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### ► **To cite this version:**

J. Lathière, D. A. Hauglustaine, A. D. Friend, N. de Noblet-Ducoudré, N. Viovy, et al.. Impact of climate variability and land use changes on global biogenic volatile organic compound emissions. *Atmospheric Chemistry and Physics*, 2006, 6 (8), pp.2129-2146. <10.5194/acp-6-2129-2006>. <hal-00328442>

**HAL Id: hal-00328442**

**<https://hal.science/hal-00328442v1>**

Submitted on 18 Jun 2008

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# Impact of climate variability and land use changes on global biogenic volatile organic compound emissions

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Received: 19 July 2005 – Published in Atmos. Chem. Phys. Discuss.: 25 October 2005

Revised: 28 February 2006 – Accepted: 22 April 2006 – Published: 20 June 2006

**Abstract.** A biogenic emissions scheme has been incorporated in the global dynamic vegetation model ORCHIDEE (Organizing Carbon and Hydrology in Dynamic Ecosystems) in order to calculate global biogenic emissions of isoprene, monoterpenes, methanol, acetone, acetaldehyde, formaldehyde and formic and acetic acids. Important parameters such as the leaf area index are fully determined by the global vegetation model and the influences of light extinction (for isoprene emissions) and leaf age (for isoprene and methanol emissions) are also taken into account. We study the interannual variability of biogenic emissions using the satellite-based climate forcing ISLSCP-II as well as relevant CO<sub>2</sub> atmospheric levels, for the 1983–1995 period. Mean global emissions of 460 TgC/yr for isoprene, 117 TgC/yr for monoterpenes, 106 TgC/yr for methanol and 42 TgC/yr for acetone are predicted. The mean global emission of all biogenic compounds is  $752 \pm 16$  TgC/yr with extremes ranging from 717 TgC/yr in 1986 to 778 TgC/yr in 1995, that is a 8.5% increase between both. This variability differs significantly from one region to another and among the regions studied, biogenic emissions anomalies were the most variable in Europe and the least variable in Indonesia (isoprene and monoterpenes) and North America (methanol). Two scenarios of land use changes are considered using the 1983 climate and atmospheric CO<sub>2</sub> conditions, to study the sensitivity of biogenic emissions to vegetation alteration, namely tropical deforestation and European afforestation. Global biogenic emissions are highly affected by tropical deforestation, with a 29% decrease in isoprene emission and a 22% increase in methanol emission. Global emissions are not significantly affected by European afforestation, but on a European scale, total biogenic VOCs emissions increase by 54%.

## 1 Introduction

The terrestrial biosphere is a major source of natural volatile organic compounds (VOCs). Isoprene and monoterpenes dominate biogenic emissions but other highly reactive chemical species, such as methanol, acetone, aldehydes and organic acids, are now also recognized to be emitted by the terrestrial vegetation at significant levels, as shown by several measurement campaigns (Kirstine et al., 1998; Schade and Goldstein, 2001; Villanueva-Fierro et al., 2004). Global VOCs emissions by the vegetation are 1150 TgC/yr (Guenther et al., 1995), corresponding to nearly 90% of global VOC emissions at the surface (including anthropogenic emissions).

Those biogenic compounds play a crucial role in tropospheric chemistry, from local to global scales, inducing increased or decreased ozone formation, depending on NO<sub>x</sub> levels (Fehsenfeld et al., 1992; Thunis and Cuvelier, 2000; Pun et al., 2002). Recently, Barkot et al. (2004) showed that isoprene photooxidation results in maximum ozone production rates for NO<sub>x</sub> concentrations in the range of about 1–10 ppbv, a typical level of anthropogenically influenced rural environments. Wang and Shallcross (2000) used a global land-surface and chemistry-transport model to show that the inclusion of isoprene emissions, estimated at 530 TgC/yr for present day conditions, has a significant impact on ozone and oxidation products, such as peroxyacetyl nitrate (PAN), in both hemispheres. Their analysis indicated that the response of ozone to isoprene emissions was predominantly governed by the spatial and temporal variations in terrestrial vegetation, with a simulated ozone increase of about 4 ppbv over the oceans and about 8–12 ppbv over mid-latitude continental areas. Sanderson et al. (2003) illustrated the impact of climate change on both isoprene emissions and ozone levels. Based on a global isoprene emission increase from 484 TgC/yr for the 1990s to 615 TgC/yr for the 2090s, as a result of climate and vegetation distribution changes, the authors calculated an

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ozone increase by 10–20 ppbv in some locations. They also noted that the changes in ozone levels were closely linked to changes in isoprene surface fluxes in regions such as the eastern U.S. or southern China. However, this effect was much less marked over the Amazon region and Africa because of lower levels of nitrogen oxides.

In addition to their importance for gas phase chemistry, the involvement of biogenic VOCs in tropospheric aerosols formation, first suggested by Went (1960), has lately been demonstrated (Griffin et al., 1999), and enforces the importance of taking into account biogenic VOC in both tropospheric gas phase reactions and particulate formation. Assuming an aerosol yield from the oxidation of biogenic VOC other than isoprene (monoterpenes and other VOC) between 5% and 40%, Andreae and Crutzen (1997) suggested that the secondary organic aerosols (SOA) formation could range between 30 and 270 Tg of organic matter per year. More recently, Tsigaridis and Kanakidou (2003) estimated, using a global chemistry-transport model, that the production of SOA from biogenic VOC might range from 2.5 to 44.5 Tg/yr. Furthermore, Kanakidou et al. (2000) estimated that the production of SOA from biogenic VOCs increased from 17–28 Tg/yr in preindustrial times to 61–79 Tg/yr for present day conditions, which was attributed to an increase in ozone and primary organic aerosol from anthropogenic sources. In addition to the recognized role of monoterpenes SOA formation (Kavouras et al., 1999; Sotiropoulou et al., 2004), recent studies (Limbeck et al., 2003; Claeys et al., 2004) also suggest the importance of isoprene in aerosol formation, leading to 2 Tg of organic matter per year (Claeys et al., 2004).

Key parameters, such as environmental conditions (mainly temperature and radiation), vegetation type and foliar area, constrain biogenic emissions, thus making them very sensitive to climate and land use changes. In addition, vegetation is not only influenced by climate change but also human activity and development, such as urbanization and agriculture expansion. Mainly initiated by Guenther et al. (1995), global biogenic emissions modeling now benefits from dynamic vegetation models, allowing the calculation of biogenic emissions for the present day, as well as studying their possible future evolution in response to vegetation and climate changes (Levis et al., 2003; Sanderson et al., 2003; Naik et al., 2004; Lathière et al., 2005). The History Database of the Global Environment (HYDE), developed by Goldewijk (2001) to estimate global land use changes over the past 300 years (1700–1990), based on historical statistical inventories and spatial analysis techniques, suggests a global increase of cropland area from  $265 \times 10^6$  ha in 1700 to  $1471 \times 10^6$  ha in 1990, and a more than six fold increase of pasture area from  $524 \times 10^6$  to  $3451 \times 10^6$  ha. Important regional differences are found: most of the land use change occurred during the 19th century in temperate regions of Canada, the United States, Russia and Oceania, whereas in most of the tropical regions, the largest rate of land use conversion occurred at the end of the last century. A

study of the impact of deforestation in Amazonia on tropospheric chemistry (Ganzeveld and Lelieveld, 2004), replacing tropical rainforest by pasture, predicted a large decrease in maximum isoprene emission fluxes, from  $12 \times 10^{15}$  to  $1.2 \times 10^{15}$  molecules  $\text{m}^{-2} \text{s}^{-1}$  ( $=11.9 \times 10^{-11}$  kgC/ $\text{m}^2/\text{s}$ ) due to a decrease in the isoprene emission factor and in the foliar density. Furthermore, a large reduction in the diurnal ozone deposition velocity and an increase of nocturnal soil deposition were predicted. Steiner et al. (2002) estimated that in East Asia, the human-induced land-cover changes, characterized by the conversion of about 30% of the land area from forests to cropland, led to a decrease of 30% in isoprene and 40% in monoterpenes annual emissions. On the other hand, forest restoration efforts are done in some regions such as boreal and temperate zones of North America and western Europe, leading to an increase in forest cover (Stanturf and Madsen, 2002), which could also affect significantly biogenic emission levels and tropospheric chemistry.

The main goal of our work is to study the sensitivity of global biogenic emissions to, first, climate and atmospheric  $\text{CO}_2$  interannual variability and, second, to vegetation distribution changes. We have incorporated a biogenic emission scheme in the dynamic global vegetation model ORCHIDEE to calculate not only isoprene and monoterpenes emissions but also methanol, acetone, acetaldehyde, formaldehyde, formic and acetic acids.

In Sect. 2 of this paper, we describe the interactive biogenic emission and vegetation model used in our study, as well as details of the simulations. Global mean estimates for the 1983–1995 period are given in Sect. 3 and compared to the results of other studies. Analysis of the impact of climate and  $\text{CO}_2$  interannual variability from 1983 to 1995 on the simulated biogenic VOC emissions is presented in Sect. 4. Finally, two scenarios of land use change, tropical deforestation and European afforestation, are considered in Sect. 5 to analyse the sensitivity of biogenic emissions to the vegetation distribution alteration.

