



<sup>7</sup>Institut Pierre Simon Laplace des sciences de l'environnement, Paris, France

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Correspondence to: P. Messina (palmira.messina@lsce.ipsl.fr)

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**Global biogenic  
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and -11 % (+3 %) for MEGAN regarding monoterpenes. We find that MEGAN is more sensitive to variation of LDF parameter than ORCHIDEE. Our results highlight the importance and the need to further explore the BVOC emission estimate variability and the interest of using models to investigate the estimate uncertainties.

## 1 Introduction

The terrestrial biosphere emits large amounts of Volatile Organic Compounds (VOCs), in particular terpenoids, such as isoprene, monoterpenes, and sesquiterpenes, and oxygenated hydrocarbons, such as methanol, acetone, formaldehyde, acetaldehyde, acetic acid, or formic acid. At the global scale, the ecosystem contribution to VOC emissions is significantly higher than the anthropogenic one, and accounts for 75–90 % to the total emission (Guenther et al., 1995; Lamarque et al., 2010). Biogenic VOCs (BVOCs) play a central role in atmospheric chemistry, influencing the oxidative capacity of the atmosphere (Arnth et al., 2011; Taraborrelli et al., 2012), leading to the formation and growth of Secondary Organic Aerosols (SOA) (Kanakidou et al., 2005; Goldstein and Galbally, 2007; Van Donkelaar et al., 2007; Acosta Navarro et al., 2014), leading to the production of tropospheric ozone in the presence of nitrogen oxides (Von Kuhlmann et al., 2003; Mao et al., 2013), and influencing the tropospheric carbon monoxide budget (Pfister et al., 2008).

In spite of the numerous measurements and the progressive understanding of the processes underlying their production, BVOC emission estimates are still highly uncertain and vary significantly among the various estimates (Steiner and Goldstein, 2007; Arnth et al., 2008; Simpson et al., 2012; Sindelarova et al., 2014).

Over the last 20–25 years, two main methods have been developed to derive BVOC inventories: a top-down approach, based on the inversion of satellite measurements and giving the possibility to derive indirectly BVOC emissions (Palmer et al., 2006; Barkley et al., 2013) and a bottom up approach. The latter one is the most largely used method for local-, regional- or global-scale studies and can be divided into two main

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categories: (i) an empirical approach where the response of leaf emissions to environmental changes is modelled using algorithms combined in a multiplicative way (Guenther et al., 2006, 2012; Lathièrè et al., 2006, 2010; Steinbrecher et al., 2009; Oderbolz et al., 2013) and (ii) a processed-based approach, where emissions are linked to the photosynthetic electron transport rate in chloroplasts (Niinemets et al., 2003a, b; Sitch et al., 2003; Keenan et al., 2009; Schurgers et al., 2009; Pacifico et al., 2011; Unger et al., 2013). The models discussed in this study belong to the first category of bottom up models.

BVOC emission modelling at the global scale is a complex issue especially because of the number of variables and processes influencing the emission of these compounds, generally characterized by a strong temporal and geographical variation. A critical point is the lack of information available at the global scale related to the various biomes, that would allow a more accurate representation of the geographical distribution and of the seasonal variation of BVOC emissions (Peñuelas and Staudt, 2010). The basal emission factor (EF) for instance, defined as the emission at the leaf level under standardized environmental conditions of temperature and solar radiation, is a key emission driver and shows a large variability from one plant species to another. The large number of measurements now available for different plants and in various sites around the world makes it possible to assign statistically significant EF values. Furthermore, there is an increasing number of field campaigns that now investigate, in addition to isoprene and bulk monoterpenes, many other important compounds for atmospheric chemistry, especially regarding the SOA formation, such as speciated monoterpenes and sesquiterpenes. More data and information are therefore available, giving the possibility to have EF estimates for a wider range of BVOCs. To calculate BVOC emissions, a single EF is usually assigned for each plant functional type (PFT), one PFT representing a group of plants having the same phylogenetic, phenological and physical characteristics (Prentice et al., 1992). Choosing one single value for each PFT is especially difficult, as one PFT can actually correspond to several plant species, and knowing that EFs show, in general, a wide range of values among

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the different plants (Kesselmeier and Staudt, 1999; Niinemets et al., 2011). Moreover several measurements even show that the emission factors are significantly sensitive to many processes and parameters difficult to isolate and linked to plants stress, such as drought periods, ozone exposure, insects, herbivores and pathogen attack (for a review see Laothawornkitkul et al., 2009 and Niinemets et al., 2010), making it more delicate to set EFs even to a single plant.

In the early works focusing on BVOCs, isoprene was the only compound considered to be both light and temperature dependent, while the other compounds were considered to be only temperature dependent. More recent papers show a growing evidence of the dependency of monoterpenes (Dindorf et al., 2006; Holzke et al., 2006; Šimpraga et al., 2013), sesquiterpenes (Hansen and Seufert., 2003) and oxygenated BVOCs (Jacob et al., 2002, 2005; Harley et al., 2007; Millet et al., 2008, 2010; Hu et al., 2011; Wells et al., 2014) on radiation. As proposed in Guenther et al. (2012), a general approach is now to consider, for each emitted compound, an emission fraction which depends on both temperature and solar light, as it is done for isoprene, and the remaining fraction depending only on temperature. Nevertheless, there is no general agreement on the exact value of this temperature and light dependent fraction.

Another crucial component in the estimation of BVOC emissions is the leaf area index (LAI) that can be either simulated using a vegetation model or prescribed using values retrieved from satellite data or field measurements. Significant differences, in terms of temporal and spatial distribution, can be noted between the LAI estimated by measurements and the LAI calculated by models, with discrepancies up to 100 % at the global scale and to more than 150 % for specific biomes types (Garrigues et al., 2008; Pinty et al., 2011; Fang et al., 2012a, b). Consequently the high uncertainty related to LAI can affect the regional and seasonal distribution of BVOC emissions.

According to our knowledge, most papers investigating the BVOC emission sensitivity focus on the response of emissions to different experimental set-ups, changing, for instance, climate forcing and land use. For example, Oderbolz et al. (2013) pointed out the importance of the differences among the land-cover inventories and the uncer-

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tainties on the classification of land cover pixel. Arneth et al. (2011) compared three vegetation models, changing the experimental set-up, such as the vegetation distribution and the climate forcings. Depending on the experiment considered, the total annual isoprene emissions were found to increase or decrease by more than 30 %.

5 Ashworth et al. (2010) investigated the impact of varying the climate forcing temporal resolution on isoprene emission in the MEGAN model, finding a variation of isoprene emissions of up to 7 % at the global scale and up to 55 % in some locations. Nevertheless very few studies investigated the impact on emissions of the uncertainty of key parameters/variables, such as LAI. One example is the work by Sindelarova et al. (2014)  
10 which performed several simulations with the MEGAN model to assess the sensitivity of isoprene emissions to many parameters and processes such as LAI, emissions factors (EFs), CO<sub>2</sub> concentration, soil moisture, and the radiation scheme. The sensitivity simulations performed showed a variation of up to 50 % of isoprene emissions at the global scale.

15 In the present work, our objectives are (i) to present the updated version of the emission module embedded in the dynamic global vegetation model ORCHIDEE, (ii) to provide present-day estimates of global BVOC emissions for several relevant compounds (isoprene, monoterpenes, sesquiterpenes, methanol, acetone, formaldehyde, acetaldehyde, acetic acid, formic acid and the main speciated monoterpenes) using  
20 the new emission scheme, (iii) to compare the ORCHIDEE results to the widely used emission model MEGAN, (iv) to explore, at the global and regional scales, the BVOC emission sensitivity to EFs, LAI, LDF in ORCHIDEE and MEGAN and to understand the reasons behind these discrepancies. Because of the lack of observations at the global scale, the best possible way to evaluate emissions models is to perform model inter-comparison and model sensitivity experiments, allowing to deeply investigate critical  
25 pattern of simulated variables and to identify the origin of uncertainty. Through model inter-comparison and sensitivity tests, we assess the limitations and uncertainties of BVOC emission estimates related to some key parameters/variables, investigating the origins of the limitation.

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In Sect. 2, the ORCHIDEE model and the updates from the previous version (Lathière et al., 2006), the MEGAN model and the technical details of the simulations are described. The comparison with other published estimates, the inter-comparison between the two models and the sensitivity tests carried out are extensively described in Sect. 3. The conclusion and discussion of the study are provided in Sect. 4.

## 2 Model developments and set-up

### 2.1 ORCHIDEE model: general description

ORCHIDEE (Organizing Carbon and Hydrology in Dynamic EcosystEm) is a dynamic global vegetation model that consists of two main parts: the carbon module STOMATE (Saclay–Toulouse–Orsay Model for the Analysis of Terrestrial Ecosystems) and the surface vegetation atmosphere transfer scheme SECHIBA (Schématisation des échanges hydriques à l'interface biosphère-atmosphère), (Krinner et al., 2005; Magnan et al., 2011).

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

In ORCHIDEE, ecosystems are represented by 13 Plant Functional Types (PFTs, listed in Table 1). Each PFT is representative of a specific set of plant species that are grouped according to plant physiognomy (tree or grass), leaf shape (needleleaf



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or broadleaf), phenology (evergreen, summergreen or raingreen) and photosynthesis type for crops and grasses (C3 or C4). The main biophysical and biogeochemical processes for each PFT are described in Krinner et al. (2005) and in Maignan et al. (2011). Activating the LPJ dynamical vegetation component of the model (Krinner et al., 2005), ORCHIDEE has also the feature to determine the surface and distribution of natural vegetation types according to climate and CO<sub>2</sub> conditions considered, while location and surfaces of agricultural PFTs are always prescribed. For our study, the global vegetation distribution is prescribed for all runs using appropriate forcings.

### 2.2 BVOCs in ORCHIDEE: module improvements

In order to consider recent findings regarding emission schemes and field measurements, the BVOC module is extensively updated. The new BVOC emission scheme is a development of the module implemented in ORCHIDEE by Lathièrre et al. (2006) and based on the model presented by Guenther et al. (2012). It now provides a multi-layer canopy model, where radiation is calculated following the scheme proposed by Spitters (1986) and Spitters et al. (1986) and the one already used in ORCHIDEE for the calculation of photosynthesis. Canopy is divided in up to 17 LAI layers (the layer number depends on the LAI value) and emissions are calculated for each layer considering the sunlit and shaded leaf fractions and the light extinction and light diffusion through the canopy and are then vertically summed, providing a single value for each PFT and grid point.

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

$$F_{icl} = LAI_{il} \cdot SLW_i \cdot EF_{icl} \cdot C_{TLi} \cdot L_c \quad (1)$$

where  $LAI_l$  is the leaf area index expressed in  $m^2 m^{-2}$  at a particular LAI layer,  $SLW$  is the specific leaf weight in  $gdm m^{-2}$ ,  $EF$  is the basal emissions at leaf level in the standard conditions of temperature ( $T = 303.15 K$ ) and photosynthetically active radiation

( $\text{PAR} = 1000 \mu\text{mol m}^{-2} \text{s}^{-1}$ ) expressed in  $\mu\text{gC gdm}^{-1} \text{h}^{-1}$ . Note that in the ORCHIDEE model, EFs depend on both the PFT and the emitted compounds considered.

$C_{\text{TL}}$  is the emission activity factor, depending on emitted compounds, that takes into account the deviation from the standard conditions related to temperature and PAR and it is extensively described in the second part of the present paragraph.  $L_c$  is the activity factor, simulating the impact of leaf age on emissions and considered for isoprene and methanol. The total emission per grid-cell is obtained by summing the emission contribution of each PFT, weighted by PFT fractional coverage. Further details on the emission module original version are given in Lathièrre et al. (2006).

