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 EcosystEm), that includes an extended list of biogenic emitted compounds, updated emission factors (EFs), a dependency on light for almost all compounds and a multi-layer radiation scheme. Over the 2000–2009 period, using this model, we estimate mean global emissions of 465 Tg C yr^{-1} for isoprene, 107.5 Tg C yr^{-1} for monoterpenes, 38 Tg C yr^{-1} for methanol, 25 Tg C yr^{-1} for acetone and 24 Tg C yr^{-1} for sesquiterpenes. The model results are compared to state-of-the-art emission budgets, showing that the ORCHIDEE emissions are within the range of published estimates. ORCHIDEE BVOC emissions are compared to the estimates of the Model of Emissions of Gases and Aerosols from Nature (MEGAN), which is largely used throughout the biogenic emissions and atmospheric chemistry community. Our results show that global emission budgets of the two models are, in general, in good agreement. ORCHIDEE emissions are 8% higher for isoprene, 8% lower for methanol, 17 % higher for acetone, 18% higher for monoterpenes and 39% higher for sesquiterpenes, compared to the MEGAN estimates. At the regional scale, the largest differences between ORCHIDEE and MEGAN are highlighted for isoprene in northern temperate regions, where ORCHIDEE emissions are higher by 21 Tg C yr^{-1}, and for monoterpenes, where they are higher by 10 and 18 Tg C yr^{-1} in northern and southern tropical regions compared to MEGAN. The geographical differences between the two models are mainly associated with different EF and Plant Functional Type (PFT) distributions, while differences in the seasonal cycle are mostly driven by differences in the Leaf Area Index (LAI). Sensitivity tests are carried out for both models to explore the response to key variables or parameters such as LAI and Light Dependent Fraction (LDF). The ORCHIDEE and MEGAN emissions are differently affected by LAI changes, with a response highly sensitive to the considered compound. LAI is scaled by a factor of 0.5 and 1.5, changing the isoprene global emission by −21% and +8% for ORCHIDEE and −15% and +7% for MEGAN, and affecting the global emissions of monoterpenes by −43% and +40% for ORCHIDEE and −11% and +3% for MEGAN. We find that MEGAN is more sensitive to variation in the LDF parameter than ORCHIDEE. Our results highlight the importance and the need to further explore the BVOC emission estimate variability and the potential for 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 (Laothawornkitkul et al, 2009; Guenther et al., 2012; Peñuelas and Staudt 2014). On the global scale, the ecosystem contribution to VOC emissions is significantly higher than the anthropogenic one, and accounts for 75–90% of the total emission (Guenther et al. 1995; Lamarque et al., 2010). BVOCs play a central role in atmospheric chemistry, influencing the oxidative capacity of the atmosphere (Arneth et al., 2011; Taraborrelli et al., 2012), 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). Additionally, BVOCs and their oxidation products lead to the formation and growth of more than 50% of the Secondary Organic Aerosols (SOA) (Kanakidou et al., 2005; Goldstein and Galbally, 2007; Van Donkelaar et al., 2007; Engelhart et al., 2008; Hallquist et al., 2009; Acosta Navarro, et al., 2014; Tsigaridis et al., 2014). Under appropriate atmospheric conditions, BVOCs can contribute to a significant fraction of particles that evolve into cloud condensation nuclei (CCN) (Riipinen et al., 2012), even enhancing the droplet number concentration in clouds (Topping et al., 2013).

Despite numerous measurements and the progressive understanding of the processes underlying their production, BVOC emission estimates are still highly uncertain and vary significantly (Steiner and Goldstein, 2007; Arneth 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, which allows BVOC emissions to be indirectly derived(Palmer et al., 2006; Barkley et al., 2013), and a bottom-up approach. The latter approach is the most widely used method for local-, regional- or global-scale studies and can be divided into two main categories:

(i) an empirical method, based essentially on Guenther et al. (1995),where the response of leaf emissions to environmental changes is modelled using algorithms combined in a multiplicative way (Guenther et al., 2006, 2012; Lathière et al., 2006, 2010; Steinbrecher et al., 2009; Oderbolz et al., 2013). Hereafter we refer to it simply as the *Guenther formulation*);

(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 strong temporal and geographical variations. A critical point is the lack of information available at the global scale related to the various biomes, that would otherwise allow a more accurate representation of the geographical distribution and of the seasonal variation of BVOC emissions (Peñuelas and Staudt, 2010). The basal EF for instance, defined as the emission at the leaf level under standardized environmental conditions of temperature and solar radiation (Guenther et al., 1995; Steinbrecher et al., 2009), shows large variability from one plant species to another. Nowadays, a large number of measurements is available for different plants and at various sites and there is an increasing number of field campaigns that 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, allowing EF estimates for a wider range of BVOCs, despite the limitations which we will discuss in Sect. 2.2.1. To calculate BVOC emissions, a single EF is usually assigned for each PFT, where one PFT represents a group of plants having the same phylogenetic, phenological and physical characteristics (Prentice et al., 1992). The choice of one single value for each PFT is especially difficult, as each PFT actually corresponds to several plant species, and EFs show, in general, a wide range of values among 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 that are difficult to isolate and linked to plant 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 for a single plant. In addition, the link between EF variation and plant phenology is in general not taken into account, or is roughly described, especially in models that adopt the empirical approach.

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 done for isoprene, with the remaining fraction dependent only on temperature. The Guenther et al. (2012) approach considers only one value per emitted compound, whilst it has been shown the LDF also depends on the plant species. For example, measurements of the diurnal cycle for monoterpenes above Amazonian rainforest (Rinne et al. 2002; Kuhn et al., 2002) suggest that emissions are dependent on both light and temperature, whilst the role of light in influencing monoterpene emissions from boreal Scot pine forest is less clear (Taipale et al., 2011). Moreover, Staudt and Seufert (1995) and Loreto et al. (1996) show that monoterpene emissions from coniferous trees are principally influenced by the temperature, while those from Holm oak are predominantly controlled by a light-dependent mechanism. Owen et al. (2002) find that, in the Mediterranean region, emissions of all compounds from Quercus sp. are light dependent, the ocimene emitted by Pinus pinea is strongly correlated to light and an apparent weak light dependency is exhibited by monoterpene emissions from Cistus incanus. Ghirardo et al. (2010) provide the fraction of light-dependent monoterpene emission, being 58% for Scots pine, 33.5% for Norway spruce, 9.8% for European larch, and 98–100% for both Silver birch and Holm oak. Shao et al. (2001) and Steinbrecher et al. (1999) attribute for Scots pine a value of 20–30% and 25–37%, respectively. Nevertheless, there is no general agreement on the exact value of the temperature- and light-dependent fraction to assign for individual compound and PFT, as it appears also from the works mentioned right above.

Another crucial component in the estimation of BVOC emissions is the LAI, which 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 are found between the LAI estimated by measurements and the LAI calculated by models, with discrepancies of up to 100% at the global scale and 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 affects the predicted regional and seasonal distribution of BVOC emissions.

According to our knowledge, most papers investigating 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 between the land-cover inventories, and of the uncertainties in the classification of land cover. 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%. Ashworth et al. (2010) investigated the impact of varying the climate forcing temporal resolution of 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. Keenan et al. (2011) investigate the effect of canopy structure using different canopy models and they conclude that larger differences in the final emissions can be attributed to the use of different canopy models, rather than different emission model approaches. Nevertheless, very few studies have 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) in which several simulations were performed with the MEGAN model to assess the sensitivity of isoprene emissions to many parameters and processes such as LAI, emission factors (EFs), CO_2 concentration, soil moisture, and the radiation scheme. The sensitivity simulations performed showed a variation in isoprene emissions of up to 50% at the global scale.

In the present work, our objectives are to (i) present the updated version of the emission module embedded in the dynamic global vegetation model ORCHIDEE, (ii) 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 the new emission scheme, (iii) compare the ORCHIDEE results to the widely used emission model MEGAN, putting the two models under the same forcing conditions, but retaining their particular characteristics (see Sect. 2.5), in particular the emission scheme, classes and distribution of PFTs and LAI processing, and (iv) explore, at the global and regional scales, the BVOC emission sensitivity to EFs, LAI and LDF in ORCHIDEE and MEGAN, and to understand the reasons behind these discrepancies. ORCHIDEE is designed to provide past, present and future scenarios of emissions from vegetation, studying the links between climate, the plant phenology and emissions. It is therefore essential that the internal variability, weaknesses and inaccuracies of the emission module are extensively investigated. The proper way to assess the correctness of a model is to evaluate it against observations, as is done, for example, for organic aerosol by Mann et al. 2014) and Tsigaridis et al. (2014) and for tropical mountain forest carbon store by Spracklen and Righelato (2014). The evaluation of BVOC emission models against observations has already been carried out at a local and regional scale (i.e. Karl et al., 2007; Kunl et al., 2007; Lathière et al., 2009; Smolander et al., 2014), demonstrating a good performance of the Guenther formulation. Nevertheless, given the ecosystem biodiversity, the huge variability of the parameters involved and the poor spatial and temporal coverage of BVOC emission observations, it is extremely difficult to infer any evaluation at global scale from these tests. In such a context we can rely on model inter-comparison and sensitivity tests in order to assess the limitations and uncertainties of BVOC emission estimates, to relate them to particular key parameters/variables and to investigate their origin. 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 conclusions and future directionsare provided in Sect. 4 and 5.

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 (Krinner et al., 2005; Magnan et al., 2011) 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 biosphere-atmosphère, in English: mapping of hydrological exchange at the biosphere/atmosphere interface).

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 layer and a deep layer (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 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). For our study, the global vegetation distribution is prescribed for all runs using appropriate forcings, as described in paragraph 2.4.

