We want to thank the reviewer for carefully reading our manuscript and for providing numerous helpful comments. The reviewers' comments are repeated in full below in black font, with our replies indicated after each comment in blue font. Text which has been changed in the manuscript is shown in red font.

Major changes

As both reviewers noted we used an incomplete definition of condensation nuclei (CN) in the text and also for our simulation. CN were defined in our model as just the smallest particles that are smaller than those counted by the optical particle counter (diameter $d_p < 0.3$ µm) instead of all particles that can be counted by a condensation nuclei counter. Usually this does not make a big difference as the smaller particles dominate in number. However, for our study, the two formulations unfortunately make a substantial difference.
We assumed that the CN presented in Campbell and Deshler (2014) were all smaller than 300 nm diameter (as this was the cutoff of their OPC). Therefore we simulated these CN as small particles and *additionally* simulated the preexisting particles surface area loss by the larger particles. This extra loss term reduced our simulated CN which made nucleation of new CN necessary in all months at all altitudes and not just during the formation of the CN layer. This is why we could derive profiles for all months; which was the major point of criticism by Reviewer 2.
We now corrected our method. As a result, we do not simulate the long warm up phase anymore, but start at the beginning of July and use the CN of Campbell and Deshler (2014) as initial particles for the first month. These CN are now the only particles in our simulation, we have no additional loss to preexisting particles anymore.
This changes our results a lot. We now do not need sulfuric acid anymore for reproducing the observed CN, except for the formation of the CN layer. This solves the major point of Reviewer 2.

We can not derive profiles of sulfuric acid for all months anymore, and therefore some of the main messages of the paper change. We focus more on the result that we can reproduce the observed CN just with coagulation and air volume compression except the CN layer formation. This is consistent with Campbell and Deshler (2014) and the general idea of the formation of the CN layer: low sulfuric acid during winter but then an increase after the onset of sunlight. However, we still derive profiles at which nucleation would occur and interpret these as upper limits (as suggested by Reviewer 2).
As the method and results changed, large parts of the paper were rewritten. We did this with the suggestions of the reviewers in mind and we also used this opportunity and changed the paper to active-writing-style. Here are the major changes:

Title: We changed the title of the paper to" Nucleation modeling of the Antarctic stratospheric CN layer and derivation of sulfuric acid profiles". This flip of the two parts reflects that the derived profiles are not our main/only result any more and this is also the order in which we present and discuss the results.

Abstract: The abstract is almost completely rewritten and describes the new results of the paper.

Introduction: We added more information on Antarctic stratospheric CN to the introduction as suggested by the reviewers. We removed the discussion of the too high CN layer altitude in the global model of Campbell et al. (2014) as we do not discuss this in the paper anymore (see below).

Methods: We rewrote and shortened this section. We shortened the SAWNUC description (Sect. 2.1) but added a short description of the most relevant processes. We removed the preexisting particle discussion from Sect. 2.2 as this is not necessary anymore. As reviewer 2 suggested the detailed discussion of the single parcel simulation is not necessary.

Results: We rewrote the Results section according to the new modelling results. We start with a simple reference case (Figure 1). We give additional information on the simulated descending air parcel paths (as both reviewers asked for this) (Figure 2a). We now show the simulated CN for all air parcels once without nucleation and then with nucleation (Figure 2b+3b). And we show the different sulfuric acid profiles and describe step-by-step how we derived them (Figure 1b+3a+4). We moved the comparison to the mid-latitude and Arctic profiles in a separate section (now 3.4) and added a comparison to the derived profile of Campbell and Deshler (2014) as reviewer 2 suggested (also 3.4).
We shortened the descriptive text of the sensitivity studies a lot and now show the effect of a 5 K temperature increase in Fig 6d.
We removed the discussion and the figure about the sulfuric acid production rates as we can not derive them anymore for all months.
We removed the comparison to the global modeling of Campbell et al. (2014) as we do not have 4 profiles anymore and as we cannot test the possible causes for the altitude shift with SAWNUC.

Discussion: We rewrote the Discussion to reflect the new results. We now state clearer what our study adds to the discussion.

We thank the reviewer for the helpful comments which significantly improved the paper.