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

Eight speciated monoterpenes ( $\alpha$ -pinene,  $\beta$ -pinene, limonene, myrcene, sabinene, camphene 3-carene, t- $\beta$ -ocimene) and bulk sesquiterpenes are now included in the updated ORCHIDEE emission module. We chose these compounds for their importance in the atmospheric chemistry, in particular regarding the secondary organic aerosol formation. EFs represent one of the greatest sources of uncertainty in the quantification of BVOC emissions (Niinemets et al., 2011). Several measurement campaigns were carried out over the last decade, giving important insights and information to re-examine thoroughly the emission factors used in the emission module and correct them accordingly. Based on an extensive review of publications, we selected the field measurements that report EFs in the standard conditions of PAR ( $1000 \mu\text{mol m}^{-2} \text{s}^{-1}$ ) and temperature ( $30^\circ\text{C}$ ). The collected EFs were gathered per PFTs and then averaged to obtain a single value per PFT, rejecting the outliers.

In Table 3 we report the new and old EFs used in the emission module and in Table 4 we present EF value for each speciated monoterpene as a percentage of the bulk monoterpene EF value. As shown in Table 3, the revision leads to the modifica-

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tion of almost all EFs, in order to take into account recent findings demonstrated by field measurements. In some cases the EF differences in comparison with the previous version, are very significant. Regarding isoprene, boreal needleleaf deciduous PFT is now recognized as a less important emitter ( $EF = 8 \mu\text{g C gdm}^{-1} \text{h}^{-1}$  in the old version and  $EF = 0.5 \mu\text{g C gdm}^{-1} \text{h}^{-1}$  in the new one) (Levis et al., 2003; Guenther et al., 2006, 2012; Karl et al., 2009; Steinbrecher et al., 2009, 2013). For monoterpenes, a significantly higher EF (from 0.8 to  $2.2 \mu\text{g C gdm}^{-1} \text{h}^{-1}$ ) is now assigned to tropical broadleaf evergreen and deciduous PFTs and for 2-Methyl-3-Buten-2-Ol (MBO), the EF for the temperate needleleaf evergreen PFT is reduced from 20 to  $1.4 \mu\text{g C gdm}^{-1} \text{h}^{-1}$  (Tarvainen et al., 2005; Hakola et al., 2006; Chang et al., 2009; Kim et al., 2010).

Our review analysis underlines a large variability of EFs, even between plants characterized by the same physiognomy, leaf shapes and photosynthesis type. Those plants are usually represented by one single PFT in global vegetation models. It is therefore difficult to choose and assign one fixed EF value for each PFT in global models, as also pointed out by Kesselmeier and Staudt (1999) and Arneth et al. (2011). Moreover, the procedure used to determine emission factors from field measurements adds an additional source of uncertainty. Indeed EFs are derived adjusting the measured flux at leaf level to standard condition of light photosynthetically active radiation (PAR) and temperature, using algorithms like Guenther et al. (1995). However there is no universal agreement on the parameterization of these algorithms (Tarvainen et al., 2005; Duhl et al., 2008; Kim et al., 2010; Bracho-Nunex et al., 2011; Fares et al., 2011).

The emission module has also been modified to include a light dependency for almost all compounds emitted. In the previous module version, isoprene was the only compound depending on both light and temperature, while the others were only depending on temperature. Most recent field campaigns highlight, for a large number of plants, the dependency of monoterpenes, sesquiterpenes and oxygenated BVOC emissions on radiation as well (Jacob et al., 2002, 2005; Hansen and Seufert, 2003; Dindorf et al., 2006; Holzke et al., 2006; Harley et al., 2007; Millet et al., 2008, 2010; Hu et al., 2011; Šimpraga et al., 2013; Wells et al., 2014). In the new emissions mod-

ule, we therefore take into account these findings adopting the approach described in Guenther et al. (2012). BVOCs are now modelled considering both light dependent (directly release through stomata) and light independent (stored in the leaf pool) emission processes and the response to temperature and light ( $C_{TL}$ ) is calculated, at each LAI layer ( $l$ ), as:

$$C_{TL}(l) = (1 - LDF)C_{TLI} + LDF \cdot C_{TLD} \cdot C_L(l) \quad (2)$$

Where LDF is the light-dependent fraction of the emission, specified for each compound emitted (Table 2),  $C_{TLI}$  is the temperature response for the emission part that is not light dependent,  $C_{TLD}$  and  $C_L$  are the temperature and light response for the light-dependent fraction, respectively.

$C_{TLI}$ ,  $C_L$  and  $C_{TLD}$  are the same functions as in the previous version of the emissions module (Guenther et al., 1995; Lathière et al., 2006) and are defined as:

$$C_{TLI} = \exp(\beta(T - T_0)) \quad (3)$$

$$C_{TLD} = \frac{\exp\left(\frac{c_T(T-T_0)}{RT_0T}\right)}{1 + \exp\left(\frac{c_M(T-T_M)}{RT_0T}\right)} \quad (4)$$

$$C_L(l) = \frac{\alpha C_L PAR(l)}{\sqrt{1 + \alpha^2 PAR(l)^2}} \quad (5)$$

$\beta$  is the empirical coefficient of the exponential temperature response (Table 2) and PAR is the photosynthetically active radiation at each LAI layer. For all the other details of the Eqs. (3)–(5) we refer to Guenther et al. (1995).

### 2.3 MEGAN description

The Model of Emissions of Gases and Aerosols from Nature (MEGAN) is a modelling system for the estimation of emission fluxes of biogenic organic compounds from the

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terrestrial vegetation. The basis of the model is a simple mechanistic approach established in Guenther et al. (1991, 1993, 1995), linking emissions with the main environmental driving factors such as solar radiation and leaf temperature. Further development of the algorithm led to the inclusion of leaf ageing, soil moisture impact on the emissions, effects of the loss and production of compounds within a forest canopy (Guenther et al., 2006). The current version of the model, MEGANv2.1, also includes a full canopy module. The model calculates light and temperature conditions inside a canopy by evaluating the energy balance on five canopy levels. Additionally, emissions of each compound are considered to have light dependent and light independent part defined by the light dependent fraction (LDF). For the detailed description of emission equations and parameterization we refer to Sect. 2 in Sindelarova et al. (2014) and Guenther et al. (2012).

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

MEGAN estimates emissions of 19 chemical compound classes, which are then re-distributed into 147 final output model species, such as isoprene, monoterpene and sesquiterpene species, methanol, carbon monoxide, alkanes, alkenes, aldehydes, ketones, acids and other oxygenated VOCs. Although the input parameters, such as vegetation description and emission potentials, can be defined by the user, MEGAN comes with a default setting of PFTs and emission factors assigned to them. The vegetation distribution is described with fractional coverage of 16 PFT classes consistent with those of CLM4 model (Lawrence and Chase, 2007). The emission potential of each modelled species is calculated based on the PFT coverage and emission factor of

each PFT category. For several VOC compounds, emission potentials can be defined in the form of input maps. Emission potential maps with global coverage and high spatial resolution for isoprene, main monoterpene species and MBO are provided together with the MEGAN code.

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

## 2.4 Model set-up and sensitivity tests

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

We carried out a total of 4 sets of runs:

1. 2 simulations for the 2000–2009 period performed by both models using each model's standard configuration, but with the same climatology (ORC\_CRU and MEG\_CRU).
2. 1 simulation for the 2000–2009 period with MEGAN using the LAI estimated by ORCHIDEE (MEG\_CRULAI)

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3. 4 simulations for the year 2006 by both models, using the ORCHIDEE LAI scaled by a factor 0.5 and 1.5, respectively (ORC\_LAI05, ORC\_LAI15, MEG\_LAI05 and MEG\_LAI15).

4. 2 simulations for the year 2006 performed by both models, where we output two test species, the first one totally dependent on light ( $LDF = 1$ ) and the second one totally independent on light ( $LDF = 0$ ) (ORC\_LDF and MEG\_LDF). The output time frequency is one hour for this run.

The run sets 3 and 4 are carried out for the year 2006, which is estimated as an averaged year regarding the BVOC emissions calculated by MEGAN and ORCHIDEE in the 10-year of simulation.

All simulations are performed at the global scale with a spatial resolution of  $0.5^\circ \times 0.5^\circ$ . We use the CRU-NCEP v5.2 meteorological forcing database providing temperature, pressure, humidity, wind speed and shortwave solar radiation. This forcing is based on the 6 hourly  $2.5^\circ$  NCEP/NCAR re-analysis (Kalnay et al., 1996) combined with the CRU TS 2.1 monthly anomalies (Mitchell and Jones, 2005).

For the ORCHIDEE model a spin-up of 20 years is first performed in order to balance the leaf stock. The spin-up is based on a 10-year loop using meteorological forcing for the year 1989, followed by a 10-year simulation from 1990 to 1999. In ORCHIDEE, the global vegetation distribution for the 13 PFTs is prescribed using the Land-Use History (LUHa.rc2) related to the year 2000 (Hurt et al., 2006). In MEGAN the distribution for the 16 PFTs is consistent with the Community Land Model v4 (Lawrence and Chase, 2007) and related to the year 2000. Table 1 gives the global surfaces covered by the different PFTs in ORCHIDEE and MEGAN.

ORCHIDEE calculates the LAI at each model time step, while MEGAN does not compute the LAI and can be driven by different inputs. We present here the results of MEGAN forced either by the LAI retrieved by MODIS (Yuan et al., 2011) or by the LAI provided by the ORCHIDEE simulation (see Table 5 for all simulation details). LAI is not processed in the same way in the two models for the emission estimate. In ORCHIDEE,



emissions are calculated firstly for each vegetation type, considering the LAI simulated online by ORCHIDEE for each PFT and then summed providing an emission value per grid cell, as described in Sect. 2.2. Whereas in MEGAN the vegetated potential is calculated over the grid cell and then it is multiplied by the related LAI, obtaining the emission per grid cell. The annual CO<sub>2</sub> concentration varies along the simulation from a value of 368 ppm for 2000 to 385 ppm for 2009. For ORCHIDEE, LDF, Ls activity factor and the  $\beta$  coefficient values are given in Table 2. For MEGAN, standard settings, as presented in Table 4 in Guenther et al. (2012), are considered.

### 3 Results

#### 3.1 Global budgets

The validation of BVOC emissions at the global scale is a complex issue because of the lack of observations available globally. Satellite observations provide very useful information, especially regarding the order of magnitude and the seasonal and regional variability of emissions, but the most abundant VOC species are not directly measured (such as isoprene and monoterpenes). Satellite measurements are also subject to large uncertainties arising from the difficulties to retrieve atmospheric concentration of short-lived compounds from space or to separate the different sources (for instance terrestrial biogenic, anthropogenic, oceanic etc.) and the various compounds themselves. Global emission estimates are generally performed using models, or applying inverse modelling techniques that combine the measurements (from satellite, ground or aircraft measurements) and models, providing emissions for compounds such as methanol (Jacob et al., 2005; Millet et al., 2008; Stavrou et al., 2009; Hu et al., 2011; Wells et al., 2012, 2014) and acetaldehyde (Jacob et al., 2002; Millet et al., 2010). Isoprene emissions have also been inferred from satellite formaldehyde concentration (Shim et al., 2005; Palmer et al., 2006; Stavrou et al., 2011, 2014; Barkley et al., 2013; Bauwens et al., 2013).



