### What are the <u>C</u>ehallenges <u>infor</u> modelling isoprene and monoterpene emission dynamics of <u>subarctic</u> plants<u>:</u> <u>? a case study from a</u> <u>subarctic tundra heath</u>

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15 Abstract. The Arctic is warming at twice the global average speed, and the warming-induced increases in biogenic volatile organic compounds (BVOC) emissions from arctic plants are expected to be drastic. The current global models' estimations of minimal BVOC emissions from the Arctic are based on very few observations and have been challenged by increasing field data. This study applied a dynamic ecosystem model, LPJ-GUESS, as a platform to investigate short-term and longterm BVOC emission responses to arctic climate warming. Field observations in a subarctic heath-tundra heath with long-20 term (13 years) warming treatments were extensively used for parameterizing and evaluating BVOC related processes (photosynthesis, emission responses to temperature and vegetation composition). We proposed an adjusted temperature (T) response curve for arctic plants with much stronger T sensitivity than the commonly-used algorithms for large-scale modelling. The simulated emission responses to  $2 \stackrel{\circ}{\overset{\circ}{\to}} C$  warming between the adjusted and original T response curves in the model were evaluated against the observed warming responses (WR) at short-term scales. Moreover, the model's responses 25 to higher levels of warming by (4 °C and 8 °C) were also investigated as a sensitivity test. The model was able to showed reasonable agreement to the observed reproduce vegetation CO2 fluxes in the main growing seasons as well as day-to-day variability of isoprene and monoterpene emissions. The observed modelled BVOC WR, especially for relatively high emission rates of BVOC as well as isoprene WR, were better captured by using the adjusted T response curve, compareding with using than by the original common one. A few days' underestimation of leaf T led towa the underestimated emission rates as well asand WR. During 1999-2012, the modelled annual mean isoprene and monoterpene emissions were 20 and 8 30 mg C m<sup>-2</sup> yr<sup>-1</sup>, with an increase in emission by 55 % and 57 % for 2 °C summertime warming, respectively. Warming by

4 °C -and 8 °C for the same period -further elevated isoprene emission for all years compared with 2 °C warming, but the impacts on monoterpene emissions levelled off at the last few years, because of a decreased decreasing coverage monoterpene emitters among the evergreen prostrate dwarf shrubs. The high WR captured by the adjusted T response curve highlight the strong T sensitivity of arctic plants.

5 At hour-day short term-scale, the WR seem to be strongly impacted by leaf T; while at day-year long term-scale, the WR are a combined effect of plant functional type (PFT) dynamics as well as and instantanewous BVOC responses to warming. The identified essential issueschallenges associated within estimating arctic BVOC emissions are: (1) correct leaf T estimation/extrapolation based on air T; (2) PFT parameterization accounting for BVOC-plant\_emission features as well as PFT<sub>2</sub>'s-physiological responses to warming; and (3) representation of long-term vegetation changes vegetation dynamics in 10 the past and the future.

#### **1** Introduction

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Biogenic volatile organic compounds (BVOC) are reactive hydrocarbons mainly emitted by plants. Emissions of these secondary metabolites are involved in plants growth, plant defence against biotic and abiotic stresses, plant communication as well as reproduction (Laothawornkitkul et al., 2009; Peñuelas and Staudt, 2010; Possell and Loreto, 2013). BVOC synthesis is regulated by enzyme activity, and many compounds are emitted in a temperature (T)- and light density (Q)dependent manner (Li and Sharkey, 2013). BVOC released into the atmosphere react with hydroxyl radicals (OH), which could reduce the atmospherice's oxidative capacity and therefore lengthen the lifetime of the greenhouse gas-methane ( $CH_{4}$ ). as a potent greenhouse gas (Di Carlo et al., 2004; Peñuelas and Staudt, 2010). An increase in BVOC emission could also elevate the tropospheric ozone  $(O_3)$  concentration when the ratio of BVOC to NO<sub>x</sub> (BVOC/NO<sub>x</sub>) is high (Hauglustaine et al., 2005), and increase the as well as secondary organic aerosol (SOA) formation (Paasonen et al., 2013). BVOC could also limit ozone formation when the BVOC/NO<sub>x</sub> ratio is low, a situation in which the regeneration of NO<sub>2</sub> can be mainly achieved by NO reacting with  $O_3$  (Hauglustaine et al., 2005).

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monoterpenes contribute most of the emissions (~70 % and 11 %, respectively, Sindelarova et al. (2014)). The modelled emission rates for isoprene are of similar magnitude as for  $CH_4$  (Arneth et al., 2008). However, the current estimates of regional emission distributions are highly uncertain for both isoprene and monoterpenes for two reasons: 1) the current emission estimates are based on field studies mainly covering tropical, temperate and boreal ecosystems (Guenther et al., 2006), lacking observational data for the Subarctic and Arctic; 2) the uncertainties in driving variables (vegetation distribution and seasonality, climate and environmental data (incl. soil water availability and the spectrum of the incoming

Global estimates of non-methane BVOC emissions are in the range of 700-1000 Tg C yr<sup>-1</sup>, of which isoprene and

30 light), and abiotic emission capacity stress, etc.) and in emission responses to these drivers (Guenther et al., 2006; Arneth et al., 2008). For instance, plants adapted to the cold environment of the Arctic appear to respond to warming differently than plants from low latitudes (Rinnan et al., 2014). Till now, the emissions from high latitudes (including the Arctic and the Subarctic) have been assumed to be minimal due to low foliar coverage. T and plant productivity (Guenther et al., 2006; Sindelarova et al., 2014). However, recent observations from the Arctic have indicated the need for revising the current assumption, as higher emissions from both plants and soils than anticipated in large-scale models have been measured (Ekberg et al., 2009; Holst et al., 2010; Potosnak et al., 2013; Rinnan et al., 2014; Schollert et al., 2014; Kramshøj

et al., 2016). Furthermore, field experiments focusing on the effects of climate warming on BVOC emissions have found unexpectedly high responses of BVOC release to a few degrees of warming (Tiiva et al., 2008; Faubert et al., 2010;

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- Valolahti et al., 2015; Lindwall et al., 2016a), which has underlined the potentially significant role of arctic BVOC emissions under changing climate. The Arctic is warming at approximately twice the global rate (IPCC, 2013) and the warminginduced drastic vegetation changes (AMAP, 2012) could impose substantial changes in BVOC emission. Both isoprene and monoterpenes are produced through the 2-C-methyl-D-erythritol 4-phosphate/1-deoxy-D-xylulose-5-
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phosphate (MEP-DOXP) pathway and are reaction products of their chief precursors, glyceraldehyde-3-phosphate (G3P) and pyruvate. G3P is produced along the chloroplastic Calvin Cycle-and serves as the chief precursor. Mechanistic models have often linked the biosynthesis of isoprene and monoterpenes with photosynthesis processes (Niinemets et al., 1999; Martin et al., 2000; Zimmer et al., 2003; Pacifico et al., 2011; Unger et al., 2013; Grote et al., 2014). In the short-term (hours-days), 15 the responses to Q and T of isoprene and part of monoterpene productions- are very similar to those of photosynthesis, but with a higher T optimum for BVOC production than photosynthesis (Guenther et al., 1995; Arneth et al., 2007). Furthermore, some monoterpenes can be emitted from storage pools in plant organs (e.g. glands or resin ducts, (Franceschi et al., 2005)). Along with the short-term responses, the long-term (days or longer) BVOC dynamics are-is affected by vegetation composition changes (Faubert et al., 2011; Valolahti et al., 2015), seasonality-vegetation phenology (Staudt et al., 2000; 20 Hakola et al., 2006), past weather conditions (Ekberg et al., 2009; Guenther et al., 2012) and vegetation growing conditions, e.g., soil water and nutrient availability (Possell and Loreto, 2013), atmospheric CO<sub>2</sub> (Wilkinson et al., 2009) and ozone levels (Loreto et al., 2004; Calfapietra et al., 2007). In comparison with empirical models (Guenther et al., 1995; Guenther et al., 2006; Guenther et al., 2012), process-based ecosystem models, explicitly representing BVOC synthesis activities, can vary with species as well as generally capture more of these long-term growing environment effects and could thus be more 25 useful in terms of predicting long-term emission responses to environmental changes (Monson et al., 2012). Usually, estimates of BVOC responses to Q and T are based on the Guenther algorithm (referred to here as G93, (Guenther et al., 1993)) and observed emission rates are often standardized to emission capacity at standard conditions (T of 30 °C and photosynthetically active radiation (PAR) of 1000 µmol  $m^{-2} s^{-1}$  using the G93 algorithm to allow for comparison with other observations. Potosnak et al. (2013) fitted leaf-level isoprene emission rates to T and Q in a moist acidic tundra and found the G93 algorithm characterized well with the T response, but not Q response. However, Ekberg et al. (2009) found that the

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T response of the G93 algorithm is not sensitive enough to capture the observed high T responses of wet tundra sedges, which was further supported by other studies in the high latitudes (Faubert et al., 2010; Holst et al., 2010). Furthermore, species-specific emission profiles (Rinnan et al., 2011; Rinnan et al., 2014; Schollert et al., 2015; Vedel-Petersen et al., 2015) have not yet been integrated into the modelling of (sub)arctic BVOC emissions (Arneth et al., 2011; Guenther et al., 2012;

Sindelarova et al., 2014).<sup>5</sup> which-<u>These</u> need to be included as a trait of plant functional types (PFTs), especially when targeting at studying the drastic impacts of climate change on vegetation composition as well as BVOC emissions in the Arctic. In addition, tundra plants with relatively dark surfaces and low growth forms (commonly less than 5 cm tall) may experience much higher leaf T than the air T at 2 m height provided by weather stations (Körner, 2003; Scherrer and Körner, 2010; Lindwall et al., 2016a), which could lead to larger emissions than anticipated in current models.

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The aim of this work was to integrate the observed emission features of arctic plants into a process-based ecosystem model in order to improve the current model estimations of arctic BVOC emissions, and to advance our understanding<del>s</del> regarding of the emission dynamics for arctic ecosystems in a warming future. The process-based dynamic ecosystem model<del>,</del> LPJ-GUESS (Lund-Potsdam-Jena General Ecosystem Simulator) (Smith et al., 2001; Smith et al., 2014)-\_was used as a platform

to distill\_simulate\_short-term and long-term responses of BVOC emissions to changes in climate for arctic plants. The model links isoprene and monoterpene productions with photosynthesis (Arneth et al., 2007; Schurgers et al., 2009). For the application to a subarctic heath tundra, the process parameterization utilized field observations of the-long-term (13 years) warming treatment effects on vegetation composition and BVOC emissions (Tiiva et al., 2008; Faubert et al., 2010; Valolahti et al., 2015). The specific objectives of this study were: (1) To capture the observed BVOC T response of BVOC emissions <u>sensitivity</u> for a subarctic ecosystem; (2) To address the importance of short-term and long-term impacts of warming on ecosystem <u>as well as</u> BVOC emissions; (3) To diagnose key model developments needed to better present BVOC dynamics for the arctic region.

#### 2 Materials and methods

#### 2.1 Study area and observational data

- The data used in this modelling study were collected at a dwarf shrub/graminoids heath tundra site-located in Abisko, northern Sweden (68°21'2N, 18°49'E). The vegetation consists of a mixture of evergreen and deciduous dwarf shrubs, graminoids and forbs. A long-term field experiment was established at this site in 1999 to investigate the effects of climate warming and increasing litter fall, resulting from the expanding tundra vegetation, on the functioning of the ecosystem. The experiment included control (C), warming (W), litter addition (L) and a-combined warming and litter addition (WL) treatments (Rinnan et al., 2008). In the current study, we only focused only on the observations from the C and W treatments. Each treatment, covering an area of 120 × 120 cm, was replicated in six blocks. The W treatments used open-top chambers (OTCs), which passively increased air T by around 2 °C, and also caused around 10 % reduction in PAR (Valolahti et al., 2015).
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During the years 2006, 2007, 2010 and 2012, BVOC emission rates were measured for all plots by sampling air from transparent polycarbonate chambers into adsorbent cartridges using a push-pull enclosure technique and analysis by gas chromatography-mass spectrometry (GC-MS). The enclosure covered a  $20 \times 20$  cm area in each plot. The isoprene emission datasets for 2006-2007 can be found in Tiiva et al. (2008) and for 2012 in Valolahti et al. (2015). For the year 2010, isoprene

emission rates were not analyzed due to technical problems (Valolahti et al., 2015). The monoterpene emissions datasets have been published by Faubert et al. (2010) for 2006-2007 and by Valolahti et al. (2015) for 2010 and 2012. Notably, BVOC in this study only refers to isoprene and monoterpenes. Closed chamber-based  $CO_2$  fluxes were measured in the same area during the same years (Tiiva et al., 2008; Valolahti et al., 2015). Species composition and coverage in the plots in the same years were estimated by point intercept-based method, in which a hit is recorded each time a plant species is touched by a pin lowered through 100 holes covering the plot area of  $20 \times 20$  cm (Tiiva et al., 2008; Valolahti et al., 2015). Species composition was measured in June for 2006, 2010 and 2012, and in June, July and August for the year 2007. Furthermore, air T and PAR inside the chamber were also monitored during the BVOC sampling time.

#### 2.2 LPJ-GUESS

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- LPJ-GUESS is a climate-driven dynamic ecosystem model with mechanistic process based representations of plant establishment, mortality, disturbance and growth as well as soil biogeochemical processes (Smith et al., 2001; Sitch et al., 2003). Vegetation in the model is defined and grouped by plant functional types (PFTs), which are based on plant phenological and physiognomic features, combined with bioclimatic limits (Sitch et al., 2003; Wolf et al., 2008). The model has been widely and successfully applied for simulating vegetation and soil carbon fluxes as well as vegetation dynamics at different spatial scales (Wolf et al., 2008; Hickler et al., 2012; Smith et al., 2014; Tang et al., 2015). In the model, individuals of each PFT in the same patch (replicate unit in the model representative of vegetation stands with different histories of disturbance and succession) can compete for light and soil resources. Plant establishment and mortality are represented as stochastic processes, but influenced by life-history, resource status and demography (Smith et al., 2014). For summergreen plants, an explicit phenological cycle is implemented, which is based on the accumulated growing degree day (GDD) sum for leaf onset and full leaf cover. In LPJ-GUESS, a generalized Farquhar photosynthesis model (Farquhar et al., 1980; Collatz et al., 1991)(Farguhar et al., 1980) for large-scale modelling is used to simulate canopy-level carbon assimilation (Collatz et al., 1991; Haxeltine and Prentice, 1996b; Haxeltine and Prentice, 1996a)-and the generalized model is built on the assumption of optimal nitrogen (N) allocation in the vegetation canopy (Haxeltine and Prentice, 1996a; Haxeltine and Prentice, 1996b) (Sitch et al., 2003). Daily net photosynthesis is estimated using a standard nonrectangular hyperbola formulation, which gives a gradual transition between the PAR-limited  $(J_E)$  and the Rubisco-limited  $(J_C)$  rates of assimilation –(Haxeltine and Prentice, 1996b). For  $C_3$  plants,  $J_E$  is a function of the canopy absorbed PAR, the intrinsic quantum efficiency for CO<sub>2</sub> uptake ( $\alpha_{c3}$ ), the CO<sub>2</sub> compensation point ( $\Gamma^*$ ) and the internal partial pressure of CO<sub>2</sub> ( $p_i$ ) (Collatz et al., 1991; Haxeltine and Prentice, 1996b).  $J_C$  is related to the maximum catalytic capacity of Rubisco per unit leaf area (Vm),  $\Gamma^*$ ,  $p_i$  and the Michaelis-Menten constant for CO<sub>2</sub> (K<sub>C</sub>)-and O<sub>2</sub>-(K<sub>O</sub>). Stomatal conductance influences the intercellular  $CO_2$ ,  $p_i$  as well as canopy transpiration.
  - 2.2.2 BVOC modelling

In LPJ-GUESS, isoprene (Arneth et al., 2007) and monoterpene (Schurgers et al., 2009) emissions are simulated as a function of the photosynthetic electron flux-used for photosynthesis. The productions  $(\underline{r})$ -of isoprene  $(\underline{E_l})$  and monoterpenes  $(\underline{E_m})$  are computed as:

$$E = \alpha I \varepsilon, \text{ where } \alpha = \frac{p_i - \Gamma^*}{6 \times (4.67 p_i + 9.33 \Gamma^*)}$$
(1)

