

Response to anonymous referee 1'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.

Abstract

Referee 1: I have my doubts that mountain-induced turbulence has the potential to mix anthropogenic aerosol and gas pollution up to 5 km height. Usually the African air is much warmer than the local one over Europe so that this strongly inhibits mixing at least at heights above 2 km. EARLINET lidar observations usually show typical pure dust characteristics in the dust plumes above 1-2 km height (see Wiegner et al., JGR, 2011, doi:20911JD016619, reporting the same dust episode, May 2008 for Munich, and may be see also Wagner et al., AMT, 2012, 29 May 2008 dust case study over Leipzig). Usually the diurnal PBL development is strongly suppressed during Saharan outbreaks towards regions north of the Alps. The internal mixing aspect (internally mixed particles versus externally mixed particles) is to my opinion speculation. Sure, it may be, but there are no clear experimental indications (unambiguously measured facts) that internal mixing really takes place so that a significant change in the dust properties follows. There was a SAMUM campaign in southern Morocco (Ansmann et al., JGR, 2008, Influence of Saharan dust on cloud glaciation. . .) and they found many cases with liquid cloud formation in the Saharan air layer clearly indicating that pure dust (with the comparably large coarse mode fraction) is a favourable CCN aerosol type, disregarding whether it is mixed with other aerosols or not as in southern Morocco. There is also the paper of Cuesta et al., JGR, 2008 (cited in the paper). I am sure that these authors also observed a lot of water cloud formation in pure dust in southern Algeria, you may ask them for their experiences. So, dust obviously always contains a certain amount of soluble material. Furthermore, it was found that immersion freezing is the dominant ice nucleation mode. So, the water phase comes first, before freezing takes place.

How is immersion freezing possible if pure dust is totally or almost hydrophobic as you try to say . . . sometimes? So, please just keep in mind for the entire discussion within the manuscript that even pure dust contains some soluble material so that dust is always (pure or contaminated) a good CCN. I agree, when mixed with pollution, dust particles may get coated and then the CCN ability may get even better. . . 14000 CCN per cm⁻³, is that really possible? Even 2000-4000 CCN per cm⁻³ is already a rather large number when taking into account that these are the particles with diameters of 80 nm and larger only. For me, 14000 CCN per cm⁻³ would be an indication that the majority of CCNs are just non-dust particles. . . any comment?

Authors : We understand the point of view of the referee. Indeed, the CCN concentration ($14\,000\text{ cm}^{-3}$) and the AOT (value equal to 1) measured over the Netherlands is very high. The referee as to consider this case study as exceptional in its intensity and the direction of the transport. Bègue¹ et al., 2012, have shown the spectacular concentration of dust emitted during this episode reaching 0.25 kg/m^2 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} (Ginoux² et al., 2004).). This dust episode represents 13% of the annual dust emission from North Africa. The dust concentrations (mass and number) obtained during this event are higher than we can obtain for an anthropogenic pollution plume over a region highly polluted. Furthermore, the AOT during high pollution event reach rarely 1 whereas the AOT can reach 10 during the strong dust storm.

You are right to mention that the dust particles can be considered slightly hydrophilic because of their mineral composition. However, the pure dust particles can act as CCN but under highly supersaturated conditions (Gibson³ et al., 2006). The proportion of mineral soluble material is estimated to 5%. In term of mass, this proportion is not enough to enhance significantly the hygroscopic properties of the fine particles. This high concentration of CCN can be explained by the partial activation of fine mode. The activation of the fine mode depends on two processes:

- Coagulation process during the transport
- Coating with soluble material

Roberts⁴ et al. (2002) have shown that a change of 20% in the amount of soluble material can impact significantly the hygroscopic properties of aerosols that initially contain less soluble material, such as dust. Our simulation results reveal that 70% of the high CCN concentration is explained by aged dust over the Netherlands.

¹ 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

² Ginoux, P.,(2004) : Long-term simulation of global distribution with the gocart model: correlation with north atlantic oscillation, *Environmental Modelling and Software*, 19, 113–128

³ Gibson, E., (2006) : Aerosol chemistry and climate : Laboratory studies of the carbonate component of mineral dust and its reaction products, *Geosph. Res. Lett*, 10.1029/2006GL026386

⁴ Roberts, G., (2002) : Sensitivity of ccn spectra on chemical and physical properties of aerosol : A case study from the amazon basin, *J. Geophys. Res.*, 107, doi:10.1029/2001JD000583

Referee 1: P40, L23: Koehler, 2011 is not given in the references. I found several other citations that are in disagreement with the references listed in the reference section.

Authors: The authors would to cite Koehler et al (2010) and not Koehler et al. (2011). It was corrected in the revised manuscript.

Referee 1: P42, L11: Koehler 2010, 2009. . . I only find Koehler 2010 in the references. .

