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

Major points

The primary issue is the authors' seeming assumption that underlying the presence of any stratospheric sulfuric acid particle concentration is a necessary sulfuric acid concentration. Yes there was a sulfuric acid profile at one time, but once the sulfuric acid condenses its concentration decreases to vanishing amounts. Because there are CN particles existent during polar winter does not mean that they were nucleated then and there. Stratospheric CN are ubiquitous in concentrations near 10 cm-3. Thus as the polar vortex forms the CN particles present at the beginning will persist in the vortex, their sedimentation quite negligible. The observations of Campbell and Deshler [2014] bear this out for much of the CN profiles they present. Here, however, the authors derive a sulfuric acid mixing ratio profile in times of the year and altitude regions where no new particle formation is observed. They then speculate on what could be causing the oxidation of so2 to obtain the sulfuric acid. Isn't this speculation on possibly an imaginary profile?
I understand when sulfuric acid particles are present there will be a residual sulfuric acid concentration; however, given that the vapor pressure of 75% by weight sulfuric acid is exceedingly low, this concentration will be quite small. Is this what is calculated by SAWNUC in the absence of new particle formation? The authors need to make clear at the outset how the observations of new particle formation are used in deriving the sulfuric acid profiles and how such profiles are obtained in the absence of new particle formation. In the latter case can there be anything more than an upper limit?

Thank you for pointing this out and we agree with all these points. As we describe above we had an additional loss term in our simulation. This loss removed particles that the model replaced with new nucleating particles. This point is corrected in the revised version and now the results match the expectations that no sulfuric acid profile is required during winter time.

The other major issue is the writing is not up to the standards expected for a scientific paper. The writing is imprecise, many details are missing, while irrelevant comments creep in. The authors should consider having some English help prior to the next submission and not keep the reader working out what is intended to be said.

We put significant effort into revising the manuscript, clarifying the structure, making the descriptions more precise, removing irrelevant and speculative comments and improving the English style.

Abstract

1.13. Reproduces what observations? Be specific here. Reproduces the new particles that are observed or . . .?

The formulations should be clearer in the new version.

1.14 As will be mentioned further, below, it is not clear how a sulfuric acid profile is obtained in July. At most this should be an upper limit.

We agree and changed the text accordingly.

1.16 “the observed magnitude of CN. . .” What does this mean? Was nucleation of new particles observed at higher temperatures?

This was about the discussion of temperatures (see below). We removed the statement from the abstract.

1.18-19. The authors have not convinced me of the need to include a sulfuric acid production rate, which is then one further step removed from the observations. Thus I see no need to go further and try and explain a winter-time production mechanism, when it has not been established that one is required.

We agree with this comment. With the correction to our calculations as stated above, a winter-time production mechanism is not needed anymore.

1.20. Why is there no comparison with the sulfuric acid profile derived by Campbell and Deshler [2014]?

Added. Thank you!

Our concentrations are about one order of magnitude higher than previously presented concentrations as our simulations consider that nucleated clusters have to grow to CN size and can coagulate with preexisting particles.

Introduction

1.23-26. This is a terrible first sentence. In fact it is not even a sentence. The clause set off with commas is a complete sentence and should stand on its own. But it is still a poor first sentence. In fact the definition of condensation nuclei (CN) is not correct. Condensation nuclei are observed by forcing small particles to grow to optically detectable sizes using a condensing fluid and a supersaturation chamber. There are a number of references available on the technique and perhaps one should be included. The size ranges vary somewhat, but typically the CN concentration is used to define the total aerosol concentration larger than some nominal size, say 10 nm. CN measurements also include particles > 300 nm, but their concentration is inconsequential compared to the total aerosol population.

We rewrote considerable parts of the introduction including the first sentence. We tried to improve the readability and to include a correct definition of CN as well as giving a proper reference.

Atmospheric aerosol particles are of interest due to their various influences on radiation, clouds, chemistry, and air quality. Condensation nuclei counters measure the number concentration of aerosol particles by growing them to optically detectable sizes by condensation (e.g. McMurry, 2000). 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.

1.27. Not all CN are formed by nucleation. It appears a significant fraction of CN are non-volatile suggesting that this fraction of CN particles appears due to the condensation of sulfuric acid on pre-existing solid cores [Borrmann et al., 2010; Campbell and Deshler, 2014].

