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# Simulated effects of changes in direct and diffuse radiation on canopy scale isoprene emissions from vegetation following volcanic eruptions

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## Abstract

Volcanic eruptions can alter the quality of incoming solar irradiance reaching the Earth's surface thereby influencing the interactions between vegetation and the Earth system. Isoprene ( $C_5H_8$ ) is a biogenic volatile organic compound emitted from leaves at a rate strongly dependent on the received flux of photosynthetically radiation radiation (PAR). We investigated the potential for volcanic eruptions to change the isoprene flux from terrestrial vegetation using canopy-scale isoprene emission simulations that vary either the relative or absolute amount of diffuse ( $I_{diff}$ ) and direct ( $I_{dir}$ ) PAR. According to our simulations, if the total amount of PAR remains constant while the proportion of  $I_{diff}$  increases, canopy-scale isoprene emissions increase. This effect increases as leaf area index increases. Simulating a decrease in the total amount of PAR, and a corresponding increase in  $I_{diff}$  fraction, as measured during the 1992 Pinatubo eruption, decreases daily total canopy-scale isoprene emissions from terrestrial vegetation by 17–19% (for leaf area indices of 6 and 2, respectively). These effects have not previously been realized or quantified. Better capturing the effects of volcanic eruptions (and other major perturbations to the atmospheric aerosol content) on isoprene emissions from the terrestrial biosphere, and hence on the chemistry of the atmosphere, therefore requires inclusion of the effects of aerosols they produce on climate and total PAR and the  $I_{diff}/I_{dir}$  ratio.

## 1 Introduction

A variety of biogenic volatile organic compounds (bVOCs) are produced and emitted by terrestrial ecosystems at the global scale, at flux rates far exceeding those of anthropogenic sources of VOCs (IPCC, 2007). The high chemical reactivity of bVOCs, coupled with their high mass emission rates, gives rise to substantial impacts on atmospheric chemistry (Laothawornkitkul et al., 2009; Arneeth et al., 2010). Estimates of above-canopy fluxes of isoprene ( $C_5H_8$ ) from terrestrial vegetation, and other reactive

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bVOCs, are required for quantitative Earth system studies. In particular, atmospheric chemistry models used to assess changes in tropospheric ozone and secondary aerosol formation (Carslaw et al., 2010) rely on good estimates of bVOC emission rates. Estimates of present day global isoprene emission totals from the terrestrial biosphere are typically 440–660 Tg C yr<sup>-1</sup> (Guenther et al., 1995, 2006; Lathiere et al., 2010). However, the fact that most models converge on a similar figure does not necessarily mean that the estimate is correct (Monson et al., 2007).

Experimental and field observations indicate that isoprene emissions from both individual leaves and the canopies of terrestrial vegetation are strongly dependent upon the amount of photosynthetically active radiation (PAR) received (Guenther et al., 1995, 2006). Empirically-based isoprene emission models therefore include a function of PAR although the partitioning of this PAR into diffuse ( $I_{\text{diff}}$ ) and direct ( $I_{\text{dir}}$ ) fractions is not considered (Guenther et al., 1995, 2006). However, radiation from the Sun is partially diffused by aerosol particles in the atmosphere, especially dust from volcanic eruptions (Farquhar and Roderick, 2003; Gu et al., 2003) via Mie scattering, elastic scattering due to particles larger than the wavelength of the radiation. Mie scattering occurs in addition to the Rayleigh scattering caused by atoms and molecules. Scattered, i.e. diffuse ( $I_{\text{diff}}$ ), radiation, gives a more uniform irradiance through the canopy and can therefore better penetrate deeper into the canopy and hence reach otherwise shaded leaves. Increases in  $I_{\text{diff}}$  therefore can enhance the gross primary productivity of vegetation at the canopy or ecosystem scales (Roderick et al., 2001; Osborne and Beerling, 2002; Gu et al., 2003; Farquhar and Roderick, 2003). At the global scale, Mercado et al. (2009) estimate that variation in  $I_{\text{diff}}$  from 1960 to 1999 led to a 25% enhancement in global gross primary productivity. Because isoprene emissions are, in part, dependent on the amount of radiation received by the leaf, this suggests that the complexity of the light environment must be considered in isoprene emissions models, rather than just the total amount of PAR received at the top of the canopy.

