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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Increase of the aerosol hygroscopicity by aqueous mixing in a mesoscale convective system: a case study from the AMMA campaign" *by* S. Crumeyrolle et al.

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We thank both reviewers for their detailed and constructive comments on our manuscript. We have revised the manuscript attempting to take into account all the comments raised by both reviewers. We apologize for the delay due to the time required to perform the requested additional analysis.

General Comments of Reviewer 2 :

1/ “Aqueous mixing” and “Cloud processing” : To improve the readability, we have changed “aqueous mixing” by “cloud processing” in the manuscript.

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Interactive Discussion
Discussion Paper

Given that you explain the observed effects on aerosol modification by cloud processing, you should give a more detailed discussion on possible processes that occur in the cloud. Are both physical and chemical processes included in the model? Can chemical processes in the cloud explain (some of) the observed aerosol modification? Cloud processing refers to several processes, including mass transfer between the gas phase and aqueous phase (e.g., via rapid aqueous oxidation of sulfur dioxide to sulfate), dissociation/ ionisation of certain dissolved species, and chemical reactions amongst various species in aqueous solution in the cloud droplets. Fewer studies have found that the aerosol particles mass have changed as a result of particle sulfate and SO2 scavenging (Hegg and Hobbs, 1981, 1982; Flossmann et al., 1985, 1987; Flossmann and Pruppaccher, 1988; O’Dowd et al., 2000).

These observations have been modeled using an air parcel model with detailed microphysics (Wurzler et al., 2000). This study shows that cloud processing of dust particles is a possible effective pathway to form soluble coatings on dust particles. Furthermore, this study shows that after one or two cycles of particles through convective clouds the contribution of gas uptake by drops and subsequent liquid phase oxidation adds considerable mass of soluble material on particles in the size range of 0.05 μm.

In our study case, the Harmattan and the monsoon flux create two distinct layers. In the higher layer or Saharan Air Layer, air masses come from desert regions and are enriched in dust particles. In lower layer or boundary layer, air masses come from the Guinea gulf and are enriched in trace gases (SO2, NO3, NH4, etc …) and in chemically modified particles. As exchanges between these two layers are weak, only the convective currents of the MCS are able to mix trace gases, modified particles and dust particles together.

The model considers only the physical and cloud processes such as aerosol sedimentation, dry deposition, and interaction between aerosol and cloud microphysics. Washout calculated in the model is based upon first order principals. In- and below-cloud impaction scavenging by cloud droplets and raindrops uses a kinetic approach

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to calculate the aerosol mass transfer as defined by Seinfeld and Pandis (1997), Pruppacher and Klett (2000) or Tost et al. (2006). The in-cloud mass aerosol transfer into rain droplets by autoconversion and accretion processes have been introduced as described by Pinty et al. (1998). The sedimentation of aerosol mass included in raindrops was solved using a time splitting technique with an upstream differencing scheme of the vertical sedimentation raindrop flux. The release of aerosols into the air due to rain evaporation is assumed to be proportional to the water evaporated (Chin et al., 2000). Indeed, aqueous phase chemistry is not included but the model is able to follow the dust mass during its collection in the cloud and its possible release after raindrops evaporation. It will be detailed in a next paper (in preparation), but results shows that about 200 μg.m-3 of dust have been released after raindrop evaporation after the MCS passage. Consequently, we rely on observations to study the changes of aerosols chemistry. The single particle analysis showed that accumulation mode particles were coated by sulfate and coarser particles by nitrate. So, chemical processes in the cloud modeled by Wurzler et al (2000) explain the surface property modifications that increase the particle hygroscopicity. We have clarified this point in the text.

2/ The term ’mixing’ is used for quite different processes. We agree that in this manuscript we used the word “mixing” to include all aerosol processes that may occur in the atmosphere. The different terms used have now been defined more accurately in the manuscript. P10059 l. 25/26 : During the aerosol transport from the Guinea Gulf to Niger, processes like coagulation, condensation, nucleation, cloud processing, dry and wet deposition may occur. Particles that have been affected by these processes have been modified. Indeed, the concentration, the size, the chemical composition, the mixing state, optical, and hygroscopic properties of each particle may change. The term “mixed” have been erased in the new version of the manuscript. p.10070, l. 9: what is meant by ’Mix particles and nitrates together’? : Because of the dissociation of Harmattan and monsoon fluxes, dust particles are never transported in the same layer as trace gases. During a convective event, the MCS downdraft and updraft mix these two layers and condensable

