJA based on
14 Aqua MODIS observations. In contrast, over the oceanic regions the relative contribution of
15 τ_a , τ_d and τ_m to the total AOD₅₅₀ exhibits a significant seasonal variability. The relative
16 contribution of anthropogenic aerosols to the total AOD₅₅₀ ranges from 27 % / 27 % in DJF to
17 50 % / 47 % in JJA based on Terra/Aqua MODIS observations. The percentages appearing
18 here are in accordance to the values appearing in Hatzianastassiou et al. (2009) where a
19 different satellite-based approach was followed. Indicatively, for the greater Athens area, an
20 average summertime anthropogenic contribution of ~ 50 % was found here based on Terra
21 MODIS data which is within the summer period range of 47-61 % indicated in the study by
22 Hatzianastassiou et al. (2009). In addition, the corresponding values for the greater
23 Thessaloniki area, Crete, Cairo and Alexandria are 53 %, 38 %, 48 % and 41 %, respectively,
24 within the range of values (57-73 %, 36-52 %, 34-56 % and 23-60 %) shown in
25 Hatzianastassiou et al. (2009). Only in the case of Ankara, our results suggest a lower
26 anthropogenic contribution (52 % versus 71-84 %). Particularly for Athens, Gerasopoulos et
27 al. (2011) following a different approach incorporating ground-based AOD observations and
28 trajectory modeling reached similar results (annual contribution of ~ 62 % from local and
29 regional sources and continental Europe which is expected to be mostly of anthropogenic
30 origin). Similarly, for Crete, Bergamo et al. (2008) using a different approach, also utilizing
31 ground-based data, found an annual anthropogenic contribution of ~ 43 %.

32 The seasonal patterns of the anthropogenic aerosols (τ_a) over the Eastern Mediterranean based
33 on MODIS Terra observations are presented in Figs. 11a, e, i and m while the seasonal

1 variability of τ_a over the whole region, over the land covered part and the oceanic part and
2 over the 9 sub-regions of interest is presented in Fig. 12. The results based on MODIS Aqua
3 observations are similar and can be found in Figs. S15a, e, i and m and Fig. S16 of the
4 Supplement. Generally, the local hot spots are detectable throughout the year; however, they
5 are becoming much more discernible in spring and especially in summer. As shown in Fig.
6 12a, τ_a nearly doubles during the warm period of the year (spring-summer) with the seasonal
7 variability being stronger over the sea (Fig. 12c) than over land (Fig. 12b). A clear peak is
8 observed in summer, August being the month with highest τ_a levels. As discussed in Sect.
9 4.3.1 the summer peak is mostly a result of three basic reasons. The first one is the deficiency
10 of wet removal processes compared to the cold period. As shown in Fig. S17, based on the
11 TRMM satellite observations, August and July are the months with the lowest precipitation
12 levels over the land covered part (a drop of $\sim 75\%$ compared to winter months) and over the
13 oceanic part (a drop of $\sim 90\%$ compared to winter months) of the Eastern Mediterranean,
14 respectively. The second reason is the enhancement of the photochemical production of
15 secondary organic aerosols in summer (Kanakidou et al., 2011) and the third reason is the
16 transport of pollution aerosols from Central Europe and biomass burning aerosols from South-
17 Western Russia and Eastern Europe during the biomass burning season in July-August
18 (Amiridis et al., 2010). The Etesians, which are persistent northerly winds that prevail over
19 the Eastern Mediterranean during summer, bring dry and cool air masses and aerosols from
20 the regions mentioned above while blocking at the same time the transport of desert dust in
21 the region and dispersing local pollution in urban areas to levels typical for rural areas (see
22 Tyrllis and Lelieveld, 2013 and references therein). As seen in Figs. 12a-l, a smaller but
23 distinct in most cases τ_a peak appears in April mostly as a result of the transport of biomass
24 burning aerosols from Russia (across the latitudinal zone 45°N - 55°N). This is in line with the
25 findings of Sciare et al. (2008) who detected traces of these biomass burning aerosols at the
26 island of Crete in Southern Greece.

27 As discussed above, the relative contribution of dust to the total AOD_{550} over land exhibits a
28 low seasonal variability ranging from 29 % in DJF to 36 % in SON based on Terra MODIS
29 observations and from 33 % in JJA to 38 % in SON based on Aqua MODIS observations (see
30 Fig. 10). Over the oceanic regions the relative contribution of dust to the total AOD_{550} ranges
31 significantly throughout a year from 26 % / 28 % in JJA to 42 % / 39 % in MAM based on
32 Terra/Aqua MODIS observations. The percentages appearing here are in accordance to model
33 and observational studies. For example, de Meij et al. (2012) using the atmospheric chemistry

1 general circulation model EMAC (ECHAM/MESSy Atmospheric Chemistry) showed that
2 dust contributes on an annual level $\sim 30\%$ to the total AOD_{550} over stations located in the
3 area of the Eastern Mediterranean. Gerasopoulos et al. (2011) found a $\sim 23\%$ percent
4 contribution of North African dust to the total AOD over Athens using ground-based AOD
5 observations and trajectory modeling. Taking into account that part of the $\sim 39\%$ local and
6 regional sources appearing in Gerasopoulos et al. (2011) is due to local dust sources,
7 especially in summer, turns out that their results are in agreement with the $\sim 33\%$ relative
8 contribution found in this work for the greater Athens area based on Terra MODIS
9 observations. The seasonal patterns of dust (τ_d) over the region based on Terra MODIS
10 observations are shown in Figs. 11b, f, j and n while the seasonal variability of τ_d over the
11 whole region, over land, over the sea and over the 9 sub-regions of interest is shown in Fig.
12 12. The corresponding results based on MODIS Aqua observations are pretty similar and can
13 be found in Figs. S15b, f, j and n and Fig. S16 of the Supplement.

14 As seen in Fig. 11f, in spring, mostly due to the strong Sahara dust events, very high τ_d values
15 appear over land regions in North Africa, Middle East, Anatolia and oceanic areas across the
16 Eastern Mediterranean (especially below 35°N). Dust loading over the sea exhibits two
17 maxima, one at the coastal zone of Libya and one across the coastal zone of Middle East. The
18 same two maxima but with much lower τ_d values appear in summer (Fig. 11j) and autumn
19 (Fig. 11n). Over land, the τ_d patterns are similar in summer and autumn, the maximum values
20 appearing over the Anatolian Plateau and areas of North Africa and Middle East. During
21 winter, dust maxima appear across the coastal zone of Northern Africa with relatively low τ_d
22 values across the coastal zone of Middle East (Fig. 11b). In winter τ_d levels are low over land
23 compared to the other seasons (Figs. 11b, f, j and n) as precipitation levels (see Fig. S17 of the
24 Supplement) and hence wet scavenging of aerosols peak. At the same time, the local
25 emissions of dust are low for regions away from the large area sources in the South (Liora et
26 al., 2015). In contrast, over the sea τ_d levels in winter are similar or slightly higher for some
27 areas than that in summer and autumn (see Figs. 11 and 12) as this is the season with the
28 second highest frequency (after spring) of strong ($\sim 21\%$) and extreme ($\sim 26\%$) desert dust
29 episodes in the region (see Gkikas et al., 2013 for details). February is by far the winter month
30 with the highest τ_d levels (see Fig. 12) in line with the findings of Pey et al. (2013) who
31 showed that the intensity of African dust episodes over stations in Greece and Cyprus peaks
32 in February. Dust exhibits a strong peak in spring, April being the month with the highest τ_d
33 levels in line with other studies (e.g. Israelevic et al., 2012; Varga et al., 2014). The peak in

1 April is a result of the high cyclonic activity over North Africa during this month as shown by
2 Flaounas et al. (2015). According to the same study, low pressure systems are responsible for
3 ~ 10-20 % of moderate and ~ 40-50 % of high and extreme Sahara dust transport events over
4 the Eastern Mediterranean. North Africa (Sharav) cyclones develop mainly in spring and
5 summer while Mediterranean cyclones develop in winter and autumn. The Mediterranean
6 cyclones are more intense than Sharav cyclones. The region, is also affected by events
7 bringing particles from dust source regions in the eastern part of the Mediterranean basin
8 (Negev desert in Israel, Sinai in Egypt, Anatolian Plateau in Turkey) and the Arabian deserts
9 (Basart et al. 2009; Pey et al., 2013; Abdelkader et al., 2015). Dust remains in the atmosphere
10 for a period of 1-4 days undergoing chemical aging before being removed (see Abdelkader et
11 al. 2015 and references therein). The seasonal variability of τ_d is much stronger and the spring
12 maxima much more prominent over the sea (see Fig. 12). This is expected, as dust is only
13 occasionally transported over the sea during episodic events, while over land, local sources
14 also contribute to the dust burden especially in summer due to the dryness of soil. For
15 example, over NBL, a broad spring-summer peak is observed, June being the month with the
16 highest τ_d levels. As one moves south (SBL, ANL and NAL) the April peak becomes more
17 prominent.

