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Editor's Comment

9 Dear Palmira Messina et al.,

10 I find the manuscript much improved. I only have one question:

11 Page 19, lines 12-14: “a) First, we select only papers that provide EFs in the proper units (μgC
12 $\text{g}^{-1} \text{h}^{-1}$) and for standard conditions such as defined in ORCHIDEE (PAR = $1000 \mu\text{mol m}^{-2}$
13 s^{-1} , temperature = $30 \text{ }^\circ\text{C}$), unless the information needed to convert the EFs accordingly was
14 given.” In here I guess you mean have only considered EFs given per leaf biomass, not per
15 leaf area? The EFs given in molar units or in grams of compound, or per hour, are trivial to
16 convert to desired unit. This could be written more explicitly.

17 Sincerely,

18 Janne Rinne

19

20

Response to Editor's Comment

22 Firstly, we thank the Editor for his time and consideration.

23

24 We agree with the Editor that we should be more specific. Actually, the crucial point is to
25 have EFs per leaf biomass and at the standard conditions of PAR = $1000 \mu\text{mol m}^{-2} \text{ s}^{-1}$ and
26 temperature = $30 \text{ }^\circ\text{C}$ or, at least, to have all the information to convert the given EFs in the
27 proper measurement unit. We changed the text accordingly and quote it below.

28

1 At page 14, lines 1-4: *“First, we select only papers that provide EFs per leaf biomass and for*
2 *standard conditions such as defined in ORCHIDEE (PAR = 1000 $\mu\text{mol m}^{-2} \text{s}^{-1}$ and*
3 *temperature = 30 °C). We do not consider papers where EFs were given per leaf area, per*
4 *area, or in different standard conditions, unless the information needed to convert the EFs*
5 *accordingly was available.”*

6

Global Biogenic Volatile Organic Compound emissions in the ORCHIDEE and MEGAN models and sensitivity to key parameters

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Abstract

A new version of the Biogenic Volatile Organic Compounds (BVOC) emission scheme has been developed in the global vegetation model ORCHIDEE (Organizing Carbon and Hydrology in Dynamic Ecosystems), that includes an extended list of biogenic emitted

1 compounds, updated emission factors (EFs), a dependency on light for almost all compounds
2 and a multi-layer radiation scheme. Over the 2000–2009 period, using this model, we estimate
3 mean global emissions of 465 Tg C yr⁻¹ for isoprene, 107.5 Tg C yr⁻¹ for monoterpenes, 38
4 Tg C yr⁻¹ for methanol, 25 Tg C yr⁻¹ for acetone and 24 Tg C yr⁻¹ for sesquiterpenes. The
5 model results are compared to state-of-the-art emission budgets, showing that the
6 ORCHIDEE emissions are within the range of published estimates. ORCHIDEE BVOC
7 emissions are compared to the estimates of the Model of Emissions of Gases and Aerosols
8 from Nature (MEGAN), which is largely used throughout the biogenic emissions and
9 atmospheric chemistry community. Our results show that global emission budgets of the two
10 models are, in general, in good agreement. ORCHIDEE emissions are 8% higher for isoprene,
11 8% lower for methanol, 17 % higher for acetone, 18% higher for monoterpenes and 39%
12 higher for sesquiterpenes, compared to the MEGAN estimates. At the regional scale, the
13 largest differences between ORCHIDEE and MEGAN are highlighted for isoprene in
14 northern temperate regions, where ORCHIDEE emissions are higher by 21 Tg C yr⁻¹, and for
15 monoterpenes, where they are higher by 4.4 and 10.2 Tg C yr⁻¹ in northern and southern
16 tropical regions compared to MEGAN. The geographical differences between the two models
17 are mainly associated with different EF and Plant Functional Type (PFT) distributions, while
18 differences in the seasonal cycle are mostly driven by differences in the Leaf Area Index
19 (LAI). Sensitivity tests are carried out for both models to explore the response to key
20 variables or parameters such as LAI and Light Dependent Fraction (LDF). The ORCHIDEE
21 and MEGAN emissions are differently affected by LAI changes, with a response highly
22 depending on the compound considered. Scaling the LAI by a factor of 0.5 and 1.5 changes
23 the isoprene global emission by -21% and +8% for ORCHIDEE and -15% and +7% for
24 MEGAN, and affects the global emissions of monoterpenes by -43% and +40% for
25 ORCHIDEE and -11% and +3% for MEGAN. Performing a further sensitivity test, forcing
26 ORCHIDEE with the MODIS LAI, confirms the high sensitivity of the ORCHIDEE emission
27 module to LAI variation. We find that MEGAN is more sensitive to variation in the LDF
28 parameter than ORCHIDEE. Our results highlight the importance and the need to further
29 explore the BVOC emission estimate variability and the potential for using models to
30 investigate the estimated uncertainties.

31

1 **1 Introduction**

2 The terrestrial biosphere emits large amounts of Volatile Organic Compounds (VOCs) in
3 particular terpenoids, such as isoprene, monoterpenes, and sesquiterpenes, and oxygenated
4 hydrocarbons such as methanol, acetone, formaldehyde, acetaldehyde, acetic acid, or formic
5 acid (Laothawornkitkul et al, 2009; Guenther et al., 2012; Penñelas and Staudt 2014). On the
6 global scale, the ecosystem contribution to VOC emissions is significantly higher than the
7 anthropogenic one, and accounts for 75–90% of the total emission (Guenther et al. 1995;
8 Lamarque et al., 2010). BVOCs play a central role in atmospheric chemistry, influencing the
9 oxidative capacity of the atmosphere (Arneth et al., 2011; Taraborrelli et al., 2012), leading to
10 the production of tropospheric ozone in the presence of nitrogen oxides (Von Kuhlmann et al.,
11 2003; Mao et al., 2013), and influencing the tropospheric carbon monoxide budget (Pfister et
12 al., 2008). Additionally, BVOCs and their oxidation products lead to the formation and
13 growth of more than 50% of the Secondary Organic Aerosols (SOA) (Kanakidou et al., 2005;
14 Goldstein and Galbally, 2007; Van Donkelaar et al., 2007; Engelhart et al., 2008; Hallquist et
15 al., 2009; Acosta Navarro, et al., 2014; Tsigaridis et al., 2014). Under appropriate atmospheric
16 conditions, BVOCs can contribute to a significant fraction of particles that evolve into cloud
17 condensation nuclei (CCN) (Riipinen et al., 2012), even enhancing the droplet number
18 concentration in clouds (Topping et al., 2013).

19 Despite numerous measurements and the progressive understanding of the processes
20 underlying their production, BVOC emission estimates are still highly uncertain and vary
21 significantly (Steiner and Goldstein, 2007; Arneth et al., 2008; Simpson, et al., 2012;
22 Sindelarova et al., 2014).

23 Over the last 20–25 years, two main methods have been developed to derive BVOC
24 inventories: a top-down approach based on the inversion of satellite measurements, which
25 allows BVOC emissions to be indirectly derived (Palmer et al., 2006; Barkley et al., 2013),
26 and a bottom-up approach. The latter approach is the most widely used method for local-,
27 regional- or global-scale studies and can be divided into two main categories: (i) an empirical
28 method, based essentially on Guenther et al. (1995), where the response of leaf emissions to
29 environmental changes is modelled using algorithms combined in a multiplicative way
30 (Guenther et al., 2006, 2012; Lathière et al., 2006, 2010; Steinbrecher et al., 2009; Oderbolz
31 et al., 2013). Hereafter we refer to it simply as the *Guenther formulation*); (ii) a processed-

1 based approach, where emissions are linked to the photosynthetic electron transport rate in
2 chloroplasts (Niinemets et al., 2003a, b; Sitch et al., 2003; Keenan et al., 2009; Schurgers et
3 al., 2009; Pacifico et al. 2011; Unger et al. 2013). The models discussed in this study belong
4 to the first category of bottom-up models.

5 BVOC emission modelling at the global scale is a complex issue, especially because of the
6 number of variables and processes influencing the emission of these compounds, generally
7 characterized by strong temporal and geographical variations. A critical point is the lack of
8 information available at the global scale related to the various biomes, making difficult an
9 accurate representation of the geographical distribution and of the seasonal variation of
10 BVOC emissions (Peñuelas and Staudt, 2010). The basal EF for instance, defined as the
11 emission at the leaf level under standardized environmental conditions of temperature and
12 solar radiation (Guenther et al., 1995; Steinbrecher et al., 2009), shows large variability from
13 one plant species to another. Nowadays, there is an increasing number of field campaigns that
14 investigate, in addition to isoprene and bulk monoterpenes, many other important compounds
15 for atmospheric chemistry, especially regarding the SOA formation, such as speciated
16 monoterpenes and sesquiterpenes. More data and information are therefore available,
17 allowing EF estimates for a wider range of BVOCs, despite the limitations for modelling
18 purposes which will be discussed in Sect. 2.2.1. To calculate BVOC emissions, a single EF is
19 usually assigned for each PFT, where one PFT represents a group of plants having the same
20 phylogenetic, phenological and physical characteristics (Prentice et al., 1992). The choice of
21 one single value for each PFT is especially difficult, as each PFT actually corresponds to
22 several plant species, and EFs show, in general, a wide range of values among different plants
23 (Kesselmeier and Staudt, 1999; Niinemets et al., 2011). Moreover, several measurements
24 show that the emission factors are significantly sensitive to many processes and parameters
25 that are difficult to isolate and linked to plant stress, such as drought periods, ozone exposure,
26 insects, herbivores and pathogen attacks, (for a review see Laothawornkitkul, et al., 2009 and
27 Niinemets et al., 2010), making it delicate to set EFs even for a single plant. In addition, the
28 link between EF variation and plant phenology is in general not taken into account, or is
29 roughly described, especially in models that adopt the empirical approach.

30 In the early works focusing on BVOCs, isoprene was the only compound considered to be
31 both light and temperature dependent, while the other compounds were considered to be only

1 temperature dependent. More recent papers show a growing evidence of the dependency of
2 monoterpenes (Dindorf et al., 2006; Holzke et al., 2006; Šimpraga et al., 2013),
3 sesquiterpenes (Hansen and Seufert., 2003) and oxygenated BVOCs (Jacob et al. 2002, 2005;
4 Harley et al., 2007; Millet et al., 2008, 2010; Hu, et al., 2011; Wells et al., 2014) on radiation.
5 As proposed in Guenther et al. (2012), a general approach is now to consider, for each emitted
6 compound, an emission fraction which depends on both temperature and solar light, as done
7 for isoprene, with the remaining fraction dependent only on temperature. The Guenther et al.
8 (2012) approach considers only one value per emitted compound, whilst it has been shown
9 the LDF also depends on the plant species. For example, measurements of the diurnal cycle
10 for monoterpenes above Amazonian rainforest (Rinne et al. 2002; Kuhn et al., 2002) suggest
11 that emissions are dependent on both light and temperature, whilst the role of light in
12 influencing monoterpene emissions from boreal Scot pine forest is less clear (Taipale et al.,
13 2011). Moreover, Staudt and Seufert (1995) and Loreto et al. (1996) show that monoterpene
14 emissions from coniferous trees are principally influenced by the temperature, while those
15 from Holm oak are predominantly controlled by a light-dependent mechanism. Owen et al.
16 (2002) find that, in the Mediterranean region, emissions of all compounds from *Quercus* sp.
17 are light dependent, the ocimene emitted by *Pinus pinea* is strongly correlated to light and an
18 apparent weak light dependency is exhibited by monoterpene emissions from *Cistus incanus*.
19 Ghirardo et al. (2010) provide the fraction of light-dependent monoterpene emission, being
20 58% for Scots pine, 33.5% for Norway spruce, 9.8% for European larch, and 98–100% for
21 both Silver birch and Holm oak. Shao et al. (2001) and Steinbrecher et al. (1999) attribute for
22 Scots pine a value of 20–30% and 25–37%, respectively. Nevertheless, there is no general
23 agreement on the exact value of the temperature- and light-dependent fraction to assign for
24 individual compound and PFT, as it appears also from the works mentioned right above.

25 Another crucial component in the estimation of BVOC emissions is the LAI, which can be
26 either simulated using a vegetation model, or prescribed using values retrieved from satellite
27 data or field measurements. Significant differences in terms of temporal and spatial
28 distribution are found between the LAI estimated by measurements and the LAI calculated by
29 models, with discrepancies of up to 100% at the global scale and more than 150% for specific
30 biomes types (Garrigues et al., 2008; Pinty et al., 2011; Fang et al., 2012a, b). Consequently,

1 the high uncertainty related to LAI affects the predicted regional and seasonal distribution of
2 BVOC emissions.

3 According to our knowledge, most papers investigating BVOC emission sensitivity focus on
4 the response of emissions to different experimental set-ups, changing, for instance, climate
5 forcing and land use. For example, Oderbolz et al. (2013) pointed out the importance of the
6 differences between the land-cover inventories, and of the uncertainties in the classification of
7 land cover. Arneth et al. (2011) compared three vegetation models, changing the experimental
8 set-up, such as the vegetation distribution and the climate forcings. Depending on the
9 experiment considered, the total annual isoprene emissions were found to increase or decrease
10 by more than 30%. Ashworth et al. (2010) investigated the impact of varying the climate
11 forcing temporal resolution of isoprene emission in the MEGAN model, finding a variation of
12 isoprene emissions of up to 7% at the global scale and up to 55% in some locations. Keenan
13 et al. (2011) investigate the effect of canopy structure using different canopy models and they
14 conclude that larger differences in the final emissions can be attributed to the use of different
15 canopy models, rather than different emission model approaches. Nevertheless, very few
16 studies have investigated the impact of the uncertainty of key parameters/variables, such as
17 LAI, on emissions. One example is the work by Sindelarova et al. (2014) in which several
18 simulations were performed with the MEGAN model to assess the sensitivity of isoprene
19 emissions to many parameters and processes such as LAI, emission factors (EFs), CO₂
20 concentration, soil moisture, and the radiation scheme. The sensitivity simulations performed
21 showed a variation in isoprene emissions of up to 50% at the global scale.

22 In the present work, our objectives are to (i) present the updated version of the emission
23 module embedded in the dynamic global vegetation model ORCHIDEE, (ii) provide present-
24 day estimates of global BVOC emissions for several relevant compounds (isoprene,
25 monoterpenes, sesquiterpenes, methanol, acetone, formaldehyde, acetaldehyde, acetic acid,
26 formic acid and the main speciated monoterpenes) using the new emission scheme, (iii)
27 compare the ORCHIDEE results to the widely used emission model MEGAN, putting the two
28 models under the same forcing conditions, but retaining their particular characteristics (see
29 Sect. 2.5), in particular the emission scheme, classes and distribution of PFTs and LAI
30 processing, and (iv) explore, at the global and regional scales, the BVOC emission sensitivity
31 to EFs, LAI and LDF in ORCHIDEE and MEGAN, and to understand the reasons behind

1 these discrepancies. ORCHIDEE is designed to provide past, present and future scenarios of
2 emissions from vegetation, studying the links between climate, the plant phenology and
3 emissions. It is therefore essential that the internal variability, weaknesses and inaccuracies of
4 the emission module are extensively investigated. The proper way to assess the correctness of
5 a model would be to evaluate it against observations, as it is done, for example, for organic
6 aerosols by Mann et al. (2014) and Tsigaridis et al. (2014) and for tropical mountain forest
7 carbon store by Spracklen and Righelato (2014). The evaluation of BVOC emission models
8 against observations has already been carried out at local and regional scales (i.e. Karl et al.,
9 2007; Kunl et al., 2007; Lathière et al., 2009; Smolander et al., 2014), demonstrating a good
10 performance of the Guenther formulation. Nevertheless, given the ecosystem biodiversity, the
11 huge variability of the parameters involved and the poor spatial and temporal coverage of
12 BVOC emission observations, it is extremely difficult to infer a robust evaluation at global
13 scale. In such a context we can rely on model inter-comparison and sensitivity tests in order to
14 assess the limitations and uncertainties of BVOC emission estimates, to relate them to
15 particular key parameters/variables and to investigate their origin. In Sect. 2, the ORCHIDEE
16 model and the updates from the previous version (Lathière et al, 2006), the MEGAN model
17 and the technical details of the simulations are described. The comparison with other
18 published estimates, the inter-comparison between the two models and the sensitivity tests
19 carried out are extensively described in Sect. 3. The conclusions and future directions are
20 provided in Sect. 4 and 5.

21

22 **2 Model developments and set-up**

23 **2.1 ORCHIDEE model: general description**

24 ORCHIDEE (Organizing Carbon and Hydrology in Dynamic EcosystEm) is a dynamic global
25 vegetation model (Krinner et al., 2005; Magnan et al., 2011) that consists of two main parts:
26 the carbon module STOMATE (Saclay-Toulouse-Orsay Model for the Analysis of Terrestrial
27 Ecosystems) and the surface vegetation atmosphere transfer scheme SECHIBA
28 (Schématisation des échanges hydriques à l'interface biosphere-atmosphère, in English:
29 mapping of hydrological exchange at the biosphere/atmosphere interface).

1 STOMATE describes processes such as photosynthesis, carbon allocation, litter
2 decomposition, soil carbon dynamics, maintenance and growth respiration. A completely
3 prognostic plant phenology including leaf critical age, maximum LAI (leaf area index),
4 senescence, plant tissue allocation, and leaf photosynthetic efficiency that varies depending
5 on the leaf age, is also taken into account. The soil water budget and the exchanges of energy
6 and water between the atmosphere and the biosphere are calculated in SECHIBA (Krinner et
7 al., 2005). The Choissnel hydrological scheme is used with a two-meter soil column
8 represented by two moisture layers: a superficial layer and a deep layer (Ducoudré et al.
9 1993). The biogenic emission scheme, of which we present a new version, is embedded in this
10 module (Lathière et al., 2006).

11 In ORCHIDEE, ecosystems are represented by 13 Plant Functional Types (PFTs, listed in
12 Table 1). Each PFT is representative of a specific set of plant species that are grouped
13 according to plant physiognomy (tree or grass), leaf shape (needleleaf or broadleaf),
14 phenology (evergreen, summergreen or raingreen) and photosynthesis type for crops and
15 grasses (C3 or C4). The main biophysical and biogeochemical processes for each PFT are
16 described in Krinner et al. (2005) and in Maignan et al. (2011). For our study, the global
17 vegetation distribution is prescribed for all runs using appropriate forcings, as described in
18 paragraph 2.4.

