SPATIAL VARIATIONS IN BIOGENIC EMISSIONS FOR LITHUANIA

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Abstract: The numerical modelling of biogenic volatile organic compounds (BVOCs) monoterpene and isoprene was carried out using three-dimensional (3D) mesoscale meteorological and photochemical atmospheric models. Emission factors, combined with land cover data represented by the appropriate 11 Biogenic Emission Inventory System (BEIS) vegetation categories, along with environmental correction factors were used to derive emission fluxes of isoprene, monoterpene and other VOCs for Lithuania. Moreover, simulated data have been combined with BEIS data using a Geographic Information System (GIS) to appropriately represent the spatial distribution of BVOCs. Calculations showed that the coniferous trees are the main sources of biogenic emissions. The highest emission fluxes of biogenic VOC are estimated to be in the region of the south-south eastern Lithuania, which has the largest forest coverage in Lithuania and the major part of these forests consist of coniferous forests. The total simulated isoprene emission flux from Lithuania (65281 km^2) reached 12710 kg h^{-1} in June and 18280 kg h^{-1} in July (approximately 34 % from coniferous forests). On the other hand, monoterpene average emission flux from Lithuania was found to be 4080 kg h^{-1} in June and 5330 kg h^{-1} in July (approximately 50% from coniferous forests).

Key words: *biogenic volatile organic compounds, emission, isoprene, monoterpene, numerical modelling, MM5 model.*

1. INTRODUCTION

Biogenic volatile organic compounds (BVOCs) play a prominent role in the chemistry of the atmosphere. Biogenic emissions of the volatile organic compounds isoprene and monoterpenes (BVOCs) can contribute to tropospheric ozone and secondary particle formation and have indirect effects on climate change. BVOCs is a rather loose term for a wide range of compounds, of which only a few are generally of great interest because they are highly reactive and thus may profoundly influence the chemical and physical properties of the atmosphere (Atkinson R. and Arey J., 2003). The determination of BVOC emission is therefore of utmost importance in regional air chemistry modelling. The most current global BVOC emission inventories are based on model results (Guenther et al., 1995, 2006). Using models, it has been estimated that globally the principal natural sources of organic compounds are woods, crops, and shrubs with a combined flux in the range of 1135 TgC $year^{-1}$, with the majority of this flux attributed to isoprene (Guenther et al., 1995). Isoprene is a highly reactive organic compound having an atmospheric lifetime of a few minutes to a few hours (Otter et al., 2002). The major sources of isoprene are emissions from deciduous trees, thus over forested areas isoprene mixing ratios can rise up to several ppbv (Apel et al., 2001). This emphasizes the importance of biogenic emissions, and inventories of BVOC emissions are thus a key issue in atmospheric sciences. During the recent decade there have been many studies aimed at quantifying BVOC emissions from forest vegetation (Kuhn et al., 2002). Studies of BVOCs have mostly been focused on forests, which are thought to be their dominant global source. The total forestland area covers 31.7 % of Lithuania. During the last 6 years it increased by 1.4 %. Coniferous stands prevail in Lithuania (58.9%) followed by soft-broadleaves (36.3%) and hard-broadleaves (4.8%).

The results presented here provide some insight into the diurnal temperature and species-specific emissions (isoprene and monoterpenes) for plant species commonly found in Lithuania.

2. METHODOLOGY

Model description

A biogenic emissions inventory for the photochemistry domain was derived, using the Biogenic Emissions Inventory System (BEIS), developed at the Environmental Protection Agency (EPA) of the USA (Pierce, 2001). The Biogenic Emissions Inventory System estimates biogenic emissions that are the result of biological activity from land-based vegetative species and nitric oxide emissions which are the result of microbial activity from certain soil types as a function of temperature, sunlight, and vegetation type. The MM5 meteorological model was applied to generate the meteorological fields used with the BEIS3 mode, Biogenic Emissions Land cover Database version 3 (BELD3) land cover database, normalized emissions profiles, and leaf area indexes (LAI) for each land type. The emission fluxes were normalized to standard environmental conditions, i. e. 30^{o}C and 1000 μmol m^{-2} s^{-1} photosynthetically active radiation (PAR). Since BELD3 database is available only for USA and Canada, the differences in the uncertainty of application can be assessed. Biogenic emission modelling system – BioModel –based on BEIS3 temporal emission computation algorithms was developed for application with the Lithuanian emission inventory (Fig. 1). The algorithms and databases used to estimate the BVOC emissions were incorporated into a geographic information system (GIS) software.