Yes, we included this now:

In the Antarctic polar vortex a background CN concentration of ~10 cm^{-3} is found (Campbell and Deshler, 2014). Volatility measurements indicate that more than half of them have a nonvolatile core which could be meteoric material (Campbell and Deshler, 2014; Curtius et al., 2005). Sulfuric acid is expected to condense on these CN (Borrmann et al., 2010; Murphy et al., 1998, 2013) […]

2.12. Where does the h2so4 in the mesosphere come from to be available for photolysis?

Potential sources are evaporation of particles and maybe oxidation some remaining OCS and SO2 in the higher stratosphere/lower mesosphere.

2.15 “performed continuously for 24 years” What does this mean every month, every day? I understood the observations are only available for a few months each year.

Added

More recently, Campbell and Deshler (2014) presented an overview of all balloon-borne CN measurements between 15 and 35 km above McMurdo Station, Antarctica (78°S), that were performed between 1986 and 2010, 2-3 times a year during winter.

2.17-20. Very awkward sentence. Try. They present monthly averaged CN concentra- tion and temperature profiles which capture the unperturbed CN, with concentrations around 10-20 cm-3 in June/July as well as the development of a layer of particles at 20-25 km, with concentrations increasing to 100 cm-3 from August until October during sunrise and warming.

Done. Thank you.

They present monthly averaged CN concentration and temperature profiles which capture the unperturbed CN, with concentrations around 10-20 cm^{-3} in June/July as well as the development of the CN layer at 21-27 km, with concentrations increasing to 100 cm^{-3} from August until October during sunrise and warming.

2.26-28. Campbell et al. [2014] did not invert the nucleation equation, they ran a three dimensional chemistry model which produces its own h2so4.

Yes, they run the 3D chemistry model. But, they also compared this to a derived sulfuric acid profile by inverting the nucleation equation. They describe this in more detail in Campbell and Deshler (2014).

Campbell and Deshler (2014) describe a method where they derive an Antarctic sulfuric acid profile from the measured CN by inverting the neutral binary nucleation equation. They used the difference between the CN before sunrise and two weeks after sunrise averaged over all years to derive a nucleation rate for all altitudes from which they derived the corresponding sulfuric acid.

2.30. Who are they?

Campbell and Deshler (2014) (This sentence was removed.)

Method

3.30. Delete the sentence, “This section describes the derivation of the Antarctic stratospheric sulfuric acid profiles” and add the information to section title, e.g. Deriving the Antarctic stratospheric sulfuric acid profiles.

Done.

4.14. “3e5 ion pairs per gram of air and second” Awkward. Use standard SI units and symbols to describe a rate.

It was communicated to us in this unit and they also use this units in their papers, so we would like to keep it like this. But we added an example conversion to ion pairs cm-3 s-1.

The ionization rate of the Antarctic stratosphere in August-September 2010 was 3e5 ion pairs per gram of air and second (Ilya Usoskin, personal communication, 2013; according to Usoskin et al., 2011) which converts to e.g. ~10 ion pairs cm^{-3} s^{-1} at 200 K and 20 hPa.

4.16-18. There should be some references for the water vapor profile and the diabatic descent rate?

Done.

The water vapor profile for July was chosen to be a linear increase from 3.0 to 6.0 ppm from 18 km to 25 km and above a constant value of 6.0 ppm up to 32 km based on MLS and hygrometer measurements in Fig. 7a in Campbell and Deshler (2014).

4.19-24. 100 nm is at the upper limit of possible pre-existing particles. What concentrations are used for these particles? Since the surface area is chosen based on reasonable sources why is the particle size important? What does it mean that the surface area was converted . . . according to the ideal gas equation? Do you mean the surface area density was adjusted to different temperatures and pressures assuming a constant mixing ratio?

The concentration was calculated from the surface area and the size. Size and concentration were necessary to calculate the coagulation rates on these preexisting particles. Yes, it was adjusted to different temperatures and pressured assuming a constant mixing ratio.

However, as we now fully simulate the preexisting particles in the model, this preexisting surface area loss is not needed any more.