Authors : In the revised manuscript, Koehler et al (2009) was added in the reference section

Referee 1: P42, L13: only slightly soluble. How do you know that more soluble material enhances the CCN potential?

May be even a very low amount of soluble material is always sufficient to become a good CCN. That's why Dusek et al. (2006) published the paper: size matters. .

Authors : The pure dust particles can act as CCN but under highly superaturated conditions (Gibson et al., 2006). It's for this reason that pure dust particles are usually not considered as good CCN (Kelly⁵ et al., 2007; Gibson⁶ et al., 2007). According to Gibson et al., (2007), the CCN activity of pure dust particle is enhances dramatically when they are mixed with a small amount of an aqueous salt. Through an experimental approach, they have shown that dust particles containing nitrate or sulfate component is 2 to 4 times more active than pure dust particles at 0.3% supersaturation. Moreover, Roberts⁷ et al., (2006) have shown that chemical composition may have an important influence on the activation of long-range transported dust. It's for the reasons previously mentioned that we wrote that mixing between dust particles and soluble material may enhance the CCN potential of dust particles.

⁵ Kelly, T., (2007) : Influence of dust composition on cloud droplet formation, *Atm. Env.*, *41*, 2904–2916

⁶ Gibson, E., (2007) : Generation of internally mixed insoluble and soluble aerosol particles to investigate the impact of atmospheric aging and heterogeneous processing on the CCN activity of mineral dust aerosol, *Aerosol. Sci. Tech.*, *41*, doi:10.108002786820701557222

⁷ Roberts, G., (2006) : North american and asian aerosols over the eastern pacific ocean and their role in regulating cloud condensation nuclei, *J. Geophys. Res.*, *111*, doi:10.1029/2005JD006661

Referee 1: P42, L21: Soluble coatings on dust are commonly observed. . . Do you have a good reference here? Is there a clear unambiguous measurement available that corroborates your point? Or is this just the conclusion from chemical analysis. . .? May be there is always almost external mixing of dust and pollution particles and marine particles only, and sure, a bit coating may take place but only introduces second order effects with respect to the CCN issue?

My point here is just to trigger a bit more critical and careful discussion and opinion making? To be clear, I do not want to stop the discussion and the development of your good and nice ideas!

Authors : We thank the reviewer for this valuable comment. In the revised manuscript this sentence was rewritten. Furthermore, several references are given to illustrate the fact that the soluble coating on dust can be observed during the long-range transport of the plume. From airborne measurements over Amazon Basin, Roberts et al. (2002) made a detailed sensitivity analysis of CCN spectra on chemical and physical properties of aerosol. They have shown that a change of 20% in the amount of soluble material can impact significantly the hygroscopic properties of aerosols that initially contain less soluble material, such as dust. Through the analysis of sample collected during brown haze and dust episode from 24 May to 21 June 2007 in Beijing, Li⁸ et al., (2009) have shown that dust particles that acquire hygroscopic nitrate coating tend to be more spherical and larger, enhancing their light scattering and CCN activity. Thus, there are unambiguous measurements reported in the previous studies which corroborate our analysis.

The reviewer is correct to mention that most cases composition plays a secondary role to size in establishing the ability of aerosol particles to act as CCN. Nevertheless, as it was revealed by Roberts et al (2002), the role of chemical processes is not negligible for particles which are weakly hydrophilic. In the manuscript, we would discuss about the importance to analysis for insoluble particles such dust how chemical change can influence their CCN activity.

⁸ Li, W.J. (2009) : Observation of nitrate coating on atmospheric mineral dust particles, Atmos. Chem. Phys., 9, 1863-1871

Referee 1: P42,L29: Levin et al, Wurzler et al., Falkovich et al., could they measure the coating of dust, or did they just conclude from other measurements (in combination with modelling). May be enhancement of hygroscopicity was introduced by the pollution itself (externally mixed sulfate particles)? So my basic question is (since I am not an expert in in situ microphysical and chemical characterization of particles): Are their clear measurement-based indications that coating of dust really takes place, and this to a considerable amount so that CCN properties can significantly be changed. Are their respective laboratory studies available that could be cited. . .?

Authors : We note that Falkovich et al. (2001) was not a good reference here. As a consequence, this reference was removed in the revised manuscript. We confirm that there are measurements indicating that coating dust really takes place. For example, we can cite the work of Roberts et al. (2002). Through the analysis of airborne measurements over the Amazon Basin, Roberts et al. (2002) quantified the impact of the change in the chemical composition of the aerosol particles on their ability to act as CCN. Based on in-situ measurements at Benjing, Li et al. (2009) shown the coating of the dust particle tend to enhancing their CCN activity. Furthermore, from measurements realized in laboratory, Gibson et al. (2007) shown that the coating of dust enhance significantly their ability to act as CCN.

