Interactive comment to Anonymous Referee #1 and Referee #2 on "Comparison Of Two Different Sea-Salt Aerosol Schemes as Implemented in Air Quality Models Applied To The Mediterranean Basin" by P. Jiménez-Guerrero et al.

The authors' gratefully acknowledge the comments of the Anonymous Referee #1 and #2, which may help to improve the quality of this manuscript and to clarify some issues that were not previously addressed.

In this sense, the manuscript has been revised after the referees' comments in order to correct errors and omissions, and to introduce the previous suggestions for improving the quality of the paper. A revision of the manuscript has already been sent to the Editorial Office for editing.

We attach an item-by-item response following to all the relevant statements of the referees.

Referee #1: […] The methods that are applied are not scientifically sound, it lacks of convincing formulations and analysis of the results. Referee #2: […] The paper is well structured and presented the current status of sea-salt simulations in atmospheric models.

First, we would like to thank the referees for the time devoted to the revision of the manuscript. In this response we will try to fix the limitations and drawbacks reported by the referees.

Referee #1: […] Equation 3 should report the formulation given in Monahan et al (1986), however the reported expression is not the formula included in Monahan publication. The original formulation is

$$dF/dr = 1.373U^{3.41}r^{-3}(1+0.057r^{1.05})x10^{1.19e(-B2)}$$

[…] dF/dr is the number of aerosol droplets per unit area of the sea surface per increment of droplet radius and not aerosol mass. Furthermore r is not the radius of the bubble at formation but the aerosol droplet radius as reported by Monahan. […] It is clearly said that the reported formulation is for "open-ocean sea-surface aerosol generation" and not from "... oceanic whitecaps along the coast".

The authors strongly apologise for the typos included. The sea salt emission functions follow the original Monahan formulation and there was a mistake in the constants considered and the description of Monahan et al. formulation when copy-editing the manuscript for LaTEX submission to the ACPD journal. All these typos and errata have been corrected in the revised manuscript. We thank the referee for identifying this mistake.

At which relative humidity the radius of the equation 3 are taken? How this has been changed to make it compatible to the nature of the aerosol particles described in CHIMERE in terms of the actual relative humidity?

The radius has been taken at 80% relative humidity. Also, this Monahan et al. (1968) scheme is valid at 80% relative humidity. To generalize it, it is expressed in terms of dry radius, which is assumed to be approximatively half the radius at 80% humidity (Gerber, 1985). For further description on the implementation of this scheme within CHIMERE, the reader is referred to Bessagnet et al. (2009).

These comments have been introduced in the manuscript.

Referee #1: […] In the following formulas Co does not appear

The definition of C^0 has been introduced in the manuscript after the reviewer suggestion.

Referee #1: it is not clear from the equation 10 of Zhang et al, how the expression 6 in this manuscript comes from. It is not clear what is the relative humidity RH, what is the relationship between the relative humidity at which the original formula of Gong is given and the relative humidity RH. How this function has been used to make it compatible with the description of aerosols in CMAQ which is modal?

Expression 6 in this manuscript comes from a revised form of eq. 10 in Zhang et al. (2005) that Zhang and co-workers derive in a reply to some comments to his paper in Atmospheric Environment (Zhang et al., 2006, Atmos. Environ. 40, 591-592), where they relate these relative humidities the referee #1 is referring to. So equation 2 of Zhang et al. (2006) correspond to the expression 6 in our manuscript. With respect to the description of model CMAQ aerosols, the reader is referred to Kelly et al. (2010). A full description of the model approach within CMAQ has not been introduced for keeping the brevity of the manuscript.

All these comments have been introduced in the paper in order to improve the clarity of the work.

Referee #1: […] the sea salt component in the aerosol at Finokalia is not at all the major component […] Furthermore it SSA may not be the major component either for Lampedusa nor for Oristano where dust outburst from Sahara can reach the islands.

After a careful revision of the assumptions made, we fully agree with the reviewer that SSA may not be the principal contributor to AOD in the Mediterranean area. Therefore, following the referee's advice, we have included all the chemical components of the aerosol fraction in the estimation of AOD from the model and compared them to the measurements from the Aeronet stations. The results obtained do not differ much from those presented before, since they all indicate an accurate behaviour of the model for reproducing the seasonal variations of AOD levels in the Mediterranean for both models.

Referee #1: Sea salt particles are very hygroscopic therefore the amount of water attached to the particles cannot be disregarded as the formula may suggests. Therefore the calculation of the modeled SSA AOD is not done correctly.

