Article

Estimating the biogenic non-methane hydrocarbon emissions over Greece.

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Abstract: Biogenic emissions affect the urban air quality as they are ozone and SOA precursors and should be taken into account when applying photochemical pollution models. The present study presents an estimation of the magnitude of Non-Methane Volatile Organic Compounds emissions (NMVOCs) emitted by vegetation over Greece. The methodology is based on computation performed with the aid of a Geographic Information System (GIS) and theoretical equations in order to develop an emission inventory on a 6x6 km^2 spatial resolution, in a temporal resolution of 1hr covering one year (2016). For this purpose, a variety of input data was used: improved satellite land-use data, land-use specific emission potentials, foliar biomass densities, temperature and solar radiation data. Hourly, daily and annual isoprene, monoterpenes and other volatile organic compounds (OVOCs) were estimated. In the area under study, the annual biogenic emissions were estimated up to 472 kt, consisting of 46.6% isoprene, 28% monoterpenes and 25.4% OVOCs. Results delineate an annual cycle with increasing values from March to April, while maximum emissions were observed from May to September, followed by a decrease from October to January.

Keywords: Biogenic emissions; Greece; Geographic Information System (GIS)

1. Introduction

Volatile organic compounds (VOC) are emitted into the atmosphere from natural sources in marine and terrestrial environment [1]. As a matter of fact, globally, biogenic sources of volatile organic compounds are estimated to exceed those from anthropogenic sources by a factor of ten to one [2]. More specifically in Europe, anthropogenic and biogenic NMVOCs emissions have comparable magnitudes: annual biogenic NMVOCs emissions are estimated at 14 Tg compared to man-made emissions of around 24 Tg [2].

A great number of VOCs are emitted from vegetation with isoprene (C_5H_8) and monoterpenes ($C_{10}H_x$) being the most abundant species. The remaining biogenic emitted species consist of a number of oxygenated compounds, such as alcohols and aldehydes and they are referred to as other VOCs (OVOCs) [1].

The calculation of their fluxes is an important input in air quality models, since they are highly reactive in the troposphere by affecting regional photochemical processes [3]. They react with the hydroxyl radical, ozone and the nitrate radical, resulting in the formation of carbon monoxide and organic species (including secondary organic aerosols) that can enhance concentrations of ozone and other oxidants in environments rich in nitrogen oxides [4]. On a global cycle, Biogenic Volatile

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Organic Compounds (BVOCs) contribute to the global carbon cycle and have a key role in the global climate [1]. Furthermore, most of them are oxidized to carbon dioxide (CO_2) into the atmosphere and determine the growth rate of atmospheric methane concentrations [1].

The BVOCs emissions depend on the different types of vegetation and meteorological conditions. Concerning the isoprene, it has been shown that it is emitted mainly from deciduous trees under high temperature and Photosynthetically Active Radiation (PAR) conditions [1]. On the other hand, monoterpenes are emitted mostly by coniferous trees. They are mainly temperature dependent except for some evergreen oaks and Norway spruce that are temperature and light dependent [5].

Presently in Greece there does not exist a detailed gridded database with recent data concerning the biogenic emissions even though it is well known that BVOCs emissions play a significant role in the creation of photochemical pollution, especially during the warm months. In addition, this region, similarly to the Mediterranean, should be studied in detail with regard to biogenic emissions due to the specificities it presents. More precisely, it has a complex vegetal biodiversity quite different from the usual northern latitude or US vegetation, it receives high fluxes of solar radiation in the summertime and is dominated by high temperatures. Finally, high ozone concentrations are often very pronounced due to primary pollutant emissions in a regional scale dominated by high radiation fluxes and temperature values [6]. The most recent studies concerning the estimation of biogenic emissions in Europe and the Balkan Peninsula, including Greece were performed by [7,8] with the aid of emission models or a Geographic Information System (GIS). Both studies referred to 2003, with outdated land use data and adopted modeled meteorological input information

The present work aims to present results of the computational system developed for estimating BVOCs emissions based on GIS technology over Greece. It covers the year 2016 and has the possibility to be regularly updated to include more years. The paper is organized as follows: in Section 2 the methodology used (the mathematical and computational model) for the estimation of the biogenic emissions is introduced. Section 3 focuses on the results of our study, presenting the spatially resolved isoprene, monoterpenes and OVOCs biogenic emissions as well as a short discussion concerning the results. A summary and conclusions are provided in Section 4.