Referee 1: P43, L1: Twohy et al. (2009) reported dust as CCN over the eastern Atlantic. . .. I believe this SAMUM paper (Ansmann, JGR, 2008) is another good example that pure dust must be assumed to be always a favorable CCN.

Authors : This sentence is not very clear and it was rewritten in the revised manuscript. Twohy et al. (2009) did not report that pure dust act as CCN. Indeed, they shown the ability of aged dust to act as CCN. This study illustrates hence the impact of the aging of Saharan dust by coating during their transport over the Atlantic Ocean. Indeed, it is well known that pure dust particles can act as good ice nuclei (IN). Moreover, the paper proposed by the referee 1 confirms that pure dust can act as good IN.

Referee 1 : P43, L15: ice nuclei (not: Ice Nuclei)

Authors : It was corrected in the revised manuscript

Referee 1 : P44, L24: IMPACT stands for Intensive Observation Period at Cabauw Tower???? May be ‘M’ stands for Measurement (rather than Observation. . .). . .?

Authors : The authors confirm that IMPACT stands for Intensive Observation Period at Cabauw Tower. Our response is confirmed by a recent study using the data measured during the EUCAARI-IMPACT campaign (Jarecka⁹ et al., 2013)

Referee 1: P45, L5: So aircraft sampling of dust particles with diameters of 10 microns was not possible. . . These are the best candidates to be coated (according to your theory. . .) because of the large and probably less curved surfaces. . .!

Authors : This remark is true. However, the dust particles with diameters of 10 microns contribute weakly to the number concentration. As a consequence, their contribution to the evolution of the CCN concentration over Cabauw is not significant.

Referee 1 : P45, L19: AMS, cutoff at 500 nm diameter, so you cover a part of the pollution particles, but not the dust coarse mode. So, no observational potential to analyze the chemical characteristics of polluted dust. Only the modeling approach is left to corroborate your basic hypothesis of dust coating. . . I my view ok?

Authors : We agree with the referee. The modeling approach is very helpful when observations are available. However, the modeling approach is also very helpful when observations are available. We think that the observational and modeling approach are complementary. Indeed, the modeling approach can reveal some results that you cannot obtain only from the observations.

Referee 1 : P45, L23: The word lidar is missing after (LNG). What does that mean: Used in the backscatter version or operated in the backscatter mode? Do you have other options? I was thinking lidar is just for 180 degree scattering?

Authors: This oversight was corrected in the revised manuscript. The authors mention that LNG is used in the backscatter version by opposition to the high spectral resolution (HSRL) version of LNG.

Referee 1 : P46, L2: You mean: Fernald, 1984! Nobody uses Fernald et al., 1972, as I know.

Authors : This is right and it has been corrected in the revised manuscript

⁹ Jarecka., D., (2013): Modeling microphysical effects of entrainment in clouds observed during EUCAARI-IMPACT field campaign, Atmos. Chem. Phys, 13, 8489-8503

Referee 1 : P46,L4: Complete set of instruments were deployed. . . . That is never possible! What does complete mean here? I have never seen a ‘perfect’ observational approach!

Authors : This sentence is rewritten in the revised manuscript.

Referee 1 : P46, L4: There is this rather complex EARLINET lidar CAELI at Cabauw for detailed dust profiling and characterization. Are there data available? Would be a pity if not during such an important campaign.

Authors: At the beginning of this study, we have contacted the scientific coordinator of EARLINET network. Nevertheless, no lidar observations at Cabauw are available during this dust event. That’s why in the previous paper (Bègue et al., 2012), we decided to use the depolarization ratio obtained from the ALS-450 LEOSPHERE lidar operated by the KNMI (Royal Netherlands Meteorological Institute) at Cabauw. During this dust event, the LEOSPHERE lidar operated continuously. Based on these lidar observations, we have characterized the presence of Saharan dust over Cabauw. The analysis of the depolarization ratio obtained from the LEOSPHERE lidar over Cabauw is discussed in detail by Bègue et al. (2012).

Referee 1: P48, L26: ORILAM also considers the dust and sea salt as externally mixed. However, in the framework of this study, the dust was introduced into the other aerosol species as internally mixed. So, this is the starting point of the paper. . . . and clearly a speculative assumption, as long as this assumption cannot be based on solid grounds of measurements. Should be clearly stated!

Authors : The internal and external mixture of species are two extreme states of aerosols. Traditionally, air quality models treats aerosols as an internal mixture. This assumption is based on the aerosols processes (such as gas-particles interactions, coagulation) drive to an internal mixing between species.

Numerous studies indicate that aerosols are generally closer to an internal mixture than an external one (except when they are close to the sources): see Liu¹⁰ et al., 2005; Klingmüller¹¹ et al., 2014.

¹⁰ Liu, X. (2005), Global modeling of aerosol dynamics: Model description, evaluation, and interactions between sulfate and nonsulfate aerosols, J. Geophys. Res., 110, D18206, doi:10.1029/2004JD005674.