19 **2.2 BVOCs in ORCHIDEE: module improvements**

20 The BVOC module is extensively updated, considering recent findings regarding emission
21 schemes and field measurements. The new BVOC emission scheme is a development of the
22 module implemented in ORCHIDEE by Lathière et al. (2006) and based on the model
23 presented by Guenther et al. (2012). It now provides a multi-layer canopy model, where
24 radiation is calculated following the scheme proposed by Spitter et al. (1986a, b) and the one
25 already used in ORCHIDEE for the calculation of photosynthesis. The canopy is considered
26 split *vertically* into several LAI layers, the number of which (up to 17) depends on the LAI
27 value. Emissions are calculated for each layer through consideration of the sunlit and shaded
28 leaf fractions and the light extinction and light diffusion through canopy. In a second step they
29 are vertically summed, providing a single value for each PFT and grid point.

1 The emission flux F of a specific biogenic compound c , for a given PFT i at a LAI layer l is
2 calculated following the Eq. (1):

$$3 \quad F_{c,i}(l) = LAI_i(l) \cdot SLW_i \cdot EF_{c,i} \cdot CTL_c(l) \cdot L_c \quad (1)$$

4 where $LAI_i(l)$ is the leaf area index expressed in $m^2 m^{-2}$ at a particular LAI layer and PFT,
5 SLW_i is the specific PFT leaf weight in $g m^{-2}$, $EF_{c,i}$ is the basal emissions at the leaf level for
6 an individual compound and PFT at standard conditions of temperature ($T = 303.15 K$) and
7 photosynthetically active radiation ($PAR = 1000 \mu mol m^{-2} s^{-1}$), expressed in $\mu gC g^{-1} h^{-1}$.
8 CTL_c is the emission activity factor, depending on the emitted compounds, that takes into
9 account the deviation from the standard conditions related to temperature and PAR, and it is
10 extensively described in the second part of the present paragraph. L_c is the activity factor
11 simulating the impact of leaf age on emissions and is considered for isoprene and methanol.
12 The total emission per grid cell is obtained by summing $F_{c,i}(l)$ over the layer l and averaging
13 the emission contribution of each individual PFT, weighted by PFT fractional land coverage.
14 Further details on the original version of the emission module are given in Lathièrè et al.
15 (2006).

16 Table 2 summarises the principal modifications compared to the previous module version. In
17 particular, we (i) added new emitted compounds, (ii) estimated the emissions using a multi-
18 layer radiation scheme that calculates diffuse and direct components of light at different LAI
19 levels, (iii) inserted a dependence on light for almost all compounds, and (iv) updated the EFs.

20 Eight speciated monoterpenes (α -pinene, β -pinene, limonene, myrcene, sabinene, camphene
21 3-carene, t- β -ocimene) and bulk sesquiterpenes are now included in the updated ORCHIDEE
22 emission module. We chose these compounds because measurements have shown that they
23 are emitted from vegetation in the greatest abundance and because of their importance in
24 atmospheric chemistry, in particular regarding secondary organic aerosol formation.

25 We mentioned that, the emission module has also been modified to include a light
26 dependency for almost all compounds emitted. In the previous module version, indeed,
27 isoprene was the only compound dependent on both light and temperature, while the others
28 were only dependent on temperature. As detailed in Sect. 1, most recent field campaigns
29 highlight, for a large number of plants, the dependency of monoterpenes, sesquiterpenes and
30 oxygenated BVOC emissions on radiation as well. To adopt a detailed parameterisation is not

1 yet possible, cause to data lacking at global scale. Therefore, in the new emission module we
2 consider the approach described in Guenther et al. (2012), even if it is rather oversimplified.
3 BVOCs are now modelled to consider both light-dependent and light-independent emission
4 processes, and the response to temperature and light (CTL) is calculated for individual
5 compounds at each LAI layer (l):

$$6 \quad CTL_c(l) = (1 - LDF_c) \cdot CTLI_c + LDF_c \cdot CTLD \cdot CL(l) \quad (2)$$

7 LDF_c is the light-dependent fraction of the emission, specified for each compound emitted
8 (Table 2). To chose the LDF value for monoterpenes, we rely on Dindorf et al. (2006), Holzke
9 et al. (2006), Guenther et al. (2012) and Šimpraga et al. (2013). Other LDF values were based
10 on Guenther et al. (2012). $CTLI_c$ is the temperature-dependent emission response that is not
11 light dependent and depends on individual compounds. $CTLD$ and CL are the temperature
12 and light responses for the light-dependent fraction, respectively, and are the same functions
13 as in the previous version of the emissions module. For all details we refer to Guenther et al.
14 (1995).and Lathièrè et al. (2006). $CTLI$ is equal to:

$$15 \quad CTLI = \exp(\beta(T - T_0)) \quad (3)$$

16 Where β is the empirical coefficient of the exponential temperature response and it is now
17 defined as in Guenther et. al (2012) (Table 2).

18 **2.2.1 Emission Factor update**

19 EF determination represents one of the greatest sources of uncertainty in the quantification of
20 BVOC emissions (Niinemets et al., 2011). Several measurement campaigns were carried out
21 over the last decade, giving important new insights and information for re-examining the
22 emission factors used in the emission module and correcting them accordingly. Nevertheless,
23 the methodology to assess EFs is still under debate within the scientific community.
24 Assigning EFs, especially on the global scale, is very tricky. In the ideal case, for each
25 compound emitted, we should consider the EFs of all plants belonging to one particular PFT
26 and the land cover of each plant. We could then, for each PFT and compound, make averages
27 weighted by plant land cover, thus obtaining an average EF for each PFT and emitted
28 compound. Unfortunately, there are not yet enough observations available to use such a
29 methodology.

1 Several aspects make it difficult to find a good strategy to assign EFs. First, sources of
2 information regarding EFs are very heterogeneous such as bibliographical reviews, article
3 presenting punctual or fairly widespread measurement campaigns, and modelling
4 experiments, making the selection of papers to use especially tricky. When a large range of
5 EF values is documented for one particular plant species, it is not obvious whether this range
6 is actually representative of a natural (geographical or species-to-species) variability, and can
7 therefore be considered as valid, or originates from technical difficulties or improvements
8 (and, in this case, if preference should be given to more recently published papers). A further
9 difficulty is linked to the high number of plant species that can be combined together into one
10 PFT, in comparison to the relatively small proportion of plant species and/or measurement
11 sites worldwide that could be investigated, despite numerous and crucial field studies.
12 Moreover, our EF review shows that EFs are highly variable from one plant to another, even
13 if the plants belong to the same PFT. In this context, it is difficult to assign a single EF per
14 each PFT which integrates this variability adequately. Lastly, the procedure itself used to
15 determine EFs from field measurements adds another source of uncertainty. Indeed, EFs are
16 derived by adjusting the measured flux at leaf level in standard conditions of
17 photosynthetically active radiation (PAR) and temperature, using algorithms such as Guenther
18 et al. (1995). However, there is no universal agreement on the parameterization of these
19 algorithms (Tarvainen et al., 2005; Duhl et al., 2008; Kim et al., 2010; Bracho-Nunex et al.,
20 2011; Fares et al., 2011).

21 All these aspects underline the challenge and uncertainty of assigning one fixed EF value for
22 each PFT in global models (Kesselmeier and Staudt, 1999, Niinemets et al., 2010 and Arneth
23 et al. 2011), also considering that the emission estimates are very sensitive to changes in EF.

24 In this particular context, we try to establish a sufficiently consistent methodology and we
25 follow the guidelines below to update the EFs in the ORCHIDEE emission scheme. All the
26 values and related references used to define the new EFs are provided in Tables S1-S10 (one
27 table for each compound) of the Supplementary Material.

28 ~~a) First, we select only papers that provide EFs in the proper units ($\mu\text{gC g}^{-1}\text{h}^{-1}$) and for~~
29 ~~standard conditions such as defined in ORCHIDEE (PAR = $1000\ \mu\text{mol m}^{-2}\text{s}^{-1}$, temperature =~~
30 ~~30 °C), unless the information needed to convert the EFs accordingly was given.~~

1 a) First, we select only papers that provide EFs per leaf biomass and for standard conditions
2 such as defined in ORCHIDEE (PAR = 1000 $\mu\text{mol m}^{-2} \text{s}^{-1}$ and temperature = 30 °C). We do
3 not consider papers where EFs were given per leaf area, per area, or in different standard
4 conditions, unless the information needed to convert the EFs accordingly was available.

5 b) When the most recent papers agree on a specific EF range, we discard the old references if
6 the EF value is significantly different. In other cases all the works collected are taken into
7 account.

8 c) First for each paper we gather all the values available per ORCHIDEE PFT and per emitted
9 compound. In there are more values per paper, we calculate the average in order to have one
10 EF per compound, PFT and paper.

11 d) Then, for each compound and each PFT, we choose an EF that is in the range of the
12 collected values and is the closest to the average and median calculated. When one EF value
13 cannot be clearly assigned, we take a value between the average or the median and the
14 previous ORCHIDEE EF values (Lathière et al., 2006). Considering the high sensitivity of the
15 emission module to EF variation, in order to avoid unreliable estimate, in the case of
16 ambiguity, for the highly emitted compounds, in particular for isoprene, a more conservative
17 approach is adopted and the EF values of the previous version are kept.

18 e) In choosing the new EFs, in the case of very few or inconclusive information, EF
19 variability between the different PFTs of the old version of ORCHIDEE (Lathière et al.,
20 2006) and/or MEGAN (Guenther et al., 2012) is taking into account.

21 f) For each compound we check *a posteriori* that the new set of EFs provides a regional
22 distribution which is consistent with the orders of magnitude expected and given in the
23 literature. Only for monoterpenes for tropical PFTs we replace the first value selected (2.5
24 $\mu\text{gC g}^{-1} \text{h}^{-1}$) with the current value (2.0 $\mu\text{gC g}^{-1} \text{h}^{-1}$).

25 Table 3 shows the new and old EFs used in the emission module, and Table 4 shows the EF
26 values for each speciated monoterpene as a percentage of the bulk monoterpene EF value. As
27 shown in Table 3, the revision leads to the modification of almost all EFs. In some cases, the
28 EF differences in comparison with the previous version are very significant. Regarding
29 isoprene, boreal needleleaf deciduous PFT is now recognized as a less important emitter (EF
30 = 8 $\mu\text{gC g}^{-1} \text{h}^{-1}$ in the old version and EF = 0.5 $\mu\text{gC g}^{-1} \text{h}^{-1}$ in the new one). The new EF is

1 decided considering the EF proposed by Guenther et al. (2006) ($0.003 \mu\text{gC g}^{-1} \text{h}^{-1}$), Guenther
2 et al. (2012) ($0.002 \mu\text{gC g}^{-1} \text{h}^{-1}$), Steinbrecher et al. (2009) and Karl et al., 2009 ($0.44 \mu\text{gC g}^{-1}$
3 h^{-1}), Smiatek and Steinbrecher (2006) ($0.10 \mu\text{gC g}^{-1} \text{h}^{-1}$) and Klinger et al. (2002) ($2.23 \mu\text{gC}$
4 $\text{g}^{-1} \text{h}^{-1}$) (more details in the Supplement). Our choice is confirmed by Ruuskanen et al.
5 (2007), who assign a contribution of less than 3% of the VOC emission to isoprene, 2-methyl-
6 3-buten-2-ol (hereafter referred to it simply as MBO) and 1,8-cineole, for larch, which is the
7 major component of boreal needleleaf deciduous PFT.

8 Furthermore, we consider boreal broadleaved deciduous trees to be a higher emitter of
9 isoprene than in the previous model version (now $\text{EF} = 18 \mu\text{gC g}^{-1} \text{h}^{-1}$, while before $\text{EF} = 8$
10 $\mu\text{gC g}^{-1} \text{h}^{-1}$), since the papers collected propose particularly high values, such as Guenther et
11 al. (2012) ($22.7 \mu\text{gC g}^{-1} \text{h}^{-1}$), Guenther et al. (2006) ($30.8 \mu\text{gC g}^{-1} \text{h}^{-1}$), Stewart et al. (2003)
12 ($33.9 \mu\text{gC g}^{-1} \text{h}^{-1}$) and Smiatek and Steinbrecher (2006) ($18.8 \mu\text{gC g}^{-1} \text{h}^{-1}$). For
13 monoterpenes, we assign a significantly higher EF (from $0.8 \mu\text{gC g}^{-1} \text{h}^{-1}$ to $2.0 \mu\text{gC g}^{-1} \text{h}^{-1}$)
14 to tropical broadleaf evergreen and deciduous PFTs. For MBO, we reduce the EF for the
15 temperate needleleaf evergreen PFT from $20 \mu\text{gC g}^{-1} \text{h}^{-1}$ to $1.4 \mu\text{gC g}^{-1} \text{h}^{-1}$ (Tarvainen et al.,
16 2005; Hakola et al., 2006; Chang et al., 2009; Kim et al., 2010).

17

18 **2.3 MEGAN description**

19 The Model of Emissions of Gases and Aerosols from Nature (MEGAN) is a modelling system
20 for the estimation of emission fluxes of biogenic organic compounds from terrestrial
21 vegetation. The basis of the model is a simple mechanistic approach established by Guenther
22 et al. (1991, 1993, 1995), which links emissions with the main environmental driving factors
23 such as solar radiation and leaf temperature. Further development of the algorithm led to the
24 inclusion of leaf ageing, soil moisture impact on the emissions, and effects of the loss and
25 production of compounds within a forest canopy (Guenther et al., 2006). The current version
26 of the model, MEGANv2.1, also includes a full canopy module. The model calculates light
27 and temperature conditions inside a canopy by evaluating the energy balance on five canopy
28 levels. Additionally, emissions of each compound are considered to have light-dependent and
29 light-independent components defined by the light dependent fraction (LDF). For a detailed

1 description of emission equations and parameterization we refer to Sect. 2 in Sindelarova et al
2 (2014) and Guenther et al. (2012).

3 MEGANv2.1 is available either as a stand-alone version or embedded in the Community
4 Land Model version 4 (CLM4) (Lawrence et al., 2011) of the Community Earth System
5 Model (CESM) (Gent et al., 2011). When operating in the stand-alone version, the driving
6 variables, such as meteorological input data, vegetation description and leaf area index, need
7 to be provided by the user. When running MEGAN inside CLM4, the input data can be
8 provided by the CESM atmospheric and land surface models on-line at each time-step. In this
9 work, we use the stand-alone model version of MEGANv2.1, hereafter simply referred to as
10 MEGAN.

11 MEGAN estimates emissions of 19 chemical compound classes, which are then redistributed
12 into 147 final output model species, such as isoprene, monoterpene and sesquiterpene species,
13 methanol, carbon monoxide, alkanes, alkenes, aldehydes, ketones, acids and other oxygenated
14 VOCs. Although the input parameters, such as vegetation description and emission potentials,
15 can be defined by the user, MEGAN comes with a default definition of PFTs and the emission
16 factors assigned to them. The vegetation distribution is described with fractional coverage of
17 16 PFT classes consistent with those of the CLM4 model (Lawrence and Chase, 2007). The
18 emission potential of each modelled species is calculated based on the PFT coverage and
19 emission factor of each PFT category. For several VOC compounds, emission potentials can
20 be defined in the form of input maps. Emission potential maps with global coverage and high
21 spatial resolution for isoprene, main monoterpene species and MBO are provided together
22 with the MEGAN code.

23 MEGAN is widely applied for the estimation of biogenic VOC emissions at both regional and
24 global scales (e.g., Guenther et al., 2006, 2012; Müller et al., 2008; Millet et al., 2010;
25 Sindelarova et al., 2014; Situ et al., 2014; Stavrakou et al., 2014) and serves for the evaluation
26 of the impact of BVOCs on atmospheric chemistry by coupling the model with chemistry
27 transport models (e.g. Heald et al., 2008; Pfister et al., 2008; Emmons et al., 2010; Fu and
28 Liao, 2012; Tilmes et al., 2015).

1 **2.4 Model set-up and sensitivity tests**

2 The objectives of the group of simulations are: (i) to provide global estimates of BVOC
3 emissions for a large variety of compounds over the 2000–2009 period, (ii) to investigate the
4 differences and similarities between the ORCHIDEE and MEGAN results regarding the
5 spatial, inter-annual and inter-seasonal variability of emissions, (iii) to analyze the response of
6 BVOC emissions to the variation of some key variables and parameters such as the LAI and
7 LDF. Table 5 summarizes the simulations performed in this study and their principal
8 characteristics.

9 We carried out a total of 5 sets of runs:

- 10 1. Two simulations for the 2000–2009 period performed by both models using each
11 model's standard configuration, but with the same climatology (ORC_CRU and MEG_CRU).
- 12 2. One simulation for the 2000–2009 period with MEGAN using the LAI estimated by
13 ORCHIDEE (MEG_CRULAI).
- 14 3. Four simulations for the year 2006 by both models, using the ORCHIDEE LAI scaled
15 by a factor 0.5 and 1.5, respectively (ORC_LAI05, ORC_LAI15, MEG_LAI05 and
16 MEG_LAI15).
- 17 4. One simulation for the year 2006 forcing ORCHIDEE with the MODIS LAI used in
18 MEGAN standard configuration.
- 19 5. Two simulations for the year 2006 performed by both models, where we output two
20 test species, the first one totally dependent on light (LDF=1) and the second one totally
21 independent on light (LDF=0) (ORC_LDF and MEG_LDF). The output time frequency is one
22 hour for this run.

23 All simulations are performed at the global scale with a spatial resolution of $0.5^\circ \times 0.5^\circ$. We
24 use the CRU-NCEP v5.2 meteorological forcing database
25 (<http://dods.extra.cea.fr/data/p529viov/cruncep>) providing temperature, pressure, humidity,
26 wind speed and shortwave solar radiation. This forcing is based on the 6-hourly 2.5°
27 NCEP/NCAR re-analysis (Kalnay et al., 1996) combined with the CRU TS 2.1 monthly
28 anomalies (Mitchell and Jones, 2005). The run sets 3 and 4 are carried out for the year 2006,
29 which is estimated as an averaged year regarding the BVOC emissions calculated by

1 MEGAN and ORCHIDEE in the 10 years of simulation. For the ORCHIDEE model a spin-up
2 of 20 years is first performed to balance the leaf stock. The spin-up is based on a 10-year loop
3 using meteorological forcing for the year 1989, followed by a 10-year simulation from 1990
4 to 1999. In ORCHIDEE, the global vegetation distribution for the 13 PFTs is prescribed using
5 the Land-Use History (LUHa.rc2) related to the year 2000 (Hurtt et al. 2006). The database
6 can be found in
7 http://dods.extra.cea.fr/work/p86ips1/IGCM/BC/SFR/OL2/PFTmap_1850to2005_AR5_LUHa
8 .rc2. In MEGAN the distribution for the 16 PFTs is consistent with the Community Land
9 Model v4 (Lawrence and Chase, 2007) and related to the year 2000. Table 1 gives the global
10 surfaces covered by the different PFTs in ORCHIDEE and MEGAN. For the present work,
11 MEGAN in the standard configuration is forced by the LAI retrieved by MODIS (Yuan et al.,
12 2011; <http://globalchange.bnu.edu.cn/research/lai/>).

