

General comments:

A better knowledge of the radiative influence of the aerosols on the Mediterranean climate is important to estimate their impact on the global warming. The Mediterranean region is rich in a variety of particles from both continental and marine sources. This paper presents the first results of an ambitious experimental campaign, based on surface and aircraft observations that propose a rather complete view of the physiochemical and optical properties of the Mediterranean aerosol. In addition, the data analysis takes benefit of the expertise from a large group of recognized scientists. Although we could discuss of the relevance of such a long paper, this manuscript brings valuable advanced results on the aerosol properties across the Mediterranean basin. However, some questions arise when reading the manuscript. First of all, if the objectives of the ChArMEx/ADRI-MED project are well-presented in the introduction, the aim and the borders of the paper should be better précised, more particularly in view of the fact that the authors continuously refer along the paper to other published results on the same topics (for instance, Section 5.4.1 mainly deals with the results of Nicolas et al. (2015) and Meloni et al. (2015)). Few parts of the manuscript looks like a compilation of results which could have been more synthesized. In particular, our understanding would gain a lot if the authors could provide a synthesis of the different results they obtained to make the reader see how to relate them, as for the AOD data in Section 5.2 or for estimates of the radiative forcing reported in Section 5.4. In addition, the authors present the analysis of the aerosol composition in Section 5.1.4 and the CHIMERE calculations in Section 6.2, but any link is made between the two sections. By the way, the comparison between the different regional models reported in Section 6 does not seem really useful for this paper since all models did not take into account aerosol species in a similar way. I also think that the manuscript could be improved thanks to a more rigorous comparison between the aerosol characteristics at the different sites. To my opinion, the major interest of the paper deals with the estimates of the local radiative forcing and the large dataset concerning the aerosol extinction provided using different instruments and methods. It is clear that this paper merits publication in ACP. I would recommend, however, a revision of the manuscript in view of the comments that I have listed below.

First, we'd like to thank the reviewer for these constructive remarks and comments on the article. We have tried to take into account most of the comments to improve the resubmitted manuscript. The detailed answers are provided hereafter.

Major concerns:

p. 19642: The description of the general meteorological conditions seems incomplete. In spite of the figure 8, too small and providing pressure by the way, the wind speed, which is the key parameter of the aerosol transport is not really given with a sufficient precision in any part of the text.

The wind intensity and direction were indicated in the Figure 7 for three different stations and in the Figure 8 at the altitude of 700 hPa but we agree that this important meteorological parameter should be more detailed. In that sense, we have included in the revised manuscript the vertical profiles of the wind intensity at the three different stations (Ersa, Lampedusa and Minorca) in the new Figure 8 for all the ChArMEx/ADRI-MED period. In addition, a new paragraph discussing the wind speed intensity during the observed period of important sea-spray concentration at Ersa has been included (part 5.2.1).

p. 19645: The comparison between the two coastal sites, i.e., Lampedusa and Ersa indicates a significantly higher mass concentration in Lampedusa. One can expect that the surplus in aerosol concentrations measured in Lampedusa is rather due to height above the sea of the aerosol acquisition, which was closer to the sea surface in Lampedusa than in Ersa. In particular, if we

consider that the sea-spray aerosols issued from breaking waves can largely contribute to the PM10 concentrations in the lower layer in such marine environments, a correction factor could be easily used for an accurate comparison by assuming an exponential decay of aerosol concentrations with altitude. The vertical profiles of aerosol concentrations can be then modelled using Toba (1965) as a kernel. The authors can also use the work of Piazzola et al. (2015) who approached the concentrations decay with altitude by a logarithmic law using vertical aerosol profiles measured in the Indian Ocean compared to data obtained from the CALIOP level2 operational products. I would suggest the authors to use this kind of corrective factor to ensure an accurate comparison between the two sites. This remarks can also be considered for the comparison between the volume distributions at four different sites reported in p. 19646.

