We would like to thank both reviewers both for their support, and for their constructive criticism. We will try and address the later in the revised manuscript as outlined below; answers to the comments (italics; in some cases these were a bit shortened) in normal font.

Rev. 1

This provocative and insightful paper investigates possible atmospheric feedbacks from changes to boreal ecosystems in a warmer future world. The study builds on the boreal-aerosol-climate feedback ideas of Kulmala et al. and Spracklen et al. and is the first to examine the impacts co-changes in boreal monoterpene emissions (SOA precursors) alongside the CO2 releases from the boreal permafrost using a state-of-the-art process-based land model (LPJ-GUESS) and global aerosol microphysics model (ECHAM5.5-HAM2). The main strengths of the paper are the presentation of the new tower measurements from the Siberian site, and the interesting analyses of the effects of BVOC emissions changes on local particle populations in a changing background-aerosol environment (Section 3.3). In addition, the study finds that when altered vegetation dynamics and composition are accounted for in LPJ-GUESS, there is a net uptake of carbon (i.e. the permafrost release of CO2 is more than offset by increases in land storage and productivity), which is a novel and important result.

Thanks for these encouraging words.

*The major problem with the study, which persists throughout the manuscript, is a lack of understanding of basic climate dynamics, for example the differences between global climate change and regional radiation budgets. A change in the radiation balance (i.e. a forcing) is directly linked to the surface temperature change on the global annual average ONLY. Local and regional changes in the radiation balance are NOT correlated with the local surface temperature response. So, for example, it is scientifically inaccurate to use the words 'cooling' and 'warming' in reference to a regional change in the atmospheric radiation budget. Regional forcing does not imply regional climate response (e.g. see review by Fiore et al., 2012). In the case of the spatially concentrated aerosols, the regional climate response does not closely follow the spatial pattern of the radiative forcing (e.g. Taylor and Penner, Nature, 1994; Shindell et al., 2007+ many others). It is not possible to know what the regional surface temperature response would be to regional changes in the longwave and shortwave radiative budgets without running a fully coupled dynamic atmosphere-ocean global climate mode*l.

The referee is right in emphasising that a change in regional forcing does not necessarily mean a proportional regional climate response. We discuss our results in the original version of the manuscript in simplified way from perspective of a change in forcing, but it was not our intention to imply a one to one match, from forcing to climate response (see eg., Fig. 3,4). (see also response to next comment & revised title).

From our understanding of the Fiore et al. review, there seems to be not yet a consistent view of how much local changes in e.g., aerosol forcing patterns would translate into a change in surface temperature – the existing studies show a complex response. Still, we will need to ensure that any misunderstanding in that respect is weeded out from the manuscript. We have revised the text accordingly, and have also added some of the references listed above to the discussion in the text. In particular, we substantially changed the abstract, and added text to the introduction and results/discussion sections (including new simulations, and a new Table 2 that shows BVOC-induced changes in CRF and CSDRF for Siberia, northern hemisphere, and globally).

If the paper is about changes to the Siberian boreal regional radiation budget, then "warming" and "cooling" need to be removed from the manuscript. Alternatively, if the authors want to talk about sign of temperature change and "warming" and "cooling", then they must instead compute the global radiative feedback effect for each component. However, in the global case, the boreal SOA effects are totally irrelevant. Moreover, based on the actual radiative feedback estimates provided in the study

(CO2 = -0.09 Wm-2; local aerosol cloud albedo effect = -0.04 Wm-2), these effects are miniscule compared to natural variability in local radiation budgets of boreal forests i.e. the aerosol-cloud-albedo results presented in the study are also negligible in terms of local energy balance for the forest. It is telling that these estimates have not been provided in the abstract.

We have changed the title ensure that no misunderstandings remain (Future biogeochemical forcing in Eastern Siberia: competing effects of CO_2 vs. secondary organic aerosols). The referee points also to the difficulty of comparing climate feedback effects that operate via well-mixed greenhouse gases and those operating through short-lived, atmospherically fast reactive compounds.