13 In ORCHIDEE, the activity factor (L_c) is kept as in Lathière et al. (2006), considering four
14 leaf age classes (new, young, mature and old leaves). For methanol, L_c is equal to 1 for new
15 and young leaves and equal to 0.5 for mature and old leaves, while for isoprene, L_c is equal to
16 0.5 for new and old leaves and equal to 1.5 for young and mature leaves. In MEGAN, the L_c
17 values are taken from Table 4 in Guenther et al. (2012); in particular, for isoprene, L_c is equal
18 to 0.05, 0.6, 1 and 0.9, and for methanol it is equal to 3.5, 3.0, 1.0, and 1.2 for the four leaf
19 age classes. For both models, no soil moisture activity factor is taken into account. The annual
20 CO_2 concentration varies along the simulation from a value of 368 ppm in 2000 to 385 ppm in
21 2009. In ORCHIDEE, the variation of CO_2 concentration can indirectly impact on the BVOC
22 emission as it affects leaf growth, while in MEGAN, a CO_2 inhibition factor on isoprene
23 emission based on Heald et al. (2009) is activated. As the CO_2 variation in this 10-year
24 simulation is low, the inhibition effect is considered insignificant (Sindelarova et al. 2014) in
25 this context. For ORCHIDEE, LDF and the β coefficient values are given in Table 2. For
26 MEGAN, the values of LDF and β are those presented in Table 4 in Guenther et al. (2012).

27 **2.5 Differences between ORCHIDEE and MEGAN emission algorithms**

28 While starting from a similar approach the ORCHIDEE and MEGAN emission modules
29 differ significantly in their parameterization and variable description and calculation. We list
30 below the main differences.

1 1) One of the principal differences in the two emission schemes is the approach on LAI.
2 ORCHIDEE calculates the LAI at each model time step for each PFT and grid cell, taking
3 into account a full plant phenology scheme and the environmental condition (temperature,
4 radiation, precipitations, CO₂, etc.), while MEGAN stand-alone version used in this study,
5 does not compute the LAI, rather, it has to be provided as an external forcing averaged over
6 the vegetated part of the grid cell.

7 2) In ORCHIDEE, the formulation of CTLD and CL is the same as in Guenther et al. (1995)
8 (see Eq. 9 and 10), while in MEGAN it is defined by Eq. (8), (9), and (10) in Guenther et al.
9 (2012). In particular in Guenther et al. (2012) the parameters of the CTLD formulation vary
10 according to the average solar radiation over the past 24h and 240h, and this dependence is
11 different for diffuse and direct radiation. We calculate the CTLD obtained with this
12 formulation considering different incoming solar radiations and we observe that the CTLD for
13 direct light is around twice that for diffuse light. In ORCHIDEE the CTLD parameters are
14 fixed and are the same for diffuse and direct radiation.

15 3) The radiation scheme in ORCHIDEE and MEGAN is based on the same approach (Spitter
16 et al., 1986 a,b), but the parameterization and formulation used are different. For example, the
17 number of vertical layers and their distribution over the LAI significantly differ between the
18 two models: up to 17 in ORCHIDEE and up to 5 in MEGAN. MEGAN also takes into
19 account the infrared radiation in emission calculation.

20 4) The PFT classes and their distribution are not the same in the two models (Table 1) and
21 they are not interchangeable without significantly modifying the models.

22 5) In ORCHIDEE, emissions are calculated for each PFT using the associated EF and LAI.
23 Next, they are averaged over the grid cell, considering the PFT land cover surface, as
24 described in Sect. 2.2. In MEGAN, vegetated emission potential (EP) is calculated over the
25 grid cell and multiplied by the average LAI over the vegetated part of the grid cell. In
26 MEGAN, vegetated potential emission maps are provided for isoprene, α -pinene, β -pinene, 3-
27 Carene, limonene, myrcene, t- β -ocimene and sabinene, while for the other compounds EPs
28 are calculated starting from the EFs per PFT and the PFT land cover distribution. This is a
29 significantly different approach. However, for ORCHIDEE, we find that global emissions
30 calculated using the EP and LAI per grid cell (the MEGAN approach) are only 5-12% lower

1 in comparison with the emissions calculated in the standard way. Isoprene presents the lowest
2 differences and monoterpenes the highest.

3 6) In the ORCHIDEE model, the dependence of the light independent emission on LAI is
4 linear, as shown in the Eq. (1) and (2) of the present work. Whereas in MEGAN, the
5 dependence on LAI is given by the γ_{LAI} factor that is equal to $(0.49 \cdot LAI)/(1+0.2 \cdot LAI^2)^{0.5}$
6 (Guenther et al., 2006). The implications of this difference are detailed in Sect. 3.4.2.

7 7) In MEGAN, leaf age classes are derived from consideration of the variation between the
8 LAI value of the current and preceding month, following a highly parameterised scheme. In
9 ORCHIDEE, leaf age classes are calculated on-line considering the plant leaf growth and leaf
10 turnover at each model time step (30 minutes).

11 8) In ORCHIDEE, hydrological processes are explicitly calculated, as briefly described in
12 Sect. 2.1.

13 9) In ORCHIDEE, the air temperature is used to compute emission, while in MEGAN the leaf
14 temperature is considered.

15

16 **3 Results**

17 **3.1 Global budgets**

18 As already discussed at the end of the introduction, the validation of BVOC emissions at the
19 global scale is a complex issue because of the poor data coverage in many regions and the
20 general lack of year-round measurements. Satellite observations provide very useful
21 information, especially regarding the order of magnitude and the seasonal and regional
22 variability of emissions, but the most abundant VOC species are not directly measured (such
23 as isoprene and monoterpenes). Satellite measurements are also subject to large uncertainties
24 arising from difficulties in the retrieval of the atmospheric concentration of short-lived
25 compounds from space or in separation of the different sources (for instance terrestrial
26 biogenic, anthropogenic, oceanic etc.) and the various compounds themselves. Global
27 emission estimates are generally performed using models, or from the application of inverse
28 modelling techniques that combine the measurements (from satellite, ground or aircraft
29 measurements) and models, providing emissions for compounds such as methanol (Jacob et

1 al. 2005; Millet et al., 2008; Stavrakou et al., 2009; Hu, et al., 2011; Wells et al., 2012, 2014)
2 and acetaldehyde (Jacob et al. 2002; Millet et al, 2010). Isoprene emissions have also been
3 inferred from satellite formaldehyde concentration (Shim et al., 2005; Palmer et al., 2006;
4 Stavrakou et al., 2011; Barkley et al., 2013; Bauwens et al., 2013; Stavrakou et al., 2014).

5 At the global scale, the main way to evaluate the results obtained in the present study is to
6 compare them with the most recent emission budgets derived either from other model runs or
7 from the inversion of satellite data. We have compared emissions from a large number of
8 estimates published so far, over the 1980–2010 period, with the global emission budgets
9 obtained from ORC_CRU and MEG_CRU simulations, the results of which are summarized
10 in Fig. 1. The emissions, calculated by the earlier version of the emission module (black
11 squares, Fig. 1) (Lathi re et al., 2006), are particularly high, as already pointed out by
12 Sindelarova et al. (2014). Methanol ($106.1 \text{ Tg C yr}^{-1}$) and acetaldehyde ($42.2 \text{ Tg C yr}^{-1}$)
13 emissions are twice as large, and formaldehyde emissions ($10.0 \text{ Tg C yr}^{-1}$) are up to 5 times
14 greater than the other estimates. The results of the new module version (ORC_CRU, green
15 stars) are more in the range of other published estimates. Although the MEG_CRU simulation
16 was carried out using the same MEGAN version as in Guenther et al. (2012) (blue hexagons,
17 Fig. 1), there is a noticeable difference between the two emission budgets (especially for
18 isoprene, monoterpenes and acetaldehyde), even when considering results for the same year
19 (e.g. 2000). Using reanalysis provided by Qian et al. (2006) as climate forcings for the year
20 2000, Guenther et al. (2012) report BVOC emissions of 472 Tg C yr^{-1} for isoprene, 124 Tg C
21 yr^{-1} for monoterpenes (considering the speciated monoterpenes accounted in this work) and
22 $11.5 \text{ Tg C yr}^{-1}$ for acetaldehyde. Our MEG_CRU simulation estimates for 2000 are 410 Tg C
23 yr^{-1} , 72 Tg C yr^{-1} , and 8.3 Tg C yr^{-1} for isoprene, monoterpenes and acetaldehyde,
24 respectively. As was already pointed out by Arneth et al. (2011), our results confirm that the
25 differences between existing meteorological forcings can lead to substantial differences in the
26 emission estimates (green triangles, first plot of Fig. 1).

27 Table 6 shows the annual emissions calculated by ORCHIDEE and MEGAN (ORC_CRU and
28 MEG_CRU simulations) at the global scale and for the northern (lat: 0–30N) and southern
29 (lat: 30S–0) tropics, the northern (lat: 30N–60N) and southern (lat: 30S–60S) temperate
30 latitudes, and the northern boreal (lat: 60N–90N) regions, averaged over the 2000–2009
31 period. At the global scale, the two models are in a good agreement. Isoprene is the main

1 compound emitted with a global amount of 465 Tg C yr⁻¹ for ORCHIDEE, accounting for
2 61% of total BVOC emissions (estimated to 757 Tg C yr⁻¹), and 428 Tg C yr⁻¹ for MEGAN,
3 accounting for 64% of total BVOCs (estimated at 666 Tg C yr⁻¹). The following most
4 abundant compounds are monoterpenes, accounting for 12% of the total for ORCHIDEE and
5 11% for MEGAN, and methanol, accounting for 5% of the total BVOC emissions for
6 ORCHIDEE and 6% for MEGAN. Acetone, sesquiterpenes and acetaldehyde each represent
7 1% to 4% of the total BVOCs for both models, while other compounds contribute to less than
8 0.5%.

9 Compared to ORCHIDEE, MEGAN global emission are 8% lower for isoprene, 8% higher
10 for methanol, 17% lower for acetone, 18% lower for monoterpenes, 39% lower for
11 sesquiterpenes and 25% for MBO. Regarding speciated monoterpenes, major differences arise
12 from α -pinene (around 40%) while the relative difference between ORCHIDEE and MEGAN
13 is between -8% and +16% for other compounds. The highest contribution to total emission is
14 attributed to the tropical regions ranging between 34% and 50% for the southern tropics and
15 between 31.5% and 39.5% for the northern tropics, depending on the compound (except
16 MBO). Both models calculate the contribution of northern temperate regions to the total
17 emission ranging from 6% to 24% and a contribution of less than 5% for southern temperate
18 regions and northern boreal regions. For MBO, field campaigns measured significant
19 emissions only for a few plant types such as Ponderosa and Scots pine (Kim et al., 2010;
20 Tarvainen et al. 2005; Harley et al., 1998). The EF values in the ORCHIDEE and MEGAN
21 models are consequently significant only for the PFTs representing these plants (TeNeEv and
22 BoNeEv), leading to notable emissions in the temperate North latitudes and contributing 88%
23 for ORCHIDEE and 63% for MEGAN of the global MBO emission.

24 At the regional scale, the largest differences between ORCHIDEE and MEGAN in terms of
25 absolute values appear in the northern temperate regions for isoprene, where emissions are 21
26 Tg C yr⁻¹ higher in ORCHIDEE. Indeed, the marked seasonal cycle of emissions for northern
27 temperate latitudes implies that the largest differences between ORCHIDEE and MEGAN
28 occur in summer. The differences between the two models are, in this case, directly linked to
29 discrepancies in the EFs and in the occupying surface of the PFTs at these latitudes (see Fig.
30 3, plots in the last row). In particular, in northern temperate region the highest discrepancies
31 are mainly due to the different PFT surface coverage for grass and crop and the higher EFs

1 values in ORCHIDDE in comparison to MEGAN. Actually, in ORCHIDEE C3Gr covers the
2 42% of vegetated surface with an $EF = 12 \mu\text{gC g}^{-1} \text{h}^{-1}$, C3Ag covers the 18% with an $EF = 5$
3 $\mu\text{gC g}^{-1} \text{h}^{-1}$, while in MEGAN the C3GrCool occupies the 20% with an $EF = 2 \mu\text{gC g}^{-1} \text{h}^{-1}$,
4 C3GrCold the 6% with an $EF = 4 \mu\text{gC g}^{-1} \text{h}^{-1}$, C3GrCool the 20% with an $EF = 2 \mu\text{gC g}^{-1} \text{h}^{-1}$
5 and Crop the 23.2% with an $EF = 0.12 \mu\text{gC g}^{-1} \text{h}^{-1}$. This example raises an important issue.
6 Considering the EF assigned to C3Gr, we lowered its value with respect to the previous
7 version, from 16 to $12 \mu\text{gC g}^{-1} \text{h}^{-1}$. These is a compromise value, chosen so that we do not
8 excessively bias the emissions in other areas. C3Gr is, indeed, strongly present in other
9 regions: 13% of northern tropical areas, 22% of southern tropical areas and 32% of the total
10 vegetation surface. A more detailed description of the different crop and grass (in other words
11 with a larger number of PFTs) could lead to more accurate results. The same consideration
12 could be done for almost all the other PFTs.

13 This illustrates the strong impact of different choices in EF allocation, not only regarding
14 global estimates, but also for geographical variation in emissions. For the other species the
15 largest differences occur in tropical regions. For example, the emission differences between
16 ORCHIDEE and MEGAN in the northern and southern tropics are $-2.2 \text{ Tg C yr}^{-1}$ and -2.1
17 Tg C yr^{-1} for methanol, 4.3 Tg C yr^{-1} and $10.2 \text{ Tg C yr}^{-1}$ for monoterpenes and 3.9 Tg C yr^{-1}
18 and 4.9 Tg C yr^{-1} for sesquiterpenes.

19 **3.2 Inter-annual and inter-seasonal emission variations**

20 Fig. 2 shows the annual and monthly global emission budgets of ORC_CRU and MEG_CRU.
21 The models have very similar annual trends and monthly variations for almost all compounds,
22 illustrating that climate variables, in particular temperature and solar radiation, are the major
23 driving factors at the global scale for inter-annual and inter-monthly variability.

24 Nevertheless large differences appear for isoprene. The emissions in ORC_CRU present a
25 clear seasonal cycle with an emission maximum in July and August that is not simulated in
26 MEG_CRU results. Indeed, the major differences can be identified in July and August, when
27 global emissions in MEG_CRU are, on average, lower by $11.5 \text{ Tg C month}^{-1}$ and 9.0 Tg C
28 month^{-1} compared with ORC_CRU. The monthly zonal average for tropical, temperate and
29 northern boreal latitudes regions are shown in Fig. 3. We observe, as mentioned in Sect. 3.1,

1 that the ORCHIDEE emissions are significantly higher in northern temperate regions
2 compared with MEGAN, with a marked seasonal cycle and the largest differences between
3 the two models occurring in summer. In July (August) in particular, calculated isoprene
4 emissions in ORC_CRU are about 4 Tg C month⁻¹ (5.5 Tg C month⁻¹) higher than in
5 MEG_CRU. In July (August), a further important contribution to the global emission peak is
6 attributed to the northern and southern tropics, where ORCHIDEE isoprene emissions are
7 higher, in total, by about 4 Tg C month⁻¹ (5 Tg C month⁻¹) in comparison to MEGAN in July
8 (August), (Fig. 3, first plot, left column).

9 MEGAN isoprene emissions are indeed dominant from the tropical regions, leading to an
10 overall stable global emission budget throughout the year (Fig. 2). The northern and southern
11 tropics have an opposite seasonal cycle, with isoprene emissions coming mostly from the
12 northern tropics between March and October and from the southern tropics for the rest of the
13 year (Fig. 3). The overall stable global emission budget is generally characteristic of the
14 compounds for which tropical regions are strong emitters all year round, such as
15 sesquiterpenes (Table 3 and Fig. 3). On the other hand, the global BVOC emissions for which
16 temperate regions are strong emitters will have a more marked seasonal cycle (Fig. 2), such as
17 for methanol and isoprene in ORCHIDEE.

18 Indeed, the two models exhibit a very different inter-seasonal variation in terms of isoprene
19 global emissions. Sindelarova et al. (2014) compared the monthly isoprene emissions time
20 series from different data-sets, showing, for some of them, an inter-seasonal variation similar
21 to ORCHIDEE, and, for others, no seasonal cycle. Based on our current knowledge, we
22 cannot establish which is the best representation because of the lack of long-term observations
23 at the global scale. However, we can extensively investigate why the differences between the
24 two models occur, performing sensitivity simulations and looking at the various processes
25 modelled. This is the main purpose of the next section.

26 Additionally, Fig. 3 shows that in northern and southern temperate and northern boreal
27 regions, the seasonal cycle is very similar between the two models, even if ORCHIDEE
28 calculates higher emissions than MEGAN, especially for isoprene.

1 3.3 Emission geographical distribution

2 The spatial patterns of BVOC emissions in winter and summer for ORC_CRU and
3 MEG_CRU simulations are presented in Figs. 5–9 for isoprene, monoterpenes, methanol,
4 acetone and sesquiterpenes. To better assess the impact of EFs on emissions, we show the
5 resulting emission potential for each grid cell, summing the EFs, each weighted by the cell
6 area occupied by each PFT. In MEGAN, emission potentials are already provided per grid
7 cell for isoprene, monoterpenes and MBO (see Sect. 2.3). Emission potentials per grid cell
8 can be interpreted as the average EFs associated with the ecosystem present in the grid cell.

9 For a particular compound, the formula to convert the ORCHIDEE EF ($\mu\text{gC g}^{-1} \text{h}^{-1}$) in the
10 potential emission ($\mu\text{g m}^{-2} \text{h}^{-1}$) consistent to those provided by MEGAN are, for emission not
11 depending on light (LDF = 0):

$$12 \quad EP = \sum_i EF_i \cdot M / M_{\text{Carbon}} \cdot LAI_{\text{REF}} \cdot SWL_i \cdot A_i \quad (6)$$

13 and for light-dependent emissions (LDF = 1):

$$14 \quad EP = \sum_i EF_i \cdot M / M_{\text{Carbon}} \cdot LAI_{\text{REF}} \cdot SWL_i \cdot A_i \cdot C_{\text{CE}} \quad (7)$$

15 where i is the index related to PFTs, M_{Carbon} and M are the molar mass of carbon and the
16 compound, respectively, LAI_{REF} equals to $5.0 \text{ m}^2 \text{ m}^{-2}$ is the LAI in MEGAN standard
17 conditions, SWL is the MEGAN specific leaf weight depending on PFTs, A is the PFT grid
18 fraction and C_{CE} is the canopy environment coefficient, a scaling factor dependent on the
19 canopy radiation module, which equals 0.57 in this MEGAN configuration (Guenther et al.,
20 2012).

