



## RESEARCH LETTER

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## Key Points:

- Human-influences on BVOCs cause net global climate forcing of  $-0.17 \text{ Wm}^{-2}$
- PI to PD net global cooling from BVOCs offsets net global warming from AVOCs
- BVOCs need to be included in assessments of anthropogenic climate forcing

## Supporting Information:

- Readme
- Table S1
- Table S2

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## On the role of plant volatiles in anthropogenic global climate change

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**Abstract** Biogenic volatile organic compound (BVOC) emissions from terrestrial ecosystems undergo rapid oxidation in the atmosphere that affects multiple warming and cooling climate pollutants. Since the preindustrial, BVOC-chemistry-climate interactions have been strongly influenced by anthropogenic changes in land cover, pollution emissions, and the physical climate state. Here, an Earth system model is applied to quantify the effects of BVOC emissions on the global radiation balance in the 1850s and 2000s including changes to tropospheric ozone, methane, and direct aerosol-radiation interactions. The net chemical forcing of global climate due to all known anthropogenic influences on BVOC emissions is  $-0.17 \text{ Wm}^{-2}$  (cooling) that offsets the  $+0.10 \text{ Wm}^{-2}$  (warming) due to anthropogenic VOC emissions from fossil fuel use and industry for this time period. BVOC emissions need to be included in assessments of anthropogenic radiative forcing.

### 1. Introduction

Biogenic volatile organic compound (BVOC) emissions from terrestrial ecosystems are recognized as a crucial quantity in chemistry-climate interactions [Fiore *et al.*, 2012]. Globally, the BVOC source exceeds 1 PgC per year [Guenther *et al.*, 2012], which is about a factor of 10 larger than the anthropogenic VOC (AVOC) source [Myhre *et al.*, 2013a]. BVOC emissions undergo rapid oxidation in the atmosphere that generates the climate pollutants tropospheric ozone ( $\text{O}_3$ ) and biogenic secondary organic aerosol (SOA) [Arneth *et al.*, 2010b; Carslaw *et al.*, 2010]. The photochemical processing of BVOC emissions influences the oxidation capacity of the atmosphere, which affects the lifetime of methane ( $\text{CH}_4$ ) and the production of other secondary aerosols including sulfate and nitrate particles whose formation rates depend upon the availability of oxidants [Unger *et al.*, 2006]. BVOC oxidation products are important for the growth of newly formed particles up to cloud condensation nuclei sizes of tens of nanometers [O'Dowd *et al.*, 2002; Riipinen *et al.*, 2012; Riccobono *et al.*, 2014] and could therefore also influence climate through cloud modification although these indirect effects are highly uncertain.

The IPCC Fifth Assessment Report (AR5) newly attributes radiative forcing in terms of primary precursor emissions because it provides a more direct link from human activities to radiative forcing [Myhre *et al.*, 2013a]. AVOC emissions are quantified as an anthropogenic radiative forcing mechanism that have contributed net global warming of  $+0.10 \text{ Wm}^{-2}$  through the sum of effects on (in  $\text{Wm}^{-2}$ ):  $\text{O}_3$  (+0.042),  $\text{CH}_4$  (+0.025), and  $\text{CO}_2$  (+0.033) [Myhre *et al.*, 2013a]. In contrast, BVOC emissions are quantified as a terrestrial biogeochemical feedback that responds to anthropogenic  $\text{CO}_2$ -emission induced surface warming [Ciais *et al.*, 2013]. Only the BVOC- $\text{O}_3$  feedback has been accounted for and has an estimated value of  $+0.03$  (+0.004 to 0.06)  $\text{Wm}^{-2} \text{ K}^{-1}$  [Arneth *et al.*, 2010a]. While the BVOC impacts on climate are frequently proffered as justification for further research, the net effect of BVOC emissions on global climate forcing in the Anthropocene is unknown. This knowledge gap is problematic for two current issues in climate policy: (1) the high ranking of forestry mitigation strategies in the international climate negotiations and (2) the ongoing debate at all levels of government regarding synergies and trade-offs between greenhouse gas and air pollutant mitigation.

