Dust-Air Pollution Dynamics over the Eastern Mediterranean
acp-2015-109
Reply to Anonymous Referee #1

by M. Abdelkader, S. Metzger, R. E. Mamouri,
M. Astitha, L. Barrie, Z. Levin, and J. Lelieveld

June 12, 2015

We thank the anonymous referee for the constructive comments – hopefully addressed satisfactorily with this reply. Modifications of the manuscript are summarized in Table 1. Please also note the modifications in Table 1-5 in our reply to referee #2.

1 AOD

Since the EMAC model distinguishes dust particles (mainly coarse mode) and anthropogenic particles (mainly fine mode), why not showing the AERONET comparisons in more detail? AERONET provides also coarse-mode and fine-mode AOD! Because the focus later on is on CUT-TEPAK comparisons, one could do these specific comparisons at least for the Cyprus AERONET station.

First of all, to clarify the complexity of the EMAC model used in this study and to answer the additional question raised by referee #2, we have modified the model description section to include the following paragraph in the revised MS (see our reply to referee #2):

Our model version distinguishes aerosol particles in 7 modes, 4 Soluble (nucleation, aitken, accumulation, coarse) and three INSoluble modes (aitken, accumulation, coarse) with the complexity of the aerosol thermodynamics as investigated in Metzger et al. (2006), by considering case F4 since ISORROPIA-II used here does not include organic salt compounds in the gas/aerosol partitioning and aerosol neutralization framework. Within EMAC, the dust particles are emitted online following Astitha et al. (2012) (e.g., governed by model dynamics, precipitation and soil moisture) in either the INSoluble accumulation and/or coarse mode and only upon aging and transport they can be transferred to the respective Soluble accumulation and/or coarse modes. The aging depends on the available condensable compounds calculated within the chemistry scheme (Sander et al., 2005). In addition, via coagulation and hygroscopic growth the size-distribution can change and small particles are transferred to larger sizes, i.e., for dust from accumulation to coarse, whereby hygroscopic growth of bulk dust and dust salt compounds is only allowed in the soluble modes. For the latter we explicitly account for the water uptake of various major mineral salt compounds, i.e., $CaSO_4$, $Ca(NO_3)_2$, $CaCl_2$, $MgSO_4$, $Mg(NO_3)_2$, $MgCl_2$, Na_2SO_4, $NaNO_3$, $NaCl$, K_2SO_4, KNO_3, KCl, whereby the mineral cations Mg^{2+}, Na^+ and K^+ are only considered as tracers for the online calculated sea salt emissions, while K^+ is additionally used for biomass burning emissions being emitted here only in the insoluble aitken mode. Thus, the dust particles can be present in our set-up in four modes, each *represented by various calcium compounds that chemically characterize the bulk dust emissions depending on the level of aging. Note that we have limited the dust neutralization reactions in this work to calcium to be able to separate the dust associated water uptake and associated aging from sea salt effects. Since our set-up is flexible, the level of aerosol neutralization complexity can/will be changed for other application tasks. For the current modeling study though, this set-up represents the dust air-pollution dynamics over the Eastern Mediterranean well.*

Lumping together a mass and number based size-distribution in one fine and coarse mode can be done, but a comparison with observations of optically derived size-distributions needs to be interpreted with great care. There are many uncertainties on both sides. Therefore we had omitted such a comparison in the manuscript. Nevertheless, to satisfy the referees request, we show the fine and coarse mode AOD for the AERONET observations and the EMAC model results for the CUT-TEPAK station, Cyprus. Figure 1 reveals that the fine mode AOD is dominating the total AOD during September 2011. The model compares well with the AERONET observations, especially during the dust outflow-2 on 28^{th}. AERONET shows an AOD of 0.4 for the coarse mode contribution to the total AOD of 0.6, which is underestimated by EMAC as a result of the steep gradient of the dust load. The dust outflow-2 was predicted to be slightly more east (see discussion of the LIDAR results below).

I know that regional dust transport models have their difficulties with correct dust uptake (emissions) by the atmosphere, and I would like to know what the result is here (EMAC), when using a global model with even coarser resolution than these regional models have.