Individual comments

From ACPD questions

While the basic description of experiments and calculations are sufficiently laid out, it would be difficult to reproduce them unless more detailed descriptions/equations of the SAWNUC model are provided, possibly in a supplementary section. Although, I understand the complexity of this box model, and that providing all details is not necessarily possible.

We added a description of the most important processes to the manuscript. The major equations of the model for the individual size bins has been described in detail by Lovejoy et al. (2004).

For this study, the basic processes simulated by SAWNUC are condensation and evaporation of sulfuric acid and coagulation for every size bin. Preexisting particles are fully simulated as particles and not just as surface area to which particles can be lost. Condensation and evaporation of sulfuric acid are the dominating processes for the formation of new particles while coagulation and condensation of sulfuric acid, if present, determine growth and reduction of existing particles.

While the overall presentation is good, there are some areas that can be better structured and written. These include grammatical errors, a cumbersome writing style, confusing section titles and organization, and a general writing fatigue in the later sections of the paper. These issues need to be fixed to improve the overall quality of the paper. I make suggestions in the annotated PDF of the paper.

Thank you for the suggestions. We followed them closely. We also changed the structure to make it clearer now. We reformulated various parts of the later sections.

There are no mathematical formulae present, but it may be nice to include some of the fundamental equations used in SAWNUC (e.g., the major nucleation equation that is inverted) possibly in a supplemental section. Showing and explaining the driving terms in the SAWNUC nucleation equation would be helpful.

See above. We added the major processes in the description of SAWNUC and in the description of our method in Sect. 3.1.

Introduction

Page 1, Line 25: This is technically not true, because Campbell and Deshler (2014) note that for balloon-borne measurements, this two-cylinder growth chamber (to grow CN measurements) is mounted vertically on top of an optical particle counter sensitive to particles> 150 nm radius [Rosen, 1964]. This CN instrument has been used to measure CN above Laramie since the early 1980s and above McMurdo since 1986. Thus their CN counters measure particles that could be larger than what you state as a 300 nm upper size threshold for a "typical CN counter". Please revise. Rosen, J. M. (1964), Vertical distribution of dust to 30 kilometers, J. Geophys. Res., 69, 4673–4676, doi:10.1029/JZ069i021p04673.

Thank you again for clarifying that point. We revised it as part of the change of the first section.

Therefore, condensation nuclei (CN) are defined as all aerosol particles that are large enough to be measured by a CN-counter, which typically can measure particles with diameters larger than ~10 nm.

Page 1, Lines 25-26: It is important to note that Campbell and Deshler (2014) results also qualify Murphy et al. [1998, 2013], where it is suggested that in polar winter, sulfuric acid primarily condenses on nonvolatile meteoritic material. Campbell and Deshler results are in general agreement with this, except in the CN layers with significant, and rapid, formation of new particles without a nonvolatile core. Its possible that the upper boundaries of the CN layer may be impacted by meteoritic material. This may be even more important to introduce here, if some type of sensitivity run is completed by adding an enhancement in theoretical meteoritic pre-existing particles are added to your study, and impacts on sulfuric acid profiles and CN layers are discussed (see comments in Sections 2 and 3). Campbell, P., and T. Deshler (2014), Condensation nuclei measurements in the midlatitude (1982–2012) and Antarctic (1986–2010) stratosphere between 20 and 35 km, J. Geophys. Res. Atmos., 119, doi:10.1002/2013JD019710. Murphy, D. M., D. S. Thomson, and M. J. Mahoney (1998), In situ measurements of organ- ics, meteoritic material, mercury, and other elements in aerosols at 5 to 19 kilometers, Science, 282, 1664–1669, doi:10.1126/science.282.5394.1664. Murphy, D. M., K. D. Froyd, J. P. Schwarz, and J. C. Wilson (2013), Observations of the chemical composition of stratospheric aerosol particles, Q. J. R. Meteorol. Soc., doi:10.1002/qj.2213.

Thank you. We included this as part of the rephrasing of the first passage and later in the introduction. In our changed method all particles could contain a nonvolatile core, except the ones that nucleate in the CN layer. Therefore, if they were measured by the CN counter, the meteoritic preexisting particles are now included in our simulation.

Page 2, Lines 11-12: Please be consistent with your "sulfuric acid" or “H2SO4” convention through the paper. It is used rather interchangeably.

