

A Study of Isoprene Emissions in Relation to Ozone Formation in the Eastern United States

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1. Introduction

Tropospheric ozone is one of gaseous species that is harmful to humane health, agriculture, and our environment. It is also a greenhouse gas that contributes to global warming. Isoprene is one of biogenic organic compounds that can enhance tropospheric ozone production under NO_x rich conditions. The relation of isoprene with ozone formation was identified by Chemidies et al. (1998) in the investigation of ozone pollution in Atlanta City. Since then, isoprene emission inventory has been developed and isoprene chemistry has been implemented in chemistry models. The BEIS (Biogenic Emission Inventory System) (Geron et al., 1994) in the United States has been developed since then. It is currently used by many chemistry models

The biogenic emission is emitted by forests and other plants. Oak tree, for example, can emit large amount of isoprene when temperature and radiation is high. The amount of emission depends on plant species, biomass of the plant, and environmental factor. In large scale modeling of biogenic emission, it is difficult to quantify the vegetation index. The LAI (leaf area index) of deciduous, evergreen forest, and agriculture in BEIS version 2 is assigned an empirical constant value based on the observations. A new method to determine LAI was developed by Gao (1995), Gao and Wesely (1995) using satellite observed reflectance and a simple radiative transfer algorithm. The satellite observations provide much more detailed information about earth surface conditions. The results showed very detailed spatial variation of LAI, and eliminated abrupt changes due to assigned constant values.

BEIS 2 was applied in OTAG (Ozone Transport Assessment Group) studies (OTAG,1997a and b). OTAG was one of the largest regional modeling activities performed over last two years. This was a policy oriented assessment of the importance of transport for reducing regional ozone over the eastern United States. The main concern is the regional ozone responses to NO_x vs. VOCs control strategies. OTAG results showed that regional ozone was responsive to NO_x controls during high concentration episodes. This work reran two days, July 7 and 8 1995, with high ozone episodes in OTAG, and replaced the isoprene emission from BEIS 2 with satellite data derived isoprene emission. The purpose is to evaluate the new isoprene emission model by comparison of ozone concentration with OTAG standard results. More importantly, this work also examines the isoprene emission in relation with ozone formation in the eastern U.S. Because emission and ozone transport are highly related to meteorology, the impact of meteorology has been studied too.

2. Description of Simulations

2.1 Derive Isoprene Emission Using Satellite Observations

Eastern United States is covered by the forests of Appalachian mountains, forests in Ozoark, and some agriculture in the midwest. Both forests contains certain amount of oaks, which are the large sources of isoprene. The plant genera distribution over the region was taken from OTAG with 12 km x 15 km resolution. It contains the percentage of 127 plant genera within each grid cell.

The amount of biogenic emission is also proportional to the LAI (leaf area index) of plants. The method using satellite observations to derived LAI was described by Gao (1995) and Gao and Wesely (1995). This method also included an simple empirical radiative transfer model to count solar irradiance that penetrated into the canopy, because the emission is affected by ambient temperature and light intensity as described by Geron et al. (1994),

$$F_I = F_i \frac{1.066\alpha I}{\sqrt{1 + \alpha I^2}} C_T \quad (1)$$

Here, F_i is the isoprene emission flux standardized to a leaf temperature at 30°C and a value of I_{PAR} , equal to 1000 $\mu\text{m}^2 \text{s}^{-1}$, and α is an empirical constant equal to 0.0027. The term C_T represents the leaf temperature adjustment factor, which can be expressed as

$$C_T = \frac{\exp[c_{T1}(T - T_s) / RT_s T]}{1 + \exp[c_{T2}(T - T_M) / RT_s T]} \quad (2)$$

Here R is the ideal gas constant (8.314 J K⁻¹ mol⁻¹), T (K) is leaf temperature, T_s is the standard temperature (303K), and T_M (314 K), and c_{T1} (95,000 J mol⁻¹) c_{T2} (230,000 J mol⁻¹) are empirical coefficients.

