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Atmospheric Chemistry and Physics Discussions

Interactive comment on "An integrated modeling study on the effects of mineral dust and sea salt particles on clouds and precipitation" *by* S. Solomos et al.

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Response to Anonymous Referee #3

General Comments: This manuscript reports a study on aerosol (specifically, mineral dust and sea salt particles) feed back to cloud dynamics and microphysics using ICLAMS (an integrated community limited area modeling system). It is an area that is gathering a lot of interest in the atmospheric science community. Through numerous sensitivity studies the authors demonstrated the importance (or sensitivity) of the interaction between aerosol activation and cloud microphysics and dynamics on the model

predicted precipitation. However there is a lack of clarity in the description of model representation of some of the key processes (particularly aerosol microphysics) and in the experiment design and setup which need to be addressed before the manuscript can be published. For example, it is not clear how aerosol particles are represented in the model: aerosol size - sectional (binned) or modal representation, aerosol mixing state - internally mixed or externally mixed (between mineral dust and sea salt, and other chemical components), processes - aerosol microphysics (condensation/evaporation, coagulation, other than the sedimentation and dry/wet deposition processes described) and secondary aerosol formation/production. It is not clear what aerosol properties are imposed and what are simulated for the various sensitivity tests. The following are some specific comments.

[REPLY] We would like to thank the reviewer for very thorough and thoughtful comments that have substantially improved the manuscript. Our responses to the issues raised are provided below.

It is not clear how aerosol particles are represented in the model: aerosol size - sectional (binned) or modal representation.

[REPLY] As mentioned in the text (page 6 lines 21-23, page 7 lines 25-26, p8 lines 1-2), dust particles are represented with eight size bins (similar to Perez et al., 2006 and Spyrou et al., 2010). Sea salt particles are represented with a bimodal lognormal distribution. In the revised manuscript these sections have been moved to appendix (lines 546-549, 570-573).

Aerosol mixing state. Internally mixed or externally mixed (between mineral dust and sea salt, and other chemical components)

[REPLY] The particles are assumed to be externally mixed and interaction between dust and salt is not treated explicitly. However, for the CCN activation scenarios we implicitly assumed various percentages of internally mixed dust / salt aerosol. This point has been made clear in the revised version.

Aerosol microphysics (condensation/evaporation, coagulation, other than the sedimentation and dry/wet deposition processes described) and secondary aerosol formation/production.

[REPLY] The present model version includes aerosol production, dry and wet deposition, CCN activation and aerosol radiative effects. Evaporation and coagulation are not parameterized in the model.

It is not clear what aerosol properties are imposed and what are simulated for the various sensitivity tests.

[REPLY] For the idealized test cases all aerosol properties are imposed. For the MEIDEX case study dust and salt mass concentrations are prognosed. Number concentrations are diagnosed from the size distribution of the particles. For sea salt, the mean diameter and geometric dispersion were fixed. For dust particles, these properties were estimated based on the relevant concentrations of the eight dust bins. This point has been made clear in the revised manuscript.

Following are the specific corrections included in the revised version of the manuscript:

Specific comments:

2. Description of ICLAMS

Is this the first publication of ICLAMS? In Table 1 the authors have highlighted the model components added to RAMS, but not all of them are given description here. Are there references for those new components that are not described, e.g., on-line multi-phase chemistry and aerosol parameterizations?

[REPLY] This is the first publication of ICLAMS. Due to limitations on the length of the manuscript some model components mostly related to atmospheric chemistry will be discussed in a subsequent publication. However they are mentioned in this table for completeness.

P5, line 135: What do you mean by “cycle” here?

[REPLY] “Cycle” here implies the production, transport and deposition processes of pollutants. This sentence has been rephrased for clarity.

2.1 Mineral dust P5, line 152-153: This seems to imply that a single landuse class is assigned to a given model grid (as opposed to multiple landuse classes with fractional coverage). Is this the case?

[REPLY] The landuse classes in RAMS/ICLAMS are assigned to fractional divisions (patches) inside each grid cell. Dust production and dry deposition processes are calculated on patch scale and then integrated over each grid cell.

P6, line 166-167: What are the three modes, and what are the median diameters and geometric standard deviations for these modes? What is the “transport mode”? Is this the only model represented in the model?

[REPLY] The three modes used are the background dust modes suggested by D’Almeida, (1987) and described also in Zender et al., 2003. The relevant references have been added in the text. The transport mode represents the particles that can be transported to long distances. This mode is represented inside the model with eight size bins similar to Perez et al., 2006a and Spyrou et al., 2010.

