

**Dear Editor and Reviewers,**

We appreciate your time and efforts for commenting on this manuscript. As both the reviewers have correctly pointed out, the comparison between model and observations was not done in the best possible way. We have solved this issue in the revision with a recalculation of emissions with the actual conditions at the time of sampling.

The other points brought up by the reviewers were also addressed, and responses to the individual comments can be found below (with grey background). The page and line numbers mentioned in the replies refer to the ones in the revised manuscript (without marked-up changes). We hope to have addressed your comments satisfactorily.

Thanks again for your great contributions.

**Best regards, on behalf of all co-authors,**

**Jing Tang**

## Replies to the 1<sup>st</sup> reviewer:

Although many comments and suggestions have been acknowledged and considered, I am sorry to say that I am not quite convinced about the argumentation not to change a thing about noon-calculations and LAI representation. This now refers to the following parts of the manuscript:

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P6, L11: The seasonality function only applies to isoprene production although (L14) the model assumes that both isoprene and monoterpenes are produced in the same pathway and respond in the same way to CO<sub>2</sub>. This is an inconsistent approach. Either the seasonality should apply on monoterpenes too or the CO<sub>2</sub> response cannot be applied on (light dependent) monoterpenes.

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Reply: Thanks for pointing out this inconsistency. We have now revised our model and applied the same seasonality function to monoterpene production. The Materials and methods section has been edited correspondingly (P6, L9-10). The impact of this change on daily and annual monoterpene emissions appeared to be very small. It plays a role only at the beginning and the end of the growing season, when emissions are generally low. The seasonality function regulates emissions of deciduous plants only, as is the case for isoprene.

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It is not clear from the description but do coniferous and herbaceous PFTs light-dependent and light-independent fractions at the same time? I guess not because then I would like to know how the epsilon<sub>s</sub> parameter is derived from measurements (Had there been an a-priori assumption about the differentiation? Have I missed this piece of information?). Please clarify in the text.

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Emissions from storages should depend on average daily temperature, not on daytime temperature. Therefore, the T values in Eq. 3 and 4 should not be the same. Has this been considered?

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Reply: In the model, BVOC synthesis uses a fraction of the photosynthetic electron flux (J in Eq. 1), and synthesized terpenoids are either entirely emitted directly (e.g. isoprene), or part of the production is stored. The latter is the case for coniferous and herbaceous PFTs, where 50% of the produced monoterpenes are put into the storage pool, from which emissions are computed based on the pool size and a temperature dependence (Eq. 4). Simulated monoterpene emissions originate hence from a combination of light-dependent and light-independent sources.

The use of 50% to distribute between emission and storage originates from Schurgers et al. (2009), and similar values were obtained from  $^{13}\text{CO}_2$  labelling by Ghirardo et al. (2010) for coniferous trees. We have not made an attempt to distinguish between light-dependent and light-independent emissions from the samples taken in this study; the strong correlation between temperature and light would require a different setup (e.g. measurements in darkness) to do this correctly. This remains a weakness of the current study, which has been addressed in the discussion (P14, L16-20).

The reviewer is correct that daily mean temperatures should be used for emissions from storage. In the model, this has actually been done correctly. We have now corrected Eq. 4 and the relevant text to show that daily temperature is used (P6, L6 and L16-18, Eq. 4).

P9, L4: I am not convinced from the response regarding using only average PAR for calculating emissions because ‘the Haxeltine and Prentice approach only describes daily photosynthesis’. Why should it not be possible ‘to compute an instantaneous flux at noon’ all the more why can an artificially calculated flux (that might be empirically increased to a reasonable degree if not calculated) not be fed into the photosynthesis model to check its response? If this would be a reason, the use of maximum temperature to calculate an upper limit for emission would also not be valid.

Reply: We agree with the reviewer that our previous attempts to compare model and observations contained unrealistic assumptions, and that the use of daily maximum air T was not good enough. We have revised our model-data comparison now, and have come up with a better solution using measured T inside the enclosure and ambient PAR values at the time of sampling, rather than using the daily climate data. In the model, the measured canopy air T and PAR were only used for re-estimating the photosynthesis fluxes and BVOC emissions at the time of sampling (for direct comparison with the observations), and these computations did not have any impact on the long-term simulation of vegetation dynamics and daily/annual BVOC emission rates. The new inputs and methods have been described on P7, L7-8 and P9, L8-12. The old Figures 4 and 5 have now been updated and integrated into one (Fig. 4, P27) with the re-estimated emission rates and WR. The result section has been changed accordingly with the new estimation of daily emission rates and WR using the measured T and PAR (P11, L1-23).

In this process of revision of the model-data comparison, we made the decision to exclude the observed monoterpene emissions in 2010 from the current paper. This was done because of gaps in the monoterpene, enclosure temperature and ambient PAR data for these measurements. In 2010, technical problems had prevented analysis of isoprene (Valolahti et al., 2015), and we have decided to take a conservative approach and not use the corresponding monoterpene data either. This enables integration of Figs 4 and 5 into one, and similar sets of observational data for both isoprene and monoterpenes.

P10, L13/14: I don't see any valid response, explanation or consideration of the fact that the modelled and measured LAI values are far off for most of the vegetation types. In figure 3, the forbs/lichens type (CLM) LAI is about 0.5 measured and 0.25 modelled, the evergreen shrubs (LSE+EDPS) are presented with a measured LAI of almost 0.4 while the model gives 0.2 (note the different axis). So how can this be called 'captured by the model'? Or was there a kind of mix up with the axis?

Reply: We can see that the large difference in measured vs. observed LAI was still poorly explained in the manuscript, and we have taken several measures to clarify and better acknowledge the issue. We can identify several reasons for the large discrepancy, which are explained below. The choice of different axis scales was not a mixed up, but a deliberate choice. They were chosen to allow for comparison of the effects of warming on the coverage of the different PFTs in the observed and modelled data. We can see that the reason was not clearly articulated, so this has been done now both where the figure is explained and in the figure legend (also see below).

In the Materials and methods, we explain the basic difference in LAI and the point-intercept-based coverage, i.e. that they "are not comparable one-to-one throughout growing seasons, since the measurement includes pin hits on different plant parts, whereas LAI only explains leaf coverage. However, the point-intercept-based coverage approaches leaf coverage when the deciduous leaves become fully developed during the growing season."

We have now edited the text in the Results (P10, L13-17) to clearly acknowledge the large underestimation of the PFT coverage by the modelled LAI compared to the observed point-intercept-based coverage. We have also added a reason for using the different axis scales "note different left and right axis scales in Fig. 3 to allow comparison of relative changes in response to warming" (P10, L14-15). This has also been amended in the legend of Fig. 3 to explain the use of different scales.

5 We also clearly acknowledge the mismatch in the discussion, already in the first sentence: "...in spite of the poor representation of the observed vegetation composition". We have edited the second sentence (P12, L26-29) to explain the most probable reason to the mismatch: "...LAI only includes the areal coverage by leaves, whereas the point intercepted-based vegetation coverage also includes coverage detected of other aboveground plant parts, like stems." The other reasons contributing to the difference (underestimation of the allocation of assimilated carbon to foliage in LPJ-GUESS and/or too low SLA values plus methodological issues in the point-intercept technique) are explained on P12, L29-31, P13, L1-5 and P13, L13-15, respectively.

10 In addition, you might consider to modify the following:  
P7, L32: Since evaluation and validation are different terms, and what has been done here is clearly 'evaluation', this should not be mixed up in the headline (in short: change validation into evaluation). (Check also throughout the manuscript.)

15 Reply: Thanks for pointing this out. We agree and have now replaced all terms "validation" with "evaluation".

## Replies to the 2<sup>nd</sup> reviewer:

I appreciate the many changes the authors have made to improve the manuscript in response to the two reviewers' comments, but I still have two major concerns. These were both concerns that I raised during my original review which I feel have not been adequately addressed.

- 5 First, and more importantly, I am still unsatisfied with the comparison of modelled daily means and maxes with the spot measurements. I see that my concerns were also shared by the other reviewer. I do appreciate that the authors have moved past the very qualitative comparison that was employed in the first version of the manuscript. But, the authors are using a model that produces only one estimate of BVOC emissions per day, and it's difficult to compare this to a relatively small number of  
10 measurements. My view is given the difficulty of acquiring the field data, these measurements are useful and I understand I can't demand the perfect dataset with more complete coverage. On the other hand, modeling is much more flexible, and more sophisticated modeling approaches can be applied. The authors can run a series of different simulations using different weather inputs. For example, they could apply a Monte Carlo approach that can give information about both mean responses and variance. This  
15 is a much more appropriate response than simply doing two runs with the mean and max values. I would be more satisfied if the authors could cite a previous study that used only daily means and maxes in a similar comparison.

- 20 Reply: Thanks for the suggestions. We have now revised the comparison between model and data and have come up with a solution using observed air T in the enclosure and ambient PAR at the time of sampling to re-compute the emission at the time of sampling (see new descriptions in Methods section, P7, L7-8 and P9, L8-12 and new figure 4 on P27)., which is the most accurate representation of expected emissions. In the model, the measured air T inside the enclosure and PAR were only used for re-estimating the photosynthesis fluxes and BVOC emissions at the time of sampling, and these computations did not have any impact on the long-term simulation of vegetation dynamics and  
25 daily/annual BVOC emission rates.

Second, and I understand this is a more minor and somewhat picky point, but the authors should more carefully read Monson et al 2012, which they cite on page 3 of the new manuscript. In particular, see section XI (Conclusions) in that reference. The main point is that so-called 'mechanistic' models, based

on the Niinemets approach, have an empirical gap due to the lack of knowledge about a number of critical processes. Again, I encourage the authors to add more nuisance to their comparison of the modelling approaches. Since the authors are not bringing anything new to the table in terms of the comparison of mechanistic vs empirical models, I don't understand the necessity of disparaging empirical models. The authors can simply state what they are using, and move forward.

Reply: Thanks for pointing this out. We have now clarified what the model can simulate in a more mechanistic way (vegetation dynamics and long-term response) and what this means for simulating BVOC emission). We also took away the unnecessary comparison between different modeling approaches, see P3, L12-14.

## References

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- Valolahti, H., Kivimäenpää, M., Faubert, P., Michelsen, A., and Rinnan, R.: Climate change-induced vegetation change as a driver of increased subarctic biogenic volatile organic compound emissions, *Global Change Biology*, 21, 3478-3488, 10.1111/gcb.12953, 2015.

# Challenges in modelling isoprene and monoterpene emission dynamics of arctic plants: a case study from a subarctic tundra heath

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**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  
15 of minimal BVOC emissions from the Arctic are based on very few observations and have been challenged ~~by~~ increasingly  
~~by~~ field data. This study applied a dynamic ecosystem model, LPJ-GUESS, as a platform to investigate short-term and long-term BVOC emission responses to arctic climate warming. Field observations in a subarctic tundra heath with long-term (13 years) warming treatments were extensively used for parameterizing and evaluating BVOC related processes (photosynthesis, emission responses to temperature and vegetation composition). We propose an adjusted temperature (T) response curve for  
20 arctic plants with much stronger T sensitivity than the commonly-used algorithms for large-scale modelling. The simulated emission responses to 2 °C warming between the adjusted and original T response curves were evaluated against the observed warming responses (WR) at short-term scales. Moreover, the model responses to warming by 4 °C and 8 °C were also investigated as a sensitivity test. The model showed reasonable agreement to the observed vegetation CO<sub>2</sub> fluxes in the main growing season as well as day-to-day variability of isoprene and monoterpene emissions. The observed relatively high  
25 ~~emission rates of BVOC as well as isoprene~~ WR were better captured by the adjusted T response curve than by the common one. During 1999-2012, the modelled annual mean isoprene and monoterpene emissions were 20 and 8 mg C m<sup>-2</sup> yr<sup>-1</sup>, with an increase 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, but the impacts on monoterpene emissions levelled off at the last few years. At hour-day scale, the WR seem to be strongly impacted by canopy air T-leaf T; while at day-year scale, the WR are a  
30 combined effect of plant functional type (PFT) dynamics and instantaneous BVOC responses to warming. The identified challenges in estimating arctic BVOC emissions are: (1) correct leaf T estimation; (2) PFT parameterization accounting for



plant emission features as well as physiological responses to warming; and (3) representation of long-term vegetation changes in the past and the future.