Thank you for this interesting remark; this is effectively right and we have now used the logarithm law proposed by Piazzola et al. (2015) for estimating the concentration of aerosols at the Ersa station for an altitude of 50 m, which is close to the altitude of Lampedusa (45 m). The calculation has been made using the value of 0.75 for the coefficient s (that corresponds to particle sizes higher than $0.5 \mu\text{m}$, see the Figure 8 of Piazzola et al., 2015) for sea-spray aerosols. By applying this correction, the corrected mean PM10 aerosol mass concentration is $12 \mu\text{g m}^{-3}$, closer to the value observed at the Lampedusa station. We have now indicated this point in the text and in the new figure (including one additional curve).

In addition, we have now also added a new paragraph on this specific point in the section 5.1.1: "In order to take into account the difference of altitudes between the two sites of Lampedusa and Ersa, we have applied a correction factor to PM10 observed at Ersa (530 m) for estimating a new PM10 concentration corresponding to the altitude of Lampedusa. In that sense, we have applied the logarithmic law reported by Piazzola et al. (2015) using a value of 0.75 for the factor s to correct the mass concentration of sea spray aerosols only. The calculated mean value of PM10 at 45 m is about $12 \mu\text{g m}^{-3}$ (Figure 13), closer to the mean value observed at Lampedusa ($21 \mu\text{g m}^{-3}$). A new caption has also been added for the Figure 13.

It should be noted that the same corrections have been now applied to the concentration of the coarse mode estimated from sun-photometer observations at Ersa. The value is now reported in the Table 6, using the value of 0.75 for the coefficient s . This point is now also mentioned in the text in the section 5.1.2.

p. 19646: The impact of the convective processes on the concentrations of anthropogenic aerosols could have been evaluated through the survey of the air-sea temperature difference. This induces a seasonal variation of the anthropogenic aerosols which can explain the differences noted with the ESCOMPTE campaign. This should be included in the analysis of meteorological conditions to produce large concentrations of polluted-smoke particles.

To our point of view, this aspect is already discussed in the parts 4.1 and 4.2 of the article, where we have demonstrated that the meteorological field (surface temperature, synoptic situations) observed during the SOP-1a campaign were not favourable to produce large concentration of secondary polluted or smoke aerosols. Such meteorological situations are indeed very different with those observed during the ESCOMPTE campaign, where AOD as large as 0.3-0.5 (in the visible range) has been observed due to the important concentration of anthropogenic-polluted particles. This point has now been reinforced in the part 5.1.4 of the new version, where comparisons with the ESCOMPTE observations are mentioned.

p. 19648-49 : By the way, the comparison of the Ersa and Lampedusa chemical analysis with the data reported during the ESCOMPTE campaign does not seems appropriate. The authors should rather compare their results to sites with quite similar character, whether it is located in the Eastern (see, Eleftheriadis et al. 2006; Bardouki et al. 2003) or in the Western Mediterranean (e.g., Piazzola et al., 2012; Sellegri et al., 2001).

This is effectively right and we have now included some comparisons with the data reported in the different references listed by the reviewer, which are more appropriated than those obtained during the ESCOMPTE project. However and as mentioned in the article, we have chosen to focus our discussions on

the BC and OC mass size distribution as such measurements are original and scarce over the Mediterranean compared to other aerosol species (as sulphates, sea-salt, nitrates or ammonium) largely referenced. In that sense, we did not use the Bardouki et al. and Sellegri et al. papers, which are mainly focused on inorganic species but we used the Eleftheriadis et al. (2006) work, which reports BC concentration at the Finokalia remote coastal site and onboard the R/V "Aegaeon". We have also used the Piazzola et al. (2012) and Mallet et al. (2011) works, which report aerosol mass size distributions of BC and OC aerosols at the Porquerolles coastal island (southeast France). Comparisons are focused on the different modes of the BC and OC mass size distributions. This point is now integrated in the article in the part 5.1.4. The following sentence has been added:

"It should be also noted that the EC concentrations observed at the Ersa station are logically (due at least to the altitude of the station and the absence of intense pollution during the SOP-1a, see section 4) lower ($0.39 \mu\text{g.m}^{-3}$) than EC concentrations (PM_{2.1}) reported by Eleftheriadis et al. (2006) from the eastern Mediterranean during the summer season ($0.60 \mu\text{g.m}^{-3}$) in July 2000. The same ascertainment is obtained on OC concentrations with higher values ($4.2 \mu\text{g.m}^{-3}$) reported by Eleftheriadis et al. (2006) compared to observations in Ersa ($1.5 \mu\text{g.m}^{-3}$). Concerning the modes of the OC and EC particle mass size distributions, the two identified modes detected in Ersa are consistent with those reported by Mallet et al. (2011) at the Porquerolles coastal island (south eastern France), who also detected two (fine and coarse) different modes of the mass size distributions for EC ($0.3\text{-}0.4 \mu\text{m}$ and $4\text{-}6 \mu\text{m}$) and OC ($0.3 \mu\text{m}$ and $5\text{-}6 \mu\text{m}$) aerosol particles."