The total emission of isoprene from the canopy can be obtained by integrating through the canopy, with the total LAI represented by L_T :

$$F = \Lambda^{-1} \int_0^{L_T} F_C dL \quad (3)$$

Here F_C can represent either F_I from Eq. (4) or F_M from Eq. (6), and Λ is the leaf area index appropriate for the canopy at peak LAI. To evaluate the integral in Eq. (7), we assume that the temperature of the canopy is uniform at $T = T_c$. For isoprene, the result is as follows:

$$F = \frac{1.066 F_i C_T}{K \Lambda} \ln \frac{\alpha + \sqrt{\alpha^2 + I_0^{-2}}}{\alpha \exp(-KL_T) + \sqrt{\alpha^2 \exp(-2KL_T) + I_0^{-2}}} \quad (4)$$

where I_0 is the PAR incident at the top of the canopy and K is the extinction coefficient for PAR in the red band. Values of Λ are those assumed in BEIS2 for maximum LAIs (Pierce et al., 1998b), e.g., 5 for deciduous forest, 3 for pines, and 7 for other conifers when leaves are abundant in summer.

The surface reflectance information was obtained from a National Oceanic and Atmospheric Administration (NOAA)–National Aeronautics and Space Administration (NASA) Pathfinder Land Data set composed monthly NDVI (normalized difference vegetative index) data at a horizontal resolution of 8 km (Agbu and James, 1994). 1 km landuse USGS (United States Geological Survey) data (USGS,1990) were used in derive of LAI.

2.2 Apply Isoprene Emission in Regional Scale Ozone Simulation

Ozone concentration over Eastern United States was relatively high on July 7 and 8, 1995. Modeling activities were conducted to study the regional scale ozone transport and reposes to emission control strategies. The meteorological model used in OTAG was RAMS (Regional Atmospheric Modeling Systems) (Pielke et al., 1992), which drove the chemistry model (OTAG, 1997a) to simulate hourly ozone, NO_x, isoprene and other thirteen photochemical gaseous species near surface. The hourly VOC emission, including isoprene emission was estimated by BEIS 2. In this study, the isoprene emission derived from satellite data replaced the VOC emission estimated by BEIS2. MM5 (The Fifth Generation of Mesoscale Model) (Grell et al., 1994) was used to drive chemistry model and emission model.

Three cases (Table 1) were rerun to simulate ozone concentration in these two days in the OTAG project. Case 1 was a standard OTAG case with the combination of BEIS2 isoprene emissions and OTAG meteorological data generated by Regional Atmospheric Modeling System (RAMS). Case 2 replaced BEIS2-derived isoprene emission with satellite-derived emission, with the intention of examining the effects of isoprene emission on ozone formation. Case 3 represented the current modeling approach of using the biogenic emission model together with MM5 simulations.

Table 1. The emission and meteorological sources for rerun and comparison of OTAG case in July 7 and 8, 1995.

Case 1	Case 2	Case 3
BEIS2 (OTAG data)	Isoprene emission derived from satellite	Isoprene emission derived from satellite

	data	data
RAMS (OTAG data)	RAMS (OTAG data)	MM5

Two domains were selected. The large domain, with 36 km by 36 km resolution, was over Eastern United States, and the small domain, 12 km by 12 km each grid cell, nested in the large domain (Fig. 2) over high ozone concentration area. MM5 was run with resolutions corresponding to both of domains. It provided basic meteorological fields. The radiation and temperature fields were coupled with biogenic emission model using satellite data to estimate hourly isoprene emission.