Anthropogenic activities since the preindustrial have influenced all aspects of BVOC-chemistry-climate interactions from the BVOC emissions themselves to the chemical oxidation pathways and conversion to biogenic SOA. BVOC emission models that include interactive representation of all known global change drivers (vegetation productivity,  $\text{CO}_2$ -inhibition, physical climate, and human land cover change) indicate that the global source is about 20 to 25% less in the present-day compared to the preindustrial world [Lathiere *et al.*, 2010; Pacifico *et al.*, 2012; Unger, 2013]. These independent model frameworks agree that

human land cover change is the dominant driver of the BVOC emission decrease since the preindustrial. A recent study found that the human land-use-driven reduction in BVOC emissions since the preindustrial has a net cooling effect on global climate through changes to atmospheric chemistry ( $-0.11 \text{ Wm}^{-2}$ ) [Unger, 2014]. That study followed the IPCC definition of global radiative forcing, the energy imbalance imposed by a single perturbation to the climate system, and as such quantified the atmospheric chemistry effects of the 1850s to 2000s human land cover change for the 2000s atmospheric background state. Therefore, in that case, the atmospheric chemistry forcing signal is fully consistent with the IPCC assessments of the surface albedo and  $\text{CO}_2$  release forcings due to human land cover change.

However, turning to the perspective of the BVOC emissions themselves, the situation is more complex because their effects on the climate pollutants depend upon the chemical and meteorological background state of the atmosphere, both of which have also been modified by human activities since the 1850s. In particular, the BVOC-chemistry-climate interactions are highly sensitive to anthropogenic emissions of nitrogen oxides ( $\text{NO}_x$ ) and existing aerosol particles available for biogenic SOA partitioning [Carlton *et al.*, 2010; Hoyle *et al.*, 2011]. To unlock new ways of thinking about BVOC emissions and modern climate change, it is necessary to depart from the strict IPCC definition and allow the global radiative forcing for BVOC emissions to include perturbations to both biogenic and anthropogenic emissions. The goals of this study are: (1) to quantify the global radiative effects of BVOC emissions in the 1850s and 2000s including the chemical effects on  $\text{O}_3$ ,  $\text{CH}_4$  and multiple aerosols, (2) to isolate the BVOC emission impacts on global radiative forcing for the 2000s–1850s anthropogenic era due to all anthropogenic influences, and (3) to calculate the global warming potentials (GWPs) on the 20 and 100 year time horizons of tropical and temperate BVOC emissions in the present-day atmospheric state.

## 2. Methods

### 2.1. Global Earth System Model

This study applies the NASA GISS ModelE2 global chemistry-climate model at  $2^\circ \times 2.5^\circ$  latitude by longitude horizontal resolution with 40-vertical layers extending to 0.1 hPa [Schmidt *et al.*, 2014]. The model framework fully integrates the land biosphere-oxidant-aerosol system so that these components interact with each other and with the physics of the climate model. The atmospheric composition model has been well tested against observations and compared with other models [e.g., Koch *et al.*, 2010; Myhre *et al.*, 2013b; Shindell *et al.*, 2013a, 2013b; Stevenson *et al.*, 2013]. The difference in radiative forcing calculated using modeled  $\text{O}_3$  versus satellite-retrieved  $\text{O}_3$  from the Tropospheric Emission Spectrometer is only  $+0.016 \text{ Wm}^{-2}$  [Shindell *et al.*, 2013a]. The model reproduces monthly mean satellite and AeroNet observations for aerosol optical depth with a normalized mean bias not larger than 15% and a correlation coefficient of  $\sim 0.6$  [Shindell *et al.*, 2013b]. The model includes  $\text{NO}_x$ -dependent biogenic SOA production from the oxidation of BVOC emissions, isoprene, monoterpene, and other VOCs (ORVOC), using a two-product scheme that describes partitioning of semi-volatile compounds between the gas and aerosol phases depending on their volatility and preexisting carbonaceous aerosol availability [Tsigaridis and Kanakidou, 2007; Tsigaridis *et al.*, 2014]. Online oxidants affect aerosol production, and online aerosols provide surfaces for chemical reactions and influence photolysis rates. The vegetation submodel is embedded within the general circulation model that provides the key meteorological drivers for the vegetation physiology. The land-surface hydrology submodel provides the grid cell level soil characteristics to the vegetation physiology. The vegetation cover is described using eight ecosystem types: tundra, grassland, shrub, savanna, deciduous, tropical rainforest, evergreen, and crop. Leaf isoprene emission is calculated as a function of electron transport-limited photosynthesis, intercellular and atmospheric  $\text{CO}_2$  concentration, and canopy temperature [Unger *et al.*, 2013]. Leaf level monoterpene emissions are simulated by defining an ecosystem-specific basal rate that is modified using a temperature-dependent algorithm [Lathiere *et al.*, 2006]. ORVOC emissions are parameterized as 44% of the isoprene emission rate where 32% of ORVOC is assumed to represent biogenic SOA precursors [Griffin *et al.*, 1999]. The vegetation biophysics module computes the photosynthetic uptake of  $\text{CO}_2$  coupled with the transpiration of water vapor and the BVOC emission rates at the 30 min physical integration time step of the global model. The model does include an interactive empirical scheme for aerosol-cloud interactions [Schmidt *et al.*, 2014], which are not considered in this work due to the large mechanistic uncertainties for biogenic SOA particles [O'Donnell *et al.*, 2011; Scott *et al.*, 2014].