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At the global scale, the main way to evaluate the results obtained in the present study is to compare them with the most recent state of the art emission budgets derived either from other model runs or from inversion of satellite data. We have compared emission from a large number of estimates published so far, over the 1980–2010 period, with the global emission budgets obtained from ORC\_CRU and MEG\_CRU simulations, the results of which are summarized in Fig. 1. The emissions, calculated by the earlier version of the emission module (black squares, Fig. 1) (Lathièrè et al., 2006), are particularly high, as already pointed out by Sindelarova et al. (2014), with methanol ( $106.1 \text{ TgCyr}^{-1}$ ) and acetaldehyde ( $42.2 \text{ TgCyr}^{-1}$ ) emissions being twice higher, and formaldehyde emissions ( $10.0 \text{ TgCyr}^{-1}$ ) being up to 5 times greater than the other estimates. The results of the new module version (ORC\_CRU, green star) are more in the range of other published estimates. Despite the MEG\_CRU simulation being carried out using the same MEGAN version than in Guenther et al. (2012) (blue hexagon, Fig. 1), there is a noticeable difference between the two emission budgets (especially for isoprene, monoterpenes and acetaldehyde), even when considering results for the same year, 2000. Using, as climate forcings, reanalysis provided by Qian et al. (2006) for the year 2000, Guenther et al. (2012) report BVOC emissions of  $472 \text{ TgCyr}^{-1}$  for isoprene,  $124 \text{ TgCyr}^{-1}$  for monoterpenes (considering the speciated monoterpenes list accounted in this work, i.e.  $\alpha$ -Pinene,  $\beta$ -Pinene, Limonene, Myrcene, Sabinene, 3-Carene, t- $\beta$ -Ocimene) and  $11.5 \text{ TgCyr}^{-1}$  for acetaldehyde. Our MEG\_CRU simulation estimates for 2000 are 410, 72, and  $8.3 \text{ TgCyr}^{-1}$  for isoprene, monoterpenes and acetaldehyde, respectively. As was already pointed out by Arneth et al. (2011), our results confirm that the differences between existing meteorological forcings can lead to substantial differences in emissions estimate (green triangles, first plot of Fig. 1).

We present in Table 6 the annual emissions calculated by ORCHIDEE and MEGAN (ORC\_CRU and MEG\_CRU simulations) at the global scale and for the northern (lat:  $0\text{--}30^\circ \text{ N}$ ) and southern (lat:  $30\text{--}0^\circ \text{ S}$ ) tropics, the northern (lat:  $30\text{--}60^\circ \text{ N}$ ) and southern (lat:  $30\text{--}60^\circ \text{ S}$ ) temperate latitudes and for the northern boreal (lat:  $60\text{--}90^\circ \text{ N}$ ) regions, averaged over the 2000–2009 period. At the global scale, the two models are in a good

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agreement, with the main compound emitted being isoprene with a global calculated source of  $465 \text{ TgCyr}^{-1}$  for ORCHIDEE, accounting for 61 % of total BVOC emissions (estimated to  $757 \text{ TgCyr}^{-1}$ ) and of  $428 \text{ TgCyr}^{-1}$  for MEGAN, accounting for 64 % of total BVOCs (estimated to  $666 \text{ TgCyr}^{-1}$ ). The following most abundant compounds are monoterpenes, accounting for 12 % of total for ORCHIDEE and 11 % for MEGAN, and methanol, accounting for 5 % of the total BVOC emissions for ORCHIDEE and 6 % for MEGAN. Acetone, sesquiterpenes and acetaldehyde represent each 1 to 4 % of the total BVOCs for both models, while other compounds contribute for less than 0.5 %.

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

At the regional scale, the largest differences between ORCHIDEE and MEGAN in terms of absolute values appear in northern temperate regions for isoprene with emissions being  $21 \text{ TgCyr}^{-1}$  higher in ORCHIDEE. Indeed, the marked seasonal cycle of emissions for northern temperate latitudes implies that the highest differences between ORCHIDEE and MEGAN occur in summer. The differences between the two models,

are, in this case, directly linked to discrepancies in the EFs and in the occupying surface of the PFTs at these latitudes (see Fig. 3, plots in the last line). This illustrates the strong impact of different choices in EF allocation, not only regarding global estimates, but also for seasonal and geographical variation of emissions. For the other species the highest differences occur in tropical regions: for example, the emission differences between ORCHIDEE and MEGAN in the northern and southern tropics are  $-2.2$  and  $-2.1 \text{ TgCyr}^{-1}$  for methanol,  $4.3$  and  $10.2 \text{ TgCyr}^{-1}$  for monoterpenes and  $3.9$  and  $4.9 \text{ TgCyr}^{-1}$  for sesquiterpenes.

### 3.2 Emission inter-annual and inter-seasonal variations

In Fig. 2 we present the annual and monthly global emission budgets of ORC\_CRU and MEG\_CRU. The models have very similar annual trends and monthly variations for almost all compounds, illustrating that climate variables, in particular temperature and solar radiation, are the major driving factors, at the global scale, for inter-annual and inter-monthly variabilities.

Nevertheless high differences appear for isoprene. The emissions in ORC\_CRU present a clear seasonal cycle with an emission maximum in July and August that is not simulated in MEG\_CRU results. Indeed, the major differences can be identified in July and August, when global emissions in MEG\_CRU, in average, are lower by  $11.5$  and  $9.0 \text{ TgC month}^{-1}$  compared to ORC\_CRU. Figure 3, where the monthly zonal average for tropical, temperate and northern boreal latitudes regions are depicted, shows that, as mentioned in Sect. 3.1, the ORCHIDEE emissions are significantly higher in northern temperate regions compared to MEGAN, with a marked seasonal cycle and the highest differences between the two models occurring in summer. In particular in July (August), calculated isoprene emissions in ORC\_CRU are about  $4 \text{ TgC month}^{-1}$  ( $5.5 \text{ TgC month}^{-1}$ ) higher than in MEG\_CRU. In July (August), a further important contribution to the global emission peak is attributed to the northern and southern tropics, where ORCHIDEE isoprene emissions are higher, in total, by about  $4 \text{ TgC month}^{-1}$

(5 TgC month<sup>-1</sup>) in comparison to MEGAN in July (August), (Fig. 3, first plot, left column).

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

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

Moreover, we observe in Fig. 3 that the MEGAN emission seasonal cycle in the tropics, especially in the South for isoprene, is more clearly marked than in ORCHIDEE. This behaviour is principally linked to the different seasonal variation between the MODIS and the ORCHIDEE LAI (Fig. 4), with the LAI calculated by ORCHIDEE presenting smaller variations between winter and summer in tropical regions, in particular in Amazonia, (Fig. 4, left column) in comparison to MODIS (Fig. 4, right column).

In northern and southern temperate and northern boreal regions, the seasonal cycle is very similar between the two models, even if ORCHIDEE calculates higher emissions than MEGAN, especially for isoprene.

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### 3.3 Emission geographical distribution

The spatial patterns of BVOC emissions in winter and summer for ORC\_CRU and MEG\_CRU simulations are presented in Figs. 5–9 for isoprene, monoterpenes, methanol, acetone and sesquiterpenes. To better assess the impact of EFs on emissions we show the resulting emission potential for each grid cell, summing the EFs, each weighted by the cell area occupied by each PFT. In MEGAN, emission potentials are already provided per grid cell, instead of EF value per PFT, for isoprene, monoterpenes and MBO (see Sect. 2.3). Emission potentials per grid cell can be interpreted as the average EFs associated to the ecosystem present in the grid cell.

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

$$\text{EP} = \sum_i \text{EF}_i \cdot M/M_{\text{Carbon}} \cdot \text{LAI}_{\text{REF}} \cdot \text{SWL}_i \cdot A_i \quad (6)$$

and for emission depending on light ( $\text{LDF} = 1$ ):

$$\text{EP} = \sum_i \text{EF}_i \cdot M/M_{\text{Carbon}} \cdot \text{LAI}_{\text{REF}} \cdot \text{SWL}_i \cdot A_i \cdot C_{\text{CE}} \quad (7)$$

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

In general, for every compound, we observe a similar geographical distribution. High emission areas are identified in Brazil, equatorial Africa, southern East Asia and southern East USA for both models, with values for ORCHIDEE (MEGAN)

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ranging between:  $5.0\text{--}12.0 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  ( $3.0\text{--}9.0 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ) for isoprene,  $0.8\text{--}2.0 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  ( $0.6\text{--}1.3 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ) for monoterpenes,  $0.3\text{--}1.2 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  ( $0.2\text{--}0.7 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ) for methanol,  $0.2\text{--}0.5 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  ( $0.1\text{--}0.3 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ) for acetone and  $0.4\text{--}0.6 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  ( $0.2\text{--}0.3 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$ ) for sesquiterpenes, respectively. For methanol, in summer, high emitting areas also appear in Europe and Russia, with values of  $0.3\text{--}0.5 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  for ORCHIDEE and of  $0.1\text{--}0.3 \times 10^{10} \text{ kg C m}^{-2} \text{ s}^{-1}$  for MEGAN. Indeed, these regions are populated by temperate and boreal needleleaf evergreen trees, which are strong methanol emitters (Table 3 and Fig. 7, last line).

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

Other remarkable differences between the two models appear in South America for isoprene, directly in relation with the EPs distribution. The pattern of isoprene emissions shows, in MEGAN, higher values in eastern Brazil, Bolivia and northern Argentina, while in ORCHIDEE it is more homogeneous with higher emissions in central Brazil. Same pattern differences can be detected in the emission potential (Fig. 5, last line on the right), and we therefore infer that the EP distribution drives the isoprene emission geographical distribution. The same conclusion is also held for monoterpenes, where lower emissions along the Amazonian river follow perfectly the lower EPs in this area. In general, comparing the emission geographical distribution for each compound and the corresponding emission potential, we can state that, in both models, emission spatial patterns are mostly affected by the EF and by the PFT distribution. Moreover, we found that the emission variation along the year is mainly driven by the

LAI, as we can observe comparing the LAI in winter and summer (Fig. 4) with the compound emitted in the corresponding seasons (Figs. 5–8).

For the compounds that are fully light dependent, such as isoprene (LDF = 1) or largely light dependent, such as methanol (LDF = 0.8), we observe that higher EP in ORCHIDEE than in MEGAN do not necessarily lead to higher emissions in ORCHIDEE. In the case of a LDF close to 1, even when the same EP value is used in both models, the emissions calculated by MEGAN are higher compared to ORCHIDEE, suggesting different response of emissions to light, in the case of BVOCs that are strongly light dependent. Indeed, this effect is less important for compounds which are less dependent on light, such as monoterpenes (LDF = 0.5) and sesquiterpenes (LDF = 0.6), and even negligible for acetone (LDF = 0.2). It therefore seems that the choice of LDF parameter can be crucial in the emission estimate and the sensitivity to EF variation. This point will be further discussed in paragraph 3.5.

### 3.4 BVOC emission sensitivity to LAI

In this section we investigate in more details the differences between the two models arising from LAI and we explore to which extent it can affect BVOC emission estimates.

We showed in Sect. 3.2 that the LAI can have an important role in driving the seasonal cycle of emissions and that some differences between the two model results and emission patterns can be attributed to the different data-sets of LAI used to run ORCHIDEE and MEGAN.