18 The relative contribution of fine mode natural aerosols to the total AOD_{550} over land exhibits
19 a very low seasonal variability ranging from 15 % in MAM and SON to 16 % in DJF and JJA
20 based on Terra MODIS observations and from 14 % in DJF and SON to 15 % in MAM and
21 JJA based on Aqua MODIS observations (see Fig. 10). The seasonal variability is also very
22 low, the highest values appearing in spring and summer (Fig. 12). Despite the generally low
23 contribution of fine mode natural aerosols to the total AOD_{550} over the Eastern
24 Mediterranean, τ_n levels are similar to τ_d levels during winter months over specific regions
25 (NBL and SBL). The low seasonal variability can also be seen in Figs. 11c, g, k and o where
26 the patterns of fine mode natural aerosols (τ_n) are presented.

27 The seasonal relative contribution of marine aerosols to the total AOD_{550} over the oceanic
28 regions of the Eastern Mediterranean is shown in Fig. 10. τ_m ranges from 20 % in MAM to 35
29 % in DJF based on Terra MODIS observations and from 21 % in MAM to 36 % in DJF based
30 on Aqua MODIS observations (see Fig. 10). Like in the case of fine mode natural aerosols,
31 the seasonal variability is very low, but here the highest values appear in winter (Fig. 12). Due
32 to the linear relation of τ_m and near surface wind speed within our algorithm (see Fig. 2) the
33 τ_m seasonal variability and patterns follow the wind speed ones (see Figs. 11d, h, l, p and

1 S14). Marine aerosol concentrations are lower close to the coastlines while the highest
2 concentrations (see Liora et al., 2015) and τ_m values within the Eastern Mediterranean appear
3 over the Aegean Sea (see Fig. 11). Overall, the τ_m patterns are in accordance to the τ_m , marine
4 particulate matter concentration and sea salt emission patterns from previous studies (Im et
5 al., 2012; Nabat et al., 2013; Rea et al., 2015; Liora et al., 2015).

6 7 **5 Summary and conclusions**

8 In this work, satellite data from MODIS Terra (3/2000-12/2012) and Aqua (7/2002-12/2012)
9 were analyzed separately in order to examine the spatial and temporal variability of aerosols
10 over the Eastern Mediterranean. A high resolution ($0.1^\circ \times 0.1^\circ$) MODIS gridded dataset was
11 compiled using a method that could be used in future regional studies. A number of tests were
12 implemented and the dataset was validated in detail using sunphotometric observations from
13 13 AERONET stations. According to the validation, the statistics appearing for MODIS Terra
14 throughout the paper could be considered more robust while areas in Northern Africa are
15 expected to be affected by the extended use of DB data which do not exhibit a very good
16 matching with the ground-based observations. It is shown that the gridding method we use
17 offers the best compromise for studying the spatial variability of aerosols on a regional or
18 local scale, preserving at the same time the representativeness of the real aerosol load over
19 each specific spot.

20 Based on MODIS observations the average AOD₅₅₀ levels over the region of the Eastern
21 Mediterranean are $\sim 0.22 \pm 0.19$ which is $\sim 45\%$ higher than the global mean. A number of
22 aerosol hot spots that coincide with megacities, large and even medium size cities, industrial
23 zones, power plant complexes, river basins, etc., can be detected on the AOD maps. A
24 number of local aerosol sources can also be seen on satellite retrieved tropospheric NO₂ and
25 planetary boundary layer SO₂ maps from OMI/AURA. This is indicative of the strong
26 presence of anthropogenic aerosols over these regions. Topography and precipitation also
27 play an important role. Generally, regions with mountain ranges are characterized by low
28 AODs while regions of low altitude are characterized by higher AODs. Regions with high
29 AOD₅₅₀ are in many cases connected to low precipitation levels and vice versa. Precipitation
30 is the major washout mechanism of atmospheric pollutants., Low removal rates from
31 precipitation contribute in preserving high the AOD₅₅₀ levels which are a result of emissions
32 and other atmospheric processes.

1 The AOD₅₅₀ patterns over the Eastern Mediterranean exhibit a significant seasonal variability
2 which is mostly driven by precipitation, photochemical production of secondary organic
3 aerosols, transport of pollution and biomass burning aerosols from Central and Eastern
4 Europe and transport of dust from the Sahara Desert and the Middle East. Differences
5 between MODIS Terra and Aqua Collection 051 AOD₅₅₀ over the Eastern Mediterranean are
6 generally small (~ -8 % over land and ~ 5 % over the sea). The comparison of the Terra-Aqua
7 differences with diurnal variabilities from the AERONET stations showed that only a part of
8 the observed differences is due to the diurnal variability of aerosols.

9 The MODIS data were combined with data from other satellites (Earth Probe TOMS,
10 OMI/AURA), reanalysis projects (ERA-Interim, MACC) and a chemistry-aerosol-transport
11 model (GOCART) to calculate the contribution of different types of aerosols to the total
12 AOD₅₅₀. The algorithm used was optimized for the Eastern Mediterranean through a number
13 of tests and comparison with LIVAS CALIOP/CALIPSO dust retrievals and AERONET
14 ground-based observations. A different approach is used for land and sea as there is not any
15 reliable satellite retrieved quantity to separate the contribution of fine and coarse mode
16 aerosols over water surfaces.

17 Overall, for the land areas, based on MODIS Terra observations, 52 % (0.112±0.087) of the
18 total AOD₅₅₀ is due to anthropogenic aerosols, 32 % (0.074±0.080) due to dust and 16 %
19 (0.034±0.026) due to fine mode natural aerosols (see Fig. 7). For the oceanic areas, 41 %
20 (0.086±0.085) of the total AOD₅₅₀ is due to anthropogenic aerosols, 34 % (0.076±0.185) due
21 to dust and 25 % (0.054±0.018) due to marine aerosols. The results based on observations
22 from MODIS Aqua are in accord with previous studies.

23 Over land, the τ_a maxima are detected over local particle pollution sources (cities, industrial
24 zones, etc.). Over the sea, τ_a is higher along the coasts being significantly lower at greater
25 distance. Very high τ_d values appear over land regions in North Africa, Middle East, Anatolia
26 and oceanic areas across the Eastern Mediterranean, especially for latitudes below 35°N. Over
27 the sea, dust loading exhibits two maxima, one at the coastal zone of Libya and one across the
28 coastal zone of the Middle East. τ_d decreases with distance from the large dust sources in the
29 South and South-East. Generally, dust reaches heights up to ~ 4-5 km in the area, the largest
30 fraction of dust mass being confined within the first 2-3 km of the troposphere. The spatial
31 variability of τ_n and τ_m is very low compared to τ_a and τ_d , following the total AOD₅₅₀ patterns
32 and the near surface wind speed patterns, respectively.

1 Over land, the relative contribution of anthropogenic aerosols, dust and fine mode natural
2 aerosols to the total AOD₅₅₀ exhibits a low seasonal variability, while over the sea the relative
3 contribution of anthropogenic aerosols, dust and marine aerosols shows a significant seasonal
4 variability.

5 τ_a nearly doubles during the warm period of the year (spring-summer), August and April
6 being the months with the highest τ_a levels. The summer peak is mostly the result of low
7 precipitation levels, enhancement of the photochemical production of secondary organic
8 aerosols and transport of pollution aerosols from Central Europe and biomass burning
9 aerosols from South-Western Russia and Eastern Europe during the biomass burning season
10 in July-August. The spring maximum in April is mostly the result of transport of biomass
11 burning aerosols from Russia in line with previous studies. Dust exhibits a strong peak in
12 spring (April), especially over the southern regions. April is the month with the highest τ_d
13 levels as a result of the high cyclonic activity over North Africa. The seasonal variability of
14 dust is much stronger and the spring maxima much more prominent over the sea as dust is
15 only occasionally transported there during episodic events, while over land, local sources
16 contribute to the dust burden, especially in summer due to the soil dryness. The seasonal
17 variability of fine mode natural aerosols is very low, the highest values appearing in spring
18 and summer. Marine aerosols also present a very low seasonal variability, the highest values
19 appearing in winter due to the high near surface wind speeds.