## 2.2. Simulations

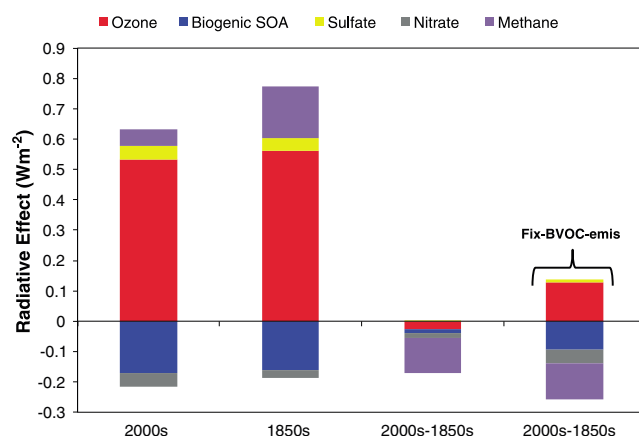
Control simulations are performed for the present-day (~2000s) and preindustrial (~1850s) atmospheres. Decadal average monthly varying sea surface temperature and sea ice climatology from the HadSST2 data set provide the physical climatic boundary conditions for the present (1996–2005) and preindustrial (1876–1885) simulations [Rayner *et al.*, 2006]. The present-day and preindustrial anthropogenic trace gas and aerosol emissions are prescribed to year 2000 and 1850 values, respectively, from the inventory developed for IPCC AR5 [Lamarque *et al.*, 2010]. The 1850s atmosphere has already been anthropogenically influenced, for instance with substantial emissions from domestic biofuel burning and deforestation burning. Vegetation cover is the standard atlas-based distribution in NASA ModelE2 [Schmidt *et al.*, 2014] that is overlaid with gridded anthropogenic land cover change for the years 2000 and 1850 [Hurtt *et al.*, 2011]. A pair of corresponding simulations is performed based on the 2000s and 1850s control runs but with the BVOC emissions selectively removed. A second set of sensitivity simulations is performed based on the control simulations (2000s and 1850s) but with the BVOC emissions held fixed at climatological monthly varying values for the 1850s. Analysis of these runs allows assessment of the relative roles of the anthropogenic versus biogenic emissions changes on the BVOC global radiative forcing (referred to as the “Fix-BVOC-emis” case). Finally, a set of two idealized perturbation simulations are performed based on the 2000s control but with BVOC emissions selectively increased by 10% in both the tropical (20°S–20°N) and temperate (30°N–60°N) biomes that have been more influenced by human land cover change than the boreal zone.

Integrations of 12 model years are completed for all control and sensitivity simulations; the first 2 years of the simulations are discarded as spin-up, and the remaining 10 years are averaged for analyses. Taking the difference between the sensitivity and control simulations allows isolation of the global radiative effects of BVOC emissions through O<sub>3</sub> and multiple aerosol species (biogenic SOA, sulfate, and nitrate) using the model’s online radiation submodel [Unger *et al.*, 2010]. The CH<sub>4</sub> radiative effect is calculated off-line based on the model changes in its chemical lifetime and accounts for the secondary long-term O<sub>3</sub> production from the CH<sub>4</sub> perturbation and the indirect stratospheric water vapor impact [Unger *et al.*, 2010]. The global warming potential (GWP) is the most widely used emission metric in policy considerations and the general standard adopted in climate assessments. To compute GWPs, the radiative effect of each agent is normalized to the total BVOC emission change for each perturbation and summed to obtain the Absolute Global Warming Potential (AGWP). The AGWP is then converted to GWP by normalizing to the 20 and 100 year CO<sub>2</sub> AGWP in Myhre *et al.* [2013a].

