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Interactive comment on "Modelling of primary aerosols in the chemical transport model MOCAGE: development and evaluation of aerosol physical parameterizations" *by* B. Sič et al.

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First of all, we would like to thank to the reviewer for a thorough review. We appreciated very constructive comments and suggestions. We have revised the text in the light of given remarks and suggestions. We would like to point out that in the response to the comments of the second reviewer we added to the manuscript figures showing the comparison against in-situ surface measurements, and the horizontal and vertical distribution of aerosols in the model in SIM1 and SIM2.

Number-wise answers to the reviewer's specific comments/suggestions/remarks are as follows.

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Specific Comments:

1) P2751L19-22: Could you include a note here about how the water uptake on the aerosol is considered?

c: In the revised manuscript, we added the description of the water take-up of the sea salt in the model.

2) P2752L7: Please specify if dry or wet diameter is used in the equation. Similarly, in the discussion of the below-cloud scavenging parameterization, please specify if wet or dry diameter is used. Also, perhaps I missed this, but was there a description of the dry deposition scheme used in the model?

c: We specified in the revised text that we consider the ambient (wet) diameter. Also, we added the more detailed description of dry depositon in the model.

3) P2753L10: I assumed here that there are separate calculations for both in-cloud and below-cloud scavenging as opposed to only one coefficient, but this could be more clearly indicated.

c: We clarified in the revised text that the scavenging coefficients consists of more components, and we labeled them accordingly.

4) P2753L23: Is Q a grid-mean value and is L_{st} an in-cloud value? Please specify? Also does Q include both liquid and ice or only liquid? Is there any temperature dependence to the in-cloud scavenging?

c: That's correct, Q is a grid mean value, and includes both liquid and solid precipitation. L_{st} is the in-cloud value. We made clarifications in the text. In the scheme itself there is no temperature dependence, but Q does depend, and that is considered in our precipitation calculations.

5) Section 3.2: I was not clear about the model's previous parameterization for the fraction of precipitating cloud cover that was used in the SIM1. I assume this section

described the parameterization for SIM2.

c: Yes, the described parameterization for the precipitating cloud fraction applies to SIM2. In the text we revised the description of the precipitation cloudiness calculations in the model. We explained in more detailes the calculations and the differences between SIM1 and SIM2.

6) P2754L15: By 'scavenging coefficient', do you mean the below-cloud or in-cloud scavenging coefficient or both?

c: It includes all components of the scavenging coefficient Λ. This is now clarified in the text.

7) P2754L24: Is Q in this equation the same as Q in Eq. 5? I think that Q in Eq. 5 is the grid mean Q based on Giorgi and Chameides (1986). However, based on the definition of L following Eq. 7, would this Q in Eq. 7 be an in-cloud value? In this equation, I would expect that both Q and L are grid-mean or both are in-cloud values, please specify this more clearly?

c: That's correct. The values has to be or both in-cloud or grid mean. Q in Eq. 7 is the same Q as in Eq. 5 and both Q and L are grid mean values. All this is now clarified in the text.

8) P2755L1: Could you mention the assumed values for the scavenging efficiency for uptake into the cloud droplets for the species considered in the model that would be used in Eq. 7?
9) P2755L4: Please check this line, do you mean that sulfate is considered as insoluble? This seems strange. Also, are all carbonaceous aerosols treated as soluble species for the purposes of wet deposition in the model? Is all dust also treated as soluble?

8/9 c: Due to a typographic error, the solubility of the species is reversed in the text. Kasper-Giebl et al. (2000) measured 'aerosol carbon' and sulphates, which are con-

sidered as insoluble and soluble species, respectively. We corrected the error in our manuscript and added the values for the scavenging efficiencies derived from the Kasper-Giebl et al. (2000) study.

10) P2755L15: The text is not clear here about the precipitation fraction used for the purposes of below-cloud scavenging. Is this based on the maximum fraction from overlying layers? This paragraph should also be clearer about the parameterization for the current and the revised model. Would introducing the names of the two primary simulations (SIM1 and SIM2) earlier in the text be helpful in making this description more readable?

c: We changed the description where below-scavenging acts. This should lead to an easier understanding of the text. Also, the references to the SIM1 and SIM2 configurations are added earlier in the text, and they are referred in whole Sect. 3. This should lead to a clearer image of the described model changes throughout the text.

11) P2757L5: Is this raindrop diameter or radius?

c: It is the diameter. We clarified this in the manuscript.

12) P2757L7: I was not clear here about when the exponential raindrop distribution is used. Is this for SIM2 and the standard model (SIM1) assumes the fixed size? I think this is the case from Table 2, but this is not clear in the text.

c: The change done for comment 10 also made clearer this point.