20 Overall, it is suggested that the AOD₅₅₀, τ_a , τ_d , τ_n and τ_m high resolution gridded dataset which
21 was compiled in this work could be used in a number of future atmospheric and biological
22 studies focusing on the region of the Eastern Mediterranean (e.g. satellite and ground-based
23 studies on aerosol-cloud-radiation interactions, experimental and field campaign studies on
24 aerosols and clouds and research on the impact of aerosols on human health and nature). It is
25 also acknowledged that a future update of the results presented here using more recent
26 releases of MODIS aerosol data (e.g. Collection 6) and aerosol reanalysis datasets (e.g.
27 NASA's Modern-Era Retrospective Analysis For Research And Applications Aerosol Re-
28 analysis) would be a useful contribution.

29

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23

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1 **Table 1.** Full name, abbreviation, geolocation, host country and type of the 13 AERONET
 2 cimel sunphotometer sites used for the validation of MODIS Terra and Aqua Collection 051
 3 observations. The common measurement period of MODIS and AERONET data and the
 4 corresponding overpass time of MODIS Terra and Aqua (*Italics*) over each station are also
 5 given.

6

AERONET Station	Lat (°N)	Lon (°E)	Period of study	Country	Type	TERRA overpass	AQUA overpass
ATHENS-NOA (ATH)	37.988	23.775	05/2008-10/2012	Greece	Urban (coastal)	9:23±22min UT	<i>11:32±22min UT</i>
Bucharest Inoe (BUC)	44.348	26.030	07/2007-09/2012	Romania	Sub-urban (coastal)	9:17±24min UT	<i>11:15±20min UT</i>
CUT-TEPAK (CUT)	34.675	33.043	04/2010-12/2012	Cyprus	Urban (coastal)	8:43±25min UT	<i>10:55±25min UT</i>
Eforie (EFO)	44.075	28.632	09/2009/12/2012	Romania	Rural (coastal)	9:09±21min UT	<i>11:04±21min UT</i>
FORTH Crete (FOR)	35.333	25.282	01/2003-08/2011	Greece	Rural (coastal)	9:12±24min UT	<i>11:25±25min UT</i>
IMS-METU-ERDEMLI (IMS)	36.565	34.255	01/2004-01/2012	Turkey	Rural (coastal)	8:39±23min UT	<i>10:48±22min UT</i>
Lecce University (LEC)	40.335	18.111	03/2003-12/2012	Italy	Sub-urban (coastal)	9:44±25min UT	<i>11:49±25min UT</i>
Nes ziona (NES)	31.922	34.789	02/2000-12/2012	Israel	Sub-urban (coastal)	8:38±24min UT	<i>10:44±25min UT</i>
SEDE BOKER (SED)	30.855	34.782	01/2000-04/2012	Israel	Rural (semi-arid)	8:30±27min UT	<i>10:50±25min UT</i>
Sevastopol (SEV)	44.616	33.517	05/2006-12/2012	Ukr.-Crimea	Urban (coastal)	8:51±21min UT	<i>10:40±21min UT</i>
Thessaloniki (THE)	40.630	22.960	09/2005-12/2012	Greece	Urban (coastal)	9:28±25min UT	<i>11:32±22min UT</i>
TUBITAK UZAY Ankara (TUB)	39.891	32.778	12/2009-04/2012	Turkey	Urban (continental)	8:48±26min UT	<i>10:56±24min UT</i>
Xanthi (XAN)	41.147	24.919	01/2008-10/2010	Greece	Rural (coastal)	9:18±25min UT	<i>11:24±21min UT</i>

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1 **Table 2.** Results of the comparison of spatially (using a spatial window around each station)
2 and temporally (± 30 min from the MODIS overpass time) collocated MODIS Terra and Aqua
3 (Italics) Collection 051 level-2 and AERONET sunphotometric (quadratically interpolated)
4 AOD₅₅₀ observations for the Eastern Mediterranean stations. The algorithms used for the
5 production of the validated MODIS data (DT and DB), the spatial window used for the spatial
6 collocation (25 x 25 km² or 50 x 50 km² window around each station) with the AERONET
7 data, the average MODIS and AERONET AOD₅₅₀ and the corresponding $\pm 1\sigma$ values, the
8 mean difference between them, the normalized mean bias (NMB) and the corresponding root
9 mean square (RMS) error, the percentage of the collocation points that fall within the
10 expected error (EE) envelope and the pre-launch expected error (pLEE) envelope (Expected
11 Uncertainty - EU envelope for DB data), the correlation coefficient R, the slope a and the
12 intercept of the regression line and the number of the collocation points are given in the table.
13 L10 denotes the use of a collocation window of 50^o x 50^o as in Levy et al. (2010) while HQ
14 denotes the use of high quality data only.

15

Alg.	Window	MODIS TERRA MODIS AQUA	AERONET	Mean Diff.	NMB %	RMS err.	in EE %	in pl EE %	R	a	b	Obs
DT	25 km	0.223±0.163	0.200±0.123	0.023±0.106	11.59	0.11	63.28	67.78	0.76	1.007	0.022	6697
<i>DT</i>	<i>25 km</i>	<i>0.247±0.173</i>	<i>0.197±0.121</i>	<i>0.050±0.109</i>	<i>25.18</i>	<i>0.12</i>	<i>57.14</i>	<i>61.87</i>	<i>0.78</i>	<i>1.113</i>	<i>0.027</i>	<i>6283</i>
DT	50 km (L10)	0.204±0.152	0.194±0.124	0.010±0.085	5.10	0.09	70.17	74.64	0.83	1.016	0.007	6054
<i>DT</i>	<i>50 km (L10)</i>	<i>0.224±0.155</i>	<i>0.194±0.125</i>	<i>0.030±0.088</i>	<i>15.34</i>	<i>0.09</i>	<i>66.76</i>	<i>70.45</i>	<i>0.82</i>	<i>1.018</i>	<i>0.026</i>	<i>5557</i>
DB	25 km	0.226±0.177	0.186±0.128	0.040±0.162	21.38	0.17	-	51.90	0.47	0.657	0.104	2580
<i>DB</i>	<i>25 km</i>	<i>0.242±0.217</i>	<i>0.182±0.118</i>	<i>0.06±0.196</i>	<i>33.03</i>	<i>0.20</i>	-	<i>55.30</i>	<i>0.44</i>	<i>0.815</i>	<i>0.094</i>	<i>5345</i>
DB _{HQ}	25 km	0.229±0.158	0.186±0.132	0.043±0.141	22.82	0.15	-	52.41	0.54	0.651	0.108	498
<i>DB_{HQ}</i>	<i>25 km</i>	<i>0.260±0.220</i>	<i>0.186±0.138</i>	<i>0.074±0.204</i>	<i>39.84</i>	<i>0.22</i>	-	<i>52.34</i>	<i>0.42</i>	<i>0.670</i>	<i>0.136</i>	<i>896</i>

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1 **Table 3.** AOD₅₅₀ levels, the corresponding $\pm 1\sigma$ values and the number of gridded values used
 2 for the calculations over Eastern Mediterranean (EMT), over the land covered part (EML),
 3 over the oceanic part and over the 9 sub-regions of Eastern Mediterranean appearing in Fig. 1
 4 based on the MODIS Terra and Aqua (Italics) observations.
 5