5 where *J* is the rate of photosynthetic electron transport, <u>and</u>  $\alpha$  converts from photon fluxes into terpenoid units and *C<sub>r</sub>* is the leaf internal CO<sub>2</sub>-concentration without water stress. The synthesis of both compounds is linked to <u>*J*</u> the photosynthetic electron transport (Niinemets et al., 1999; Niinemets et al., 2002); and <u>-a</u>A fraction ( $\varepsilon$ ) of the electron transport contributing to terpenoid productions (Eq. 2) is determined from a plant-specific fraction under standard conditions ( $\varepsilon_s$ , usually at a T of 30 °C and a PAR of 1000 µmol m<sup>-2</sup> s<sup>-1</sup>) which is adjusted for leaf T, <u>seasonality leaf phenology</u> ( $\sigma$ ), and atmospheric CO<sub>2</sub> concentration:

$$\boldsymbol{\varepsilon} = f(T)f(\boldsymbol{\sigma})f(\mathrm{CO}_2)\boldsymbol{\varepsilon}_{\mathcal{S}}$$
<sup>(2)</sup>

The standard fraction  $\varepsilon_s$  is computed from the often reported standard emission rate (emission capacity) together with the simultaneously estimated photosynthetic electron flux under these standard conditions <u>(standard T and PAR)</u> in the model. The choice of different T and PAR as standard conditions will influence the value for  $\varepsilon_s$ , and then the estimated emission rate at different conditions. The T response corrects for the T optimum from for terpenoid synthesis, which is higher than that for photosynthesis:

$$f(T) = e^{\alpha_r(T - T_s)} \tag{3}$$

The parameter  $a_r$  represents the T sensitivity and <u>standard temperature ( $T_s$ )</u> is often 30 <u>°</u><sup>e</sup>C <u>(adjusted to 20 °C in this study)</u>. In the model, daily mean T (model input) has been adjusted to daylight hours T based on daylength as well as daily T range (Arneth et al., 2007) and the daytime T is used for calculating daily emission rates. However, <u>F</u>for the study in the Subarctic, the <u>often</u>-used reference  $T_s$  of 30 °C as standard conditions as well as and the T responses ( $a_r$ ) were adjusted based on the observation data and will be discussed below. The seasonality function,  $f(\sigma)$ , only applies to isoprene production and is <u>based</u> on a degree-day method in Spring, T and daylength thresholds in Autumn <u>based</u> on the work by (Arneth et al., 2007; Schurgers et al., 2009) which was later modified by Schurgers et al. (2011). The atmospheric CO<sub>2</sub> concentration enhances terpenoid synthesis when the concentration is lower than ambient, and vice versa, which <u>are is</u> represented by the function  $f(CO_2)$  (Arneth et al., 2007). The model assumes that both isoprene and monoterpenes are produced in the same pathway and that they respond to CO<sub>2</sub> concentration in the same way.

For monoterpenes, a storage pool (*m*) is assigned to represent the specific (long-term) storage of monoterpenes within a leaf (Schurgers et al., 2009). The storage pool is only implemented for coniferous and herbaceous PFTs (see Table S1). The emission of monoterpenes from the storage ( $\underline{E}_{MS}$ ) is a function of T and *m* with an average residence time ( $\tau$ ).  $\underline{\tau} \underline{\tau}_{S^{\sigma}}$  is the residence time at the standard T of 30 °C (adjusted to 20 °C in this study, consistent with the modification on the T responses

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of terpenoid synthesis). The residence time  $\tau$  is adjusted based on the standard condition  $\tau_{\underline{S}}$  for T responses with a  $Q_{10}$ -relationship.

$$E_{Ms} = m / \tau$$

$$\tau = \frac{\tau_s}{Q_{10}^{(T-T_s)/10}}$$
(4)

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In LPJ-GUESS, the BVOC response to light resides in the photosynthesis processes (light-dependence of *J* in Eq. 1). Additionally, considering the high sensitivity of BVOC production to leaf T, the model applies a computation of leaf T based on air T and energy balance constraints (Arneth et al., 2007; Schurgers et al., 2009). further extended the computation of leaf T from air T to model BVOC emission. The calculation of leaf T in the model was based on solving leaf energy balance, where the incoming shortwave and longwave radiation is balanced by the outgoing longwave radiation and sensible heat fluxes as well as latent heat loss. The existing leaf energy balance equations appeared to underestimate the incoming longwave radiation relative to clear-sky condition (Sedlar and Hock, 2009). The estimated leaf T, rather than air T, was used for both photosynthesis and BVOC synthesis. Water loss (latent heat fluxes) is regulated by stomatal conductance and soil water content, which is also linked to leaf T estimation in the model. For this study, the representation of incoming longwave radiation was modified by specifically considering the cloud emission of longwave radiation relative to clear sky condition (Sedlar and Hock, 2009).

#### 2.3 Simulation setup

#### 2.3.1 Input data

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The daily climate data of air T, air T range and precipitation for the period 1984-2012 (Callaghan et al., 2013; Tang et al., 2014) were provided by the Abisko scientific research station (Abisko Naturvetenskapliga Station, ANS). Four gaps in daily radiation data from ANS (during the periods of 1984/01/01-1984.06-30/06/1984, 2006/06/09/06-2006/06/16/06/2016, 2007/02/13/02-2007/02/15/02/2007, 2011/07/23/07-2011/08/17/08/2011) were filled by-with the Princeton reanalysis dataset (Sheffield et al., 2006) for the grid cell nearest Abisko. The annual CO<sub>2</sub> concentrations for the whole study period (1984-2012) were obtained from McGuire et al. (2001) and TRENDS (http://cdiac.esd.ornl.gov/trends/co2/contents.htm).

#### 2.3.2 Plant functional types

25 The dominant plant species from the observations (Valolahti et al., 2015) were divided into 7 PFTs (Table 1). The PFT parameters (see Table S1) were mainly derived from previous studies for the arctic region using LPJ-GUESS (Wolf et al., 2008; Miller and Smith, 2012; Tang et al., 2015), but they also extend the arctic PFT lists were extended to consider BVOC emission characteristics. The low summergreen shrubs (LSS) were divided into a *Salix*-type (SLSS; high isoprene emitter) and a non-*Salix*-type (NSLSS; e.g., *Betula nana*-dominance, predominantly monoterpenes rather than isoprene emitters)

(Schollert et al., 2014; Vedel-Petersen et al., 2015). Furthermore, due to the abundance of prostrate dwarf shrubs (PDS) in the study area, distinguishing PDS (canopy height lower than 20 cm) from low shrubs (canopy height lower than 50 cm) was implemented through adjusting parameters controlling vegetation height. The PDS-type was further divided into two PFTs with evergreen and deciduous phenology. Moss, widely appearinged in the study area, was not distinguished from forbs and lichens, due to limited data for parameterizing moss physiognomic features and their preferable growing conditions.

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In LPJ-GUESS, the <u>crown canopy</u> of each tree is divided into thin layers (original value is 1.0 m in a forest canopy) in order to integrate PAR received by each tree. The <u>depth-thickness</u> of this layer was reduced to 10 cm in this study to better capture the vertical profile of low and prostrate shrubs. In addition, the original specific leaf area (SLA, m<sup>2</sup> kg C<sup>-1</sup>) values in LPJ-GUESS were estimated based on a fixed dependency on leaf longevity (Reich et al., 1997). In our study, a fixed SLA was assigned to each PFT (Oberbauer and Oechel, 1989) to improve the simulated leaf area index (LAI) for arctic plants. Emission capacities for the PFTs were determined from available leaf-level measurement data from the Subarctic and Arctic.

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# The details about the data sources for parameterizing emission capacity at 30 °C ( $I_{E_{ISS30}}$ ) and 20 °C ( $E_{ISS20}$ ) can be found in Table S2 and the averaged emission capacities (among all literature data in Table S2) for each PFT as well as the representative plant species can be found in Table 1. The emission rates from the literature are generally provided as standardized emission capacities at 30 °C using G93 algorithm and these values were further rescaled to 20 °C using the adjusted T response curve from this study (Fig. 1).

#### 2.3.3 Model calibration and validation

The modelled CO<sub>2</sub> fluxes, LAI as well as the BVOC T response were firstly calibrated before evaluating the modelled daily BVOC emission rates. Two (from the year 2006 and 2007)-out of four years' (2006 and 2007) elosed chamber measured net ecosystem production (NEP), ecosystem respiration (ER) and the estimated gross primary production (GPP) as well as point intercept-based species composition were used for calibrating. T-and the data for the -otherrest two years' data- (from the year-2010 and 2012) were used for validating -the simulated carbon cycle processes. Previous studies focusing on light responses of NEP for arctic plants (Shaver et al., 2013; Mbufong et al., 2014) have reported relatively low quantum efficiencies ( $a_{c3}$ ) caused by overall low sun angle conditions and low leaf area. A thorough sensitivity study of parameters used in LPJ-GUESS (Pappas et al., 2013) has found that  $a_{c3}$  is the most influential parameter in terms of the simulated vegetation carbon fluxes. Also, a pre-validation of the modelled CO<sub>2</sub> fluxes with the observations in this study using the default  $a_{c3}$  value (0.08) has found a large overestimation of both GPP and ER (not shown). Therefore, a sampling of  $a_{c3}$ (using the range of 0.02 to 0.125 µmol CO<sub>2</sub> µmol photons<sup>-1</sup>, proposed by Pappas et al. (2013)) was conducted to find the best value to depict the observed GPP, ER and LAI of the years 2006 and 2007 for the subarctic ecosystem (Fig. S1). After calibration, the model was evaluated The evaluation of the-calibrated  $a_{c3}$  onwith the simulated CO<sub>2</sub> fluxes and vegetation composition-was conducted using the observed CO<sub>2</sub> fluxes and the point intercept-based plant coverage data from 2010 and 2012, respectively.

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The daytime air T in the study area is often below 20 °C (Ekberg et al., 2009), and standardization of terpenoid emissions to 20 °C, instead of 30 °C, has been suggested for modelling in boreal and arctic ecosystems (Holst et al., 2011, Ekberg et al., 2009) due to plants<sup>2</sup> adaptation to low T environment. In the model, the photosynthetic electron fluxes under standardized conditions are simulated in order to convert the input emission capacity to the standard fraction ( $\varepsilon_s$ , see Eq. 2). The choice of

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the standardized T (used in Eq. 3 as well as in estimating photosynthesis rates at this T) will influence the estimated fraction of electron fluxes for BVOC synthesis. In this study, a data fitting to the suggested standard T of 20 °C was conducted using the observed ecosystem-level isoprene emission rates in July together with measurement chamber air T from the control-C plots. The observations were mostly conducted during daytime with relatively high PAR values, and therefore the response of the emission rates to light was not specifically considered in the current data fitting. Potential feedbacks from the 10 variations in the atmospheric CO<sub>2</sub> concentration of emissions were ignored for the three years with isoprene sampling (a) rough model estimation of ~3% reduction in emissions between 2006 and 2012). The data collected from different blocks were separated for the curve fitting and the parameters controlling T response ( $\alpha_{\tau}$  in Eq. 3) were determined (Fig. 1). -An adjusted  $\alpha_{\tau}$  value of 0.23 was chosen after fitting all the data from July over three years' measurements. Apart from the low  $R^2$  value for block 1, the data were well captured by the exponential shape ( $R^2 \ge 0.8$ ) of the T response curve. The calibrated T responses were used for standardizing leaf-level emission rates (see  $M_{IS20}$ , Table 1) as well as estimating 15 emission rates in the model.

After calibrating the modelled carbon cycle processes as well as the  $\alpha_{r}$  value for BVOC T response, Tthe vegetation abundance of each PFT composition was evaluated using simulated LAI against the point intercepted-based species composition. The species were grouped into the corresponding PFTs for comparingison and the point intercept-based hits within the same PFT group were summed. The summed hits were divided with 100 pin hits to compare with the modelled LAI. After <u>calibrating</u> the modelled CO<sub>2</sub> fluxes and LAI, the modelled isoprene and monoterpene emission rates were compared with the observations. The simulated emissions, represented as daily daytime average values ( $\mu$ g C m<sup>-2</sup> h<sup>-1</sup>, daytimeily emission rates divided by day length) may not allow accurate comparisons with the actual emission rates which were typically conducted in the middle of the day (between  $\frac{10.9}{10.9}$  am - 52 pm). Therefore, the emission rates at noon (maximum hourly emissions) were also estimated by an additional computation of the emission, applying the leaf T computed from the daily maximum air T for photosynthesis and BVOC emissions. Due to- the daily process used in the model (Sitch et al., 2003), it is not possible to compute an instantaneous photosynthesis flux at noon. Therefore, the daily average PAR was used for estimating the emission rates at noon. Both daily noon and daytime average rates were shown in order to present the range (instead of direct comparison with the observed emission at each specific hour) of the modelled daily emissions relative to the observed emissions, We expect the emission rates at noon to be more comparable with the observations than the daily mean emissions, considering diurnal dynamics of emissions (Lindwall et al., 2015).

The model's performance in modelling BVOC emissions was evaluated by Willmott's index of agreement (A) (Eq. 5) and mean bias error (B) (Eq. 6). The index A describes the agreement between the modelled fluxes ( $E_i$ ) with the observed ( $O_i$ )

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and a value close to 1 indicates a good agreement. The index B estimates the mean deviation between the modelled and <u>observed values</u> (Willmott et al., 1985) and values close to 0 indicates models' good agreement to observations.

$$A = 1 - \frac{\sum_{i=1}^{N} |E_i - O_i|}{\sum_{i=1}^{N} (|E_i - \overline{O}| + |O_i - \overline{O}|)}$$
(Eq. 5)  
$$B = \frac{\sum_{i=1}^{N} (E_i - O_i)}{N}$$
(Eq. 6)

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where  $\overline{O}$  is the observed mean value, N is total number of data records.

#### 2.3.4 Effect of wWarming experiment

To simulate the observed warming responses from the OTCs, <u>a</u> warming of 2 °C was imposed in the model for the growing season (the period with OTC warming) (Tiiva et al., 2008; Valolahti et al., 2015). The modelled warming responses (WR, <u>emission differences between C and W treatments</u>) using the original T response ( $\alpha_{\tau} = 0.1$ ,  $T_{S} = 30$  °C,  $E_{I30}$  and  $E_{M30}$  °C, Eq. 3) and the adjusted T response ( $\alpha_{\tau} = 0.23$ ,  $T_{S} = 20$  °C,  $E_{I20}$  and  $E_{M20}$  Eq. 3 and Fig. 1) were compared with the observed WR. Furthermore, <u>apart from the settings with 2 °C warming</u>, additional simulations with a warming <u>byof</u> 4 °C and 8 °C, reflecting the range of climatic projections in this region (IPCC, 2013), were also conducted to test<u>for</u> the anticipated ecosystem-scale responses <del>of to</del> different levels of warming.

#### **3 Results**

#### 15 **3.1 Modelled CO<sub>2</sub> fluxes and vegetation composition**

The simulated ecosystem CO<sub>2</sub> fluxes and LAI were sensitive to the parameter value chosen for The parameter  $a_{c3}$ , which describes determining the efficiency in converting solar radiation to earboncarbohydrates, and which varied between 0.02 to 0.125 µmol CO<sub>2</sub> µmol photons<sup>-1</sup> following Pappas et al. (2013), showed strong impacts on the modelled ecosystem CO<sub>2</sub> fluxes and LAI for the growing seasons 2006 and 2007 (Fig. S1). For CO<sub>2</sub> fluxes, the lowest root mean square error (RMSE) values occurred at 0.035 µmol CO<sub>2</sub> µmol photons<sup>-1</sup> for GPP and ER, while the lowest RMSE value for LAI was 0.051 µmol CO<sub>2</sub> µmol photons<sup>-1</sup> when comparing with the observations for 2006 and 2007. A value of 0.04, consistent with the study by Shaver et al. (2013) was selected for  $a_{c3}$  to limit the RMSE values of both the modelled CO<sub>2</sub> fluxes and LAI. Using this value for  $a_{c3}$  calibrated with observation data from 2006 and 2007, the model captured the observed day-to-day variations as well as the magnitudes of the chamber-based GPP, ER and NEP from for 2010 and 2012, though with an slight overestimation of CO<sub>2</sub> fluxes (particularly for the early growing seasons, Fig. 2), and an underestimation of LAI (Fig. 3). For

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the year 2012, the model showed large overestimations of the observed GPP and ER for the limited number of measurements in this growing season.

