Response to anonymous referee 2's comments

First of all, the authors acknowledge the referee for his constructive comments and suggestions. In the revised manuscript, the authors made an effort to improve the quality of the discussion by adding information concerning lidar depolarization ratio and the size of the particle. Furthermore, we made also an effort to say clearly what is coming from observation and model. The modifications are indicated by italic and bold fonts in the revised manuscript.

Major comments:

Referee 2 : 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.

Authors : We understand the point of view of the referee and that's why the conclusion of the section 4.1 was rewritten in the revised manuscript. Indeed, at this stage of the argumentation we cannot confirm a hygroscopic modification whereas we haven't shown the hygroscopic modification yet. Furthermore, the goal of this section is not to explain why dust are highly hygroscopic after long-range. The real purpose of this section is to determine the nature and quantify the amount of the anthropogenic aerosol mixed with the dust particles. The last paragraph of the section 4.1 should be a transition in order to introduce the next section which the goal is to show a hygroscopic modification by coating. In order to reduce the confusion concerning the real motivation of the section 4.1, we decided to move this last paragraph at the beginning of the section 4.2 (see revised manuscript).

Referee 2 : Also, the dust plume is located in a layer between 2 to 5 km. 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.

Authors : We thank the referee for this valuable comment. The surface measurements were used on the one hand to evaluate the simulation results and on the other hand they were used

to depict the spatial and temporal distribution of anthropogenic aerosol over Europe. In the revised manuscript the last figure was used to prove that our model is doing a fairly good job and then we have used it to better understand the modifications of dust particles. In order to clarify this part of the manuscript, the paragraph was rewritten (see revised manuscript).

Referee 2 : 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.

Authors : We thank the referee for this relevant comment. The comparison between the AMS measurements and Meso-NH were realized in taking into account secondary chemical species. Furthermore, we took into account the Aitken and accumulation mode of Meso-NH. According to lognormal formulation the particle with diameter greater than 500 nm correspond to 8% of the spectrum. Thus, we think that the comparison between the AMS measurements and Meso-NH are based reasonably on the same parameters. However, in order to improve the quality of the comparison model/observations, we remove the 8% of the particles greater than 500 nm. A sentence has been added in the revised manuscript to precise these elements. Furthermore, we plotted the figures in taking into account this correction in the revised manuscript.

Referee 2 : 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 hygroscopicity parameter.

Authors : With respect to Fig 10 in the manuscript, below is shown the corresponding SMPS size distribution plot, measured on the ATR-42. Most of the aerosol particles are found in a dominating and extremely broad accumulation mode (containing significant aerosol number

concentrations up to 500 nm in the SMPS scan). In general most of the particles (70-90%) have sizes beyond 50 nm. Only for two smaller periods, one shortly after 13:15 and the other around 13:55, smallest aerosol sizes in high number concentration (may stem from nucleation event) are observed. Comparing CCN-0.2%/CN50nm to CCN-0.2%/CN10nm these two small periods of course have very significant impact on the respective ratios. All the rest of the discussed 1h time period the calculation of both ratios generates slightly larger CCN-0.2%/CN50nm ratios as compared to CCN-0.2%/CN10nm since the particle fraction below 50 nm is reasonably small with respect to total particle concentration beyond 10 nm (CPC3010). We plotted the figure in taking into account this correction in the revised manuscript.

