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Isoprene emission estimates and uncertainties for the Central African EXPRESSO study domain

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Abstract. A global three-dimensional (3-D) chemistry and transport model was used to demonstrate that a factor of 2 decrease in isoprene and monoterpene emissions results in significant (10-30%) changes in predicted concentration distributions of compounds such as OH, MPAN, NO_x, H_2O_2, O_3, and CO. Isoprene and monoterpenes were predicted to have a particularly strong impact on tropical regions, including central Africa. The 1996 Experiment for Regional Sources and Sinks of Oxidants (EXPRESSO) study included a number of experiments that improved our ability to predict isoprene and monoterpene emissions from central Africa. The results of these experiments have been incorporated into an isoprene emission model that predicts hourly emissions on a spatial scale of about 1 km^2. The model uses procedures that are suitable for estimating global emissions but uses regional measurements to accurately parameterize the model. Comparisons with above-canopy aircraft and tower flux measurements demonstrate that the model can estimate emissions within a factor of 2 for regions where ground measurements of model inputs are available. The annual central African isoprene emission predicted by our revised model (35 Tg C) is only 14% less than that predicted by our earlier model, but there are considerable differences in estimates of individual model variables. The models differ by more than a factor of 5 for specific times and locations, which indicates that there are large uncertainties in emission estimates for at least some locations and seasons. The good agreement obtained for the EXPRESSO study field sites, however, suggests that the model can predict reasonable estimates if representative field measurements are used to parameterize the model.

1. Introduction

The chemical composition of the atmosphere is an important component of the global environment and observed trends in trace gas concentrations demonstrate that it is changing rapidly [*Brasseur et al.*, 1998]. Surface emissions have a major role in determining atmospheric trace gas concentrations, and an understanding of the processes controlling emissions is needed to predict future chemical composition. The biogenic emission of isoprene from the leaves of some plant species has been shown to have a strong influence on atmospheric chemistry in regions as diverse as the remote Amazon Basin [*Jacob and Wofsy*, 1988] and the highly populated eastern United States [*Pierce et al.*, 1998]. Isoprene oxidation influences OH and ozone concentrations, has a significant role in CO production, the formation of organic acids, and the photochemical conversion of NO_y species. Since isoprene can influence concentrations of ozone and gases that are removed by OH, including methane, isoprene emissions may indirectly impact climate.

The need for accurate estimates of biogenic volatile organic compound (VOC) emissions as inputs to global chemistry and transport models (CTMs) led to the development of the global model of *Guenther et al.* [1995] under the International Global Atmospheric Chemistry/Global Emissions Inventory Activity (IGAC/GEIA) program. This effort involved experts specializing in many different aspects of biogenic emission modeling. The IGAC-GEIA group produced a model for estimating emissions using the limited data that were available. The model predicted that over 90% of the global annual nonmethane VOC emission is produced by vegetation and that isoprene contributes about half of the global total, making it the single largest VOC emission into the atmosphere [*Guenther et al.*, 1995].

The initial IGAC-GEIA natural VOC model estimated monthly average isoprene emissions on a 0.5° latitude X 0.5° longitude global grid as a function of foliar density, an emission capacity (the emission at specified environmental conditions), and an emission activity factor that accounted for variations due to environmental conditions [*Guenther et al.*, 1995]. Foliar density was based on a simple model constrained by the global estimates of *Box* [1981]. Emission activity was calculated using algorithms that were based on results from greenhouse-grown North American plant species. Isoprene emission capacities were based on field measurements reported in 20 publications. Eighty percent of these field measurement studies were conducted in North America or Europe, which together contribute only 12% of the global isoprene flux. Conversely, South America and Africa accounted for almost two thirds of the global total but were represented by only one study in South America. *Guenther et al.* [1995] reported a strong need for measurements from these continents, particularly in the tropics.

The Experiment for the Regional Sources and Sinks of Oxidants (EXPRESSO) was an international and multi-disciplinary research program designed to investigate processes controlling the chemical composition of the tropical troposphere above central Africa and to consider their impact on the global atmosphere. An overview of the experiment and its objectives is

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given by *Delmas et al.* [this issue]. The objectives included an improved ability to predict isoprene emissions within the EXPRESSO study domain, which ranges from savanna landscapes in the north to tropical rain forest landscapes in the south. The field experiments included enclosure, tower and aircraft isoprene flux measurements [*Klinger et al.*, 1998; D. Serça et al., unpublished data, 1999 (hereinafter referred to as S99); L. Vierling et al., unpublished data, 1999 (hereinafter referred to as V99); *Greenberg et al.*, this issue]. In this paper, we synthesize the results of the EXPRESSO study and other recent work to improve estimates of isoprene emission from central Africa. We begin by analyzing the sensitivity of a global chemistry and transport model to changes in isoprene emission. This model sensitivity study is followed by a description of the methods used to produce improved isoprene emission estimates for the EXPRESSO domain. We then present the results from this model and evaluate its uncertainties by comparison with alternative modeling methods and with EXPRESSO field measurements.

2. Chemistry and Transport Model Sensitivity

A global three-dimensional chemistry and transport model, IMAGES [*Muller and Brasseur*, 1995], was used to characterize the importance of accurate isoprene emission estimates for investigating the distributions of the chemical compounds responsible for the oxidation capacity of the troposphere. The IMAGES model was designed to study the oxidation capacity of the atmosphere and includes over 40 compounds, 125 chemical reactions, and 26 photodissociations (see *Muller and Brasseur* [1995] for more details). Three model scenarios were run for this analysis: (1) no biogenic VOC, (2) biogenic VOC emissions from the IGAC-GEIA database [*Guenther et al.*, 1995], and (3) biogenic VOC emissions at 50% of the IGAC-GEIA database.

The distributions of ozone, CO, NO_x, peroxymethacrylyl nitrate (MPAN), OH, H_2O_2, HO_2, and other compounds predicted by IMAGES were sensitive to the magnitude of biogenic VOC emissions. As expected, the change in the predicted concentration distributions is greatest near terrestrial surfaces where biogenic VOC are emitted. OH concentrations near the surface decrease (and HO_2 and H_2O_2 increase) by 30 to 90% when biogenic VOC are included in IMAGES (compared to the no-biogenic VOC case). As a result, the model predicts that the lifetime of isoprene, or any other compound that exists primarily in the atmospheric boundary layer above terrestrial surfaces and is primarily removed by OH, increases by a factor of 5 when biogenic VOC are included. A factor of 2 reduction in biogenic VOC emissions decreases the lifetime of isoprene by a factor of 1.6. Plate 1 illustrates the model prediction of a substantial decrease (10 to 20%) in OH concentrations at altitudes between 7 and 12 km in the tropics when biogenic VOC emissions are added. This is important because it demonstrates that changes in biogenic VOC emissions could influence the lifetimes of longer-lived compounds such as methane. The predicted decreased OH at high altitudes apparently results from an increase in NO_x (Plate 1) due to the transport of MPAN into this region. The predicted increased formation and transport of MPAN is a result of increased biogenic VOC concentrations in the boundary layer. The addition of biogenic VOC results in a CO increase of 15 to 20% at heights up to 15 km in the Southern Hemisphere. The impact is less in the more polluted Northern Hemisphere.

Plate 2 shows that IMAGES predicts that biogenic VOCs increase ozone concentrations by more than 10% in the atmosphere directly above most terrestrial surfaces and that five regions have near-surface ozone increases of 25 to 50%. Two are temperate regions, in the southeastern United States and eastern China, where anthropogenic NO_x emissions strongly influence ozone levels. The other three regions are the tropical regions of South America, Australia/Indonesia, and Africa. The increased ozone production associated with biogenic VOC in tropical regions is a result of IMAGES predictions of sufficient NO_x in these areas due to biomass burning. Field investigations are needed in these five regions to determine if the general patterns predicted by IMAGES are realistic and to improve our characterization of biogenic emissions for these locations. Many biogenic VOC emission studies have been conducted in the southeastern United States, but very little work has been done in the other four regions [*Guenther et al.*, 1995]. The need for observations in Africa, which had received the least attention of any of these five regions, was an important motivation for the EXPRESSO study.

There are significant changes in the distribution of chemical constituents, such as ozone, predicted for a factor of 2 change in isoprene emission, which is similar to the level of uncertainty associated with current emission estimates. The changes are greatest in the atmospheric boundary layer above terrestrial surfaces, where the model predicts that a factor of 2 increase in isoprene emission results in a 10 to 20% decrease in OH, a 15 to 35% increase in H_2O_2, and a 5 to 15% increase in O_3. The model also predicts that this factor of 2 increase in isoprene emission results in a 6 to 10% increase in CO throughout the Southern Hemisphere (up to 18 km above ground level), and a 5 to 15% decrease in OH at heights between 7 and 14 km above ground level in the tropics. We recognize that there are other major uncertainties associated with global CTM predictions of trace gas distributions, but this analysis suggests that biogenic VOC emission estimates are a significant contributor to the overall uncertainty.