The details about the representative species for each PFT can be found in Table 1. For the 5 <u>PFT</u> groups of PFTs, the modelled growing season LAI values for the year 2010 and 2012 were generally lower than the point intercept-based coverage estimations from the field observations (note different left and right ax<u>ies</u> scales in Fig. 3), except for the *Salix*-type summergreen shrubs and deciduous prostrate dwarf shrubs (SLSS+SPDS). The two-most dominant vegetation groups in the C plots, forbs/lichens and evergreen shrubs, were captured by the model. However, the large coverage of graminoids (GRT) and non-*Salix*-type deciduous shrubs (NSLSS) was largely underestimated by our model.

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In response to 2 °C warming, the modelled LAI for the shrub PFTs (SLSS+SPDS, NSLSS, LSE+EPDS) showed an increase, while the modelled LAI for graminoids and forbs/lichens largely decreased. For the two groups of shrubs (NSLSS and LSE+EPDS), the modelled increase is in agreement with the observations. Based on the observations, vegetation changes have been to the same directions for two groups of shrubs (NSLSS and LSE+EPDS). However, an the observed large increase of the coverage of forbs/lichens as well as a decreased coverage of graminoids in the W treatments for the year 2010 and 2012 were not captured by the model.

#### 15 3.2 Modelled BVOC emissions

#### **3.2.1 Seasonal variations**

BVOC emissions are closely linked to leaf as well as ecosystem development.s and sSimulating dynamic vegetation seasonal variations in leaf area and vegetation composition enables us to assess the model's performance in representing short-term emission changes in response to T and PAR, as well as long-term changes in vegetation development and distribution. The seasonal variations of the modelled BVOC emissions as well as the span of all BVOC samplings over four years are presented in Fig. S2. The span of the BVOC measurements/samplings covered the main growing seasons over four years. The modelled emission rates in the C plots showed pronounced day to day as well as seasonal variations (Fig. 4). The modelled emissions of isoprene and monoterpenes were low in the spring and autumn, and peaked on warm days during the summer. The day to day variations in the emissions agreed well with the variations of T and PAR. When both T and PAR were high through the growing season, the peaks of both isoprene and monoterpene emissions occurred and the deviations between the modelled daily average and noon emissions became larger. The observed magnitude of isoprene emissions during daytime showed large spatial variations between the blocks for the days with the observed high average emission rates (blue error bars in Fig. 4) and the observed average rates (blue squares) were well captured by the modelled noon emissions. For monoterpene emissions, the modelled daily average was closer to the observation, especially for the years with generally low emissions (2006, 2010 and 2012). The observed high monoterpene emissions for a few days were relatively better captured by the modelled noon emission. The emissions of monoterpenes remained more constant towards the end of the growing season (not fully represented here).

#### 3.2.12 Daily emissions

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#### • Emission rates at in the control (ambient) conditions

The observed daily variations in isoprene emissions were mostly-generally\_captured by the model (Fig. 4, Fig. 45). The observed isoprene emission rates (Fig. 45a) lay between the modelled daily daytime\_average and daily noon emission rates, with the exception of a few days with much lower higher observed simulated noon emission rates from the observations than the observed modelled noon emissions (22/08/-August 2006, 10/07/-July 2007 and 05/08/-August 2007). For these dates, the observed chamber air T were higher than the modelled daily noon leaf T (squares in Fig. 45c) and were also higher than or close to 20 °C. For 06 July 2007, when the simulated noon leaf T was higher than 20 °C, the model captured the observed high emissions well. NoticeablyNotably, the model used air T at 2 m height from the ANS station to extrapolate the leaf T, while the measured T is the air T inside the measurement chamber. Over three growing seasons, the observed air T inside the modelled daily-daytime\_average leaf T and 3.4 °C warmer than the modelled daily noon and the observed daytime emission rates were 9.1, 25.8 and 25.5 µg C m<sup>-2</sup> h<sup>-1</sup>, respectively (all numbers averaged for the days on which measurements were made) and the modelled daily noon isoprene emission rates demonstrated better representation (A = 1.02 and B = -1.28) of the observed daytime emission rates than the daily average (A = 1.37 and B = -19.05).

- For monoterpenes, the modelled da<u>ytimeily</u> average emission rates in the C plots (light grey bars in Fig. <u>56a</u>) showed relatively-closer values to the observations (A = 1.07 and B = -0.36), compared to the modelled noon emission rates (A = 0.47 and B = 5.09) (dark grey bars in Fig. <u>56a</u>). Over four sampling growing seasons-, the modelled da<u>ytimeily</u> mean, daily noon and observed daytime emission rates were 2.4, 7.9 and 2.5  $\mu$ g C m<sup>-2</sup> h<sup>-1</sup>, respectively (all numbers averaged for the days on which measurements were made). The modelled da<u>ytimeily</u> mean showed better agreements with the observed low monoterpene emissions for 2006, 2010 and 2012, but underestimated the observed high emission rates for the year 2007. In 2007, the highest emission rates observed on 06 July were not captured by the modelled da<u>ytimeily</u> average, but were of similar magnitude as the modelled daily noon emission rates (with certain overestimations). Whereas the observed emissions showed great variations between years (1.3, 8.1, 0.3, 0.5  $\mu$ g C m<sup>-2</sup> h<sup>-1</sup>, for the four years measured, respectively), the
- simulated daily noon emissions were more similar between years (2.2, 3.0, 2.1 and 2.4  $\mu$ g C m<sup>-2</sup> h<sup>-1</sup>, respectively).

Emission responses to 2 °C warming

In response to 2 °C warming, the modelled leaf T increased on average by 2 °C, while the observed chamber air T in the W plots increased by 1.8 °C relative to the C plots on an average of averaged of the four growing seasons with observations. For isoprene, the modelled WR (Fig. 45b) were relatively generally lower than the observed WR, especially for a few days with strong observed WR. Averaging over three years, the modelled da<u>ytime</u>ily average, noon and the observed WR were 5.7, 15.2 and 29.3  $\mu$ g C m<sup>-2</sup> h<sup>-1</sup>, respectively and warming increased the modelled da<u>ytime</u>ily average isoprene emission rates by 63 %, the daily noon by 59 % and the observed emissions by 115 % (all numbers averaged for the days on which measurements were made). Over three years, the observed strong WR for a few dates (e.g., 22/08/-August-2006, 10/07/-July

2007, 05/08/-August 2007 and 14/06/-June-2012) were underestimated by the modelled noon WR when the observed chamber air T in the C was close to or higher than 20 °C, but the modelled leaf noon T was below this level. However, for the day when both daily leaf T and chamber air T were over 20 °C (e.g., 13 June 2006, 06 July 2007), the observed WR were relatively higher than the modelled daily average, but lower than the modelled daily noon WR.

For monoterpenes, the modelled dailytime average, noon and the observed WR were 2.0, 6.0 and 2.5 µg C m<sup>-2</sup> h<sup>-1</sup>, 5 respectively. The averaged WR from the modelled noon emissions were much higher than the observations. The modelled daily daytime average WR showed better agreement with the observations. For one day with extremely high WR (06 July 2007), the modelled noon WR better captured the strong responses. Averaging over four growing seasons, warming increased the modelled daily average monoterpene emissions by 81 %, the daily noon emission by 76 % and the observed 10 emissions by 98 %.

The modelled daily noon WR using the adjusted BVOC T response ( $\alpha_r = 0.23$ ,  $T_s = 20$  °C, Eq. 3) were further compared with the simulation using the original T response ( $\alpha_t = 0.1, T_s = 30$  °C, Eq. 3). For isoprene (Fig. <u>67</u>a), the simulation using the adjusted T response showed a substantial increase of the modelled WR as well as a better agreement to the observations (A =1.24, B = -11.85) than the simulation using the original T response (A = 1.47, B = -25.98). The modelled WR using the

- 15 original T response generally underestimated showed limitation in capturing the the observed high WR. Averaging through three years, the modelled isoprene WR using the original T response only gave represented 11 % of the observed WR, while the modelled WR using the new T response captured 52 % of the observed WR. For monoterpenes, the modelled WR using the original T response showed relatively closer values to the observations for the years with the observed low WR (2006, 2010 and 2012). For the year 2007, the observed high monoterpene WR were better captured by the simulated WR with the
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new T response. For both T responses, the modelled WR wereis generally lower than the observed WR for isoprene, but higher than the observed WR for monoterpenes.

#### 3.2.3-2 Annual emissions

A comparison of the simulated annual BVOC emissions from the C and W treatments demonstrated that the 2 °C warming during the growing seasons increased both isoprene and monoterpene annual emissions. Averaging over 13 years, the warming by 2<sup>-o</sup>C during the growing seasons this temperature increase increased annual isoprene and monoterpene emissions by 55 % and 57 %, respectively (p < 0.01, Mann-Whitney test). The modelled emissions showed strong inter-annual variations in response to warming (Fig. 78). For the warmest year (2011), the W treatment increased annual isoprene and monoterpene emissions by 99 % and 94 %, respectively. The mean annual isoprene and monoterpene emissions in the C for 1999-2012 were 20 and 8 mg C m<sup>-2</sup> yr<sup>-1</sup>, respectively. For the four years with BVOC sampling, the modelled average WR were 58 % and 70 % for annual isoprene and monoterpene emissions, respectively. The modelled annual WR at the annual scale were at of the similar magnitudes as the modelled daily average WR for the days with BVOC samplings (63 % for isoprene and 81 % for monoterpenes).

The simulations imposing the warming by 4 °C or 8 °C during the same period as the 2 °C warming increased annual isoprene emissions by 120 % and 247 %, respectively (p < 0.01, Mann-Whitney test) and annual monoterpene emissions by 87 % and 167 %, respectively (p < 0.01, Mann-Whitney test). For isoprene, the strongest WR of all levels of warming appeared in 2011. Higher levels of warming further elevated isoprene emissions for all years, but the impact on monoterpene emissions levelled off for due to a decreasinged coverage of evergreen prostrate dwarf shrubs (EPDS) with 8 °C warming. The decrease in coverage of EPDS only occurred for the last few years with 4 °C warming. The different levels of warming generally increased shrub growths, but largely decreased the coverage of forbs/lichens and graminoids (CLM and GRT) (data not presented). At annual scale, the long-term vegetation changes associated with warming by 4 °C or 8 °C showed strong impacts on BVOC emissions.

#### 10 4 Discussion

#### 4.1 Emission rates

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The modelled day-to-day variations of ecosystem  $CO_2$  fluxes (Fig. 2) and BVOC emissions generally followed the observationswas able to reproduce vegetation CO<sub>2</sub> fluxes (Fig. 2) as well as the main dynamics of isoprene and monoterpene emissions (Fig. 4), in spite of deficiencies in the representation of the observed vegetation composition (Fig. 3). The mismatch between the modelled LAI and the point intercepted-based vegetation coverage may be caused by an underestimation of the allocation of assimilated carbon to foliage in LPJ-GUESS and/or too low SLA values (Table S1). In LPJ-GUESS, the carbon allocation among different living tissues follows four allometric equations (see Eqs. 1-4 in Sitch et al. (2003))-to control the structural development of each modelled plant individual (see Eqs. 1-4 in Sitch et al. (2003)). The allometric parameters for some of the arctic PFTs used in this study were validated by Wolf et al. (2008) derived for a model applying a quantum efficiency  $\alpha_{c3}$  of 0.08 at the regional scale, which may require further justification after the reduction in  $\alpha_{c3}$  that was applied here to match the observed daily CO<sub>2</sub> fluxes. The reduced quantum efficiencies reflect the growth environment with low T as well as and low sun angle in high latitudes (Shaver et al., 2013), but more observations are still needed to better quantify light use efficiency of arctic plants (Dietze et al., 2014). Furthermore, Van Wijk et al. (2005) found a close linkage between total foliar nitrogen (N) content and LAI for arctic plants, which was further supported by Campioli et al. (2009) for an arctic ecosystem dominated by *Cassiope tetragona*. However, the current simulations neither include C-N interactions -nor consider potential impacts of N limitations on plants developments (Smith et al., 2014), which need to be improved in future model simulations in this region (Michelsen et al., 2012). The subdivision of arctic PFTs into smaller groups to specifically consider isoprene and monoterpene emission features was has shown the importance to be important for capturing the emission dynamics in this heath tundra ecosystem.  $\frac{1}{2}$  but tThe development of parameterizations for arctic PFTs also poses challenges torequires considering the phenological and physiognomic features of mosses (currently aggregated in the CLM-type PFT, Table S1), which may bring additional uncertainties to the modelled LAI. At the same time, tThe current evaluation of the modelled LAI with the point intercept-based measurements of plant coverage cannot

disregard uncertainties from the field method itself, such as subjective judgement of species from each hit, and the potential influences from hits on stems as well as sampling inclining angles (Wilson, 2011). Also, the seasonal variation in leaf development as well as the randomly selected blocks from the heterogeneous landscape may further complicate the comparison of the simulated LAI with the local observations. Capturing the start of the growing season in the model is also

5 crucial for depicting the dynamics of seasonal  $CO_2$  fluxes (Tang et al., 2015). The overestimated GPP in the beginning of growing seasons (Fig. 2a) suggests uncertainties in modelling the time of this-its start. The current algorithm for detecting start of growing season in large scale applications (Sykes et al., 1996) may not be sensitive enough for prediction of budburst of arctic plants (Pop et al., 2000).

The modelled annual isoprene and monoterpene emissions, 20 and 8 mg C m<sup>-2</sup> yr<sup>-1</sup> for 1999-2012, correspond to less than

- 10 0.1 % of the modelled GPP. The modelled emission rates are not only linked to the modelled photosynthesis fluxes, but also to determined by the emission capacity assigned for to each PFT (see Tables 1 and S2). For some PFTs (e.g., the Salix-type and prostrate summergreen shrubs, SLSS and SPDS), the emission capacities in Table 1 are of similar magnitude as observed values that are applied in large-scale models for boreal forests (see Table 2 in Rinne et al. (2009)). The relatively low-observed relatively low emissions in comparison with lower latitudes (Arneth et al., 2011; Sindelarova et al., 2014) are 15 mainly caused by low T and plant biomass, and not by low emission capacities (Holst et al., 2010). Notably, t
- The numbers for the estimated annual emissions are still highly uncertain, considering the dissimilarities to the observations in the modelled LAI as well as early season CO<sub>2</sub> fluxes. Furthermore, the T response in this study (Fig. 1) may be more robust for isoprene than monoterpenes, which may also contribute to the uncertainties in annual emissions. Also, there are more studies about  $CO_2$  inhibition on isoprene emissions (Arneth et al., 2007), but less on monoterpenes. Generally, the
- emission responses of monoterpene could become less clearer than isoprene due to potential emissions from storage pools (Peñuelas and Staudt, 2010). Therefore, more laboratory experiments in controlled conditions testing BVOC responses (especially monoterpenes) of arctic plants to different environmental variables could largely reduce the abovementioned uncertainties. Based on the current estimation, the relative magnitude of isoprene and monoterpene emissions from this site may not contribute significantly to the global number. However, the highly reactive compounds emitted by plants could
- 25 undergo chemical reactions in the local/regional atmosphere and provide feedbacks to the climate. Furthermore, the warming-induced strong increase of emissions could indicate an increasing role of BVOC in the local atmospheric chemistry and also global emission magnitudes for future conditions.