Regarding the CO2 vs. aerosol effects: the numbers had not been omitted from the original abstract because of their small magnitude, as suspected by the reviewer. Our logic was more as follows:

1) One of the goals of the paper is to emphasise that –in principle- a number of potentially opposing processes need to be covered when assessing the full terrestrial-atmosphere feedbacks, using a study in boreal Siberia, and CO2 vs. BVOC/SOA as an exemplar.
2) We are well aware of the fact that a number of additional processes were not included in the model simulations and analysis, due to model constraints. In fact the missing aspects are not limited to albedo changes, but would also include: changes in the hydrology (which affects CH4 and N2O vs. CO2 fluxes), changes in the N budget, changes in Bowen ratio, etc. Thus, adding two out of many "numbers" to the abstract didn't seem to be defensible.
3) What is more, the data-driven, simple extrapolation and the microphysics model lead to quite different results. Both approaches are fraught with uncertainties, which we discuss in the text.

Based on the above reasoning, we did not want to give too much weight on the estimated forcings by placing them into the abstract. We had obviously not managed to point this discrepancy out in sufficient detail and revise the ms accordingly, with textual changes as already mentioned in the response to the first point..

There is a major disconnect between the use of sophisticated land ecosystem and aerosol microphysics models, and then using an inappropriately semi-quantitative back-of-the-envelope calculation for the aerosol indirect effect. Why isn't the global climate model employed to calculate the aerosol-cloud radiative changes? The back of the envelope AIE forcing is littered with unqualified assumptions including "calculated CCN values correspond to changes in cloud droplet number" and "Ac = 0.65". The albedo forcing is linearly dependent on Ac, so choice of Ac has a large impact on the results. Where does the 0.65 value come from and does it have anything to do with reality in the Siberian boreal? There are other methodological concerns described in the section below.

We agree with the reviewer on the level of detail on the indirect effect estimates. Due to computational costs, we could not quantify the radiative effects of each sensitivity experiment. Hence, we have come to an intermediate solution, nevertheless providing in the revisions a more sophisticated and suitable measure for the radiative effects. While we have kept the simulations from the previous manuscript, we have now calculated the indirect and direct aerosol forcing of the main experiments from ECHAM-HAM simulations (new Table 2,& revised methods and results. This will help the reader to put the overall BVOC-aerosol-climate sensitivity into perspective of the other results, and the CCN results from other sensitivity tests can be reflected onto the given radiative forcing.

To move forward, I recommend to re-write the paper in the framework of: 'the atmospheric aerosol impacts of future boreal ecosystem emissions'. A single paragraph could be included in the discussion at the end of the paper about possible climatic feedbacks from these atmospheric changes.

It was not entirely clear to us exactly what the reviewer suggested here. Since there was no major criticism raised on the sections regarding vegetation and BVOC observations/modelling, and the observational parts of aerosols we left these sections (that make up the largest part of the paper) more or less untouched. We did, however, revise the aerosol modelling section (including new simulations, revised Figures; see above) and hope we can remove the reviewers concerns in that respect.

Other major comments:
1. Page 19151. "We provide a comparative assessment of how the magnitude of the BVOC-SOA cooling compares with forcing from changes in the CO2 balance of Eastern Siberia." Why? What is the rationale for comparing CO2 and local aerosol radiative effects? The CO2 forcing is global and the local temperature and weather will respond to this global forcing through dynamic atmosphere and ocean feedbacks in the system.

Up to now, studies that investigate the role of terrestrial vegetation dynamics and carbon cycle in the climate system typically account solely for CO2, while studies that look at BVOC-climate interactions ignore other processes (eg, CO2). However, understanding the full range of interactions between atmospheric composition, climate change and terrestrial processes we need a much more integrative perspective. The reviewer is correct that it poses a challenge to combine effects of well mixed greenhouse gases and locally constrained, short-lived substances. By combining modelling/observations of BVOC, SOA, vegetation composition and CO2 exchange we seek to provide an example for such an approach – including a focus on a very remote area, where such data have not yet been obtained before.

We will revise/add the text in order to make this point stronger.

2. There is no mention of the local climate impacts of the albedo decrease associated with the regional enhancements woody vegetation and primary productivity. In addition, how do the associated biophysical land-surface changes impact the aerosol production and lifetime?

