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Responses to Editor's and Referees' Comments

Summary for the Editor

We thank the Editor and the Referees for the worthy and constructive comments that helped us to significantly improve the manuscript.

We summarize below the major changes made to the manuscript in order to address Editor's and Referees' major concerns.

Following Editor's and Referees' suggestion:

- we add a new section (2.2.1) dedicated to EFs where we discuss the difficulties related to the EF setting at global scale. We describe the general approach used to assign new EFs and we give some examples on how we proceed in updating the emission factor;
- we insert a new section (2.5) where we detail the differences between emission module formulations in ORCHIDEE and in MEGAN;
- we better detail the arguments supporting the usage of the ORCHIDEE LAI as input for the emission scheme. We also point out the problems still to be solved and the open questions;
- we re-organise section 3.4, differentiating more clearly the discussion on LAI uncertainties and the impact on BVOC emission estimates of LAI seasonal cycle and size. We add new sub-sections: *"3.4.1 LAI seasonal cycle impact"* and *"3.4.2 LAI size impact"*.
- we better describe the LDF parameter used in both models and the implications of the simplified assumptions of this parameterization;
- we add a new section (5) where we discuss the possible developments and the impact of our findings.
- The manuscript was read by two English native speakers to reduce, as much as possible, the grammar mistakes.

Responses to Editor's Comments

We thank Editor for his time and consideration. We closely examine the insightful and constructive comments, that have helped us to improve the manuscript.

Editor's comments are quoted in bold. Authors' answers are in regular font and authors' changes in the manuscript are quoted in italic.

We refer to the marked-up manuscript version for section numbers and pages.

Responses to General Comment

I have a comment on the light dependent fraction (LDF) of monoterpene emission, as included in Equation (2) of the manuscript. The LDFs used in the study are different for different compounds emitted, being 1 for isoprene and 0.6 for monoterpenes. However, these seem to be held constant across all plant functional types. I question the validity of this latter assumption which seems to be copied from predecessor models. According to both the light dependencies of measured monoterpene emissions (Staudt and Seufert 1995; Steinbrecher et al., 1999; Kuhn et al., 2002; Rinne et al 2002; Taipale et al., 2011) and stable isotope labeling experiments (Loreto et al., 1996; Ghirardo et al., 2010), the LDF for monoterpenes to vary from 1 for many broadleaf trees to less than 0.5 for many conifers. This issue and its implications should be at least discussed in the paper.

Authors: We are aware that this is a rather crude approximation. Nevertheless, there are currently not enough observations to define a solid global scale parameterization of LDF, which presents similar difficulties as setting EFs (first paragraph of section 3.5). Therefore, as a first step, we decided not to further detail the LDF modelling and we have chosen a single LDF value for all PTFs, as proposed in Guenther et al. 2012. We discuss this issue in more details in section 1, leaning on the references suggested by the Editor (see §1).

The sensitivity tests that we have performed prove that emissions can vary significantly depending on the LDF used. They also provide an evaluation of the error associated with a different selection of LDF. This is detailed below in §2.

At the end of section 3.5 we better detail the implication related to LDF incertitude and we change the last sentence (see §2), which in the original version leads to misinterpretations. Moreover, we add a point about this issue in section 5 (see §3).

§1 *"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 to individual compounds and PFTs, as it also appears from the works mentioned right above."*

§2 *"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."*

§3 *"- 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;"*

Response to Technical Comment

Why is value for beta defined for isoprene emission, when the term where it appears in Equation (2) is zero when LDF is 1?

Authors: In the code we set arbitrary beta = 0.9 for MBO and isoprene, but indeed the beta value can be any, since the CTLI term is zero when LDF is set to1 and there is no need to specify it. We therefore remove the beta value for isoprene and MBO from the table 2 and we put "-".

References from Editor

Ghirardo A, Koch K, Taipale R, Zimmer I, Schnitzler J-P, Rinne J. 2010. Determination of de novo and pool emissions of terpenes from four common boreal/alpine trees by 13CO2 labelling and PTR-MS analysis. Plant, Cell & Environment, 33: 781-792.

Kuhn, U. S. Rottenberger, T. Biesenthal, A.Wolf, G. Schebeske, P. Ciccioli, E. Brancaleoni, M. Frattoni, T.M. Tavares, and J. Kesselmeier Isoprene and monoterpene emission of Amazonian tree species during the wet season: direct and indirect investigations on controlling environmental functions Journal of Geophysical Research, 107 (D20), 8071, doi:10.1029/2001JD000978, (2002)

Loreto, F., Ciccioli, P., Cecinato, A., Brancaleoni, E., Frattoni, M.,Fabozzi,C.,Tricoli,D.,1996. Evidence of the photosynthetic origin of monoterpenes emitted by Quercus ilex L. leaves by 13C labeling. Plant Physiology 110, 1317–1322.

Rinne, H.J.I., A.B. Guenther, J.P. Greenberg, P.C. Harley: Isoprene and monoterpene fluxes measured above Amazonian rainforest and their dependence on light and temperature. Atmospheric Environment 36 (2002) 2421–2426

Staudt M., Seufert G. (1995) Light-dependent emissions of monoterpenes by Holm oak (Quercus ilex, L.), Naturwissenschaften 82, pp.89-92.

Steinbrecher R., Hauff K., Hakola H. & Rössler J. (1999) A revised parameterization for emission modelling of isoprenoids for boreal plants. In Biogenic VOC Emissions and Photochemistry in the Boreal Regions of Europe (eds T. Laurila & V. Lindfors), pp. 29–43. Air Pollution Research Report 70. Commission of European Communities, Luxembourg.

Taipale, R., M. K. Kajos, J. Patokoski, P. Rantala, T. M. Ruuskanen, and J. Rinne. Role of de novo biosynthesis in ecosystem scale monoterpene emissions from a boreal Scots pine forest. Biogeosciences, 8, 2247-2255, doi:10.5194/bg-8-2247-2011, 2011

References in the answers:

Shao, M., Czapiewski, K. V., Heiden, A. C., Kobel, K., Komenda, M., Koppmann, R., and Wildt, J.: Volatile organic compound emissions from Scots pine: Mechanisms and description by algorithms, J. Geophys. Res., 106(D17), 20 483–20 492, 2001.

Responses to Referee #1's Comments

We thank Referee #1 for his/her time and consideration. We closely examined his/her insightful and constructive comments which have considerably helped us to improve the manuscript.

Referee #1's comments are quoted in bold. Authors' answers are in regular font and authors' changes in the manuscript are quoted in italic.

We refer to the marked-up manuscript version for section numbers and pages.

Response to General Comments

(Initial paragraph or section evaluating the overall quality of the discussion paper)

1. This paper describes the updates in the BVOC emission module in the ORCHIDEE model. It further compares the predicted emissions by ORCHIDEE with the emissions predicted by the widely used emission model MEGAN. The paper also provides sensitivity test results to various parameters. The authors find that the spacial patterns of various BVOCs depend mostly on the allocated emission factor, while the seasonal patterns depend mostly on the leaf area index. The scope of the paper fits the journal well. The topic is timely, since there is a continuous need to intercompare models and a strong need to test their sensitivity towards input parameters, though there is also a strong need to evaluate models against measurements. These needs arise as there are great variations in individual plant species and that these species that have very different potential to emit VOCs (both in amounts and in the distribution of individual VOCs) are only covered by a few emission potentials in global models - as is also emphasised by the authors. The authors also highlight that there are very many factors (both physical, but especially biological) that affect the emissions, hence one has to be cautions when making conclusions based on global emission estimates. Unfortunately, the paper is quite messy and there are many mistakes – both in the grammar, but more importantly in the use of symbols. This naturally has to be corrected. My main concerning is how the emission potentials (EF) are allocated to the plant functional types (see also below). The handling of the EF is more detailed in the new emission module of ORCHIDEE (include higher degree of light dependency), but the justification of EF to different PFT has not improved and is also not properly justified. As the authors also specify, the impact of EF is dominant on the predicted emission and I therefore do not understand why the authors did not try to improve this part.

Authors: Referee #1 points out a crucial issue in BVOC modelling: the emission potential (EF) allocation. Following also the Referee #2's comment, we discuss more carefully the inherent difficulties in setting EFs at global scale. We add a new section (2.2.1) where we describe the general approach used to assign new EFs and its limitations.

In addition, accordingly Referee #1's comments, we re-organise some parts of manuscript, seeking a clearer exposition, and we correct the use of notation in equations (1), (2) and (3) (for the details see points 51). The paper was read by two English native speakers to reduce, as much as possible, the grammar mistakes.

*"**2.2.1. Emission Factors 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 (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 2–methyl–3–buten–2–ol (hereafter we refer to it simply as 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)."

Response to Specific comments:

(Section addressing individual scientific questions/issues)

2. *) P33970L6-9: you could potentially supplement with some references.

Authors: We add at first paragraph of section 1 the references as follows.

"(Laothawornkitkul et al, 2009; Guenther et al., 2012; Penũelas and Staudt 2014)"

3. *) P33970L14: maybe you could also mention some of the papers that first showed the contribution of BVOCs to SOA formation and growth.

Authors: We change the text, in section 1, page 3.

"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)."

4. *) P33970L9-18: Maybe you could also mention the contribution to CCN and clouds and hence climate.

Authors: We add in section 1, page 3:

"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)."

5. *) P33971L15-18: Maybe you could add some references?

Authors: We add in section 1, page 4, two references in which EF is defined.

"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),…"

6. *) P33972L1-6: Also mention that phenology is not included in these models (e.g. MEGAN and ORCHIDEE), which is a big lack in order to describe the emission of VOCs.

Authors: Actually, ORCHIDEE includes an explicit description and calculation of plant phenology, as briefly described in section 2.4. MEGAN, on the other hand, does not explicitly calculate it. What is not taken into account in both models is the variation of EF in relation with plant phenology.

We add it in section 1, page 5.

"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."

7. *) P33972L16-17: I agree, but maybe it is also worth mentioning that the temperature and light dependency not only depends on the VOC of interest, but also the plant species considered (e.g. Ghirardo et al., 2010) due to the different production paths.

Authors: In section 1, page 5, we detail more this important point, also following the Editor remark:

"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."

8. *) You discuss the sensitivity of the models with respect to LAI and also mention the discrepancy between modelled and measured LAI, but there is no word on biomass. I did a quick test with MEGAN and the emission scales proportionally with the biomass and non-linearly with LAI, and the emission is much more sensitive to the biomass than to the LAI. The effect of biomass has to be discussed in the paper. It would also improve the manuscript if there was at least some discussion on other canopy characteristics (canopy height, depth, age, ...).

Authors: The current version of the MEGAN model, which is the one used in this study, does not use the biomass density as an input parameter. It defines emission potentials (EPs) of the model grid cell (in case of using the emission potential maps) or EFs of each PFT (when using the look up table) as emission per surface ($\mu g\ m^{-2}\ h^{-1}$). Therefore the amount of emitting biomass is, in fact, accounted in the value of LAI. Even in the ORCHIDEE emission module, the input parameter is LAI, that is obtained by multiplying the leaf biomass per PFT grid cell area ($gC\ m^{-2}$) to the PFT

specific leaf occupying area (SLA) (m^2 gC^{-1}). The biomass is calculated in the phenology module, while SLA is a parameter vector.

About the other canopy characteristics in MEGAN (i.e canopy height, canopy depth, leaf width, leaf length), they are principally used to determine the leaf temperature. As the leaf temperature is not calculated in ORCHIDEE, there is no correspondence about these parameters between the two models.

In MEGAN leaf age classes, are derived considering the variation between LAI value of the current and preceding month, following a highly parameterised scheme. In ORCHIDEE leaf age classes are calculated considering the plant leaf growth and leaf turnover at each model time step (30 minutes) and are not directly correlated with LAI. The comparison between these two variables and the implementation of sensitivity tests to assess the impact of these different approaches are not straightforward. It would be a very interesting investigation and we mention it as future development of this work in the new section 5 (see below).

It is worth to mention these differences between both models. We add them in the new section 2.5 related to the differences between ORCHIDEE and MEGAN emission algorithms (see point 10.).

"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."

9. *) P33973L23-29: I do not agree with the authors at all! You cannot asses the correctness of a model by inter-comparing it with another model. This will only tell you how the models differ. As the authors mention earlier in the intro, more and more field measurements are done and it must be those that the models have to be evaluated against in order to evaluate their uncertainty. Unfortunately this has not been done enough, but there is a strong need to do so! A few examples are Tsigaridis et al., 2014, Mann et al., 2014 and Spracklen & Righelato 2014.

Author: The sentence indeed was not properly formulated. We change it in section 1, page 7 as follows:

"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."

10. *) It would improve the paper significantly if you included a section that clearly describes what is the difference between your emission module and the MEGAN module. To me it seems that the largest difference is the land cover, which is anyway not predicted by your emission module, but LPJ (or LUH – not clear which). Otherwise it seems to me that you just changed the light dependency and emission factors and this you might as well just have been done in MEGAN.

Authors: We add a new section 2.5, where the differences between the two models and the emission schemes are better explained (see §1). We also change the paragraph where the γ_{LAI} is described (see §2) in section 3.4.2.

We have also noticed that the explanation about the land cover database is ambiguous. Since simulations were performed only for ten years, we do not use a dynamical calculation of vegetation land cover but, rather, land cover fixed data provided by Hurtt et al. (2006). There is, thus, no need to mention this ORCHIDEE module. Therefore we have removed the last sentence about the LPJ model at the end of section 2.1. We also add in section 2.4, page 18, the reference path for the land cover files used (see §3).

§1 ***"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 equation 9 and 10), while in MEGAN it is defined by equations (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 section 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 section 2.1;

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

§2 *"Such differences are detailed in point 6 of section 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* $m^2\ m^{-2}$*."*

§3 *"The database can be found in:*

http://dods.extra.cea.fr/work/p86ipsl/IGCM/BC/SFR/OL2/PFTmap_1850to2005_AR5_LUHa.rc2."

11. *) P33976L1-2: I guess that this is also the case in MEGAN?

Authors: In MEGAN there are two options:

(1) EFs are set per PFT as in ORCHIDEE using look up table (for the values see Table 2 in Guenther et al., 2012);
(2) high resolution emission potential (EP) maps are used, which do not depend on PTFs. These maps are compiled considering information from different plant species flux measurements and land cover plant species distribution. EPs are provided for isoprene, α-pinene, β-pinene, 3-Carene, limonene, myrcene, t-β-ocimene and sabinene.

It is now more explicit in the text in the new section 2.5 (see §1, point 5).

12. *) P33976L17-19: I guess the real argument is also that these are the compounds that have been measured to be emitted from vegetation in the greatest abundance?

Authors: We definitely agree with the Referee #1. We add the comment in section 2.2, page 10.

"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."

13. *) P33976L25-26: Have you somehow taken the landcover of various species within a PFT into consideration when doing the averaging? If not, I am sceptical.

Authors: We have taken into account the possibility of making an average weighted on land cover of various species within a PFT. But we remarked that the values of EF collected are often related

to a small number of species and often come from the same measurement sites. This is one of the major limitation in the allocation of EFs, which cannot be reduced by the weighted average. In theory, if we knew the EF of all plant species belonging to the same PFT and their land cover, the weighted average method would be statistically robust and absolutely preferred. Unfortunately, all this information is not currently available. We agree that it is a very coarse procedure. The low accuracy of the EF parameter used in the model, as well as other parameters/variables, is the principal reason which prompted us to perform a sensitivity study.

We detail the EF limitation in the new section 2.2.1 (see also point 1).

14. *) P33977L3-6: Please also mention the large change in boreal broadleaved deciduous trees, which I somehow doubt.

Authors: We mention in the text (section 2.2.1, page 15) the new EF value, related to boreal broadleaved deciduous trees, and the papers in which particularly high values for this PFT are provided.

"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})."

15. *) P33977L13-15: But one could look into e.g. forest inventories or similar in order to get a better idea.

Authors: We agree with Referee #1, considering anyway the limitations which may arise and that we discuss in the point 13. Following Referee #2's remark too, we detailed in the new section 2.2.1 the limitation in assigning an individual EFs per PFT (see point 1).

16. *) P33977L25-29: Please also mention that the light dependency of a compound also depends on which plant it is emitted from (e.g. Ghirardo et al., 2010).

The Referee #1 raises an important point. Also accordingly with the Editor's comment, we detail in section 1 (page 5) the discussion about the light dependency (see §4) and we mention it in section 2.2 , page 11 (see §5), following the 2 comment, too. The text is reformulated in the following way:

§3 *"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 and 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."

§4 *"As detailed in section 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."*

17. *) P33978L3: I do not agree that light dependency only means "directly released through stomata" and that temperature dependency only means "stored in the leaf pool". It is much more complicated than that and refers in great part to the production of the compounds. So please reformulate or leave out.

Authors: we removed it from the text.

18. *) P33982L1-5: It would be very very interesting to see what emissions ORCHIDEE and MEGAN would produce if the approach of the other model was used.

Authors: Referee #1 raises a very interesting point. However, doing this in MEGAN would require considerable changes in the code and this is out of the scope of the current paper. In fact, main objective is to characterise how the particular characteristics of the two models impact on the BVOC emission estimates, by putting them under the same forcing conditions (see §5). We specify it in section 1 (page 7).

However, we have performed this test in ORCHIDEE. We found that the annual global emissions calculated with the usual method are 5-12% higher than those calculated adopting the MEGAN approach (using EP and LAI averaged over the grid cell). Isoprene presents the lowest differences and monoterpenes the highest. This test gives indeed an interesting insight and we mention it in section 2.5, point 5 (see §6).

On a general ground, we are interested in performing sensitivity tests on variables/parameters which are likely to be objects of future developments of ORCHIDEE. For example: EFs, LDFs or LAI. In our opinion, this is less likely to be the case for the use of emission potential maps.

§5 *"…(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 section 2.5), in particular the emission scheme, classes and distribution of PFTs and LAI processing, …"*

§6 *"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."*

19. *) P33982L11-12: Guess the point is that some areas (e.g. Europe and the US) are covered quite well (though there is definitely a lack of year-round measurements), while there exists no or close to no data in order regions.

Authors: We agree with Referee #1 and we change the text at the beginning of section 3.1 as follows, considering also the point 9:

"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."

20. *) P33983L22-24: Reading this and looking at Fig. 1, it seems to me that what should really be tested/improved is the met forcings, since that seems to have much greater impact than the emission module.

Authors: We are aware that the emission variations resulting from differences in meteorological forcing can be significant and we are also aware that this is a very important issue. However, in the manuscript we focus more on the internal source of variability, on the weakness of the emission

module and how to improve it. In fact, the ORCHIDEE model is also designed to be used in future emission scenarios studies. We stress this point at the end of section 1, page 7 (see below).

Arneth et al. (2011) have already provided very interesting and quite exhaustive study on this issue. We, therefore, only mention some results confirming their conclusions and we do not investigate this topic further (section 3.1, page 22).

Regarding the meteorological forcing data, we are not directly involved in the meteorological forcing production. We have used for this study the state of the art forcing files available for the model.

We change the text in section 1 (page 7) as follows:

"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."

21. *) P33989L3-13: Wouldn't it be better to move this to Sec. 3.5? After reading this short paragraph, I am wondering why the emission response is different, since you use the same emission algorithm (Guenther/MEGAN).

Authors: We moved the paragraph to section 3.5.

The two emission algorithms are similar, but there are some differences that can be crucial. In the new section 2.5, we point out the principal discrepancies. In particular, the discrepancy mentioned in the paragraph is extensively explained in section 3.5.

22. *) Table 2: Where does the LDF and Beta values come from? Why is LDF and Beta 0.6 for total monoterpenes, but no values are assigned for the individual monoterpenes? Or is the LDF and Beta values also 0.6 for all the individual monoterpenes? This is not clear. There is information about the light dependency of the individual monoterpenes, which seems to be quite large – e.g. sabinene and ocimene seems to be very light dependent (e.g. Owen et al., 2002). Please indicate what "MBO" is. Compound names should not be in capital.

Authors: LDF and beta are indeed not exhaustively described in the text. We add more details in section 2.2, page 12 (see §5 and §6).

This is the first time that speciated monoterpenes are inserted in the code and, as a first step, we decided not to further detail the modelling of individual monoterpenes and choose a single

LDF/Beta value (LDF=0.6 and Beta=0.1) for all monoterpenes. We are aware that it is a rather crude approximation, but there are currently not enough observations to assign a sufficiently solid parameterization at global scale. We have chosen the Beta value referring to Guenther et al. (2012) and the LDF value relying on Dindorf et al., 2006; Holzke et al., 2006; Guenther et al., 2012; Šimpraga et al., 2013.

MBO is the 2–methyl–3–buten–2–ol and it is often referred as MBO (e.g. Hakola et al., 2006; 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). We mention more clearly in section 2.2.1 (page 15) that we refer to 2–methyl–3–buten–2–ol as MBO (see §7). Considering the other compounds, we put the name in minuscule, checking throughout the text.