The geographical distribution and global average of ORCHIDEE LAI (solid black line) and MODIS LAI (red line) (Yuan et al., 2011), depicted in Figs. 4 and 11, exhibit big differences between the two models. As illustrated in Fig. 11, the global monthly mean LAI calculated by ORCHIDEE is higher by  $1.5\text{--}2\text{ m}^2\text{ m}^{-2}$  compared to the LAI used in MEGAN and based on MODIS data-sets. In the tropics, the MODIS LAI exhibits a quite clear seasonal cycle, especially in Amazonia, Central Africa and Indonesia, that is not simulated by ORCHIDEE (Fig. 4). The differences between these estimates of LAI are significant. In a validation study, using satellite derived vegetation index time series,

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Maignan et al. (2011) already pointed out some model weaknesses in the ORCHIDEE ability to model correctly the LAI, especially in the equatorial forest (Amazonia, central Africa, Indonesia), where a poor correlation of model output with satellite data was shown. On the other hand, inferring values for LAI at small or large regional scales is particularly challenging, and data available, either coming from field or satellite data, also have some significant uncertainties. Satellites, for instance, measure the effective LAI, obtained from indirect optical methods, and strongly determined by the a priori assumptions that have to be done to allow the inversion procedure. Even starting from the same input reflectance, diverse retrieval methods can lead to LAI values that are highly different (Garrigues et al., 2008; Fang et al., 2013). The effective LAI can be very dissimilar to the LAI directly measured in situ and relative differences can reach 100 % (Fang et al., 2012a, b). Field and satellite data bring very useful and complementary information regarding the order of magnitude, the seasonal and the geographical variability of LAI, but performing a robust evaluation of model ability to simulate the LAI, especially at the global scale, still remains challenging, and is also beyond the objective of our study.

We deeply examine the response of (i) MEGAN when forced with LAI provided by ORCHIDEE, (ii) ORCHIDEE and MEGAN to the same LAI variations, considering changes that have the same order of magnitude than the LAI uncertainties. We therefore perform 2 more simulations for each model, using the ORCHIDEE LAI multiplied by a factor of 0.5 or 1.5.

To fulfil the first objective, we perform a 10-year simulation following the same configuration as in the previous runs, but forcing MEGAN with the ORCHIDEE LAI (MEG\_CRULAI simulation, Table 5). This simulation confirms that LAI is one of the key drivers regulating the seasonal cycle. Generally, for every compound, we observe a better agreement between the MEG\_CRULAI and the ORC\_CRU simulations than between MEG\_CRU and ORC\_CRU, especially in the tropical regions. In the Fig. 10, where the tropical zonal means of monthly isoprene emissions are depicted, we observe that the variability, along the year, calculated in MEGAN is closer to the one de-



terminated by ORCHIDEE. Comparing the MEG\_CRULAI (Fig. 10) with the MEG\_CRU simulation (Fig. 3, first plot), we can see that the emission peak in northern tropics in April and May and the emission decrease in southern tropics in June and July are less marked.

Nevertheless, the global and zonal emission budgets (Table 7) in MEG\_CRULAI simulation are not significantly different than the ones determined in MEG\_CRU, even if the ORCHIDEE LAI is significantly higher than MODIS one. At the regional scale, in boreal and temperate regions the MEG\_CRULAI emissions are slightly higher than the MEG\_CRU ones, and in the tropics they are even slightly lower for some compounds. As proposed by Sindelarova et al. (2014), a possible reason for the emission decrease calculated in the tropics by MEGAN, can be attributed to the strengthened effect of leaf self-shading caused by the increase in LAI in locations characterized by a dense vegetation (e.g. in central Africa or Amazon). This effect can be predominant for compounds for which biogenic emissions are strongly dependent on light, such as isoprene or methanol.

Indeed, for the other compounds the MEG\_CRU and MEG\_CRULAI emission budgets are very similar. We could foresee that these results are linked to the leaf self-shading effect on leaf temperature. In contrast to ORCHIDEE where the air temperature is used, in MEGAN the leaf temperature is calculated for shaded and sunlit leaves. If the leaf self-shading effect was crucial even for light-independent compounds, we would expect a much higher leaf temperature for sunlit leaves than for shaded leaves. Calculating the difference of hourly leaf temperature between sunlit and shaded leaves in the case of dense vegetation (TrBrEv and TrBrDe), we estimate a differences of about 1–1.5 °C, that could unlikely be high enough to explain such differences in emissions. Lathière et al. (2006), for instance, found that an increase of the global surface temperature by 1 °C leads to an increase of isoprene emissions at most of 11 %. We, therefore, doubt that the only mechanism, behind the not-increasing BVOC emissions for light-independent compound, is the leaf self-shading.

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The LAI is a strong driver of emissions, it is therefore important to investigate in more details if models show the same response to a particular change in LAI. In Fig. 12, we present four simulations that are performed forcing both MEGAN and ORCHIDEE with the ORCHIDEE LAI (Fig. 11) multiplied by a factor of 0.5 (MEG\_LAI05 and ORC\_LAI05) and by a factor of 1.5 (MEG\_LAI15 and ORC\_LAI15) for the year 2006 (details in Table 5). Only the zonal average for tropics South and northern temperate area, for isoprene and monoterpenes are displayed, but are representative examples for other regions.

Regarding isoprene, we observe that ORCHIDEE and MEGAN present a similar response to LAI variation. When the LAI is multiplied by a factor of 0.5 (1.5), change in emissions compared to the reference runs (MEG\_CRULAI, ORC\_CRU) reaches  $-18\%$  ( $+12\%$ ) for MEGAN and  $-21\%$  ( $+8\%$ ) for ORCHIDEE in the southern tropics and reaches  $-34\%$  ( $+21\%$ ) for MEGAN and  $-32\%$  ( $+16\%$ ) for ORCHIDEE in northern temperate areas. In the tropics especially, the emissions calculated by the two models are little sensitive to the LAI increase. Indeed as isoprene is a light-dependent compound, the contribution of lowest LAI layers to emissions is insignificant beyond a certain LAI threshold, as there is no more light available. We observe that MEGAN is less sensitive than ORCHIDEE to LAI increase, implying much likely that the MEGAN canopy model includes a solar light decrease inside the canopy slightly stronger than in ORCHIDEE. Monoterpene emissions show a different response in term of sensitivity to LAI. In the southern tropics, the relative difference of monoterpene emission budget between ORC\_LAI05 (ORC\_LAI15) and ORC\_CRU is  $-43\%$  ( $+40\%$ ), and  $-9\%$  ( $+3\%$ ) between MEG\_LAI05 (MEG\_LAI15) and MEG\_CRULAI. In northern temperate regions, relative difference in monoterpene emission budget between ORC\_LAI05 (ORC\_LAI15) and ORC\_CRU is  $-44\%$  ( $+40\%$ ), and  $-14\%$  ( $+6\%$ ) between MEG\_LAI05 (MEG\_LAI15) and MEG\_CRULAI. These simulations confirm a much small emission impact of LAI variation on emission in MEGAN, even for compounds not fully depending on light, such as monoterpenes (LDF = 0.6).

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In Table 8 we provide the total emission budget calculated for MEG\_LAI05, ORC\_LAI05, MEG\_LAI15 and ORC\_LAI15 simulations for every compounds. In general in ORCHIDEE, the lower the light dependence, the higher the sensitivity to LAI is, while for MEGAN, the sensitivity to LAI does not significantly change with LDF. The explanation for this difference in emission response lies in the different formulation used in the two models for light independent emissions. Indeed in the ORCHIDEE model, the light independent emission from a particular PFT shows a clear linear dependence on LAI, as shown in the Eqs. (1) and (2) in the present paper. In MEGAN however, the light independent emission is calculated using, among other factors,  $\gamma_{LAI}$ , which is the factor accounting for the relation between LAI and emission, equal to  $(0.49 \cdot LAI)/(1 + 0.2 \cdot LAI^2)^{0.5}$  (Guenther et al., 2006). This factor increases almost linearly for LAI lower than  $2 \text{ m}^2 \text{ m}^{-2}$ , followed by a more moderate increase rate for LAI of  $2\text{--}5 \text{ m}^2 \text{ m}^{-2}$  and is then almost stable for LAI higher than  $5 \text{ m}^2 \text{ m}^{-2}$ . The light-independent emission descriptions in the two models therefore respond differently to LAI variation, with differences more striking when LAI is greater than  $2 \text{ m}^2 \text{ m}^{-2}$ , while the ORCHIDEE emissions keep increasing linearly with LAI, the MEGAN emissions do not increase as strongly anymore. In this case, the different modelling choices bring significant discrepancies in emission sensitivity between the two models.

### 3.5 BVOC emission sensitivity to LDF

As described in Sect. 2.2, LDF parameter sets the light-dependent fraction of emissions for each compound. Many experimental studies point out for several plant species that, if emissions can be totally light-independent for some BVOCs, the emissions of most of them are actually light dependent, with a more or less high dependency depending on the considered compound (Jacob et al., 2002, 2005; Hansen and Seufert, 2003; Dindorf et al., 2006; Holzke et al., 2006; Harley et al., 2007; Millet et al., 2008, 2010; Hu et al., 2011; Wells et al., 2014). As the results of these studies are highly heterogeneous, assigning a single LDF value for each compound is as difficult as assigning the EFs for each PFT (Sect. 2.2). Hence the LDF uncertainty could be even higher than

the one associated to EFs, as there are less quantitative studies about this subject published so far.

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

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

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

In Fig. 13 the daily profile averaged over each month of the LDF index is presented to investigate the daily and annual variations. At the global scale (left panel) we observe that the LDF index associated to MEGAN is much higher (up to 20 %), than the one associated to ORCHIDEE. At the regional scale, in the southern tropics for example (second panel) it reaches up to 70 % and is the double of the index calculated for

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ORCHIDEE. The light-dependent part of emissions in MEGAN is therefore more important than the ORCHIDEE one, with important impacts on emission estimate. Firstly, we show that based on the same EF value, the MEGAN emissions are higher than in ORCHIDEE for compounds associated to high LDF, as expected from Sect. 3.3. Secondly, as the light dependent and independent part are combined in an additive way to produce emissions Eq. (2), *orcldf0* or *megldf0* compound can be figured out as the independent part of emission and *orcldf1* or *megldf1* as the dependent part. According to these results, starting from the same light independent emissions in the two models, if the LDF parameter is increased, the MEGAN emission increases faster than ORCHIDEE ones, as the light-dependent component of emission in MEGAN is higher in comparison with ORCHIDEE. MEGAN therefore results more sensitive to LDF variation than ORCHIDEE.

## 4 Conclusions

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

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

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The ORCHIDEE emission estimates are within the range of the published emission budgets. The ORCHIDEE global budgets averaged over the investigated period (2000–2009) are  $465 \text{ TgCyr}^{-1}$  for isoprene,  $108 \text{ TgCyr}^{-1}$  for monoterpenes,  $38 \text{ TgCyr}^{-1}$  for methanol,  $25 \text{ TgCyr}^{-1}$  for acetone and  $24 \text{ TgCyr}^{-1}$  for sesquiterpenes. The global emission budgets are, in general, in good agreement between the two models, with the ORCHIDEE emissions being 8 % higher for isoprene, 8 % lower for methanol, and 17 % higher for acetone, 18 % higher for monoterpenes and 39 % higher for sesquiterpenes compared to MEGAN results. At the regional scale, the largest differences in term of spatial emission distribution between ORCHIDEE and MEGAN, occur in the northern temperate region for isoprene. This different behaviour is directly linked to differences in the EF and PFT distribution in this area.

As a more general statement, considering the emissions geographical distribution for each compound and the correspondent emission potential, we show that, in both models, EF and PFT distribution are the main drivers of emission geographical pattern. In term of seasonal variation, the differences between the two models in the tropics are mostly due to the different seasonal cycle of LAI between MODIS and ORCHIDEE, while the high discrepancy in northern temperate regions is attributed to differences in the EF distribution.