13) P2757L13: Is it possible to directly compare a field measurement and a theoretical value for the scavenging coefficient due to confounding dynamical factors?

c: We were not completely sure that we well understood this question. Especially what the reviewer meant by the confounding dynamical factors. If the remark was that it is difficult to make a direct comparison of observed and parameterized values, because of the incertainties in the both values of the scavenging coefficient, we agree with it. Our intention was to follow indications by different studies that also the phoretic and

electric effects could be important in the scavenging. They could be added to the semi-empiric parameterisation from Slinn (1977) that we used. To investigate what that would bring to the results of our model, we made an enlarged SIM2 configuration with also implemented the mentioned effects. In the end our findings were that these effects did not change significantly the performance of our model.

14) Section 3.2.4: Were you able to isolate the impact of the addition of evaporation between SIM1 and SIM2? Can you comment on this?

c: The return of the re-evaporation is 10% of aerosols back. In the revised paper, we comment on this, and we compared and comment on the influence of the re-evaporation compared to the influence of the change of the precipitating cloud cover.

15) Section 3.3: Are the emissions of carbonaceous aerosols changed between SIM1 and SIM2? This looks to be the case from Table 4 but I was not clear about the changes when reading this section.

c: Thank you to note this. The participation in ACCMIP project lead to introduction of ACCMIP emissions in the use into the model. In the first version of manuscript, we overlooked that for black and organic carbon in the SIM1 we used the AeroCom emissions. We added the description of AeroCom emissions in Sect. 3 and make clearer what we use for each simulation.

16) P2762L5: The text mentions that hygroscopic growth affects optical properties, sedimentation and dry deposition. Please state clearly if this Eq. 20 is the equation used in the model to obtain the wet radius for all aerosols for optical properties and deposition and including also for below-cloud scavenging.

c: Yes it is, Eq. 20 is the equation which we use to obtain the wet diameter for hydrofilic aerosols and we clarified this in the revised manuscript.

17) P2763L5: Are you able to comment on how this change in interpolation influenced the wind fields? Why did you choose to make this change? A comparison of the pre-

cipitation fields with GPCP is shown, but are you able to comment on the comparison with observations for the wind fields used? Also the text could be clearer here about which interpolation method used for SIM1 and SIM2, respectively.

c: The change of interpolation method for the wind field influenced the desert dust emission considerably. We suppose that it is linked with the effect of the wind threshold velocity, where the emission does not depend linearly on the wind, and where the small changes can have a big impact on the emitted quantities. This is the case because, with the achieving of the threshold, the emissions are triggeres and already a considerable amount of dust can be emitted. The change in the model was motivated by the possible impact on the Asian dust sources, where we expected that because of quite sharp orographic changes in the region and the model relatively coarse resolution, the wind interpolation method could have an impact on the emissions. In the end, the change for the Saharan desert dust sources was as important as for the Asian deserts. Also, in the revised text, the model wind fields are compared with the QuikSCAT satellite measurements. The comparison figure is added to the manuscript and commented.

18) Section 4: Do you filter the model data using the same criteria as applied to the MODIS data? Also do you sample the model exactly at the satellite overpass times?

c: The model data that are used for the comparison are sampled only at the location of the available MODIS data. The MODIS data is the level 3, combined Aqua and Terra data, where a unique overpass time is impossible to apply. The model data is taken at 12UT for each day.

19) P2764L8: Is there any filtering process applied to SEVIRI data?

c: We performed SEVIRI filtering as it is recommended by Carrer et al. (2010) and this is now explained in the text.

20) P2765L14: Can you quantify what you mean by 'significant improvement'?

c: We quantified these improvements in the text by adding some statistics in the new

version of the paper.

21) Figure 1: I found the colors on the right side panels to be counter-intuitive. Would it be better to show (Model – MODIS), then a negative bias as mentioned in P2765L27 would appear as a negative number on the figure.

c: We considered this remark and changed the colors in this figure by subtracting now the model from the observations.

22) Can you explain why you chose to present 'bias' as your metric opposed to something else such as fractional mean bias, which also allows for error in the observations, as outlined in Boylan and Russell (2006), Atmos.Environ.? Please explicitly provide the definition for your bias metric and perhaps consider presenting an alternative metric. Could a global metric be included perhaps on Fig. 1?

c: Statistics are changed in figures and tables. Instead of bias we use modified normalized mean bias (mnmb), and instead of root mean square error we use fractional gross error (fge). Appendix is added which explains all used statistical indicators. The global metrics for MODIS data are available in Fig 2 (figure number in the original manuscript).

23) P2768L12: Table 4 seems to indicate that the SIM2 burden does not correspond as well to the AeroCom mean as for SIM1, please check this sentence.

c: We corrected this sentence.