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gases are in contact with dust particles. Then, the condensation process may happen. In this sentence the word “mix” means “condensate”. It has been corrected in the manuscript. p. 10077, l. 5: Is it simply ’due to the presence of aged marine and anthropogenic aerosols’? We agree. The fact that particles in the BL are more hygroscopic than in the SAL, is not only a consequence of the presence of marine and anthropogenic aerosols. In one hand this is due to the presence of initial aerosols more hygroscopic than in the SAL. In the other hand, this is due to soluble vapour gases that condensate on particles during this long-range transport that enhance the hygroscopicity of particles. The observed results shown in figures 5 and 6 confirm that the CCN concentration and fraction are more important in the boundary layer before the MCS passage. p. 10079, l. 6: The MCS leads to a mixing of the air masses. However, the aerosol must undergo some kind of processing (chemical or physical) in order to change their properties. Indeed, the MCS leads to a mixing of soluble aerosols from the boundary layer with insoluble aerosols from the SAL. Then, this mixing appears to be in an aqueous phase. Indeed, the cloud and ice mixing ratio is between 0.4 and 4 g.kg-1 (see fig. 15). Many processes occur inside the MCS. First of all, the more hygroscopic particles activate to become droplets. Soluble compounds of activated particles are dissolved in the liquid water. Then, insoluble particles are collected by droplets. Upon water evaporation, dissolved compounds are precipitated on the core of the droplet. Initial insoluble particles are coated by soluble elements and their hygroscopic properties increase.

3/ What has been really determined in the single particle analysis? The single particle analysis is based mainly on the morphological and chemical analysis of individual particles found on both stages of the impactor. Samples were imaged firstly under a digitized transmission electron microscope (TEM, Hitachi H-7650) to obtain high resolution images of the particles. Then, the same fields were located again under a scanning electron microscope (SEM, JEOL, JSM-5910LV) coupled to an energy dispersive X-ray spectroscope (EDS, Princeton Gamma-Tech, Prism2000), in order to obtain the elemental composition of the individual particles (Na, Mg, Al, Si, P, S, Cl, K, Ca, Mn,

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Fe). Elements such as C, N and O were not included in the quantification of the relative atomic ratio. For more details on the single particle analysis, see Matsuki et al. (2008). This reference has been added in the manuscript. Finally, “sulfur” has been replaced by “sulfate” in the manuscript.

Specific comments of Reviewer 2 :

Abstract, l. 18: We added (CCN) at line 10 in order to define CCN.

p. 10062, l. 11: What is the uncertainty on the supersaturations? Recently it has become evident in several CCN studies that the actual supersaturation values can differ significantly from nominal values. Was this deviation taken into account? This is true that the uncertainties on the supersaturations are significant. Leaitch et al. (1999) and Bilde and Svenningsson (2004) reported effective and nominal supersaturations. The effective over the nominal supersaturation (Seff /Snom) ratios reported in these two studies were 0.75 and 0.78, respectively. Thus, the uncertainty on the supersaturations for the Wyoming CCN chamber is about 25%.

During the AMMA campaign, we measured the CCN concentration at different supersaturations (0.2, 0.4, 0.6, 0.8, 1). We plotted the CCN/CN for each supersaturation and the tendencies were exactly the same. The goal of this paper was not to carefully quantify the CCN fraction at one accurate supersaturation but to show the relative evolution of the CCN fraction after the MCS passage.

p. 10065, l. 4: This sentence has been completed.

p. 10066, l. 7/8, l. 18: Both sentences have been modified as suggested.

p. 10068, l. 22-24: Where all these compounds, including sea-salt, externally or internally mixed, i.e., does ’fraction of particles’ refer to ’number fraction’ or ’mass fraction’? As the results are provided by the single particle analysis, the fraction of particles refer to number fraction. We clarified it in the text. Furthermore, we were talking about different types of particle. Thus,

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the sampled particles were at 26% (in number) composed by sulfate and 13% were composed by potassium and sulfate.

p. 10070, l. 12-15: Even though it becomes clear in Figure 6, also the text should reflect why the CCN fraction increases. Is it an addition of new CCN or remains the CCN number the same and the fraction only increases because the total CN number decreases due to sedimentation of insoluble particles? We agree that this point is probably not explained enough in the manuscript, and we have added a part of this discussion in the manuscript. The CN and CCN concentrations plotted on Fig.5 show that the CN concentration decreases due to wet deposition. Indeed, during the MCS passage the precipitation rate involve processes like aerosol scavenging, aerosol collection by cloud/ rain droplets. Before and after the MCS passage, other processes like coagulation and dry deposition occurs. All these processes involve a CN concentration decreasing. Another result shown on Fig.5 is that the CCN concentration increases after the MCS passage. So, the CCN fraction increase shown in Fig.6 is due to a decrease of CN concentration and an addition of new CCN into the atmosphere.

p. 10074, l. 9: Is the 5h shift between observations and model taken into account in Fig. 10? According to the Figure, the 5 h delay cannot be ’clearly retrieved’. The time delay between observations and model is not taken into account in Fig. 10. This figure is used to illustrate this 5h shift. We agree that this time delay cannot be “clearly retrieved” on the temperature and the pressure time series because of the daily evolution of both parameters. But, regarding the wind speed and the cumulated precipitation rate we can “clearly retrieved” this 5h shift. Indeed, the first modeled wind speed peak is shifted 5 hours before the observed wind speed peak. The second and third modeled wind speed peaks correspond to the MCS formed east of Banizoumbou (see Fig.9). These peaks did not appear in the observations because of a different MCS trajectory in the reality compared to the simulation.

