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REASSESSMENT OF BIOGENIC VOLATILE ORGANIC COMPOUND EMISSIONS IN THE ATLANTA AREA

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Abstract---Localized estimates of biogenic volatile organic compound (BVOC) emissions are important inputs for photochemical oxidant simulation models. Since forest tree species are the primary emitters of BVOCs, it is important to develop reliable estimates of their areal coverage and BVOC emission rates. A new system is used to estimate these emissions in the Atlanta area for specific tree genera at hourly and county levels. The U.S. Department of Agriculture, Forest Service Forest Inventory and Analysis data and an associated urban vegetation survey are used to estimate canopy occupancy by genus in the Atlanta area. A simple canopy model is used to adjust photosynthetically active solar radiation at five vertical levels in the canopy. Leaf temperature and photosynthetically active radiation derived from ambient conditions above the forest canopy are then used to drive empirical equations to estimate genus level emission rates of BVOCs vertically through forest canopies. These genera-level estimates are then aggregated to county and regional levels for input into air quality models and for comparison with (1) the regulatory model currently used and (2) previous estimates for the Atlanta area by local researchers. Estimated hourly emissions from the three approaches during a documented ozone event day are compared. The proposed model yields peak diurnal isoprene emission rates that are over a factor of three times higher than previous estimates. This results in total BVOC emission rates that are roughly a factor of two times higher than previous estimates. These emissions are compared with observed emissions from forests of similar composition. Possible implications for oxidant events are discussed.

Key word index: Isoprene, monoterpene, foliar mass, forest inventory, oxidant.

INTRODUCTION

Biogenic volatile organic compounds (BVOCs) are important local and regional influences on photochemical oxidant formation (Fehsenfeld *et al.*, 1992). According to the National Research Council (1991), the ozone problem in many parts of the United States has been exacerbated by a policy that has emphasized control of anthropogenic volatile organic compound (VOC) emissions (such as those from motor vehicles and industrial sources) and has neglected the contribution that BVOC emissions make to ozone formation. Although these emissions occur naturally, they must be considered in deciding whether anthropogenic NO_x (the oxides of nitrogen, NO and NO₂) vs anthropogenic VOC should be controlled to reduce ambient ozone levels. Chameides *et al.* (1988) were the first to demonstrate the importance of BVOC emissions in the Atlanta area. They concluded that when natural BVOC emissions were considered in the Empirical Kinetics Modeling Approach (EKMA) (Seinfeld, 1988), anthropogenic NO_x control was preferred over anthropogenic VOC control to reduce episodic summertime ozone concentrations in the Atlanta area.

Current models such as the Urban Airshed Model and the Regional Oxidant Model use empirical algorithms such as the Biogenic Emissions Inventory System (BEIS) for estimating BVOC emissions from general (deciduous and coniferous) forest categories (Pierce *et al.*, 1990). Forests are thought to emit approximately 90% of BVOC, with agricultural and scrub lands contributing the balance (Lamb *et al.*, 1987). BVOC emissions from forested areas are typically estimated by multiplying an emission factor expressed as micrograms of carbon per gram foliar dry mass per hour (μ g-C g⁻¹ h⁻¹) by biomass density and land area factors. For crops, the emission rates are

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Table 1. Foliar densities $(g m^{-2})$ for each forest type, divided among emission categories (from Lamb *et al.*, 1987). Note that a mixture of forest types are assumed to occur within each emission class. For instance, the oak (*Quercus*) forests are dominated by high isoprene emitting species but are also assumed to contain a smaller fraction of lower emitting species such as those in the maple (*Acer*) and pine (*Pinus*) genera

Forest type	High isoprene deciduous	Low isoprene deciduous	Nonisoprene deciduous	Nonisoprene coniferous
Oak	185	60	60	70
Other deciduous	60	185	90	135
Coniferous	39	26	26	559

Table 2. Forest emission factors [μ g-C (g-foliar dry mass)⁻¹h⁻¹] standardized for bright sunlight ($\ge 800 \ \mu$ mol m⁻²s⁻¹) and 30°C (from Pierce *et al.*, 1990)

Chemical species	High isoprene deciduous	Low isoprene deciduous	Nonisoprene deciduous	Nonisoprene coniferous	
Isoprene (C ₅ H ₈)	13.0	5.82	0.0	0.0	
α -Pinene ($C_{10}H_{16}$)	0.11	0.04	0.06	1.00	
Other monoterpenes	0.10	0.04	0.06	1.14	
Other VOCs	2.86	1.55	1.69	1.22	

expressed as chemical mass per unit time per unit land area. Lamb *et al.* (1993b) estimates the U.S. annual total BVOC emission flux at 19 to 50 Tg (10^{12} g), with a "best" estimate of 29 Tg. These BVOC emissions are equal to or exceed estimated VOC emissions from anthropogenic sources on a national basis.