4.28-31. Is there really a trajectory of an air parcel here, or do the authors just mean variations in the various parameters required by the SAWNUC model? The language is confusing. If the latter then I think the more appropriate words would be box model simulation rather than air parcel trajectory. If the former then where does the air parcel start, what altitude, location, dimensions? What does it mean to “connect” to the monthly maximum of the measured CN? Do the authors mean the trajectory which reproduces the monthly maximum CN concentration? The next sentence is equally awkward, “It is assumed that this maximum of the particle concentration resides in a single air parcel that descends inside the polar vortex.” The maximum of the particle concentration doesn’t reside in the air parcel until the nucleation has occurred. What the authors are trying to suggest, as far as I can understand, is the optimum trajectory to reproduce the observed maximum in CN concentration. The phrase “maximum trajectory” does not convey this idea and I would recommend changing this language to something more representative of what is being simulated. Something like optimum trajectory to reproduce the observed maximum in CN concentration. This maximum in CN concentration is height dependent. Is this height dependence consistent with the diabatic descent in the model?

Thank you for pointing out these issues and the imprecise wording in our explanation. We rewrote the whole description and tried to include these points. We also added a Figure which shows the assumed descent pathways of the simulated air parcels.

We assume that the CN maximum of each month resides in a single subsiding air parcel, and place the other air parcel trajectories around this CN maximum pathway (Fig. 2a).

5.1. I believe the authors mean the sulfuric acid concentrations were varied on a monthly basis? Search implies that these concentrations exist on some data base or?

Yes, we don't use the word search anymore.

5.4-6. Awkward and unnecessary detail. The authors are merely iterating the model until they achieve a match with the measured CN by varying the sulfuric acid concentrations. If so say this. It is not the sulfuric acid concentration that corresponds to the monthly CN concentrations, it is the simulated CN concentrations based on the sulfuric acid concentrations. The writing here and elsewhere is awkward and requires the reader to do a lot of guessing as to what is meant.

Thank you. We rewrote the methods and the results section and rearranged the structure to make this much clearer in the new description.

5.14-18, Fig. 1b. The model is initialized with a CN concentration of zero. Is this realistic? There will be some residual particles, CN, persisting in the air as the polar vortex forms. It seems more plausible to assume these residual particles appear as the background of about 10 cm-3 shown in Campbell and Deshler [2014] outside of the CN layers than to assume a concentration of zero. If the model is allowed to initialize to 110% of the June/July CN maximum, then there would not be any further new particle formation in the June/July period. So no additional sulfuric acid is required. This point needs to be clarified.

Thank you for pointing that out. We changed our method of the initial particles so that this should not be a problem anymore. Now we just use the June/July measurements as initial preexisting particles for the following months.

5.18-19. Since the model is already at 110% of the CN maximum, why is further searching required? Is sulfuric acid required to maintain the pre-existing particles? Are these particles losing sulfuric acid? I understood these particles change size due to fluctuations in water based on temperature but not on the loss of sulfuric acid.

This was again because of the wrong additional losses. So some CN had to be produced again.

It is not clear how the subsequently derived sulfuric acid profile for June/July is obtained. How does it differ from the sulfuric acid profile that is required to build up the CN from zero to 110% of June/July values? How does the work here then take advantage of the measurements which show a maximum of CN near 30 km above a possible background of around 10 cm-3? If there is no new particle formation in June July then the speculation on the source of a sulfuric acid profile is misplaced.

Yes, we changed our method and now have no July sulfuric acid profile any more.

5.26. Please show the mean CN values on Fig. 1 a. It would also be instructive to include on both panels the observations which are being reproduced.

We agree. But as we removed this Figure as suggested by the reviewer this is not necessary anymore.

5.27-28. If there is a decrease from June/July – August as could be argued from Campbell and Deshler [2014] since the CN maximum concentration does not change in their Fig. 1, then it appears again that no new particles are formed so only an upper limit can be placed on the sulfuric acid concentration.

We agree. We clarify in the revised version that we can only give an upper limit for the sulfuric acid concentration.

5.32. How is this month long time interval different than the previous ones? Why is the sulfuric acid concentration now strongly increased? In Campbell and Deshler [2014] there is almost no change in CN concentration between September and October in the CN layer, arguing again for little information on a sulfuric acid profile.

It is not different. It had to be strongly increased because some nucleation was needed (due to the additional CN loss) and temperature is much higher in October.