2.2 Sea salt spray P7, line 199 – 202: Is this bi-modal distribution of sea salt particles mapped onto the same 8-bin structure as in the case of dust particles? How is the particle mixing state represented in the model (e.g., externally mixed, internally mixed, or something else)?

[REPLY] Sea salt is treated with two mass bins inside the model, each one representing a lognormal mode. The particles are externally mixed and there is no explicit interaction between dust and salt. However, for the case study over Eastern Mediterranean several scenarios of internally mixed aerosol were considered in a parameterized way. Specifically, we assumed various percentages of dust particles to contain soluble material (sea salt) in order to represent different stages of aerosol ageing. This point has been clarified in the revised version.

2.4 Wet deposition P8, line 234: Are you talking about in-cloud removal (rain-out) or below-cloud removal (wash-out)? The equations (8 – 10) are for below-cloud scavenging of particles by precipitation.

[REPLY] Both in-cloud and below-cloud removal are treated in the model. The parameterization for in-cloud scavenging (Eq.8) has been adopted from the CAMx model (Environ, 2006), similar to Spyrou et al., 2010. For brevity, the sections referring to dust and salt production and deposition processes have been moved to the appendix in an attempt to limit the length of the manuscript.

2.6 Cloud droplet nucleation parameterization P10, line 297-299: What are the processes included in the calculation of aerosol properties, e.g., hygroscopic growth, uptake of condensable gases, coagulation?

[REPLY] The dry and wet deposition processes are size-dependent. For this reason, the relative concentrations of the eight dust bins vary from place to place. For example, bigger particles exist near sources and at low tropospheric layers while away from sources and at higher levels the dust spectrum is shifted towards smaller particles. The properties of dust particles distribution are estimated based on the prognostic mass concentrations of the eight dust bins at every grid point and timestep. We agree with the reviewer that explicit calculation of the interactions between aerosols and gases is also an important issue. Given the length of this study, it would be very challenging to explore such interactions, and defer them to a future study

P11, line 305-306: Is IN explicitly linked to modelled dust particle concentration in ICLAMS? This is not obvious from the reference given. It would be helpful to provide the actual formulation used as well as references.

[REPLY] The ice nucleation parameterization used in this study is that of Meyers et al.,

(1992). The IN number at the original formulation is considered homogeneous for the entire model domain. In the present version of the model several options have been added to improve this approach. These options include the increase of IN by an order of magnitude at the grid points where dust is present and also considering the total amount of prognosed dust particles as IN. This is still a simplistic approach on the link between IN and dust and we are currently working on a forthcoming version of the model that will include a comprehensive ice nucleation parameterization scheme that considers the competition of homogenous and heterogeneous freezing for water vapor.

3. Clouds and precipitation in an environment with natural particles 3.1 Idealized simulations What kind of lateral boundary condition is used for these idealized tests? For these tests only FNS parameterization is used for aerosol activation. It would be useful to include a run with the original droplet nucleation scheme in RAMS to look at the impact of the new scheme, or has this comparison been done elsewhere?

[REPLY] Zero gradient lateral conditions were used, that is common for this type of applications. The purpose here was mainly to illustrate the role of aerosol on cloud processes. The original RAMS scheme does not include such features. Such comparisons have been performed during preliminary model tests (e.g. Solomos et al., AGU 2008) but we do not consider worth including these results here.

P12, 334: what is the soluble fraction assumed for these two cases?

[REPLY] The soluble fraction was set to 33% for the first mode and 95% for the second.

P12, line 347: the maximum droplet number concentration 130 cm-3 is greater than the total number concentration of dust particles prescribed for this case (100 cm-3), any explanation?

[REPLY] Once cloud droplets are formed they can be transported inside the model domain. Also they can grow to rain sizes and similarly rain droplets can evaporate to produce again cloud droplets. So the number of cloud droplets at each grid point and at each timestep is governed by various other processes apart from the initial CCN number.

P12, line 358: similarly the maximum droplet number concentration 2133 cm-3 is greater than the total number concentration of dust particles prescribed for this case (1500 cm-3),which again seems odd.

[REPLY] See previous comment.

P12, line 359 – 360, and Table 3: the rain mixing ratio at hour 2 for the two cases are 0.47 g/kg for "pristine" and 0.37 g/kg for "hazy". The difference is significant but not huge. In contrast, the difference in rain droplet concentration between the two cases is huge (27.65 L-1 vs. 2.2 L-1). Does this make sense?

[REPLY] These are maximum values and they do not necessarily occur at the same grid point. These values are rather indicative of the difference in the initiation of precipitation between the two simulations.

P13, line 367: figure 5 is redundant. It is simply a graphic presentation of column 3 of Table 3.

[REPLY] Figure 5 has been removed.