To satisfy this request, we include a quantitative model comparison of our EMAC results with DREAM (`http://www.bsc.es/earth-sciences/mineral-dust/catalogo-datos-dust`) in this reply. Figure 2 shows the dust load and outflow for the 28^{th} September at noon. Interestingly, both models predict a dust outflow and a dust front which just has touched on Cyprus. These dust-fronts are associated with a very steep gradient of the dust concentrations, which strongly increases a few longitudes to the East. But, compared to the LIDAR observations, both models may not have captured fully the outflow dynamics along the south side of the Troodos mountains, which are up to 2000 meters high and a natural barrier for the dust layer, which peaks around 1500 meters (see discussion of the LIDAR results below). Of course, matching one station on a small island at a given time is an issue that is almost impossible to be accurately resolved by any global model, especially by a climate model (despite sophisticated nudging technique and the relatively high resolution). For this task, higher resolution regional models must be used.

The advantage of regional models clearly is the higher resolution, which can resolve the orography with grid boxes down to a few kilometers, although for atmospheric chemistry applications a resolution of 25-50 km is typical. This advantage cannot be met with a global chemistry-climate model such as EMAC. Even not closely with our high resolution set-up, i.e., T255, which is with a grid box size of $\approx 50\ km$ not only computationally very demanding, but also beyond the state-of-the-art that has been applied for this topic so far. But on the other hand, the advantage of our EMAC version is that dust emissions are calculated online, i.e., fully coupled with meteorology and radiation (being nudged towards observation), without any limitation and assumptions on the moisture, dust and air pollution fluxes at the regional domain boundaries. The latter is also relevant here as the dust is transported over large distances. And this advantage can compensate for the disadvantage of using a lower resolution in case of dust-air pollution dynamics. Ideally, a consistent coupling of equally complex models that represent at least the details currently considered in our study would be required for localized forecasts of individual stations, especially if located around a coast-line of islands with relatively high mountains. Clearly, such applications or a detailed comparison goes far beyond the current work – it remains a challenging subject for follow-up studies.

2 Vertical profiling

Lidar provides the potential to distinguish dust and fine-mode particles, too. There is an EARLINET polarization lidar at Limassol, and I found in AMT a paper (Mamouri et al., 2014) dealing with the same time period as shown here (27-29 September 2011). Why do you not include these observations in this paper in the frame of a thorough comparison? I have long experience with lidar/model comparisons in the case of dust and know that these comparisons usually show large discrepancies between lidar and modeled dust profiles. I assume (speculate) that this is similar here, and I further speculate that this is the reason for ignoring the lidar observations. Nevertheless, progress in science arises from discrepancies! So, please show these comparisons, at least for the published strong dust outbreak (28-29th September).

Although a comparison is not straight forward, mainly because of the assumptions on the treatment of extinction of dust particles at both sides, we have prepared such comparison which will be included in the revised manuscript.

The following section will be included in the revised manuscript:
(with the attached Figure 3 numbered as Figure 10 in the revised MS)

Section 3.3.2 – EMAC versus ground based LIDAR profiles

*The vertical structure of the dust outflow-2 is compared to ground based LIght Detection And Ranging (LIDAR) measurements of the Cyprus University of Technology (CUT) in Limassol, Cyprus, which also hosts the AERONET station. The LIDAR observations have been recently used to study dust outflows over the EM, including the dust outflow-2 considered in our study (*Mamouri et al., 2013; Nisantzi et al., 2014; Mamouri and Ansmann, 2014, 2015*). For a consistent comparison, the LIDAR observations are averaged within the model vertical grid box.*

Figure 10 shows the simulated and observed total and dust only extinction at CUT-TEPAK. The model results are shown at three different longitudes: 33 °E at CUT-TEPAK (EMAC), 34°E (EMAC-1) and 35°E (EMAC-2), with all longitudes referring to the latitude of the CUT-TEPAK station (34.675°N). The comparison shows that EMAC captures the LIDAR signal, but 2° (about 200 km) more to the east. This underestimation decreases with each profile further east, indicating a steep gradient of the model dust layer concentration that is associated with the front of the dust-outflow-2 (shown in Fig. 6 and 7). Although the magnitude of the model extinction is predicted lower at the CUT-TEPAK station at both days, EMAC captures the observed peak at 1.5 km height (2nd day) with a 1.8 km dust layer height well, given the relatively coarse vertical grid resolution of the model (which is 500 m at that height).

