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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source contributions to 2012 summertime aerosols in the Euro-Mediterranean region" *by* G. Rea et al.

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We thank the reviewer for the careful review of this manuscript and valuable comments to improve its quality. Please find below our point-by-point replies.

In the manuscript "Source contributions to 2012 summertime aerosols in the EuroMediterranean region", the authors try to quantify the relative contribution of emissions to surface PM and AOD over Europe and the Mediterranean regions in summer 2012. They use a regional model CHIMERE driven offline by the WRF mesoscale model and model results are compared to in situ measurements and satellite observations. The paper is a good contribution to the understanding of the aerosol budget over those specific regions. Even if the results are not very surprising (anthropogenic

emissions dominate surface PM in the northern part of the domain and dust particles dominate the aerosol budget in the western Mediterranean region), the relative contributions are quantified in this paper. Although the simple methodology of switching off emissions from different sources can lead to uncertainties due to the non-linearities of the physical and chemical processes leading to the formation and loss of particulate matter (rather than a tagging method), I find the paper of good quality and I recommend it for publication. The performance of the model is quite well evaluated both at the surface and in the vertical column. However, the last section (Section 6) of the paper, which turns out to be the most relevant part of this study, needs significant revision since the results on the causes of the exceedances are really difficult to interpret.

Main comments :

1. Sect. 6 is the key section of the paper because the authors try here to determine the relative contributions to the aerosol budget when the mass concentrations exceed the limits of the health based standards. But this section is treated in a rather qualitative style, and it is thus difficult to understand the main results. How often does the model overestimate dust mass concentration, hence simulates unexisting exceedances ? Is there any reason for that (issue in the dust emission routine, transport patterns not well reproduced, underestimation of wet/dry deposition...) ? How often does the model capture the exceedences linked to anthropogenic sources or fires emissions? It would be relevant to add some statistics to assess how often the model reproduces the observed exceedances as a function of the main contributions : naturally-occuring and not naturally-occuring exceedences...

The conclusions about the amount of exceedances due to dust pollution events might be uncertain, due to the remaining discrepancies between the number of exceedances detected by the model and the observations. In the new version of the manuscript, the contribution of each source on exceedances only seen by the model is analysed (new

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Figure 14), and a new method to avoid the bias at each station is made, decreasing the total number of exceedances detected by the model to almost the half. The new paragraph with the explanation of the method is (Section 6): "First, the bias on background levels is evaluated at each measurement station. The background is defined as the baseline concentration, on days when no significant peak is measured. It is generally associated with anthropogenic and biogenic sources, which have relatively low variability during the summer compared to dust and fire emissions. The average "background bias" is estimated to -6.3 μg m^{-3} for PM_{10} and -1.9 μg m^{-3} for $PM_{2.5}$ in MED-We, -0.93 μg m^{-3} and 1.3 μg m^{-3} in NEU-We, and -3.2 μg m^{-3} and -0.2 μg m^{-3} in NEU-Ce, on average over all stations. Then for the sporadic sources, i.e. for the dust and fire, the resulting bias at each point of a peak is subtracted only for contribution concentrations of the corresponding source. This processing is performed only when the model overestimates the concentrations with respect to observations, but not when a peak is detected only by the observations." The contributions are then analysed on this basis, making the results more robust. The proportion of exceedances that happened exclusively from a single source is quantified, and also the proportion for the other exceedances: "In total, the only sources that result exclusively (i.e. when their contribution alone on PM concentration is more that the threshold of 50 μg m^{-3}) in an exceedance are dust (in 59% of the cases, i.e. 294 exceedances on the 498 observed and simulated), fires (in 1% of the cases, i.e. 5 exceedances), and anthropogenic sources (2 exceedances). The other 197 exceedances are due to mixing between several sources: anthropogenic sources contribute to 9.2% (MED-We) to 27.9% (NEU-Ce) of the concentrations, fires from 14.2% to 17.8%, and dust from 49.5 to 67.1%.

For $PM_{2.5}$ exceedances, the only sources that account exclusively in concentrations above 25 μg m^{-3} are anthropogenic sources (69 exceedances on the 374 observed and simulated at the same time, i.e. 18.5%) and fires (5 exceedances). The 300 other exceedances are a mix between anthropogenic sources (from 32.9% to 41.1%), biogenic sources (12.1% to 24%), fires (3.4% to 11.5%) and mineral dust (6.1% to 16.1%)."

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2. Is there also any standard for PM2.5 proposed by EU legislation (P 8212, l 11) ? PM2.5 values are used along the paper and or unfortunately not considered in this final section. There are nevertheless more relevant than PM10 for human health. Doing the same study for PM2.5 would be relevant and would give a better insight on the understanding of the main causes of PM exceedances in this region.

