We thank both reviewers for their detailed comments on our manuscript. We have responded to all the comments and made modifications to our manuscript. Our detailed response to the review is below (reviewer comments in red, our responses in black).

Reviewer 1:

In this manuscript, the authors use a global aerosol microphysics model and an offline radiative transfer model to study how the manner in which secondary organic aerosols (SOA) is added to the aerosol size distribution affects simulated changes in CCN-sized particles and the radiative effects of biogenic SOA. They show that the first aerosol indirect effect (AIE) due to biogenic SOA is sensitive to the SOA model treatment. This work highlights the importance of better representation of the impact of SOA on particle size distributions. The content of this work is within the scope of ACP. The following comments should be properly addressed before I can recommend it for final publication in ACP.

Major comments:

1. The authors used two different methods to treat the partitioning of biogenic organics into particle phase ("kinetic" and "equilibrium" approaches). In the "kinetic" approach (Eq. 9), all organics from the oxidation of monoterpene were assumed to be nonvolatile. This may significantly over-predict the growth rate of freshly nucleated particles as only the extremely low volatile part (a small fraction) of oxidation products can directly condense on the nucleated particles. In the "equilibrium" approach (Eq. 10), all SOA were also assumed to be non-volatile and the re-partition of SOA into gas phase were not allowed. This can't really be considered as "equilibrium" approach because it was not "equilibrium" at all (no reparation, no temperature-dependence). The authors should at least discuss the uncertainties associated with the non-volatile assumption of all oxidation products. A figure showing the global distribution of modeled Sorg based on the two approaches is needed to help the reader better understand the results of the work.

   We agree with the reviewer that our mass-based partitioning approach does not represent an "equilibrium" approach; we refer to this approach as "mimicking the thermodynamic" approach (p4152, lines 12-15), we do not specifically call it "equilibrium". To clarify this further we have changed the terminology throughout the manuscript to refer to this as the "mass-based" approach, rather than "thermodynamic"

We also agree that the 100% kinetic approach will overestimate the amount of very low volatility material that may condense in this way. However, this has been the standard approach used in our own model (Spracklen et al., 2006; Mann et al., 2010; Scott et al., 2014) and other global microphysics models (Makkonen et al., 2009; Pierce and Adams, 2009; Makkonen et al., 2012; D'Andrea et al., 2015).

We now emphasise that many models make this assumption, after line 7 on p4153:

"We note that the KinSOA simulation matches the approach used in a wide range of recent global aerosol models (Spracklen et al., 2006; Makkonen et al., 2009; Pierce and Adams, 2009; Mann et al., 2010; Makkonen et al., 2012; D'Andrea et al., 2015)."

We have included a sensitivity study to examine the impact of a 50%:50% split between the amount of SOA mass partitioned according to the particle surface area and according to the pre-existing organic mass (i.e., only allowing half of the SOA to condense kinetically). Our results suggest non-linear behaviour in terms of the fraction of SOA distributed kinetically and the impact on cloud droplet number. Future work from our group will examine in more detail the effect on particle growth rates of distributing a more realistic amount (between 0% (i.e. our mass-based approach) and 50% (our split approach)) of the SOA kinetically, based on new laboratory studies on the production of extremely low volatility organic oxidation products. We already discuss this uncertainty in the conclusions of our paper.

To isolate the impact of the two different approaches to partitioning, we calculate the amount of SOA condensing in the same way under each approach (i.e., determined by the available condensation sink); accordingly the amount of S_{org} present under the two different approaches will vary only as the size distribution changes. We now clarify this in our paper (at p4152, line 8). Because the amount of organic condensation is identical in our simulations, we don't feel that a figure of S_{org} concentrations would be of benefit to the paper.

2. Another concern is related to the uncertainties of this study associated with several other simplifications and assumptions: (1) Offline chemistry ("prescribe six-hourly mean offline oxidant"); (2) No nitrate and ammonium; (3) "secondary organic material is generated at a fixed molar yield (13 %) from the oxidation of monoterpenes". The formation of atmospheric particles and their growth to CCN sizes depend on the concentrations of precursors involved. The authors showed that the method to deal with organics lead to large difference in CCN and AIE. The above

simplifications and assumptions will surely impact CCN and AIE. The authors need to justify the usage of these simplifications in this study and discuss how these may affect the conclusions of this work.