3. Emission Model Description

Isoprene emissions were estimated using the Global Biosphere Emissions and Interactions System (GLOBEIS). GLOBEIS is a flexible modeling framework that estimates foliar emissions as

$$\text{emission} = [\varepsilon\,]\,[D_p D_f]\,[\gamma_P \gamma_T \gamma_A]\,[\,\rho\,], \qquad (1)$$

where ε is a landscape average emission capacity, D_p is the annual peak foliar density, D_f is the fraction of foliage present at a particular time of year, the emission activity factors γ_P, γ_T, and γ_A account for the influence of photosynthetic photon flux density (PPFD), temperature, and leaf age, respectively, and ρ is an escape efficiency that represents the fraction of the isoprene emitted by the canopy that is released into the above-canopy atmosphere. Emissions were predicted for the EXPRESSO study region (4°S to 12°N latitude and 8°E to 32°E longitude) in central Africa. This region has a land surface area of 4.54×10^6 km^2, which is 15% of the African continent and 3% of global land surface area. The location of the emission model domain on the African continent and the land cover distribution in this region are shown in Plate 3. GLOBEIS was used to estimate hourly isoprene emissions for 1996 (the year of the main EXPRESSO field campaigns) using the modeling procedures described in this paper (referred to as EXP96) and estimates based on the

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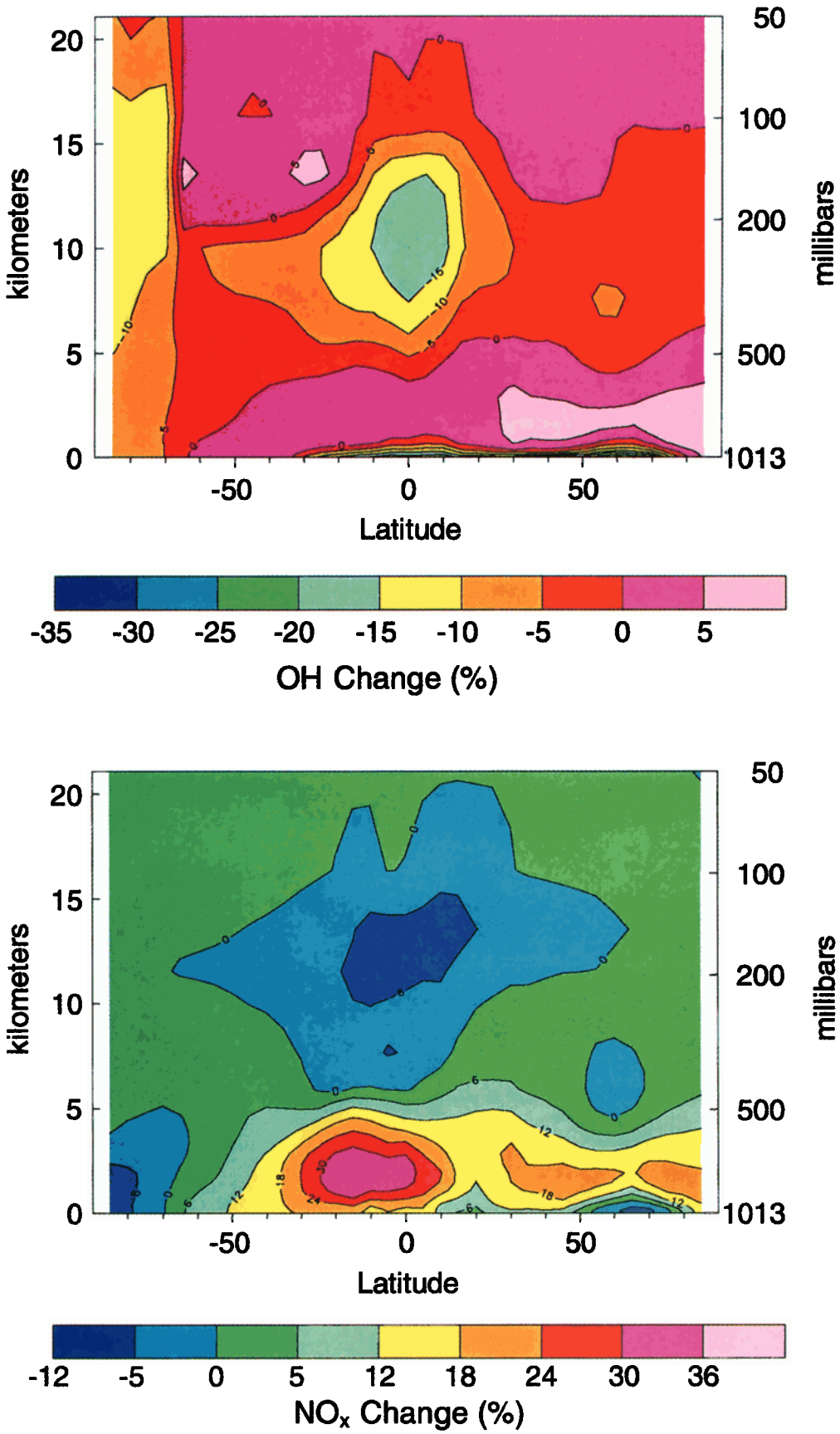

Plate 1. Percent difference in the July zonal average vertical distribution of OH (A) and NO_x (B) predicted by IMAGES for scenarios with isoprene and without isoprene.

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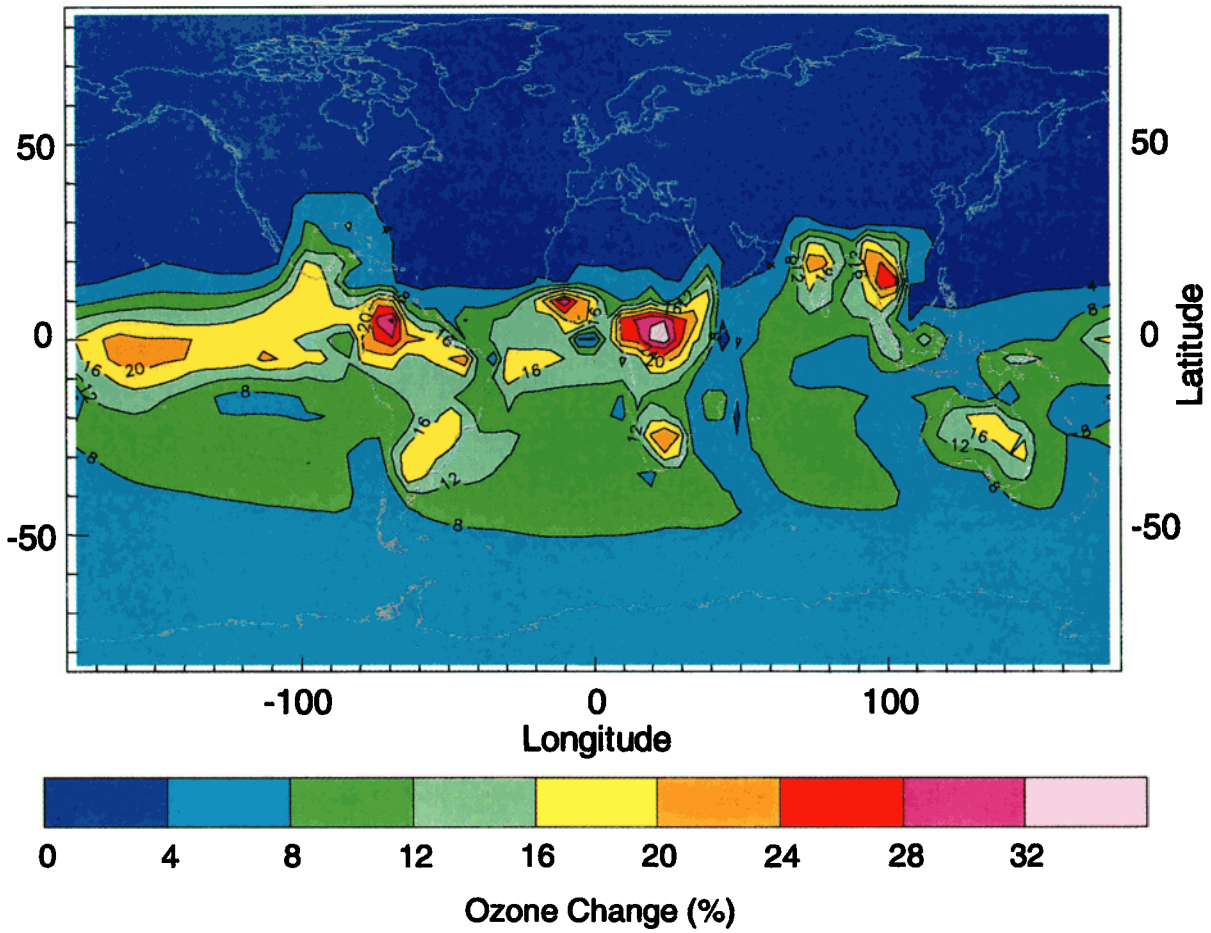

Plate 2. Percent difference in near-surface July ozone distribution predicted by IMAGES for scenarios with isoprene and without isoprene.

modeling procedures of *Guenther et al.* [1995] (referred to as G95). EXP96 used a spatial resolution of 0.01° latitude and longitude (about 1 km^2 with 3.84×10^6 grid cells).