P. 19653: The authors explained the low values of the SSA measured in Lampedusa by the contribution of the coarse mode to the total size distribution, which is attributed to the dust aerosols. Would it be possible that the sea-spray production at the air-sea interface (see next comment) also contributes to the decrease of the SSA through the injection of coarse and giant particles in the MABL?

This is effectively right and the presence of the coarse mode of sea salt aerosols could effectively contribute to the decrease of SSA in the solar spectral region. We have now added this specific point in the new version.

p. 19657: The southwest episodes allowing dust transport in the Northern Mediterranean is also often characterized by the occurrence of strong sea-spray injection in the lower part of the Marine Atmospheric Layer through breaking waves in addition to deposition fluxes of the dust particles advected from the Saharan region. This is confirmed by the LNG surface observations reported in Section 5.3.2. Can we consider that the AOD values measured in these conditions should be due to the combination of dust and strong sea-spray flux occurring at the sea surface? Could the authors use more the Angstrom coefficient to provide a better analysis?

Effectively, the AOD measured on 19 June and presented in the Section 5.3.2 using LNG observations is the combination of sea-salt aerosols produced at the sea surface associated with the presence of mineral dust transported above the MBL. This point is mentioned in the document:

"The aerosol extinction is found to be significant around 41 to 41.5° N that could be due to sea-salt particles generated in south Corsica Island due to the local acceleration of the wind occurring between the Corsica and Sardinia islands (not shown). This increase of the aerosol loading in the MBL associated with dust aerosol transported to higher altitudes results in an increase of total AOD at these latitudes."

In addition, the remark of the reviewer is interesting as we can effectively observe a difference in the daily Angstrom Exponent (AE, calculated between 440 and 870 nm) between the Ersa (AE of 1.0) and the Minorca (AE of 0.5) stations, which are affected by this dust event. However pure sea salt and desert dust have AE in the same range (<0.5) and we cannot conclude that such a difference is due to a larger contribution of sea-salt at Ersa.

p. 19666: The main objectives of the ten ChArMex project is to investigate how the modifications of the radiative budget due to aerosols affect the sea-surface evaporation fluxes. Concerning the sea-

spray aerosols, could the impact on the sea-surface evaporation fluxes and relative humidity profiles be estimated ?

This interesting question is difficult to answer. To our point of view, the AOD contribution due to sea-salt is likely too moderate on average for affecting significantly the sea surface temperature, O-A fluxes and relative humidity profiles, but a firm conclusion would need an important work with specific simulations which we consider outside of the scope of this paper. The possible impact of sea-salt radiative forcing on the sea-surface evaporation fluxes and relative humidity profiles should indeed be studied using specific simulations including only marine aerosols in a coupled Ocean-Atmosphere model.

P. 19658 If a strong contribution of dust aerosols is indeed noted all along the campaign, do these results allowed to say if it is different from the past, especially if we consider that the measurement period is known to be the good one (with the autumn season) for Saharan dust intrusion in the Northern Mediterranean.

The representativity of our observations obtained during the 2013 summer period compared to other years and seasons is an interesting scientific question. A specific work-package of the ChArMEx project dedicated to the variability and trends over the Mediterranean is on-going to investigate this point. Nevertheless and as mentioned in the part 4.2 of the article, we have integrated the AOD anomalies of summer 2013 compared to summer AOD derived from MODIS and MISR data (for the 2000 to 2013 period). Our conclusions were that the aerosol concentrations observed during the SOP-1a were slightly lower but in the same range of magnitude that usually observed during summer period over the western Mediterranean. This point is now detailed in the section 4.2.

p. 19651: The results reported in Section 5.2.1, 5.2.2 and 5.2.3 should deserve to be synthesized. ***As much as possible, we have tried to synthesize and reduce the size of the three different sections in the new version.***

p. 19663: The comparison of the COSMO-MUSCAT with other regional models which does not have the same characteristics (Table 8) seems inappropriate since all models did not take into account aerosol species in a similar way. I am not sure that this part of the paper is very useful.