Volcanic eruptions inject enormous amounts of dust into the atmosphere and cause a change in the ratio  $I_{\text{diff}}/I_{\text{dir}}$ . In fact, instrumental records showed attenuation of  $I_{\text{dir}}$

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and enhancement of  $I_{\text{diff}}$  solar radiation, and a peak global cooling of 0.4 K after the Pinatubo eruption (McCormick et al., 1995; Olmo et al., 1999). Given these observations, we suggest that changes to the relative amounts of  $I_{\text{dir}}$  and  $I_{\text{diff}}$  following a major volcanic eruption might alter the rate of canopy-scale isoprene emissions to the atmosphere, with potentially important consequences for the oxidative capacity of the troposphere (Telford et al., 2010).

Here we investigate this hypothesis using a theoretical treatment that involves modifying a radiative transfer scheme of an existing isoprene emissions model (Guenther et al., 1995, 2006) with the DePury and Farquhar (1997) treatment of  $I_{\text{diff}}$  and  $I_{\text{dir}}$  irradiating a vegetation canopy. DePury and Farquhar (1997) developed a photosynthesis model permitting calculation of the  $I_{\text{dir}}$  and  $I_{\text{diff}}$  falling on the sunlit and shaded fractions of the canopy. We undertook simulations for two case studies with this coupled model, both for an arbitrary  $1 \text{ m}^2$  area of land, with a range of leaf area indices (LAI). In Case 1, we consider the effects of varying the proportion of  $I_{\text{diff}}$  with the total PAR flux remaining constant. In Case 2, we simulate the effects of observed changes in  $I_{\text{diff}}/I_{\text{dir}}$  following the Pinatubo volcanic eruption on canopy-scale isoprene emissions with a range of leaf indices.

## 2 Materials and methods

Canopy-scale isoprene emission rates were calculated, following the method established by Guenther et al. (1995, 2006), as

$$\text{Emission rate} = \varepsilon\gamma \quad (1)$$

where  $\varepsilon$  is an emission factor for a specific plant functional type at standard conditions (in  $\text{mg C m}^{-2} \text{ h}^{-1}$ ), and  $\gamma$  is the emission activity factor (dimensionless) which modifies the emission rate with functions of climate, environment and vegetation. It is the product of a factor for each variable considered:

$$Y = Y_t \times Y_{\text{LAI}} \times Y_p \quad (2)$$

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where  $\gamma_t$ ,  $\gamma_{LAI}$  and  $\gamma_p$  are the factors for temperature, LAI and PAR, respectively. Other factors can be included (soil moisture and leaf age, for example), but our calculations here address the specific hypothesis that different relative amounts of  $I_{dir}$  and  $I_{diff}$  can effect isoprene emission rates. We use the function for  $\gamma_t$  given in Guenther et al. (2006) which depends upon an hourly and daily average temperature. For PAR and LAI we consider the canopy to be divided into sunlit and shaded fractions which are differently affected by direct and diffuse radiation (DePury and Farquhar, 1997). We thereby calculate isoprene emission rates for the sunlit and shaded fractions separately. Then for a given value of LAI, the sunlit fraction is given by Guenther et al. (1995):

$$f_{sun} = LAI \times [1 - \exp(-0.5 \times f / \sin\beta)] \times \sin\beta \quad (3)$$

and the shaded LAI is therefore simply:

$$f_{shade} = LAI - f_{sun} \quad (4)$$

where  $\beta$  is the solar angle which we obtain from solar geometry equations for a given latitude, day of the year and hour of the day. We then calculate  $\gamma_{LAI}$  according to Guenther et al. (2006).