The main components of GLOBEIS include (1) procedures for processing model inputs, (2) a landscape model that estimates canopy environment and vegetation characteristics, (3) the biogenic emission model described by equation (1), and (4) procedures used to summarize emission estimates and generate output in the format required for a specific CTM. GLOBEIS simulates landcover characteristics (ε and D) on an annual time step, seasonal variations (D_f, and γ_A) with a monthly time step, and diurnal variations (γ_P, γ_T, and ρ) with an hourly time step. The specific procedures used for the EXP96 estimates are described below. The G95 procedures are briefly summarized in section 1 and are described in more detail by *Guenther et al.* [1995].

3.1. Annual Variations

GLOBEIS requires a spatially distributed land cover characteristics database which includes emission capacity (μg of carbon per g dry weight foliar mass per hour (μg C g^{-1} h^{-1})) and peak foliar density (g dry mass of foliage per m^2 of land surface area). The EXP96 database was produced by assigning land cover characteristics to the IGBP-Discover Seasonal Land Cover Regions (SLCR) database (version 1.2). About two thirds of the 197 SLCR land cover types defined for the African continent occurred within the EXPRESSO domain. The database is derived from 1 km AVHRR data spanning April 1992 through March 1993 and is described, and available in digital format, on the Internet (http://edcwww.cr.usgs.gov/landdaac/glcc/af_int.html). We assumed that these 1992/1993 data are representative of our 1996 scenario.

The peak LAI and foliar densities assigned to each land cover type are based on the estimates of *Box* [1981], *Box et al.* [1989], *Guenther et al.* [1996], *Braswell et al.* [1996], and *Klinger et al.* [1998]. *Klinger et al.* [1998] estimate that total LAI in central Africa ranges from less than 2 in the grassland savannas of Chad to over 6 in the tropical rain forests of the Congo. Their estimates of the fraction of LAI associated with woody vegetation ranges from less than 20% in grassland savannas to over 95% in tropical rain forests. *Box et al.* [1989] report that semideciduous tropical forests have much less foliage (LAI = 4) than evergreen rain forests (LAI=9-12). *Klinger et al.* [1998] measured LAI ranging from 5.5 to 6.3 at several EXPRESSO tropical forest sites. It is not clear if the LAI estimation method (LAI-2000) used by *Klinger et al.* [1998] underestimates LAI in canopies with high LAI or if these forests have lower LAI than other rain forests.

Previous studies [e.g., *Guenther et al.*, 1994, 1996; *Klinger et al.*, 1998] defined an isoprene emission capacity (also called emission potential or basal emission rate) as the isoprene emission rate that occurs when a leaf is exposed to specified PPFD and leaf temperature conditions. As is discussed below, we now recognize the need to account for other factors, such as leaf age and the degree to which the leaf is temperature and

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Table 1. Percentage of isoprene-emitting foliage in African landscapes

Region	Landscape	Isoprene Emitters	Averaging Species Method	Diversity	Reference
South Africa	Mopane savanna	3%	weighted	low	*Guenther et al.* [1996]
South Africa	Combretum savanna	9%	weighted	low	*Guenther et al.* [1996]
South Africa	*Acacia nigrescens* savanna	64%	weighted	low	*Guenther et al* [1996]
South Africa	*Acacia tortilis* savanna	9%	weighted	low	*Guenther et al.* [1996]
South Africa	Burkea savanna	69%	weighted	low	*Guenther et al.* [1996]
Central Africa	mixed upland rainforest	10%	weighted	high	*S99*
Central Africa	Gilbertiodendron rainforest	85%	weighted	low	*S99*
Central Africa	primary rain forest	12%	equal	high	*Klinger et al.* [1998]
Central Africa	secondary rain forest	22%	equal	high	*Klinger et al* [1998]
Central Africa	transition woods	20%	equal	high	*Klinger et al.* [1998]
Central Africa	Isoberlinia savanna	38%	equal	low	*Klinger et al.* [1998]
Central Africa	other savanna	20%	equal	low	*Klinger et al.* [1998]

shade-adapted. This has been accomplished by introducing an additional emission activity factor, γ_A, which accounts for leaf age, and by modifying the algorithms used to account for environmental conditions by including past and current conditions. The result is that the emission capacity reported by previous studies is not directly comparable with the emission capacity used for this study.

Enclosure measurement studies in Africa [*Guenther et al.*, 1996; *Klinger et al.*, 1998; *S99*] report that plants emit isoprene at rates ranging from less than 0.1 to greater than 100 μg C g^{-1} h^{-1}. It is difficult to estimate emission capacities from these measurements since the influence of leaf age and past environmental conditions were not characterized during these studies. The emission activity factor that accounts for leaf age, γ_A, can be less than 0.1, early or late in the life of a leaf. The influence of PPFD is accounted for in most enclosure measurement studies by using the measured PPFD during the time of measurement and the PPFD-dependent algorithm of *Guenther et al.* [1993]. However, the influence of growth environment (i.e., Sun or shade-adapted leaves) is not easily characterized and has not been considered in most emission survey studies. *Harley et al.* [1997] report that the emission capacity of shade-adapted leaves growing 3-5 m below the top of a temperate forest canopy is about half that of Sun-adapted leaves growing at the top of the canopy. Most enclosure measurement emission surveys have primarily used leaves growing near the ground, which, particularly in dense forest, tend to be shade adapted, in which case they would be expected to have a low γ_P. Recent evidence suggests that the emission activity of shade leaves may increase slowly over time when maintained in moderate to high light. For example, *S99* collected branches accessible from the ground from two tree species in central Africa, maintained them in high light for between 1 and 5 hours, measuring the emission rate at 30°C and 1000 μmol m^{-2} s^{-1} repeatedly. When first measured, emissions were almost undetectable, but after a lag time (15 min in one species and 2 hours in the second), emissions began to increase exponentially, reaching rates of more than 60 μg C g^{-1} h^{-1} after two additional hours and still increasing. Thus within a few minutes (or in some cases hours) of illumination, shade-adapted leaves may have a very low value (< 0.1) of γ_P; in the extreme, isoprene emissions may be undetectable, causing an isoprene-emitting species to be characterized as a nonemitter. *Lerdau and Throop* [1999] report similar observations in a tropical forest in Panama where no isoprene emission was observed for three tree species growing in extreme shade even though these same species were high emitters when growing in full Sun.

Klinger et al. [1998] placed plant species into two categories: "high" emitters with an emission rate of 0.37 μg C cm^{-2} h^{-1} and "low" emitters with an emission rate of 0.03 μg C cm^{-2} h^{-1}. If we assume that the "high" emitters were a mixture of Sun and shade leaves (γ_P = 0.5, specific leaf mass = 80 g m^{-2}) and the "low" emitters tended to be shade leaves from the bottom of the canopy (γ_P = 0.05, specific leaf mass = 50 g m^{-2}) then the emission capacity of both categories is about 100 μg C g^{-1} h^{-1}. This is in agreement with the mean isoprene emission capacity (99 μg C g^{-1} h^{-1}) observed by *Guenther et al.* [1996] for isoprene-emitting plant species in southern African savannas, when their reported branch-level emission capacities are converted to leaf-level conditions using the factor recommended by *Guenther et al.* [1994]. Although we expect emission capacities to differ among plant species, the existing measurement database is not suitable for characterizing this variation, and so we use a leaf level emission capacity (ε_0) of 100 μg C g^{-1} h^{-1} for all isoprene-emitting plants.