## 1 Introduction

Biogenic volatile organic compounds (BVOC) are reactive hydrocarbons mainly emitted by plants. Emissions of these secondary metabolites are involved in plant 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 atmospheric oxidative capacity and therefore lengthen the lifetime of methane (CH<sub>4</sub>), 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<sub>3</sub>) concentration when the ratio of BVOC to NO<sub>x</sub> (BVOC/NO<sub>x</sub>) is high (Hauglustaine et al., 2005), and increase 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<sub>3</sub> (Hauglustaine et al., 2005). Global estimates of non-methane BVOC emissions are in the range of 700-1000 Tg C yr<sup>-1</sup>, of which isoprene and 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<sub>4</sub> (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-~~uding~~ soil water availability and the spectrum of the incoming light, abiotic and biotic stress) 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; Valolahti et al., 2015; Kramshøj et al., 2016; 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 warming-induced 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-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. 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; Grote et al., 2014). In the short-term (hours-days), the responses to Q and T of isoprene and monoterpene production 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 is affected by vegetation composition changes (Faubert et al., 2011; Valolahti et al., 2015), vegetation phenology (Staudt et al., 2000; Hakola et al., 2006), past weather conditions (Ekberg et al., 2009; Guenther et al., 2012) and 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). ~~Here, we use In comparison with empirical models (Guenther et al., 1995; Guenther et al., 2006; Guenther et al., 2012), a process-based ecosystem model s, to explicitly representing BVOC synthesis activities and emissions. The model, can simulate vary with species dynamically simulate vegetation composition dynamically and represents as well as~~ long-term growing environment effects, ~~and could thus be more and is thus~~ 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  $\mu\text{mol m}^{-2} \text{s}^{-1}$ ) 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 ~~that~~ the G93 algorithm characterized emissions well with the T response, but not Q response. However, Ekberg et al. (2009) found that the 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 arctic BVOC emissions (Arneth et al., 2011; Guenther et al., 2012; Sindelarova et al., 2014). These need to be included as a trait of plant functional types (PFTs), especially when 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.

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 regarding emission dynamics for arctic ecosystems in a warming future. The process-based dynamic ecosystem model LPJ-GUESS (Lund-Potsdam-Jena General Ecosystem Simulator) (Smith et al., 2001; Smith et al., 2014) was used as a platform to

simulate short-term and long-term responses of BVOC emissions to changes in climate for arctic plants. The model links isoprene and monoterpene production 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 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 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; (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/graminoid heath tundra located in Abisko, northern Sweden (68°21'N, 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 combined warming and litter addition (WL) treatments (Rinnan et al., 2008). In the current study, we only focused 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).

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. The enclosure covered a 20 × 20 cm area in each plot. The air T inside the enclosure and PAR in ambient conditions were measured during the sampling. The isoprene emission datasets For 2006-2007, the datasets for isoprene emission can be found in Tiiva et al. (2008) and those for monoterpenes in Faubert et al. (2010). ~~and for 2012 in Valolahti et al. (2015).~~ For the year 2012, ~~isoprene emission rates were not analyzed due to technical problems (Valolahti et al., 2015).~~ isoprene and 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<sub>2</sub> fluxes were measured in the same area for 2006, 2007, 2010 and 2012 during the same years (data from 2006 and 2007 were published in Tiiva et al. (2008), whilst data from 2010 and 2012 have not been published) (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 × 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.

## 2.2 LPJ-GUESS

### 2.2.1 LPJ-GUESS general framework

LPJ-GUESS is a climate-driven dynamic ecosystem model with mechanistic 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 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) 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). 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  $CO_2$  uptake ( $\alpha_{c3}$ ), the  $CO_2$  compensation point ( $\Gamma^*$ ) and the internal partial pressure of  $CO_2$  ( $p_i$ ) (Collatz et al., 1991; Haxeltine and Prentice, 1996b).  $J_C$  is related to the maximum catalytic capacity of Rubisco per unit leaf area ( $V_m$ ),  $\Gamma^*$ ,  $p_i$  and the Michaelis-Menten constant for  $CO_2$  and  $O_2$ . 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. The productions of isoprene ( $E_I$ ) and monoterpenes ( $E_M$ ) are computed as:

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

where  $J$  is the rate of photosynthetic electron transport and  $\alpha$  converts photon fluxes into terpenoid units. The synthesis of both compounds is linked to  $J$  (Niinemets et al., 1999; Niinemets et al., 2002) and a fraction ( $\epsilon$ ) of the electron transport contributing to terpenoid production (Eq. 2) is determined from a plant-specific fraction under standard conditions ( $\epsilon_s$ , usually at a T of 30 °C and a PAR of 1000  $\mu\text{mol m}^{-2} \text{s}^{-1}$ ) which is adjusted for leaf T, seasonality ( $\sigma$ ), and atmospheric  $CO_2$  concentration:

$$\varepsilon = f(T)f(\sigma)f(\text{CO}_2)\varepsilon_s \quad (2)$$

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 (standard T and PAR) 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 for terpenoid synthesis, which is higher than that for photosynthesis:

$$f(T) = e^{\alpha_\tau(T-T_s)} \quad (3)$$

The parameter  $\alpha_\tau$  represents the T sensitivity and the standard temperature ( $T_s$ ) is often 30 °C (adjusted to 20 °C in this study). In the model, daily mean T ( $T_d$ , 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. For the study in the Subarctic, the often-used reference  $T_s$  of 30 °C as well as the T responses ( $\alpha_\tau$ ) were adjusted based on the observation data and will be discussed below. The seasonality function,  $f(\sigma)$ , ~~only was applies-applied~~ to both isoprene and monoterpene production and is based on a degree-day ~~method-sum~~ in Spring, ~~T~~ and a daylength thresholds in Autumn (Arneth et al., 2007; Schurgers et al., 2009). The atmospheric CO<sub>2</sub> concentration enhances terpenoid synthesis when the concentration is lower than ambient, and vice versa, which is represented by the function  $f(\text{CO}_2)$  (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 ( $E_{Ms}$ ) is a function of ~~-T~~  $T_d$  and  $m$  with an average residence time ( $\tau$ ).  $\tau_s$  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 of terpenoid synthesis). The residence time  $\tau$  is adjusted based on the standard condition  $\tau_s$  for  $T_d$  responses with a Q<sub>10</sub>-relationship.

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

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

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). ~~-~~The calculation of leaf T in the model was based on solving the leaf energy balance, where the incoming shortwave and longwave radiation ~~is-are~~ 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 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. 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.

## 2.3 Simulation setup

### 2.3.1 Input data

5 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 01/01-30/06/1984, 09/06-16/06/2016, 13/02-15/02/2007, 23/07-17/08/2011) were filled 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  
10 (<http://cdiac.esd.ornl.gov/trends/co2/contents.htm>). The air T inside the enclosure and ambient PAR at canopy level were also used as the model inputs for each measuring day- (Tiiva et al., 2008; Faubert et al., 2010; Valolahti et al., 2015).

### 2.3.2 Plant functional types

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.,  
15 2008; Miller and Smith, 2012; Tang et al., 2015), but 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  
20 through adjusting parameters controlling vegetation height. The PDS-type was further divided into two PFTs with evergreen and deciduous phenology. Moss, widely appearing 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.

In LPJ-GUESS, the crown 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 thickness of this layer was reduced to 10 cm in this study to better capture the  
25 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. The details about the data sources for parameterizing emission capacity at 30 °C ( $E_{IS30}$ ) and 20 °C ( $E_{IS20}$ ) can be found in  
30 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~~evaluation

The modelled CO<sub>2</sub> fluxes, LAI as well as the BVOC T response were first calibrated before evaluating the modelled daily BVOC emission rates. Two out of four years' (2006 and 2007) measured net ecosystem production (NEP), ecosystem respiration (ER) and estimated gross primary production (GPP) as well as point intercept-based species composition were used for calibrating. The data for the other two years (2010 and 2012) were used for evaluating ~~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 ( $\alpha_{c3}$ ) 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  $\alpha_{c3}$  is the most influential parameter in terms of the simulated vegetation carbon fluxes. Also, a pre-~~evaluation~~validation of the modelled CO<sub>2</sub> fluxes with the observations in this study using the default  $\alpha_{c3}$  value (0.08) has found a large overestimation of both GPP and ER (not shown). Therefore, a sampling of  $\alpha_{c3}$  (using the range of 0.02 to 0.125  $\mu\text{mol CO}_2 \mu\text{mol photons}^{-1}$ , 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 with the simulated CO<sub>2</sub> fluxes and vegetation composition using the observed CO<sub>2</sub> fluxes and the point intercept-based plant coverage data from 2010 and 2012, respectively.

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 plant 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 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 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 variations in the atmospheric CO<sub>2</sub> concentration -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 ( $\alpha_t$  in Eq. 3) were determined (Fig. 1). An adjusted  $\alpha_t$  value of 0.23 was chosen after fitting all the data from July over three years' measurements. Apart from the low R<sup>2</sup> value for block 1, the data were well captured by the exponential shape (R<sup>2</sup>  $\geq$  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 emission rates in the model. This adjusted T



response was also evaluated with the observed enclosure air T and monoterpene emission rates in July ( $R^2 = 0.66$  for all blocks).

The abundance of each PFT was evaluated using simulated LAI against the point intercepted-based ~~species-vegetation~~ composition. The species were grouped into the corresponding PFTs for comparison 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. ~~As the point-intercept-based species abundances and LAI cannot be compared one-to-one throughout growing seasons, comparison since the measurement could include pin hits on different plant parts, but whereas LAI only explains leaf coverage. However, the point-intercept-based measured coverage could become close to approaches leaf coverage when the deciduous leaves become fully developed during the growing season the relative differences in abundances between PFTs are similarly described by both measurements.~~

After calibrating the modelled  $\text{CO}_2$  fluxes and LAI, the modelled isoprene and monoterpene emission rates were compared with the observations. The simulated daytime average emissions ~~represented as daytime average values~~ ( $\mu\text{g C m}^{-2} \text{ h}^{-1}$ , daytime emission rates divided by day length) ~~may do~~ not allow an accurate comparison with the actual-observed emission rates, which were typically ~~conducted-obtained~~ in the middle of the day (between 9 am – 5 pm). Therefore, an additional estimate of the emission rates ~~noon (maximum hourly emissions) were also estimated for the conditions prevailing during the sampling was made. This was done by computing an additional computation of the emission, through applying the leaf the measured air T inside the enclosure and PAR during the sampling time computed from the daily maximum air T for photosynthesis and BVOC emissions. This computation was performed twice: Additionally, the modelled emission rates for each measuring day once using the original T response ( $\alpha_T = 0.1$ ,  $T_S = 30^\circ\text{C}$ ,  $E_{J30}$  and  $E_{M30}$ , Eq. 3) and once with the adjusted T response ( $\alpha_T = 0.23$ ,  $T_S = 20^\circ\text{C}$ ,  $E_{J20}$  and  $E_{M20}$ , Eq. 3 and Fig. 1) were compared.~~ 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, 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 ( $O_i$ ) and a value close to 1 indicates a good agreement. The index B estimates the mean deviation between the modelled and observed values (Willmott et al., 1985) and values close to 0 indicates models' good agreement to observations.

$$A = 1 - \frac{\sum_{i=1}^N |E_i - O_i|}{\sum_{i=1}^N (|E_i - \bar{O}| + |O_i - \bar{O}|)} \quad (\text{Eq. 5})$$



$$B = \frac{\sum_{i=1}^N (E_i - O_i)}{N} \quad (\text{Eq. 6})$$

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

### 2.3.4 Effect of warming

To simulate the observed warming responses from the OTCs, a 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, difference between C and W treatments) using the original T response ( $\alpha_t = 0.1$ ,  $T_s = 30^\circ\text{C}$ ,  $E_{130}$  and  $E_{M30}$ , Eq. 3) and the adjusted T response ( $\alpha_t = 0.23$ ,  $T_s = 20^\circ\text{C}$ ,  $E_{120}$  and  $E_{M20}$ , Eq. 3 and Fig. 1) were also compared with the observed WR. Furthermore, additional simulations with a warming by 4 °C and 8 °C, reflecting the range of climatic projections in this region (IPCC, 2013), were also conducted to test for the anticipated ecosystem-scale responses to different levels of warming.