Our objective was to understand the impact of partitioning of biogenic organics on aerosol and global radiative effects. We therefore chose to isolate this effect whilst keeping other variables constant (e.g., offline chemistry, fixed SOA yields). We now highlight these simplifications and add discussion as suggested by the referee. We have added the following (at p4153, line 3):

“To isolate the impact of the condensation of secondary organic material the SOA yield, the oxidant chemistry and the SOA condensation rate was identical in all simulations.”

Our treatment of SOA is similar to many other global aerosol models. We have added the following at p4148, line 11:

“The treatment of SOA in global aerosol models is summarised by Tsigaridis et al. (2014).”

To address the specific points:

1) We accept that the use of offline chemistry is a simplification and in the future we intend to examine the impact of coupled chemistry on our treatment of SOA. The lack of online chemistry here means that we are missing the feedback on oxidant concentrations due to BVOC emission, however, we are looking at the difference between two ways of distributing the SOA, once formed via BVOC oxidation, so we do not expect this aspect of our results to be very sensitive to our use of offline oxidants.

We have added the following at line 9, p4150:

“The use of offline oxidants means that we do not simulate the feedback on oxidant concentrations due to monoterpene emission.”

2) The version of GLOMAP-mode used here does not include ammonium or nitrate aerosol; a version of GLOMAP-mode containing ammonium and nitrate is under development but was not available at the start of this work. In the absence of nitrate and ammonium, our total mass concentrations are likely to be biased low – however, the focus of this study was to compare the two methods of distributing secondary organics across the size distribution.

We have added the following to our model description at line 2, p4150 to clarify that we do not include nitrate or ammonium:

"Material in the particle-phase is classified into four components: sulphate (SU), black carbon (BC), particulate organic matter (POM) and sea-salt (SS); this version of GLOMAP-mode does not include nitrate or ammonium aerosol."

3) Generating secondary organics from the oxidation of BVOCs, using a fixed molar yield, is a simple approach based on empirical relationships between the amount of organic material found to reside in the aerosol phase and the level of BVOC emission an air parcel has been exposed to (e.g., Tunved et al., 2004; Tunved et al., 2006). In reality these yields would be sensitive to the specific precursors and oxidants involved, but are poorly constrained.

Whilst the simulated amount of SOA formed could be different with different/variable yields, we were interested in the relative difference between two approaches that distribute the same amount of SOA across the size distribution.

3. GLOMAP-mode was employed for this study. The mode approach uses several longnormal to describe particle size distributions. Since this work focuses on the effect of particle growth associated with SOA on particle size distributions and CCN concentrations, I was wondering why the authors didn't use their more accurate GLOMAP-bin aerosol model for the work.

As with many aerosol-climate models, it is the modal version of GLOMAP that is to be included within HadGEM-ES (the UK's Earth System Model) so, in terms of global radiative effects, we wanted to explore this process within the modal scheme. In previous work we have demonstrated that the modal model is able to reproduce much of the behaviour of the sectional (GLOMAP-bin) model. We have added the following after line 10, p4150.

"In previous work (Mann et al., 2012) it has been demonstrated that the modal version of the model we use here is able to accurately simulate the aerosol size distribution simulated by a sectional version of the model (Spracklen et al., 2005)."

However, we agree that there is a need to explore these processes using global models with a spectrum of complexity; as the reviewer points out the process of particle growth is a particularly important process to examine using our more detailed sectional scheme and we will be addressing this in future work.

4. Page 4150, Equation (2). How [NucOrg] was calculated in the model? Is NucOrg the same as Sorg (Eq. 4)? You only mentioned that NucOrg represents monoterpene oxidation products but monoterpene oxidation should have many

products of different volatilities. Some details on the calculation of [NucOrg] and global distribution of [NucOrg] should be given.

Yes NucOrg is the same as S_{org}. We have corrected Equation 2 to clarify this. This was the approach taken when the Metzger scheme was originally implemented in GLOMAP (Metzger et al. 2010) and in Scott et al. 2014. However, we agree that monoterpene oxidation will produce products of differing volatilities and that only a fraction of these will be able to participate in the earliest stages of new particle formation. We have added a sensitivity study exploring the impact of a 50:50 split – see earlier response. Future work from our group will assess the impact of allowing differing amounts of the secondary organics (or those produced from particular pre-cursors via particular reactions) to participate in new particle formation.

