

We would like to thank both reviewers for their overall encouraging statements about our manuscript, and for their constructive suggestions on how to improve it. In the following, we detail how we have addressed their comments in the manuscript revision.

The revised manuscript has track changes enabled which should make it easier for the reviewers and editor to see where revisions have been made.

Reviewer one:

The paper is generally very difficult to read, and I would suggest the authors to make substantial editing to the manuscript. As CO₂ increases and SOA increases are quite different subjects, it is understandable that the paper covers a lot of different aspects. But this also requires the authors to pay attention to the organization of the paper (see some of the specific comments).

The modeling part and the observation part should be better integrated. Although the authors argued that the paper provided an example to study different process in a consistent observational and modeling framework, I feel the paper needs further work to better integrate observations and modeling results (see also some of the specific comments).

We have separated in the results section the observation and modelling aspects of the papers because of the different time scales covered (present-day, one growing season, vs. century perspective of the modelling), and because of the differences in geographic domain (forest vs. Siberia as a whole). This separation we would like to keep. Still, we hope that in response to the reviewer's points below (as well as in response to reviewer #2) the revised text has now a better "flow".

Section 2: The descriptions and discussions in this section are generally too long. I would suggest to move some of these discussions into Section 3 &4 and combine them with discussions in these two sections.

Re-reading the methods section, the reviewer is perfectly correct in that some of the text would fit better to the results section. Accordingly, we moved part of the text and adopted it to fit into Section 3 and Section 4; for instance, lines 280-86, 410-37. We also shortened parts of the remaining text.

Line 158: the model is set up to test CO₂-inhibition hypothesis. It is not clear to me how this is achieved.

We have revised the sentence to make clearer to the reader how this is done (lines 157-163).

Line 247: where does 1.9 come from? Also, from Section 3.1, from line 274 to line 283, the range of E from 1.9 to 9.6 is also discussed. I feel it is better to move some of the discussion in Section 3.1 to Section 2.

We have moved most of the text on E*, and related text on how it is implemented in the DGVM (lines 410-427) to the discussion section, as suggested by the reviewer (we leave some in the results section as it is necessary when showing the range of simulated total BVOC emissions for Siberia/Table 1). In the methods/aerosol modelling, we refer the reader to the results section (lines 250-252); the reviewer is correct that the value without this reference is confusing.

Line 252, CCN at 1%. 1% seems too large.

We have decided to focus discussion on CCN at 1.0% supersaturation, since it reflects the changes in Aitken mode concentrations and local changes in precursor emissions (added to lines 256-58). We have now included an additional figure (A3) in appendix to indicate changes in CCN(0.2%). (see also response to a query by reviewer no. 2, corresponding to CCN supersaturation)

Line 271: For the "larch" plant functional type, an emission potential of E=2.4. But it in 247, E=1.9?

The value of 2.4 is an "old" value in the paper by Guenther et al., who at that time could not draw on any measurements on larch. We have moved this aspect to the discussion section and hope this has become less confusing (lines 421-27). For the aerosol modelling we had adopted the conservative, low end of measured E* (lines 495-96).

Table 1: description texts: the range of E from 1.9 to 9.6, and then a weighted one 6.4. Which one is used?

(Now Table 2) This is described in the table caption (slightly revised, for more clarity), and in the subscripts to the values given in the table (Total_{MT_{BNS}}): Simulations for monoterpene emissions for the boreal needleleaf summergreen (BNS) plant functional type compared three cases (indicated as subscripts for Total_{MT_{BNS}}): using maximum (9.6 $\mu\text{g}_C \text{ g}^{-1} \text{ h}^{-1}$) and minimum (1.9 $\mu\text{g}_C \text{ g}^{-1} \text{ h}^{-1}$) values for E^* measured in Spasskaya Pad (see text), and $E^* = 6.2 \mu\text{g}_C \text{ g}^{-1} \text{ h}^{-1}$ as a weighted average from all observations at the Spasskaya Pad location.

Section 3: Observations and modeling results are not well integrated. For example, why not compare simulated aerosol concentrations with observed present aerosol concentrations? ...I would like to see how present day BVOC and aerosols are simulated compared with observations.

We have added a comparison of present-day daily BVOC model results for the Spasskaya Pad location against observations, using site-specific data, and some examples from literature (lines 293-301), and also included a data-model comparison in lines 464-473.