Done

Method

Page 3, Lines 9 – 11: This is rather different then in Campbell et al. (2014), where their CARMA model divides sulfate aerosols amongst 30 size bins in CARMA, with sulfate mass increasing by a factor of 2.4 between adjacent bins. The dry radii of these 30 aerosol size bins range from 0.343nm to 2.17 μm.

Yes, it is rather different. The design with the linear bins first is one of the key elements of SAWNUC to accurately simulate the nucleation and is deeply implemented in the code. This can not be changed. However, we did change the geometric scale factor for the larger bins to 2.4 and thereby increased the size of the largest bin. It had only a very small effect on the derived profiles.

Page 4, Line 9: Is it technically true to call the SAWNUC output CN concentrations, as you have previously defined CN from a measurement perspective? The output from SAWNUC contains concentrations of particles that are not observable in CN counters at the supersaturations employed in Campbell and Deshler, 2014 (~ < 3 - 10 nm). Maybe just state "Particle concentrations and sizes...."

We agree. Done.

Page 4, Lines 12-13: What is the chosen 5 nm detection limit based on here? Is this an arbitrary cutoff selection for the model, or do you have appropriate reference for this? Campbell and Deshler (2014) report that their CN measurements contain all particles r > 3 - 10 nm in size, dependent on pressure altitude.

We did choose it as a lower limit of the 50 % cutoff (as 6 nm diameter was reported as 75% cutoff). However, during the redesign of the paper we now changed it to the reported 20 nm diameter for all altitudes and use 6 nm for all altitudes in the sensitivity test.

Page 4 Lines 16 – 17: Provide justification for this H2O profile. Show references, such as "...based on July MLS and hygrometer measurements in Figure 7a in Campbell and Deshler (2014)", or if you used a combination of different measurements to get your assumed profile.

Done.

Page 5, Lines 9 – 11: What altitude(s) is (are) represented in the model "maximum trajectories" in Figure 1? This information is very important to describe and show. If these "maximum trajectory" altitudes are representative of the matched maximum CN concentrations in Campbell and Deshler (2014) Figure 1, then better explanation is needed in this section.

Thank you, we added a Figure that shows the trajectories and rewrote the text.

Page 5, Lines 17 -21: Why a decrease in10% of CN from the May 1 - June 15 period to the beginning of June/July period? Can this possibly be based on limited CN measurements in June/July from Campbell and Deshler (2014)? Is this an arbitrary value?

It was a somewhat arbitrary choice. Some decrease is expected because of coagulation and we chose it to be 10%. We redesigned the preexisting particles simulation to avoid this choice.

Page 6, Lines 7 – 8: What are these "assumed functions"? Are they derived from Campbell et al. (2014), for example in Figure 3 they show time series of CN concentration and maximum CN altitude with time for McMurdo, derived from many years of measurements at different julian days.

No, they were really assumed; laid around the monthly values and chosen so that the monthly mean matches to the measurements.

Page 6, Lines 32-33: Does this statement really justify that neglecting temperatures < 190 K introduces little uncertainty. Suggest rewording, if not additional discussion as to the uncertainty it really introduces to the nucleation rate. See Figure 5d.

(The referred phrase was moved to Sect. 2.2.) We agree. We now use the sensitivity test to show the influence in Fig 6d.

This introduces some uncertainty which is estimated in our sensitivity test of a 5 K temperature increase (Sect. 3.3).

However, this temperature sensitivity has to be considered when interpreting the July and August profiles at low altitudes as there the temperature was below SAWNUC's lower temperature range of 190 K (maximum 5 K below, see Fig. 1a).

Page 7, Lines 2-3: What part of the design does not allow for these three trajectories in Sept? Rather a mystery here for the reader.

Thank you. We discuss it now in more detail in the rewritten results section.

In September above the CN layer at ~28 km, too many CN are simulated even without sulfuric acid being present (Fig. 2b). This is the result of an air volume compression in the subsiding air parcels from 31 km in August to 28 km in September which increases the CN concentration by ~60 %. Here, coagulation is not efficient enough to reduce the monthly mean CN to the observed value.

Results

Page 7, Line 28: What is the total stratospheric sulfur value based upon?

We agree that a reference would be needed. However, according to the new modelling results this discussion was changed anyhow.