## 2 The interactive biogenic emission and vegetation model

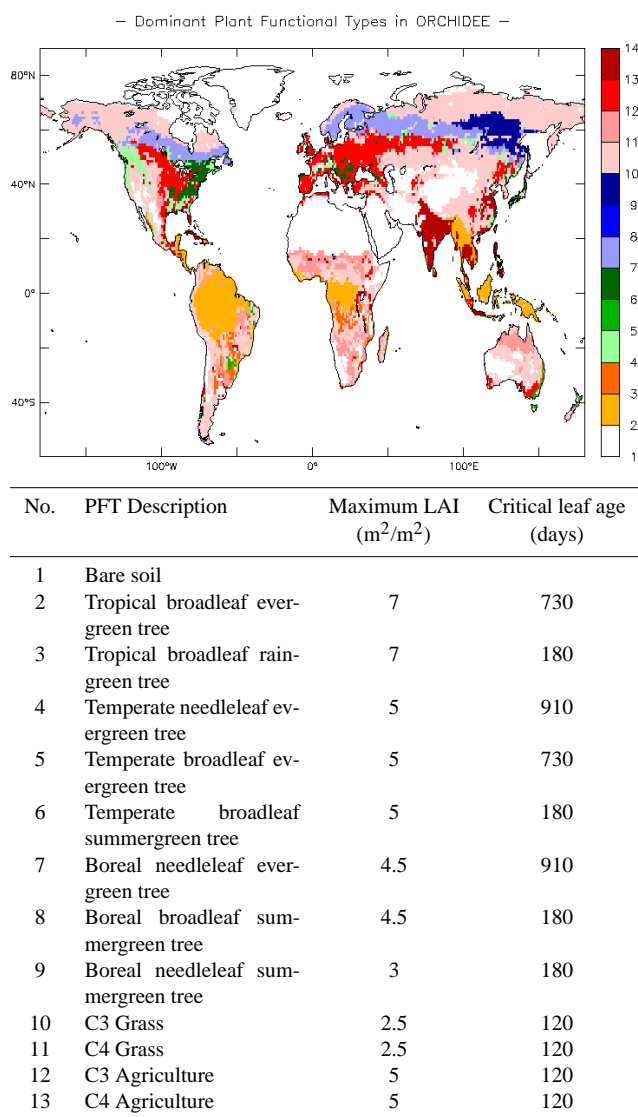
### 2.1 General description

A biogenic emission scheme, based on Guenther et al. (1995) parameterizations, has been incorporated into the dynamic global vegetation model ORCHIDEE (Organizing Carbon and Hydrology in Dynamic Ecosystems) (Krinner et al., 2005). This terrestrial biosphere model consists of the surface-vegetation-atmosphere transfer scheme SECHIBA (Schématisation des échanges hydriques à l'interface biosphere-atmosphère), (Ducoudré et al., 1993; De Rosnay and Polcher, 1998) and of the carbon module STOMATE (Saclay-Toulouse-Orsay Model for the Analysis of Terrestrial Ecosystems), which includes the dynamic

vegetation component of the LPJ (Lund-Potsdam-Jena) dynamic model (Sitch et al., 2003).

SECHIBA calculates processes characterized by short time-scales, ranging from a few minutes to hours, such as energy and water exchanges between the atmosphere and the terrestrial biosphere, photosynthesis, as well as the soil water budget. SECHIBA has a time step of 30 min in order to simulate the diurnal cycle of the terrestrial biosphere. STOMATE treats daily processes such as carbon allocation, litter decomposition, soil carbon dynamics, phenology, maintenance and growth respiration. Important parameterizations, allowing the dynamic simulation of vegetation distribution, have been taken from the global model LPJ: processes such as fire, sapling establishment, light competition, tree mortality and climatic criteria for the introduction and elimination of plant functional types are integrated into ORCHIDEE, with a time step of one year.

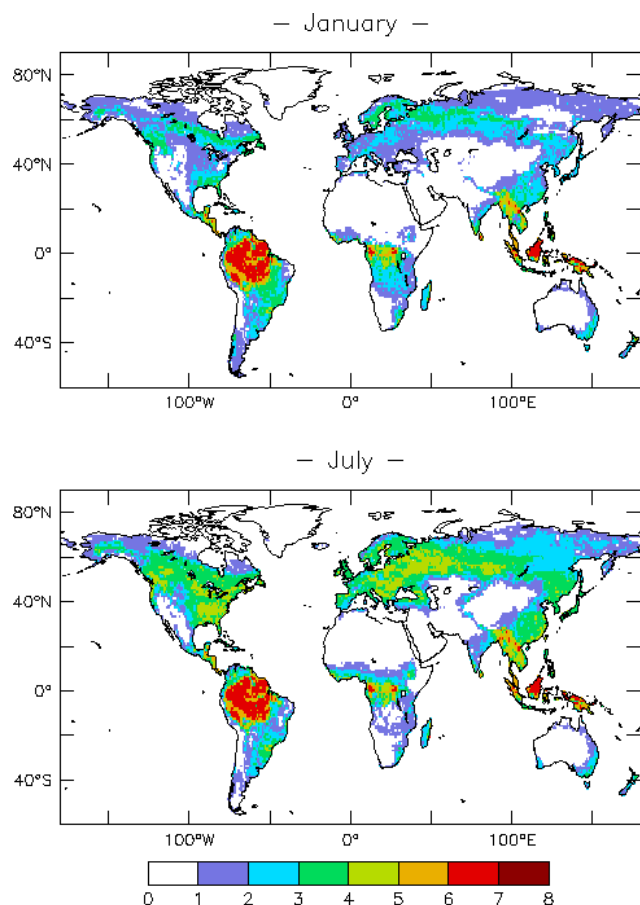
In the ORCHIDEE model, the land surface is described as a mosaic of twelve plant functional types (PFTs) and bare soil (Fig. 1). The definition of PFT is based on ecological parameters such as plant physiognomy (tree or grass), leaves (needleleaf or broadleaf), phenology (evergreen, summergreen or raingreen) and photosynthesis type for crops and grasses (C<sub>3</sub> or C<sub>4</sub>). Relevant biophysical and biogeochemical parameters are prescribed for each PFT (Krinner et al., 2005). This distinction is of great importance since the nature and the amount of the biogenic VOCs emitted are very different from one vegetation type to another. In the current version of ORCHIDEE there is no explicit terrestrial nitrogen cycle. However, nitrogen is implicitly taken into account in the photosynthesis and carbon allocation calculations. For carbon allocation, a nitrogen limitation is parameterized as a function of monthly soil moisture and temperature, because nitrogen availability of a plant depends on microbial activity in soil, and thus on soil moisture and temperature. Photosynthesis is parameterized as an exponential function decreasing with canopy depth, with an asymptotic minimum limit of 30% of the maximum efficiency, to take into account vertical variation of photosynthetic capacity based on leaf nitrogen content. Leaf onset and senescence are calculated, depending on the PFT, by applying warmth and/or moisture stress criteria to the meteorological conditions. Carbon allocation to leaves, and to other compartments such as roots and sap wood, depends on external constraints such as temperature, moisture or light. The more stress a tissue will suffer, the more carbon it receives. In addition, a maximum leaf area index (LAI) value, above which no carbon will be allocated to leaves, is prescribed for each PFT: 7 m<sup>2</sup>/m<sup>2</sup> for tropical trees, 5 m<sup>2</sup>/m<sup>2</sup> for temperate trees, 3 and 4.5 m<sup>2</sup>/m<sup>2</sup> for boreal forests, 2.5 m<sup>2</sup>/m<sup>2</sup> for grasses and 5 m<sup>2</sup>/m<sup>2</sup> for crops (Fig. 1). The leaf photosynthetic efficiency varies during its life cycle: small when the leaf is young, it increases quickly with the leaf age up to a maximum value, lasting until the leaf age equals half its critical value, and then decreases. To take into account this feature, a leaf age structure has been imple-



**Fig. 1.** Dominant plant functional types (PFTs) in ORCHIDEE, corresponding maximum leaf area index (LAI in m<sup>2</sup>/m<sup>2</sup>) and critical leaf age (in days).

mented in the ORCHIDEE model. In the current model version, 4 leaf age classes are considered, each corresponding to various leaf efficiency regimes and of equal duration (=critical leaf age/number of leaf age classes). The critical leaf age is 910 days for needleleaf evergreen trees, 730 days for broadleaf evergreen trees, 180 days for raingreen and summergreen trees and 120 days for grasses and crops. This feature is critical in accounting for the leaf age influence on biogenic emissions (MacDonald and Fall, 1993; Guenther et al., 2000).

ORCHIDEE can either be forced by archived climate fields or driven by a general circulation model and can run in various modes, depending on the activated model components. For instance, the natural PFT distribution can either be



**Fig. 2.** Monthly mean Leaf Area Index (LAI,  $\text{m}^2/\text{m}^2$ ) calculated by ORCHIDEE for January and July.

prescribed from an inventory (static mode, LPJ not activated) or entirely simulated by the model (dynamic mode, LPJ activated), depending on climate conditions. The fraction of the grid box occupied by agricultural PFTs is always prescribed so that crop extent is not affected by the dynamic vegetation change. The atmospheric  $\text{CO}_2$  levels influence photosynthesis, and consequently, the LAI, and can thus indirectly affect VOC biogenic emissions. It is thus really important to consider the relevant  $\text{CO}_2$  atmospheric level for each year and take into account its evolution throughout the period analysed.

Validation studies showed that the ORCHIDEE model accurately simulates water, energy and carbon exchanges, as well as vegetation processes on short and long time-scales. A more detailed description of the ORCHIDEE model and its validation is presented in Krinner et al. (2005), and its good skills for interannual variability have been illustrated by Ciais et al. (2005).

## 2.2 Biogenic emission model

In addition to isoprene and monoterpenes, we also explicitly estimate the emissions of methanol, acetone, acetaldehyde, formaldehyde, formic and acetic acids, which are usually considered as a family of compounds and estimated as bulk emissions. VOC biogenic emissions are calculated every 30 minutes, based on the Guenther et al. (1995) parameterizations, and integrate additional feature such as the leaf age influence on isoprene and methanol emissions.

The generic formula is:

$$F = LAI \times s \times E_f \times C_T \times C_L \times L_a \quad (1)$$

where  $F$  is the flux of the considered biogenic species, given in  $\mu\text{gC}/\text{m}^2/\text{h}$  and LAI the leaf area index in  $\text{m}^2/\text{m}^2$ , calculated at each time step by the model (Fig. 2). The specific leaf weight  $s$  in  $\text{gdm}/\text{m}^2$  (dm: dry matter) is prescribed in ORCHIDEE depending on the considered PFT.  $E_f$  is the emission factor in  $\mu\text{gC}/\text{gdm}/\text{h}$  prescribed for each PFT and biogenic species (Table 1). Isoprene, and monoterpenes emission factors are based on Guenther et al. (1995) and adapted to the PFTs considered in ORCHIDEE. For methanol, we use emission factors from Guenther et al. (2000) and MacDonald and Fall (1993), crops being the highest methanol emitters. For the other VOCs (acetone, aldehydes and acids), we use emission factors from Kesselmeier and Staudt (1999) and Janson and De Serves (2001).

