We thank the referee for the constructive comments. Our replies to the comments and our actions taken to revise the paper (in blue) are given below (the original comments are copied here in *Italic*).

Anonymous Referee #3

This paper compares modeled and measured nucleation events in spring and summer at nine locations in North America. Two nucleation parameterizations are used in the model: ion-mediated (IMN) and the CLOUD BioOxOrg empirical parameterization. The paper is very clearly written and the figures and tables are clearly presented. I recommend this paper should be published subject to a few revisions.

We appreciate the reviewer's positive comments about the manuscript.

I have some general recommendations below:

The authors need to clearly present what the novelty is of this study. In the current manuscript, this is not clearly stated. For example, it is well known that new particle formation occurs more frequently in the spring than the summer at these types of locations, and that models have varying degrees of success at accurately predicting this seasonality. Arguably, the large overprediction in nucleation events by the BioOxOrg simulation is the more novel result, so this should be emphasized over the comparison with IMN. The paper seems to take for granted that the IMN mechanism should be considered the "base case" simulation. Also, one of the main objectives of the paper, model-measurement comparison of nucleation events, has been performed many times, including by this research group. I have not found any evidence that this model (APM) is used very widely in the community, so the authors need to justify the scientific significance of evaluating it.

We would like to point out that model (APM) evaluation is not one of the main objectives of the paper. As stated in the last paragraph of Introduction (also first sentence of Abstract), the primary objective of this study is to evaluate the potential role of oxidation products of biogenic VOCs in NPF in the real atmosphere. To achieve the goal, we analyze NPF events and non-events based on PSDs measured over nine forest areas in North America (NA) and compare them to model simulations with and without including organics in the nucleation rate calculation. Since biogenic VOC emissions and their oxidation are strongest in the summer, we use the observed spring and summer contrast in NPF events to study the possible role of organics in NPF and evaluate our current understanding of NPF processes in the atmosphere. The IMN scheme is employed mainly for the comparison purpose (see Section 2.2). We show that "Both Nucl-Org and Nucl-IMN schemes capture the observed high frequency of NPF in spring, but the Nucl-Org scheme significantly over-predicts while the Nucl-IMN scheme slightly under-predicts NPF and particle number concentrations in summer". Our study also indicates that the two schemes predict quite different nucleation rates (including their spatial patterns), concentrations of cloud condensation nuclei, and aerosol first indirect radiative forcing in North America.

We agree with the referee that the large overprediction in nucleation events (in the summer) by the BioOxOrg simulation demonstrated in this study is novel. Other novel features of this study includes: (1) PSDs measured over nine NA forest areas have been compiled and used to evaluate the possible role of oxidation products of biogenic VOCs in NPF; (2) Both Nucl-Org and Nucl-IMN schemes capture the observed high frequency of NPF in spring but their predictions differ dramatically in summer; (3) The two schemes predict quite different nucleation rates (including their spatial patterns), concentrations of cloud condensation nuclei, and aerosol first indirect radiative forcing in North America, both in spring and summer. We think that these novel points have already been clearly reflected in the abstract and in the main text.

In this study the IMN scheme is employed mainly for the comparison purpose (see Section 2.2). We don't think that we say or imply anywhere in the text that the IMN mechanism should be considered the "base case" simulation. Our study shows that the Nucl-IMN scheme slightly under-predicts NPF and particle number concentrations in summer.

The authors should also view there model with a more critical eye. They should specifically justify their SOA mechanism and nucleation mechanism. Have they evaluated their organic aerosols concentrations against AMS data in the past? If so, mention this. It is also possible that the modeled LV-SOG (alpha-pinene) is not representative of actual ambient low volatility organic aerosol, something that is briefly mentioned by the authors but warrants further explanation.

As pointed out in our reply to the previous comment, model evaluation is not the focus of this paper. The reasons for the choice of nucleation mechanisms have been given in Section 2.2 (also Introduction). As to the SOA mechanism, GEOS-Chem uses the two-product SOA formation model originally developed by Chung and Seinfeld (2002). In this study, the extended two-product model that considers the successive oxidation aging of secondary organic gases in the atmosphere, as described in Yu (2011), has been employed. Yes, we have evaluated organic aerosols concentrations against AMS data in Yu (2011) and this has been mentioned in the revised manuscript.

In Section 4, we point out that "while both LV-$SOG_{\alpha\text{-pinene}}$ in the model and BioOxOrg in the chamber studies are later-generation oxidation products of biogenic monoterpenes, it is possible that only a subset of LV-$SOG_{\alpha\text{-pinene}}$ may act as BioOxOrg vapors that are involved in nucleation". This was offered as one of the possible reasons behind the significant overprediction of NPF events and particle number concentrations in summer by the Nucl-Org scheme. What we meant is that LV-$SOG_{\alpha\text{-pinene}}$ in the model may be not representative of BioOxOrg vapors involved in nucleation in the chamber studies. We have clarified this in the revised manuscript.

Specific comments:
1) p. 21274, line 4-5. The authors cite 80-95% total contribution of NPF to CN concentrations, and 50-80% contribution to CCN. Only a previous paper from this group, Yu and Luo 2009, is cited. A more exhaustive review of the literature on NPF contribution to CCN is warranted here.

Also, the definition of "contribution" is also important here, as many of these papers are actually sensitivity studies.

The representative works ("Spracklen et al., 2008; Pierce and Adams, 2009; Yu and Luo, 2009") on the contribution of NPF to CN and CCN number abundance were cited in the first part of sentence. To address the reviewer's concern about the exact percentages and avoid the confusion with regard to the definition of "contribution", we have deleted second part of the sentence and combined the first part of the sentence with the sentence followed:

"Secondary particles formed via nucleation dominate the global total particle number abundance (Spracklen et al., 2008; Pierce and Adams, 2009; Yu and Luo, 2009) and global simulations indicate that the aerosol IRF is quite sensitive to nucleation parameterizations (Wang and Penner, 2009; Kazil et al., 2010; Yu et al., 2012)."

2) p. 21274, line 10. Sulfuric acid should be mentioned here

Added as suggested.

3) p. 21275 line 10. The Egbert mention should include a citation of Pierce et al. (2014) ACP.

Done.

4) p. 21275 line 24 and p. 21276 line 3. Overuse of the phrase "state of the art"

To address the reviewer's concern, we have deleted "state of the art" from the sentence.

5) p. 21277 line 26. Can the coarse resolution model grid (2x2.5) accurately represent nucleation at a specific point?

Yes when nucleation events are regional rather than local. Nevertheless, the coarse resolution could contribute to some of the difference between model results and simulations (Fig. 4).

6) Results section, Fig 3 and 4. Is 10 days and up to one month enough data to make the conclusion that the BioOxOrg parameterization may not be applicable? If the data for more spring and summer months are available, that analysis would make the paper stronger.

In this study, we attempted to use data from multiple forest sites to increase the representativeness of the comparisons. The comparison of 10 days shown in Fig. 3 was an example. Our conclusions are based on comparisons of one spring month at 4 sites and one summer month at 9 sites (Fig. 4). While obviously more data would be helpful, we think that we have enough data to show that the BioOxOrg parameterization as given in Riccobono et al. (Science, 2014) may not be applicable in the summer.