Aerosol observations and growth rates in the tropical tropopause layer

Reply to reviewer 1

"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?"

We will amend the introduction by adding a paragraph at the end of section 1.1: *Current models of nucleation and growth of aerosol in the TTL have not been properly tested against measurements and there is substantial disagreement in the literature. This disagreement stems from the nucleation and growth mechanism being not precisely known. There are a number of competing theories on how aerosol are produced and grow: binary nucleation, ternary nucleation, ion recombination (e.g. Wilhelm et al., 2004; Sorokin et al., 2006; Sorokin and Arnold, 2007) and organic compounds (e.g. Kulmala et al., 2006). Furthermore, the final two processes are also expected to interact with first two, causing further difficulty for modellers. There are also varying models within each scheme (English et al. 2011); all of this combines to give disagreement between in situ aerosol observations and model data. This is particularly apparent in the nucleation regime where different models can give results that vary by orders of magnitude (Vehkamaki et al. 2002). In this paper we examine aerosol measurements downwind of a convective anvil in order to derive the growth rate of aerosol particles into the size range greater than 10 nm. These will be compared with a simple model for aerosol nucleation and growth to see if the model is consistent with our results.*

"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."

For this paper, we have chosen to focus only on cases that demonstrate clear cases of convective outflow. Indeed, this was the motivation for the study: to determine the timescale for aerosol nucleation and growth after convective degassing. The outflow timescales in this work were short enough to discount significant amounts of external entrainment (page 2372, line 25). Therefore, volcanic influence was not relevant to this paper. Furthermore there were no significant eruptions near to the campaign area within a recent time.

"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."

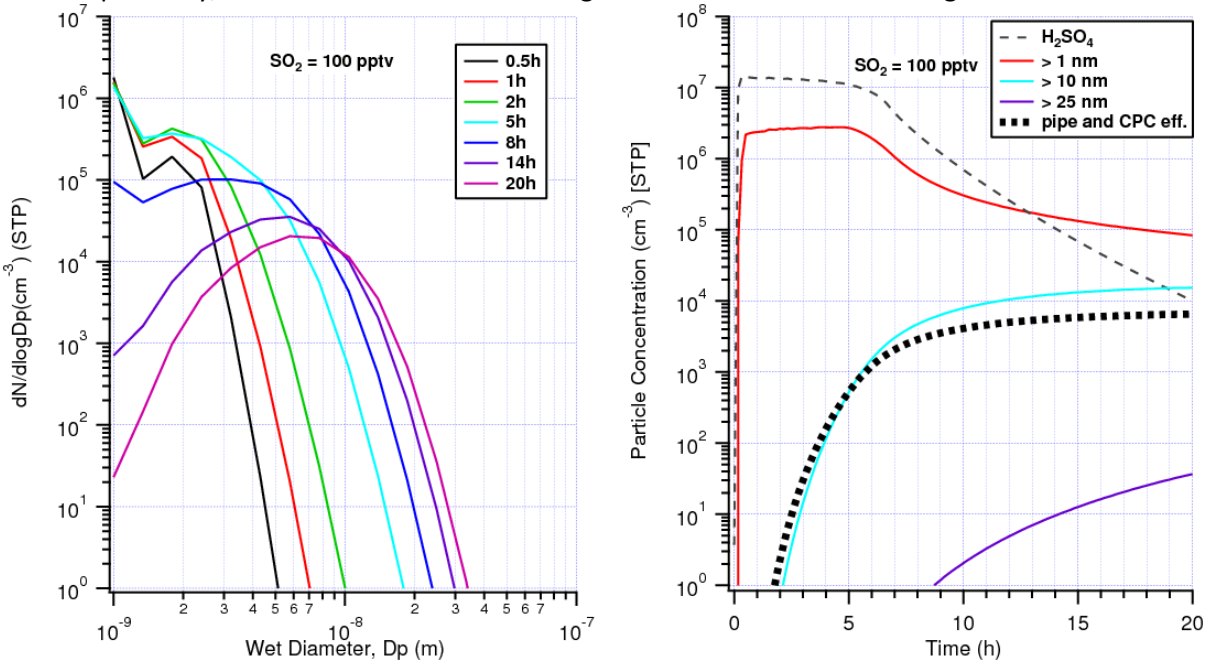
The ACTIVE flight of the 23 January 2006 was used due to a combination of favourable meteorological conditions - a large convective system - and an Egrett flight downwind of the anvil. Nearly all other Egrett flights were made either inside anvils or in their immediate vicinity; a few survey flights were also conducted but these sampled air that had not encountered convection for