For  $\gamma_p$  we follow Guenther et al. (2006), where the equation is dependent upon hourly ( $P$ ), daily average ( $P_{24}$ ) and the previous 10 day average ( $P_{240}$ ) values of radiation, with a different constant ( $P_0$ ) used for sunlit and shaded parts.

$$\gamma_p = C_p [(\alpha \times P) / \sqrt{(1 + \alpha^2 \times P^2)}] \quad (5)$$

$$\alpha = 0.004 - 0.0005 \times \ln(P_{240}) \quad (6)$$

$$C_p = 0.0468 \times \exp(0.0005 \times (P_{24} - P_0)) \times P_{240}^{0.6} \quad (7)$$

Derivation of the equations for the calculation of direct, diffuse, sunlit and shaded PAR is given in DePury and Farquhar (1997), and these authors include terms for reflected and scattered PAR.

## 2.1 Model calculations

**Case 1. Fixed  $I_{\text{tot}}$  variable  $I_{\text{diff}}$ .** We first consider canopy isoprene emissions for differing  $I_{\text{dir}}$  and  $I_{\text{diff}}$  using calculated values of radiation, as per DePury and Farquhar (1997). In this case, we varied the relative proportion of  $I_{\text{diff}}$  from 0 to 40% of the total PAR, but held the total PAR photon flux density constant ( $1076 \mu\text{mol m}^{-2} \text{s}^{-1}$ ). We calculated isoprene emissions over a single day (choosing the 195th day of the year) for  $1 \text{ m}^2$  area of land surface, at latitude of  $55^\circ$ , temperature of 290 K, and  $\varepsilon = 10 \text{ mg isoprene m}^{-2} \text{ h}^{-1}$  and LAIs 3, 4 and  $6 \text{ m}^2 \text{ leaf m}^{-2}$  land surface. These environmental conditions, and the emission factor, represent a deciduous broad-leaved forest on a summer day in the Northern Hemisphere.

**Case 2. Pinatubo. Variable  $I_{\text{tot}}$  and  $I_{\text{diff}}$ .** Gu et al. (2003) reported measurements of direct and diffuse radiation in the years following the eruption of Mount Pinatubo in 1991, which was the largest eruption in the last 100 years, and which injected vast amounts of aerosol particles into the troposphere and stratosphere (equivalent to  $10^{10} \text{ t}$  of magma, and  $2 \times 10^7 \text{ t}$  of sulphur dioxide, some of which would oxidize in the atmosphere to form sulphate aerosol). We therefore used a second alternative approach to simulating the effect of changes in  $I_{\text{dir}}$  and  $I_{\text{diff}}$  on canopy-scale isoprene emissions using curves fitted to the data of Gu et al. (2003) obtained during (1992) and after (1994) the Pinatubo eruption under cloudless skies in a northern hardwood forest ( $42.5^\circ \text{ N}$ ,  $72.2^\circ \text{ W}$ ) (Fig. 1). PAR for the sunlit and shaded LAI fractions and modelled canopy emissions were computed as before. These data indicate  $I_{\text{diff}}$  increases by 50% to 70% and  $I_{\text{dir}}$  decreases by 30% to 9% between dawn/dusk and midday (Fig. 1). Environmental and isoprene emissions rates as for Case 1.

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### 3 Results and discussion

Our sensitivity analyses indicate that for Case 1, increasing the amount of  $I_{\text{diff}}$  radiation relative to the total PAR decreases isoprene emissions from the sunlit fraction of the canopy (Fig. 2a) and increases emissions from the shaded fraction (Fig. 2b).

Overall the differential effects of increases in the relative amount of  $I_{\text{diff}}$  on the sunlit and shaded fractions result in a small net increase in the daily total isoprene emissions; this effect increases with an increasing proportion of diffuse radiation (Fig. 2c). The strength of this effect increases as LAI increases and generates more shaded leaf area per unit area of land. This result is intuitive given the change in the shaded leaf area of a canopy resulting from changes in  $I_{\text{diff}}$  radiation which better penetrates the canopy, reaching a greater fraction of the shaded leaves, than direct radiation (Roderick et al., 2001; Farquhar and Roderick, 2003). These calculations indicate that sustained changes in the quality of solar radiation over the lifetime of plants could alter isoprene emissions from canopies, in addition to their well established effects on photosynthesis and productivity (Roderick et al., 2001; Osborne and Beerling, 2002; Gu et al., 2003; Farquhar and Roderick, 2003).