2.2 BVOCs in ORCHIDEE: module improvements

The BVOC module is extensively updated, considering recent findings regarding emission schemes and field measurements. The new BVOC emission scheme is a development of the module implemented in ORCHIDEE by Lathière 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 Spitter et al. (1986a, b) and the one already used in ORCHIDEE for the calculation of photosynthesis. The canopy is considered split *vertically* into several LAI layers, the number of which (up to 17) depends on the LAI value. Emissions are calculated for each layer through consideration of the sunlit and shaded leaf fractions and the light extinction and light diffusion through canopy. In a second step they are 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):

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

where $LAI_i(l)$ is the leaf area index expressed in $m^2\ m^{-2}$ at a particular LAI layer and PFT, SLW_i is the specific PFT leaf weight in g m^{-2}, $EF_{c,i}$ is the basal emissions at the leaf level for an individual compound and PFT at standard conditions of temperature (T = 303.15 K) and photosynthetically active radiation (PAR = 1000 μmol $m^{-2}\ s^{-1}$), expressed in μgC $g^{-1}\ h^{-1}$. CTL_c is the emission activity factor, depending on the 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 is considered for isoprene and methanol. The total emission per grid cell is obtained by summing $F_{c,i}(l)$ over the layer *l* and averaging the emission contribution of each individual PFT, weighted by PFT fractional land coverage. Further details on the original version of the emission module are given in Lathière et al. (2006).

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

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

We mentioned that, the emission module has also been modified to include a light dependency for almost all compounds emitted. In the previous module version, indeed, isoprene was the only compound dependent on both light and temperature, while the others were only dependent on temperature. As detailed in Sect. 1, most recent field campaigns highlight, for a large number of plants, the dependency of monoterpenes, sesquiterpenes and oxygenated BVOC emissions on radiation as well. To adopt a detailed parameterisation is not yet possible, cause to data lacking at global scale. Therefore, in the new emission module we consider the approach described in Guenther et al. (2012), even if it is rather oversimplified. BVOCs are now modelled to consider both light-dependent and light-independent emission processes, and the response to temperature and light (CTL) is calculated for individual compounds at each LAI layer (l):

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

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

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

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

2.2.1 Emission Factor update

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 for re-examining thoroughly the emission factors used in the emission module and correcting them accordingly. Nevertheless the methodology to assess EFs is still under debate within the scientific community.

Assigning EFs, especially on the global scale, is very tricky. In the ideal case, for each compound emitted, we should consider the EFs of all plants belonging to one particular PFT and the land cover of each plant. We could then, for each PFT and compound, make averages weighted on plant land cover, thus obtaining an average EF for each PFT and emitted compound. Unfortunately, there are not yet enough observations available to use such a methodology.

There are several factors that make it difficult to find a good strategy to assign EFs valid for all compounds:

1. depending on the compound and the PFT, the number of measurements available differs considerably, and the statistical accuracy of the EFs may therefore be very variable;

2. in some cases, the most recent measurements contradict the older ones, therefore it is reasonable to consider only the most recent data. However, in other cases the difference between recent and older measurements is not so clear, therefore it is not easy to understand if it is better to consider less recent measurements in the evaluation of EFs;

3. considering the values of EFs that we collected from the literature, we note that they are actually often related to a small number of plant species from mostly the same measurement sites. The values found could not be considered as a significant representative set for the PTFs at the global scale;

4. in many papers focussing on modelling, the EFs presented are either taken directly from previous models, or are based on a review or on measurements available. In this context, it is very difficult to make consistent averages and understand which values found should be taken into account.

Taking all this into account we decided to proceed as follows.

As general rule, and based on an extensive review of publications, we select papers, in which it is possible to convert the EFs into the units and at the standard conditions that are considered in ORCHIDEE (PAR = 1000 μmol m^{-2} s^{-1}, temperature = 30 °C). We do not always perform an average over all values collected, but we use a qualitative and comparative method to justify the EFs.

In the case of isoprene, we principally consider the most recent papers, the ones that present new measurements or original review. The review carried out for EFs confirms that the values used in the previous version (Lathière et al., 2006) are consistent with the latest measurements. Only for certain PFTs it is necessary to change the value of EF. Indeed, isoprene has already been widely measured for several years, while other BVOCs have been documented only more recently.

In the case of the other compounds, since there are fewer papers and the information is not so well consolidated, we adopt a similar strategy but we are less restrictive in paper choice. In general, we perform averages considering the different values from all papers collected, and we compare these averages to the older values in ORCHIDEE. Whenever big differences between the new value and the old one were found, we look in detail at the various papers to see if there are some outliers, and if so, we do not consider them in the EF evaluation.

Table 3 show the new and old EFs used in the emission module and Table 4 presents EF values for each speciated monoterpene as a percentage of the bulk monoterpene EF value. As shown in Table 3, the revision leads to the modification of almost all EFs. 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 μgC g^{-1} h^{-1} in the old version and EF = 0.5 μgC g^{-1} h^{-1} in the new one). We based the choice on papers focussing on reviewed or measured EFs, such as Guenther et al. (2006) (EF = 1.44 μgC g^{-1} h^{-1}), Guenther et al. (2012) (0.002 μgC g^{-1} h^{-1}), Steinbrecher et al. (2009) (EF = 0.44 μgC g^{-1} h^{-1}), and Smiatek and Steinbrecher (2006) (EF = 0.09 μgC g^{-1} h^{-1}) and Klinger et al. (2002) (EF = 0.52 μgC g^{-1} h^{-1}). All these values are much lower than those assigned by Lathière et al. (2006), and their average is 0.5 μgC g^{-1} h^{-1}, which we set as the new value. In this case, we do not consider the other papers where EFs are directly taken from previous models or for which the source of information was not clear. Our choice is confirmed by Ruuskanen et al. (2007), who assign a contribution of less than 3% of the VOC emission to isoprene, 2-methyl-3-buten-2-ol (hereafter we refer to it simply as MBO) and 1,8-cineole, for larch, which is the major component of boreal needleleaf deciduous PFT.

Furthermore, we now consider boreal broadleaved deciduous trees to be a higher emitter of isoprene than in the previous model version (now EF = 18 μgC g^{-1} h^{-1}, while before EF = 8 μgC g^{-1} h^{-1}), since most of the papers collected propose particularly high values, such as Levis et al. (2003) (24 μgC g^{-1} h^{-1}), Arneth et al. (2011) (45 μgC g^{-1} h^{-1}), Guenther et al. (2006) (42.3 μgC g^{-1} h^{-1}) and Guenther et al. (2012) (22.7 μgC g^{-1} h^{-1}). For monoterpenes, a significantly higher EF (from 0.8 μgC g^{-1} h^{-1} to 2.2 μgC g^{-1} h^{-1}) is now assigned to tropical broadleaf evergreen and deciduous PFTs. For MBO the EF for the temperate needleleaf evergreen PFT is reduced from 20 μgC g^{-1} h^{-1} to 1.4 μgC g^{-1} h^{-1} (Tarvainen et al., 2005; Hakola et al., 2006; Chang et al., 2009; Kim et al., 2010).

Our review analysis confirms a large variability in EFs, even among plants that are usually represented by one single PFT in global vegetation models (characterized by the same physiognomy, leaf shapes and photosynthesis type). It is therefore a source of high uncertainty to 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 by adjusting the measured flux at leaf level at a standard conditions of light photosynthetically active radiation (PAR) and temperature, using algorithms such as 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).

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 terrestrial vegetation. The basis of the model is a simple mechanistic approach established by Guenther et al. (1991, 1993, 1995), which links 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, and 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 components defined by the light dependent fraction (LDF). For a 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 the 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 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 simply referred to as MEGAN.

MEGAN estimates emissions of 19 chemical compound classes, which are then redistributed 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 definition of PFTs and the emission factors assigned to them. The vegetation distribution is described with fractional coverage of 16 PFT classes consistent with those of the 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 applied 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 the 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 group of 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. two 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. one simulation for the 2000–2009 period with MEGAN using the LAI estimated by ORCHIDEE (MEG_CRULAI)

3. four 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. two 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 ORCHDEE in the 10 years of simulation.

All simulations are performed at the global scale with a spatial resolution of 0.5° × 0.5°. We use the CRU-NCEP v5.2 meteorological forcing database (http://dods.extra.cea.fr/data/p529viov/cruncep) providing temperature, pressure, humidity, wind speed and shortwave solar radiation. This forcing is based on the 6-hourly 2.5° 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 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 (Hurtt et al. 2006). The database can be found in http://dods.extra.cea.fr/work/p86ipsl/IGCM/BC/SFR/OL2/PFTmap_1850to2005_AR5_LUHa

.rc2.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.

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).

In ORCHIDEE, the activity factor (L_c) is kept as in Lathière et al. (2006), considering four leaf age classes (new, young, mature and old leaves). For methanol, L_c is equal to 1 for new and young leaves and equal to 0.5 for mature and old leaves, while for isoprene, L_c is equal to 0.5 for new and old leaves and equal to1.5 for young and mature leaves. In MEGAN, the L_c values are taken from Table 4 in Guenther et al. (2012); in particular, for isoprene, L_c is equal 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 age classes. For both models, no soil moisture activity factor is taken into account. The annual CO_2 concentration varies along the simulation from a value of 368 ppm in 2000 to 385 ppm in 2009. In ORCHIDEE, the variation of CO_2 concentration can indirectly impact on the BVOC emission as it affects leaf growth, while in MEGAN, a CO_2 inhibition factor on isoprene emission based on Heald et al. (2009) is activated. As the CO_2 variation in this 10-year simulation is low, the inhibition effect is considered insignificant (Sinderalova et al. 2014) in this context. For ORCHIDEE, LDF and the β coefficient values are given in Table 2. For MEGAN, the values of LDF and β are those presented in Table 4 in Guenther et al. (2012).

2.5 Differences between ORCHIDEE and MEGAN emission algorithms

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

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

direct light is around twice that for diffuse light. In ORCHIDEE the CTLD parameters are fixed and are the same for diffuse and direct radiation;

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

3) the PFTs classes and their distribution are not the same in the two models (Table 1) and they are not interchangeable without significantly modifying the models;

4) LAI is considered in a different way in the two models. ORCHIDEE calculates the LAI at each model time step for each PFT and grid cell, taking into account a full plant phenology scheme. MEGAN, on the other hand, does not compute the LAI, rather, it has to be provided as an input averaged over the vegetated part of the grid cell;

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

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

7) in MEGAN, leaf age classes are derived from consideration of the variation between the LAI value of the current and preceding month, following a highly parameterised scheme. In

ORCHIDEE, leaf age classes are calculated on-line considering the plant leaf growth and leaf turnover at each model time step (30 minutes);

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

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

3 Results

3.1 Global budgets

As already discussed at the end of the introduction, the validation of BVOC emissions at the global scale is a complex issue because of the poor data coverage in many regions and the general lack of year-round measurements. 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 difficulties in the retrieval of the atmospheric concentration of short-lived compounds from space or in separation of the different sources (for instance terrestrial biogenic, anthropogenic, oceanic etc.) and the various compounds themselves. Global emission estimates are generally performed using models, or from the application of 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; Stavrakou 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; Stavrakou et al., 2011; Barkley et al., 2013; Bauwens et al., 2013; Stavrakou et al., 2014).