more than a day. The results from 23 January showed distinct and contrasting regions of high cloud particle number concentrations/low aerosol particle number concentrations and vice versa. In other cases where measurements were made downwind of convection, the anvils were so numerous (viewed from infra-red satellite data) that disentanglement of air parcel origin, using wind projections, was unreliable. This argument can be added as an additional paragraph to the end of section 1.1 in order to clarify the choice of 23 January.

"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)…."

First of all (and this will be mentioned in the text: pages 2372-2373) the AEROFOR model was initiated at 220K and not sub-200K as was the concern in the paper by Vehkamaki et al. (2002). The AEROFOR model, which has been tested successfully (such as Clement et al. 2006), was chosen as a suitably time saving option. It is simple enough to implement and to test varying SO_2 and humidity concentrations. Clement et al. (2006) point out that the updated Vehkamaki et al. (2002) parameterisation differs little from the original Kulmala et al. (1998) parameterisation at upper tropospheric conditions. English et al. (2011)'s model results were insensitive to the nucleation scheme used; the coagulation scheme, however, had a greater influence on the 10 nm particles and larger – for UTLS conditions. We recognise that the nucleation parameterisation is highly uncertain, but as we show below, the results of the paper are not sensitive to the nucleation rate. We chose the Clement et al. (2006) model as a reasonably simple representation of coagulation and particle growth in a binary system, and show that this is not consistent with the observations. Clearly therefore a different set of assumptions is needed – either a different model or a different aerosol composition.

Due to the comment concerning the nucleation scheme used, our AEROFOR model was updated to use the parameterisation by Vehkamaki et al. (2002). Below are the plots of our re-run model using the latest nucleation scheme. For the size ranges that we consider in this paper (the > 10nm particles) the blue curve is almost identical to the original model; the curve appears after approximately 2 hours and the maximum concentration is just over 10^4 cm^-3, as in the original run. More importantly, the CPC modulation curve is again almost identical to the original run.



"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 particle, the authors refer to Boy et al (1998). Boy and co-workers investigated new particle formation in a forest environment on the leeward side of the Rocky Mountains at an elevation of 2900m. 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."

It is true, as the reviewer mentioned, that the paper by Boy et al. (2008) refers to data taken from boundary layer forests and that the expected critical cluster would have a higher critical radius, and therefore a higher number of sulphuric acid molecules, than that of a critical nucleation particle in the cold upper troposphere. This was NOT included in the AEROFOR model parameterisation and was merely used as a preliminary estimate for the approximate number of SO_2 molecules required to produce the maximum aerosol concentration we did. We chose 25% sulphuric acid ratio (by volume) in 10nm aerosol as an arbitrary number in the simple number calculation on page 2370 line 15. We will remove the reference to Boy et al. (2008) in the revised version to avoid confusion.

"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"

We accept the suggestion for the title.

"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)"

Accept suggestion. We will mention the broader scope of the CARIBIC campaign and include suggested references.

"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"

There is a small drop in CO as the aircraft enters cloud (with accompanying RHi increase) at 13.65 hours.

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

Accepted. The number of nucleation events (115) will be explicitly mentioned as an outcome from our nucleation identification methodology (include at the end of 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!"

It is correct that the Fiedler et al (2011) campaign place over the Atlantic Ocean. This will be clarified in the text (page 2358, line 16).

Page 2371 line 15 delete one "could"

Corrected

"Figure 2 can be combined with Figure 6"

We would prefer to keep these two figures apart for clarity.

"Figure 8 Numbers for the color shading are missing"

Figure has been amended

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

Figure has been amended

"Page 2379, line 30 Typoo Möhler instead of Möhlerr"

Amended

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