Answers to reviewer III:

We want to thank the reviewer for the insightful and detailed comments on our manuscript. Based on the observation and suggestions for improvement, we have revised our manuscript; the details of the revisions are given below along with answers to the questions the reviewer raised.

The paper presents a study about the variation of natural aerosol production due to biogenic emissions from the boreal forest with ambient temperature. With regard to the expected and partly already observed climatic change the topic is of general interest. However, the study is based mainly on model results, only measurements of aerosol number size distributions are used. Thus, the main criticism from my side is that the authors have no evidence to what extent the observed particles consist really of biogenic material. Maybe there are other studies from the same sites with chemical measurements which can be used to strengthen the results. Otherwise it is much speculation about the composition of the particles. More detailed points are listed below. However, if the authors address the points carefully I can suggest the paper for publication in ACP.

Answer: *The available chemical measurements at the SMEAR II station show that submicron particles consist mainly of organic material during the summer season. In summer, the average ratio between organic and elemental carbon at this site is >10 (data not yet published), which suggest very small contribution by anthropogenic combustion sources. This gives indirect evidence on the very important role biogenic organic matter in submicron particles at this station. When looking at ultrafine particles (<100 nm in diameter), as in our study, the contribution of biogenic organics is clearly the dominant one. This has been shown both by direct measurements of growing particle at the SMEAR II site (Pennington et al. 2013, Atmos. Chem. Phys. 13, p. 10215) and by different particle growth rate analysis made for the SMEAR I and II site (see our detailed responses below).*

Comments in detail:

Abstract:

The abbreviation BVOC should be explained or not used here.

Answer: *We have changed the term in the revised version.*

Introduction:

A better separation between your work and that from Tunved et al would be nice. Could you please explain briefly the method and results from Tunved and the relation to your study. From this introduction it is not clear, what is new here.

Answer: *The idea behind this study is similar to that of Tunved et al.'s two studies (2006 and 2008), namely to classify observations at field stations by the amount of time air masses have spent over boreal forest, and then estimate the emission of biogenic aerosol precursor vapours, and then connect the obtained numbers to aerosol observations. In these studies, a clear connection was established, and in the 2008 study Tunved et al. also provided a parameterization, which described the effect of a temperature increase on the aerosol size distribution was taken into account. The parameterization assumed that temperature only affected the VOC emission; however, it is likely that a change in temperatures will also change the VOC conversion efficiency (aerosol yield) into aerosol particles. In this study, we attempt to use similar methods to establish whether this temperature effect on the aerosol yield can be observed in the data. In addition, the studies of Tunved et al. did not account for particle losses during transport to the site. By estimating these losses, we can obtain an upper limit estimate for the conversion rate of emitted VOC into aerosol during transport to the site. The text will be modified accordingly to explain these differences to the readers.*

Measurement and methods:

Please explain the method here in more detail. How can you be sure that really monoterpenes are responsible for particle formation and growth?

Answer: *Several papers have analyzed the growth of nucleated particles at the measurement sites, and overall summaries are presented in the paper by Nieminen et al. (2014, Boreal Env. Res., in press) for the SMEAR II station and by Kyro et al. (2014, Atmos. Chem. Phys. 14, p.4383) for the SMEAR I station.*
The most plausible anthropogenic trace gas contributing to the particle growth is sulphuric acid, which on average explains less than 10% of the growth in SMEAR II and a bit more in SMEAR I. So low-volatile organic compounds give the dominant contribution to the particle growth in air masses arriving at these sites outside the winter period. There are other studies that suggest biogenic emissions to be by far the most important source of such low volatile organics at these sites. So, while we cannot rule out the anthropogenic contribution to the growth caused by organic compounds, this contribution is very likely small during summertime in our study area. It is likely that other biogenic precursors (eg. sesquiterpenes) besides monoterpenes give some contribution to the particle growth as well, but the magnitude of this contribution is impossible to estimate using the available information. Laboratory studies (eg. Mentel at al., 2009, 2013) have shown that the mixture of VOC, even the mixture of different monoterpenes, influences the aerosol yield and formation potential. As our ability to currently quantify the emission patterns is still limited, current approaches use monoterpenes as a kind of a proxy for aerosol-forming VOC, and include the complexity of the VOC mixture and aerosol formation microphysics as a single coefficient, the aerosol yield. As our results show, the aerosol mass yield and particle formation potential at different temperatures is variable; this may be partly due to variation of the VOC emission pattern and partly due to other microphysical reasons. The result highlights the need to include yield parameters with a functional dependence on eg. temperature in secondary aerosol formation descriptions.

Page 33316, line 1-4: Do you think this calculation of ground level temperature is realistic? I would be skeptical and expect large inaccuracies. Can you try to validate this method by ground based measurements? There should be a few stations on the trajectory way measuring temperature.