Interestingly, the calculated vertical extent of the dust layer is wider than the LIDAR signal, which indicates that the total aerosol layer is thicker, at least a few hundred kilometers eastward. This might be related to flow disturbance by the orography, and/or a result of the contribution of other compounds that are considered in our model simulation. The vertically integrated dust extinction is similar to the total extinction profiles for both EAMC and the observations, but the predicted concentration maximum of the dust layer is closer to the observations for the second day. For the first day, EMAC does not capture the observed dust signal, but the total AOD (integral of the area under the profile) is comparable to the AERONET AOD shown in Fig. 4 for both days.

Let me start with Figure 6: The satellite images indicate a lot of dust over Cyprus on 28th September as well as on 29 September. Figure 7: Why did you calculate trajectories for Limassol (22 Sep) and then for the northeastern peak of Cyprus (28th Sep, evening)? Why not again Limassol? This is strange and seems to be arbitrarily selected. In the evening of 28th Sep there was probably dust everywhere over Cyprus according to the images in Figure 6. So, please show the trajectories for Limassol, only!

The objective is to study the dynamics of the dust air-pollution interaction during two dust events. The MODIS satellite image (Figure 6 p. 7526 in the MS) shows dust outflow over the EM, including Cyprus. The model results (Fig. 9 p. 7529 in the MS) resolve the dust outflow over the EM, however, with a strong dust gradient over the eastern part of Cyprus on 28th, September, which is also predicted by the DREAM model (see Fig. 2). On the other hand, the HYSPLIT back trajectories (Fig. 7 pg 7527 of the MS), are based on the NCEP reanalysis data which has a resolution of 270 km. Such coarse resolution seems in this case insufficient as is does not capture the strong gradient associated with the atmospheric dynamics over Cyprus. As a result, the calculated HYSPLIT back trajectories at Limassol (CUT-TEPAK) indicated an air mass that has reached CUT-TEPAK originating from Turkey, instead from the Arabian Desert as indicated by the MODIS image (Figure 6 p. 7526) and our EMAC model results (shown in Fig. 9 p. 7529) and the results of the DREAM model (see Fig. 2). For this reason, we have calculated the back trajectories at the eastern part of Cyprus, where our model dust load is also more consistent with the MODIS image. In order to be consistent with the back trajectories, back trajectories could be calculated using the meteorological field driven by regional and high resolution model for better representation of the topography with an advanced back trajectory model such as FLEXPART. But this is beyond this study.

Figures 8, 9 show very nice model results covering the two outbreak situations. Height- longitude plots of dust distribution are shown. You emphasize the CUT-TEPAK station, so these dust cross sections are for 34.5 North? Right? Please state that clearly! What does "zonal dust" then mean in this context? A dust value at 700 hPa, for 30E and 34.5N describes just one value for the given location, not for a zonal belt?

We apologize for the typo. The figure shows the meridional mean of the dust at CUT-TEPAK location (34.675^{o}N). The figure caption is changed to:

Time series of (top) dust load and (bottom) meridional mean for the first event at CUT-TEPAK location (34.675° N). The green shaded area represents the orography.

Now, a comparison between the Limassol lidar versus EMAC dust profile (dust extinction, overall aerosol extinction profiles) is required. Maybe the best time for comparison is the morning of 29 September (according to the images in Figure 6). I realize from Figure 9 that there are strong horizontal dust inhomogeneities (in the model results), especially close to Limassol, so the lidar comparison may reflect that by strong deviations between observation and modeling. This must be discussed. But at least the geometrical structure (base and top heights of the dust layer) should match, I speculate.

We have addressed this point by the new Section 3.3.2 which will show a comparison of our EMAC results with the suggested LIDAR observations at CUT-TEPAK. The results of this comparison (and the new Section 3.3.2) have been presented above.

Finally, Figure 10: I appreciate very much that CALIPSO data are included in the comparison. This is not just an easy task! CALIPSO provides backscatter coefficients. How did you get the extinction values? What lidar ratios were applied? Probably 40sr for pure dust and 60sr for polluted dust, and 20sr for marine!

As shown in the manuscript (p. 7507 line 10) CALIPSO Level 2 version 3.01, 5 km aerosol profile (APro-Prov) product is used. The lidar ratios used to calculate the extinction of pure dust, polluted dust and marine aerosols are: 40 sr, 65 sr and 20 sr, respectively (Winker et al., 2009). Fig. 4 (below) shows the ratios and size distribution as used in CALIPSO aerosol model.