## 3. Results

### 3.1. Global Radiative Effect of BVOC Emissions in the 1850s and 2000s

The interactive BVOC emission model responds to all anthropogenic changes in physical climate and meteorology, land cover, atmospheric CO<sub>2</sub> levels, and vegetation productivity between the 1850s and 2000s. In the 1850s control, the global BVOC source strength is 777 TgC/yr (isoprene = 485 TgC/yr; monoterpene = 205 TgC/yr; ORVOCs = 87 TgC/yr), which is 30% higher than for the 2000s control simulation that simulates a global BVOC source of 607 TgC/yr (isoprene = 404 TgC/yr; monoterpene = 132 TgC/yr; ORVOCs = 70 TgC/yr). BVOC emissions substantially influence Earth’s global radiation budget in the 1850s and 2000s through multiple warming and cooling climate pollutants, which demonstrates the large lever that BVOC emissions represent in the global climate system (Figure 1 and Table S1). The global radiative effects are similar in both climatic states and are characterized by radiative warming due to increases in O<sub>3</sub> and CH<sub>4</sub> and decreases in sulfate aerosol that are partially offset by radiative cooling due to increases in biogenic SOA and nitrate aerosol. The BVOC-O<sub>3</sub> global radiative effect (+0.5 to +0.6 Wm<sup>-2</sup>) may be larger than the O<sub>3</sub> anthropogenic global radiative forcing [Myhre *et al.*, 2013a]. The BVOC-CH<sub>4</sub> global radiative effect is smaller than that of BVOC-O<sub>3</sub> in both climatic states. The biogenic SOA direct global radiative effect in 2000s (−0.17 Wm<sup>-2</sup>) compares well to published estimates from other global models: −0.26 Wm<sup>-2</sup> [O’Donnell *et al.*, 2011] and −0.08 to −0.78 Wm<sup>-2</sup> [Scott *et al.*, 2014]. In the 1850s and 2000s, BVOC emissions exert a substantial net warming global radiative effect of +0.4 to +0.6 Wm<sup>-2</sup>. Holding the BVOC emissions fixed at the 1850s values in the 2000s world (Fix-BVOC-emis case) strongly enhances the BVOC-O<sub>3</sub> and biogenic SOA global radiative effects by 33% and 44%, respectively, and dampens the BVOC-CH<sub>4</sub> global radiative effect by 50% (Table S1). Thus, increases in anthropogenic pollution emissions intensify the BVOC global radiative effects, even in the absence of any changes to the BVOC emissions themselves.



**Figure 1.** Global radiative effects of biogenic volatile organic compound (BVOC) emissions through multiple warming and cooling climate pollutants in the 2000s and 1850s. The difference (2000s minus 1850s) defines the global radiative forcing of BVOC emissions for this time period. Also shown are the radiative forcing results for the sensitivity case that considers changes to anthropogenic emissions only (Fix-BVOC-emis) in which BVOC emissions do not change between the 1850s and 2000s. Uncertainty estimates (<10%) computed from the standard error of the mean for 10 years of model output based on internal variability in the climate model are provided in Table S1.

### 3.2. Global Radiative Forcing of BVOC Emissions 2000s Minus 1850s

Figure 1 shows the difference between the BVOC global radiative effects in each climatic state that defines the anthropogenic radiative forcing of BVOC emissions (2000s minus 1850s). The value accounts for all human-driven impacts, including increases in anthropogenic pollution emissions, decreases in BVOC emissions, and changes to the physical climate state, on the BVOC-chemistry-climate interactions across this time period. The most substantial difference between the time periods is a 3 times larger CH<sub>4</sub> radiative effect in the 1850s versus the 2000s, which results in a CH<sub>4</sub> forcing of  $-0.12 \text{ Wm}^{-2}$ . The anthropogenic radiative forcing of BVOC emissions (2000s minus 1850s) exerts smaller global cooling through O<sub>3</sub> ( $-0.03 \text{ Wm}^{-2}$ ) and biogenic SOA ( $-0.012 \text{ Wm}^{-2}$ ). In

contrast, the human land cover change single perturbation and resultant AVOC emissions reduction (in the absence of changes to anthropogenic pollution emissions or physical climate state) result in a larger and opposite sign biogenic SOA radiative forcing ( $+0.09 \text{ Wm}^{-2}$ ) [Unger, 2014]. In sum, all anthropogenic influences on BVOCs are responsible for a substantial net cooling global radiative forcing of  $-0.17 \text{ Wm}^{-2}$  that completely offsets the globally warming AVOC radiative forcing [Myhre et al., 2013a].

