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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Effect of surface reaction on the cloud nucleating properties of mineral dust: AMMA aircraft campaign in summer 2006" *by* A. Matsuki et al.

Anonymous Referee #2

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General comments

The paper addresses the question of mineral dust heterogeneous processing with or without clouds within the scope of AMMA, which is in the scope of ACP. New data material is presented from samples collected during an AMMA special observation period. Electron microscopy was used to analyze a significant number of particles. The authors conclude from their data on the processing history of the particles. While the overall conclusions are justified, some details remain challengeable. The used analytic methods are established, but a discussion of measurement uncertainties is missing (though a detailed error discussion would not be in the scope of this publication). A

relevant number of related works is properly credited, title and abstract reflect the paper's content. The overall structure is fine, however, some paragraphs found in the results section should be rearranged into the methods section. In general, the use of the language is adequate.

Specific comments

"General specific comment": It is surely not within the scope of this work (and not to be assessed by the methods used), but it should be mentioned (maybe in the introduction), that, of course, also organic materials play a (unknown?) role in the dust-biomass burning-cloud interactions. Their presence might bias the results of the inorganic interpretation.

(notation in page number, line(s))

1799, 2-3: Could be more specific, e. g. "The Saharan desert with the Sahelian region is the largest global source".

1799, 11-14: Omit this sentence, as it contradicts the next paragraphs, in which works are listed which were dealing with dust-cloud-interactions.

1799, 28: Dust composition is also dependent on the geological situation, of which the soils derive.

1801, 1: Maybe some introductory remarks should be given on the specifics of the region, where the campaign was performed (why there?) or reference to an AMMA overview paper, if there is any.

1801, 15: Insert reference for the CAI inlet

1801,18: It is quite unclear at this point, why each cloud element should release (exactly?) one residual particle, though it doesn't seem to have significant impact on the conclusions.

1802, 2: Surely, the sampling condition was not 1013 hPa: is it a mass flow rate given

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(which would impact the impactor's cutoff sizes with changing pressure) or a volume flow rate?

1802, 26-29: How was the quantification of the elemental concentrations done? By the manufacturer's software (with standards or standard-less), or by a custom model?

1803, after 12: Later in the manuscript, a criterion table for particles classification is given. That together with the according text should be incorporated into the methods section here.

1803, 16-17: change to "Presence of a _silicate_ dust particle..." and exclude the Al content for quartz particles (which is grouped into the silicates later)?

1803, 19: Looking on Fig. 1, the particles referred are very rounded, but not exactly spherical in a geometric way, like for instance fly ashs can be (in the limits of measurement error). Maybe this could be misinterpreted, e.g., by people doing optical calculations? Would spheroidal (or nearly spherical) be a better expression?

1804, 17-24: Rephrase for better understanding.

1804, 25-27: Does that mean that they are dried droplets? Or are they still liquid or ductile in the electron microscope?

1805, 8: Insert a reference for the meteorological models or give some properties and limitations, especially with respect to 1806, 20-25 where their validity for convective cases is criticized.

1805, 16-1806, 8: Integrate this part into the method section; at least the instrumentation should not occur in the results section for the first time.

1806, 13-27: The conclusion onto the rather local particles sources seems to be rather weak: You mention model weaknesses for convective events and 28 % of Ca-rich minerals are found, which rather point to the Saharan region, as the Sahelian does not have very high Ca contents in soil (i. e., Claquin et al. 1999, JGR 104, 22243-22256,

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plate 1).

1807, 2-3: How was determined whether particles were of biomass burning origin? What is the possible source of the sulfate particles?

1807, 7: How many particles were analyzed in total?

1807, 12-19: Move this description to the method section.

1809, 18: Gypsum is CaSO4 * 2 H2O.

1809, 22: Is there a better way to cite this (as "Tobo, personal communication")?

1810, 1-3: Clear-sky conditions are referencing the time of sampling, but the aerosol particles definitely originate from the soil, so they have already a certain age (probably days). From the given data, we do not know, whether they have been cloud-processed before. Thus, the conclusion of dry uptake of HNO3 in this case cannot be drawn on base of the given data.