The LAI calculated by ORCHIDEE is higher by  $1.5\text{--}2 \text{ m}^2 \text{ m}^{-2}$  than the LAI retrieved by MODIS. We examine how these discrepancies can infer on the BVOC estimates, exploring the emission sensitivity to LAI. Sensitivity tests are then performed multiplying the ORCHIDEE LAI by a factor of 0.5 and 1.5. ORCHIDEE and MEGAN emissions present a similar response to LAI variation for isoprene if LAI is multiplied by a factor of 0.5 (1.5), with global emission budget, varying by  $-21 \%$  ( $+7.8 \%$ ) with respect to ORCHIDEE reference run and by  $-14 \%$  ( $+6.6 \%$ ) for MEGAN. On the other hand, for monoterpenes, ORCHIDEE is much more sensitive to LAI variation, with global emission budget changing by  $-43 \%$  ( $+40 \%$ ), while MEGAN presents a global emission budget changing by  $-12 \%$  ( $+6 \%$ ). These discrepancies are linked to differences in the light independent emission formulation between the two models. ORCHIDEE con-

siders a linear dependence of emissions on LAI, while MEGAN assign a quasi-linear dependence of emissions for LAI up to  $2 \text{ m}^2 \text{ m}^{-2}$  and an asymptotic increase with almost no change for LAI greater than  $5 \text{ m}^2 \text{ m}^{-2}$ .

We investigate the contribution of the light-dependent and light-independent part of emissions and consequently the impact that a different choice of LDF can have on emissions. In MEGAN, the light independent part of emissions is more important than in ORCHIDEE reaching a factor of two in the southern tropics. Increasing the LDF parameter, the MEGAN emissions increase faster than in ORCHIDEE, implying that the MEGAN estimates are more sensitive to LDF variation than ORCHIDEE.

Our results underline that because of the high uncertainties of the involved variables/parameters and the different choices in modelling processes, high uncertainties still affecting the BVOC emission estimates. Model inter-comparison and sensitivity tests can be extremely useful to understand to which parameters/variables BVOC emissions are most affected, why they are so sensitive, and which is the most effective way to improve their evaluation. Our results highlight the importance and the need to further explore the BVOC emission estimate variability.

Models such as ORCHIDEE or MEGAN, among many others, are widely used to investigate biosphere-atmospheric chemistry interactions at different scales, from local to global. Those models are usually built on the same or very similar empirical approaches, but can differ substantially on some aspects, for instance in the number and diversity of plant species taken into account, the EF allocation, the description of the LAI (either simulated, when using a vegetation model, or prescribed), the use of air or leaf temperature to calculate emissions. BVOC emission estimates provided throughout the literature can therefore exhibit different patterns in term of global estimates, geographical or seasonal variation. Field campaigns and laboratory experiments provide very valuable information, enabling the modellers to complement the description of BVOCs in emission scheme, in order to take into account the improvement of our knowledge. Such data are also very challenging to acquire, especially on the long-term. The modelling community therefore still faces the problem of BVOC emission

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model evaluation, with not yet any robust and satisfying way to properly examine our results in term of global numbers or regional/seasonal/inter-annual variations.

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**Table 1.** Plant Functional Types in ORCHIDEE and MEGAN and corresponding occupied surfaces in  $10^{12} \text{ m}^2$ .

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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**Table 2.** Comparison between the old and new versions of the biogenic emission module in ORCHIDEE: list of emitted compounds, principal parameters for emission equations, radiation model type and compounds for whom 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, $\alpha$ -pinene, $\beta$ -pinene, Limonene, Myrcene, Sabinene, Camphene, 3-Carene, t- $\beta$ -Ocimene, Other monoterpenes, Sesquiterpene, MBO, Other VOCs	Isoprene, MBO	1.0	0.9	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, $\alpha$ -pinene, $\beta$ -pinene, Limonene, Myrcene, Sabinene, Camphene	0.6	0.6		
		3-Carene, t- $\beta$ -Ocimene, Other monoterpenes				
		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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**Table 3.** Emission Factors (EFs,  $\mu\text{g C gdm}^{-1} \text{h}^{-1}$ ) for each PFT for the main compounds emitted, in the previous (first line) and actual (second line, 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	8.0	16.0	45.0	8.0	8.0	8.0	16.0	24.0	5.0	5.0	He et al. (2000), Klinger et al. (2002), Levis 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. (2009), Steinbrecher et al. (2009, 2013), Tsui et al. (2009), Lathièrre et al. (2010), Leung et al. (2010), Arneth et al. (2011), Fu and Liao (2012), Oderbolz et al. (2013)
	<b>24.0</b>	<b>24.0</b>	<b>8.0</b>	<b>16.0</b>	<b>45.0</b>	<b>8.0</b>	<b>18.0</b>	<b>0.5</b>	<b>12.0</b>	<b>18.0</b>	<b>5.0</b>	<b>5.0</b>	
Monoterp.	0.800	0.800	2.400	1.200	0.800	2.400	2.400	2.400	0.800	1.200	0.200	0.200	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), Guenther et al. (2006, 2012), 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)
	<b>2.000</b>	<b>2.000</b>	<b>1.800</b>	<b>1.400</b>	<b>1.600</b>	<b>1.800</b>	<b>1.400</b>	<b>1.800</b>	<b>0.800</b>	<b>0.800</b>	<b>0.220</b>	<b>0.220</b>	
Sesqiterp.	–	–	–	–	–	–	–	–	–	–	–	–	Guenther et al. (2006, 2012), Duhl et al. (2008), Matsumaga 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)
	<b>0.450</b>	<b>0.450</b>	<b>0.130</b>	<b>0.300</b>	<b>0.360</b>	<b>0.150</b>	<b>0.300</b>	<b>0.250</b>	<b>0.600</b>	<b>0.600</b>	<b>0.080</b>	<b>0.080</b>	
Methanol	0.600	0.600	1.800	0.900	0.600	1.800	1.800	1.800	0.600	0.900	2.000	2.000	Schade and Goldstein (2001), Karl et al. (2004, 2005, 2009), Hayward et al. (2004), Guenther et al. (2006, 2012), Smiatek and Steinbrecher (2006), Harley et al. (2007), Chang et al. (2009), Steinbrecher et al. (2009), Bracho-Nunez et al. (2011), Fares et al. (2011)
	<b>0.800</b>	<b>0.800</b>	<b>1.800</b>	<b>0.900</b>	<b>1.900</b>	<b>1.800</b>	<b>1.800</b>	<b>1.800</b>	<b>0.700</b>	<b>0.900</b>	<b>2.000</b>	<b>2.000</b>	
Acetone	0.290	0.290	0.870	0.430	0.290	0.870	0.870	0.870	0.290	0.430	0.070	0.070	Janson et al. (1999), Janson and De Serves (2001), Schade and Goldstein (2001), Karl et al. (2004, 2005, 2009), Villanueva-Fierro et al. (2004), Guenther et al. (2006, 2012), Smiatek and Steinbrecher (2006), Chang et al. (2009), Steinbrecher et al. (2009), Bracho-Nunez et al. (2011), Fares et al. (2011)
	<b>0.250</b>	<b>0.250</b>	<b>0.300</b>	<b>0.200</b>	<b>0.330</b>	<b>0.300</b>	<b>0.250</b>	<b>0.250</b>	<b>0.200</b>	<b>0.200</b>	<b>0.080</b>	<b>0.080</b>	

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**Table 3.** Continued.

	TrBrEv	TrBrDe	TeNeEv	TeBrEv	TeBrDe	BoNeEv	BoBrDe	BoNeDe	C3Gr	C4Gr	C3Ag	C4Ag	References
Acetaldeh.	0.100 <b>0.200</b>	0.100 <b>0.200</b>	0.300 <b>0.200</b>	0.150 <b>0.200</b>	0.100 <b>0.250</b>	0.300 <b>0.250</b>	0.300 <b>0.160</b>	0.300 <b>0.160</b>	0.100 <b>0.120</b>	0.150 <b>0.120</b>	0.025 <b>0.035</b>	0.025 <b>0.022</b>	Kesselmeier et al. (1997), Janson et al. (1999), Martin et al. (1999), Schade and Goldstein (2001), Hayward et al. (2004), Karl et al. (2004, 2005), Villanueva-Fierro et al. (2004), Guenther et al. (2006, 2012), Smiatek and Steinbrecher (2006), Chang et al. (2009), Steinbrecher et al. (2009), Fares et al. (2011)
Formaldeh.	0.070 <b>0.040</b>	0.070 <b>0.040</b>	0.200 <b>0.080</b>	0.100 <b>0.040</b>	0.070 <b>0.040</b>	0.200 <b>0.040</b>	0.200 <b>0.040</b>	0.200 <b>0.040</b>	0.070 <b>0.025</b>	0.100 <b>0.025</b>	0.017 <b>0.013</b>	0.017 <b>0.013</b>	Kesselmeier et al. (1997), Janson et al. (1999), Martin et al. (1999), Villanueva-Fierro et al. (2004), Guenther et al. (2006, 2012), Smiatek and Steinbrecher (2006), Chang et al. (2009), Karl et al. (2009), Steinbrecher et al. (2009)
Acetic Acid	0.002 <b>0.025</b>	0.002 <b>0.025</b>	0.006 <b>0.025</b>	0.003 <b>0.022</b>	0.002 <b>0.080</b>	0.006 <b>0.025</b>	0.006 <b>0.022</b>	0.006 <b>0.013</b>	0.002 <b>0.012</b>	0.003 <b>0.012</b>	0.001 <b>0.008</b>	0.001 <b>0.008</b>	Kesselmeier et al. (1997, 1998), Martin et al. (1999), Staudt et al. (2000), Villanueva-Fierro et al. (2004), Guenther et al. (2006, 2012), Smiatek and Steinbrecher (2006), Chang et al. (2009), Karl et al. (2009), Steinbrecher et al. (2009)
Formic Acid	0.010 <b>0.015</b>	0.010 <b>0.015</b>	0.030 <b>0.020</b>	0.015 <b>0.020</b>	0.010 <b>0.025</b>	0.030 <b>0.025</b>	0.030 <b>0.015</b>	0.030 <b>0.015</b>	0.010 <b>0.010</b>	0.0150 <b>0.010</b>	0.0025 <b>0.008</b>	0.0025 <b>0.008</b>	Kesselmeier et al. (1997, 1998), Martin et al. (1999), Staudt et al. (2000), Villanueva-Fierro et al. (2004), Guenther et al. (2006, 2012), Smiatek and Steinbrecher (2006), Chang et al. (2009), Karl et al. (2009), Steinbrecher et al. (2009)
MBO	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	20.000 <b>1.4</b>	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	0.000 <b>0.14</b>	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	0.000 <b>0.00002</b>	Baker et al. (1999), Schade and Goldstein (2001), Tarvainen et al. (2005), Guenther et al. (2006, 2012), Hakola et al. (2006), Chang et al. (2009), Kim et al. (2010)

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

	TrBrEv	TrBrDe	TeNeEv	TeBrEv	TeBrDe	BoNeEv	BoBrDe	BoNeDe	C3Gr	C4Gr	C3Ag	C4Ag
$\alpha$ -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
$\beta$ -Pinene	11	11	14.6	12.2	8.7	14.6	6.3	15.0	12.3	8.0	15.4	15.4
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
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
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
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
3-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
t- $\beta$ -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
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

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

Simulation Name	Model	Climate Forcing	EFs	LDF	LAI	<i>T</i>	Period	Output Frequency
ORC_CRU	ORCHIDEE	CRU	Standard version	Standard version	ORCHIDEE LAI	<i>T</i> air	2000–2009	1 month
MEG_CRU	MEGAN	CRU	Standard version	Standard version	Modis Lai	<i>T</i> leaf	2000–2009	1 month
MEG_CRULAI	MEGAN	CRU	Standard version	Standard version	ORCHIDEE LAI	<i>T</i> leaf	2000–2009	1 month
ORC_LAI05	ORCHIDEE	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 0.5	<i>T</i> air	2006	1 month
ORC_LAI15	ORCHIDEE	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 0.5	<i>T</i> air	2006	1 month
MEG_LAI05	MEGAN	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 0.5	<i>T</i> leaf	2006	1 month
MEG_LAI15	MEGAN	CRU	Standard version	Standard version	ORCHIDEE LAI multiplied by 0.5	<i>T</i> leaf	2006	1 month
ORC_LDF	ORCHIDEE	CRU	EFs = 1	LDF = 1 and 0	ORCHIDEE LAI	<i>T</i> air	2006	1 h
MEG_LDF	MEGAN	CRU	EFs = 1	LDF = 1 and 0	ORCHIDEE LAI	<i>T</i> air	2006	1 h

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**Table 6.** Emission budget ( $\text{Tg C yr}^{-1}$ ) averaged over the 2000–2009 period for the ORC\_CRU (bold lines) and MEG\_CRU simulations at the global scale, for northern and southern tropics, for northern and southern temperate area and for northern boreal region.