Further insights are obtained by considering the results for the sensitivity simulations that hold the BVOC emissions fixed to 1850s values (Fix-BVOC-emis case; Figure 1 and Table S1). This case isolates the effects of the anthropogenic pollution emission increases on the BVOC-chemistry-climate interactions. In the absence of BVOC emissions changes, the effects of the anthropogenic pollution emission increases on BVOCs between the 1850s and 2000s drive a large positive O<sub>3</sub> radiative forcing ( $+0.13 \text{ Wm}^{-2}$ ) and a substantial biogenic SOA forcing ( $-0.10 \text{ Wm}^{-2}$ ). However, coupling the anthropogenic-driven 30% loss of BVOC emissions to the anthropogenic pollution emission increases between the 1850s and 2000s reverses the sign of the BVOC-O<sub>3</sub> climate impact and substantially dampens the biogenic SOA response. In this SOA model, the effects of anthropogenic-driven increases in oxidation capacity and existing aerosol particles onto which biogenic SOA can partition are large enough to offset the loss of the primary precursor BVOC emissions resulting in a slightly enhanced direct radiative effect in the present day relative to the preindustrial.

The CH<sub>4</sub> forcing response is similar for all anthropogenic effects on BVOC emissions and the Fix-BVOC-emis case. Thus, it is deduced that the CH<sub>4</sub> forcing is mostly driven by the effects of anthropogenic emissions increases (NO<sub>x</sub>). Indeed, the CH<sub>4</sub> signal reflects the larger relative influence of BVOC emissions on the CH<sub>4</sub> chemical lifetime in the cleaner preindustrial world. The net global radiative forcing for the Fix-BVOC-emis case ( $-0.12 \text{ Wm}^{-2}$ ) is dampened by about 30% relative to the effects of all anthropogenic impacts ( $-0.17 \text{ Wm}^{-2}$ ) mostly due to the different sign O<sub>3</sub> climate impact.

### 3.3. GWPs of Tropical and Temperate BVOC Emissions

The 10% uniform increase in tropical BVOC emissions amounts to an additional 37 TgC/yr ( $\Delta$  isoprene = 27 TgC/yr;  $\Delta$  monoterpene + ORVOCs = 10 TgC/yr). The 10% uniform increase in temperate BVOC emissions amounts to an additional 15 TgC/yr ( $\Delta$  isoprene = 9 TgC/yr;  $\Delta$  monoterpene + ORVOCs = 6 TgC/yr). In the present-day climate, the characteristic radiative response to tropical and temperate BVOC emission injections is similar

with the largest AGWPs for changes to (in  $\text{mWm}^{-2}/\text{TgC}$ ):  $\text{O}_3$  (+0.5 to +0.6; warming), biogenic SOA (−0.3 to −0.4; cooling), and  $\text{CH}_4$  (+0.2; warming) (Table S2). Increases in tropical and temperate BVOC emissions have smaller AGWPs through (in  $\text{mWm}^{-2}/\text{TgC}$ ): sulfate (+0.05 to +0.13; warming), nitrate (−0.03 to −0.05; cooling), and  $\text{CO}_2$  (+0.01 to +0.02; 20 year warming). For both ecosystems, the largest BVOC AGWP is through increases to  $\text{O}_3$ . The strong positive  $\text{O}_3$  signal in the tropics may seem surprising because, under low  $\text{NO}_x$  conditions, BVOC emissions tend to reduce ambient  $\text{O}_3$  levels. Indeed, BVOC emission increases do drive local surface reductions in  $\text{O}_3$  in low  $\text{NO}_x$  regions including the tropical rainforest. However, the BVOC injection always results in an overall increase in  $\text{O}_3$  at the global scale. At the global-scale, the cooling biogenic SOA response offsets about half of the  $\text{O}_3$  warming for tropical BVOCs and up to 80% for temperate BVOCs. The  $\text{CH}_4$  AGWP is similar for both tropical and temperate BVOC emissions injections. Remarkably, increases in tropical BVOCs have a 40% larger global warming impact per unit emission than increases in temperate emissions because temperate emissions have a lower  $\text{O}_3$  efficiency but a higher biogenic SOA efficiency than tropical emissions. Hence, the 20 year GWP for tropical BVOC emissions is 24 and for temperate emissions is 17. The 100 year GWP is 7 for tropical emissions 5 for temperate emissions. In comparison, AVOC 20 year GWPs are in the range of 15–30 (depending on emission region) that decrease to 4–9 on the 100 year time horizon [Fry *et al.*, 2012, 2014; Collins *et al.*, 2013]. To put the BVOC emission results into perspective, a 25% increase in tropical BVOC emissions would have approximately the same global warming impact as the entire aviation sector,  $\sim +50 \text{ mWm}^{-2}$  [Lee *et al.*, 2009].

