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Airborne measurements of isoprene and monoterpene emissions from southeastern U.S. forests

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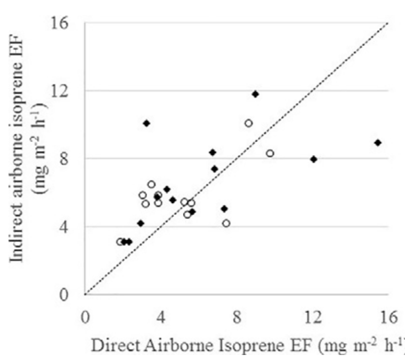
HIGHLIGHTS

- Aircraft eddy covariance and inverse modeling BVOC flux measurements agree.
- Models should account for differing emission factors in upper and lower canopy.
- Modeled and measured emissions agree for landscapes with high emitters.

GRAPHICAL ABSTRACT



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ABSTRACT

Isoprene and monoterpene emission rates are essential inputs for atmospheric chemistry models that simulate atmospheric oxidant and particle distributions. Process studies of the biochemical and physiological mechanisms controlling these emissions are advancing our understanding and the accuracy of model predictions but efforts to quantify regional emissions have been limited by a lack of constraints on regional distributions of ecosystem emission capacities. We used an airborne wavelet-based eddy covariance measurement technique to characterize isoprene and monoterpene fluxes with high spatial resolution during the 2013 SAS (Southeast Atmosphere Study) in the southeastern United States. The fluxes measured by direct eddy covariance were comparable to emissions independently estimated using an indirect inverse modeling approach. Isoprene emission factors based on the aircraft wavelet flux estimates for high isoprene chemotypes (e.g., oaks) were similar to the MEGAN2.1 biogenic emission model estimates for landscapes dominated by oaks. Aircraft flux measurement estimates for landscapes with fewer isoprene emitting trees (e.g., pine plantations), were about a factor of two lower than MEGAN2.1 model estimates. The tendency for high isoprene emitters in these landscapes to occur in the shaded understory, where light dependent isoprene emissions are diminished, may explain the lower than expected emissions. This result demonstrates the importance of accurately representing the vertical profile

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of isoprene emitting biomass in biogenic emission models. Airborne measurement-based emission factors for high monoterpene chemotypes agreed with MEGAN2.1 in landscapes dominated by pine (high monoterpene chemotype) trees but were more than a factor of three higher than model estimates for landscapes dominated by oak (relatively low monoterpene emitting) trees. This results suggests that unaccounted processes, such as floral emissions or light dependent monoterpene emissions, or vegetation other than high monoterpene emitting trees may be an important source of monoterpene emissions in those landscapes and should be identified and included in biogenic emission models.

1. Introduction

It is widely recognized that terrestrial ecosystems produce and emit a large variety of biogenic volatile organic compounds (BVOCs) into the atmosphere and this flux dominates the global budget of VOC emissions from all sources combined (Guenther et al., 1995; Kesselmeier and Staudt, 1999; Goldstein and Galbally, 2007; Lamarque et al., 2010). Most BVOC, including isoprene and monoterpenes, are highly reactive and have significant roles in both daytime and nighttime atmospheric chemistry (Zhang et al., 2000; Brown et al., 2009). This includes their contribution to the formation of Secondary Organic Aerosol (SOA) (Carlton et al., 2009), which is a major air quality issue and a significant uncertainty in climate change simulations (Pachauri et al., 2014). Furthermore, BVOC are an important precursor to ground level ozone in rural and populated urban areas (Chameides et al., 1988; Guo et al., 2012). As one of the six Criteria Air Pollutants regulated under the National Ambient Air Quality Standards, ground level ozone poses substantial threats to human health and public welfare in the U.S. (U.S. Environmental Protection Agency, 2014).