2. Methodology

2.1. The mathematical model

In the present study, the mathematical model for estimating isoprene, monoterpenes and OVOCs emissions in Greece was incorporated into the GIS platform. The mathematical model used for all types of vegetation, describing the emissions flux on an hourly basis is that of [9]:

$$\text{Flux}\left(\mu g\ m^{-2} yr^{-1}\right)=\int \varepsilon . D . \gamma\ dt \tag{1}$$

where ε is the emission potential ($\mu g\ g^{-1} h^{-1}$) for any particular species, D is the foliar biomass density (g dry weight foliage m^{-2}), and γ is a unit less environmental correction factor representing the effects of short-term (e.g. hourly) temperature and solar radiation changes on emissions.

Concerning the estimation of the isoprene emissions, [10] showed that, to a very good approximation, the short-term (e.g. hourly) variations in emissions could be described by the product of a light-dependent factor and a temperature-dependent factor. So, the environmental correction factor for the isoprene emission is expressed as:

$$\gamma_{iso}=C_L . C_T \tag{2}$$

The light-dependent factor is given by:

$$C_{L_{iso}}=\frac{a\ C_{L_1} L}{\sqrt{1+a^2 L^2}} \tag{3}$$

where a=0.0027 and C_{L_1}=1.066 are empirical constants, and L is the PAR flux (μmol photons (400-700nm)$m^{-2}s^{-1}$).

The temperature-dependent factor is given by:

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$$C_{T_{iso}}=\frac{\exp\left(\frac{C_{T_1}(T-T_s)}{R\,T_s\,T}\right)}{1+\exp\left(\frac{C_{T_2}(T-T_M)}{R\,T_s\,T}\right)} \quad (4)$$

where R is the gas constant (=8.314 J K^{-1} mol^{-1}), and C_{T_1}(=95000 J mol^{-1}), C_{T_2}=(230000 J mol^{-1}), and T_M(=314 K) are empirical coefficients based upon measurements of three plant species: eucalyptus, aspen, and velvet bean, but which seem to be valid for a variety of different plant species [10] and finally, T_s(=303K) is the standard temperature.

Concerning the estimation of the monoterpene emissions, the environmental correction factor suitable for most of the plants is parameterized using the following equation [10]:

$$\gamma_{mts}=\exp\left(\beta(T-T_s)\right) \quad (5)$$

where β(=0.09 K^{-1}) is an empirical coefficient based on non-linear regression analysis of numerous measurements present in the literature.

Recent studies, proved that monoterpene emissions from some evergreen oaks, and also Norway spruce show a light-dependency, which seems to be well described by the isoprene environmental correction factor [5].

Since the environmental conditions controlling emissions of OVOCs are not entirely understood compared to isoprene and monoterpenes and given the lack of other information, OVOCs emissions are considered temperature dependent and the use of Equation (5) is recommended for the estimation of their emissions [11].

2.2 The computational model

In order to produce the NMVOCs emission inventory for Greece on a 6x6 km^2 spatial and a 1hr temporal resolution covering one year, the GIS software (ArcView v10) was used in order to combine a variety of input data: improved satellite land-use data, land-use specific emission potentials, foliar biomass densities, temperature and solar radiation data. For the calculation of the hourly biogenic emissions, detailed meteorological data for the time period of a whole year (2016) were used. After calculating the hourly emission fluxes, daily, monthly and yearly emission values were also estimated.