P13, line372 – 373: it is not clear how the role of melting hydrometeors is illustrated in Figure 4.

[REPLY] During this stage of cloud development there was limited formation of new cloud droplets from activation of CCN. Precipitation at this stage comes from the ice phase of the cloud. The increased amounts of ice at the upper cloud layers provided the source for precipitating condensates. This sentence has been rephrased.

P13, line 388 – 389: the explanation given is not sound. The possible reason may be that the presence of giant CCNs in the "hazy" case drove down the maximum supersaturation reached in the updraft which prevented the activation of small particles.

In the "pristine" case, because of the relative low number concentration of CCN, the maximum supersaturation may not be affected as significantly by the presence of a few giant CCNs. But of course, this will all depend on parameterization used in the model.

[REPLY] The formulation used in the model is the one suggested by Barahona et al., (2010) which considers the effect of giant CCN on droplet formation. In the "pristine" case, the clouds contained limited number of droplets which allowed them to grow fast to rain droplets. Adding a few GCCN for this case did not significantly change the cloud droplet spectrum in the model, hence rainfall was not affected. Similar to other published studies (e.g. Teller and Levin, 2006) we found precipitation rate to be mainly affected by the number of cloud droplets.

P13 – p14: other than the visual illustration of Figure 7, can we look at domain total precipitation to see if the "hazy" cases always result in reduced precipitation in comparison to their respective "pristine" counterparts?

[REPLY] This is a very useful comment. The domain total precipitation for each case has been included in the revised manuscript and the corresponding paragraph has been rephrased. "Pristine" clouds in general produce more rain. However this is not always the case. Increased CCN may under some circumstances lead to more precipitation. As an example, the "hazy complex terrain" case produced more accumulated precipitation than the "pristine complex terrain". Similar examples were simulated during the "real case" runs. Under realistic conditions the response of aerosols on precipitation is not monotonic and should be examined per case.

3.2 Case study Some of the specifics for the simulation setup are missing, such as, the length of simulation (or simulation period), the chemical tracers (gaseous and particulate) carried in the simulation, the initial and lateral boundary conditions used for the chemical tracers. Again, it is not clear whether mineral dust and sea salt aerosols are treated as externally mixed aerosols and if the model allows internal mixing (through coagulation, for example).

[REPLY] This information has been included in the revised manuscript. The model was run for a two day spin-up period (26-27 January 2003) prior to the actual event to allow the establishment of an aerosol background. The test case run started at 28 January 00:00 UTC and lasted for 48 hours. During this simulation only dust and sea salt particles were included. The model does not allow explicit interaction between dust and sea salt. For the cloud activation process we assumed internally mixed aerosol in a parameterized way. Fixed percentages of dust particles were assumed to be coated by sea salt.

P16, line 463 – 465: are any of these aging processes represented in the model simulation?

[REPLY] Aging is not included in an explicit way. What we have done is to assume fixed percentages of soluble dust particles that represent average conditions.

P16, line 468: Figure 10 is not a good illustration. It may be more effective using simple 2D plots to illustrate. Also, what model runs are shown in Figure 10 and 11 (15km, 3km, or 750m resolution runs)?

[REPLY] Figure 10 has been removed.

P16 bottom paragraph carrying over to p17: is particle number concentration a prognostic variable or diagnosed from mass concentration? Again, are dust and sea salt particles treated separately as externally mixed particles in ICLAMS?

[REPLY] Particle number concentration is diagnosed from mass concentration. Dust and sea salt are treated separately in ICLAMS.

P17, bottom paragraph: for EXP3 IN concentration was multiplied by 10 in presence of mineral dust – this implies that IN is not linked to modelled particles in ICLAMS (which is somewhat in conflict to the statement made at the end of section 2.6.

[REPLY] We recognize this issue; this simulation was carried out as a first-order approximation of the impact of dust against the “background” IN concentrations that are

used in the Meyers parameterization. In a future implementation of the model, a much more sophisticated approach will be used, where size distribution and aerosol type will be considered in the calculation of the IN spectrum.

P17, line 517 – p18, line 527 (discussion on aerosol size spectrum): I am somewhat confused on how particles are modelled in ICLAMS. Is the particle size distribution simulated or imposed? Also, are sea salt particles included as CCNs in the droplet nucleation calculation? Is the soluble fraction of dust particles treated as sodium chloride or ammonium sulphate in these experiments?

[REPLY] In ICLAMS dust and sea salt mass is a prognostic variable for each bin. Number concentrations are diagnosed from mass concentrations. For sea salt the distribution properties are fixed. For dust the distribution properties are estimated from the relative mass concentrations of the eight dust bins. Sea salt particles are included as CCN in the droplet nucleation mechanism. The soluble fraction of dust particles during all the tests for the 28th January 2003 case is considered to be sodium chloride.

