

We would like to thank both reviewers for their positive and helpful comments on our manuscript. Below, we have addressed the specific points raised:

Reviewer 1:

This manuscript presents a very well-performed analysis on the radiative effects of aerosols resulting from biogenic organic precursor emissions and associated uncertainties. I recommend accepting this paper for publication in *Atmospheric Chemistry and Physics* after the authors have addressed the following, relatively minor, issues.

Section 2.1.1, first paragraph. Is there a specific reason for selection the yields 13% and 3% as base case values?

There is considerable uncertainty associated with the magnitude of BVOC emission and the amount of SOA produced from their oxidation. In our study, the yield of SOA production is varied in order to represent the uncertainty in both parameters (by altering the total amount of SOA being produced).

We have modified Section 2.1.1, p16966, line 6 onwards, to include further details on the selection of these base case values:

“Monthly emissions of monoterpenes and isoprene are taken from the Global Emissions Initiative (GEIA) database (Guenther et al., 1995), giving total emissions of 127 and 503 Tg(C) a⁻¹ respectively. Considerable variability is observed in the yield of SOA generated by BVOC oxidation (e.g. Yu et al., 1999; Kroll et al., 2006; Shilling et al., 2009). Here we generate SOA at fixed molar yields of 13% and 3% for the oxidation of monoterpenes and isoprene respectively; monoterpenes are prescribed the reaction characteristics of α -pinene and oxidation reactions proceed at the rates given in Table 1. The use of a 13% molar yield for the production of monoterpene SOA follows the approach of Spracklen et al. (2006), which was based on Tunved et al. (2004). Tunved et al. (2006) revised this estimate downwards to an apparent mass yield of between 5 and 10%, suggesting that the atmospheric yield may be lower than the 13% molar yield (equivalent to a 14.3% mass yield) used in GLOMAP. The 3% molar yield (equivalent to a 3.3% mass yield) for the production of isoprene SOA is based on the yields obtained by Kroll et al. (2005, 2006) over a range of experimental conditions (0.9 – 5.5% mass yield).

The global production of biogenic SOA is poorly constrained, with estimates ranging between 12 Tg(SOA) a⁻¹ and 1820 Tg(SOA) a⁻¹ (Griffin et al., 1999; Kanakidou et al., 2005; Goldstein and Galbally, 2007; Heald et al., 2010; Heald et al., 2011; Spracklen et al., 2011b). Using molar yields of 13% and 3% (for monoterpenes and isoprene respectively) produces approximately 37 Tg(SOA) a⁻¹ in our model, which is towards the lower end of current estimates. Consequently, it may be the case that the yields applied here are too low (i.e. do not produce enough SOA), or too high (Tunved et al., 2006). We explore this uncertainty by varying the SOA production yield in a series of sensitivity experiments.”

Section 2.1.2. The authors could add which year the primary emissions are supposed to represent.

Thanks, we've added further details to Section 2.1.2, p16967, lines 15-18:

“Annual mean emissions of BC and POM from fossil and bio-fuel combustion are based on energy data from 1996 and taken from Bond et al. (2004); monthly wildfire emissions for the year 2000 are from the Global Fire Emissions Database (GFED) inventory (van der Werf et al., 2004).”

Section 2.1.3, first paragraph. Note that also BHN rate is calculated from a parameterization. The main differences between the four parameterizations are the assumed nucleating species and data source (theory, lab experiments, field measurements) from which the parameterization has been derived. A slight modification could be made to avoid confusion by the reader.

Thanks, we have modified the text in Section 2.1.3, p16967, line 22 onwards to read:

“Since new particle formation has been shown to strongly affect CCN concentrations (Spracklen et al., 2008b) and simulated aerosol indirect radiative effects (Bellouin et al., 2013), we quantify the impact of BVOC emissions using four nucleation mechanisms. In all experiments we include a parameterisation to represent the binary homogeneous nucleation (BHN) of sulphuric acid and water (Kulmala et al., 1998a) which occurs mainly in the free troposphere (e.g. Spracklen et al., 2005a). New particle formation rates in the boundary layer cannot be adequately explained by BHN alone so we examine the impact of BVOC emissions when three additional parameterisations for new particle formation are applied in the boundary layer, in combination with BHN.”