Following the Editor remark too, we add a paragraph in section 1, where the LDF issue is discussed (see §8).

§5: *"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)."*

§6: *"β is the empirical coefficient of the exponential temperature response and it is now defined as in Guenther et. al (2012)"*

§7: *"…2-methyl-3-buten-2-ol (hereafter we refer to it simply as MBO)..."*

§8: *"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."

23. *) Table 3: I understand that you have to limit, but there are much more papers available.

Authors: We have collected many more papers than those included in table 3, but we have rejected many of them as they do not report EFs at standard conditions of PAR and temperature or there are not enough details for reporting EFs at the standard conditions. In addiction we realize that they are often related to the same number of species and come from the same measurement sites. For further details see point 1 and section 2.2.1 in the manuscript.

24. *) Table 4: where do these ratios come from?

Authors: This ratio comes from the paper review carried out for EFs. The only difference is that we represent it as percentage of speciated monoterpene EFs with respect to the PFT bulk monoterpene EF. The references used are those listed for monoterpenes in Table 3. We are now more precise and we add the references per individual monoterpene in Table 4.

25. *) Table 6: Why are there no estimates from MEGAN concerning limonene, myrcene, 3-carene and ocimene?

Authors: Accordingly to Referee #1's remark we add in Table 6 and 7 the limonene, myrcene, 3-carene and ocimene emission estimates.

26. *) I am very sceptical that you predict so high isoprene emissions (especially compared to the monoterpene emissions) in northern temperate and boreal areas. This also seem to be one of your largest differences to the MEGAN model. I fear that this high isoprene emission is due to the fact that you have not considered which northern plants emit isoprene and which do not.

Authors: The largest differences in emissions are actually related to the different choice of the EF values, to the different PFT land cover and to the different PFT categorisation between the two models.

To show this, we compare ORCHIDEE EFs with MEGAN ones (provided in Table 2 by Guenther et al. 2012), after converting them in the ORCHIDEE units. They are not exactly the EFs used to perform simulation in MEGAN, as in the case of isoprene the emission potential map is directly

provided. However, by calculating the emission potential using these EFs (Guenther et al. 2012) we get EPs comparable to those of the provided map (differences around 10%).

In the boreal zone the percentage of main PFTs calculated with respect to the surface occupied by vegetation in boreal region and the related EFs are:

In ORCHIDEE

- C3Gr (51.5%) with an EF = 12 μgC g^{-1} h^{-1}
- BoBrDe (21.3%) with an EF = 18 μgC g^{-1} h^{-1}
- BoNeEv (16.5%) with an EF = 8 μgC g^{-1} h^{-1}
- BoNeDe (7%) with an EF = 0.5 μgC g^{-1} h^{-1}

In MEGAN

- BoSbDe (48%) with EF = 9.9 μgC g^{-1} h^{-1},
- BoNeEv (24%) with EF = 6.2 μgC g^{-1} h^{-1}
- C3Cold (16.5%) with EF = 4 μgC g^{-1} h^{-1}
- BoBrDe (2%) with EF = 22.7 μgC g^{-1} h^{-1}

We see that the main differences come from the different choice of EF value for grass and the different PFT classes land cover. For MEGAN in boreal areas there is a strong presence of BoSbDe which is not represented in ORCHIDEE, where it is replaced by BoBrDe and C3Gr.

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.

Considering the BoBrDe EF, we set new value relying on Levis et al. (2003), Arneth et al. (2011), Guenther et al. (2006) and Guenther et al. (2012) but the choice is critical as there is no shrub in ORCHIDEE.

In the case of the temperate zones we identify similar problems. The percentage of main PFTs and the related EFs are:

In ORCHIDEE

- TeNeEv (9.4%) with EF = 8 μgC g^{-1} h^{-1}
- TeBrDe (12%) with EF = 45 μgC g^{-1} h^{-1}
- C3Gr (42%) EF = 12 μgC g^{-1} h^{-1}

- C3Ag (18%) with EF = 5 μgC g^{-1} h^{-1}

- BoNeEv (7.8%) EF = 8 μgC g^{-1} h^{-1}

In MEGAN :

- TeNeEv (8.4%)

- BoNeEv (20%) EF = 6.2 μgC g^{-1} h^{-1}

- TeBrDe (7.6%) with EF = 31 μgC g^{-1} h^{-1}

- C3GrCold (6%) with EF = 4 μgC g^{-1} h^{-1}

- C3GrCool (20%) with EF = 2 μgC g^{-1} h^{-1}

- Crop (23.2%) with EF = 0.12 μgC g^{-1} h^{-1}

In this case the discrepancies, between the two models, are mainly linked to the different values of the EFs and the different PFT surface coverage for grass and crop.

In the north temperate zones, there is an additional reason. The global average of bare soil in ORCHIDEE is half compared to MEGAN, while the north temperate average of bare is a third. This can contribute to higher emissions of ORCHIDEE than MEGAN.

From this comparison the limitations in the use of the PFTs come up. The most critical PFTs are the grass and the crop and in boreal region the BoBrDe and BoSbDe. Further studies would be useful to understand if the chosen values should be modified. It appears rather clear that a parameterisation with a larger number of PFTs could lead to more accurate results. We raise this point in the conclusions.

Lowering the ORCHIDEE EF values for the concerned PFTs we could reduce the differences between ORCHIDEE and MEGAN emissions. Nevertheless this is out of the scope of the article, which is focused on quantifying the variability of BVOC related to certain parameters. Moreover similarities of the outcomes of the two models may be accidental. For example, in the paper we show that ORCHIDEE is very sensitive to variations of LAI and with can make its emissions much more similar to those of MEGAN by halving the LAI (ORC_LAI05 simulation, Fig. 12, right column). Thus, if MODIS LAI would eventually turn out to be the correct one, we would have to reduce the LAI ORCHIDEE by a factor 2 (Fig. 10), and we would get similar emissions to those of simulation ORC_LAI05. However, at present, given the high uncertainties on LAI MODIS, we are not allowed to conclude that ORCHIDEE LAI is incorrect. In conclusion, we cannot say, at the moment, whether it is better to correct LAI or the EF or both.

We add in section 3.1 a part of this important issue.

"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."

27. *) You predict higher sesquiterpene emissions in the tropics than MEGAN – why is this so? Just because the EF is increased in your simulations?

Authors: The largest differences in emissions are actually related to the differences of PFTs distribution and EFs between the two models, in particular the differences that rise from crop and grass. The higher EFs related to these PFTs contribute to higher sesquiterpenes emissions in ORCHIDEE in comparison to MEGAN.

In the Tropics the percentage of main PFTs calculated with respect to the surface occupied by vegetation in that region and the related EFs are:

In ORCHIDEE:

- C3Gr (17.6%) EF = 0.6 μgC g^{-1} h^{-1}
- C4Gr (27%) EF = 0.6 μgC g^{-1} h^{-1}
- C4Ag (8%) EF = 0.08 μgC g^{-1} h^{-1}
- TrBrEv (24%) EF = 0.45 μgC g^{-1} h^{-1}
- TrBrDe (13%) EF = 0.45 μgC g^{-1} h^{-1}

In MEGAN

- C3GrWarm (22.4%) EF = 0.01 μgC g^{-1} h^{-1}
- C3GrCool (11%) EF = 0.01 μgC g^{-1} h^{-1}
- Crop 14% (14%) EF = 0.002 μgC g^{-1} h^{-1}

- TrBrEv (24%) EF = 0.46 μgC g^{-1} h^{-1}

- TrBrDe (13%) EF = 0.46 μgC g^{-1} h^{-1}

The EFs, set in ORCHIDEE, are based on Matsunaga et al. (2009), Steinbrecher et al. (2009), Ortega et al. (2008), Duhl et al. (2008), where the values are considerably higher than in MEGAN.

We do not add the above examples to the manuscript since we consider that those provided in the answer to point 26 already give a sufficient insight in the matter.

28. *) It would be good if you added a section in the end that would also discuss the impact of your findings? And maybe hold this together with previous studies on e.g. meteorology.

Authors: We add a new section in the end of manuscript, discussing the possible developments and the impact of our findings.

"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."

Response to Technical Corrections:

(Compact listing of purely technical corrections)

29. *) There are many places where the language could be improved (not by fancy words, but just correct English – e.g. the article is sometimes missing.). I have indicated some mistakes, but there are more.

Authors: We thank the Referee #1 for checking the English of manuscript. We examine the text, also following the Referee #2's comment, in particular the use of article "the", the present and past tenses, some prepositions (in, at, for, with…), the misspelled of needleleaf. We uniform all the text in UK English. The manuscript text has been read by two English native speakers.

30. *) You use the unit "gdm" - it is not clear to me what this means.

Authors: It means gram of dry matter. Actually "gdm" is misleading and can be better replaced by "g". We change in "gdm" in "g" everywhere.

31. *) Please provide the full institutional addresses in the affiliations.

Authors: We change the text in:

"[1]{Laboratoire des Sciences du Climat et de l'Environnement, LSCE-IPSL, CEA/CNRS/OVSQ, Université Paris-Saclay, CEA-Orme des Merisiers, 91191 Gif-sur-Yvette, France}

[2]{Laboratoire Atmosphères, Milieux, Observations Spatiales, LATMOS-IPSL, UPMC/CNRS/OVSQ, UPMC 4 Place Jussieu, 75252, Paris, France}

[3]{Department of Atmospheric Physics, Faculty of Mathematics and Physics, Charles University in Prague, Ke Karlovu 3, 121 16 Prague, Czech Republic}

[4]{Cooperative Institute for Research in Environmental Sciences, University of Colorado, 216 UCB, Boulder, Colorado 80309, USA}

[5]{National Oceanic and Atmospheric Administration (NOAA) Earth System Research Laboratory, Chemical Sciences Division, 325 Broadway R/CSD, Boulder, Colorado, 80305-3337, USA}

[6]{Max Planck Institute for Meteorology, Bundesstraße 53, 20146, Hamburg, Germany}

[7]{Institut Pierre Simon Laplace des sciences de l'environnement, UPMC 4 Place Jussieu, 75252, Paris, France}"

32. *) Please spell out "BVOC" first time this acronym is used (first line in abstract) and not in the intro. Same comment for "PFT" (P33969L21), which is currently first spelled out in the intro.

Authors: Text corrected.

33. *) P33969L27: It took me some time before I realised that you scaled LAI by 0.5 and 1.5. Please write this more clearly.

Authors: We reformulate the text in the Abstract:

"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."

34. *) P33970L2: "to variation of LDF" → "to variation in the LDF".

Authors: Text corrected.

35. *) P33970L26: "*largely*" → "*widely*".

Authors: Text corrected.

36. *) At many places, you mix UK and US English. Please homogenise.

Authors: The text checked and homogenise.

37. *) P33971L11: delete "a" and replace "variation" by "variations".

Authors: Text corrected.

38. *) P33971L15: Don't spell out "EF" again, you have already done so. Same goes for "LAI" on P33972L19. In general: please check the whole manuscript for places where you have spelled out acronyms more than once.

Authors: Text checked and corrected.

39. *) P33971L17: delete "is a key emission driver", since that does not make sense – it IS the actual emission at standard conditions. Also delete "a" in front of "large variability".

Authors: Text corrected.

40. *) P33971L28: "as one PFT can actually correspond to" → "as one PFT is actually corresponding to", since there are always more than one species covered in a PFT.

Authors: Text corrected.

41. *) P33971L27-P33972L1: this sentence sounds broken or that something is missing.

Authors: Text changed in section 1 as follows:

"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)."

42. *) P33972L25: Replace "can affect" with "affects" and add "modelled" or "predicted" or "calculated or similar in front of "regional and seasonal distribution".

Authors: Text corrected in section 1 as follows:

"Consequently, the high uncertainty related to LAI affects the predicted regional and seasonal distribution of BVOC emissions."

43. *) Is there no reference to STOMATE available?

Authors: The references Krinner et al., 2005 and Magnan et al., 2011 cover both STOMATA and SECHIBA module. We have shifted their position so that it is clearer (section 2.1).

"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:…"

44. *) P33975L4: What is "LPJ"? Is it the LPJ model that provides the surface areas provided for ORCHIDEE in Table 1?

Authors: For our study (see also point 10), the global vegetation distribution is prescribed for the runs using appropriate forcing (the Land-Use History - LUHa.rc2) related to the year 2000 as described in section 2.4. These files provide the surface areas in Table 1. The module mentioned is a part of ORCHIDEE and related to the dynamical vegetation component (LPJ). We did not activate this part. We therefore delete this paragraph that is misleading and we keep only, in section 2.1:

"For our study, the global vegetation distribution is prescribed for all runs using appropriate forcings, as described in paragraph 2.4."

45. *) P33975L16: Add "The" before "canopy". "Divided in up to 17 LAI layers" - this sounds very weird – I guess you mean that you split the total LAI into different canopy layers – please reformulate.

Authors: We reformulated the text in section 2.2.

"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."

46. *) P33975L25: Add "the" before "leaf level". Replace "in the" with "at". Now I will try to stop making note on this grammar stuff – please check it yourself. There are many following mistakes.

Authors: Text checked and corrected.

47. *) P33976L3: Add "the" before "emitted". Since CTL depends on the emitted compounds, why is it not CTL*i* instead?

Authors: We changed it. We also changed notation in equation (1) and (2) in order to be clearer (see point 51).

48. *) P33977L1: "in order to take into..."???

Authors: The phrase is deleted because the concept is explained shortly before.

49. *) P33977L3: "needleaf" is misspelled here and later.

Authors: Changed in the text.

50. *) P33977L8: "2-Methyl-3-Buten-2-Ol" → "2-methyl-3-buten-2-ol".

Authors: Changed in the text

51. *) P33978L6-10: I am very confused by your symbols. Is CTLI in Eq. 1 then not supposed to be CTL(l)?

We changed the use of notation in equation (1) and (2) and the text in section 2.2, to be clearer.

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

$$F_{c,i}(l) = LAI_i(l) \cdot SLW_i \cdot EF_{c,i} \cdot CTL_c(l) \cdot L_c \quad (2)$$

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…"

"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):

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

LDF_c is the light-dependent fraction of the emission, specified for each compound emitted (Table 2)."

52. *) P33978L13-18: Either you write out the meaning of the symbols, or you leave out the equations and only refer to Guenther et al., 1995.

Authors: We deleted the equations.

53. *) P33980L20+L24: "2" and "1" → "two" and "one". Other places also with same mistake.

Authors: We changed the text.

54. *) P33981L12: Any reference or website to CRU-NCEP?

Authors: We inserted the website in section 2.4 (page 16).

"(http://dods.extra.cea.fr/data/p529viov/cruncep)"

55. *) P33982: "Ls"??? You have used Lc before – is that now the same?

Authors: The Referee #1 is right, we write now *"L_c"*

56. *) P33983L19-20: compounds does not start with a capital letter – please change it here and also other places in the text where you wrote it like that.

Authors: We changed it in the text.

57. *) P33988L19: Sure it's not "western Brazil"?

Authors: The Referee #1 is right. We changed the text.

58. *) P33989L1: "...can observe comparing..."?

Authors: The sentence does not exist anymore.

59. *) P33989L6-9: You start and end the line with emphasising that this is important for light dependent emission – I think you don't have to add "in the case of BVOCs that are strongly light dependent".

Authors: We deleted it in the text.

60. *) P33989L21-22: Don't write about the "solid black line" and "red line", since you confuse the reader, cause there are no such lines in Fig. 4.

Authors: We deleted it in the text.

61. *) P33990L20: "for each" → "with each".

Authors: Text corrected.

62. *) P33990L24-27: Please add reference to the figures that shows this.

Authors: The sentence does not exist anymore.

63. *) P33994L9: "non light-dependent" = "light independent".

Authors: Text corrected

64. *) P33995L14-20: maybe past tense works better.

Authors: We changed the text in:

"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."

65. *) Table 5: "Modis Lai" = "MODIS LAI"? You must have a mistake in the LAI info column for the simulations where you multiply LAI with 1.5! Is it really so that you used the air temperature for MEG_LDF and not the leaf temperature?

Authors: The LAI provided by MODIS is used as forcing in the MEG_CRU simulation. We corrected the mistakes related to ORC_LAI15, MEG_LAI15 in LAI column. We indeed use the leaf temperature in MEG_LDF simulation. We put "*T leaf*" in the T column.

66. *) Figure 11: The unit is not supposed to be in italic? From your figure text "The thick and thin dashed line represent..." → "The thick and thin dashed lines represent...". Maybe also worth to mention that the LAI peaks at different times in ORCHIDEE and MEGAN and why this is so. Also, Fig. 11 should be listed before Fig. 10, since it is mentioned in the text before Fig. 10.

Authors: We corrected the figure and related text and we reversed the order of Fig. 10 and 11.

We mention in section 3.4 (page 27) in the text that LAI peaks at different times in ORCHIDEE and MEGAN, giving a possible explanation.

"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 most likely 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."

67. *) Figure 10: Please include the results from MEG_CRU in this figure too. It helps for the comparison.

Authors: Figure changed.

References from Referee #1:

Ghirardo et al., Plant, Cell and Environment, 33, 781, 2010.

Mann et al., Atmos. Chem. Phys, 14, 4679, 2014.

Owen et al., Atmos. Environ., 36, 3147, 2002.

Spracklen and Righelato, Biogeosciences, 11, 2741, 2014.

Tsigaridis et al., Atmos. Chem. Phys., 14, 10845, 2014.

References in the answers:

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010.

Responses to Referee #2's Comments

We thank Referee #2 for his/her time and consideration. We closely examined his/her insightful and constructive comments which have considerably helped us to improve the manuscript.

Referee #2's comments are quoted in bold. Authors' answers are in regular font and authors' changes in the manuscript are quoted in italic.

We refer to the marked-up manuscript version for section numbers and pages.

This is an interesting manuscript presenting an alternative global emission model to MEGAN as well as some insights into the mechanisms of such models. It is well suited to the journal and overall well written although grammar and language should be improved in some occasions. There are, however, a couple of open questions/problems that should be clarified along with a number of minor issues before accepting it for ACP.

Author: We checked the text, also following the Referee #1's remarks, in particular the use of article "the", the present and past tenses, some prepositions (in, at, for, with…), the misspelled of needleleaf. We uniform all the text in UK English. The manuscript was read by two English native speakers to reduce, as much as possible, the grammar mistakes.

First, I disagree with the argument that the current available information about Emission Factors is sufficient for statistical significance as stated in the introduction (P33971, L19). As far as I can see this is only valid for very few species while for many others only very few measurements can be found. The question is, however, if the available EFs are sufficient to characterize the representative species for a PFT. Although the authors point out the difficulties of PFT parameterisation (and among these I miss the one that PFTs are of variable species composition) they are obviously of the opinion that they have overcome these difficulties. But how were these PFT specific EFs actually derived? All what is presented is Table 3 showing one EF per PFT and a list of references with varying detail. I would like to illustrate this point: In the ORCHIDEE model description, the authors say they have determined an isoprene emission factor of 0.5 for the boreal needleleaf deciduous PFT (=Larches) based on Levis et al. 03 (EF 0.0), Guenther et al. 06 (EF 0.7), Karl et al. (EF 0.0), Steinbrecher et al. 09 (EF 0.0), and Steinbrecher et al. 13 (only oaks in here). So how does this work out? One of the problems seems to be that only secondary sources are used which in

turn partly use the same original investigations. It would be more logical to fall back on primary literature sources – preferably new ones or at least complemented by new ones (e.g. Ruuskanen et al. 07, Ghirardo et al. 2010). So, which measurements from which species were used to derive which PFT and how is it done? This is probably an issue for a supplement.

Author: We agree with the Referee #2. Our argumentation is overstated considering the methodology used in our paper. Nowadays we can only say that there are more observations and a larger number of compounds measured. We change the text consequently in section 1, page 4 (see §1).

Assigning EFs, especially for the global scale purposes, is a very tricky issue, and the methodology to be used is still under debate within the scientific community. In an ideal case, for each compound emitted, we should consider 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 getting an average EF for each PFT and emitted compound. Unfortunately, there are not yet enough observations available to use such a methodology.

The approach used to derive EFs is not fully detailed in the manuscript. It represents an important part of the upgrade of the emission module, but it is not the central issue of the work. All values used are available in the papers and listed in Table 3. We do not use a new statistical methodology or consider original measurement that deserves to be particularly detailed. Actually we use a qualitative and comparative method to attribute the EF values. However, we agree with the Referee #2 that it is important to be more precise in the manuscript, in order to better understand the procedure used and the difficulties encountered in assigning the new EFs. We specify more clearly why, at present, in the modelling community, statistically valid methods to assess the EF have not been developed yet. To put more attention on this issue we dedicate the new section 2.2.1 (see §2).