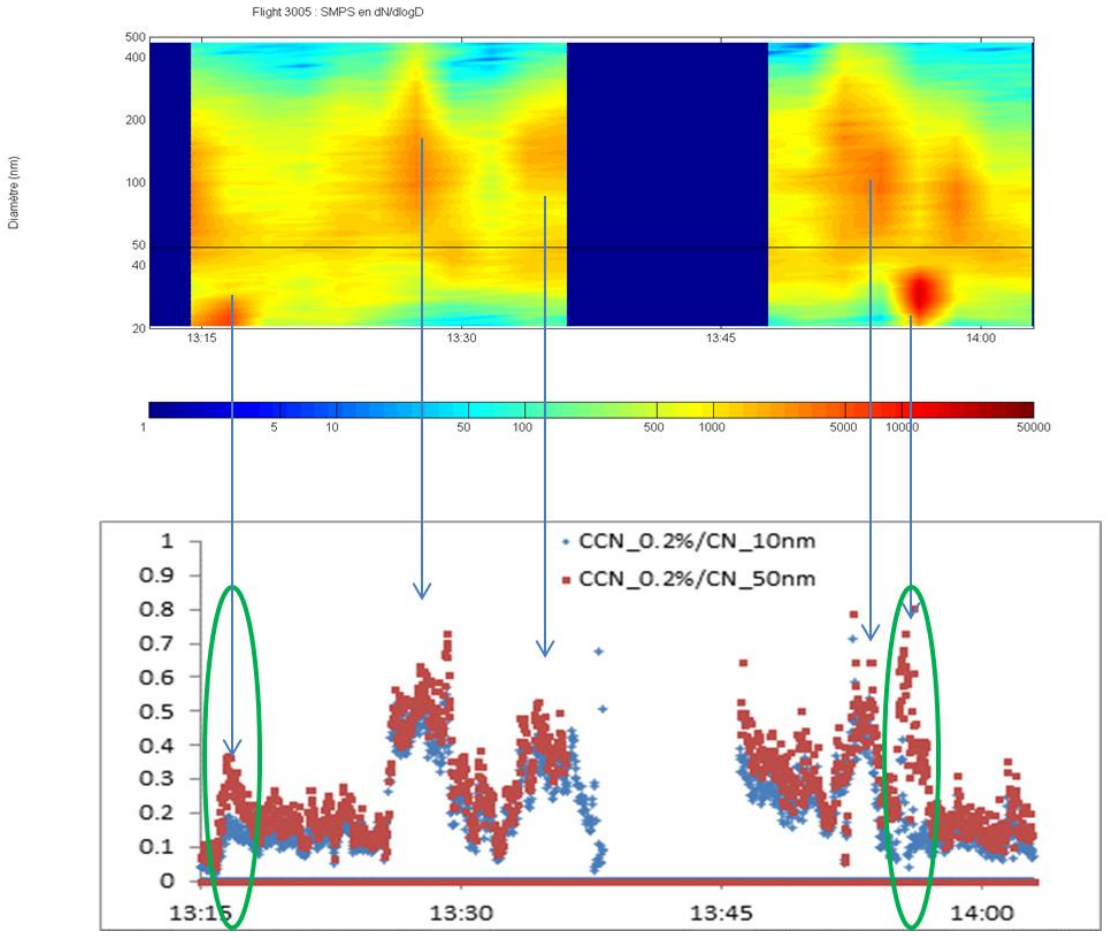

Figure 2.1 *: (top) Evolution of aerosol size distribution fits obtained from SMPS during the flight of the ATR-42 (dN/dlog D). (bottom) evolution of the CCN/CN_10nm and CCN/CN_50nm ratio at 0.2% supersaturation.*

The referee is right to mention the kappa. Our opinion is that the measurements recorded aboard the ATR-42 are not enough to retrieve trustworthy values of kappa. It is for this reason that we prefer to use the CCN/CN ratio (in taking into account the size limitation) as hygroscopicity parameter.

Refree 2 : 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.

Authors : The referee is right. This under-estimation is a consequence of the horizontal resolution of Meso-NH which induces a smoothing of the structure. The comparisons between the CCN calculated and CCN observed following the altitude range (Fig. 2.2) reveal that the model is doing a good job over the boundary layer (1-2.4 km and 2.5-3 km) with a correlation coefficient greater than 0.8. However, in the boundary layer where the concentrations are more sensitive to steep gradient of the surface emission, the CCN concentration is weakly reproduced by Meso-NH (coefficient correlation around to 0.4). In order to improve the results in the surface, the simulation should be run with better horizontal resolution (around 1 km) in agreement to the heterogeneities of the sources. Unfortunately, we haven't got a trustworthy emission inventory at these high resolutions. We prefer to constrain our study to the effect induce by the transport at large scale.

The results obtained for chemical species look fairly similar to those obtained with the CCN concentration. Thus, the model is doing a good job over the boundary layer (1-2.4 km and 2.5-3 km) with a correlation coefficient greater than 0.7 whereas below 1 km the correlation coefficient is less than 0.5.