24) P2768L15: Do you mean that the sea salt lifetime shows improved agreement with the AeroCom mean, despite the poorer agreement with the burden? Could the text provide information at some point about how the sea salt mass is distributed differently among the 6 size bins between SIM1 and SIM2 and how this impacts the various scavenging processes?

c: We made these sentences clearer. In the model output we do not expect that the model updates of wet deposition and sedimentation affect the mass between bins. The in-cloud scavenging schemes in SIM1 and SIM2 are not size dependent. The below-

cloud scavenging scheme from Slinn (1977) is used in SIM1 and SIM2 with different subcomponents, but the size dependency is not altered. The mass between bins in our changes is significantly changed only by the changes in the emissions by the way how we defined them. For these reasons, we did not look closely to the changes in the mass between bins in the different simulations.

25) P2769L2: Is this 22

c: This is the decrease in total, taking into account all species.

26) P2769L27: The text is rather vague about increases and decreases depending on location, could you summarize the most relevant changes more explicitly?
27) P2769L29: There is mention of 'high precipitating cloud fraction' in SIM2_WDEP, but this is not explicitly presented. How much difference was there in this fraction between the simulations? Also were you able to isolate whether this cloud fraction or the addition of re-evaporation had a greater impact on your results?
28) Figure 6b, thinking of the above point and the related discussion of this figure here, would you be able to color-code the points in the scatter plot somehow to indicate the tropical regions where evaporation is expected to be more important?

c26,c27,c28 c: We added and explicitly explained the differences between SIM2 and SIM_WDEP simulations between the Tropical Pacific where re-evaporation should be important and the South Pacific where changes in cloud cover should be more important. Also these two regions are represented in a subgraph added to Fig. 6b (figure number in the original manuscript), where we can see the differences between effects on different regions. As previously discussed the effects of re-evaporation and changes in cloud cover and wet deposition calculations have different effects in these regions, and now this is highligthed in the graph. Also, we commented on the degrees of changes of re-evaporation and the cloud cover.

29) P2770L23: Dust overestimation found by Zhang et al. (2012) does not seem consistent with your findings of underestimation. Please check this. Also here in regard

to the Jaegle et al. (2011) paper is the sea salt underestimation value quoted for the tropics or global?

c: When comparing our findings with the findings obtained by other models, we were mainly emphasizing the degree of the differences (their absolute value) between models and observations. In this sense, the results from Zhang et al. (2012) vary to a similar degree as found in MOCAGE in this study. The quoted difference in the paper of Jaegle at al. is a global value. We changed text to make these points clearer.

30) P2771L15: Could the apportioning between the bins also contribute to this discrepancy? Could the text more explicitly discuss how this is different between the different simulations and how this influenced the results for dust and sea salt in particular.

c: Yes, the effects on the bin apportioning in the model were explored by Martet et al. (2009), and the ranges of the bins were chosen so that are adapted to the efficiency of the sinks. We do not believe that it is the strongest effect in the explanation of the differences. Additionally, we also explored another possibility, that the introduction of secondary aerosols and sea salt chemical reactions could also change the quantities of sea salt in the model. The secondary aerosol module is in the process of implementation in the model. The first tests showed that the dechlorination could be particularly important effect on the sea salt burden (and lifetimes) in the model.

31) P2772L3: "longer mean atmospheric residence time" relative to what? Please specify.

c: We specified in the revised manuscript that it is compared to the AeroCom average.

32) P2772L13: "complex and balanced". . . are there any relevant patterns that you could mention here?

c: We specified some of the results mentioned in Sect. 7.

33) P2773L19-22: Please quantify what you mean by 'significantly improve' and 'correspond better'.

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c: We added the references to the tables where the data is available and which confirm the statements.

34) Table 4: Why is OC missing from the table?

c: In AeroCom data we find POM (particulate organic matter), and it is suggested that it can be converted to OC by using a fixed presumed POM/OC ratio. We decided not to put OC in this table because, first, we believe that the AeroCom OC that we could calculate from POM and the fixed presumed POM/OC ratio, does not reflect exactly the ACCMIP OC. POM in the AeroCom models used to calculate median values in the model inter-comparison, are treated in variety of different ways, and the fixed presumed POM/OC ratio does not necessarily represent well the models' POM/OC relation. Second, we presume that the BC comparison reflect well the difference in the model performance of the carbonaceous aerosols in SIM1 and SIM2.

35) Figure 2 and 6: Could any color-coding be added to the scatter plots to provide more information, perhaps by geographic region?

c: The colored regions are added to Fig 6b (original manuscript) as an addition. Otherwise, the division of the whole field into regions and its coloring would impact the readability of the plots.

36) Figure 5: Why was only one day chosen? Were than any other days when the primary aerosols were dominant that could make the comparison more robust?

c: The SEVIRI dataset used for the creation of the presented one-day plot, covered a limited time period during the considered year 2007. Other days did not clearly show the dominance of the primary aerosols.

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