Relative to isoprene emission, the magnitude of monoterpene emissions was much lower since the species in the study area were mostly considered to be isoprene emitters (Tiiva et al., 2008; Faubert et al., 2010). The observed monoterpene

emissions were generally low for the sampling days (see Fig. S2), and the validation of the modelled emissions with these low rates could indicate that the modelled outputs were only validated for low emission rates, but was generally lack of evaluation of potentially high emission rates. On one hand, tThe model showed certain limitations in representing the observed low monoterpene emission rates (mainly for the year 2010 and 2012), which could be attributed to the prescribed value for splitting the produced monoterpenes into direct emissions (50 %) and emissions from storage pools (50 %)

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(Schurgers et al., 2009). This split determines the distribution of monoterpene emissions over the year, since and an allocation of the monoterpenes into storage pools results in a more gradual distribution of emissions. At the same time, the overestimated monoterpene emissions during the evaluationng periods (mainly growing seasons) may also indicate that the implemented storage residence time is too short (maybe larger storage size). The adjusted temperature response may not be

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equally accurate for monoterpenes, and/or the temperature dependence of monoterpene emissions from the storage pool (Eq. 4) is too strong for arctic plants. This may be due to leaf anatomy specialized to the arctic conditions (Schollert et al., 2015). On the other hand, the push-pull enclosure technique used for BVOC emission measurements can also bring uncertainties to the measurement data: the choice of sampling time and flow rates influences temperature and humidity inside the enclosure and this, as well as potential gas concentration changes within the enclosure, may impact the plant physiological status. The

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impacts also depend on the ecosystem emission rate (Niinemets et al., 2011). The current observations of BVOC emissions only covered the main growing season. Sampling over a longer season (Holst et al., 2010) could help to improve the parameterizationing of this the portioning partitioning over direct emission and storage, as well as the T response of emission rates from storage pools. Furthermore, ongoing <sup>13</sup>C labeling experiment focusing on arctic mesocosm (Lindwall, I-Ghirardo et al., unpublished data) could also help to identify the fractions of monoterpene emissions from production or storage. 15 FinallyAdditionally, evaluating the modelled daily emission rates with the field observed emission rates were measured at eertain different time points periods of a day and the evaluation of daily emission with these time points observations cannot avoid potential impacts from and are thus influenced by the diurnal dynamics of BVOC emissions, which has been found to be strong in the Arctic (Lindwall et al., 2015).

#### 4.2 Responses to warming

20 The modelled increases of shrub coverage in response to the W treatment mostly followed the field-observations (Valolahti et al., 2015) and are-is consistent with the general trend from other arctic studies-in the Arctic (Wahren et al., 2005; Elmendorf et al., 2012). However, the observed increase of bryophytes is rather site-specific, which was not captured by the model. In contrast, the modelled W-induced decreased coverage of graminoids and forbs/lichens agrees well with the large-scale trend identified by Elmendorf et al. (2012) who conducted a global synthesis of 61 tundra warming experiments. 25 The decreasing soil moisture in W treatments (excluding wet ecosystems) is one of the main constraints on bryophyte coverage (Lang et al., 2012).

Along with vegetation community responses alterations, the short-term T responses of the vegetation are central for accurately depicting daily BVOC emission responses to the warming W treatment. Through enhancing adjusting the BVOC T sensitivity (from  $\alpha_{\tau} = 0.1$ ,  $T_s = 30$  °C to  $\alpha_{\tau} = 0.23$ ,  $T_s = 20$  °C in Fig. 1), the simulated BVOC WR (63 % for isoprene and 81 % for monoterpenes) became comparable to the observed responses (115 % for isoprene and 98 % for monoterpenes). The underestimation of a few days' strong isoprene WR could be partly attributed to the-lower leaf T estimations derived

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from 2 m air T measured at the ANS station, which was lower than the observed daytime chamber air T in the low canopy (Fig. 45c). The low-statured plants in dry to mesic tundra ecosystems (Schollert et al., 2014; Lindwall et al., 2016b) are efficient in absorbing heat and thus prone to have a high canopy T on a sunny day. This can directly elevate BVOC emissions as well as WR (Lindwall et al., 2016a).- Furthermore, for other regions with underlying permafrost (not the case in this study site) in the Arctic, the potentially low ecosystem evapotranspiration can increase both ground and canopy T. Also, plants acclimated to cold environment may drive larger emissions responses once they are exposed to warmer T (Rinnan et

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al., 2014). TOn the other hand, tT he observed strong WR can also be partly due to the potential side effects of the OTCs in the W treatment, e.g., reduced wind speed (De Boeck et al., 2012), drying of the surface soil -and increased frequency of high-temperature events (Bokhorst et al., 2013). At annual to decadal timescales, the warming in the experimental plots caused changes in total plant biomass and species coverage which were found to contribute to the increase in BVOC emissions after 13 years of treatments (Valolahti et al., 2015). These indirect effects on BVOC emissions were not yet 10 identified after 7-8 years of warming for the yearsin 2006 and 2007 (after 7-8 years of treatments) (Tiiva et al., 2008; Faubert et al., 2010), which points outhighlights the importance of accurately representing the temporal dynamics of vegetation as a driver of BVOC emissions. The modelled annual emissions in response to different degrees of warming (Fig. 78) have clearly elucidated the combined effects from of the direct responses to summer warming as well as with the indirect responses from vegetation changes, although the model still has limitations in representing the observed vegetation composition in detail (Fig. 3). Furthermore, these combined effects also suggest a non-linear response of BVOC emissions to

#### different levels of warming.

The adjusted T response curve better represents subarctic plants' isoprene emission responses (mainly isoprene emissions) to warming better than the original curve which has been parameterized for global simulations (Fig. 67). It further supports the earlier suggested stronger T sensitivity of BVOC emissions from arctic plants compared to plants from other regions (Ekberg

- 20 et al., 2009; Holst et al., 2010; Rinnan et al., 2014). The commonly-used T response in Guenther's algorithm (Guenther et al., 1993) is based on the Arrhenius-type dependence of enzyme activities with an optimum T around 40 °C, and the shape of the Guenther's response is very close to the exponential curve with  $\alpha_{\tau}$  value of 0.13 (using standard T of 30 °C) when leaf T is lower than 30 degrees. The high  $\alpha_{\tau}$  value found in this study indicates that a slight T increase during summertime could cause a large increase of isoprene and monoterpene emissions from the studied cold subarctic ecosystem (Faubert et al., 2010;
- 25 Holst et al., 2010). Furthermore, the adjusted T response is based on the data fitting of the observed canopy air T with hourly isoprene emission rates, and this response is used to estimate daytime emissions in the model. The inconsistent temporal resolution calls for further adjustment for this T response for arctic plants.

The pronounced high T responses in this tundra ecosystem further indicates that T responses of BVOC emissions could vary spatially (Niinemets et al., 2010) and points out the importance of applying locally optimized parameters for global estimates (Ekberg et al., 2009).

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#### 4.3 Suggestions for other models and potential measurements further work

For extrapolating the current model developments to large-scale (regional) applications, we suggest to-addressing the following issues: 1) The emission responses to T of arctic plants could be further tested based on laboratory experiments in controlled conditions; 2) The strong decoupling of leaf T from air T and <u>the</u> strong dependence of BVOC emissions on leaf T (Lindwall et al., 2016a) point to a need <u>for accurately</u> capturinge leaf T accurately in models. Long-term parallel observations of both leaf and air T will be useful for the algorithm developments focusing on arctic vegetation (Rinnan et al., 2014); 3) The subdivision of the existing PFTs into groups featuring isoprene and monoterpene emissions are encouraged for other relevant modelling studies (Grote et al., 2014), though-and additional data may be required for characterizing the new subgroups, such as bioclimatic limitations; 4) The potential impacts of seasonal dynamics of vegetation as well as phenology on emission capacities should be further identified with whole-season BVOC sampling (Staudt et al., 2000); 5) <u>Arctic PFT's</u> <u>The</u> responses and/or acclimation <u>of arctic PFTs</u> to warmer climate should be better paramet<u>erizriz</u>ed in the model to-<u>better</u> represent improve the representation of long-term vegetation effects on BVOC emissions.

#### 10 5 Conclusions

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This study has demonstrated the model's ability to depict the observed isoprene and monoterpene emission rates as well as daily variations in the <u>BVOC</u> emission of a subarctic tundra ecosystem. The modelled warming responses using the <u>a</u> adjusted <u>T response</u> curve with adjusted for a stronger <u>BVOC</u>. T response showed good agreements with the observations, especially for the days with the observed strong emission responses to warming. Short-term underestimations were most likely linked to the underestimated leaf T during the daytime. In the long-term (days-years), a mismatch in the modelled vegetation composition could also bring uncertainty in the simulation of emission responses to warming. The model estimated the mean annual isoprene and monoterpene emissions to be 20 and 8 mg C m<sup>-2</sup> yr<sup>-1</sup>, with around 55 % and 57 % increase in annual emissions in response to a 2 °C warming for the period 1999-2012. For the warmest year, the 2 °C warming during the growing season resulted in 99 % and 94 % increase of isoprene and monoterpene emissions. These strong warming responses of arctic BVOC emissions have hitherto not been specifically described in large-scale models and are therefore suggested to be included, especially in estimating regional emissions from the pan-Arctic.

#### Author contribution

J. Tang, G. Schurgers and R. Rinnan designed this research project. J. Tang did simulation runs, model developments and comparisons with the observation. G. Schurgers largely contributed to the research questions, model processes development and calibration. R. Rinnan contributed to the research questions, data collection and project financial supports. H. Valolahti, P. Faubert, P. Tiiva and A. Michelsen provided field data used in this study. J. Tang wrote the manuscript and all authors critically read, commented, corrected and finally approved the manuscript.

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#### **Tables and figures**

Table 1 Plant functional types (PFTs) and representative species in the study area. The emission capacity of isoprene ( $E_{IS}$ ,  $\mu$ g C gdw<sup>-1</sup> h<sup>-1</sup>) and monoterpenes ( $M_S$ ,  $\mu$ g C gdw<sup>-1</sup> h<sup>-1</sup>) at 20 °C (in bold and italics) using the adjusted temperature response curve are presented as  $I_{S20}$ , whilst the averaged literature values based on the Guenther's algorithms with 30 °C as the standard temperature. The values are based on the available growing season leaf-level measurements from the Arctic.

mmunis; Vaccinium
; Salix myrsinites
, Dryas octopetala;
eticulata
a, Carex rupestris,
Festuca ovina, Poa
, Bartsia alpina,
oina, Gymnadenia
pponica, Pinguicula
emorus, Saussurea
ocomium splendens
reberi, Sphagnum
lis, Cladonia spp.

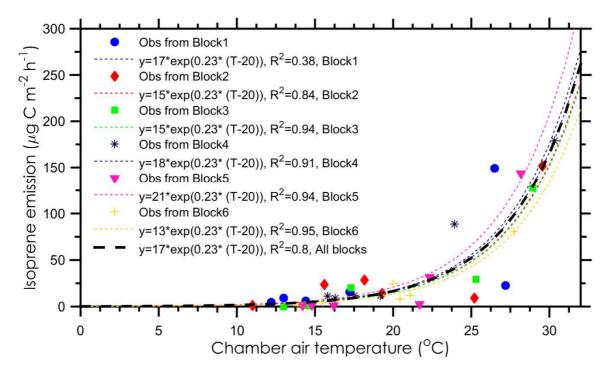


Figure 1 The observed isoprene emission rates in relation to the chamber air temperature in July over three field <del>campaign</del> seasons (2006, 2007, 2012) in the Abisko heath-tundra heath.

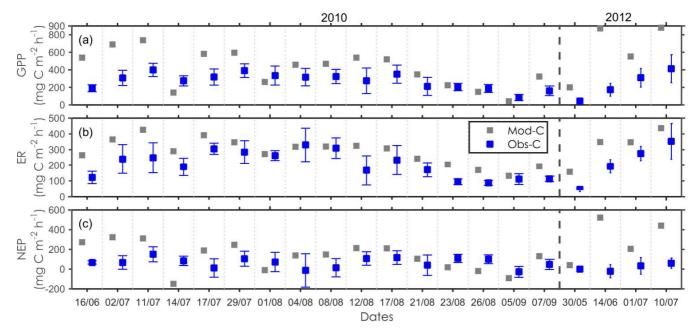


Figure 2 Modelled (grey) and observed (blue) gross primary production (GPP, (a)), ecosystem respiration (ER, (b)), and net ecosystem production (NEP, (c)) for the growing season of 2010 and 2012 in the control plots at the Abisko heath-tundra heath. Error bars indicate the standard deviation for the six replicates

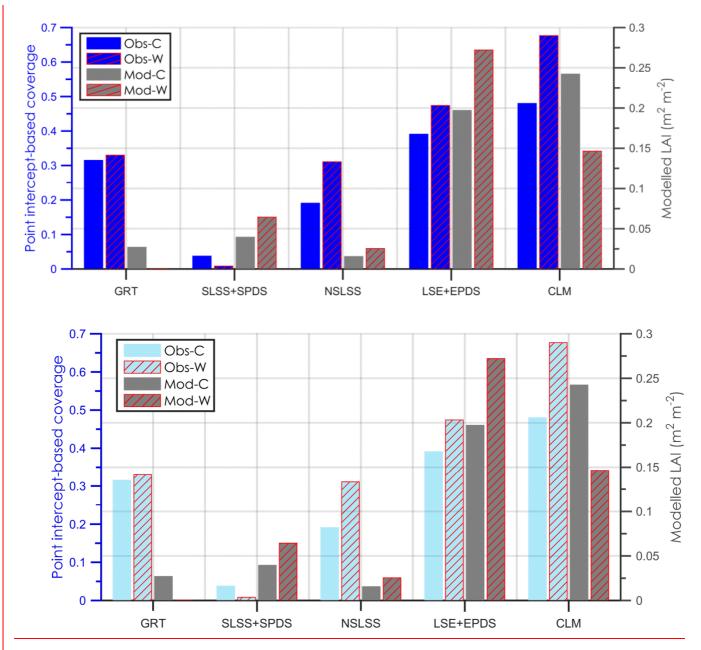


Figure 3 Point-intercept based vegetation coverage (during the growing season) and modelled growing season-leaf area index (LAI, m<sup>2</sup> m<sup>-2</sup>) averaged for the <u>year-growing season</u> 2010 and 2012 for the control (C) and warming (W) treatments in the Abisko heath tundra heath. Different y axes are used for the observed (Obs) and the modelled (Mod) coverage. GRT: Graminoid tundra; SLSS: *Salix*, low shrubs summergreen; SPDS: Summergreen prostrate dwarf shrubs; NSLSS: Non-*Salix*, low shrubs summergreen; LSE: Low shrubs evergreen; EPDS: Evergreen prostrate dwarf shrubs; CLM: Cushion forbs, lichens and moss tundra.

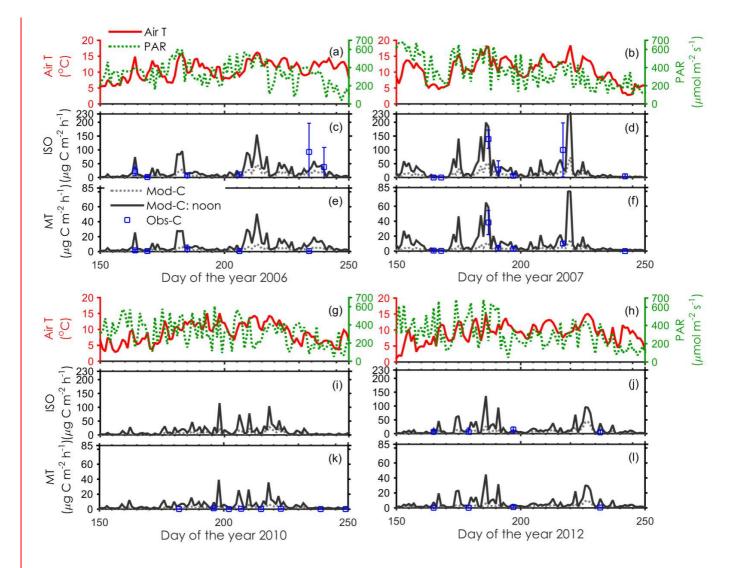
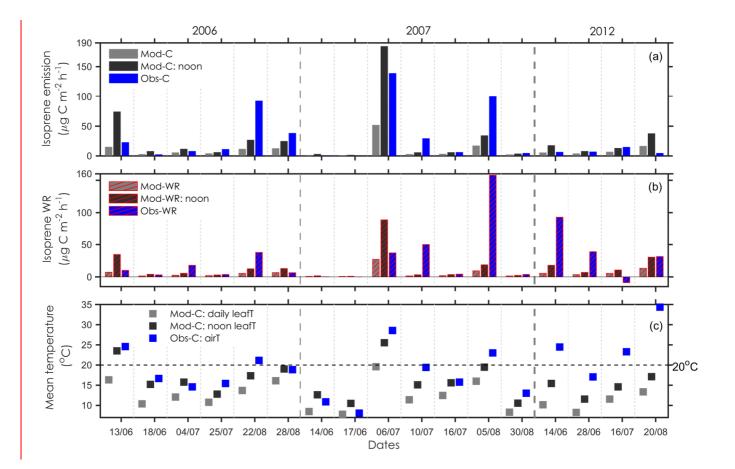


Figure 4 Time-series of the air temperature (Air T) at 2 m height, photosynthetically active radiation (PAR), the modelled isoprene (ISO) and monoterpene emissions (MT) ( $\mu$ g C m<sup>-2</sup> h<sup>-4</sup>) for the days 150-250 in 2006, 2007, 2010 and 2012 in the Abisko heath tundra. Both modelled and observed fluxes are from the control (C) conditions and the modelled daily average (Mod-C) and daily noon (Mod-C: noon) emissions are presented. Error bars indicate the standard deviation for the six replicates. For the year 2010, isoprene emission rates were not analyzed due to technical problems.



