The author thanks the referee for the critical reviewing of the manuscript and constructive comments. My point-to-point replies to the comments are given below (the original comments are copied here in *Italic*). The manuscript has been revised accordingly.

General Comments
As the title states, this manuscript describes a "new" model that attempts to improve the capability of simulating secondary organic aerosol on a global scale, a topic well within the purview of ACP. The text is generally clear and easy to follow, and the abstract is appropriate for the contents of the manuscript. However, based on the specific comments discussed subsequently, I am unable to recommend its publication. I do not find that previous work is cited adequately. There are several figures that could be removed or require clarification. Most importantly, I do not find the model approach to be terribly novel, and I believe that there may be some errors in the model formulation. Therefore, it is impossible to say whether the results (and therefore the interpretation of these results) are correct.

I don't think that I claim in the manuscript that the model approach is very novel. That's why I named it "extended" model (not "new" model) as it built on the existing 2-product (or 2p) model and recent findings about the importance of the aging of SOGs in the atmosphere. The main objectives of this study are to extend the 2p SOA formation model in GEOS-Chem so that it can take into account SOGs aging process and the kinetic condensation of low-volatile SOGs, without adding substantially to the computational burden of the model. To my knowledge, this work represents the first global size-resolved aerosol model that considers the explicit kinetic condensation of low volatile SOGs. Based on several specific comments given below, the referee appears to have misunderstood the extended SOA model described in this manuscript and main objectives of this manuscript.

The SOA formation process is very complex and some kind of simplification and approximation for global representation are unavoidable. This extended model is not aimed to solve all the problems associated with SOA formation in the global model, instead it is to improve or extend the existing representation of SOA formation in the GEOS-Chem. I think that the results presented in the manuscript provide some useful insights which can further our understanding of SOA formation process and its representation in the global aerosol model. In addition, the scheme can reasonably account for the well-recognized contribution of low volatile organics to the condensational growth of nucleated particles, which is not treated in most of the global aerosol models (as far as I know).

Errors in the SOA model formulation are possible and uncertainties are unavoidable. I have been trying very hard to identify such errors or uncertainties and correct or reduce them and will continue to do so in the future studies. I hope that the schemes of the extended SOA formation described in the manuscript are clearly enough for others to reproduce the results and check the possible errors in the schemes. In addition, after the publication of this manuscript, I will seek to include this extended scheme for inclusion in the standard version of GEOS-Chem that will be released to public. Then other interested researchers can check for possible errors and continue to advance the representation of SOA formation in the global model.

Specific Comments
Page 19814, Line 18. This statement is not exactly true. According to the Odum papers, the 2-product model has 2 products in it as a result of using least-squares fitting of experimental versus calculated yields in chamber experiments. Having an additional product showed no benefit with respect to the ability of the model to fit the data and therefore was superfluous mathematically.

It appears that the referee misunderstood the extended scheme described in the manuscript. The additional product (LV-SOG) added in the extended scheme is not to re-interpret or re-fit the chamber data but to consider the aging of 2 products (MV-SOG and SV-SOG) in the atmosphere after first generation of oxidation (See discussion in section 2).

Page 19815, Line 28, a reference is needed here (perhaps to Pankow or to the volatility basis set papers).

A reference to Pankow and Barsanti (2009) has been added.

Page 19816, first paragraph. This introduction is woefully incomplete. The author gives the impression that the 2-product model developed by Odum _15 years ago is the only model used to simulate secondary organic aerosol. However, an incredible amount of work has been done to improve upon the original 2-product model (all described in USEPA documentation as well as in recent papers from that group). In addition, other model approaches are completely ignored. For example, the volatility basis set (VBS, from the CAPS group at Carnegie Mellon) is not mentioned beyond the relationship between a partitioning coefficient (Pankow) and a saturation concentration (Donahue). Pankow's new model is note mentioned at all either. Semi- explicit (CACM/MPMPO of Pun, Griffin, and co-workers) and completely explicit (based on the Master Chemical Mechanism or self-generating mechanisms such as that of Aumont) are not even mentioned. This is completely inappropriate given that the "novel" model described in this manuscript is very similar in some aspects to these other approaches. For example, the conversion of material into less volatile material is not a new idea, as it is one of the key features of the USEPA improvements to the 2-product model, the VBS, CACM/MPMPO, and the explicit approaches.

Points are well taken. I have added a paragraph in the introduction to review other approaches of simulating SOA formation mentioned by the referee.