$C_T$  (Eq. 2) and  $C_L$  (Eq. 3) are adjustment factors which account for the influence of leaf temperature and photosynthetically active radiation (PAR) on biogenic emissions and depend on the considered biogenic species. For isoprene, both light and temperature dependencies are considered and the radiation extinction inside the canopy is also taken into account. The leaf area index, used in Eq. (1), is divided into a sunlit and a shaded fraction and a separate calculation is applied to each to estimate the incident PAR flux, used in Eq. (3). The direct PAR is considered to only reach sunlit leaves whereas diffuse PAR is received by both sunlit and shaded leaves (Guenther et al., 1995). The total flux of isoprene is then the sum of emissions from both sunlit and shaded leaves. The PAR is calculated from the surface incident short-wave radiation given in the forcing file (see below). For other chemical species only a temperature dependency is considered as in Guenther et al. (1995):

$$C_T = \frac{\exp\left(\frac{CT1(T-T_s)}{RT_sT}\right)}{1 + \exp\left(\frac{CT2(T-T_M)}{RT_sT}\right)} \text{ for isoprene,}$$

$$C_T = \exp(\beta(T-T_s)) \text{ for all other compounds} \quad (2)$$

$$C_L = \frac{\alpha CL1Q}{\sqrt{1 + \alpha^2 Q^2}} \text{ considered for isoprene only} \quad (3)$$

$T$  is the leaf temperature (K),  $Q$  is the incident flux of PAR ( $\mu\text{mol.phot.m}^{-2}\text{s}^{-1}$ ),  $T_s$  is the leaf temperature at standard conditions ( $=303\text{ K}$ ),  $R$  is the ideal

**Table 1.** Biogenic emission factors in the ORCHIDEE model ( $\mu\text{gC/gdm/h}$ ).

PFT	Isoprene	Monoterpenes	Methanol	Acetone	Acetaldehyde	Formaldehyde	Formic acid	Acetic Acid
Tropical broadleaf evergreen tree	24	0.8	0.6	0.29	0.1	0.07	0.01	0.002
Tropical broadleaf raingreen tree	24	0.8	0.6	0.29	0.1	0.07	0.01	0.002
Temperate needleleaf evergreen tree	8	2.4	1.8	0.87	0.3	0.2	0.03	0.006
Temperate broadleaf evergreen tree	16	1.2	0.9	0.43	0.15	0.1	0.015	0.003
Temperate broadleaf summergreen tree	45	0.8	0.6	0.29	0.1	0.07	0.01	0.002
Boreal needleleaf evergreen tree	8	2.4	1.8	0.87	0.3	0.2	0.03	0.006
Boreal broadleaf summergreen tree	8	2.4	1.8	0.87	0.3	0.2	0.03	0.006
Boreal needleleaf summergreen tree	8	2.4	1.8	0.87	0.3	0.2	0.03	0.006
C3 grass	16	0.8	0.6	0.29	0.1	0.07	0.01	0.002
C4 grass	24	1.2	0.9	0.43	0.15	0.1	0.015	0.003
C3 agriculture	5	0.2	2	0.07	0.025	0.017	0.0025	0.0005
C4 agriculture	5	0.2	2	0.07	0.025	0.017	0.0025	0.0005

gas constant ( $=8.314 \text{ JK}^{-1} \text{ mol}^{-1}$ );  $C_{T1}$  ( $=95\,000 \text{ Jmol}^{-1}$ ),  $C_{T2}$  ( $=230\,000 \text{ Jmol}^{-1}$ ),  $T_M$  (314 K),  $\beta$  ( $=0.09 \text{ K}^{-1}$ ),  $\alpha$  ( $=0.0027$ ) and  $C_{L1}$  ( $=1.066$ ) are empirical coefficients.

Air temperature is often considered as an approximation of leaf temperature in biogenic emission models when a more detailed canopy energy balance is not available. The choice of the temperature used to calculate emissions is critical since temperature is a major factor governing biogenic emissions levels. Leaf temperature measurement is rather complex and published studies, analysing the difference between leaf and air temperature, are quite contrasted. Loreto et al. (1996) indicated that leaf temperature did not strictly follow changes in air temperature. Pier and McDuffie (1997) measured median leaf-to-air temperature differences of about  $2^\circ\text{C}$  at the canopy top and of  $0.5^\circ\text{C}$  or less within the canopy. Kirstine et al. (1998) found, on a pasture site, that the difference between leaf and air temperature, which tends to zero under freely evaporating conditions, increases under drought-stressed conditions by up to  $5\text{--}10^\circ\text{C}$ . Martin et al. (1999) measured, over a two-month period, that the leaf temperature exceeded air temperature by more than  $2^\circ\text{C}$  on 10% of the daylight hours and the excess could go beyond  $6^\circ\text{C}$ . Since the vegetation model ORCHIDEE does not calculate a separate leaf temperature, we use the surface temperature, which is the radiative budget temperature, including soil surface and canopy. However, the surface temperature can differ by up to  $5^\circ\text{C}$  or more from air temperature. In order to avoid a huge emissions overestimation, we thus impose that the temperature used in our biogenic emission model cannot differ by more than  $2^\circ\text{C}$  from air temperature, which is the most observed difference.

Several studies (MacDonald and Fall, 1993; Guenther et al., 2000) underline the impact of leaf age on the emission rate. They highlight that young leaves have a smaller emission capacity for isoprene, but a higher one for methanol, than mature leaves, and that old leaves have a strongly reduced emission yield. To account for the influence of leaf age, we assign a biogenic emission activity factor  $L_a$  (Eq. 1)

for isoprene and methanol emissions, depending on the four leaf age classes given in ORCHIDEE. For methanol, we assume that mature and old leaf emission (leaf age classes 3 and 4) efficiency is half that of young leaves, and as the emission factors prescribed are typical of young leaves, we thus assign a leaf efficiency factor of 1 for leaf age classes 1 and 2 and of 0.5 for leaf age classes 3 and 4 (MacDonald and Fall, 1993). For isoprene, we consider that the emission factors given in Guenther et al. (1995) correspond to a medium-age vegetation emission capacity and assume that young and old leaves emit 3 times less than mature leaves. Therefore, we assign a leaf efficiency of 0.5 for leaf age classes 1 and 4 and of 1.5 for leaf age classes 2 and 3 (Guenther et al., 1999a).

### 2.3 Simulations

The model is run with a resolution of  $1^\circ$  in longitude and  $1^\circ$  in latitude and forced every 3 h by the ISLSCP-II (International Satellite Land-Surface Climatology Project, Initiative II Data Archive, NASA) satellite based climate archive (Hall et al., 2005). This forcing includes incident longwave and shortwave radiation at the surface, rainfall rate, ambient air temperature, surface pressure, near-surface wind speed, ambient air specific humidity and snowfall rate. These near-surface meteorology data are based on atmospheric reanalyses from the NCEP/DOE (National Center for Environmental Predictions/Department of Energy) Reanalysis 2. For most of the fields, the reanalysis data has been hybridised with observational data or corrected for differences in elevation between the reanalysis model topography and the ISLSCP-II initial topography. The simulations are performed in static mode, which means that the vegetation distribution is prescribed using a global map. The variability of climate conditions and the atmospheric  $\text{CO}_2$  increase will thus not affect the vegetation distribution, but will impact the vegetation growth, and especially photosynthesis activity and carbon allocation to leaves. For our study, we use a present-day global map (Fig. 1) based on Loveland et al. (2000) and corrected for crops by Ramankutty and Foley

**Table 2.** CO<sub>2</sub> atmospheric levels and temperature conditions for the 1983–1995 interannual variability simulation.

Year	CO <sub>2</sub> mixing ratio (ppm) and increase from 1983 (%)	Global mean air temperature (°C)
1983	342.7	13.7
1984	344.5 (+0.52%)	13.3
1985	345.7 (+0.87%)	13.3
1986	347.3 (+1.34%)	13.5
1987	348.9 (+1.81%)	13.6
1988	351.5 (+2.57%)	13.8
1989	353.1 (+3.03%)	13.6
1990	353.9 (+3.27%)	13.9
1991	355.7 (+3.79%)	13.8
1992	356.4 (+3.99%)	13.4
1993	356.9 (+4.14%)	13.6
1994	358.6 (+4.63%)	13.7
1995	360.3 (+5.13%)	14.1
Mean	351.9	13.6

(1999) and for anthropogenic grasses by Goldewijk (2001) (De Noblet-Ducoudré and Peterschmitt, unpublished work). Initial conditions are taken from a 10-year equilibrated simulation based on 1983 climate and CO<sub>2</sub> conditions. The interannual simulation, run for the 1983–1995 period, considers not only climate change but also annually varying CO<sub>2</sub> atmospheric levels (Table 2): the CO<sub>2</sub> mixing ratio increases from 342.7 ppmv in 1983 up to 360.3 ppmv in 1995, a 5% increase over the 1983–1995 period (Keeling and Whorf 2005). The global and annual mean air temperature ranges from 13.3°C to 14.1°C (Table 2). The impact of key parameters, such as radiation extinction or temperature, on biogenic emissions is assessed running two sensitivity simulations, based on the 1983 climate and atmospheric CO<sub>2</sub> conditions.

In order to evaluate the impact of human activities on ecosystems distribution and hence on biogenic emissions, we perform a second set of simulations: tropical deforestation and European afforestation. For the tropical deforestation simulation, PFTs 2 and 3 (tropical broadleaf evergreen and raingreen trees), located in large regions of South America, Africa and Indonesia, were replaced by 50% tropical crops and 50% tropical grasses (PFTs 11 and 13) while for the European afforestation simulation, we substituted crops (PFTs 12 and 13) with temperate broadleaf summer-green trees (PFT 6) in the regions between 37° N–70° N and 12° W–44° E. For these simulations, we have only addressed the issue of human induced land cover change and ignored the potential impacts of climate and CO<sub>2</sub> changes and of natural vegetation redistribution. We have then run one specific year using only the 1983 climate and CO<sub>2</sub> conditions. Although these simulations do not include dynamic vegetation response to projected future climate and land use change,

they do represent predicted regional land use tendencies and their impact on regional and global biogenic emissions.