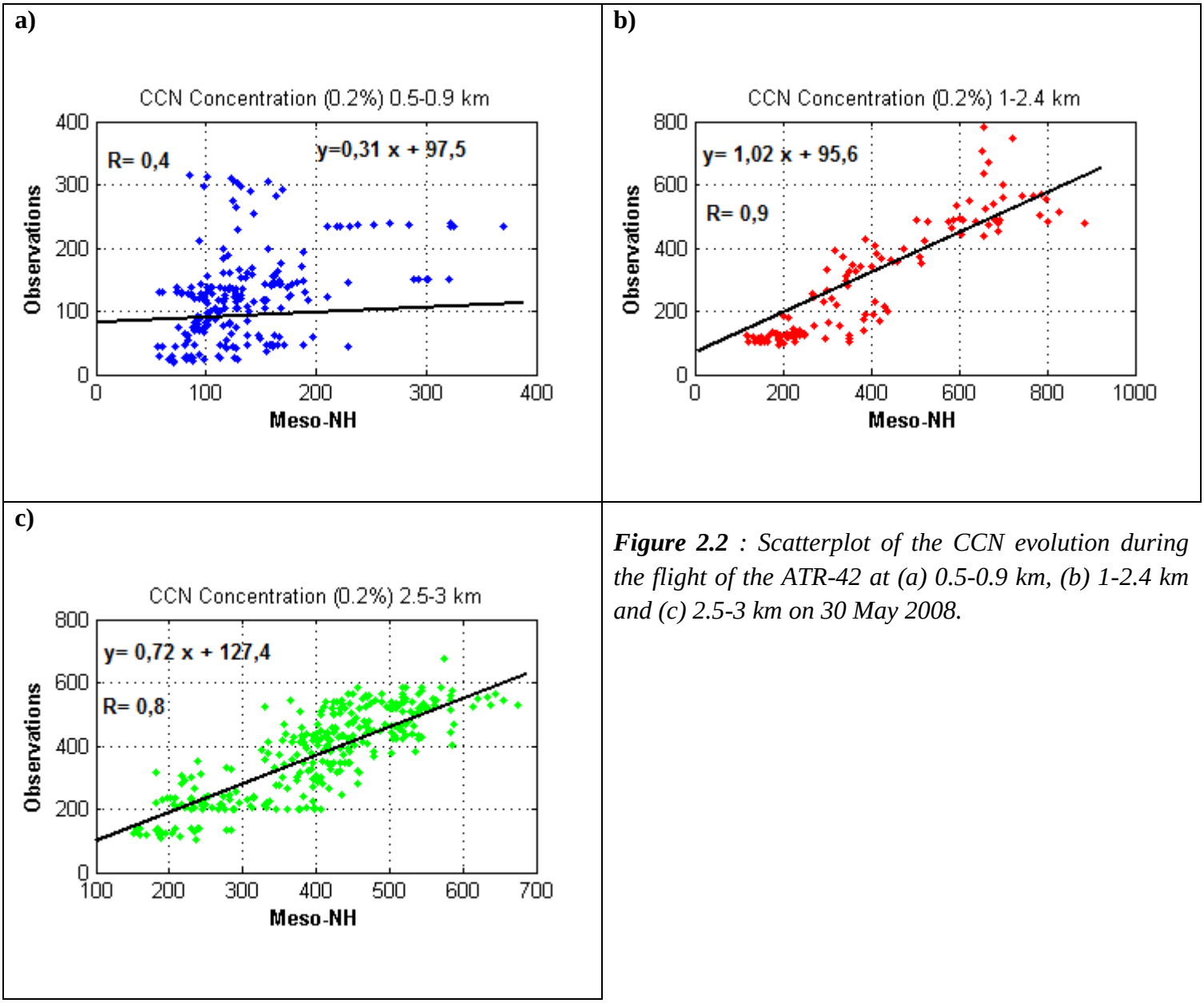

Figure 2.2 *: Scatterplot of the CCN evolution during the flight of the ATR-42 at (a) 0.5-0.9 km, (b) 1-2.4 km and (c) 2.5-3 km on 30 May 2008.*

The figure 2.2 (CCN comparison) have been added to the revised manuscript.

Minor remarks

Referee 2 : Format : Why is there a mix of normal and italic fonts ? Be consistent throughout the paper with the acronyms ATR-42 not ATR42

Authors : The italic fonts correspond to the revisions wondered by the editor for the publication of the manuscript in ACPD. We remind that the italic and bold fonts in the revised manuscript refer only to the revisions asked by the two referees. Moreover, we were more consistent throughout the revised manuscript with the acronym ATR-42.

Referee 2 : P53 L22-27 : The vertical structures of clouds is not well reproduced. What does that change for you study case?

Authors : The cloud are not well reproduced explicitly by the model with a grid spacing of 25 km. It is true for the entire of the Numerical Weather Predict (NWP) model. It is a consequence of the size of the clouds under grid opposite to grid spacing of the model. No explicit cloud (Liquid water content) reproduced by a model does not mean that the model does not take into account their mixing and radiative effect. It is the role of the implicit parameterization of clouds. The implicit parameterizations used by Meso-NH are Kain and Fritsch[1] (1993) and Betchtold[2] et al., (2001) for the deep convection whereas the EDKF scheme is used for the shallow convection (Pergaud[3] et al., 2009). Furthermore, the fractional cloudiness is parameterized following Chaboureau and Bechtold[4] (2002).