In this paper, we apply a new scheme (Guenther *et al.*, 1994; Geron *et al.*, 1994) for estimating hourly BVOC emissions from forest vegetation in the Atlanta area. The study area covers the original 11 county ozone nonattainment area discussed by Chameides *et al.* (1988). Our objectives are (1) to describe the use of the U.S. Department of Agriculture, Forest Service Forest Inventory and Analysis (FIA) data from Thompson (1989) and Cost* in determining a forest emission source inventory for the Atlanta area, (2) to estimate hourly BVOC emissions for an Atlanta ozone nonattainment day (4 June 1984), and (3) to compare these estimates with those from Chameides *et al.* (1988) and the BEIS.

METHODS

The approach currently used to estimate BVOC emissions on a regional basis (BEIS) combines foliar mass estimates derived from Lamb *et al.* (1987) with land use coverage from the Geoecology database (Olson *et al.*, 1980). Foliar density (grams of dry foliage per square meter) is estimated for each county by multiplying foliar density factors with forest area derived from the Geoecology data. Foliar densities are provided for only three forest types to be consistent with available emission factors. Likewise, 106 natural community types reported in the Geoecology data are simplified to either oak forests, other deciduous forests, coniferous forests, grassland/range, or barren lands. Foliar densities for the three forest types are based primarily on data from the International Biological Program as reported by Leith and Whitaker (1975), National Academy of Science (1975), Rasmussen (1972), and Dasmann (1976). Foliar densities are apportioned into emission classes and reflect estimated emission class compositions for each forest type as shown in Table 1.

Biogenic emission fluxes were generated from the computed foliar mass using emission factors adapted (as geometric means) from Lamb *et al.* (1987). These factors, shown in Table 2, are given for each forest emission category and were based upon a synthesis of available emission measurements. BEIS also includes a simple model to simulate the vertical profiles of leaf temperature and solar radiation in a forest canopy (Lamb *et al.*, 1993b). BVOC emission rates increase exponentially (up to approximately 38°C for isoprene) with increasing leaf temperature, while isoprene emission rate also increases with increasing light intensity up to approximately $1000 \,\mu$ mol m⁻² s⁻¹ (Guenther *et al.*, 1993; Tingey *et al.*, 1979, 1980).

Chameides *et al.* (1988) assumed that forests in the 11county Atlanta ozone nonattainment area emit no isoprene at night, increased linearly to a rate of 2100 μ g-Cm⁻²h⁻¹ at noon, remained constant until 3 PM local time, and then decreased linearly to zero at 7 PM local time. Nonisoprene BVOC emissions were assumed to be emitted at a rate of 1400 μ g-Cm⁻²h⁻¹ throughout the diurnal cycle. This simple model will be referred to hereafter as model GTECH.

General emission rate categories in models GTECH and BEIS were the only feasible approaches at the time these systems were developed, since relatively few emission rates were measured from dominant vegetation types, and the vegetation databases were not specific enough to provide coverage by individual species or genera. More recently, however, a comprehensive database of genus-specific emission rate estimates has been developed (Guenther *et al.*, 1994). In addition, the FIA has compiled county-level forest inventory data in consistent formats for Georgia (Thompson 1989). Using these databases, BVOC fluxes are calculated using the system described by Geron *et al.* (1994), which will be referenced in this paper as BEIS2.

^{*} Noel Cost, U.S. Department of Agriculture, Forest Service, Southeastern Forest Experiment Station, Asheville, North Carolina, unpublished data.