BioModel implements OpenGIS specified grid coverage interfaces, involves many GIS typical low-level procedures written in assembly language and using processor multimedia technologies (MMX, SSE), and uses a graphic subsystem to visualize spatial emission data. BioModel consists of inputs to CORINE land cover database, providing information on the different Lithuania land cover data in Geographic Tagged Image File Format (GeoTIFF), MM5

meteorology data in netCDF format and produces spatial-temporal biogenic emissions data, from where the data are directly extracted as standard GRID fields, afterwards statistically analysed and stored in the netCDF format.

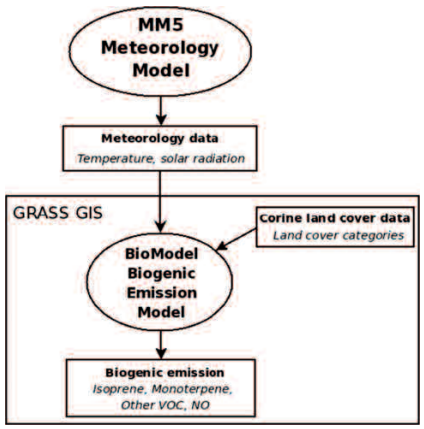

Figure 1. Diagram of biogenic emission modelling system.

Static normalized biogenic emission profiles and leaf area index per land use category, obtained from BEIS3 data, are contained in executable module. BioModel consists of input file in MODELS3 air quality modelling system. Geographic Resources Analysis Support System (GRASS), a raster- and vector-based Geographic Information System image processing system module r.bio.emis, calculating isoprene, monoterpene and other volatile organic compounds, was additionally developed. Module r.bio.emis was formatted to input into GRASS software system and supports native GRASS spatial data format. Thus, spatial data can be imported into GRASS geospatial database from various formats and exported to various formats supported by GRASS.

The isoprene emission flux mostly depends on photosynthetically active radiation and environment temperature. BioModel involves a complex canopy model to split photosynthetically active radiation to direct and diffuse beams and compute fractions of leaves that are sunlit and shaded according to pressure, solar radiation, sun altitude, and leaf area index, and finally uses Guenther A. B. et al. (1993) equation to estimate the light correction factor for isoprene emission flux computation taken from BEIS3.

Light and temperature controls on emissions

Biogenic emission is widely spread and has impact on background atmosphere chemical processes. BVOC emission was found to vary from one species to another. For region-scale air quality modelling, biogenic emission researches are concentrated on volatile organic compounds from vegetation and nitrogen oxides from soil. Temperature regulates isoprene (Monson et al., 1992) and monoterpene (Fischbach et al., 2001) synthase activities, thus controlling the synthesis of most isoprenoids. According to Guenther et al. (1997), there are four major factors controlling natural BVOC emissions: landscape average (species-specific) emission potential ε ($\mu g\ g^{-1}\ h^{-1}$), foliar biomass density D (gm^{-2} d.w. - dry weight), an environmental correction factor γ (nondimensional). Emission fluxes ($\mu g\ m^{-2}\ h^{-1}$) can then be modelled by

$$F = \varepsilon D \gamma. \qquad (1)$$

The emission potential and the foliar biomass density are species-specific properties, and they should be assessed individually for each tree genera and subspecies. A non-dimensional environmental correction factor includes the effect of temperature and light conditions.

For appropriate modelling, in-canopy profiles of temperature and radiation must be considered. The emission potential ε accounts for genetic controls of isoprene and monoterpene production. It represents the emission rate per unit foliar mass for a specific plant species normalized to leaf surface temperature $Ts = 30^0$C and photosynthetically active radiation PAR = 1000 $\mu mol\ m^{-2}\ s^{-1}$.

The environmental correction factor γ describes the diurnal variation of the biogenic VOC emissions. Several numerical algorithms have been developed to simulate the effect of light and temperature on isoprene and monoterpene emissions (Guenther et al., 1993, 1991; Lamb et al., 1993, 1996). In this work we have adopted the algorithms proposed by Guenther et al. (1993), which have been shown to perform extremely well when applied to different vegetation types and environmental conditions. According to this approach, the terpene emissions are controlled by the volatilisation of hydrocarbons from storage pools inside the leaf (temperature control), while isoprene is emitted directly after it has been synthesized by the plant (light and temperature control).