To quantify the impact of isoprene and monoterpenes on climate and air quality, accurate prediction of the spatiotemporal distributions of emissions is needed. Numerical approaches, including the Model of Emissions of Gases and Aerosols from Nature (MEGAN, Guenther et al., 2006, 2012), are dependent on the availability of accurate landcover distributions including the foliage area per unit ground area (Leaf Area Index, LAI, $m^2\ m^{-2}$) of vegetation covered surfaces, which we refer to as LAIv, and the relative composition of different plant chemotypes, which are plant species with differing capacities to emit BVOCs. MEGAN BVOC emissions are driven by global LAIv and chemotype fractions data that are estimated using satellite based landcover distributions calibrated using ground based vegetation measurements. This may require high resolution, regional specific information to supplement global datasets in heterogeneous landscapes such as urban areas (Kota et al., 2015). In addition to the need for distributions of chemotypes, it is necessary to specify the emission factors associated with each chemotype. The lack of direct measurements has made it difficult to evaluate the isoprene emission factors (EFs) used in regional to global emissions models (Warneke et al., 2010; Zare et al., 2012). These EF databases have primarily been based on measurements from enclosure experiments at leaf or branch level and have been evaluated by comparison to boundary layer concentrations and tower based eddy flux measurements (Guenther et al., 2006). Evaluations of isoprene EF databases in the eastern US have reported differences of about a factor of two between observed and predicted isoprene concentrations and note that this is within the uncertainty of the indirect measurement approach (Carlton and Baker, 2011; Kota et al., 2015; Warneke et al., 2010). A recent study by Kota et al., 2015; compared the global MEGAN BVOC emission model with tower eddy flux measurements in an urban area and concluded that the EF were too high and the LAI values were too low demonstrating the need for better inputs in heterogeneous areas such as urban landscapes. The eddy covariance flux measurement approach, which provides a direct measurement of isoprene emissions and largely simplifies the interpretation of chemical losses and boundary layer dynamics, has recently been extended from canopy to regional scales using airborne eddy flux measurement techniques (Karl et al., 2009). Previous applications of this technique have quantified the VOC emissions of individual shale gas facilities in the southeastern US (Yuan et al., 2015) and have demonstrated that the MEGAN model can accurately simulate isoprene EF distributions across oak woodlands and other landscapes in California (Misztal et al., 2014, 2016).

In this study, we derive high resolution isoprene and monoterpene flux estimates from aircraft eddy covariance observations collected in the southern US during the 2013 SAS (Southeast Atmosphere Study) and compare direct and indirect flux measurement approaches. We assess the landcover and environmental data available for relating emission rates to emission factors and consider the uncertainties involved in both. The emission factors calculated for high emission chemotypes are described and compared with the values used for MEGAN and the implications for regional BVOC emission modeling are discussed.

2. Methods

2.1. Landcover and meteorological data

2.1.1. LAI foliar distributions

LAI refers to projected one-sided green leaf area per unit ground surface area (m^2 of leaf area / m^2 of ground area). The LAI of vegetation covered surfaces, LAIv, is defined as

$$\mathrm{LAIv} = \mathrm{LAI}/\mathrm{f_c} \tag{1}$$

where f_c is the area fraction that is covered by living vegetation. For this study, 8 day averaged LAI data for North America were retrieved from the MODIS (MOderate Resolution Imaging Spectroradiometer) satellite product Collection 5 (MCD15A2) (Fang et al., 2013) for the study period and combined with climatological maximum green vegetation fraction data (Broxton et al., 2014) based on MODIS remote sensing products MCD12Q1 and MOD13A2. Spatial resolution of the LAIv data is approximately 900 m and the temporal resolution is 8 days. A multiyear LAIv database (years 2003 to 2013) was compiled to investigate the importance of using LAIv for the specific campaign period.

2.1.2. Vegetation type distributions

Vegetation species have vastly different capabilities to produce and emit BVOC and thus information on the distributions of the abundance of high and low emitting plant species is needed for realistic regional BVOC emission estimates (Rasmussen, 1970). The MEGAN2.1 model adopted the 16 CLM (Community Land Model) Plant Functional Type (PFT) schemes (Oleson et al., 2013) as the first step in categorizing spatial variations in BVOC emission chemotypes. PFTs are broad categories of vegetation (e.g. temperate broadleaf deciduous trees and boreal evergreen shrubs) with average emission factors that differ substantially. Emission capacities of different plant species within a PFT can still vary considerably. For this project, the global PFT database described by Guenther et al. (2012) was updated to produce a 30 m resolution PFT database, following the CLM4.5 PFT scheme, for the contiguous US.

The CLM 16 PFT scheme was designed to represent variability in land-surface exchange processes, including water and energy fluxes, but it does not fully capture BVOC emission variations. For example,