### 3 Global mean biogenic emissions for the 1983–1995 period

#### 3.1 Comparison with other studies

Calculated global mean emissions for the 1983–1995 period are summarized and compared to previous estimates in Table 3. On a 13 years-mean basis, the global biogenic emission source calculated by our model totals 752 TgC/yr of which 460 TgC/yr are isoprene, 117 TgC/yr monoterpenes, 106 TgC/yr methanol, 42 TgC/yr acetone, 15 TgC/yr acetaldehyde, 10 TgC/yr formaldehyde, 1.5 TgC/yr formic acid and 0.3 TgC/yr acetic acid. Our results are within the range found in the literature. The slight underestimation when compared with Guenther et al. (1995) estimate of 506 TgC/yr for isoprene global emission results from our accounting of the leaf age influence. In a sensitivity study, we have indeed neglected this parameter and found a global isoprene emission of 506 TgC/yr, identical to Guenther et al. (1995) and closer to the values reported by Wang and Shallcross (2000) and Levis et al. (2003). Monoterpenes emission calculated by ORCHIDEE is close to the 127 TgC/yr given by Guenther et al. (1995). Monoterpenes and other VOC emissions may also be dependent on the leaf age, although, to the best of our knowledge, this effect has only been studied for isoprene and methanol emissions. Global emissions calculated for aldehydes and acids are within the range given by Wiedinmyer et al. (2004). Biogenic emissions of acids total 1.8 TgC/yr from which formic acid emissions contribute to 1.5 TgC/yr, 3 times less than the production of organics by ozonolysis reaction, estimated to 5 TgC/yr (Baboukas et al., 2000).

Our estimate of global isoprene emission is similar to the 454 TgC/yr reported by Naik et al. (2004). Naik et al. (2004) considered a potential vegetation map with no agricultural land, which should lead to higher emissions than ours. However, they assumed that grasses are not a major emitter of isoprene (emission factor of 0) while we use emissions factors of 16 and 24 μgC/gdm/h for C<sub>3</sub> and C<sub>4</sub> grasses, respectively (Guenther et al., 1995), that results in an additional emission of 90 TgC/yr into the atmosphere.

Sanderson et al. (2003) calculated global emissions of 484 TgC/yr for isoprene, a little higher than our results, which can be explained again by the difference in emission factors. Based on a sensitivity analysis, we found that adopting the emission factors proposed by Sanderson et al. (2003) would lead to a global isoprene source about 485 TgC/yr.

Tao and Jain (2005) estimated isoprene global biogenic emissions to 601 TgC/yr, taking into account the leaf age influence. The emission factors were not assigned on a biome basis but depended on the grid cell considered and

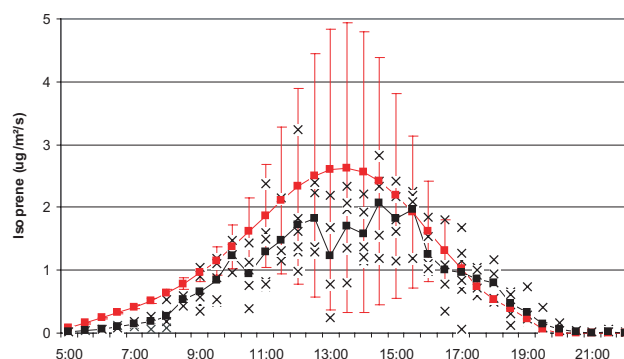
**Table 3.** Global mean biogenic emissions (TgC/yr) and contribution to total biogenic VOCs (%) over the period 1983–1995.

TgC/yr	Isoprene	Monoterpenes	Methanol	Acetone	Acetaldehyde	Formaldehyde	Formic acid	Acetic acid	TOTAL
This study	460 (61%)	117 (15%)	106 (14%)	42 (6%)	15 (2%)	10 (1.5%)	1.5 (0.2%)	0.3 (0.05%)	752
Galbally and Kirstine (2002)			37.5						
Guenther et al. (1995)	503	127							1150
Horowitz et al. (2003)			107						
Levis et al. (2003)	507	33							692
Naik et al. (2004) (potential vegetation)	454	72							
Sanderson et al. (2003)	484								
Tie et al. (2003)			108						
Wang and Shallcross (2000)	530								
Wiedinmyer et al. (2004)			50–250	10–50	10–50	2–10	0.4–2	0.4–2	

thus showed a strong variability throughout the world, which is not the case in our study.

If the results calculated with our model compare well with other estimates on a global basis, they do emphasize the sensitivity of computed biogenic emissions to the chosen emission factors. Indeed, an emission factor allocation based only on PFTs may not be sufficient to take into account the large emission factor variability from one ecosystem to another and to provide a realistic regional distribution of biogenic compounds, which can be of great importance, especially for atmospheric chemistry studies. Serça et al. (2001) investigated isoprene emission in a forest-site of the Republic of Congo, as part of the EXPRESSO program, and found that of the 28 species from the mixed forest, only 3 were high isoprene emitters (emission rate over  $16 \mu\text{gC/g/h}$ ) while in the monospecific forest, 2 species had high emission rates (45 and  $62 \mu\text{gC/g/h}$ ), for the same conditions of temperature and PAR. Growth environmental conditions such as long-term variation in light, temperature and water stress presumably regulate emissions capacity as observed for isoprene (Harley et al., 1999; Hanson and Sharkey, 2001a; Pétron et al., 2001) and monoterpenes (Staudt et al., 2003). VOC biogenic emissions calculation should then integrate those parameters and a more complex emission factors prescription, depending not only on PFT but also on the geographical location should then be used, as proposed by Guenther et al. (2006) in the MEGAN model.

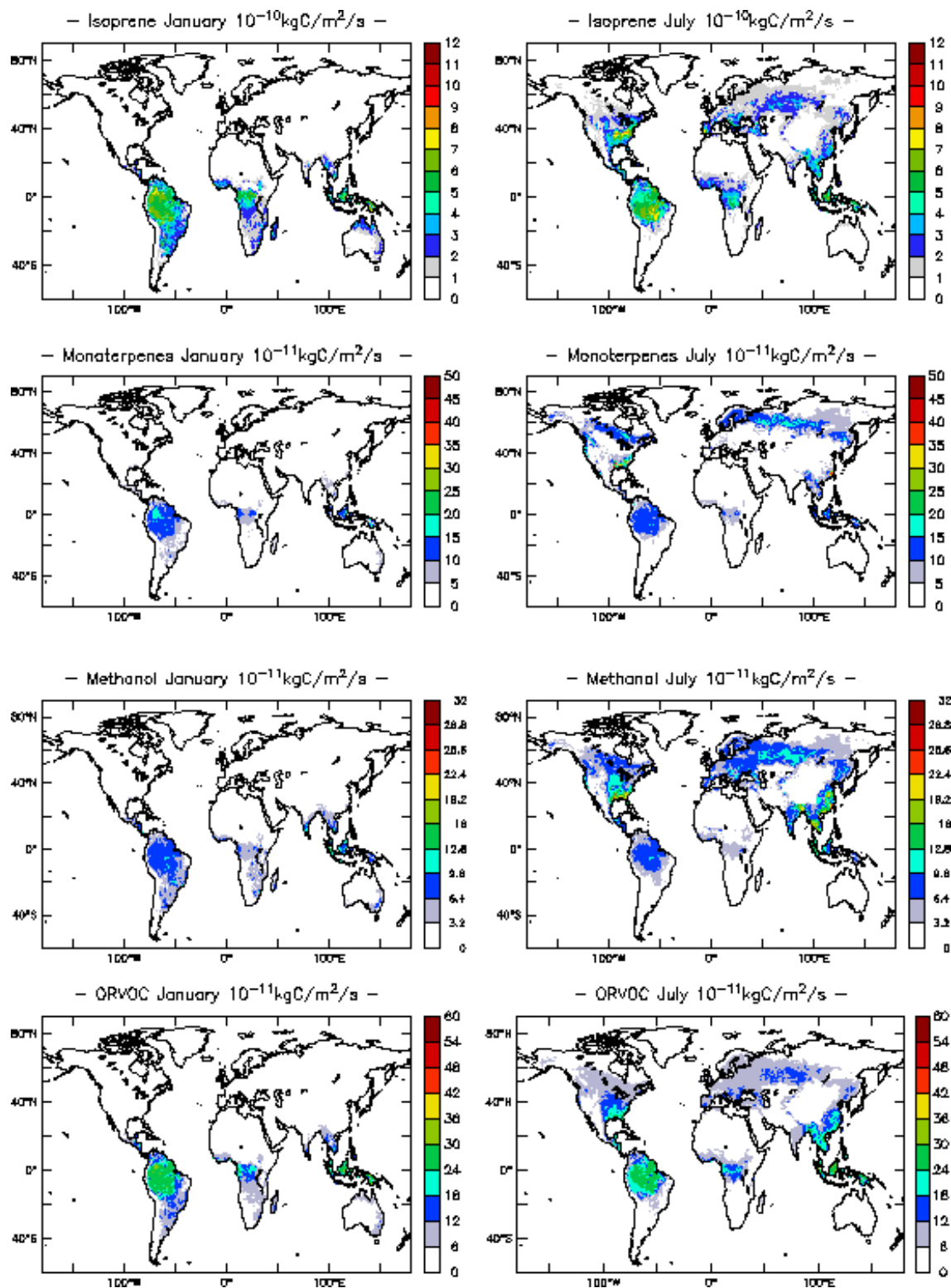
Galbally and Kirstine (2002) estimated methanol emissions by flowering plants to 100 Tg of methanol per year ( $37.5 \text{ TgC/yr}$ ), based on plant structure and metabolic properties. This work, which leads to significantly lower emissions compared to ours, uses a completely different approach based on a plant physiology model. Horowitz et al. (2003) estimated biogenic methanol emissions of  $108 \text{ TgC/yr}$ , based on the emission ratio to isoprene found by Guenther et al. (2000) for North America. Tie et al. (2003) studied the impact of biogenic methanol on tropospheric oxidants as-



**Fig. 3.** Comparison of the ORCHIDEE results with measurements of the ECHO campaign (Spirig et al., 2005) for isoprene ( $\mu\text{g/m}^2/\text{s}$ ). Monthly mean ORCHIDEE isoprene emissions for July (red squares) and variance (red bars) are compared to the mean diurnal cycle (black squares) measured during the ECHO campaign, and to the fluxes reported day-by-day (black crosses).

suming a global surface methanol source of  $117 \text{ TgC/yr}$ , of which 92% is due to biogenic sources, that is  $108 \text{ TgC/yr}$ , which is very similar to our results.