Page 16974, lines 4-6. This statement is unclear. Practically all the regions are coincident with some sort of primary emissions, so it is rather question of the magnitude of primary emissions. Please modify the sentence a bit.

We agree and have modified the text in Section 3.1, p16974, lines 4-6 to read:

“The largest increases in absolute CCN concentration are simulated in regions experiencing high levels of primary particulate emissions, particularly tropical regions affected by biomass burning. This suggests an important interaction between SOA and the presence of primary particles.”

Page 16977, lines 3-5. The statement "When monoterpene oxidation products are allowed to participate directly in nucleation, the contribution of biogenic SOA to CCN concentrations is substantially greater " needs to be clarified a bit further. The survival of particles nucleated at diameter d^* up to the diameter 3 nm is an exponential function (equation 4), and therefore the survival probability is sensitive to the particle growth rate, condensation sink and, importantly, to d^* as well. The authors should investigate whether larger enhancements in CCN concentrations in case of organic nucleation are due to higher nucleation rate in those cases or, rather, due to larger assumed values of d^* . In practice, assuming larger value of d^* for an organic nucleation mechanism is equal to assuming that organics participate in both nucleation and initial steps of nuclei growth, so one might also consider stating "When monoterpene oxidation products are allowed to participate in the very early steps of new particle formation, the contribution".

We agree, this point could benefit from clarification; we have modified the following to Section 3.2, p16977, lines 3 onwards to read:

“When monoterpene oxidation products are allowed to participate in the very early stages of new particle formation, the contribution of biogenic SOA to CCN concentrations is substantially greater. The large increase in CCN concentrations when the Org1 mechanism is used can be attributed to the fact that in the absence of BVOC emissions new particle formation in the boundary layer does not occur. In a test simulation, the value for d^* in the Org1 experiments was reduced from 1.5 nm to 0.8 nm (to match ACT but not intended to represent a real scenario). This reduced the percentage increase in global annual mean CCN concentration (upon inclusion of biogenic SOA) from 45% to 36%, indicating that most of the enhancement in CCN concentration comes from the inclusion of organics in the nucleation rate itself, but some is due to the increased survival probability of newly formed particles associated with assuming a larger value for d^* .”

Page 16982, lines 23-29. Besides the two papers mentioned here, also comparison to the few other measurement-based estimates of the AIE could be added here (see Lihavainen et al. 2009 in the reference list, and references therein). Those measurement-based studies could also be added to introduction (page 16964, lines 25-27). The paper by Lihavainen et al. predicts much lower direct radiative effect compared with AIE over boreal forests, which seems to be in line with the results in figure 7. The authors should discuss this issue as well in this paragraph.

Thanks for spotting this – it was an oversight to only include the Lihavainen et al., (2009) values for the DRE and not the AIE in the introduction, we have added this paper to Section 1, p16964, line 21 onwards:

“Several studies suggest a large first AIE from biogenic SOA over the boreal forests at high northern latitudes. Using a global aerosol microphysics model, Spracklen et al. (2008) simulated a doubling of regional summertime CCN concentrations as a result of monoterpene emissions, and a subsequent regional first AIE of between -1.8 and -6.7 W m^{-2} of boreal forest. Using measurements made at Pallas in Finland, Lihavainen et al. (2009) estimated a local summertime first AIE of between -3.2 and -6.4 W m^{-2} over the boreal forest region. A stronger annual first AIE (locally between -5 and -14 W m^{-2}) was calculated by Kurten et al. (2003) using measurements taken at a Hyytiälä in southern Finland.”