## 3 Results

### 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  $\alpha_{c3}$ , which describes the efficiency in converting solar radiation to carbohydrates, and which was varied between 0.02 to 0.125  $\mu\text{mol CO}_2 \mu\text{mol photons}^{-1}$  following Pappas et al. (2013) (Fig. S1). For CO<sub>2</sub> fluxes, the lowest root mean square error (RMSE) values occurred at 0.035  $\mu\text{mol CO}_2 \mu\text{mol photons}^{-1}$  for GPP and ER, while the lowest RMSE value for LAI was 0.051  $\mu\text{mol CO}_2 \mu\text{mol photons}^{-1}$  when comparing with the observations for 2006 and 2007. A value of 0.040, consistent with the study by Shaver et al. (2013) was selected for  $\alpha_{c3}$  to limit the RMSE values of the modelled CO<sub>2</sub> fluxes and LAI. Using this value for  $\alpha_{c3}$ , the model captured the observed day-to-day variations as well as the magnitude of the chamber-based GPP, ER and NEP for 2010 and 2012, with an overestimation of CO<sub>2</sub> fluxes (particularly for the early growing seasons, Fig. 2), and a largen underestimation of LAI (Fig. 3). For the year 2012, the model showed large overestimations of the observed GPP and ER for the limited number of measurements in this growing season.

For the 5 PFT groups, the modelled growing season LAI values for 2010 and 2012 were generally-much lower than the point intercept-based coverage estimations from the field observations (note different left and right axis scales in Fig. 3 to allow comparison of relative changes in response to warming), except for the *Salix*-type summergreen shrubs and deciduous prostrate dwarf shrubs (SLSS+SPDS). The two-dominance of two vegetation groups in the C plots, forbs/lichens and evergreen shrubs, were was consistent between the modelled and the observed-captured by the model. However, the coverage of graminoids (GRT) and non-Salix-type deciduous shrubs (NSLSS) was underestimated by our model.

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 (Fig. 3). For the two groups of shrubs (NSLSS and LSE+EPDS), the modelled increase is in agreement with the observations. However, 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.

### 3.2 Modelled BVOC emissions

BVOC emissions are closely linked to leaf as well as ecosystem development. Simulating seasonal variation in leaf area and vegetation composition 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 seasonal variations of the modelled daily BVOC emissions as well as the span of all BVOC samplings over four-three years are presented in Fig. S2.

#### 3.2.1 Daily emissions

- Emission rates in the control (ambient) conditions

The observed air T and PAR showed day-to-day variations through the sampling periods (Fig. 4e), which resulted in strong daily variations in the observed BVOC emissions (Fig. 4a and 4c). These observed ~~observed daily~~ variations in isoprene and monoterpene emissions were generally captured by the model (Fig. 4) for 2006 and 2007. For the year 2012, the model overestimated both isoprene and monoterpene emission rates over the three sampling days. The observed isoprene emission rates (Fig. 4a) lay between the modelled daytime average and daily noon emission rates, with the exception of a few days with much lower simulated noon emission rates than the observed (22/08/2006, 10/07/2007 and 05/08/2007). For these dates, the observed chamber air T were higher than the modelled daily noon leaf T (squares in Fig. 4c) and 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. Noticeably, the model used air T at 2 m height from the ANS station to extrapolate the leaf T for estimating daily BVOC emissions (Fig. S2), while the measured T is the observed air T and PAR during the sampling hours were used for modelling emissions to directly compare with the observed. The modelled high emission rates for a few days (e.g., 10/07/2007, 14/06/2012) were directly linked to the observed high T and PAR (Fig. 4e). Averaging over all measuring days in 2006 and 2007, the modelled and observed isoprene emission rates were 46.6 and 34.7  $\mu\text{g C m}^{-2} \text{ h}^{-1}$ , and the modelled and observed monoterpene emission rates were 8.5 and 5.3  $\mu\text{g C m}^{-2} \text{ h}^{-1}$ , respectively. For the year 2012, the modelled emission rates (80.4 and 14.9  $\mu\text{g C m}^{-2} \text{ h}^{-1}$  for isoprene and monoterpenes, respectively) were much higher than the observed (9.1 and 0.5  $\mu\text{g C m}^{-2} \text{ h}^{-1}$ , for isoprene and monoterpenes, respectively). Over three growing seasons, the observed air T inside the chambers was on average 7.2 °C warmer than the modelled daytime average leaf T and 3.4 °C warmer than the modelled daily noon leaf T. The modelled daytime average, daily noon and the observed daytime emission rates were 9.1, 25.8 and 25.5  $\mu\text{g C m}^{-2} \text{ h}^{-1}$ , 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 daytime average emission rates in the C plots (light grey bars in Fig. 5a) showed 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. 5a). Over four sampling growing seasons, the modelled daytime mean, daily noon and observed daytime emission rates were  $2.4$ ,  $7.9$  and  $2.5 \mu\text{g C m}^{-2} \text{h}^{-1}$ , respectively (all numbers averaged for the days on which measurements were made). The large overestimation by the model in the year 2012 was also seen for GPP and ER (Fig. 2). The modelled daytime mean showed better agreement 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 daytime 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 ( $1.3$ ,  $8.1$ ,  $0.3$ ,  $0.5 \mu\text{g C m}^{-2} \text{h}^{-1}$ , for the four years measured, respectively), the simulated daily noon emissions were more similar between years ( $2.2$ ,  $3.0$ ,  $2.1$  and  $2.4 \mu\text{g C m}^{-2} \text{h}^{-1}$ , respectively).

- Emission responses to  $2^\circ\text{C}$  warming

In response to  $2^\circ\text{C}$  warming by the OTCs, the modelled leaf T increased on average by  $2^\circ\text{C}$ , while the observed chamber enclosure air T in the W plots increased by  $2.14.8^\circ\text{C}$  relative to the higher than that in the C plots averaged over the four three growing seasons with observations. For isoprene, the modelled-observed magnitudes of WR (Fig. 4b) were generally quite close to lower than the observed WR captured reasonably well by the model, except for 5 August 2007 especially for a few days with strong observed WR. For this day, the air T in the W was higher than in the C plots, but the PAR value was lower in the W than in the C plots (Fig. 4e). Averaging over three years, the modelled-simulated daytime average, noon and the observed isoprene WR were  $19.65.7$ ,  $15.2$  and  $28.4 \mu\text{g C m}^{-2} \text{h}^{-1}$ , respectively. and Warming increased the observed isoprene emissions by  $1195\%$ ; but only increased the model-modelled emissions by  $37\%$  simulations overestimate this by a warming effect of  $37\%$  led daytime average isoprene emission rates by  $63\%$ , the daily noon by  $59\%$  and the observed emissions by  $1165\%$  (using the averaged WR divide with the averaged emissions all numbers averaged for the days on which measurements were made). Over three years, the observed strong WR for a few dates (e.g., 22/08/2006, 10/07/2007, 05/08/2007 and 14/06/2012) were underestimated by the modelled noon WR when the observed chamber air T in the C was close to or higher than  $20^\circ\text{C}$ , 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  $20^\circ\text{C}$  (e.g., 13/06/2006, 06/07/2007), the observed WR were higher than the modelled daily average, but lower than the modelled daily noon WR.

For monoterpenes, the modelled daytime average, noon and the observed WR were  $2.0$ ,  $6.06.1$  and  $42.05 \mu\text{g C m}^{-2} \text{h}^{-1}$ , respectively. The averaged WR from the modelled noon emissions were much higher than the observations. The modelled daytime average WR showed better agreement with the observations. For one day with extremely high WR (06/07/2007), the modelled noon WR better captured the strong responses. Averaging over four three growing seasons, warming increased the

observed monoterpene emissions by 93%, and the modelled ~~daily average monoterpene~~ emission by 81 %, ~~the daily noon emission by 76 % and the observed emission by 98-63 %~~ (all numbers averaged for the days on which measurements were made).:-

These modelled ~~daily noon~~ WR ~~using obtained with~~ the adjusted BVOC T response ( $\alpha_t = 0.23$ ,  $T_s = 20$  °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. ~~6a5a~~), the simulation using the adjusted T response showed a substantial increase ~~of in~~ the modelled WR as well as a better agreement ~~to with~~ the observations ( $A = 1.1624$ ,  $B = -0.844.85$ ) than the simulation using the original T response ( $A = 1.47$ ,  $B = -275.2698$ ). The modelled WR using the original T response ~~generally largely~~ underestimated the observed high WR. Averaging ~~through over~~ three years, the ~~modelled~~ isoprene WR ~~modelled~~ using the original T response (~~used at a global scale~~) only gave 44.4 % of the observed WR, while the ~~modelled~~ WR ~~modelled~~ using the new T response captured 52-69 % of the observed WR (~~using the modelled average WR to divide with the observed average WR~~). For monoterpenes, ~~the modelled~~ WR modelled using the adjusted T response ( $A = 0.80$  and  $B = 2.13$ ) showed a moderate improvement as compared to using the original T response ( $A = 1.35$  and  $B = -2.83$ ). ~~†~~The modelled WR using the original T response (~~used at a global scale~~) ~~showed closer values~~ underestimated the observed WR by 72%, but the modelled WR using the adjusted T response overestimated the observed WR by 53%. ~~to the observations for the years with the observed low WR (2006, 2010 and 2012).~~ For the year 2007, the observed high monoterpene WR was ~~asere~~ better captured by the simulated WR with the ~~new adjusted~~ T response. ~~As for the modelled emission rates, the overestimation of the observed WR also mainly occurred in 2012. For both T responses, the modelled WR were generally lower than the observed WR for isoprene, but higher than the observed WR for monoterpenes.~~

### 3.2.2 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, this warming 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. ~~67~~). 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~~ three years with BVOC sampling, the modelled average WR were 58 % and 70 % for annual isoprene and monoterpene emissions, respectively. The modelled annual WR were of ~~the~~ similar magnitude as the modelled daily average WR (data not shown) 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 due to a decreasing 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 growth, 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.

## 4 Discussion

### 4.1 Emission rates

The modelled day-to-day variations of ecosystem CO<sub>2</sub> fluxes (Fig. 2) and BVOC emissions (Fig. 4) generally followed the observations, in spite of ~~deficiencies in the~~ poor representation of the observed vegetation composition (Fig. 3). The mismatch between the modelled LAI and the ~~point-intercepted-based-observed~~ vegetation coverage ~~may be~~ likely partly due to that LAI only includes the areal coverage by leaves, whereas the point-intercepted-based vegetation coverage also includes coverage detected of other aboveground plant parts, like stems. Further, the mismatch may also 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 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 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 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 limitation on plant development (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 shown to be important for capturing the emission dynamics in this heath tundra ecosystem. The development of parameterizations for arctic PFTs also requires 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. The 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 ~~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 crucial for depicting the dynamics of seasonal CO<sub>2</sub> fluxes (Tang et al., 2015). The overestimated GPP in the beginning of growing seasons (Fig. 2a) suggests uncertainties in modelling the time of 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).

5 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 0.1 % of the modelled GPP. The modelled emission rates are not only linked to the modelled photosynthesis fluxes, but also determined by the emission capacity assigned 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 observed relatively  
10 low emissions in comparison with lower latitudes (Arneth et al., 2011; Sindelarova et al., 2014) are mainly caused by low T and plant biomass, and not by low emission capacities (Holst et al., 2010).