#### 4. Discussion and Conclusions

A state-of-the-science global Earth system model has been applied to show that anthropogenic influences on BVOC emissions drive a global climate forcing that is net cooling ( $-0.17 \text{ Wm}^{-2}$ ). The quantitative error assessment values ( $<10\%$ ) presented in Table S1 are due to internal variability in the climate model based on the standard error of the mean for 10 years of model output. Larger uncertainties exist due to limitations in several key areas. The BVOC emission response to global change factors, in particular the treatment of the  $\text{CO}_2$ -inhibition effect that has ramifications for the relative importance of human land cover change as a driver, is a source of uncertainty [Monson *et al.*, 2007]. New measurements suggest that the instantaneous  $\text{CO}_2$ -inhibition of isoprene emissions is no longer effective under higher temperatures [Sun *et al.*, 2013]. While important recent progress has been made in improving understanding of the BVOC chemical oxidation pathways under both pristine and polluted conditions, uncertainties do still persist [Paulot *et al.*, 2009; Crouse *et al.*, 2011; Mao *et al.*, 2012, 2013]. Future research needs to examine possible nonlinear behavior in the BVOC emission global radiative forcing across different  $\text{NO}_x$  regimes. A large source of uncertainty is from the biogenic SOA production [Hallquist *et al.*, 2009; Carslaw *et al.*, 2010]. Current generation global models significantly underestimate the amount of organic aerosol present in the atmosphere and are unable to reproduce the variability in the measurements [Tsigaridis *et al.*, 2014]. Let us consider that the biogenic SOA negative direct radiative effect would need to be a factor of 3 times larger to compete with the large positive BVOC- $\text{O}_3$  radiative effect. This level of underestimate seems unlikely. However, there may be additional anthropogenic influences on the SOA production not considered here that could result in a larger 2000s minus 1850s SOA radiative forcing, which would drive a larger overall net cooling BVOC global climate forcing. Finally, a recent estimate of the biogenic SOA aerosol indirect effect that accounts for new particle formation [Scott *et al.*, 2014] does compete with the magnitude of the BVOC- $\text{O}_3$  radiative effect and therefore could potentially change the sign of the BVOC emission global radiative effect in the 1850s and 2000s climatic states to net negative (cooling). However, the 2000s minus 1850s anthropogenic BVOC radiative forcing results presented here are not sensitive to the biogenic SOA aerosol indirect effect because the preindustrial to present-day change in biogenic SOA direct forcing is so small, about a factor of 10 smaller than for the BVOC- $\text{CH}_4$  forcing. A direct aerosol forcing of this magnitude ( $-0.012 \text{ Wm}^{-2}$ ) does not result in a statistically significant global aerosol indirect effect forcing in a global climate model relative to natural internal variability [Unger *et al.*, 2010]. As knowledge of the BVOC emissions, oxidation pathways, SOA production mechanisms, and cloud interactions improves, these processes should be incorporated into the analysis.

Previous research has highlighted the importance of BVOC emissions in regional climate change [Pitman *et al.*, 2012]. This work provides the first quantitative evidence that BVOC emissions may play a role in modern

climate change at the global scale. While BVOC emissions represent a deceptively negligible flux in the global carbon cycle (1% of gross carbon assimilation), changes in these potent natural emissions can have a disproportionately large impact on the global radiation budget. BVOC emissions need to be included in future IPCC assessments of anthropogenic radiative forcing.

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Data supporting Figure 1 and the GWP calculations are available as Supporting Information Table S1 and Table S2.

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