Page 7, Lines 29 – 30: Issue regarding your definition of a "typical CN Counter" and the statements made here. Typical CN counters in Campbell and Deshler (2014) and Campbell et al. (2014) measure all particles with r > 3 - 10 nm (dependent on pres- sure/altitude), and thus could technically count particles above 300 nm. Thus, I do not understand why the derivation mechanism fails here.

We agree. See discussion at the beginning.

Page 8, Lines 3 – 5: Also Murphy et al. (2013) suggested that in polar winter, sulfuric acid primarily condenses on nonvolatile meteoritic material. The results of Campbell and Deshler (2014) are in general agreement with this at higher altitudes, except in the CN layers with significant, and rapid, formation of new particles without a nonvolatile core. Murphy, D. M., K. D. Froyd, J. P. Schwarz, and J. C. Wilson (2013), Observations of the chemical composition of stratospheric aerosol particles, Q. J. R. Meteorol. Soc., doi:10.1002/qj.2213.

Thank you, we added this to the introduction and discussion sections.

Page 8, Lines 8 – 21: This is an intriguing comparison to measured mid-latitude profiles indeed. However, couldn't the SAWNUC inversion method also be applied to derive a mid-latitude sulfuric acid profile using the mid-latitude seasonal CN/temperature profiles shown in Figure 1a and 1b of Campbell and Deshler (2014)? I would like to see an additional derived mid-latitude sulfuric acid profile since it is more a direct comparison to the mid-latitude measurements, and also does not incur the same altitude/adiabatic expansion shifts when comparing polar to mid-latitude regions. The summary of Figure 1 in Mills et al. (2005) show sulfuric acid measurements mainly during September and October, so you could use the representative fall season CN/temp profiles in Campbell and Deshler Figure 1a and 1b to derive a "fall mid-latitude sulfuric acid profile". This can provide supplemental information on the performance of the SAWNUC inversion method used.

(This comparison was moved to Sect. 3.4) Thank you for the suggestions. Unfortunately, as the CN layer above Wyoming is expected to form in the polar region and then is transported to the mid-latitudes we can not model its nucleation properly. But we added a discussion about how the Antarctic profiles can be compared to mid-latitude profiles in Sect. 3.4.

Page 8, Lines 31 – 33: You should add that these results also further support Campbell and Deshler (2014) findings that the majority of the observable polar CN layer is due to neutral nucleation.

Added in the discussion

In agreement with Campbell and Deshler (2014) we find that the development of the CN layer can be explained by neutral sulfuric acid-water nucleation.

Page 9, Line 8: What (if any) sensitivity runs were performed due to holding H2O profile constant? Were different H2O profiles tested? It is likely a small uncertainty in the nucleation rate, but may be worth investigating and mentioning.

Yes, we also tested 5 ppm everywhere and it results in only very little change.

Additional sensitivity studies (not shown here) showed that the exact amount of ions or water molecules (e.g. 5 ppm everywhere) has only a small influence on the derived profiles because the ion concentrations are high enough so that they are not a limiting factor, and the few parts per million stratospheric water vapor uncertainty is too small.

Page 9, Lines 11 – 14: Since the profiles derived are ~> 20 km, then why is the cutoff > 5 nm chosen, which is more representative of lower atmosphere conditions. Shouldn't the profiles be derived using a cutoff closer to > 20 nm (as reported in Campbell and Deshler, 2014)?

Yes, we changed this in the new method and in the text.

The measured CN were compared with the simulated CN by summing over all simulated particles with diameter above 20 nm, as Campbell and Deshler (2014) reported a detection limit of their CN counters of 6-20 nm diameter.

Page 9, Lines 15 – 16: If the main growth mechanism to a higher observable cutoff size was due to coagulation, then does the sulfuric acid concentrations have to increase in the nucleation areas? In other words, what effect does coagulation rates have on this sensitivity? Also, the profiles are nearly the same above about 28 km between June/July and September, regardless of cutoff size.

Here the coagulation is not the main effect but condensation of sulfuric acid on the small clusters. Therefore more sulfuric acid is needed. This effect reduces with higher sulfuric acid amounts as at these high concentrations, the clusters grow quickly after nucleation.