The land use/ land cover (LULC) data used in the present study was provided by the United States Geological Survey (USGS) Global LULC version 2.0 Database derived from the 1 km Advanced Very High Resolution Radiometer (AVHRR) data spanning April 1992- March 1993. The USGS classification system includes 25 land cover categories, only 14 of which are found in the area under study (Figure 1). The different land use classes emitting BVOC are characterized by one ecosystem type (e.g. Grassland) or a combination of two of them (e.g. Mixed Scrubland/Grassland). The area was divided into cells using a spatial resolution of 6x6 km^2 with Lambert Conic Conformal projection. Each cell was checked separately and correction of the LULC category was made if necessary based on the work done by [6,12].

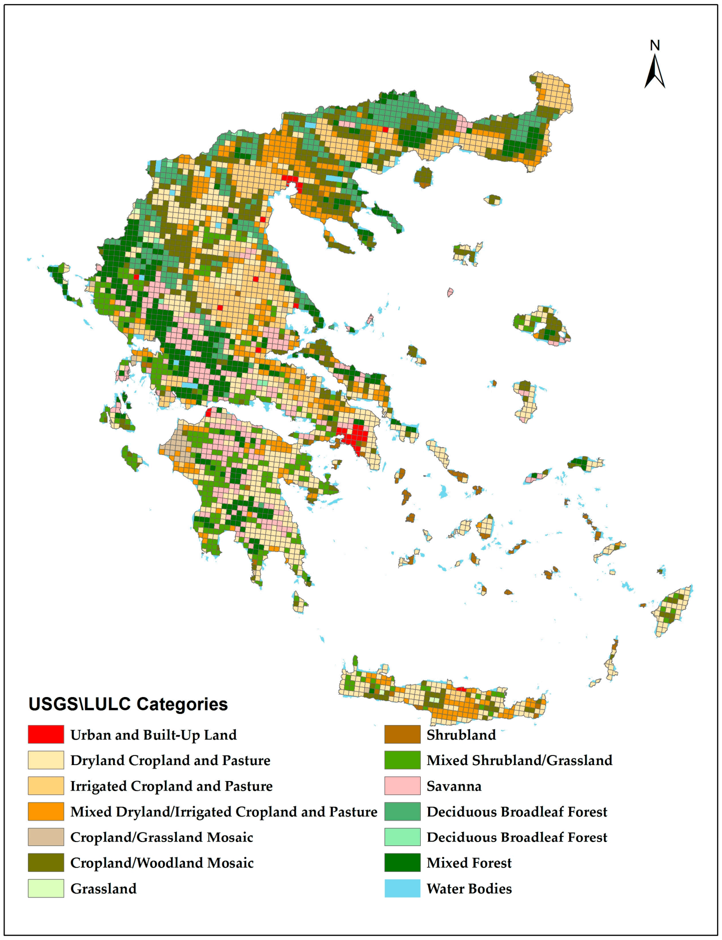

Figure 1. Greece with the land use categories attributed at each cell.

The employment of land-use specific emission potentials and foliar biomass densities for every month covering the whole year is essential for the estimation of isoprene, monoterpenes and OVOCs. The main references used for the selection of these values were from the recent study of [8] under the NatAir program (Improving and Applying Methods for the Calculation of Natural and Biogenic Emissions and Assessment of Impacts on Air Quality) for the region of Europe and the neighboring ones. According to this study, foliar biomass densities and emission potentials are assigned to commonly observed European vegetation species. Furthermore, the fact that the foliar biomass densities are not constant during the year was taken into account. In order to describe the seasonal variation of the foliar biomass densities, it was necessary to use corrective factors which vary between the different vegetation species according to the study of [7]. So, appropriate monthly foliar biomass densities were assigned to each land-use category (Table 1). When a land-use class was characterized by a combination of different vegetation species, it was assumed that the monthly average foliar biomass density is equal to the mean value of the foliar biomass densities of all vegetation types within the land use category [7]. Finally, the specific emission potentials for the land use classes that are a combination of different vegetation types were calculated using the formula:

$$\varepsilon = \frac{\sum(\frac{\varepsilon_i D_i}{n})}{\sum(\frac{D_i}{n})} \qquad (6)$$

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where ε_i and D_i are the emission potentials and the foliar biomass densities of each vegetation type within the land use category and n is the number of vegetation types within the land use category [7].