Results from the calculations under the conditions given by Case 2 driven by these observations represent a first-order simulation of the possible effects the eruption of Mount Pinatubo on isoprene emissions from vegetation arising solely from changes in the nature and quality of solar radiation. Figure 3 shows the results of our canopy isoprene emission calculations for this second more realistic case undertaken with LAI = 2 and 6, in representative dusty (1992) and clear (1994) atmospheres. We report the hourly variations in isoprene emissions because the proportion of  $I_{\text{diff}}$  radiation varies with time of day, being greater at dawn and dusk than at midday (Roderick et al., 2001). Total amounts of isoprene emitted per day were 14.4 and 11.7 mg C m<sup>-2</sup> for LAI = 2 in clear and dusty atmosphere, respectively, and 26.3 and 21.8 mg C m<sup>-2</sup> for LAI = 6. The percentage changes are quite similar at both low and high LAI, -19% for LAI = 2 and -17% for LAI = 2.

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Our results (Fig. 3) indicate that total canopy-scale isoprene emissions are greatest in the case of clear skies, as a result of the total radiation being higher, with more isoprene emitted from the shaded fraction of the canopy at higher LAI. The relative change in emissions going from clear to dusty sky, i.e. [(emissions in dusty sky – emissions in clear sky)/emissions in clear sky] are given in Fig. 4. These results show that going from a clear to a dusty atmosphere reduces isoprene emissions from the sunlit fraction of the canopy more than those from the shaded fraction. For a low LAI, emissions from the shaded fraction increase around midday, even though in a dusty atmosphere the total amount of radiation reaching the canopy has decreased. The sunlit fraction of the canopy has the same percentage change throughout the day and this only marginally increases with LAI = 6. For the shaded fraction of the canopy, the difference in relative change in emissions is much more affected by the time of day and there is much more of an effect due to LAI (Fig. 4). So there are interesting changes due to clear or dusty atmosphere with the isoprene emissions from the shaded part of the canopy, with respect to LAI and time of day.

It is interesting to compare our results with those of Telford et al. (2010), who calculated changes in isoprene emissions from the terrestrial biosphere, and their impacts on atmospheric chemistry, following the Pinatubo eruption in 1991. Telford et al. (2010) modelled a 9% reduction in global isoprene emissions, caused by the cooler, drier climate following Pinatubo and a reduction in total PAR. Our results indicate that explicit consideration of the reduction in  $I_{dir}$  and increase in  $I_{diff}$  in the aftermath of the eruption of Mount Pinatubo (McCormick et al., 1995) would lead to additional reductions in isoprene emissions from forests not yet considered, further enhancing the sink for methane through increased tropospheric hydroxyl abundance (the dominant sink for methane). This suggests that earlier estimates of the enhanced sink for methane after 1990 due to reductions in isoprene fluxes from the terrestrial biosphere of up to  $5 \text{ Tg}(\text{CH}_4)\text{yr}^{-1}$  (Telford et al., 2010) may represent an underestimate for indirect vegetation effects on the reduced growth rate of atmospheric methane (Dlugokencky et al., 2003).

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## 4 Conclusions

Our canopy-scale simulations highlight the potential for changes in the quality of incoming solar radiation following major perturbations resulting from atmospheric aerosol loading can exert effects on isoprene emissions from terrestrial vegetation. These effects have not previously been realized or quantified. We show that actual changes in  $I_{\text{diff}}$  and  $I_{\text{dir}}$  following the eruption of Mount Pinatubo, due to the injection of diffusing particles into the atmosphere, could have exerted significant reductions (17–19%) in canopy isoprene emissions, and that these effects vary with canopy LAI and time of day. Variations in the  $I_{\text{diff}}$  and  $I_{\text{dir}}$  following volcanic eruptions therefore likely represent a missing component in modelling the emissions of biogenic VOCs and their interaction with the Earth system. The calculated effects we report here may be amplified if changes in  $I_{\text{diff}}$  alter the basal emission factors for leaves in shaded or sunlit fractions of the canopy. We are not aware of any such differences but suggest our model calculations point to the need for experimental investigations in this area.

