Interactive comment on “Aerosol processing and CCN formation of an intense Saharan dust plume during the EUCAARI 2008 campaign” by N. Bègue et al.

Anonymous Referee #2

Received and published: 8 January 2015

This paper describes the dust modification occurring during transport from Sahara to Netherlands. The paper is based on Meso-NH simulations validated by in-situ measurements from EUCAARI campaign. They used a large variety of instruments (AMS, CPC, CCNC, MAAP) that were located at the ground or on-board the ATR-42.

The paper is well written and the topic fits the ACP’s scope. Therefore, this paper can be published in ACP after major revisions. As reviewer #1 stated, the discussion should be more sensitive as no observational evidences could be provided to support their conclusions. As reviewer #1 focused on the remote sensing measurements, I focused all my comments on the in-situ measurements and their comparison with simulation
results.

Major comments There is a structure issue in this manuscript. Indeed, in the section 4.1, the authors are concluding on the dust hygroscopic modifications. However, the evidences of these modifications are shown in the section 4.2. If your goal is to explain why dust are highly hygroscopic after long-range transport, I strongly suggest to show Figure 10 and Figure 11 first and then explain what could explain this huge enhancement of the CCN concentrations. Also, the dust plume is located in a layer between 2 to 5km. The authors present several plots showing surface measurements. Those plots are presented to evaluate the simulations results, but are mixed throughout the paper with the actual results highlighting the dust modifications. The last figure is for example depicting the CCN concentration measured at the surface. The mix of results from the surface and the dust layer (that are not related right?) is confusing. Either you want to use this data set to prove that your model is doing a fairly good job and then you have to change the title either you prove that the model is doing a fairly good job and then you use it to better understand the modifications of dust particles throughout the transport.

AMS measurements are limited to refractive aerosol with diameter lower than 500nm. This instrument is thus not designed for dust measurements. You can use it to determine the chemical composition evolution of the Aitken and accumulation mode of refractive aerosols and validate your model. The comparison simulation/observations should be done by taking into account the size and chemical properties of the aerosol (refractive/non-refractive) limitations. Otherwise you are just comparing two different parameters that cannot be compared. The authors never clearly write those limitations in this paper. Could you differentiate dust from urban particles? Could you differentiate the organics or inorganics which have condensate on dust to organics/inorganics which have condensate on non dust particles? This information would be priceless and would help to convince the reader.

To calculate the CCN/CN ratio, the aerosol concentration (CN) should correspond to
aerosol that can be activated. The scientific community commonly use CN>50nm. Indeed, particles smaller than 50nm are not CCN active but are numerous. That could mislead the CCN/CN interpretation. Instrumentation on board the ATR-42 included particle sizers providing aerosol concentration from 10nm to the inlet cut-off. You have to use a CN concentration from 50nm-5um. Besides the CCN/CN parameter is not anymore used to avoid the size issue, kappa (see Petters and Kreidenweis, ACP, 2007) is nowadays the hygroscopocity parameter.

The CCN concentrations are always under-estimated by the model. Any idea why ? I would like to see a plot CCN calculated from MESO-NH as a function of the observed CCN color-coded with the altitude. It seems like the Model is doing a bit better for the lower altitude than the elevated layers. The similar plot would be interesting for the sulphate, ammonium, OC and nitrate.

Minor remarks :

Format : Why is there a mix of normal and italic fonts ? Be consistent throughout the paper with the acronyms ATR-42 not ATR42

P53 L22-27 : The vertical structures of clouds is not well reproduced. What does that change for you study case?

P57 L14-17 : Is the model able to condense sulphate, nitrate or organics on dust particles ? Because knowing that sulphate, BC and Dust were simultaneously in the same layer doesn’t mean that dust would be modified. Could you quantify, using the relative humidity, the amount of material that could have condensate on dust ?

P57 L17- 19: Should it be a new paragraph ? or should it be the introduction of the 4th section ?

P58 L16-17 'the aerosol layer is continuously masked by cloud layer' This affects the CALIPSO measurements. Thus the comparison with Meso-NH is quite questionable…

P58 L17-20 : Both sentences mean the same thing. Remove one of it.
So according to Figure 6 you can differentiate dust particulate mass from the rest. The extinction coefficient plotted on Figure 7 is due to total aerosol loads right? Can you add dashed lines to represent dust extinction coefficient on Figure 2, 3 and 7? That would be helpful to see where the dust are located. CALIPSO can provide you information about the type of particles. So basically you could retrieve the dust contribution to the total extinction.

The AMS is a powerful instrument but it has a lot of limitations. The comparison you performed looks good BUT did you compare the same thing (non-refractory and smaller than 500nm particles)? The simulated concentrations correspond to total aerosol load or to aerosol with diameter smaller than 500nm? The authors have to state clearly what parameters they are using.

What kind of observations were you looking for? At Cabauw there was an AMS, AIS and SMPS... Which is more than enough to compare model and observations... The SOA measurements were performed at Cabauw, so did you mean that all these instruments were not working properly during this study case or that you didn’t find the data?

Please rephrase ‘with more half’

Meaning that the mixing was not efficient with dust... That would be really interesting to estimate the sulphate and nitrate that may have been mixed with dust particles.

You confirmed here that the hygroscopicity modification is due to a coating of sulphate or nitrate BUT you haven’t shown the hygroscopic modification yet.

CCNC means Cloud Condensation Nuclei Counter.

As previously said, the aerosol concentration (CN) of the CCN/CN corresponds to aerosol that can be activated. In this case you used the total CN including aerosol from 10nm to 50nm. Even composed of soluble compounds, these aerosols are not
acting as CCN because as said by the referee 'size matters'. You should use a CN concentration from 50nm-5um or Kappa...

P 63: So the CCN concentration measured at the surface are really high (up to 14000#.cm-3) and generally over 4000#.cm-3. These concentrations are certainly not representative of a dust event. CCN concentrations measured on-board the ATR-42 are not exceeding 1000#.cm-3. The events occurring at the surface have nothing to do with the dust plume and that needs to be said clearly in the manuscript. Why even showing it?

P65 L15-16: The value of the CCN/CN over Netherland was greater than those observed over the Saharan region. How do you know? Did you use measurements over the Saharan region when the dust were lifted up to say so? Is it just your model that give this information? Please be clear. Dealing with Observations and Model is fine but you need to clearly say what is coming from the observation or from the model.

Figure 1: From what I see, you used a column integrated aerosol concentration. Why didn’t you use a more classical parameter like AOD. Moreover you could have compared those results with MODIS or AERONET measurements. That will convince the readers that the model is doing a great job.

Figure 2: Figure’s quality need to be improved.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 27039, 2014.