Thanks for the quick response. I appreciate it!

I have a comments on a few of the points below.

*Reviewer: The paper has several points that are confusing (e.g. the "maximum threshold of reactant concentration" and the "SOG nucleation) as well as claims that this paper is the first to evaluate SOG nucleation. I requested clarification of these issues as well as citation of the previous modelling work that included SOG nucleation during the quick review (pre-ACPD) stage. The authors chose to not make any of these revisions at the quick review stage even though *none* of these changes were scientific (beyond clarification of techniques), changed the conclusions of their paper, or would have taken much time to revise. Yet, these changes would have (1) enhanced my ability to review the paper in this round by allowing me to better understand their approach and (2) prevented the authors from falsely claiming they were the first to consider SOG nucleation in a model. Because I will need to consider what the authors are actually doing with the "maximum threshold" and "SOG nucleation" approaches once they have been explained to me more clearly, I will require at least 1 more set of reviews (or maybe this could be done by iterated responses on the ACPD discussion site). Otherwise, while a have a decent number of revisions, I feel that they are generally on the "minor" side of things in that I don't think new model simulations are required.*

Authors: We would like to thank the reviewer for his comments on the manuscript. We are happy to enter into the on-line discussion of the manuscript now. We believe that this is the right time and place during the review process for manuscripts to be published in ACP where e.g. techniques described in the manuscript can be clarified or more references can be added. According to the guidelines for the review process in ACP, technical corrections can be suggested (typing errors, clarification of figures, etc.) before publication in ACPD. However, further requests for revision (e.g. clarification of methods) of the scientific contents are not allowed. They shall be expressed in the interactive discussion following publication in ACPD. In our opinion, these guidelines help to avoid modifications of the original ACPD manuscript, so that the published ACPD manuscript only reflects the author's opinion (although this might need revisions), and not already that of the reviewers. However, by insisting to stick to these rules, we never questioned the value of the reviewer's comments and appreciate the time and effort the reviewer has spent to read the manuscript and provide constructive comments to improve the manuscript. Below we address the major and minor comments in detail.

The exact quote from the "Review Process" page on the ACP site is "The Co-Editor is asked to evaluate whether the manuscript is within the scope of the journal and whether it meets a basic scientific quality. If necessary, he may ask independent referees of his choice for support. The Co-Editor can suggest technical corrections (typing errors, clarification of figures, etc.) before publication in ACPD. Further requests for revision of the scientific contents are not allowed at this stage of the review process but shall be expressed in the interactive discussion following publication in ACPD."

I do not see any explicit mention of "clarification of methods" not being allowed. I would interpret "clarification of methods" as following "typing errors, clarification of figures, etc." and not "revision of the scientific contents" (why would clarification of figures be allowed but not clarification of methods? both aid in understand the manuscript). I don't see how any of my requests would have altered the scientific contents. I did not ask for you to change your methods, your results, your interpretation of your results etc..

Regarding preserving authors opinions and not including the reviewers opinions (which isn't explicitly addressed on the Review Process page anyways), I agree that me requesting citations to be added reflects my opinions on what literature is relevant for the background and that this probably wasn't appropriate at the initial review stage (I won't do this in the future). However, I'm not sure how clarifying your nucleation and oxidation schemes would have reflected my opinion in any way (it would only explain your work more clearly). The same for asking to fix the false claim that no one had modelled SOG nucleation.

Thus, unless I am way off with my interpretation of what is written on the Review Process page, revising based on my request was not against any rules (except for the request for additional references).

Major comments:
1. SOG nucleationReviewer: 1a. First, there are at least 2 papers that have included SOG nucleation in global modelling studies: - The Metzger et al., 2010 paper that the authors already cite has global modelling showing the impact of organics on nucleation globally. - Scott, C. E., Rap, A., Spracklen, D. V., Forster, P. M., Carslaw, K. S., Mann, G. W., Pringle, K. J., Kivekäs, N., Kulmala, M., Lihavainen, H., and Tunved, P.: The direct and indirect radiative effects of biogenic secondary organic aerosol, Atmos. Chem. Phys. Discuss., 13, 16961-17019, doi:10.5194/acpd-13-16961-2013, 2013. Please remove the claims that no one has tested this before and cite/discuss their work.