Section 3.3: Is CO-inhibition implemented in the sensitivity tests? This is not clear to me from reading Section 3.3. But line 158 seems to suggest this was tested in the paper.

We added clarifying statements to the revised section 3.3 (lines 360, 376)

Section 4: How is CO₂ forcing calculated?

The method of calculating CO-radiative forcing follows the widely applied, standard methodology used eg. in each of the IPCC reports; for simplicity we assume an ocean uptake of 50% such as done in Stich et al. (see references given in the methods, lines 211-12)

Line 399: the sensitivity of CCN to E. This is not clear to me.

We agree that this is confusing to the reader, and is indeed not really visible in the Figures we have chosen to show. We removed the sentence.

Lines 424-434: This paragraph needs further editing. For example, it is not clear why the authors want to discuss changes in wildfire emissions and anthropogenic SO₂ emissions. Later, it seems that the effects of BVOC are only assessed by assuming wildfire and anthropogenic CO₂ unchanged. Many more paragraphs have similar issues. This is why the paper is really hard to read.

The challenge of understanding changes in aerosol load arise from competing factors that can either lead to enhancing or lowering aerosol burden. The chief reason why we need to add the discussion on wildfire and SO₂ arise from the fact that the background load and size distribution are important for whether or not the enhanced BVOC lead to enhanced SOA and CCN production or not. Wildfires, in particular, are an important feature of boreal forests. However, the reviewer is correct that the paragraph had been confusing – in fact, we had at some stage in the analysis attempted to look at changing wildfire emissions but then realized that this would make the manuscript very complex (w.r.t. to the SOA analysis) and distract from the main story of the paper. We have revised the paragraph and hope that this has now become easier to understand. Overall, we believe that (in response to the reviewer's suggestions) the revised manuscript has become easier to read (lines 505 onwards).

Technical corrections:

Line 29: miss “.” Between “effect” and “Combining”.

Thanks, well spotted.

Line 32-35: The sentence of “On the global level, . . .” needs some further work.

The sentence has been revised.

Line 97: Make sure the citation format is correct.

We couldn't find an error with the format, but will check again in the type-set version.

Line 102: “BVOCs e.g.,” → “BVOCs (e.g.,” ?

Thanks, corrected.

Lines 120-123: What are the units of Q , CGR, CS?

The units for these variables have now been added to the revised manuscript (lines 121-124 & Figure caption Figure 2).

Line 213: "components black carbon" → "components of black carbon"

Sentence has been revised.

Reviewer two:

I would suggest the authors to improve the readability of the manuscript through more diagrams and tables: - A schematic diagram summarizing the investigated interaction mechanisms; - A table with details of model configuration and highlighted new/important parameterization/treatment; - A table summarizing uncertain processes or those not sufficiently considered.

In response to this suggestion we have included in the methods section a new table 1, which summarises some of the critical model features. We contemplated also, as suggested, a table that summarises the missing processes, but felt in the end that this would not add crucial new knowledge, and keep this in the text at the end of the conclusion section.

Page 27142 line 24, "Aerosol particles were continuously monitored with a Scanning mobility particle sizer (SMPS) located at the foot of the eddy covariance tower ... ", how about the inlet system for aerosol measurements, were the aerosols sampled also from 30 m above the ground? If not, a difference between above and below canopy will be expected challenging the results of Fig. 2. To test the difference, I would suggest the authors to make similar comparison (as Fig. 2) but for a single species below and above the canopy, e.g., monoterpene (or other VOCs, trace gases).

A comparison of below and above canopy measurements is unfortunately not possible since neither for the BVOC nor for the aerosol concentration measurements along canopy vertical profile was possible in this extremely remote location. However, the way the aerosol sampling was conducted is similar to the well-established methodology applied in many aerosol monitoring locations (Nieminen et al., 2014). Since aerosols are monitored regarding the change in number and size distribution (not their flux) and the typical life-time of particles in the studied size range is close to a day or more, there is no difference expected between an inlet below or above the canopy. Among others, e.g. Nieminen et al., 2014 have shown that nucleation events are regional phenomena that can be measured at the same time within the space of several hundreds of square km.

Page 27142 line 24, please define the acronym "DGVM"

Thanks for spotting this, is now defined in line 127.

Page 27144 line 4, "A recent data-base estimate was 191, 495, and 1024 GtC in the 0–30, 0–100 and 0–300cm soil layer, of permafrost-affected soils, respectively". Are these estimates from observations or from models? What is the geographical coverage of these values (Eastern Siberia or above 40 degree N)?

