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Interactive comment on “Microphysical simulations of new particle formation in the upper troposphere and lower stratosphere” by J. M. English et al.

J. M. English et al.

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Dear F. Khosraw: Please see your comments below with our responses and corresponding modifications to the paper. We have attached the modified paper as a pdf supplement that includes your suggestions as well as the other two reviewers.

Commenter, F. Khosraw: The study by English et al. contributes to the understanding of new particle formation in the UT/LS. These processes are still not well understood and thus studies as the present one are important to improve our understanding. However, some parts of the paper are not well written and could be improved. First of all, their major finding is not that the aerosol distribution for particles greater than 10 nm is

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controlled by coagulation and not nucleation. It is generally known that nucleation does only affect the small size bins. Further, it is also generally known that coagulation is only dependent on the size of the particle and not on the composition. Thus, it is clear that coagulation is independent of the nucleation process and gets as stronger as more particles are newly produced (e.g. after a volcanic eruption like Pinatubo one will find the strongest effect of coagulation on the particle distribution). This is a result one gets as soon as microphysical model simulations considering nucleation and coagulation are performed (e.g. Khosrawi and Konopka, 2003; Khosrawi et al., 2010). However, it is generally not stated that clearly in previous publications as it is done by English et al. There is nothing wrong with this statement, but they should include some references since this finding is not really new.

Authors' Response: We agree that numerous other papers have results that support our conclusion that coagulation is more important than nucleation at sizes greater than 10 nm. However, we present the first time this conclusion is clearly stated and supported by observations in the UTLS region. We added a reference to Snow-Kropala et al (2011) at line 60 which is relevant to ion nucleation. Your references (Khosrawi and Konopka, 2003; Khosrawi et al 2010) provide good theoretical relationships between atmospheric mixing, nucleation, and coagulation, but we do not see how they relate to the focus of the paper.

The major findings of English et al. are that (1) IMN is quite important in the UT/LS which is in agreement with the findings by Lee et al. (2003) and (2) that one gets very different results dependent on which BHN scheme is used. However, here it would be nice if they could give a recommendation which BHN scheme is in better agreement with measurements. For the evaluation of their model they only perform a comparison with one of the schemes, but it would also be interesting to know how the other BHN scheme performs compared to measurements.

Authors' Response: We respectfully disagree with the above statements. While it is true that nucleation rates differ significantly between schemes, there is very little dif-

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ference between aerosol properties when comparing number concentrations above 10 nm – see Figures 8, 10, 11, 13, 14, 15, and 16. Although we structured the paper by beginning with model validation of the Zhao BHN scheme, one can see from these same Figures that we could have evaluated the model with any of the other schemes, because comparison to observations is not driven by nucleation scheme. To support our choice to start with just one nucleation scheme, we have added at line 218: “We have found that for most particle properties all three nucleation schemes produce nearly identical results. Therefore, below we first compare simulations using one nucleation scheme (Zhao BHN) with observations. Later we highlight where the schemes differ.” To clarify our findings we added at line 492 “Lee et al. (2003) suggested that ion nucleation was important in the UTLS, on the basis of their ability to match observed size distributions with a model based on ion clusters. In contrast, we find that identical size distributions are produced for each type of nucleation, due to the dominance of coagulation. Hence fitting the size distribution is not diagnostic of the type of nucleation occurring (binary or ion). Unfortunately, the rate of nucleation is not easily determined from data either because all of the particle properties for particles larger than 10nm are not altered even for two order of magnitude changes in the nucleation rate.”

Another general point of criticism is that too many figures are included compared to the length of the text. Especially, since most of the figures concern the evaluation of the model which as is not the main focus of this paper. I would thus suggest that the authors check if some of the figures can be omitted.

Authors' Response: We wanted to evaluate the model against as many observations as possible, and believe that each of the figures helps to characterize our calculations. The other reviewers did not suggest that any figures be removed.

Some specific comments: p12442: It would be interesting to know which altitudes were considered. Which altitudes are considered here as UT/LS?

Authors' Response: UTLS is not defined by a standard altitude region. We com-

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pared our model to observations constrained to the region the observations were measured. In a few instances we also looked at the middle and upper stratosphere, so we have changed line 14 to: “. . .we studied aerosol formation and microphysics in the upper troposphere and lower stratosphere (UTLS) as well as the middle and upper stratosphere. . .”. To clarify our general definition of UTLS, we have added at line 40: “The UTLS region does not have a standard spatial definition, but we generally refer to the region between 50 and 500 hPa.”

p12442, l4: Here you write “tropical” UT/LS, but in section 4.2 also the UT/LS of the midlatitudes and polar regions is considered.