3. The effects of future changes to atmospheric water vapor and humidity are not mentioned anywhere in the manuscript. Do they influence aerosols and Ac?

Points 2 and 3 are perfectly valid, and would require a fully coupled climate model run with dynamic vegetation switched on, interactive carbon cycle, aerosol physics and air chemistry. For instance, applying a regional coupled model experiment (including LPJ-GUESS) recently showed that responses to climate warming in the arctic yielded not only treeline advance, but also densification, and changes in dominant PFTs (which we see here as well) which fed back to climate, via altered albedo and evapotranspiration, to change local temperatures with a pronounced seasonal signal, which in turn altered the C cycling. This biophysical feedback would certainly have changed BVOC emissions too, had they been in the model – let alone effects on aerosol chemistry and physics. But such a comprehensive coupled model analysis was beyond the scope of the project (hence the original title specifically stating "biogeochemical"). The authors are in fact not aware of whether any modelling groups has so far managed to provide runs with such fully dynamic, coupled models. Still, albedo had been mentioned briefly at the end of the original manuscript. In the revised version, we draw more attention also to other processes that so far cannot be easily (if at all) quantified with coupled model simulations.

4. The link between LPJ-GUESS model output and ECHAM5 simulations is not clear. The models are not dynamically coupled. Are the BVOC emissions computed in LPJGUESS and then input off-line into ECHAM5.5-HAM2?

Yes that is exactly how it was done (see also references to previous papers that applied such an approach). We have revised the methods section for further clarification.

5. It appears that the aerosol microphysics model does not account for the effects of future changes to gas-phase chemistry and the oxidative environment (e.g. background and local ozone levels), which will have a huge impact on SOA loading. A 15% yield is assumed regardless of oxidative environment. The authors need to comment on the influence of changing oxidation chemistry.

The reviewer is correct in that the aerosol-chemistry model assumes unchanged oxidant concentrations between years 2000 and 2100. Although an evident oversimplification, the assessment of changing oxidation fields is out of the scope of the current manuscript. However, we have added a comment on the revised manuscript on the assumptions of oxidant fields.

6. The future BVOC emissions will have an influence on ozone and methane, which contribute important large-scale radiative changes that are not mentioned in the paper. For example, ozone is an important forcing in the Arctic region (e.g. Shindell, Local and remote contributions to Arctic warming, GRL, 2007).

7. Natural and anthropogenic NOx emission changes and effects are not discussed. Even in "clean environments" NOx is present from soils and fires. NOx changes could have large impacts on the results. For example, even in "clean environments", where the ozone production is NOx-limited, an increase in VOCs still results in an increase in the total amount of ozone produced.

Points 6 and 7: these are all very true. We were not in a position to provide a comprehensive measurement and modelling exercise of all climatically relevant processes. As mentioned above, having a fully coupled aerosol/chemistry/climate model with vegetation dynamics (and biogeochemistry, and biophysics) enabled is a non-trivial request. Moreover, in addition to the computational constraints *per se*, parameterisation would also be a challenging task. There are in fact many difficulties of obtaining "only" the BVOC and aerosol measurements in remote locations such as Eastern Siberia - measurements that are crucial to provide at least some basis for the modelling. And just like there has so far been no observations of BVOC from these regions, there also is no data e.g., for soil NOx fluxes, let alone within canopy atmospheric chemistry.

While one could run fully coupled models to explore all interactions, we did not have the means to do so in the project (see response to Points 2 and 3). In fact, I am not aware of any of the present-day ESM would have all required processes dynamically represented (including dynamic hydrology and wetlands for emissions of CH4, and N2O; vegetation dynamics fully enabled, all gaseous chemistry and aerosol processes, and their interactions, etc.; and these to be run over centuries.). Many of the parameterisations in these models would not be backed by observations (see NOx as an example: without observational evidence, one could certainly question whether e.g., the Yienger and Levy soil NOx algorithm, or adoptions thereof, would produce realistic emissions).

Moreover, with the current state of system understanding one could ask if every study that seeks to explore interactions between atmosphere, climate and the land surface was to be required to encapsulate all possible exchanges. This would make it very difficult to isolate the importance of individual processes and in my view would, at our current state of modelling, not help to provide understanding.

