Atmos. Chem. Phys. Discuss., 15, C1673–C1679, 2015
www.atmos-chem-phys-discuss.net/15/C1673/2015/

Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

Received and published: 19 April 2015

In the manuscript "Source contributions to 2012 summertime aerosols in the Euro-Mediterranean 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 phys-

ical 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...

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.

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

ACPD
15, C1673–C1679, 2015

role of dust on pollution peaks might be too strong.

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.

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 ?

Other specific comments :

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.

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

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

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

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 ?

ACPD
15, C1673–C1679, 2015

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.

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.

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.

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.

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%). 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?

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.

ACPD
15, C1673–C1679, 2015

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.

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

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.

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.

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.

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

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.

24. Tables 2 and 3 : Please define MFE and MFB in the manuscript. I don’t understand

ACPD
15, C1673–C1679, 2015

why |MFB| is not the absolute value of MFB... Please do not forget the unit (ug/m3) in the tables.

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?

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.

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.

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

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 contributions.

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

Technical comments :

In the manuscript, the authors should harmonize the expression “summer 2012”. It is sometimes also written “summer of 2012” or “Summer of 2012”.

p 8194, l 18 : at many locations

P 8199, l 25 : than surface measurements

ACPD

15, C1673–C1679, 2015

P 8205, l 21: since these emissions cannot be controlled

P 8208, l 1 : overestimation

p 8208, 10 : two datasets

P 8210, l 17 : Fig. 10 is introduced before Fig. 9 (only mentioned p 8211, l21). It should be probably better to permute the two figures.

P 8216, l 7 : sea salts

p 8216, l 24 : acknowledge

p 8216, l 29 : availability

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