

Interactive comment on “Primary aerosols and secondary inorganic aerosols budget over the Mediterranean basin during 2012 and 2013” by Jonathan Guth et al.

Anonymous Referee #2

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This study is a potentially interesting work on the aerosol budget over the Mediterranean during the years 2012 and 2013 but as mentioned by the other reviewer also, the observations from the campaign are not used for the model evaluation, so the only links to the ChArMEx experiment are finally the simulation period and the study area.

The manuscript is well structured but the use of English requires some polishing to be at the level of ACP publications. It could be suitable for publication in ACP after such polishing and a number of further improvements as outlined below.

Îž miss citations in the introduction or comparison of the model results with earlier studies that have shown aerosol budgets in the area like for instance the following

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modeling studies:

an de Brugh, J. M. J., Schaap, M., Vignati, E., Dentener, F., Kahnert, M., Sofiev, M., Huijnen, V., and Krol, M. C.: The European aerosol budget in 2006, *Atmos. Chem. Phys.*, 11, 1117–1139, <https://doi.org/10.5194/acp-11-1117-2011>, 2011.

Im U., Daskalakis N., Markakis K., Vrekoussis M., Hjorth J., Myriokefalitakis S., Gerasopoulos E., Kouvarakis G., Richter A., Burrows J., Pozzoli L., Unal A., Kindap T., Kanakidou M., Simulated Air Quality and Pollutant Budgets over Europe in 2008, *Science of Total Environment*, 10.1016/j.scitotenv.2013.09.090, 470–471, 270–281, 2014.

Im U., S. Christodoulaki, K. Violaki, P. Zarbas, M. Kocak, N. Daskalakis, N. Mihalopoulos and M. Kanakidou, Atmospheric deposition of nitrogen and sulfur over Europe with focus on the Mediterranean and the Black Sea, *Atmospheric Environment*, 81, 660–670, <http://dx.doi.org/10.1016/j.atmosenv.2013.09.048>, 2013

Mising also reference/comparison to experimental studies like the following data analysis studies that discuss the importance of dust aerosol for the background aerosol in the Mediterranean at surface: Querol, X., Pey, J., Pandolfi, M., Alastuey, A., Cusack, M., Pérez, N., Moreno, T., Viana, M., Mihalopoulos, N., Kallos, G. και Kleanthous, S.: African dust contributions to mean ambient PM10 mass-levels across the Mediterranean Basin, *Atmos. Environ.*, 43(28), 4266–4277, doi:10.1016/j.atmosenv.2009.06.013, 2009.

Querol, X., Alastuey, a., Pey, J., Cusack, M., Pérez, N., Mihalopoulos, N., Theodosi, C., Gerasopoulos, E., Kubilay, N. and Koçak, M.: Variability in regional background aerosols within the Mediterranean, *Atmos. Chem. Phys. Discuss.*, 9(2), 10153–10192, doi:10.5194/acpd-9-10153-2009, 2009.

There are also studies that investigate the entire tropospheric column over the Mediterranean, based both on ground based observations and satellite observations.

How the results of the present study compare with these earlier studies?



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

Specific comments:

Abstract: line 6: annual AEROSOL burden

Section 2.1 MOCAGE model: Information is missing on how the aerosol size is parameterized in the model. Since later in the discussion the authors refer to the aerosol size to explain the model results, it is appropriate to briefly outline what aerosol sizes are taken into account in the model and how they change during atmospheric aging.

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

Section 3.1

In the comparisons with the databases (MODIS, AERONET, AIRBASE / AQeR, EMEP), a seasonal comparison would be more useful than an annual.

lines 14-15: comment on the underestimate of dust aerosol, what are the potential reasons?

line 20 : domains

Line 24 : simulate

Page 6, line 3: numbers

Line 19 classes 1 to 5 are kept in order to remove ...

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

Page 8, line 5: Δ burden remove 'is'

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Section 4.2.1 you need to define the exact 'box' in which you calculate the budget both horizontal extent and vertical extent of the area.

ACPD

Page 9, line 1 in the region;

Page 9, line 2 define SIA

Page 9, line 3: replace provide by add up to

Page 9, line 5: once can also note...

Page 9, 1st paragraph: please explain what are the chemical loss terms for nitrate and ammonium aerosol.

Page 9, line7, add after 'whatever the thermodynamic conditions are' ' because of the very low vapor pressure of the sulfuric acid'. And remove the sentence after that.

Page 9, lines 15-18: Nitrate aerosol partitioning to the aerosol phase is very sensitive to the aerosol pH (Guo, H., et al. (2016), Fine particle pH and the partitioning of nitric acid during winter in the northeastern United States, J. Geophys. Res. Atmos., 121, doi:10.1002/2016JD025311) and NH₄NO₃ is semi volatile. How well is this computed in your model? Do you consider dust aerosol alkaline components in ISORROPIA calculations?

In Tables 10 and 11, the percentages 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?

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

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double checked and the reason of this difference has to be clarified.

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Page 10, lines 7 and 8 replace 'season' by 'dust period'

Page 10, line 10, over North Africa

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

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

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

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

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

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

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2017-670>, 2017.

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