Atmos. Chem. Phys. Discuss., 14, C9462–C9470, 2014
www.atmos-chem-phys-discuss.net/14/C9462/2014/

Atmospheric Chemistry and Physics Discussions

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 #1

Received and published: 24 November 2014

General

The paper deals with long-range transport of desert dust towards northern Europe and the potential change of the hygroscopic properties of aged dust by coating by anthropogenic substances which in turn enhances the potential of dust particles to serve as (better) CCN. High-quality airborne observations of aerosols obtained during a field campaign are included in the study which also uses aerosol-focusing simulation models to interpret the observations. The topic is state-of-the-art, the methods and approaches applied are reasonable, the paper is well written, and thus appropriate for ACP.

Nevertheless, I have a long list of remarks. Most of the argumentation in the paper is to my opinion speculative, i.e., trustworthy observational evidence for the main hypothesis can not be provided. Therefore the discussion should be more sensitive, indicating the speculative character of the message (of all the conclusions made) of the publication. My comments may help to reflect the argumentations and thus may help to improve this good paper..

Details:

Abstract:

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?

Introduction:

Page 27040 = P40, L1 = line 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.

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

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. . ...

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!

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. . .?

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.

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

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

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. . ..!

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 modelling approach is left to corroborate your basic hypothesis of dust coating. . . I my view ok? 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?

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

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!

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 pitty if not during such an important campaign.

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 can not be based on solid grounds of measurements. Should be clearly stated!

P52, L23: Bangaert. . .

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.

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?

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

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 non-dust 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!

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

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.

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 10000 per cm-3, is that really compatible with the occurrence of lofted dust from Africa. Such higher

CCN number concentrations, may be found in Bejing, in China, but in lofted African plumes over the Netherlands????

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

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. . .

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.

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 to1800 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. . .

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.

I hope my impact leads to a more careful discussion with save statements, I would like to see a clear separation between solid observational facts and all the findings that

are based on the consistency between observations and state-of-the-art simulations. Many of the conclusions are to my opinion speculative. Unfortunately, the shown lidar profiles are rather noisy and not just trustworthy. Depolarization ratio height profiles would be very important for the entire discussion regarding the degree of dust mixing with other aerosols, but not shown.

To the figures. . .

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

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).

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

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.

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 2km height. So, I do not understand this phenomenon.

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.

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?

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