For polluted cases, ORILAM always model the anthropogenic aerosols in an internal mixture for (see Tulet et al., 2005¹² ; 2006¹³). This is true that ORILAM also considers the natural aerosols such as dust and sea salt as external. But this assumption is dedicated to study the transport of aerosols without strong interaction with pollution (case of desert region or over non polluted marine areas, see papers using ORILAM during AMMA campaign; Kocha¹⁴ et al., 2012; Tulet¹⁵ et al., 2008).

In our case, we assume that the period of the mixing between species is upper than a day. To our opinion this is largely enough to assume as acceptable an aerosol in an internal mixing.

Referee 1: P52, L23: Bangaert. .

Authors : The name of the author (Bangert) was correctly written in the revised manuscript.

Referee 1: P54: Fig4, surface concentrations and surface wind fields, why is that shown, when you deal with lofted dust? . . . and if one keeps in mind in addition that vertical mixing is always strongly reduced during Saharan dust advection (a rather warm dust African air is advected above the colder European air close to ground, i.e., the lowest 1-1.5 km). During strong dust outbreaks such as observed here, the PBL (space left for vertical mixing) is usually just 500 to 1000 m in depth. Please keep that in mind of all the discussion and discrepancies mentioned in the next pages P55-57.

Authors : We would to describe the temporal and spatial evolution of the anthropogenic aerosol in subsection 3.2. Because of the anthropogenic activities, the anthropogenic pollution aerosols are mainly located near the surface. It's for this reason that we show surface

¹¹ Klingmüller., K., (2014) : Sensitivity of aerosol radiative effects to different mixing assumptions in the AEROPT 1.0 submodel of the EMAC atmospheric-chemistry-climate model, *Geosci. Model Dev.*, 7, 2503–2516

¹² Tulet, P.,(2005) : ORILAM, a three moment lognormal aerosol scheme for mesoscale atmospheric model. on-line coupling into the mesonh-c model and validation on the escompte campaign, *J. Geophys. Res.*, 110, D18201, doi:10.1029/2004JD005716

¹³ Tulet, P., (2006) : ORILAM-SOA: A computationally efficient model for predicting secondary organic aerosols in 3D atmospheric models., *J. Geophys. Res.*, 111, D19205, doi:10.1029/2006JD007152

¹⁴ Kocha, C.,. (2012). High-resolution simulation of a major West African dust-storm: comparison with observations and investigation of dust impact. *Quarterly Journal of the Royal Meteorological Society*, 138(663), 455-470.

¹⁵ Tulet, P., (2008) : The 7-13 March, 2006, dust 1121 storm over West Africa : generation, transport and vertical stratification., *J. Geophys. Res.*, 113, D00C08, doi:10.1029/2008JD009871

concentrations and surface wind fields in Figure 4. We agree with the referee 1 concerning the PBL height. As it is shown in Figure 1.1 (see next response), the PBL height over Europe is located in average to 1.2 km during this strong dust outbreak. The question about the vertical mixing is discussed in details in the following response.

Referee 1 : P56, L28: carbonaceous component extends from the surface to 5 km. . . . How is that possible? A potential upward mixing over Europe is simply impossible. So may be the carbon particles originated from Africa? The PBL over northern Africa can easily reach heights of 4-5km above ground. . . . Any comment?

Authors : This statement sounds strange. There is no statement in the text which indicate that the concentrations of primary aerosols simulated May 28 between 3 and 5 km asl result from a local development of the PBL. PBL (see TKE cross section below; the gradient mark the top of the PBL, Fig. 1.1) does not exceed the 1.2 km altitude on the area of the vertical section of Figure 6. The situation from 25 to 28 is marked by the passage of deep convective systems over the Mediterranean (see Bègue et al., 2012). The species simulated above the PBL are the residual pollution produced the previous days and coming from the southeast of France, as it was shown by Bègue et al (2012). The plume was transported vertically by convection resulting from the convergence of surface flows southeast and northwest.