We have also added the Lihavainen et al. (2009) reference to our discussion in Section 6, p16982, line 23 onwards:

“Over boreal forests, regional annual first AIEs of between -0.1 W m^{-2} and -0.5 W m^{-2} are calculated with the ACT mechanism (Figure 7 (right)) and between -0.1 W m^{-2} and -1.5 W m^{-2} with the Org1 nucleation mechanism. As illustrated in Figure 9, much of the boreal region experiences a summertime (JJA) mean first AIE of between -1 W m^{-2} and -5 W m^{-2} , when the Org1 mechanism is used, matching the large cooling effect over these forest regions calculated by previous studies (Kurten et al., 2003; Spracklen et al., 2008; Lihavainen et al., 2009). However, the strongest regional radiative effect (up to -9 W m^{-2}) is simulated over the North Atlantic ocean ($50 - 60^\circ\text{N}$) which, whilst experiencing a smaller CDNC increase than the land regions at the same latitude, has higher cloud coverage in the ISCCP dataset (i.e. cloud fraction of 50-70% as compared to 0-30% over the land). In agreement with Lihavainen et al. (2009) we find that over the boreal region, the simulated first AIE is substantially greater than the DRE (Figure 7).”

Page 16983, line 1. Are the authors referring to oceanic regions surrounding boreal forest? Please be more specific here:

Yes, we agree that this is vague. The text in Section 6, p16982, line 29 onwards has been modified and now reads:

“However, the strongest regional radiative effect (up to -9 W m^{-2}) is simulated over the North Atlantic ocean ($50 - 60^\circ\text{N}$) which, whilst experiencing a smaller CDNC increase than the land regions at the same latitude, has higher cloud coverage in the ISCCP dataset (i.e. cloud fraction of 50-70% as compared to 0-30% over the land).”

Section 6. It would be interesting to see a brief discussion on how the relative importance of DRE and AIE due to biogenic SOA varies spatially. Based on Figure 7, there appears to be large differences between different world regions, yet purely visual inspection of the figure provides only qualitative information on this issue.

Thanks, that's a good point; we have added the following text to the end of Section 6, p16983, following line 16:

“There is a large difference between the spatial pattern of the DRE and first AIE due to biogenic SOA. This is mainly driven by the distribution of clouds since a high cloud fraction will suppress any potential DRE from changes to particle number and size below, but will enhance any potential AIE from changes to changes to particle number and size. Consequently the DRE tends to be stronger over the relatively less cloudy (based on the ISCCP clouds for the year 2000) land regions and the first AIE tends to be stronger over the relatively more cloudy ocean regions. For example, in the ACT simulation shown in Figure 7, the DRE is more than a factor of 5 greater than the first AIE over central Africa and South America, whereas the first AIE is more than a factor of 5 greater than the DRE over high latitude northern and southern hemisphere ocean regions.”

Section 8, second paragraph. Referring to my previous comment, the authors should check out whether larger enhancements in CCN in case of organic nucleation are really due to larger nucleation rates, or whether larger assumed values of d^* are the primary reason

[See response to previous comment.](#)

Reviewer 2:

The manuscript presented by Scott et al. provides an extensive analysis of the microphysical processes involved with CCN production from biogenic precursor emissions and the resulting radiative effects. The thoroughness of the paper combined with a concise and compelling writing style made the manuscript easy to follow. The results presented in the manuscript are highly relevant for both the modeling and understanding of the effects of biogenic SOA formation and the impact of SOA on global and regional climate. Scientifically, I find nothing wrong with the approach taken and therefore I suggest this manuscript for publication upon addressing some (very) minor revisions explained in more detail below.

Section 2.1.1. It is mentioned that there is substantial uncertainty in the yield of SOA generated by BVOC oxidation, but there is no discussion on why the specific molar yields (13% and 3%) were chosen for monoterpenes and isoprene.

[See response to Reviewer 1 on this point.](#)

Section 2.1.3. There is no mention of some form of ternary homogeneous nucleation as a possible nucleation mechanism. Possibly include a brief explanation as to why BHN was used as the base mechanism and not a more recent mechanism (or no nucleation whatsoever).