3. Results and Discussion

3.1 Comparison with Harvard Forest Data

Simulations of isoprene emission rates were compared to measurements at Harvard Forest, a 50-to-70-year-old deciduous forest located in Massachusetts (42°32'N, 72°11'W) (Goldstein et al., 1998). The isoprene emission rates were reported as 45-min averages found by application of the flux-gradient method to measurements of mean concentration differences above the forest. For the simulations, observations were used of PAR at a height of 29 m above the ground (about 9 m above the top of the canopy) and of air temperature at a height of 22.6 m above the ground. In Eq. (4), the observed value of $L_T = 3.5$ for midsummer at Harvard Forest and the BEIS2 assumed value of $\Lambda = 5$ for deciduous forest were used. Then the total isoprene flux was calculated as the sum of the fluxes from the prevalent tree species weighted by their relative coverages: oak (mostly red), 36.4%; red maple, 21.8%; red pine, 16.2%; hemlock, 15.6%; birch, 3.5%; white pine, 2.9%, and cherry, 2.3% (Goldstein et al., 1998).

Goldstein et al. (1998) illustrated several well-known features of isoprene emission. During the summer, the rates peaked near mid-day and decreased to extremely small values at night. To compare measured rates with simulated rates, the data for July, August, and September of 1995 were arranged as weekly 4-hour averages for three periods per day: 0600-1000 hours Eastern Standard Time (EST), 1000-1400 hours, and 1400 to 1800 hours. As is shown in Fig 1, the modeled emissions matched the measurements fairly well for the morning and midday periods, but tended to be too small for the weeks with relatively large emission rates. Goldstein et al. (1998) had also found that the peaks tended to be underestimated by BEIS2 and suggested that the emission factor in BEIS2 for oak trees of $70 \mu\text{g C h}^{-1}$ per gram of foliar dry mass (corresponding to $29750 \mu\text{g isoprene m}^{-2} \text{h}^{-1}$ and $437.5 \mu\text{mol m}^{-2} \text{h}^{-1}$) is too low. Over the past

several years, measured or inferred emission factors of $100 \mu\text{g C g}^{-1} \text{ h}^{-1}$ and larger have been reported for oak (Guenther et al., 1996a, b). The rate used in the current model for additional simulations is $86 \mu\text{g C g}^{-1} \text{ h}^{-1}$, near the middle of the range of the reported values as suggested by Geron et al. (2000). Other factors that could have contributed to the underestimation of the emission rates in the simulation for Harvard Forest are that emissions were measured at point sources while the percentage of plant composition are local averaged, and that air temperature rather than canopy temperature was used in Eq. (4). For the afternoon, in contrast to the cases for morning and midday, the simulated values shown in Fig. 1 are consistently too large. This discrepancy could have been caused by a suppression of leaf-air exchange through leaf stomata during the small relative humidities at the leaf surface that typically occur during afternoons, an effect that is not currently considered in the parameterizations of isoprene emissions.

3.2 Ozone Concentration over Eastern United States

Simulations for all the three cases were carried out for 24-hour periods on 7 and 8 July. Fig. 2 shows an example of Cases 1 and 2 results on surface isoprene emissions and O_3 concentrations in the lowest 50-m layer of the atmosphere at 1500 hours EST for 8 July 1995. The simulated isoprene emission rates generated in Case 2 (Fig. 2b) with the present biogenic emissions scheme using satellite data tend to be larger than those from Case 1 (Fig. 2a) in high density forest areas, lower in coast areas. In northern portion of the domain, such in West Virginia, Kentucky in Case 2 (Fig. 2d) has correspondingly slightly higher concentrations of O_3 than Case 1 (Fig. 2c). The peaks in O_3 concentration seen in the northeastern U.S. appear to correspond to the higher levels of isoprene emission simulated with Case 2. Above the mixed forests of the southern Appalachian Mountains (e.g., Mississippi, Alabama, Tennessee) and parts of the forested areas in the Ozarks (in Missouri and Arkansas), however, a significant increase O_3 concentration is not seen for Case 2 despite local increases by factor of about four in isoprene emissions. This situation suggests that the simulated production of O_3 tend to be limited by relatively low concentrations of NO_x in the southern portions of the domain.