Fig. 1. County level total BVOC emission rates (numeric values in mg-C m⁻² h⁻¹) from forests and urban tree canopies averaged over the entire county land area. These emissions are corrected for canopy environment, ambient temperatures of 30 °C, and full sunlight (1500 μ mol m⁻²s⁻¹). The points indicate forested plot locations, while plus marks represent nonforest biomass sample plot locations within nonforest landuse types. Color shades indicate the percentage of county area covered by tree canopy. The 11-county Atlanta study area is outlined in blue.

Forest extent and crown cover

The forest statistics used in this study are a subset of those developed in Geron *et al.* (1994) for the 11county Atlanta study area. Forest extent, species composition, and tree diameter distribution data used in this approach are obtained from the most recently available FIA data for the state of Georgia (Thompson, 1989). These data are collected on approximately 10-year cycles by the U.S. Forest Service.

U.S. Forest Service estimates of forest area, timber volume, and tree species composition are based on a two-phase sampling strategy. In Phase 1, individual 0.4 hectare sample plots are systematically spaced on aerial photographs covering the entire state of Georgia. Each sample plot is classified using aerial-photo interpretation methods. In Phase 2, a subsample of classified forest plots are visited by FIA field survey teams, where detailed measurements of forest vegetation are taken. The Phase 1 sample provides the forest area estimates that are associated with each Phase 2 sample plot. Phase 2 measurements provide information about conditions that can be observed only on the ground and provide a correction for misclassification and change between the time the photography was taken and the inventory is being taken. For the 11-county Atlanta study area, the sampling intensity was approximately 1 plot per 1400 ha of land area.

The species composition and diameter distributions derived from the FIA data are used to approximate regional forest canopy coverage and foliar mass by genus as described in Geron *et al.* (1994). The percentage of horizontal canopy area occupied by each genus is assumed to be equal to the percentage of total stand basal area (sum of cross-sectional area of stems at 4.5 ft (1.37 m) above ground) of the respective genera with minor adjustments to account for crown width variation among genera (Geron *et al.*, 1994).

The U.S. Forest Service survey methods described above apply only to contiguous forest tracts of at least 1 acre (0.4 ha). In urban areas in general and Atlanta in particular, significant tree cover exists in smaller tracts within other landuse types (e.g. residential, low density developed). To account for such tree cover, a separate FIA nonforest database (Cost) was analyzed. This database consists of points determined from aerial photography to be of landuse types other than forest. The percentage ground surface covered by tree canopy on a 1 acre (0.4 ha) tract about each point was estimated from the photos and recorded to the nearest 10%. Of these plots, 30% (those with plot numbers ending in 0, 2, and 6) were selected for field measurements. On the plots with canopy cover estimated to be greater than zero, vegetation was identified and stem diameters measured on the ground. The total nonforest canopy cover determined from the aerial photo analysis was allocated to each genus in proportion to the fraction of total basal area each

genus accounted for on the ground plots. The sum of the nonforest area represented by these points is equal to and exhaustive of the total nonforest area within the 11-county study area. The combined forest and nonforest area data were exhaustive of the area as a whole and exclusive of each other (i.e. were nonoverlapping). We therefore have an estimate of area occupied by each genus in the 11-county area. Within this area, 604 forest and nonforest sample points were ground-sampled for tree genus and stem size (see Fig. 1).

Forest foliar mass

Foliar mass estimation is described in Geron *et al.* (1994). For the Atlanta area, the values are 700 g m⁻² for Pinus and Juniperus, and 375 g m⁻² for deciduous stands. These foliar density values are very similar to those used by Lamb *et al.* (1987) and in the BEIS. We assume that foliage occupies the respective horizontal canopy area at these densities for each class. Allocating foliar mass in this manner allows urban woodlots, open woodland, and understocked stands to assume less foliar mass and canopy area per hectare than fully stocked stands, while maintaining realistic maximum foliar masses for fully stocked, closed canopies during the period of peak foliation.