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 as well as the overestimation of daily isoprene and monoterpene emissions of a few days. The observed low values of CO<sub>2</sub> fluxes (GPP and ER) and BVOC emissions in 2012 could be due to harmful effects of an insect outbreak in the nearby birch forest (Hanna Valolahti, personal observation). However, the potential impacts from insect outbreaks have not been explicitly included in the model. When both T and PAR were high (e.g., on 06/07/2007), the model tended to overestimate the emission rates, which could suggest that the stronger T sensitivity that was obtained in this study does not extend to these high temperature values. Furthermore, the estimated emission rates the T response in this study (Fig. 1) may be more robust for isoprene than for monoterpenes, because 1) the adjusted T response curve was only applied for monoterpene production, and there is a lack of data for evaluating T responses of monoterpene emissions from storage pools (Eq. 4); , which may also contribute to the uncertainties in annual emissions. Also, 2) there are more studies about supporting CO<sub>2</sub> inhibition on isoprene emissions (Arneth et al., 2007), but less than 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  
25 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 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  
30 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), ~~which could bias the evaluation, and the validation evaluation of the modelled emissions with these low rates could indicate that the modelled outputs were only~~

~~evaluated/validated for low emission rates, but was generally lack of evaluation of potentially high emission rates. More observations in the higher T range would enhance our confidence in the new T response function, specifically for monoterpenes. Furthermore, On one hand, the 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 %) (Schurgers et al., 2009). This split determines the distribution of monoterpene emissions over the year, since 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 evaluation periods may also indicate that the implemented storage residence time is too short (maybe larger storage size). The adjusted temperature response may not be 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 impacts also depend on the ecosystem emission rate (Niinemets et al., 2011). Furthermore, tTTThe current observations of BVOC emissions only covered the main growing season. Sampling over a longer season (Holst et al., 2010) we could help to improve the parameterization of the 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 mesocosms (Lindwall, Ghirardo et al., unpublished data) could also help to identify the fraction of monoterpene emissions from production or storage. Finally, the observed emission rates were measured at different time points of a day and the evaluation of daily emission with these time points observations are thus influenced by the diurnal dynamics of BVOC emissions, which has been found to be strong in the Arctic (Lindwall et al., 2015). On the other hand, the 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 in addition to potential gas concentration changes within the enclosure, may impact the plant physiological status. The impacts also depend on the ecosystem emission rate (Niinemets et al., 2011) as well as and sampling time of a day, considering the strong diurnal dynamics of BVOC emissions in the Arctic (Lindwall et al., 2015). The model evaluation using these half-hour-long samplings cannot avoid the influence of changed conditions inside the enclosure and of plant adaption to these conditions.~~

## 4.2 Responses to warming

The modelled increase of shrub coverage in response to the W treatment mostly followed the observations (Valolahti et al., 2015) and is consistent with the general trend in the Arctic (Wahren et al., 2005; Elmendorf et al., 2012). However, the observed increase of bryophytes is rather site-specific, and 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. 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 alterations, the short-term T responses of the vegetation are central for accurately

depicting daily BVOC emission responses to the W treatment. Through adjusting the BVOC T sensitivity (from  $\alpha_T = 0.1$ ,  $T_S = 30^\circ\text{C}$  to  $\alpha_T = 0.23$ ,  $T_S = 20^\circ\text{C}$  in Fig. 1), the simulated BVOC WR ( $19.6 \mu\text{g C m}^{-2} \text{h}^{-1}$  63% for isoprene and  $81\% \cdot 6.1 \mu\text{g C m}^{-2} \text{h}^{-1}$  for monoterpenes) became comparable to the observed responses ( $28.4 \mu\text{g C m}^{-2} \text{h}^{-1}$  145% for isoprene and  $98\% \cdot 4.0 \mu\text{g C m}^{-2} \text{h}^{-1}$  for monoterpenes).

The adjusted T response curve represents subarctic plants' isoprene emission responses to warming better than the original curve which has been parameterized for global simulations (Fig. 56). It further supports the earlier suggested stronger T sensitivity of BVOC emissions from arctic plants compared to plants from other regions (Ekberg et al., 2009; Holst et al., 2010; Rinnan et al., 2014; Kramshøj et al., 2016). 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  $40^\circ\text{C}$ , and the shape of the Guenther's response is very close to the exponential curve with  $\alpha_T$  value of 0.13 (using standard T of  $30^\circ\text{C}$ ) when leaf T is lower than 30 degrees. The high  $\alpha_T$  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; 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 both the emission rates at sampling hour and also daytime emissions in the model. The inconsistent different temporal resolution for estimating daytime emissions calls for further adjustment for of this T response for arctic plants.

The underestimation of ~~a few days's~~ strong isoprene WR on 5 Aug 2007 ( $157.8 \mu\text{g C m}^{-2} \text{h}^{-1}$ ) cannot be directly linked to the T and PAR differences between the C and W plots during the sampling time. The modelled emission at the C plot was 24% lower than the observed, caused by slightly different meteorological conditions during the sampling, but the modelled WR was 73.94% lower than the observed on this date. The observed strong WR could be linked to strong elevation of leaf T, could be partly attributed to the leaf T estimations derived 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. 4c). 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 leaf canopy T on a sunny day (Schollert et al., 2014; Lindwall et al., 2016b). This can directly elevate BVOC emissions as well as and WR (Lindwall et al., 2016a), and . ~~The observed strong decoupling of leaf T from 2 m air T for low-statured plants (Körner, 2003; Lindwall et al., 2016a). further indicates the current algorithms used for estimating leaf T in the model needs to adjust for arctic plants.~~ 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~~ leaf T. Also, plants acclimated to cold environment may drive larger emission responses once they are exposed to warmer T (Rinnan et al., 2014). The 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 identified after 7-8 years of warming in 2006 and 2007 (Tiiva et al., 2008; Faubert et al., 2010), which highlights 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. 7) clearly elucidated the combined effects of the direct responses to summer warming 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 represents subarctic plants' isoprene emission responses to warming better than the original curve which has been parameterized for global simulations (Fig. 6). It further supports the earlier suggested stronger T sensitivity of BVOC emissions from arctic plants compared to plants from other regions (Ekberg 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  $\alpha_*$  value of 0.13 (using standard T of 30 °C) when leaf T is lower than 30 degrees. The high  $\alpha_*$  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; 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.~~

### 4.3 Suggestions for further work

For extrapolating the current model developments to large-scale (regional) applications, we suggest 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 the strong dependence of BVOC emissions on leaf T (Lindwall et al., 2016a) point to a need for accurately capturing leaf T in models. Long-term parallel observations of both leaf and air T will be useful for the algorithm development 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), 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) The responses and/or acclimation of arctic PFTs to warmer climate should be better parameterized in the model to improve the representation of long-term vegetation effects on BVOC emissions.

## 5 Conclusions

This study has demonstrated the model's ability to depict the observed isoprene and monoterpene emission rates as well as daily variations in the BVOC emission of a subarctic tundra ecosystem. The modelled warming responses using a response curve adjusted for a stronger T response showed good agreements with the observations, especially for the days with the observed strong emission responses to warming. Short-term underestimations of the observed peak of WR ~~wasere~~ 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 process development and calibration. R. Rinnan contributed to the research questions, data collection and project financial support. 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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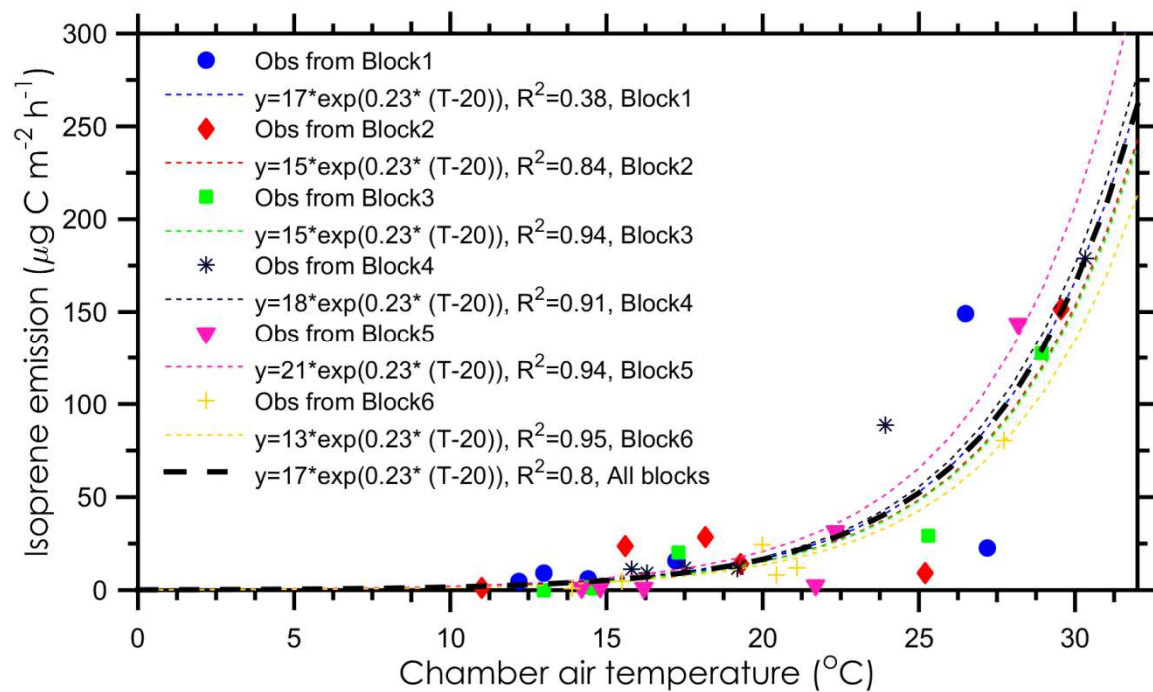
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Tables and figures

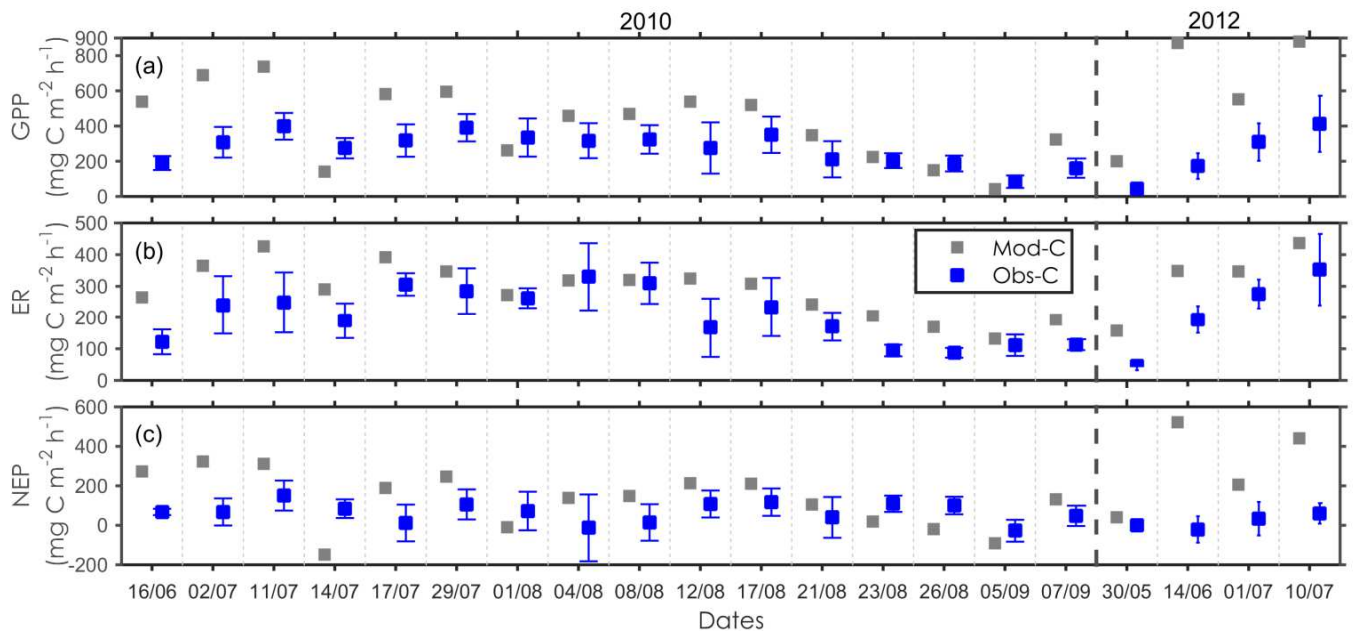
Table 1 Plant functional types (PFTs) and representative species in the study area. The emission capacity of isoprene ( $E_{IS}$ ,  $\mu\text{g C gdw}^{-1} \text{h}^{-1}$ ) and monoterpenes ( $M_S$ ,  $\mu\text{g C gdw}^{-1} \text{h}^{-1}$ ) at 20 °C (in bold and italics) using the adjusted temperature 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.

