Interactive comment on "Aerosol sources in the western Mediterranean during summertime: A model-based approach"

by Mounir Chrit et al.

Anonymous Referee #3
Received and published: 30 January 2018

This study presents the sensitivity of aerosols and their chemical composition over the Eastern Mediterranean as calculated by different model simulations performed in the framework of their ChArMEx experiment. The manuscript is very well organized and easy to follow, with a good level of English language. The manuscript is suitable for publication in ACP provided that the below minor comments are addressed in a revised version.

Material and Methods

1) Is the WRF model configured with 1-way or 2-way nested?

The WRF model was configured with 1-way nesting method.

This sentence is added in the revised version "…. To simulate WRF meteorological fields over the Mediterranean domain, 1-way nested WRF simulations are conducted on two nested domains …"

2) It would be good if more background is provided on how these different configurations for meteorology are designed.
The sentence:
"Before conducting the sensitivity study relative to meteorology (section 3) by using two different meteorological datasets, WRF is run with a number of different configurations, which are compared to measurements in section 3." is replaced by
"Before conducting the sensitivity study relative to meteorology (section 3) by using two different meteorological datasets, WRF is run with a number of different configurations, which are compared to measurements in section 3. In these configurations, the same physical parameterisations are used, but with different horizontal coordinates."
Furthermore, the description of the WRF configuration is added in the revised version:
"The WRF configuration used for this study consists of the Single Moment-5 class microphysics scheme (Hong et al., 2004), the RRTM radiation scheme (Mlawer et al, 1997), the Monin-Obukhov surface layer scheme (Janjic, 2003), and the NOAA Land Surface Model scheme for land surface physics (Chen et al., 2001). Sea surface temperature update, surface grid nudging (Liu et al., 2012, Bowden et al., 2012) options are activated. In the first configuration (WRF-Lon-Lat), horizontal …"

3) It is not clear from the text that both EMEP emissions and HTAP emissions are used in the simulations. Refer to Table 1.
The following sentence is added in the revised paper.
"… higher in HTAP emission inventory). HTAP emissions are used in the reference simulation and EMEP emissions for a sensitivity study as shown in Table 1.
Gaseous anthropogenic …"

4) Give more information on how the aerosols and organics are calculated in the model. Is it VBS that is used? Is the aerosol module operating on modal or sectional bins?

The aerosol modeling in this study is not based on the VBS approach but on a surrogate approach, and the aerosol size distribution uses a sectional approach.

A description of aerosol modeling is added in the revised paper after the description of the sensitivity studies on meteorology and sea-salt emissions: "…sodium and 4.22% of sulfate.

The SIze REsolved Aerosol Model (SIREAM; Debry et al., 2007) is used for simulating the dynamics of the aerosol size distribution by coagulation and condensation/evaporation. SIREAM uses a sectional approach and the aerosol distribution is described here using 20 sections of bound diameters: 0.01, 0.0141, 0.0199, 0.0281, 0.0398, 0.0562, 0.0794, 0.1121, 0.1585, 0.2512, 0.3981, 0.6310, 1.0, 1.2589, 1.5849, 1.9953, 2.5119, 3.5481, 5.0119, 7.0795 and 10.0 µm. The condensation/evaporation of inorganic aerosols is determined using the thermodynamic model ISORROPIA (Nenes et al., 1998) with a bulk equilibrium approach in order to compute the partitioning between the gaseous and particle phases of aerosols. Because the concentrations and the partitioning between gaseous and particle phases of inorganic aerosols (chloride, nitrate, ammonium) is strongly affected by condensation/evaporation and reactions with other pollutants, sensitivities of inorganic concentrations to hypothesis used in the modeling (thermodynamic equilibrium, mixed sea-salt and anthropogenic aerosols) are also performed (section 4.4.2)
For organic aerosols, the gas–particle partitioning of the surrogates is computed using SOAP assuming bulk equilibrium (Couvidat and Sartelet, 2015). The gas–particle partitioning of hydrophobic surrogates is modeled following Pankow (1994), with absorption by the organic phase (hydrophobic surrogates). The gas– particle partitioning of hydrophilic surrogates is computed using the Henry's law modified to extrapolate infinite dilution conditions to all conditions using an aqueous-phase partitioning coefficient, with absorption by the aqueous phase (hydrophilic organics, inorganics and water). Activity coefficients are computed with the thermodynamic model UNIFAC (UNIversal Functional group; Fredenslund et al., 1975). After condensation/evaporation, the moving diameter algorithm is used for mass redistribution among size bins. As detailed in Chrit et al. (2017), anthropogenic intermediate/semi-volatile organic compounds (I/S-VOC) emissions are emitted as three primary surrogates of different volatilities (characterized by their saturation concentrations C∗: $\log(C*) = -0.04, 1.93, 3.5$). The ageing of each primary surrogate is represented through a single oxidation step, without NOx dependence, to produce a secondary surrogate of lower volatility ($\log(C*) = -2.4, -0.064, 1.5$ respectively) but higher molecular weight. Gaseous I/S-VOC emissions are missing from emission inventories, they are estimated here as detailed in Zhu et al. (2016) by multiplying the primary organic emissions (POA) by 1.5, and by assigning them to species of different volatilities. A sensitivity study where I-S/VOC emissions are not taken into account is also performed.
"

5) Make it clear that dust emissions are not calculated in the model but only provided from the boundaries. Also provide information on how the boundary conditions are calculated. It is only in the results section that MOZART model is mentioned.