About the isoprene EF related to boreal needleleaf deciduous tree, we consider the following EF values, that we converted (if necessary) into the proper units used in ORCHIDEE:

- EF = 0.44 μgC g^{-1} h^{-1} (in original unit: 0.5 μg g^{-1} h^{-1}) in the supplement of Steinbrecher et al. (2009), (Table 2). The values come from an extensive review.
- EF = 0.44 μgC g^{-1} h^{-1} (in original unit: 0.5 μg g^{-1} h^{-1}) in Karl et al. (2009) (Table 5). They based their values on Steinbrecher et al. (2009).
- EF = 0.0 μgC g^{-1} h^{-1} in Levis et al. (2003). They assign for needleleaf deciduous trees the emission rates recommended for larch species by Guenther et al. (1994).

- EF = 8.0 μgC g^{-1} h^{-1} in Fu and Liao. (2012) taken from the literature, but they do not say from where.
- EF = 1.44 μgC g^{-1} h^{-1} (in original unit: 0.7 mg m^{-2} h^{-1}) in Guenther et al. (2006). They assign an average over all needleleaf deciduous tree.
- EF = 0.09 μgC g^{-1} h^{-1} (in original unit: 0.1 μg g^{-1} h^{-1}) in Smiatek and Steinbrecher (2006). The value is for larch only.
- EF = 0.002 μgC g^{-1} h^{-1} (in original unit: 1 μg m^{-2} h^{-1}) in Guenther et al. (2012). The value comes from a review.
- EF = 0.52 μgC g^{-1} h^{-1}(= 221 μgC m^{-2} h^{-1}) in Klinger et al. (2002). The value comes from measurements.
- EF = 1.44 μgC g^{-1} h^{-1} in Lathière et al., 2010. They use the same value as in Guenther et al. (2006).
- EF = 8.0 μgC g^{-1} h^{-1} in Arneth et al. (2011). They adopt this value for the model inter-comparison.

In these papers the values, which come from a review or measurements, are: Guenther et al. (2006) (EF = 1.44 μgC g^{-1} h^{-1}), Guenther et al. (2012) (EF = 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 more lower than the ones assigned by Lathière et al. (2006) (EF = 8.0 μgC g^{-1} h^{-1}) and the 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 because they based their choice only taking into account other previous model setting or the source of information was not clear. We report a part of this discussion, as an example of the procedure used in section 2.2.1 (see §2).

In our review we did not consider the paper of Ruuskanen et al. (2007). They assign a contribution of less than 3% of the VOC emission to isoprene, 2-methyl-3-buten-2-ol (MBO) and 1,8-cineole, for larch. Anyway, they confirm that boreal needleleaf deciduous PFTs, which are mostly composed by larch, are very low isoprene emitters. We mention this work in section 2.2.1 (see §2).

§1 *"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 section 2.2.1."*

§2 *"**2.2.1. Emission Factors 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 $\mu gC\ g^{-1}\ h^{-1}$ in the old version and EF = 0.5 $\mu gC\ g^{-1}\ h^{-1}$ 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 $\mu gC\ g^{-1}\ h^{-1}$), Guenther et al. (2012) (0.002 $\mu gC\ g^{-1}\ h^{-1}$), Steinbrecher et al. (2009) (EF = 0.44 $\mu gC\ g^{-1}\ h^{-1}$), and Smiatek and Steinbrecher (2006) (EF = 0.09 $\mu gC\ g^{-1}\ h^{-1}$) and Klinger et al. (2002) (EF = 0.52 $\mu gC\ g^{-1}\ h^{-1}$). All these values are much lower than those assigned by Lathière et al. (2006), and their average is 0.5 $\mu gC\ g^{-1}\ h^{-1}$, 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 (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 $\mu gC\ g^{-1}\ h^{-1}$, while before EF = 8 $\mu gC\ g^{-1}\ h^{-1}$), 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 2–methyl–3–buten–2–ol (hereafter we refer to it simply as 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).”

Second, I am a bit surprised that LAI is more or less stated to be wrong in ORCHIDEE already in 2011 (P33990, L1) but has not been improved since although the deviation to measurements is very large and it is discussed (and demonstrated) to be a very important driver for emission. There is a bit of discussion about uncertainties in measurements but I feel that the paper doesn’t dare to claim that the ORCHIDEE simulations are as valid as the MODIS derived values. However, if the MODIS data are considered ‘state of the art’, then I see three options to proceed: 1. Improve the LAI simulations, 2. Improve the argumentation to a degree that the reader can accept ORCHIDEE simulations as equally likely as MODIS data, or 3. Run all simulations with MODIS derived values only. Option 3 seems the most feasible to me.

Author: Our current state of knowledge does not allow us to say which of the two methods give more realistic LAI values: the retrieval from the MODIS satellite or ORCHIDEE calculation. The satellite actually measures the *effective* LAI and not the *real* LAI (Pinty et al. 2011; Fang et al. 2012a; Fang et al., 2012b). LAI is obtained from indirect optical methods and strongly determined by the *a priori* assumptions necessary to perform the inversion procedure. We detailed the

uncertainties of LAI satellite measurements and ORCHIDEE estimation in the first part of section 3.4, pages 28-29 (see §3).

In addition, we mention that ORCHIDEE is designed to provide future scenarios of emissions from vegetation, studying the links among climate, the plant phenology and emissions. Therefore, the main concern is rather to study weaknesses in LAI modelling and eventually improve it and not to force it with other LAI estimates. A new version of ORCHIDEE model is going to be developed, including a more detailed hydrological scheme, a complete nitrogen cycle and an higher number of forest PFT, where any possible weaknesses of LAI estimates could be solved. We specify it more clearly at the end of section 1 (see §4).

§3 *"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."

§4 "*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."*

Third, I would like to see a bit more model descriptions and information about setups. For ORCHIDEE, the activity factor is mentioned to depend on leaf age but it is not clear how it is derived and how it is different for different PFTs? It is not used in the comparisons of model simulations although it may pose a difference to MEGAN, particularly if it is decreasing the emission of PFTs with high leaf longevity. Furthermore, it is clear that drought and CO2 is changing in the simulations but it is not clear if one or both are considered for emission calculations. Regarding MEGAN, respective functions exist as options because emission is quite sensitive to both (e.g. Seco et al. 15, Acosta-Navarro et al. 14). With respect to the setup, I think that given the large differences in the PFT covered areas between the MEGAN and ORCHIDEE runs it would make sense running the models with each other's land-cover scheme to demonstrate the effect of this issue separately.

Author: At the end of section 2.4 we add more information about L_c and leaf age, telling which values are used, for which species and citing related references (see §5). Even if the absolute values

are different, the L_c choice in the two models is quite similar, as both models consider higher emissions for new and young leaves for methanol and lower emissions for isoprene. Considering leaf class, things are more complex. Leaf age classes, in MEGAN, are derived considering the variation between LAI value of the current and preceding month, following an highly parameterised scheme. In ORCHIDEE leaf age classes are calculated considering the plant leaf growth and leaf turnover at each model time step (30 minutes) and are not directly correlated with LAI. The comparison between these two variables and the implementation of sensitivity tests to assess the impact of these different approaches are not straightforward. It would be a very interesting investigation and we mention it in section 5, page 39, as future development of this work (see §6).

Furthermore, we add some more information about the humidity/drought effect and about the CO_2 inhibition factor in influencing emission (see §5), as they are taken into account in the two models.

Considering the last point (running the models with each other's land-cover), the aim of the paper is to compare the two models, putting them under the same forcing conditions, but retaining their own particular characteristics (detailed now in new section 2.5 of paper): the emission scheme, parameterization setting, PFT distribution, radiative scheme. We stress this point in section 1, page 7 (see§7). In addition, PFT distribution is not interchangeable without significantly modifying the models since PFT classification are not the same in the two models. We clearly detail it in the new section 2.5, page 19, point 3 (see §8).

§5 *"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."*

§6 *"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."

§7 *(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 section 2.5), in particular the emission scheme, classes and distribution of PFTs and LAI processing,…"*

§8 *"…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; "*

In addition, I would recommend avoiding repetitions throughout the manuscript (e.g. P33977 last paragraph, P33983 L18/19, P33996 last paragraph) and re-structure the analysis of LAI impacts, i.e. differentiating more clearly between the effect of size vs. dynamic and between emission area and light (and temperature) modifying impact (see also Keenan et al. 11). In this context, it is perhaps critical to state that some LAI are so large that there 'is no more light available' (P33992, L17). If this would be true, photosynthesis couldn't work and leaves wouldn't make any sense at all.

Author: We reformulate the text in section 2.2, page 11 (see §9), section 3.1, page 21 (see §10) and section 4, page 37 (see §11) to avoid repetition.

We re-structure the LAI analysis differentiating more clearly the discussion on LAI uncertainties, impact of LAI seasonal cycle and size on BVOC emission estimates (see the manuscript, section 3.4), adding two sub-sections: *"3.4.1 LAI seasonal cycle impact"* and *"3.4.2 LAI size impact"*.

About the statement *"there is no more direct light available"*, we omitted the essential word *"direct"*. The phrase is indeed incorrect without it. In section 3.4.2, page 32, we change the text accordingly (see §12).

In addition, we mention the very interesting work of Keenan et al. (2011) in section 1 (see §13).

§9 *"As detailed in section 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."*

§10 *"(considering the speciated monoterpenes accounted in this work)"*

§11 *"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}."*

§12 *"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."*

§13 *"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."*

References from Referee #2:

Ghirardo A, Koch K, Taipale R, Zimmer I, Schnitzler J-P, Rinne J. 2010. Determination of de novo and pool emissions of terpenes from four common boreal/alpine trees by 13CO2 labelling and PTR-MS analysis. Plant, Cell & Environment, 33: 781-792.

Keenan T, Grote R, Sabaté S. 2011. Overlooking the canopy: The importance of canopy structure in scaling isoprenoid emissions from leaf to canopy. Ecological Modelling, 222: 737-747.

Ruuskanen TM, Hakola H, Kajos MK, Hellen H, Tarvainen V, Rinne J. 2007. Volatile organic compound emissions from Siberian larch. Atmospheric Environment, 41: 5807- 5812.

Seco R, Karl T, Guenther A, Hosman KP, Pallardy SG, Gu L, Geron C, Harley P, Kim S. 2015. Ecosystem-scale VOC fluxes during an extreme drought in a broad-leaf temperate forest of the Missouri Ozarks (central USA). Global Change Biology, 21: 3657-

3674.

References in the answers:

Guenther, A., P. Zimmerman, and M. Wildermuth, Natural volatile organic compound emission rate estimates for U.S. woodland landscapes, Atmos. Environ., 28, 1197– 1210, 1994.

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), ~~including~~ 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 ~~For the~~ 2000~~–~~2009 period, using this model, we estimate ~~with this model,~~ 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 ~~between the two models~~. 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~~with the~~ ORCHIDEE emissions are~~being~~ higher by 21 Tg C yr^{-1}, and for monoterpenes, where they are ~~being~~ 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. ~~When the~~ LAI is scaled by a factor of 0.5 and ~~(~~1.5~~)~~, changing the isoprene global emission by ~~change is~~ –21% and ~~(~~+8%~~)~~ for ORCHIDEE and –15% and ~~(~~+7%~~)~~ for MEGAN ~~regarding isoprene~~, and affecting the global emissions of monoterpenes by ~~is~~ –43% and ~~(~~+40%~~)~~ for ORCHIDEE and –11% and ~~(~~+3%~~)~~ for MEGAN. ~~regarding monoterpenes.~~ We find that MEGAN is more sensitive to variation in the ~~of~~ LDF parameter than ORCHIDEE. Our results highlight the importance and the need to further explore the BVOC emission estimate variability and the ~~interest~~ potential for~~of~~ using models to investigate the estimate uncertainties.

1 Introduction

The terrestrial biosphere emits large amounts of Volatile Organic Compounds (VOCs)~~,~~ in particular terpenoids, such as isoprene, monoterpenes, and sesquiterpenes, and oxygenated hydrocarbons~~,~~ such as methanol, acetone, formaldehyde, acetaldehyde, acetic acid, or formic acid (Laothawornkitkul et al, 2009; Guenther et al., 2012; Penũelas and Staudt 2014). On~~At~~ the global scale, the ecosystem contribution to VOC emissions is significantly higher than the anthropogenic one, and accounts for 75–90% of~~to~~ the total emission (Guenther et al. 1995; Lamarque et al., 2010). ~~Biogenic VOCs (~~BVOCs~~)~~ play a central role in atmospheric chemistry, influencing the oxidative capacity of the atmosphere (Arneth et al., 2011; Taraborrelli et al., 2012), ~~leading to the formation and growth of Secondary Organic Aerosols (SOA) (Kanakidou et al., 2005; Goldstein and Galbally, 2007; Van Donkelaar et al., 2007, Acosta Navarro, et al., 2014.),~~ leading to the production of tropospheric ozone in the presence of nitrogen oxides (Von Kuhlmann et al., 2003; Mao et al., 2013), and influencing the tropospheric carbon monoxide budget (Pfister et al., 2008). 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).

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

(i) an empirical method~~approach~~, 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*); ~~and~~

(ii) a processed-based approach, where emissions are linked to the photosynthetic electron transport rate in chloroplasts (Niinemets et al., 2003a, b; Sitch et al., 2003; Keenan et al., 2009; Schurgers et al., 2009; Pacifico et al. 2011; Unger et al. 2013).

The models discussed in this study belong to the first category of bottom-~~-~~up models.

BVOC emission modelling at the global scale is a complex issue, especially because of the number of variables and processes influencing the emission of these compounds, generally characterized by ~~a~~ strong temporal and geographical 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 ~~emission factor (~~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), ~~is a key emission driver and~~ shows ~~a~~ large variability from one plant species to another. Nowadays, a~~The~~ large number of measurements is~~now~~ available for different plants and at~~in~~ various sites ~~around the world makes it possible to assign statistically significant EF values~~ and~~. Furthermore,~~ there is an increasing number of field campaigns that ~~now~~ investigate, in addition to isoprene and bulk monoterpenes, many other important compounds for atmospheric chemistry, especially regarding the SOA formation, such as speciated monoterpenes and sesquiterpenes. More data and information are therefore available, allowing~~giving the possibility to have~~ 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 ~~plant functional type (~~PFT~~)~~, where one PFT represent~~sing~~ a group of plants having the same phylogenetic, phenological and physical characteristics (Prentice et al., 1992). ~~Choosing~~ The choice of one single value for each PFT is especially difficult, as ~~one~~ each PFT ~~can~~ actually corresponds to several plant species, and ~~knowing that~~ EFs show, in general, a wide range of values among ~~the~~ different plants (Kesselmeier and Staudt, 1999;

Niinemets et al., 2011). Moreover, several measurements even show that the emission factors are significantly sensitive to many processes, and parameters that are difficult to isolate and linked to plants stress, such as drought periods, ozone exposure, insects, herbivores and pathogen attack (for a review see Laothawornkitkul, et al., 2009 and Niinemets et al., 2010), making it more delicate to set EFs even forto 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 it is done for isoprene, withand the remaining fraction dependenting only on temperature. Nevertheless, there is no general agreement on the exact value of this temperature and light dependent fraction. 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 ~~leaf area index (~~LAI~~),~~ ~~that~~ 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~~, can be noted~~ between the LAI estimated by measurements and the LAI calculated by models, with discrepancies of up to 100% at the global scale and ~~to~~ more than 150% for specific biomes types (Garrigues et al., 2008; Pinty et al., 2011; Fang et al., 2012a, b). Consequently, the high uncertainty related to LAI ~~can~~ affects the predicted regional and seasonal distribution of BVOC emissions.

According to our knowledge, most papers investigating ~~the~~ BVOC emission sensitivity focus on the response of emissions to different experimental set-ups, changing, for instance, climate forcing and land use. For example, Oderbolz et al.~~,~~ (2013) pointed out the importance of the differences between~~among~~ the land-cover inventories, and of the uncertainties ~~o~~in the classification of land cover ~~pixel~~. Arneth et al. (2011) compared three vegetation models, changing the experimental set-up, such as the vegetation distribution and the climate forcings. Depending on the experiment considered, the total annual isoprene emissions were found to increase or decrease by more than 30%. Ashworth et al. (2010) investigated the impact of varying the climate forcing temporal resolution of~~n~~ 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 ~~performed~~ several simulations were performed with the MEGAN model to assess the sensitivity of isoprene emissions to many parameters and processes such as LAI, emission~~s~~ 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% ~~of isoprene emissions~~ at the global scale.

In the present work, our objectives are to (i) ~~to~~ present the updated version of the emission module embedded in the dynamic global vegetation model ORCHIDEE, (ii) ~~to~~ provide present-day estimates of global BVOC emissions for several relevant compounds (isoprene, monoterpenes, sesquiterpenes, methanol, acetone, formaldehyde, acetaldehyde, acetic acid, formic acid and the main speciated monoterpenes) using the new emission scheme, (iii) ~~to~~ 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) ~~to~~ 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. ~~Because of the lack of observations at the global scale, the best possible way to evaluate emissions models is to perform model inter-comparison and model sensitivity experiments, allowing to deeply investigate critical pattern of simulated variables and to identify the origin of uncertainty. Through model inter-comparison and sensitivity tests, we assess the limitations and uncertainties of BVOC emission estimates related to some key parameters/variables, investigating the origins of the limitation.~~ 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 directions ~~and discussion of the study~~ are 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)~~, (Krinner et al., 2005; Magnan et al., 2011)~~.

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

In ORCHIDEE, ecosystems are represented by 13 Plant Functional Types (PFTs, listed in Table 1). Each PFT is representative of a specific set of plant species that are grouped

according to plant physiognomy (tree or grass), leaf shape (needleleaf 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. ~~Activating the LPJ dynamical vegetation component of the model (Krinner et al., 2005), ORCHIDEE has also the feature to determine the surface and distribution of natural vegetation types according to climate and CO_2 conditions considered, while location and surfaces of agricultural PFTs are always prescribed. For our study, the global vegetation distribution is prescribed for all runs using appropriate forcings.~~

2.2 BVOCs in ORCHIDEE: module improvements

~~In order to consider recent findings regarding emission schemes and field measurements, the~~ 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.~~Canopy is divided in up to 17 LAI layers (the layer number depends on the LAI value) and emissions are calculated for each layer considering the sunlit and shaded leaf fractions and the light extinction and light diffusion through the canopy and are then vertically summed, providing a single value for each PFT and grid point.~~

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

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

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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 gdmg m^{-2}, $EF_{c,i}$ is the basal emissions at the leaf level for an individual compound and PFT in theat standard conditions of temperature (T = 303.15 K) and photosynthetically active radiation (PAR = 1000 μmol m^{-2} s^{-1}), expressed in μgC gdmg^{-1} h^{-1}. Note that in the ORCHIDEE model, EFs depend on both the PFT and the emitted compounds considered.

$CTL_{TL,c}$ 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 original version are given in Lathière et al. (2006).

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

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 for their importance in the atmospheric chemistry, in particular regarding the secondary organic aerosol formation.

EFs represent one of the greatest sources of uncertainty in the quantification of BVOC emissions (Niinemets et al., 2011). Several measurement campaigns were carried out over the last decade, giving important insights and information to re-examine thoroughly the emission factors used in the emission module and correct them accordingly. Based on an extensive

review of publications, we selected the field measurements that report EFs in the standard conditions of PAR (1000 μmol m^{-2} s^{-1}) and temperature (30 °C). The collected EFs were gather per PFTs and then averaged to obtain a single value per PFT, rejecting the outliers.

In Table 3 we report the new and old EFs used in the emission module and in Table 4 we present EF value for each speciated monoterpene as a percentage of the bulk monoterpene EF value. As shown in Table 3, the revision leads to the modification of almost all EFs, in order to take into account recent findings demonstrated by field measurements. In some cases the EF differences in comparison with the previous version, are very significant. Regarding isoprene, boreal needleaf deciduous PFT is now recognized as a less important emitter (EF = 8 μgC gdm^{-1} h^{-1} in the old version and EF = 0.5 μgC gdm^{-1} h^{-1} in the new one) (Levis et al., 2003; Guenther et al., 2006, 2012; Karl et al., 2009; Steinbrecher et al, 2009; Steinbrecher et al, 2013) For monoterpenes, a significantly higher EF (from 0.8 μgC gdm^{-1} h^{-1} to 2.2 μgC gdm^{-1} h^{-1}) is now assigned to tropical broadleaf evergreen and deciduous PFTs and for 2-Methyl-3-Buten-2-Ol (MBO), the EF for the temperate needleaf evergreen PFT is reduced from 20 μgC gdm^{-1} h^{-1} to 1.4 μgC gdm^{-1} h^{-1} (Tarvainen et al., 2005; Hakola et al., 2006; Chang et al., 2009; Kim et al., 2010).