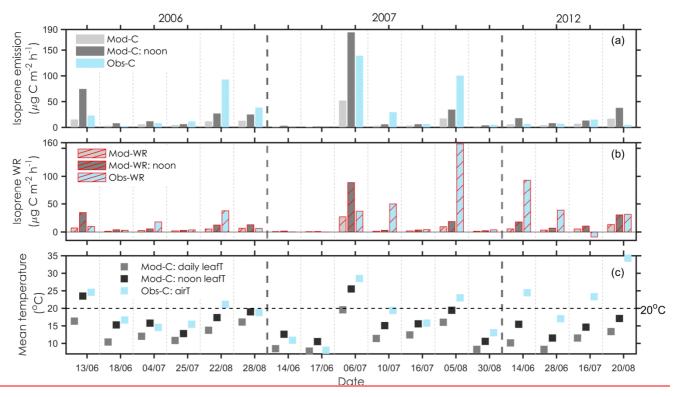


Figure <u>45</u> Comparison of the modelled daily average and noon isoprene emission rates with the observations in the control (C) plots (a) and evaluation of modelled warming responses (WR) with the observed WR (b) at the Abisko <u>heath-tundra heath</u>. The modelled daily average (daily leafT) and noon leaf temperatures (noon leafT) in the C plots were compared with the observed chamber air temperature (airT) averaging over sampling times during daytime. Mod: Modelled; Obs: Observed.

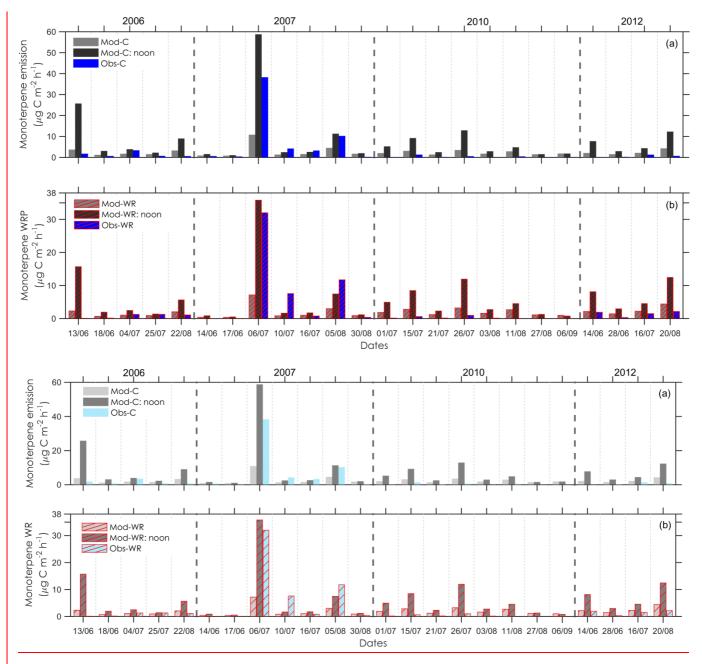


Figure <u>56</u> Comparison of the modelled daily average and noon monoterpene emission rates in the control (C) plots (a) and the evaluation of modelled warming responses (WR) with the observed WR (b) at the Abisko <u>heath</u>-tundra <u>heath</u>. Mod: Modelled; Obs: Observed.

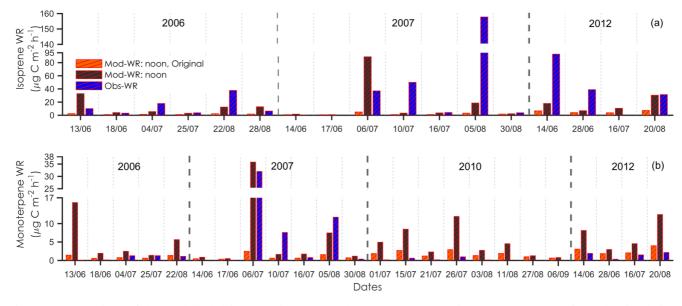


Figure 7 Comparison of the modelled daily noon isoprene and monoterpene warming responses (WR) from the simulation with the original temperature responses (Mod WR: noon, Original) and the simulation with the adjusted temperature responses (Mod WR: noon). The observed WR (Obs WR) are also presented.

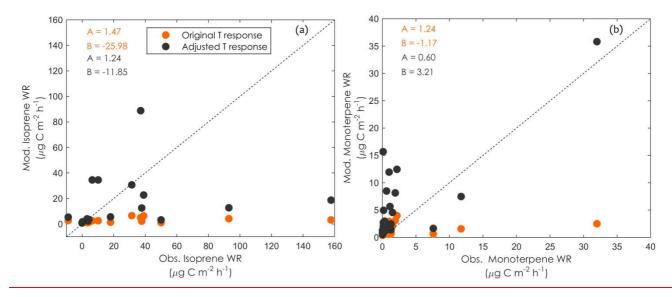


Figure 6 Scatter plot of the modelled (Mod.) and the observed (Obs.) warming response (WR) for both isoprene (a) and monoterpene (b), using the adjusted and the original T response.

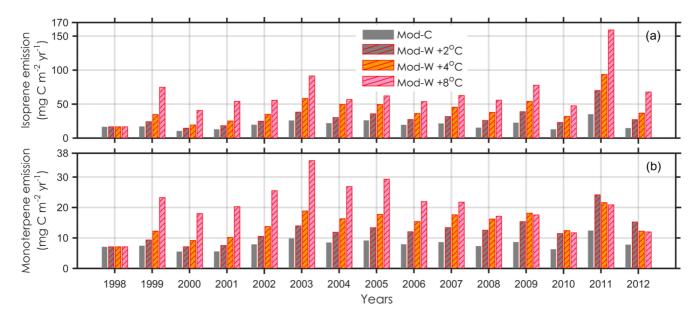


Figure 8–7\_Modelled annual isoprene and monoterpene emissions for the period 1998-2012 at the Abisko heath tundra. The warming (W) treatment started in 1999 and three levels of warming (+2 °C, +4 °C and +8 °C) were applied during summertime. The modelled annual emissions in the control (C) plots are also presented.

Thanks for the reviewer's constructive comments and suggestions for improving this manuscript. All comments have been answered (with grey colour background). Below, we first give general answers to the reviewer's comments and the detailed comments will be addressed separately point by point.

## Changes made in the revised manuscript have now been indicated (with green colour background). The page and line numbers mentioned in the changes refer to the ones in the revised manuscript.

#### Major comments

The subject matter of this paper is important. The Arctic environment is changing rapidly. Because of BVOC impacts on air chemistry, it's important to have models that can successfully predict the response of BVOC emissions. This paper makes an important contribution by employing a model with a dynamic vegetation component. As they warm, Arctic ecosystems are expected to see a shift towards woody plants, and this should change the capacity of the ecosystems to emit BVOCs. The paper has strengths, but also needs substantial improvements before publication. The basic modelling approach is sound, and it's helpful that the authors include the investigators that actually made the measurements. The paper demonstrates a good understanding of many of the ecosystem processes that should be captured by the model. Overall, I thought the discussion section was strong. Among weaknesses, the comparison of the model to the observations need to be improved. First, much of the discussion is qualitative. The model is said to fit the observations well in many instances, but there is no quantitative analyses: no goodness of fit metrics, and no statistics. Need to formally compare model to observations with statistics. More specifically, using the max and the daily average as a basis for comparison doesn't make much sense. What is the point of a daily average, especially since the meaning of the daily average changes with the long diurnal cycles in the Arctic? Why not just use the times of day that cover the range of the observations? Also, the figures could be improved by consolidation. The same data are presented in multiple figures in two different instances. The figures would also be easier to interpret if instead of presenting the max/daily average, just one metric was used for comparison to the observations. Also, there is very little acknowledgement of potential for experimental error in observations (one mention at the very end). Given the technical challenges with experiments in the Arctic, the potential for measurement error should be addressed.

The employed model is touted as being a mechanistic model, but then an empirical method is used for its calibration to the dataset. This is not itself a problem per se, but the paper states that mechanistic models are better than empirical models. If so, why is such an empirical calibration necessary? Also, a serious deficiency with the model is that it does not account for the effect of previous weather conditions (example, 24 hours and 10 days) on the capacity to emit BVOCs. This effect is potentially very important in the Arctic.

Finally, the list below of minor comments and technical corrections is extensive.

Response: (1) We agree with the reviewer that the comparison of modelled and observed variables should include some statistics and will add Willmott's index of agreement (describing models' prediction with pairwise-matched observations) as well as mean bias error (describing mean deviations between modelled and observed values) for each comparison.

Changes: The model's performances have now been evaluated by Willmott's index of agreement and mean bias error, see the descriptions in Page 10, Equation 5 and 6. These statistics have now been added in the results section.

(2) The reason why we present both daytime average and daily maximum values to compare with the observed is that the BVOC sampling on each field plot was conducted at a certain time point of a day for 30 min, while the modelled processes are at daily scale. Considering the strong diurnal cycle of BVOC emissions (for an arctic example, please see (Lindwall et al., 2015)), neither daily average nor daily maximum were accurate enough to directly compare with the observations. We reason that by presenting both rates, so we can see the range of the modelled daily emissions.

Changes: Both rates are still included in the manuscript, but clarification of the abovementioned reasons has now been added (see Page 9, lines 26-30).

(3) There is some data repetition in Figures 4-6, and separating them in different figures was aiming to explain different perspectives. In the revised manuscript, we will move Figure 4 to the supplementary to reduce data repetition. There will be no data repetition in Figure 5 and 6.

Changes: Figure 4 has been moved to the Supplementary Figure S2, together with previous description in the result section (see Page 11, lines 21-33 and Supplementary Page 6).

(4) We agree that the discussion about potential uncertainties from the measurements should be addressed in depth, mainly covering point intercept-based coverage and side effects from OTC-chambers.

Changes: The side effects of the OTC-chambers on warming responses were described in Page 17, Lines 5-7. The discussion about the point-intercept-based measurement was on Page 15, Lines 1-2. Discussion about the used technique for BVOC measurement has now been added on Page 16, Line 7-10.

(5) Regarding to the empirical method used in calibrating T response, we agree with the reviewer that there is empirical element in the parameter estimation, which, however, reflects some processes understanding, e.g. underlying enzyme activation. Even for mechanistic models in bio/geosciences fields, it sometimes cannot avoid of using empirical relationships determined from observations where multiple processes may anticipate. In our case, the calibrated T response (empirically) is used for influencing fraction of photosynthetic electron transport contributing to isoprene and monoterpene production, which is internally linked to other processes and potentially reflect more dynamics than other empirical models.

Changes: no changes have been made. Please see the above response.

(6) The current model did not consider the past weather conditions, rather, it emphasizes the enzymatic acclimation to short-term climate. As we do not have observed data to support which period of past weather should be taken into consideration, adding emission acclimation to the past weather will bring additional uncertainties to the modelled fluxes and complicate the current comparison. Ekberg et al. (2009) has fitted their observational data to obtain a relationship between past weather conditions (48 h) and isoprene emissions (not for monoterpene) from wetland sedges. We would like to further address this issue in the future model development with assistance of available climate data on measurement sites (past 24-96 h temperature as well as leaf level BVOC sampling data).

Changes: no changes have been made. Please see the above response.

Minor comments and technical corrections

Title: The title suggestions that the article will focus generally on modelling subarctic plants, but instead the article is about one specific effort using one specific model formulation. While of course some of the manuscript is more general, it is also uses data from just one field site.

Response: We can well understand the reviewer's concern about including only one study site and will therefore add a sub-title to specify it. The title will be changed to "Challenges in modelling isoprene and monoterpene emission dynamics of subarctic plants: A case study from a tundra heath".

Changes: New title: "Challenges in modelling isoprene and monoterpene emission dynamics of arctic plants: a case study from a subarctic tundra heath"

Page 1, line 14 – page 2, line 4: The abstract could be clearer. There are some specific recommendations below, but more generally the abstract should be condensed and just the highlights presented.

Response: Thanks for the comments. The abstract will be adjusted to condense the length and to make points clearer.

Changes: The abstract has been condensed by deleting a few sentences, See page 1, Lines 29-30 and Page 2, Lines 2-4.

Page 1, line 14: Title says "subarctic" while abstract goes back and forth between arctic and subarctic. Make sure each use is intentional. Further in the manuscript, (sub)arctic is used. Again, make sure this is all consistent.

Response: We will carefully consider each mention of "subarctic" and "arctic" through the whole manuscript. We will check and correct to use the most precise term in each place. The observational data originate at the Subarctic, but many ecosystem processes and components function similarly both in the Subarctic and the Arctic, and many of the issues handled are similar in both regions.

Changes: We have changed the title by stressing that the paper is presenting challenges for modelling BVOC in the Arctic based on this case study in a subarctic tundra.

Page 1, line 23: "higher levels of warming" instead of "higher levels' warming".

Response: Accepted.

Changes: Changed.

Page 1, lines 24-26: The sentence should be written. Do you mean the "measured" BVOC WR, not modeled? If you do mean modeled, what was the standard "were better captured"? Also, "compared" instead of "comparing".

Responses: Yes, it should be "measured". The suggested changes will be implemented.

Changes: Statistic has now been added in the main text (Page 12, lines 14-15).

Page 1, line 26: This sentence relays an interesting result, but there is not enough context to warrant inclusion in the abstract. Please remove it.

Response: The reason behind is the underestimated leaf T is one of the main factors influencing short-term BVOC fluxes. The sentence sounds a bit misleading, but we will clarify it.

Changes: The sentence has been changed to: A few days' underestimation of leaf T is one of the main causes responsible for the underestimated emission rates as well as WR.

Page 1, lines 30-31: This sentence can be removed, since it's a circular argument. The high WR led to the high adjustment T curve.

Response: The adjustment of T curve was only based on the emission rates from the control plots under naturally varying weather conditions within the growing season, and no emission rates from the warming plots were used. So, it is not high WR, which led to the high T curve. The improved ability of capturing the observed WR by the adjusted T curve indicates a better representation for arctic plants.

Changes: This sentence has been deleted to condense the abstract a bit.

Page 2, line 3: remove "extrapolation".

Response: Changed.

Changes: changed.

Page 2, lines 3-4: How do points (2) and (3) differ? Isn't "PTF's responses to warming" a subset of "representation of vegetation dynamics in the past and future"?

Response: we agree that these two points sound similar. Here, in point (2) "PFT's responses to warming", we mainly mean plant's physiological adaption to warmer climate, while in point (3) "representation of vegetation dynamics in the past and future", we mainly refer to long-term vegetation development, e.g., composition changes, disturbances, and expansion, etc. We will clarify the sentence.

Changes: Point 2 and 3 has been changed to: (2) PFT parameterization accounting for plant emission features as well as PFTs' physiological responses to warming; and (3) representation of long-term vegetation changes in the past and the future.

Page 2, line 7: "plant" instead of "plants" or include an apostrophe.

Response: Accepted.

Changes: changed

Page 2, lines 10-13: First, need to include that BVOCs don't solely react with OH. In particular, ozone is another important reaction partner for some BVOCs. Second, in a low-NOx environment, BVOC emissions can lead to a reduction in tropospheric ozone concentrations.