Finally, for the OVOCs, due to lack of reliable experimental data on their emissions, [7] recommended the use of the uniform emission rate of 1.5 μg $g^{-1}h^{-1}$ for all tree species.

Table 1. Foliar biomass densities (g dry weight foliage m^{-2}) and emission potentials (μg $g^{-1}h^{-1}$) for the observed land use categories in Greece for the month of July.

Land Use Category	Foliar Biomass Density	Isoprene Emission Potential	Monoterpene Emission Potential
Urban and Built-Up Land	100	2	1
Dryland Cropland and Pasture	100	0.5	0.5
Irrigated Cropland and Pasture	300	0.5	0.5
Mixed Dryland/Irrigated Cropland and Pasture	325	1.85	1.56
Cropland/Grassland Mosaic	175	0.5	0.5
Cropland/Woodland Mosaic	200	1.63	1.63
Grassland	50	0.5	0.5
Shrubland	350	3	2.5
Mixed Shrubland/Grassland	200	2.69	2.25
Savanna	75	3.5	3.5
Deciduous Broadleaf Forest	340	30	0.5
Evergreen Needleleaf Forest	700	1	2.5
Mixed Forest	500	7	3
Water Bodies	0	0	0

Hourly temperature values were provided by the National Observatory of Athens (www.meteo.gr) from 292 meteorological stations for 2016. The typical temperature diurnal variation for all the stations was produced by calculating the average hourly temperature values of each month of the year. Hourly temperature maps were constructed using the technique of Inverse Distance Interpolation (IDW), thus providing a continuous temperature field covering the area under study.

The solar radiation data were estimated with the aid of a new research project for the development of SOLar Energy Applications (SOLEA, www.solea.gr) [13]. It is based on solar irradiance spectra produced via a synergy of neural networks and radiative transfer simulations. The Photosynthetically Active Radiation (PAR) was calculated for every month of 2016 having 0.05^o latitude by 0.05^o longitude spatial resolution and 1hr temporal resolution. Then, with the aid of GIS, these radiation values were adjusted to the area of interest with a spatial resolution of 6x6 km^2.

Initially, the hourly biogenic emissions were calculated for a typical day of every month of 2016. Then, the monthly biogenic emissions were estimated by summing up the daily emissions of isoprene, monoterpenes and OVOCs per month in Greece.

3. Results

The temperature and light dependency of the biogenic emissions determine their magnitude. This leads to an increase of BVOC emissions during the daytime, with observed maximum values at midday. For that reason, a cell was selected where high biogenic emissions were expected in order to observe the diurnal variation of the emission rates. This cell is located at mount Parnitha which is a densely forested mountain range north of Athens and also, the highest at the Attica peninsula. As we can observe (Figure 2), isoprene emissions occur only during daytime because of their strong light dependency and on the other hand, monoterpenes and OVOCs emissions occur both during daytime and nighttime. The maximum values are observed during midday (13.00 UTC) with isoprene being the most abundant of the species.