About 30% of the approximately 250 African plant species investigated by *Guenther et al.* [1996], *Klinger et al.* [1998], and S99 emit isoprene. This percentage is similar to that observed for North American vegetation [*Guenther et al.*, 1994]. The preferred method for estimating isoprene emissions is to combine a plant species composition database with emission data characterizing each species. While this can be accomplished for the United States [*Geron et al.*, 1994] and specific locations in Africa [*Guenther et al.*, 1996], these data are not available for most of central Africa. Instead, we have defined vegetation types with different percentages of foliage that emit isoprene. The percentage of isoprene emitters in the various African landscapes investigated by *Guenther et al.*, [1996], *Klinger et al.*, [1998], and S99 are shown in Table 1. If all species have an equal probability of being an isoprene emitter, then we would expect that areas with a high species diversity would have values more similar to the overall average percentage of isoprene emitters (about 30% for woody plants) than areas with low species diversity. For example, if a single plant species comprises 80% of the foliage in a landscape, then the percentage of isoprene emitters in that landscape will be less than 20% if that species is not an isoprene emitter and greater than 80% if that species is an isoprene emitter. The emission surveys shown in Table 1 indicate that landscapes with high species diversity have a factor of 2 range (10% to 22%) in the abundance of isoprene emitters,

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Table 2. Model variables Assigned to Prevalent Landcover Types in the EXPRESSO Domain

SLCR	Area	Description	ϕ_w	$\phi_{w,i}$	$\phi_{w,h}$
184	6.4%	degraded lowland forest/savanna	0.4	0.2	0.005
154	5.8%	tropical forest (semideciduous)	0.95	0.12	0.005
174	5.3%	high density Sudanian woodland	0 9	0 5	0.005
141	5.0%	tropical rain forest	1.0	0.22	0.005
186	5 0%	degraded trop. forest, Sudanian wood	0.6	0 36	0.005
197	4.8%	water	0.0	0.0	0.0
150	4.3%	dense tropical rain forest	1.0	0 22	0.005
145	4.1%	tropical forest (semi-deciduous)	0.95	0 12	0.005
164	3 7%	tree savanna	0.7	0 2	0.005
102	3.4%	Sudanian woodland with crops	0.45	0.5	0.005

SLCR is the land cover code assigned by IGBP-DIS Area indicates the portion of the EXPRESSO domain covered by the land cover type; ϕ_w is the fraction of total foliage contributed by woody plants; $\phi_{w,i}$ and $\phi_{w,h}$ are the fraction of woody and herbaceous plant foliage, respectively, which are isoprene emitters

while landscapes with low species diversity range from 3% to 85% isoprene emitters.

The model variables used to calculate the landscape average emission capacity, ε in equation (1), in the 10 most prevalent SLCR land cover types within the EXPRESSO domain are shown in Table 2. The landscape average emission capacity is calculated as

$$\varepsilon = \varepsilon_0 (\phi_w \phi_{w,i} + \phi_h \phi_{h,i}) \quad (2)$$

where ε_0 (=100 $\mu g\ g^{-1}\ h^{-1}$) is the emission capacity of isoprene-emitting plants, ϕ_w and ϕ_h are the fraction of total foliage contributed by woody and herbaceous plants, respectively, and $\phi_{w,i}$ and $\phi_{h,i}$ are the fraction of woody and herbaceous foliage, respectively, which are isoprene emitters. We consider plants to be either woody or herbaceous, that is, $\phi_h = 1 - \phi_w$. Although there are relatively few measurements of isoprene emission from herbaceous vegetation, these data suggest that very few of these plants are isoprene emitters in comparison to woody plants. We use a single value ($\phi_{h,i} = 0.005$) in all landscapes to represent the fraction of herbaceous plants that emit isoprene. This value is very uncertain and could be an order of magnitude greater or less in at least some regions. The estimated fraction of foliage associated with woody vegetation in each SLCR land cover type is based on the vegetation descriptions given by IGBP-DIS (e.g., degraded lowland forest/savanna, Sudanian woodland with crops, dense tropical rain forest) and the values given in Table 1. For example, SLCR categories 154, 184, and 164 are based on the *Klinger et al.* [1998] data for primary rain forest, transition woods, and other savannas. Some land cover types were assumed to be a mosaic of several of the landscapes included in Table 1.

3.2. Seasonal Variations

Monthly variations in foliage fraction (D_f) and the emission activity associated with leaf age (γ_A) were estimated for each location in the model domain. Foliage fraction is the ratio of the foliage present at a specific time of year to the peak foliage during the year. It was estimated using the average monthly LAI data for the two years (1987 and 1988) in the ISCLCP LAI database. This database has a resolution of 1° latitude × 1° longitude and is described by *Sellers et al.* [1994]. The ISCLCP LAI estimates were derived from satellite measurements of an index (simple ratio) that is related to LAI due to the very different optical properties of green leaves in the red (high absorption) and infrared (high reflection) bands. *Sellers et al.* [1994] used the simple ratio to interpolate between minimum and maximum LAI values assigned to different global vegetation types.

Guenther [1997] notes that young and old leaves emit isoprene at substantially lower rates and suggests that this could be simulated using estimates of seasonal variations in foliage (D_f) as well as temperature (or precipitation for drought deciduous vegetation). Increasing D_f indicates a higher proportion of young leaves and decreasing D_f indicates old leaves. We use a simple model that describes the landscape average leaf age emission activity as

$$\gamma_A = [A_1 \Delta D_f] + [A_2 (1-\Delta D_f)], \quad (3)$$

where A_1 is the average emission activity of young and old leaves, A_2 is the fraction of mature foliage present during the month of peak foliar density, and ΔD_f is the absolute difference between the foliar fraction of the current month and the previous month divided by the maximum foliar fraction of the two months. ΔD_f represents the foliage change (increase or decrease) during the month. We assume that the changing foliage still has some mature leaves and so assign A_1 a value of 0.33. We expect A_2 to be slightly less than 1 in most landscapes, because of the continuous presence of at least a few young or old leaves, and assign a value of 0.95. These coefficients are expected to vary among landscapes, but there are no data available for developing these relationships.

3.3. Diurnal Variations

Diurnal variations in escape efficiency ρ and emission activity factors γ_T and γ_P are estimated for each location in the model domain. The escape efficiency is the ratio of the canopy emission rate to the above canopy flux We use the model of *Jacob and Bakwin* [1991] which predicts ρ as a function of the deposition and canopy ventilation rates. We assume that the canopy ventilation rate is proportional to the mean wind speed within the canopy. Our estimate of deposition rate is based on observations of microbial consumption of isoprene in soils reported by *Cleveland and Yavitt* [1997].

Guenther *et al.* [1993] account for the influence of PPFD using an algorithm that predicts that isoprene emission increases with increasing PPFD up to a saturation point, where emissions level off

$$\gamma_P = (\alpha C_L Q)/[(1+\alpha^2 Q^2)^{0.5}], \quad (4a)$$

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Table 3. Comparison of Emission Model Predictions for Central Africa

	G95	EXP96	Difference
Land area (10^6 km^2)	4 52	4 54	+0 4%
Escape efficiency (ρ)	1	0 95	-5%
Foliar density			
peak (D_P, g m^{-2})	1000	594	-41%
seasonal fraction (D_f)	0 59	0.67	+14%
annual average (D_P x D_f)	590	437	-26%
Emission capacity and activity			
emission capacity (ε, μg g^{-1} h^{-1})	16.5	19 8	+20%
temperature (γ_T)	0.513	0 571	+11%
PPFD (γ_P)	0.220	0.251	+12%
age (γ_A) 1	0.84	-16%	
combined (ε x γ_T x γ_P x γ_A)	3.70	4.28	+16%
Annual isoprene emission (Tg C)	41 1	35.4	-14%

where Q is current PPFD (μmol m^{-2} s^{-1}), and α and C_L are empirical coefficients. Guenther *et al.* [1993] assumed constant values, α = 0.0027 and C_L =1.066, for these coefficients, but recent studies have shown that these coefficients vary with canopy depth, presumably because of differences in past PPFD levels [*Harley et al.*, 1996, 1997]. On the basis of these data, we assume that

$$\alpha = 0.001 + 0.00085\ \mathrm{LAI}, \tag{4b}$$

$$C_L = 1.42\ \exp(-0.3\ \mathrm{LAI}), \tag{4c}$$

where LAI is the cumulative leaf area index above the leaf. This algorithm predicts that a leaf near the top of the canopy (LAI depth of less than 0.5) will have a γ_P of about 1 when exposed to a PPFD of 1000 μmol m^{-2} s^{-1}, while a leaf lower in the canopy (LAI depth of greater than 5) will have a lower value ($\gamma_P < 0.31$) when exposed to the same PPFD.

The influence of leaf temperature is simulated as

$$\gamma_T = E_{opt}\, C_{T2} \exp(C_{T1}\, x)/[C_{T2}-C_{T1}\{1-\exp(C_{T2}\, x)\}] \tag{5a}$$

where $x = [(1/T_{opt})-(1/T)]/R$, T is current leaf temperature (K), R is the gas constant (=0.00831), E_{opt} is the maximum normalized emission capacity, T_{opt} is the temperature at which E_{opt} occurs, and the empirical coefficients C_{T1} (=95) and C_{T2} (=230) represent the energy of activation and deactivation, respectively. Equation 5 is nearly equivalent to the algorithm of *Guenther et al.* [1993] for E_{opt} = 1.9 and T_{opt} = 312.5 K and we use these values for the EXP96 estimates. There is some evidence that the coefficients in equation (5a) are dependent on the temperature that the leaf has been exposed to during the past several days and weeks [*Sharkey* 1999], G. Petron et al., Seasonal temperature variations influence isoprene emission, submitted to Geophysical Research Letters. As a sensitivity study, we consider the potential influence of past temperature on these coefficients by estimating

$$E_{opt} = 1.9 \times \exp(0.125\ (T_d - 301)) \tag{5b}$$

and

$$T_{opt} = 312.5 + 0.5\ (T_d - 301) \tag{5c}$$

where T_d is the mean temperature (K) of the past 15 days.