21 In general, for every compound, we observe a similar geographical distribution. High
22 emission areas are identified in Brazil, equatorial Africa, southern East Asia and southern
23 East USA for both models, with values for ORCHIDEE (MEGAN) ranging between:
24 $5.0\text{--}12.0 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ($3.0\text{--}9.0 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$) for isoprene, $0.8\text{--}2.0 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$
25 ($0.6\text{--}1.3 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$) for monoterpenes, $0.3\text{--}1.2 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ($0.2\text{--}0.7 \cdot 10^{10} \text{ kg C}$
26 $\text{m}^{-2} \text{ s}^{-1}$) for methanol, $0.2\text{--}0.5 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ($0.1\text{--}0.3 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$) for acetone and
27 $0.4\text{--}0.6 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ($0.2\text{--}0.3 \cdot 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$) for sesquiterpenes, respectively. For
28 methanol, in summer, high emitting areas also appear in Europe and Russia, with values of

1 0.3–0.5 10^{10} kg C m⁻² s⁻¹ for ORCHIDEE and 0.1–0.3 10^{10} kg C m⁻² s⁻¹ for MEGAN.
2 Indeed, these regions are populated by temperate and boreal needleleaf evergreen trees, which
3 are strong methanol emitters (Table 3 and Fig. 7, last row).

4 In southeast China and south-eastern USA, for methanol, acetone and, to a lesser extent,
5 monoterpenes, ORCHIDEE emission estimates are higher than MEGAN. This is directly
6 linked to the larger fraction of temperate needleleaf evergreen trees (TeNeEv) in ORCHIDEE
7 in comparison to MEGAN (not shown), which are strong emitters of these compounds. The
8 emission potentials (last row, Figs. 6–8) show the same geographical pattern that is mainly
9 driven by the PFT distribution in these regions.

10 Other notable differences between the two models appear in South America for isoprene,
11 directly in relation with the EP distribution. The pattern of isoprene emission in MEGAN has
12 higher values in western Brazil, Bolivia and northern Argentina, while in ORCHIDEE the
13 values are more homogeneous, with higher emissions in central Brazil. The same pattern
14 differences are detected in the emission potential (Fig. 5, last row on the right), and we
15 therefore infer that the EP distribution drives the isoprene emission geographical distribution.
16 The same conclusion also holds for monoterpenes, where lower emissions along the
17 Amazonian river follow perfectly the lower EPs in this area. In general, comparing the
18 emission geographical distribution for each compound and the corresponding emission
19 potential, we can state that, in both models, emission spatial patterns are mostly affected by
20 the EF and PFT distributions.

21 **3.4 BVOC emission sensitivity to LAI**

22 In this section, we investigate in detail the differences between the two models arising from
23 LAI and we explore to what extent LAI can affect BVOC emission estimates.

24 Figs 4 and 10 show large differences in the geographical distribution and global average of
25 ORCHIDEE LAI and MODIS LAI (Yuan et al., 2011). As illustrated in Fig. 10, the global
26 monthly mean LAI calculated by ORCHIDEE is 1.5–2 m² m⁻² higher compared to the LAI
27 used in MEGAN and based on MODIS data-sets. In addition the LAI peaks at different times
28 throughout the year in ORCHIDEE and MEGAN. We investigate the contribution of different
29 areas and we observe that, whilst in northern temperate region the MODIS LAI peaks in July
30 and afterwards decreases quite fast, the ORCHIDEE LAI peak in both July and August.

1 Furthermore, in the boreal region, the ORCHIDEE LAI peaks one month later (August) than
2 the MODIS LAI (July). Therefore, the time shift observed globally is due to the greater
3 persistence of the growing season provided by ORCHIDEE in the northern temperate area
4 and its delay in the northern boreal region compared with what is detected by MODIS.

5 Furthermore, in the tropics, the MODIS LAI exhibits quite a clear seasonal cycle, especially
6 in Amazonia, Central Africa and Indonesia that is not simulated by ORCHIDEE (Fig. 4).

7 The differences between these LAI estimates are significant, but our current state of
8 knowledge does not allow us to establish which estimate is more reliable. Field and satellite
9 data bring very useful and complementary information regarding the order of magnitude, the
10 seasonal and the geographical variability of LAI. Nevertheless, inferring values for LAI on
11 small or large regional scales is particularly challenging, and data available from, either field
12 or satellite measurements also have significant uncertainties. Satellites, for instance, do not
13 measure the *real* LAI, but the *effective* LAI obtained from indirect optical methods and
14 strongly determined by the *a priori* assumptions necessary for the inversion procedure. Even
15 starting from the same input reflectance, diverse retrieval methods can lead to LAI values that
16 are highly different (Garrigues et al., 2008; Fang et al., 2013). The effective LAI can be very
17 dissimilar to the LAI directly measured in situ and relative differences can reach 100% (Fang
18 et al. 2012a, b).

19 The transition from *effective* to *real* LAI is possible only when additional information about
20 the vegetation structure is available (Pinty et al. 2011), increasing the risk of inaccuracy. The
21 sources of uncertainties are numerous (Garrigues et al., 2008). First, foliage clumping is, in
22 general, not taken into account, leading to underestimates of LAI of up to 70% over the
23 coniferous forest. Second, the forest understory is not systematically taken into account since
24 the satellite LAI product is derived from a vertical integrated radiation signal. Third, in dense
25 canopies, such as broadleaf tropical forests, the optical signal can saturate, leading to an
26 underestimate of the effective LAI in comparison with the true value with a saturation limit of
27 $3.0 \text{ m}^2 \text{ m}^{-2}$ (Pinty et al. 2011). Forth, the presence of ice and snow can strongly upset the
28 retrieval, making it very difficult to estimate LAI in boreal and mountain regions.

29 Conversely, in a validation study using satellite-derived vegetation index time series,
30 Maignan et al. (2011) pointed out some weaknesses in the ability of ORCHIDEE to correctly
31 model the LAI seasonal cycle, especially in the equatorial forest (Amazonia, central Africa,

1 Indonesia) where a poor correlation of model output with satellite data was demonstrated. In
2 general, quite large and comparable uncertainty is found when different LAI databases are
3 compared. Krinner et al. (2005) found that the difference between ORCHIDEE and MODIS
4 satellite LAI (Myneni et al., 2002) is as much as the difference between the satellite data that
5 they used and an alternative satellite vegetation cover data set (Tucker et al., 2001). Therefore
6 given the many existing limitations, we cannot precisely estimate to which extent
7 ORCHIDEE LAI is reliable. It is likely that the ORCHIDEE LAI modelisation has room for
8 improvement and a possible component to be upgraded is the allocation of the different
9 carbon stocks, but further investigations are needed. Performing a robust evaluation of the
10 model's ability to simulate the LAI, especially at the global scale, still remains challenging,
11 and is beyond the scope of our study.

12 In this context, model inter-comparison and sensitivity tests give an essential insight to assess
13 the impact of different LAI estimates and their uncertainties on BVOC emissions.

14 **3.4.1 LAI seasonal cycle impact**

15 LAI has an important role in driving the seasonal cycle of emissions. To show this, we
16 perform an extra 10-year simulation following the same configuration as in the previous runs,
17 but forcing MEGAN with the ORCHIDEE LAI (MEG_CRULAI simulation, Table 5) and we
18 compare the results with MEG_CRU and ORC_CRU simulations.

19 First of all, we observe that, for the MEG_CRU simulation, the isoprene emission seasonal
20 cycle in the tropics (particularly in the South) is more marked than for ORC_CRU simulation
21 (Fig. 11). This behaviour is principally related to the differences in seasonal variation between
22 the MODIS and the ORCHIDEE LAI (Fig. 4), since the ORCHIDEE LAI presents smaller
23 variations between winter and summer in tropical regions, in particular in Amazonia, (Fig. 4,
24 left column) in comparison with MODIS LAI (Fig. 4, right column). Whereas, the two models
25 have a similar inter-seasonal variability when they are driven by the same LAI
26 (MEG_CRULAI and ORC_CRU). Moreover, MEG_CRULAI simulation gives a lower peak
27 in the northern tropics April and May emission than MEG_CRU (Fig. 11), being more similar
28 to ORC_CRU.

1 Generally, for every compound, we observe a better agreement between the MEG_CRULAI
2 and the ORC_CRU simulations than between MEG_CRU and ORC_CRU, especially in the
3 tropical regions.

4 **3.4.2 LAI range**

5 The global and zonal emission budgets (Table 7) in the MEG_CRULAI simulation are not
6 significantly different than those determined in MEG_CRU, even if the ORCHIDEE LAI is
7 significantly higher than MODIS LAI, suggesting a low sensitivity of MEGAN to LAI size
8 Indeed, at the regional scale, in boreal and temperate regions, the MEG_CRULAI emissions
9 are slightly higher than those in MEG_CRU, and in the tropics they are even slightly lower
10 for some compounds. As proposed by Sindelarova et al. (2014), a possible reason for the
11 emission decrease calculated in the tropics by MEGAN is to the strengthened effect of leaf
12 self-shading caused by an increase in LAI in locations characterized by a dense vegetation
13 (e.g. in central Africa or Amazonia). This effect can be predominant for compounds for which
14 biogenic emissions are strongly dependent on light, such as isoprene or methanol.

15 Indeed, for the other compounds the MEG_CRU and MEG_CRULAI emission budgets are
16 very similar. We could foresee that these results are linked to the leaf self-shading effect on
17 leaf temperature. In contrast to ORCHIDEE, where the air temperature is used, in MEGAN
18 the leaf temperature is calculated for shaded and sunlit leaves. If the leaf self-shading effect
19 was crucial even for light-independent compounds, we would expect a much higher leaf
20 temperature for sunlit leaves than for shaded leaves. Calculating the difference in hourly leaf
21 temperature between sunlit and shaded leaves in the case of dense vegetation (TrBrEv and
22 TrBrDe), we estimate differences of about 1–1.5 °C, which would unlikely be high enough to
23 explain such differences in emissions. Lathière et al. (2006), for instance, found that an
24 increase in the global surface temperature by 1°C leads to an increase of isoprene emissions
25 of at most 11%. We therefore doubt that the only mechanism behind the static BVOC
26 emissions for light-independent compounds is leaf self-shading.

27 We, therefore, investigate in more detail whether models show the same response to a
28 particular change in LAI. We perform two extra simulations for each model, using the
29 ORCHIDEE LAI multiplied by a factor of 0.5 or 1.5. The scaling factor considered are
30 consistent with the LAI uncertainties (see the beginning of Sect. 3.4). Fig. 12 shows the four

1 simulations: MEG_LAI05, ORC_LAI05 (ORCHIDEE LAI multiplied by 0.5) and
2 MEG_LAI15 and ORC_LAI15 (ORCHIDEE LAI multiplied by 1.5), for the year 2006
3 (details in Table 5). Only the zonal average for the tropics and southern and northern
4 temperate areas, for isoprene and monoterpenes are displayed, but they are also representative
5 of other regions.

6 Regarding isoprene, we observe that ORCHIDEE and MEGAN present a similar response to
7 LAI variation. When the LAI is multiplied by a factor of 0.5 (1.5), change in emissions
8 compared to the reference runs (MEG_CRULAI, ORC_CRU) reaches -18% ($+12\%$) for
9 MEGAN and -21% ($+8\%$) for ORCHIDEE in the southern tropics, and reaches -34%
10 ($+21\%$) for MEGAN and -32% ($+16\%$) for ORCHIDEE in northern temperate areas. In the
11 tropics especially, the emissions calculated by the two models are little sensitive to the LAI
12 increase. Indeed isoprene is a light-dependent compound thus, beyond a given LAI threshold,
13 the contribution of the highest LAI layers is very low, as there is no more or very little direct
14 light available. We observe that MEGAN is less sensitive than ORCHIDEE to an LAI
15 increase, which is likely due to the different parameterisation of CTLD factor in the two
16 models as described Sect. 2.5. In more details, as LAI increases, the growth of sunlit leaves
17 fraction is dumped by an exponential factor as in Spitter et al. (1986b), implying lower
18 contribution of sunlit leaves with respect to shaded leaves for high LAI values. In MEGAN,
19 for equal incoming radiation, the relative contribution of sunlit leaves, with respect to shaded
20 leaves, is roughly twice than in ORCHIDEE. This explains the different sensitivity of the two
21 models.

22 Monoterpene emissions show a different response in terms of sensitivity to LAI. In the
23 southern tropics, the relative difference in monoterpene emission budget between
24 ORC_LAI05 (ORC_LAI15) and ORC_CRU is -43% ($+40\%$), and -9% ($+3\%$) between
25 MEG_LAI05 (MEG_LAI15) and MEG_CRULAI. In northern temperate regions, the relative
26 difference in the monoterpene emission budget between ORC_LAI05 (ORC_LAI15) and
27 ORC_CRU is -44% ($+40\%$), and -14% ($+6\%$) between MEG_LAI05 (MEG_LAI15) and
28 MEG_CRULAI. These simulations confirm a much smaller emission impact of LAI variation
29 on emissions in MEGAN, even for compounds not fully dependent on light, such as
30 monoterpenes (LDF=0.6).

1 Table 8 shows the total emission budget calculated for MEG_LAI05, ORC_LAI05,
2 MEG_LAI15 and ORC_LAI15 simulations for every compound. In general in ORCHIDEE,
3 the lower the light dependence, the higher the sensitivity to LAI, while for MEGAN, the
4 sensitivity to LAI does not significantly change with LDF. The explanation for this difference
5 in emission response lies in the different formulation for light independent emissions in the
6 two models. Such differences are detailed in point 6 of Sect. 2.5. In particular, in
7 ORCHIDEE, the light independent emission linearly depends on LAI whereas, in MEGAN it
8 is determined by the γ_{LAI} factor and it varies almost linearly for low LAI ($< 2 \text{ m}^2 \text{ m}^{-2}$) and
9 then more and more slowly up to become almost constant for LAI higher than $5 \text{ m}^2 \text{ m}^{-2}$. The
10 light-independent emission descriptions in the two models therefore respond differently to
11 LAI variation, with differences more striking when LAI is greater than $2 \text{ m}^2 \text{ m}^{-2}$, while the
12 ORCHIDEE emissions keep increasing linearly with LAI, the MEGAN emissions do not
13 increase as strongly anymore. In this case, the different modelling choices bring significant
14 discrepancies in emission sensitivity between the two models.

15 **3.4.3 MODIS LAI**

16 Considering the high sensitivity of BVOC emissions to the LAI and the high differences
17 between ORCHIDEE and MODIS LAI, we perform an additional simulation, forcing
18 ORCHIDEE with the LAI provided by MODIS (ORC_CRUMOD) for the year 2006. Details
19 of ORC_CRUMOD are provided in Table 5. In Fig. 13, we present the differences between
20 the seasonal averages of ORC_CRUMOD and ORC_CRU for monoterpenes and isoprene
21 emissions. In ORC_CRUMOD, isoprene emissions significantly decrease in the tropics, up to
22 $3\text{--}6 \cdot 10^{-10} \text{ kgC m}^{-2} \text{ s}^{-1}$ in Brazil, in the African savanna, India and Northern Australia, while
23 they increase up to $0.75\text{--}1 \cdot 10^{-10} \text{ kgC m}^{-2} \text{ s}^{-1}$ in some areas of South America, Australia, and
24 Africa and up to $1\text{--}3 \cdot 10^{-10} \text{ kgC m}^{-2} \text{ s}^{-1}$ in equatorial Africa. The monoterpene emissions
25 decrease almost everywhere, especially in many tropical and equatorial areas and northern
26 temperate and boreal areas (up to $0.5 \cdot 10^{-10} \text{ kgC m}^{-2} \text{ s}^{-1}$).

27 Fig. 13 also illustrates the seasonal variation for both isoprene and monoterpene emissions in
28 the tropics, and clearly shows that the use of MODIS LAI implies a seasonality in tropical and
29 equatorial emissions which is almost not present in ORC_CRU simulation. Confirming the
30 results presented in section 3.4.2, monoterpene emissions show higher sensitivity to LAI

1 variations than isoprene, with the monoterpene annual global budget for ORC_CRUMOD
2 being 32% lower than for ORC_CRU, while for isoprene, the annual global budget is 6%
3 lower. Considering the other species, the impact of using the MODIS LAI is stronger for
4 species with a lower LDF. The relative difference between ORC_CRUMOD and ORC_CRU
5 is -4% for methanol, -30% for acetaldehyde, formaldehyde, acetic acid and formic acid,
6 -36% for acetone and -44% for MBO.

7

8 **3.5 BVOC emission sensitivity to LDF**

9 As described in Sect. 2.2, the LDF parameter sets the light-dependent fraction of emissions
10 for each compound. Many experimental studies point out for several plant species that, if
11 emissions can be totally light-independent for some BVOCs, the emissions of most of them
12 are actually light-dependent to a degree that depends on the compound (Jacob et al. 2002,
13 2005; Hansen and Seufert, 2003; Dindorf et al., 2006; Holzke et al., 2006; Harley et al., 2007;
14 Millet et al., 2008, 2010; Hu, et al., 2011; Wells et al., 2014). Since the results of these studies
15 are highly heterogeneous, assigning a single LDF value to each compound is as difficult as
16 assigning the EFs to each PFT (Sect. 2.2). Hence, the LDF uncertainty could be even higher
17 than the uncertainties associated with EFs, as there have been fewer less quantitative studies
18 on this subject published to date.

19 The objective of this section is to quantify, for both ORCHIDEE and MEGAN, the relative
20 contribution of the light-dependent and light-independent part to the total emissions, and
21 consequently to determine the impact of LDF-attributed values on emission estimates, giving
22 clues to better understand the different behaviours of the two models.

23 For the fully light dependent (isoprene: LDF=1) or largely light dependent compounds
24 (methanol: LDF=0.8) (Fig. 5 and Fig. 7), we observe that a higher EP in ORCHIDEE than in
25 MEGAN does not necessarily lead to higher emissions in ORCHIDEE. In the case of a LDF
26 close to 1, even when the same EP value is used in both models, the emissions calculated by
27 MEGAN are higher compared to ORCHIDEE, suggesting a different emissions response to
28 light. Indeed, this effect is less important for compounds which are less dependent on light
29 such as monoterpenes (LDF = 0.5) (Fig. 6) and sesquiterpenes (LDF = 0.6) (Fig. 9), and
30 indeed are even negligible for acetone (LDF = 0.2) (Fig. 8). It therefore seems that the choice

1 of LDF parameter can be crucial in the emission estimate and in the sensitivity to EF
2 variation.