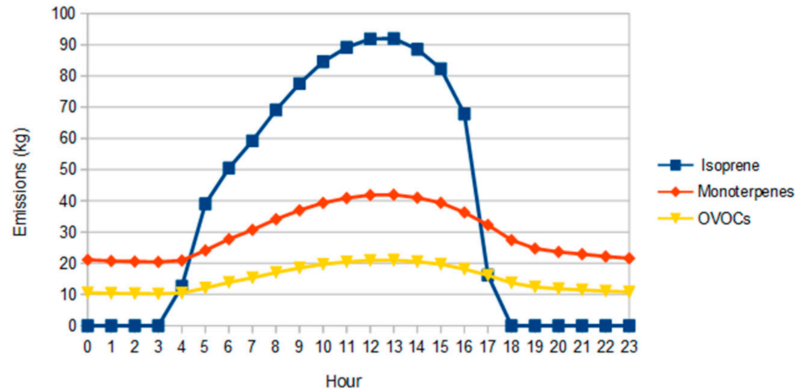
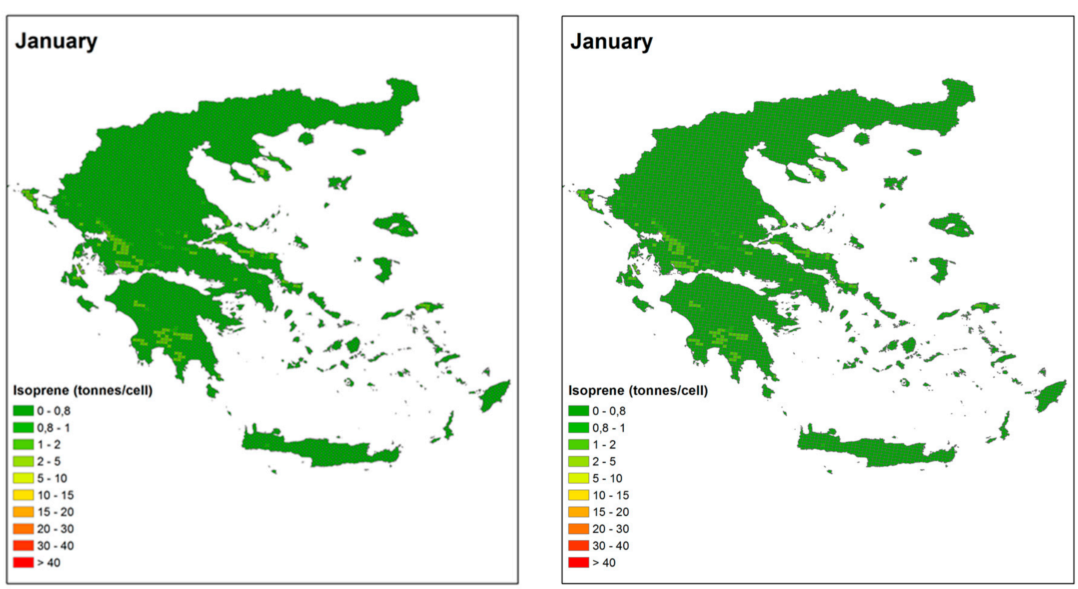


Figure 2. Diurnal variation of BVOCs emission rates above a selected grid cell (Parnitha, Attica).

As it was expected, the BVOC emissions take their maximum values during summertime and more precisely, in July. A winter month (January) and a summer month (July) were chosen to be presented in the present study. More precisely, isoprene emissions during summer reach the 148.71 tones per cell, while on the other hand, in January much lower isoprene emissions were estimated as expected, (Figure 3). The land-use categories that are characterized by maximum isoprene emissions are the mixed forests and the deciduous broadleaf forests because of their high emission potential and foliar biomass density.

Figure 3. Monthly isoprene emissions on (a) January and on (b) July.



(**a**) (**b**)

The isoprene emissions during summer are higher than the monoterpene and OVOCs ones, but during winter the monoterpenes and OVOCs emissions are higher because of the dependency of isoprene emissions on solar radiation. Indicatively, in July, the isoprene emissions contribute up to

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53.6% to the total biogenic emissions, with monoterpenes contribution being only up to 24.4% and OVOCs' s up to 22% (Figure 4). On the other hand, in January, monoterpenes contribution is the highest one (46.8%), followed by OVOCs' s (42.7%) and isoprene's (10.5%).