8. Page 19150. "and hence have a net cooling effect at clean-air locations (Arneth et al., 2010; Makkonen et al., 2012b; Paasonen et al., 2013)." Do you mean net negative effect on the regional radiation budget? Have the effects on other important long-lived radiatively active agents (ozone and methane) been properly accounted for in these analyses?

This statement is on secondary aerosols (and not including all other potential interactions), and we rewrote, hoping that in the revised version the meaning is better phrased.

9. What about the direct radiative effect of the SOA change? Is that completely irrelevant for boreal ecosystems?

We have included an estimate of the direct radiative effect of changing BVOC in the revised manuscript.

10. There are several problems with Methods on page 19156:

(a) *"The generated climate was adjusted to the CRU period between 1960 and 1990". How was the generated climate 'adjusted'? Which variables? Did you force with CRU for present day and ECHAM5.5 RCP8.5 for future*?

Correct, the originally statement lacked an important reference, which is now included. We have also expanded the methods in response to the reviewer's questions.

(b) "Half of the SOA is assumed to be of such low volatility that it can partition to particles already at nucleation size (2 nm), increasing the formation of 2–3nm particles and hence the modelled total number concentration. However, most of the oxidized organic mass will be distributed to larger aerosols, increasing the coagulation and condensation sink." *These 2 sentences are confusing. Isn't it the remaining half of the SOA mass and not "most of the oxidized"? Please clarify*.

We have clarified the text in the revised manuscript.

(c) "All simulations are initiated with a six months spin-up, followed by one year simulation for analysis. The model is nudged towards year 2000 meteorology, reducing the noise arising from differing meteorological fields." This method is confusing and does not make sense. Firstly, a one-year simulation is not sufficient for an aerosol-climate sensitivity simulation. Meteorology has extremely high interannual variability in the boreal high-latitudes (e.g. Hawkins and Sutton, GRL, 2012; Mahlstein et al., ERL, 2011). Therefore, an estimate of interannual variability needs to be included in the paper as an uncertainty estimate (may be larger than the naked value computed from one simulation year e.g. -0.04 Wm-2). Secondly, exactly what nudging is applied? Is the ECHAM5.5-HAM2 model nudged to large-scale winds? Nudging the future climatic conditions at 2100 towards year 2000 meteorology is a wrong approach. The noise can be reduced by using consistent meteorology from the 2000s of the RCP8.5 simulation for the present day.

We understand the issues raised by the reviewer, but the approach taken here is widely used in aerosol-climate assessments. Firstly, many similar assessments are approached with chemistry-transport models, which inherently apply reanalysis meteorology products. A similar approach is used with global aerosol-climate models to constrain internal model variability and hence ease the comparison of simulated model experiments. Secondly, a one-year simulation after 3-6 month spin-up time is very often used for nudged experiments, although we agree that this approach does not reveal inter-annual variability in the results. Also, nudging future climate simulations with present-day meteorological winds is done in order to compare with present-day results with the artificial assumption that e.g. the cloudiness, winds etc. are not changing. We have clarified these in the revised manuscript.

(d) The present day run should use the year 2000 anthropogenic emissions developed for the IPCC AR5 inventory that have been properly harmonized with the future RCP8.5 scenario (LaMarque et al., 2010). The AEROCOM emissions used in this study are not consistent with the RCP8.5.

We agree with the reviewer that the inventories are not consistent. From the BVOC-aerosol-climate effect point of view we believe that this is not crucial for the manuscript. One of the main results regarding BVOC-aerosol-climate coupling is done with two simulations, present-day and future BVOC emissions, but keeping other emissions constant. This commonly used approach addresses the aerosol and radiative effects of BVOC change itself, but is unaffected by emission inventory inconsistency. In addition to this, we have simulated the sensitivity of aerosol fields to emission potential uncertainty in present-day, which is also unaffected by the inconsistency. We do address also the concurrent changes in BVOC emission and anthropogenic emission. However, we believe that other uncertainties in the underlying assumptions on anthropogenic emissions exceed the uncertainties from somewhat inconsistent emission inventories. Globally, the AEROCOM inventory (141.8 Tg(SO_2)/yr) exceeds present-day SO_2 emissions of RCP8.5 scenario of 107 Tg(SO2)/yr. Hence, the current simulations would

generally overestimate the contrast between years 2000 and 2100. However, it is not evident, to which extent this applied in the domain of interest in the present study.