PFT	$E_{IS30}$	$E_{IS20}$	$E_{MS30}$	$E_{MS20}$	Representative species names
Low Shrubs Evergreen (LSE)	1.751	<b><i>1.737</i></b>	0.089	<b><i>0.088</i></b>	<i>Empetrum hermaphroditum</i> ; <i>Juniperus communis</i> ; <i>Vaccinium vitis-idaea</i>
<i>Salix</i> , Low Shrubs Summergreen (SLSS)	11.305	<b><i>11.213</i></b>	0.300	<b><i>0.297</i></b>	<i>Salix phylicifolia</i> ; <i>Salix glauca</i> ; <i>Salix hastata</i> ; <i>Salix myrsinites</i>
Non- <i>Salix</i> , Low Shrubs Summergreen (NSLSS)	2.512	<b><i>2.492</i></b>	1.208	<b><i>1.199</i></b>	<i>Vaccinium uliginosum</i> ; <i>Betula nana</i>
Evergreen Prostrate Dwarf Shrubs (EPDS)	1.411	<b><i>1.400</i></b>	1.312	<b><i>1.301</i></b>	<i>Vaccinium oxycoccus</i> ; <i>Cassiope tetragona</i> ; <i>Dryas octopetala</i> ; <i>Saxifraga oppositifolia</i> ; <i>Andromeda polifolia</i>
Summergreen Prostrate Dwarf Shrubs (SPDS)	14.117	<b><i>14.003</i></b>	0.428	<b><i>0.425</i></b>	<i>Salix arctica</i> , <i>Arctostaphylos alpinus</i> , <i>Salix reticulata</i>
Graminoid Tundra (GRT)	9.898	<b><i>9.818</i></b>	0.000	<b><i>0.000</i></b>	<i>Calamagrostis lapponica</i> , <i>Carex parallela</i> , <i>Carex rupestris</i> , <i>Carex vaginata</i> , <i>Eriophorum vaginatum</i> , <i>Festuca ovina</i> , <i>Poa alpigena</i>
Cushion forbs, Lichens and Moss tundra (CLM)	1.198	<b><i>1.188</i></b>	0.030	<b><i>0.029</i></b>	<i>Astragalus alpinus</i> , <i>Astragalus frigidus</i> , <i>Bartsia alpina</i> , <i>Cerastium alpinum</i> , <i>Charmorchis alpina</i> , <i>Gymnadenia conopsea</i> , <i>Leucorchis albida</i> , <i>Pedicularis lapponica</i> , <i>Pinguicula vulgaris</i> , <i>Bistorta vivipara</i> , <i>Rubus chamaemorus</i> , <i>Saussurea alpina</i> , <i>Silene acaulis</i> , <i>Tofieldia pusilla</i> , <i>Hylocomium splendens</i> , <i>Tomentypnum nitens</i> , <i>Pleurozium schreberi</i> , <i>Sphagnum warnstorffii</i> , <i>Peltigera aptosa</i> , <i>Cetraria nivalis</i> , <i>Cladonia spp.</i>

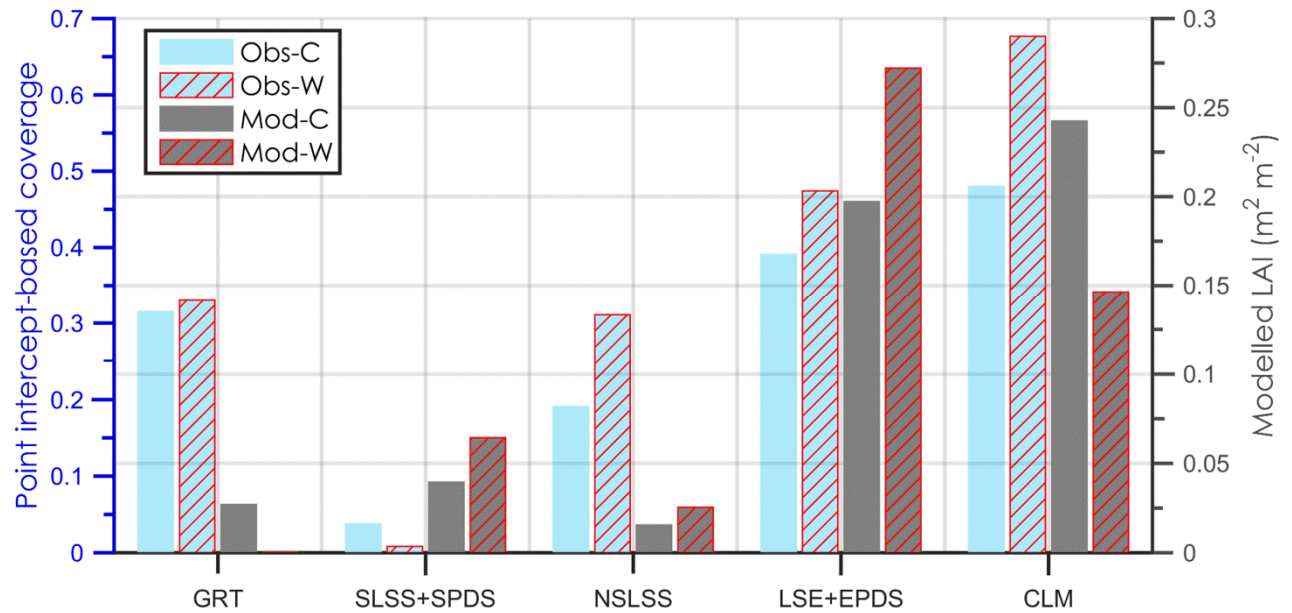




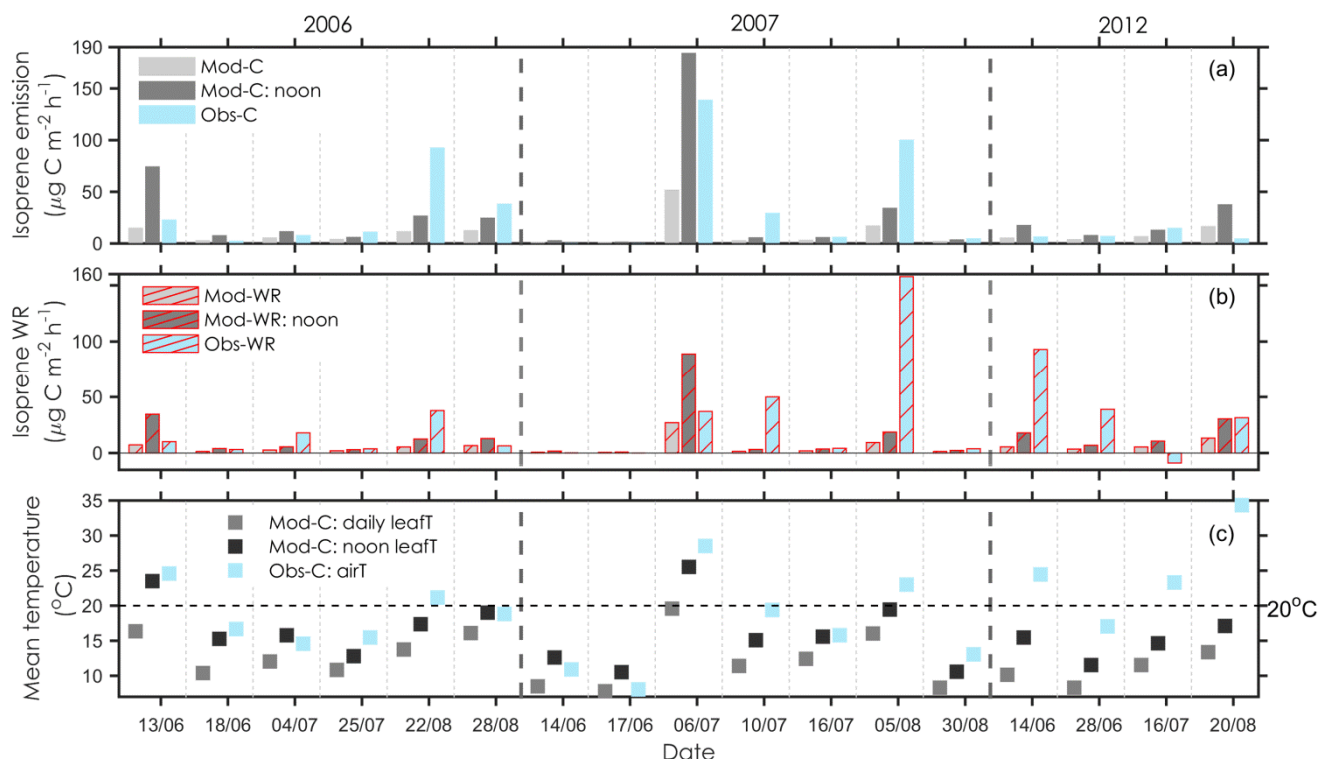
**Figure 1** The observed isoprene emission rates in relation to the chamber air temperature in July over three field seasons (2006, 2007, 2012) in the Abisko tundra heath.

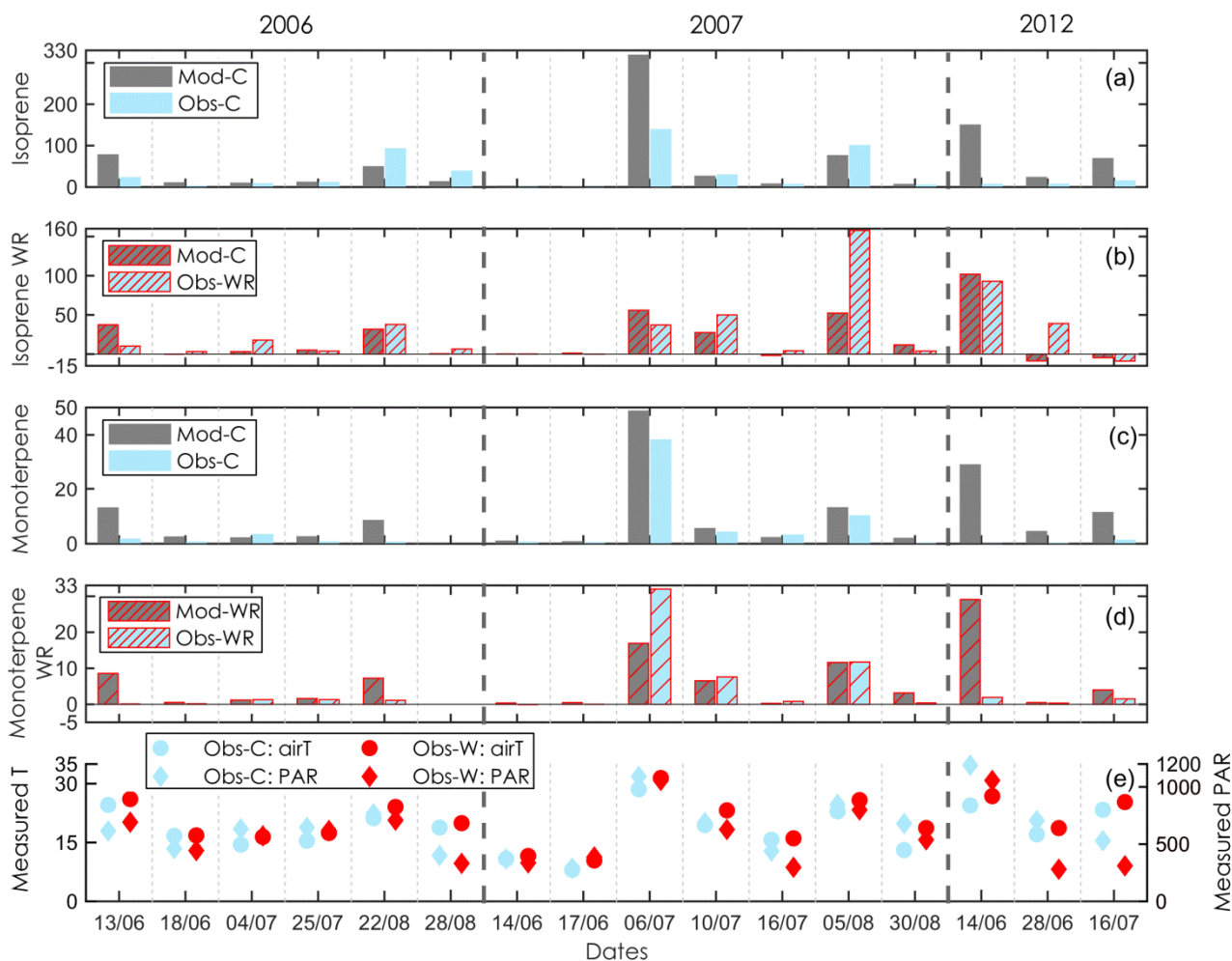


**Figure 2** Modelled (grey) and observed (blue) gross primary production (GPP, (a)), ecosystem respiration (ER, (b)), and net ecosystem production (NEP, (c)) for the growing season of 2010 and 2012 in the control plots at the Abisko tundra heath. Error bars indicate the standard deviation for the six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