5.10-6.4 and Fig. 1a). Is all of this necessary to understand what is done, and do the authors really want to explain the subtleties of Fig. 1a? In the end more realistic time steps are used and profiles created. I don't really think the reader is benefitting any from the previous discussion and it raises many questions such as listed above.

We agree. Especially as now as no sulfuric acid is necessary in July and August (and October) this is not needed anymore. We shortened this section but extended the description of the modelled air parcel paths to clarify our approach and the results.

6.17-24. This paragraph is confusing by suggesting that the sulfuric acid is dependent on other processes in the model rather than just the amount needed to create the CN maximums. Here the authors claim that the sulfuric acid is dependent on sunlight, which we know it is, but I didn't think this was considered by the SAWNUC model.

We wanted to show that the profiles agree with the big picture. But we see that this could be rather confusing as we did not produce these results. We agree that it needs to be clarified which parts are simulated by SAWNUC and which are the assumed process that could lead to the simulated values. We considered this during the rewriting of the text and concentrate on the results that can be simulated by SAWNUC.

6.21. What is meant by, "the chemical lifetime of SO2 by OH"?

We meant how fast the SO2 is oxidized by OH. The lifetime is the SO2 concentration divided by the loss rate. However, this paragraph was removed.

6.23. Since the authors are now dealing with monthly averages, how can they make this claim, "Starting in September, the sulfuric acid amount increases first slowly and then strongly from September to October"? Monthly averages cannot supply this information.

When there is only a small change from August to September but a big change from September to October, then a bigger change is expected later in September. But we agree that this argumentation could be problematic and removed the statement.

Fig. 1b. What is the explanation for the fast rise in CN near the beginning of September, while the sulfuric acid only increases at the end of September? What is causing the CN formation if not the sulfuric acid? There is a disconnect here.

Well observed. The sulfuric acid was longer on higher levels in the beginning of September compared to the months before. So there was a sulfuric acid increase even though it was only a small one. This allowed new small particles to form and to grow to observable size.

However, now we don't show this anymore.

6.26. "To complement the profiles" What profiles? So far the authors have been calculating the conditions required to reproduce the maximum in observed CN concentrations. This would then only be a sulfuric acid concentration at one altitude in one month. Perhaps the authors intend to say they will now create profiles. If so the title of this section is misleading.

We agree, that to this point we only had one value in each month. Hope this is clearer in the new method description where we just describe the method once for all simulations.

7.3 "in September" is redundant and should be deleted.

Done.

3 Results

Section 2.2.3 and Fig. 2. This section and figure raise again my main question about this work. How is a sulfuric acid profile derived if there is no new particle formation? The CN layers in the CN profiles shown by Campbell and Deshler [2014] are limited in altitude to a fraction of the 18-40 km to be investigated here. Thus, for example in September above 27 km there is no evidence of new particle formation. So can anything more than an upper limit be placed on the sulfuric acid profile? Yyet Fig. 2 shows the September sulfuric acid profile to continue to increase above 27 km. How are profiles created to 40 km when there are no observations above 35 km?

The profiles only go up until 32 km. The production outside the CN layer was needed because of the wrong additional CN loss. This is corrected now.

7.10-15. Here again the authors are discussing results suggesting that the sulfuric acid profile is derived from sources outside the matching of the SAWNUC model with observations of CN.

We wanted to show that the profiles are generally in agreement with the expected bigger picture. But we agree that this could be rather confusing as we did not produce these results. In the revised version we only discuss the direct results from SAWNUC.

7.17 "the shapes of both profiles are very similar" The authors discuss the temperature profile as if it were shown in Fig. 2, but it is not. Thus the readers cannot follow this point. The temperature profile should be included in Fig. 2.

We agree and now show the temperatures profiles in the revised version (Figure 1).

7.19-21. I don't understand. Do the authors mean there is a narrow sulfuric acid concentration range leading to the nucleation of particles in the 10-100 cm-3 range? There is no reason particles cannot exist at these concentration levels if they are already there. The nucleation rate is not in a small window. The nucleation rate is determined by the concentration and the temperature. The small window is determined by both temperature and sulfuric acid concentration. Far too many or too few CN for what? I thought the particle concentrations were used to derive the sulfuric acid profile given the temperature, so if there are more or less CN this just affects the sulfuric acid concentration.