Authors’ Response: The tropical region is where nucleation occurs, but some microphysics and comparisons occur at mid and high latitudes. We agree that this is confusing and have removed the word “tropical” from line 14.

p12442, l10: As you write it here it is quite confusing and misleading. With only reading the abstract it sounds like that nucleation is not important in the upper troposphere. The problem is that you first describe that you apply three nucleation schemes and when suddenly describe one of your results concerning the further development of the particle distribution without describing the results of the comparison of the nucleation schemes. Further, it is also not stated what you mean with atmospherically relevant processes. Thus, I would suggest to add some more details and move this text part further down.

Authors’ Response: We have shown that differences in nucleation rates of up to 2 orders of magnitude are not important to atmospherically relevant processes. We have clarified atmospherically relevant by adding (line 21): “Therefore, based on this study, processes relevant to atmospheric chemistry and radiative forcing in the UTLS are not sensitive to the choice of nucleation schemes.”

p12442, l18: Similar as above. Your main focus is the comparison of the nucleation schemes, but in the abstracts nothing concerning the results on the performance of

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the nucleation schemes is mentioned, but it is discussed what happens with the particle distribution after the particles have been formed. In general, I would say that the abstract needs some more structure and a clear line what has done and what are the results of this study.

Authors' Response: We have added to line 18: "None of the nucleation schemes is superior at matching the limited observations available at the smallest sizes. However, it is found that coagulation, not nucleation, controls number concentration at sizes greater than approximately 10 nm. " We also added to line 492: "Lee et al. (2003) suggested that ion nucleation was important in the UTLS, on the basis of their ability to match observed size distributions with a model based on ion clusters. In contrast, we find that identical size distributions are produced for each type of nucleation, due to the dominance of coagulation. Hence fitting the size distribution is not diagnostic of the type of nucleation occurring (binary or ion). Unfortunately, the rate of nucleation is not easily determined from data either, because all of the particle properties for particles larger than 10nm are not altered even for two-order-of-magnitude changes in the nucleation rate." It is unfortunate that observations in the UTLS are so limited that we cannot constrain which nucleation scheme is best at matching observations of number concentration at the smallest sizes (<10 nm).

p12442, I21: Why are now the results of the upper stratosphere discussed? The paper focuses on the UT/LS, why then mention aerosol properties at 30 km in the abstract?

Authors' Response: The paper focuses on new particle formation in the tropical UTLS, but a model evaluation that includes comparisons to observations at all latitudes and in the middle and upper stratosphere too. We have added to line 14 "...we studied aerosol formation and microphysics in the upper troposphere and lower stratosphere (UTLS) as well as the middle and upper stratosphere...".

p12442, I25: I doubt that particles that are produced in the UT/LS will make the way down to the boundary layer. Even vice versa only a minority of particles succeeds to

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reach up to the UT/LS (the ones that e.g. do not serve as CCN). Further, especially sulphuric acid/water particles will melt on their way down due to the increasing temperatures. p12242, l25: Check the structure of the sentence, something went wrong here

Authors' Response: We have added "possibly descend" this low, and changed "into" to "to". This is a published theory by Clarke; we are not defending or attacking it, just mentioning it. Line 39: "These particles may cross the tropopause and accelerate stratospheric ozone destruction via heterogeneous chemistry (Hofmann and Solomon, 1989), impact climate by modifying cirrus cloud properties in the upper troposphere (Jensen et al., 1996), and possibly descend to the marine boundary layer and act as cloud condensation nuclei (CCN) there (Clark, 1993).

p12443, l1: I would suggest to include "on the surface of Polar Stratospheric Clouds (PSC) particles" after "heterogeneous chemistry".

Authors' Response: The ozone destruction can result from heterogeneous chemical reactions on PSCs, as well as on the sulfate particles themselves; therefore we have not modified this sentence.

p12446, l8-10: One should differentiate here between pure organics as found in the boundary layer and sulphate-organics as found in the upper tropical troposphere (Froyd et al., 2009). Further, some references should be added and differences in UT compositions at different latitudes should be discussed. Further, the kind of aerosols that are found in the LS is still unclear since there are not so many measurements characterising aerosols in this region.

Authors' Response: Murphy et al. have found that sulfates are the primary component above the tropopause. We have added the Froyd et al. reference (line 133-137): "Although other aerosols, such as organics, are known to compose a significant fraction of the sulfate aerosol mass in the UTLS (Froyd et al., 2009, Murphy et al., 2007), sulfates are believed to be the primary source of new particles in this region, and the

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primary aerosol in the lower and middle stratosphere (Murphy et al., 2007)."

p12448, l26-27: Somewhat earlier you write that this nucleation scheme gives unrealistic results in the middle and upper stratosphere, but now it is stated above the tropopause. If these values are really not reliable in the lower stratosphere why then using this scheme for a UT/LS study? If the problems really occurs at the altitude regions considered here it should be more clearly stated. p12449, l10: same comment as for p12448, l26-27.