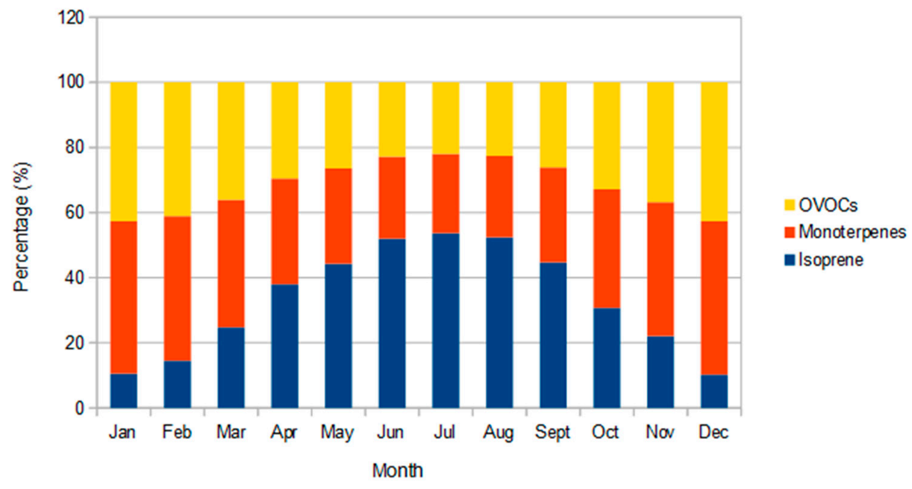

Figure 4. Percentage of contribution of each hydrocarbon to the total monthly biogenic emissions.

Finally, the annual biogenic emissions were estimated as the sum of the total monthly emissions for 2016 per hydrocarbon. Figures 5 and 6 illustrate the spatial distribution of annual isoprene and monoterpene emissions over Greece. Maximum isoprene emissions are about 545.11 tones per cell. Spatially, these maximum values are observed above mixed forests and deciduous broadleaf forests located in the area of Thrace, West Thessaly, Peloponnese, Epirus (Pindus Mountains), Lesvos and Macedonia. Concerning the monoterpenes, peak emission values are about 188.7 tones per cell. The land-use categories characterized by high monoterpenes emissions are the mixed forests and the mixed dryland/irrigated cropland and pasture and are located to the Western Epirus, Central Greece, Euboea, Lesvos, central Peloponnese and Thrace. Finally, the maximum OVOCs emission observed to the area of Greece is up to 112.9 tn/cell. This maximum value is the lowest compared to the maximum values of isoprene and monoterpenes emissions. The areas characterized by high OVOCs emissions are the same as the monoterpenes' s ones.

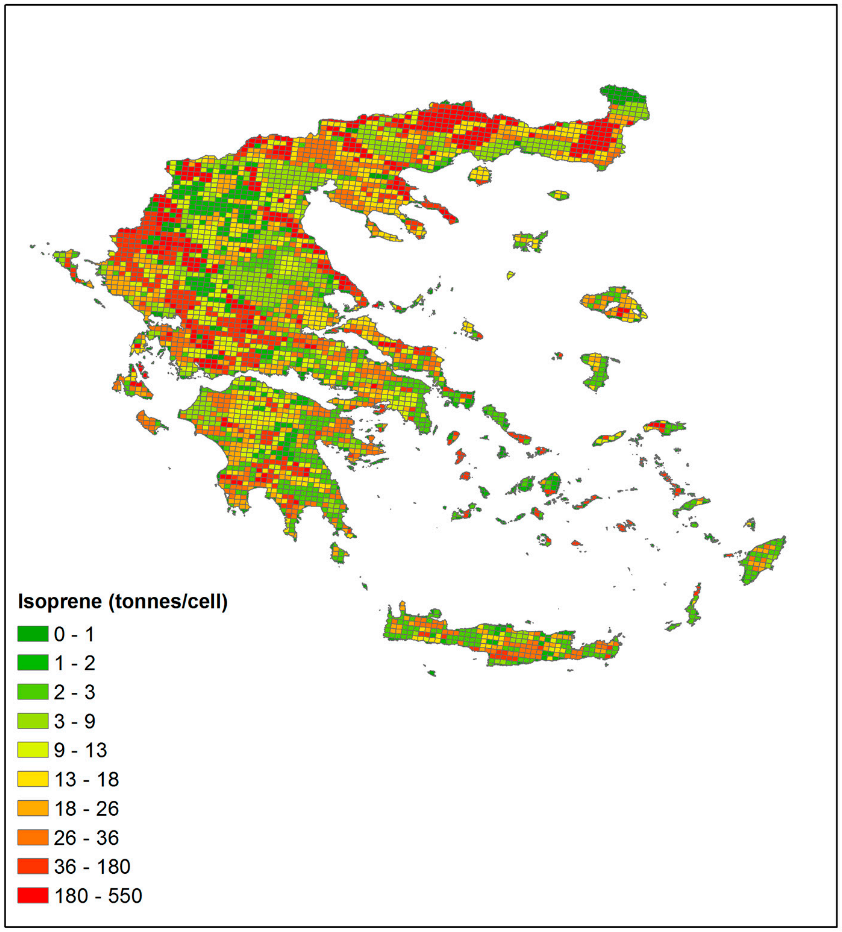

Figure 5. Spatial distribution of total annual isoprene emissions over Greece.

