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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Derivation of Antarctic stratospheric sulfuric acid profiles and nucleation modeling of the polar stratospheric CN layer" *by* Steffen Münch and Joachim Curtius

Anonymous Referee #1

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Review For ACP-2016-583: Derivation of Antarctic stratospheric sulfuric acid profiles and nucleation modeling of the polar stratospheric CN layer

Summary and Recommendations:

The paper ACP-2016-583 derives sulfuric acid profiles over McMurdo Station, Antarctica, and investigates nucleation in regards to the observed stratospheric CN layer that has a global extent. It is closely tied to the CN layer work presented in Campbell and Deshler (2014) and Campbell et al. (2014), where they presented a method of inverting the binary homogeneous nucleation rate equation to get "derived" sulfuric acid profiles that are based on measured CN profiles.

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Discussion paper

The authors of this paper were intrigued by the inversion method of Campbell et al. (2014), and thus extended the nucleation inversion idea by using the more robust SAWNUC model to include more processes and refine the sulfuric acid profiles over Antarctica (for which no measurements exist). Overall, the results in Section 4 compare favorably with Campbell et al. (2014), despite some differences in magnitude and altitude. The paper provides useful information to the scientific community regarding sulfuric acid profiles and a concentration range representative of the winter-spring polar vortex over Antarctica.

Overall, the paper was generally well written and laid out, the methods are sound, and the results are scientifically significant. The paper does, however, include some technical flaws, relatively weak arguments and statements, and suffers from considerable presentation and writing fatigue, especially towards the later sections of the paper. Sentence structure and organization can also be improved. Finally, some of the analysis and presentation of the results needs work. While my formal recommendation is to accept this paper with minor revisions, it is noted that a fair number of changes (with a few technical corrections) should be made before publication. Please see specific comments included here and in the annotated PDF that contains additional writing details for help in these areas.

General Comments:

1. Does the paper address relevant scientific questions within the scope of ACP?

Yes, the paper addresses the scientific question and challenge of deriving sulfuric acid concentration profiles over Antarctica, where no measurements exist. This topic does fit into the scope of material appropriate for ACP.

2. Does the paper present novel concepts, ideas, tools, or data?

The paper does not present an entirely novel concept, as it was first suggested and discussed in Campbell et al. (2014). The paper does, however, extend and refine the

work of Campbell et al. (2014) using a more robust nucleation model, while providing adequate citations and discussions of the Campbell et al. (2014) work.

3. Are substantial conclusions reached?

Yes, the conclusions are substantial in that they provide refined sulfuric acid profiles from a box model over a region where no measurements exist. I envision these profiles can be used in further modeling studies of the Antarctic stratosphere, while also helping to place any potential future model results or observations into context. Furthermore, the results are also used to confirm the validity of the model used to predict the global extent of the CN layer in Campbell et al. (2014).

4. Are the scientific methods and assumptions valid and clearly outlined?

The scientific methods appear valid, however, the choice of some assumptions and approximations used in the modeling need to be further clarified or presented more clearly in regards to the available measurements.

5. Are the results sufficient to support the interpretations and conclusions?

Yes, I agree that the results generally support the interpretations and conclusions stated in this paper.

6. Is the description of experiments and calculations sufficiently complete and precise to allow their reproduction by fellow scientists (traceability of results)?

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.

7. Do the authors give proper credit to related work and clearly indicate their own new/original contribution?

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Discussion paper

Yes, there is sufficient credit and citations given, and the authors clearly indicate their new/original contribution.

8. Does the title clearly reflect the contents of the paper?

Yes.

9. Does the abstract provide a concise and complete summary?

Yes.

10. Is the overall presentation well structured and clear?

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.

11. Is the language fluent and precise?

Yes, the language is fluent, but could be more precise in many areas. I make suggestions in the annotated PDF of the paper.

12. Are mathematical formulae, symbols, abbreviations, and units correctly defined and used?

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.

13. Should any parts of the paper (text, formulae, figures, tables) be clarified, reduced, combined, or eliminated?

There are some areas of clarification needed regarding text and figure presentation

(see specific comments), but I see nothing that need to be completely eliminated. In fact, the addition of some more figures and explanation may further improve the paper.

14. Are the number and quality of references appropriate?

There are some places in the paper where additional references citing relevant work are needed to give credit, and to give further basis for approximations/assumptions made.

15. Is the amount and quality of supplementary material appropriate?

Not necessarily. See comment 12 above.

Specific Comments (please see annotated PDF for additional writing suggestions and corrections):

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.

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 organics, 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.

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

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.

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 ($\sim$ < 3 - 10 nm). Maybe just state "Particle concentrations and sizes...."

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.

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.

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.

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?

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.

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.

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.

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

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 pressure/altitude), and thus could technically count particles above 300 nm. Thus, I do not understand why the derivation mechanism fails here.

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.

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.

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.

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.

Page 9, Lines 11 – 14: Since the profiles derived are $\sim>$ 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)?

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.

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?

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

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.

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"

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.

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."

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)

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

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

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.

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?

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)?

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.

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.

Page 13, Lines 16 – 18: This argument could be improved from my previous com-

ment regarding derived SAWNUC sulfuric acid profiles over mid-latitudes compared to measurements over mid-latitudes.

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?

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/acp-2016-583/acp-2016-583-RC1-supplement.pdf

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