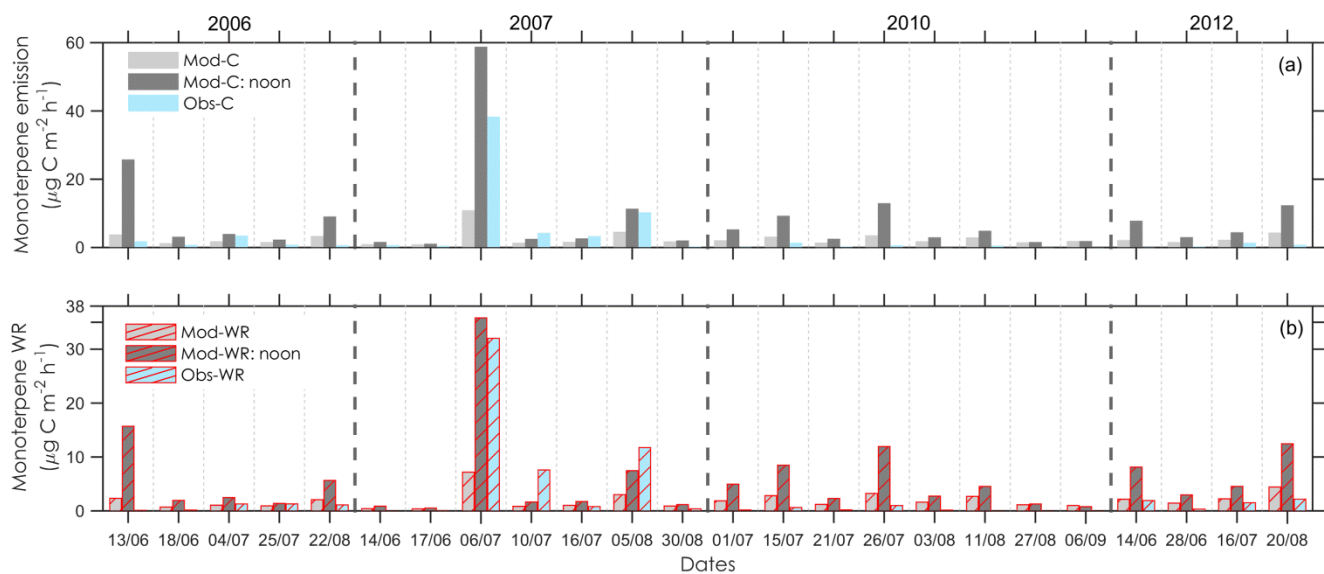


**Figure 3** Point-intercept based vegetation coverage and modelled leaf area index (LAI,  $\text{m}^2 \text{m}^{-2}$ ) averaged for the growing season 2010 and 2012 for the control (C) and warming (W) treatments in the Abisko tundra heath. Different y axes are used for the observed (Obs) and the modelled (Mod) coverage to allow comparison of warming effects. 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.

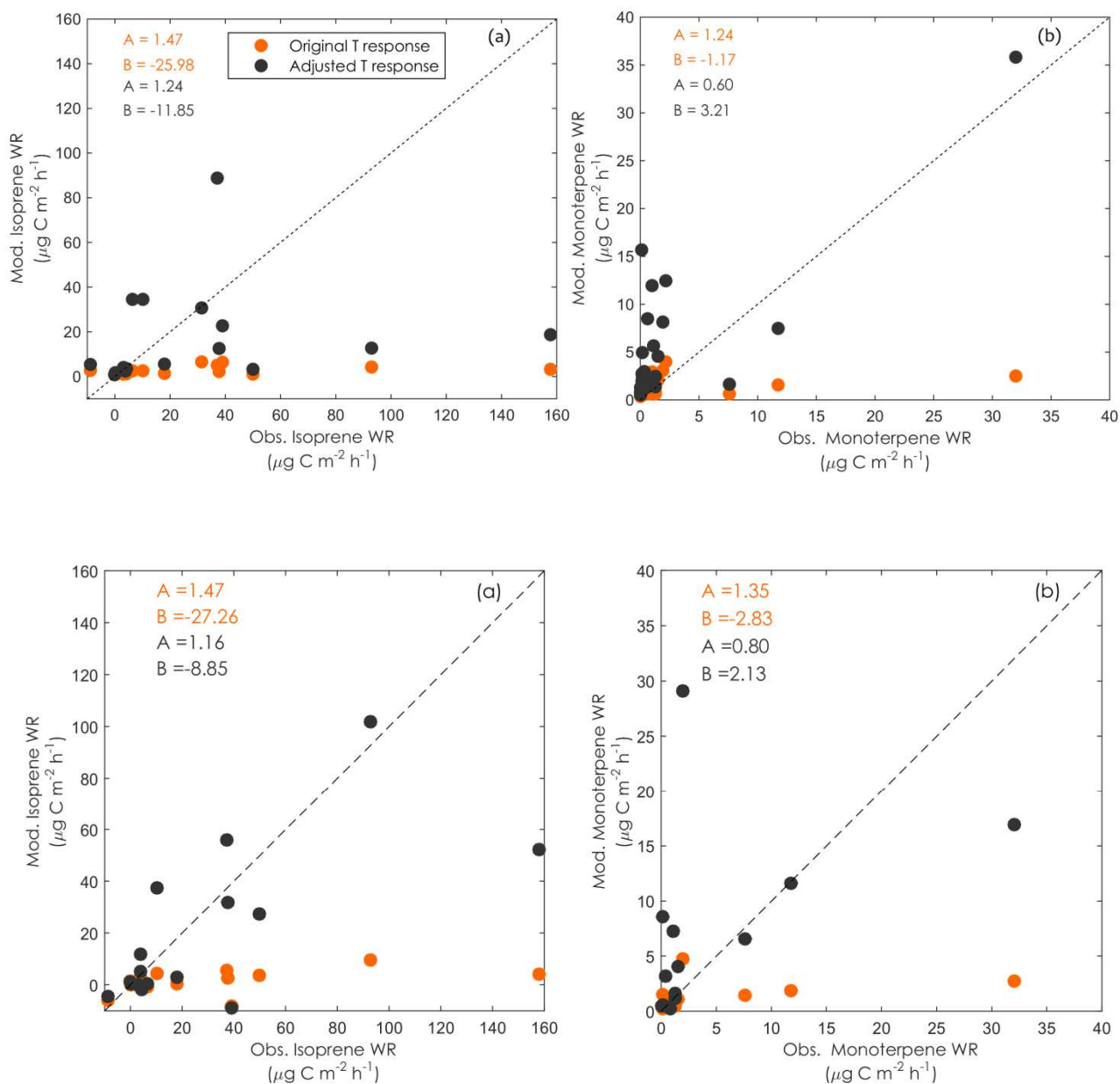




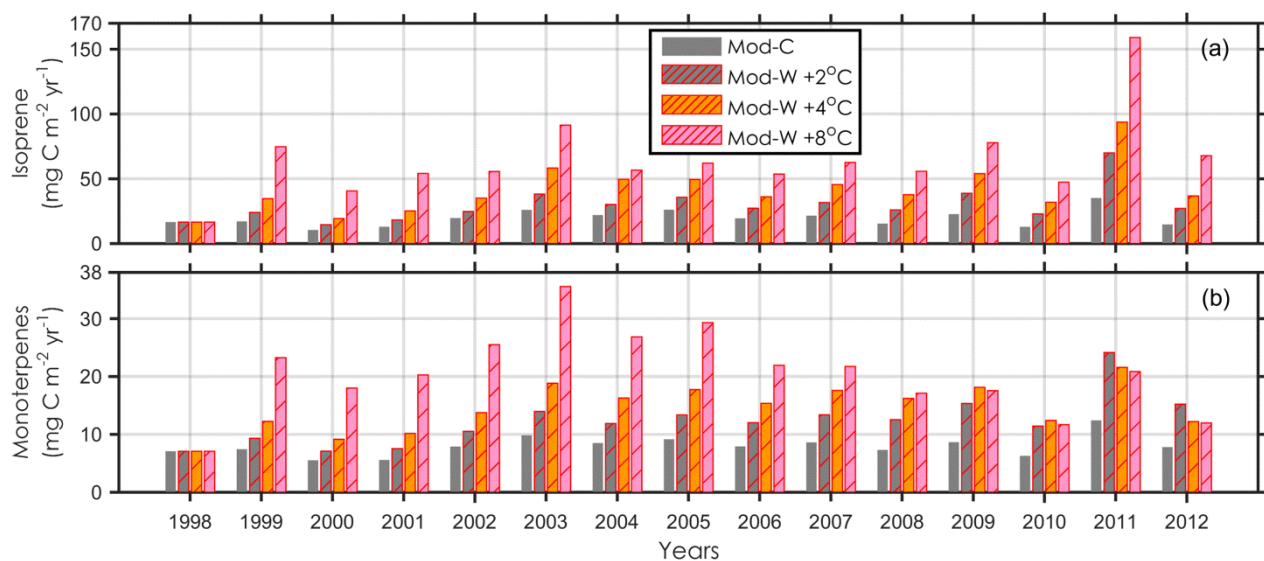
**Figure 4** Comparison of the modelled ~~daytime average and noon~~ (a) isoprene and (c) monoterpene emission rates with the observations in the control (C) plots ~~(a and c)~~ and evaluation of modelled warming responses (WR) with the observed WR (b and d) at the Abisko tundra heath. The ~~modelled daytime average (daily leafT) and noon leaf temperatures (noon leafT) in the C plots~~ were compared with the observed chamber enclosure air temperature (airT) and PAR outside the enclosure are displayed in (e). Mod: Modelled; Obs: Observed.



**Figure 5 Comparison of the modelled daytime 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 tundra heath. Mod: Modelled; Obs: Observed.**



**Figure 6-5** 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 67** 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.