A part of this study is the comparison between the annual biogenic emissions with the anthropogenic ones. For Greece, several studies have been done in order to estimate the anthropogenic NMVOCs emissions due to their importance in the creation of photochemical pollution. The annual anthropogenic NMVOCs emissions were derived from the study [6] concerning the years 2006 to 2012. For the comparison, the emissions for the year 2012 were used. The annual biogenic emissions were estimated up to 472 kt and the anthropogenic ones up to 325 kt for Greece. We observe that the annual biogenic emissions exceed the anthropogenic emissions by a fact of 1.45, fact that underlines the study of the biogenic emissions in the Mediterranean.

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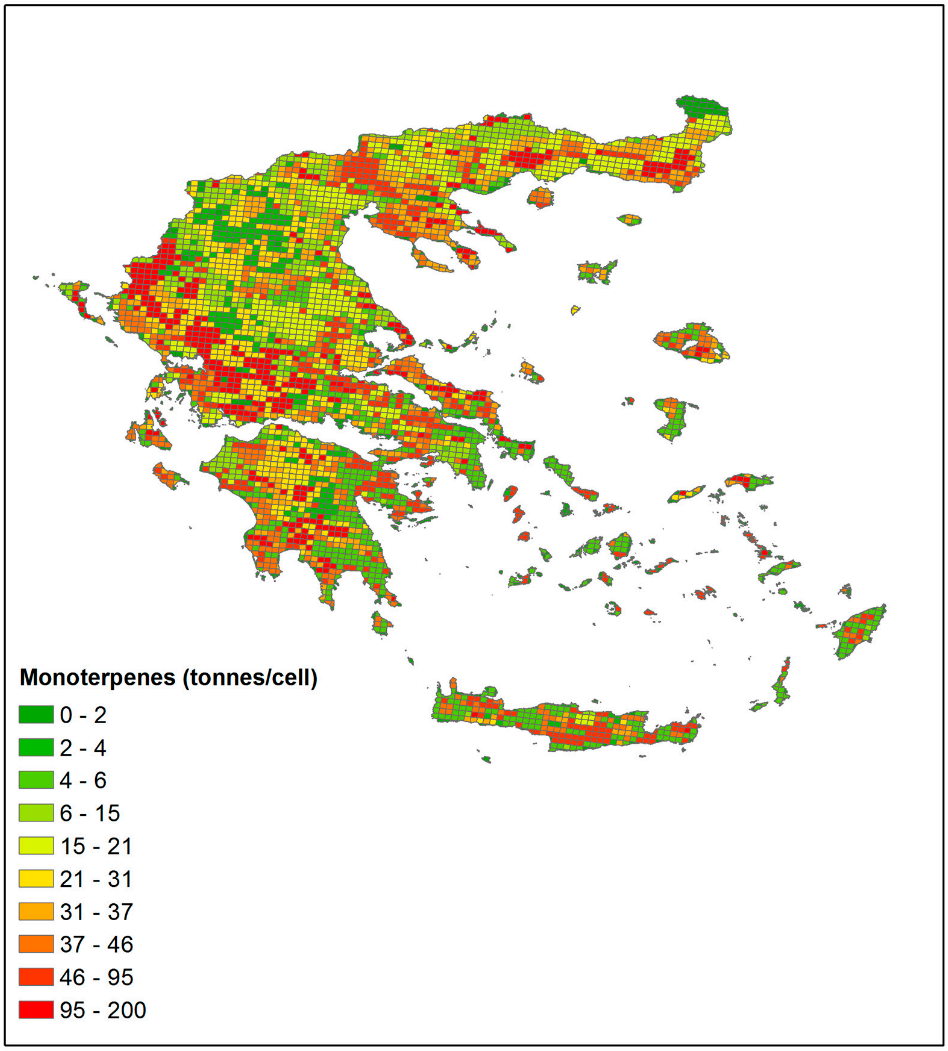

Figure 6. Spatial distribution of total monoterpene emissions over Greece.