The boundary conditions are calculated from the simulation over Europe. The boundary conditions for the European simulation are calculated from the global model MOZART4.

Details about dust emission and boundary conditions are added after the paragraph describing the parameterization used for sea-salt emissions. "The boundary conditions for the European simulation are calculated from the global model MOZART4 (Horowitz et al., 2003) (https://www.acom.ucar.edu/ wrf-chem/mozart.shtml),, and those for the Mediterranean domain are obtained from the European simulation. Mineral dust emissions are not calculated in the model, but are provided from the boundaries."

6) Give more details on the SSE calculations. Is the surf zone included, how, or is only the open sea emissions calculated?
The sea-salt parameterizations used in chemistry transport models are built on open sea measurements. However, the same parameterization is used independently of the zone. This remark is added in the revised paper: "Sea-salt emissions are parameterized using Jaeglé et al. (2011) in the reference simulation and using the commonly-used Monahan et al. (1986) for a sensitivity study. These two parameterizations are based on open-sea measurements, but they are different in terms of the source function, "

7) Table 1 and 2 should be explained in the text and provide the motivation and the reasons for these different scenarios more clearly.
The different parameters, which are particularly attached to uncertainties, are detailed in the introduction. The section 2.1 aims to describe not only the set up but also alternative parameters/parameterizations commonly used in modelling. To clarify this point, the following sentence is added at the beginning of section 2.
"In order to simulate aerosol formation over the western Mediterranean, the Polair3d/Polyphemus air quality model is used, with the set-up described in Chrit et al. (2017) and summarized here. For parameters/parameterizations that are particularly attached to uncertainties (anthropogenic emissions,

meteorology, sea-salt emissions, modelling of condensation/evaporation), the alternative parameters/ parameterizations that are used in the sensitivity studies are also detailed. "
Section 2.1 is renamed "Simulation set-up and alternative parameterizations".

Results

8) Why the time series analyses for 2012 shown in the appendix? This would also show the extend of the dust break contribution to the levels.

All time series are shown in Appendix as specific pollution episodes are not studied. There was indeed a dust episode in 2012, but this episode is not discussed in details here, and a reference to the paper of Nabat et al. (2015) is added.

9) Explain Table 7 more clearly (in the caption maybe). What are the differences showing, period mean? What is the background of using normalized RMSE to show the sensitivity of the different inputs?
The periods used for the comparisons to measurements are now detailed in the caption of Table 7: "Comparisons of simulated PM_{10}, PM_1 and OM_1 daily concentrations to observations (concentrations and RMSE are in $\mu g\ m^{-3}$) during the summer campaign periods of 2012 (between 09 June and 03 July) and 2013 (between 07 June and 03 August)." We used a normalized RMSE rather than the RMSE to show the sensitivity of the different inputs in order to be able to compare the sensitivity of different pollutants which have different concentrations. For a pollutant with a relatively low concentration, the RMSE can be low but the sensitivity can be high compared to another pollutant with a higher concentrations. Normalizing the RMSE allows us to overcome this problem and to compare the sensitivity of the different pollutants.

10) As Table 7 shows, majority of the sensitivity simulation target 2013. Therefore, please also show the composition of PM in 2013 too to assist the discussions.
In Figure 3, the compositions of PM_{10} and PM_1 for 2012 are replaced by the composition for 2013.

11) Add that these are observed composition in Figure 3 caption.

We added in the revised paper and in the text that Figure 3 shows the simulated compositions of PM_{10} and PM_1.

" presented above. Figure 3 shows the simulated composition of …" and in the caption of Figure 3.

12) What does the 0.04±0.03 show in the Table 8?

This was a mistake. 0.04±0.03 is now removed from Table 8 in the revised version.

13) Page 15, Line 2. Normalized RMSE varies between 44 to 267%, not 48?
Yes, therefore 44% is replaced by 48 % in the revised paper.

Conclusions

What is the general conclusion of the study?

Particles are made of different compounds. The particle sources and the parameters influencing the concentrations are different depending on the compounds. These sources and parameters are identified here through 4 main conclusions for the different compounds/parameters.
The following sentence is added at the beginning of the conclusion section to clarify the aim of the study: "This work presents a sensitivity study to different input data and model parameterizations to better understand aerosol sources over the Mediterranean and the parameters influencing the aerosol concentrations. Aerosol sources are different depending on the aerosol chemical compounds. Comparisons to observations are performed at the ERSA station to estimate how realistic are the concentrations simulated with the different parameters. "
The conclusion was also rewritten to clarify the sources and the parameters influencing the concentrations and to better show the 4 main conclusion points:

- Sulfate originates mostly from maritime traffic. Furthermore, maritime traffic leads to the formation of oxidants that in turn enhance the formation of biogenic aerosols, with the potential formation of organic nitrate and organo sulfate.

- Organics are mostly from a biogenic origins. Even if the contribution of sea-salt emissions to organic concentrations is low, organic concentrations are strongly influenced by sea-salt emissions, because they partition between the gas and the particle phases and they are hydrophilic. This underlines the need to better characterize the properties (affinity with water) of secondary organic aerosols.
- Secondary pollutants, such as nitrate, ammonium and chloride, as the particle-phase concentrations are strongly influenced by the gas/particle phase partitioning, because a high percentage of their concentration is in the gas phase. This underlines the need to develop aerosol models able to represent accurately this gas-phase partitioning.
- There is a high sensitivity of secondary pollutants (inorganics and organics) to meteorology, stressing the importance of accurate meteorological modeling and the potential strong influence of climate change on the concentrations of these secondary pollutants.
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