P18, line 533: what does “less polluted cloud” mean here? The dust particles in EXP1 are less hygroscopic (or less) aged than those in EXP2. To clarify, by “5% of dust particles were hygroscopic” (referring to line 510 on p17) do you mean that all dust particles are composed of 5% soluble material and 95% insoluble material?

[REPLY] Less polluted refers to clouds that have been developed in an environment with fewer CCN. The soluble fraction of a hygroscopic dust particle during all runs is set constant (33%). What changes throughout the cases is the percentage of dust particles that are assumed to contain soluble material.

P18, line 555 – 557: does this translate to greater accumulated precipitation, which is in contrast to the idealized cases, i.e., more CCNs lead to reduced precipitation?

[REPLY] This often is the case for warm clouds. The response of precipitation to aerosol variations is generally not monotonic. For example, in the topography tests, increased CCN leads to higher clouds with more ice condensates. These clouds may either evaporate or produce heavy rainfall depending on the ambient conditions.

P19, line 565 – 566: "particle concentration was a prognostic variable" – number concentration or mass concentration, size-resolved or bulk?

[REPLY] Mass concentrations of dust and sea salt are prognostic variables in ICLAMS. Number concentrations are diagnosed. Dust is represented with eight mass bins as in Perez et al., 2006a and Spyrou et al., 2010. Sea salt is represented with a bimodal lognormal mass distribution.

P19, line 578 – 5580: can you indicate the number of observations available for each of these thresholds (somewhere in Figure 16 or 17)?

[REPLY] Figure 16 (Figure 13 in the revised manuscript) has been redrawn and this information has been included.

P19, 584 – 587: this statement is not well supported by Figure 16, e.g., case 7 and 9 overpredicted at the higher end. Again how many observations over these threshold ranges?

[REPLY] As stated also in the text, the limited time period as well as the limited number of stations does not allow the extraction of robust statistical results. The results presented here should be treated mostly as an indicator of the significant variability on precipitation that can arise from aerosol. However the online treatment of aerosol as CCN in general improved model performance for most thresholds but still not for all cases. For example, enhancing the hygroscopicity of dust particles (Case 8) leaded to degraded model performance. In our opinion, this means that including such interactions in modeling experiments can improve our understanding on several atmospheric processes but the results should be examined with care and always within the framework of the specific case study.

P20, line 601 – 603: the better average biases can be a result of compensating errors

between over and under prediction as seen in Figure 16. What can be established here from these tests is sensitivity to the interaction between aerosol and cloud microphysics and dynamics. The improvement in the score may be fortuitous due to uncertainties in modeled/imposed aerosol properties and possible compensating errors.

[REPLY] The "prognostic aerosol" cases overall scored better biases than the "fixed aerosol" ones. However we agree with the reviewer that these results mostly indicate the variability and non-linear nature of these processes. Figure 16 has been removed from the revised manuscript.

4. Concluding remarks P20, line 609 – 610: "Aerosol partitioning ... such as aging particles" needs rephrasing.

[REPLY] This paragraph has been removed.

P20, line 613 – 615: there is also significant uncertainty in modeled aerosol properties.

[REPLY] Sure. However this sentence has been removed from the conclusions section in the updated version.

P21, line 640: what is the increase of 15% in soluble dust concentration referring to? Again, it is not clear if the soluble fraction refers to the fraction in number concentration or in particle composition. My understanding of the Fountoukis and Nenes scheme is that the soluble fraction is referring the latter (i.e., the fraction in composition assuming internally mixed aerosols).

[REPLY] All hygroscopic dust particles contain 33% Sodium Chloride. The increase of 15% refers to the fraction in number concentration of dust that is assumed to be hygroscopic.

P21, line 645 – 646: again the improved precipitation score may be fortuitous due to compensating errors.

[REPLY] Precipitation was in general under-predicted by the model. Bias score was less than one for all thresholds except two cases (Case7 and Case9) that over-predicted the 54mm threshold. However, even excluding the 54mm threshold from the calculation of average bias the prognostic cases still score better biases. These results are not presented here to show model improvement but to indicate the importance of these processes in clouds and precipitation. This paragraph has been rephrased and the scores now refer to specific thresholds.

P21, line 647 – 650: similar caveat to this statement as above, as well as uncertainties in the combination of simulated and imposed aerosol properties in this study.

[REPLY] The uncertainties related to aerosol properties are significant since there are a variety of natural mechanisms that we still do not understand or are poorly parameterized. However all model runs were performed with exactly the same configuration apart from the fraction of hygroscopic dust particles to allow for a valid comparison between the several cases.

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