

Response to anonymous Referee number 3 review

The authors would like to thank referee #2 for his/her useful comments. Each response to the referee's question is organized as follows: (1) comment from the referee in bold, (2) authors' response and changes in the manuscript in normal font. The changes in the revised manuscript, except the small edit corrections, are in green color in the revised manuscript. Moreover, the manuscript has been proofread by a native English speaker.

- 1. Perhaps the authors should clarify/justify the aim of the work. As a part of that, try to better explain their choice of the study region (the red rectangle) for the “Mediterranean basin” (this is only mentioned for dust on p. 12, line 22) and also 50 m wide coast line. I'd recommend that the authors make a better effort to show the added value of the obtained results and their scientific and/or practical usefulness and implication. The authors should make more clear the connection to the ChArMEx and the choice of the years, since they have not at all made any use of the campaign's data.**

Some of the points of this remark will be addressed later in this document. The choice of the study domain is addressed in points 2 and 6.

We agree that the paragraph in the introduction describing the aims of the work was not clear enough and also the link to ChArMEx. The paragraph of the introduction providing the aims of the paper has been changed.

« The past studies focused on summer season. Here we go a step further by analysing the aerosols over the Mediterranean region based on a two year long simulation that includes the intensive periods (2012 and 2013). Our objective is to establish the budget of the primary aerosols and secondary inorganic aerosols in this region for these two years including an analysis of its seasonal variability. Because particulate pollution is an issue there, we also analyse, from a sensitivity simulation, the contribution of the anthropogenic emissions from in the Mediterranean coast and from international shipping emissions to the aerosol budget. The years 2012 and 2013 having different mean meteorological conditions, this allows us to quantify the impact of this year-to-year meteorological variability on the aerosol budget. This work being part of the ChArMEx project, the choice of the years 2012 and 2013 for this study was linked to the possible availability of ChArMEx data, such as aerosol composition, on the long term that could be compared to the simulation results. Unfortunately, to day, these data are not available. Nevertheless, these years are of interest because of their different meteorology and also it makes possible to link our results to other ChArMEx studies published in the present issue. »

The conclusion has also been changed to make clearer the objectives of this study.

«This study aimed at establishing the budget of the primary aerosols and secondary inorganic aerosols on the Mediterranean basin based on numerical simulations of the years 2012 and 2013 using the MOCAGE model. We also studied its seasonal variability, its year-to-year variability with meteorological conditions, and the contribution of local anthropogenic emissions from the populated coastal area and from international shipping in the Mediterranean basin. Firstly, [...] ».

- 2. I'd recommend to use (international) shipping or ship emissions instead of anthropogenic emissions from the sea. By the way, would not it be more useful to assess separately the role of international shipping and land-based emission in the aerosol pollution in the region?**

The Mediterranean basin is a region of high economic activity linked to the shipping business. The development of both coastal land-based and international shipping economy are linked and should not be dissociated according to the authors. The reference to international shipping emissions has been corrected.

- 3. Please, specify what were aerosol sizes included in the study (p.3 l.30-31), in particular for sea salt and dust (p.4)**

The aerosol sizes in the model have been added in the model description.