Answer: *Temperature from trajectory calculations was from GDAS (Global Data Assimilation System) reanalysis archive which has been combined all the available ground measurements. Many previous studies have also shown that meteorological data from GDAS is applicable in scientific studies. We used a constant lapse rate to estimate the ground temperature, which was the same approach applied in Tunved et al., (2008). In this way, our results would be comparable to Tunved et al., (2008).*

Following paragraph: How do you know the monoterpene emissions, also its seasonal dependence? Can you please give a reference or an explanation?
Page 33317: Again concerning the monoterpene emissions: Is there any chance to validate these calculated emissions with measurements?

Answer: *Tarvainen et al. (2005) has shown the seasonal dependence of monoterpene emissions over boreal forest environment. When calculating monoterpene emissions, we used a well-established emission algorithm. Monoterpene emissions have been measured during different field campaigns at the SMEAR II site, and the performance of the emission algorithm has been found to be quite reasonable. We added a few lines into the text to discuss the accuracy of emission calculation in more detail.*

Results and discussion:

Page 33318, line 23: How accurate is the average trajectory temperature? Please give an uncertainty range here?

Answer: *The meteorological dataset including e.g. trajectory temperature, mixing height in the trajectory calculations was from GDAS archive for dataset after 2004, and from FNL archive for dataset before 2004, provided by the NCEP (National Centre of Environmental Predictions). GDAS allocates observations to a gridded, 3-D, model space, which include surface observations, balloon data, wind profiler data, aircraft reports, radar observations, and satellite observations. Therefore, the temperature uncertainty from the trajectory has been minimized by combining all the available measurements. Previous studies have shown that meteorological data from GDAS is applicable in scientific studies.*

Page 33318, line 27: Please define 'MT', I think I know what you mean, but it helps to have it defined when it is used for the first time.

Answer: *We have added monoterpene (MT) in the revised text.*

Page 33320, lines 1-6: What's about new particle formation at the sites? We all know the nice banana-plots from Hyytiälä, but your figures look quite different? How does new particle formation at the site influence your results?

Answer: *Figures 4 and 5 integrate over all "clean" air masses entering the sites, including those in which nucleation and growth is very weak compared with the most pronounced nucleation and growth events shown by the banana plots. The cases when nucleated particle have grown to larger size when arriving our measurement site dominate the cases when we see small freshly-nucleated particles at our site, which explains the feature that the growth appears to start from between about 10-20 nm in Figures 4 and 5. The same integration of air masses explains also the lower particle growth rates in Figures 4 and 5 as compared with typical growth rates observed during the banana-type nucleation events. The few case studies of particle growth in air mass transport between 2 measurement stations investigated systematically by Väänänen et al. (2013, Atmos. Chem. Phys. 13, p.) support this interpretation.*

Following paragraph: I also think that the growth rate is very low and your following discussion does not convince me. Maybe the growth rate is highly variable between ground and higher altitudes? At ground sites usually higher values are observed. If you think you deviation in the GR comes from taking the whole data set, can you calculate it for individual cases only?

See our previous answer. We have made a few airborne measurements of particle growth using a Zeppeling around the SMEAR II station (data not yet published). These measurements have shown that the particle growth rates observed higher up in the boundary layer are not essentially different from those measured at the ground level.

Section 3.2.2: The used monoterpene concentrations are obtained from the model, right? It is a bit confusing to, because these modelled MT values are used together with measured particle concentrations.

Answer: *Yes, monoterpene concentrations were obtained from the model. We used a well-established MT emission algorithm, as it is not possible to obtain all the monoterpene concentrations along trajectories, due to limited ground measurements. The performance of such MT emission algorithm has been found to be very reasonable. Since we attempted to investigate the general trend of temperature influence on the natural aerosol yield, the modelled MT concentrations used together with measured particle concentrations should be sufficient to reveal such result.*

Page 33324, line 20-25: Why is the aerosol yield higher for lower temperatures? This is not obvious to me.

Answer: *This is one of the main results of our study. As explained earlier, the higher yield may occur due to differing reasons: at colder temperatures, VOC mixtures with higher aerosol-forming potential are emitted, and therefore, more of the total VOC is converted to aerosol mass. Another possible explanation is that temperature has an effect on the gas-to-particle conversion process. The observation that new particle formation is more efficient at lower temperatures points towards more low-volatile vapours being present during this time, which could be due to different plant emission pattern.*

Summary and Conclusion:

From this section it sounds that the new particle formation always starts above the ocean and particles grow over land and are finally measured at the site. Whats about nucleation over land?

Answer: *What we mean here is that nucleation takes preferably place in originally clean marine air masses AFTER they arrived to the boreal forest area, i.e. over land. We will modify the text a bit to avoid misunderstandings.*

I expect also other compounds to contribute to particle growth beside the biogenic emissions. Can you quantify this or is this really negligible?

Answer: *Several papers have analysed the growth of nucleated particles at the measurement sites, and overall summaries are presented in the paper by Nieminen et al. (2014, Boreal Env. Res., in press) for the SMEAR II station and by Kyrö et al. (2014, Atmos. Chem. Phys. 14, p.4383) for the SMEAR I station.*
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