CALIPSO uses the linear iterative method to derive profiles of volume backscatter and extinction coefficients from both clouds and aerosols. In the CALIPSO extinction analysis, features are analyzed or solved (their particulate backscatter and extinction profiles retrieved) using lidar ratios appropriate to the type of cloud or aerosol layer of which they are composed. As a scene, or any profile for that matter, can contain features of different types. Different lidar ratios are used in different regions of the scene or profile. The lidar ratio is considered to be constant over certain intervals within each backscatter profile, as determined by the layer detection and scene classification algorithms. A unique feature of the CALIPSO lidar analysis is that it attempts to average signals in atmospheric regions where the optical properties are uniform and the signal strengths are comparable (Vaughan et al., 2009). Perhaps the single most difficult task among the scene classification algorithms is determining the appropriate lidar ratio to be used in the optical analyses of aerosol layers as a results of the very high ground speed of the satellite (7 km s^{-1}) compared to the stationary ground based LIDAR. Field measurements of aerosol lidar ratios in the surface-attached aerosol layers (SAL) vary from 15 sr to in excess of 120 sr (Vaughan et al., 2004). Lidar-only measurements of lidar ratios require a Raman lidar or a high spectral resolution lidar (HSRL). CALIPSO therefore determines a value for Sa using a model-matching scheme: the optical, geophysical (e.g., latitude, longitude), and temporal (season) characteristics of the 532 layer are used as decision points to navigate through a flow chart that ultimately selects a most likely aerosol model for each aerosol layer.

And what I know from the Kanitz paper (AMT 2014?) the jumps (in color) in the extinction values are artefacts and due to the jumps in the extinction values when you go from laser foot prints over the Mediterranean Sea (lidar ratio 20) to land surfaces (lidar ratio of 40/60) and back to sea surfaces (20) again. These strong aretfacts should be mentioned.

The comparison between CALIPSO and the EMAC model (upper panel of Fig. 10 p. 7530 in the MS) includes only the CALIPSO pure dust classification. Artifacts of marine aerosol layers that extend over land may indeed produce a mixed signal potentially affecting the CALIPSO classification over land (Fig. 10, lower panel). For our comparison this is less relevant, since we focus on a distinct dust layer over the Mediterranean sea. And the CALIPSO classification is most safe for dust cases. An even more detailed combination of model results, ground-based and satellite observations would be desired to better distinguish between pure dust and polluted dust events. But such consistent data is not available. Nevertheless, our model results can give additional insight in the effects of mixing of dust and air-pollution, and may be used to challenge results such of Kanitz et al. (2014). Again, this is beyond the scope of this study. In this respect, we see our modeling study as one contribution to an overall very challenging effort – both from an observational point of view as well as from our modeling point. We look forward to a closer collaboration which will help improving our understanding of this very important subject.

The following text is added to the revised MS:

In addition, the CALIPSO aerosol type classification could overestimate the extinction and the optical depth for marine aerosols. This may add artifacts to the mixed dust classification over land (Kanitz et al., 2014).

Figure 10: I cannot see (or distinguish) brown and black isolines. . .?

We apologize that brown iso-lines are not distinguishable. Unfortunately, this is a result of the graphic conversion during the typesetting procedure, which converted vector graphics to raster images to reduce the file size. But we will keep the quality of the vector images in the final manuscript, so that the details are also visible without zooming into the image.

References

Astitha, M., Lelieveld, J., Abdel Kader, M., Pozzer, A., and de Meij, A.: Parameterization of dust emissions in the global atmospheric chemistry-climate model EMAC: impact of nudging and soil properties, Atmospheric Chemistry and Physics, 12, 11 057–11 083, doi:10.5194/acp-12-11057-2012, URL `http://www.atmos-chem-phys.net/12/11057/2012/`, 2012.

Kanitz, T., Ansmann, A., Foth, A., Seifert, P., Wandinger, U., Engelmann, R., Baars, H., Althausen, D., Casiccia, C., and Zamorano, F.: Surface matters: limitations of CALIPSO V3 aerosol typing in coastal regions, Atmospheric Measurement Techniques, 7, 2061–2072, doi:10.5194/amt-7-2061-2014, URL `http://www.atmos-meas-tech.net/7/2061/2014/`, 2014.

Mamouri, R. E. and Ansmann, A.: Fine and coarse dust separation with polarization lidar, Atmospheric Measurement Techniques, 7, 3717–3735, doi:10.5194/amt-7-3717-2014, URL `http://www.atmos-meas-tech.net/7/3717/2014/`, 2014.