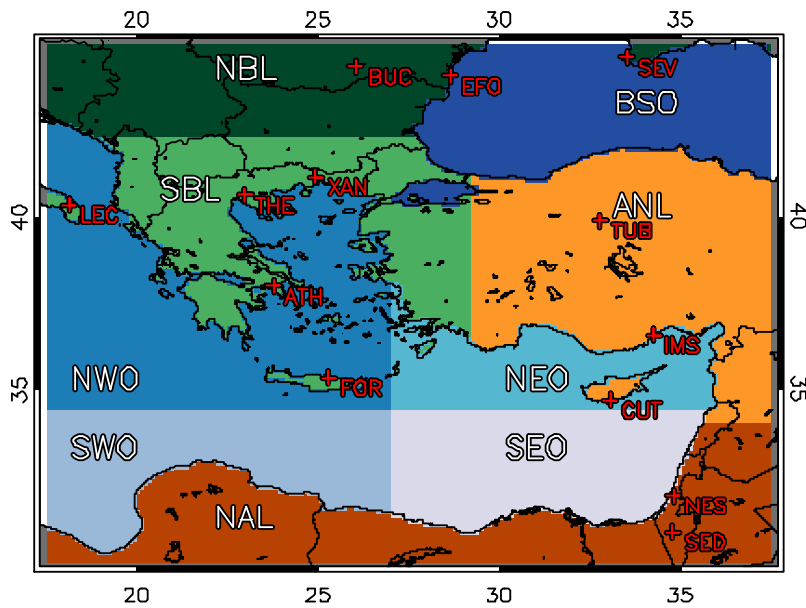
Region	MODIS TERRA AOD ₅₅₀	Num. of values	<i>MODIS AQUA</i> <i>AOD₅₅₀</i>	Num. of values
EMT	0.215±0.187	61496654	<i>0.217±0.199</i>	<i>49522934</i>
EML	0.219±0.165	25923766	<i>0.239±0.189</i>	<i>21008713</i>
EMO	0.213±0.201	35572888	<i>0.202±0.205</i>	<i>28514221</i>
NBL	0.183±0.163	5563495	<i>0.187±0.162</i>	<i>3853688</i>
SBL	0.197±0.152	7345829	<i>0.207±0.152</i>	<i>5272449</i>
ANL	0.223±0.146	7948817	<i>0.228±0.148</i>	<i>5539261</i>
NAL	0.282±0.192	5065625	<i>0.306±0.238</i>	<i>6343315</i>
BSO	0.198±0.150	6433951	<i>0.183±0.134</i>	<i>5262438</i>
NWO	0.209±0.162	11645069	<i>0.197±0.154</i>	<i>9231630</i>
SWO	0.226±0.266	6202893	<i>0.223±0.310</i>	<i>4925665</i>
NEO	0.214±0.196	4807910	<i>0.199±0.166</i>	<i>3896554</i>
SEO	0.221±0.236	6483065	<i>0.210±0.239</i>	<i>5197934</i>

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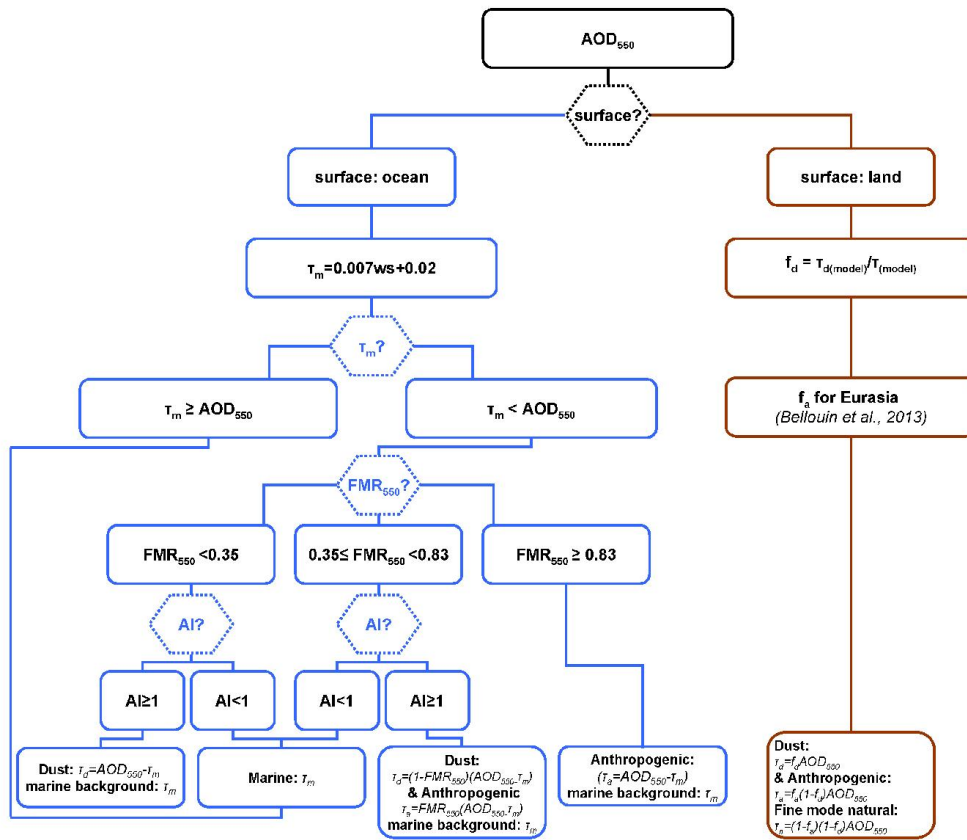
1 **Table 4.** Relative contribution of anthropogenic aerosols, dust, fine mode natural and marine
2 aerosols to the total AOD₅₅₀ (bold) and the corresponding τ_a , τ_d , τ_n , τ_m levels with their $\pm 1\sigma$
3 values (in parentheses) over Eastern Mediterranean (EMT), over the land covered part (EML),
4 over the oceanic part and over the 9 sub-regions of Eastern Mediterranean appearing in Fig. 1
5 based on the MODIS Terra and Aqua (Italics) observations. The sum of the aerosol type
6 AODs per region does not necessarily correspond to the total AOD₅₅₀ values appearing in
7 Table 3 as these results were for the total of the days with aerosol retrievals even for days
8 when our aerosol type separation algorithm was not applicable.
9

Region	Satellite	Contribution to MODIS TERRA/AQUA AOD ₅₅₀			
		Anthropogenic	Dust	Fine mode natural	Marine
EML	TERRA	52 % (0.112±0.087)	32 % (0.074±0.080)	16 % (0.034±0.026)	-
	AQUA	50 % (0.117±0.093)	35 % (0.090±0.102)	15 % (0.035±0.028)	-
EMO	TERRA	41 % (0.086±0.085)	34 % (0.076±0.185)	-	25 % (0.054±0.018)
	AQUA	40 % (0.079±0.080)	33 % (0.070±0.181)	-	27 % (0.054±0.018)
NBL	TERRA	59 % (0.108±0.101)	23 % (0.042±0.046)	18 % (0.032±0.030)	-
	AQUA	59 % (0.110±0.100)	24 % (0.045±0.047)	17 % (0.033±0.030)	-
SBL	TERRA	55 % (0.109±0.088)	28 % (0.056±0.058)	17 % (0.033±0.026)	-
	AQUA	55 % (0.113±0.088)	29 % (0.060±0.060)	16 % (0.034±0.026)	-
ANL	TERRA	51 % (0.113±0.075)	34 % (0.076±0.068)	15 % (0.034±0.023)	-
	AQUA	50 % (0.114±0.075)	35 % (0.079±0.070)	15 % (0.034±0.023)	-
NAL	TERRA	50 % (0.113±0.083)	35 % (0.083±0.085)	15 % (0.034±0.025)	-
	AQUA	48 % (0.118±0.091)	38 % (0.099±0.108)	14 % (0.035±0.027)	-
BSO	TERRA	53 % (0.108±0.103)	22 % (0.044±0.101)	-	25 % (0.051±0.016)
	AQUA	51 % (0.094±0.087)	22 % (0.042±0.085)	-	27 % (0.051±0.016)
NWO	TERRA	41 % (0.087±0.090)	33 % (0.071±0.142)	-	26 % (0.055±0.020)
	AQUA	40 % (0.079±0.083)	32 % (0.066±0.127)	-	28 % (0.055±0.020)
SWO	TERRA	32 % (0.071±0.070)	42 % (0.097±0.257)	-	26 % (0.058±0.018)
	AQUA	32 % (0.093±0.288)	41 % (0.072±0.080)	-	27 % (0.059±0.018)
NEO	TERRA	48 % (0.098±0.094)	28 % (0.061±0.144)	-	24 % (0.050±0.016)
	AQUA	46 % (0.086±0.082)	28 % (0.057±0.115)	-	26 % (0.050±0.016)
SEO	TERRA	36 % (0.079±0.070)	39 % (0.087±0.224)	-	25 % (0.055±0.016)
	AQUA	36 % (0.075±0.071)	38 % (0.080±0.217)	-	26 % (0.055±0.016)