Simulations for Case 3 were carried out with the emissions generated by current biogenic emissions model using satellite remote sensing data with MM5 meteorological outputs. Fig. 3 shows the ozone concentration on July 7 at 12:00, July 8 at 16:00 in Case 1 and 3. Compare Fig. 3c with Fig. 3d, it shows that ozone concentration pattern is very similar. No noticeable changes in ozone concentration can be observed in northeastern U.S, except near large NO_x emission sources. In the snortheastern of U.S., ozone concentration in Case 3 was slightly higher than that in Case1. Also, Case 3 did not show the high ozone concentration along northeastern coast area. The differences in the spatial pattern seen in Case 3 versus Case 1 (Figs. 3c and 3d) are similar to those seen in Case 2 versus Case 1 (Figs. 2c and 2d), and thus can be attributed to the differences in simulated isoprene emission rates. For 8 July, the peak O_3 concentration in Case 3 was 115 ppb, very close to the value of 116 ppb in Case 1, but the peak in Case 3 occurred in Tennessee about two hour later than the Case 1 peak, which was located in North Carolina.

Fig. 3a and 3b are the ozone concentration spatial distribution on July 7 in Case 1 and Case 3. Large differences can be seen in the concentration as well as peak ozone concentrations. The peak O₃ concentration in the domain was about 103 ppb in Case 1 vs. 176 ppb in Case 3 on July 7. This is mostly caused by the meteorology fields on July 7. FDDA (four dimensional data assimilation) was incorporated in the simulation of MM5 for 8 July, not for 7 July. This indicates that meteorology has significant impact on regional ozone transport.

4. Summary

Isoprene emission in the eastern United States was estimated by a biogenic emission model. The algorithm for isoprene emission is based on the integration of leaf-level isoprene emission with respect to whole-canopy leaf area index. The unique feature of this algorithm is the use of satellite-observed reflectance to derive the leaf area index, which results in considerable detail in the spatial patterns. To evaluate model results, a comparison was made to observations at Harvard Forest. The model-estimated emission rates matched observations reasonably well. Further studies were conducted by simulation of ozone concentrations over eastern United States for an Ozone Transport Assessment Group (OTAG) case by using two methods of estimating isoprene emissions: a standard application of Biogenic Emissions Inventory System version 2 (BEIS2) and an application using remote sensing data from satellite. The Mesoscale Meteorological Model version 5 (MM5) was coupled with the dry deposition and biogenic emission model to generate emissions for July 7 and 8, 1995, days with significant ozone episodes. The satellite-derived isoprene emission derived by using meteorological data from MM5 tended to be higher than from results BEIS2 in the southwestern part of the domain but slightly lower along the East Coast. To examine the effect of uncertainties in isoprene emission on ozone formation in this region, three cases were examined. Case 1 was a standard OTAG case with the combination of BEIS2 isoprene emissions and OTAG meteorological data generated by Regional Atmospheric Modeling System (RAMS). Case 2 replaced BEIS2-derived isoprene emission with satellite-derived emission, with the intent of examining the effects of isoprene emission on ozone formation. Case 3 represented the current modeling approach of using the dry deposition and biogenic emission model together with MM5 simulations. The results for Case 2 showed that isoprene emissions affected ozone concentration in the northeastern United States, but had no noticeable impact in the southeastern United States except near large NO_x point sources. A comparison of Case 3 and Case 1 indicated that the pattern of ozone concentration derived by using the current modeling approach of isoprene emission simulation and MM5-generated meteorological fields was very similar to the Case 1 OTAG standard results involving BEIS2 and RAMS meteorological data. The maximum value of ozone concentration was 115 ppb for Case 3 versus 116 ppb for Case 1 on July 8, 1995. The meteorological fields, however, had very strong effects on spatial patterns of ozone concentrations. For example, the maximum value of ozone concentration was as large as 176 ppb when the MM5 simulations were made without four-dimension data assimilation, versus 116 ppb for the OTAG case on July 7, 1995.

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CD-ROM, EROS Data Center, U. S. Geological Survey, National Mapping Division, Sioux Falls, South Dakota; and Center for Advanced Land Management Information Technology, Conservation and Survey Division, University of Nebraska, Lincoln (updated version available on the Web at http://edcdaac.usgs.gov/glcc/nadoc1_2.html#usgs)