BVOC EMISSION RATES

Standardized emission rates

The database of Guenther et al. (1994) provides emission rates of isoprene and monoterpenes [µg-C $(g-foliar dry mass)^{-1}h^{-1}$ standardized for bright sunlight (1000 μ mol m⁻² s⁻¹) and 30 °C for dominant forest tree genera in North America. Standardized emission rates for species within a genus have generally been found to fall within \pm 50% of these emission rates. A third emission rate class aggregates all other volatile organic compounds (OVOCs) and is currently assumed to be $1.5 \,\mu g$ -C(g-foliar dry mass)⁻¹h⁻¹ with a range of 0.5-5.0 μ g-C g⁻¹h⁻¹ (Guenther et al., 1994). Individual compounds in this class vary greatly in reactivity, having lifetimes from a few hours to > 100 d (Guenther et al., 1994). The isoprene emission factor of 70 μ g-C (g-foliar dry $(mass)^{-1}h^{-1}$ is considerably higher than previous leaf-level emission factors for high isoprene emitting genera such as Quercus, Liquidambar, and Populus. This is largely due to interpretation of photosynthetically active radiation (PAR) levels associated with past branch-level enclosure measurements. Guenther et al. (1994) noted that, for branches with leaf areas indices near $3 \text{ m}^2 \text{ m}^{-2}$, leaves at the top of the enclosure would emit at a rate 75% greater than the average for the entire branch due to light extinction (shading) of foliage at the lower leaf layers within the enclosure. In addition, geometric mean emission factors are used in models BEIS and GTECH, while higher arithmetic mean emission rates are currently used in BEIS2.

Genus	Isop	Mono	ovoc	%Canopy	Common name
Pinus	0.1	3.0	1.5	34.4	Pine
Quercus	70.0	0.6	1.5	26.4	Oak
Liquidambar	70.0	3.0	1.5	9.2	Sweetgum
Fraxinus	0.1	0.1	1.5	7.5	Ash
Liriodendron	0.1	0.2	1.5	6.7	Yellow poplar
Cornus	0.1	1.6	1.5	6.5	Dogwood
Carya	0.1	1.6	1.5	5.5	Hickory
Fagus	0.1	0.6	1.5	3.7	American beech
Acer	0.1	1.6	1.5	3.3	Maple
Oxydendrum	0.1	0.6	1.5	1.9	Sourwood
Diospyros	0.1	0.1	1.5	1.8	Persimmon
Nyssa	14.0	0.6	1.5	1.2	Blackgum
Juniperus	0.1	0.6	1.5	1.1	Eastern red cedar
Prunus	0.1	0.1	1.5	1.0	Cherry
Ulmus	0.1	0.1	1.5	0.9	American Elm
Carpinus	0.1	1.6	1.5	0.3	Hornbeam
Magnolia	0.1	3.0	1.5	0.3	Magnolia
Platanus	35.0	0.1	1.5	0.3	Sycamore
Betula	0.1	0.1	1.5	0.2	Birch
Cercis	0.1	0.1	1.5	0.1	Redbud
Ilex	0.1	0.2	1.5	0.1	Holly
Malus	_	_	1.5	0.1	Apple
Morus	0.1	0.6	1.5	0.1	Mulberry
Salix	35.0	0.1	1.5	0.1	Willow

Table 3. BVOC emission factors $[\mu g-C (g-foliar dry mass)^{-1} h^{-1}]$ standardized for bright sunlight (1000 μ mol m⁻² s⁻¹) and 30°C for genera in the Atlanta area

Note: Isop = Isoprene, Mono = monoterpenes, and OVOC = other VOC. The percent contribution to canopy coverage is listed as % canopy. Total % canopy for the study area exceeds 100% due to rounding.

Table 3 lists the genera found in the Atlanta area, their percentage of total tree canopy coverage, and standard BVOC emission rates.

Canopy PAR correction

Since PAR strongly controls isoprene emission rate, a simple algorithm was applied to reduce PAR at lower levels within forest canopies. Specific leaf weight (SLW, in grams per square meter dry weight) is also adjusted according to the equation of Jurik (1986) so that deciduous leaves in the top canopy layer have SLWs nearly twice that of the lowest layer, since leaves in the upper canopy are twice as heavy per unit area (Monk et al., 1970; Jackson, 1966; Jurik, 1986). The foliar mass is therefore skewed toward the upper canopy which receives more sunlight. The combined effect of these adjustments is to reduce isoprene emissions by roughly 30-50% of that predicted without accounting for vertical PAR and SLW reduction downward through the forest canopy. These effects are discussed in Geron et al. (1994) and Lamb et al. (1993a, b).