At the global scale, the main way to evaluate the results obtained in the present study is to compare them with the most recent emission budgets derived either from other model runs or from the inversion of satellite data. We have compared emissions 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) (Lathière et al., 2006), are particularly high, as already pointed out by Sindelarova et al. (2014).Methanol (106.1 Tg C yr^{-1}) and acetaldehyde (42.2 Tg C yr^{-1}) emissions are twice as large, and formaldehyde emissions (10.0 Tg C yr^{-1}) are up to 5 times greater than the other estimates. The results of the new module version (ORC_CRU, green stars) are more in the range of other published estimates. Although the MEG_CRU simulation was carried out using the same MEGAN version as in Guenther et al. (2012) (blue hexagons, 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 (e.g. 2000). Using reanalysis provided by Qian et al. (2006) as climate forcings for the year 2000, Guenther et al. (2012) report BVOC emissions of 472 Tg C yr^{-1} for isoprene, 124 Tg C yr^{-1} for monoterpenes (considering the speciated monoterpenes accounted in this work) and 11.5 Tg C yr^{-1} for acetaldehyde. Our MEG_CRU simulation estimates for 2000 are 410 Tg C yr^{-1}, 72 Tg C yr^{-1}, and 8.3 Tg C yr^{-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 the emission estimates (green triangles, first plot of Fig. 1).

Table 6 shows the annual emissions calculated by ORCHIDEE and MEGAN (ORC_CRU and MEG_CRU simulations) at the global scale and for the northern (lat: 0–30N) and southern (lat: 30S–0) tropics, the northern (lat: 30N–60N) and southern (lat: 30S–60S) temperate latitudes, and the northern boreal (lat: 60N–90N) regions, averaged over the 2000–2009 period. At the global scale, the two models are in a good agreement. Isoprene is the main compound emitted with a global amount of 465 Tg C yr^{-1} for ORCHIDEE, accounting for 61% of total BVOC emissions (estimated to 757 Tg C yr^{-1}), and 428 Tg C yr^{-1} for MEGAN, accounting for 64% of total BVOCs (estimated at 666 Tg C yr^{-1}). The following most abundant compounds are monoterpenes, accounting for 12% of the 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 each represent 1% to 4% of the total BVOCs for both models, while other compounds contribute to 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 α-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 compound (except MBO). Both models calculate the 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 a few plant types such as 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 significant only for the PFTs representing these plants (TeNeEv and BoNeEv), leading to notable emissions in the temperate North latitudes and contributing 88% for ORCHIDEE and 63% for MEGAN of the global MBO emission.

At the regional scale, the largest differences between ORCHIDEE and MEGAN in terms of absolute values appear in the northern temperate regions for isoprene, where emissions are 21 Tg C yr^{-1} higher in ORCHIDEE. Indeed, the marked seasonal cycle of emissions for northern temperate latitudes implies that the largest 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 row). In particular, in northern temperate region the highest discrepancies are mainly due to the different PFT surface coverage for grass and crop and the higher EFs values in ORCHIDDE in comparison to MEGAN. Actually, in ORCHIDEE C3Gr covers the 42% of vegetated surface with an EF = 12 μgC g^{-1} h^{-1}, C3Ag covers the 18% with an EF = 5 μgC g^{-1} h^{-1}, while in MEGAN the C3GrCool occupies the 20% with an EF = 2 μgC g^{-1} h^{-1}, C3GrCold the 6% with an EF = 4 μgC g^{-1} h^{-1}, C3GrCool the 20% with an EF = 2 μgC g^{-1} h^{-1} and Crop the 23.2% with an EF = 0.12 μgC g^{-1} h^{-1}. This example raises an important issue. Considering the EF assigned to C3Gr, we lowered its value with respect to the previous version, from 16 to 12 μgC g^{-1} h^{-1}. These is a compromise value, chosen so that we do not excessively bias the emissions in other areas. C3Gr is, indeed, strongly present in other regions: 13% of northern tropical areas, 22% of southern tropical areas and 32% of the total vegetation surface. A more detailed description of the different crop and grass (in other words with a larger number of PFTs) could lead to more accurate results. The same consideration could be done for almost all the other PFTs.

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

3.2 Inter-annual and inter-seasonal emission variations

Fig. 2 shows 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 large 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 are, on average, lower by 11.5 Tg C month^{-1} and 9.0 Tg C month^{-1} compared with ORC_CRU. The monthly zonal average for tropical, temperate and northern boreal latitudes regions are shown in Fig. 3. We observe, as mentioned in Sect. 3.1, that the ORCHIDEE emissions are significantly higher in northern temperate regions compared with MEGAN, with a marked seasonal cycle and the largest differences between the two models occurring in summer. In July (August) in particular, calculated isoprene emissions in ORC_CRU are about 4 Tg C month^{-1} (5.5 Tg C 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 Tg C month^{-1} (5 Tg C month^{-1}) in comparison to MEGAN in July (August), (Fig. 3, first plot, left column).

MEGAN isoprene emissions are indeed dominant from the tropical regions, leading to an overall stable global emission budget throughout the year (Fig. 2). The 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 for 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 strong emitters will have a more marked seasonal cycle (Fig. 2), such as for methanol and isoprene in 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 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 extensively 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.

Additionally, Fig. 3 shows that 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.

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 with the ecosystem present in the grid cell.

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

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

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

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

where i is the index related to PFTs, M_{Carbon} and M are the molar mass of carbon and the compound, respectively, LAI_{REF} equals to 5.0 m^2 m^{-2} is the LAI in MEGAN standard conditions, SWL is the MEGAN specific leaf weight depending on PFTs, A is the PFT grid fraction and C_{CE} is the canopy environment coefficient, a scaling factor dependent on the canopy radiation module, which equals 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) ranging between: 5.0–12.0 10^{10} kg C m^{-2} s^{-1} (3.0–9.0 10^{10} kg C m^{-2} s^{-1}) for isoprene, 0.8–2.0 10^{10} kg C m^{-2} s^{-1} (0.6–1.3 10^{10} kg C m^{-2} s^{-1}) for monoterpenes, 0.3–1.2 10^{10} kg C m^{-2} s^{-1} (0.2–0.7 10^{10} kg C m^{-2} s^{-1}) for methanol, 0.2–0.5 10^{10} kg C m^{-2} s^{-1} (0.1–0.3 10^{10} kg C m^{-2} s^{-1}) for acetone and 0.4–0.6 10^{10} kg C m^{-2} s^{-1} (0.2–0.3 10^{10} kg C m^{-2} s^{-1}) for sesquiterpenes, respectively. For methanol, in summer, high emitting areas also appear in Europe and Russia, with values of 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. Indeed, these regions are populated by temperate and boreal needleleaf evergreen trees, which are strong methanol emitters (Table 3 and Fig. 7, last row).

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

Other notable differences between the two models appear in South America for isoprene, directly in relation with the EP distribution. The pattern of isoprene emissionin MEGAN has higher values in western Brazil, Bolivia and northern Argentina, while in ORCHIDEE the values are more homogeneous, with higher emissions in central Brazil. The same pattern differences are detected in the emission potential (Fig. 5, last row on the right), and we therefore infer that the EP distribution drives the isoprene emission geographical distribution. The same conclusion also holds 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 PFT distributions.

3.4 BVOC emission sensitivity to LAI

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

Figs 4 and 10 show large differences in the geographical distribution and global average of ORCHIDEE LAI and MODIS LAI (Yuan et al., 2011). As illustrated in Fig. 10, the global monthly mean LAI calculated by ORCHIDEE is 1.5–2 m^2 m^{-2} higher compared to the LAI used in MEGAN and based on MODIS data-sets. In addition the LAI peaks at different times throughout the year in ORCHIDEE and MEGAN. We investigate the contribution of different areas and we observe that, whilst in northern temperate region the MODIS LAI peaks in July and afterwards decreases quite fast, the ORCHIDEE LAI peak in both July and August. Furthermore, in the boreal region, the ORCHIDEE LAI peaks one month later (August) than the MODIS LAI (July). Therefore, the time shift observed globally is due to the greater persistence of the growing season provided by ORCHIDEE in the northern temperate area and its delay in the northern boreal region compared with what is detected by MODIS.

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

The differences between these LAI estimates are significant, but our current state of knowledge does not allow us to say which estimate is correct. Field and satellite data bring very useful and complementary information regarding the order of magnitude, the seasonal and the geographical variability of LAI. Nevertheless, inferring values for LAI on small or large regional scales is particularly challenging, and data available from, either field or satellite measurements also have significant uncertainties. Satellites, for instance, do not measure the *real* LAI, but the *effective* LAI obtained from indirect optical methods and strongly determined by the *a priori* assumptions necessary for 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).

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

Conversely, in a validation study using satellite-derived vegetation index time series, Maignan et al. (2011) pointed out some weaknesses in the ability of ORCHIDEE to correctly model the LAI, especially in the equatorial forest (Amazonia, central Africa, Indonesia) where a poor correlation of model output with satellite data was demonstrated. In general, quite large and comparable incertitude is found when different LAI databases are compared. Krinner et al. (2005) found that the difference between ORCHIDEE and MODIS satellite LAI (Myneni et al., 2002) is as much as the difference between the satellite data that they used and an alternative satellite vegetation cover data set (Tucker et al., 2001). Therefore given the many existing limitations, we cannot conclude which LAI estimate is more reliable (LAI obtained from MODIS satellite or calculated by ORCHIDEE). It is likely that the ORCHIDEE LAI could be improved and a possible component to be upgraded is the allocation of the different carbon stocks, but further investigations are needed. Performing a robust evaluation of the model's ability to simulate the LAI, especially at the global scale, still remains challenging, and is beyond the scope of our study.

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

3.4.1 LAI seasonal cycle impact

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

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

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.