Model	Area	Isoprene	Methanol	Acetone	Acetald	Formald	Acetic Acid	Formic Acid	MBO	Sesqiter.	Monoter	$\alpha$ -Pinene	$\beta$ -Pinene	Limonen	Myrcene	Sabinene	3-Carene	T- $\beta$ -Ocimene
<b>ORCHIDEE</b>	<b>Global</b>	<b>464.6</b>	<b>37.8</b>	<b>24.6</b>	<b>8.6</b>	<b>1.9</b>	<b>1.1</b>	<b>0.7</b>	<b>1.3</b>	<b>24.3</b>	<b>91.3</b>	<b>40.9</b>	<b>12.2</b>	<b>10.7</b>	<b>7.2</b>	<b>8.19</b>	<b>6.5</b>	<b>9.3</b>
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			36.7		
<b>ORCHIDEE</b>	<b>Tro North</b>	<b>176.3</b>	<b>12.9</b>	<b>8.6</b>	<b>2.9</b>	<b>0.6</b>	<b>0.4</b>	<b>0.2</b>	<b>0.1</b>	<b>9.6</b>	<b>32.8</b>	<b>14.8</b>	<b>4.3</b>	<b>4.0</b>	<b>2.7</b>	<b>2.9</b>	<b>2.0</b>	<b>3.5</b>
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			13.9		
<b>ORCHIDEE</b>	<b>Tro South</b>	<b>217.7</b>	<b>13.0</b>	<b>10.9</b>	<b>3.8</b>	<b>0.8</b>	<b>0.5</b>	<b>0.3</b>	<b>0.0</b>	<b>12.3</b>	<b>42.7</b>	<b>19.6</b>	<b>5.5</b>	<b>5.1</b>	<b>3.6</b>	<b>3.7</b>	<b>2.4</b>	<b>4.6</b>
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			16.4		
<b>ORCHIDEE</b>	<b>Tem North</b>	<b>51.6</b>	<b>9.1</b>	<b>3.6</b>	<b>1.3</b>	<b>0.3</b>	<b>0.2</b>	<b>0.1</b>	<b>1.2</b>	<b>1.6</b>	<b>10.9</b>	<b>4.3</b>	<b>1.6</b>	<b>1.1</b>	<b>0.6</b>	<b>1.2</b>	<b>1.6</b>	<b>0.8</b>
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			4.4		
<b>ORCHIDEE</b>	<b>Tem South</b>	<b>5.4</b>	<b>0.6</b>	<b>0.3</b>	<b>0.1</b>	<b>0.03</b>	<b>0.01</b>	<b>0.01</b>	<b>0.0</b>	<b>0.2</b>	<b>1.0</b>	<b>0.5</b>	<b>0.1</b>	<b>0.1</b>	<b>0.06</b>	<b>0.1</b>	<b>0.04</b>	<b>0.1</b>
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			0.5		
<b>ORCHIDEE</b>	<b>Boreal</b>	<b>4.4</b>	<b>1.5</b>	<b>0.6</b>	<b>0.2</b>	<b>0.05</b>	<b>0.03</b>	<b>0.02</b>	<b>0.03</b>	<b>0.2</b>	<b>2.0</b>	<b>0.9</b>	<b>0.3</b>	<b>0.2</b>	<b>0.1</b>	<b>0.2</b>	<b>0.3</b>	<b>0.15</b>
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			0.7		

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**Table 7.** Mean emission budgets ( $\text{TgCyr}^{-1}$ ) for the 2000–2009 period estimated in MEG\_CRULAI simulation.

MEGAN	Isoprene	Methanol	Acetone	Acetald	Formald	Acetic Acid	Formic Acid	MBO	Sesquiterp	Monoterp	$\alpha$ -Pinene	$\beta$ -Pinene	Other Monoterp
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	36.4
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	13.6
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	16.1
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	4.5
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	0.5
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	0.8

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**Table 8.** Annual emission budgets ( $\text{TgCyr}^{-1}$ ) for the year 2006 in ORC\_CRU, MEG\_CRULAI, taken as reference, and in the LAI sensitivity tests (ORC\_LAI05, ORC\_LAI15, MEG\_LAI05 and MEG\_LAI15).

Simulation	Isoprene	Methanol	Acetone	Acetald	Formald	Acetic Acid	Formic Acid	MBO	Sesquiter	Monoter	$\alpha$ -Pinene	$\beta$ -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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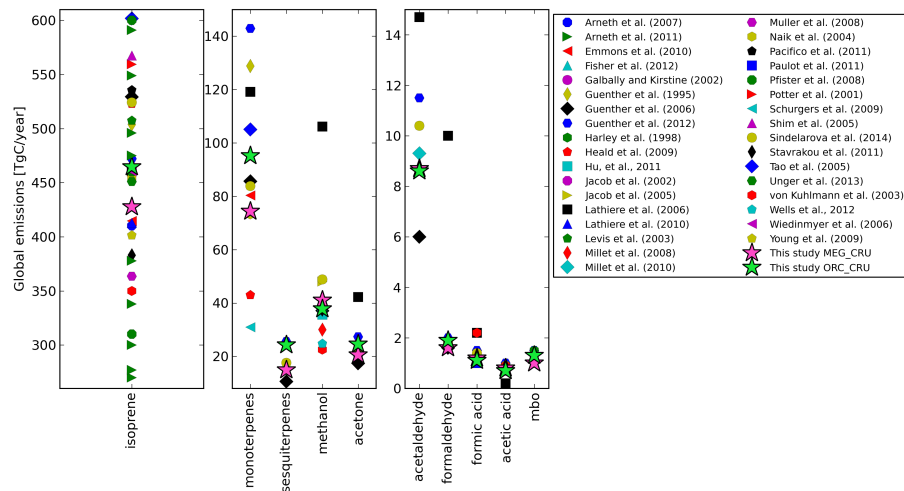
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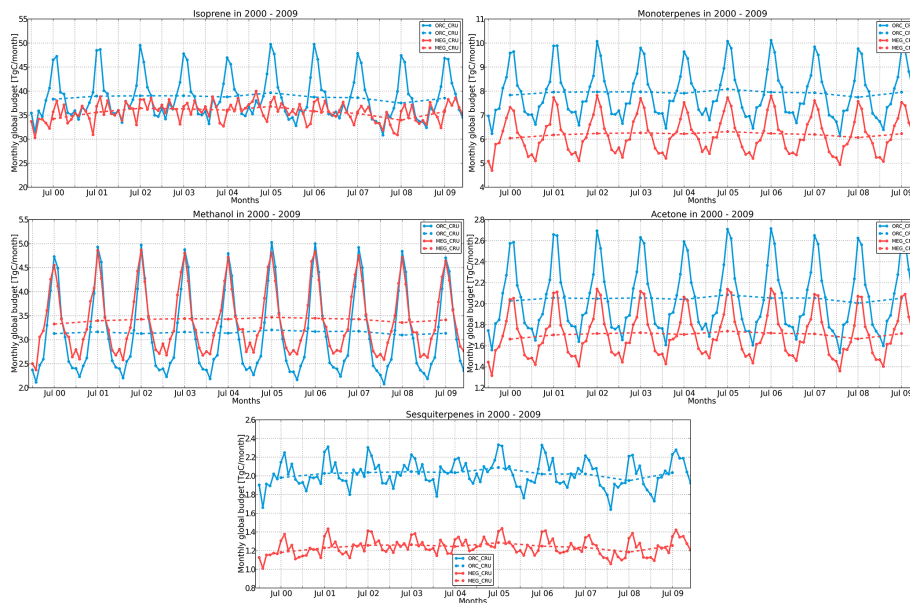
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**Figure 1.** Global emission budgets (TgCyr<sup>-1</sup>) calculated by ORCHIDEE (ORC\_CRU simulation, green stars) and MEGAN (MEG\_CRU simulation, pink stars), compared with published estimates for the main BVOCs presented in this work. Note that the vertical axes have different scales in the three plots.

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**Figure 2.** Monthly global (solid lines) and yearly averaged (dashed lines) emission budgets in  $\text{TgCmonth}^{-1}$  for ORC\_CRU and MEG\_CRU simulations for isoprene, monoterpenes, methanol, acetone and sesquiterpenes.

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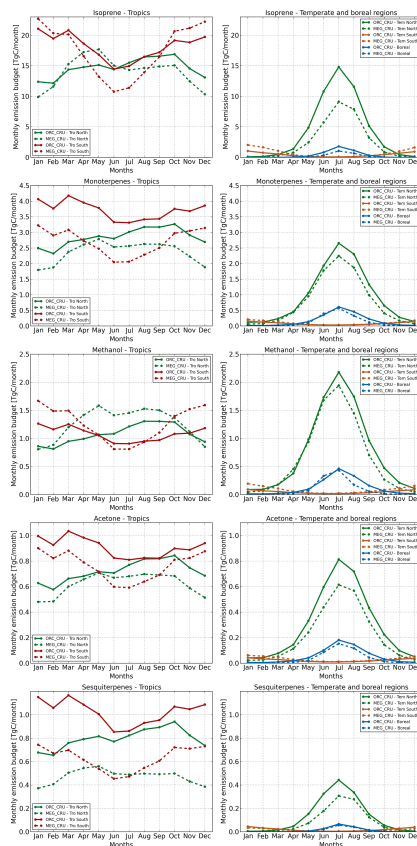
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**Figure 3.** Zonal mean for northern and southern tropics (left column), northern and southern temperate and northern boreal latitudes (right column) of the monthly emission budget ( $\text{TgCmonth}^{-1}$ ) averaged over the simulation period (2000–2009), in ORCHIDEE and MEGAN runs for isoprene, monoterpenes, methanol, acetone and sesquiterpenes, respectively.