Response: The text will be clarified. Thanks for the points.

Changes: The description has been changed to: An increase in BVOC emission could also elevate the tropospheric ozone (O3) concentration when the ratio of BVOC to NOx (BVOC/NOx) is high (Hauglustaine et al., 2005), and also increase secondary organic aerosol (SOA) formation (Paasonen et al., 2013). BVOC could also limit ozone formation when the BVOC/NOx ratio is low, a situation in which the regeneration of NO2 can be mainly achieved by NO reacting with O3 (Hauglustaine et al., 2005).

Page 3, line 3: "from" instead of "along". Also, why is G3P the "chief precursor" if pyruvate is also required?

Response: We have changed the words. The description of G3P as the chief precursor was not accurate and will be corrected.

Changes: The text has been changed to: both G3P and pyruvate serve as the chief precursor.

Page 3, line 6: "part of monoterpene productions" should be clarified.

Response: This is not accurate. It will be changed to "monoterpene productions".

Changes: Changed to "monoterpene productions".

Page 3, line 8: remove the inner set of parentheses.

Response: Corrected.

Changes: changed.

Page 3, lines 13-15: This is a contentious statement, and there is no reference. A more nuanced statement is necessary, and could reference Monson, R.K., Grote, R., Niinemets, U., Schnitzler, J.P., 2012. Modeling the isoprene emission rate from leaves.New Phytologist 195, 541-559.

Response: We will add the description about process-based models can represent BVOC synthesis activities in chloroplasts and vary between species and leaf long-term growing environment. The suggested reference will be added.

Changes: process-based ecosystem models, representing BVOC synthesis activities, can vary with species as well as long-term growing environment effects and could thus be more useful in terms of predicting long-term emission responses to environmental changes (Monson et al., 2012)

Page 3, line 16: "referred to here" instead of "referred here"

Response: Accepted.

Changes: changed.

Page 3, line 17: remove space before comma

Response: Corrected.

Changes: changed.

Page 3, lines 18-20: Should reference Potosnak et al 2013 here. While dwarf willow's T response was OK compared to G93, the light response was more linear than expected.

Response: We will add this description. Thanks.

Changes: The suggested descriptions is added: Potosnak et al. (2013) fitted leaf-level isoprene emission rates to T and Q in a moist acidic tundra and found the G93 algorithm characterized well with the T response, but not Q response.

Page 3, line 25: Should also include low transpiration rates. Because of permafrost, transpiration rates can be low, which also leads to the high ground temperatures.

Response: Based on the observations, there is no permafrost at the studies plots. The issue is relevant for other arctic regions.

Changes: Some discussions about other regions with permafrost underlying have been added on Page 17, Lines 2-4.

Page 3, line 30: Give what LPJ-GUESS stands for.

Reponses: the full name will be added.

Changes: The full name is: Lund-Potsdam-Jena General Ecosystem Simulator

Page 4, lines 2-4: The objectives could be clarified. To me, (1) "capture the observed BVOC T sensitivity" is the same as part of (2) "To address short-term and long-term impacts of warming on ecosystem BVOC emissions." Be more specific about your study goals, or further differentiate the difference between 1 and 2.

Responses: Thanks for point out this part. The first aim will be clarified by changing it to "capture the observed T responses of BVOC emissions for a subarctic ecosystem", to clarify that we mainly tackle questions about emission responses to temperature. The second mainly aim to compare short-term and long-term warming effects on the whole ecosystem. We will clarify both aims.

Changes: The text has been changed to: The specific objectives of this study were: (1) To capture the observed T response of BVOC emissions for a subarctic ecosystem; (2) To address the importance of short-term and long-term impacts of warming on ecosystem as well as BVOC emissions;

Page 4, line 8: use straight single quote for minutes symbol.

Response: corrected.

Changes: changed.

Page 5, line 1: You have already defined PFTs above, so don't redefine.

Response: corrected.

Changes: changed.

Page 5, lines 2-4: Is this statement true for Arctic-specific PFTs? Please indicate this.

Response: Two of the cited references were conducted studies in high latitudes. So yes, this statement is true for arctic PFTs.

Changes: no changes have been made.

Page 5, line 5: What does "large-scale" mean here? I consider the base Farquhar equations to be leaf-level. Do you mean canopy-scale?

Response: The "large-scale" here mainly refers to the spatial scale. LPJ-GUESS uses the canopy-level photosynthesis calculation based on Haxeltine and Prentice (1996a), where a set of canopy-level equations were developed from the Farquhar leave-level equations. We will clarify our description in the main text.

Changes: The descriptions have now changed to "In LPJ-GUESS, a generalized Farquhar photosynthesis model (Farquhar et al., 1980; Collatz et al., 1991) for large-scale modelling is used to simulate canopy-level carbon assimilation and the generalized model is built on the assumption of optimal nitrogen (N) allocation in the vegetation canopy (Haxeltine and Prentice, 1996a; Haxeltine and Prentice, 1996b)"

Page 5, lines 4-12: I assume transpiration & stomatal conductance are also modelled to get pi? Maybe you'll talk about this further down, but it would be important for understanding discrepancies between air and leaf temperature.

Response: Yes, in the model, the stomatal conductance influences transpiration as well as intercellular CO2 concentration. We will clarify this in the descriptions.

Changes: See page 5, lines 30-31 and page 7, lines 13-14.

Page 5, line 18: How is Ci different from pi defined on line 11. Just concentration vs. partial pressure? What does "without water stress" really mean? This is probably tied to my comment above.

Response: Thanks for pointing out. It should be  $p_i$  and  $p_i$  is influenced by stomatal opening, so we will correct both.

Changes: changed. See page 6, Equation 1 and lines 5-6.

Page 5, line 26: "optimum from terpenoid synthesis" should be "optimum for terpenoid synthesis"

Response: corrected.

Changes: changed

Page 6, lines 1-2: Give a reference for the co2 response in the model, as you've done for the other responses.

Response: corrected.

Changes: The reference of Arneth et al., (2007) has been added.

Page 6, lines 12-15: Again, this gets back to my comments above about transpiration and conductance. It would make more sense to move this discussion to the general description of the model, before discussing biogenics. Also, more detail on this part is necessary. What are the details here? This can be done by references to the literature, if it has been described by LPJ-GUESS before. What is the coupling between estimating leaf temp, internal co2, transpiration and stomatal conductance? Or is a more empirical algorithm used?

Response: Using leaf T instead of air T for photosynthesis was developed in this study, not in other LPJ-GUESS studies. The reason for putting the description of leaf T development after BVOC process description is that the development of leaf T algorithm mainly considers the strong sensitivity of BVOC to leaf T. We agree with the reviewer that the description of leaf T as well as its linkage to the transpiration and stomatal conductance should be extended. We will also stress the leaf T rather than air T was used for photosynthesis in this study.

Changes: See changes from Page 7 lines 9-13.

Page 7, line 4: Fix grammar: either "appearing" or change sentence structure."

Response: changed.

Changes: changed.

Page 7, line 4-5: I agree there is insufficient data, but mosses may make a large contribution to BVOC emissions in some Arctic ecosystems. So, it's fine to incorporate them into a larger PFT, but are you capturing their emissions? That is, do the emission factors for this PFT reflex the mosses?

Response: The emission factors for the CLM have considered the observed moss emission rates. At the ecosystem level, we cannot distinguish how much emission was from the moss relative to the other species.

Changes: The measured emission rates from mosses had been integrated into the CLM group (see Table S2). No changes have been made.

Page 7, line 17: first, not firstly.

Response: corrected.

Changes: changed.

Page 7, line 20: "other" instead of "rest"

Response: corrected.

Changes: changed.

Page 7, lines 18-21: Given the lack of data for the Arctic, it's justifiable to use two years data for calibration and two years for validation. But, the sensitivity of this procedure should be assessed by flopping the years: how different are the results if the second two years are used for calibration, and the first two used for validation?

Response: We did sensitivity testing using data from other two years (2010 and 2012) to calibrate, but this resulted only in slight effects on the best values for the checked LAI, GPP and ER, and the trend was consistent with that in Fig. S1. Further, it did not affect the selection of 0.04  $\mu$ mol CO<sub>2</sub>  $\mu$ mol photons <sup>-1</sup>.

### Changes: no changes have been made. Please see our explanation in the above response.

Page 8, lines 9-11: The goodness of fit here is a bit deceptive. The fit is entirely driven by the relatively few points that are above 23 deg C. Since everything below that is relatively close to zero, there is little new information added. For example, blocks 5 and 6 only have one observation each above 23 deg C, so the individual fits are very good. I don't see the added value in the doing the individual fits for each block. It seems all that info comes out of the overall fit. Finally, you should understand the justification for using 20 instead of 30. Yes, this makes sense conceptually and certainly for measurements, but realizes that mathematically, using your formulation, there is no difference between using 20 and 30, because of the laws of exponents. That is, you'll get the same r2 for the fits with each. This isn't true with more complicated formulations of the T response; for example, the T response in isoprene emission for G93.

Response: The reason of adding block fitting is to illustrate the general fitting to the whole dataset also worked for each block (except for block 1) providing stronger evidence for the general trend. From the exponential equations along, we agree with the reviewer that there is no difference between using 20 or 30 degrees as reference temperature. However, the reference T in the model is not only used as BVOC T response equation, but also used in estimating photosynthesis electron flow rates at this reference T to convert the input emission capacity to fraction (see P7, line 35- P8, line1-2). The photosynthesis responses to the reference T of 20 and 30 degrees are not exponential. More explanation about the difference causing by different reference T will be added.

## Changes: Please see the added clarification at Page 6, line 14-15 and Page 9, line 5.

Page 8, lines 16-23: This is confusing. Your goal is to compare your measurements to the model. So, yes, using daily averages isn't appropriate. But why discuss them in the first place? I think you'll use them for another purpose, but that's not clear. Why do you use max T & PAR? Wouldn't an average around the measurement time make more sense? And again, your last sentence here is obvious. Particularly in the Arctic, with low sun angles for much of the day, this isn't a strong statement.

Response: We agree that the issue indeed was a bit confusing because of our wording. We will clarify that not all measurements were only between 10 am- 2 pm, but also with a few sampling between 9 am - 5 pm, and this is the reason why we still keep the daily average in the results. Since we don't use hourly inputs, it is not possible to average emissions for the time period of days with the measurements. We used theoretical maximum T as the input for extracting daily maximum emission rates. Generally, it is not a very difficult problem to compute the maximum PAR, but we cannot really compute an instantaneous photosynthesis flux at noon (or any other time) with Haxeltine and Prentice approach. Lindwall et al. (2015) has shown strong diurnal cycle of BVOC emissions in the Arctic. This reference will be added to support our statement in the last sentence.

Changes: see changes in Page 9, lines 26-31.

Page 8, lines 27-28: Again, examine Equation 3. You'll see that changing from 30 to 20 only introduces a constant.

Response: As explained in the previous responses, the reference temperature is not only used for the exponential equation (Eq. 3), but also used in LPJ-GUESS to link the photosynthesis rates at 20 degree.

Changes: Please see the added clarification at Page 6, line 14-15 and Page 9, line 5.

Page 9, line 4: In Fig. S1, the figure legend should indicate what the dashed vertical line denotes at the value of 0.4 in both panels. The text explains this, but the figure caption should too.

Response: Corrected.

Changes: changed.

Page 9, line 10: Do you expect to see a one-to-one correspondence between the point intercept info and the LAI values? This surely doesn't hold as LAI gets closer to 1 (and exceeds it), but you should share your expectation here. Do you assume that there is no overlap with cover, and therefore there should be a one-to-one relationship? If so, state that.

Response: From the model side, LAI is the most relevant variable which can be used to compare with the point intercept measured plant coverage. The point intercept-based method does count numbers of plants that pin hits, not only the top canopy layer. So it should not have problem in comparing with LAI when LAI get larger or closer to 1. In this context, we did not assume no overlap with cover.

Changes: No change has been made, but please see the discussion regarding to uncertainties in pin-point measurement (page 14, Line 33 to Page 15, line 1-2).

Page 9, lines 18-22: You discussed the LAI response to warming, but not the GPP/NEP/ER response. Why?

Response: We did compare GPP response to warming as well and it showed that an underestimated vegetation  $CO_2$  fixation to warming in most cases, which can also been seen in the evaluating LAI response to warming (the absolute difference of total LAI between C and W plots). So we only included the LAI response to warming in the manuscript. Although the plant  $CO_2$  fluxes are also linked to BVOC emission, we consider the warming responses of LAI more relevant as they are aggregated effects of both photosynthesis responses and vegetation composition changes (directly linked to the changes in emitted compounds and relative magnitudes).

## Changes: no changes have been made.

Page 9, line 29 – page 10, line 1: This analysis isn't adding much to your argument. Of course you see this, because your model is driven by PAR and T. You don't need to cover this result. It follows directly from your model formation (Equations 1-3). Second, I don't understand the relevance of relating mean daily ISO/MT production to the noontime values. What do you learn from this?

Response: Thanks for your points. We will move figure 4 in the supplementary instead. We think if reader is interested to see the seasonality of BVOC emissions in this region as a general picture about temporal dynamics

of emissions, they can still reach it. As we explained in the earlier responses, due to the strong diurnal variations of BVOC emissions during a day, sampling time is crucial for determining the emission magnitudes. Although many samplings were conducted during the period of 10 am - 2 pm, there were also samplings conducted beyond this period. Through presenting both daily average and maximum values, we can conduct an approximate evaluation of how the model performs by checking if the measured rates were in the modelled range.

Changes: Figure 4 has been moved to Figure S2. The clarifications about why we present both daily noon and maximum values have been added in Page 9, lines 26-31.

Page 10, lines 3-4: For "the observed average rates (blue squares) were well captured by the modelled noon emissions" you need to present some statistics to back up this statement. You should do an xy plot of this data and see what the fit looks like. Even if you don't present the plot as a figure, you should report the statistics of the fit.

Response: Thanks for pointing this out. We will add Willmott's index of agreement (A) as well as mean bias error (B) to describe the model's performance.

Changes: See the added statistics on Page 12, lines 14-15.

Page 10, line 10: As mentioned below, the same data is presented in Figs 4 and 5a. And now you've made the same statement about fit as above. This should be consolidated, and again there needs to be a statistical analysis of the goodness of fit.

Response: We will move Figure 4 and relevant descriptions into the Supplementary. So in this case, we will not have much data overlap in the figures. We will add statistics to the remaining figures.

Change: Figure 4 has been removed and statistics have been added to text and Figure 6.

Page 10, lines 13-18: Yes, the temperature drives these emissions, but this is a bit complicated because of the chamber observations. There are two issues: one, the model's ability to predict leaf T; second, the increase in air T because of the chamber used to measure BVOCs. Only the first is important for extrapolating your results.

Response: very good point. In this context, we can only discuss what we have considered in terms of leaf temperature estimation. However, the side effects of measurement chambers on leaf temperature were considered to be minor. As tested by De Boeck et al. (2012), the main side effects from chambers on leaf temperature is related to reduced wind speed. In our case, the measuring chamber has a fan to mix air during sampling time, in which we do not expect large impacts on the observed leaf temperature, but could have some impacts on chamber air temperature. However, for the warming treatment where the OTCs were installed to passively increase surrounding temperature, the OTC-resulted reduction of wind speed may elevate leaf T more than the expected on air temperature warming and could be considered as one reason for why the modelled WR was generally lower than the observed (Please see Section 4.2, first paragraph).

Changes: no changes have been made. Please see the explanation above.

Page 10, lines 25-27: Again, need statistics to back up these contentions.

Response: We will add Willmott's index of agreement (A) as well as mean bias error (B).

Changes: added. See changes on Page 13, lines 13-14.

Page 11, line 30: After not using any statistics comparing the model to observations, why would you use a statistic in this case, when you are comparing the model to itself?

Response: When we evaluated the modelled emission rates at daily scale, we focused on presenting the absolute differences from the observed. But as the reviewer suggested, we will add statistics for the model-data comparison. For the modelled annual emissions, we don't have the observed data and to illustrate warming effects on the emissions, the Mann-Whitney test was applied.

Changes: The added statistics have been indicated in previous answers.