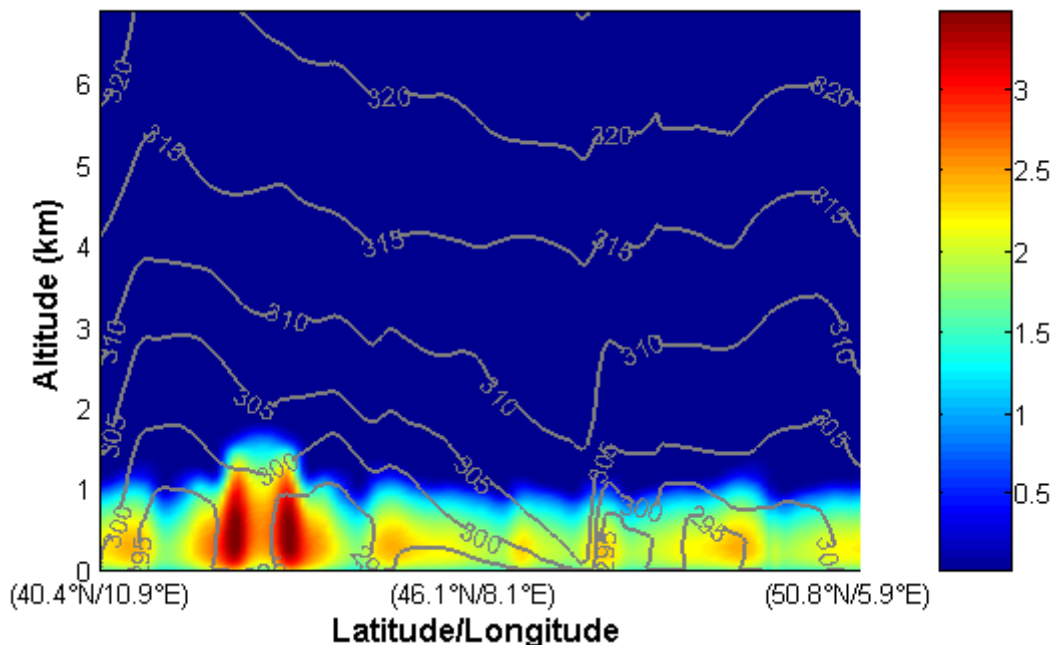


Figure 1.1 : Vertical cross section of TKE (Turbulence Kinetic Energy) with the potential temperature following the dust plume trajectory over Europe between (40.4°N ; 10.9°E) and (56.6°N ; 8.0°E) obtained from the simulation

Moreover, the model cannot simulate anthropogenic aerosol particles from Africa because the emission inventory used is limited to Europe. Only the desert dust aerosols are coming from Africa. A sentence has been added in the text to clarify this point. Note that this reinforces the interest to use mesoscale model in addition to observation which integrates the transport and the chemical processes during several days for reconstructing a 3D distribution of the pollution.

Referee 1: P57, L16: Now we PROPOSE to quantify the impact of this mixing on the hygroscopic and CC properties. That means Sect.4 is just speculative, you discuss hypotheses and consequences!

Authors : The purpose of the Section 3 is to demonstrate the presence of mixing zone between the plume and the pollution aerosols over Europe. In particular, the purpose is to localize the geographical region where the mixing took place from Meso-NH simulation. In Section 4, through the use of AMS observations and the numerical simulation, the chemical composition of the mixing is analyzed. In a second step, the calculation of the CCN concentration was presented at four supersaturation level in Section 4. Inside this section, the analysis is made combining airborne, ground-based observations and numerical simulation. Moreover, it is noticeable that this dust event is reproduced correctly by Meso-NH. Thus, the simulation can be used in trust for the analysis. As a consequence, we think that Section 4 is not really just purely speculative.

Referee 1 : P58: In this discussion you need to show polarization lidar profiles, i.e., profiles of the particle linear depolarization ratio to convince the reader that there is almost pure dust at heights above 1.5km, and how much dust in terms of backscatter, extinction, and derived mass concentration. If the particle depolarization ratio is 30%, you have pure dust, if the depolarization ratio is 15-25% you have an external mixture of dust and nondust components (marine particles, urban haze). So, please provide CALIPSO particle linear depolarization ratios (you may have to average 50 to 100 CALIPSO shots!) However, even by applying such an averaging, the profiles will be very helpful for the study here. You probably need to average the available cross and parallel backscatter coefficients (available in the CALIOP data base, and to calculate afterwards the particle depolarization ratio from the cross-to-parallel backscatter ratio)! I hope, that in addition CAELI lidar depolarization ratio profiles are available, too. Without such depolarization ratio profiles, the paper is based on rather weak grounds!

Authors : The discussion concerning the depolarization and color ratio were added in the revised manuscript. Small depolarization and color ratios derived from LNG measurements shows that the extinction coefficient was mainly due to the Saharan dust. It is seen in CALIPSO lidar observations that the particulate depolarization ratio is about 15 % at latitudes higher than 53° N and altitudes below 3km outside clouds. However, as we wrote previously no lidar observations at Cabauw are available during this dust event. As consequence, we based our discussion only on the CALIPSO and LNG observations. Furthermore, the CALIPSO particle linear depolarization ratio picture was added in the revised manuscript.

Referee 1: P60, L 14: considered as internally mixed, so again, this is speculation..

Authors : Traditionally, air quality models treats aerosols as an internal mixture. This assumption is based on the aerosols processes (such as gas-particles interactions, coagulation) drive to an internal mixing between species. Furthermore, numerous studies indicate that aerosols are generally closer to an internal mixture than an external one (expect when they are close to the sources): (Liu et al., 2005; Klingmüller et al., 2014).