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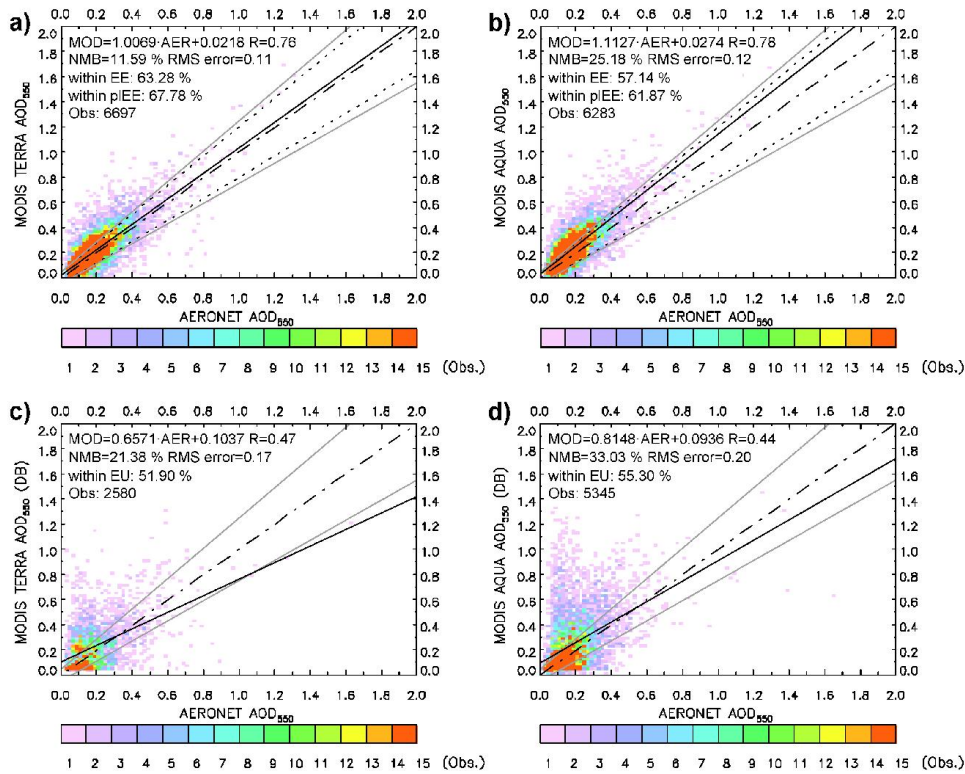


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 2 **Figure 1.** Eastern Mediterranean map with the 9 sub-regions selected for the generalization of
 3 our results and the location of the AERONET stations used for the validation of MODIS
 4 satellite data. The 9 sub-regions are: NBL (Northern Balkans Land), SBL (Southern Balkans
 5 Land), ANL (Anatolia Land), NAL (Northern Africa Land), BSO (Black Sea Oceanic), NWO
 6 (North-Western Oceanic), SWO (South-Western Oceanic), NEO (North-Eastern Oceanic) and
 7 SEO (South-Eastern Oceanic). The full names and the geolocation of the 13 AERONET
 8 stations appearing in the map are available in Table 1.
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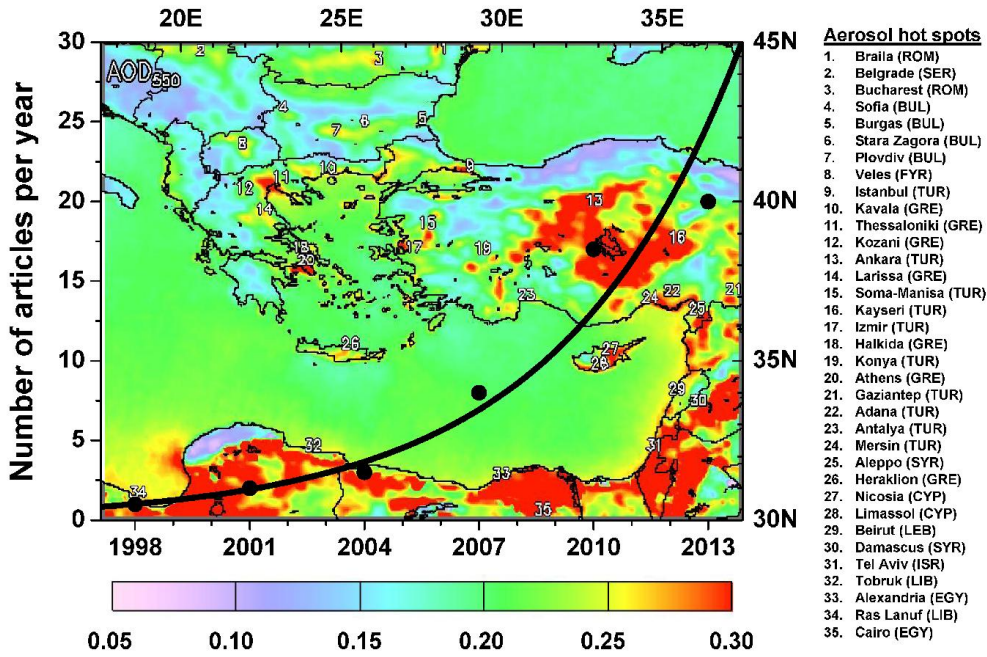
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Figure 2. Flowchart with the methodology followed for the calculation of the anthropogenic aerosol, dust and marine aerosol optical depths (τ_a , τ_d and τ_m) over the sea (blue color) and the anthropogenic aerosol, dust and fine mode natural aerosol optical depths (τ_a , τ_d and τ_n) over land (brown color).

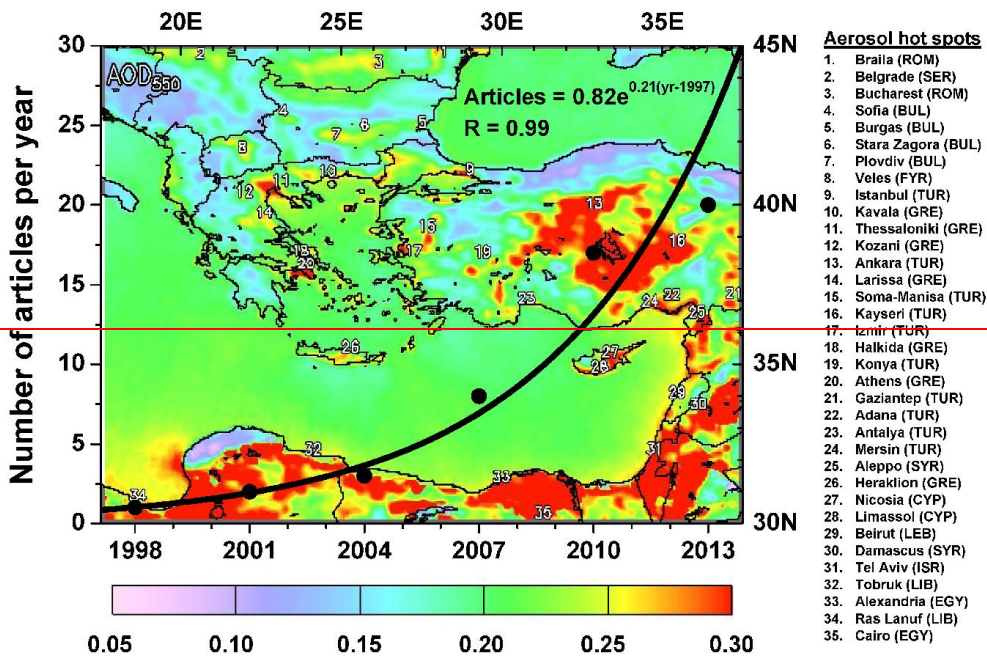


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3 **Figure 3.** Comparison of spatially (using a 25 x 25 km² window around each station) and
4 temporally (± 30 min from the MODIS overpass time) collocated MODIS Collection 051
5 level-2 and AERONET sunphotometric (quadratically interpolated) AOD₅₅₀ observations for
6 the Eastern Mediterranean stations: (a) for MODIS Terra DT data, (b) for MODIS Aqua DT
7 data, (c) for MODIS Terra DB data and (d) for MODIS Aqua DB data. The color scale
8 corresponds to the number of MODIS-AERONET collocation points that fall within 0.02 x
9 0.02 grid boxes. The solid line is the regression line of the MODIS-AERONET observations,
10 the dashed-dotted line is the 1:1 line, the dotted lines represent the expected error (EE)
11 envelope and the grey lines the pre-launch expected error (pLEE) envelope (Expected
12 Uncertainty - EU envelope for DB data). The slope and the intercept of the regression line, the
13 correlation coefficient R, the normalized mean bias (NMB), the root mean square (RMS)
14 error, the percentage of the collocation points that fall within the EE and pLEE and the number
15 of all the collocation points.

