Response to Reviewer 3

We thank the reviewer for his/her careful review and positive and constructive comments. The reviewer comments are in plain font, the responses in *Italics*

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.

As submitted, the paper contained 15 references to the work of Clarke and coworkers. In accordance with the reviewer's suggestion, we have added the suggested four new references to Clarke's work and included the following paragraph in the introduction: "Based on observations over the remote Pacific and supported by extensive subsequent investigations, Clarke and coworkers proposed an 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 (Clarke, 1992; 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."" In section 3.7, we are contrasting our model to that of Clark and other workers in several important aspects, e.g., the role of organics vs sulfates and the mechanism of downward transport. See also our response to the first comment by Reviewer 2.

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

Done.

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?

We have a considerable amount of evidence for the transport of BC and other aerosol constituents from Africa to the Amazon Basin from several campaigns. Recently, we have published a modeling study on this topic (Wang et al., 2016). We are currently preparing a paper in which we are documenting the transport of biomass smoke from Southern Africa to the Amazon during ACRIDICON-CHUVA. This has also been observed in previous campaigns, e.g., Andreae et al. (1994). We have added the following text to section 3.4.4:

"Transport of biomass smoke containing BC and other constituents from Africa to South America has been documented previously, e.g., from Northern Africa during the wet season (Talbot et al., 1990; Wang et al., 2016) and from Southern Africa during the dry season (Andreae et al., 1994). A detailed study on the transport of Southern African aerosols to the Amazon during ACRIDICON-CHUVA is in preparation and will be published elsewhere."

Clarke, A. D., Atmospheric nuclei in the remote free troposphere: J. Atmos. Chem., 14, 479-488, doi:10.1007/bf00115252, 1992.

Clarke, A. D., Atmospheric nuclei in the Pacific midtroposphere - their nature, concentration, and evolution: J. Geophys. Res., 98, 20,633-20,647, doi:10.1029/93jd00797, 1993.

Clarke, A. D., Varner, J. L., Eisele, F., Mauldin, R. L., Tanner, D., and Litchy, M., Particle production in the remote marine atmosphere: Cloud outflow and subsidence during ACE 1: J. Geophys. Res., 103, 16,397-16,409, doi:10.1029/97jd02987, 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., 59, 363-382, 2002.

Wang, Q., Saturno, J., Chi, X., Walter, D., Lavric, J. V., Moran-Zuloaga, D., Ditas, F., Pöhlker, C., Brito, J., Carbone, S., Artaxo, P., and Andreae, M. O., Modeling investigation of light-absorbing aerosols in the Amazon Basin during the wet season: Atmos. Chem. Phys., 16, 14,775-14,794, doi:10.5194/acp-16-14775-2016, 2016.

Andreae, M. O., Anderson, B. E., Blake, D. R., Bradshaw, J. D., Collins, J. E., Gregory, G. L., Sachse, G. W., and Shipham, M. C., Influence of plumes from biomass burning on atmospheric chemistry over the equatorial Atlantic during CITE-3: J. Geophys. Res., 99, 12,793-12,808, 1994.