Referee 1 : P60, L18: More than 50% of the mixture was made up of dust . . . in terms of mass concentrations? Again, please provide particle depolarization ratio profiles to support your speculation.

Authors : The discussion about the particle depolarization ratio was included in the revised manuscript.

Referee 1 : P60, L29: . . . was found to be consistent with previous studies of atmospheric processing of mineral dust. . . this statement is not just convincing to me. Consistency sounds nice, but is that sufficient to conclude that internally mixed dust particles were present and that the CCN ability of dust was significantly enhanced? Can you really draw such conclusions? You found partly rather large CCN concentrations above 10 000 per cm⁻³, is that really compatible with the occurrence of lofted dust from Africa. Such higher CCN number concentrations, may be found in Beijing, in China, but in lofted African plumes over the Netherlands????

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 25 May). 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 (Figure 1.2) and 25 000 cm⁻³ for the fine mode (Figure 1.3).

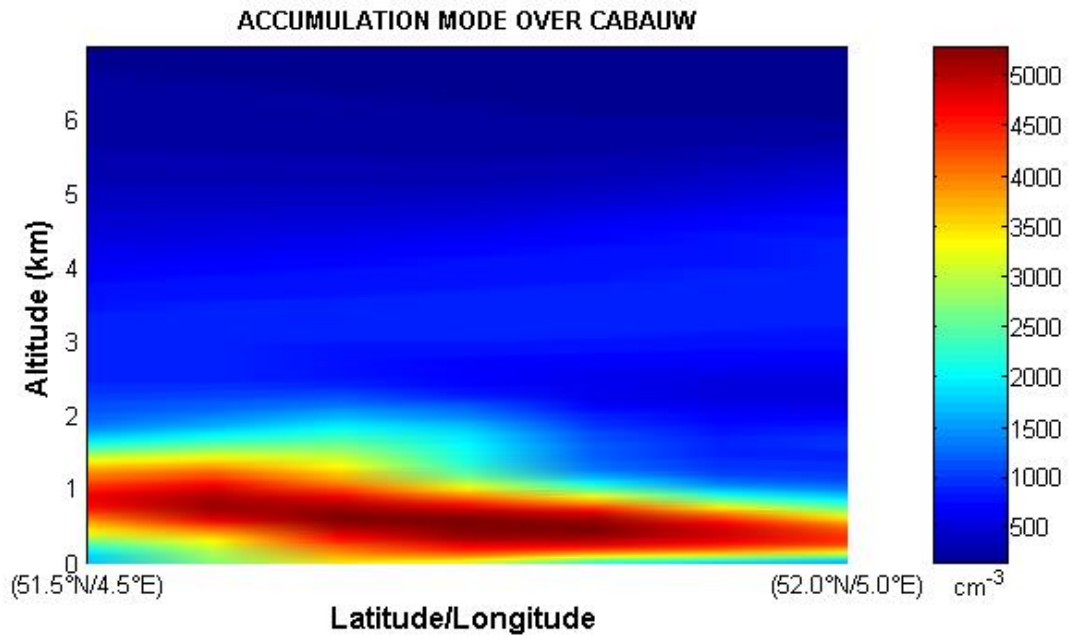


Figure 1.2 : Vertical cross section of the accumulation mode concentration (particle.cm^{-3}) over Cabauw obtained from Meso-NH simulation.