Mamouri, R. E. and Ansmann, A.: Estimated desert-dust ice nuclei profiles from polarization lidar: methodology and case studies, Atmospheric Chemistry and Physics, 15, 3463–3477, doi:10.5194/acp-15-3463-2015, URL `http://www.atmos-chem-phys.net/15/3463/2015/`, 2015.

Mamouri, R. E., Ansmann, A., Nisantzi, A., Kokkalis, P., Schwarz, A., and Hadjimitsis, D.: Low Arabian dust extinction-to-backscatter ratio, Geophysical Research Letters, 40, 4762–4766, doi:10.1002/grl.50898, URL `http://dx.doi.org/10.1002/grl.50898`, 2013.

Metzger, S., Mihalopoulos, N., and Lelieveld, J.: Importance of mineral cations and organics in gas-aerosol partitioning of reactive nitrogen compounds: case study based on MINOS results, Atmospheric Chemistry and Physics, 6, 2549–2567, doi:10.5194/acp-6-2549-2006, URL `http://www.atmos-chem-phys.net/6/2549/2006/`, 2006.

Nisantzi, A., Mamouri, R. E., Ansmann, A., and Hadjimitsis, D.: Injection of mineral dust into the free troposphere during fire events observed with polarization lidar at Limassol, Cyprus, Atmospheric Chemistry and Physics, 14, 12 155–12 165, doi:10.5194/acp-14-12155-2014, URL `http://www.atmos-chem-phys.net/14/12155/2014/`, 2014.

Sander, R., Kerkweg, A., Jckel, P., and Lelieveld, J.: Technical note: The new comprehensive atmospheric chemistry module MECCA, Atmospheric Chemistry and Physics, 5, 445–450, doi:10.5194/acp-5-445-2005, URL `http://www.atmos-chem-phys.net/5/445/2005/`, 2005.

Vaughan, M. A., Young, S. A., Winker, D. M., Powell, K. A., Omar, A. H., Liu, Z., Hu, Y., and Hostetler, C. A.: Fully automated analysis of space-based lidar data: an overview of the CALIPSO retrieval algorithms and data products, pp. 16–30, doi:10.1117/12.572024, URL `http://proceedings.spiedigitallibrary.org/proceeding.aspx?articleid=851227`, 2004.

Vaughan, M. A., Powell, K. A., Winker, D. M., Hostetler, C. A., Kuehn, R. E., Hunt, W. H., Getzewich, B. J., Young, S. A., Liu, Z., and McGill, M. J.: Fully Automated Detection of Cloud and Aerosol Layers in the CALIPSO Lidar Measurements, Journal of Atmospheric and Oceanic Technology, 26, 2034–2050, doi:10.1175/2009JTECHA1228.1, URL `http://journals.ametsoc.org/doi/abs/10.1175/2009JTECHA1228.1`, 2009.

Winker, D. M., Vaughan, M. A., Omar, A., Hu, Y., Powell, K. A., Liu, Z., Hunt, W. H., and Young, S. A.: Overview of the CALIPSO Mission and CALIOP Data Processing Algorithms, Journal of Atmospheric and Oceanic Technology, 26, 2310–2323, doi:10.1175/2009JTECHA1281.1, URL `http://journals.ametsoc.org/doi/abs/10.1175/2009JTECHA1281.1`, 2009.

Table 1: Revised manuscript – Modifications for referee#1. Please also note the modifications in Table 1-5 in our reply to referee #2.

no.	P. no.	Line no.	Before	Correction
1	7493	Authors	M. Abdelkader, S. Metzger, M. Astitha, Z. Levin, and J. Lelieveld	M. Abdelkader, S. Metzger, R. E. Mamouri, M. Astitha, L. Barrie, Z. Levin, and J. Lelieveld
2	7507	8		New Section 3.3.2 (and adjusted labeling of the following sections)
3	7507	20	Added text before "Therefore, the CALIPSO data .....":	In addition, the CALIPSO aerosol type classification could overestimate the extinction and the optical depth for marine aerosols. This may add artifacts to the mixed dust classification over land (Kanitz et al., 2014).
4	7514	16	Added text to acknowledgment:	The authors thank the Remote sensing and Geo-Environment Research laboratory of Cyprus University of Technology (CUT) for providing the LIDAR extinction profiles at the CUT-TEPAK station.
5	7528-9	caption	Time series of (top) dust load and (bottom) vertical cross-sections at the CUT-TEPAK station for the first event, the green shaded area represents the orography.	Time series of (top) dust load and (bottom) meridional mean for the first event at CUT-TEPAK location (34.675°N). The green shaded area represents the orography.
6	–	–	Added Fig. 3 of this reply	In the MS, labeled Figure 10 (adjusted labeling of figures).
7	–	–	No Supplement.	Figure 1 will be included in a Supplement.