We propose to let in place this part related to the COSMO-MUSCAT model in the article. We understand the remark but although COSMO-MUSCAT doesn't take into account all aerosol species (especially secondary inorganic), this model simulated the dust sources, emission fluxes, size distribution, vertical profiles and (dry/wet) deposition of dust aerosols (which are the major species in most of the SOP-1a aerosol events) in a different way than the two other RCM (RegCM and ALADIN) or the CTM CHIMERE models. In that context, it appears important to us presenting the results obtained with this regional model.

p. 19669: Some question also deals with the radiative impact of Mediterranean aerosols, the TOA simulations presented in Fig. 29 at the end of the manuscript, we could expect the authors to relate their results to the potential changes of the radiative budget due to aerosols in the Mediterranean or compare them to the work of Nicolas et al. (2015) and Meloni et al. (2015).

This is a very interesting remark and we have now added some comparisons with the results obtained from 1-D radiative transfer calculations. In that sense, we focused our discussion on the Nicolas et al. (2015) work, who performed different simulations using different surface albedo based on the ATR-42 flights above the Balearic islands (flight 29) and the Granada (flight 30) station, which are characterized by two distinct surface albedo. The inclusion of high surface albedo (0.27 at 870 nm) in the 1-D radiative transfer model compared to sea-surface albedo (0.02 at 870 nm) decreases the Top Of Atmosphere radiative effect from forcings weakly (-4 W m^{-2}) to significant negative (-10 W m^{-2}) values, for the Granada and Minorca simulations, respectively. Such results are consistent with the 3-D simulations presented in Fig. 29 and we have now added a sentence on this point in the part 6.5.2:

"Such results are consistent with the study of Nicolas et al. (in prep.), who performed two different simulations using different surface albedo (from marine to continental), based on the ATR-42

observations above the Balearic Islands and the Granada station. The inclusion of high surface albedo (0.27 at 870 nm) in the 1-D radiative transfer model compared to low sea-surface albedo (0.02 at 870 nm) contributes to decrease the TOA radiative effect at Granada."

p. 19671: The authors concluded "Non negligible aerosol extinctions (about 50 Mm⁻¹) have also been observed within the Marine Boundary Layer (MBL), due to the presence of polluted or marine aerosols." Maybe I have missed something, but I did not see anything in the manuscript that permits this conclusion.

This is effectively right and we have now modified this sentence by removing the term "polluted" and including the possible contribution of sea-spray aerosols to the aerosol extinction in the MBL. The new sentence is the following:

"Aerosol extinctions measured on-board the ATR-42 show local maxima reaching up to 150 Mm⁻¹ within the dust plume, associated to extinctions of about 50 Mm⁻¹ within the Marine Boundary Layer (MBL) possibly due to the presence of sea-spray aerosols."

Minor concerns:

p. 19619 and others: I would replace "sea-salt" by "sea-spray."

This is now done in the new version.

p. 19635: A comparison of the aerosol extinction vertical profiles with satellite data, as the CALIOP outputs could have been interesting.

This specific comparison is proposed in the work of Nicolas et al. (in prep. in this special issue), which will be submitted to the ChArMEx special issue as well as the Léon et al. (2015), which will be re-submitted.

p. 19646: In parallel, the lowest concentrations are observed at the Ersa station, near the anthropogenic sources of the southern France and Italy. This is well consistent with the absence of intense polluted photochemical or smoke aerosol events during the SOP-1a.

This sentence was effectively not very clear. We have now rephrased it in the new version: "In parallel, the lowest concentrations are observed at the Ersa station due to the absence of intense polluted photochemical or smoke aerosol events over southern France and Italy during the SOP-1a."

p. 19699: I don't know if it is due to my printed version of the manuscript, but the figures are too small to be clear.

We think this is due to the printed version. All the figures have been provided in .eps or .pdf, with an adapted format.

References

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