5. Page 4150, Equation (2). Riccobono et al. (Science, 2014) gave a more updated formula for the organically mediated nucleation. Why not use the formula given by Riccobono et al.? What is the uncertainty associated with the usage of specific nucleation parameterizations? A figure showing the global distributions of nucleation rates predicted by ACT and ORG schemes should be provided.

The start of this work predated the Riccobono publication so we have used the organically mediated mechanism given by Metzger et al. 2010. Our intention here is to compare the two different treatments of SOA partitioning and we use the Metzger scheme as an example of an organically mediated mechanism. To demonstrate the sensitivity to nucleation parameterization we also completed simulations using the activation nucleation mechanism. Both the activation and organically mediated nucleation parameterizations showed consistent behaviour; we would not expect the Riccobono mechanism to differ greatly. We highlight the consistent response, in terms of CDNC change, between the activation and Metzger mechanisms in Section 3.1. The nucleation rates themselves are not the focus of this study; therefore we do not feel that a figure of global nucleation rates is necessarily of relevance to this paper.

6. Figure 1 shows the simulated and observed particle size distributions at a boreal forest site. The conclusion of this work is for global. Therefore, it is necessary to compare simulated CN and CCN number concentrations with those observed at different sites around the globe.

Our intention here was not to evaluate the model extensively (which we have done in other studies) but to compare these two different treatments using an example observed size distribution to put the modelled results into context. We have added a sentence on previous model evaluation after line 10, p4150.

"We have previously evaluated GLOMAP against observations of CN (Spracklen et al., 2010) and CCN (Spracklen et al., 2011)."

We have now added a comparison against measurements collected at a pristine Amazon rainforest location (Manaus, Brazil) to Figure 1 and related discussion to Section 3.

Minor comments:

1. Pages 4147-4148. As I understand, both kinetic approach (for low volatile organics) and equilibrium (for semi-volatile organics) were used in in the work of Yu (2011) cited here. This should be reflected and correctly described in the review of previous work in the Introduction.

We already include the following description at line 18 p4148:

"Yu (2011) found that allowing successive stages of oxidation to occur, and the generation of non-volatile products, increased simulated surface level CCN concentrations by 5-50%, over a version of the same model in which the thermodynamic assumption was applied."

So we have expanded this slightly to clarify that this involved using both a kinetic and thermodynamic approach:

"*Yu (2011) found that allowing successive stages of oxidation to occur, and the generation of non-volatile products which were distributed according to the kinetic approach, increased simulated surface level CCN concentrations by 5-50% over a version of the same model in which only an equilibrium approach was taken*."

2. Page 4149, line 12. Offline radiative transfer model was used. Please add some details on what aerosol (and gas) fields at what temporal resolutions were saved for the offline RT calculation.

We have added extra details to Section 2.2 (p4193, line 9 onwards) to clarify that our radiative transfer calculations are based on monthly mean aerosol fields and provide other further details. This section now reads:

"The changes to CDNC due to the inclusion of biogenic SOA (i.e., *KinSOA* - *NoSOA*) are calculated from the monthly mean aerosol distribution, using the parameterisation developed by Nenes and Seinfeld (2003), Fountoukis and Nenes

(2005) and Barahona et al. (2010); assuming a globally uniform updraught velocity of 0.2 m s^{-1}.

The first AIE due to biogenic SOA is then determined using the offline radiative transfer model of Edwards and Slingo (1996) with nine bands in the longwave and six bands in the shortwave; a monthly mean climatology based on ECMWF reanalysis data and cloud fields for the year 2000 from the ISCCP-D2 archive (Rossow and Schiffer, 1999) are used. To calculate the first AIE, a uniform control cloud droplet effective radius (r_{e1}) of 10 µm is assumed to maintain consistency with the ISCCP derivation of liquid water path, and for each perturbation experiment the effective radius (r_{e2}) is calculated as in Eqn. 11, from monthly mean cloud droplet number fields $CDNC_1$ and $CDNC_2$ respectively (where $CDNC_1$ represents the simulation including SOA, and $CDNC_2$ represents the simulation with no SOA).