The data are extrapolated from observations and are for high latitude soils affected by permafrost globally (ie, not including permafrost soils in mountains). The sentence is revised for clarification (now in lines 274-276).

Page 27145 line 1, " Multiple interacting processes can thus lead to enhanced global monoterpene emissions in future, or -if the "CO2 inhibition" is included- yield emissions that are more or less similar to present-day or even slightly smaller ". Can you elaborate how the CO2 inhibition was parameterized in your model?

We have added a short explanation to the methods text, see also response to reviewer one.

Page 27148 line 11, "The assumption of unchanging oxidant fields induces some uncertainty . . . The model climate is nudged towards ERA-40 reanalysis year 2000 meteorology, an approach that is widely used in aerosol-climate assessments". Did you use the same oxidative capacity field and nudging meteorology field for the year 2000 and 2100?

The oxidant fields and nudging meteorology are same for both 2000 and 2100. This allows us to study the sensitivity of CCN to BVOC emissions while assuming no changes in oxidative capacity or general circulation. We made this point more obvious, and moved it to the discussion section (see also response to request by reviewer one), lines 486-492.

Such treatment is OK for a sensitivity study but I am not sure if it is appropriate when you draw conclusions for a specific year 2100. Changes of land uses and VOCs will change the distribution of oxidants and meteorology.

The reviewer is correct in that we should be careful in some places for using a specific year for the ECHAM SOA and radiative effects.– which in fact wasn't really intended, even though we probably used "2100" in a too simplified way. In some cases this was correct, especially when referring to BVOC emissions that were input to the SOA simulations since these were indeed taken from a specific year from LPJ-GUESS transient 20th-21st century simulation. But in other cases we have revised the text, indicating that we mean to simulate conditions towards the end of the 21st century.

Page 27148 line 27, the authors used 1% supersaturation CCN. The atmospheric relevance of supersaturation depends on the aerosol concentration and updraft velocity. It will be better if the authors could give CCN at multiple supersaturations (e.g., in the supplement).

Page 27155 line 15, " What is more, SOA formation only partly enhances the survival of small particles by providing additional growth (Makkonen et al., 2012a), but partly also suppresses it by increasing the coagulation sink for small particles (Fig. A2, lower left panel; see also O'Donnell et al.,2011)".

CCN is defined for a specific supersaturation. Larger particles are better CCN (activated at lower supersaturation) than smaller ones. Though the coagulation removes smaller particles reducing the CCN number concentrations at high supersaturation, it increases the particle size leading to more good CCNs (activated at lower supersaturation). This is another mechanism affecting the indirect radiative effect of aerosols.

The manuscript uses CCN at 1.0% supersaturation to estimate potential effect on cloud properties. We agree that "realistic" supersaturations are generally lower than 1.0%, hence, CCN(1.0%) concentration as such provides the upper limit for CCN concentration. In the simulations, one must also bear in mind that in aerosol model M7, CCN(1.0%) might correspond to a cut-off diameter inside Aitken mode, whereas CCN(0.2%) cut-off is generally between Aitken and accumulation modes. CCN at 0.2% reflects larger aerosols, hence the changes in CCN(0.2%) likely seen further away from aerosol and precursor sources. We have included a new(CCN(0.2%)) figure in the appendix (Figure A3) corresponding to summer concentration changes averaged over 5 years. We also include explanatory text in lines 517-521.

Page 27150 Section 3.2, It is not clear for me how this section is linked to the modeling part of this study. Do you use it in the model parameterizations?

We have included the measurements to demonstrate that there are clear links between the concentrations of MT and SOA formation (e.g., Section heading and Figure 2). While this has been shown for a number of boreal forest sites we felt it important to also demonstrate this effect for Eastern Siberia, as an important basis for the model experimental set-up, because of its strong focus on BVOC emission effects on aerosol. We had also highlighted later in the text that an extrapolation of observation-based radiative forcing estimates to future forcings over large regions is not possible (lines 452-463). Data were not used for model parameterization in case of aerosol modelling.

Page 27151 line 14, "Hence, the poor relation between the source rate of condensing vapour and levels of radiation (Fig. 2b) indicates that OH-radical concentration did not have a major impact on Q. This agrees with the findings by Ehn et al. (2014) that ozone instead of OH. is an important, if not the main, atmospheric agent oxidizing organic vapours into a chemical form that condenses on particle surfaces."