Environmental correction factor for isoprene and monoterpene

Isoprene is not stored in plants and emitted in sunlight during photosynthesis. However, monoterpenes can be stored in plants; hence, they are emitted both during day and night. Basically, one has to distinguish between terpenes, which after biosynthesis, will be emitted from leaves more or less immediately and will be stored in corresponding pools prior to emission. If the inner leaf is the location of actual production or storage pools, then the release of terpenes is most likely via leaf stomata. The environmental correction factor for isoprene emissions is thus:

$$\gamma_{ISO} = C_T C_L, \tag{2}$$

here C_T is the temperature correction and C_L is the light correction.
The light correction has the form:

$$C_L = \frac{\alpha C_{L1} L}{\sqrt{1+\alpha^2 L^2}}, \tag{3}$$

here L is the photosynthetically active photon flux density (PPFD), μmol photons m^{-2} s^{-1}, α = 0.0027 and C_{L1} = 1.066 are empirical coefficients (Guenther, 1997).
The temperature correction is given by Guenther et al. (1993):

$$C_T = \frac{\exp(\frac{C_{T1}(T-T_S)}{RT_S T}}{C_{T3} + \exp(\frac{C_{T2}(T-T_M)}{RT_S T}}, \tag{4}$$

here T is the leaf temperature in K, T_S *is* the leaf temperature under standard conditions (303.15 K), R is the universal gas constant, C_{T1} = 95 000 J mol^{-1}, C_{T2} = 230 000 J mol^{-1}, C_{T3} = 0.961, and T_M = 314 K are empirical coefficients given by Guenther (1997).

The environmental correction for monoterpene emissions is:

$$\gamma_{TERP} = \exp(\beta(T - T_S)), \tag{5}$$

here β = 0.09 C^{-1} is an empirical coefficient.

This correction factor is also generally used for other VOCs (Oxygenated volatile organic compounds (OVOCs)), because experimental data on the OVOC emissions are still too scarce to facilitate the development of specific emission algorithms (Guenther, 1994, Simpson, 1999).

3. RESULTS AND DISCUSSION

Emission flux of monoterpene, isoprene and other VOCs mostly depends on photosynthetically active radiation and environment temperature. The highest biogenic emission fluxes in Lithuania were expected during warm season, therefore biogenic emission for June-July 2004 was simulated. Biogenic emission (monoterpene, isoprene, other VOCs) fluxes attributable to 11 land use categories in Lithuania were simulated using the above model, parameterized and modified using the model input values of the database of biomass, emission flux factors, temperature, and PAR. The model results provide us the emissions for June-July months of the year 2004. The examples of model results of simulated species monthly emissions for June are given in Fig. 2 (a, b, c).

The total simulated isoprene emission flux from Lithuania (65281 km^2) reached 12714 kg h^{-1} in June and 18282 kgh^{-1} in July (approximately 34% from conifer forests). The highest emissions occur in the forested eastern and southern parts of the country with peak emissions of over 1096 g $km^{-2}h^{-1}$. The highest isoprene emission fluxes during June (Fig. 2a) – July were produced by conifer forests – 753 and 521 g km^{-2} h^{-1}, respectively. Conifer forests take area of 7248 km^2 (11 % of Lithuania area) and appear to be the most important isoprene emitters. Other high flux isoprene emission producers were mixed and deciduous forest, which cover 7196 km^2 (11 %) and 4138 km^2 (6.3 %) area of Lithuania and average emission fluxes were estimated 368 g $km^{-2}h^{-1}$ and 346 g $km^{-2}h^{-1}$ (June), and 528 g $km^{-2}h^{-1}$ and 498 g $km^{-2}h^{-1}$ (July), respectively. In the southeastern part of Lithuania where a high share of all type forests exists isoprene emissions dominate, as well in the same part monoterpene emissions prevail as a result of conifer forests.

Thus, all areas in the southern and eastern parts of the country are strong monoterpene emitters with peak emissions of over 559 g $km^{-2}h^{-1}$. The highest monoterpene emission fluxes were produced by conifer forests: average 276 g $km^{-2}h^{-1}$ in June (Fig. 2b) and 361 g $km^{-2}h^{-1}$ in July. The total simulated monoterpene average emission flux from Lithuania was found to be 4083 kg h^{-1} in June and 5330 kg h^{-1} in July (approximately 50 % from conifer forests). Although conifer forests produced highest OVOC emission fluxes (average 115 g $km^{-2}h^{-1}$ in June (Fig. 2c) and 150 g $km^{-2}h^{-1}$ in July), the total highest average emission flux was produced by dry crop, which takes 22233 km^2 (34 %) area in Lithuania, i.e 1938 kg h^{-1} in June and 2506 kg h^{-1} in July. Total average OVOC emission flux from Lithuania was 5755 kg h^{-1} in June and 7484 kg h^{-1} in July (approximately 33 % from dry crops).