The first AIE of biogenic SOA is then calculated by comparing net (SW + LW) radiative fluxes using the varying r_{e2} values derived for each perturbation experiment, to those of the control simulation with fixed r_{e1}. In these offline experiments, we do not calculate the second aerosol indirect (cloud lifetime) effect.

To determine the DRE, following the methodology described in Rap et al. (2013), the radiative transfer model is used to calculate the difference in net top-of-atmosphere all-sky radiative flux between experiments including SOA and the equivalent experiments without SOA. Aerosol optical properties are calculated from the monthly mean aerosol distribution. The refractive index for each mode is calculated as the volume-weighted mean refractive index of the components present (values for which are given at 550 nm in Table A1 of Bellouin et al., 2011), including water for the soluble modes. The optical properties (scattering and absorption coefficients, together with the dimensionless asymmetry parameter) are then obtained from look-up tables of all realistic combinations of refractive index and Mie parameter, as described in Bellouin et al. (2013).”

Reviewer 2:

The authors have investigated the effects of thermodynamic equilibrium and kinetic condensation treatments for partitioning biogenic SOA on the magnitude of the direct and first indirect effects. The effects of two new particle formation schemes – one based on activation rate of H2SO4 clusters and the other based on organically mediated cluster formation rate – are also investigated. While the topic of this study is within the scope of ACP, I am unable to recommend it for final publication in the current form due to several issues listed below.

Comments:

1. I had some difficulty in assessing what major insights this study provides that were not already provided by Riipinen et al., ACP, 2011 – that kinetic condensation leads to an increase in CCN while thermodynamic partitioning suppresses it. While this study computes CDNC from CCN and the radiative effects, I am not convinced that is enough to qualify it as a unique scientific contribution, especially when the thermodynamic approach used here seems somewhat flawed (see next comment). Comparing the effects of two new particle formation schemes is new, but that does not seem to be the main focus of the study.

We respectfully disagree with the reviewer here as we have made the first assessment on the global direct and indirect radiative effects. We demonstrate that the treatment of condensation is an important uncertainty in the calculation of global indirect effects, and this is the first time this process has been examined using the GLOMAP model. We feel that this is a sufficient contribution to justify our manuscript. This is highlighted in the Introduction and we now also highlight this novelty in the Conclusions.

2. The thermodynamic approach used in this study is not the one typically used in a thermodynamic partitioning model. It simply partitions the SOA mass (predicted by the kinetic approach) according to organic mass (MOA), and does not let it evaporate, undergo further oxidation in the gas phase, and repartition in subsequent time steps. While I understand the rationale behind this approach, it would be incorrect to call it thermodynamic approach.

We agree and although we had originally used this terminology to maintain consistency with Riipinen et al., 2011 and D'Andrea et al., 2013, we have changed the terminology throughout the manuscript to refer to this as the “mass-based” partitioning approach instead.

3. Comparison of observed size distribution with model predictions is shown for only one site. More comparisons at different locations are necessary as this is a global study.

Our intention here was not to evaluate the model extensively but to compare these two different treatments using an example observed size distribution to put the modelled results into context. In previous work we have made extensive evaluations of the model – we now detail this after line 10, p4150.

We have now added a comparison against measurements collected at a pristine Amazon rainforest location (Manaus, Brazil) and related discussion to Section 3. These two locations are chosen as they are relatively far from pollution sources and biogenic SOA is likely a dominant component of fine aerosol mass.

4. This study focuses biogenic SOA, but uses only monoterpenes as precursor VOCs. What about isoprene, which is also an important biogenic VOC that can produce substantial amounts of SOA in certain regions of the globe? Other sources such as anthropogenic VOCs and biomass burning VOCs can be important as well. Ignoring these key SOA precursors would lead to underprediction of global SOA budget and skew the magnitudes of direct and indirect effects, and hence the main results of this study

In this study we wanted to investigate the relative impact of two different approaches to partitioning for a given amount of SOA, which is formed here from a precursor with the spatial/temporal emission pattern of monoterpenes. But we agree with the reviewer that this approach will likely lead to an underestimate in the amount of SOA produced in our simulations and we accept that the amount of SOA included will affect the magnitude of the DRE and AIE we calculate.