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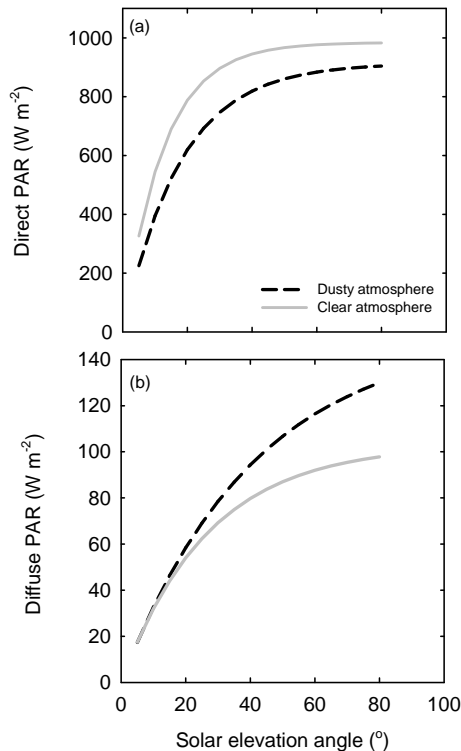
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**Fig. 1.** Variation of direct **(a)** and diffuse **(b)** PAR as a function of solar angle in dusty (1992) and clear (1994) atmospheres. The curves are given by **(a)** Direct PAR, dusty atmosphere =  $914.5 (1 - e^{-0.05664\beta})$ , **(b)** Diffuse PAR, dusty atmosphere =  $151.7 (1 - e^{-0.0423\beta})$ , **(c)** Direct PAR, clear atmosphere =  $984.4 (1 - e^{-0.08061\beta})$ , **(d)** Diffuse PAR, clear atmosphere =  $103.1(1 - e^{-0.03712\beta})$ , where  $\beta$  is the solar angle. Based on data from Gu et al. (2003).

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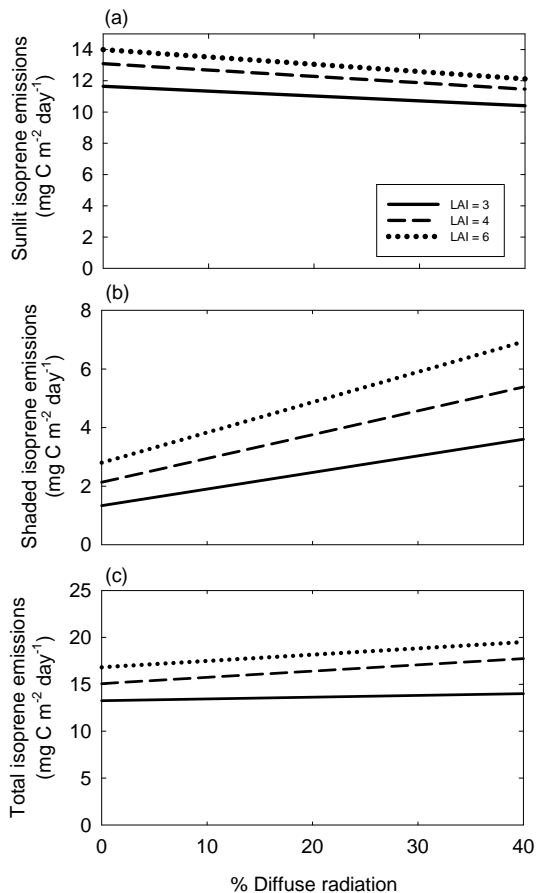
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**Fig. 2.** Calculated daily variations in **(a)** sunlit, **(b)** shaded, and **(c)** total canopy isoprene emissions as the proportion of  $I_{\text{diff}}$  radiation increases. Results are shown for a range of leaf area indices (LAIs). In these simulations, total PAR is kept fixed and all other conditions are as given in main text.