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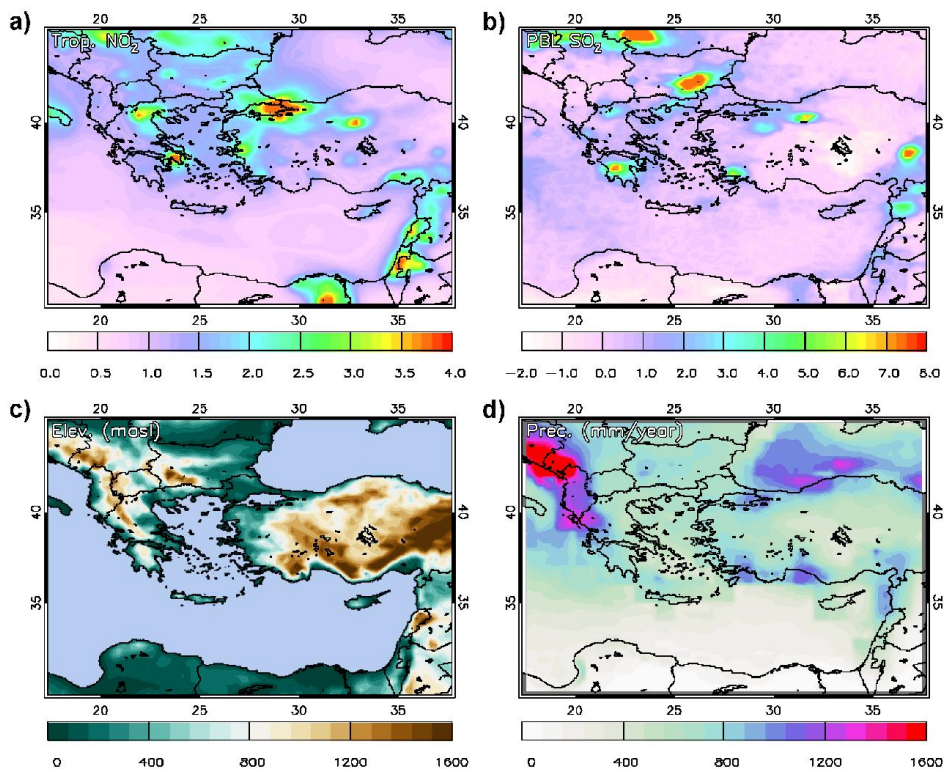
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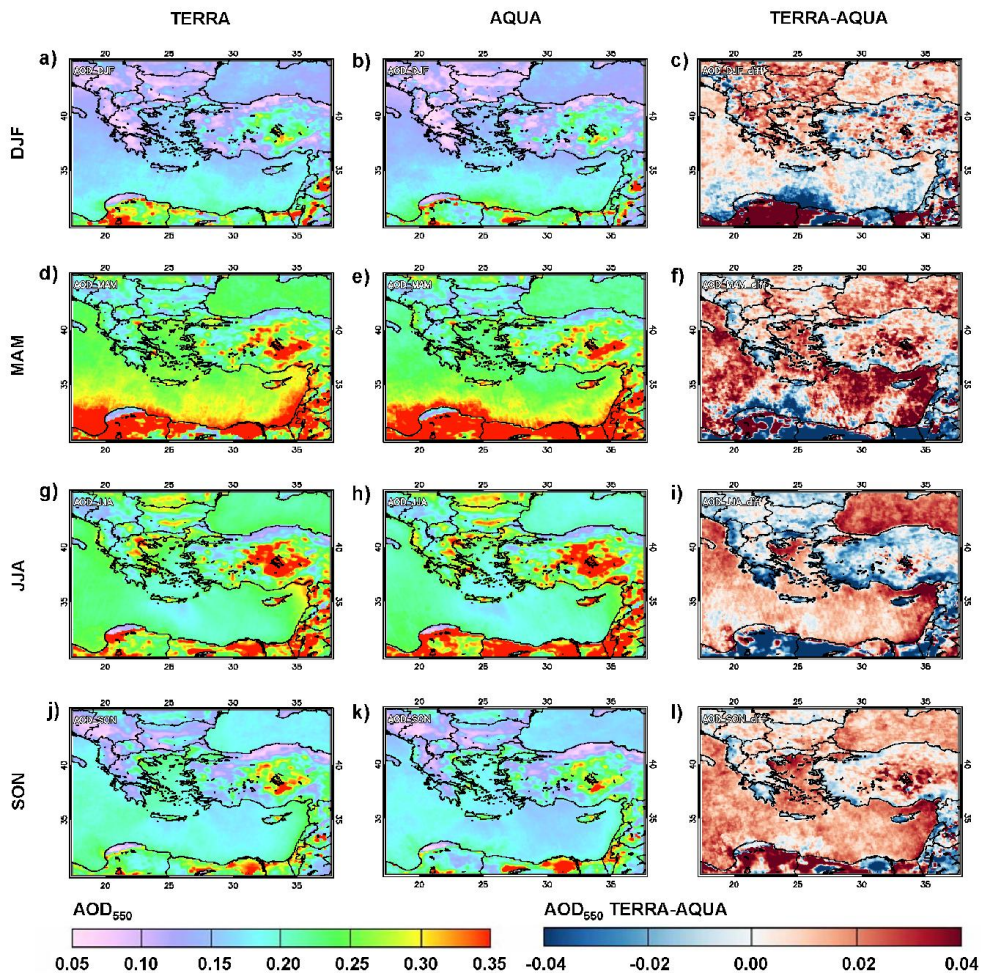
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1 **Figure 4.** AOD₅₅₀ patterns over Eastern Mediterranean as seen by MODIS Terra during the
 2 period 3/2000-12/2012 (3/2000-12/2007 for regions of North Africa covered by DB data
 3 only). The colorscale corresponds to the AOD₅₅₀ levels while the top x-axis and the right y-
 4 axis correspond to the longitude (°E) and latitude (°N), respectively. The position of 35
 5 aerosol hot spots is marked on the map (numbers from 1 to 35) while the names of the places
 6 and the countries where the hot spots are located appear on the right of the map. In the same
 7 figure the exponential growth of the number of satellite-based articles focusing on aerosols
 8 over the greater Eastern Mediterranean from 1997 to 2014 is shown (black line). The black
 9 dots represent the number of articles published within three year intervals. The bottom x-axis
 10 and the left y-axis correspond to the years and the number of published articles, respectively.
 11 The exponential growth corresponds to a near doubling of the publication rate every 3 years.

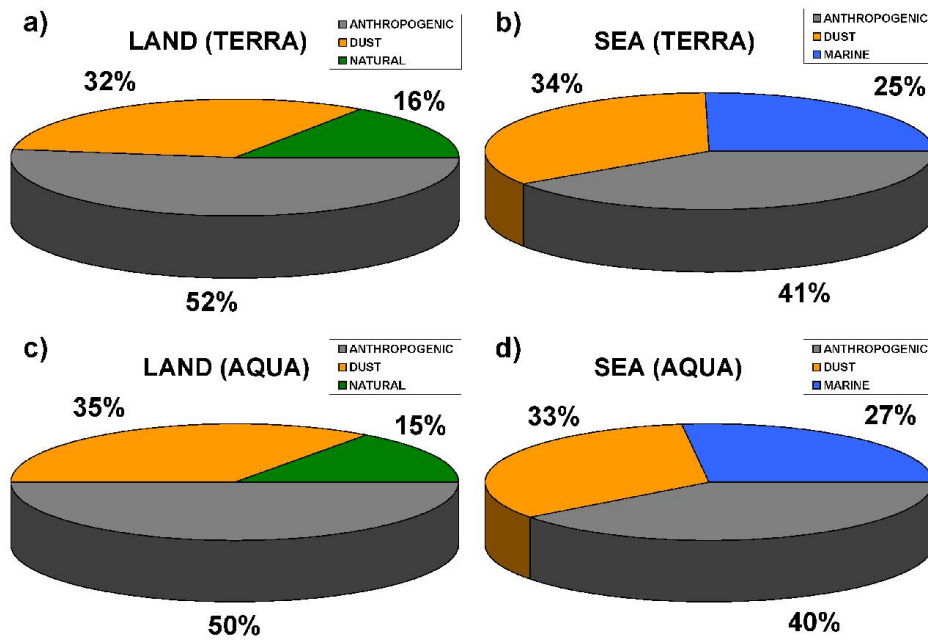


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 14 **Figure 5.** (a) Tropospheric NO₂ levels and (b) Planetary boundary layer SO₂ levels (in 10¹⁵
 15 molecules/cm²) over the Eastern Mediterranean as seen from OMI/AURA (2005-2012), (c)
 16 Topography (GTOPO elevation data in meters above sea level) and (d) Annual precipitation