## Supplementary material

### S1 PFTs simulated for the study area

**Table S1 Detailed description of the PFT parameters for the study area (Abisko, tundra heath). LSE: low shrubs evergreen; SLSS: *Salix*, low shrubs summergreen; NSLSS: non-*Salix*, low shrubs summergreen; EPDS: evergreen prostrate dwarf shrubs; SPDS: summergreen prostrate dwarf shrubs; GRT: graminoid tundra; CLM: cushion forbs, lichens and mosses tundra; S: shrub; G: grass; NL: needleleaf; BL: broadleaf; Max.: maximum; Min.: minimum; EG: evergreen; SG: summergreen; GDD5: growing degree days above 5 °C; GDD0: growing degree days above 0 °C; ~~Min.: Minimum; Max.: Maximum.~~**

Parameters	LSE	SLSS	NSLSS	EPDS	SPDS	GRT	CLM
Growth form	S	S	S	S	S	G	G
Leaf physiognomy	NL	BL	BL	NL	BL	BL	BL
Fraction of roots in the upper (0.5 m)/lower (1 m) soil layer	0.8/0.2	0.8/0.2	0.8/0.2	0.8/0.2	0.8/0.2	0.9/0.1	0.9/0.1
Max. leaf:root carbon mass ratio	1	1	1	0.5	0.5	0.2	0.2
Min. canopy conductance (mm/s)	0.3	0.5	0.5	0.5	0.5	0.5	0.5
Phenology types	EG	SG	SG	EG	SG	any	any
Longevity of leaves (years)	3	0.5	0.5	3	0.5	1	1
Leaf turnover rate (year <sup>-1</sup> )	0.33	1	1	0.5	1	1	0.6
Root turnover rate (year <sup>-1</sup> )	0.7	0.7	0.7	0.7	0.7	0.5	
Sapwood turnover rate (year <sup>-1</sup> )	0.01	0.01	0.01	0.01	0.01	-	-
Fire resistance (0-1)	0.12	0.12	0.12	0.12	0.12	0.5	0.5
Min. forest floor PAR establishment (KJ <del>m<sup>-2</sup></del> /day <sup>-1</sup> )	1000	1000	1000	1250	1250	1250	1250
Interception coefficient <sup>ε</sup>	0.06	0.02	0.02	0.04	0.02	0.01	0.01
Parameter for relationship between crown area and stem diameter	10	10	10	10	10	-	-
Allometry parameter (k_allom2) related vegetation height and stem diameter	4	4	4	1	1	-	-
Allometry parameter (k_allom3) related vegetation height and stem diameter	0.67	0.67	0.67	0.67	0.67	-	-
Constant in crown area and stem diameter relationship	1.6	1.6	1.6	1.6	1.6	-	-
Max. tree crown area (m <sup>2</sup> )	1	1	1	1	1	-	-
Tree leaf to sapwood area ratio	125	125	125	100	100	-	-
Sapwood and heartwood density (kg C m <sup>-3</sup> )	200	200	200	200	200	-	-

Growth efficiency threshold ( $\text{kg C m}^{-2}\text{-leaf yr}^{-1}$ )	0.012	0.012	0.012	0.01	0.01	-	-
Max. establishment rate ( $\text{samplings m}^{-2}\text{ yr}^{-1}$ ) <sup>*</sup>	0.6	0.8	1	0.8	0.8	-	-
Recruitment shape parameter <sup>†</sup>	10	10	7	10	10	-	-
Mean non-stress longevity (yr)	25	25	25	30	30	-	-
GDD5 required to obtain full leave cover	0	50	50	0	50	50	1
Photosynthesis min. temperature ( $^{\circ}\text{C}$ )	-4	-4	-4	-4	-4	-4	-4
Approximate lower range of temperature optimum for photosynthesis	10	10	10	10	10	10	10
Approximate upper range of temperature optimum for photosynthesis	30	30	30	25	25	25	25
Photosynthesis max temperature ( $^{\circ}\text{C}$ )	38	38	38	38	38	38	38
Min. temperature of coldest month for survival	-32.5	-40	-40	-1000	-1000	-1000	-1000
Min. temperature of coldest month for establishment	-32.5	-32.5	-32.5	-1000	-1000	-1000	-1000
Max. temperature of coldest month for establishment	1000	1000	1000	1000	1000	1000	1000
Min. temperature of warmest month for establishment	-1000	-1000	-1000	-1000	-1000	-1000	-1000
Min. GDD5 for establishment	100	100	100	0	0	0	0
Min. GDD0 for reproduction	300	300	300	150	150	150	50
Max. GDD0 for reproduction <sup>‡</sup>	-	-	-	1500	-	1400	-
Min. snow cover (mm)	-	-	-	20	20	-	50
Maintenance respiration coefficient	1	1	1	1	1	1	1
Min. fraction of available soil water in upper soil layer during growing season	0.1	0.1	0.1	0.01	0.01	0.01	0.01
Max. evapotranspiration rate	5	5	5	5	5	5	5
Litter moisture flammability threshold (fraction of available water holding capacity)	0.3	0.3	0.3	0.3	0.3	0.2	
Sapwood C:N mass ratio	330	330	330	330	330	-	-
Fine root C:N mass ratio	29	29	29	29	29	29	29
Maximum nitrogen uptake per fine root ( $\text{kg N kg C}^{-1}\text{ day}^{-1}$ )	0.0028	0.0028	0.0028	0.0028	0.0028	0.00551	0.00551
Half-saturation concentration for N uptake ( $\text{kg N l}^{-1}$ )	1.477E-06	1.477E-06	1.477E-06	1.477E-06	1.477E-06	1.886E-06	1.886E-06
Fraction of sapwood or root for N long-term storage	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Specific leaf area ( $\text{m}^2\text{ kg C}^{-1}$ )	12.56	24.25	24.25	12.56	24.25	19.75	25.8
Isoprene emission capacity ( $\mu\text{g C g}^{-1}\text{ h}^{-1}$ ) $E_{IS20}$	1.751/1.737	11.305/11.213	2.512/2.492	1.411/1.400	14.117/14.003	9.898/9.818	1.198/1.188
Isoprene emissions show a seasonality (1) or not (0)	0	1	1	0	1	1	0

Monoterpene emission capacity ( $\mu\text{g C g}^{-1} \text{h}^{-1}$ ) $E_{MS20}$	0.089/0.088	0.300/0.297	1.208/1.199	1.312/1.301	0.428/0.425	0.000/0.000	0.030/0.029
Fraction of monoterpene production that go into storage pool	0.5	0.5	0.5	0.5	0.5	0.5	0
Aerodynamic conductance ( $\text{m s}^{-1}$ )	0.04	0.04	0.04	0.03	0.03	0.03	0.03

\*,  $\xi$ : the values were adjusted based on the point intercepted-based observations to increase/decrease relative abundance;

€: a dimensionless biome-dependent proxy for rainfall region (Gerten et al., 2004).

Γ: relates to life history class of plant functional types. High values of this parameter represent a steeper decline in establishment rate as shading reduces potential seedling growth.

**Table S2 Detailed description of literature values used for parameterizing PFT emission capacities, isoprene ( $I_S$ ,  $\mu\text{g C gdw}^{-1} \text{h}^{-1}$ ) and monoterpene ( $M_S$ ,  $\mu\text{g C gdw}^{-1} \text{h}^{-1}$ ) emissions at 20 °C and 30 °C. For some PFTs, the multiple data values from the same study are from different sampling dates in the original publications.**

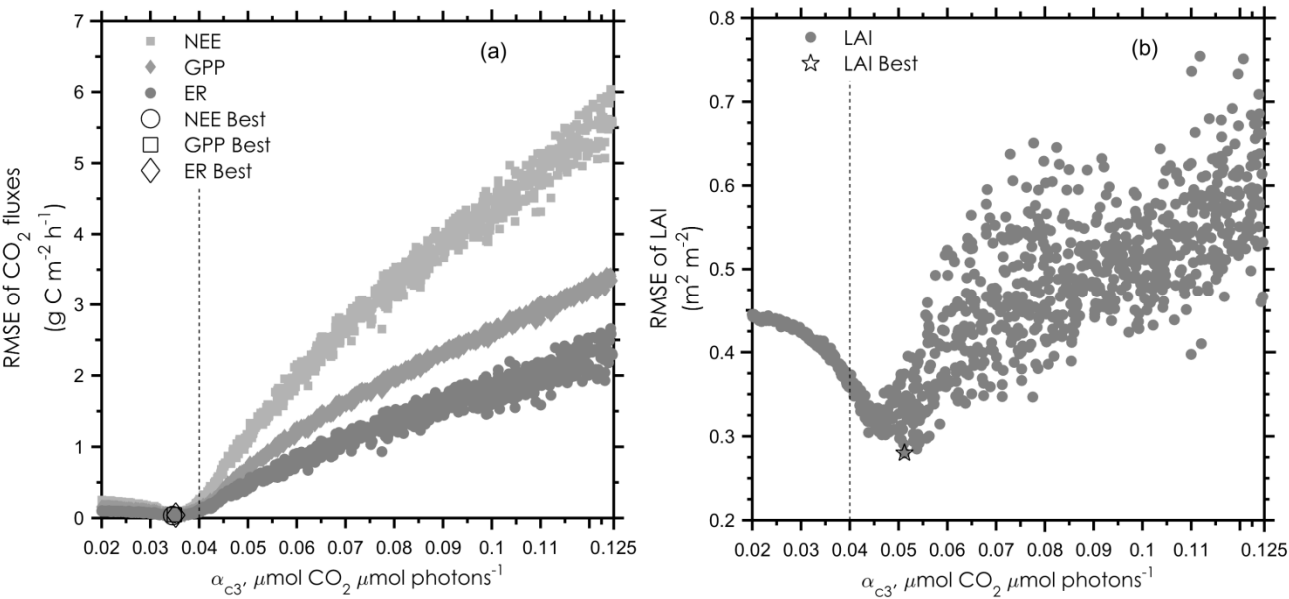
Plant functional types (PFTs)	Species name	Emission potentials ( $\mu\text{g C gdw}^{-1} \text{h}^{-1}$ )		Reference	Emission potentials ( $\mu\text{g C gdw}^{-1} \text{h}^{-1}$ )		Reference
		$E_{IS30}$	$E_{IS20}$		$E_{MS30}$	$E_{MS20}$	
Low Shrub evergreen (LSE)	<i>Empetrum hermaphroditum</i>	8.050	7.985	(Schollert et al., 2015)	0.029	0.029	(Schollert et al., 2015)
		0.700	0.694		0.066	0.065	
		0.000	0.000	(Vedel-Petersen et al., 2015)	0.020	0.020	(Vedel-Petersen et al., 2015)
		0.004	0.004		0.110	0.109	
		0.003	0.003		0.198	0.218	
	<b>Average</b>	<b>1.751</b>	<b>1.737</b>		<b>0.089</b>	<b>0.088</b>	
Salix, Low Shrubs Summergreen (SLSS)	<i>Salix phylicifolia</i>	14.160	14.045	(Rinnan et al., 2011)	0.910	0.903	(Rinnan et al., 2014)
		2.050	2.033	(Vedel-Petersen et al., 2015)	0.048	0.048	(Vedel-Petersen et al., 2015)
	<i>Salix glauca</i>	12.670	12.567		0.130	0.129	
		16.340	16.207		0.110	0.109	
	<b>Average</b>	<b>11.305</b>	<b>11.213</b>		<b>0.300</b>	<b>0.297</b>	
Non-Salix group of summergreen shrubs (NSLSS)	<i>Vaccinium uliginosum</i>	0.000	0.000	Schollert unpublished data			
		0.000	0.000	(Rinnan et al., 2011)	1.070	1.061	(Rinnan et al., 2011)
		19.480	19.322	(Schollert et al., 2015)	1.730	1.716	(Schollert et al., 2015)
	<i>Betula nana</i>	1.870	1.855		0.680	0.674	
		0.000	0.000	Schollert unpublished data			
		0.000	0.000				
		0.000	0.000	(Vedel-Petersen et al., 2015)	2.400	2.381	(Vedel-Petersen et al., 2015)
		0.990	0.982		0.840	0.833	
		0.267	0.265		0.530	0.526	
	<b>Average</b>	<b>2.512</b>	<b>2.492</b>		<b>1.208</b>	<b>1.199</b>	
Evergreen Prostrate Dwarf Shrub (EPDS)	<i>Cassiope tetragona</i>	0.132	0.131	(Schollert et al., 2015)	1.800	1.785	(Schollert et al., 2015)
		7.315	7.255		0.110	0.109	
		0.000	0.000		0.033	0.033	
		2.430	2.410		0.190	0.188	
		0.000	0.000		0.029	0.029	
		0.000	0.000	(Rinnan et al., 2011)	3.160	3.134	(Rinnan et al., 2011)
	<b>Average</b>	<b>1.411</b>	<b>1.400</b>		<b>1.312</b>	<b>1.301</b>	
Summergreen Prostrate Dwarf Shrub (SPDS)	<i>Salix arctica</i>	27.350	27.128	(Rinnan et al., 2014)			
		2.240	2.222	(Schollert et al., 2015)	0.330	0.327	(Schollert et al., 2015)
		21.960	21.782		0.930	0.922	
		6.030	5.981		0.025	0.025	

Graminoid (GRT)	<i>Salix arctophila</i>	4.640	4.602	(Vedel-Petersen et al., 2015)	0.430	0.427	(Vedel-Petersen et al., 2015)
		13.260	13.152		0.720	0.714	
		23.340	23.151		1.700	1.686	
	<b>Average</b>	<b>14.117</b>	<b>14.003</b>		<b>0.428</b>	<b>0.425</b>	
		20.240	20.076	(Ekberg et al., 2009)	0.000	0.000	(Ekberg et al., 2009)
	<i>Eriophorum angustifolium</i>	10.001	9.920				
		0.735	0.729				
		3.463	3.435				
		27.359	27.137				
		26.432	26.217				
Cushion forbs, lichens, and moss tundra (CLM)	<i>Carex rostrata</i>	14.832	14.712				
		0.080	0.080				
		0.266	0.264				
		1.704	1.690				
		6.150	6.100				
		7.521	7.460				
	<b>Average</b>	<b>9.898</b>	<b>9.818</b>		<b>0.000</b>	<b>0.000</b>	
	<i>Sphagnum cuspidatum</i>	1.160	1.151	Tiiva unpublished data			
	<i>Sphagnum fuscum</i>	0.864	0.857	(Hanson et al., 1999)			
	<i>Sphagnum balticum</i>	2.034	2.017				
		2.108	2.091	(Ekberg et al., 2011)			
		3.216	3.190				
		1.703	1.689				
	<i>Warnstorfia exannulata</i>	0.132	0.131	(Tiiva et al., 2007)	0.010	0.009	(Faubert et al., 2010)
	<i>Aulacomnium palustre</i>	2.860	2.837	Tiiva unpublished data			
	<i>Dicranum polysetum</i>	0.043	0.043	(Hanson et al., 1999)			
	<i>Hylocomium splendens</i>	0.011	0.011				
	<i>Ptilidium ciliare</i>	0.024	0.024				
	<i>Sphagnum*</i>	0.220	0.218	(Janson and De Serves, 1998)	0.050	0.050	(Janson et al., 1999)
	<b>Average</b>	<b>1.198</b>	<b>1.188</b>		<b>0.030</b>	<b>0.029</b>	

\*There is no species name in the original publication.