A canopy environment model, similar to that of G95, is used by EXP96 to estimate leaf temperature and PPFD distributions within a canopy. The major differences are (1) improved methods of calculating light scattering and the penetration of diffuse light within the canopy, (2) specific leaf mass (mass per unit leaf area) that decreases with canopy depth, and (3) the addition of a leaf energy balance model that accounts for the influence of wind speed, humidity, and solar radiation on leaf temperature. The methods used to calculate leaf temperature and light scattering and penetration of diffuse light within the canopy are based on the model of *Goudriaan and van Laar* [1994]. The specific leaf weight algorithm is similar to that described by *Geron et al.* [1994].

The hourly average above-canopy environmental conditions (temperature, direct and diffuse PPFD, wind speed, and humidity) used as inputs to EXP96 were estimated from the (NCEP/NCAR) reanalysis (described on the internet at www.scd.ucar.edu/dss/pub/reanalysis) of the NCEP global model data. Hourly variations were estimated from the 4 times daily NCEP data using the procedures described by *Goudriaan and van Laar* [1994].

4. Emission Model Results and Comparisons

4.1. Regional Totals

The G95 model predicts that about 41 Tg C of isoprene is emitted annually from the EXPRESSO domain in central Africa. This is about a third of the total isoprene flux that G95 estimates for all of Africa, although this region comprises only about 15% of the total land surface area of the African continent. The EXP96 and G95 model results are compared in Table 3. The coarser spatial resolution of G95 results in a slight (0.4%) overestimate of the land surface area within the EXPRESSO domain. G95 has higher estimates of three of the model variables in equation (1) (D_p, γ_A, and ρ) and lower estimates of four variables (D_f, ε, γ_P, and γ_T). The net result of these differences is that the annual isoprene flux estimated by EXP96 is only 14% lower than the estimate of G95.

The average D_p predicted by EXP96 is 41% lower than that estimated by G95. A major reason for this difference is the lower peak foliar density assigned to savanna regions by EXP96. The G95 estimates of D_p (= 800 g m^{-2} for savannas) are solely based on the recommendation of *Box* [1981]. *Guenther et al.* [1996] investigated emissions from savannas in southern Africa and report that these savannas have a much lower peak foliar density. While this adjustment should improve the accuracy of emission estimates in some savanna regions, there are still large uncertainties in estimates of D_p due to limited measurements.

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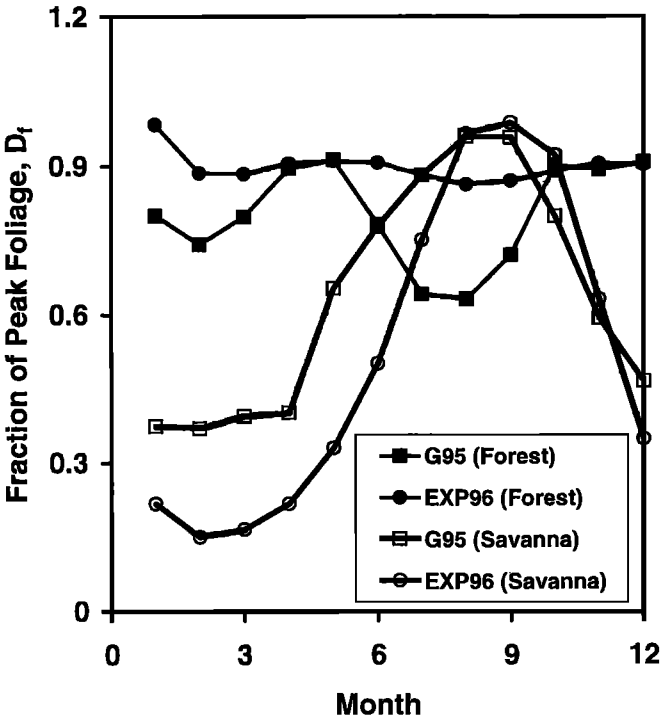

Figure 1. Monthly variations in the fraction of foliage present, D_f, within savanna and forest regions.

The annual average D_f (the fraction of the peak foliar density present at a particular time of year) estimated by EXP96 is 14% higher than the average value estimated by EXP96. As discussed in the following section, this is primarily due to the low D_f values predicted by G95 for rain forest landscapes. The net result of the differences in D_p and D_f predicted by the two models is that the annual average foliar density estimated for the entire EXPRESSO domain by EXP96 is 26% less than that estimated by G95.

The PPFD- and temperature-dependent emission activity factors γ_P and γ_T, estimated by EXP96, are higher than the values estimated by G95. There are two factors that dominate the difference in predicted γ_T. The leaf energy balance model used by EXP96 predicts higher leaf temperatures than G95 in the top of the canopy where most isoprene emission occurs. Secondly, the diurnal temperature variation used for EXP96 results in higher emissions than that of G95, which used only the monthly average temperature. Because daytime temperatures are considerably higher than daily average temperatures, and due to the non-linear relationship between emissions and temperature, the lack of diurnal temperature variation results in significantly underestimated emissions.

Keller and Lerdau [1999] characterized the temperature and light response of isoprene emission from leaves near the top of a tropical forest canopy in Panama. Their light response is similar to that predicted by equation (4) for an LAI < 1. They observed a relationship between temperature and isoprene emission that could be described by the algorithm of *Guenther et al.* [1993] by using different coefficients. They note that the original coefficients significantly underestimate isoprene emissions at high temperatures. However, our comparison of isoprene emissions estimated for central Africa using the two sets of coefficients indicates that the difference is less than 5%. This is because the coefficients suggested by *Keller and Lerdau* [1999] predict higher emissions at temperatures above 30°C but lower emissions at temperatures below 30°C. The use of coefficients that are dependent on the temperature of the past 15 days (equations 5b and 5c) results in a similarly small difference in estimated annual isoprene emission from this region. The annual average air temperature for the EXPRESSO domain is about 26°C and average daytime leaf temperatures are around 30°C which minimizes the influence of the temperature response algorithms which account for deviations from 30°C.

There are four factors that contribute to the difference in γ_P estimated by EXP96: LAI, above canopy PPFD, canopy light environment model, and the numerical algorithm describing the relationship between PPFD and emission. The decreased foliage predicted by EXP96 results in a higher canopy average PPFD (i.e., lower LAI increases the fraction of the canopy that is in direct sunlight) and therefore an increased canopy average γ_P. The above canopy PPFD, and fraction that is diffuse PPFD, used for EXP96 are 1996 estimates from the NCEP global model while those used for G95 were estimated with a simple model as a function of solar angle and a 30 year climatological cloud cover database. The G95 estimates predict considerably lower PPFD, particularly with clear skies.

As discussed above, the isoprene emission capacities used by the two models are not directly comparable. However, one notable difference in the methods used by the two models is that EXP96 estimates a greater area of cropland vegetation, which has a lower isoprene emission capacity. EXP96 introduces two additional factors (ρ and γ_A) which were not used by *Guenther et al.* [1995]. For the purpose of comparison, we indicate that both factors are set equal to 1 for G95. The escape efficiency factor is introduced to account for the difference in the isoprene flux from the canopy and what is released into the atmosphere above the canopy. This factor has a relatively small effect, a 5% reduction in emissions. EXP96 also introduces an emission activity factor that accounts for differences due to changes in leaf age and phenology. The addition of this factor results in a 16% decrease in emissions.

4.2. Annual and Diurnal Variations

Annual variations in foliar density (D_f) predicted by EXP96 and G95 for savanna (site E in Plate 3) and rain forest (site A in Plate 3) regions are compared in Figure 1. EXP96 predicts that D_f at this savanna location ranges from about 0.15 in February to almost 1 in September. The savanna D_f estimated by G95 for the same location follows the same general pattern and is of the same magnitude during June to December but is about a factor of 2 greater during January to May. The annual average rain forest D_f predicted by EXP96 is nearly constant (standard deviation of 3%) with a mean of 0.9. The G95 rain forest D_f estimate has a lower annual mean (0.8) and higher standard deviation (13%). As a result, the estimates for some months are as much as 25% lower. The D_f estimates for both models are based on measurements by the NOAA advanced very high resolution radiometer (AVHRR) satellite. The database used for EXP96 [*Sellers et al.*, 1994] uses a technique, discussed above, that should result in better estimates than those used for G95. Another difference is that the D_f estimates for EXP96 are based on an average of data for 1987 and 1988, while G95 uses 1990 AVHRR measurements.