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

We mentioned that, tThe 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 dependenting on both light and temperature, while the others were only dependenting on temperature. Most recent field campaigns highlight, for a large

~~number of plants, the dependency of monoterpenes, sesquiterpenes and oxygenated BVOC emissions on radiation as well (Jacob et al. 2002, 2005; Hansen and Seufert, 2003; Dindorf et al., 2006; Holzke et al., 2006; Harley et al., 2007; Millet et al., 2008, 2010; Hu, et al., 2011; Šimpraga et al., 2013; Wells et al., 2014). In the new emissions module, we therefore take into account these findings adopting the approach described in Guenther et al. (2012).~~ 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~~ing~~ both light~~-~~ dependent ~~(directly release through stomata)~~ and light~~-~~independent ~~(stored in the leaf pool)~~ emission processes, and the response to temperature and light (C$TL_{~~TL~~}$) is calculated for individual compounds~~,~~ at each LAI layer (l)~~, as~~:

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

~~Where~~ 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).~~,~~ C$TLI_{~~TLI~~}$ is the temperature-dependent emission response ~~for the emission part~~ that is not light dependent and depends on individual compounds.~~,~~ C$TLD_{~~TLD~~}$ and C$L_{~~L~~}$ 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).

C$TLI_{~~TLI~~}$ is equal to:

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

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

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

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

Commento [PM1]: We delete these two formula, referring only to Guenther 1995

Where β is the empirical coefficient of the exponential temperature response and it is now defined as in Guenther et. al (2012) (Table 2). and PAR is the photosynthetically active radiation at each LAI layer. For all the other details of the formula (3-5) we refer to Guenther et al. (1995).

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 the terrestrial vegetation. The basis of the model is a simple mechanistic approach established in by Guenther et al. (1991, 1993, 1995), linking 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 part components defined by the light dependent fraction (LDF). For the 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 the CLM4, the input data can be provided by the CESM atmospheric and land surface models on-line at each time-step. In this work, we use the stand-alone model version of MEGANv2.1,. Hhereafter we refer to it 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 setting 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 ~~used~~ 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. ~~2~~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~~1~~ simulation for the 2000~~-~~–2009 period with MEGAN using the LAI estimated by ORCHIDEE (MEG_CRULAI)
3. four~~4~~ simulations for the year 2006 by both models, using the ORCHIDEE LAI scaled by a factor 0.5 and 1.5, respectively (ORC_LAI05, ORC_LAI15, MEG_LAI05 and MEG_LAI15).

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

The run sets 3 and 4 are carried out for the year 2006, which is estimated as an averaged year regarding the BVOC emissions calculated by MEGAN and 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 ~~in order~~ to balance the leaf stock. The spin-up is based on a 10-year loop using meteorological forcing for the year 1989, followed by a 10-year simulation from 1990 to 1999. In ORCHIDEE, the global vegetation distribution for the 13 PFTs is prescribed using the Land-Use History (LUHa.rc2) related to the year 2000 (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).

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

~~calculated firstly for each vegetation type, considering the LAI simulated online by ORCHIDEE for each PFT and then summed providing an emission value per grid cell, as described in Sect. 2.2. Whereas in MEGAN the vegetated potential is calculated over the grid cell and then it is multiplied by the related LAI, obtaining the emission per grid cell. The annual CO_2 concentration varies along the simulation from a value of 368 ppm for 2000 to 385 ppm for 2009.~~ 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~~, Ls activity factor~~ and the β coefficient values are given in Table 2. For MEGAN, the values of LDF and β are those ~~standard settings, as~~ presented in Table 4 in Guenther et al. (2012)~~, are considered~~.

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, t~~T~~he 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. ~~of the lack of observations available globally.~~ Satellite observations provide very useful information, especially regarding the order of magnitude and the seasonal and regional variability of emissions, but the most abundant VOC species are not directly measured (such as isoprene and monoterpenes). Satellite measurements are also subject to large uncertainties arising from ~~the~~ difficulties in the retrieval of the ~~to retrieve~~ atmospheric concentration of short-lived compounds from space or in separation of ~~to separate~~ the different sources (for instance terrestrial biogenic, anthropogenic, oceanic etc.) and the various compounds themselves. Global emission estimates are generally performed using models, or from the application of ~~applying~~ inverse modelling techniques that combine the measurements (from satellite, ground or aircraft measurements) and models, providing emissions for compounds such as methanol (Jacob et al. 2005; Millet et al., 2008; 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 ~~state of the art~~ 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)~~, with m~~Methanol (106.1 Tg C yr^{-1}) and acetaldehyde (42.2 Tg C yr^{-1}) emissions ~~being~~ are twice as large~~higher~~, and formaldehyde emissions (10.0 Tg C yr^{-1}) ~~being~~ 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 ~~Despite~~ the MEG_CRU simulation was ~~being~~ carried out using the same MEGAN version as~~than~~ 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~~, as climate forcings,~~ 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 ~~list~~ accounted in this work~~, i.e. α-Pinene, β-Pinene, Limonene, Myrcene, Sabinene, 3-Carene, t-β-Ocimene~~) 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~~s~~ estimates (green triangles, first plot of Fig. 1).

~~We present in~~ 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 ~~for~~ 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 ~~with~~ the main compound emitted ~~being isoprene~~ with a global ~~calculated source~~amount of 465 Tg C yr^{-1} for ORCHIDEE, accounting for 61% of total BVOC emissions (estimated to 757 Tg C yr^{-1}), and ~~of~~ 428 Tg C yr^{-1} for MEGAN, accounting for 64% of total BVOCs (estimated at~~to~~ 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 ~~each~~ 1% to 4% of the total BVOCs for both models, while other compounds contribute ~~for~~ 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 ~~considered~~ compound (except MBO). Both models calculate ~~a~~ 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~~, for instance,~~ Ponderosa and Scots pine (Kim et al., 2010; Tarvainen et al. 2005; Harley et al., 1998). The EF values~~,~~ in the ORCHIDEE and MEGAN models~~,~~ are consequently ~~set~~ significant only for the PFTs representing these plants (TeNeEv and BoNeEv), leading to notable emissions in the temperate North latitudes and contributing ~~for~~ 88% for ORCHIDEE and 63% for MEGAN ~~to~~ 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~~with~~ emissions are~~being~~ 21 Tg C yr^{-1} higher in ORCHIDEE. Indeed, the marked seasonal cycle of emissions for northern temperate latitudes implies that the ~~high~~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~~line~~). 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 ~~seasonal and~~ geographical variation ~~of~~in emissions. For the other species the ~~high~~largest differences occur in tropical regions.~~:~~ ~~f~~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 ~~Emission li~~nter-annual and inter-seasonal emission variations

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

Nevertheless ~~high~~ 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, ~~i~~on average, ~~are~~ lower by 11.5 Tg C $month^{-1}$ and 9.0 Tg C $month^{-1}$ compared with~~to~~ ORC_CRU. ~~Fig. 3, where~~ T~~t~~he monthly zonal average for tropical, temperate and northern boreal latitudes regions are shown in Fig. 3.~~depicted, shows that,~~ We observe, as mentioned in Sect. 3.1, that the ORCHIDEE emissions are significantly higher in northern temperate regions compared ~~to~~ with MEGAN, with a marked seasonal cycle and the ~~high~~largest differences between the two models occurring in summer. In ~~particular 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 ~~emitted~~ dominant~~ly~~ from the tropical regions, leading to an overall stable global emission budget throughout the year (Fig. 2). The n~~N~~orthern 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 ~~a~~ strong emitters will have a more marked seasonal cycle (Fig. 2), such as for methanol and isoprene ~~for~~ 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 ~~some~~ others, no seasonal cycle. Based on our current knowledge, we cannot establish which is the best representation~~,~~ because of the lack of observations at the global scale. However, we can extensively ~~deeply~~ investigate why the differences between the two models occur, performing sensitivity simulations and looking at the various processes modelled. This is the main purpose of the next section.

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

Additionally, Fig. 3 shows that ~~I~~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 ~~to~~ with the ecosystem present in the grid cell.

For a particular compound, the formula to convert the ORCHIDEE EF (μgC ~~gdmg~~$^{-1}$ 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):

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

and for light-dependent emission~~s depending on light~~ (LDF = 1):

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

where i is the index related to PFTs, M_{Carbon} and M are the molar mass of carbon and the compound, respectively, LAI_{REF} ~~being~~ 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~~ing~~ on the canopy radiation module, which equals ~~to~~ 0.57 in this MEGAN configuration (Guenther et al., 2012).

In general, for every compound, we observe a similar geographical distribution. High emission areas are identified in Brazil, equatorial Africa, southern East Asia and southern East USA for both models, with values for ORCHIDEE (MEGAN) 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 ~~of~~ 0.1–0.3 10^{10} kg C m^{-2} s^{-1} for MEGAN. Indeed, these regions are populated by temperate and boreal ~~needleaf~~needleleaf evergreen trees, which are strong methanol emitters (Table 3 and Fig. 7, last row~~line~~).

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

Other ~~remarkable~~ notable differences between the two models appear in South America for isoprene, directly in relation with the EP~~s~~ distribution. The pattern of isoprene emission~~s shows,~~ in MEGAN has~~,~~ higher values in western ~~eastern~~ Brazil, Bolivia and northern Argentina, while in ORCHIDEE the values are ~~it is~~ more homogeneous, with higher emissions in central Brazil. The ~~S~~same pattern differences are ~~can be~~ detected in the emission potential (Fig. 5, last ~~line~~ row on the right), and we therefore infer that the EP distribution drives the isoprene emission geographical distribution. The same conclusion ~~is~~ also holds~~held~~ for monoterpenes, where lower emissions along the Amazonian river follow perfectly the lower EPs in this area. In general, comparing the emission geographical distribution for each compound and the corresponding emission potential, we can state that, in both models, emission spatial patterns are mostly affected by the EF and ~~by the~~ PFT distributions. ~~Moreover, we found that the emission variation along the year is mainly driven by the LAI, as we can observe comparing the LAI in winter and summer (Fig. 4) with the compound emitted in the corresponding seasons (Figs. 5-8).~~

~~For the compounds that are fully light dependent, such as isoprene (LDF=1) or largely light dependent, such as methanol (LDF=0.8), we observe that higher EP in ORCHIDEE than in~~

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

3.4 BVOC emission sensitivity to LAI

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

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

Figs 4 and 10 show large differences in ~~T~~the geographical distribution and global average of ORCHIDEE LAI ~~(solid black line)~~ and MODIS LAI ~~(red line)~~ (Yuan et al., 2011)~~, depicted in Figs 4 and 11, exhibit big differences between the two models~~. As illustrated in Fig. 10~~1~~, the global monthly mean LAI calculated by ORCHIDEE is~~ higher by~~ 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, i~~I~~n the tropics, the MODIS LAI exhibits ~~a~~ quite a clear seasonal cycle, especially in Amazonia, Central Africa and Indonesia~~,~~ that is not simulated by ORCHIDEE (Fig. 4). ~~The differences between these estimates of LAI are significant. In a validation study,~~

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

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.

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

~~To fulfil the first objective, we perform a 10-year simulation following the same configuration as in the previous runs, but forcing MEGAN with the ORCHIDEE LAI (MEG_CRULAI simulation, Table 5). This simulation confirms that LAI is one of the key drivers regulating the seasonal cycle)~~. ~~Generally, for every compound, we observe a better agreement between the MEG_CRULAI and the ORC_CRU simulations than between MEG_CRU and ORC_CRU, especially in the tropical regions. In the Fig. 10, where the tropical zonal means of monthly isoprene emissions are depicted, we observe that the variability, along the year, calculated in MEGAN is closer to the one determined by ORCHIDEE. Comparing the MEG_CRULAI (Fig. 10) with the MEG_CRU simulation (Fig. 3, first plot), we can see that the emission peak in northern tropics in April and May and the emission decrease in southern tropics in June and July are less marked.~~

3.4.2 LAI size impact

~~Nevertheless, t~~The global and zonal emission budgets (Table 7) in the MEG_CRULAI simulation are not significantly different than ~~the ones~~those determined in MEG_CRU, even if the ORCHIDEE LAI is significantly higher than MODIS ~~one~~LAI, suggesting a low sensitivity of MEGAN to LAI size~~.~~ Indeed, a~~A~~t the regional scale, in boreal and temperate regions, the MEG_CRULAI emissions are slightly higher than ~~the~~ those in MEG_CRU ~~ones~~, and in the tropics they are even slightly lower for some compounds. As proposed by Sindelarova et al. (2014), a possible reason for the emission decrease calculated in the tropics by MEGAN~~, can be attributed~~is to the strengthened effect of leaf self-shading caused by ~~the~~ 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 ~~of~~ in hourly leaf temperature between sunlit and shaded leaves in the case of dense vegetation (TrBrEv and TrBrDe), we estimate ~~a~~ differences of about 1–1.5 °C, ~~that could~~which would unlikely be high enough to explain such differences in emissions. Lathière et al. (2006), for instance, found that an increase ~~of~~ in the global surface temperature by 1°C leads to an increase of isoprene emissions of at most ~~of~~ 11%. We~~,~~ therefore~~,~~ doubt that the only mechanism~~,~~ behind the ~~not increasing~~static BVOC emissions for light-independent compound~~s,~~ is ~~the~~ leaf self-shading.

~~The LAI is a strong driver of emissions, it is~~We, therefore, ~~important to~~ investigate in more detail~~s~~ ~~if~~ 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).~~In~~ Fig. 12~~,~~ shows ~~we present~~ the four simulations~~: that are performed forcing both MEGAN and ORCHIDEE with the ORCHIDEE LAI (Fig. 11) multiplied by a~~

~~factor of 0.5 (MEG_LAI05 and ORC_LAI05) and by a factor of 1.5 (MEG_LAI15 and ORC_LAI15)~~ 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 ~~S~~southern and northern temperate areas, for isoprene and monoterpenes are displayed, but they are also representative ~~examples for~~ 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 ~~as~~ isoprene is a light-dependent compound thus, beyond a given LAI threshold, ~~,~~ the contribution of ~~lowest~~ the highest LAI layers ~~to emissions~~ is ~~insignificant~~ very low ~~beyond a certain LAI threshold~~, 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. ~~implying much likely that the MEGAN canopy model includes a solar light decrease inside the canopy slightly stronger than in ORCHIDEE.~~ 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 ~~of~~ 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 dependenting on light, such as monoterpenes (LDF=0.6).

~~In~~ Table 8 shows ~~we provide~~ the total emission budget calculated for MEG_LAI05, ORC_LAI05, MEG_LAI15 and ORC_LAI15 simulations for every compound~~s~~. In general in ORCHIDEE, the lower the light dependence, the higher the sensitivity to LAI ~~is~~, while for MEGAN, the sensitivity to LAI does not significantly change with LDF. The explanation for this difference in emission response lies in the different formulation ~~used in the two models~~ for light independent emissions 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}. ~~Indeed in the ORCHIDEE model, the light independent emission from a particular PFT shows a clear linear dependence on LAI, as shown in the Eq. (1) and (2) in the present paper. In MEGAN however, the light independent emission is calculated using, among other factors, γ_{LAI}, which is the factor accounting for the relation between LAI and emission, equal to $(0.49 \cdot LAI)/(1+0.2 \cdot LAI^2)^{0.5}$ (Guenther et al., 2006). This factor increases almost linearly for LAI lower than 2 m^2 m^{-2}, followed by a more moderate increase rate for LAI of 2 – 5 m^2 m^{-2} and is then almost stable 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 ~~, with a more or less high dependency depending~~ on the ~~considered~~ compound (Jacob et al. 2002, 2005; Hansen and Seufert, 2003; Dindorf et al., 2006; Holzke et al., 2006; Harley et al., 2007; Millet et al., 2008, 2010; Hu, et

al., 2011; Wells et al., 2014). ~~As~~ Since the results of these studies are highly heterogeneous, assigning a single LDF value ~~for~~ to each compound is as difficult as assigning the EFs ~~for~~ to each PFT (Sect. 2.2). Hence, the LDF uncertainty could be even higher than the ~~one~~ uncertainties associated ~~to~~ with EFs, as there ~~are~~ have been fewer less quantitative studies ~~about~~ on this subject published to date~~so far~~.

The objective of this ~~paragraph~~ 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 ~~between~~ 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 ~~one~~ defined as ~~non~~ light -independent (LDF = 0) and the second ~~one~~ defined as totally light-dependent (LDF = 1). All EFs are set to 1 µgC ~~gdmg~~$^{-1}$ h^{-1} for ~~every~~ each PFT~~s~~. 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, ~~that~~ 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 ~~to~~ with MEGAN is much higher (up to 20%), than the ~~one~~ index associated ~~to~~ with ORCHIDEE. At the regional scale, in the southern tropics for example (second panel) the index~~it~~ reaches up to 70% and is the ~~double~~ twice as large ~~of~~ the index calculated for ORCHIDEE. The light-dependent part of the emissions in MEGAN is therefore more important than ~~the~~ ORCHIDEE ~~one~~, 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 ~~to~~ with high LDF, as expected from Sect. 3.3. ~~Secondly, as the light dependent and independent part are combined in an additive way to produce emissions Eq. (2), orcldf0 or megldf0 compound can be figured out as the independent part of emission and orcldf1 or megldf1 as the dependent part. According to these results, starting from the same light independent emissions in the two models, if the LDF parameter is increased, the MEGAN emission increases faster than ORCHIDEE ones, as the light dependent component of emission in MEGAN is higher in comparison with ORCHIDEE. MEGAN therefore results more sensitive to LDF variation than ORCHIDEE.~~

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

~~As a more general statement~~More generally, considering the emissions geographical distribution for each compound and the correspondinge~~nt~~ emission potential, we show that, in both models, EF and PFT distribution are the main drivers of ~~emission~~ 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 ~~high~~ large discrepancy in northern temperate regions is attributed to differences in the EF distribution.

The LAI calculated by ORCHIDEE is ~~higher by~~ 1.5–~~-~~2 m^2 m^{-2} higher than the LAI retrieved by MODIS. We examined ~~how~~ what these discrepancies can ~~infer~~ impact on the BVOC estimates~~, exploring the emission sensitivity to LAI~~. Sensitivity tests are then performed forcing both models with ~~multiplying the~~ 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. ~~for isoprene if LAI is multiplied by a factor of 0.5 (1.5), with global emission budget, varying by −21% (+7.8%) with respect to ORCHIDEE reference run and by −14% (+6.6%) for MEGAN. On the other hand~~Conversely, for monoterpenes, ORCHIDEE is much more sensitive to LAI variations, ~~with global emission budget changing by −43% (+40%), while~~ in comparison to MEGAN. ~~presents a global emission budget changing by −12% (+6%).~~ These discrepancies are ~~linked~~ due to differences in the light–independent emission formulation between the two models. In ORCHIDEE ~~considers a linear~~ the dependence of emissions on LAI is linear, while in MEGAN ~~assign~~ for LAI up to 2 m^2 m^{-2}~~a~~ is quasi-linear ~~dependence of emissions for LAI up to 2 m^2 m^{-2}~~, then progressively reducing the increase up to become nearly constant ~~and an asymptotic increase with almost no change~~ 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 ~~Increasing the LDF parameter, the MEGAN emissions increase faster than in ORCHIDEE, implying that the~~ ORCHIDEE ~~MEGAN~~ estimates are more sensitive to LDF variation than– MEGAN~~ORCHIDEE~~. Moreover, we show that overestimation (underestimation) in LDF value leads to emission underestimation (overestimation).

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

~~Models such as ORCHIDEE or MEGAN, among many others, are widely used to investigate biosphere-atmospheric chemistry interactions at different scales, from local to global. Those models are usually built on the same or very similar empirical approaches, but can differ substantially on some aspects, for instance in the number and diversity of plant species taken into account, the EF allocation, the description of the LAI (either simulated, when using a vegetation model, or prescribed), the use of air or leaf temperature to calculate emissions. BVOC emission estimates provided throughout the literature can therefore exhibit different patterns in term of global estimates, geographical or seasonal variation. Field campaigns and laboratory experiments provide very valuable information, enabling the modellers to complement the description of BVOCs in emission scheme, in order to take into account the improvement of our knowledge. Such data are also very challenging to acquire, especially on the long term. The modelling community therefore still faces the problem of BVOC emission model evaluation, with not yet any robust and satisfying way to properly examine our results in term of global numbers or regional/seasonal/inter-annual variations.~~

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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References

Acosta Navarro, J. C., Smolander, S., Struthers, H., Zorita, E., Ekman, a. M. L., Kaplan, J. O., Guenther, a., Arneth, a. and Riipinen, I.: Global emissions of terpenoid VOCs from terrestrial vegetation in the last millennium, J. Geophys. Res. Atmos., 119, 6867–6885, doi:10.1002/2013JD021238, 2014.

Arneth, A., Monson, R. K., Schurgers, G., Niinemets, Ü., and Palmer, P. I.: Why are estimates of global terrestrial isoprene emissions so similar (and why is this not so for monoterpenes)?, Atmos. Chem. Phys., 8, 4605-4620, doi:10.5194/acp-8-4605-2008, 2008.