The lower cutoff leads to lower sulfuric acid concentrations as the nucleated CN do not have to grow as large by sulfuric acid condensation to be counted (Fig. 6a). This effect decreases with increasing sulfuric acid as at higher concentrations, the clusters grow quickly once they are nucleated.

Page 9, Lines 21-22: Ultimately, it would be best to avoid using a constant CN measurement cutoff size, since it is clearly dependent on altitude. Can this be varied for the multiple trajectories used in SAWNUC to derive the sulfuric acid profiles, possibly arriving at a "best guess" sulfuric acid profiles over McMurdo?

Yes, the cutoff could be varied but more information is needed for that e.g. a cutoff vs. altitude dependence. We are not sure how we could arrive at a best guess as there are too many unknowns. Note, however, that in the revised version the cutoff uncertainty is not as high anymore as we assume the CN outside the layer to be large particles.

Page 10, Line 10: Does both coagulation and condensation rates both have little influence?

Yes, it is now clearer.

A reduction of all coagulation and condensation rates by 20% increases all profiles only a little […]

Page 10, Line 16: What is this inter-annual CN increase based on? Campbell and Deshler (2014) indicate a range in CN concentrations dependent on altitude in Figure 3, and for the CN layer maximum level in Figure 4 for McMurdo.

We agree that this was not a proper choice as the actual CN variation is more complex. We decided to remove the CN increase and just vary temperature in the sensitivity test.

Page 10, Line 28: This whole section seems rather out of place in the paper, and would be better placed as the last summary part for Section 3, and named "3.4 Summary and discussion of model uncertainties"

Yes, we agree. We now only use the sensitivity tests in the comparison and discussion sections to interpret the results.

Page 11, Line 1: I don't see a compensation between the monthly values and cutoff size, as they are in the same direction for both June/July to September and in October. Additional clarification is needed.

Yes, they are in the same directions. But the monthly values already were an overestimation and the cutoff showed that the values should be higher. So this matches. However, as we now use a different cutoff size, this text is not needed anymore.

Page 11, Lines 5 - 6: If accepting my previous comment about moving this whole section to the last part in Section 3, this statement could then be made more robust and stated such as the following: "The June/July profiles have additional uncertainty due to rather unknown sulfuric acid production rates from processes that do not require sunlight over Antarctica in the winter. This uncertainty is compounded by sparse CN measurements made during June/July (and prior)

over Antarctica."

Good suggestion, thank you. The production rates were removed from the paper.

Page 11, Lines 7-8: This should not be a separate paragraph if that was what was intended. Also, I do not know if this can be quantitatively determined from the results presented. Please make sure to state that this is a qualitative approximation. Finally, from the discussion of the sulfuric acid production rates for June/July, it seems that this period may have even greater uncertainty compared to the October period, especially considering the much less CN measurements during June/July compared to October (see Figure 3a in Campbell and Deshler, 2014)

We agree that this was not quantitative. We removed the statement.

Page 11, Lines 17 – 18: Shouldn't this be "therefore this production term cannot represent any sulfuric acid that evaporates from preexisting particles"

No, the idea was that everything that is not explicitly simulated could be included in this production term. (This section was removed.)

Page 12, Lines 2 – 3: Please clarify what you mean by "one additional step away from the measurements"

The idea was that going from CN measurements to sulfuric acid concentration is one step which involves the processes simulated by SAWNUC. Determining the production rates represents a second step as this could also include processes that are not simulated by SAWNUC, which makes these results more uncertain. (This section was removed.)

Page 12, Lines 3 – 4: This is a confusing argument. You note that the derived sulfuric acid profiles in Figure 6 could be an overestimate (by a max of one order of magnitude) in this model, but then wouldn't the inclusion of these processes further increase the sulfuric acid production rates, and thus lead to a possibly even greater overestimate (although understandably unknown). In essence, it may indeed be difficult to compare to the limited measurements of Krieger and Arnold over the Arctic, and may in fact be underestimated in this model for the Antarctic June/July period. Please revise this statement.

We agree that this sentence was poorly phrased but the whole section was removed anyhow.

4 Comparison with global modeling

Page 12, Lines 8 -12: Why not generate a plot to show these temperature, sulfuric acid, and CN comparisons. It would be much clearer for the reader to associate the altitude discrepancies between this work and Campbell et al. (2014) during early July and late October, and beneficial

to the discussions that follow in this section.