3.4.2 LAI size impact

The global and zonal emission budgets (Table 7) in the MEG_CRULAI simulation are not significantly different than those determined in MEG_CRU, even if the ORCHIDEE LAI is significantly higher than MODIS LAI, suggesting a low sensitivity of MEGAN to LAI size

Indeed, at the regional scale, in boreal and temperate regions, the MEG_CRULAI emissions are slightly higher than those in MEG_CRU, 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 is to the strengthened effect of leaf self-shading caused by an increase in LAI in locations characterized by a dense vegetation (e.g. in central Africa or Amazonia). 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 in hourly leaf temperature between sunlit and shaded leaves in the case of dense vegetation (TrBrEv and TrBrDe), we estimate differences of about 1–1.5 °C, which would unlikely be high enough to explain such differences in emissions. Lathière et al. (2006), for instance, found that an increase in the global surface temperature by 1°C leads to an increase of isoprene emissions of at most 11%. We therefore doubt that the only mechanism behind the static BVOC emissions for light-independent compounds is leaf self-shading.

We, therefore, investigate in more detail whether models show the same response to a particular change in LAI. We perform two extra simulations for each model, using the ORCHIDEE LAI multiplied by a factor of 0.5 or 1.5. The scaling factor considered are consistent with the LAI uncertainties (see the begging of Sect. 3.4).Fig. 12 shows the four simulations: MEGLAI05, ORC_LAI05 (ORCHIDEE LAI multiplied by 0.5) and MEG_LAI15 and ORC_LAI15 (ORCHIDEE LAI multiplied by 1.5), for the year 2006 (details in Table 5). Only the zonal average for the tropics and southern and northern temperate areas, for isoprene and monoterpenes are displayed, but they are also representative of 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 isoprene is a light-dependent compound thus, beyond a given LAI threshold, the contribution of the highest LAI layers is very low , as there is no more or very little direct light available. We observe that MEGAN is less sensitive than ORCHIDEE to an LAI increase, which is likely due to the different parameterisation of CTLD factor in the two models as described Sect. 2.5. In more details, as LAI increases, the growth of sunlit leaves fraction is dumped by an exponential factor as in Spitter et al. (1986b), implying lower contribution of sunlit leaves with respect to shaded leaves for high LAI values. In MEGAN, for equal incoming radiation, the relative contribution of sunlit leaves, with respect to shaded leaves, is roughly twice than in ORCHIDEE. This explains the different sensitivity of the two models.

Monoterpene emissions show a different response in terms of sensitivity to LAI. In the southern tropics, the relative difference in 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, the relative difference in the 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 smaller emission impact of LAI variation on emissions in MEGAN, even for compounds not fully dependent on light, such as monoterpenes (LDF=0.6).

Table 8 shows the total emission budget calculated for MEG_LAI05, ORC_LAI05, MEG_LAI15 and ORC_LAI15 simulations for every compound. In general in ORCHIDEE, the lower the light dependence, the higher the sensitivity to LAI, 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 for light independent emissions in the two models. Such differences are detailed in point 6 of Sect. 2.5. In particular, in ORCHIDEE, the light independent emission linearly depends on LAI whereas, in MEGAN it is determined by the γ_{LAI} factor and it varies almost linearly for low LAI (< 2 $m^2 m^{-2}$) and then more and more slowly up to become almost constant for LAI higher than 5 $m^2 m^{-2}$. 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 m^2 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, the 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 to a degree that depends on the 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). Since the results of these studies are highly heterogeneous, assigning a single LDF value to each compound is as difficult as assigning the EFs to each PFT (Sect. 2.2). Hence, the LDF uncertainty could be even higher than the uncertainties associated with EFs, as there have been fewer less quantitative studies on this subject published to date.

The objective of this section 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 estimates, giving clues to better understand the different behaviours of the two models.

For the fully light dependent (isoprene: LDF=1) or largely light dependent compounds (methanol: LDF=0.8) (Fig. 5 and Fig. 7), we observe that a higher EP in ORCHIDEE than in MEGAN does 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 a different emissions response to light. Indeed, this effect is less important for compounds which are less dependent on light such as monoterpenes (LDF = 0.5) (Fig. 6) and sesquiterpenes (LDF = 0.6) (Fig. 9), and indeed are even negligible for acetone (LDF = 0.2) (Fig. 8). It therefore seems that the choice of LDF parameter can be crucial in the emission estimate and in the sensitivity to EF variation.

To isolate the signal related to the LDF, we investigate the hourly variation of two "test compounds", the first defined as light independent (LDF = 0) and the second defined as totally light-dependent (LDF = 1). All EFs are set to 1 μgC g^{-1} h^{-1} for each PFT. 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 compounds 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, which 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 with MEGAN is much higher (up to 20%), than the index associated with ORCHIDEE. At the regional scale, in the southern tropics for example (second panel) the index reaches up to 70% and is the twice as large the index calculated for ORCHIDEE. The light-dependent part of the emissions in MEGAN is therefore more important than ORCHIDEE, with important impacts on emission estimates. Firstly, we show that based on the same EF value, the MEGAN emissions are higher than in ORCHIDEE for compounds associated with high LDF, as expected from Sect. 3.3.

Secondly, the variable orcldf0 (megldf0) represents the emissions when LDF is zero while orcldf1 (megldf1) represents the emissions when LDF is one; thus, they define the interval spanned by emissions as LDF varies. Therefore, a low LDF index is associated with a greater variability of emissions for equal light-independent emissions. Consequently, ORCHIDEE results more sensitive to LDF variation than MEGAN, as the ORCHIDEE LDF index is lower than the MEGAN index. Furthermore, the LDF index provides an evaluation of error due to a diverse choice of LDF values. The LDF index is always less than 100, meaning that the light-independent component of the emission is always bigger than the light-dependent part. Therefore, if LDF in the model is greater than it should be, emissions will be underestimated, while if it is less, emissions will be overestimated. At regional scale, tropical areas, that are associated to high LDF index, will be less sensitive to LDF variation than other regions.

4 Conclusions

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

The ORCHIDEE emission estimates are within the range of the published emission budgets. The ORCHIDEE global budgets averaged over the period investigated (2000–2009) are 465 Tg C yr^{-1} for isoprene, 108 Tg C yr^{-1} for monoterpenes, 38 Tg C yr^{-1} for methanol, 25 Tg C yr^{-1} for acetone and 24 Tg C yr^{-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, 17% higher for acetone, 18% higher for monoterpenes and 39% higher for sesquiterpenes compared to the MEGAN results. At the regional scale, the largest differences in terms 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.

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

The LAI calculated by ORCHIDEE is 1.5–2 m^2 m^{-2} higher than the LAI retrieved by MODIS. We examined what these discrepancies can impact on the BVOC estimates. Sensitivity tests are then performed forcing both models with the ORCHIDEE LAI multiplied by a factor of 0.5 and 1.5. ORCHIDEE and MEGAN emissions present a similar response to these LAI variations. Conversely, for monoterpenes, ORCHIDEE is much more sensitive to LAI variations, in comparison to MEGAN. These discrepancies are due to differences in the light-independent emission formulation between the two models. In ORCHIDEE the dependence of emissions on LAI is linear, while in MEGAN for LAI up to 2 m^2 m^{-2} is quasi-linear , then progressively reducing the increase up to become nearly constant for LAI greater than 5 m^2 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. We find that ORCHIDEE estimates are more sensitive to LDF variation than MEGAN. Moreover, we show that overestimation (underestimation) in LDF value leads to emission underestimation (overestimation).

5 Future directions

Model inter-comparison and sensitivity tests are extremely useful to define which parameters/variables mainly affect BVOC emissions, which is the cause of this sensitivity, and how estimates can be improved. Previous works have already investigated the impact of different experimental set-ups (climate forcing and vegetation distribution) (Arneth et al., 2011), differences in the canopy structure description (Keenan et al., 2011) and land cover classification (Oderbolz et al., 2013) on emissions.

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

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

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

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

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Table 1. Plant Functional Types in ORCHIDEE and MEGAN and corresponding occupied surfaces in 10^{12} 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	C3 Grass	C3 Grass Cold	37.00	4.20
	C3GrCool		C3 Grass Cool		12.55
C4Gr		C4 Grass		14.89	11.025
C3Ag	Crop	C3 Agriculture	Crop	10.19	14.58
C4Ag		C4 Agriculture		4.88	
-	TeSbEv	-	Temperate shrub evergreen	-	0.074
-	TeSbDe	-	Temperate shrub deciduous	-	5.39
-	BoSbD	-	Boreal shrub deciduous	-	8.02

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 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

Table 3. Emission Factors (EFs, µgC g^{-1} 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**	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; 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; Lathière et al., 2010; Leung et al., 2010; Arneth et al 2011; Fu and Liao., 2012; Oderbolz et al. 2013.
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**	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.
Sesqiterp.	- **0.450**	- **0.450**	- **0.130**	- **0.300**	- **0.360**	- **0.150**	- **0.300**	- **0.250**	- **0.600**	- **0.600**	- **0.080**	- **0.080**	Guenther et al., 2006, 2012; 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.
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; 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.
Acetone	0.290 **0.250**	0.290 **0.250**	0.870 **0.300**	0.430 **0.200**	0.290 **0.330**	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 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.
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; 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 **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; 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 **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; 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 **0.015**	0.010 **0.015**	0.030 **0.020**	0.015 **0.020**	0.010 **0.025**	0.030 **0.025**	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; 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 **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**	Baker et al., 1999; Schade and Goldstein, 2001; Tarvainen et al., 2005; Guenther et al., 2012; Hakola et al., 2006; Chang et al., 2009; Kim et al., 2010.

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 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;
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**	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;
t-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; 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.

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_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

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 global scale, for northern and southern tropics, northern and southern temperate areas and northern boreal regions.