Authors: Thanks for this advice. In the revised manuscript we will for sure remove the sentences 'To our knowledge, three-dimensional modelling studies on SOG nucleation are not yet available.' in the introduction and '. . . but to our knowledge they have not yet been evaluated in three-dimensional atmosphere-chemistry aerosol model simulations.' in the conclusions. Please note, however, that nucleation is not the primary focus of the manuscript, considering that nucleation events at Puy-de-Dôme were rare during the investigated period.

Thanks

Reviewer: 1b. How are the authors actually doing "SOG nucleation". They say that their nucleation scheme is Vehkamäki et al. (2002); however, this nucleation scheme is a H2SO4+H2O binary scheme (that only predicts nucleation under cold, free-tropospheric conditions in most models... not in the boundary layer).

Authors: The parameterisation for sulfuric acid-water nucleation of Vehkamäki et al. (2002) has been developed for tropospheric and stratospheric conditions, valid for a temperature range from 230.15-300.15 K, relative humidities of 0.01-100% and total sulphuric acid concentrations of 104-1011 cm-3. As aerosol microphysical parameterisations of the regional model REMOTE (Langmann et al., 2008) are based on those of ECHAM5-HAM (Stier et al., 2005), it is the basic nucleation scheme of the model, despite potential limitations.

Reviewer: Thus, it is not clear how the authors are doing SOG nucleation. I have thought of two possibilities of what they might be doing: (1) They are treating LV-SOG as the same as H2SO4 and using [LV-SOG]+[H2SO4] as an input to Vehkamäki et al. (2002) as opposed to just [H2SO4]. However, there is no basis for this method.

Authors: In recent years, the awareness that nucleation parameterisations involving only H2SO4 (and water) do not yield satisfying results triggered the development of nucleation parameterisations including organic vapours. Paasonen et al. (2010) (and Kerminen et al. (2010)) proposed eight different empirical nucleation parameterisations derived from combining data from four measurement sites. Inspired by these ideas, the simple assumption applied in the current manuscript is to use the scheme of Vehkamäki et al. (2002) for two nucleation pathways: 1. H2SO4+H2O nucleation and 2. LV-SOG+H2O nucleation. For the second pathway H2SO4 concentrations were replaced by LV-SOG concentrations in the nucleation scheme. We certainly agree with the reviewer that such an approach represents a simplification (maybe even an oversimplification), as e.g. interactions between H2SO4-SOG nucleation are not considered.

Nevertheless, we would like to emphasis that – even in a simple way – nucleation of organic vapour is taken into account in the model simulations presented in the manuscript.

Thanks for this clarification. However, since there is no experimental or theoretical basis as to why replacing H2SO4 with LV-SOG in the Vehkamaki scheme would give realistic nucleation rates (even though we know that LV-SOG can participate in nucleation), this needs to be clearly discussed in the manuscript. It's ok for us modellers to be creative with our approaches (I'm sure I've done crazier approaches than this), but we need to do our best to be clear about when we are making our own approximations and what the potential limitations of these approximations are.

Reviewer: (2) They are using Vehkamäki et al. (2002) as just H2SO4+H2O, but since they are condensing LV-SOG onto the nucleation mode, they are calling this "SOG nucleation". However, this latter approach is not SOG nucleation at all, it is binary (H2SO4+H2O) nucleation followed by condensation of LV-SOG to already-nucleated, stable aerosols. If this is the case, the authors should not be calling this SOG nucleation at all. Furthermore, this would be in no way novel as many papers have used condensation of non-volatile SOA onto freshly nucleated particles (see any global aerosol microphysics modelling paper out of the following groups since about 2008 or 2009: Ken Carslaw, Dom Spracklen, Risto Makkonen, Peter Adams, Jeff Pierce... there are probably _25 papers that already do this... furthermore Riipinen et al. 2011 is entirely about how important this initial LV-SOG condensation is in new-particle growth). I'm curious as to why the authors did not just use the scheme published in Metzger et al. 2010 for SOG nucleation that explicitly has SOG in the scheme? This seems like the easiest way to have SOG nucleation in a model at this time.