The diurnal and annual variations predicted by EXP96 can be evaluated by comparison to the above-canopy isoprene fluxes measured with the tower-mounted relaxed eddy accumulation (REA) system described by S99. The observed measurements are

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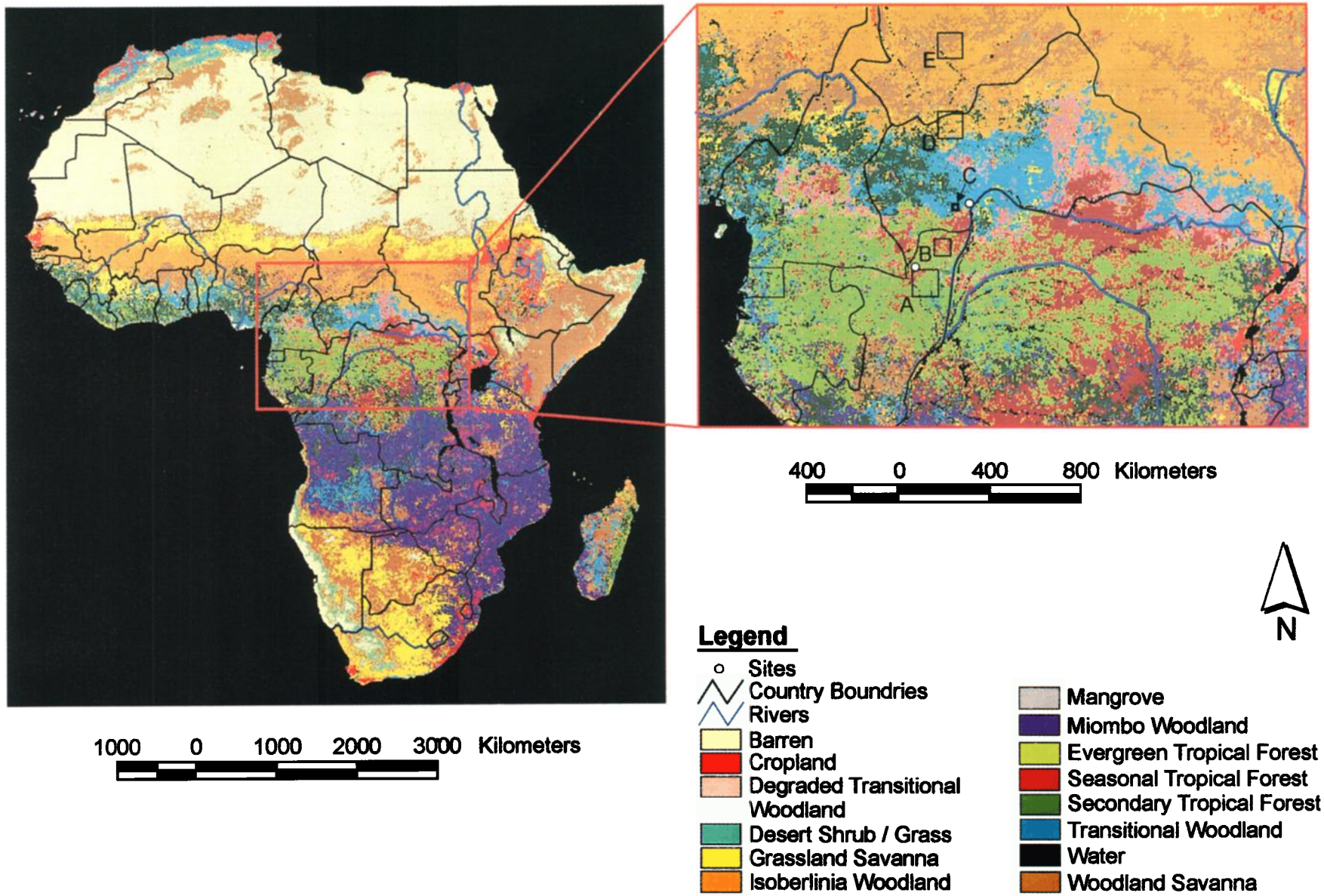

Plate 3. Landcover distribution within the EXPRESSO model domain. The locations labled A – E correspond to the sites described in Table 4.

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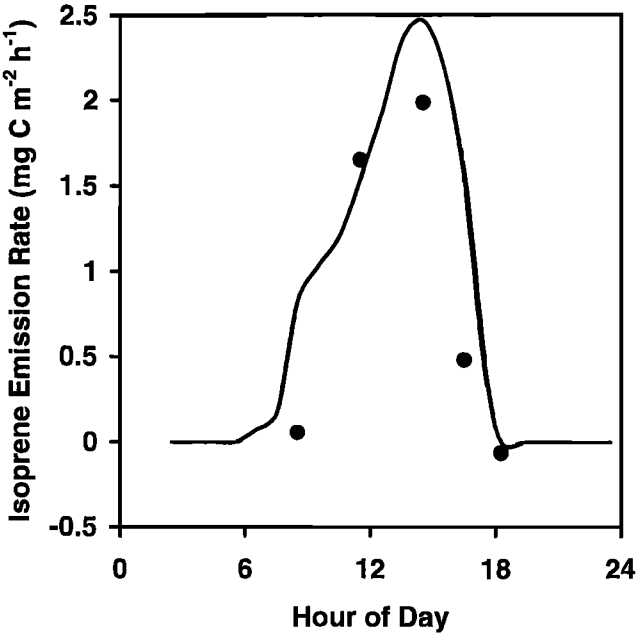

Figure 2. Diurnal variation in emission (mg C m^{-2} h^{-1}) predicted (solid line) and observed (filled circle) at the EXPRESSO tropical forest field site on March 18 1996.

representative of the mixed upland rain forest landscape within a few hundred meters of the tower, which is located in a minimally disturbed region in the Nouabale-Ndoki National Park, Republic of Congo (2°12'39"N, 16°23'51"E). For this comparison, fluxes were predicted using the results of the emission survey at this site, shown in Table 1, and the NCEP meteorological estimates which represent a much larger area (about 5×10^5 km^2). The observed fluxes shown in Figure 2 were estimated from nine 30 min average fluxes that were measured on March 18, 1996, and averaged to estimate hourly average fluxes. Both observed and predicted fluxes follow a pattern of negligible fluxes at night, increasing rapidly in the morning to a maximum after noon, and then decreasing rapidly in the evening. Model predictions of diurnal variations of above-canopy fluxes have been validated for temperate forest canopies using larger data sets [e.g., *Guenther and Hills*, 1998]. The above-canopy flux measurements of S99 do not suggest any major differences for tropical forest canopies, but the limited observations do not exclude the possibility of significant differences. The model predicts that about half of the daily total isoprene emission occurs between the hours of 1100 and 1500 and that the flux during each of these hours is about 13% of the daily total. The predicted ratios between hourly and daily total emission were used to estimate daily total emissions from midday (1100 to 1500) 30-min observations. The isoprene fluxes measured by S99 include 18 midday measurements during March and 9 midday measurements during November and December 1996. Figure 3 shows that the mean daily total isoprene flux estimated from the March field experiments is about a factor of 2 higher than the estimate for the November/December observations. Figure 3 also shows the predicted day-to-day changes in emissions due to changes in temperature and PPFD. The predicted variations in daily total emission rates are about a factor of 2 and can occur over a timescale of about a week. The above-canopy PPFD and temperature conditions estimated by the NCEP model for the March and the November/December field study periods are similar. This suggests that the observed differences in isoprene emissions are due to factors other than above-canopy PPFD and temperature. One possible reason for the difference is a lower stomatal conductance in March (the end of the dry season) than in November (the end of the wet season), causing a higher leaf temperature, which would result in a higher isoprene emission. S99 observed much lower transpiration rates in March than in November. This indicates a lower stomatal conductance, probably because of decreased leaf water potential, although this was not measured during the experiment. It is also interesting to note that CO_2 fluxes were considerably less in March, than in November, which is expected if stomatal conductance were lower and leaf temperature higher. This is because, for conditions typical of the tropical forest canopy, increasing leaf temperature results in increasing isoprene emission but decreasing photosynthesis. These results suggest that our leaf energy balance model should be modified to account for changes in leaf water potential.

4.3. Spatial Distributions

The predicted latitudinal gradients of LAI shown in Figure 4 are averaged over a longitude of 17°E to 20°E, which is near the center of the EXPRESSO domain. The LAI estimated by EXP96 is in good agreement with the ISCLCP database [*Sellers et al.*, 1994] in the region of degraded woods and cropland, between 4° and 8°N latitude, but is much higher in the tropical forest and northern savanna regions. The ISCLCP estimate for the tropical forest is probably too low, but it is not clear which is the best estimate for the northern savannas. The estimate used for EXP96 is based on the IGBP-DIS land cover database, which classifies a significant fraction of this region as "high-density Sudanian

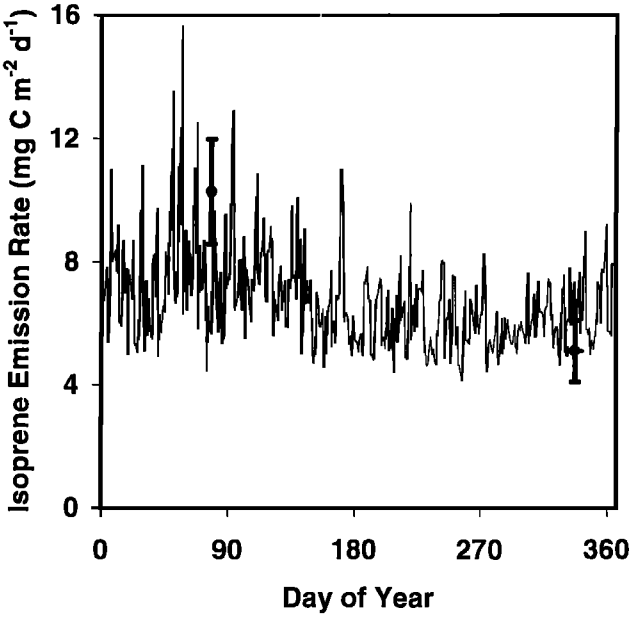

Figure 3. Annual variation in the daily total emission (mg C m^{-2} d^{-1}) predicted (solid line) and observed (mean=filled circle, ± standard deviation=vertical bar) at the EXPRESSO tropical forest field site (Ndoki, Congo) in 1996.