Comparison of the ORCHIDEE monthly mean emissions fluxes over the 1983–1995 with a limited compilation of measurements is given in Table 4 and show that the results of our model are broadly within the range of selected measurements. In the Fig. 3, the diurnal isoprene emissions cycle calculated by our model is compared to measurements of the ECHO campaign (Spirig et al., 2005), which took place in North-West Germany in July 2003, and we can see that in this case, the diurnal variation of isoprene fluxes is quite well captured by our model. It is of course difficult to evaluate a global model based only on a few comparisons with measurements, and a more detailed validation is required. Nevertheless, the examples shown in Table 4 and Fig. 3 underline that our model is generally consistent with the measurements.



**Fig. 4.** Global mean biogenic emissions over the 1983–1995 period in  $\text{kgC/m}^2/\text{s}$  in January (left column) and July (right column) for isoprene, monoterpenes, methanol and ORVOC.

**Table 4.** Comparison of the ORCHIDEE 1983–1995 mean biogenic emissions with on-site measurements ( $\text{mgC}/\text{m}^2/\text{h}$ ).

Sources	Location	Compound	Period	Measurement	This study
Pattey et al. (1999)	Canada	Isoprene	July	1.10–4.58	0.43
			September	0.4–1.67	0.13
Westberg et al. (2001)	USA	Isoprene	August	9–10.5 (warm period) 0.9–1.8 (cool period)	1.8
Apel et al. (2002)	USA	Isoprene	Summer	9 (max)	
Karl et al. (2003)	USA	Isoprene	Summer	6.7 (max)	2.2
		Methanol	Spring	0.4 (mean) 0.7 (max)	0.11
		Acetone	Fall	0.3 (mean) 1 (max)	0.1
Greenberg et al. (1999)	Congo	Isoprene	November	0.6–1.3	1.9
Rinne et al. (2002)	Brazil	Isoprene	July	1.8	2.2
			May	0.32	0.25
Ciccioli et al. (2003)	Italy	Monoterpenes	July	2.09	0.5
			November	0.16	0.14
Rinne et al. (1999)	Finland	Monoterpenes	Summer	0.5–0.6 (max)	0.3
Spanke et al. (2001)	Finland	Monoterpenes	Spring and Summer	0.063–0.3 (max)	0.2–0.3

### 3.2 Attribution of global emissions to PFTs and VOC components

Three major biogenic compounds are emitted: isoprene, which contribute to more than 61% to the global annual biogenic emissions, monoterpenes (15%) and methanol (14%) (Table 3). However, even if other compounds such as acetone (6%), acetaldehyde or formaldehyde (2% and 1.5%), formic (0.2%) and acetic (0.05%) acids are emitted in smaller individual quantities, these other species total a significant amount of reactive compounds emitted to the atmosphere, of almost 70 TgC/yr.

Figure 4 shows the geographical distribution of calculated emissions for isoprene, monoterpenes, methanol and OR-VOC (defined as other reactive VOCs with lifetime smaller than 1 day, and calculated with a constant emission factor of  $1.5 \mu\text{gC}/\text{gdm}/\text{h}$  for every PFTs, Guenther et al., 1995). In January, significant emissions of  $2\text{--}8 \times 10^{-10} \text{ kgC}/\text{m}^2/\text{s}$  for isoprene and  $5\text{--}20 \times 10^{-11} \text{ kgC}/\text{m}^2/\text{s}$  for monoterpenes are calculated in the southern hemisphere and tropical regions. Isoprene is primarily emitted in tropical regions, because tropical vegetation (PFTs 2 and 3) are characterized by both high leaf area index up to  $7\text{--}8 \text{ m}^2/\text{m}^2$  (Fig. 2) and strong emissions factors (Table 1). High isoprene emissions occur all year in these regions. In July, high isoprene emissions by  $3\text{--}10 \times 10^{-10} \text{ kgC}/\text{m}^2/\text{s}$  are also calculated locally in the east coast of the US and in southern Spain due to a high density of temperate broadleaf summergreen trees (PFT 6). Monoterpenes emission levels are also significant in tropical regions, but do not extend as much as isoprene emissions to

the south of the Amazon region. In July, high monoterpenes emissions of  $10\text{--}20 \times 10^{-11} \text{ kgC}/\text{m}^2/\text{s}$  are found in Siberia and Canada, whereas tropical regions contribute much less to the global monoterpenes emission. The biogenic emissions of other VOCs such as acetone, acids and aldehydes have the same pattern because their emissions factors and emission parameterization are similar to monoterpenes. Methanol emissions are particularly high in tropical regions such as India or the southern Asia, due to a high density of crops, a strong methanol emitter, in those regions. In July, methanol emissions range from 6 to over  $20 \times 10^{-11} \text{ kgC}/\text{m}^2/\text{s}$  in Indonesia.

Table 5 shows the contribution of various PFTs and regions to the global biogenic emissions on a 1983–1995 mean basis. The contribution of tropical vegetation (PFTs 2 and 3) to the global emissions are 61% for isoprene, 53% for monoterpenes but only 32% for methanol. Crops (PFTs 12 and 13) are significant methanol emitters, contributing to 44% of global methanol emissions, but are much less important for isoprene and other VOCs (6–6.5% of global emissions). Tropical regions are a major contributor to global annual biogenic emissions: 83% of isoprene, 69% of monoterpenes and 66% of methanol global annual emissions originate from tropical regions ( $30^\circ \text{S}$ – $30^\circ \text{N}$ ). In our study, grasses (PFTs 10 and 11) are a significant source of isoprene contributing to 20% of the global yearly emission. This result is in contrast with other studies (Levis et al., 2003; Naik et al., 2004), which assume that grasses do not emit isoprene at a significant level. Even if the maximum leaf area index for grasses is only  $2.5 \text{ m}^2/\text{m}^2$ , we consider quite high isoprene

**Table 5.** Contribution of PFTs and various regions to global biogenic emissions in TgC/yr (and %) on a 1983–1995 mean basis. Tropical regions are defined between  $\pm 30^\circ$  and  $0^\circ$ , higher latitude regions are defined between  $\pm 30^\circ$  and  $\pm 90^\circ$ .

PFT	Isoprene	Monoterpenes	Methanol	Acetone
Tropical broadleaf evergreen tree	258 (56%)	58 (50%)	31 (30%)	21 (50%)
Tropical broadleaf raingreen tree	23 (5%)	3.5 (3%)	2 (2%)	1.5 (3%)
Temperate needleleaf evergreen tree	7 (2%)	13 (11%)	7 (7%)	5 (11%)
Temperate broadleaf evergreen tree	12 (3%)	4.5 (4%)	2.5 (2%)	2 (4%)
Temperate broadleaf summergreen tree	32 (7%)	3 (2%)	1.5 (1%)	1 (2%)
Boreal needleleaf evergreen tree	5 (1%)	11 (9%)	6 (5%)	4 (9%)
Boreal broadleaf summergreen tree	1.5 (0.33%)	2 (2%)	1 (1%)	0.8 (2%)
Boreal needleleaf summergreen tree	1 (0.2%)	1 (0.9%)	1 (0.6%)	0.4 (0.9%)
C3 Grass	44 (10%)	8 (7%)	4.5 (4%)	3 (7%)
C4 Grass	46 (10%)	7 (6%)	4 (4%)	3 (6%)
C3 Agriculture	15 (3%)	3 (3%)	22 (22%)	1 (3%)
C4 Agriculture	16 (3.5%)	3 (3%)	23 (22%)	1 (3%)
Tropics	381 (83%)	82 (69%)	70 (66%)	30 (70%)
Tropical South	213 (46%)	45 (38%)	33 (31%)	16 (38%)
Tropical North	168 (37%)	37 (31%)	37 (35%)	13.5 (31%)
Higher South	8 (2%)	2 (2%)	3 (2%)	0.8 (2%)
Higher North	71 (15%)	34 (29%)	33 (31%)	12 (29%)
Europe	14 (3%)	5 (4%)	7 (6%)	2 (4%)
Global	460	117	106	42

emission factors, as given in Guenther et al. (1995) for hot and cool grass/shrub, and the spatial coverage and density of grasses is often very high. It is thus a critical point to know if grasses can be considered as an isoprene emitter or not since a very different regional isoprene emissions distribution would arise. Several studies emphasized the low isoprene emission level of grasses but the number of species of grass that emit isoprene is highly uncertain (Holzinger et al., 2002; Wiedinmyer et al., 2004). Moreover, high isoprene emission capacities have been measured for shrubs (Guenther et al., 1999b), which are included in PFTs 10 and 11. Grasses are of less importance for other VOCs emissions, for which their contribution is between 8 and 13%.

### 3.3 Sensitivity tests to parameters choices

Additional simulations were conducted for the year 1983 to assess the sensitivity of biogenic emissions to key parameters such as temperature, leaf age or radiation extinction within the canopy (Eq. 1). We estimated that the radiation extinction considered for isoprene emissions leads to the halving of isoprene emission, decreasing from 983 TgC/yr without radiation extinction to 478 TgC/yr. Accounting for the leaf age influence leads to a decrease in global emissions of 10% for isoprene and 27% for methanol. Increasing the surface temperature (used as a surrogate of leaf temperature) in the biogenic emissions model globally by  $1^\circ\text{C}$  leads to an increase in the global emission of 11% for isoprene and 9% for

other compounds (478 TgC/yr to 529 TgC/yr for isoprene, 119 TgC/yr to 129 TgC/yr for monoterpenes, 106 TgC/yr to 116 TgC/yr for methanol and 43 TgC/yr to 47 TgC/yr for acetone) resulting in a substantial additional total emission of VOCs to the atmosphere of 78 TgC/yr. Considering the difficulty in estimating leaf temperature and the canopy energy balance highlights the large uncertainty in calculated biogenic emissions. The next step for biogenic emission model improvement would be to calculate the leaf temperature for sunlit and shaded leaves, as suggested by measurements given by Harley et al. (1997).