Authors: As outlined above and hopefully described in a more clear and understandable way, nucleation of organic vapour is taken into account in a simplified way in the model simulations presented in the manuscript. Condensation of LV-SOG is considered in addition (see Fig. 1 of the manuscript).

Yes, thank you.

2. "maximum threshold of reactant concentration"
Reviewer: "To further simplify the approach of Yu (2011) we determine the mass conserving trans-formation rate of MV-SOG -> SV-SOG and SV-SOG -> LV-SOG by prescribing a maximum threshold of the reactant concentration being available for oxidation. This way we avoid determining the saturation vapour pressure of the oxidised SOG compounds, which greatly simplifies the procedure proposed by Yu (2011). Results with 1% and 10 % threshold values (in the latter case an additional requirement is that OH concentrations exceed 0.1 ppt thereby excluding night-time aging processes) are presented in Sect. 4.2." I'm not exactly sure what the authors are doing here. My best guess is that if "k" is the aging rate constant, they are predicting d[LV-SOG]/dt by... d[LV-SOG]/dt = k[OH]*(0.1*[SV-SOG]) for the 10% threshold or d[LV-SOG]/dt = k*[OH]*(0.01*[SV-SOG]) for the 1% threshold rather than d[LV-SOG]/dt = k*[OH]*[SVSOG] (and similar for the aging of MV-SOG to SV-SOG). However, if this is the case, isn't this the same as scaling the aging rate constant down by 10% and 1%. I'm not sure what the basis of this would be. My only guess is that perhaps the un-scaled rate constant created chemistry too fast for their modelled time step (causing negative concentrations under some conditions) and rather than reducing the timestep, they reduced the rate constant. However, I am only speculating because I really don't follow the reasoning for the maximum threshold. Also, why does this procedure avoid determining the saturation vapor pressure of the oxidized SOG components? The authors have these pure-value vapor pressures in Table 1 for SV and MV (and the authors can calculate the sat vap pressures over a mixture from partitioning theory), and LV the authors assume to be non-volatile, so I'm not sure why the authors need to avoid determining*

them. And I also don't know why this max threshold would allow the authors to avoid determining the saturation vapor pressures.
The discussion of the "maximum thresholds" needs to be clarified.

Authors: The reviewer understood the concept of thresholds correctly regarding the introduction of the threshold value in the aging reaction equations. However, we strongly reject speculations about a too large time step and negative concentration – both do not appear in the model simulations described in the manuscript. The reason for the implementation of thresholds is a conceptual one. As LV-SOG represents the lowest volatile SOG, further aging is not considered (see Figure 1 of the manuscript), and the concept with maximum thresholds is only applied for MV-SOG and SV-SOG aging. The concept with maximum thresholds for MV-SOG and SV-SOG aging has been introduced into the model, to take into account that only a fraction of the respective SOG's will reach saturation vapour pressures low enough to move into the next category (MV-SOG into SV-SOG and SV-SOG into LV-SOG). As the oxidation products are represented by only one component in each category, this way the spread of saturation vapour pressures of different oxidation products is implicitly considered. Other authors limit the category jump by e.g. assuming that each OH oxidation adds one oxygen atom and reduces C by 1.5 decade (Yu, 2011) or introduce C* bins and determine the mass yields of products in each C* bin (Donahue et al., 2006). We will clarify the concept of thresholds in the aging equations in the revised manuscript according to the above written explanations and also by better pointing out that this empirical approach makes use of the Puy-de-Dôme measurements (shown in Fig. 6 of the manuscript) for scaling.*

Ok, this explanation is much better. I think the concept of "the fraction of oxidation products that have a volatility low enough to move to the lower volatility class" is solid, and it is fine for publication once it is explained clearly as you describe here.