**S2 Sensitivity testing of  $\alpha_{c3}$**

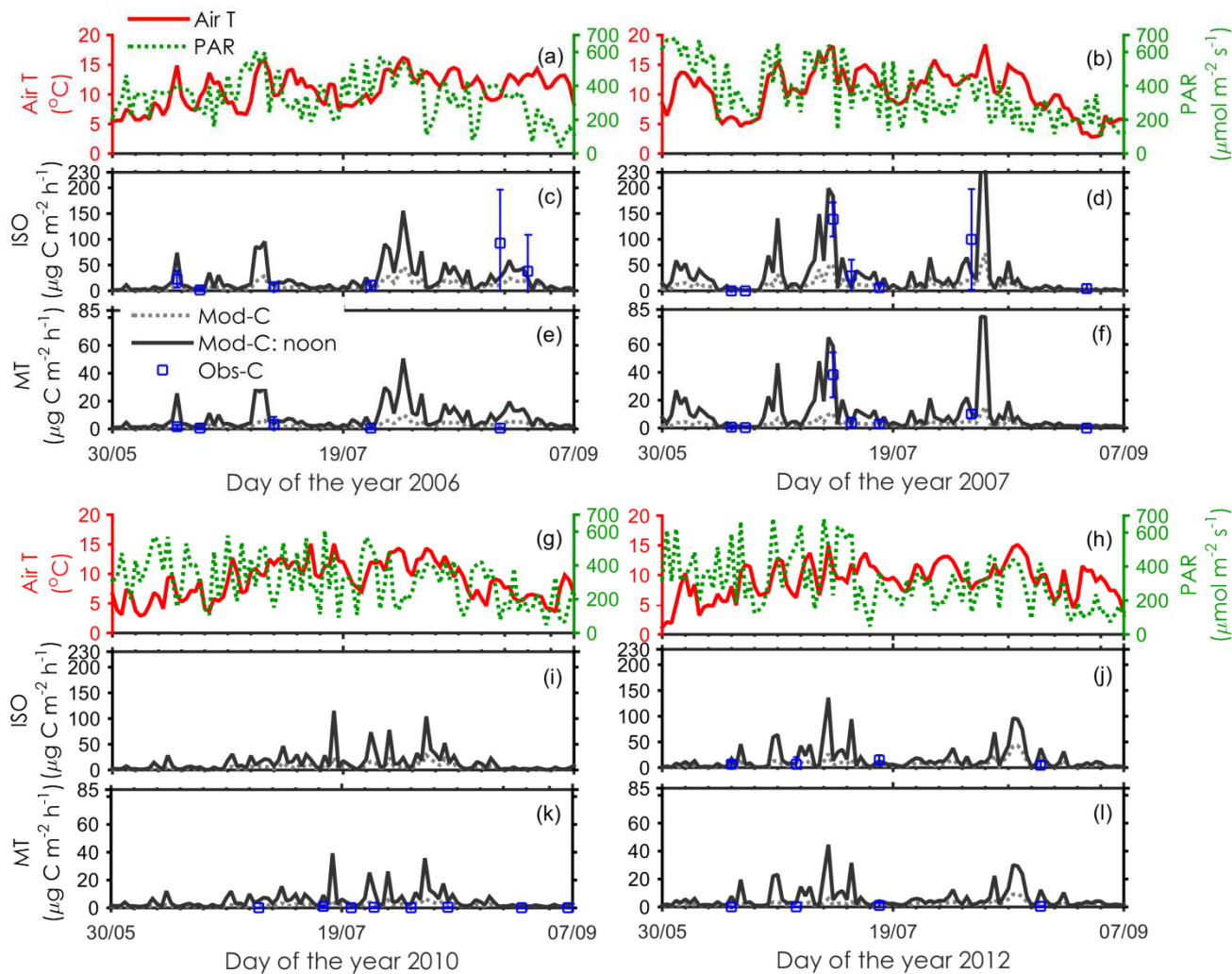
A uniform sampling of the parameter  $\alpha_{c3}$  (1000 times, under the range of 0.02 to 0.125  $\mu\text{mol CO}_2 \mu\text{mol photons}^{-1}$ ) was implemented and the model simulations with different  $\alpha_{c3}$  values were conducted to investigate how the modelled GPP, ER and LAI are influenced by the parameter  $\alpha_{c3}$ . Closed-chamber based  $\text{CO}_2$  fluxes ~~as well as~~ point intercepted-based plant coverage in the control plots were compared with the simulated outputs. Modelled GPP, ER, NEP and LAI were largely influenced by the parameter value of  $\alpha_{c3}$  (Fig. S1). The root mean square error (RMSE) values of the four investigated variables showed a slight decrease, followed by a sharp increase with increasing  $\alpha_{c3}$ . For the RMSE of GPP, ER and NEP, the first quantile occurs at the lowest value range of  $\alpha_{c3}$ , with the RMSE of LAI spreading between 0.03 and 0.07. The parameter values with the lowest RMSE (Best) for GPP, ER, LAI are 0.034, 0.037 and 0.051  $\mu\text{mol CO}_2 \mu\text{mol photons}^{-1}$ , respectively.



**Figure S1** The root mean square error (RMSE) of the modelled net ecosystem production (NEP), gross primary production (GPP), ecosystem respiration (ER), (a) and leaf area index (LAI), (b) related to the observations for the years 2006 and 2007. The parameter values with the lowest RMSE (Best, in the legend) are marked. The dashed lines point out the  $\alpha_{c3}$  selected for this study.

### S3 Seasonal variation of BVOC emissions

The span of the BVOC measurements covered the main growing seasons over ~~three~~<sup>four</sup> years. The modelled daily average emission rates in the C plots showed pronounced day-to-day ~~as well as~~<sup>and</sup> seasonal variations (Fig. S2). The modelled emissions of isoprene and monoterpenes were low in ~~the spring~~<sup>Spring</sup> and ~~autumn~~<sup>Autumn</sup>, and peaked on warm days during the ~~summer~~<sup>Summer</sup>. 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 variation between the blocks for the days with the observed high average emission rates (blue error bars in Fig. S24) ~~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 observations, especially for the years with generally with low emissions, (2006, 2010 and 2012). The observed high monoterpene emissions for a few days were better captured by the modelled noon emission.~~ The emission of monoterpenes remained more constant than that of isoprene towards the end of the growing season (not fully presented here).





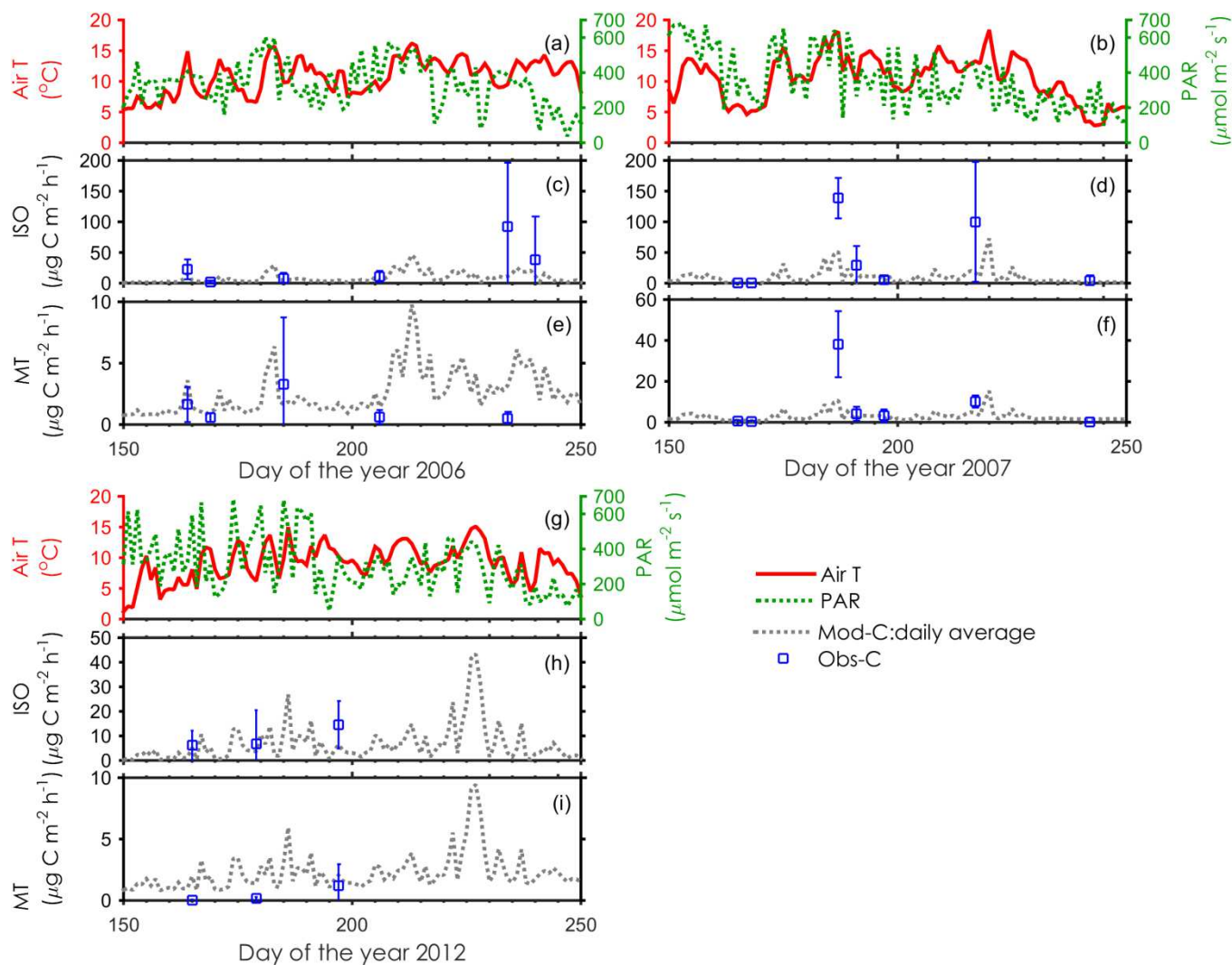


Figure S2 Time-series of the air temperature (Air T) at 2 m height, photosynthetically active radiation (PAR), the modelled isoprene (ISO) and monoterpene emissions (MT) for the days 150-250 in 2006, 2007, ~~2010~~ and 2012 in the Abisko tundra heath. Both modelled and observed fluxes are from the control (C) conditions ~~and the modelled daily average (Mod-C: daily average) 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.~~

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