[BHN was used as the base case mechanism to maintain consistency with previous studies using the GLOMAP model \(e.g. Spracklen et al., 2011a; Schmidt et al., 2012\). As the focus of this paper was the role of biogenic SOA, ternary nucleation involving ammonia/amines was not explored here but as shown recently by Almeida et al., \(2013\) this may be an important process in the boundary layer. We have added the following text to Section 2.1.3, p16969, before line 6:](#)

[“As the focus of this study is to investigate the role of biogenic SOA, we do not consider ternary homogeneous nucleation, but as has been shown recently by Almeida et al. \(2013\), ternary nucleation involving amines may be an important process in the boundary layer.”](#)

Section 3.1, paragraph 4. The authors could add a brief explanation of the decrease in CCN seen over eastern Asia in Figure 1.

[The decrease in CCN simulated over eastern Asia \(using the ACT mechanism\) is due to the suppression of new particle formation \(via activation of H₂SO₄ clusters\) in the boundary layer due to the enhanced condensation sink. This region has high SO₂ emissions \(in the emission dataset used here\); in the absence of SOA the available H₂SO₄ is sufficient to form new particles and grow them to CCN size. When biogenic SOA is included, the impact of the enhanced condensation sink outweighs the contribution of SOA to the growth of newly formed particles and a decrease in CCN concentration is simulated. The same decrease is not observed in the BHN only experiment as there is little new particle formation occurring in the boundary layer \(and therefore little to be suppressed\). We have inserted the following text to Section 3.1, p16975 at line 24:](#)

[“Additionally, in regions with particularly high SO₂ emission \(e.g. eastern Asia; Figure 1\), and therefore high H₂SO₄ concentrations, the enhanced condensation sink may also suppress new particle formation and growth as a route for CCN generation in the boundary layer, reducing CCN concentrations in the presence of biogenic SOA.”](#)

Section 3.1, paragraph 5. The suppression of nucleation over ocean regions is part of the reason there is a decrease in CCN downwind of continental regions, however it is worth mentioning that wet deposition will efficiently remove CCN as well (with fewer small particles to grow to CCN sizes due to nucleation suppression earlier over the continents).

That is correct, we mention here the process of nucleation scavenging but have rephrased the text at Section 3.1, p16976, line 1 to clarify that this leads to wet deposition:

“Therefore in the presence of SOA, particles grow more quickly to a size where they may be removed from the atmosphere by wet deposition”

Section 3.1, paragraph 6. The authors could provide more details explaining why the CCN response saturates to SOA production yield.

True, we were quite brief here. Beyond a certain point (in terms of increasing yield), which will vary spatially and temporally, the generation of “new” CCN (via particle growth) becomes limited by the enhanced condensation sink and suppression of new particle formation. Although it is worth noting that the increased yield experiments are conducted in the absence of organically-mediated new particle formation, which would alter this response. We have added briefly to Section 3.1, paragraph 6, p16976, line 16 onwards:

“This is confirmed by the variation in CCN change observed for Experiments 4 to 7, where a factor of 5 increase in the SOA production yield results in less than a doubling of absolute and fractional CCN changes. This occurs because the generation of additional CCN (via condensation of secondary organic material) becomes limited by the enhanced condensation sink (due to larger particle sizes) and consequent suppression of new particle formation.”

Section 8. As a general comment, the authors could briefly discuss how the results of the numerous simulations performed could provide some guidance for future model development regarding biogenic SOA, especially for large-scale global modeling where the balance between computational costs and accuracy must be considered.

We've added the following text to Section 8, p16988, after line 12:

“We have shown here that the role of organic oxidation products in new particle formation is important in determining, particularly the indirect, radiative effect of biogenic SOA. However, our understanding of which secondary organic species contribute to each stage of new particle formation and growth at different particle sizes is far from complete (e.g. Kulmala et al., 2013). In the absence of a detailed understanding, and bearing in mind computational costs, future model development regarding biogenic SOA could proceed on the basis that at least some (as yet undetermined) fraction of the secondary organic oxidation products contribute to new particle formation.

Our simulations also highlight the important interaction between secondary organics and primary particles from combustion, particularly from fires in the tropics. This suggests that the (simulated) ageing of insoluble particles should be determined by the availability of condensable material, rather than prescribed (i.e. transferring insoluble particles to the soluble distribution after a fixed period of time), in order to capture the sensitivity of CCN concentration to changing levels of secondary organics.”

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