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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Aerosol characteristics and particle production in the upper troposphere over the Amazon Basin" *by* Meinrat O. Andreae et al.

**Anonymous Referee #3**

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## Summary

The manuscript presents the results of the ACRIDICON-CHUVA aircraft campaign over the Amazon basin in which the characteristics of aerosol particles were determined in 11 flights that reached the upper troposphere (altitude > 8 km). Comprehensive instrumentation on board of the aircraft collected data of aerosol particle size, number, and composition, in addition to gas phase composition and cloud microphysical properties and meteorological data. The authors analyze and interpret the measurements with respect to the formation mechanism of aerosol in the upper troposphere and with regard to the upper troposphere as an aerosol source for the Amazon basin.



The introduction provides the background and references on upper tropospheric aerosol measurements, discusses the upper troposphere as an aerosol source, and gives a brief overview of aerosol nucleation mechanisms. The "Methods" section documents and explains the instrumentation used in the aircraft campaign, the data collection and analysis procedures, and the analysis of air mass history using back-trajectory modeling. The "Results and Discussion" section guides the reader systematically through the observations and the insights they provide. First the synoptic meteorological situation and air mass history in the Amazon region during the campaign are discussed, followed by a characterization of the atmospheric chemical composition. The authors then present and discuss the vertical distribution of aerosol particle concentration and the differences between upper- and lower-tropospheric aerosol in terms of different particle properties. Very high number concentrations and a composition dominated by organics of upper tropospheric aerosol particles stand out among the numerous findings. The authors proceed to characterize the relationship of deep convection, high aerosol number concentrations, and new particle formation in the upper troposphere. A special case of a deep convective cloud that interacts with a (fresh) biomass burning plume with new particle formation in the cloud outflow is discussed separately. Finally, a conceptual model for the aerosol life cycle in the Amazon basin is proposed. In the conceptual model, deep convection lifts boundary layer air into the middle and upper troposphere, where it is released. Preexisting aerosol particles are removed by cloud scavenging, while aerosol nucleation precursors molecules, likely biogenic volatile organic compounds, are released by the convective outflows. These nucleation precursors are oxidized to molecules that drive aerosol nucleation, and account for the very high concentrations of aerosol particles observed in the middle and upper troposphere. Large scale subsidence and downdrafts surrounding deep convection would then bring these newly formed aerosol particles into the boundary layer. The upper troposphere in the Amazon basin would therefore, in particular in pristine conditions, serve as an important and possibly dominant source of aerosol particles.

**Comments**

The present work gives a comprehensive account of aerosol properties in the atmospheric column from the boundary layer to the upper troposphere over the Amazon basin, facilitated by a well-instrumented aircraft with high-altitude and long distance capability. The results support the transport of boundary layer air containing aerosol nucleation precursors by deep convection into the upper troposphere, with subsequent new particle formation in convective outflow and growth involving organics as a key aerosol source in the region. The work is an important contribution to the understanding of the aerosol life cycle over tropical landmasses.

The text is written in an accessible and organized style with an appropriate amount of detail. A thorough description of the campaign, the instruments, measurements, procedures, and results is given. Limitations of the measurements (such as the lower aerosol size measurement cutoff of 20 nm, which hampers identification of new particle formation) and associated uncertainties in the interpretation of the results are well accounted for in the discussion. Figures are informative and integrate themselves seamlessly into the narrative.

The manuscript is nearly ready for publication, except for several points that I would like the authors to address.

1) The conceptual aerosol life cycle model in which convection lifts boundary layer air with nucleation precursor molecules into the upper troposphere, where nucleation takes place in the detrainment zone, followed by aerosol growth and descent through the troposphere into the boundary layer, has been to the best of my knowledge first formulated by A. D. Clarke (1992) based on observations and supported by subsequent investigations (e.g. Clarke, 1993; Clarke et al., 1998). These measurements were carried out over the oceans and implied sulfuric acid, likely from dimethyl sulfide and sulfur dioxide oxidation, as the molecule driving aerosol nucleation. Clarke and Kapustin (2002) wrote that "the tropics commonly have low aerosol mass but very high



number concentrations in the upper free troposphere (FT) that appear to form from sulfuric acid (nucleation) in convective regions and near cloud edges. These age and subside to become effective cloud condensation nuclei (CCN) when mixed into the marine boundary layer." This conceptual model is applied in the present manuscript to a pristine tropical continental region with organic molecules as the likely nucleation precursor. References to works by Clarke et al. and their context do, however, not provide due credit. I would like to ask the authors to add a brief paragraph in which their analysis and findings are placed into the context of this previously developed aerosol life cycle model and which provides credit to A. D. Clarke for its development with the below references.

Clarke, A. D., Atmospheric nuclei in the remote free troposphere, J. Atmos. Chem., 14, 479-488, 1992.

Clarke, A. D., Atmospheric nuclei in the Pacific midtroposphere: Their nature, concentration, and evolution, J. Geophys. Res., 98(D11), 20633-20647, doi:10.1029/93JD00797, 1993.

Clarke, A. D., J. L. Varner, F. Eisele, R. L. Mauldin, D. Tanner, and M. Litchy, Particle production in the remote marine atmosphere: Cloud out-flow and subsidence during ACE 1, J. Geophys. Res., 103, 16,397-16,409, 1998.

Clarke, A. D. and Kapustin, V. N.: A Pacific aerosol survey. Part I: A decade of data on particle production, transport, evolution, and mixing in the troposphere, J. Atmos. Sci., 52, 363-382, doi:10.1175/1520-0469(2002)059<0363:APASPI>2.0.CO;2, 2002.

2) Line 78-79: " ... or upward into the Tropical Transition Layer (TTL) and the lower stratosphere (Weigel et al., 2011; Randel and Jensen, 2013) ..."

Please add a reference to Brock et al. (1995), who identified the role of upper tropospheric aerosol nucleation for stratospheric aerosol concentrations.

C. A. Brock, P. Hamill, J. C. Wilson, H. H. Jonsson, K. R. Chan: Particle Formation

in the Upper Tropical Troposphere: A Source of Nuclei for the Stratospheric Aerosol, Science, 1650-1653, 1995

3) Line 719-723: "Interestingly, these concentrations over the Amazon Basin are only slightly higher than the values measured over the tropical Western Atlantic during the Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment (SALTRACE), June/July 2013: ca. 0.2 ug m-3 in the LT and ca. 0.001 ug m-3 in the FT (Schwarz et al., 2017), which suggests that a significant fraction of the rBC is entering the basin by long-range transport from Africa."

It is not clear that one can make this statement simply by comparing BC mass concentrations from two campaigns that are more than year apart, without analyzing transport and the contribution of local BC sources. Can you add a supporting discussion or evidence that would corroborate the point, or instead, formulate the statement hypothetically?

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