Here we discussed how much the sulfuric acid would change if we want to nucleate only some more CN. We reformulated the description of our approach significantly to make this point clearer.

7.22. "20 or 50 particles present" Are these particles already existing or nucleated particles? I assume the authors mean an observation of 20 – 50 new particles has a small influence on the derived sulfuric acid.

Yes. An observation of 20 or 50 new particles.

7.25-26. This is confusing. If the CN layer were not observed then there would be no attempt to derive the sulfuric acid profile. Above and below the CN layer there is not enough information to derive the sulfuric acid profile, only upper bounds, determined by the temperature, can be placed on it.

We agree.

7.30. This statement is not correct CN instruments measure all particles above some nominal size, typically about 10 nm. In addition I believe Campbell and Deshler report observations explicitly of particles > 150 nn using a second instrument, and no enhancements of these particles were observed in the CN layers.

Yes. This was a misunderstanding on our side.

7.27-29. These statements should give the authors pause to consider if there is an error somewhere rather than trying to explain these high sulfur amounts.

As stated above, there was an error in our assumptions. Now this is corrected and there is no need to explain the high sulfur amounts anymore.

7.30-32. This argument is incorrect. The water vapor concentration is low throughout the stratosphere. The particles are not composed of water but a sulfuric acid water mixture and the temperature determines how much water is retained by the highly acidic particle. The water vapor concentration has nothing to do with it, only temperature. There are plenty of water molecules.

We agree with this statement in general. The argumentation of 7.30-7.32 was removed in the revised version.

7.32. "To compensate there high . . ." For the reasons above I don't' understand this argument. Isn't the nucleation of particles dependent on the gathering of critical clusters of the acid not the water?

See above

8.5-7. Why is this assumption made? Campbell and Deshler, Fig. 5 show directly that the non-volatile fraction above 25 km is similar to that below 20 km, about 60%. It is not at all clear how the authors derive a sulfuric acid profile when there is no obvious nucleation. Again there can only be an upper bound.

Again, agreed and corrected.

Fig. 3. Why isn't the profile derived by Campbell and Deshler [2014] included in this figure?

The profile of C&D 2014 is included now.

8.33 "However, in the areas of higher temperatures the CN are almost exclusively produced by ion-induced nucleation." This assumes that CN are nucleated, but there is really no evidence for this, thus it is questionable whether any CN are nucleated in these areas. Sentence should be rewritten to reflect that, if particles nucleated they would require ion-induced nucleation, but in fact there is no evidence that such particles are nucleated.

Yes, we agree.

10.29-30. What is over/under estimated? Why is it clearly related to the months? I believe the authors mean the size cutoff measurement uncertainty.

This referred to the discussion of the 5 day periods. It was removed in the revised version.

11.7-8. This seems somewhat obvious as all the nucleation occurs in August, September. When there is no new particle formation this approach can only provide an upper bound on the sulfuric acid.

We agree.

11.15-16. Sulfuric acid evaporating from freshly forming particles? How does this occur? My understanding is that the vapor pressure of sulfuric acid is very low, thus it condenses readily and then stays condensed. Same question about pre-existing particles. At the temperatures used in the modeling I would not believe that the pre-existing particles would do much more than adjust their water content to the temperature changes.

Yes, in most altitude regions and for most times they do not evaporate. Only at the highest altitudes and highest temperatures in October there is some evaporation of sulfuric acid predicted.

11.14-18. If I understand correctly the production rate is simulated by continually adding the necessary h2so4 molecules to simulate the observations without specifying where the molecules come from e.g. so2, or particles. Thus all this discussion about the possible sources is speculation without basis and should be eliminated.

Yes, this was speculation. It is removed from the revised manuscript as this source is not needed anymore.

11.22-25. I am again confused as to why there is production of h2so4 in June July when the CN particle concentration is not changing from the model initialization. Much of the rest of this paragraph is again speculation and should be strictly limited.

Removed.

12.1-4. This is all redundant. Overall the paper would not suffer if section 3.3 and figure 6 were removed.

Yes we agree. Removed.

Summary

12.13-15. It is not clear why SAWNUC is predicting a sulfuric acid mixing ratio at this time in this region.

Removed. Not an issue anymore, due to the revised loss calculation.

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