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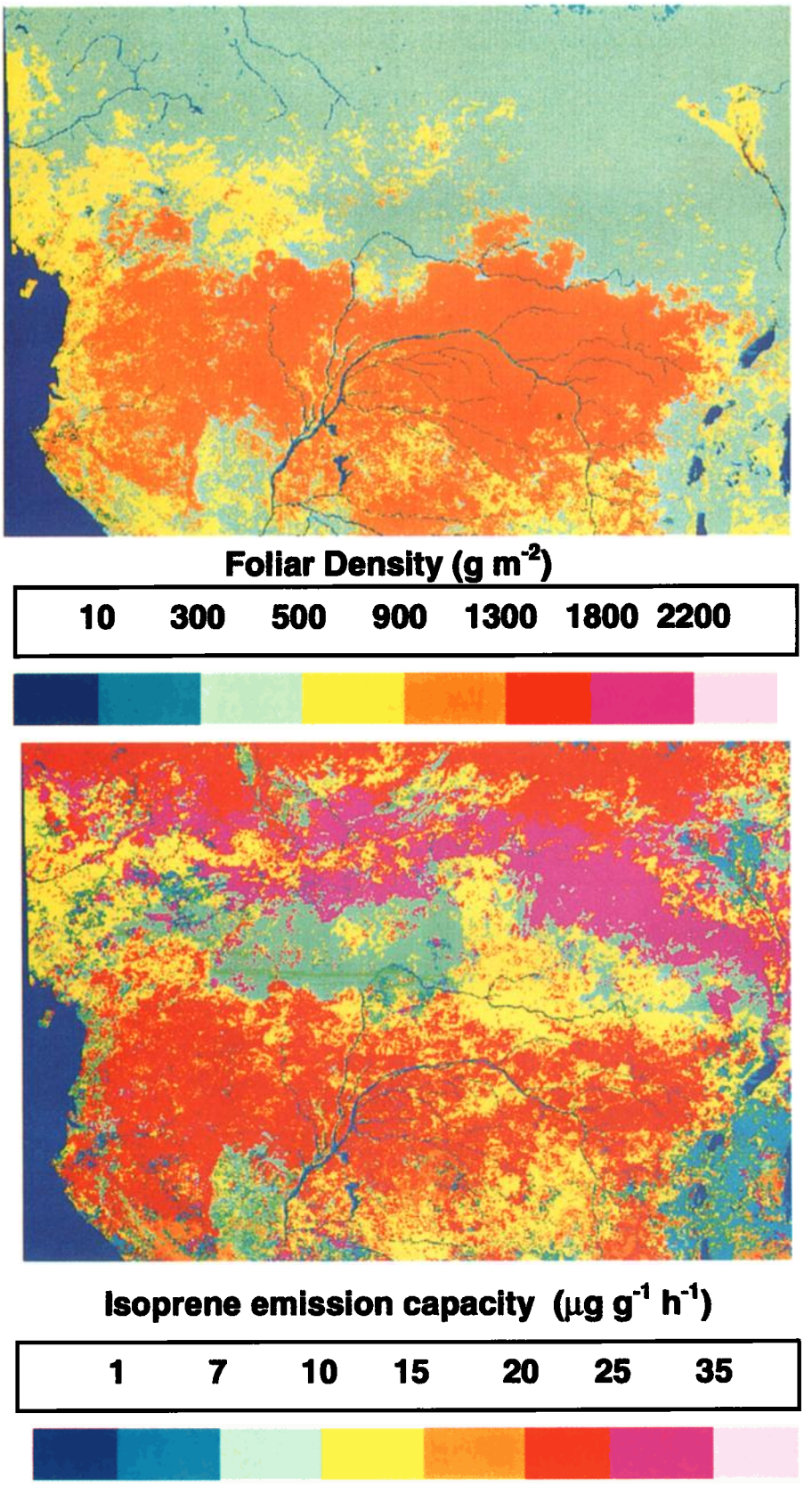

Plate 4. Spatial distribution of foliar density (top, g m^{-2}) and emission capacity (bottom, µg C g^{-1} h^{-1}) predicted by EXP96.

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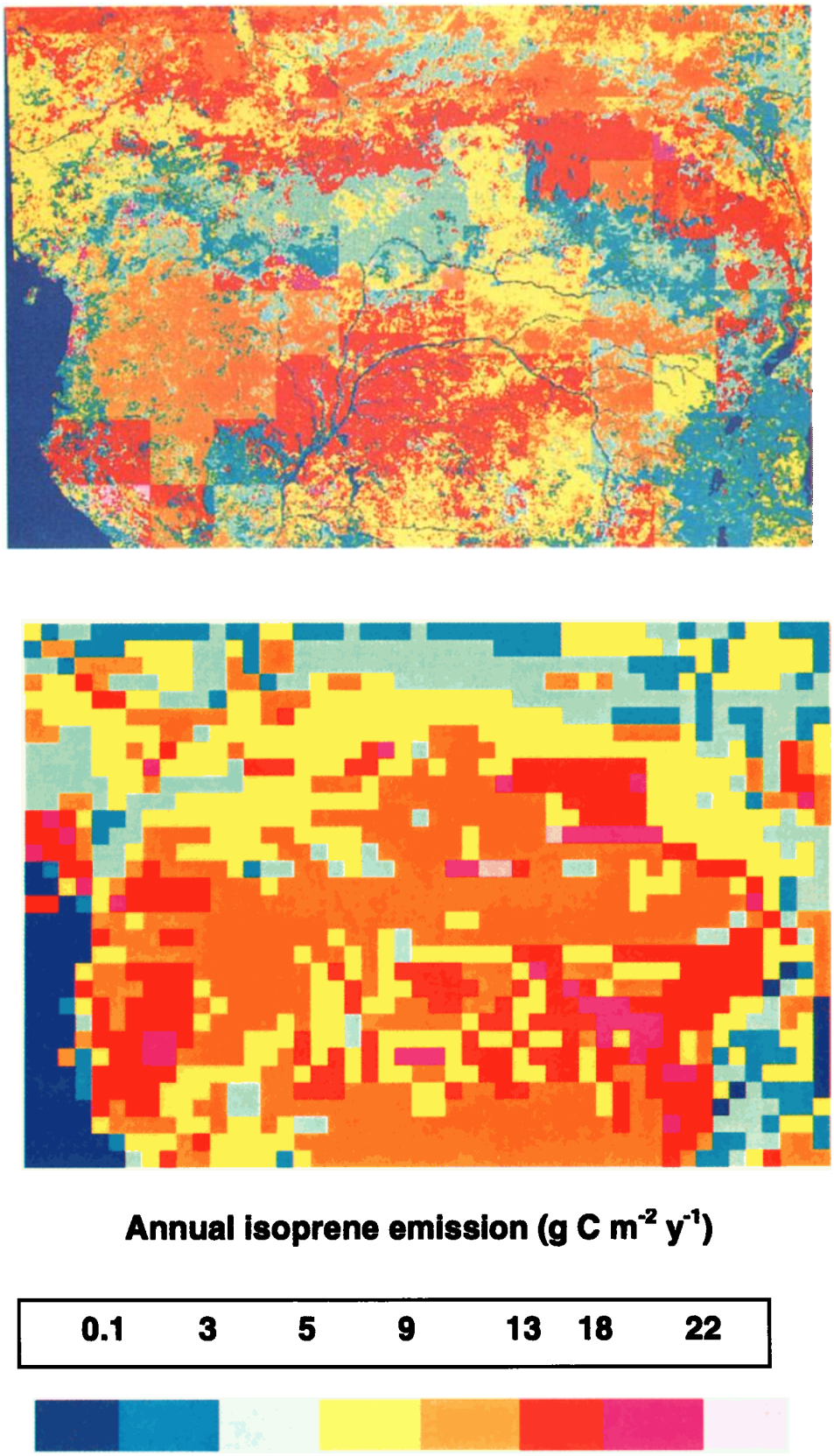

Plate 5. Spatial distribution of annual isoprene emission (g C m^{-2} y^{-1}) predicted by EXP96 (top) and by G95 (bottom) for the EXPRESSO model domain.