(e) How is it possible to isolate the impacts of changing wildfire emissions on CCN by holding both wildfire and anthropogenic pollution emissions fixed to year 2000?

There are two separate issues related to wildfires in the manuscript. First, we show that the behaviour of BVOC oxidation products varies between gridboxes depending on the pre-existing aerosol size distribution, which is in the Siberian domain modulated by fire occurrence. The presence of wildfire in a certain model gridbox generally leads to SOA condensation on larger particles, leading to reduced new particle formation. Second, we want to underline the uncertainty of wildfire emission estimates. Generating new scenarios and modules for wildfire evolution in Siberia is out of the scope of this manuscript, but simulations with either estimated future wildfire or by keeping present-day wildfire distribution reveal some sensitivity of CCN to wildfire in Siberia.

(f) Line 22. E = 1.9. It is only later in the results section that we actually find out what this variable is, including units. Please include units and definition here too*.

Has been added, thanks for spotting this.

Minor comments:
Page 19155 Line 21 repetition of "Larch, in this model setup"

Has been corrected, thanks for spotting it.

Rev. 2:

This is an interesting paper with results from an understudied region. The paper makes an important point that the climate change feedbacks that are typically considered may not be the dominant ones and impacts such as BVOC emission changes need to be quantified. While this is a good first step in a qualitative assessment of the potential impacts in this region it is not really quantitative because not enough is known about these processes and the uncertainties are too high. This paper shows the potentially important role of BVOC but the authors need to be very clear on how little is known of the key processes and rates here.

Thanks for these positive statements.

I recommend the paper be published after addressing the following points. General: In order to determine if changing BVOC can be important for climate, it is necessary to show that BVOC are emitted at rates that can impact particle concentrations. There is large range in emission factors (1.9 to 9.6 ugCm-2h-1) and the resulting impact on particles is sensitive to the assumed emission magnitude. *The authors should provide some assessment of the BVOC emission factors by comparing with ambient data. How do these leaf level emissions compare with observed monoterpene concentrations and the aerosol growth rate shown in Figure 2?*

The reviewer raises an interesting question, but using above canopy monoterpene concentration measurements as constraint for leaf emissions is a complex observation/modelling exercise in itself – and is unfortunately beyond what we can reasonably do with our limited data set. Scaling leaf-emission fluxes to above-canopy concentration requires "in-between" a detailed canopy exchange and chemistry model, information about mixing in the near-canopy air layer, as well as information on (among others) emissions of nitrous oxides and concentrations of OH and O_3 (both the mixing, the chemical reactions, and the gradient of light and temperature need to be accounted for) – none of which are available from the Siberian field site. Moreover, the leaf-scale measured emissions were derived at leaves close to top

of the canopy, and there will be a gradient in leaf-emissions that responds to gradients in e.g., temperature and light. The observed concentrations thus are the results of horizontal, vertical (tree:tree differences) as well as seasonal variability in emissions, plus variability in the overall chemical and physical environment.

Still, figure 2D shows that BVOC concentration is a significant contributor to the growth rate. While formation and growth of secondary organic aerosol has been shown in many studies to take place via condensation of organic vapour (Carslaw et al., 2010), the required landscape scale emissions and surface concentrations do not need to be at their seasonal maximum (which also implies substantial variability in leaf-level emissions). For instance, most nucleation events in a Scots pine dominated landscape in Finland have been found in spring, when measured monoterpene concentrations in the near-surface were about one tenth of the summer time maximum (ca. 60 ppt, *vs*. up to 500 ppt; Haapanala et al., 2007;Lappalainen et al., 2009). We found here MT concentrations of similar magnitude to these.

What evidence is there that monoterpene emissions, especially emissions from stored pools, will be higher in a warmer environment. There is no question that emissions are higher if temperatures increase for a few hours but what do we know about monoterpenes emitted from trees growing in a warmer environment. There could be limits based on the pool size or perhaps plants adapt to a future climate. Discuss what is known about this and how the results depend on the assumptions made here.