There is no legislation in Europe for $PM_{2.5}$, only an annual aim of less than 25 μg m^{-3}. However, in other states and according to WHO recommendations, the 24h-averaged concentration must be lower than 25 μg m^{-3}. The same study as PM_{10} exceedances is thus been made in the new version of the manuscript, as suggested by the reviewer.

3. Surprisingly, the correction on threshold added when dust contribution is larger than 60% does not lead to any significant improvement of the model performance to simulate the exceedances (P 8214, 5-7). How do the authors therefore explain the large remaining discrepancies during pollution events ? The final conclusions on the role of dust on pollution peaks might be too strong.

A new method has been used in the new version of the manuscript, giving more robust results (see answer to main comment 1).

4. P 8214, l 12-13 : To check this assertion, I would expect also a comparison of the modeled AOD with observations for those specific exceedances in order to discuss if dust is overestimated at the surface only or if the whole simulated profile can partly explain the discrepancy.

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The assertion that the event of 20-21 June is not well captured by the model has already been analyzed in term of AOD in Section 4.1, P.8207 l.29 : "However, there is no associated overestimation of AOD values by the model compared to the observations (MODIS or AERONET). This suggests that the overstimation in surface PM may be due to an overestimated transport at low altitudes, i.e. a wrong vertical distribution of the dust plumes. However, it may also be explained by an excess in total mass and a shift in the aerosol size distribution towards finer particles, as highlighted by Menut et al. (2015) through comparisons of CHIMERE simulations with AERONET retrievals of the size distribution. An excessive PM concentration would then result in a correct AOD due to the variation of the extinction efficiency with the size of the aerosol."

5. Most of the exceedances reported by CHIMERE do not seem realistic, as they are ascribed by the authors to overestimated dust events (for more than 90% of cases). CHIMERE model is currently used (as part of PREVAIR) in Europe to forecast air quality and pollution peaks. Which main message and recommendations may the authors deliver in terms of directions for improvement and in terms of operational purposes ?

In terms of operational purpose, the modelling of $PM_{2.5}$ and PM_{10} is globally in coherence with observations, as suggested by the section 4 of the paper. However, discrepancies and difficulties in considering air quality standards exceedances, particularly because of uncertainties on peak values must be taken into consideration. We advice to analyse the PM speciation in CHIMERE outputs and to focus the confidence on biogenic and anthropogenic particulate matter. Due to their importance, mineral dust source is though necessary for the analysis of exceedances, but must be considered with precaution.

Other specific comments:

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6. p 8195, l 18-20 : Has it already been done in other studies over the Mediterranean region? If yes, how do the authors' results compare with previous work ? If not, it should be clearly said in the introduction.

To the knowledge of the authors, no other study was made with a model to estimate the contributions of all these different sources over the Mediterranean basin. However, some studies cited in the introduction have analyzed specific locations or emissions: "The majority of the studies previously cited have been made at specific experimental sites and sometimes for specific case studies or sources." A study has also been added and compared to the results for impact of anthropogenic sources on $PM_{2.5}$ in the Eastern Basin: "Another study by Im and Kanakidou (2012) showed that regional anthropogenic emissions contributes to 47% of PM2.5 in the Eastern Basin (Greece, 110 Turquey) in summer, with 4% from Istanbul".

7. Sect. 2.1 : What is the uncertainty associated to PM2.5 and PM10 observations from AirBase ?

We added these sentences in Section 2.1, first paragrah: "The diversity of instruments and sampling, associated with certain meteorological conditions (humidity) contribute to the uncertainty of the database. The quality standards on the measurements for the AirBase network is less than 25%."

8. P 8199, l 4-5 : "the uncertainties are well-documented". Please give numbers corresponding to the AERONET products used in this work.

The precision of the AOD measurement, found in the literature, is given in the sentence: "The estimated precision of the direct AOD measurements is $\sim$0.01 (at 440 nm) for a recently calibrated instrument (Holben et al., 1998)"

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9. p 8200 : The authors say that exceptional high AOD were observed in June 2012. Do intense dust events explain it ?

A strong dust event actually happened in June 2012 (Nabat et al., 2015). This precision has been added in the sentence Section 2.2): "Exceptions appears in June where the AOD was higher than the climatology in 330 North Africa and over the Atlantic Ocean near North African coasts (due to a strong dust event (Nabat et al., 2015))".

10. P 8202, l 7-8 : Only 18 vertical levels between the surface and 200 hPa are used. Since most of the discussion is related to surface PM, the authors should detail the number of levels in the PBL. What is the point of using a regional model in such a large area if the number of vertical levels is lower than in a global model ? Do the authors see any improvement in the model performance in comparison to the LMDz-INCA model used at the boundaries ?

