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Evaluation of MEGAN predicted biogenic isoprene emissions at urban locations in Southeast Texas

Sri Harsha Kota [a, 1], Gunnar Schade [b], Mark Estes [c], Doug Boyer [c], Qi Ying [a, *]

[a] Zachry Department of Civil Engineering, Texas A&M University, College Station, TX 77843, USA
[b] Department of Atmospheric Sciences, Texas A&M University, College Station, TX 77843, USA
[c] Texas Commission of Environmental Quality, Austin, TX 78704, USA

HIGHLIGHTS

- CMAQ over-predicted isoprene concentrations using MEGAN generated emissions.
- Predicted isoprene oxidation products, MACR and MVK, were higher than observations.
- MEGAN significantly over-predicted isoprene concentrations in urban Houston area.
- Gridded EF of isoprene in urban Houston area in MEGAN input was overestimated.

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ABSTRACT

Summertime isoprene emissions in the Houston area predicted by the Model of Emissions of Gases and Aerosol from Nature (MEGAN) version 2.1 during the 2006 TexAQS study were evaluated using a source-oriented Community Multiscale Air Quality (CMAQ) Model. Predicted daytime isoprene concentrations at nine surface sites operated by the Texas Commission of Environmental Quality (TCEQ) were significantly higher than local observations when biogenic emissions dominate the total isoprene concentrations, with mean normalized bias (MNB) ranges from 2.0 to 7.7 and mean normalized error (MNE) ranges from 2.2 to 7.7. Predicted upper air isoprene and its first generation oxidation products of methacrolein (MACR) and methyl vinyl ketone (MVK) were also significantly higher (MNB = 8.6, MNE = 9.1) than observations made onboard of NOAA's WP-3 airplane, which flew over the urban area. Over-prediction of isoprene and its oxidation products both at the surface and the upper air strongly suggests that biogenic isoprene emissions in the Houston area are significantly overestimated. Reducing the emission rates by approximately 3/4 was necessary to reduce the error between predictions and observations. Comparison of gridded leaf area index (LAI), plant functional type (PFT) and gridded isoprene emission factor (EF) used in MEGAN modeling with estimates of the same factors from a field survey north of downtown Houston showed that the isoprene over-prediction is likely caused by the combined effects of a large overestimation of the gridded EF in urban Houston and an underestimation of urban LAI. Nevertheless, predicted ozone concentrations in this region were not significantly affected by the isoprene over-predictions, while predicted isoprene SOA and total SOA concentrations can be higher by as much as 50% and 13% using the higher isoprene emission rates, respectively.

1. Introduction

Plants emit significant amounts of biogenic volatile organic compounds (BVOCs) into the atmosphere (Guenther, 1995). Many of the BVOCs are highly reactive unsaturated alkenes, such as isoprene and volatile terpenes, that can effectively react with hydroxyl radicals (Zhang et al., 2000), ozone (Warneke et al., 2004) and nitrate radical (Brown et al., 2009, 2013) in atmospheric photochemical reactions. BVOCs are the dominant precursors to global secondary organic aerosol (SOA) loading (Carlton et al., 2009) and thus can have significant impacts on global climate

* Corresponding author.
E-mail address: qying@civil.tamu.edu (Q. Ying).
[1] Now at: Department of Civil Engineering, Indian Institute of Technology, Guwahati, 781903, India.

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(Pacifico et al., 2009). In urban areas with significant biogenic influences, BVOCs can also contribute significantly to regional SOA concentrations (Budisulistiorini et al., 2013; Lin et al., 2013). In addition to their role in aerosol formation, BVOCs are important precursors of tropospheric ozone. For example, the incremental ozone reactivity of isoprene is approximately 20% higher than that of ethylene (Carter, 1994; Derwent et al., 1996). Consequently, isoprene may dominate ozone formation in rural areas (Dreyfus et al., 2002) but also contribute significantly in densely populated areas (Guo et al., 2012) In a previous study covering Southeast Texas, Ying and Krishnan (2010) showed that contributions of biogenic emissions to ozone formation are ~20% higher than contributions from anthropogenic emissions.

Evaluation of the impacts on air quality and global climate due to biogenic emissions depends on accurate isoprene and other BVOC emissions estimations from different vegetation types. A number of models have been developed to estimate biogenic emissions in regional and global scales, such as the Biogenic Emission Inventory System (BEIS) (Pierce et al., 1998), the Model of Emissions of Gases and Aerosols from Nature (MEGAN) (Guenther et al., 2006, 2012), and the Global Biosphere Emissions and Interactions System (GloBEIS3) (Yarwood et al., 2002). These emission models have been directly evaluated with leaf-level and ambient flux measures (Kaser et al., 2013; Langford et al., 2010), and indirectly with observed concentrations (Warneke et al., 2010). Model inter-comparison studies have also been reported. For example, Carlton and Baker (2011) used the Community Multiscale Air Quality (CMAQ) model to simulate concentrations of BVOCs and their oxidation products in a high emission region in central United States using BEIS and MEGAN, and found that while MEGAN significantly over-estimated isoprene and monoterpene ambient mixing ratios, BEIS under-estimated them. Warneke et al. (2010) derived emission rates of isoprene from ambient measurements and compared with MEGAN and BEIS estimations, and came to similar conclusions. Stavrakou et al. (2010) compared predicted isoprene concentrations in the eastern US from a number of difference isoprene oxidation schemes using MEGAN generated isoprene emission, and showed that all schemes significantly overpredicted isoprene concentrations. Park et al. (2010) evaluated the isoprene emission model against flux measurements at a Houston urban location and concluded that model output is satisfactory only when accurate local emitter density and leaf area are used in the model.