3 To isolate the signal related to the LDF, we investigate the hourly variation of two “test
4 compounds”, the first defined as light independent (LDF = 0) and the second defined as
5 totally light-dependent (LDF = 1). All EFs are set to $1 \mu\text{gC g}^{-1} \text{h}^{-1}$ for each PFT. The other
6 settings are specified as in the reference run and are the same for the two test compounds (for
7 further details see Table 5). We refer in the text to the first compound as *orcldf0* if it is
8 calculated by ORCHIDEE and as *megldf0* if it is calculated by MEGAN, while we refer to the
9 second compounds as *orcldf1* and *megldf1*, respectively.

10 In order to quantify the contribution of the light-dependent part in comparison to the light-
11 independent one, we use the LDF index, which we define as the ratio between the light-
12 dependent and the light-independent test compound, multiplied by 100 (*orcldf1/orcldf0*·100,
13 *megldf1/megldf0*·100). Using the LDF index we can easily compare the behaviour of the two
14 models, avoiding the complication arising from the mismatch between the two land covers.
15 Indeed, the direct comparison of the absolute values of *orcldf* and *megldf* compounds could
16 be affected by the differences between the PFT distributions in the two models, and the signal
17 due to LDF change could therefore not be well isolated.

18 In Fig. 14 the daily profile averaged over each month of the LDF index is presented to
19 investigate the daily and annual variations. At the global scale (left panel), we observe that the
20 LDF index associated with MEGAN is much higher (up to 20%), than the index associated
21 with ORCHIDEE. At the regional scale, in the southern tropics for example (second panel)
22 the index reaches up to 70% and is the twice as large the index calculated for ORCHIDEE.
23 The light-dependent part of the emissions in MEGAN is therefore more important than
24 ORCHIDEE, with important impacts on emission estimates. Firstly, we show that based on
25 the same EF value, the MEGAN emissions are higher than in ORCHIDEE for compounds
26 associated with high LDF, as expected from Sect. 3.3.

27 Secondly, the variable *orcldf0* (*megldf0*) represents the emissions when LDF is zero while
28 *orcldf1* (*megldf1*) represents the emissions when LDF is one; thus, they define the interval
29 spanned by emissions as LDF varies. Therefore, a low LDF index is associated with a greater
30 variability of emissions for equal light-independent emissions. Consequently, ORCHIDEE
31 results more sensitive to LDF variation than MEGAN, as the ORCHIDEE LDF index is lower

1 than the MEGAN index. Furthermore, the LDF index provides an evaluation of error due to a
2 diverse choice of LDF values. The LDF index is always less than 100, meaning that the light-
3 independent component of the emission is always bigger than the light-dependent part.
4 Therefore, if LDF in the model is greater than it should be, emissions will be underestimated,
5 while if it is less, emissions will be overestimated. At regional scale, tropical areas, that are
6 associated to high LDF index, will be less sensitive to LDF variation than other regions.

7

8 **4 Conclusions**

9 The main objectives of this study were to (i) present the new version of the BVOC emission
10 module embedded in the ORCHIDEE model, (ii) provide BVOC emission estimates for the
11 2000–2009 period for a large diversity of compounds, (iii) compare the ORCHIDEE model
12 results to emissions calculated by MEGAN in terms of global, regional and seasonal patterns,
13 and (iv) investigate how the uncertainty linked to some key variables or parameters such as
14 the LAI and the LDF could affect the BVOC emission estimate in the two models.

15 The new ORCHIDEE emission module now considers many speciated monoterpenes and
16 bulk sesquiterpenes, that have been shown to be important regarding SOA formation, uses
17 updated EFs and includes developments in the physical processes related to BVOC formation,
18 such as the emission dependence on light for almost all compounds, a multi-layer calculation
19 of diffuse and direct radiation, and shaded and sunlit leaves over LAI layers.

20 The ORCHIDEE emission estimates are within the range of the published emission budgets.
21 The ORCHIDEE global budgets averaged over the period investigated (2000–2009) are 465
22 Tg C yr⁻¹ for isoprene, 108 Tg C yr⁻¹ for monoterpenes, 38 Tg C yr⁻¹ for methanol, 25 Tg C
23 yr⁻¹ for acetone and 24 Tg C yr⁻¹ for sesquiterpenes. The global emission budgets are, in
24 general, in good agreement between the two models, with the ORCHIDEE emissions being
25 8% higher for isoprene, 8% lower for methanol, 17% higher for acetone, 18% higher for
26 monoterpenes and 39% higher for sesquiterpenes compared to the MEGAN results. At the
27 regional scale, the largest differences in terms of spatial emission distribution between
28 ORCHIDEE and MEGAN occur in the northern temperate region for isoprene. This different
29 behaviour is directly linked to differences in the EF and PFT distribution in this area.

1 More generally, considering the geographical distribution of emissions for each compound
2 and the corresponding emission potential, we show that, in both models, EF and PFT
3 distributions are the main drivers of the geographical emission pattern. In terms of seasonal
4 variation, the differences between the two models in the tropics are mostly due to the different
5 seasonal cycles of LAI between MODIS and ORCHIDEE, while the large discrepancy in
6 northern temperate regions is attributed to differences in the EF distribution.

7 The LAI calculated by ORCHIDEE is $1.5\text{--}2\text{ m}^2\text{ m}^{-2}$ higher than the LAI retrieved by MODIS.
8 We examined how these discrepancies can impact the BVOC estimates. Sensitivity tests
9 performed forcing both models with the ORCHIDEE LAI multiplied by a factor of 0.5 and
10 1.5 showed that, for isoprene, ORCHIDEE and MEGAN emissions present a similar response
11 to these LAI variations. Conversely, for monoterpenes, ORCHIDEE is much more sensitive
12 to LAI variations in comparison to MEGAN. These discrepancies are due to differences in the
13 light-independent emission formulation between the two models. In ORCHIDEE, the
14 dependence of emissions on LAI is linear, while in MEGAN it is quasi-linear for LAI up to 2
15 $\text{m}^2\text{ m}^{-2}$, then the increase is progressively reduced to become nearly constant for LAI greater
16 than $5\text{ m}^2\text{ m}^{-2}$. The sensitivity test performed forcing ORCHIDEE with MODIS LAI,
17 confirmed that in tropical areas the principal differences between ORCHIDEE and MEGAN
18 BVOC estimation come from the LAI, and that compounds with lower LDF show a higher
19 sensitivity to LAI variation.

20 We investigated the contribution of the light-dependent and light-independent part of
21 emissions and consequently the impact that a different choice of LDF can have on emissions.
22 In MEGAN, the light-independent part of emissions is more important than in ORCHIDEE,
23 reaching a factor of two in the southern tropics. We find that ORCHIDEE estimates are more
24 sensitive to LDF variation than MEGAN. Moreover, we showed that overestimation
25 (underestimation) of the LDF value leads to emission underestimation (overestimation).

26

27 **5 Future directions**

28 Model inter-comparison and sensitivity tests are extremely useful to define which
29 parameters/variables mainly affect BVOC emissions, what is the cause of this sensitivity, and
30 how estimates can be improved. Previous works have already investigated the impact of

1 different experimental set-ups (climate forcing and vegetation distribution) (Arneeth et al.,
2 2011), differences in the canopy structure description (Keenan et al., 2011) and land cover
3 classification (Oderbolz et al., 2013) on emissions.

4 In the present work, we focused on the impact of LAI, LDF, EFs and PFT distribution. Our
5 results underline that the high uncertainties in the involved variables/parameters and the
6 different choices in modelling processes result in a high variability of BVOC emission
7 estimates. The outcome of this analysis provides some guidelines for future developments of
8 BVOC emission models at the global scale. In particular, the following issues should be
9 carefully addressed:

- 10 - LAI uncertainties are still extremely high and have a considerable impact on
11 emissions. Improvements in LAI modelling or estimation at the global scale are
12 essential;
- 13 - EF allocation is a big concern because of its high variability. A proper way to assign
14 statistically robust values at a global scale has not yet been found. Significant
15 improvement can be achieved only by increasing the observation data coverage of
16 many regions and performing long-term measurements;
- 17 - LDF parameterisation is still oversimplified and has a significant impact on emissions.
18 Future developments should, therefore, improve LDF parameterization accuracy, for
19 example, by including PFT dependency. As for EFs, more reliable results can be
20 achieved only by increasing observation coverage;
- 21 - the rather low number of PFTs in global models is a limiting factor in accurate
22 emission estimates;

23 Further analysis will certainly be needed to include other important parameters/variables in
24 the investigation, for example, leaf temperature versus air temperature usage, leaf age classes,
25 parameters in the Guenther formulation, and the soil moisture activity factor.

26 Finally, it is worth mentioning that, besides model inter-comparison, there is a strong need to
27 evaluate model results against emission observations. This has already been done in other
28 domains, for example in atmospheric chemistry modelling (Mann et al., 2014; Tsigaridis et
29 al., 2014). In the case of BVOC, however, observational data are very challenging to acquire,

1 especially on the long-term scale. Therefore, for BVOC emission modelling, a robust
2 validation of model results against observations is still lacking.

3

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13

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1 Table 1. Plant Functional Types in ORCHIDEE and MEGAN and corresponding occupied
 2 surfaces in 10^{12} m².

PFT acronym		PFT full name		PFT surface	
ORCHIDEE	MEGAN	ORCHIDEE	MEGAN	ORCHIDEE	MEGAN
	BaSo		Bare soil	21.43	40.30
	TrBrEv		Tropical broadleaf evergreen tree	12.84	11.40
	TrBrDe		Tropical broadleaf deciduous tree	7.49	5.82
	TeNeEv		Temperate needleleaf evergreen tree	4.50	3.43
	TeBrEv		Temperate broadleaf evergreen tree	4.04	1.81
	TeBrDe		Temperate broadleaf deciduous tree	5.79	4.45
	BoNeEv		Boreal needleleaf evergreen tree	5.74	9.71
	BoBrDe		Boreal broadleaf deciduous tree	5.14	1.68
	BoNeDe		Boreal needleleaf deciduous tree	1.98	1.47
C3Gr	C3GrCold C3GrCool	C3 Grass	C3 Grass Cold C3 Grass Cool	37.00	4.20 12.55
	C4Gr		C4 Grass	14.89	11.025
C3Ag C4Ag	Crop	C3 Agriculture C4 Agriculture	Crop	10.19 4.88	14.58
-	TeSbEv	-	Temperate shrub evergreen	-	0.074
-	TeSbDe	-	Temperate shrub deciduous	-	5.39
-	BoSbD	-	Boreal shrub deciduous	-	8.02

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1 Table 2. Comparison between the old and new versions of the biogenic emission module in
 2 ORCHIDEE: list of emitted compounds, principal parameters for emission equations,
 3 radiation model type and compounds for which the leaf emission activity is activated.

	Output Species	Light (LDF) and temperature dependence (Beta) function		Radiation model type	Species with leaf age activation
		Species	LDF Beta		
ORCHIDEE new version	methanol, acetone, acetaldehyde, formaldehyde, acetic acid, formic acid, total monoterpene, α -pinene, β -pinene, limonene, myrcene, sabinene, camphene, 3-carene, t- β -ocimene, other monoterpenes, sesquiterpene, MBO, Other VOCs	isoprene, MBO	1.0 -	Light multilayer vertical profile to calculate radiation extinction inside the canopy for both sunlit and shaded leaves	isoprene methanol
		acetaldehyde, formaldehyde, acetic acid, formic acid	0.8 0.10		
		acetone	0.2 0.10		
		methanol	0.8 0.8		
		total monoterpene, α -pinene, β -pinene, limonene, myrcene, sabinene, camphene 3-carene, t- β -ocimene, other monoterpenes	0.6 0.6		
		total sesquiterpene	0.5 0.17		
ORCHIDEE old version	methanol, acetone, acetaldehyde, formaldehyde, acetic acid, formic acid, total monoterpene, MBO, other VOCs	isoprene, MBO	1.0 0.9	One layer	isoprene methanol

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1 Table 3. Emission Factors (EFs, $\mu\text{gC g}^{-1} \text{h}^{-1}$) for each PFT for the main compounds emitted, in the previous (first line) and actual (second line,
 2 in bold) version of the ORCHIDEE emission module. The list of references used to set the new values is provided in the last column.

	TrBrEv	TrBrDe	TeNeEv	TeBrEv	TeBrDe	BoNeEv	BoBrDe	BoNeDe	C3Gr	C4Gr	C3Ag	C4Ag	References
Isoprene	24.0 24.0	24.0 24.0	8.0 8.0	16.0 16.0	45.0 45.0	8.0 8.0	8.0 18.0	8.0 0.5	16.0 12.0	24.0 18.0	5.0 5.0	5.0 5.0	He et al., 2000; Klinger et al., 2002; Levis e al., 2003; Stewart et al., 2003; Padhy and Varshney, 2005; Bai et al., 2006; Geron et al., 2006; Guenther et al., 2006, 2012; Smiatek and Steinbrecher, 2006; Karl et al., 2007, 2009; Steinbrecher et al. 2009; Tsui et al., 2009; Lathière et al., 2006; Leung et al., 2010; Bracho-Nunez et al., 2011; .
Monoterp.	0.800 2.000	0.800 2.000	2.400 1.800	1.200 1.400	0.800 1.600	2.400 1.800	2.400 1.400	2.400 1.800	0.800 0.800	1.200 0.800	0.200 0.220	0.200 0.220	He et al., 2000; Janson and De Serves, 2001; Stewart et al., 2003; Hayward et al., 2004; Karl et al., 2004, 2007, 2009; Bai et al., 2006; Geron et al., 2006; Hakola et al., 2006; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Helmig et al., 2007; Ortega et al., 2008; Steinbrecher et al. 2009; Kim et al., 2010; Bracho-Nunez et al., 2011; Fares et al., 2011; Guenther et al., 2012.
Sesquiterp.	- 0.450	- 0.450	- 0.130	- 0.300	- 0.360	- 0.150	- 0.300	- 0.250	- 0.600	- 0.600	- 0.080	- 0.080	Lathière et al., 2006; Helmig et al., 2007; Duhl et al., 2008; Matsunaga et al., 2009; Steinbrecher et al. 2009; Karl et al., 2009; Ortega et al., 2008; Bracho-Nunez et al., 2011; Hakola et al., 2006; Kim et al., 2010; Fares et al., 2011; Guenther et al., 2012.
Methanol	0.600 0.800	0.600 0.800	1.800 1.800	0.900 0.900	0.600 1.900	1.800 1.800	1.800 1.800	1.800 1.800	0.600 0.700	0.900 0.900	2.000 2.000	2.000 2.000	Schade and Goldstein, 2001; Geron et al., 2002; Karl et al., 2004, 2005, 2009; Hayward et al., 2004; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Harley et al., 2007; Steinbrecher et al. 2009; Bracho-Nunez et al., 2011; Fares et al., 2011; Guenther et al., 2012.
Acetone	0.290 0.250	0.290 0.250	0.870 0.300	0.430 0.200	0.290 0.300	0.870 0.300	0.870 0.250	0.870 0.250	0.290 0.200	0.430 0.200	0.070 0.080	0.070 0.080	Janson and De Serves 2001; Schade and Goldstein, 2001; Karl et al., 2004, 2005, 2009; Villanueva-Fierro et al., 2004; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Steinbrecher et al. 2009; Bracho-Nunez et al., 2011; Fares et al., 2011; Guenther et al., 2012.
Acetaldeh.	0.100 0.200	0.100 0.200	0.300 0.200	0.150 0.200	0.100 0.250	0.300 0.250	0.300 0.160	0.300 0.160	0.100 0.120	0.150 0.120	0.025 0.035	0.025 0.022	Kesselmeier et al., 1997; Schade and Goldstein, 2001; Hayward et al., 2004; Karl et al., 2004, 2005, 2009; Villanueva-Fierro et al., 2004; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Steinbrecher et al. 2009; Fares et al., 2011; Guenther et al., 2012.
Formaldehy.	0.070 0.040	0.070 0.040	0.200 0.080	0.100 0.040	0.070 0.040	0.200 0.040	0.200 0.040	0.200 0.040	0.070 0.025	0.100 0.025	0.017 0.013	0.017 0.013	Kesselmeier et al., 1997; Janson et al., 1999; Villanueva-Fierro et al., 2004; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Chang et al., 2009; Karl et al., 2009; Steinbrecher et al. 2009; Guenther et al., 2012.
Acetic acid	0.002 0.025	0.002 0.025	0.006 0.025	0.003 0.022	0.002 0.080	0.006 0.025	0.006 0.022	0.006 0.013	0.002 0.012	0.003 0.012	0.001 0.008	0.001 0.008	Kesselmeier et al., 1997, 1998; Staudt and Kesselmeier, 2000; Villanueva-Fierro et al., 2004; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Karl et al., 2009; Steinbrecher et al. 2009; Guenther et al., 2012.
Formic Acid	0.010 0.015	0.010 0.015	0.030 0.020	0.015 0.020	0.010 0.025	0.030 0.015	0.030 0.015	0.030 0.015	0.010 0.010	0.0150 0.010	0.0025 0.008	0.0025 0.008	Kesselmeier et al. 1997, 1998; Staudt and Kesselmeier, 2000; Villanueva-Fierro et al., 2004; Lathière et al., 2006; Smiatek and Steinbrecher, 2006; Chang et al., 2009; Karl et al., 2009; Steinbrecher et al. 2009; Guenther et al., 2012.
MBO	0.000 0.00002	0.000 0.00002	20.000 1.4	0.000 0.00002	0.000 0.00002	0.000 0.14	0.000 0.00002	0.000 0.00002	0.000 0.00002	0.000 0.00002	0.000 0.00002	0.000 0.00002	Tarvainen et al., 2005; Hakola et al., 2006; Lathière et al., 2006; Chang et al., 2009; Kim et al., 2010; Guenther et al., 2012.

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1 Table 4. Percentage of speciated monoterpene EFs with respect to the PFT bulk monoterpene EF (forth line, in bold the Table 3) in the new
 2 version of the ORCHIDEE emission module.