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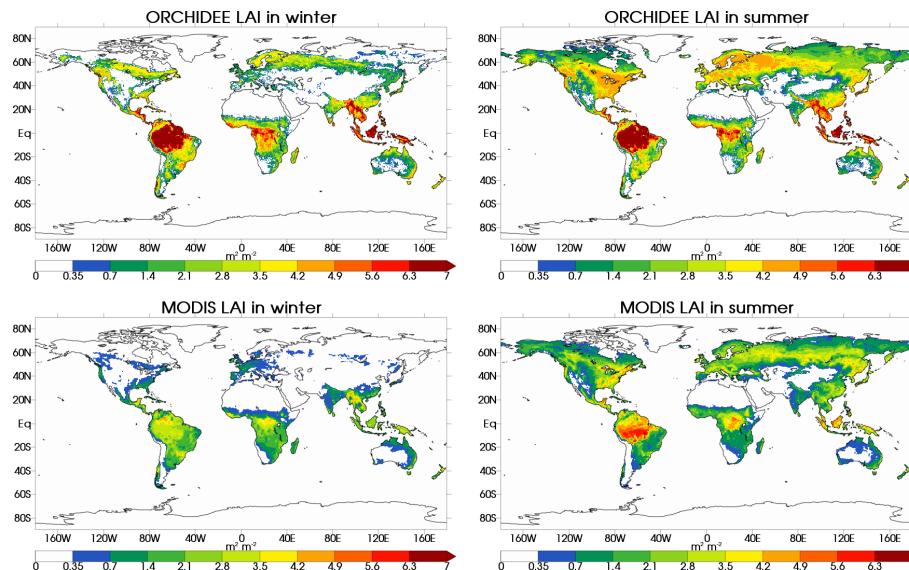
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**Figure 4.** Leaf area index (LAI) considered for BVOC emission estimate in ORCHIDEE (LAI calculated on line) and in MEGAN (MODIS retrieval) in summer (June, July, August) and winter (December, January, February), averaged over the 2000–2009 period ( $\text{m}^2 \text{m}^{-2}$ ).

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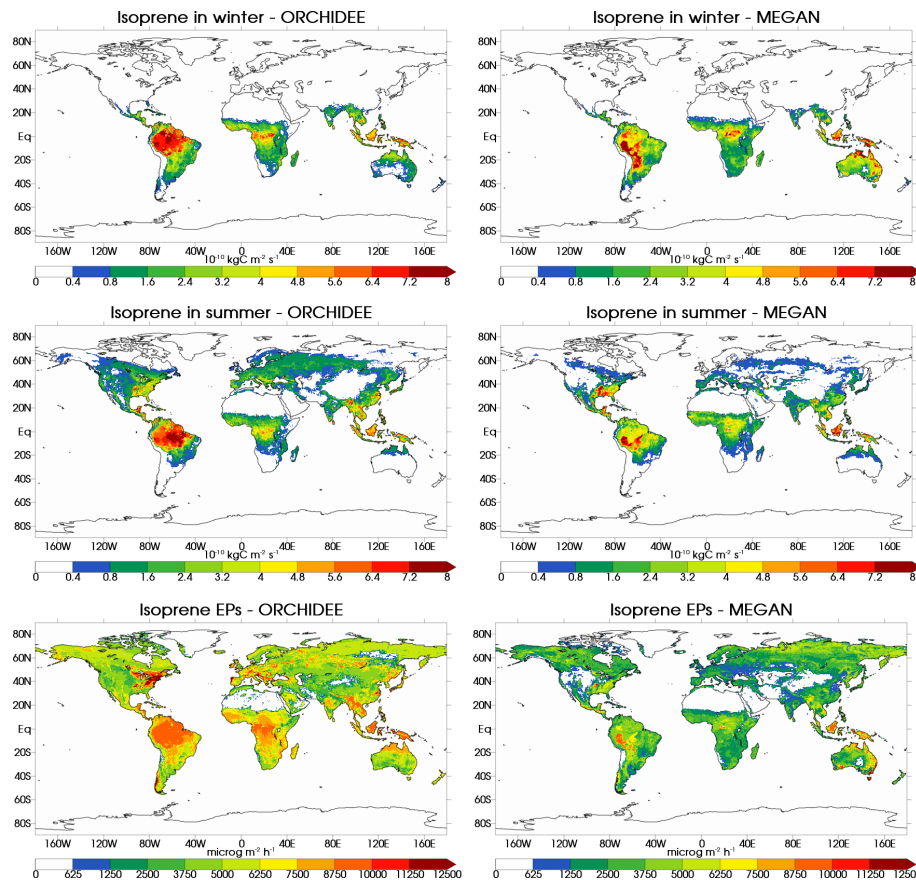
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**Figure 5.** Emissions in winter (first line) and summer (second line) in  $10^{-10} \text{ kg C m}^{-2} \text{ s}^{-1}$  and emission potentials (EPs) (third line) in  $\mu\text{g m}^{-2} \text{ h}^{-1}$  for ORCHIDEE (ORC\_CRU, left-hand column) and MEGAN (MEG\_CRU, right-hand column) for isoprene.

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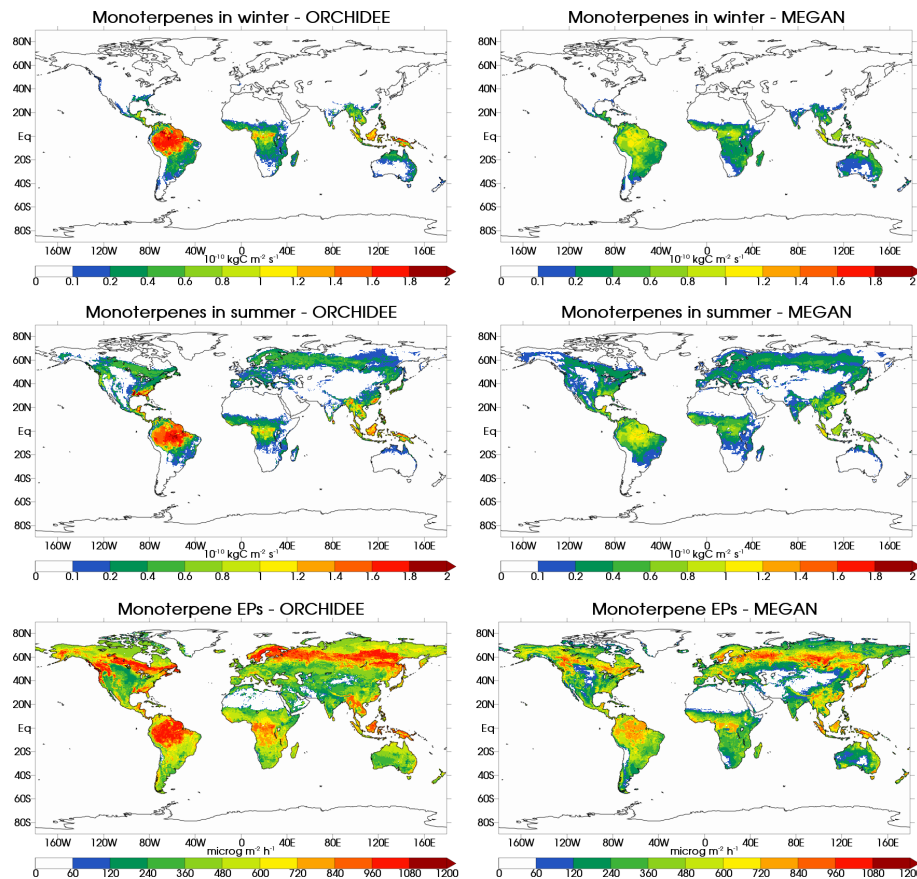


Figure 6. The same as Fig. 5, but for monoterpenes.

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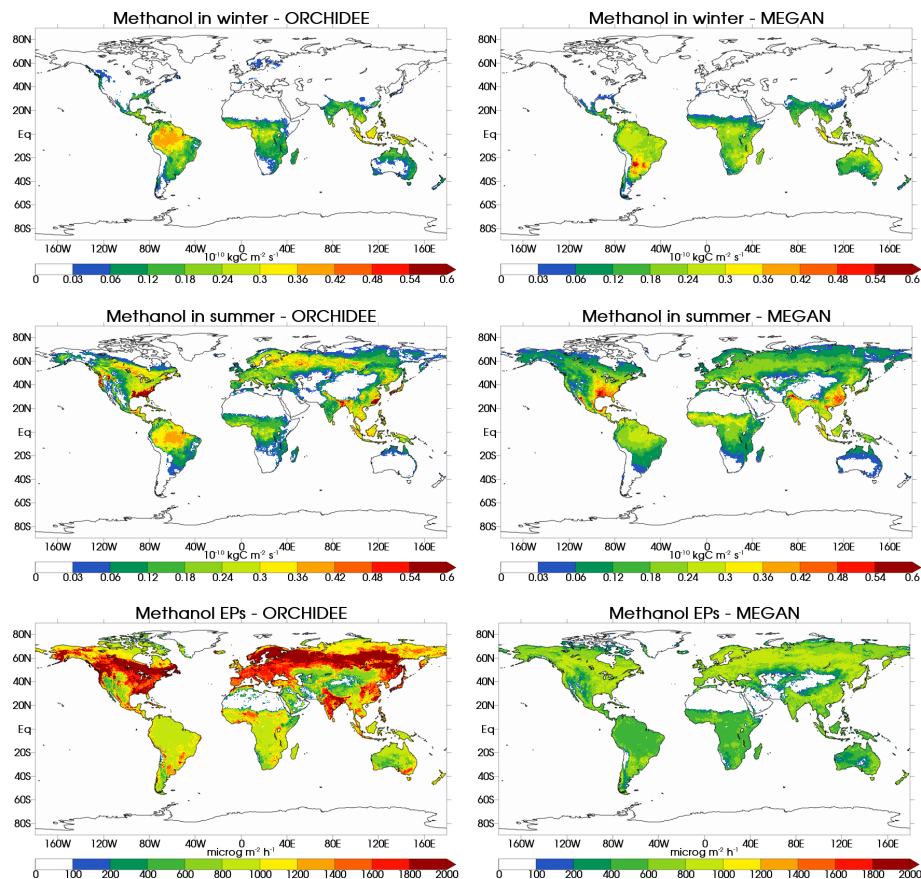


Figure 7. The same as Fig. 5, but for methanol.

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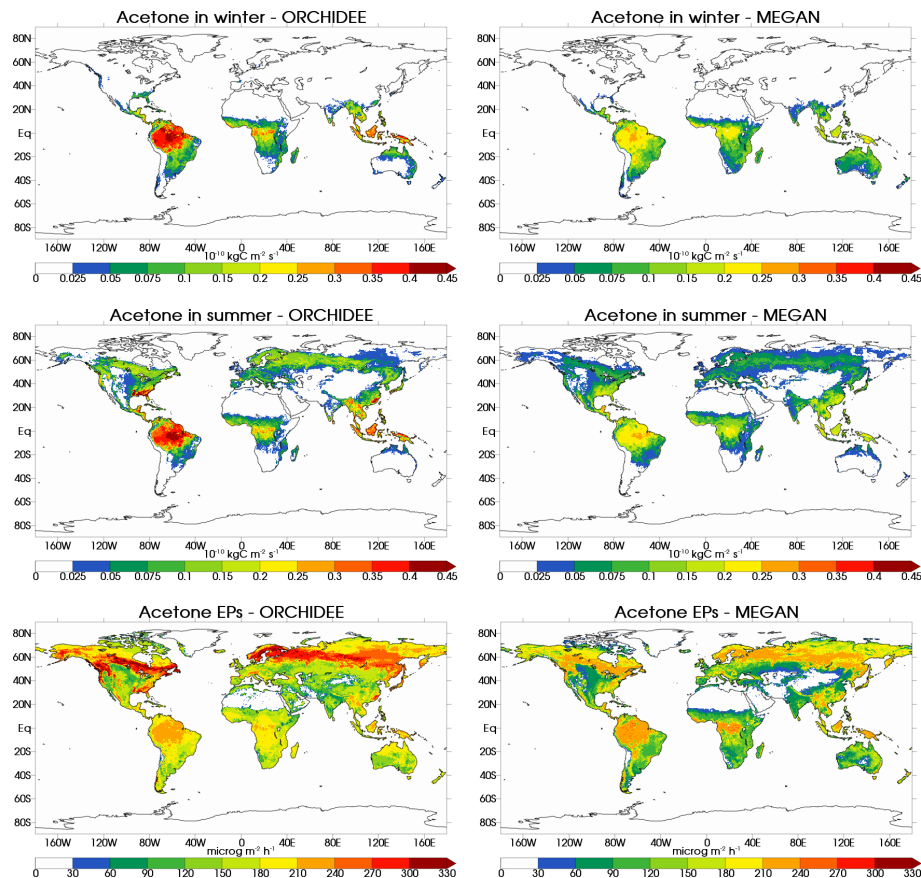


Figure 8. The same as Fig. 5, but for acetone.