RESULTS AND DISCUSSION

Forest canopy cover

Using the methodology described in the previous section, forest area, foliar density, and BVOC emission fluxes have been calculated for the 11-county Atlanta study area. The analysis of the forest area data (circa 1990) indicates that 53% of the 11-county study area is currently classified as forest. This is in excellent agreement with an assessment of 53% from recent (circa 1992) LANDSAT thematic mapper imagery [North Carolina State University Computer Graphics Center (NCSU-CGC), unpublished data] for the same area. Likewise, Chameides *et al.* (1988) found the area to be 57% forested from 1985 satellite imagery, and noted that forest loss in the area was estimated to occur at a rate of approximately 2% per year due to urbanization.

In analysis of the tree canopy cover in the nonforest landuse classes, we find that an additional 6% of the area is occupied by tree canopy, for a regional total of 59%. This is in good agreement with Pierce *et al.* (1993). Although all of these estimates are in relatively good agreement (53-59%), we believe that the upper end of this range is a reasonable recent total for tree cover found in both forest and nonforest landuse classes.

Chameides et al. (1988) estimated that, of the 57% forested area, 23% was deciduous, 18% mixed, and 16% coniferous forest. The estimates from the NCSU-CGC are 19, 23, and 11%, respectively. Since our estimates are at the genus level we are able to break down the "mixed" forest class into its deciduous and coniferous components. Of the 59% forest area, we

estimate that 39% is occupied by deciduous canopy and 20% by coniferous, again indicating reasonable agreement between the independent estimates.

The tree canopy cover in the study area falls into 24 genera (Table 3). The majority of the canopy cover (62.7%) falls into the lowest isoprene emitting category [0.1 μ g-C (g-foliar dry mass)⁻¹ h⁻¹]. However 35.6% (or 21% of the total study area) is occupied by genera in the highest isoprene emitting class. *Quercus* (oak) and *Liquidambar* (sweetgum) represent the only genera in this class sampled in the Atlanta area. The remaining 1.7% falls into the two intermediate categories. Quantitative isoprene and monoterpene emission rate data have been compiled by Guenther *et al.* (1994) for all genera except *Malus* (i.e. apple), which is estimated to compose less than 0.1% of the canopy cover.

BVOC emissions

Figure 2a shows the model results from the three systems for 4 June 1984. On this day ozone concentrations in Atlanta peaked at 0.147 ppmv, exceeding the National Ambient Air Quality Standard (NAAQS) of 0.120 ppmv. During mid-afternoon (2 to 3 PM LST) the BEIS2 estimates nearly twice as much total BVOC $(6.3 \text{ mg-C m}^{-2} \text{h}^{-1})$ emission as GTECH $(3.5 \text{ mg-Cm}^{-2} \text{h}^{-1})$ and over 2.5 times that estimated from BEIS $(2.5 \text{ mg-C m}^{-2} \text{ h}^{-1})$. Of these peak total BVOC emission estimates, isoprene composed 60, 67, and 86% of the total from GTECH, BEIS, and BEIS2, respectively. Diurnal total BVOC emissions for the study area from GTECH, BEIS, and BEIS2 are 49, 24, and 56 kg km⁻² d⁻¹, respectively. Isoprene composed 51, 31, and 77% of the respective diurnal totals. Chameides et al. (1988) estimated that BVOC emission rates in excess of 50 kg km⁻² d⁻¹, in the presence of anthropogenic NO_x emissions, could result in ozone (O_3) levels exceeding the NAAQS of 0.12 ppmv even if there were no local anthropogenic VOC sources. The high emission rates for the region estimated from BEIS2 are therefore potentially significant, especially when one considers that a much higher proportion of the estimated diurnal total flux occurs during the mid-afternoon, when high O₃ levels typically occur. Given the likelihood of a major contribution of BVOC emissions in the Atlanta area, ongoing analysis from the Southern Oxidant Study (SOS) in 1992 should help address the role that BVOCs play in ozone formation.