	TrBrEv	TrBrDe	TeNeEv	TeBrEv	TeBrDe	BoNeEv	BoBrDe	BoNeDe	C3Gr	C4Gr	C3Ag	C4Ag	
α-Pinene	39.5	39.5	35.4	46.3	32.6	35.4	31.6	66.2	23.1	20.0	27.7	27.7	Janson et al., 1999; He et al., 2000; Janson and De Serves 2001; Schade and Goldstein, 2001; Greenberg et al. 2004; Villanueva-Fierro et al., 2004; Tarvainen et al 2005; Geron et al., 2006; Ortega et al., 2008; Smiatek and Steinbrecher, 2006; Dominguez-Taylor et al., 2007; Karl et al., 2007, 2009; Steinbrecher et al. 2009; Guenther et al., 2012
β-Pinene	11	11	14.6	12.2	8.7	14.6	6.3	15.0	12.3	8.0	15.4	15.4	Janson et al., 1999; He et al., 2000; Janson and De Serves 2001; Villanueva-Fierro et al., 2004; Tarvainen et al 2005; Geron et al., 2006; Smiatek and Steinbrecher, 2006; Dominguez-Taylor et al., 2007; Karl et al., 2007, 2009; Ortega et al., 2008; Steinbrecher et al. 2009; Guenther et al., 2012
Limonene	9.2	9.2	8.3	12.2	6.1	8.3	7.1	3.7	14.6	28.0	9.2	9.2	Janson et al., 1999; He et al., 2000; Janson and De Serves 2001; Villanueva-Fierro et al., 2004; Bai et al., 2006; Geron et al., 2006; Smiatek and Steinbrecher, 2006; Dominguez-Taylor et al., 2007; Karl et al., 2007, 2009; Ortega et al., 2008; Steinbrecher et al. 2009; Guenther et al., 2012
Myrcene	7.3	7.3	5.0	5.4	2.8	5.0	1.9	2.5	6.2	5.7	4.6	4.6	Janson et al., 1999; He et al., 2000; Janson and De Serves 2001; Villanueva-Fierro et al., 2004; Geron et al., 2006; Smiatek and Steinbrecher, 2006; Karl et al., 2007, 2009; Ortega et al., 2008; Steinbrecher et al. 2009; Guenther et al., 2012
Sabinene	7.3	7.3	5.0	8.3	0.4	5.0	26.3	3.0	6.5	5.0	6.2	6.2	He et al., 2000; Tarvainen et al 2005; Smiatek and Steinbrecher, 2006; Karl et al., 2007, 2009; Ortega et al., 2008; Steinbrecher et al. 2009; Guenther et al., 2012
Camphene	5.5	5.5	4.2	4.9	0.4	4.2	0.5	2.3	5.4	5.3	3.1	3.1	Janson et al., 1999; He et al., 2000; Janson and De Serves 2001; Tarvainen et al 2005; Bai et al., 2006; Geron et al., 2006; Smiatek and Steinbrecher, 2006; Karl et al., 2007, 2009; Ortega et al., 2008; Steinbrecher et al. 2009; Guenther et al., 2012;
β-Carene	4.8	4.8	17.5	1.0	2.4	17.5	1.3	4.2	6.5	5.7	20.0	20.0	Janson et al., 1999; He et al., 2000; Janson and De Serves 2001; Villanueva-Fierro et al., 2004; Tarvainen et al 2005; Bai et al., 2006; Hakola et al., 2006; Smiatek and Steinbrecher, 2006; Dominguez-Taylor et al., 2007; Karl et al., 2007, 2009; Steinbrecher et al. 2009; Ortega et al., 2008; Guenther et al., 2012;
<i>t</i>-β-Ocimene	9.2	9.2	5.4	4.4	11.3	5.4	10.5	2.8	13.8	12.0	3.1	3.1	Steinbrecher et al. 2009; Karl et al., 2009; Ortega et al., 2008; Guenther et al., 2012
Other Monoterpene	6.2	6.2	4.6	5.3	5.3	4.6	14.5	0.3	11.6	10.3	10.7	10.7	Janson et al., 1999; He et al., 2000; Janson and De Serves, 2001; Stewart et al., 2003; Hayward et al., 2004; Karl et al., 2004, 2007, 2009; Spirig et al., 2005; Tarvainen et al., 2005; Bai et al., 2006; Geron et al., 2006; Hakola et al., 2006; Smiatek and Steinbrecher, 2006; Helmig et al., 2007; Ortega et al., 2008; Steinbrecher et al. 2009; Kim et al., 2010; Bracho-Nunez et al., 2011; Fares et al., 2011; Guenther et al., 2012.

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1 Table 5. Configuration of simulations performed by ORCHIDEE and by MEGAN.

Simulation Name	Model	Climate Forcing	EFs	LDF	LAI	T	Period	Output frequency
ORC_CRU	ORCHIDEE	CRU	Standard version	Standard version	ORCHIDEE LAI	T air	2000-2009	1 month
MEG_CRU	MEGAN	CRU	Standard version	Standard version	MODIS LAI	T leaf	2000-2009	1 month
MEG_CRULAI	MEGAN	CRU	Standard version	Standard version	ORCHIDEE LAI	T leaf	2000-2009	1 month
ORC_LAI05	ORCHIDEE	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 0.5	T air	2006	1 month
ORC_LAI15	ORCHIDEE	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 1.5	T air	2006	1 month
MEG_LAI05	MEGAN	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 0.5	T leaf	2006	1 month
MEG_LAI15	MEGAN	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 1.5	T leaf	2006	1 month
ORC_CRUMOD	ORCHIDEE	CRU	Standard version	Standard version	MODIS LAI	T air	2006	1 month
ORC_LDF	ORCHIDEE	CRU	EFs = 1	LDF = 1 and 0	ORCHIDEE LAI	T air	2006	1 hour
MEG_LDF	MEGAN	CRU	EFs = 1	LDF = 1 and 0	ORCHIDEE LAI	T leaf	2006	1 hour

1 Table 6. Emission budget (Tg C yr⁻¹) averaged over the 2000–2009 period for the ORC_CRU (gray lines) and MEG_CRU simulations at the
 2 global scale, for northern and southern tropics, northern and southern temperate areas and northern boreal regions.

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Model	Area	Isoprene	Methanol	Acetone	Acetald	Formald	Acetic Acid	Formic Acid	MBO	Sesqiter.	Monoter	α -Pinene	β -Pinene	Limonen	Myrcene	Sabinene	3-Carene	T- β -Ocimene
ORCHIDEE	Global	464.6	37.8	24.6	8.6	1.9	1.1	0.7	1.3	24.3	91.3	40.9	12.2	10.7	7.2	8.19	6.5	9.3
MEGAN	Global	427.6	40.9	20.5	8.7	1.6	1.2	0.8	1.0	14.9	74.4	24.6	13.1	6.9	2.1	5.5	4.8	17.4
ORCHIDEE	Tro North	176.3	12.9	8.6	2.9	0.6	0.4	0.2	0.1	9.6	32.8	14.8	4.3	4.0	2.7	2.9	2.0	3.5
MEGAN	Tro North	1685	15.1	7.4	3.2	0.6	0.5	0.3	0.2	5.7	28.4	9.5	5.1	2.6	0.7	2.2	1.9	6.5
ORCHIDEE	Tro South	217.7	13.0	10.9	3.8	0.8	0.5	0.3	0.0	12.3	42.7	19.6	5.5	5.1	3.6	3.7	2.4	4.6
MEGAN	Tro South	209.6	15.1	9.1	4.2	0.7	0.5	0.3	0.1	7.5	32.5	10.5	5.5	2.9	0.8	2.6	1.6	8.5
ORCHIDEE	Tem North	51.6	9.1	3.6	1.3	0.3	0.2	0.1	1.2	1.6	10.9	4.3	1.6	1.1	0.6	1.2	1.6	0.8
MEGAN	Tem North	30.7	7.9	2.6	0.8	0.2	0.1	0.1	0.6	1.0	9.2	3.1	1.8	1.0	0.5	0.4	1.0	1.4
ORCHIDEE	Tem South	5.4	0.6	0.3	0.1	0.03	0.01	0.01	0.0	0.2	1.0	0.5	0.1	0.1	0.06	0.1	0.04	0.1
MEGAN	Tem South	9.1	1.0	0.4	0.1	0.03	0.02	0.01	0.01	0.2	1.2	0.5	0.2	0.14	0.03	0.06	0.06	0.25
ORCHIDEE	Boreal	4.4	1.5	0.6	0.2	0.05	0.03	0.02	0.03	0.2	2.0	0.9	0.3	0.2	0.1	0.2	0.3	0.15
MEGAN	Boreal	2.2	1.1	0.4	0.1	0.02	0.01	0.01	0.02	0.1	1.6	0.5	0.3	0.19	0.07	0.07	0.18	0.23

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1 Table 7. Mean emission budgets (Tg C yr⁻¹) for the 2000–2009 period estimated in MEG_CRULAI simulation.

MEGAN	Isoprene	Methanol	Acetone	Acetald	Formald	Acetic Acid	Formic Acid	MBO	Sesquiterp	Monoterp	α -Pinene	β -Pinene	Limonen	Myrcene	Sabinene	3-Carene	T- β -Ocimene
Global	422.7	41.1	20.2	8.5	1.5	1.2	0.8	1.1	14.5	74.0	24.5	13.0	6.9	2.1	5.7	4.8	17.0
Tro North	162.5	14.8	7.2	3.2	0.6	0.4	0.3	0.2	5.5	28.0	9.3	5.0	2.5	0.7	2.1	1.8	6.4
Tro South	210.1	15.0	8.9	4.1	0.7	0.5	0.3	0.1	7.3	32.0	10.5	5.4	2.8	0.7	2.5	1.5	8.2
Tem North	30.9	8.2	2.8	0.8	0.2	0.1	0.1	0.7	1.1	9.6	3.2	1.8	1.0	0.5	0.4	1.1	1.5
Tem South	9.2	1.1	0.4	0.1	0.03	0.02	0.01	0.01	0.2	1.3	0.5	0.2	0.15	0.03	0.07	0.06	0.26
Boreal	2.4	1.3	0.5	0.1	0.02	0.01	0.01	0.02	0.15	1.8	0.6	0.3	0.21	0.08	0.08	0.20	0.27

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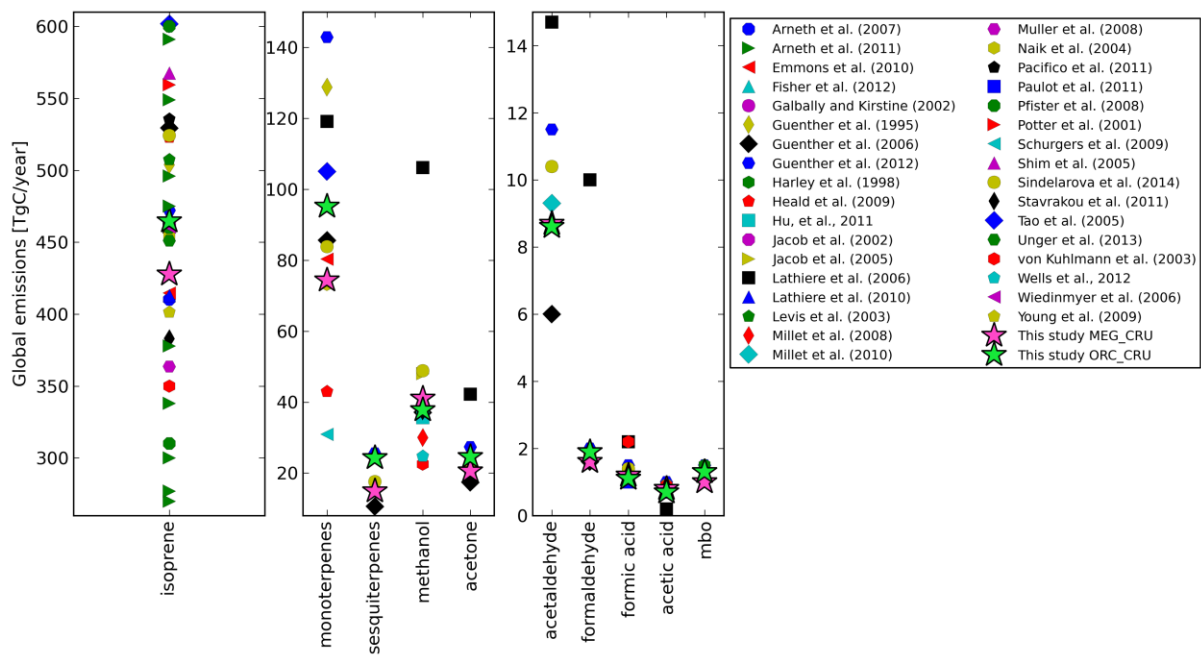
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1 Table 8. Annual emission budgets (Tg C yr⁻¹) for the year 2006 in ORC_CRU,
 2 MEG_CRULAI (taken as reference) and in the LAI sensitivity tests (ORC_LAI05,
 3 ORC_LAI15, MEG_LAI05 and MEG_LAI15).

Simulation	Isoprene	Methanol	Acetone	Acetald	Formald	Acetic Acid	Formic Acid	MBO	Sesquiter	Monoter	α -Pinene	β -Pinene	Other Monoter
ORC_CRU	464.9	38.0	24.6	8.6	1.9	1.1	0.7	1.4	24.2	95.4	41.0	12.2	42.3
ORC_LAI05	365.3	23.3	12.7	5.3	1.1	0.7	0.4	0.7	13.5	54.1	23.2	6.9	23.9
ORC_LAI15	501.1	50.4	36.5	11.4	2.5	1.5	1.0	2.1	34.2	133.8	57.4	17.1	59.2
MEG_CRULAI	422.5	41.4	20.3	8.6	1.6	1.2	0.8	1.1	14.5	74.2	24.6	13.1	36.5
MEG_LAI05	360.9	34.4	18.3	7.6	1.4	1.0	0.7	1.0	13.5	66.4	21.5	11.7	33.2
MEG_LAI15	450.2	45.1	20.7	8.9	1.6	1.2	0.8	1.2	14.6	76.8	25.8	13.5	37.5

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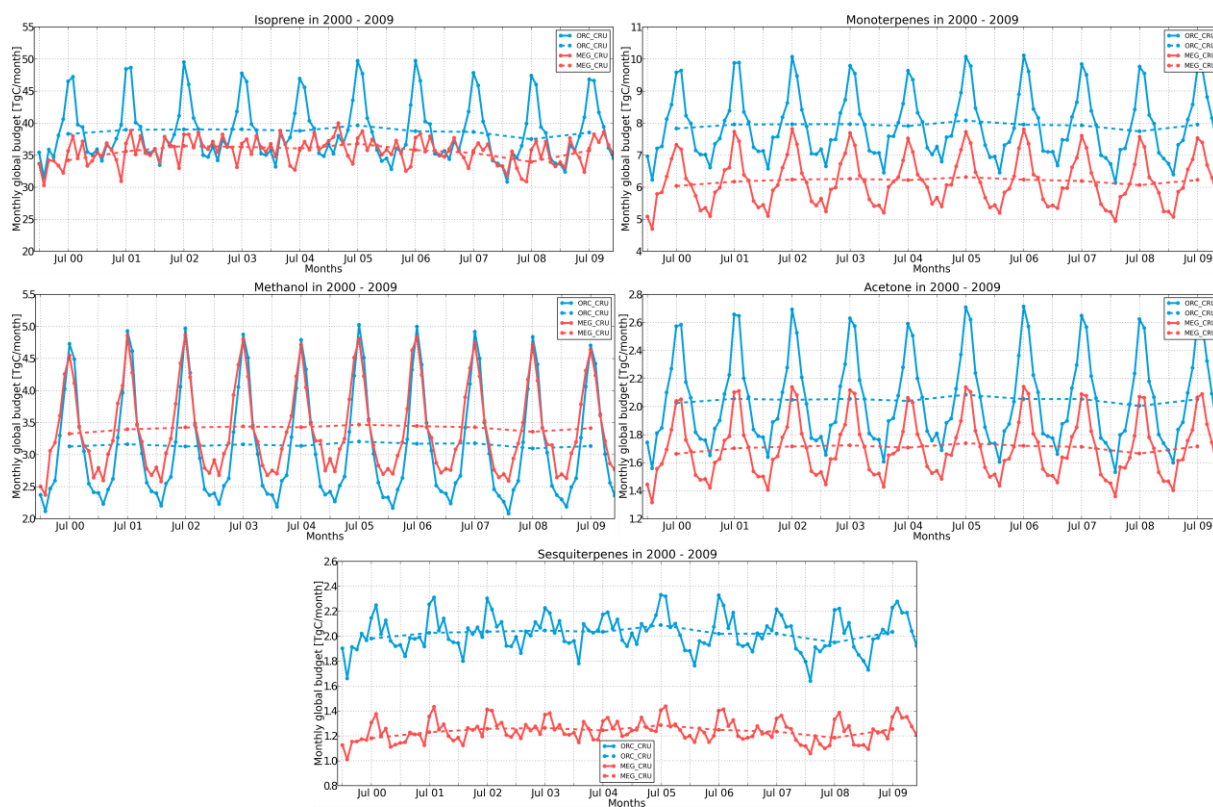
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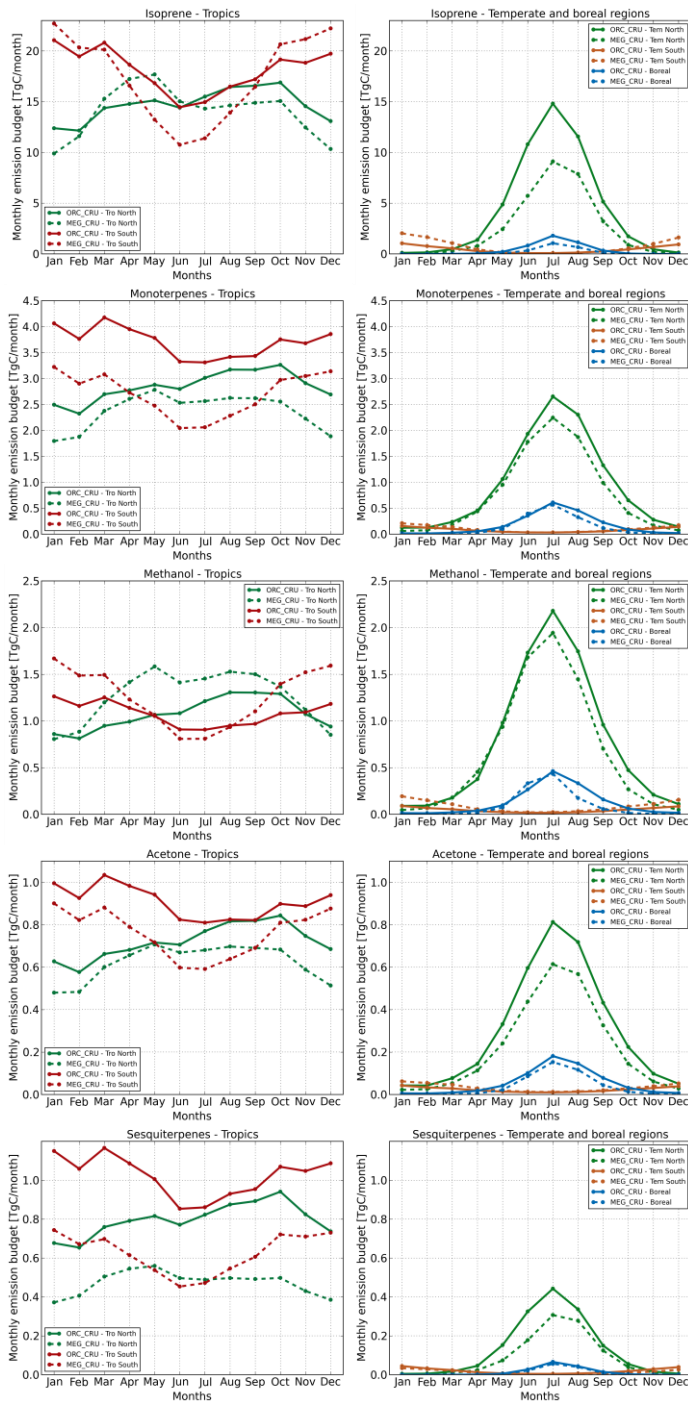
2 Figure 1. Global emission budgets (Tg C yr^{-1}) calculated by ORCHIDEE (ORC_CRU
 3 simulation, green stars) and MEGAN (MEG_CRU simulation, pink stars), compared with
 4 published estimates for the main BVOCs presented in this work. Note that the vertical axes
 5 have different scales in the three plots.