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

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

Table 8. Annual emission budgets (Tg C yr^{-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	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

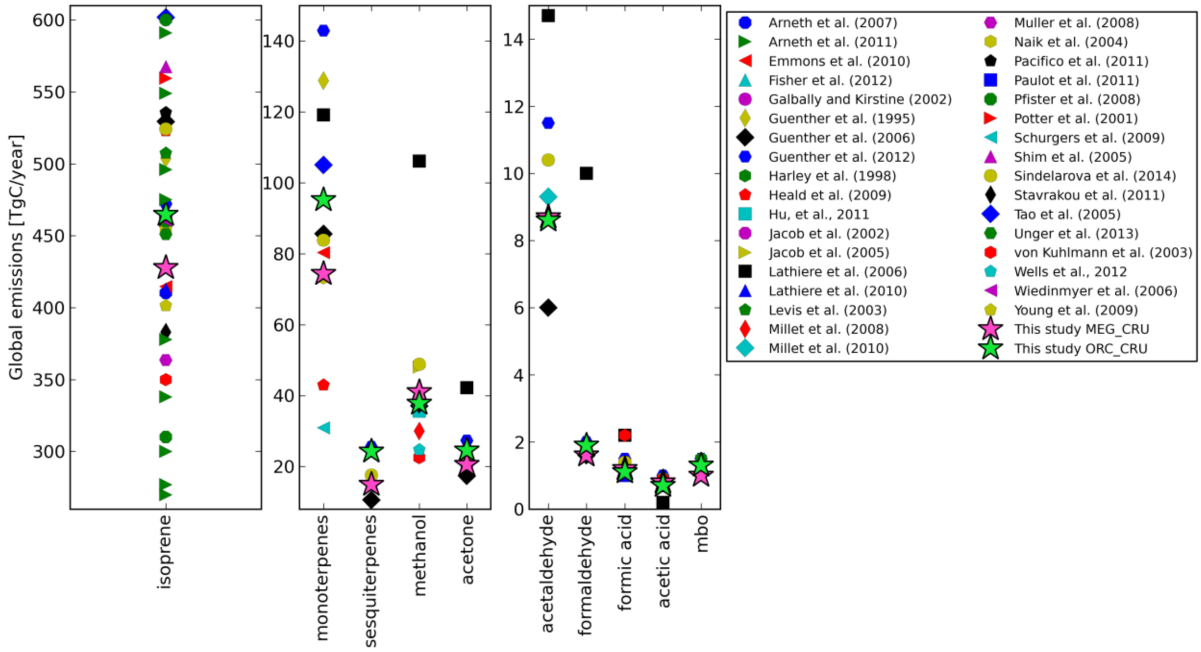

Figure 1. Global emission budgets (Tg C yr^{-1}) calculated by ORCHIDEE (ORC_CRU simulation, green stars) and MEGAN (MEG_CRU simulation, pink stars), compared with published estimates for the main BVOCs presented in this work. Note that the vertical axes have different scales in the three plots.

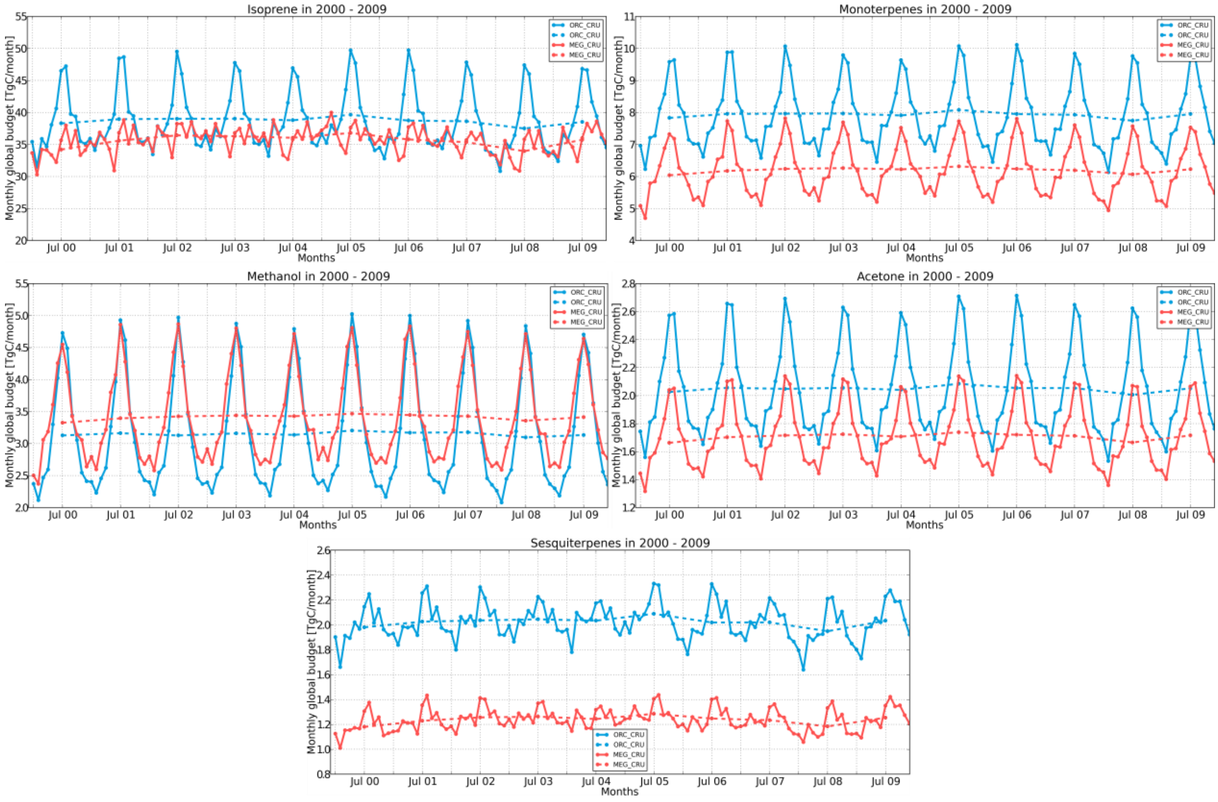

Figure 2. Monthly global (solid lines) and yearly averaged (dashed lines) emission budgets in Tg C month^{-1} for ORC_CRU and MEG_CRU simulations for isoprene, monoterpenes, methanol, acetone and sesquiterpenes.

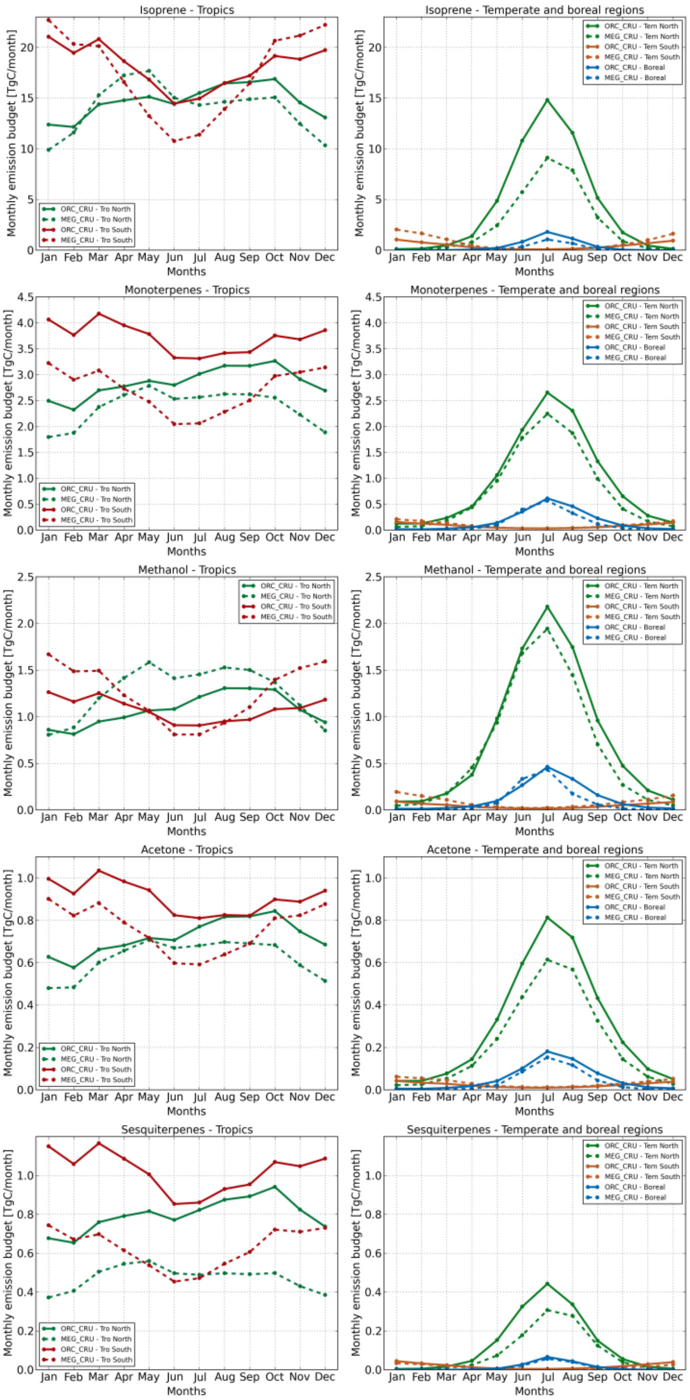

Figure 3. Zonal mean for northern and southern tropics (left column), northern and southern temperate and northern boreal latitudes (right column) of the monthly emission budget (Tg C month^{-1}) averaged over the simulation period (2000–2009) in ORC_CRU and MEG_CRU runs for isoprene, monoterpenes, methanol,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