As commented before, the calculation of the modelled AOD has been re-done in order to introduce all components. Furthermore, corrections for the RH have been introduced in the estimation of AOD as advised by Referee #1. It has been clearly stated in the manuscript as follows:

> "*The mass concentration for each species is directly obtained from CHIMERE and CMAQ. A relative humidity correction factor (Tang et al., 1981; Tang, 1996) takes into account that the growth and phase change of hygroscopic particles affect their light-scattering efficiency (Malm et al., 1994). The factor f(RH) is parameterized from data published by Tang et al. (1981) as a function of the relative humidity, taken from the WRF-ARW meteorological model, f(RH) varies between 1 (at low RH) and 21 (at RH=99%).*"

Reviewer #1: Finally the structure of the manuscript is confusing. Formulations of sea salt function as well as deposition parameterisations are described only after the evaluation of the model results.

Following the reviewer's suggestions, the manuscript has been re-written and the structure of the manuscript changed in order to present the parameterisations before the evaluation of the model results.

Referee #2: […] If an over-estimate of surface wind was predicted in the model, one would expect an over-estimate of sea-salt fluxes from the sea-salt emission schemes, which should result in an overestimates of the surface concentrations of sea-salt aerosols, which the models failed to predict [,,,] […]Both dry and wet depositions were analyzed from the models. However, it seems the analysis did not provide any information about the model performance with respect to the inconsistency from point (1) above.

The referee raises a very interesting aspect of the study. However, as a first guess, there is not a direct relationship between an overestimation of the wind and an overestimation of the fluxes. Even if we do not take into account the rest of the processes involved in a model, the sea salt fluxes respond to certain parameterizations and empirical or semi-empirical relationships that may not accurately represent the sea salt fluxes for the conditions studied in this work. In other words, overestimating the wind speed does not necessarily lead to an overestimation of the flux, estimated, for instance, as

$$dF/dr = 1.373U^{3.41}r^{-3}(1+0.057r^{1.05})\times 10^{1.19e(-B2)}$$

Moreover, the slight underestimations observed for SSA in both models can be caused by a large number of circumstances: errors in the prediction of the mixing height (leading to a larger dilution), problems with the horizontal or vertical discretization of the model, overestimations in the dry and wet deposition (which is hard to evaluate over the Mediterranean Sea), misrepresentations of the aerosol radius, uncertainties in the settling velocitites… Therefore, it becomes nearly impossible to isolate one determined process as causing these negative biases for the models.

Hence, in this comparison we have tried to find the global causes for the differences between schemes as implemented in a CTM, not between the schemes themselves (here, a box-model would be much more useful to compare theoretical schemes).

Referee #1: […] Measurements of SS concentrations are used for a statistical analysis of the modes' results, but they are not used to discuss the ability of the model in terms of spatial or seasonal patterns; Referee #2: Another concern for this paper is the lack of evaluation of the model performance for simulating sea-salt aerosol in terms of size distributions before its application to the calculation of AOD […].

As stated in the definition of the objectives of the manuscript, the main goal of the paper is not to provide a comprehensive evaluation of the model behavior, such as done in other works like Manders et al. (2010) and Pay et al. (2010) in Atmospheric Environment, or the recently paper published in ACPD by Tsyro et al., Atmos. Chem. Phys. Discuss., 11, 11143–11204 (2011). We should bear in mind that our main objective is to highlight how different sea-salt schemes, implemented in different CTMs, intercompare when applied to the Mediterranean area.

A detailed evaluation of the CALIOPE system for reproducing PM levels over Europe can be found in Pay et al. (2010) and we have considered that it would be redundant to present such a detailed evaluation in this manuscript. Nonetheless, we agree with the referees that there is a need to assure the correct performance of the models in a paper devoted (as we have done in the evaluation section of the manuscript). An evaluation devoted to sea-salt aerosol would deserve an entire paper on that topic (such as the works previously mentioned).

However, we fully agree with the need for a more detailed evaluation of the spatio-temporal patterns of the models. What's more: the suggestions of the referees led us to develop an evaluation with a deeper detail. Hence, a paper in preparation by Basart et al. entitled "Aerosols in the CALIOPE-EU air quality modelling system: validation and analysis of PM levels, optical depths and chemical composition over Europe" will be submitted shortly for peer review, presenting a largest validation of AOD and PM levels all over Europe for the CALIOPE system.

All these comments have been introduced in the manuscript in order to clarify the evaluation section. In addition, following the comments of the referees, the main findings of Basart and Pay works have been included in the section devoted to the model evaluation to complement the present findings of this work.

Referee #2: The Summary and Conclusion needs more scientific founding on the model inter-comparison […] What have we learnt?

The authors fully agree on the referee #2 comments; and hence this section has been modified in the revised version of the manuscript in order to try to highlight the most sounding points of the results.

Thank you for considering this manuscript for publication in Atmospheric Chemistry and Physics.

Dr. Pedro Jiménez-Guerrero (on behalf of all the co-authors)
University of Murcia, Spain.
Ed. CIOyN, Campus de Espinardo, Of. 1.21, 30100 Murcia (Spain)
pedro.jimenezguerrero@um.es