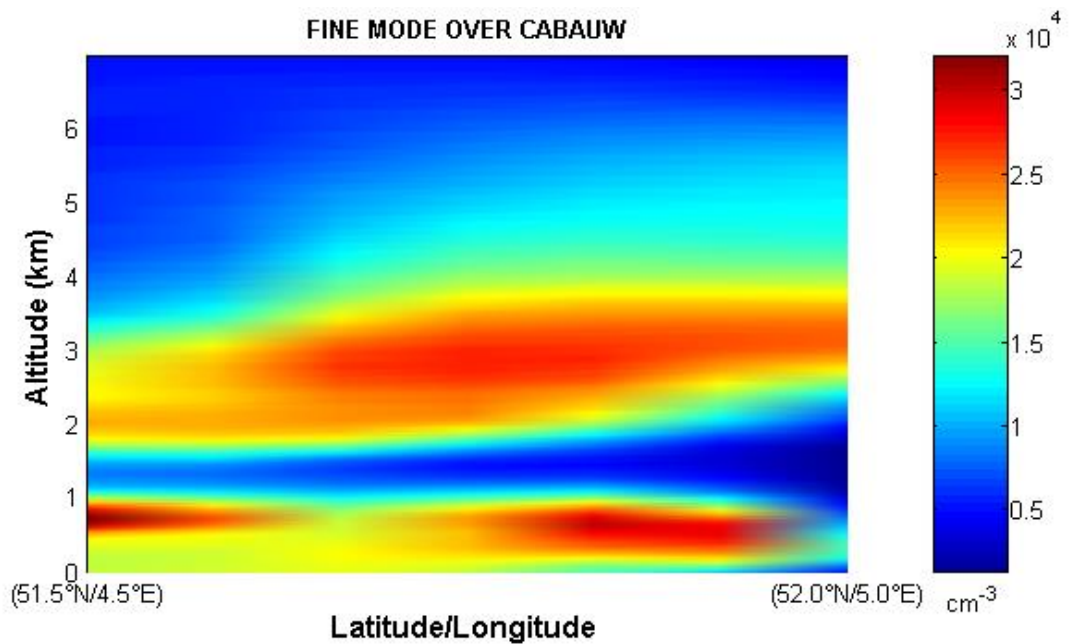


Figure 1.3 : Vertical cross section of the fine mode concentration (particle.cm^{-3}) over Cabauw obtained from Meso-NH simulation.

Referee 1 : P61, L3: It is reasonable to assume. . . , yes I understand this statement, but it remains speculation!

Authors : This conclusion is formulated from the simulation results which were found in agreement with the observation. As a consequence, we think that it is not really speculation.

Referee 1 : P61, L16: The results mentioned above confirm that the atmospheric processing of this Saharan dust led to an evolution of its hygroscopicity. Again, can you really state that? I would prefer a more tentative character of all your statements. Yes, you provide new arguments in this complex discussion. I appreciate that! That is the reason, that I want to see this paper published. But being in such good position, you can decide how to contribute to the discussion, less speculative. . . I would prefer. . .

Authors : In order to reduce the speculative nature of the discussion, the authors discussed about the depolarization ratio and the color ratio in the revised manuscript.

Referee 1 : P61, L20-25: It is interesting to read that chemical composition appears to play a significant role again. Dusek et al., 2006 stated that number concentration matters. . .,and chemical composition can be regarded as second order effect.

Authors : The reviewer is correct to mention that recent studies, such as Dusek et al., (2006), suggest that most cases composition plays a secondary role to size in establishing the ability of aerosol particles to act as CCN. Nevertheless, as it was previously mentioned, Roberts et al (2002) have shown that the role of chemical processes is not negligible for particles which are weakly hydrophilic. For this type of particles the coating processes with soluble materials allow to initiate the CCN activation. The coating processes are important for the insoluble particles such dust at the beginning of the CCN activation processes. In the manuscript, we would discuss about the importance to analysis for insoluble particles such dust how chemical change can influence their CCN activity. Furthermore, information concerning the size of the particles was added in the revised manuscript.

Referee 1 : P62, L7-10: You measured CCN conc. of 80 cm⁻³, and 700 to 900 cm⁻³ higher up, and the measurements show also, that the total number concentrations (CN) were only between 400 to 1800 cm⁻³ for these cases? So, you had almost marine pristine conditions, but on the other hand a comparably high CCN concentration? I am puzzled by these numbers. . . and later on you found up to CCN up to 14000 cm⁻³ (for 0.63% super saturation) during the same dust episode. I am really confused? All this points to the idea, that anthropogenic pollution was the main contributor to CCN, and not the dust. .

Authors : We thank the reviewer for this relevant comment. Firstly, we note that the CN concentration measured on board ATR-42 range from 800 to 4500 particles per cm⁻³ (Figure 1.4).

Secondly, it is also worth noticing 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.

Thirdly, it can be observed that the CN evolution is fairly reproduced by Meso-NH (Figure 1.4). Furthermore, our 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.