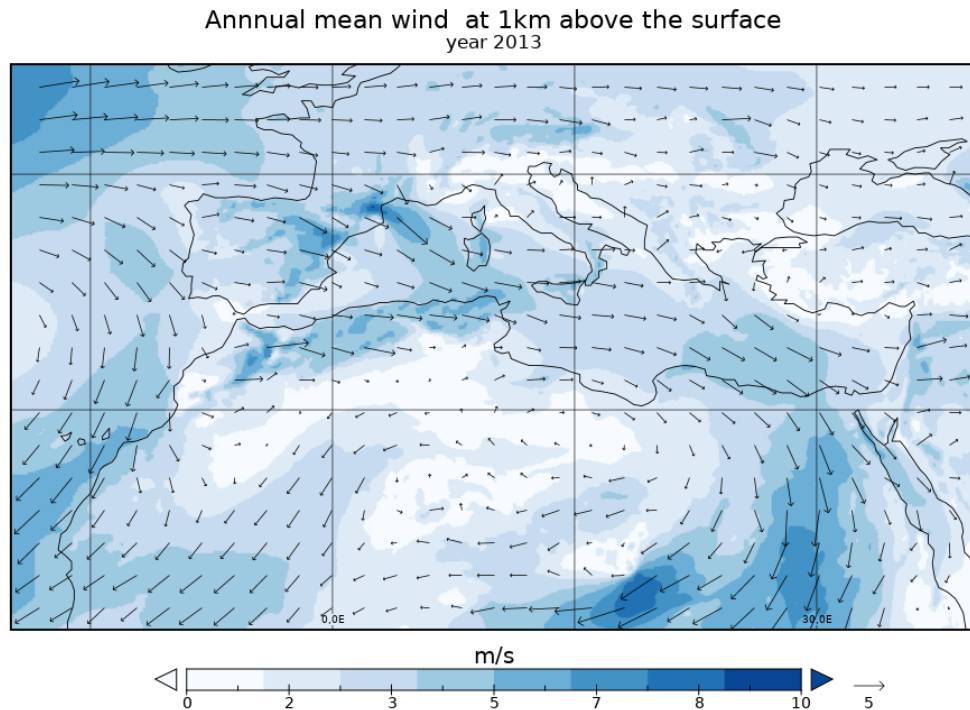


Figure 1. Yearly mean of wind vectors at 1000m above the surface for the year 2013.

4. **Why have not you limited your model evaluation with EMEP data to secondary inorganics. Why would not you also evaluate PM10, PM2.5, elemental carbon, Na+ (perhaps even dust)?**

The PM10 and PM2.5 data are already evaluated using the AQeR database which has a much larger number of stations than the EMEP database. Following your suggestion, we added the comparison for elemental carbon and organic carbon and sodium in the manuscript.

5. **to p. 9. African dust is often transported at higher levels in BL and free troposphere. Please justify the use of 200m high winds for describing dust transport here. Have you looked at 3d dust concentrations/vertical profiles?**

Indeed desert dust can be transported in the free troposphere. The wind map at 200m is representative of the global wind in the low troposphere. Figure 1 present the wind map at 1 km above the surface for the year 2013. The wind vectors in this figure presents the same patterns and structure as at 200m above the surface (Fig. 6 in the manuscript). Moreover the 200m level is closer to the surface, hence it is more representative for the desert dust emissions behaviour.

6. **How sensitive the results of the study to the choice of the study area (the location and size of the red rectangle)? See p. 12 line 22**

We chose a domain that contained all the basin and the coast to account for the inseparable maritime and harbour activities. The complex pattern of the seaside coast forces us to sometimes take a large part of the inland (e.g. in North Africa). Moreover we wanted a rectangle domain to ease the computation and the analysis regarding meteorological parameters which would have not been possible with such a discontinuous domain. We have not tested the sensibility of the location of the limit of the domain used.

Nevertheless, we expect the sensitivity to vary from one aerosol to another. It is important for desert dust, as we have seen, the southern boarder of the study domain crosses an emission area. For sea salt for example we expect the results to be less sensitive as they are not transported far away due to their large size. For primary organic carbon and black carbon, we also expect the result to be not very sensitive as the export essentially is linked to high concentrations in the Middle East associated with a western flux exporting them.

5

7. **Almost half of the Conclusions section is actually Outlook. I'd recommend to improve the Conclusions, avoid general statements and highlight your findings.**

The conclusion has been partially rewritten and completed.

8. **Decide whether to use "sea salt" or "sea salts"; better to use import/export than importation/ exportation**

10

The text has been changed to use sea salt and import/export.

9. **Page 1 L 4-5: why not write like 29° N , 10° W**

ACP's author guidelines only claim coordinates need a degree sign and a space when naming the direction. It is the author choice to write it this way.

10. **L 8: this is quite "normal" seasonal variation – not sure it should be in Abstract**

15

The sentence has been removed.

11. **L11: "with high values for some of them" - specify or drop out; Maybe better to give a range of values for different aerosols than just BC example (40%).**

The sentence has been removed.

12. **L 17-18: I'm not sure it's quite true for dust, since you approach doesn't distinguish between export and import.**

20

Here, the sentence only applies to anthropogenic emissions as written line 15. Since it seemed not clear enough we added it also line 16.