It is impossible to remove this error to the other possible sources of numerical error inside a model composed with large sets of atmospheric parameterization. The NWP models are evaluated by comparison to observations. In the previous article (Bègue[5] et al., 2012), a comparison of the precipitations simulated was realized with the TRMM observations. This comparison revealed that the precipitations were fairly reproduced by Meso-NH.

Referee 2 : 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 ?

Authors : This is true. This is the main limitation of the internal mixing for aerosols. This assumption is probably questionable close to the emission sources. Our opinion is at long distance (case of our study), the different processes such as coagulation, absorption will drive

[1] Kain, J., and Fritsch, J (1993) : Convective parameterization for mesoscale models: The kain-fitsch scheme, In: The representation of cumulus convection in numerical models. Eds: K.A. Emanuel and D.J. Raymond. AMS. Monographs, 201 Charles Street Providence, RI 02904-2294 USA, 46, 165–170

[2] Bechtold, P., (2001) : A mass-flux convection scheme for regional and global models, *Quart. J. Roy. Meteor. Soc.*, *127*, 869–886

[3] Pergaud, J., (2009). A parameterization of dry thermals and shallow cumuli for mesoscale numerical weather prediction. Boundary-layer meteorology, 132(1), 83-106.

[4] Chaboureau, J. P., & Bechtold, P. (2002). A simple cloud parameterization derived from cloud resolving model data: Diagnostic and prognostic applications. *Journal of the atmospheric sciences*, *59*(15), 2362-2372.

[5] Bègue, N., (2012) : Long-range transport of saharan dust over northwestern europe during eucaari 2008 campaign : Evolution of dust optical properties by scavenging, *J. Geophys. Res.*, *117*, doi:10.1029/2012JD07611

the aerosol composition close to this type of mixing. A major part of the CTM models uses internal mixing when they parameterizes the gas particles interactions by thermodynamics equilibrium such as ISORROPIA, EQSAM, MARS, ARES, MPMPO etc..

Referee 2 : P57 L17- 19: Should it be a new paragraph ? or should it be the introduction of the 4th section ?

Authors : It should be the introduction of the 4th section. These sentences have been moved at the beginning of the 4th in the revised manuscript.

Referee 2 : 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: : :

Authors : We agree with this comment. We are limited to compare ours models results out of the clouds regions. This is a limitation of our study. Our opinion is that the few areas of comparisons are enough to analyses and validate the plume spread.

Referee 2 : P58 L17-20 : Both sentences mean the same thing. Remove one of it.

Authors : We have removed one of it in the revised manuscript

Referee 2 : P58 : So according to Figure 6 you can differenciate 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.

Authors : It is right. No selection is made on lidar signal and CALIPSO identifies 6 types of aerosols, depending on several a priori location criteria latitude, altitude) and scattering (backscatter and depolarization), see Omar[6] et al., 2009.

Retrieving the dust contribution in the CALIPSO signal is possible, but only in the sense of depolarization attribution, which means two types of aerosol particles using known reference values, following Tesche[7] et al., 2009 and Jouan[8] et al., 2014. That is to say, knowing the

[6] Omar, A. H., (2009). The CALIPSO automated aerosol classification and lidar ratio selection algorithm. Journal of Atmospheric and Oceanic Technology, 26(10), 1994-2014.

[7] Tesche, M., (2009): Vertical profiling of Saharan dust with Raman lidars and airborne HSRL in southern Morocco during SAMUM. Tellus, 61B, 144–164

depolarization ratio of dust (dp1) and non-dust (dp2) backscattering coefficients for the two types of particles supposed to be mixed, and measuring the total depolarization dpt, one can retrieve the equivalent fraction of dust as in Eq. 1.

$$\beta_{p,1} = \beta_{p,t} \frac{(\delta_{p,t} - \delta_{p,2})(1 + \delta_{p,1})}{(\delta_{p,1} - \delta_{p,2})(1 + \delta_{p,t})} \quad \text{(Eq.1)}$$

Assuming the non-dust particles are not depolarizing dp2=0, one can derive the fraction Fd of dust to total backscatter as :

Fd = (dpt/dp1)*((1+dp1)/(1+dpt)

dp1 is very close to dpt in the orbit of Fig. 2, except to north of 47N, where it drops to half its initial value, and Fd then decrease from 1 to 0.54. On the second day, in the orbit of Fig. 3, this is also observed north of 50N.