Arneth, A., Schurgers, G., Lathiere, J., Duhl, T., Beerling, D. J., Hewitt, C. N., Martin, M., and Guenther, A.: Global terrestrial isoprene emission models: sensitivity to variability in climate and vegetation, Atmos. Chem. Phys., 11, 8037-8052, doi:10.5194/acp-11-8037-2011, 2011.

Ashworth, K., Wild, O., and Hewitt, C. N.: Sensitivity of isoprene emissions estimated using MEGAN to the time resolution of input climate data, Atmos. Chem. Phys., 10, 1193-1201, doi:10.5194/acp-10-1193-2010, 2010.

Bai, J., Baker, B., Liang, B., Greenberg, J. and Guenther, A.: Isoprene and monoterpene emissions from an Inner Mongolia grassland, Atmos. Environ., 40, 5753–5758, doi:10.1016/j.atmosenv.2006.05.019, 2006.

Baker, B., Guenther, A., Greenberg, J., Goldstein, A., and Fall, R.: Canopy level fluxes of measurements of 2-methyl-3-buten-2-ol by relaxed eddy accumulation: field data and model comparison, J. Geophys. Res., 104, 26 104–26 114, 1999.

Barkley, M. P., Smedt, I. De, Van Roozendael, M., Kurosu, T. P., Chance, K., Arneth, A., Hagberg, D., Guenther, A., Paulot, F., Marais, E. and Mao, J.: Top-down isoprene emissions over tropical South America inferred from SCIAMACHY and OMI formaldehyde columns, J. Geophys. Res. Atmos., 118, 6849–6868, doi:10.1002/jgrd.50552, 2013.

Bauwens, M., Stavrakou, T., Müller, J.-F., De Smedt, I., and Van Roozendael, M.: Satellite-based isoprene emission estimates (2007–2012) from the GlobEmission project, in: Proceedings of the ACCENT-Plus Symposium, Atmospheric Composition Change – Policy

Support and Science, Session on Short lived pollutants, climate and air quality, Urbino, Italy, 17–20 September, 2013.

Bracho-Nunez, A., Welter, S., Staudt, M., Kesselmeier, J.:Plant- specific volatile organic compound emission rates from young and mature leaves of Mediterranean vegetation, J. Geophys. Res., 116, D16304, doi:10.1029/2010JD015521, 2011.

Chang, K. H., Yu, J. Y., Chen, T. F. and Lin, Y. P.: Estimating Taiwan biogenic VOC emission: Leaf energy balance consideration, Atmos. Environ., 43(32), 5092–5100, doi:10.1016/j.atmosenv.2009.06.038, 2009.

~~Ducoudré, N. I., Laval K., and. Perrier A: SECHIBA, a new set of parameterizations of the hydrologic exchanges at the land-atmosphere interface within the LMD atmospheric general circulation model, J. Clim., 6(2), 248–273, dx.doi.org/10.1175/1520-0442(1993)006<0248:SANSOP>2.0.CO;2~~

Dindorf, T., Kuhn, U., Ganzeveld, L., Schebeske, G., Ciccioli, P., Holzke, C., Köble, R., Seufert, G. and Kesselmeier, J.: Significant light and temperature dependent monoterpene emissions from European beech (Fagus sylvatica L.) and their potential impact on the European volatile organic compound budget, J. Geophys. Res., 111, 1–15, doi:10.1029/2005JD006751, 2006.

Dominguez-Taylor, P., Ruiz-Suarez, L.G., Rosas-Perez, I., Hernandez-Solis, J.M., Steinbrecher, R.: Monoterpene and isoprene emissions from typical tree species in forests around Mexico City, Atmos. Environ., 41, 2780–2790, 2007.

Ducoudré, N. I., Laval K., and. Perrier A: SECHIBA, a new set of parameterizations of the hydrologic exchanges at the land-atmosphere interface within the LMD atmospheric general circulation model, J. Clim., 6(2), 248–273, dx.doi.org/10.1175/1520-0442(1993)006<0248:SANSOP>2.0.CO;2

Duhl, T. R., Helmig, D., and Guenther, A.: Sesquiterpene emissions from vegetation: a review, Biogeosciences, 5, 761–777, doi:10.5194/bg-5-761-2008, 2008.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model

for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, doi:10.5194/gmd-3-43-2010, 2010.

Engelhart, G. J., Asa-Awuku, A., Nenes, A., and Pandis, S. N.: CCN activity and droplet growth kinetics of fresh and aged monoterpene secondary organic aerosol, Atmos. Chem. Phys., 8, 3937-3949, doi:10.5194/acp-8-3937-2008, 2008.

Fang, H., Wei, S. and Liang, S.: Validation of MODIS and CYCLOPES LAI products using global field measurement data, Remote Sens. Environ., 119, 43–54, doi:10.1016/j.rse.2011.12.006, 2012a.

Fang, H., Wei, S., Jiang, C. and Scipal, K.: Theoretical uncertainty analysis of global MODIS, CYCLOPES, and GLOBCARBON LAI products using a triple collocation method, Remote Sens. Environ., 124, 610–621, doi:10.1016/j.rse.2012.06.013, 2012b.

Fang, H., Jiang, C., Li, W., Wei, S., Baret, F., Chen, J. M., Garcia-Haro, J., Liang, S., Liu, R., Myneni, R. B., Pinty, B., Xiao, Z. and Zhu, Z.: Characterization and intercomparison of global moderate resolution leaf area index (LAI) products: Analysis of climatologies and theoretical uncertainties, J. Geophys. Res. Biogeosciences, 118, 529–548, doi:10.1002/jgrg.20051, 2013.

Fares, S., Gentner D.R, Park J.-H., Ormeno, E., Karlik,J., Goldstein, A.H.: Biogenic emissions from Citrus species in California, Atmos. Environ., 45, 4557–4568 doi:10.1016/j.atmosenv.2011.05.066, 2011.

Fu, Y. and Liao, H.: Simulation of the interannual variations of biogenic emissions of volatile organic compounds in China: Impacts on tropospheric ozone and secondary organic aerosol, Atmos. Environ., 59, 170–185, doi:10.1016/j.atmosenv.2012.05.053, 2012.

Garrigues, S., Lacaze, R., Baret, F., Morisette, J. T., Weiss, M., Nickeson, J. E., Fernandes, R., Plummer, S., Shabanov, N. V., Myneni, R. B., Knyazikhin, Y. and Yang, W.: Validation and intercomparison of global Leaf Area Index products derived from remote sensing data, J. Geophys. Res. Biogeosciences, 113(2), doi:10.1029/2007JG000635, 2008.

Gent, P. R., Danabasoglu, G., Donner, L. J., Holland, M. M., Hunke, E. C., Jayne, S. R., Lawrence, D. M., Neale, R. B., Rasch, P. J., Vertenstein, M., Worley, P. H, Yang, Z.-L., and

Zhang, M.: The Community Climate System Model version 4, J. Climate, 24, 4973–4991, doi:10.1175/2011JCLI4083.1, 2011.

Geron, C., Guenther, A., Greenberg, J., Karl, T. and Rasmussen, R.: Biogenic volatile organic compound emissions from desert vegetation of the southwestern US, Atmos. Environ., 40, 1645–1660, doi:10.1016/j.atmosenv.2005.11.011, 2006.

Ghirardo, A., Koch, K., Taipale, R., Zimmer, I., Schnitzler, J.-P., and Rinne, J.: Determination of de novo and pool emissions of terpenes from four common boreal/alpine trees by $^{13}CO_2$ labelling and PTR-MS analysis, Plant Cell Environ., 33, 781–792, doi:10.1111/j.1365-3040.2009.02104.x, 2010.

Greenberg, J. P., Guenther, A. B., Petron, G., et al.: Biogenic VOC emissions from forested Amazonian landscapes, Global Change Biol., 10(5), 651–662, doi: 10.1111/j.1529-8817.2003.00758.x, 2004.

Goldstein, A.H., and Galbally, I.E. : Known and Unexplored Organic Constituents in the Earth's Atmosphere, Environmental Science and Technology, 41, 5, 1514 - 1521, 2007.

Guenther, A. B., Monson, R. K., and Fall, R.: Isoprene and monoterpene emission rate variability: observations with eucalyptus and emission rate algorithm development, J. Geophys. Res., 96, 10799–10808, 1991.

Guenther, A. B., Zimmerman, P. R., Harley, P. C., Monson, R. K., and Fall, R.: Isoprene and monoterpene emission rate variability: model evaluations and sensitivity analyses, J. Geophys. Res., 98, 12609–12617, 1993

Guenther, A, Hewitt, C. N., Erickson, D., Fall, R., Geron, C., Graedel, T., Harley, P., Klinger, L., Lerdau, M., Mckay, W. a, Pierce, T., Scholes, B., Steinbrecher, R., Tallamraju, R., Taylor, J. and Zimmerman, P.: A Global-Model of Natural Volatile Organic-Compound Emissions, J. Geophys. Res., 100(94), 8873–8892, doi:10.1029/94JD02950, 1995.

Guenther, A. B., Karl, T., Harley, P., Wiedinmyer, C., Palmer, P. I., and Geron, C.: Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature), Atmos. Chem. Phys., 6, 3181–3210, doi:10.5194/acp-6-3181-2006, 2006.

Guenther, A. B., Jiang, X., Heald, C. L., Sakulyanontvittaya, T., Duhl, T., Emmons, L. K. and Wang, X.: The model of emissions of gases and aerosols from nature version 2.1 (MEGAN2.1): An extended and updated framework for modeling biogenic emissions, Geosci. Model Dev., 5, 1471–1492, doi:10.5194/gmd-5-1471-2012, 2012.

Hakola, H., Tarvainen, V., Bäck, J., Ranta, H., Bonn, B., Rinne, J., and Kulmala, M.: Seasonal variation of mono- and sesquiterpene emission rates of Scots pine, Biogeosciences, 3, 93-101, doi:10.5194/bg-3-93-2006, 2006.

Hallquist, M., Wenger, J. C., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N. M., George, C., Goldstein, A. H., Hamilton, J. F., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M. E., Jimenez, J. L., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, Th. F., Monod, A., Prévôt, A. S. H., Seinfeld, J. H., Surratt, J. D., Szmigielski, R., and Wildt, J.: The formation, properties and impact of secondary organic aerosol: current and emerging issues, Atmos. Chem. Phys., 9, 5155-5236, doi:10.5194/acp-9-5155-2009, 2009.

Hansen, U. and Seufert, G.: Temperature and light dependence of β-caryophyllene emission rates, J. Geophys. Res., 108, 1–7, doi:10.1029/2003JD003853, 2003.

Harley, P., Fridd-Stroud, V., Greenberg, J., Guenther, A. and Vasconcellos, P.: Emission of 2-methyl-3-buten-2-ol by pines: A potentially large natural source of reactive carbon to the atmosphere, J. Geophys. Res., 103, 25479, doi:10.1029/98JD00820, 1998.

Harley, P., Greenberg, J., Niinemets, Ü., and Guenther, A.: Environmental controls over methanol emission from leaves, Biogeosciences, 4, 1083-1099, doi:10.5194/bg-4-1083-2007, 2007.

Hayward, S., Tani, A., Owen, S. M. and Hewitt, C. N.: Online analysis of volatile organic compound emissions from Sitka spruce (Picea sitchensis)., Tree Physiol., 24, 721–728, doi:10.1093/treephys/24.7.721, 2004.

He, C., Murray, F. and Lyons, T.: Monoterpene and isoprene emissions from 15 Eucalyptus species in Australia, Atmos. Environ., 34, 645–655, doi:10.1016/S1352-2310(99)00219-8, 2000.

Heald, C. L., Henze, D. K., Horowitz, L. W., Feddema, J., Lamarque, J. F., Guenther, A., Hess, P. G., Vitt, F., Seinfeld, J. H., Goldstein, A. H., and Fung, I.: Predicted change in global secondary organic aerosol concentrations in response to future climate, emissions, and land use change, J. Geophys. Res.-Atmos., 113, D05211, doi:10.1029/2007JD009092, 2008.

Heald, C. L., Wilkinson, M. J., Monson, R. K., Alo, C. A., Wang, G., and Guenther, A.: Response of isoprene emission to ambient CO2 changes and implications for global budgets, Global Change Biol., 15, 1127–1140, 2009.

Helmig, D., Ortega, J., Duhl, T., Tanner, D., Guenther, A., Harley, P., Wiedinmyer, C., Milford, J. and Sakulyanontvittaya, T.: Sesquiterpene emissions from pine trees - Identifications, emission rates and flux estimates for the contiguous United States, Environ. Sci. Technol., 41, 1545–1553, doi:10.1021/es0618907, 2007.

Holzke, C., Dindorf, T., Kesselmeier, J., Kuhn, U. and Koppmann, R.: Terpene emissions from European beech (Fagus sylvatica L.): Pattern and emission behaviour over two vegetation periods, J. Atmos. Chem., 55, 81–102, doi:10.1007/s10874-006-9027-9, 2006.

Hu, L., Millet, D. B., Mohr, M. J., Wells, K. C., Griffis, T. J., and Helmig, D.: Sources and seasonality of atmospheric methanol based on tall tower measurements in the US Upper Midwest, Atmos. Chem. Phys., 11, 11145–11156, doi:10.5194/acp-11-11145-2011, 2011.

Hurtt, G. C., Frolking, S., Fearon, M. G., Moore, B., Shevliakova, E., Malyshev, S., Pacala, S. W. and Houghton, R. A.: The underpinnings of land-use history: three centuries of global gridded land-use transitions, wood-harvest activity, and resulting secondary lands, Glob. Chang. Biol., 12(7), 1208–1229, doi:10.1111/j.1365-2486.2006.01150.x, 2006.

Jacob, D. J., Field, B. D., Jin, E., Bey, I., Li, Q., Logan, J., Yantosca, R. M. and Singh, H. B.: Atmospheric budget of acetone, J. Geophys. Res.-Atmos., 107, doi:10.1029/2001JD000694, 2002.

Jacob, D. J., Field, B. D., Li, Q., Blake, D. R., de Gouw, J., Warneke, C., Hansel, A., Wisthaler, A., Singh, H. B. and Guenther, a.: Global budget of methanol: Constraints from atmospheric observations, J. Geophys. Res.-Atmos., 110, 1–17, doi:10.1029/2004JD005172, 2005.

Janson, R., De Serves, C. and Romero, R.: Emission of isoprene and carbonyl compounds from a boreal forest and wetland in Sweden, Agric. For. Meteorol., 98-99, 671–681, doi:10.1016/S0168-1923(99)00134-3, 1999.

Janson, R. and De Serves, C.: Acetone and monoterpene emissions from the boreal forest in northern Europe, Atmos. Environ., 35, 4629–4637, doi:10.1016/S1352-2310(01)00160-1, 2001.

Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G.,Woollen, J., Zhu, Y., Chelliah, M., Ebisuzaki, W., Higgins, W., Janowiak, J., Mo, K. C., Ropelewski, C.,Wang, J., Leetmaa, A., Reynolds, R., Jenne, R., and Joseph, D.: The ncep/ncar 40-year reanalysis project, B. Am. Meteorol. Soc., 77, 437–471, 1996.

Kanakidou, M., Seinfeld, J. H., Pandis, S. N., Barnes, I., Dentener, F. J., Facchini, M. C., Van Dingenen, R., Ervens, B., Nenes, A., Nielsen, C. J., Swietlicki, E., Putaud, J. P., Balkanski, Y., Fuzzi, S., Horth, J., Moortgat, G. K., Winterhalter, R., Myhre, C. E. L., Tsigaridis, K., Vignati, E., Stephanou, E. G., and Wilson, J.: Organic aerosol and global climate modelling: a review, Atmos. Chem. Phys., 5, 1053-1123, doi:10.5194/acp-5-1053-2005, 2005.

Karl, T., Potosnak, M., Guenther, A., Clark, D., Walker, J., Herrick, J. D. and Geron, C.: Exchange processes of volatile organic compounds above a tropical rain forest: Implications for modeling tropospheric chemistry above dense vegetation, J. Geophys. Res. Atmos., 109, D18306, doi:10.1029/2004JD004738, 2004.

Karl, T., Harley, P., Guenther, A., Rasmussen, R., Baker, B., Jardine, K., and Nemitz, E.: The bi-directional exchange of oxygenated VOCs between a loblolly pine (Pinus taeda) plantation and the atmosphere, Atmos. Chem. Phys., 5, 3015-3031, doi:10.5194/acp-5-3015-2005, 2005.

Karl, T., Guenther, A., Yokelson, R. J., Greenberg, J., Potosnak, M., Blake, D. R. and Artaxo, P.: The tropical forest and fire emissions experiment: Emission, chemistry, and transport of biogenic volatile organic compounds in the lower atmosphere over Amazonia, J. Geophys. Res. Atmos., 112, D18302, doi:10.1029/2006JD008539, 2007.

Karl, M., Guenther, A., Köble, R., Leip, A. and Seufert, G.: A new European plant-specific emission inventory of biogenic volatile organic compounds for use in atmospheric transport models, Biogeosciences, 6, 1059–1087, doi:10.5194/bg-6-1059-2009, 2009.

Keenan, T., Niinemets, Ü., Sabate, S., Gracia, C., and Peñuelas, J.: Process based inventory of isoprenoid emissions from European forests: model comparisons, current knowledge and uncertainties, Atmos. Chem. Phys., 9, 4053-4076, doi:10.5194/acp-9-4053-2009, 2009.

Kesselmeier, J., Bode, K., Hofmann, U., Müller, H., Schäfer, L., Wolf, A., Ciccioli, P., Brancaleoni, E., Cecinato, A., Frattoni, M., Foster, P., Ferrari, C., Jacob, V., Fugit, J. L., Dutaur, L., Simon, V. and Torres, L.: Emission of short chained organic acids, aldehydes and monoterpenes from Quercus Ilex L. and Pinus Pinea L. in relation to physiological activities, carbon budget and emission algorithms, Atmos. Environ., 31(97), 119–133, doi:10.1016/S1352-2310(97)00079-4, 1997.

Kesselmeier, J., Bode, K., Gerlach, C. and Jork, E. M.: Exchange of atmospheric formic and acetic acids with trees and crop plants under controlled chamber and purified air conditions, Atmos. Environ., 32(10), 1765–1775, doi:10.1016/S1352-2310(97)00465-2, 1998.

Kesselmeier, J. and Staudt, M.: Biogenic volatile organic compounds (VOC): An overview on emission, physiology and ecology, J. Atmos. Chem., 33, 23–88, doi:10.1023/A:1006127516791, 1999.

Kim, S., Karl, T., Guenther, A., Tyndall, G., Orlando, J., Harley, P., Rasmussen, R., and Apel, E.: Emissions and ambient distributions of Biogenic Volatile Organic Compounds (BVOC) in a ponderosa pine ecosystem: interpretation of PTR-MS mass spectra, Atmos. Chem. Phys., 10, 1759-1771, doi:10.5194/acp-10-1759-2010, 2010.

Klinger, L. F., Li, Q. J., Guenther, a. B., Greenberg, J. P., Baker, B. and Bai, J. H.: Assessment of volatile organic compound emissions from ecosystems of China, J. Geophys. Res. Atmos., 107, 4603, doi:10.1029/2001JD001076, 2002.

Krinner, G., Viovy, N., de Noblet-Ducoudré, N., Ogée, J., Polcher, J., Friedlingstein, P., Ciais, P., Sitch, S. and Prentice, I. C.: A dynamic global vegetation model for studies of the coupled atmosphere-biosphere system, Global Biogeochem. Cycles, 19, 1–33, doi:10.1029/2003GB002199, 2005.

Kuhn, U., Rottenberger, S., Biesenthal, T., Wolf, A., Schebeske, G., Ciccioli, P., Brancaleoni, E., Frattoni, M., Tavares, T. M., and Kesselmeier, J.: Isoprene and monoterpene emissions of Amazonian tree species during the wet season: direct and indirect investigations on

controlling environmental functions, J. Geophys. Res., D107, 8071, doi:10.1029/2001JD000978, 2002.

Kuhn, U., Andreae, M. O., Ammann, C., et al.: Isoprene and monoterpene fluxes from Central Amazonian rainforest inferred from tower-based and airborne measurements, and implications on the atmospheric chemistry and the local carbon budget, Atmos. Chem. Phys., 7, 2855–2879, doi:10.5194/acp-7-2855-2007, 2007.

Lamarque, J. F., Bond, T. C., Eyring, V., Granier, C., Heil, a., Klimont, Z., Lee, D., Liousse, C., Mieville, a., Owen, B., Schultz, M. G., Shindell, D., Smith, S. J., Stehfest, E., Van Aardenne, J., Cooper, O. R., Kainuma, M., Mahowald, N., McConnell, J. R., Naik, V., Riahi, K. and Van Vuuren, D. P.: Historical (1850-2000) gridded anthropogenic and biomass burning emissions of reactive gases and aerosols: Methodology and application, Atmos. Chem. Phys., 10, 7017–7039, doi:10.5194/acp-10-7017-2010, 2010.