p. 10075, l. 15-21: Can you give any reason why the BL thickness differ by a factor of

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2? In MesoNH the MCS is modeled 5 hours before the observed one. Nevertheless to be comparable, it is important to have the same transport of the aerosol that came from the MCS in the model and in reality. The comparison between the modeled and observed aerosol mass profile need to take into account this time difference. So we plotted on Figure 11, the observed concentration measured during the ATR flight between 12:00 UTC and 14:00 UTC, and the concentration modeled between 07:00 UTC and 10:00 UTC. If this time delay permits to give a good comparison of the aerosol profile in the intermediate layer (aerosol that came from the MCS), it causes some problems in the boundary layer. The compared meteorological situations are different due to the time lag between the observed and modeled periods. Indeed, due to the difference of solar radiative flux, the boundary layer is thicker in the morning than at noon. At 7 UTC the boundary layer thickness is 2 times lower than at 12 UTC.

p. 10077, l. 15: Does the increase in CCN/CN fraction and decrease on CN number mean that the air mass that was down-transported contains a lower particle concentration, diluting the current particle concentration? Behind the MCS updraft, the precipitation falls create density currents. These density currents down transported the air masses from the group 3 trajectories (see Fig.12). During this significant air masses descent, particles from these air masses underwent cloud processing, which was described in the general comment 2 (activation, collection and evaporation). These processes lead to an increase of the CCN concentration in the atmosphere. In the same time, as a part of the precipitation rate was not evaporated (see Fig.15), the particle scavenging by rain droplets explains that the CN concentration is decreasing.

p. 10079, l. 12 and 18: Do you mean ’modified particles’ or ’newly formed particles’. The good expression is modified particles. We corrected it.

p. 10080, l. 9: It has been modified as suggested.

p. 10080, l. 24/25: Is an increase of the volume concentration of the coarse mode

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due to (chemical?) mass addition in clouds? Assuming all particles are spherical, the volume concentration is calculated from the number concentration. In the Boundary layer, the volume concentration increase is not due to chemical mass addition in the cloud but to the generation of dust particles by the MCS density currents with diameter much bigger than those initially present in the BL.

10080, l. 26: We agree and we have corrected it in the manuscript.

p. 10081, 8/9: ’hygroscopic characteristics’ have been replaced by ’hygroscopicity’

Figures 5 and 6: Could you add the legends to the plots? What are the different symbols, triangles (Fig, 5), squares and dots? It might also help if you could add a horizontal line marking the BL. The legends have been added to Figures 5 and 6. The addition of two (before and after the MCS) horizontal lines marking the BL on Fig.5 overloads this figure and makes it hardly readable, but it has been done on Fig.6.

Technical comments

p. 10058, l. 13, p. 10060, l. 26, p. 10062, l. 5, p. 10062, l. 20, p. 10063, l. 1, p. 10063, l. 12, p. 10066, l. 8/9, p. 10067, l. 28, p. 10068, l. 14 : Language mistakes have been corrected, and as suggested by Reviewer 2, this manuscript was carefully checked for proper English by a native English person.

p. 10072, l. 11, p. 10073, l. 7, p. 10073, l. 24: Revised as suggested

p. 10074, l. 12: It has been done

p. 10074, l. 24: We removed ’modeled’

p. 10075, l. 9: Contradiction: is it 12-15 h after the passage or do you refer to 7:00-11:00 UTC? In our modelisation, 12-15 h after the passage corresponds exactly to 7:00-11:00 UTC.

p. 10075, l. 12, p. 10076, l. 5/6, p. 10076, l. 27: These points have been corrected.

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p. 10078, l. 4: In fact both terms may be valuable for the explanation given in this sentence. We really want to say behind (i.e. locally) the MCS where the precipitation occurs. In Fig.14, which is a picture of surface dust concentration, we can clearly divided all convective cells in two parts. The convective part of the cell where there is high surface wind speeds, and the second part of the MCS where density currents occur due to precipitation rate. But, it is true that after (i.e. temporal) the MCS the precipitation rate increases the soil moisture and then inhibits the dust generation as well.

p. 10078, l. 13, p. 10078, l. 17, p. 10078, l. 19, p. 10078, l. 20, p. 10078, l. 20, p. 10079, l. 13: These six words have been changed as suggested.

p. 10080, l. 5, p. 10081, l. 10-12: Revised as suggested.

p. 10082, l. 10, p. 10082, l. 23, p. 10082, l. 26: The reference mistakes have been corrected.

Figure 7/8/10: These three points have been corrected.

Figure 12/15: We added x-axis label to the first figure and plotted the second one figure with larger font for axis label.

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