13. **L 22: on weather (meteorology is a scientific study)**

Corrected

14. **L 23: What do you mean by "sensitive to atmospheric pollution"? In what terms? What is the difference here between atmospheric pollution and air quality?**

25

We agree that we have misused the word sensitive. What we wanted to say is that the Mediterranean basin is subject to atmospheric pollution issues. The difference between atmospheric pollution and air quality is that air quality is one of the many issues that can be related to atmospheric pollution.

15. **Page 2: L 1-2: experiences periodic/sporadic pollution from forest fires? Complex topography? Check the sentence – geography/topography associated with the flows? Are you sure/What do mean by "it's especially sensitive to climate change"?**

30

Again, the word "sensitive" was misused. The idea here was to say that the climate simulations tend to show that the climate of the Mediterranean basin will become dryer and warmer, especially during summer.

16. **L 7: in summer months in 2012 and 2013**

35

Corrected.

17. **L 13-14: strange sentence**

The sentence has been simplified.

18. **L 9-19: not sure these details about weather during those short periods are relevant, but more information about “data collected” could be.**

The introduction has been completed with details about measurements made during the ChArMEx related campaigns.

19. **L 20-24: aerosol contribution to what? Edit: composed of; were the second large contributor. anthropogenic emissions were the major part of PM2.5 composition.**

The sentences have been changed.

20. **L 25-26: re-write the sentence**

Done.

21. **L 28: aerosols.. were dominated by. . .**

Corrected.

22. **L 32: ..two years .. include the the intensive periods. . .**

Corrected

23. **Page 5 L 15: the importance of SOA - inconsistent with p. 3 (31032)**

We do not understand where the inconsistency is. Page 5 L15 claims SOA can "represent a significant part of the fine mode aerosols" while page 3 l 31-32 claims that "the fine mode aerosol contribution [to **total mass**] is low". We added the words "to total mass" in the manuscript to be more precise.

24. **L 22 (also p.6 l.3): Are you discussing here MOCAGE vs CHIMERE? Is it in the paper’s scope?**

No we are comparing our results to a recent study over similar domain and period.

25. **L 31: for 2012 and 2013**

Corrected.

26. **Page 6 L 8-14: not sure it’s needed if just only the data format was changed, unless there were more essential changes affecting the data consistency**

These sentences present the data used in the study. There is now only one sentence about the change between AIRBASE and AQeR. The first three paragraphs have been moved to the Appendix following referee 2.

27. **L 15: periurban?**

Corrected to peri-urban (<https://www.eea.europa.eu/themes/sustainability-transitions/urban-environment/urban-green-infrastructure-glossary-for-urban-green-infrastructure>)

28. **L 16: for model evaluation..;**

Corrected.

29. **L 18-19: rural station; 1 to 5 are kept in order to assure a set of representative sites (or whereas 6 to 10 that are not representative are removed)**

Corrected.

30. **L 22: “similar behaviour” - awkward formulation; aerosols should be PM10 and PM2.5**

In this sentence we talk about the years 2012 and 2013, not PM10 and PM2.5. The sentence has been changed to be clearer.