Page 13, lines 23-25: This is an interesting contention. But, the emissions for the storage pool are generally regarded as being due to the physical process of evaporation of the MTs. Why would this change for Arctic plants?

Response: Parameterization of Eq. 4 is based on global scale study by Schurgers et al. (2009). It may be the case that for arctic plants, there is larger storage pools for MTs or different leaf anatomy which could influence release from storage pools. We are lacking of knowledge to quantify these effects on MTs emission, but we will clarify our discussion here.

Changes: see changes on Page 16, lines 2-6.

Page 13, lines 28-29: Yes, and that is why restricting your modelling to the times of day when measurements occurred would help.

Response: Please see the previous response regarding to daily processes in the model.

Changes: No changes have been made due to the modelled process at daily scale, but a change has been made about the measured time range (see Page 9, line 24).

Page 14, lines 2-3: Do you mean the bryophyte decrease due to drying is an artifact of the experimental warming and shouldn't be captured by the model? Please elaborate.

Response: The observations found an increase of bryophyte coverage, but our model predicted a decrease of the coverage. The decreasing trend in response to warming is consistent with the study by Elmendorf et al. (2012) where they summarized 61 tundra warming experiments. Elmendorf et al. (2012) elaborated that drying of soil moisture is one of the reasons of declining of bryophyte coverage, which was captured by our model.

Changes: no changes have been made. Please see the explanation above.

Page 14, lines 5-7: Yes, because you used the observed data to fit your model. Remind readers of that point.

Response: As mentioned in the earlier reply, we only used the emission rate and T at control plots to get the response curve, but compared the modelled WR with the observed, which has shown the improvement.

### Changes: no changes have been made. Please see the explanation above.

Page 14, lines 11-13: Could also mention the drying that was noted above for species responses (bryophytes).

Response: changed.

Changes: added on Page 17, line 6.

Page 14, lines 18-19: Need some more analysis here. Yes, the two responses are very important. But, you should re-emphasize that your dynamic vegetation model isn't doing a great job of getting the vegetation changes correct. Therefore, the results in Fig 8 are illustrative of the impact, but the details are not certain.

Response: Thanks for the good points. We agree with the reviewer that we only illustrate a potential impact, but that there are still uncertainties in capturing the vegetation dynamics in detail.

Changes: The uncertainties about capturing vegetation dynamics have now been added in page 17, Lines 14-15. We also emphasized the uncertainties from the emission responses to environmental variables (Page 15, Lines 16-21)

Page 14, lines 25-30: I agree with most of this logic, but since this particular study is looking at whole system measurements, there is potentially an interaction between the true T response of the plants and the issue of canopy temperature described earlier. You should at least discuss the possibility that some of this T response is not at the enzymatic level, as suggested here, but is due to a non-linear increase in leaf T with increasing air T due to canopy warming. Perhaps some of the references cited are leaf-level measurements which could clarify this point?

Response: The decoupling of leaf T from air T at 2 m height may partly contribute the observed strong warming responses. It may also relate to arctic plant species. Discussion about the strong decoupling of leaf T from air T as well as its linkage to potential T response will be added.

Changes: Discussion about leaf T decoupling from air T has been added on Page 16, Lines, 4-5. Also, the discussion about the potential effects of different time resolution in the measured data and observed data has been added on Page 16, Lines 15-15 and Page 17, Lines 25-27.

Page 15, lines 1-3: Yes, this is important, but it also brings in the issue of drought stress. Drought stress can occur frequently in some Arctic ecosystems due to relatively shallow soils above the permafrost. To understand canopy heating, it will be necessary to understand canopy water dynamics.

Response: Thanks for the reviewer for pointing this out. In LPJ-GUESS, leaf energy balance has been considered and the evapotranspiration is a function of soil water content. For this particular site, there is no permafrost and the soil is relatively moist. As the reviewer correctly pointed out, drought stress for some other arctic regions are possible, which may lead to further increase in canopy surface temperature.

Changes: discussion about potential drying in the arctic permafrost region has been added on Page 17, lines 2-4.

Page 15, lines 15-16: Great this is stated clearly in the conclusion, but this point should also be made in the discussion.

Response: changed.

Changes: see discussions on Page 17, lines 14-16 and page 15, line 16-17.

Figures 2, 4, 5 and 6: For Figs 2, 5 and 6, use you DD/MM on the time axis, but day of year for Fig 4. Be consistent, and I prefer day of year.

Response: We will change to DD/MM for Fig. 4

Changes: see Figure S2.

Figures 5, 6: The top panels (a) of each figure are the same data presented in Figure 4. These results shouldn't be presented twice.

Response: We will only keep Figure 5 in the main text.

Changes: Figure 4 has been removed from the main text.

Figure 7: There is also a lot overlap with Figures 5b and 6b: two of the three sets of data have already been shown. In addition, why is there a break in the y-axis, when mostly the same data have been presented in Figures 5b and 6b without a break?

Response: I guess you mean Figure 6b and 7b. The reason for adding a break in y axis in Fig. 7 is the modelled WR from the simulation with the original T response is really low, which is only presented here. In the revised version, we will use scatter plot to compare the differences between two T responses curves.

Changes: A new scatter plot has been added to replace the original Figure 7.

Figure 9: Why include the higher-T scenarios? I understand they are (unfortunately) realistic due to the IPCC estimates of climate change. But, you don't discuss them much, and there are obviously some weird things happening with the vegetation change (for example, lower +8 compared to +2 for MTs in 2012). Since the vegetation changes predicted by the model are suspect, the results of the +4 and +8 runs are highly speculative.

Response: The purpose of having Fig. 8 (Not figure 9) in the manuscript is to illustrate factors influencing BVOC emissions at long-term scale, highlighting that vegetation changes can affect the response at decadal timescales, and also illustrating that this can lead to a change in the response over time, or a non-linear response to the level of warming. We agree that these estimates are uncertain, also given the fact that the model does not capture all observed vegetation trends. We will expand the description of the results and discuss the underlying response.

Changes: The purpose of having higher-T scenarios had been described in Section 2.3.4 (Page 10, Lines 12-13). Discussion regarding to the annual emission estimates has been added on Page 15, Lines 16-21. In the results section, we have addressed that vegetation is responsible for very different emission responses at 4 °C or 8 °C warming.

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This is a nicely written manuscript which addresses an important question in BVOC estimation – namely the representation of cold environments in global estimates and the uncertainties of modelling in this respect. It is also well timed since a lot of new information has recently been published about this topic and the implementation of this knowledge into a model is overdue. However, I feel like I have to urge the authors to be more careful in what they regard as 'good agreement' between measurement and simulation or at which point they conclude that the model's suitability has been 'demonstrated'. Overall, I see a lot of model deficiencies and uncertainties in this study which should probably be the prime focus of the investigation. In this respect, I would welcome figures or statistics that show the actual relation between measurements and simulations rather than column- or point diagrams. Apart from this, I think that the model description part needs some elaboration.

Response: Thanks for the reviewer's suggestion. Briefly here, we will address the model's agreement with observations using a Willmott's index of agreement as well as mean bias error. Apart from BVOC related processes, a description of general photosynthesis processes will be added to Section 2.2.

Changes: Statistics have been added in the results section (see Page 12, lines, 14-15 and lines 17-18) and the model description has been extended in the section 2.2.1 (see Page 5, lines 16-21).

Specific comments:

P1, L22: 'Short time scales' not only need to be defined, mentioning them here is also irritating. In fact, the question about simulations and observations referring to different time periods is troubling me throughout the manuscript.

Response: The term refers to a period of hours to a few days versus long-term scale of months to years. The clarification will be added in the main text. Since the simulated results from the model were at daily scale and the measured fluxes could be at any time point of a day, presenting the modelled daily average and maximum values aimed to bridge the differences in the time periods.

Changes: The definition of different scales has been clarified in the abstract as well as in the main text. The reason why we present both daily maximum and average has been clarified on Page 9, Lines 28-30.

P1, L24: The model 'was able' to reproduce carbon fluxes for the majority of the vegetation period but showed considerable weakness in representing the seasonality, probably due to mismatch of phenological phases. This should be recognized.

Response: The modelled  $CO_2$  fluxes do show some uncertainties in representing fluxes at the beginning of growing reason, which is discussed (see P13, L6-8) and related to phenological phases (the start of growing season). We agreed with the reviewer and will add the time period when the model did captured the observation. Also, we will change the term "was able to" to "showed reasonable agreement to".

Changes: The abovementioned changes have been added in the abstract. See Page 1, Line 26.

P1, L26: The difference of effective temperature in model and observation is certainly one reason for a mismatch in emission simulations which has been correctly acknowledged here. However, giving this as the only reason for a possible deviation is misleading at this point.

Response: Thanks for point out. The sentence will be clarified by stressing that leaf T is one potential main cause, but not the only reason for mismatches between model and observations.

Changes: The sentence has been deleted to condense the abstract.

P2, L17ff: Major uncertainties are also other driving factors for emissions that are usually not considered in models, namely air chemistry, soil water availability, UV light and biological stress impacts. Also the representation of seasonality (which is composed of phenology and enzymatic activity changes) is a point worth mentioning here. The authors are mentioning most of these points at a later stage but I feel that it needs mentioning here.

Response: Thanks for the great point. More details will be added in the introduction, paragraph 2.

Change: The suggested uncertainties have been added on Page 2, Lines 29-30.

P3, L5: I think that in the Pacifico and Unger papers, the Niinemets approach is used. So this is to some degree a repetition here.

Response: Agreed, we have reduced the references to unique implementations.

Changes: Two references: Pacifico et al., 2011 and Unger et al., 2013 have been removed.

P3, L10: seasonality and/or past weather conditions? In fact this is the same problem. You might differentiate into effects of phenology and enzymatic activity shifts though.

Response: We will change "seasonality" to "vegetation phenology" to differentiate relatively short-term acclimation (past weather condition) with vegetation phenological phases.

Changes: "Seasonality" has been changed to "vegetation phenology".

P4, L15: From the later remarks I take it that the BVOC emissions were not taken round the clock so the time or time period during the day when the measurements were made should be mentioned.

Response: A detailed description about measuring time will be added into the Section 2.3.3.

Changes: The update on the measuring time has been added on Page 9, line 24.

P5, L7ff: I am a bit irritated here. The Haxeltine and Prentice photosynthesis approach is for seasonal or annual photosynthesis estimation, assuming a kind of optimal adjustment to average environmental conditions. Nevertheless, the model seems to work on daily timesteps here. The description given about the model itself looks very much like the Collatz approach – so what is taken from Haxeltine here? Regarding the description, many abbreviations are introduced here that seem not to be used later on –please check.

Response: Thanks for pointing this out. We agree that the description (mainly references) of the photosynthesis processes was unclear. Though Haxeltine and Prentice model use monthly data as input, but it still have daily time step photosynthesis processes, which is what LPJ-GUESS is based on. The original simplified Farquhar model used in Haxeltine and Prentice is developed by Collatz et al. (1991) approach which works at sub-daily

scale. The model upscaling of leaf-level calculation to canopy scale is based on the Haxeltine's approach. The abbreviations which are not used later on will be deleted.

Changes: The descriptions have now changed to "In LPJ-GUESS, a generalized Farquhar photosynthesis model (Farquhar et al., 1980; Collatz et al., 1991) for large-scale modelling is used to simulate canopy-level carbon assimilation and the generalized model is built on the assumption of optimal nitrogen (N) allocation in the vegetation canopy (Haxeltine and Prentice, 1996a; Haxeltine and Prentice, 1996b)"

P5, L14ff: Since emissions depend on temperature in a highly non-linear fashion, I think it is generally acknowledged that calculating them with daily average values is necessarily not capturing the dynamics. Regarding the Niinemets model, for example Unger et al. used a 15 minutes time steps. From the description it sounds like LPJ feeds daily photosynthesis results into daily emissions. Can you elaborate on the problem? Also, I think that the reference temperature used in equation 3 and/or the parameter in the response function needs to be adjusted because the model is not using them as an immediate response value anymore but as parameter for daily average emission. (30 degrees as an average value throughout the day would probably exhaust the emission apparatus so that the response curve would not be valid anyway.)

Response: Thanks for the good points. The simulations in this manuscript used daily climate inputs and therefore the model works on daily scale, resulting in daily emissions. To overcome (the largest part of) the problem rightly raised by the reviewer, we compute a daytime mean (rather than daily mean) temperature to simulate BVOC emissions (details in (Arneth et al., 2007)). This will be stressed in the revised manuscript. Still, the reviewer is correct that an average daytime temperature may still yield an underestimation of the emissions with the convex shape of the temperature response, certainly if the temperature variations during daytime are large. We will add discussion on this problem. To make our outputs comparable to a few time points measurements during a day, we came up this idea of presenting both daytime average and also daily maximum emission rate.

About the fitted curve with reference temperature of 20 degree, we are now aware of potential uncertainties caused by different time resolutions. In an ideal case, if we have more frequent BVOC samplings in a day as well as in the main growing season, we could average daytime T and emission rates before do the curve fitting. However, the current dataset is too few to support us to implement this parameter adjusting. The reviewer is correct and we will address this issue in the discussion at well. Thanks!

Changes: As explained above, the mismatches between of time resolution between the modelled and the measured cannot be completely solved due to the daily scale applied in the model. So no changes have been in this part. However, the daytime temperature, instead of daily average temperature was used in the model, and further clarified in the revised manuscript (see changes in Page 6, Line 19-20).

As above-mentioned reasons, we cannot adjust our temperature curve based on daily averaged data, due to the limited data availability. But we are aware of potential uncertainties caused by different temporal resolution, and related discussion has therefore been added, see changes on Page 16, lines 15-18.

P5, L15: Instead of using I for isoprene as well as monoterpenes shouldn't you use Ei and Em or similar? This can further be modified for storage (e.g. Ems) in equation 4.

Response: The equations will be modified based on the suggestions.

## Changes: The suggested symbol has been used, see Equation 1 and 4 on Pages 6-7.

P5, L22: Here, the influence is named 'phenology' while later the same function refers to 'seasonality' (L30). Since these are two different things – is this a lumped index? Specific or specifically parameterized for PFTs? Empirical or dependent on weather or climate?

Response: The use of "phenology" here is indeed not correct,  $f(\sigma)$  represents the seasonality of the emissions caused by variations in enzyme activity. The effect of phenology (represented in the model as the abundance of leaves) is captured separately by affecting the amount of absorbed radiation. We will correct the sentence.

Changes: Please see the corrections on Page 6, line 5 and line 16. The model description regarding to plant phenology has been added on Page 6, line 9 and lines 22-23.

P5, L27ff: see also comment from L14ff. It seems that the reduction of reference temperature is rather a necessity from applying the model on a daily time step than a particular feature of arctic plants.

Response: Applying the reference temperature of 20  $^{\circ}$ C is of relevance for arctic plants since in most cases, the daytime T is close to or below 20  $^{\circ}$ C. We used the measured hourly BVOC fluxes with temperature in July to get the fitted temperature curve (see Fig. 1). The fitted response has been directly used in the model.

Changes: The clarification of using 20 °C for also computing photosynthesis fluxes has been added on Page 6, Lines 14-15, Page 8, Line 5. Potential uncertainties brought by different temporal resolution have been added on Page 16, line 15-18.

P5, L29: it is stated that the reference temperature is changed. This is to 20 oC as elaborated on later, correct?

Response: yes, we used the reference temperature of 20 °C. This will be clarified in the text.

Changes: See page 9, the 1<sup>st</sup> paragraph under Section 2.3.3.

P6, L2: fCO2 according to? Since it seems that variable CO2 air concentrations are used, it would be helpful to know to which degree CO2 might be responsible for differences between the years (probably small, but anyhow).

Response: Yes, the changes are small indeed, as f(CO2) varies with the inverse of the CO2 concentration. This gives a reduction of ~3% between 2006 and 2012.

Changes: see changes on Page 9, Line 11.

P6, L14: If the energy balance calculation was modified specifically for this study and is not published elsewhere, this modification should be explained.

Response: The development we had in this manuscript was essentially based on the work by Sedlar and Hock (2009) and therefore we did not include more details than just citing the original paper. But we will add more details about what are the main effects of adjusting the longwave radiation calculation.