## 4 Interannual variability of biogenic emissions over the period 1983–1995

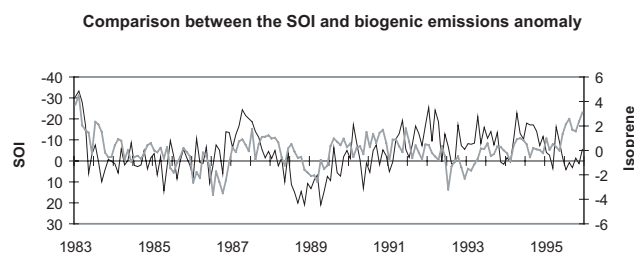
In this section, the impact of the variability in climate conditions and of increasing atmospheric  $\text{CO}_2$  concentrations on biogenic emissions is investigated over the period 1983–1995. As shown in Table 6, the total global emission of all biogenic compounds ranges from 717 TgC/yr in 1986 to 778 TgC/yr in 1995, that is a 8.5% increase between both extreme values, and has a variability of  $\pm 16$  TgC/yr over the 1983–1995 period. In order to assess the impact of atmospheric  $\text{CO}_2$  mixing ratio increase on biogenic emissions, we performed a second 1983–1995 simulation considering the 1983  $\text{CO}_2$  atmospheric level (342.7 ppmv) for the whole period (Table 6). In 1995, the emissions difference between the

**Table 6.** Interannual variability of biogenic emissions over the 1983–1995 period (TgC/yr).

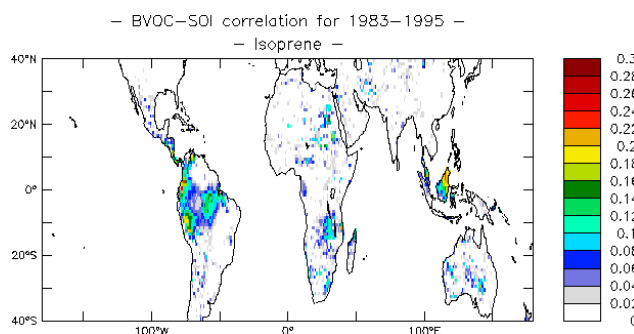
	Isoprene		Monoterpenes	Methanol	Acetone	Acetaldehyde	Formaldehyde	Formic acid	Acetic acid	TOTAL	
	1983 CO <sub>2</sub>	Increasing CO <sub>2</sub>								1983 CO <sub>2</sub>	Increasing CO <sub>2</sub>
1983	478	478	118	106	43	14.8	10.2	1.48	0.295	772	772
1984	459	459	116	105	42	14.5	10.0	1.45	0.291	747	748
1985	454	454	116	105	42	14.6	10.0	1.46	0.291	743	743
1986	435	436	113	102	41	14.1	9.7	1.41	0.283	714	717
1987	465	467	118	106	42	14.7	10.1	1.47	0.293	754	759
1988	454	457	117	106	42	14.6	10.0	1.46	0.292	742	748
1989	451	453	117	106	42	14.6	10.0	1.46	0.292	738	744
1990	465	468	121	109	44	15.1	10.4	1.51	0.30	762	769
1991	459	463	120	107	43	14.9	10.3	1.49	0.299	752	760
1992	445	449	115	103	41	14.4	9.9	1.44	0.287	725	734
1993	451	456	117	106	42	14.6	10.0	1.46	0.291	736	747
1994	458	462	119	107	43	14.8	10.2	1.48	0.297	747	758
1995	472	476	122	109	44	15.2	10.4	1.52	0.304	768	778

“constant CO<sub>2</sub>” simulation and the “increasing CO<sub>2</sub>” simulation reaches 4 TgC/yr for isoprene and 10 TgC/yr for the total VOC, which corresponds to 0.8% and 1.3% difference, respectively, linked to an increase in foliar biomass under increasing atmospheric CO<sub>2</sub> conditions.

An important control on climate conditions in tropical regions is the occurrence of El Niño and La Niña events, which lead to major warming and cooling cycles in the eastern and central Pacific. The Southern Oscillation Index (SOI), defined as the pressure difference between Tahiti and Darwin, provides the intensity of El Niño and La Niña episodes. As shown in Table 6, biogenic emissions are generally higher during El Niño years (1983, 1987, 1990–1991 and 1994–1995), and lower during La Niña years (1984–1985, 1988–1989). A similar finding was obtained by Naik et al. (2004) over the period 1971–1990. However there is an exception for the 1992–1993 weak El Niño event for which global biogenic emissions are lower than average. For those two years, the El Niño effect on the temperature increase in the western part of South America is very small (with a maximum increase of 0.4°C in northern part of South America compared to 1983–1995 mean temperature of 26–28°C in this region), and does not impact significantly on biogenic emissions levels, whereas a temperature decrease can be noted southward of Brazil and in Central Africa, leading to a decrease in biogenic emissions. The comparison between the SOI and the calculated monthly global isoprene emissions anomalies (Fig. 5) indicates a negative correlation. The correlation coefficient ( $r$ ) between the SOI and monthly global biogenic emissions anomalies, calculated for the period 1983–1995, is  $-0.28$  for isoprene,  $-0.14$  for monoterpenes and  $-0.01$  for methanol. These values are in qualitative agreement with Naik et al. (2004) but of smaller absolute correlation ( $-0.54$  for isoprene and  $-0.24$  for monoterpenes for 1971–1990, in Naik et al., 2004). The correlation coefficients obtained



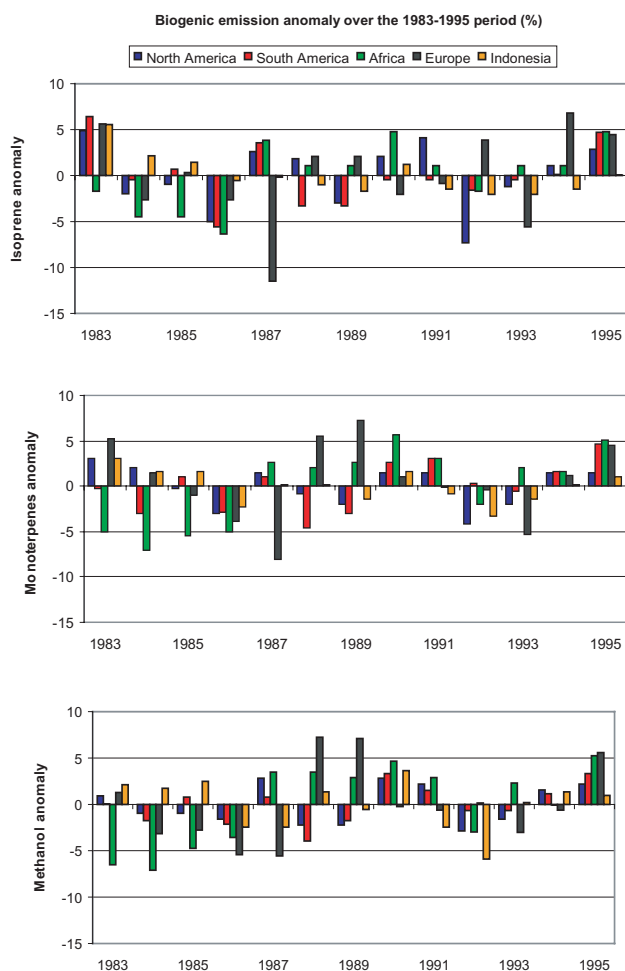
**Fig. 5.** Comparison between the Southern Oscillation Index SOI (black line) and the simulated anomalies of global monthly biogenic isoprene emissions (grey line, TgC/month) for the 1983–1995 period.



**Fig. 6.** Square correlation coefficient ( $r^2$ ) between the Southern Oscillation Index and biogenic isoprene emissions for the 1983–1995 period.

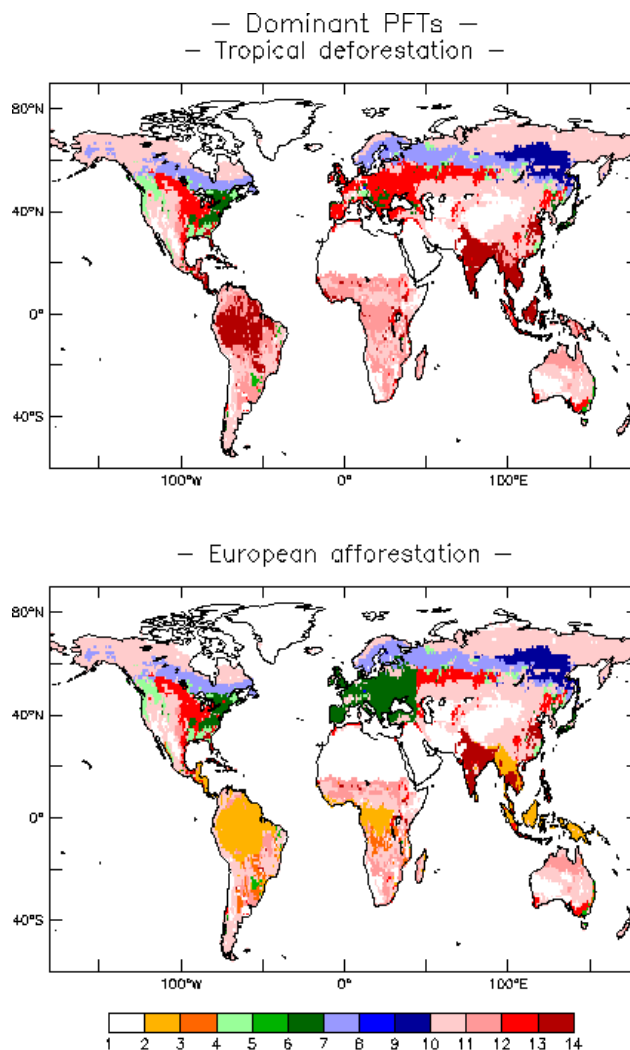
are rather small but it is however interesting to note that the correlation between biogenic emissions and SOI is somewhat higher in tropical regions compared to other regions, as shown in Fig. 6 for isoprene.

