

Interactive comment on “Temporal and spatial variations of aerosol physical and chemical properties over West Africa: AMMA aircraft campaign in summer 2006” by A. Matsuki et al.

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Response to M. Pósfai (Referee2)

First of all, we would like to thank M. Pósfai for carefully going through the article, as well as providing very constructive comments and suggestions for improving our English. Please find our answers to each of your comments below, and now the article is revised accordingly.

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Major comments:

*This study describes the results of the physical and chemical characterization of
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aerosol particles and cloud elements collected before and after the onset of the monsoon period in West Africa. Aerosol particle concentrations, size distributions and their CCN activities were measured, along with the analysis of individual particle compositions and morphologies using transmission electron microscopy. The combination of bulk size and volatility measurements with TEM single-particle analysis is an extremely effective, albeit labor-intensive procedure for obtaining comprehensive information on the particles. The paper could serve as a good reference for the properties of aerosol particles that dominate in the distinct atmospheric layers that are typical in West Africa. It is a surprising result that particles in the 'monsoon layer', arriving from the ocean, are less CCN-active than particles in the free troposphere and in the Saharan aerosol layer. Another noteworthy result is the identification of a significant fraction of submicron dust particles. The paper deserves to be published in ACP and needs only minor adjustments.

- Volatility measurements. The results of volatility measurements are given for the 20-300 nm fraction. Since volatility data are mentioned to be available for larger sizes, it would be useful to show them.

See also response to reviewer 1:

The fitting of the size distributions has been extended to 4 lognormal modes. Using these fitted size spectra we present now volatility behavior of the entire range of submicrometer aerosol particles up to 1 μm as proposed by the reviewer 1. Indeed the SMPS and OPC instruments connected to the thermo desorption column measure the response of the entire size spectra to elevated temperature. The major information of figure 5 is that submicrometer aerosol is much less volatile during dry season (SOP1), as compared to SOP2a1 (monsoon onset) and SOP2a2 (monsoon fully developed). The higher volatility of the aerosol after the onset of the monsoon seems to increase from SOP2a1 to SOP2a2. The volatile aerosol stems particularly from the 100-300nm accumulation mode particles. Nevertheless, also during SOP2a1 and 2a2, a fraction of the particles seems to be as refractory as almost the entire submicrometer aerosol

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population during SOP1. A look at the volatility behavior of aerosol particles in the size range 1-2 μm does not show the same volatility trends as observed for submicrometer aerosol. Indeed, these supermicrometer particles are much more refractory with respect to temperature (and thus, less volatile) and show no clear trends when comparing periods SOP1, 2a1, and 2a2. The latter result is certainly due to the fact that supermicron particles are predominantly composed of non-volatile coarse mode mineral and sea salt particles (Matsuki, 2010).

- Identification of particles from biomass burning (p4478, top paragraph). C and O were not analyzed, and only the K and S contents of the particles were taken as evidence for a biomass burning origin. This procedure may be slightly misleading, since many biomass smoke particles, particularly tar balls, do not contain significant K or S. On the other hand, some non-smoke sulfates can also contain K. The authors mention the volatility of some K/S-bearing particles, which may also indicate that the analyzed particles are primarily sulfates that may not necessarily originate from biomass smoke. (According to my own experience, organic particles from biomass burning are not spectacularly volatile in the TEM – see, for example, Pósfai et al. (2003), JGR 108, D13, 8483.) Thus, the results should be interpreted with caution, and there may be some chance that the number of particles from biomass burning is overestimated.

We do see peaks of C and O in our spectra, but relied more on the detection of potassium as evidence for a biomass burning origin since the collodion substrate itself contains some C and O, and due also by the difficulties related to the quantification of such lighter elements. Our definition stems simply from the idea that excess fine potassium not attributable to mineral dust or sea salt is an indicator of biomass burning (e.g. Andreae, 1983, Science, 220). Although other combustion processes (e.g. fossil fuel burning and waste incineration) may also result in the formation of non smoke sulfates containing K, considering the remoteness from major industries, we believe our assumption may not lead to a serious misinterpretation. We did find some particles resembling tar balls, but as mentioned in your own work, distinction between tar balls

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and other biomass burning particles (e.g. organic) are sometimes arbitrary. Our tar ball like particles are also included in the biomass burning fraction since they also contained slight amounts of K and S. We avoid stating that the biomass burning particles are “enriched” in K but instead they “contain” K. We also mention about the potential overestimation of biomass burning fraction related to our definition.

We were not actively searching for biomass smoke or haze conditions and our results may be more representative of background aerosols over the studied region. Now, we are convinced that the susceptibility to the electron beam mostly originates from the higher sulfate content found in many of our particles as compared to more freshly emitted biomass burning particles found in other reports. In the revised version, we mention that the biomass burning particles are initially stable under electron beam by referring to your work, so the susceptibility of many of our particles suggests presence of volatile organic acids or significant mixing with sulfate.

- Concerning the low volatility of the particles in the monsoon layer, could at least some of the particles come from urban emissions from the large coastal cities in West Africa? Particles from urban pollution are, in general, more hydrophobic than organic particles from savanna or forest fires. Are “biomass burning” particles meant to include those from household emissions, which could be also quite significant in that region? Some discussion on these points would be welcome.