The vertical levels of CHIMERE are concentrated in the lower troposphere: there are 18 levels and the majority is near the ground. The regional model is thus more representative of the surface levels and allows a more precise analysis of surface concentrations. 18 levels are enough to describe the lower troposphere. The global model LMDZ-INCA is used to simulate the stratosphere. A sentence is added in the new version of the manuscript (Section 3.2): "The vertical discretization consists in 18 uneven levels in hybrid sigma-pressure coordinates, from the surface up to 200 hPa and with 8 levels under 800 hPa."

11. P 8203, l 4-5 : This is a mistake : the AOD does not vary linearly with wavelength. Please recalculate the simulated AOD using an Angström exponent.

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The calculation of the AOD has been revised using a log-scale interpolation with the Angstrom exponent: "In this study, the simulated AOD from CHIMERE at 500 and 550 nm are used and extracted using the Angström coefficient from the output wavelengths" (end of the Section 3.2).

The results of the manuscript (evaluation of AOD and contributions) have been modified, but the differences are not significant.

12. P 8204, l 24-29 : Are the fire emissions set at the surface level ? Or do the authors use a plume injection height to distribute them vertically ? It can strongly impact the contribution from fires.

Fire emissions are injected homogeneously onto the boundary layer. We thus added the sentence (Section 3.3): "Fire emissions are injected homogeneously onto the boundary layer for this study. This parametrization can lead to uncertainties on the results as it impacts the transport of polluted plumes."

13. p 8205, l 20-22: I don't really understand the purpose of these two sentences. The authors may want to say they only consider emissions inside their regional domain.

corrected

14. p 8210, l 4-9 : The surface concentrations due to fire emissions may be overestimated, given that the injection heights do not seem to be taken into account in this paper.

See comment number 12.

15. P 8210, l 10-16 : There is a huge contrast between the PM10 contributions of sea salts over the Atlantic Ocean (> 80%) and over the Mediterranean sea (< 10%).

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It is probably caused by the online Monahan scheme based on surface winds. Is the contribution over the Mediterranean sea realistic ? Did the authors validate the surface winds from WRF?

The contribution of sea salt on PM10 concentrations is bigger in proportion over the Atlantic Ocean, but it is equivalent in absolute contribution. Over the Atlantic Ocean, almost only sea salts contribute in the concentration of surface PM10, but this concentration is low (up to 15 μg m^{-3}).

16. Sect. 5.2 : The section is relevant for the paper since it is an attempt to link surface PM and AOD. In this reviewer's opinion, it might be interesting to check if the vertical distribution of aerosols simulated from CHIMERE is realistic, because this discussion is only based on modeling results. Different vertical profiles may lead to a same AOD. An averaged profile of CALIOP over summer 2012 (on clear sky days) would be really valuable to support the discussion of this section. Also, AOD from AERONET and MODIS are only available when clouds are not present in the vertical column, whereas surface PM are available most of the time. The comparisons between model and observations are not performed on the same collocated data. Therefore, the conclusions based on a surface PM underestimation associated with an AOD quite well reproduced should be done with caution, since clouds may be rather associated to the presence of certain aerosol types.

We agree that an evaluation of the vertical distributions of aerosols would be very helpful for the discussion provided in Section 5.2. A comparison between the simulated profiles and the CALIOP lidar observations would therefore be valuable. It has already been conducted in previous studies using CHIMERE. For example, Vuolo et al. (2009) used CALIOP observations to evaluate the vertical structures of transported dust plumes (during June to September 2006 and January to March 2007). They showed that the model has a vertical overspread of 50% in summer, but less multilayers dust situations. This bibliography has been added in the new version of the paper

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in Section 5.2.

Since this kind of analysis requires a precise analysis of a large dataset (due to the relatively low coverage of the CALIOP observation), as well as a complex study of the associated uncertainties, we have chosen not to include it in the present paper for the summer of 2012, which already integrates many datasets.

17. P 8211, l 22 : "the contribution of sea salts remains very low (< 1%), due to a low radiative impact". Did the authors indeed evaluate the radiative impact of sea salts in this study? Many studies highlighted the fact that the surface radiative impact on sea salts might be important over the sea due to their contribution to the activation of droplets in stratocumulus.

We did not evaluate the radiative impact of sea salts in this study, we thus removed this sentence in the new version.

18. P 8211, l 25 : I didn't find the figure illustrating this temporal variability.

We decided to not include the figure of AOD temporal time series of contribution (like Figure 10 for PM) because it is similar to the Figure 10. But for the new version, we added it on Figure 10 (now Figure 11 in the new manuscript) to allow a better understanding of Section 5.2.

19. P 8212, l 2 : "contributions from fine particles to AOD are larger". Please give numbers for fine and coarse AOD provided by AERONET and MODIS.

