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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol observations and growth rates in the tropical tropopause layer" *by* D. A. Waddicor et al.

Anonymous Referee #1

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This paper describes a case study of Aitken and accumulation mode aerosol observed downwind of the anvils of deep tropical thunderstorms during the ACTIVE field campaign in January 2006. Using particle counter measurements and back trajectories the authors estimate the time the aerosol concentration reaches its peak maximum and examine furthermore the hypothesis if the strong increase in aerosol concentrations can be explained by production of sulphuric acid from SO2 followed by particle nucleation and coagulation alone.

Overall, I found the manuscript represents a quite solid piece of work combining measurements and model simulations for a selected case study with an interesting result. There are a couple of things that need to be clarified and/or more elaborated, so I recommend publication after the authors have addressed the comments given below.

First of all, I think the introduction part needs some reworking. The motivation is somehow hidden in the middle of the introduction (page 2358, lines 1-2). The authors give quite nicely an overview about the existing aerosol measurements, about a couple of hypothesis and explanations and a very extended one of the modeling work from Clement et al (2006) but then the reader is left alone with this knowledge. For me it remains unclear, why despite all this explanations and theories the mechanism is still unclear.

Furthermore, why is this specific case study important and necessary? Do you gain now additional information with your measurements which were not available before? The authors do not mention anything about the role of silent degassing or small eruptive volcanoes. Recent studies have shown the import influence of tropospheric volcanic eruption an increase of the stratospheric aerosol e.g. Solomon et al. (2011), so they might play a role for the TTL aerosol as well.

The selection of the date of the case study is also not completely clear to me. Why did you choose the 23 January? What makes this date special or typical? Are the results for this day more specific and or can they be considered in a more general framework.

The authors discuss in their paper the uncertainties with respect to the sulphuric acid formation rate, but they do not tackle one other critical point, if the applied model is suitable/applicable for their specific model study. For their model studies the authors uses the AEROFOR model (Pirjola, 1999) which has been designed and successfully applied for aerosol studies under tropospheric conditions. To save computer time in AEROFOR a parameterized value for the sulphuric acid mole fraction of the critical nucleus as a function of temperature, relative humidity and relative acidity (Kulmala et al, 1998) is used. As discussed in Vehkamaeki et al. (2002) this parameterization is valid only between 233 K and 298 K and for relative humidities between 10% and 100% and can not be applied globally e.g. the upper tropical troposphere with temperatures below 200 K. Hence, uncertainties in the nucleation rate can be more than one order of magnitude and therefore contribute to the existing differences between model and

observations.

The authors use for their different estimates often values which were gained under quite different conditions (forests) or heights (boundary layer, middle troposphere). For example for the amount of sulphuric acid in the freshly nucleated particl, the authors refer to Boy et al (1998). Boy and coworkers investigated new particle formation in a forest environment on the leeward side of the Rocky Mountains at an elevation of 2900 m. The question therefore naturally arises how representative are these values for the TTL and maritime conditions. I would like to see a more careful discussion here and throughout the text.

Minor comments Title: I think the title is too broad and more suitable for an overview paper than for a specific case study. It should be more specific e.g. Aerosol observations and growth rates downwind of the anvils of a deep tropical thunderstorm

Page 2357, lines 5-124, CARIBIC measurements were also taken at the INDIC route (Hermann et al, 2003) and over the North Atlantic (Hermann et al, 2012)

Page 2366, line 12, I do not see a change in the CO when the aircraft entered the cloud just a slow decrease with height

Page 2368 line 17, The number 115 should be listed as outcome of the identification of nucleation events in section 4.2

Page 2370 lines 5-8, I am confused, I thought the work of Fiedler et al (2011) consider African biomass burning plumes over the Atlantic. Please clarify !

Page 2371 line 15 delete one “could”

Figure 2 can be combined with Figure 6

Figure 8 Numbers for the color shading are missing

Figure 13 One can reduce the size range of the x axis to 10ˆ-7m

Page 2379, line 30 Typoo Möhler instead of Möhlerr

References: Boy, M., et al.: New particle formation in the Front Range of the Colorado Rocky Mountains, Atmos. Chem. Phys., 8, 1577–1590, doi:10.5194/acp-8-1577-2008, 2008. 25 2359, 2371

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