As indicated by Fig. 6, the variability in biogenic emissions over the 13 years of the simulation varies from one



**Fig. 7.** Variability of annual biogenic emissions anomaly based on 1983–1995 mean emissions calculated for North America, South America, Africa, Europe and Indonesia for isoprene (upper panel), monoterpenes (middle panel) and methanol (lower panel).

region to another. To further illustrate this feature, Fig. 7 shows the annual emission anomaly in %, compared to 1983–1995 mean emissions, calculated for isoprene, monoterpenes and methanol for 5 regions: North America (150 W–52 W, 12 N–70 N), South America (85 W–34 W, 60 S–15 N), Africa (18 W–51 E, 35 S–37 N), Europe (10 W–50 E, 35 N–72 N) and Indonesia (60 E–160 E, 11 S–40 N). Biogenic emissions variability is strongly affected by the evolution of environmental conditions such as radiation, temperature or leaf area index. In Europe, in relation to a strong air temperature variability over the period 1983–1995, the emission anomaly has the largest variability from  $-11.5\%$  to  $+6.8\%$  for isoprene, from  $-8.1\%$  to  $+7.2\%$  for monoterpenes and from  $-5.6\%$  to  $+7.2\%$  for methanol. In 1987, biogenic emissions have a strong negative anomaly in Europe, where annual mean air temperature differs the most from 1983–1995 mean temperature compared to other regions studied, especially for



**Fig. 8.** PFTs distribution considered for the Tropical deforestation (upper panel) and European afforestation (lower panel) scenarios.

isoprene ( $-11.5\%$ ) and monoterpenes ( $-8.1\%$ ). The amplitude of the isoprene emission anomaly is about 10% for North and South America and Africa, but smaller for Indonesia (7.5%). Variation in the emission anomaly is lowest in Indonesia ( $-3.3\%$  to  $+3\%$ ) and North America ( $-4.2\%$  to  $+3.1\%$ ) for monoterpenes and in North ( $-2.9\%$  to  $2.8\%$ ) and South ( $-3.9\%$  to  $+3.3\%$ ) America for methanol.

## 5 Impacts of tropical deforestation and European afforestation on global biogenic emissions

### 5.1 Tropical deforestation

Figure 9 illustrates the change in total annual mean leaf area index between the tropical deforestation scenario and the 1983 control run (1983 simulation with non-modified present vegetation map). In the tropical regions with low

**Table 7.** Global biogenic emissions (TgC/yr) and changes (%) for the tropical deforestation simulation and European afforestation simulation compared to the control run (nc: no change).

TgC/yr (%)	1983 Control Run			Tropical deforestation		European afforestation	
	Global	Tropics	Europe	Global	Tropics	Global	Europe
Isoprene	478	396	15	339 (−29%)	257 (−35%)	497 (+4%)	33 (+126%)
Monoterpenes	118	81	5	84 (−29%)	48 (−41%)	118 (−0.1%)	6 (+13%)
Methanol	106	70	7	130 (+22%)	93 (+33%)	102 (−3.5%)	3 (−51%)
Acetone	43	29	2	30 (−29%)	17 (−42%)	43 (−0.2%)	2 (+10%)
Acetaldehyde	15	10	0.64	10 (−29%)	6 (−42%)	15 (nc)	0.7 (+16%)
Formaldehyde	10	7	0.44	7 (−30%)	4 (−44%)	10 (nc)	0.5 (+14%)
Formic acid	1.5	1	0.06	1 (−29%)	0.6 (−44%)	1.48 (nc)	0.07 (+14%)
Acetic acid	0.29	0.20	0.013	0.21 (−27%)	0.12 (−40%)	0.29 (nc)	0.015 (+15%)
TOTAL	772	595	30	602 (−22%)	426 (−20%)	787 (+2%)	46 (+54%)

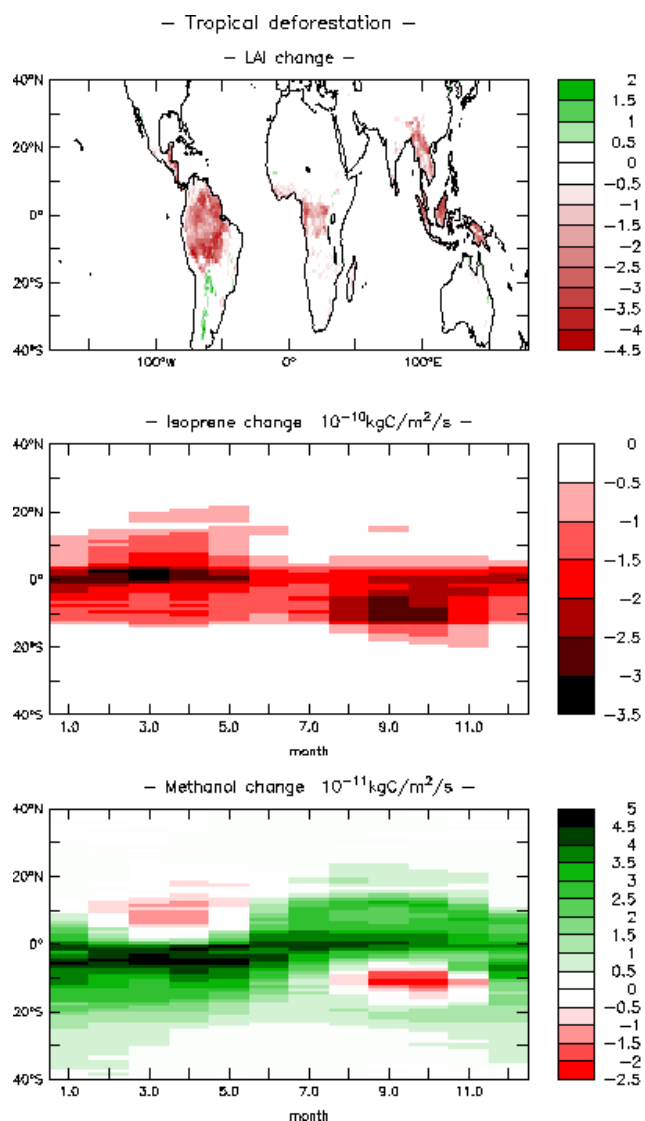
precipitation, such as the southern part of Brazil, the LAI is quite small in the control run ( $1\text{--}3\text{ m}^2/\text{m}^2$ ). Deforestation leads to a small increase in LAI in this region between 1 and  $1.5\text{ m}^2/\text{m}^2$ . In significant parts of Amazonia, Central Africa and Indonesia, a large decrease in LAI in the range  $2\text{--}4.5\text{ m}^2/\text{m}^2$  is modelled.

Tropical deforestation has a major impact on biogenic emissions, as shown in Table 7. On a global basis, methanol emissions increase by 22%, in relation to crops coverage expansion, whereas isoprene and other VOC emissions decrease by 27–30%. If we consider tropical regions only, a 33% increase in methanol emissions occurs and a 35–44% decrease is predicted for isoprene and other VOCs. The isoprene emission decrease calculated for East Asia reaches 26%, which is closed to the 30% decrease calculated by Steiner et al. (2002). Moreover, as illustrated in Fig. 9 by the zonal mean difference of biogenic emissions between the tropical deforestation simulation and the control run, the seasonality of emissions is also affected by the tropical deforestation. The decrease in isoprene zonal mean emissions, governed by a reduction in both LAI and emission factors under tropical deforestation conditions, is significant all year long ( $-0.5\times 10^{-10}$  to  $-2\times 10^{-10}\text{ kgC/m}^2/\text{s}$ ) and peaks in March north of the Equator and in September south of the Equator ( $-2\times 10^{-10}$  to  $-3.5\times 10^{-10}\text{ kgC/m}^2/\text{s}$ ). Methanol zonal mean emissions increase by  $0.5\text{--}4\times 10^{-11}\text{ kgC/m}^2/\text{s}$  throughout the year, and by  $4\text{--}5\times 10^{-11}\text{ kgC/m}^2/\text{s}$  from January to September between  $0^\circ$  and  $10^\circ\text{ S}$ . A decrease in zonal mean methanol emissions is also modelled north of the equator in March, and south of the equator in September ( $-0.5$  to  $-2.5\times 10^{-11}\text{ kgC/m}^2/\text{s}$ ). This reduction in methanol emissions is due to the large decrease in LAI in these regions, and occurs despite the significantly larger methanol emission factors of crops compared to tropical trees (Table 1). These results illustrate the importance of tropical regions as a major and very sensitive biogenic emissions source, under pressure from rapid and strong land use change.

## 5.2 European afforestation

Under the European afforestation scenario, the total annual mean LAI tends to decrease by  $1\text{ m}^2/\text{m}^2$  over large regions of Europe, and up to  $2\text{ m}^2/\text{m}^2$  in Great-Britain, where the highest LAI decrease occurs. LAI increases locally by up to  $2.5\text{ m}^2/\text{m}^2$  in the centre of Europe, northward of the Black Sea (Fig. 10). This result, which can seem surprising, is linked to the fact that we turn perennial crops into a summergreen vegetation, both characterized by a maximum LAI of  $5\text{ m}^2/\text{m}^2$ . We know that perennial crops are not realistic and this is being changed in ORCHIDEE (Gervois et al., 2004). Nevertheless, this bias in the representation of crops only marginally affects the results discussed here.