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 2 Figure 2. Monthly global (solid lines) and yearly averaged (dashed lines) emission budgets in
 3 Tg C month⁻¹ for ORC_CRU and MEG_CRU simulations for isoprene, monoterpenes,
 4 methanol, acetone and sesquiterpenes.

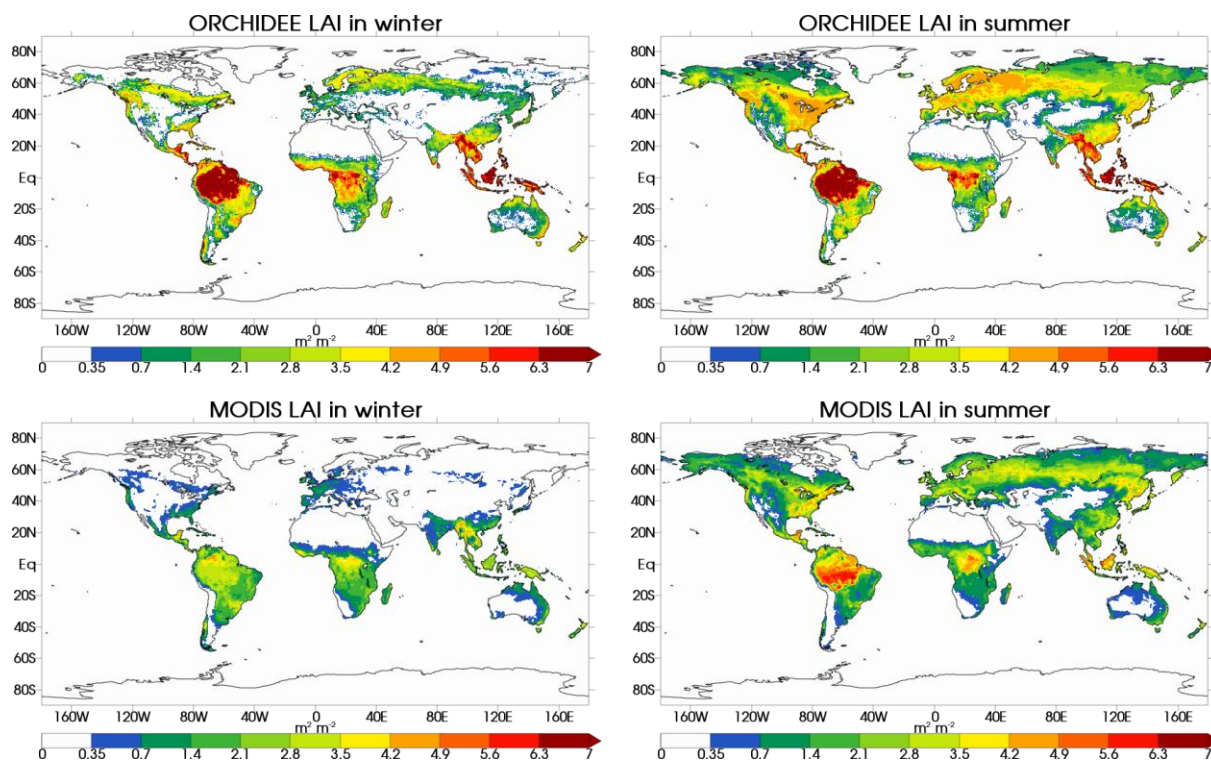
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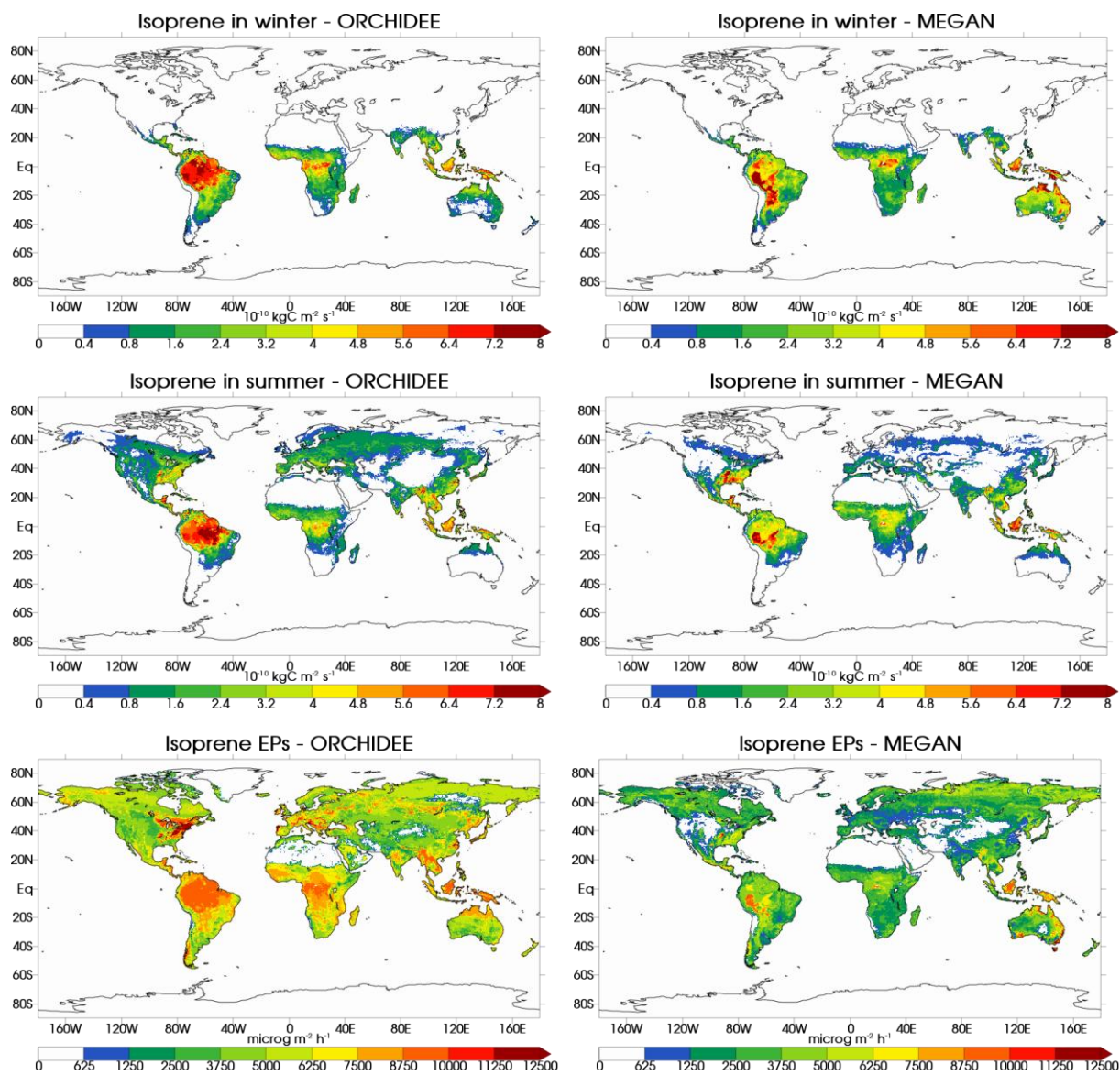
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2 Figure 3. Zonal mean for northern and southern tropics (left column), northern and southern
 3 temperate and northern boreal latitudes (right column) of the monthly emission budget (Tg C
 4 month⁻¹) averaged over the simulation period (2000–2009) in ORC_CRU and MEG_CRU
 5 runs for isoprene, monoterpenes, methanol, acetone and sesquiterpenes, respectively.

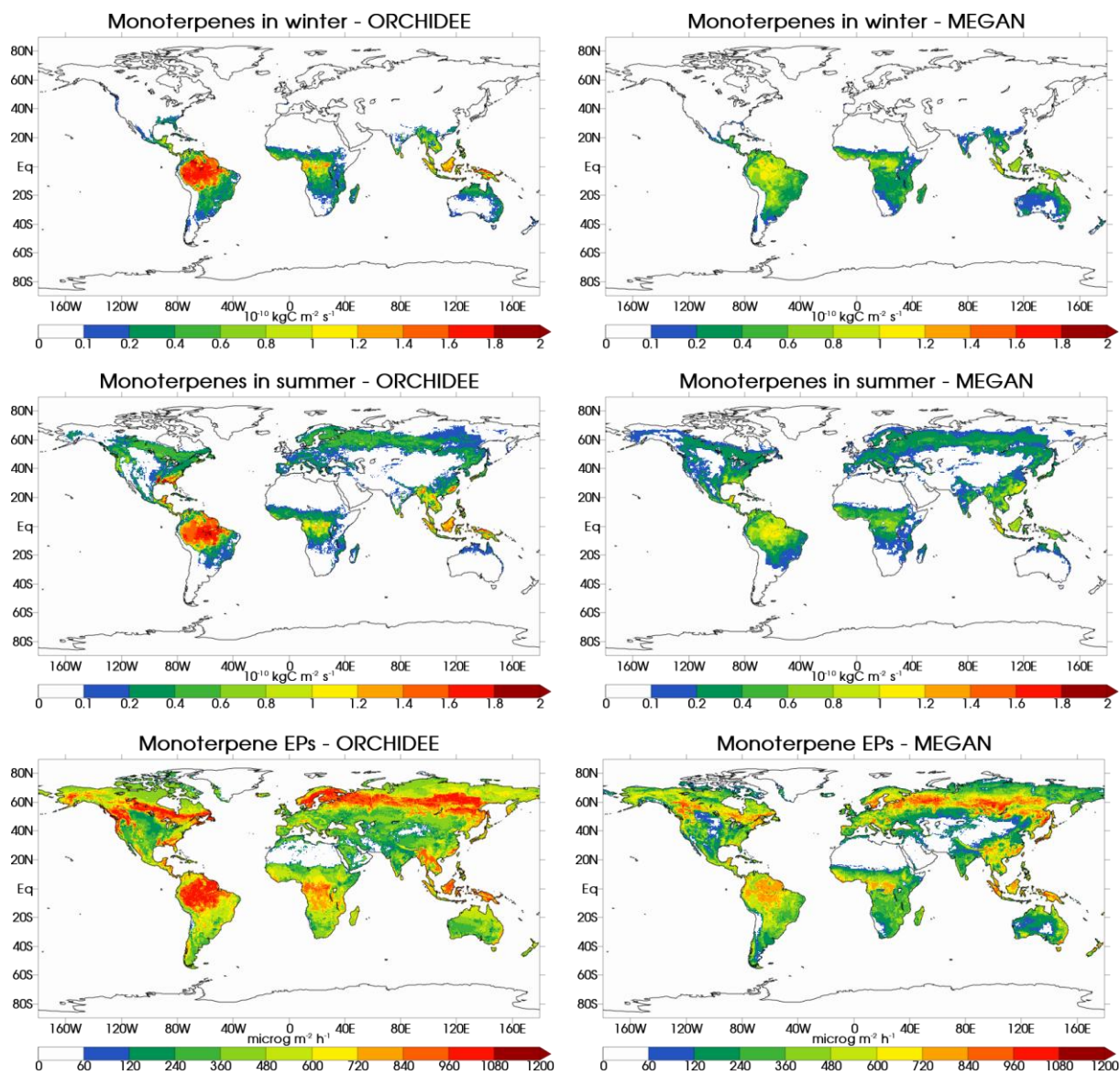
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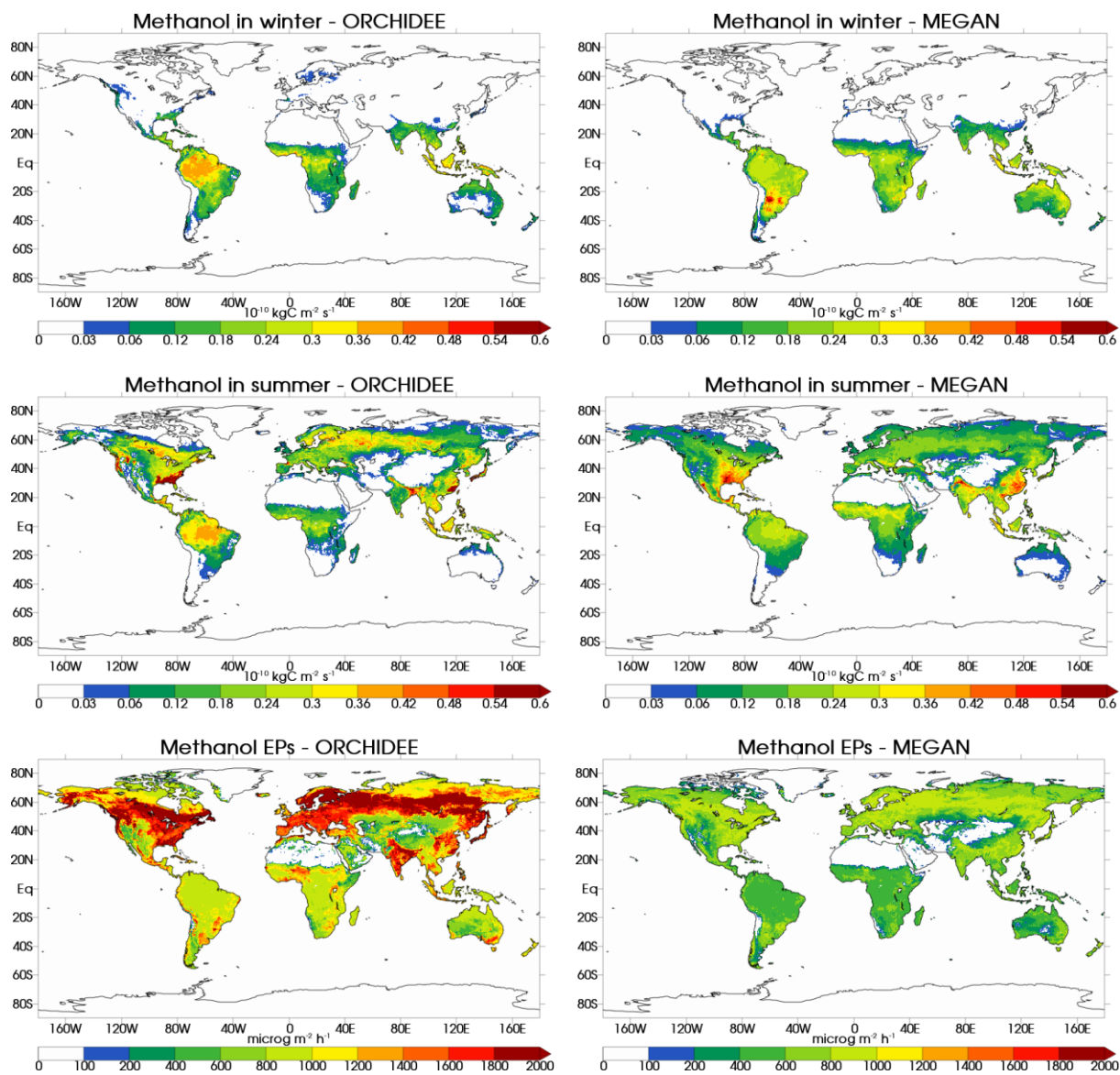
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 2 Figure 4. Leaf area index (LAI) considered for BVOC emission estimates in ORCHIDEE
 3 (LAI calculated on line) and in MEGAN (MODIS retrieval) in summer (June, July, August)
 4 and winter (December, January, February), averaged over the 2000–2009 period ($\text{m}^2 \text{m}^{-2}$).
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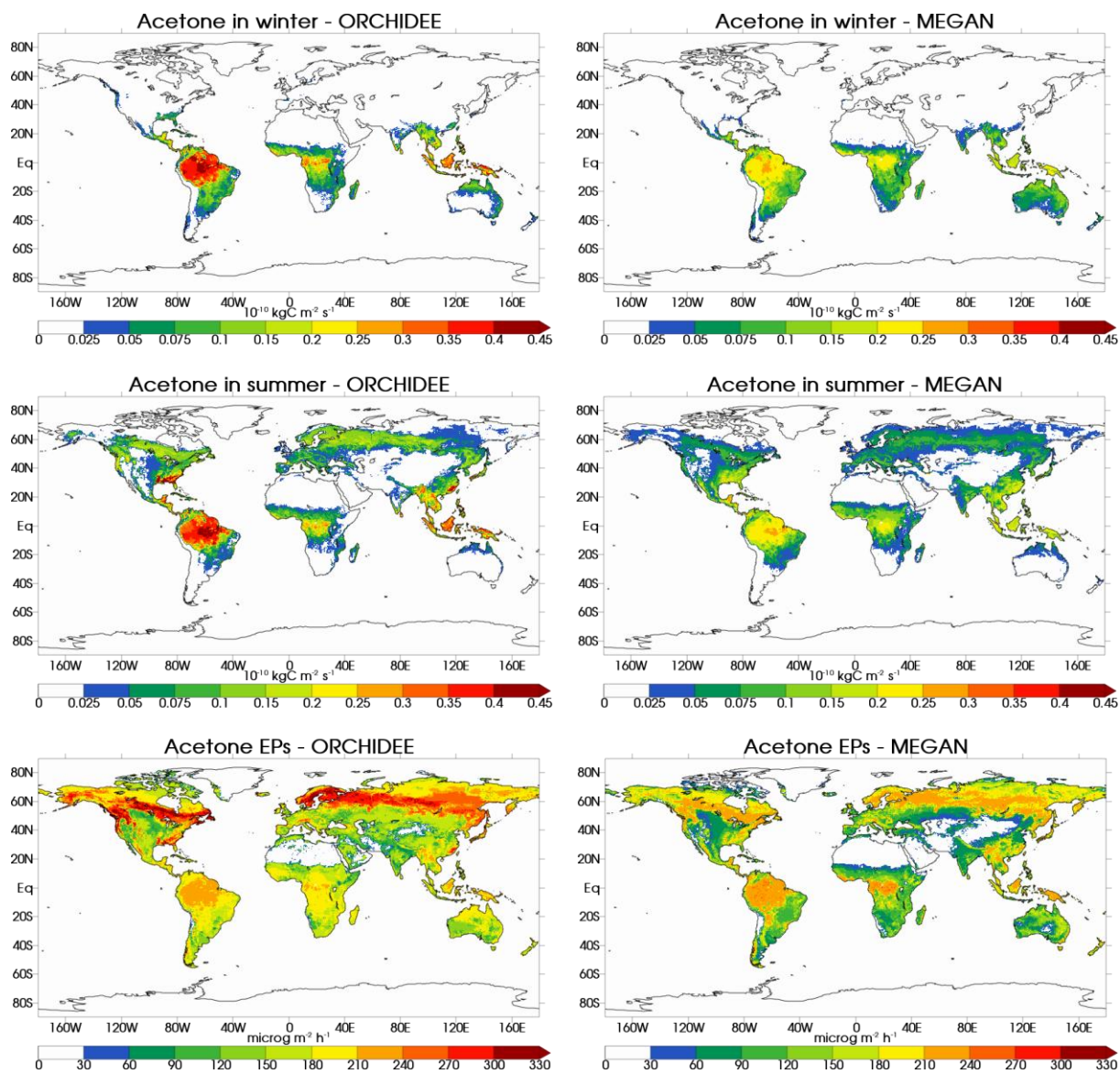
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 2 Figure 5. Emissions in winter (first row) and summer (second row) in $10^{-10} \text{ kg C m}^{-2} \text{ s}^{-1}$ and
 3 emission potentials (EPs) (third row) in $\mu\text{g m}^{-2} \text{ h}^{-1}$ for ORCHIDEE (ORC_CRU, left column)
 4 and MEGAN (MEG_CRU, right column) for isoprene.
 5