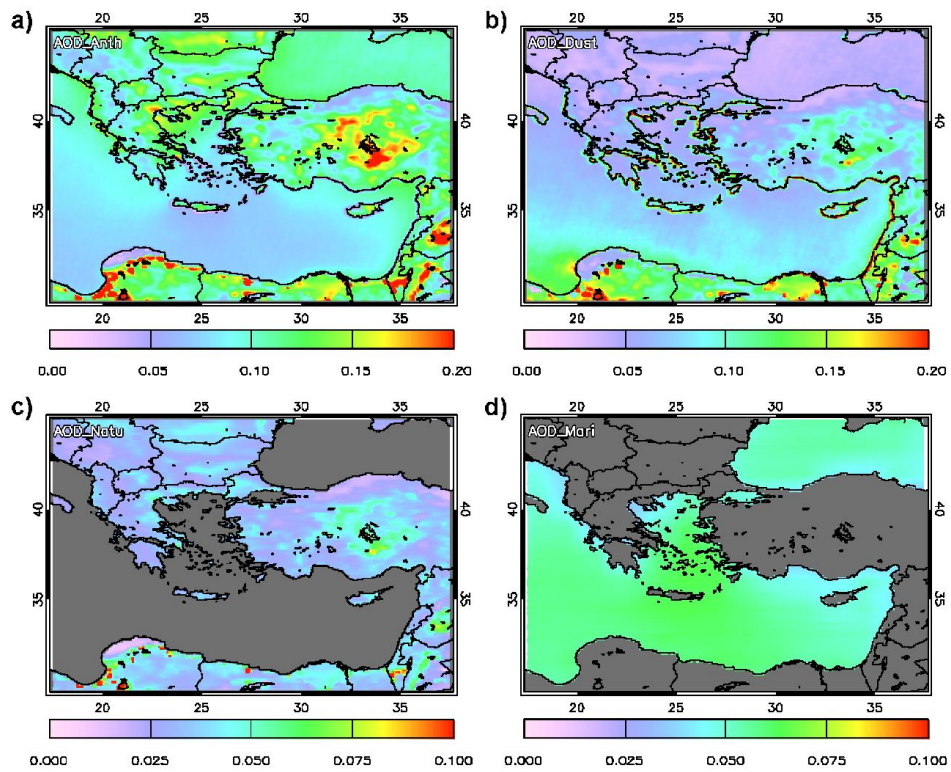
1 levels (in mm/year) from 3B43 TRMM and Other Sources Monthly Rainfall Product (2000-
 2 2012).



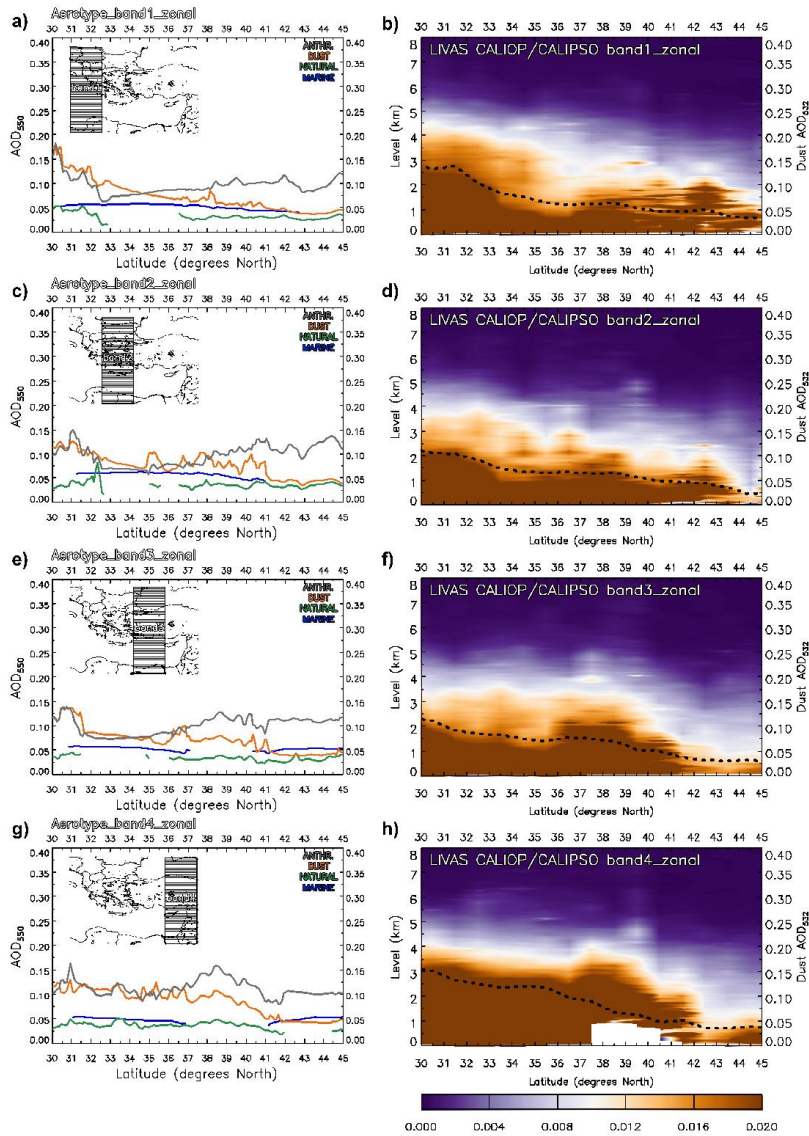
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 5 **Figure 6.** Seasonal AOD₅₅₀ patterns over the Eastern Mediterranean as seen by MODIS Terra
 6 (left column) during the period 3/2000-12/2012 (3/2000-12/2007 for regions of North Africa
 7 covered by DB data only) and MODIS Aqua (middle column) during the period 7/2002-
 8 12/2012. The differences between MODIS Terra and Aqua AOD₅₅₀ on a seasonal basis appear
 9 on the right column.



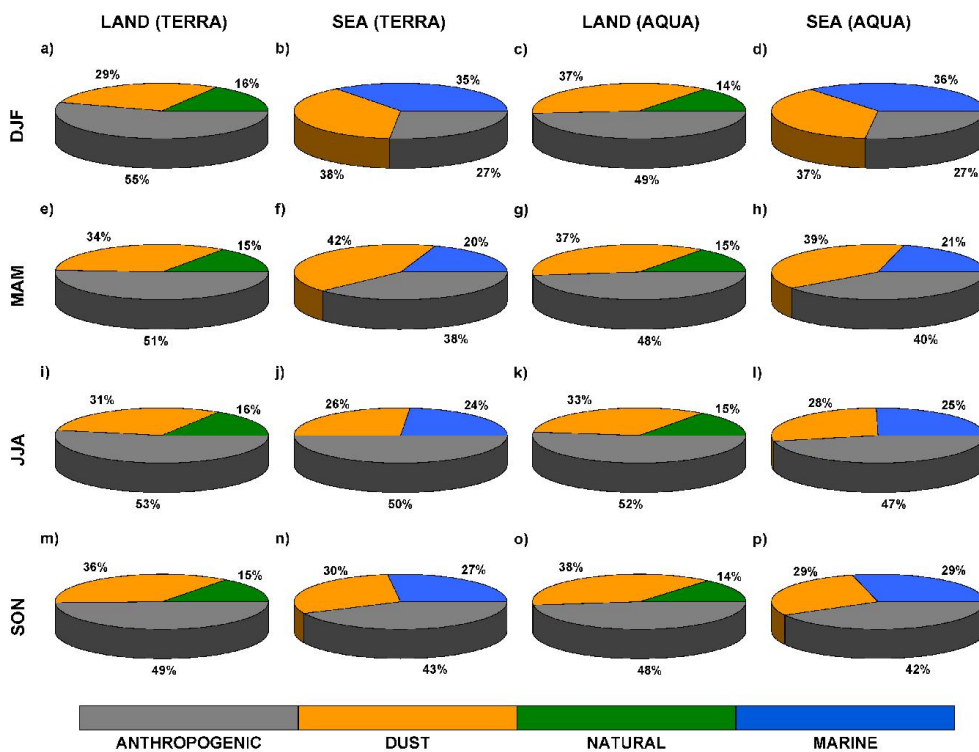
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3 **Figure 7.** Relative contribution of anthropogenic aerosols, dust and fine mode natural
4 aerosols to the total AOD₅₅₀ over the land covered part of Eastern Mediterranean based on
5 MODIS Terra (a) and MODIS Aqua (c) observations and relative contribution of
6 anthropogenic aerosols, dust and marine aerosols to the total AOD₅₅₀ over the oceanic part of
7 Eastern Mediterranean based on MODIS Terra (b) and MODIS Aqua (d) observations.