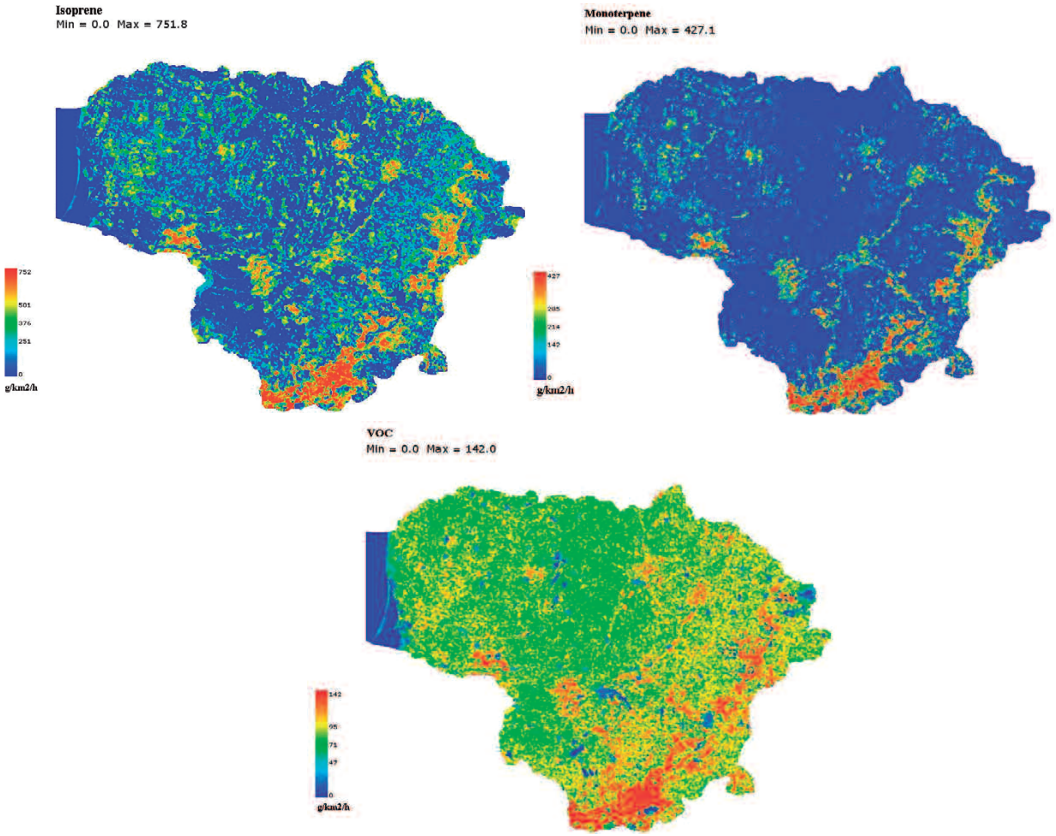

Figure 2. Biogenic emission distribution in Lithuania for June 2004: a) isoprene, b) monoterpene, c) VOCs.

The impact of light intensity and of temperature on isoprene emissions were examined. The variation of isoprene emissions closely follows the temperature variability, and daily emissions reflect the great sensitivity of the isoprene to the environmental conditions (Eqs. 3 and 4). Isoprene levels demonstrated an exponential increase with the temperature. Isoprene emission is also strongly dependent on temperature, increasing exponentially up to a maximum emission at approximately 24º C (June) and 27º C (July). Regression analysis revealed significant correlations at the 99% confidence level between temperature and isoprene (r=0.78, N=1412, June), (r=0.84, N=1466, July).

4. CONCLUSIONS

The highest isoprene emission fluxes during June–July were produced by conifer forests – 521 and 753 $gkm^{-2}h^{-1}$, respectively. Conifer forests take area of 7248 km^2 (11% of Lithuania area) and appear to be the most important isoprene emitters. Other high flux isoprene emission producers were mixed and deciduous forest, which cover 7196 km^2 (11%) and 4138 km^2 (6.3%) area of Lithuania and average emission fluxes were estimated 368 $gkm^{-2}h^{-1}$ and 346 $gkm^{-2}h^{-1}$ (June), and 528 $gkm^{-2}h^{-1}$ and 498 $gkm^{-2}h^{-1}$ (July), respectively. The highest monoterpene emission fluxes were produced by conifer forests: average 276 $gkm^{-2}h^{-1}$ in June and 361 $gkm^{-2}h^{-1}$ in July.

The temperature dependence of isoprene emissions is reflected in diurnal emissions patterns, as well as solar radiation. Isoprene levels demonstrated an exponential increase with the temperature.

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