Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2017-697-SC1, 2017

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulation of fine organic aerosols in the western Mediterranean area during the ChArMEx 2013 summer campaign" *by* Arineh Cholakian et al.

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Received and published: 12 December 2017

Dear editor, dear referee,

In this reply, we would like to respond to some of the fundamental issues raised by one referee (the other being positive about the paper requesting only minor modifications) on novelty and also similarities to the paper by Chrit et al.. We thank the referee for initiating this discussion, but we hope and think being able to show the originality of our work. We will respond to the detailed remarks of both referees in a later stage. The referee's statements are numbered and are in quotes.

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Discussion paper

1 – “In addition, the authors do not give appropriate credit of the work of Chrit et al. (2017) who simulated this same data, using a different approach. The authors indicate that the concentrations and properties are well simulated – so this calls into question the need for the current paper. What does this new study tell us that the work of Chrit et al. (2017) did not?”

Although the two papers were based on exploiting results from the same measurements campaign, there are fundamental differences between them. Apart from obvious differences like different models with different inputs, the approach taken in the two articles regarding the simulation of organic aerosols is quite different. In our article, we use the VBS scheme with modifications that take into account formation of non-volatile SOA and fragmentation processes. In Chrit et al. (2017), they use a surrogate based approach where SOA are divided into three types, hydrophilic, hydrophobic or both (Couvidat et Sartelet 2015) with modifications added to take into account ELVOCs. In our mind, it is important to compare all these different schemes to detailed measurement data and not limit this comparison to one particular scheme.

Our paper goes significantly beyond the Chrit et al. (2017) paper. In our work, we use in addition to data from Cap Corsica measurements from a second site at Mallorca for the same period. The two sites have different characteristics as it is seen in the results of 14C measurements and PMF results, the one at Mallorca being more strongly influenced by anthropogenic emissions. Since the goal of the article was to evaluate the performance of different SOA simulation schemes in the western Mediterranean area, it is more representative to take into account campaign data from the two sites. Measurements used for Mallorca station have not been compared to simulations in other articles (neither the OA concentration, 14C measurements, nor the PMF results), so this point is clearly novel. In addition, our work provides, in addition to that of OA, a detailed comparison of meteorological inputs, SOA precursors (isoprene and mono-terpenes) and intermediate compounds (MVK+MAC) for both sites and thus goes beyond Chrit et al. (2017).

For the Cap Corsica station, because of its unique geographical characteristics not well represented by CTMs, an orographical representativeness error study is performed in our work, which is actually sited in Chris et al. (2017) to indicate an error for organic aerosol (Chrit et al. 2017, Page 7, section 4, sub-section 2). The novelty of this approach is discussed in more detail below.

A source apportionment study for the western Mediterranean basin at two different altitudes (surface and 300-450m altitutes) is also included in our study, which was not included in Chrit et al. (2017). This study shows the impact of biogenic and anthropogenic sources for the formation of OA not only on the surface, but also for a higher altitude.

As a side note, the two articles were written at almost the same time period. While Chrit et al. (2017) was accepted for publication on 23 October 2017, this article was submitted for discussion on 26 august 2017.

2 – "As a result of these facts, I do not find this manuscript very novel."

Here, we wish to show that our work is indeed novel and original in several important aspects.

The Cap Corsica site has unique geographical characteristics which raises issues when comparing simulations to measurements. Indeed, it is located on the northerly edge of a crest and surrounded from the west, north and east by slopes falling rapidly to the sea. Its altitude is at 530 m, while in CHIMERE simulations, even at a 1 km horizontal resolution, it is represented at 360 m height. This height difference might induce differences between simulated and observed concentrations, which needs to be assessed, especially in a marine environment with low boundary layer heights. Therefore, a novel approach was developed in this work to calculate an orographic representativeness error, based on comparing simulated concentrations at horizontally very close grid cells located at different altitudes (from 100 to 500 m). The method allows attributing representativeness error bars to different species, large for primary

compounds especially emitted in the marine boundary layer like NOx and BC, smaller for (mainly) secondary compounds as O3 and OA, which are compared to simulation observation differences. As far as to our knowledge, this approach was not taken anywhere else for the estimation of errors produced by orographic representativeness of a site. This method can be applied to other sites with the same characteristics (high altitude remote sites with strong change of altitude compared to nearby terrain). The results obtained from this approach are also cited in Chrit et al 2017 (Page 7, section 4, sub-section 2). We would argue that this development, while not the primary goal of the paper, is clearly novel.