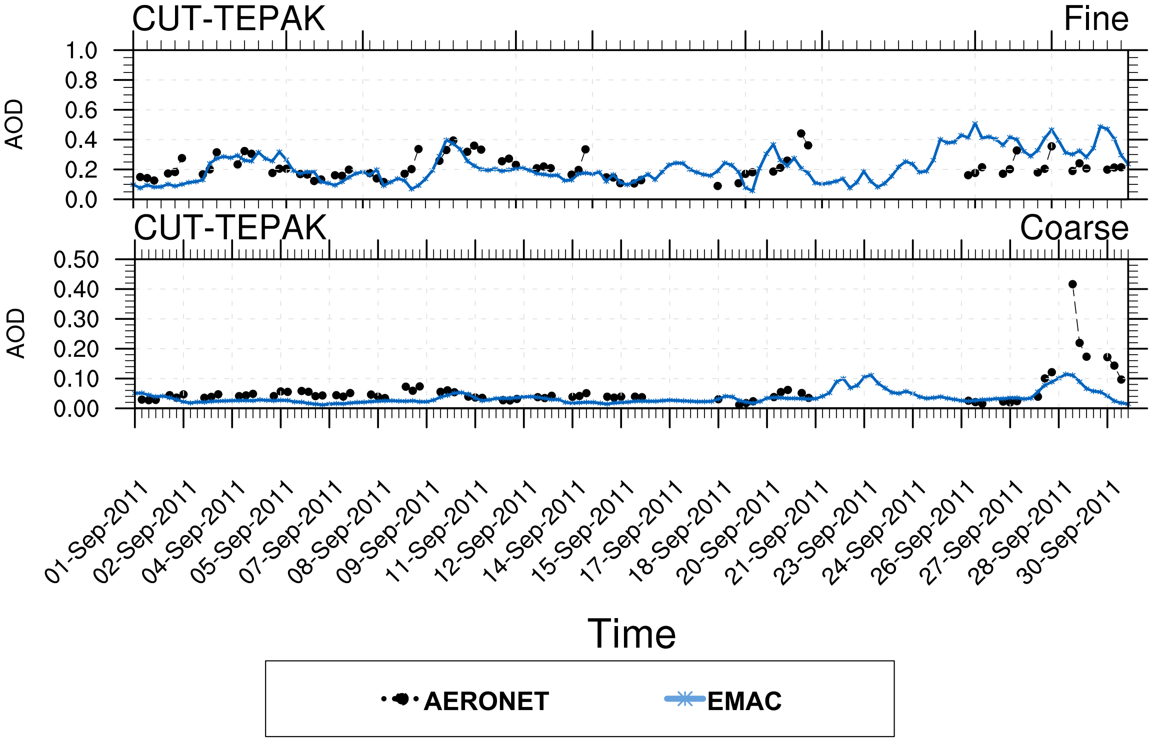

Figure 1: Aerosol fine mode (top) and coarse mode (bottom) AOD: EMAC model (blue lines) versus AERONET station observations (black lines). This figure is added to the supplement.