Laothawornkitkul, J., Taylor, J. E., Paul, N. D. and Hewitt, C. N.: Biogenic volatile organic compounds in the Earth system: Tansley review, New Phytol., 183, 27–51, doi:10.1111/j.1469-8137.2009.02859.x, 2009.

Lathière, J., Hauglustaine, D. A., Friend, A. D., De Noblet-Ducoudré, N., Viovy, N., and Folberth, G. A.: Impact of climate variability and land use changes on global biogenic volatile organic compound emissions, Atmos. Chem. Phys., 6, 2129-2146, doi:10.5194/acp-6-2129-2006, 2006.

Lathière, J., Hewitt, C. N., and Beerling, D. J.: Sensitivity of isoprene emissions from the terrestrial biosphere to 20th century changes in atmospheric CO_2 concentration, climate, and land use, Global Biochem. Cy., 24, GB1004, doi:10.1029/2009GB003548, 2010.

Lawrence, P. J. and Chase, T. N.: Representing a new MODIS consistent land surface in the Community Land Model (CLM 3.0), J. Geophys. Res.-Biogeo., 112, G01023, doi:10.1029/2006JG000168, 2007.

Lawrence, D. M., Oleson, K. W., Flanner, M. G., Thornton, P. E., Swenson, S. C., Lawrence, P. J., Zeng, X., Yang, Z.-L., Levis, S., Sakaguchi, K., Bonan, G. B. and Slater, A. G.: Parameterization improvements and functional and structural advances in version 4 of the Community Land Model, J. Adv. Model. Earth Syst., 3, M03001, doi:10.1029/2011MS000045, 2011.

Leung, D. Y. C., Wong, P., Cheung, B. K. H. and Guenther, A.: Improved land cover and emission factors for modeling biogenic volatile organic compounds emissions from Hong Kong, Atmos. Environ., 44, 1456–1468, doi:10.1016/j.atmosenv.2010.01.012, 2010.

Levis S., Wiedinmyer, C., Bonan, G. B., and Guenther, A.: Simulating biogenic volatile organic compound emissions in the Community Climate System Model, J. Geophys. Res., 108, 4659, doi:10.1029/2002JD003203, 2003.

Loreto, F., Ciccioli, P., Cecinato, A., Brancaleoni, E., Frattoni, M., Fabozzi, C., and Tricoli, D.: Evidence of the photosynthetic origin of monoterpenes emitted by Quercus ilex L. leaves by ^{13}C labeling, Plant Physiol., 110, 1317–1322, 1996.

Maignan, F., Bréon, F.-M., Chevallier, F., Viovy, N., Ciais, P., Garrec, C., Trules, J., and Mancip, M.: Evaluation of a Global Vegetation Model using time series of satellite vegetation indices, Geosci. Model Dev., 4, 1103-1114, doi:10.5194/gmd-4-1103-2011, 2011.

Mann, G. W., Carslaw, K. S., Reddington, C. L., Pringle, K. J., Schulz, M., Asmi, A., Spracklen, D. V., Ridley, D. A., Woodhouse, M. T., Lee, L. A., Zhang, K., Ghan, S. J., Easter, R. C., Liu, X., Stier, P., Lee, Y. H., Adams, P. J., Tost, H., Lelieveld, J., Bauer, S. E., Tsigaridis, K., van Noije, T. P. C., Strunk, A., Vignati, E., Bellouin, N., Dalvi, M., Johnson, C. E., Bergman, T., Kokkola, H., von Salzen, K., Yu, F., Luo, G., Petzold, A., Heintzenberg, J., Clarke, A., Ogren, J. A., Gras, J., Baltensperger, U., Kaminski, U., Jennings, S. G., O'Dowd, C. D., Harrison, R. M., Beddows, D. C. S., Kulmala, M., Viisanen, Y., Ulevicius, V., Mihalopoulos, N., Zdimal, V., Fiebig, M., Hansson, H.-C., Swietlicki, E., and Henzing, J. S.: Intercomparison and evaluation of global aerosol microphysical properties among AeroCom models of a range of complexity, Atmos. Chem. Phys., 14, 4679-4713, doi:10.5194/acp-14-4679-2014, 2014.

Mao, J., Paulot, F., Jacob, D. J., Cohen, R. C., Crounse, J. D., Wennberg, P. O., Keller, C. a., Hudman, R. C., Barkley, M. P. and Horowitz, L. W.: Ozone and organic nitrates over the eastern United States: Sensitivity to isoprene chemistry, J. Geophys. Res. Atmos., 118(11), 11256–11268, doi:10.1002/jgrd.50817, 2013.

Martin, R. S., Villanueva, I., Zhang, J. and Popp, C. J.: Nonmethane hydrocarbon, monocarboxylic acid, and low molecular weight aldehyde and ketone emissions from vegetation in central New Mexico, Environ. Sci. Technol., 33, 2186–2192, 1999.

Matsunaga, S. N., Guenther, A. B., Greenberg, J. P., Potosnak, M., Papiez, M., Hiura, T., Kato, S., Nishida, S., Harley, P. and Kajii, Y.: Leaf level emission measurement of sesquiterpenes and oxygenated sesquiterpenes from desert shrubs and temperate forest trees using a liquid extraction technique, Geochem. J., 43, 179–189, doi:10.2343/geochemj.1.0017, 2009.

Millet, D. B., Jacob, D. J., Custer, T. G., de Gouw, J. A., Goldstein, A. H., Karl, T., Singh, H. B., Sive, B. C., Talbot, R. W., Warneke, C., and Williams, J.: New constraints on terrestrial and oceanic sources of atmospheric methanol, Atmos. Chem. Phys., 8, 6887-6905, doi:10.5194/acp-8-6887-2008, 2008.

Millet, D. B., Guenther, A., Siegel, D. A., Nelson, N. B., Singh, H. B., de Gouw, J. A., Warneke, C., Williams, J., Eerdekens, G., Sinha, V., Karl, T., Flocke, F., Apel, E., Riemer, D. D., Palmer, P. I., and Barkley, M.: Global atmospheric budget of acetaldehyde: 3-D model analysis and constraints from in-situ and satellite observations, Atmos. Chem. Phys., 10, 3405-3425, doi:10.5194/acp-10-3405-2010, 2010.

Mitchell, T. D. and Jones, P. D.: An improved method of con- structing a database of monthly climate observations and as- sociated high-resolution grids, Int. J. Climatol., 25, 693–712, doi:10.1002/Joc.1181, 2005.

Müller, J.-F., Stavrakou, T., Wallens, S., De Smedt, I., Van Roozendael, M., Potosnak, M. J., Rinne, J., Munger, B., Goldstein, A., and Guenther, A. B.: Global isoprene emissions estimated using MEGAN, ECMWF analyses and a detailed canopy environment model, Atmos. Chem. Phys., 8, 1329–1341, doi:10.5194/acp-8- 1329-2008, 2008.

Myneni, R. B., Hoffman, S., Knyazikhin, Y., Privette, J. L., Glassy, J., Tian, Y., Wang, Y., Song, X., Zhang, Y., Smith, G., Lotsch, A., Friedl, M., Morisette, J. T., Votava, P., Nemani, R. R., and Running, S. W.: Global products of vegetation leaf area and fraction absorbed PAR from year one of MODIS data, Remote Sens. Environ., 83, 214–231, 2002.

Naik, V., Delire, C., and Wuebbles, D. J.: Sensitivity of global biogenic isoprenoid emissions to climate variability and atmospheric CO 2 , J. Geophys. Res.-Atmos., 109, D06301, doi:10.1029/2003JD004236, 2004.

Niinemets, Ü.: Controls on the emission of plant volatiles through stomata: Differential sensitivity of emission rates to stomatal closure explained, J. Geophys. Res., 108, 1–17, doi:10.1029/2002JD002620, 2003a.

Niinemets, Ü.: Controls on the emission of plant volatiles through stomata: A sensitivity analysis, J. Geophys. Res., 108, 1–10, doi:10.1029/2002JD002626, 2003b.

Niinemets, Ü., Arneth, A., Kuhn, U., Monson, R. K., Peñuelas, J. and Staudt, M.: The emission factor of volatile isoprenoids: Stress, acclimation, and developmental responses, Biogeosciences, 7, 2203–2223, doi:10.5194/bg-7-2203-2010, 2010.

Niinemets, Ü., Kuhn, U., Harley, P. C., Staudt, M., Arneth, A., Cescatti, A., Ciccioli, P., Copolovici, L., Geron, C., Guenther, a., Kesselmeier, J., Lerdau, M. T., Monson, R. K. and Peñuelas, J.: Estimations of isoprenoid emission capacity from enclosure studies: Measurements, data processing, quality and standardized measurement protocols, Biogeosciences, 8, 2209–2246, doi:10.5194/bg-8-2209-2011, 2011.

Oderbolz, D. C., Aksoyoglu, S., Keller, J., Barmpadimos, I., Steinbrecher, R., Skjøth, C. A., Plaß-Dülmer, C. and Prévôt, A. S. H.: A comprehensive emission inventory of biogenic volatile organic compounds in Europe: improved seasonality and land-cover, Atmos. Chem. Phys., 13(4), 1689–1712, doi:10.5194/acp-13-1689-2013, 2013.

Ortega, J., Helmig, D., Daly, R. W., Tanner, D. M., Guenther, A. B. and Herrick, J. D.: Approaches for quantifying reactive and low-volatility biogenic organic compound emissions by vegetation enclosure techniques - Part B: Applications, Chemosphere, 72, 365–380, doi:10.1016/j.chemosphere.2008.02.054, 2008.

Owen, S. M., Harley, P., Guenther, A., and Hewitt, C. N.: Light dependency of VOC emissions from selected Mediterranean plant species, Atmos. Environ., 36, 3147–3159, 2002.

Pacifico, F., Harrison, S. P., Jones, C. D., Arneth, A., Sitch, S., Weedon, G. P., Barkley, M. P., Palmer, P. I., Serça, D., Potosnak, M., Fu, T-M., Goldstein, A., Bai, J., and Schurgers, G.: Evaluation of a photosynthesis-based biogenic isoprene emission scheme in JULES and simulation of isoprene emissions under present-day climate conditions, Atmos. Chem. Phys., 11, 4371–4389, doi:10.5194/acp-11-4371-2011, 2011.

Padhy, P. K. and Varshney, C. K.: Isoprene emission from tropical tree species, Environ. Pollut., 135, 101–109, doi:10.1016/j.envpol.2004.10.003, 2005.

Palmer, P. I., Abbot, D. S., Fu, T.-M., Jacob, D. J., Chance, K., Kurosu, T. P., Guenther, A., Wiedinmyer, C., Stanton, J. C., Pilling, M. J., Pressley, S. N., Lamb, B. and Sumner, A. L.: Quantifying the seasonal and interannual variability of North American isoprene emissions using satellite observations of the formaldehyde column, J. Geophys. Res., 111, 1–14, doi:10.1029/2005JD006689, 2006.

Peñuelas, J. and Staudt, M.: BVOCs and global change, Trends Plant Sci., 15(January), 133–144, doi:10.1016/j.tplants.2009.12.005, 2010.

Pfister, G. G., Emmons, L. K., Hess, P. G., Lamarque, J. F., Orlando, J. J., Walters, S., Guenther, A., Palmer, P. I. and Lawrence, P. J.: Contribution of isoprene to chemical budgets: A model tracer study with the NCAR CTM MOZART-4, J. Geophys. Res. Atmos., 113, 1–21, doi:10.1029/2007JD008948, 2008.

Pinty, B., Andredakis, I., Clerici, M., Kaminski, T., Taberner, M., Verstraete, M. M., Gobron, N., Plummer, S. and Widlowski, J. L.: Exploiting the MODIS albedos with the Two-Stream Inversion Package (JRC-TIP): 1. Effective leaf area index, vegetation, and soil properties, J. Geophys. Res. Atmos., 116(9), 1–20, doi:10.1029/2010JD015372, 2011.

Prentice, I. C., Cramer, W., Harrison, S. P., Leemans, R., Monserud, R. A., and Solomon, A. M.: A global biome model based on plant physiology and dominance, soil properties and climate, J. Biogeogr., 19, 117–134, 1992.

Qian, T., Dai, A., Trenberth, K. E., and Oleson, K.W.: Simulation of global land surface conditions from 1948 to 2004, Part I: Forcing data and evaluations, J. Hydrometeorol., 7, 953–975, 2006.

Riipinen, I., Yli-Juuti, T., Pierce, J. R., Petäjä, T., Worsnop, D. R., Kulmala, M., and Donahue, N. M.: The contribution of organics to atmospheric nanoparticle growth, Nat. Geosci., 5, 453–458, doi: 10.1038/ngeo1499, 2012.

Rinne, H. J. I., Guenther, A. B., Greenberg, J. P., and Harley, P. C.: Isoprene and monoterpene fluxes measured above Amazonian rainforest and their dependence on light and temperature, Atmos. Environ., 36(14), 2421–2426, 2002.

Ruuskanen, T. M., Hakola, H., Kajos, M. K., Hellén, H., Tarvainen, V., and Rinne, J.: Volatile organic compound emissions from Siberian larch, Atmos. Environ., 41, 5807–5812, 2007.

Schade, G. W. and Goldstein, A. H.: Fluxes of oxygenated volatile organic compounds from a ponderosa pine plantation, J. Geophys. Res., 106(D3), 3111–3123, 2001.

Schurgers, G., Arneth, A., Holzinger, R. and Goldstein, A.: Process-based modelling of biogenic monoterpene emissions: sensitivity to temperature and light, Atmos. Chem. Phys. Discuss., 9, 271–307, doi:10.5194/acpd-9-271-2009, 2009.

Shao, M., Czapiewski, K. V., Heiden, A. C., Kobel, K., Komenda, M., Koppmann, R., and Wildt, J.: Volatile organic compound emissions from Scots pine: Mechanisms and description by algorithms, J. Geophys. Res., 106(D17), 20 483–20 492, 2001.

Shim, C., Wang, Y., Choi, Y., Palmer, P. I., Abbot, D. S., and Chance, K.: Constraining global isoprene emissions with Global Ozone Monitoring Experiment (GOME) formaldehyde column measurements, J. Geophys. Res.-Atmos., 110, D24301, doi:10.1029/2004JD005629, 2005.

Sitch, S., Smith, B., Prentice, I. C., Arneth, A., Bondeau, A., Cramer, W., Kaplan, J. O., Levis, S., Lucht, W., Sykes, M. T., Thonicke, K., and Venevsky, S.: Evaluation of ecosystem dynamics, plant geography and terrestrial carbon cycling in the LPJ dynamic global vegetation model, Glob. Change Biol., 9, 161–185, 2003.

Šimpraga, M., Verbeeck, H., Bloemen, J., Vanhaecke, L., Demarcke, M., Joó, E., Pokorska, O., Amelynck, C., Schoon, N., Dewulf, J., Van Langenhove, H., Heinesch, B., Aubinet, M. and Steppe, K.: Vertical canopy gradient in photosynthesis and monoterpenoid emissions: An insight into the chemistry and physiology behind, Atmos. Environ., 80, 85–95, doi:10.1016/j.atmosenv.2013.07.047, 2013.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., and Wind, P.: The EMEP MSC-W chemical transport model – technical description, Atmos. Chem. Phys., 12, 7825-7865, doi:10.5194/acp-12-7825-2012, 2012.

Sindelarova, K., Granier, C., Bouarar, I., Guenther, A., Tilmes, S., Stavrakou, T., Müller, J.-F., Kuhn, U., Stefani, P., and Knorr, W.: Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years, Atmos. Chem. Phys., 14, 9317-9341, doi:10.5194/acp-14-9317-2014, 2014.

Situ, S., Wang, X., Guenther, A., Zhang, Y., Wang, X., Huang, M., Fan, Q. and Xiong, Z.: Uncertainties of isoprene emissions in the MEGAN model estimated for a coniferous and broad-leaved mixed forest in Southern China, Atmos. Environ., 98, 105–110, doi:10.1016/j.atmosenv.2014.08.023, 2014.

Smiatek, G. and Steinbrecher, R.: Temporal and spatial variation of forest VOC emissions in Germany in the decade 1994-2003, Atmos. Environ., 40, 166–177, doi:10.1016/j.atmosenv.2005.11.071, 2006.

Smolander, S., He, Q., Mogensen, D., Zhou, L., Bäck, J., Ruuskanen, T., Noe, S., Guenther, A., Aaltonen, H., Kulmala, M., and Boy, M.: Comparing three vegetation monoterpene emission models to measured gas concentrations with a model of meteorology, air chemistry and chemical transport, Biogeosciences, 11, 5425-5443, doi:10.5194/bg-11-5425-2014, 2014.

Spirig, C., Neftel, A., Ammann, C., Dommen, J., Grabmer, W., Thielmann, A., Schaub, A., Beauchamp, J., Wisthaler, A., and Hansel, A.: Eddy covariance flux measurements of biogenic VOCs during ECHO 2003 using proton transfer reaction mass spectrometry, Atmos. Chem. Phys., 5, 465-481, doi:10.5194/acp-5-465-2005, 2005.

Spitters, C. J. T., Toussaint, H. A. J. M. and Goudriaan, J.: Separating the Diffuse and Direct Component of Global Radiation and Its Implications for Modeling Canopy Photosynthesis, Part I. Components of Incoming Radiation, Agric. For. Meteorol., 38, 217–229, 10.1016/0168-1923(86)90060-2, 1986a.

Spitters, C. J. T.: Separating the diffuse and direct component of global radiation and its implications for modeling canopy photosynthesis Part II. Calculation of canopy photosynthesis, Agric. For. Meteorol., 38, 231–242, doi:10.1016/0168-1923(86)90061-4, 1986b.

Spracklen, D. V. and Righelato, R.: Tropical montane forests are a larger than expected global carbon store, Biogeosciences, 11, 2741-2754, doi:10.5194/bg-11-2741-2014, 2014.

Staudt, M. and Seufert, G.: Light-dependent emission of monoterpenes by holm oak (Quercus ilex 1.), Naturwissenschaften, 82, 89–92, 1995.

Staudt, M., Wolf, A. and Kesselmeier, J.: Influence of environmental factors on the emissions of gaseous formic and acetic acids from orange (Citrus sinensis L.) foliage, Biogeochemistry, 48, 199–216, doi:10.1023/A:1006289120280, 2000.

Stavrakou, T., Müller, J.-F., De Smedt, I., Van Roozendael, M., van der Werf, G. R., Giglio, L., and Guenther, A.: Global emissions of non-methane hydrocarbons deduced from SCIAMACHY formaldehyde columns through 2003–2006, Atmos. Chem. Phys., 9, 3663–3679, doi:10.5194/acp-9-3663-2009, 2009.

Stavrakou, T., Guenther, A., Razavi, A., Clarisse, L., Clerbaux, C., Coheur, P.-F., Hurtmans, D., Karagulian, F., De Mazière, M., Vigouroux, C., Amelynck, C., Schoon, N., Laffineur, Q., Heinesch, B., Aubinet, M., Rinsland, C., and Müller, J.-F.: First space-based derivation of the global atmospheric methanol emission fluxes, Atmos. Chem. Phys., 11, 4873-4898, doi:10.5194/acp-11-4873-2011, 2011.

Stavrakou, T., Müller, J.-F., Bauwens, M., De Smedt, I., Van Roozendael, M., Guenther, A., Wild, M., and Xia, X.: Isoprene emissions over Asia 1979–2012: impact of climate and land use changes, Atmos. Chem. Phys., 14, 4587–4605, doi:10.5194/acp-14-4587-2014, 2014.

Steinbrecher, R., Hauff, K., Hakola, H., and Rössler, J.: A recise parameterization for emission modeling of isoprenoids for Boreal plants, in: Biogenic VOC Emissions and Photochemistry in the Boreal Regions of Europe – Biphorep, edited by: Laurila, T. and Lindfors, V., Air Pollution Research Report No. 70, Commission of the European Communities, Luxembourg, 29–43, 1999.

Steinbrecher, R., Smiatek, G., Köble, R., Seufert, G., Theloke, J., Hauff, K., Ciccioli, P., Vautard, R. and Curci, G.: Intra- and inter-annual variability of VOC emissions from natural and semi-natural vegetation in Europe and neighbouring countries, Atmos. Environ., 43(7), 1380–1391, doi:10.1016/j.atmosenv.2008.09.072, 2009.

Steinbrecher, R., Contran, N., Gugerli, F., Schnitzler, J. P., Zimmer, I., Menard, T. and Günthardt-Goerg, M. S.: Inter- and intra-specific variability in isoprene production and photosynthesis of Central European oak species, Plant Biol., 15, 148–156, doi:10.1111/j.1438-8677.2012.00688.x, 2013.

Steiner, A. H. and Goldstein, A. L.: Biogenic VOCs, in: Volatile Organic Compounds in the Atmosphere, edited by R. Koppmann, Blackwell Publishing Ltd., 82-128, 2007.