Page 19818, Line 14. I am troubled by the lack of inclusion of HV-SOG. If LV-SOG are not formed directly (perhaps they should be?), whatever material has been reacted goes to either SV, MV, or HV. With known alpha parameters for SV and MV and assumed molecular weights for these products, it is possible to calculate the yield of HV material. This is a feature in each of the SOA models described above yet not dis-cussed in this manuscript. There are also examples in the literature of what would likely be considered HV material forming aerosol after an additional generation of oxidation. This model does not account for this at all. The author should also use caution in ascribing physical properties (SV or MV) to products that are completely hypothetical

(or that are based solely on a fitting procedure; these products are not unique, and another different set of parameters would give only slightly different results in the fitting procedure).

I apologize for forgetting to define clearly the HV-SOG in the manuscript. As detailed in the text, SV-SOG and MV-SOG correspond to the "two products" in the widely used 2p SOA model. HV-SOG refers to those high volatile oxidation products of VOCs (i.e, products other than those SOGs indicated in Equations 3 and 4 which experimental yielding information are available). Many HV-SOGs may be further oxidized to CO_2 (or other volatile carbon species) but some unknown fractions may be end up as SOA. As I understand, the widely used 2-product model (at least the one implemented in GEOS-Chem) does not track HV-SOG. To avoid confusion, I have deleted the discussion of HV-SOG in the revised manuscript.

I am not convinced that Equations (6) and (7) are exactly correct; they also raise several questions about the model formulation that are not addressed in the manuscript. These equations represent the time rates of change of the concentrations of MV and SV SOG. However, the reactive loss term only includes the fraction of the material that would be converted into the less volatile species upon reaction, yet in theory all of the material should react. Does this then mean that for each time step, the predicted concentration of MV or SV material is too large since the reactive loss term is too small? It is appropriate for this factor (ξ) to appear in the formation term for the products. What happens to the material that has reacted yet does not move down in volatility? Does it just float around in its volatility class ad infinitum or does it not react? If the same material were to be oxidized multiple times, should that material either move down (by addition of O) or up (by fragmentation) in volatility (as discussed recently by Kroll, Donahue, et al.). Additional model questions that are not addressed: How are the loss rates to partitioning and condensation calculated? Is it appropriate to use one aging rate constant when the molecular structure is very likely to change as you move from HV to MV to SV?

The referee raised some good points here.

It is true that "the reactive loss term only includes the fraction of the material that would be converted into the less volatile species upon reaction, yet in theory all of the material should react". However, this does not mean that "for each time step, the predicted concentration of MV or SV material is too large since the reactive loss term is too small". The reason that we introduce the variable fraction (ξ) in our formulation is due to the large range of saturation vapor pressures for each category of SOGs (from different VOCs and at different temperatures). In the real atmosphere, all SOGs should react but only a fraction of SOGs in the category (MV-SOG, SV-SOG) have saturation vapor pressure lower enough to be moved to the next category (i.e, MV-SOG to SV-SOG or SV-SOG to LV-SOG). As discussed in the text (pages 19819-19920), ξ values depend on the decrease of SOG vapor pressure due to oxidation and the ratios of C^*_{MV} to C^*_{SV} (or C^*_{SV} to C^*_{LV}) which differ for different SOGs and vary with temperature.

The material that has reacted but does not move down in volatility (category) will not float around in its volatility class forever. The volatility change within the category is taken into account by assuming that the log-normal distribution of each SOG category is always maintained after a faction of the SOG in the left-tail (lowest C*) of log-normal distribution has been moved down to the next category. Therefore, the same material will have chance to be moved down after being oxidized multiple times. This approximation enables us to represent the C* changes associated with oxidation aging in the global model while keep the computational cost at a reasonable level. This has been clarified in the revised manuscript.

The loss rate to condensation is calculated via condensation growth equation (see for example, Eq. 10 in Yu, ACP, 6, 5193–5211, 2006) and to partitioning is based on the scheme described in Chung and Seinfeld (J. Geophys. Res., 107, 4407, 2002). The possible value of aging rate constant is another uncertainty of the model and the value used in this study is based on Jimenez et al. (Science, 326, 1525, 2009). The uncertainty can be reduced if more laboratory data of aging rate constant become available.

Page 19820, prior to section 3. There are no comparisons to controlled/chamber data for this model. Is it capable of matching chamber data? Has the model been evaluated? Because this is the first publication of this extended model, a model evaluation (besides a comparison to one site!) is required. On page 19825, it is stated that this will be done in a future publication using global AMS datasets. It is my opinion that this needs to be done in the first paper describing this model.

This is another indication that the referee might have misunderstood the extended SOA model described in this manuscript which is built on the existing 2p model and recent findings about the importance of the aging of SOGs in the atmosphere. The 2p model already implemented in GEOS-Chem directly uses various chamber data as detailed in the references cited in the manuscript (Chung and Seinfeld, 2002; Liao et al., 2007). The additional product (LV-SOG) added in the extended scheme is not to re-interpret the chamber data but to consider the aging of 2 products (i.e., MV-SOG and SV-SOG) in the atmosphere after first generation of oxidation (See second paragraph on page 19815, section 2, figure 2). Following the suggestion of Dr. Napelenok, we have revised the title to avoid the confusion.