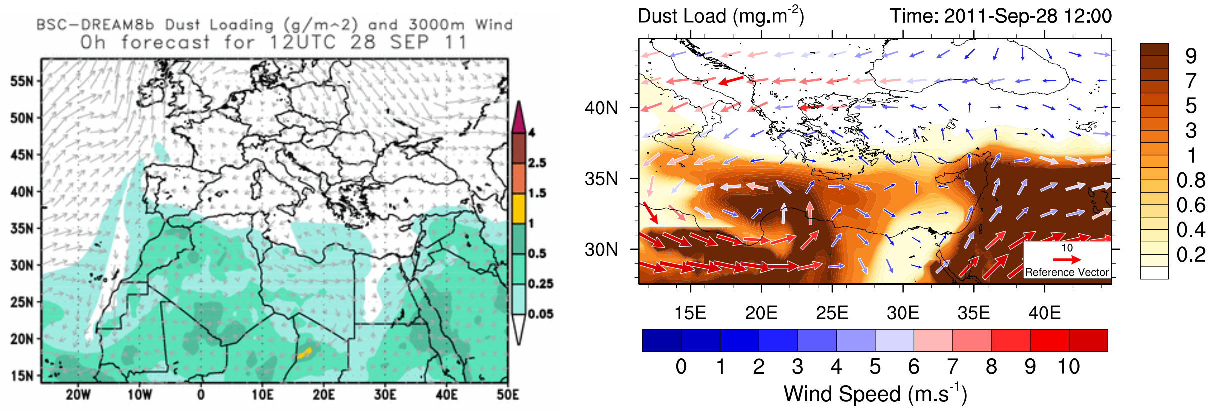

Figure 2: Dust load on 28th September 2011 for DREAM (left) and EMAC model (right).

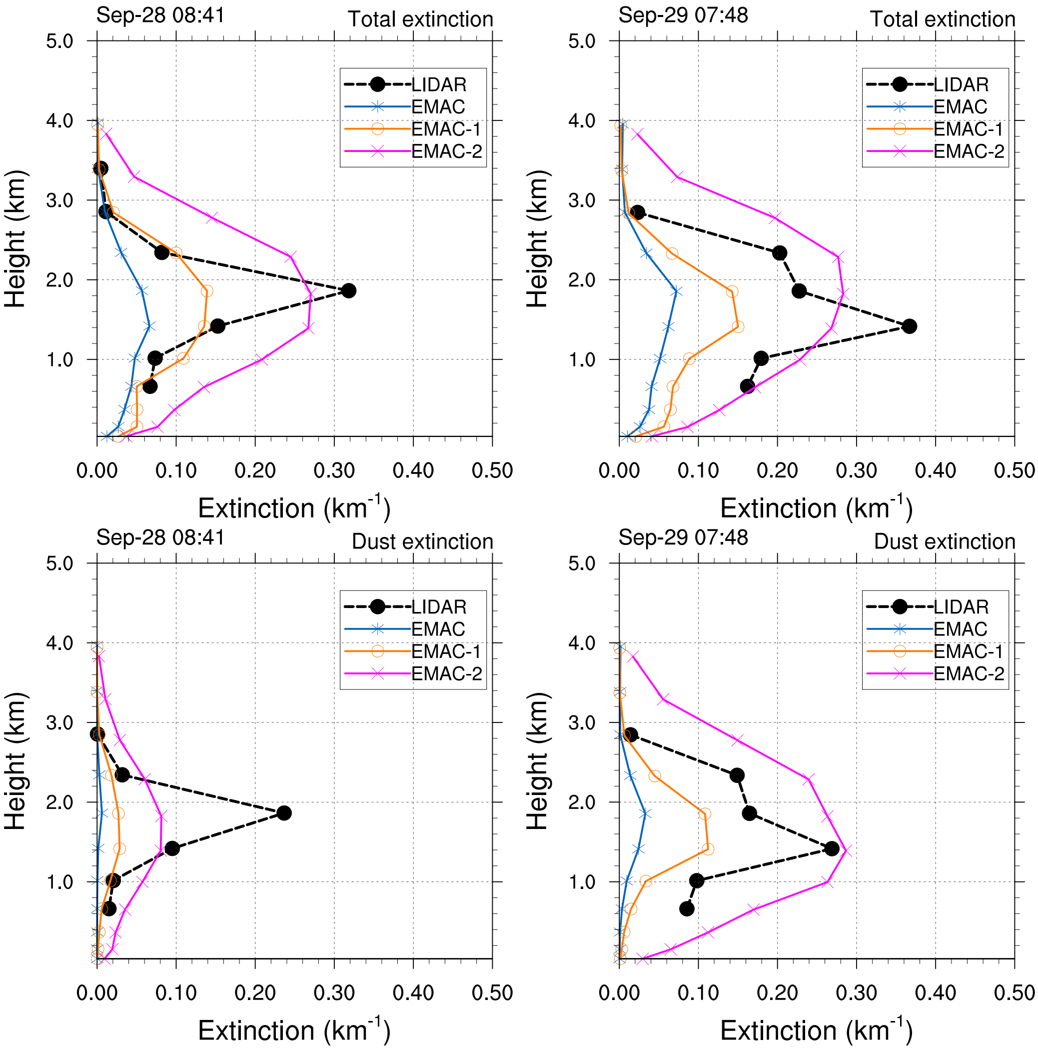

Figure 3: Comparison between the modeled and the observed total and dust only extinction at CUT-TEPAK station at different longitudes: 33 °E (EMAC) at CUT-TEPAK station, 34°E (EMAC-1) and 35°E (EMAC-2); all longitudes refer to the latitude of CUT-TEPAK (34.675°N). This figure is included in the revised MS.