Stewart, H. E., Hewitt, C. N., Bunce, R. G. H., Steinbrecher, R., Smiatek, G., and Schoenemeyer, T.: A highly spatially and temporally resolved inventory for biogenic isoprene and monoterpene emissions: Model description and application to Great Britain, J. Geophys. Res., 108, 4644, doi:10.1029/2002JD002694 , 2003.

Taipale, R., Kajos, M. K., Patokoski, J., Rantala, P., Ruuskanen, T. M., and Rinne, J.: Role of de novo biosynthesis in ecosystem scale monoterpene emissions from a boreal Scots pine forest, Biogeosciences, 8, 2247-2255, doi:10.5194/bg-8-2247-2011, 2011.

Taraborrelli, D., Lawrence, M. G., Crowley, J. N., Dillon, T. J., Gromov, S., Groß, C. B. M., Vereecken, L. and Lelieveld, J.: Hydroxyl radical buffered by isoprene oxidation over tropical forests, Nat. Geosci., 5(3), 300–300, doi:10.1038/ngeo1433, 2012.

Tarvainen, V., Hakola, H., Hellén, H., Bäck, J., Hari, P., and Kulmala, M.: Temperature and light dependence of the VOC emissions of Scots pine, Atmos. Chem. Phys., 5, 989-998, doi:10.5194/acp-5-989-2005, 2005.

Tilmes, S., Lamarque, J.-F., Emmons, L.K., Kinnison, D.E., Ma, P.-L., Liu, X., Ghan, S., Bardeen, C., Arnold, S., Deeter, M., Vitt, F., Ryerson, T., Elkins, J.W., Moore, F., Spackman, J.R. and Val Martin, M.: Description and evaluation of tropospheric chemistry and aerosols in the Community Earth System Model (CESM1.2), Geoscientific Model Development, 8, 1395–1426, 2015.

Topping, D., Connolly, P., and McFiggans, G.: Cloud droplet number enhanced by co-condensation of organic vapours, Nature Geosci., 6, 443–446, 2013.

Tsigaridis, K., Daskalakis, N., Kanakidou, M., Adams, P. J., Artaxo, P., Bahadur, R., Balkanski, Y., Bauer, S. E., Bellouin, N., Benedetti, A., Bergman, T., Berntsen, T. K., Beukes, J. P., Bian, H., Carslaw, K. S., Chin, M., Curci, G., Diehl, T., Easter, R. C., Ghan, S. J., Gong, S. L., Hodzic, A., Hoyle, C. R., Iversen, T., Jathar, S., Jimenez, J. L., Kaiser, J. W., Kirkevåg, A., Koch, D., Kokkola, H., Lee, Y. H, Lin, G., Liu, X., Luo, G., Ma, X., Mann, G. W., Mihalopoulos, N., Morcrette, J.-J., Müller, J.-F., Myhre, G., Myriokefalitakis, S., Ng, N. L., O'Donnell, D., Penner, J. E., Pozzoli, L., Pringle, K. J., Russell, L. M., Schulz, M., Sciare, J., Seland, Ø., Shindell, D. T., Sillman, S., Skeie, R. B., Spracklen, D., Stavrakou, T.,

Steenrod, S. D., Takemura, T., Tiitta, P., Tilmes, S., Tost, H., van Noije, T., van Zyl, P. G., von Salzen, K., Yu, F., Wang, Z., Wang, Z., Zaveri, R. A., Zhang, H., Zhang, K., Zhang, Q., and Zhang, X.: The AeroCom evaluation and intercomparison of organic aerosol in global models, Atmos. Chem. Phys., 14, 10845-10895, doi:10.5194/acp-14-10845-2014, 2014.

Tsui, J. K.-Y., Guenther, A., Yip, W.-K. and Chen, F.: A biogenic volatile organic compound emission inventory for Hong Kong, Atmos. Environ., 43, 6442–6448, doi:10.1016/j.atmosenv.2008.01.027, 2009.

Tucker, J. C., Slayback, A. D., Pinzon, E. J., Los, O. S., Myneni, B. R. and Taylor, G. M.: Higher northern latitude normalized difference vegetation index and growing season trends from 1982 to 1999, Int. J. Biometeorol., 45(4), 184–190, doi:10.1007/s00484-001-0109-8.

Unger, N., Harper, K., Zheng, Y., Kiang, N. Y., Aleinov, I., Arneth, a., Schurgers, G., Amelynck, C., Goldstein, a., Guenther, a., Heinesch, B., Hewitt, C. N., Karl, T., Laffineur, Q., Langford, B., McKinney, K. a., Misztal, P., Potosnak, M., Rinne, J., Pressley, S., Schoon, N. and Serça, D.: Photosynthesis-dependent isoprene emission from leaf to planet in a global carbon-chemistry-climate model, Atmos. Chem. Phys., 13, 10243–10269, doi:10.5194/acp-13-10243-2013, 2013.

Van Donkelaar, A., Martin, R. V., Park, R. J., Heald, C. L., Fu, T. M., Liao, H. and Guenther, A.: Model evidence for a significant source of secondary organic aerosol from isoprene, Atmos. Environ., 41, 1267–1274, doi:10.1016/j.atmosenv.2006.09.051, 2007.

Villanueva-Fierro, I., Popp, C. J., and Martin, R. S.: Biogenic emissions and ambient concentrations of hydrocarbons, carbonyl compounds and organic acids from ponderosa pine and cottonwood trees at rural and forested sites in Central New Mexico, Atmos. Environ., 38(2), 249–260, 2004.

Von Kuhlmann, R., Lawrence, M. G., Crutzen, P. J. and Rasch, P. J.: A model for studies of tropospheric ozone and nonmethane hydrocarbons: Model description and ozone results, J. Geophys. Res., 108, doi:10.1029/2002JD002893, 2003.

Wells, K. C., Millet, D. B., Hu, L., Cady-Pereira, K. E., Xiao, Y., Shephard, M. W., Clerbaux, C. L.,Clarisse, L., Coheur, P.-F., Apel, E. C., de Gouw, J., Warneke, C., Singh, H. B., Goldstein, A.H., and Sive, B. C.: Tropospheric methanol observations from space: retrieval

evaluation and constraints on the seasonality of biogenic emissions, Atmos. Chem. Phys., 12, 5897–5912, doi:10.5194/acp-12-5897-2012, 2012.

Wells K. C., D. B. Millet, K. E. Cady-Pereira, M. W. Shephard, D. K. Henze, N. Bousserez, E. C. Apel, J. de Gouw, C. Warneke, and H. B. Singh. Quantifying global terrestrial methanol emissions using observations from the TES satellite sensor. Atmos. Chem. Phys., 14, 2555-2570, 2014.

Yuan, H., Dai, Y., Xiao, Z., Ji, D. and Shangguan, W.: Reprocessing the MODIS Leaf Area Index products for land surface and climate modelling, Remote Sens. Environ., 115(5), 1171–1187, doi:10.1016/j.rse.2011.01.001, 2011.

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	0.9	Light multilayer vertical profile to calculate radiation extinction inside the canopy for both sunlit and shaded leaves	Isoprene Methanol
		Acetaldehyde, Formaldehyde, Acetic acid, Formic acid	0.8	0.10		
		Acetone	0.2	0.10		
		Methanol	0.8	0.8		
		Total monoterpene, 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 ~~gdmg~~$^{-1}$ 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.

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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.

Formattato: Sinistro: 3,5 cm, Destro 2 cm, Superiore: 2,5 cm, In basso: 2,5 cm, Larghezza 29,7 cm, Altezza: 21 cm

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

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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, ~~for~~ northern and southern temperate areas and ~~for~~ 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

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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

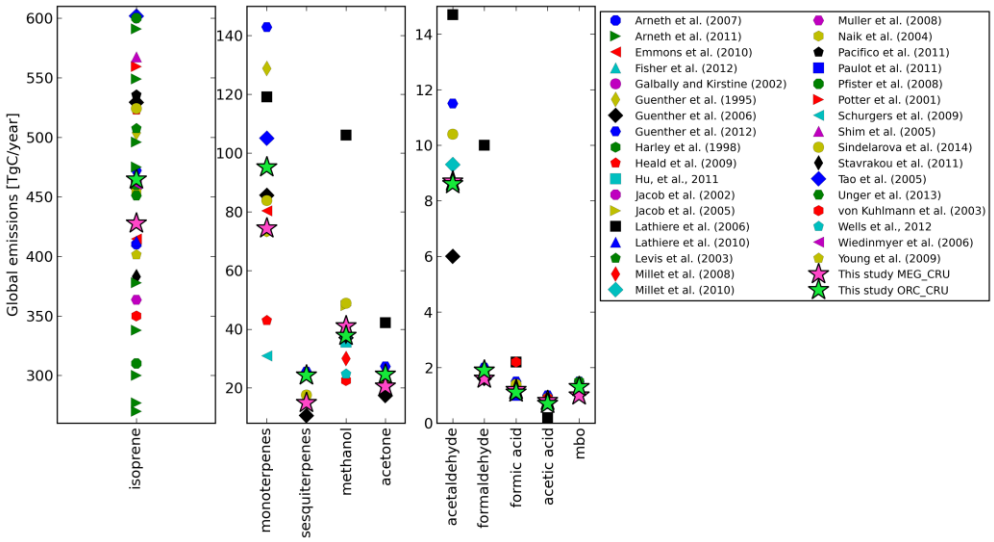


Figure 1. Global emission budgets (Tg C yr^{-1}) calculated by ORCHIDEE (ORC_CRU simulation, green stars) and MEGAN (MEG_CRU simulation, pink stars), compared with published estimates for the main BVOCs presented in this work. Note that the vertical axes have different scales in the three plots.

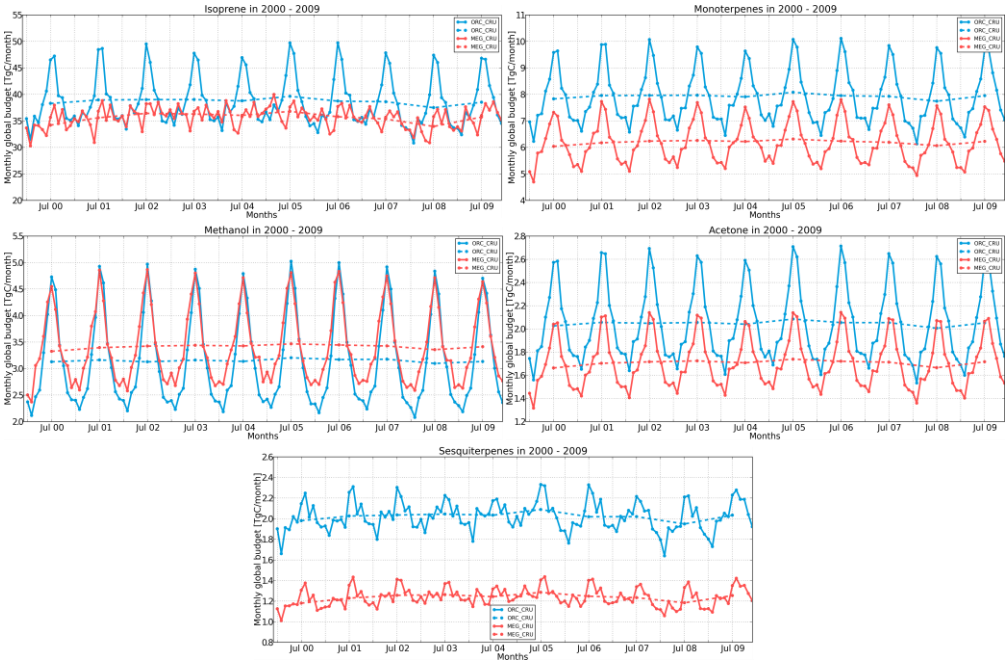


Figure 2. Monthly global (solid lines) and yearly averaged (dashed lines) emission budgets in Tg C month^{-1} for ORC_CRU and MEG_CRU simulations for isoprene, monoterpenes, methanol, acetone and sesquiterpenes.

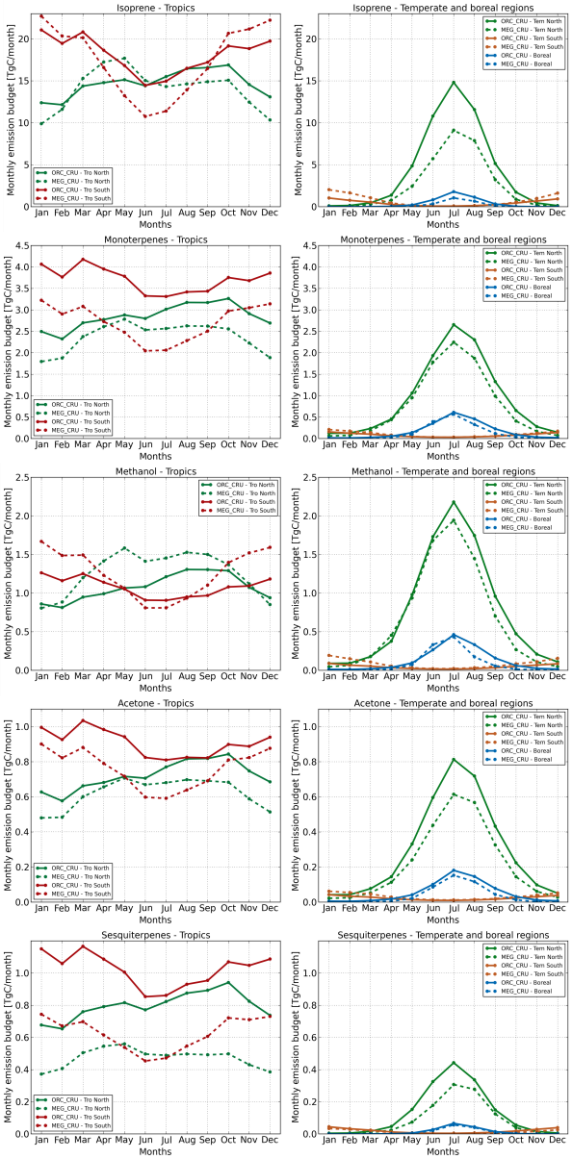


Figure 3. Zonal mean for northern and southern tropics (left column), northern and southern temperate and northern boreal latitudes (right column) of the monthly emission budget (Tg C month^{-1}) averaged over the simulation period (2000–2009)~~,~~ in ORC_CRU and MEG_CRU runs for isoprene, monoterpenes,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

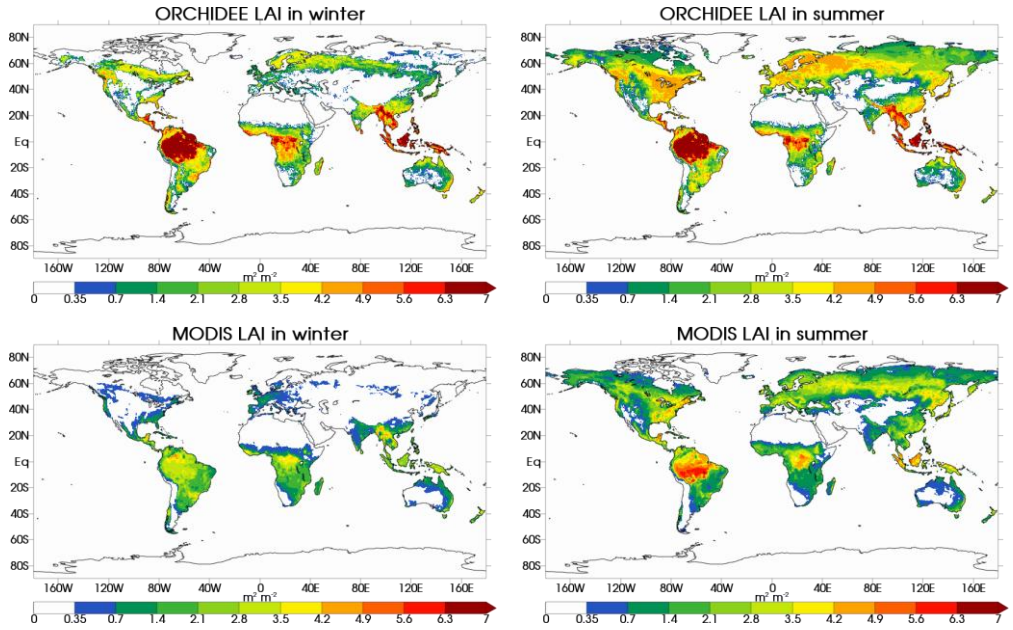


Figure 4. Leaf area index (LAI) considered for BVOC emission estimates in ORCHIDEE (LAI calculated on line) and in MEGAN (MODIS retrieval) in summer (June, July, August) and winter (December, January, February), averaged over the 2000–2009 period (m^2 m^{-2}).

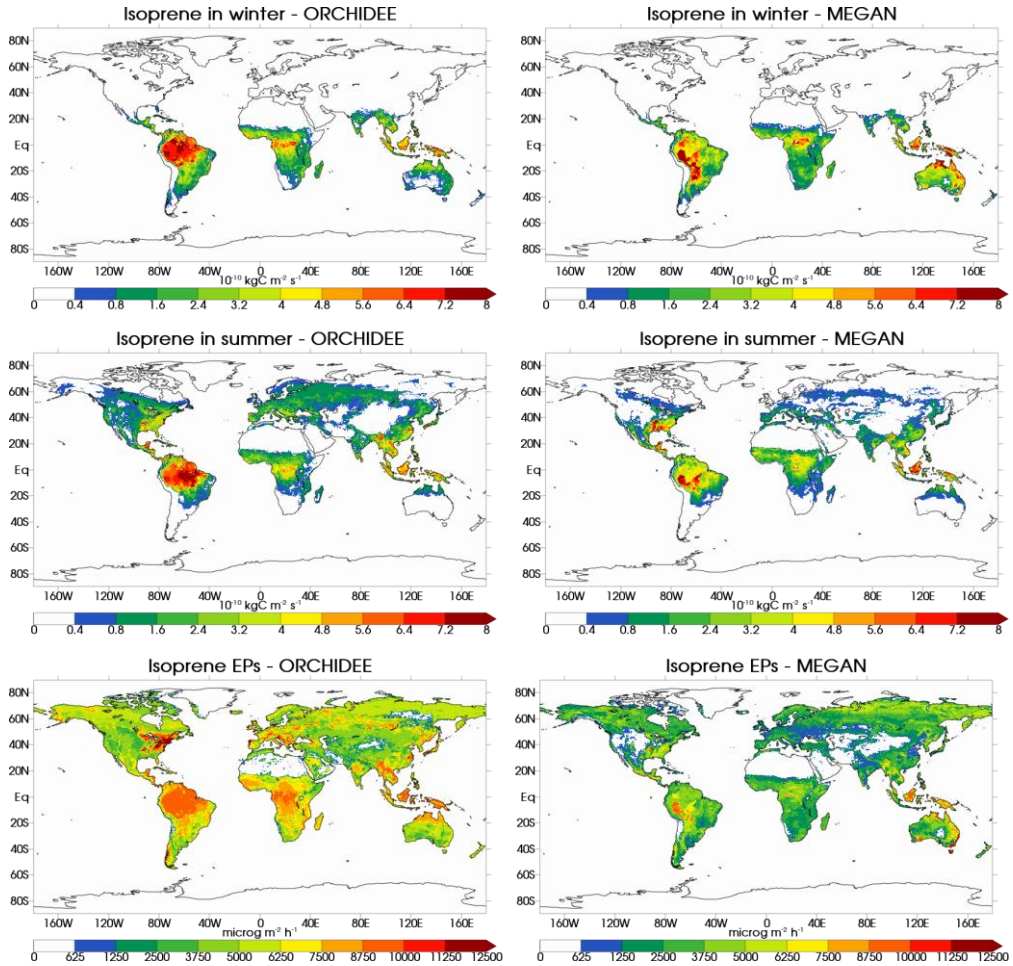


Figure 5. Emissions in winter (first ~~line~~row) and summer (second ~~line~~row) in 10^{-10} kg C m^{-2} s^{-1} and emission potentials (EPs) (third ~~line~~row) in µg m^{-2} h^{-1} for ORCHIDEE (ORC_CRU, left ~~hand~~ column) and MEGAN (MEG_CRU, right ~~hand~~ column) for isoprene.