1810, 21-23 and Fig. 6: The figure shows a trend of increasing S+Cl atomic fraction for increasing humidity. Taking into account the uncertainties in quantification of the elemental composition and uncertainties in the relative humidity (we do not know, for fractionated clouds, whether a particle comes from inside or outside a cloud) it is impossible to conclude more from this 25 scattered data points. Everything else is speculative, especially the "excellent" exponential relationship.

1811, 16: What about the nitrate? It should also add to this competition.

1812, 7-1813, 9 and Fig. 7: On which level of significance it can be excluded, that the values of Aug 06/14 are the same as the average, taking into account the standard deviation given in the figure, the (undocumented) uncertainties derived from counting statistics and the errors in elemental quantification? The detailed discussion appears over-interpreted with respect to only two data points while in fact no conclusion can be derived. Although it looks like a lot of work was put into this question, the section

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should maybe be shortened.

1815, 27-1816,8: The work of Dusek et al. is not well comparable to this work, so there is not really a contradiction. Dusek et al. investigated the total number population of aerosol and, thus, concentrated on the submicron fraction close to the CCN activation border (see also the size distribution plots in their work). In the submicron regime, the particle size is indeed the main (though not the only, as Dusek et al. also state) controlling factor (which we should have suspected since Köhler). In contrast, this present work focuses on details of the supermicron particles, where chemistry obviously plays a role. However, in the present work it is not quantified, how many of the mineral dust particles remain interstitial between the cloud droplets. So we do not know whether the influence of chemistry on activation is important for a large number fraction of the supermicron particles. The argumentation around the supersaturation is vague ("it is said").

1823: If "gypsum" really was classified as "calcite", shouldn't this have a significant bias on the result interpretation, as calcite probably is at the beginning of a processing chain, and gypsum rather at the end? How frequent was gypsum observed? Are there minimum Al and Si concentrations for a particle to be sorted into group 2 or 3?

Technical corrections and phrasing suggestions

1798, 2-9: Shorten sentences.

1798, 10: Omit "Both" and "later"

1798, 11: "using a transmission" and "and a scanning"

1798, 12: "with an energy" and "spectroscopy"

1798, 20: "A surprisingly"

1798, 22: "hygroscopicity, resulting from their reaction"

1798, 24: "there are sufficient"

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1800, 12: change "Implication" to "A result"

1800, 16: "point of their emission"

1800, 24-28: Shorten

1801, 7-8: Omit "In order to ... acting as CCN"

1802, 15: "flight, the particle-laden"

1802, 20-22: "About 10 randomly chosen fields of view per sample were imaged at a constant magnification (3000x) at 120 kV acceleration voltage."

1803, 2-3: "and low atmospheric"

1803, 3-5: "Samples failing to have more than 30 analyzed particles (6 out of 37) were excluded from this work due to the lack of representativeness."

1803, 8-9: "..., sample substrates were prepared using the above-mentioned grids with additional nitron (C20H16N4) coating."

1803, 21: Omit "Other"

1804, 1: "and different clay minerals"

1804, 6: Omit "found in this study often"

1804, 9: "collodion film showed a spherical/spheroidal"

1804, 11-13: "At least 60 % up to 90 % of the Ca-dominated particles on the reactive nitron reagent film contained NO3-. It becomes evident ..."

1804, 18: "of the atmospheric irregular dust particles"

1804, 18-24: reword

1805, 29: Omit "probably due to the interstitial particles"

1806, 1: "counted in the gaps of the cloud"

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Interactive Comment

1807, 7: "1495 of the total supermicron particles (nnnn) were ..."

1807, 15-16: "Particles enriched in Ca were termed Calcite (CaCO3) and those enriched in Ca and Mg Dolomit (CaMg(CO3)2)." (as it is found in the table)

1808, 15-16: Omit "as geologists ... silicates)."

1809, 1: "roll" –> "role"

1809, 10-11: "Cl/Na ratio is"

1809, 21: "The possibility"

1813, 24: "pathway (3.) may"

1829: "Characters (a) to (e) show averages for the peculiar air-mass trajectories identified in Fig. 4" (?)

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 1797, 2009.