The total biogenic NMVOCs emissions over the study area are estimated to be 473 kt, consisting of 220 kt of isoprene, 132 kt of monoterpenes and 120 kt of OVOCs. Comparing the present results with those of other studies for Greece or for an extender area including Greece, it was found that our results differ by a factor of 0.58 from the results of [7], and by a factor of 1.92 from the study of [14]. Given the fact that the estimated uncertainty level of annual global biogenic emissions is a factor of 3 [1] which is the lower limit of accuracy of annual biogenic emissions for Europe, it was assumed that the results from this study are in good agreement with already existing studies.

5. Concluding Remarks

In the present study, the biogenic emissions in Greece were estimated with the aid of a Geographic Information System (GIS), existing equations and detailed meteorological and solar radiation data. Hourly, daily and annual isoprene, monoterpenes and OVOCs emissions were estimated for the area under study with a spatial resolution of 6x6 km^2.

In the study domain, annual biogenic NMVOCs emissions were estimated to 472 kt, composed of 46.6% isoprene, 28% monoterpenes and 25.4% OVOCs. The annual cycle of biogenic emissions is characterized by higher values during summertime which is in agreement with the influence of high temperature and solar radiation values on the emissions. It is estimated that 89.3% of the annual isoprene emissions occur between May and September due to their dependency on temperature and radiation fluxes. On the other hand, the corresponding percentage for monoterpenes and OVOCs is lower (75% for both types of hydrocarbons).

As it was expected, the different land use categories emit different quantities of hydrocarbons as they are characterized by different foliar biomass densities and emission potentials. It was observed that the areas exhibiting peak isoprene emissions are covered either by forest with broadleaved deciduous trees or by forest with evergreen and deciduous trees, mostly present at western and northern Greece. Annual isoprene emissions contribute 46.3% to the annual biogenic emissions. Concerning the monoterpene emissions, the areas emitting the most are covered by forests with evergreen and deciduous trees. The annual monoterpene emissions contribute 28.2% to the total annual emissions. Finally, the annual OVOCs emissions are estimated to contribute 25.5% to the total annual emissions.

Further work has to be done concerning the estimation of biogenic emissions. More precisely, the use of isoprene, monoterpene and OVOCs emission potentials is accompanied by a high level of uncertainty. In the present study, the emission potentials derived from the database given by [8] which was the most recent study for the European vegetation, since previous studies used. Already existing databases are based on the American vegetation types. Then, for the land-use categories characterized by a combination by two or more ecosystems, we do not have a certain emission potential. Therefore, more work is required for these land-use categories for Europe and more precisely, for the Mediterranean regions, as this area contributes significant to the total European annual biogenic emissions. Furthermore, the use of a fixed emission potential for the OVOCs emissions, due to lack of experimental measurements concerning the OVOCs, is a source of uncertainty.

Concerning the foliar biomass densities used for each month covering one year, it is underlined that they are, also, a source of uncertainty because of the use of corrective factors in order to describe the seasonal variation of the foliar biomass densities. Concerning the satellite land-use data, despite the fact that they are characterized by high resolution, they can be easily misinterpreted. In the present study, the satellite land-use data were checked and tested for the valid representation of land use categories covering Greece. At this point, it should be mentioned that hourly meteorological data have been used for certain periods, instead of climatological data, affecting our estimations, a fact which reduces the uncertainty in the calculation of the environmental correction factors.

In conclusion, given the importance of biogenic emissions in atmospheric photochemistry in Greece, it is essential to continuously monitor, record and improve the methods estimating the emissions from natural sources as well as the study of their interaction with the anthropogenic emissions and finally, their overall contribution to ozone and to aerosols matter formation.

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