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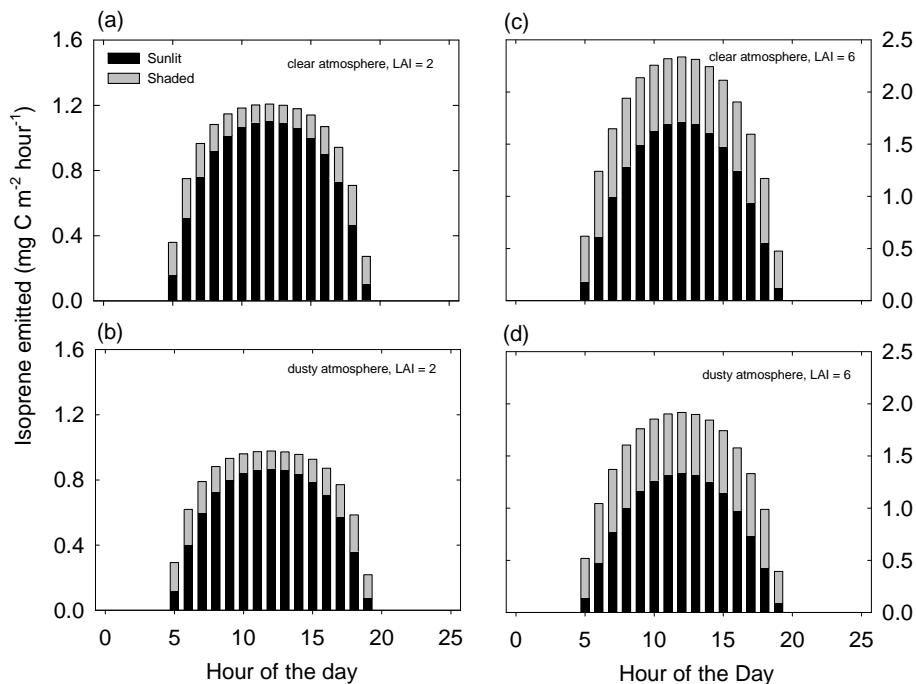
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**Fig. 3.** Hourly variation in sunlit and shaded isoprene emission for 1 m<sup>2</sup> using Eqs. (8) to (11) to calculate direct and diffuse PAR. **(a)** and **(b)** are for LAI = 2 in a dusty and clear atmosphere, respectively, **(c)** and **(d)** are the corresponding plots for an LAI = 6. Other conditions as stated in main text.

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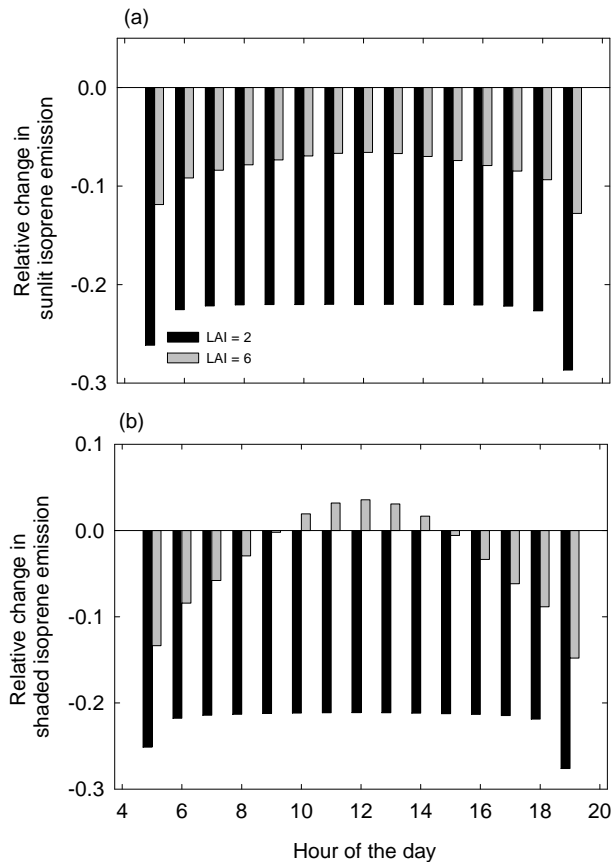
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**Fig. 4.** Relative change in isoprene emission for 1 m<sup>2</sup> over the course of a day for **(a)** sunlit and **(b)** shaded fractions of the canopy, calculated for an LAI = 2 and 6. Relative change is given as [(emissions in dusty sky – emissions in clear sky)/emissions in clear sky].