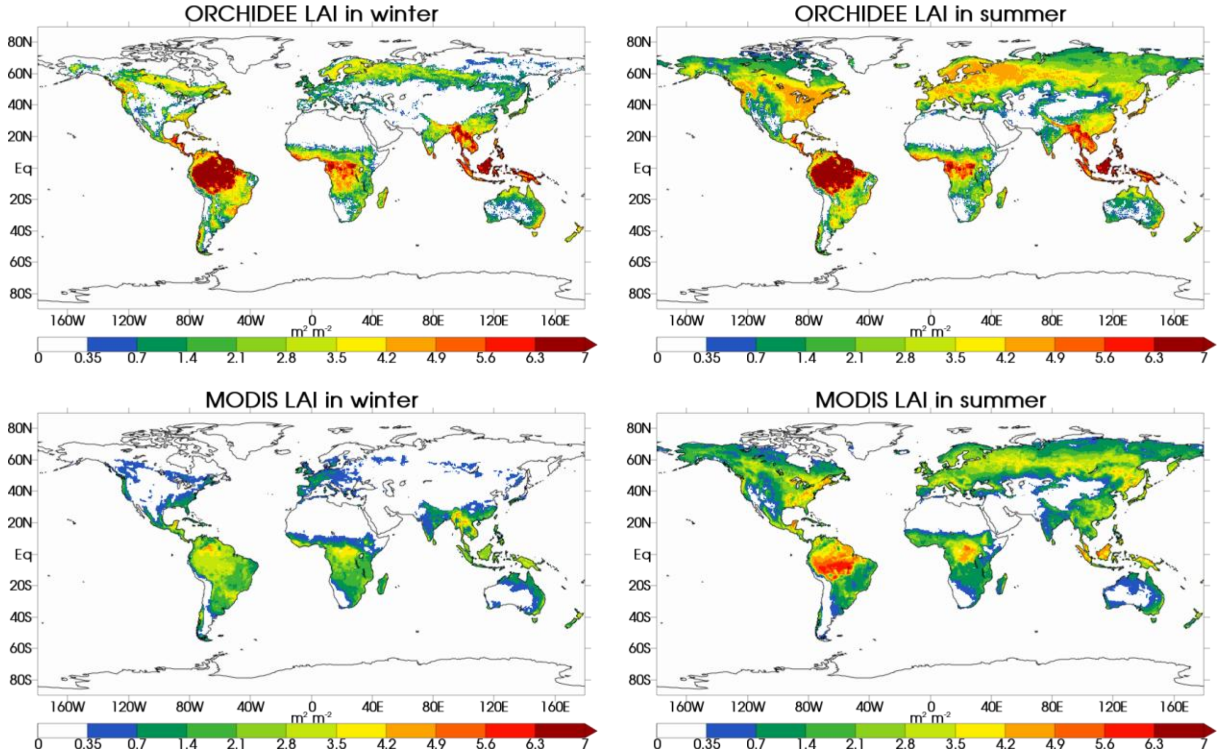

Figure 4. Leaf area index (LAI) considered for BVOC emission estimates in ORCHIDEE (LAI calculated on line) and in MEGAN (MODIS retrieval) in summer (June, July, August) and winter (December, January, February), averaged over the 2000–2009 period ($m^2\ m^{-2}$).

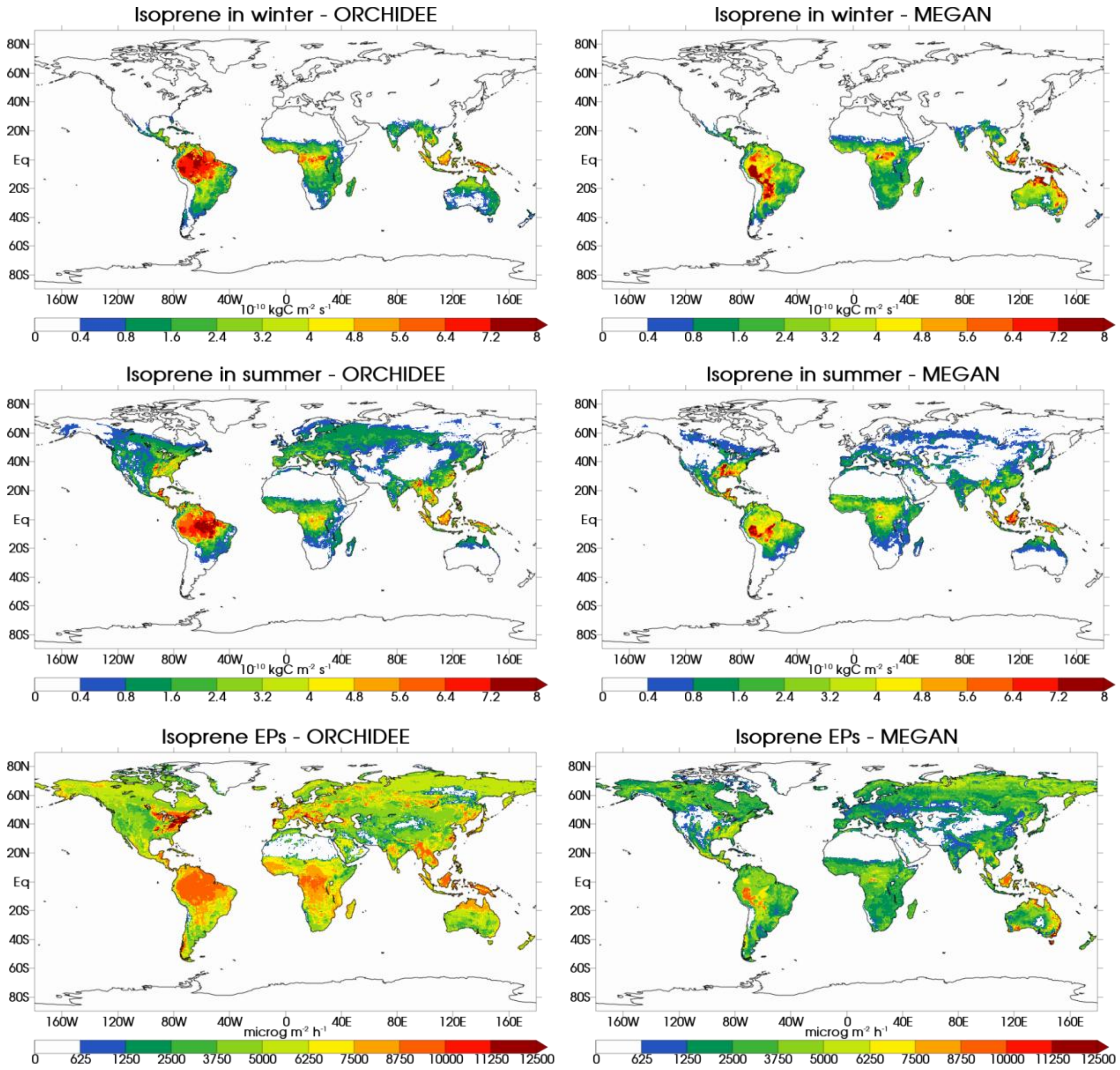

Figure 5. Emissions in winter (first row) and summer (second row) in 10^{-10} kg C m^{-2} s^{-1} and emission potentials (EPs) (third row) in μg m^{-2} h^{-1} for ORCHIDEE (ORC_CRU, left column) and MEGAN (MEG_CRU, right column) for isoprene.

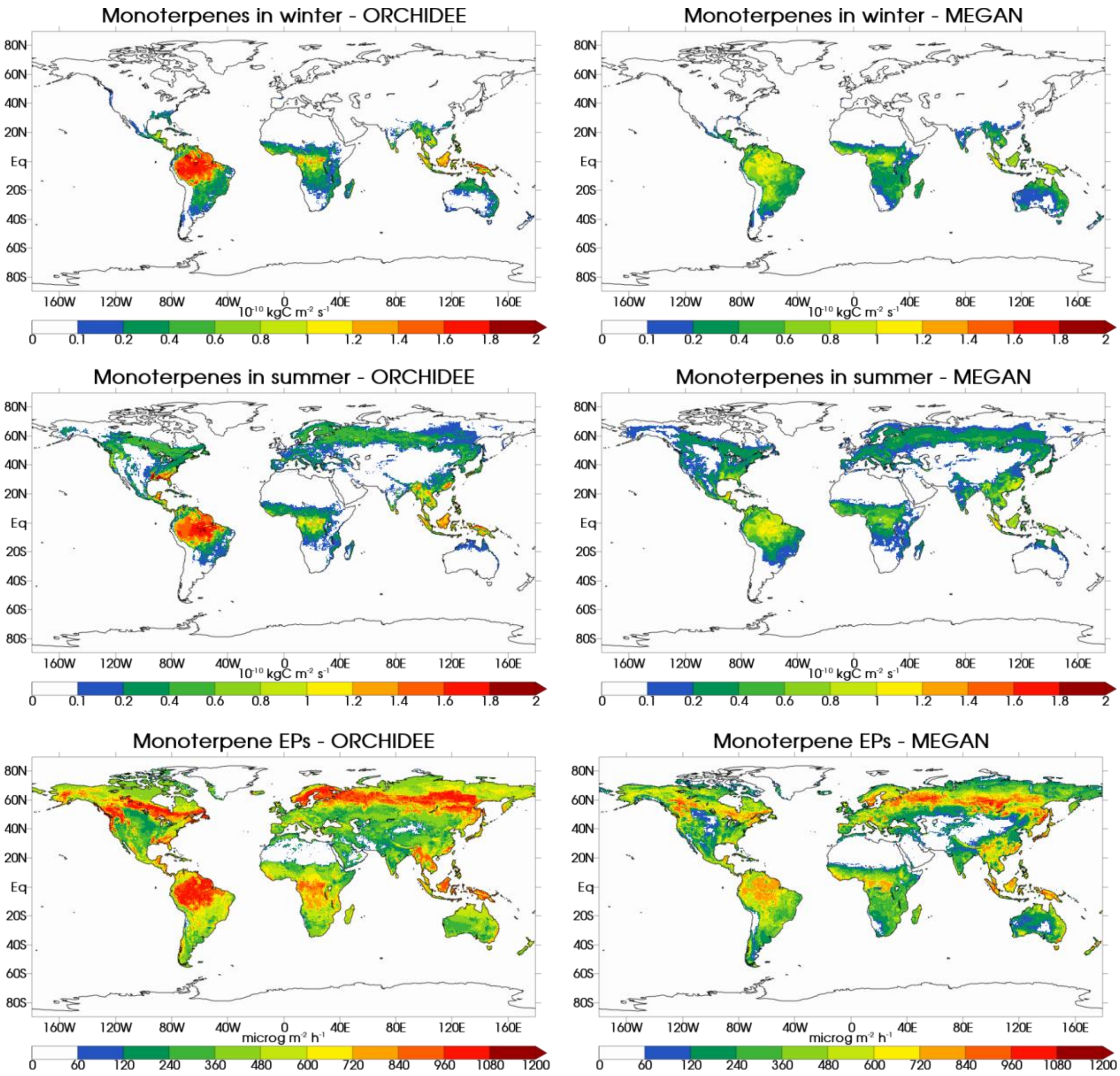

Figure 6. The same as Fig. 5, but for monoterpenes.