The primary difference between the three systems is the emission rate of isoprene. Models BEIS and GTECH use standardized emission rates determined in studies where the light environment about individual leaves in branch enclosures was poorly understood. As a result, leaf mean PAR exposure levels within the branch enclosures was probably overestimated, resulting in underestimation of the standardized isoprene emission rate by perhaps a factor of two (Lamb *et al.*, 1993b; Ciuenther *et al.*, 1994). They also assume that forested areas are composed of a uniform mix of forest types. This likely misrepresents the relative abundance of key genera such as Ouercus. Diurnal peak isoprene emissions from GTECH, BEIS, and BEIS2 were 2.1, 1.7, and 5.4 mg-C $m^{-2} h^{-1}$, respectively. Figure 2b illustrates the diurnal trends estimated from the respective systems. The GTECH and BEIS estimates are similar in magnitude and exhibit a profile nearly identical in form, while BEIS2 yields dramatic increases in diurnal isoprene emission estimates, especially in the afternoon of the sample day when ambient temperatures increase to approximately 32°C. Data collected from forests of similar composition in Oak Ridge, Tennessee, during the summer of 1992 (Lamb et al., 1993a) reveal reasonable agreement with the emissions we estimate for the Atlanta area using BEIS2.

Model GTECH assumes that a constant 1.4 mg- $Cm^{-2}h^{-1}$ flux of nonisoprene BVOC is emitted throughout the 24 h period. This accounts for the higher nighttime and morning emissions estimated by GTECH. BEIS and BEIS2 estimate emissions of these compounds as a function of temperature and are in good agreement with each other. There remains considerable uncertainty in these estimates, however. These diurnal patterns of nonisoprene BVOC emissions from the three models are demonstrated in Fig. 2c.

The findings presented here reinforce the photochemical significance of BVOC emissions presented by Chameides et al. (1988). They also present a strong case for additional improvements to BVOC emission rate models. Results presented in this paper and in Geron et al. (1994) indicate that relatively few genera are estimated to be responsible for a large proportion of BVOC flux (especially in the case of isoprene) on a regional basis. Since large variation (\pm 50%) is currently associated with genus-level emission rates, variability between and within species in important genera (e.g. Quercus, Liquidambar, and Pinus) could further improve BVOC emission models. In order to reduce this variability, it is likely that a better understanding of biochemical and physiological linkages to BVOC emission rates will be needed.

The OVOC category used currently assumes that all vegetation emits other VOCs at basically the same rate. This is obviously a gross simplification. Mac-Donald and Fall (1993) documented significant quantities of methanol emissions from various vegetation types. Goldan et al. (1993) also noted significant amounts of the C₅ alcohol methyl butenol in remote air over a western coniferous forest. In current BVOC models, these classes of compounds are poorly accounted for. As a crude illustration of their potential importance, if we assume a mean diurnal methanol emission rate of 13.2 μ g (g-foliar dry mass)⁻¹ h⁻¹ (or 5 μ g methanol carbon (g-foliar dry mass)⁻¹ h⁻¹) from MacDonald and Fall (1993) from forest vegetation in the Atlanta area, peak area average total BVOC rates increase by approximately 20% to over 7.4 mg- $Cm^{-2}h^{-1}$, and the daily total BVOC emission rate

increases to 80 kg km^{-2} . Although methanol is not considered reactive, its immediate reaction products (HO₂ radicals and formaldehyde) are reactive and influence photochemistry (MacDonald and Fall, 1993). Such findings emphasize the importance of screening dominant vegetation types in a nonintrusive

manner for other carbon containing compounds, including polar and oxygenated substances. Further disaggregation of the OVOC class by reactivity would also be beneficial to photochemical modeling exercises. This may require advanced analytical techniques capable of quantitating such compounds.







Fig. 2. (Continued).



Fig. 2(a)-(c). Area average isoprene, nonisoprene, and total BVOC emission rates (mg-C m⁻² h⁻¹), respectively, from forest and urban tree canopies. Emissions from model GTECH are adjusted as a step function of time of day as described in the text. Emissions from models BEIS and BEIS2 are corrected for canopy environment, ambient temperature, and PAR observed in the area on 4 June 1984.

Recent work by Sharkey et al. (1992), Kuzma and Fall (1993), and Monson et al. (1994) indicates that there are strong seasonal and developmental effects on emission rates, even after leaves have fully emerged in the spring. Isoprene emission rates are very low for several weeks following leafout, and likewise decrease during leaf senescence. Experimental data and models are needed to quantify the seasonal variation in these emission rates.

Forest canopy architecture and microenvironment models must also be tested and developed which account for leaf angle, energy balance, and moisture at landscape scales. These will greatly influence the light and temperature environment of BVOC emitting foliage, which have been illustrated as the primary controls of BVOC emissions (Lamb *et al.*, 1993a, b).

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