This sentence was ambiguous: the authors wanted to say that the contributions from the different sources observed in the temporal variability of AOD followed the contributions of sources on column $PM_{2.5}$ concentrations (not shown) rather than the contributions of sources on column PM_{10}. The sentence has been re-written in the

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new version of the manuscript (see answer to comment 20).

20. P 8212, l 4 : "more active optically". I am not convinced by this comment : particles in the accumulation mode may indeed be more optically efficient (in terms of scattering efficiency) than larger mineral dust particles due to the size parameter (diameter over wavelength), but the impact of dust particles on aerosol extinction coefficient or AOD still remain significant as their surface size distributions have a very large contribution. To support the discussion of this section, it would be interesting to give here the relative contributions to AOD of dust versus anthropogenic emissions.

This paragraph has been re-written, with relative contributions of AOD (Section 5.2, last paragraph): "Compared to the total column (vertically integrated) concentrations of PM (not shown), source contributions to AOD follow contributions to $PM_{2.5}$ concentrations. At the considered wavelength (500 nm), fine particles from anthropogenic or local fire emissions, even concentrated in the first layers, contribute more significantly (34% for mean anthropogenic contribution) than larger mineral dust particles (23%) at the considered wavelength (500 nm) to the calculated AOD."

21. P 8212, l 9 : Current legislation provides different thresholds. Please clarify if it is a reference level for daily/annual mean or a limit value for human health.

We specified: "For PM_{10}, the air quality limit value is set, for health based considerations, to a daily mean of 50 μg m^{-3} and an annual mean of 40 μg m^{-3}."

22. P 8213, l 27 : where does this number of 60 % come from ? Does it correspond to a specific level . How is it chosen ?

The contribution of 60% was chosen arbitrarily to select peaks with "predominant" dust contribution. In the new version of the manuscript, a new method to optimize

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the source contributions on exceedances is computed (see comment 1) and then this arbitrary number is not used for extracting the peaks.

23. P 8216, l 10-11 : Fine particles (like BC) have generally diameters much smaller than 500 nm, so the efficiency itself is not at its maximum. But, more significantly, the surface size distributions have a larger impact than the scattering efficiency. The sentence given by the authors is not supported by any calculations in the manuscript. As I suggested previously, the authors should at least provide modeled and observed fine and coarse AOD contributions and AOD from dust/anthropogenic particles to conclude.

This wrong justification has been removed.

24. Tables 2 and 3 : Please define MFE and MFB in the manuscript. I don't understand why |MFB| is not the absolute value of MFB... Please do not forget the unit (ug/m3) in the tables.

The formulas has been added for MFE and MFB (Section 4.1):

$$MFB = \frac{1}{N}\sum_{i=1}^{N}\frac{(C_{mod} - C_{obs})}{(C_{mod} + C_{obs})/2} \quad (1)$$

$$MFE = \frac{1}{N}\sum_{i=1}^{N}\frac{|C_{mod} - C_{obs}|}{(C_{mod} + C_{obs})/2} \quad (2)$$

|MFB| is the mean absolute MFB among all the stations of the sub-region. A sentence has been added to explicit it in the new version. Units have been added in Table 2 and 3.

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25. Fig. 1 : is it really a daily average as indicated by the caption, or a monthly average of daily values ? Also "daily maximum" : is it a monthly maximum?

corrected

26. Fig. 4 : Harmonize the dimensions for the emission fluxes. Some of them are in molecules/cm2/s and others in g/cm2/s. It is therefore difficult for the reader to have an idea of the relative importance from the different sources. Also, the color scale is badly chosen for dust emissions, and some source regions appear in the "white" zone.

Figure 4 (Figure 5 in the new version) has been re-computed in the new version of the manuscript with a new scale for dust emissions, and with all the fluxes in the same unit ($g/cm^2/s$).

27. Fig. 5 : As indicated in this figure, the observed PM and CHIMERE results are daily means averaged over all the stations for each region. Please add the corresponding error bars and observed and modeled PM. They have been forgotten in the current version.

Figure 5 (Figure 6 in the new version) has now error areas corresponding to the standard deviation of the observations available within each sub-region.

28. Fig. 6 : Please also add the error bars.

Error bars have also been added in Figure 6 (Figure 7 in the new version).

29. Fig 12 is presented p 8212, l 17 in the manuscript but the thresholds used to build this figure have been modified according to p 8213, l 28. The authors should present both results on Fig 12 : the threshold of 50 ug/m3 to see the real performance of the model, and then the red curve where the thresholds are corrected from dust

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contributions.

In Figure 12, results from CHIMERE and CHIMERE "optimized" with the new method are both indicated in red and dashed blue line.

30. Fig. 13 : Representing the same figure also in PM2.5 would give more weight to the paper.

All the figures and discussions have been extended to $PM_{2.5}$ exceedances of the WHO recommandation of 25 μg m^{-3} (no current legislation in the European Union).

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8191, 2015.