Page 12, Lines 16 – 17: While this is indeed possible, is there any evidence of OH production of sulfuric acid over the Antarctic region? I would be very careful in drawing such suggestions that cannot be backed by measurements. Also, you make suggestions regarding July underestimate by global model compared to SAWNUC, but what about the October overestimate?

We agree, but as we decided to drop the comparison it is no longer relevant.

Page 12, Lines 18 – 23: This argument seems to suffer from some writing fatigue and is rather weak. No major assessments are made as to why this altitude shift of nucleation controlling variables occurs in the global model. Please improve this paragraph and make better supporting statements/arguments. Are you (and the satellite obs. by Hopfner et al., 2013) suggesting too high SO2 at hight altitudes in the global model compared to observations and SAWNUC? And what can be inferred as to the cause of this using your SAWNUC model (e.g., enhanced SO2 subsidence from mesosphere)?

Thank you. While we think that a comparison of the global model SO2 with the satellite could improve our understanding, we removed it as there is no direct relation to the SAWNUC model.

The idea was that in the satellite data the high SO2 values penetrate deeper in the Antarctic stratosphere than they do in the global model. Therefore there should be more SO2 at lower altitudes according to the satellite, which then could lead to the formation of the CN layer at lower altitudes. Therefore it looked like this downward transportation of SO2 and maybe the downward transport of everything else was not strong enough in the global model. However, this is speculation and we would like to be cautious and therefore removed the comparison.

Page 12, Lines 24 – 28: Section 3 and Figures 1 and 3 in Campbell et al. (2014) already support the ability of the global model to reproduce the observed temperature, water vapor, and CN profiles Antarctica and mid-latitudes, with the exception of an altitude shift in CN over Antarctica, and possible underprediction in magnitude of sulfuric acid. Again, can you provide potential reasoning into why this altitude shift occurs for all nucleation controlling variables against SAWNUC? Have you checked to make sure that there are no discrepancies between altitude definitions in the global model and how they are defined in SAWNUC to ensure an apples-to-apples comparison? Remember that the global model is an Eulerian framework, while the trajectories predicted in SAWNUC are Lagrangian.

Yes we made sure that we compare the right altitudes. In Campbell et al. (2014) Figure 1b it looked to us as if the temperature was underpredicted by the global model at higher altitudes. Which could result in the nucleation there. Then both controlling variables would have the altitude shift. But yes, we cannot support or model this properly with SAWNUC and therefore we removed the comparison with the global model from the paper.

Summary

Page 13, Lines 6 – 7: This is misleading as stated, because it implies that the global model in Campbell et al. (2014) was run with their derived sulfuric acid profiles, while in fact, it was run with the WACCM sulfuric acid profiles, and the derived profiles were used for further insight and to assess possible uncertainties in the WACCM profiles. Please revise.

We agree that this was misleading. We removed the reference to Campbell et al. (2014) from the summary as we do not discuss the global modeling any more.

Page 13, Lines 16 – 18: This argument could be improved from my previous comment regarding derived SAWNUC sulfuric acid profiles over mid-latitudes compared to measurements over mid-latitudes.

Yes, we added a short discussion about the mid-latitudes in the comparison section.

Page 14, Lines 2- 3: While this section adequately summarizes the findings presented here, more discussion on what SAWNUC can answer as to why the global model predicts sulfuric acid at too high an altitude, will make the discussion and results more robust in this paper. What are some of the potential processes leading to this altitude shift?

Unfortunately, SAWNUC can not answer more than Campbell et al. (2014) already did. We can just confirm the expectation of Campbell and Deshler (2014) that their profiles are an underestimation. But they already discussed the consequences of this for the global modeling in detail in Campbell et al. (2014). This is why we removed the comparison to the global model. If we should speculate for reasons for the altitude shift, we would suggest either the downward transport as mentioned above. But maybe a too fast oxidation of SO2 could lead to the same results? This question can not be answered with our box model, but only with a global model maybe by analyzing tracers… We changed the Conclusions section so that we now not only summarize but also say what knowledge we add to existing one.

We thank the reviewer again for the helpful comments which significantly improved the paper.