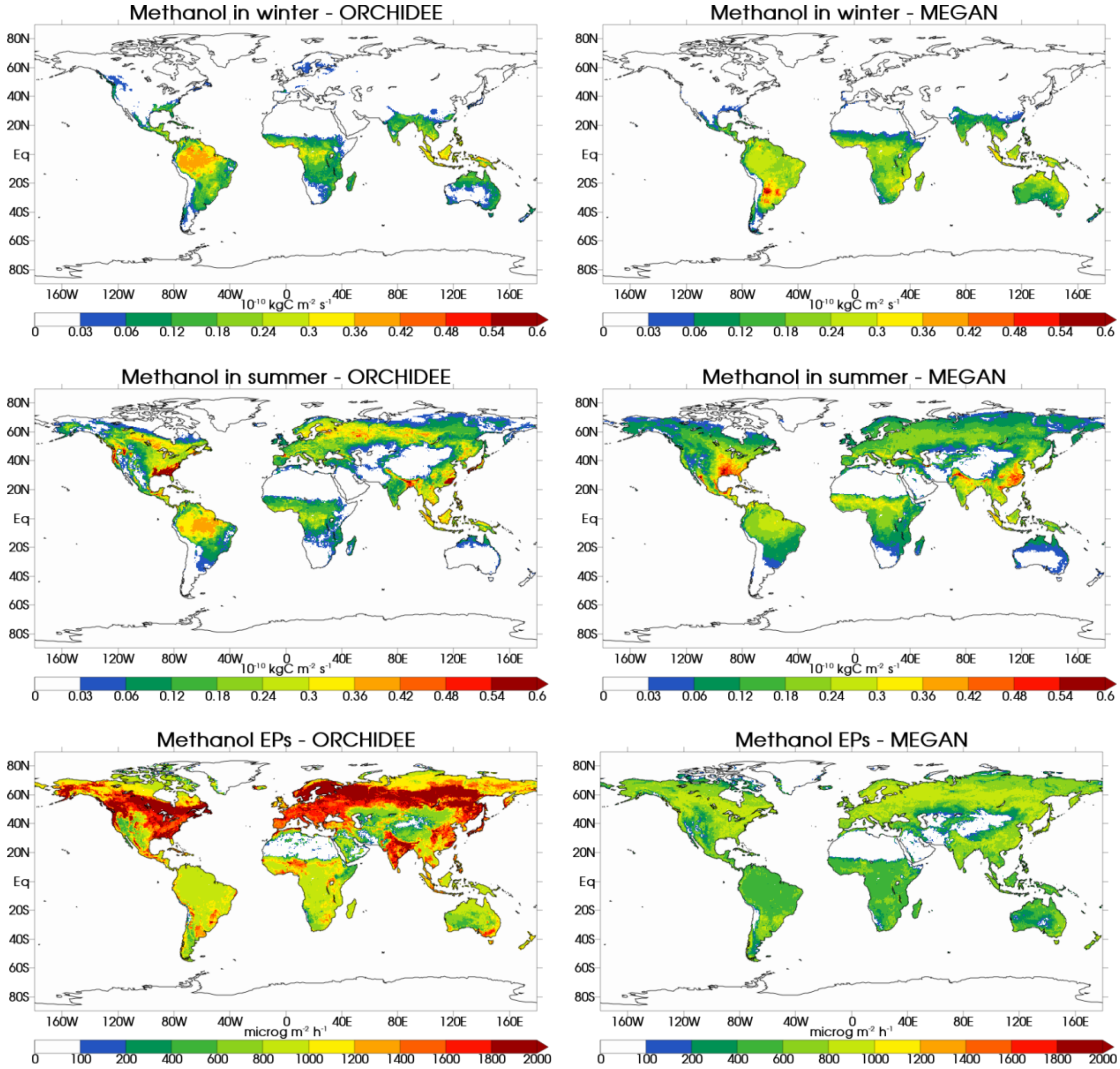

Figure 7. The same as Fig. 5, but for methanol.

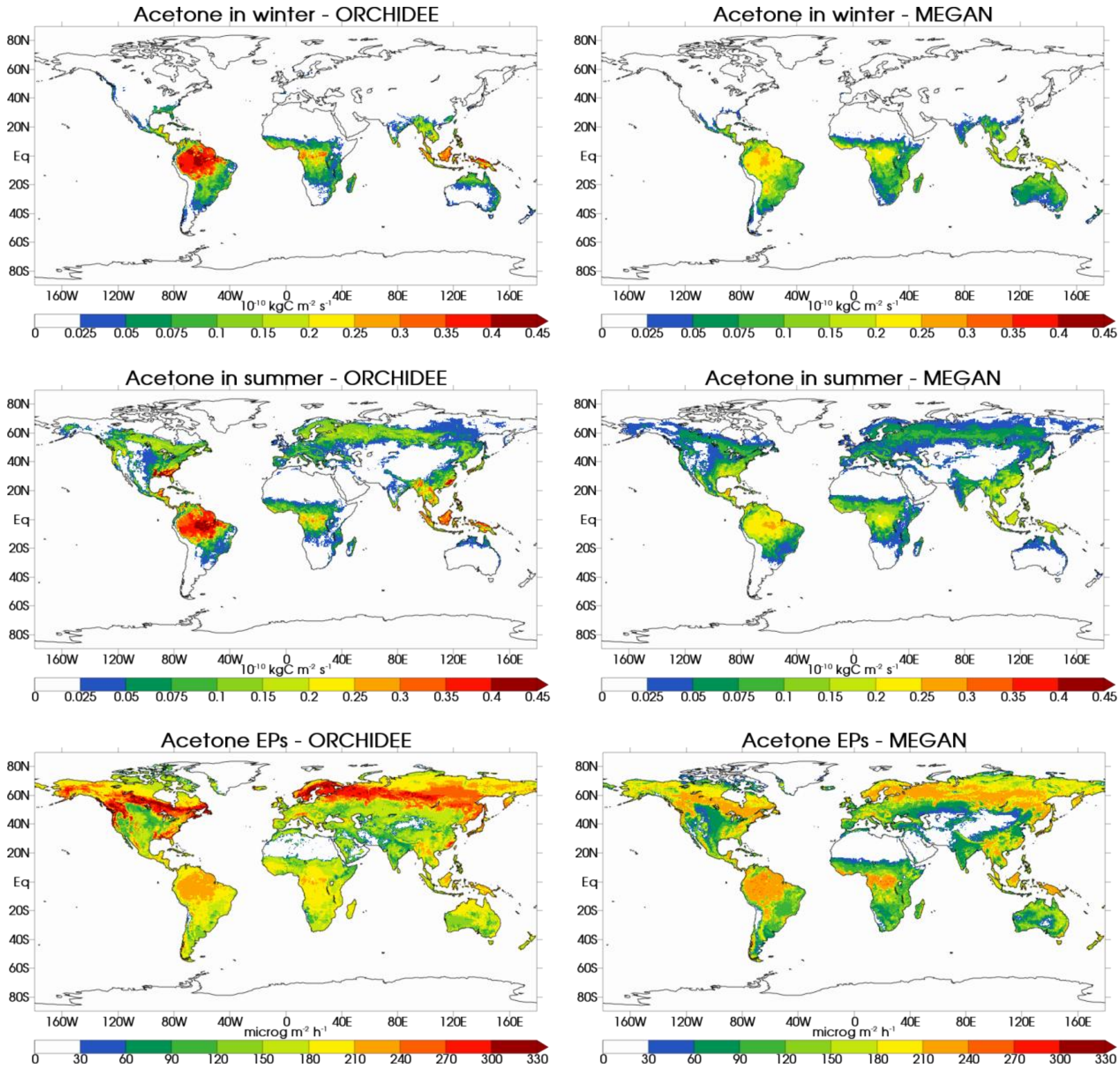

Figure 8. The same as Fig. 5, but for acetone.

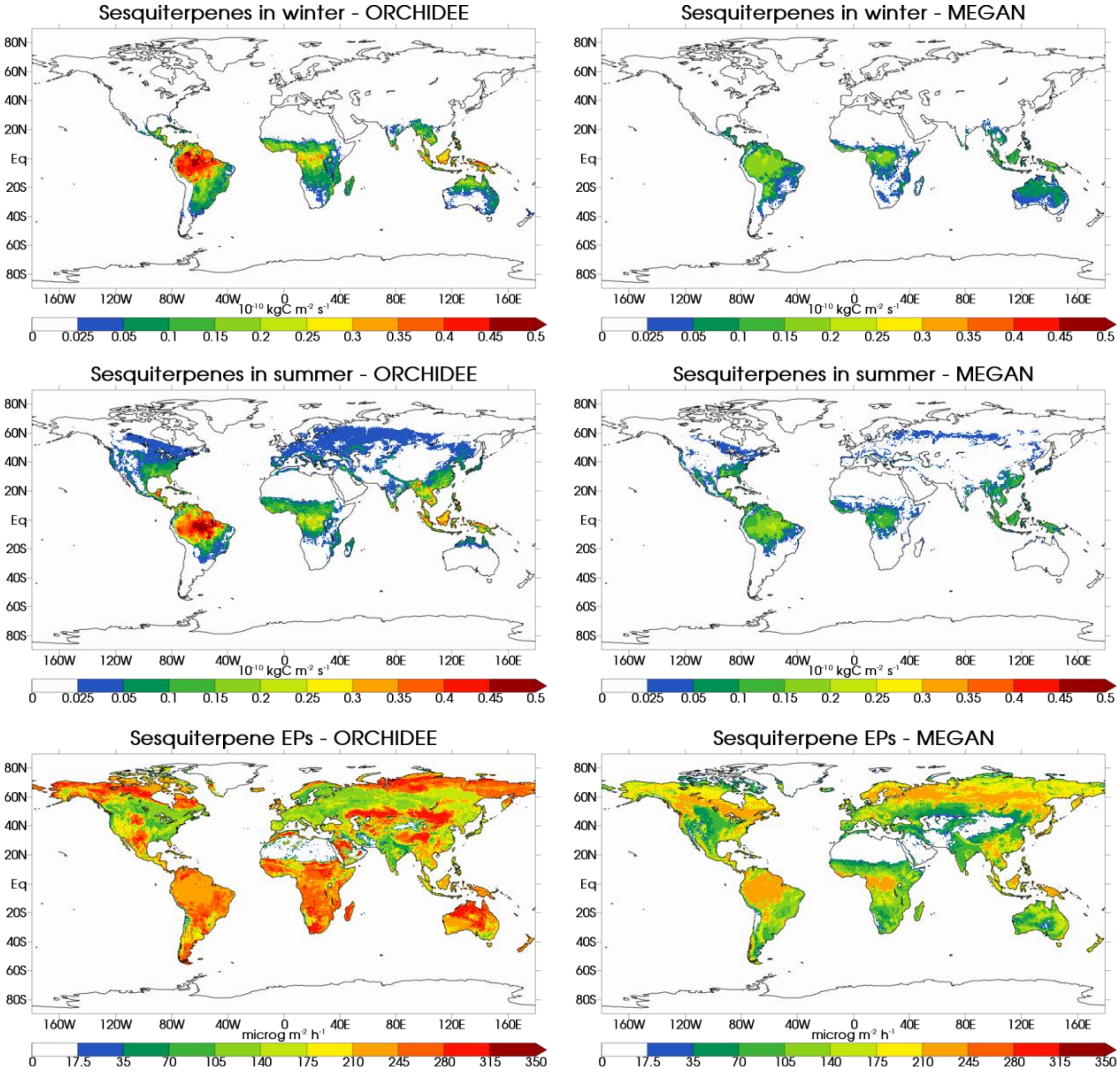

Figure 9. The same as Fig. 5, but for sesquiterpenes.