While these studies provided important evaluation of the emission models, they are usually carried out over densely forested areas with rather uniform vegetation types and coverage. Capability of the biogenic emission models in heterogeneous urban/suburban regions with less vegetation cover has not been satisfactorily evaluated. In addition, recent studies have pointed out that the anthropogenic contributions to isoprene concentrations in urban and rural environment can be significant. For example, Borbon et al. (2001) used principal component analysis on measured hydrocarbons in an urban region and concluded that motor vehicle contributions to isoprene were non-negligible in summer and became more significant in winter. Song et al. (2008) studied the differences between observed and CAMx-predicted concentrations of isoprene in Southeast Texas using GloBEIS, and concluded that under-predictions of anthropogenic emissions might be a reason for under-predictions of isoprene at urban locations. Park et al. (2011) summarized past findings and concluded from their urban isoprene flux measurements in Houston that traffic emissions could make non-negligible contributions to isoprene emissions, especially at night time and during early morning rush-hour. If anthropogenic emissions were a significant contributor to total isoprene concentrations, traditional observation and modeling based techniques, which use total isoprene concentrations from all sources, could not be directly used to evaluate the performance of the biogenic emission inventories in urban areas.

In this study, the capability of the most recent version of the MEGAN model (MEGAN2.1) in estimating isoprene emissions in Southeast Texas during a relatively wet summer ozone episode is evaluated using high temporal resolution isoprene concentration data collected at a number of urban sites as well as in the upper air near these urban locations. Uncertainties in the predicted isoprene concentrations due to emission factors, the distribution of plant functional types (PFTs) and leaf area index (LAI) are discussed.

2. Model description

The Community Multiscale Air Quality Model (CMAQ, version 4.7.1) (Byun and Schere, 2006; Carlton et al., 2010; Foley et al., 2010) was used as a framework to incorporate a source-oriented version of the SAPRC-99 gas phase photochemical mechanism (Carter, 2000) to directly predict isoprene concentrations due to biogenic and anthropogenic emissions. In the source-oriented CMAQ model, emissions, transport, gas phase chemistry, dry/wet deposition of isoprene from biogenic sources are tracked separately from isoprene emitted from anthropogenic sources using reactive source-tagged species. In summary, isoprene emissions from biogenic sources are represented in the model using ISOPRENE_X1 while isoprene emissions from anthropogenic sources are represented using ISOPRENE. Reactions of ISOPRENE_X1 are exactly the same as the reactions of the original ISOPRENE. For example, reaction set (R1) shows their reactions with hydroxyl radical (OH):

$$\begin{aligned}&\mathrm{ISOPRENE} + \mathrm{OH} \rightarrow 0.23\mathrm{MACR} + 0.32\mathrm{MVK} + \ldots\\&\mathrm{ISOPRENE\_X1} + \mathrm{OH} \rightarrow 0.23\mathrm{MACR\_X1} + 0.32\mathrm{MVK\_X1} + \ldots\end{aligned} \quad (\mathrm{R1})$$

Reaction products MACR and MVK also carry the same source-tag as their parent isoprene species. In this way, contributions of biogenic and anthropogenic sources to isoprene and its first generation oxidation products can be directly determined. The upwind isoprene entering the domain from model boundaries is lumped with the anthropogenic isoprene source category. Due to the short atmospheric lifetime, isoprene from upwind sources is not expected to travel long distance to affect concentrations at urban sites in the middle of the model domain.

While more detailed source-oriented SAPRC-99 mechanisms have been used in the past to track VOC sources (Ying and Krishnan, 2010; Zhang et al., 2013), the current version is greatly simplified to only track primary emissions of isoprene from two sources (biogenic and anthropogenic). This is sufficient for the purpose of this study to evaluate biogenic isoprene emission inventories and can greatly reduce simulation time and output file size. By separately tracking the isoprene from biogenic and anthropogenic emission sources, comparisons of observations with predictions can be made at times when predicted isoprene concentrations at monitoring sites are dominated by biogenic emissions. This allows a more strict evaluation of the performance of the underlying biogenic emission model. A similar approach was used in a previous study that evaluated vehicle emissions of CO and NO_x in Southeast Texas (Kota et al., 2014).

3. Model application

The source-oriented CMAQ model was applied to predict the isoprene concentrations during a three-week summer ozone episode in Southeast Texas during the 2006 Texas Air Quality Study