Referee 2 : P59-60 : 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.

Authors : We thank the reviewer for this relevant comment. A detail response was given previously (see major comment). We remind just that we took into account the Aitken and accumulation mode of Meso-NH. According to lognormal formulation the particle with diameter greater than 500 nm correspond to 8% of the spectrum. in order to improve the quality of the comparison model/observations, we remove the 8% of the particles greater than 500 nm (see revised manuscript).

[8] Jouan, C., (2014) : On the relationship between Arctic ice clouds and polluted air masses over the North Slope of Alaska in April 2008, Atmos. Chem. Phys., 14, 1205–1224, doi:10.5194/acp-14-1205-2014.

Referee 2 : P60 L26 : 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 ?

Authors : It is right to mention that various set of instruments very helpful to compare model and observations were deployed at Cabauw. However, we had some difficulties to find some of them. As a consequence, this sentence (P60-L26: lack of SOA observations over the Netherlands) refer to the fact that we did not find the data.

Referee 2 : P60 L19 : please rephrase 'with more half'

Authors : It was rephrased in the revised manuscript.

Referee 2 : P60 L21-22 : 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.

Authors : The amounts of sulphate and nitrate were estimated from the simulation results and reported in the revised manuscript.

Referee 2 : P61 L3-18 : 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: : :

Authors : We thank the reviewer for this valuable comment. The purpose of this paragraph is to discuss about a possible hygroscopic modification of the dust particles in taking into account the amount of anthropogenic aerosol mixed with them. At this stage of the paper we can just conclude to a possible hygroscopic modification in agreement with similar previous study. Thus, the referee is right to mention that we cannot confirm a hygroscopic modification whereas we haven't shown the hygroscopic modification yet. As a consequence, this part of the paragraph was rephrased in the revised manuscript.

Referee 2 : P61 L27 : CCNC means Cloud Condensation Nuclei Counter.

Authors : It is right. The CCNC means Cloud Condensation Nuclei Counter.

Referee 2 : P62 : 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 referee1 'size matters'. You should use a CN concentration from 50nm-5um or Kappa...

Authors : A response is given previously (see Major comment). Furthermore, the CCN/CN ratio was calculated in taking into account this size limitation in the revised manuscript.

Referee 2 : 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 ?

Authors : Reviewer as to considers this case study as exceptional in its intensity and the direction of the transport. Bègue et al., 2012, have shown the spectacular concentration of dust emitted during this episode reaching 0.25 kg/m2 in the northern Africa for a total mass estimated to 185 Tg (49 Tg on the Mai, 25). These values have to be compared to the annual total mass of dust emitted of 1400 $Tg.yr^{-1}$ (Ginoux et al., 2004).). This dust episode represents 13% of the annual dust emission from North Africa. The number concentration simulated above Cabauw ranging between 100 and 5000 cm^{-3} for the accumulation mode and 25 000 cm^{-3} for the fine mode.

Moreover, it is also worth noting that the CCN concentrations depicted in Figure 10a and Figure 11 are not observed at the same place. Indeed, the figure 11 depicts the CCN concentration at Cabauw (51.97°N; 4.93°E) whereas the figure 10a depicts the CCN concentration over an area extends between 52.57°N and 51.88°N latitude and 6.34°E and 4.99°E longitude. Thus, it is not amazing to observe CCN concentrations fairly different on the figures 11 and 10a.

Then, the numerical simulations reveal that the CN concentration is mainly due to the presence of dust. It is for the reasons mentioned above that we support the fact that aged dust is the main contributor to CCN.

Referee 2 : 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.

Authors : We have concluded that the value of the CCN/CN over Netherland was greater than those observed over the Saharan region from the value usually reported in the literature

over Saharan region (Crumeyrolle[9] et al., 2008; Matsuki[10] et al., 2010). Moreover, It was clearly stated in the revised manuscript.

FIGURES

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.

Authors : We have already used the AOD parameter in the first paper that we wrote concerning this dust event (see Bègue et al., 2012). In order to change with our previous study we decided to use another parameter.

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

Authors : We made an effort to improve the quality of the figure in the revised manuscript

[9] Crumeyrolle, S., (2008) : Increase of the aerosol hygroscopicity by cloud processing in a mesoscale convective system: a case study from the AMMA campaign., *Atmos. Chem. Phys.*, *8* (23), 6907–6924.

[10] Matsuki, A., (2010) : Cloud processing of mineral dust : direct comparison of cloud residual and clear sky particles during amma aircraft campaign in summer 2006, *Atmos. Chem. Phys.*, *10*, 1057–1069