Can you explain better how this "greatly simplifies the procedure proposed by Yu (2011)"? It seems like the procedures are similar, but you just scale your production rates by 0.01 or 0.1.

Reviewer: P26767 L7-9: Why couldn't the authors apply the quasi-steady-state approximation here? Please clarify.

Authors: P26768? The equilibrium approach assumes that the secondary organics in the particle phase and gas phase are always in instantaneous equilibrium. This represents a good approximation for organics with relatively high saturation vapour pressure. However, when ignoring secondary organics in the gas phase during transport, and only transporting the organics in the particle phase, the approach is better valid for low volatile species with only low gas phase concentration. Therefore, we argue that for both, secondary organics in the particle phase and gas phase, transport processes should be taken into account, because otherwise the mass of secondary organics in the gas phase is lost (see Fig. 4 of the manuscript (green line)).

Yes, 26768, sorry.

I'm still confused here. The quasi-steady-state approximation is not the same as assuming instantaneous equilibrium (the quasi-equilibrium approach). In the paper you say quasi-steady-state approximation, but in the response, you only mention equilibrium. The quasi-steady-state concentration of a species will be out of gas-particle equilibrium if there is net production or loss of that species (but mass transfer is balancing production, so you get essentially a constant concentration in the gas phase). In general the quasi-steady-state approximation is used for species with low-vapor pressure such as sulfate (e.g. Pierce, J.R., Adams, P.J., A computationally efficient aerosol nucleation/condensation method: Pseudo-steady-state sulfuric acid, Aerosol Science and Technology, 43, 216-226, 2009.). In the case of low-volatility material, there is not much in the vapor phase for transport. Thus, I'm not sure why QSS wouldn't work.

Did you mean to say "quasi-equilibrium approximation" in the manuscript? If yes, than things make sense to me.

Reviewer: P26767 L19-21: Vehkamäki et al. (2002) is just for H2SO4 and H2O, not SOG. Why not Metzger et al. (2010), which actually accounts for LV-SOG concentrations?

Authors: see answers above

When you add the description of how LV-SOG to Vehkamaki, can you add discussion of why you chose to do this method rather than using Metzger?

Reviewer: P26772 L21-25 and Figure 6: Are the authors comparing the modelled LVSOA to the measured LV-OOA and comparing the modelled SV-SOA+MV-SOA to the measured SV-OOA? I didn't find this explicitly stated. The authors should use some caution here because OOA in the AMS can be aged POA. Since the model doesn't have aged POA in this comparison, this could be a source of error.

Authors: As written in the manuscript, we compare modeled LV-SOC to measured low-volatile SOC and the modeled sum of MV-SOC and SV-SOC to measured semi-volatile SOC (page 26772 lines 21-23). We will correct the y-label of Fig. 6 (SOC instead of SOA) and add the above description to the figure label as well. Concerning POC, we agree with the reviewer, that AMS measurements may include aged POC, however, this contribution cannot be separated from SOC. We will add a sentence about this source of uncertainty to the revised manuscript.

The AMS technically doesn't measure SOC (or SOA), what you are using, I believe, is the SV-OOA and LV-OOA (oxidized organic aerosol) or the carbon-only portion of these (SC-OOC and LV-OOC). I assume you equating SOA and OOA. This is why I said "I didn't find this explicitly stated" because it wasn't clear what the "measured SOC" was. Please also describe this explicitly along with the aged POC that you mentioned.

Reviewer: L26772 L28: Why wasn't 100% aging tested? I assume this would the same as Yu... but this goes back to me not understanding the "maximum thresholds".

Authors: see answers above

Would 100% aging be the same as Yu? I'm thinking yes, but the "greatly simplifies the procedure proposed by Yu (2011)" comment in the methods section makes me think that perhaps something else changed.