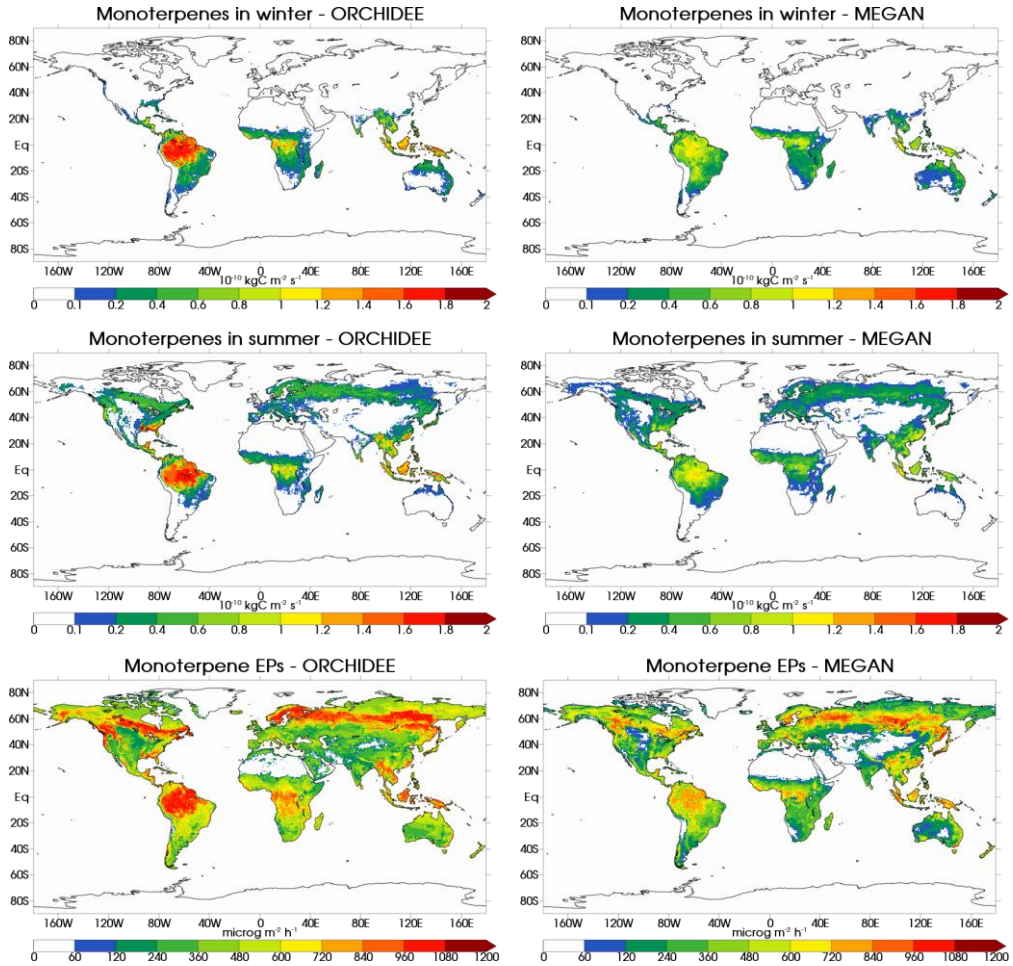


Figure 6. The same as Fig. 5, but for monoterpenes.

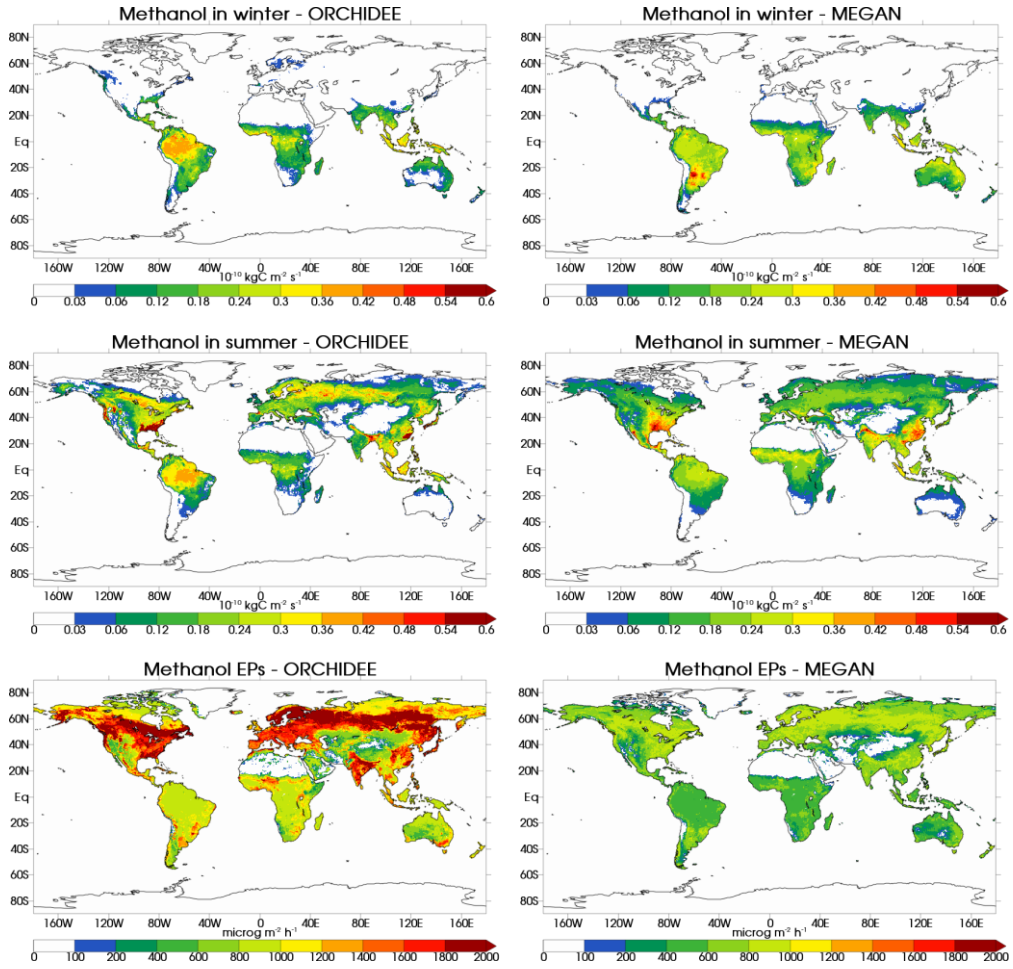


Figure 7. The same as Fig. 5, but for methanol.

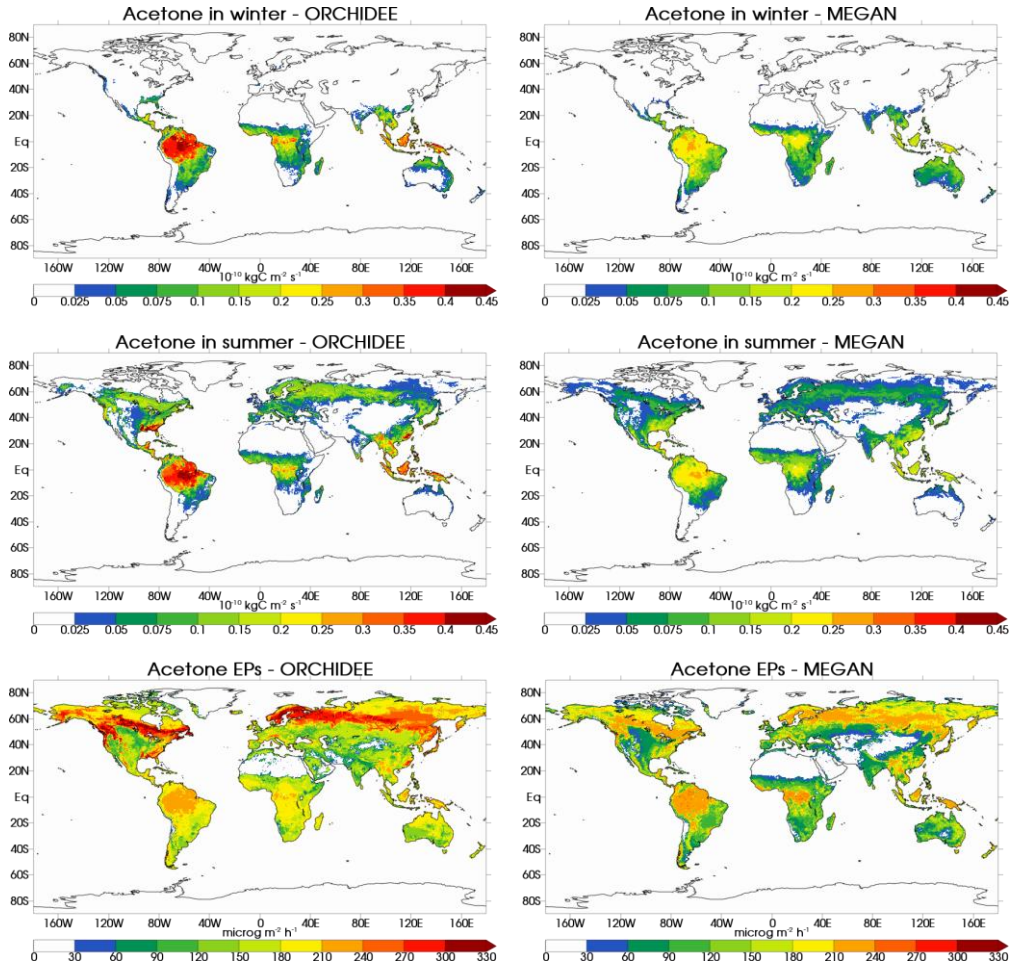


Figure 8. The same as Fig. 5, but for acetone.

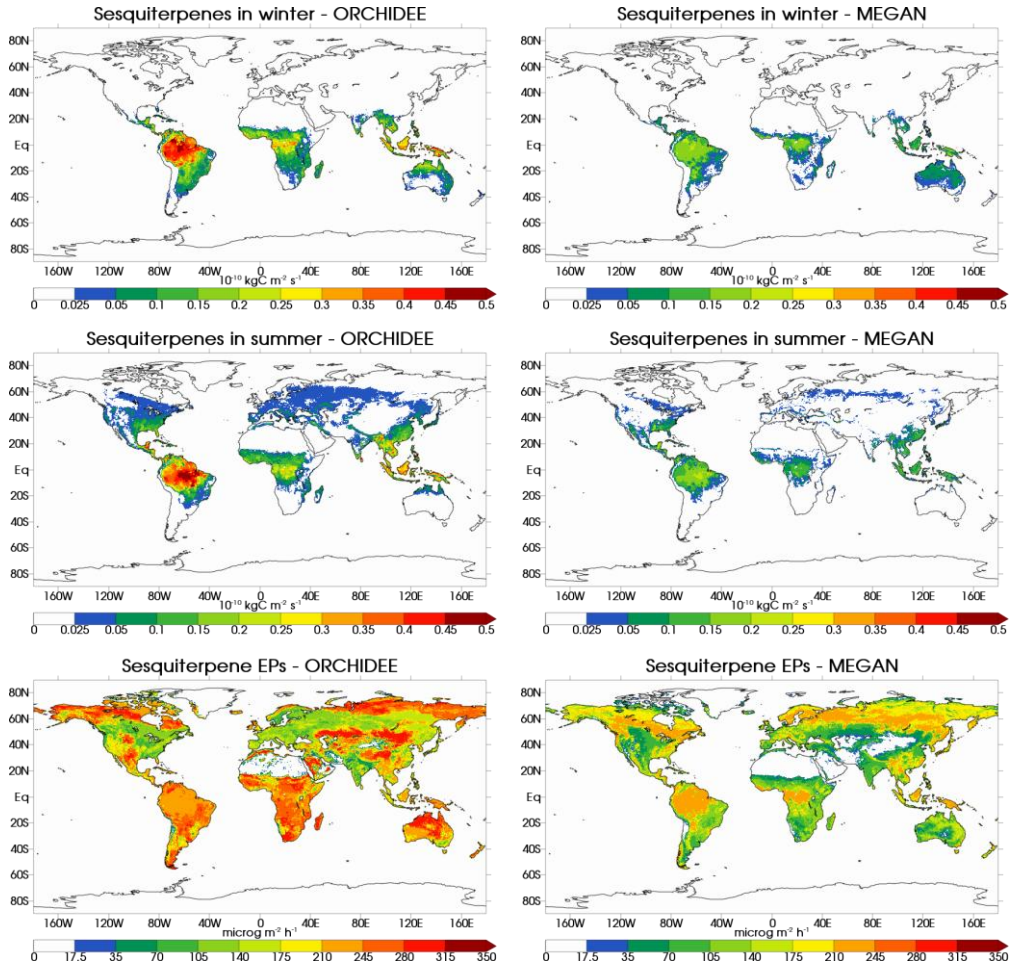


Figure 9. The same as Fig. 5, but for sesquiterpenes.

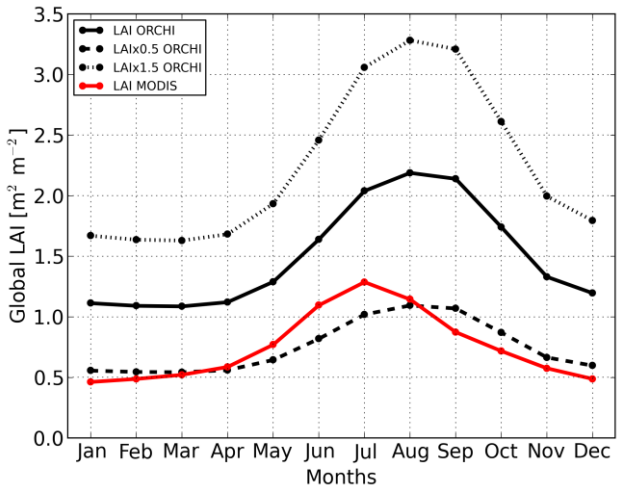
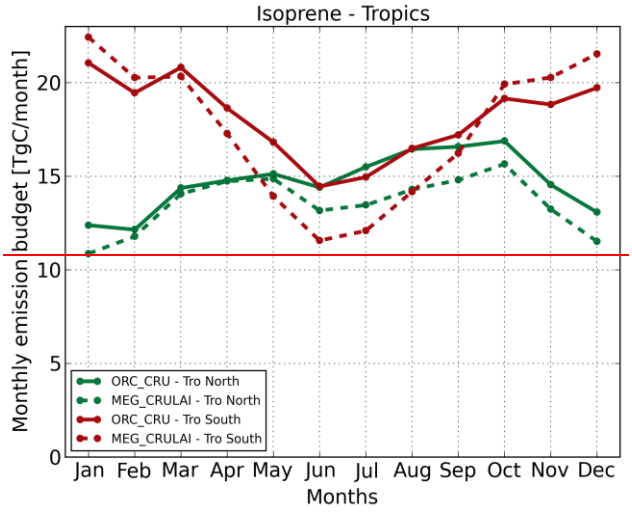
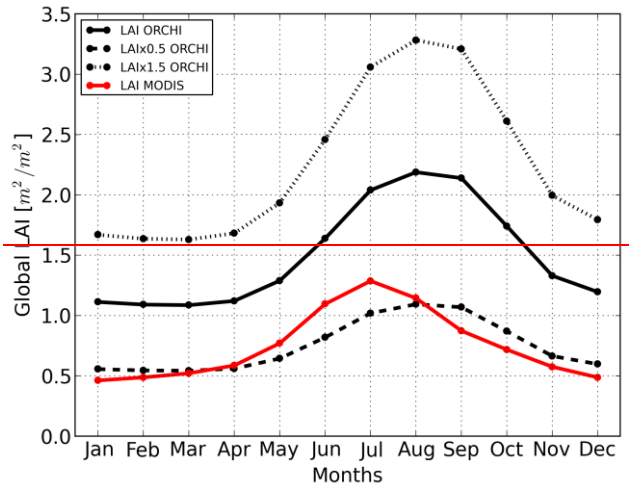


Figure 10. Global monthly mean LAI (m^2 m^{-2}) calculated by ORCHIDEE (solid black line) and retrieved from MODIS measurements (red line). The thick and thin dashed lines represent the LAI from ORCHIDEE multiplied by a factor 0.5 and 1.5, respectively.



~~Figure 10. 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 11. 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 line represent the LAI from ORCHIDEE multiplied by a factor 0.5 and 1.5 respectively.~~

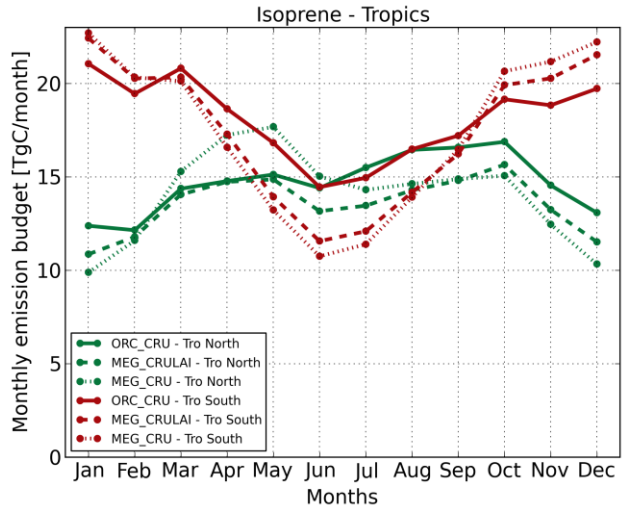


Figure 11. Zonal mean of monthly emission budgets (Tg C $month^{-1}$), averaged over the simulation period (2000–2009) for the northern and southern tropics, in ORC_CRU (solid line) and MEG_CRULAI (dashed line) simulations for isoprene.

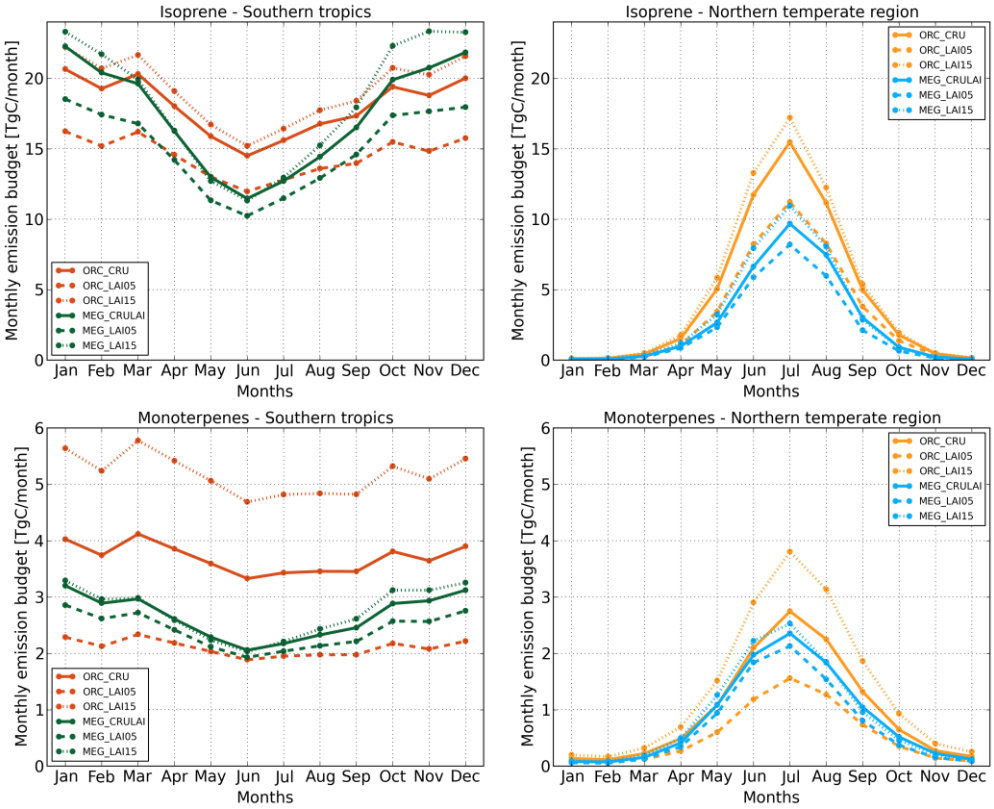


Figure 12. Zonal average of changed emissions in the different LAI sensitivity tests: ORC_CRU and MEG_CRULAI using ORCHIDEE LAI (solid line), ORC_LAI05 and MEG_LAI05 using ORCHIDEE LAI·0.5 (thin dashed line) and ORC_LAI15 and MEG_LAI15 using ORCHIDEE LAI·1.5 (thick dashed line) in the year 2006, for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

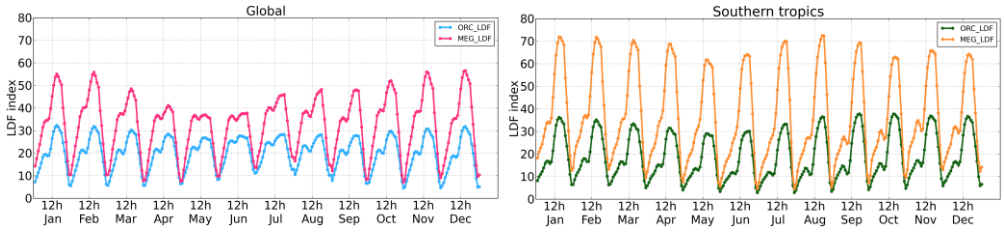


Figure 13. Global (left plot) and southern tropical (right plot) average of the LDF index for ORCHIDEE and MEGAN. The LDF index is provided as the hourly daily profile averaged over each month.