Changes: The details have been added on Page 7, line 3-6. "The existing leaf energy balance equations appeared to underestimate the incoming longwave radiation under overcast conditions, which has been updated by

specifically considering the cloud emission of longwave radiation relative to clear-sky condition (Sedlar and Hock, 2009) The estimated leaf T, rather than air T, was used for both photosynthesis and BVOC synthesis."

P8, L15: I don't get how this can give you LAI values. Could you elaborate a bit? Looking at figure 3 there seems to be a difference between Lai and what is measured but the measurements are nevertheless used for evaluation. So how are the two related?

Response: The point intercept-based measurement gives a description of plant coverage (Finzel et al., 2012). During the growing season, the chances that the pins hit on leaves are generally higher and therefore we link these measured data with LAI which describes leaf coverage per ground area. It is not one-to-one relationship to compare (influenced by sampling inclining angles, sampling time, hits on stems etc., see discussion in section 4.1), but we think the modelled LAI is the closest variable we can compare with the measurement.

Changes: See the above explanation. No change has been made.

P8, L21: I agree that model results in daily resolution might not be comparable to measurements done at noon. This seems to be a general problem as mentioned above. I also agree that you can calculate noon temperature from average temperature to get a representative value of noon emission – but why don't you do the same with PAR? Instead of using the average value which is definitely wrong you can estimate maximum PAR from average PAR (e.g. Berninger F (1994) Simulated irradiance and temperature estimates as a possible source of bias in the simulation of photosynthesis. Agric. Forest Meteorol. 71:19-32)? Have you estimated the sensitivity of this error on the results?

Response: Thanks for the great point. Generally, it is not a very difficult problem to compute the maximum PAR, but we cannot really compute an instantaneous photosynthesis flux at noon (or any other time) with Haxeltine and Prentice approach, because it describes daily photosynthesis. It also becomes difficult to estimate potential sensitivity from different PAR values.

Changes: Please see the explanation above. Clarification has been made on Page 9, lines 26-27.

P9, L3: Check wording. I think it should be the modelled co2 fluxes that are sensitive to a change of parameter. This should also be indicated in some kind of measure, i.e. the degree to which the parameter was varied.

Response: The wording will be altered to clarify the text. The range for the parameter  $\alpha_{c3}$  was based on a previous study by Pappas et al. (2013) and the changes of modelled CO<sub>2</sub> fluxes as well as LAI, responding to the parameter  $\alpha_{c3}$  were illustrated in Figure S1. From Fig. S1, we can clearly see how the modelled CO2 and LAI varied with the parameter  $\alpha_{c3}$ . Before running sensitivity testing of  $\alpha_{c3}$ , we have selected several parameters to do sensitivity testing and then estimate Sobol sensitivity index to quantify the explained ability of each parameter to the modelled CO<sub>2</sub> fluxes as well as LAI.

Changes: Please see the changed wording on Page 10, Line 16-18. The pre-test/measure of varied degree of each parameter was tested but not presented.

P9, L9ff: In fact, the deviations are considerable. Not only GPP and thus emission is considerably overestimated in both years early seasons – which should be quantified and considered in annual estimates – but LAI is totally wrong in all PFTs except LSE+EPDS and CLM under current climate where the overestimation is a mere 10-15

percent. In L15/16 it is stated that these are the most important PFTs but in the next sentence the other PFTs are described to have a 'large coverage'. Are there any numbers that I have missed that give an objective picture about the abundances?

Response: Since the  $CO_2$  fluxes are not continuously measured, quantification of the overestimated  $CO_2$  fluxes of early season in annual estimates is unfortunately not possible. Considering that the modelled LAI and the point intercept-based may be not one-to-one relationship, the relative abundance of different PFTs coverage was evaluated. The measured coverage can be influenced by hits on non-leaf parts, pin size, subjective judgement of species and sampling inclining angles (see Discussion 4.1). We agree with the reviewer that the wording was at times confusing, e.g. the words "dominated" and "large coverage" and we will correct it.

Changes: No changes have been made in terms of estimating the overestimated GPP & emissions in annual emission due to limited observations. The pointed confusing sentence has been changed to "The two most dominant vegetation groups in the C plots, forbs/lichens and evergreen shrubs, were captured by the model. However, the coverage of graminoids (GRT) and non-Salix-type deciduous shrubs (NSLSS) was underestimated by our model. "Since we mainly look at compare the relative abundance of the modelled PFT LAI with the observed, no absolute numbers were compared between the modelled and the observed.

P10, L5: Monoterpene emissions seem to be met particularly because measurements occurred mostly on days with low emissions (according to figure 4). This is a problem because the high simulated emissions practically lack evaluation that should be addressed. I can certainly imagine other ways of representation or statistical analysis that can be used to elaborate on the point.

Response: Thanks for pointing this out. We will add discussion on the potential lacking evaluation of high monoterpene emission rates. We will also add the statistics for the comparisons and Figure 7 has changed to scatter plot to illustrate the modelled and the observed WR.

Changes: Please see the added discussion on Page 15, Line 29-32.

P10, L10: Similarly, I have large difficulties agreeing that figure 5 supports the statement that isoprene emissions were mostly captured by the model.

Response: This sentence actually pointed out that model is doing fairly good job on describing day-to-day variations of isoprene emission, though still have some discrepancies in capturing absolute magnitudes for some days. We will change our wording here and add statistic to support our description.

Changes: The sentence has been changed to: The observed daily variations in isoprene emissions were generally captured by the model (Fig. 4). The statistic has been added on Page 12, Lines 14-15.

P11, L26ff: The simulated annual emissions include the largely wrong response of LAI as well as the wrong response in early season emission, right? Can the error somehow be estimated? I have the feeling that these calculations might be too far off to be considered here.

Response: As mentioned in an earlier response, the closed-chamber based CO2 fluxes were not continuous measurements. The concluded overestimated  $CO_2$  fluxes during the early seasons were based on very few measured data points. To further consider their influence on the annual estimate is difficult without continuous

data support. The simulated annual estimate is uncertain considering the mismatch in LAI and early season  $CO_2$  fluxes, and we will clearly point out the uncertainty in the revised manuscript. However, presenting annual emissions in this manuscript is to look at longer timescales despite the discrepancies found in the evaluations.

Changes: As we mentioned, it may not be one-to-one relationship between the modelled LAI and the point intercept-based coverage. Instead of comparing the absolute values between these two, we mainly focused on the the modelled and the observed relative abundance as well as their response to warming. In this way, the model did fairly good job. So no changes have been made, but some wordings. About uncertainties of annual emissions, we have added more discussions on Page 15, lines 16-23.

P12, L14ff: The discussion seems to be overall comprehensive. Still, as for example in the first line, I think the authors are overenthusiastic about their results. This also applies for the conclusions.

Response: we will adjust the wording.

Changes: The sentence has been changed to: The modelled day-to-day variations of ecosystem CO2 fluxes (Fig. 2) and BVOC emissions generally followed the observations. See page 14, lines 12-13.

P14, L23ff: The comparison with common parameterization should not only be concentrated on the arctic environment but also on the problem with the time resolution (see above).

Response: The time resolution could be a possible cause. As mentioned in an earlier reply, the model has used daytime temperature, instead of daily temperature, which could reduce potential differences caused by two time scales. We will add discussion about potential influences of time resolution on emission T response in Section 4.2.

Changes: The clarification of daytime temperature used in the model has been added on Page 6, lines 19-20. The discussion about uncertainties from different temporal resolutions has been added on Page 16, Lines 15-18.

## Reference

- Arneth, A., Niinemets, Ü., Pressley, S., Bàck, J., Hari, P., Karl, T., Noe, S., Prentice, I. C., Serça, D., Hickler, T., Wolf, A., and Smith, B.: Process-based estimates of terrestrial ecosystem isoprene emissions: Incorporating the effects of a direct CO2-isoprene interaction, Atmospheric Chemistry and Physics, 7, 31-53, 2007.
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- Sedlar, J. and Hock, R.: Testing longwave radiation parameterizations under clear and overcast skies at Storglaciären, Sweden, The Cryosphere, 3, 75-84, 10.5194/tc-3-75-2009, 2009.

This paper presents a very valuable and interesting work, focusing on isoprene and monoterpene emissions from subarctic plants, a topic that has not been investigated or published much so far. I really appreciate the originality of this study, which helps to improve our understanding regarding emission estimates. However, as also raised by the two other referees, I think that this manuscript would really benefit from a deeper and more detailed presentation, of the result analysis and discussion especially, which would help to appreciate more clearly the validity of the conclusions of this work. Here are some feedbacks and corrections that would need to be considered before publication in BG, that I warmly support.

Thanks for the reviewer's valuable comments. We have now added the responses to each comment (shown with grey colour background).

Changes made in the revised manuscript have now been indicated (with green colour background). The page and line numbers mentioned in the changes refer to the ones in the revised manuscript.

Abstract: in "evaluating BVOC related processes", which processes for instance do you refer to, photosynthesis?

Response: Here, we referred to photosynthesis, BVOC temperature responses and vegetation composition. The clarification in the abstract will be added.

Changes: the abovementioned processes have been added. See Page 1, Lines 21.

Generally in the manuscript, the analysis is rather qualitative than quantitative and should be more detailed and specified. Some elements giving more precise information on the context could also be added. For instance, what is the estimated contribution of subarctic plants to global isoprene and monoterpene emissions? This could be specified for both the present-day case and the different warming scenarios, giving more perspective to the work carried out, and is important to be discussed, especially in section 4.

Response: Thanks for these good points. A statistical analysis of the model performance will be added. Regarding the contribution of subarctic plants' contributions to the global emissions, we would like to address in a coming manuscript where we will integrate multiple sites BVOC emission in the Arctic. We think it is a bit risk to estimate the contribution to global emissions based on one site study. We think the contribution to local atmospheric chemistry is potentially more important than the contribution to global emission (reactive compounds) and the warming-induced strong increase of emissions in this region is very important to address at global perspectives. We will add these two points in the Section 4.

Changes: The added discussions related to the contribution to global number as well as local atmosphere chemistry are on Page 15, lines 23-27.

Page 5, section 2.2.2 BVOC modelling: Could you please detail what the seasonality function used in isoprene production calculation stands for?

Response: The seasonality of isoprene production reflects observed changes in the availability of the enzyme for terpenoid synthesis, and is calculated based on a degree-day method in spring and a decrease in autumn based on temperature and day length. The details will be added.

## Changes: Please see the added details on Page 6, lines 23-24.

Page 6, section 2.2.2 BVOC modelling: Works published so far agree on the CO2 inhibition effect regarding isoprene emissions, but not regarding other BVOC emissions. Is the f(CO2) function considered in the model only for isoprene or for every BVOCs? On which work is it based and is the same parameterization considered for every compounds?

Response: In the model, we used the same  $CO_2$  response function for both isoprene and monoterpenes (and we do not use other BVOCs than those two) and the  $f(CO_2)$  response is based on the work by Arneth et al. (2007). We assume in the model that isoprene and monoterpenes are produced in the same pathway and assume both responses to  $CO_2$  in the same way. We agree with the reviewer that more work agreed on the  $CO_2$  inhibition on isoprene. In the work by Peñuelas and Staudt (2010), they listed some studies (in the supplementary) with  $CO_2$  inhibition effects on monoterpenes. We will extend our discussion on this topic and indicate this response is more robust for isoprene than monoterpenes.

Changes: The Arneth et al., (2007) paper has been added. The discussion about potential higher uncertainty for the modelled monoterpene than isoprene emissions has been added on Page 15, lines 16-23.

Page 9, line 25: What do you mean exactly with "dynamic vegetation" in "simulating dynamic vegetation enables us to assess the model performance"? Day-to-day variability? Higher frequency? Indeed the term of dynamic vegetation can also refer in vegetation modeling to long-term changes in vegetation distribution due to climate and CO2 changes.

Response: With the term "dynamic vegetation", we wanted to stress the model's ability to capture seasonal variations in leaf area as well as annual-decadal changes in vegetation composition. The sentence will be adjusted to clarify this in the revised manuscript.

Changes: The sentence has been changed to: BVOC emissions are closely linked to leaf as well as ecosystem developments. Simulating vegetation seasonal variations in leaf area as well as vegetation compositions enables us to assess the model performance in representing short-term emission changes in response to T and PAR, as well as long-term changes in vegetation development and distribution.

The model/data comparison would also really benefit from a deeper analysis. If isoprene and monoterpene emission estimates fall into the data values, it is however difficult to come to a clear conclusion, as data are not that numerous, and as model estimates are given either as daily average or for noon. At what time were emission data collected and how are they compiled for model-data comparison?

Response: Thanks for the great points. Model-data comparison will be further analysed by adding statistics. For each data point, it is an average of six replicates in the field which were measured at different time points of a day (between 9 - 17). Since the model is running at daily time step, it is not possible to average the modelled emission rates at sampling time. We saw this limitation and therefore used the daily average and maximum as an indication about the model's performance.

Changes: Please see the added statistic equations 5 and 6 on Page 10 and the numbers in the result section (Page 12, lines 14-15 and Page 13, lines 13-14). The clarification about why we use both daily average and noon have been added on Page 9, Lines 28-30.

The parameterization is calibrated and adjusted in order to better represent BVOC emission from Arctic plants. This is a crucial and one major contribution of this work and yet it is only very quickly mentioned in section 4.2. It is important to add a more detailed and quantitative analysis of the emission improvement, both in the results section and in the discussion part.

Response: Thanks for point out. We agreed with the reviewer that we should discuss the derived new temperature curve in a more detail. We will add more discussion regarding to parameterizing T response for arctic plants.

Changes: In the result section, the comparison between the original T curve and the new one has been quantified by using scatter plot (see Figure 6). In the discussion section, we have added some explanation and discussion of this strong T response (using the adjusted T curve) (Page 17, Lines 2-7). The associated high T during sunny days was explained in the one sentence before. Also, the uncertainties associated the data as well as method we used for deriving the T curve have been added on Page 17, Lines 25-27.

Specific comments:

Page 1, line 23: change "the model's responses" to "the model responses"

Response: changed.

Page 1, line 23: change "higher levels' warming" to "higher warming levels"

Response: changed to higher levels of warming.

Page 2, line 4: change "Â'IPFT's reponses" to "PFT responses"

Response: changed to "physiological responses of PFTs"

Page 2, line 6: change "Biogenic volatile organic compounds (BVOC)" to "Biogenic volatile organic compounds (BVOCs)"

Response: Through the manuscript, we use BVOC as a plural term.

Page 2, line 11: change "atmosphere's oxidative capacity" to "atmosphere oxidative capacity"

Response: Changed.

Page 2, line 15: change "(... respectively (Sindelarova et al., 2014))" to "(... respectively; Sindelarova et al., 2014)"

Response: Changed.

Page 3; line 29: change "and to advance our understandings of the" to "and our understanding regarding"

#### Response: changed.

Page 3, line 30: remove comma in "ecosystem model, LPJ-GUESS"

Response: we will remove comma and add full name for the model.

Changes: the full name has been added: Lund-Potsdam-Jena General Ecosystem Simulator

Page 7, line 28: please change "LAI of the year 2006 and 2007" to "LAI of the years 2006 and 2007"

Response: changed.

Page 7, line 33, change "due to plants' adaptation" to "due to plant adaptation"

#### Response: changed.

Page 8, line 20-21: change "Due to lacking of data about the daily maximum" to "Due to the lack of data regarding the daily maximum"

Response: changed.

Page 9, line 26: change "to assess the model's performance" to "to assess the model performance"

Response: changed.

Page 15, line 11: change "the model's ability" to "the model ability"

Response: changed.

Figures:

It is hard to distinguish the observations from the emission estimates. Could you please trying using another color?

Response: We will modify the figures colors to make it contrast.

Changes: Figure 4 and 5 have changed colours and Figure 6 has changed from bar plot to scatter plot.

# Reference

Arneth, A., Niinemets, Ü., Pressley, S., Bàck, J., Hari, P., Karl, T., Noe, S., Prentice, I. C., Serça, D., Hickler, T., Wolf, A., and Smith, B.: Process-based estimates of terrestrial ecosystem isoprene emissions: Incorporating the effects of a direct CO2-isoprene interaction, Atmospheric Chemistry and Physics, 7, 31-53, 2007.

Peñuelas, J. and Staudt, M.: BVOCs and global change, Trends in Plant Science, 15, 133-144, 2010.