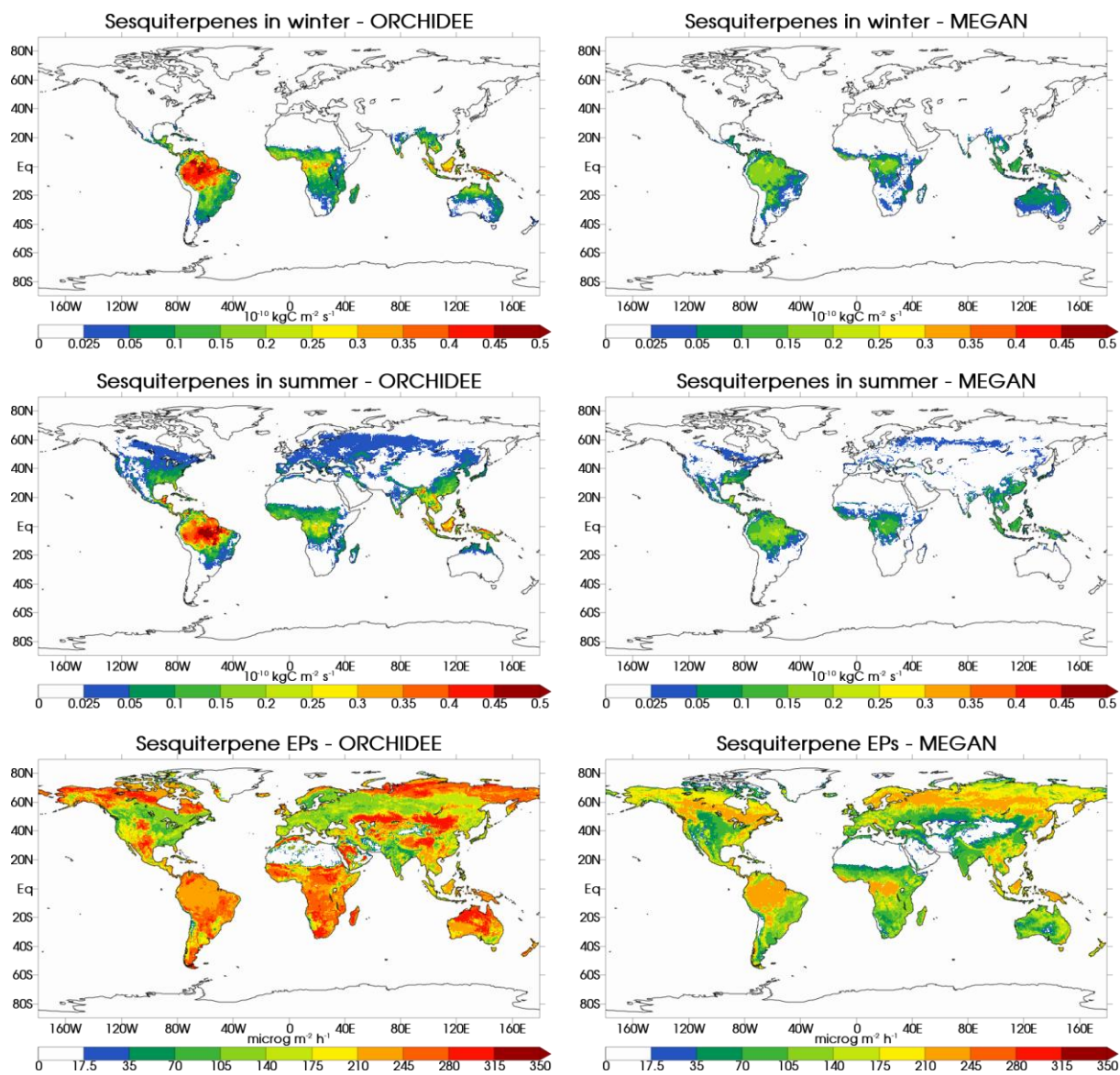
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 2 Figure 6. The same as Fig. 5, but for monoterpenes.
 3



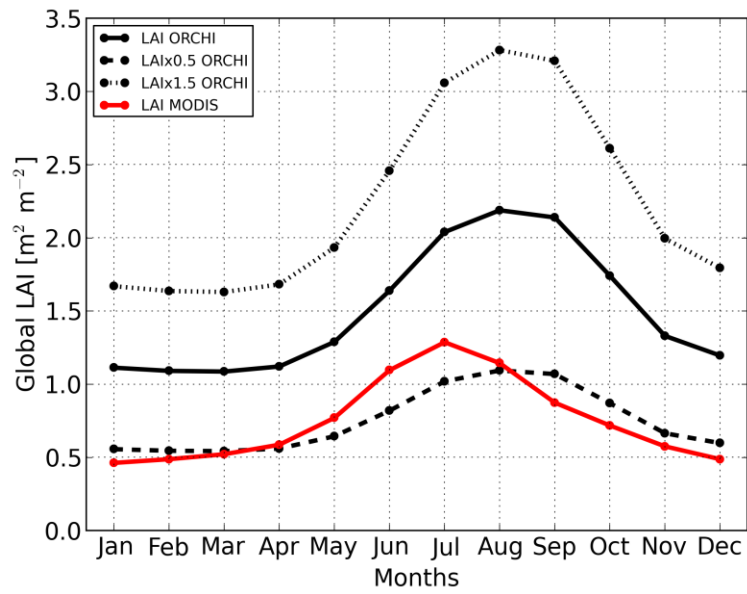
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 2 Figure 7. The same as Fig. 5, but for methanol.
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 2 Figure 8. The same as Fig. 5, but for acetone.
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 2 Figure 9. The same as Fig. 5, but for sesquiterpenes.
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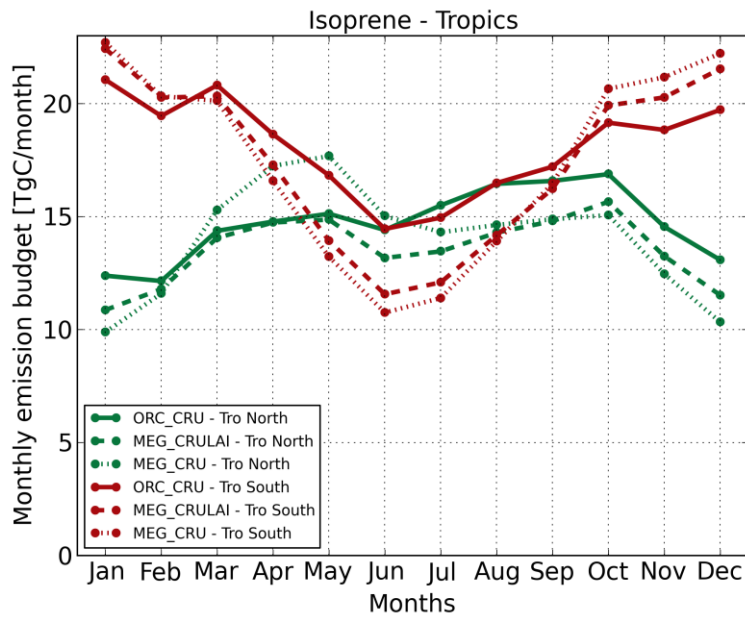


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2 Figure 10. Global monthly mean LAI ($\text{m}^2 \text{m}^{-2}$) calculated by ORCHIDEE (solid black line)
 3 and retrieved from MODIS measurements (red line). The thick and thin dashed lines represent
 4 the LAI from ORCHIDEE multiplied by a factor 0.5 and 1.5, respectively.

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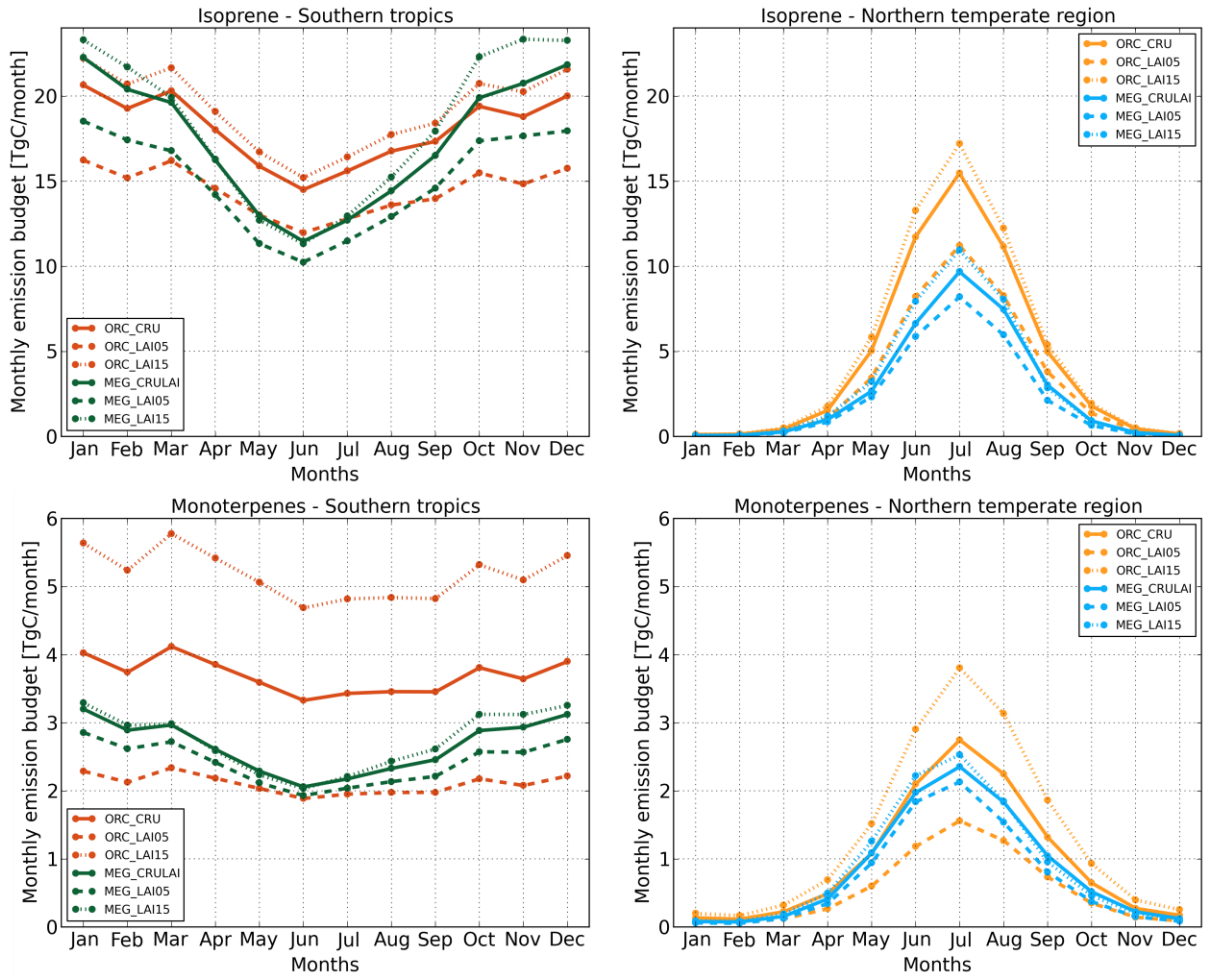


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3 Figure 11. Zonal mean of monthly emission budgets (Tg C month^{-1}), averaged over the
4 simulation period (2000–2009) for the northern and southern tropics, in ORC_CRU (solid
5 line), MEG_CRULAI (thick dashed line) and MEG_CRU (thin dashed line) simulations for
6 isoprene.

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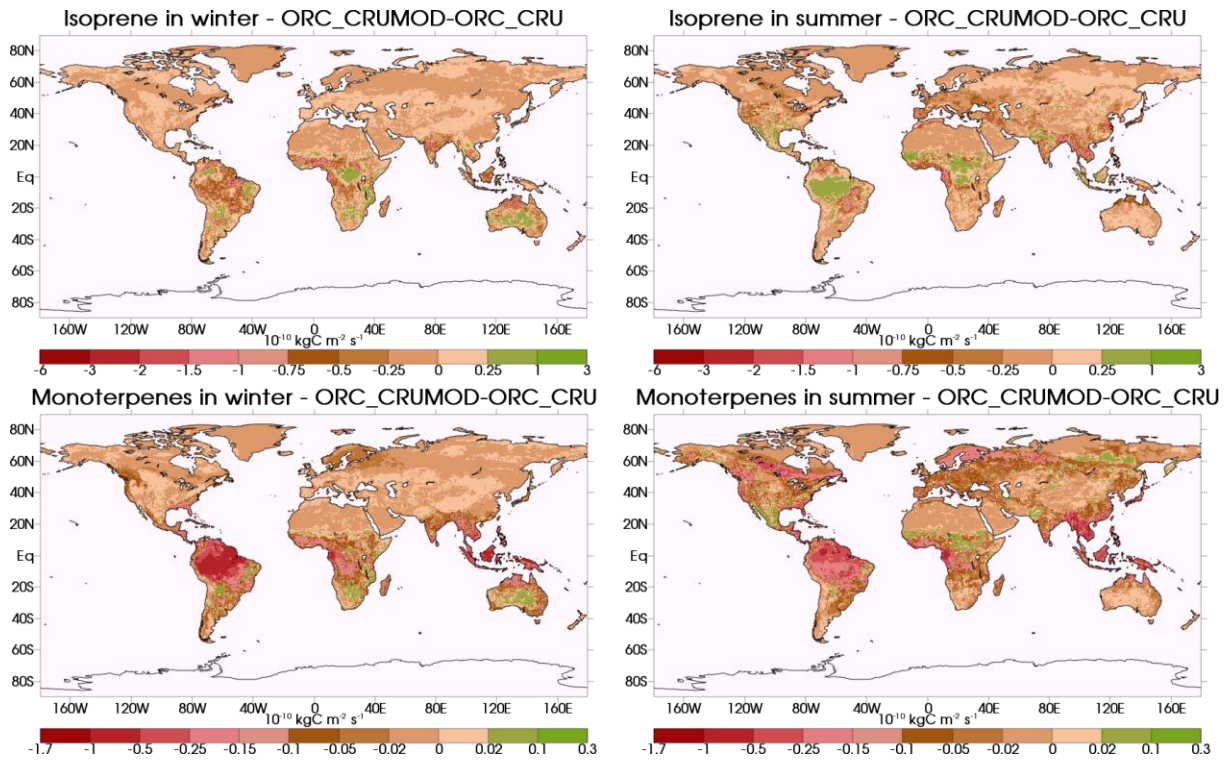


1

2 Figure 12. Zonal average of changed emissions in the different LAI sensitivity tests:
 3 ORC_CRU and MEG_CRULAI using ORCHIDEE LAI (solid line), ORC_LAI05 and
 4 MEG_LAI05 using ORCHIDEE LAI·0.5 (thick dashed line) and ORC_LAI15 and
 5 MEG_LAI15 using ORCHIDEE LAI·1.5 (thin dashed line) in the year 2006, for the southern
 6 tropical (left column) and northern temperate regions (right column) for isoprene and
 7 monoterpenes. Emissions are given in Tg C month^{-1} .

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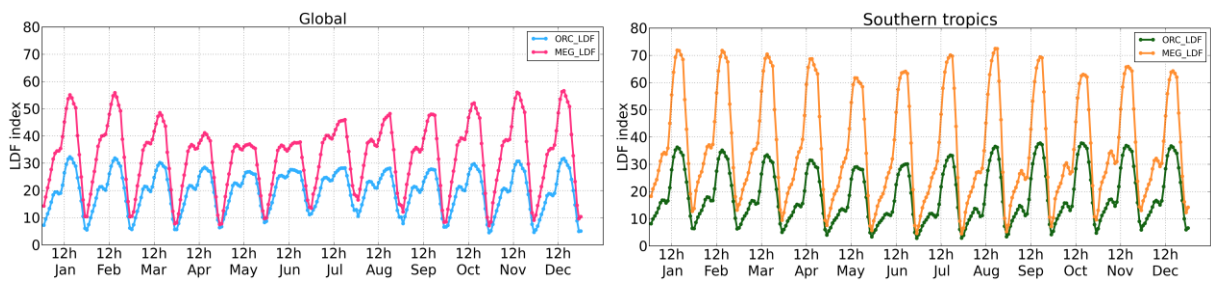
3

4 Figure 13. Differences between the ORC_CRUMOD and ORC_CRU simulation for isoprene
5 and monoterpenes emissions in summer and winter for 2006.

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3 Figure 14. Global (left plot) and southern tropical (right plot) average of the LDF index for
4 ORCHIDEE and MEGAN. The LDF index is provided as the hourly daily profile averaged
5 over each month.

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