I am not convinced by this argument because OH has a short life time and may have a large difference below and above the canopy, how about the correlation between Q and O₃?

Hens et al. (2013) have shown that the OH concentrations below and above canopy are indeed quantitatively different, but have a clear correlation and variability thus agrees qualitatively. The strong variation in the OH concentrations, which can be more than an order of magnitude within the matter of hours around sunrise and sunset (e.g. Petäjä et al., 2009 and Hens et al., 2013), should have a clear impact on Q if OH was the dominant oxidant converting MT to condensable vapors. And since the OH concentration is strongly affected by radiation (also below the canopy; Petäjä et al., 2009), the impact of OH concentration on Q should also be visible in Fig. 2b.

Unfortunately, we could not inspect the correlation between Q and O₃ (or Q and MT ozonolysis rate), because the remoteness of the site limited measurement capacity, and ambient ozone concentration measurements were not conducted during the campaign. However, even if we assumed that O₃ is the only oxidizer affecting on Q, we

would not expect a clear correlation between Q and O₃, because the relative variation in O₃ concentrations is much smaller than that of monoterpenes (during the campaign by Hens et al. (2013), the variation of O₃ concentration was within a factor of three, whereas the observed variation in MT concentrations reached factors of 10 to 100). The correlation we show in Fig. 2b suggests that Q is not heavily affected by the oxidant concentrations, which supports the assumption of O₃ being the main oxidant. We have made further clarifications in the manuscript, regarding to OH concentrations above and below canopy (line 417) and relative variations in concentration levels of OH, MT and O₃ (line 337-344).

What's the time resolution of data in Fig. 2b and 2d, there seem to be much less data than those in Fig. 2c.

Q in panels (b) and (d) were calculated for those periods for which the growth rate of particles was possible to determine. In panel (c) all the monoterpene measurements are presented, and those corresponding with events of determined growth rate are encircled. We have added an explanation to caption of Figure 2 to explain the reason for the different data points that are displayed in the panels.

Page 27151 line 27, "An overall C loss of 100 PgC assumed to be in the form of CO₂", what's the reference year?

Added clarification (compared to today's levels; seen also in Table 1; line 353).

Table 1: what's the unit of "NPP_global"?

Thanks, is clarified in the Table caption. (now Table 2)

To improve the readability, "BVOC in Tg C a⁻¹", should be "BVOC emissions" also correct that in the table. For the unit "ug_C g⁻¹ h⁻¹", what's the meaning of subscript "C" (instead of C in Pg C)? Overall, it is difficult to connect the table with its caption.

We have removed the subscript format in "C". The table caption was edited (also in response to reviewer one), and we hope it is easier to read now.

Table 2: The paper is an assessment of competing effects of CO₂ and SOA. The negative forcing of SOA effect has been evaluated and given. For comparison, what's the forcing due to a change of CO₂?

Table is now Table 3. Since CO₂ is a well-mixed greenhouse gas its value can only be given globally and is stated in the text (403). We feel it would look odd to add this single number into the table. Moreover, we were severely criticized in an earlier version of the manuscript for over-interpreting our radiative forcing estimates, which is why we would prefer to leave the table unchanged.

Figure 1: For "maximum summer leaf area index", do you refer to seasonal, monthly or daily data? "a,b" are used in the figure caption while "A,B" are used in figure labeling.

Thanks, we clarified. We also use capital letters for Figure and caption.

Figure 3: "Areas with statistical significant changes in CCN are indicated.", can you elaborate on the statistical analysis?

The figure 3 right panel indicates areas with significant changes with dots. The statistical analysis is based on monthly average CCN concentrations from 5-years of simulated data, and statistical significance of the CCN anomaly is evaluated using a two-sample t test, without assuming equal variance between the two populations. Added explanatory text to the Figure caption.

Figure 4: Can you also show the absolute increase (in the supplement) for reference?

Showing absolute changes in SOA mass in different size modes is unfortunately not possible from our analysis, since aerosol masses and the respective changes are dominated by accumulation and coarse modes, whereas the climate effects would be sensitive to new CCN from growth from nucleation and Aitken mode. However, we revised the Figure caption of fig. 4 to add more detail.

Figure A1: CO₂ on/off labels seem missing in top panels.

That is intentional, since the CO₂ inhibition is compared to today's atmospheric CO₂ as a reference = simulations with on and off would not show a large difference; the difference becomes only visible in the future simulations. Is clarified in the revised figure caption.

References:

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