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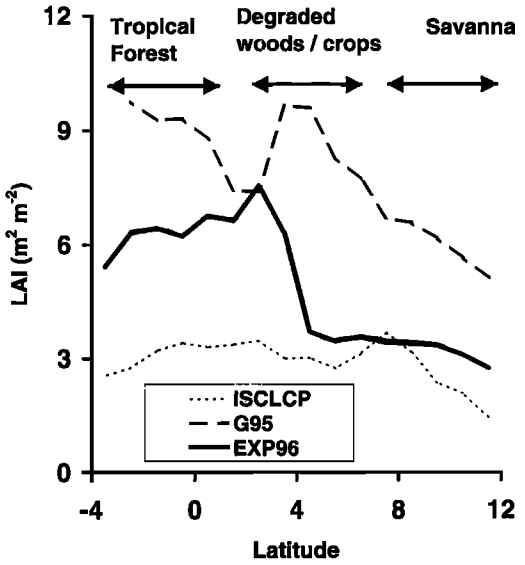

Figure 4. Latitudinal variation of LAI within a longitude zone of 17 to 20 E.

woodland." The G95 estimates of LAI appear to be overestimates for most of the EXPRESSO domain.

Five sites, located in Plate 3 along a latitudinal transect, were chosen to represent the major landscape types within the EXPRESSO domain. Emission rates estimated for these sites by the G95 and EXP96 models and by an aircraft relaxed eddy accumulation flux system [*Greenberg et al.*, this issue] are compared in Table 4. The emission rates are averaged over the aircraft sampling periods, which were primarily in late mornings of November and December 1996. The emission rates predicted by EXP96 range from 70% higher to 50% lower than the estimates of G95 and the mean value is about 20% lower. The EXPRESSO aircraft measurements were made at two of these sites (B and C in Plate 3). The isoprene emission rates predicted by EXP96 are about 30% higher than the aircraft flux measurements for the degraded forest/woodland site and about 120% higher for the tropical forest site. The aircraft flux measurements may underestimate surface fluxes due to flux divergence [*Greenberg et al.*, this issue] which, if corrected, would result in better agreement with the model. As shown in Figure 3, the model overpredicts the isoprene flux at the end of the wet season, which is when the aircraft flights occurred, but not at the end of the dry season. If this behavior is representative of the entire region, then EXP96 may underestimate during some seasons (e.g., in March) and overestimate during other seasons (e.g.. November) but provide a reasonable estimate of the annual average emission.

The spatial distribution of foliar density, emission capacity, and annual emission rates predicted by EXP96 are shown in Plates 4 and 5 G95 predicts higher estimates of foliar density over a large portion of the model domain and the emission capacities predicted by the two models differ considerably. As a result, the spatial patterns in annual total emission shown in Plate 5 are considerably different, although both models predict a similar range of emission rates, from 1 to 20-25 g C m^{-2} yr^{-1}, which occur throughout the domain. The highest emission rates (25 g C m^{-2} yr^{-1}) predicted by G95 are somewhat greater than those (20 g C m^{-2} yr^{-1}) predicted by EXP96.

Table 4. Comparison of Model and Aircraft REA Estimates of Isoprene Emission Rates

Site	Location	SLCR	Description	Isoprene Emission (mg C m^{-2} h^{-1})		
				G95	EXP96	Aircraft
A	1.5°N, 17.0° E	150,149	dense tropical forest	1.9	3.3	no data
B	3.1°N, 17.8°E	154	tropical forest (semideciduous)	2.9	2.2	1.0
C	4.7°N, 18.2° E	184	degraded forest/woodland	1.6	0.8	0.6
D	7.5°N, 18°E	174	Sudanian woodland	3.4	3.1	no data
E	10.5°N, 18°E	116,98	woodland/savanna, grass/shrub	4.9	2.4	no data

Site corresponds to the labels given in Plate 3. SLCR and description indicate the most common land cover types at the sites. Isoprene emission is the average rate estimated for the hours of the aircraft sampling periods (primarily late morning during November/December) by the models G95 [*Guenther et al.* 1995] and EXP96 (this work) and the aircraft REA flux estimates of *Greenberg et al.* [this issue].

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5. Discussion and Conclusions

The task of estimating isoprene emission rates with the accuracy desired for a global CTM is challenging. A sensitivity study demonstrated that a factor of 2 decrease in biogenic VOC emissions results in 5 to 35% changes in concentrations of OH, H_2O_2, and O_3 near terrestrial surfaces and 5 to 15% changes in OH and CO at heights between 7 and 14 km in the tropics. There are a number of major uncertainties associated with global CTMs, but the uncertainty in isoprene emission estimates (greater than a factor of 2) significantly contributes to the overall uncertainty associated with CTM predictions of the distributions of the trace gases that control the oxidation capacity of the atmosphere.

The isoprene emission modeling procedures described and evaluated in this paper are presented as a framework that can be improved and extended with additional measurements. These procedures are not specific for central Africa but are designed for global emission modeling procedures that are reasonable if model inputs for a particular region are based on reliable measurements. The results are current best estimates that can be improved as additional data are provided to improve the model parameterizations.

Characterizing the degree to which leaves are shade-adapted is important for predicting isoprene emissions. It is particularly important that we characterize these factors when making the leaf enclosure measurements used to estimate emission capacities. The enclosure measurements of Klinger et al. tend to represent shade-adapted leaves that were briefly exposed to sunlight. The large difference between the isoprene emission capacities reported here, and those of *Klinger et al.* [1998] is primarily because Klinger et al. assumes that the emission capacity of shade adapted leaves is representative of the entire canopy. The influence of leaf age has a lesser impact but should be considered and efforts made to characterize the leaves used in emission enclosure studies. We recommend that measurements of mature, sun-adapted leaves be used to establish isoprene emission capacities. However, we recognize that this is not always possible and suggest that canopy depth (cumulative LAI above the measured leaf or branch) be used in equation (4) to provide a rough adjustment to the emission activity of a leaf that has not been growing in full sunlight.

A quantitative estimate of the uncertainty associated with these estimates would require more data than are currently available. Instead, it is possible to discuss the general level of uncertainty associated with these estimates such as that indicated by comparing the results of the G95 and EXP96 models. The differences shown in Table 3 are somewhat representative of the minimum level of uncertainties in each of the model variables. The difference in the estimated annual total isoprene emission was -14%, although the differences in individual model variables ranged from -41% to 20%. Some variables are correlated and others are anticorrelated. An example of an anticorrelation is the reduction in foliar density (decreased D_p) which resulted in an increase in γ_P, the PPFD-dependent emission activity factor.

The above-canopy measurements available for evaluating model estimates for central Africa include a tower REA system (S99) which averaged over an area of about 1 km^2 and an aircraft REA system [*Greenberg et al.*, this issue] which characterized two regions of about 100 km^2. The reasonable agreement (within about a factor of 2) of the model predictions and field estimates is similar to the results of model evaluations reported for North America [e.g., *Guenther and Hills,* 1998]. These results show that the model produces reasonable estimates of fluxes for areas that have been characterized with ground measurements (e.g., LAI and enclosure emission measurement surveys), but it does not provide an indication of our ability to predict fluxes from other landscapes.

Three of the model variables (ρ, γ_T, and γ_P) influence diurnal variations, while four of them (D_f, γ_A, γ_T, and γ_P) determine annual variations. The uncertainty in isoprene emission estimates for a specific location and time is expected to be much larger than the uncertainty associated with the annual total for the entire EXPRESSO domain. This is because the limited data available for parameterizing the model are somewhat representative of the mean values for all landscapes and seasons but may not be representative of specific locations and times. For example, we are more certain that about 30% of the foliage of all woody plants in the EXPRESSO domain emit isoprene than we are of our estimate of this parameter at a specific location.

A general ranking of the uncertainties associated with each of the model variables in equation (1) can be assumed from model sensitivity tests, such as that shown in Table 3. Escape efficiency ρ is not likely to make a significant contribution to the uncertainty in annual isoprene estimates, although it may be important for predicting diurnal variations. The uncertainty associated with foliar densities (D_p and D_f) is at least 50% for the annual regional total and is probably more than a factor of 3 for a specific time and location. The total uncertainty associated with emission activities and emission capacity is probably similar to or higher than the uncertainties in foliar density estimates. Future improvements in satellite remote sensing technology could significantly improve these estimates. Advances in efforts to characterize vegetation canopies, above-canopy environmental conditions, and plant physiological processes should reduce uncertainties in our estimates of emission capacity and emission activity. Investigations in temperate landscapes can improve our ability to model isoprene emission in all regions, but there is a strong need for field studies in the tropics. The results of S99 and V99 suggest that our general modeling framework is acceptable but that field measurements are needed to parameterize the model for tropical landscapes.

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