

Response to anonymous Referee number 2 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. **How the results of the present study compare with these earlier studies?**

We added comparisons to previous studies at several places in the text. In particular, we compared the results of our budget calculation to previous budget over Europe and found generally good consistency, such as the fact that the zone in a net source of pollutants as already shown by Aan de Brugh et al., (2011) and Im et al., (2014).

2. **Also in all tables and throughout the manuscript where reference is made to budget, it has to be clarified they concerne PM10 aerosol (or is it bulk aerosol in the model?)**

All the budgets presented in the manuscript are for total aerosol mass, not PM10. This has been clarified in section 4.2.

3. **Abstract line 6: annual AEROSOL burden**

Added to the manuscript.

4. **Section 2.1 MOCAGE model Information is missing on how the aerosol size is parametrized in the model. Since later in the discussion the authors refer to the aerosol sizes are taken into account in the model and how they change during atmospheric aging.**

The model uses a sectional representation with six size bins for each aerosol type. The sizes are ranging from to 2nm to 50µm and there is no effect of ageing on aerosol size in the model. We added this information and also added the size parametrization used for the desert dust that was missing in the manuscript (Kok, 2011).

5. **Section 2.1 last paragraph - here the authors could refer to observationnally based studies (Querol et al) the estimates the importance of dust aerosol for the background aerosol levels in the Mediterranean.**

The reference to the work of Querol at al., (2009) has been added to the text.

6. **Section 3.1. In the comparison with the database (MODIS, AERONET, AIRBASE/AQeR, EMEP), a seasonal comparison would be more useful than an annual.**

Following your remark, we added, when it was informative (MODIS and AQeR) a seasonal comparison to the evaluation of the simulation in the manuscript.

7. **line 14-15: comment on the underestimate of dust aerosol, what are the potential reasons?**

A potential explanation maybe that the desert dust aerosols are not transported far enough, the vertical injection and the following transport being hard to model properly.

8. **line 20: domains**

Corrected.

9. **line 24: simulate**

Corrected.

10. **Page 6, line 3: numbers**

Corrected.

11. **line 19 classes 1 to 5 are kept in order to remove ...**
Corrected.
- 5 12. **Section 3.4: it seems that no data from the Mediterranean region are included: this has to be explicitly mentioned in the text since it is a limitation of the comparison**
The text mention the "lack of this type of measurements outside Europe" highlighting the limitation of the comparison. To be more precise, we added a sentence saying that EMEP network only allows us only to characterize the north west part on the Mediterranean and that this is a limitation of the comparison.
- 10 13. **Page 8, line 5: Δ burden remove 'is'**
Corrected.
14. **Section 4.2.1 you need to define the exact 'box' in which you calculate the budget both horizontal and vertical extent of the area** Done in section 4.2
15. **Page 9, line 1 in the region;**
Corrected.
- 15 16. **Page 9, line 2 Define SIA**
Added to the manuscript.
17. **Page 9, line 3: Replace provide by add up to**
Corrected.
18. **Page 9, line 5: One can also note...**
20 Corrected.
19. **Page 9, 1st paragraph, please explain what are the chemical loss terms for nitrate and ammonium aerosol.**
For ammonium, nitrate and sulphate there are no emissions in the model. The first columns corresponds to the quantity of aerosol condensed. In the same way, the column "chemical loss" shows the evaporation of the aerosols. This has been added to the manuscript.
- 25 20. **Page 9, line 7, add after 'whatever the thermodynamic conditions are' 'because of the very low vapor presure of the sulfuric acid'. And remove the sentence after that:**
Done.
- 30 21. **Page 9, line 15-18: Nitrate aerosol partitioning to the aerosol phase is very sensitive to the aerosol pH (Guo et al., 2016) and NH₄NO₃ is semi volatile. How well is this computed in your model? Do you consider dust aerosol alkaline components in ISORROPIA calculations?**
35 The partitioning with the pH is treated by ISORROPIA (we are using version 2 of ISORROPIA), the input being the the ions concentrations and the water content. Although the pH has not been validated extensively in our model, it has been shown by Guo at al, (2015) that ISORROPIA II predicts well the pH. The water content input is provided by the numerical weather prediction simulation, in which data assimilation is made. Therefore, we assume the input information is as precise as possible. Concerning, the alkaline components are not yet considered in this study. A work is in progress to include it into the ISORROPIA implementation in MOCAGE. The inclusion of these compounds would increase pH and shift the partitioning of HNO₃ to the aerosol phase.

22. **Table 10 and 11, the percentage do not add up to 100% per species (no closure - could a mass balance issue?) and the differences from the 100% seem to be larger in 2012 than in 2013. Why there are differences. is it a question of spin up time for the model stabilization? Is there a significant change in the burden ? and if yes is such change justified and by what?**

The tables in the manuscript were not clear. The reader might have thought, for the first line of table 10, that $33.3 + 38 - 27.4$ should be equal to 100. In order to make the manuscript clearer, we changed the signs of the numbers in the tables. All sinks are now negative.

Moreover, the term of transportation is computed as the difference between the mass before and after the advection. Hence this term includes the error made during the advection which remain small but can become not negligible as we are using a semi-lagrangian scheme which has a smoothing effect on the strong peak in aerosols concentrations leading to a small loss of mass. This is discussed in section 4.1. This can be seen for example with desert dust where the residual is the higher. Moreover, the difference between 2012 and 2013 can be explained by the difference in the concentration field to be advected. For example, the desert dust emissions are higher in 2013 where the residual mass is slightly higher too. There is no spin up effect since the two years of simulation are made the same way, with the same 3 months spin-up. This has been added to the manuscript.

23. **Tables 14 and 15 are produced from Tables 8 and 9 (all emissions) when compared to Tables 12 and 13 (no anthropogenic emissions over sea and at the coasts). However, the percentages provided in Table 14 and 15 are similar but not the same as those calculated when using numbers from the relevant Tables. The numbers have to be double checked and the reason of this difference has to be clarified.**

It comes from the fact that the numbers in the tables 14 and 15 are computed with the real values of the budget, not the round numbers. For example for the very first term of the table 15 values in the manuscript are 1.07 and 0.76 which lead to $(0.76 - 1.07)/1.07 * 100 = -28.97\%$. But the "real" values are 0.763 and 1.073 which leads to $(0.763 - 1.073)/1.073 = -28.89\%$. The text has been changed to explain why the reader could find some small differences.

24. **Page 10, lines 7 and 8: replace season by dust period:**

Corrected.

25. **Page 10, line 10: over North Africa:**

Corrected.

26. **Figure 9 could be in the supplement since it is a global model result.**

Since this figure would be the only material in the supplement we prefer keep it in the manuscript.

27. **Page 10, line 22: annual cycle with maximum burden in summer:**

Changed.

28. **Page 10, line 28: from which boarder(s) of the domain the export takes place?**

With our computation method we can not have the exact position where the exportation takes place. Based on concentration and wind maps, we showed for example that desert dust aerosols are exported out of the study domain through the southern boundary, while carbonaceous aerosols are mainly exported out of the study domain through the eastern boundary. This has been added to the text.

29. **Page 12; 1st paragraph: also nitrate partitioning depends on the aerosol pH (see earlier comment)**

We added a sentence in the text to add this argument.

30. **Page 12, line 10 - mention also that many major cities in this area are coastal**

Added.

31. **Page 6, the values reported for Table 4 are different in the text than in the table**

This has been corrected.