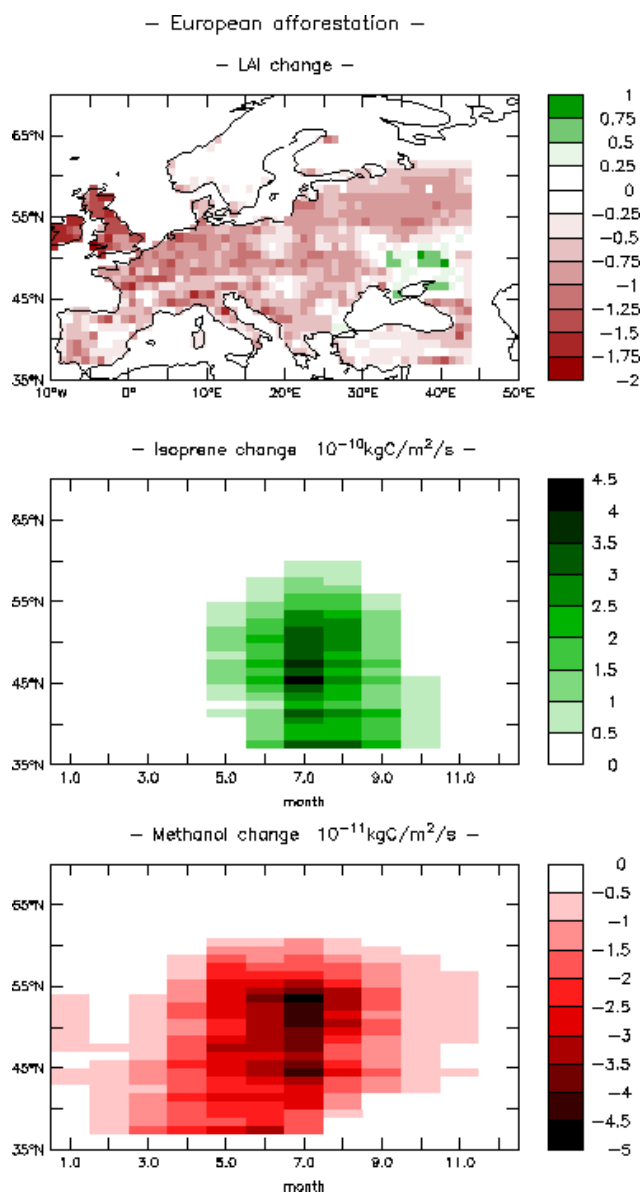
Globally, emissions are not significantly affected by European afforestation: a 4% increase is calculated for isoprene and a 3.5% decrease for methanol, and less than 0.2% for other VOCs is computed. However, at the European scale, the impact is very significant, with a 51% decrease in methanol emissions, an increase of 126% for isoprene and 10–15% for other VOCs, leading to an increase in total biogenic emissions in Europe of 54%. The very large increase in the isoprene emission factor (multiplied by 9 compared to crops) leads to a large overall increase in biogenic emissions, despite the small decrease in LAI. The combined effect of total LAI and emission factor reduction leads to a large decrease in methanol emissions, lasting through the year since methanol crops emissions also occur in winter in the control run while there is no emission by PFT 6 (summergreen trees) during winter. The change in zonal mean emissions, given in Fig. 10, shows a  $0.5\times 10^{-10}$  to  $4.5\times 10^{-10}\text{ kgC/m}^2/\text{s}$  increase in zonal mean isoprene emissions from April to November. Zonal mean methanol emissions decrease by  $0.5\times 10^{-11}\text{--}1.5\times 10^{-11}\text{ kgC/m}^2/\text{s}$ , between January and March and after October, and by  $1.5\times 10^{-11}\text{--}5\times 10^{-11}\text{ kgC/m}^2/\text{s}$ , during spring and summer. For both isoprene and methanol, the maximum change in zonal mean emissions occurs in July



**Fig. 9.** Impact of a tropical deforestation scenario on total annual mean LAI (m<sup>2</sup>/m<sup>2</sup>) (upper panel), zonal mean isoprene (middle panel) and methanol (lower panel) emissions compared to the 1983 control run.

between 42° N and 52° N. Even though global biogenic emissions are not significantly affected by European afforestation, its impact on regional emissions is very large and could strongly influence tropospheric chemistry mechanisms both within and outside Europe.

The vegetation distribution change considered in our study is only intended to be a sensitivity experiment and probably overestimates the future changes. Nevertheless, the results obtained in the tropical deforestation and European reforestation simulations underline the strong impact of vegetation distribution alteration on VOC biogenic emissions as well as the high dependency of emission levels to the evolution of land management.



**Fig. 10.** Impact of a European afforestation scenario on total annual mean LAI (m<sup>2</sup>/m<sup>2</sup>) (upper panel), zonal mean isoprene (middle panel) and methanol (lower panel) emissions compared to the 1983 control run.

## 6 Conclusion

A biogenic emission scheme has been integrated in the global vegetation model ORCHIDEE in order to calculate biogenic emissions of isoprene, monoterpenes, methanol, acetone, acetaldehyde, formaldehyde, as well as formic and acetic acids at the global scale. In order to study the impact of climate and land use changes on biogenic emissions, several simulations are performed. Global mean emissions of 460 TgC/yr for isoprene, 117 TgC/yr for monoterpenes, 106 TgC/yr for methanol and 42 TgC/yr for acetone are calculated over the

period 1983–1995. Tropical regions are identified as the primary source of global biogenic VOCs, contributing to 83% of isoprene, 69% of monoterpenes and 66% of methanol global annual emissions. The interannual simulation predicts a substantial variability in biogenic emissions over the 1983–1995 period. The total global and annual emissions of the summed VOCs range from 718 TgC/yr (1986) to 778 TgC/yr (1995), i.e. an 8.4% variability over 1983–1995. The variability in emissions is markedly different from one region to another. Among the regions studied, biogenic emissions variation compared to 1983–1995 mean emissions was found to be the highest in Europe and the smallest in Indonesia (isoprene and monoterpenes) and North America (methanol).

Two scenarios of land use change, tropical deforestation and European afforestation, have been considered to examine the sensitivity of biogenic emissions to the vegetation distribution alteration. On the global scale, our calculations indicate that the tropical deforestation leads to a methanol emission increase by 22%, whereas isoprene and other VOCs emissions decrease by 27–30%. If we consider tropical regions only, a 33% increase is calculated for methanol emissions, and a 35–44% decrease is estimated for isoprene and other VOCs. Global emissions are not significantly affected by the European afforestation, but at the European scale, the impact is much more important, with a 51% decrease for methanol emissions and an increase of 126% for isoprene and 10–15% for other VOCs, leading to an increase in the total biogenic emissions in Europe of 54%.

Our results are generally consistent with previous studies. It should be noted that our study also takes into account features such as the leaf age for isoprene and methanol emissions. Based on a sensitivity test, we estimated that accounting for the leaf age influence leads to a decrease in global emissions of 10% for isoprene and 27% for methanol. Future biogenic emission studies need to consider if grasses can be considered as a significant isoprene emitter or not as this could greatly affect regional isoprene emissions distribution. Indeed, we calculate that grasses contribute to nearly 20% of the isoprene global and annual emission whereas recent studies such as Levis et al. (2003) and Naik et al. (2004) assume that the grass isoprene emission factor is negligible. Depending on this choice, our emission estimates for land-use change scenarios, such as European afforestation and Tropical deforestation, could be significantly different.

Air temperature above the canopy is used to calculate biogenic emissions when a more accurate canopy energy balance, estimating leaf temperature is not available. Pier and McDuffie (1997) observed, comparing measurements and model emissions, that calculating biogenic emissions based on air temperature leads to a slight underestimation of high emission rates. Hanson and Sharkey (2001b) pointed out that biogenic emission models should be adjusted to account for variation in response to temperature and light levels during vegetation growth, which could be done simply by altering the emission capacity (other coefficients used in light and

temperature emissions dependencies were not found to be sensitive to environmental growth conditions). Increasing globally the leaf temperature used in the biogenic emissions model by 1°C leads to an increase in the global emission of 11% for isoprene and 9% for other compounds (478 TgC/yr to 529 TgC/yr for isoprene, 119 TgC/yr to 129 TgC/yr for monoterpenes, 106 TgC/yr to 116 TgC/yr for methanol and 43 TgC/yr to 47 TgC/yr for acetone) resulting in a substantial additional emission of VOCs to the atmosphere of 78 TgC/yr. The difficulty to estimate the leaf temperature and the canopy energy balance appears as a major uncertainty in calculating biogenic emissions. A next step for biogenic emissions models improvement would be to calculate the leaf temperature for sunlit and shaded leaves, as suggested by the measurements of Harley et al. (1997).

The distribution, as well as the diversity, of vegetation throughout the world is under pressure from both direct anthropogenic changes, such as deforestation and agriculture expansion, driven by population growth, and indirect changes, also affected partly by human activities, including atmospheric CO<sub>2</sub> and climate evolution. Alcamo et al. (1996) suggested that in the next 50 years, land-cover conversion may occur predominantly in the tropical and subtropical regions, in response to projections of demographic growth and economic activity. The consequences of such changes not only on biogenic emissions but also on the tropospheric gas phase and particulate chemistry could be significant. The use of dynamic global vegetation models, for future scenarios, and more generally for climate and land use change studies, is an essential next step. Nevertheless, important points, such as the pattern and dependency of emissions on environmental conditions for compounds other than isoprene and monoterpenes, and the response of foliar density and biogenic emission factors to CO<sub>2</sub> and climate change, are still under debate. Rosenstiel et al. (2003) showed that under increased atmospheric CO<sub>2</sub> level from 430 ppmv to 800 and 1200 ppmv, the isoprene production was reduced by 21% and 41% while above-ground biomass accumulation was enhanced by 60% and 82%. We can reasonably consider that considering this influence in our study would not change significantly the estimates calculated over the 1983–1995 period, characterized by a 5% increase of the atmospheric CO<sub>2</sub> but could however be subsequent on longer time-scales. The influences of those various parameters could affect emissions levels significantly, and more information is needed to reduce the estimates uncertainty and improve our understanding of biosphere-atmosphere interactions.

*Acknowledgements.* We thank J.-Y. Peterschmitt for his help in the preparation of the global vegetation maps. Useful comments and discussions on this work by P. Friedlingstein, D. Serça, G. Krinner, A. Guenther and S. Gervois are gratefully acknowledged. Computer time has been provided by the C.C.R.T under project p24. This work was partly funded by the European projects RETRO (EVK2-CT-2002-00170) and ENSEMBLES (GOCE-

CT-2003-505539). G. Folberth acknowledges support provided by the Canadian Centre for Climate Modelling and Analysis, Meteorological Service of Canada.

Edited by: U. Lohmann

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