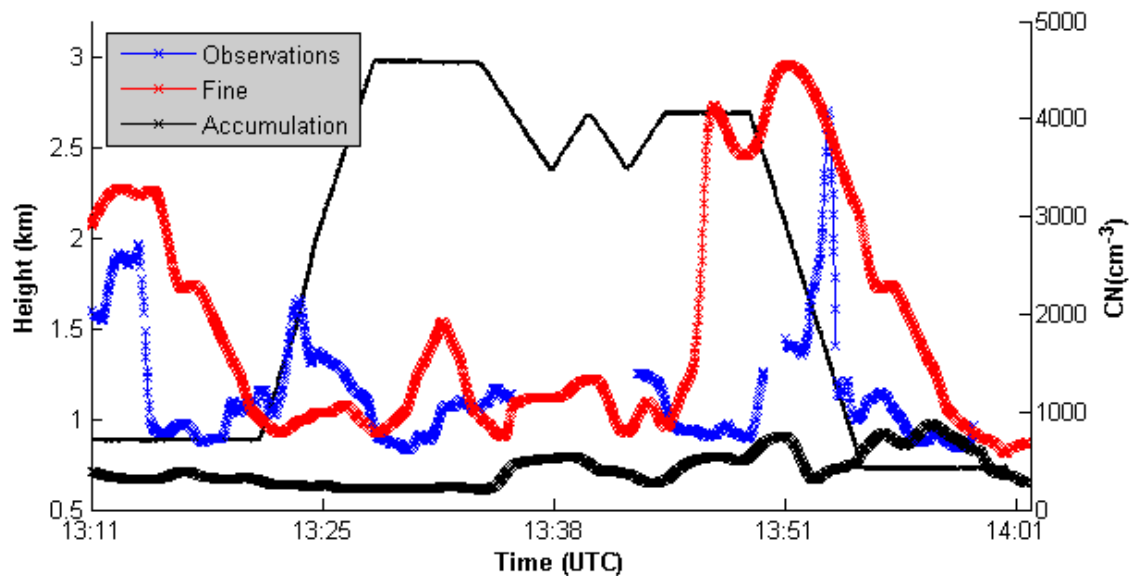


Figure 1.4 : Evolution of the CN concentration measured (blue line) and simulated (red and black line) by Meso-NH between 13:11 and 14:01 on 30 May 2008 over the Netherlands between (52.36°N/6.35°E) and (51.83°N/5.11°E).

Referee 1 : P63, L2: CCN/CN ratios of 15% found during AMMA sounds much more realistic, and what were the CCN and the CN numbers over the AMMA area? Please state for better comparison.

Authors : The CCN number at 0.6 % supersaturation reported by Crumeyrolle et al (2008) is ranging between 300 and 100 cm⁻³ whereas the CN number is ranging from 2100 to 900 cm⁻³. This information was added in the revised manuscript.

Figures...

Figure 1: P, C, M, H must be explained in the figure caption. Copy and paste from Figure 4 caption...

Authors : An explanation is given in the revised manuscript

Figure 2: Poor CALIPSO signal quality, can we be sure, that below the dense (more yellow) upper parts of the dust plumes there is really less dust, or is this just also a shadowing effect as caused by several clouds in the same figure. . . . When the Klett formalism is used in the forward mode, the solutions can be very erroneous with increasing range (with lower and lower heights).

Authors : No clouds are detected on May 28th, except very north (52° N) so that results are not really perturbed by clouds. One can see that on most of the track (up to 47° N) pure dust aerosols are detected.

It is right to mention that When the Klett formalism is used in the forward mode, the solutions can be very erroneous with increasing range (with lower and lower heights). However, a specific procedure is applied on CALIPSO processing which limits instability (Young and Vaughan¹⁶, 2009)

Figure 3: Again a lot of shadowing by clouds in the right part of the CALIPSO color plot.

Authors : On May 29th, high level clouds prevent to observe below, although at 48° N one can see a cloud (high backscatter and depolarization) but below 45° N and above 50° N, aerosol particles can be detected which depolarization remains lower than pure dust.

Figure 5: So these are also surface data. . . , far below the dust layers. . . , the period is up to 29 May, not up to 31 May as stated.

Authors : Although the data do not cover all the study period and far below the dust layers, we think that this figure is helpful to describe the temporal and spatial evolution of the anthropogenic aerosol over Europe. Moreover this figure is used to illustrate that your model is doing a fairly good job on the simulation of the anthropogenic aerosol over Europe. The good period was added in the revised manuscript.

¹⁶ Young, S. A., & Vaughan, M. A. (2009). The retrieval of profiles of particulate extinction from Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations (CALIPSO) data: Algorithm description. *Journal of Atmospheric and Oceanic Technology*, 26(6), 1105-1119.

Figure 6: Strange layering of BC and OC mass concentrations up to 5 km in the beginning (28 May). How can that be explained? African origin? At the same time there must be rather stable layering at the base of the dust layer between 1.5 and 2 km height. So, I do not understand this phenomenon.

Authors : An explanation is given previously (see page 9)

Figure 7: All the blue areas in the upper plot (lidar) below the yellow and red areas higher up are areas without data, the same is true for the blue areas below the cloud layer (red area around 700-1000m height). That must be clearly stated in the caption.

Authors : This information was added in the revised manuscript

Figure 10: Observations show strong jumps when the aircraft is moving up and moving down. Is that a systematic effect (bias)? Air flow corrections, effects caused by normalization to standard pressure and temperature conditions?

Authors : The ATR-42 aerosol inlet is an isokinetic inlet, controlled for isokinetic sampling (automatic and manual control mode: due to altitude changes and related pitch angle). When looking at the SMPS data (90 s of time resolution, compared to 1s for CN counters) we clearly see the particle plume that the aircraft crossed during its climb between 1326 UTC and 1329 UTC for example. Likewise during the descent! Particle plumes that are more or less filamented, in particular at its' borders, can create considerable fluctuations in the CN concentrations, particularly seen in aircraft data, that are subject to the aircraft speed and corresponding aerosol heterogeneity in the aerosol concentrations (10 CN measurement data points over 1km).