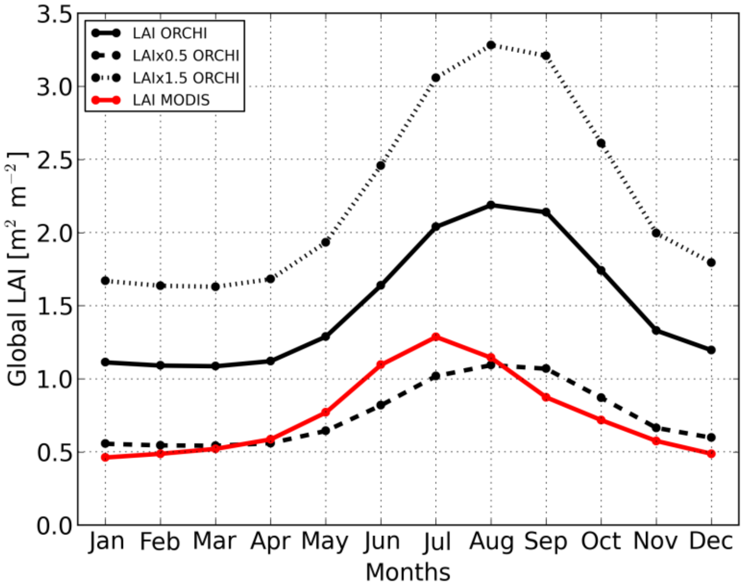

Figure 10. Global monthly mean LAI (m^2 m^{-2}) calculated by ORCHIDEE (solid black line) and retrieved from MODIS measurements (red line). The thick and thin dashed lines represent the LAI from ORCHIDEE multiplied by a factor 0.5 and 1.5, respectively.

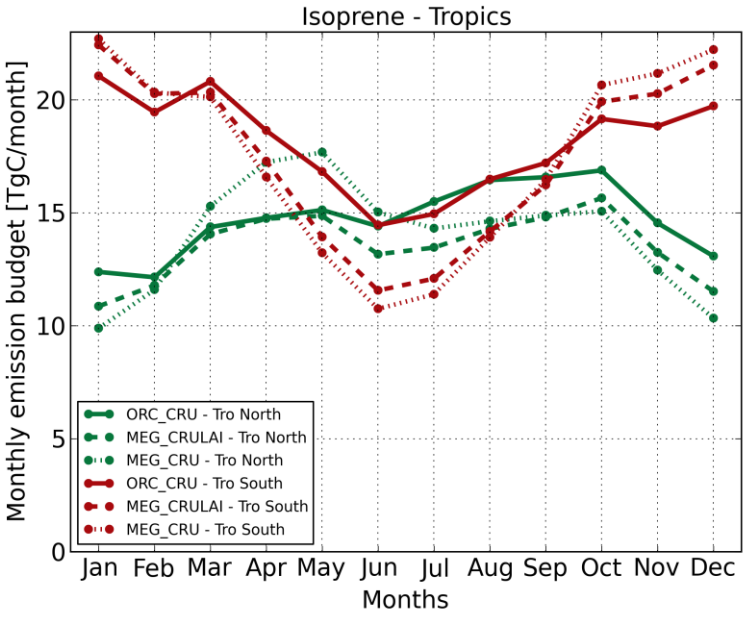

Figure 11. Zonal mean of monthly emission budgets (Tg C month^{-1}), averaged over the simulation period (2000–2009) for the northern and southern tropics, in ORC_CRU (solid line) and MEG_CRULAI (dashed line) simulations for isoprene.

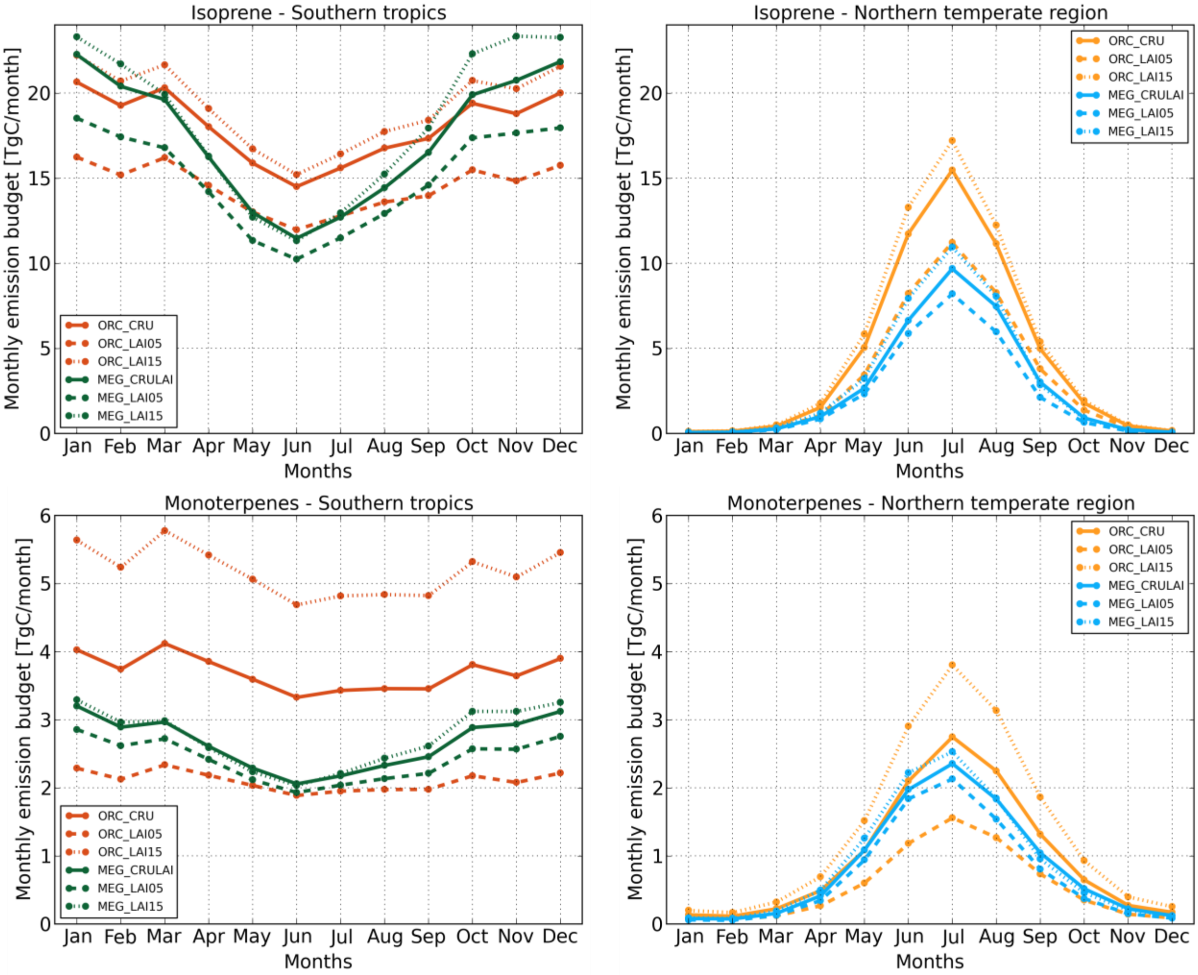

Figure 12. Zonal average of changed emissions in the different LAI sensitivity tests: ORC_CRU and MEG_CRULAI using ORCHIDEE LAI (solid line), ORC_LAI05 and MEG_LAI05 using ORCHIDEE LAI·0.5 (thin dashed line) and ORC_LAI15 and MEG_LAI15 using ORCHIDEE LAI·1.5 (thick dashed line) in the year 2006, for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

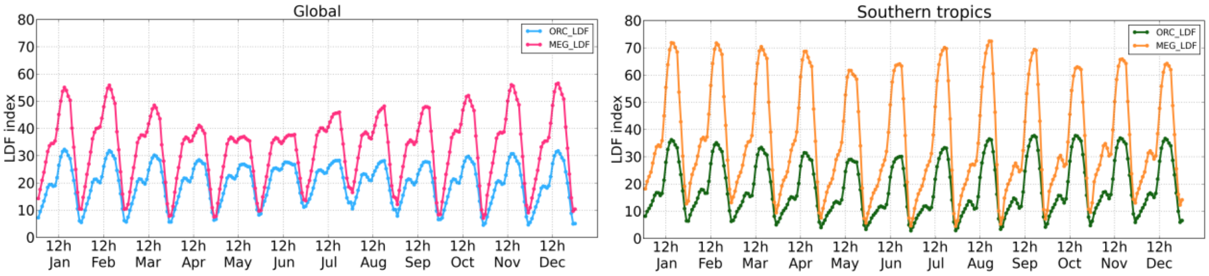

Figure 13. Global (left plot) and southern tropical (right plot) average of the LDF index for ORCHIDEE and MEGAN. The LDF index is provided as the hourly daily profile averaged over each month.