In Scott et al., 2014, we showed that the global annual mean AIE (simulated using an activation nucleation mechanism) was very similar for simulations containing monoterpenes only (-0.07 W m^{-2}), isoprene only (-0.06 W m^{-2}) and both monoterpenes and isoprene (-0.06 W m^{-2}). However, we showed that increasing the amount of SOA further (by multiplying the yields at which the SOA is generated by a factor of 2 or 5) reduced the magnitude of the AIE (because the growth of smaller particles to CCN size was suppressed due to the enhanced condensation sink), but also reduced our model’s ability to capture observed N_{80} and CCN concentrations. This demonstrates that the magnitude of the AIE does not scale linearly with SOA formation.

Accordingly we have added the following to our model description at line 9 of p4150:

"*The inclusion of monoterpene emissions only will likely lead to an underestimate in the amount of SOA produced in our model simulations.*"

5. Page 4149, line 25: It is stated that 5 log-normal size modes are used in the model, implying that the model uses some sort of modal dynamics approach to simulate aerosol size distribution. This could be problematic if the mode widths are assumed to remain fixed after kinetic condensation, which produces a characteristic narrowing the mode. Please provide some details on the modal dynamics approach used and how this issue is handled in the model?

GLOMAP-mode uses a two-moment modal scheme, within which mode number and mass concentrations may vary but mode width is held fixed. We agree with the reviewer that the modal scheme may not fully capture the evolution of the aerosol size distribution during condensation.

Following condensation, the particle number concentration for a mode is held constant and the new geometric mean diameter for the mode is calculated according to the new particle volume (due to added mass). The standard deviation of the mode is fixed so the mode width would not change to reflect any change to the distribution of material within a mode.

In previous work we have demonstrated that our 2 moment modal model closely matches results from a sectional version of the model (see response to Reviewer #1). Our group also will be using the more detailed, sectional version of our model to examine particle growth due to the condensation of secondary organics.

To clarify this issue we have added the following before line 1, p4153:

"GLOMAP-mode uses a two-moment modal approach, within which mode number and mass concentrations vary but the mode width is held fixed. Following condensation, the particle number concentration for a mode is held constant and the new geometric mean diameter for the mode is calculated according to the updated volume per particle (Mann et al., 2010). Since the mode width is held fixed, this approach will not capture potential changes to the distribution of material within a mode following condensation."

6. Page 4150, line 4: It is mentioned that six-hourly mean offline oxidant concentrations are used to oxidise monoterpenes to form SOA at 13% molar yield. What time splitting interval is used to perform coagulation and condensation calculations? Also, in which order are these calculations done? And are the results sensitive to the order?

A flowchart of the microphysical operations performed within GLOMAP is given in Spracklen et al., 2005. Within each chemistry or microphysical time step (15 mins), there are 5 competition sub-steps, during which nucleation and condensation compete for the available sulphuric acid (and organic oxidation products when an organic nucleation mechanism is used). Condensation is performed first (then nucleation), then coagulation. Spracklen et al. (2005) found that shortening the length of the time-step, and changing the order of operations altered particle concentrations by less than 5%.

We have added the following to our model description, at line 4, p4150:

"Operator splitting is used to calculate the different microphysical processes: condensation is calculated followed by nucleation then coagulation with a time step of 3 minutes. Spracklen et al. (2005) found that shortening the length of the time-step, and changing the order of operations altered particle concentrations by less than 5%."

7. Editorial comment: Abstract, line 17: Change "medicated" to "mediated".

Thank you for pointing this out, we have now corrected this.

Additional changes:

The results presented initially for the ORG simulation were generated using a rate coefficient (*k* in Eq. 2) of 5 x 10^{-12} s^{-1} (not 5 x 10^{-13} s^{-1} as given on line 22 of p4150). We have updated our results throughout to reflect the lower rate coefficient of 5 x 10^{-13} s^{-1} specified in Metzger et al. (2010), but include the original results in Table 2 as a sensitivity study representing a faster nucleation rate. The effect of applying a mass-based approach, rather than a kinetic approach, on the global annual mean AIE is the same using either value for *k* (i.e., a 24% reduction).

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