These are very interesting points. Although we couldn't find features that can be directly linked with urban emission, we cannot exclude the possibility that some of the particles come from the major coastal cities (e.g. Abidjan, Accra, Cotonou, Lagos, Lome) since the accelerated monsoon flux and decoupling of SAL and PBL together might favor more direct transport of particles from the coastal regions. At the same time, there is a significant zonal variability in the land cover along the way (forests, Savanna, cropland and Sahel), and it is very challenging to determine the sources from forest fires, agricultural burning or household emissions. Our term “biomass burning” therefore should be interpreted in broader sense such that there may be significant con-

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tribution from household emissions. We added few lines describing these possibilities in the conclusion.

Minor (mostly technical) comments: Abstract, 3rd line from bottom: 'after the onset of the monsoon period'

Corrected.

4465, line 22: 'spatial variation in the aerosol..'

Corrected.

4466, line 24: 'to be contrasted with...'

Corrected.

4466, line 26: delete 'the' after 'aircraft'

Corrected.

4467, line 23: 'descents'

Corrected.

4468, line 8: 'when the monsoon was at its peak'

Corrected.

4468, line 14: 'evaporation'

Corrected.

4469, line 13: 'one CCN spectrum'

Corrected.

4470, top paragraph: 'samples were collected' instead of 'made', in two places

Corrected.

4470, line 18/19: 'spectrometer' instead of 'spectroscopy'

Corrected.

4471, line 20: Gulf of Guinea

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Corrected.

4472, line 8: 'in Niamey in 2006'

Corrected.

4472, line 23: distributions

Corrected.

4472, lines 24/28: convoluted sentence, please rewrite

The sentence now appears in the revised manuscript as:

“Since the trajectories arriving in the highest altitude range (4-6km) showed markedly different (vertical) transport patterns as compared to those in the underlying SAL (Fig. 3), the related aerosol size distributions were averaged separately and denoted as those representing the “free troposphere”.”

Table 1: Particle sizes should be in nm, not μm

Corrected.

4474, lines 18: 'overlying' (also in many places elsewhere in the paper), and 'If any different, then..'

Corrected.

4474, last paragraph: It may be useful to state here again that the volatility measurements were performed on particles in the 20 to 300 nm fraction (i.e., most mineral grains were not included), otherwise it is difficult to understand how the volatility of particles in the SAL layer could be larger than in the monsoon layer.

Now the volatility measurements accounts for sizes 20 to 1000 nm in the revised manuscript. The last sentence in the paragraph now appears in the revised manuscript as:

“Since the particle volatility shown in Fig.5 is limited to 20-1000nm range, this finding suggests that the fine particle composition may be significantly different between the monsoon layer and SAL especially during the wet period (SOP2a2).”

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4477, line 3/4: 'understand' or 'study' instead of 'to confirm or rule out', and please delete 'supposed'

Corrected.

4477, line 11: 'in a companion paper'

Corrected.

4477, line 15: aerosol

Corrected.

4477, line 18: 'consisting' instead of 'constituting'

Corrected.

4477, line 23: 'showed characteristic morphology.'

Corrected.

4477, line 24: comma instead of period

Corrected.

4477, line 26: '...it was difficult to distinguish the mineral species from the...'

Corrected.

4479, lines 14/16: meaning of the sentence unclear

The sentence now appears in the revised manuscript as:

"Whereas large compositional difference between the monsoon layer and SAL as suggested by the particle volatility measurements during SOP2a2 (Fig. 5) also is confirmed by the remarkably large fraction of biomass burning particles in the monsoon flux (Fig. 9)."

4480, line 25: On the contrary

Corrected.

4481, lines 3/4 and 6/7: meaning unclear

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This part now appears in the revised manuscript as:

“This is because fresh particles contain much less secondary organic acids (e.g. oxalate, formate, acetate) or secondary inorganic species (e.g. ammonium, sulfate, nitrate), as compared to aged ones (Reid et al., 2005). The volatilized volume is largely dependent on the abundance of such secondary species. Therefore, small volatilized volume fractions observed in the monsoon layer provide further indication that the dominant biomass burning particles were still relatively fresh.”

4481, line 10: 'which are believed..'

Corrected.

4481, lines 15/18: Are CCN activity data for the dust fraction available for this study? If yes, it might be worth showing them.

It would be very interesting if we can show them. Unfortunately, CCN counter was not available during the dry period when dust particles were predominant.

4482, line 5: analyses

Corrected.

4482, line 11: proceeded

Corrected.

4482, line 17: compositions

Corrected.

4482, line 24/25: please reword sentence

The sentence now appears in the revised manuscript as:

“The mineral dust fraction gradually decreased as the season changed from dry to monsoon period, and eventually the biomass burning and sulfate particles together became dominant.”

4483, line 6: 'hydrophobic' instead of 'hydrophilic'

Corrected.

4483, line 14: 'how to explain that the particle chemistry.'

Corrected.

4484, line 1: compositions

Corrected.

4484, line 8: delete 'out'

Corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 4463, 2010.

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