For the eastern Mediterranean area, an abundance of literature is available both for chemical composition (Bardouki et al., 2003 ; Koçak et al., 2007b ; Koulouri et al., 2008 ; Hildebrandt et al., 2010) and also for OA comparisons to simulations (Fountoukis et al., 2014). However, for the western part of the basin, much less studies exist for the chemical composition of the area (for example Querol et al., 2009 ; Pey et al., 2013), but these data have not been used to the best of our knowledge specifically for CTM model evaluation. Thus, detailed model -observation comparisons of different aspects of OA (total concentrations, oxidation states, modern/fossil fractions from 14C) including comparisons of precursors, intermediary compounds, meteorological conditions and other gaseous/particulate species for two different sites, as presented in our paper, have not been performed before for the western Mediterranean area. More limited comparisons for OA and for one site were done by Chrit et al. (2017); however the comparisons for the Mallorca station are not used in any other articles, therefore are clearly new. Also, articles comparing different SOA simulation schemes have not yet focused on this western part of the basin with its particular set-up and processing different emission sources including continental, coastal, and marine anthropogenic and biogenic emissions.

Source apportionment for the western Mediterranean area is also a subject that has not been much explored before. Of course, source apportionment measurements have

been presented for different sites of the Mediterranean basin (Koçak et al., 2007 ; Querol et al., 2009 ; Minguillón et al., 2011 ; Pey et al., 2013) and also for the European area there are studies that discuss the simulated effects of biogenic emissions (Sartelet et al., 2012) on OA, but none of them give the results of the simulated source apportionment of OA for the whole western basin. This part of our work, answers one of the principal questions raised by the ChArMEx campaign, with the goal of exploring the source apportionment of the OA over the western Mediterranean area. We think that it makes an important contribution to the ACP /AMT special ChArMEx section.

3 – "However, no mention of cloud processing in the aerosol modules is mentioned. Why not link the updated VBS with a cloud processing module?"

While adding cloud processing modules to the model is certainly a good and novel idea, it would be a completely different work apart from the questions that this work seeks to answer to.

4 – "However, these types of evaluations were done during development of those models."

Actually, these schemes were not tested and compared to measurements for the Mediterranean area. They were tested in the US (Robinson et al., 2007 ; Lane et al., 2008), different parts of Europe (Petetin et al., 2014 ; Zhang et al., 2013 ; Fountoukis et al., 2014) and in South America (Shrivastava et al., 2011 ; Hodzic and Jimenez, 2011 ; Shrivastava et al., 2013 ; Shrivastava et al., 2015). Only in Chrit et al (2017) a scheme for the simulation of organic aerosols is tested for the western Mediterranean, but the evaluated scheme is different from the four evaluated schemes in our work (mainly VBS derived schemes in our work versus a one-step surrogate scheme in Chrit et al. 2017).

Concluding remarks:

As a conclusion, our work answers some of the key questions raised by the ChArMEx

project such as evaluation of a CTMs and different organic aerosol schemes and source apportionment of OA in the western Mediterranean basin. It uses for this the wealth of experimental data available from the ChArMEx campaign. We think that these results are novel and important for the valorization of the project. In addition, one of the far reaching goals of the ChArMEx project is to evaluate the modifications in atmospheric composition induced by climate change in this region. Future studies dealing with this issue could build on results of our current study, in terms of evaluation of the CHIMERE and the different organic aerosol schemes. In a second reply and revised version, we will make more ample references to Chrit et al. (2017), which is now published. We will better insist on the novel aspects of our paper as argued in this reply and certainly respond to specific and technical points mentioned in both reviews.

Response written by : Arineh Cholakian et Matthias Beekmann

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2017-697, 2017.