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3 **Figure 8.** (a) Anthropogenic aerosol (τ_a), (b) dust (τ_d), (c) fine mode natural aerosol (τ_n) and
4 (d) marine aerosol (τ_m) patterns over the Eastern Mediterranean based on MODIS Terra
5 observations during the period 3/2000-12/2012 (3/2000-12/2007 for regions of North Africa
6 covered by DB data only).

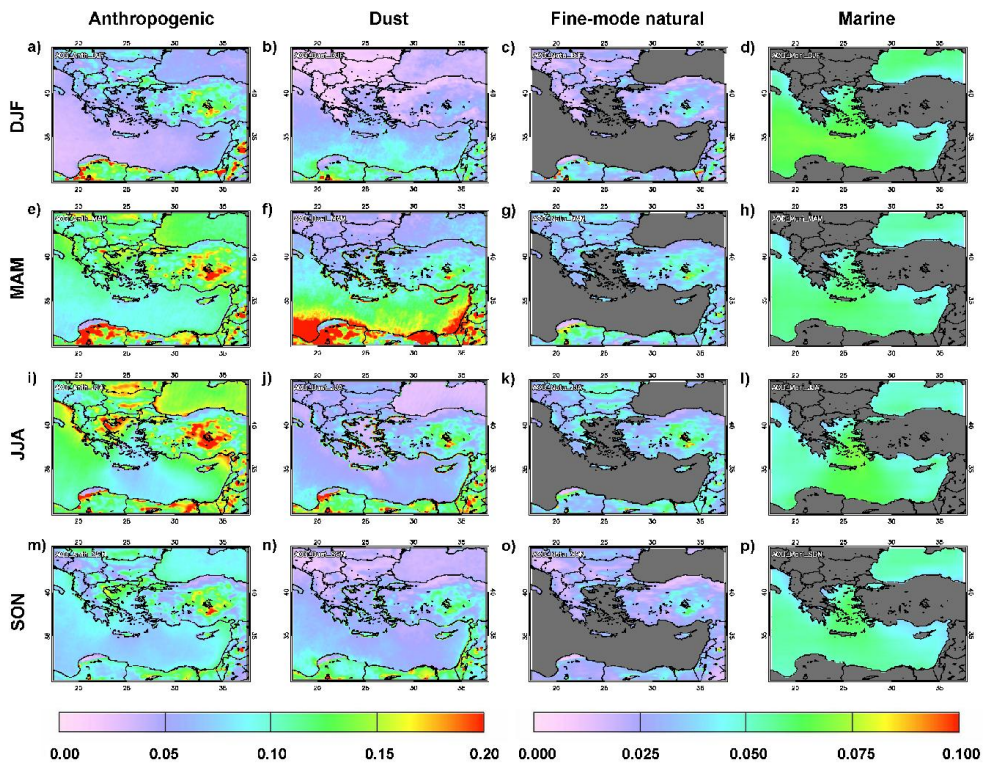


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3 **Figure 9.** Left column: Latitudinal variability of anthropogenic aerosols (τ_a), dust (τ_d), fine
4 mode natural aerosols (τ_n) and marine aerosols (τ_m) for four 5-degree longitudinal bands (see
5 embedded maps) covering Eastern Mediterranean based on MODIS Terra observations. Right
6 column: Latitudinal variability of dust extinction coefficients at 532 nm in km⁻¹ (colorscale
7 corresponds to the extinction coefficients and left y-axis to the atmospheric levels) and dust
8 aerosol optical depth at 532 nm (dotted line corresponding to the right-y-axis) for the same
9 four bands from LIVAS CALIOP/CALIPSO observations.

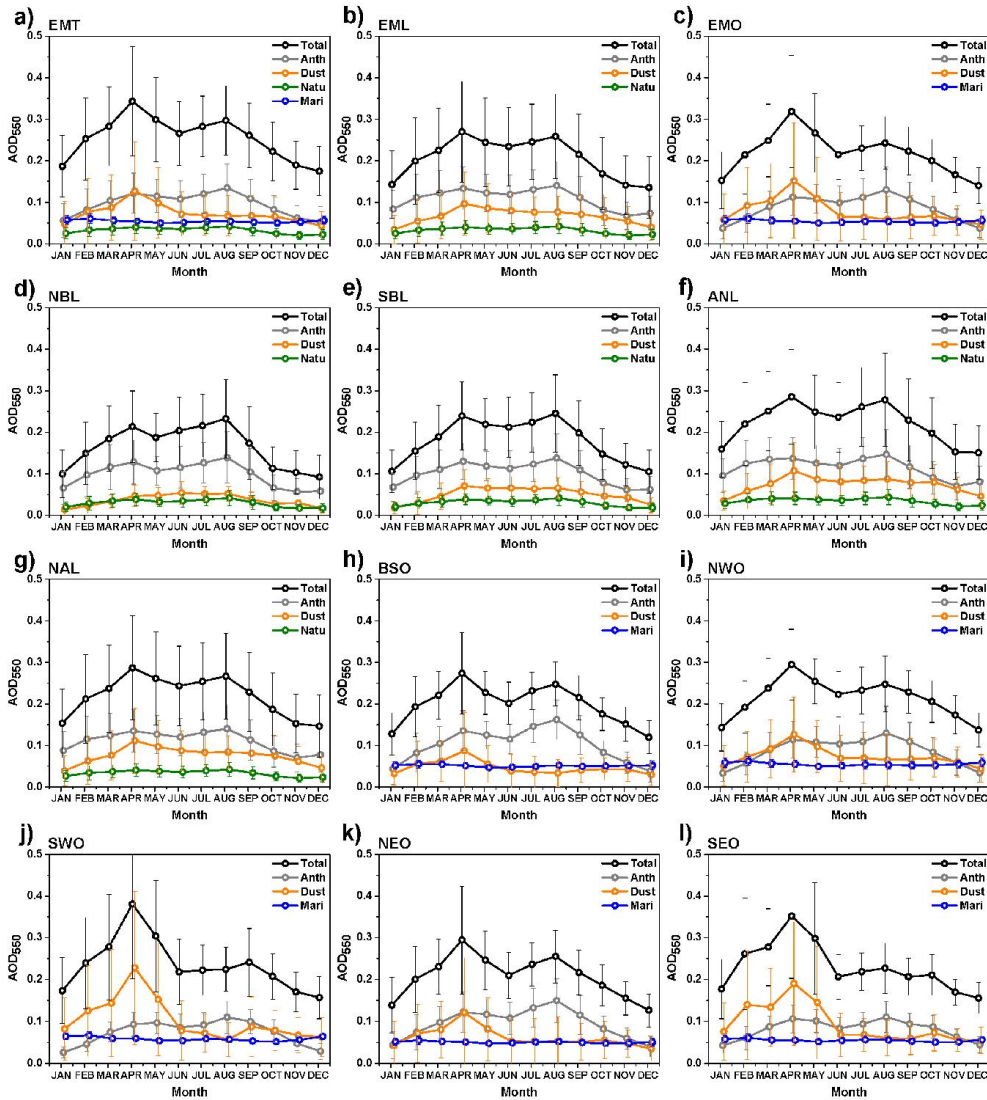


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3 **Figure 10.** Seasonal relative contribution of anthropogenic aerosols, dust and fine mode
 4 natural aerosols to the total AOD₅₅₀ over the land covered part of Eastern Mediterranean
 5 based on MODIS Terra (a, e, i, m) and MODIS Aqua (c, g, k, o) observations and seasonal
 6 relative contribution of anthropogenic aerosols, dust and marine aerosols to the total AOD₅₅₀
 7 over the oceanic part of Eastern Mediterranean based on MODIS Terra (b, f, j, n) and MODIS
 8 Aqua (d, h, l, p) observations.



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3 **Figure 11.** Seasonal (a, e, i, m) anthropogenic aerosol (τ_a), (b, f, j, n) dust (τ_d), (c, g, k, o) fine
4 mode natural aerosol (τ_n) and (d, h, i, p) marine aerosol (τ_m) patterns over the Eastern
5 Mediterranean based on MODIS Terra observations during the period 3/2000-12/2012
6 (3/2000-12/2007 for regions of North Africa covered by DB data only).



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3 **Figure 12.** Seasonal variability of anthropogenic aerosols (τ_a), dust (τ_d), fine mode natural
4 aerosols (τ_n) and marine aerosols (τ_m) over Eastern Mediterranean (EMT), over the land
5 covered part (EML), over the oceanic part (EMO) and over the 9 sub-regions of the Eastern
6 Mediterranean appearing in Fig. 1 based on MODIS Terra observations. The error bars
7 represent the $\pm 1\sigma$ values calculated from monthly gridded data.