The reviewer is perfectly correct –acclimation is a real challenge for global modelling; and it is so far not well tackled. Not only for BVOC emissions but also for other important ecosystem processes (e.g., temperature optimum of photosynthesis, respiration…). We have added a paragraph that discusses these important aspects.

Specific:
Section 2.1 BVOC measurements: The first sentence says " Leaf and canopy BVOC emissions fluxes. . .. were measured" but I don't see the canopy BVOC flux measurements described anywhere. Monoterpene concentration data are shown in Figure 2 but I don't see where there are described in section 2.1

Good point and an oversight from our side, thanks for spotting this. We have removed reference to the fluxes (since indeed these haven't been used), and added information on how concentrations were measured to the methods section.

Page 19154, lne 3: delete 'was"
Thanks, revised.

Page 19155, line 7-10: This does not seem to agree with what is reported by Kajos et al. Also, it does not agree with the monoterpene concentration data in Figure 2. If there are light dependent monoterpenes then the lower PAR values should have lower emissions but the figure shows the opposite.

We have slightly revised the sentence to avoid misunderstanding (lines 167-171). Lower concentration do not automatically correspond to lower emissions in case of reactive substances– the observations could also reflect a faster chemistry taking place at the high light conditions.

Page 19155, line 15: reword "allows to"

Sentence is revised: As a result, the model's PFTs can be mapped to tree-species…

Page 19155, line 17-19: reword this sentence. "applying the necessary BVOC emission capacities" is repeated

Done, thanks.

Page 19155, line 20-21: reword this sentence. "Larch, in this model setup" is repeated

Done

Page 19156, line 10: reword this sentence. "to allow establishment" is repeated
Is corrected, thanks.

Page 19155, line 28: insert "the" in between "from" and "biosphere"
Done.

Page 19157, line 1: Why 15%? Provide some justification for this number.
The 15% yield is based on AeroCom recommendation (Dentener et al., 2006), and is currently being used by most aerosol-climate models (see e.g. Tsigaridis et al., 2014). Although certainly unrealistic in terms of realistic SOA formation, the 15% SOA yield serves as a reference value and the study presented in our manuscript does not address the question of SOA yield uncertainty itself.

Page 19157, line 17: It is not noise that is being reduced.
The text regarding nudging is rewritten in the revised manuscript.

Page 19159, line 5: reword "the thus"
True, sounds odd. Removed "thus".

Page 19159, line 22: Solar radiation can be, but is not always, a good surrogate for OH concentration. The authors should provide some results from their modeling to demonstrate

Rohrer and Berresheim (2006) show very strong correlation (R=0.94) between solar ultraviolet radiation and OH concentration in a 5-year data set at Hohenpeissenberg, Germany. Furthermore, Hens et al. (2014) showed that especially the day-time OH concentrations depend on the photolysis rate also in boreal forest region (Fig. 8 in Hens et al., 2014). Even though there are several other factors affecting the OH-concentration, we find that 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. If such relation would exist, the data points with high radiation levels should deviate from those with very low radiation levels.

To clarify these points in a better way, we have added text & literature references.

Page 19160, line 27: recent studies have shown that many conifers emit some light dependent monoterpenes
That is correct and we were not entirely sure about the comment here, since we had added the comment about hybrid emission patterns from larch already in the previous version. But we have revised the sentence to clarify this further.

sPage 19161: Was there any change in emissions simulated with change in species composition within a PFT? Were all conifers assigned the same emission factor? What was the impact on BVOC emissions

due to the change in PFT distributions?

We used here only the shade intolerant summergreen type PFT for mapping to larch-parameters (see methods, clarified in revised version). Emission factors were varied for larch to cover range of leaf-scale measurements (see table, simulated emission totals).

Page 19162, line 13: is "are" supposed to be "area"?
Yes! – thanks, corrected.

Page 19163: are these changes significant (for example, the CCN increase of 5-10 cm-3).
We have included details to the revised manuscript (included also in revised Figure 3).

Page 19163, line 17: what is "CCN (0.2%)"? I can't tell what 0.2% represents.

Thanks for highlighting this oversight, has been clarified in the revised manuscript .