Authors' Response: Although the tables were originally designed for the troposphere, they should be accurate in the UTLS. In the middle stratosphere and above, input conditions are outside the bounds of the table. The paper now reads at line 201: "However, there may remain some unrealistic nucleation rates in the middle stratosphere and above due to the boundary conditions of the lookup tables. The tables should be used with caution in these regions."

p12449, l13: Measurements are usually "validated", but models are usually "evaluated".

Authors' Response: We have changed the section heading to "Model Evaluation" at line 217.

p12449, l14: From what you write about the other schemes, it feels a bit like cheating now. For the evaluation the scheme without any restrictions is used, but how the results will be affected by the schemes with restrictions is not further discussed.

Authors' Response: In writing this paper, we have conducted many simulations, including a comparison of all three nucleation schemes, both with and without the VW correction. In all comparisons, the nucleation schemes lead to very similar number concentrations > 10 nm, aerosol mass, area, volume, extinctions, and effective radius. We added at line 218 "We have found that for most particle properties all three nucleation schemes produce nearly identical results. Therefore, below we first compare simulations using one nucleation scheme (Zhao BHN) with observations. Later we

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highlight where the schemes differ.”

p12451, I1: Why do the sulphate aerosols evaporate? This should be explained a bit more.

Authors' Response: We have added to line 260: “It is so warm and dry above 35 km that the sulfuric acid vapor pressure exceeds the total (gas + particle) mixing ratio of sulfuric acid; hence the particles completely evaporate.”

p12455, I10-13: Is that a really good solution? In general the problems with the unrealistic nucleation rates occurs in the middle and upper stratosphere, but here the lower stratosphere is considered, thus the effect on the present results should be low. It should be more clearly stated why these nucleation schemes are applied for the lower stratosphere though they were developed for the troposphere, what the errors are and how this affects the present results.

Authors' Response: We have added at line 381: “However, an analysis of the input parameters has found that the tables behave well in the UTL region and below.”

p12456, I1: Which time period has been considered?

Authors' Response: In our model, we looked at all simulation output across the year – we write at line 394: “Simulation ‘data’ points include values for 360 days in the third simulation year.” We have added a caveat to this comparison at line 396: “Additionally, since the model output is across the entire year, while the aircraft data are obtained on specific days, differences may be due to temporal variability.”

p12456, I2-4: These results could be expected and are in agreement with the theory. Some references are definitely missing here.

Authors' Response: This might be expected, but as far as we know only Lee et al have discussed this.

p12459, I2-5: These results are not really new and also here some references are

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missing. Further, I would suggest to move the text starting in line 9 to line 5, so that the discussion on the effects of coagulation on the particle distribution is not splitted into two different parts. Finally, a study which might be of interest for the present study presented here is the comparison of different nucleation models by Korhonen et al. (2003).

Authors' Response: We had referenced Kazil and Yu in section 4.1, but will repeat them here on line 483. Also, we have added your recommended Korhonen reference on line 485 and in section 4.1, and added it to the list of references. Finally, we have restructured the sentences per your suggestion. The paper now reads at line 482: "Calculations suggest that ion-mediated nucleation rates in the UTLS are 25% higher than binary only, consistent with some studies (e.g. Kazil et al., 2010) but different from others (e.g., Yu et al., 2010). However, the two binary schemes vary by two orders of magnitude, consistent with other studies (Korhonen et al., 2003, Yu et al., 2010). More importantly, it is found that coagulation, not nucleation, controls number concentration at sizes greater than approximately 10 nm in the UTLS and the middle and upper stratosphere. The dominance of coagulation over other microphysical processes is consistent with other recent work using microphysical models; Pierce and Adams (2009) found coagulation to be more important than nucleation in tropospheric studies, and Timmreck et al. (2010) found coagulation to drive stratospheric particle sizes from the eruption of Mount Toba to much larger values than previously assumed. Lee et al. (2003) suggested that ion nucleation was important in the UTLS, on the basis of their ability to match observed size distributions with a model based on ion clusters. In contrast, we find that identical size distributions are produced for each type of nucleation, due to the dominance of coagulation. Hence fitting the size distribution is not diagnostic of the type of nucleation occurring (binary or ion). Unfortunately, the rate of nucleation is not easily determined from data either, because all of the particle properties for particles larger than 10nm are not altered even for two-order-of-magnitude changes in the nucleation rate."

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Thanks for helping us improve this paper.

Sincerely, Jason English, Brian Toon, Michael Mills, and Fangqun Yu

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/11/C6803/2011/acpd-11-C6803-2011-supplement.pdf>

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12441, 2011.

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