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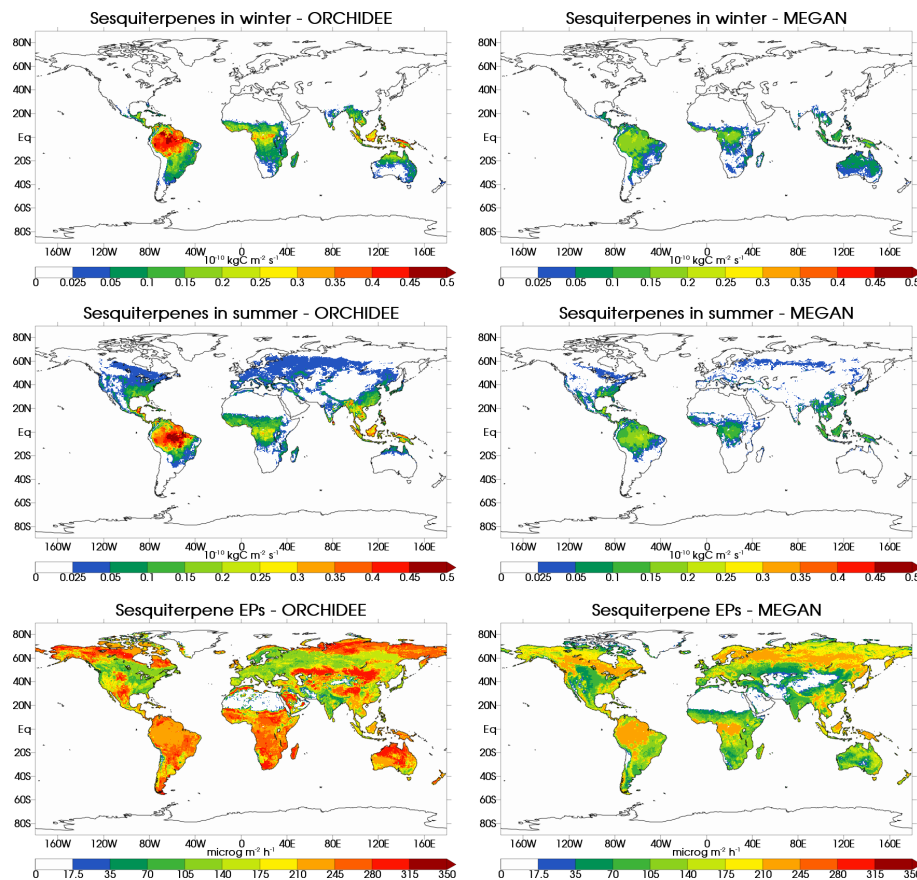
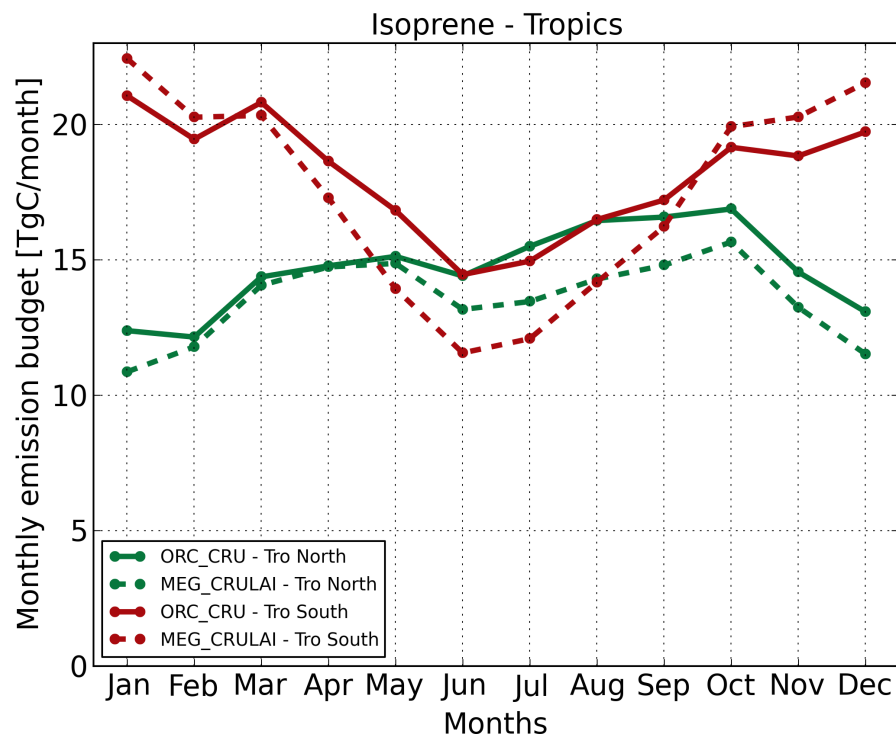


Figure 9. The same as Fig. 5, but for sesquiterpenes.

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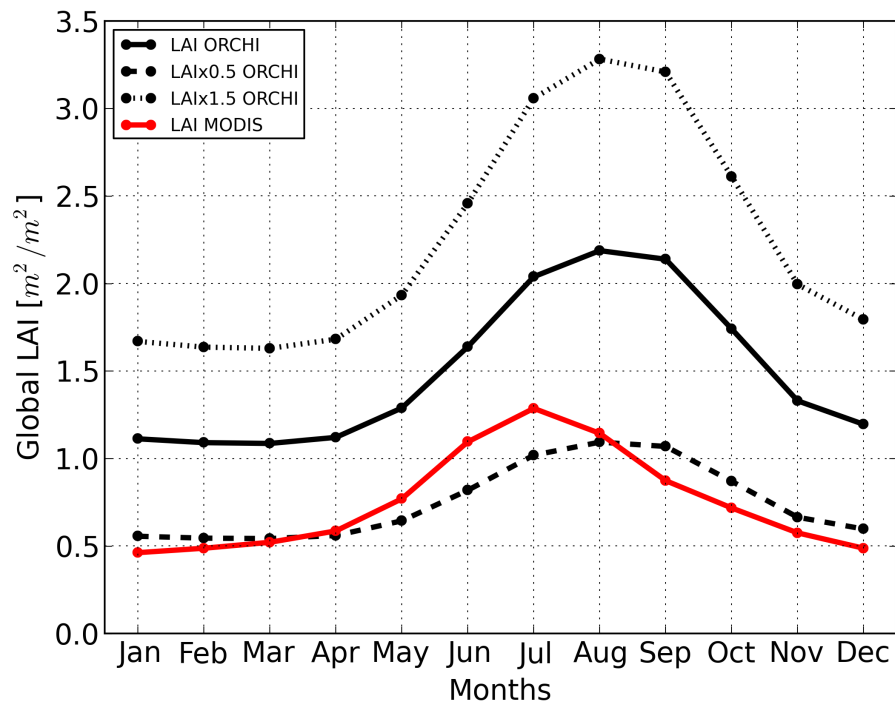
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**Figure 10.** Zonal mean of monthly emission budgets ( $\text{TgC month}^{-1}$ ), averaged over the simulation period (2000–2009) for the northern and southern tropics, in ORC\_CRU (solid line) and MEG\_CRULAI (dashed line) simulations for isoprene.

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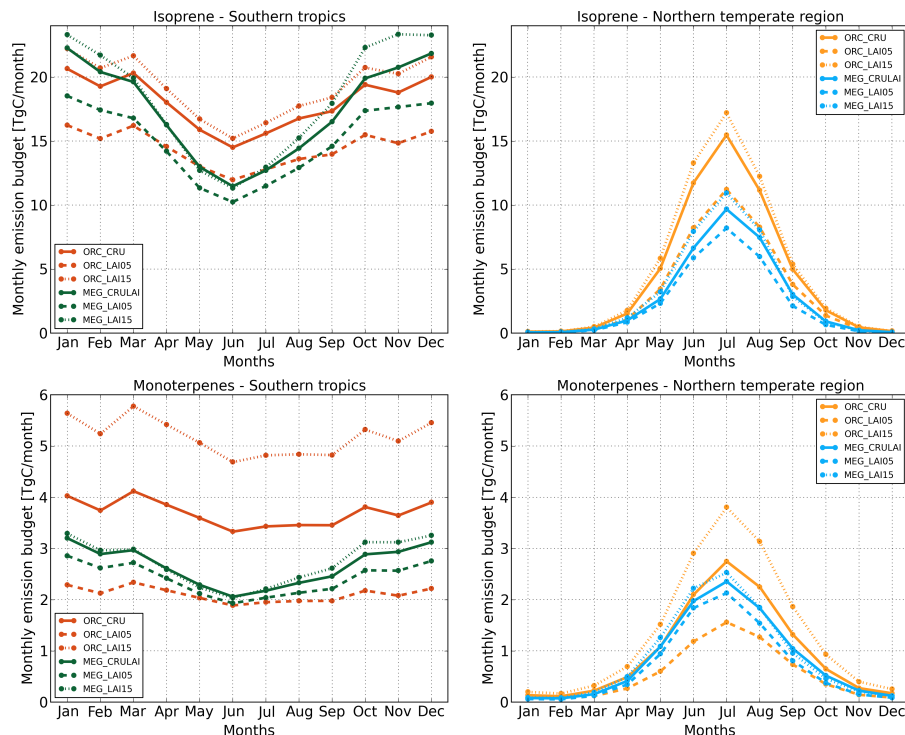
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**Figure 11.** Global monthly mean LAI ( $\text{m}^2 \text{m}^{-2}$ ) calculated by ORCHIDEE (solid black line) and retrieved from MODIS measurements (red line). The thick and thin dashed line represent the LAI from ORCHIDEE multiplied by a factor 0.5 and 1.5 respectively.

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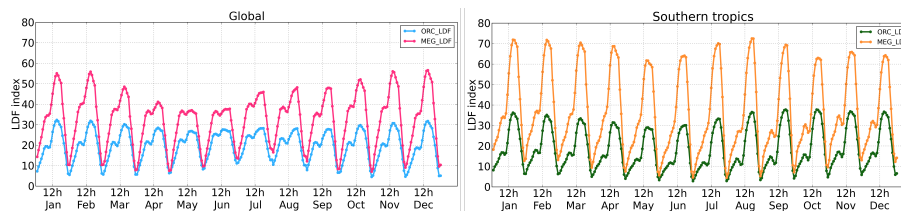
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**Figure 12.** Zonal average of changed emissions in the different LAI sensitivity tests: ORC\_CRU and MEG\_CRULAI using ORCHIDEE LAI (solid line), ORC\_LAI05 and MEG\_LAI05 using ORCHIDEE LAI·0.5 (thin dashed line) and ORC\_LAI15 and MEG\_LAI15 using ORCHIDEE LAI·1.5 (thick dashed line) in the year 2006, for the southern tropical (left column) and northern temperate regions (right column) for isoprene and monoterpenes. Emissions are given in  $\text{TgCmonth}^{-1}$ .

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**Figure 13.** Global (left plot) and southern tropical (right plot) average of the LDF index for ORCHIDEE and MEGAN. The LDF index is provided as the hourly daily profile averaged over each month.