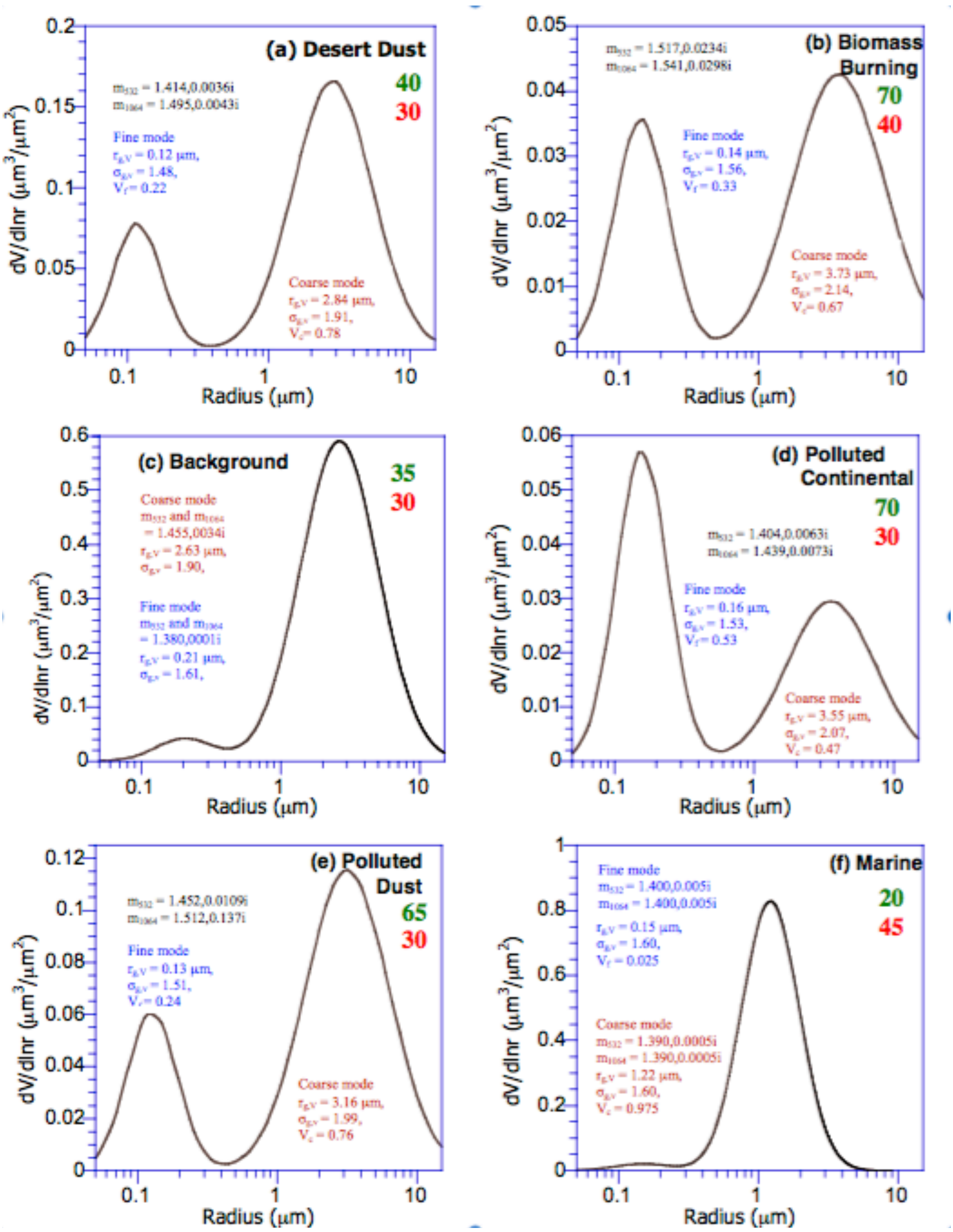

Figure 4: The size distribution and microphysical properties of the CALIPSO aerosol models. For each model, extinction/backscatter ratio S_a at 532nm (green) and 1064 nm (red) are shown.