Following the referee's suggestion, I have added a figure to compare the simulated SOA mass concentrations with those observed by AMS around the globe (as compiled and reported in Jimenez et al., 2009).

Page 19823, top of the page. The author speculates that anthropogenic SOA could significantly affect the results shown. Alpha parameters and emissions inventories are available for many anthropogenic species; these could be included (as opposed to having speculations made about them) assuming that the model formulation is corrected.

The main objectives of this study are to extend the 2p SOA formation model implemented in GEOS-Chem to take into account aging process in the atmosphere and to represent the kinetic condensation of low-volatile SOGs on atmospheric particles in GEOS-Chem. To add emissions and reaction schemes of anthropogenic organic species into GEOS-Chem is beyond the scope of

this study. To address the referee's concern, I have changed the speculation sentence to "Further research is needed to investigate how the oxidation aging of anthropogenic VOCs may affect [LV-SOG] in their source regions."

Page 19823, Line 26. I would not say that is can be seen clearly in Figure 5 that the new model improves the capability to predict number concentrations. In fact, I would prefer a quantitative description of model performance (and improvement over the previous SOA model capabilities).

To address the referee's concern and to show the improvement more clearly and quantitatively, we have added an additional figure which give a comparison of monthly mean size distributions along with the size-dependent correlation coefficients and normalized mean bias. This additional figure shows clearly the improvement in the GEOS-Chem's ability in capturing the particle formation and growth processes when the extended SOA formation scheme is used.

Page 19825, Line 17. It is nearly impossible to see the overlain values for the surface sites on Figure 7a.

To make the overlain values clearer, we have reduced the map lines and improve the resolution of the figure so that one can zoom in (say by 400%) and see the overlain values.

Page 19826, Lines 6-8. I would argue that you need to validate modules used in global models before putting them into the global models (see earlier comment), not just use both aerosol mass and number.

Please refer to reply to earlier comment.

Page 19826, second paragraph. No information at all is given in the manuscript regarding the CCN activity parameters of the SOA material. Because the LV material was not included in previous modeling efforts, it is imperative to include it here.

CCN activity parameters were based on scheme and data given in Petters and Kreidenweis (ACP, 7, 1961-1971, 2007). Has made this clear in the revised manuscript.

Page 19827, first paragraph. The calculations are admittedly crude. However, this end point seems to be a major conclusion of the paper – that this new model shows how important this SOA could be to the indirect effect of particles on climate. Without addressing the concerns previously raised, it is nearly impossible to put any credence in the result presented.

Please see my replies to the concerns raised above.

Figure 1 is not necessary. Saturation concentration increases with temperature. This is known. Figure 2 is also unnecessary. In Figure 3, mass concentrations are converted to particle number concentrations using an assumed molecular weight. Is molecular

Figure 1 is provided not only to show the increase of saturation concentration with temperature but also to give quantitative values of saturation concentration at different temperatures for various SOGs which are used to guide the design of extended SOA model described in the paper. Nevertheless, we agree with the referee that the saturation concentration as a function of temperature can be easily calculated. To address the referee's concern, we have removed Figure 1.

Figure 2. I feel that this schematic illustration is useful to facilitate the discussion and help the readers to understand the particle formation and growth process considered in the GEOS-Chem. One key objective of the extended SOA scheme is to improve the representation of particle formation and growth process in the GEOS-Chem.

Figure 3. Mass concentrations are converted to molecular (not "particle") number concentrations of gas species with the assumed molecular weight (mass of one molecule). Diameter and density of the molecules are not needed for the conversion.

Figure 8. As stated in the text, the difference (or improvement) is small. Following the suggestion given in Dr. Napelenok's review, NME and NMB values have been reported to quantify the difference.

Typographical Errors
In general, the paper was well written, but there are several examples
where greater care is necessary in the editing.
For example (and I believe this list is incomplete): Page 19812, Line 16, fact should be factor. Page 19812, Line 26, of should be to. Page 19813, Line 17, mass should be masses. Page 19813, Line 22, yielding should be yields. Page 19814, Line 3, phase should be plural. Page 19817, Line 21, the should be removed. Page 19827, Line 14, the should be inserted between of and major.

The referee's careful reading is appreciated. These typos have been corrected in the revised manuscript. In order to minimize typographical errors, I have also asked an English native speaker (Kevin Bartlett) to proofread the revised manuscript.