31. **L 24: PM2.5 is better. . .**
This sentence has been changed.
32. **L 26: you mean that the natural aerosols are accurately calculated?**
We added a sentence to clarify that in the region where measurements of the AQeR database are made natural aerosols do not play an important role.
33. **L 28-29: “slightly less good” = worse**
5 Corrected.
34. **Page 7 L 1-5: If you used EMEP monitoring data (from ebas.no data base), you should write that explicitly. There is no need to list US, Asian etc networks/databases.**
Corrected.
35. **L 6: ..use measurements of secondary inorganic aerosols. . .**
10 Modified.
36. **L 23: can you specify the errors?**
The MOCAGE model does not represent SOA, so that we do not have available quantity to estimate the errors.
37. **Page 8 L 8 and 14: Inconsistency: All terms...directly computable, but indirect estimation Ttran ???**
The word "prognostic" has been changed to "estimated" to correct the sentence.
- 15 38. **Page 9, L 3: primary carbonaceous**
Corrected
39. **L 5-7: since you are discussing the hypothesis, how sound/unsound you mean it its, how much uncertainty in the results it causes?**
20 The authors are not discussing the hypothesis made. They are simply explaining why the destruction of sulphate aerosols is equal to zero. According to Fountoukis and Nenes (2007), "Sulfuric acid, sodium and crustal species have a very low vapor pressure and are assumed to exclusively reside in the aerosol phase". The sentence has been changed in the manuscript for a better understanding.
40. **L 15: What about the export from the Atlantic? Could it be that import/export are considerable, but compensate each other in the budget?**
25 The authors wrote: "We can then consider the global behaviour of sea salt is that there is almost no flux." This means that over the domain considered, the total import/export of sea salt is balanced. We can not give more information with our method used for computing the budget since we cannot differentiate import from export.
41. **L 19: to further facilitate the analysis...**
Corrected.
- 30 42. **L 22: “north of the southern boundary” sounds strange, maybe along the southern boundary of the domain;**
Corrected.
43. **L 23: I do not understand how E/N-E winds can transport dust from the south.:**
They transport desert dust, emitted into the study domain, towards the south, outside the domain of study. The sentence has been rewritten to be clearer.

- 35 44. **L 24-25: for carbonaceous aerosols; across the eastern border**
Corrected.
45. **L 26: Explain that you compare the precipitation amount and wind speeds in 2012 and 2013**
This was mentioned in line 21, but we added it in line 26 also.
46. **L 30: sea salt aerosol levels**
5 Changed.
47. **L 31: less precipitation, in exactly which area – impossible to tell from Fig. 6**
Corrected.
48. **L 33: higher speeds of the wind advecting/bringing transporting the pollution**
Corrected.
- 10 49. **Page 10 L 2-3: the monthly variations of budgets, better use the Figure present or show**
Corrected.
50. **L 5-6: may be to talk about larger and smaller emissions, or higher and lower dust levels instead of active/less active season**
It has been corrected by talking about dust levels.
- 15 51. **L 10. winds were stronger in North Africa. . .**
Corrected.
52. **L 14: not in summer, but in July month**
The sentence has been corrected by saying that the burden is higher or equivalent despite lower emissions.
53. **L 19: likely reason for the enhanced BC concentrations in July 2013**
20 The authors are not sure about the question asked here. We assume the question is "Why does not BC also have enhanced concentrations in July 2013?". The enhanced concentrations of OC in July 2013 originate from biomass burning emissions in North America. The emission inventory used gives much more OC emissions than BC for these events, explaining the difference of behaviour.
- 25 54. **L 26: bad formulation. Suggested: there are considerable differences in the tropospheric (?) load of the different aerosols.**
Corrected.
55. **L 29-31: does not the same (emissions and meteorological conditions within the domain) applies to sea salt; also explain “meteorological conditions within the domain”**
30 Since the emissions are dependent of the 10m wind speed, they have spatial variations according to the spatial pattern of the wind field. Hence, they are not the same inside and outside the domain of study. The “meteorological conditions within the domain” are the meteorological conditions inside the domain. This has been changed in the manuscript.
56. **L 33: Re-write sentence starting with Another source...**
Done.
57. **Page 12 L 17. The model does not equally “well represents the aerosols”. Please, make more precise summary.**
This part of the conclusion has been rewritten considering the changes made in the manuscript.

58. **L 21: ..all considered in the study aerosols are exported**
Changed.
- 5 59. **L 22-25: Re-write awkward sentences: We observed an annual cycle. . . (what's new about this?) and The annual cycle can also be affected.. (has to be more specific; what difference in primary emissions?. . .) L 31: Re-write the last sentences**
The conclusion has been partially rewritten and completed.
60. **Fig 2: make the circles visible**
Done.
- 10 61. **Figs 4-5: difficult to see the differences. Maybe to plot the difference instead?**
Both figures are important for the analysis and the colortable has been chosen to highlight the differences when they exist. There are already a lot of figures and tables. Adding the difference would not add information and would increase the size of the manuscript.
62. **Fig 7: correct caption: sea salt**
Corrected.
63. **Table 7 caption: comparison MOCAGE simulations with EMEP observations**
Corrected.