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A model for European Biogenic Volatile Organic Compound emissions: Software development and first validation

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ABSTRACT

A grid-oriented Biogenic Emission Model (BEM) has been developed to calculate Non-Methane Volatile Organic Compound (NMVOC) emissions from vegetation in high spatial and temporal resolutions. The model allows the emissions calculation for any modeling domain covering Europe on the basis of: 1) the U.S. Geological Survey 1-km resolution land-use database, 2) a land-use specific, monthly isoprene, monoterpene and Other Volatile Organic Compound (OVOC) emission potentials and foliar biomass densities database, 3) temperature and solar radiation data provided by the mesoscale meteorological model MM5. The model was applied for Europe in 30-km spatial resolution for the year 2003. The European total emissions for 2003 consist of 33.0% isoprene, 25.5% monoterpenes and 41.5% OVOC. BEM results are compared with those from the well-documented global Model of Emissions of Gases and Aerosols from Nature (MEGAN). The BEM total emissions compare well with the MEGAN ones. In July 2003, the results of both models agree within a factor of 1.2 for total isoprene emissions and within a factor of 2 for total monoterpene emissions. The comparison of the spatial distributions of the July 2003 isoprene and monoterpene emissions calculated with BEM and MEGAN shows that, in the greater part of the study area, the differences are below the current uncertainty limit for the estimation of spatially-resolved biogenic VOC emissions in Europe being equal to about ±600 kg km^{-2} $month^{-1}$. Differences that are above this limit are found mainly in the eastern European countries for isoprene and in the Mediterranean countries for monoterpenes.

1. Introduction

Vegetation naturally releases organic compounds in the atmosphere, which are collectively referred to as Biogenic Non-Methane Volatile Organic Compounds (BNMVOC). BNMVOC play a prominent role in the chemistry of the atmosphere and more particularly in the formation of tropospheric ozone (Curci et al., 2009; Wang et al., 2008; Bell and Ellis, 2004; Zerefos et al., 2002) and secondary organic aerosols (Kleindienst et al., 2007; Kanakidou et al., 2005). Curci et al. (2009) simulated an average 5% increase in summer daily ozone maxima over Europe due to BNMVOC emissions with peaks over Portugal and the Mediterranean region (+15%). Brasseur et al. (2003) demonstrated that BNMVOC may produce 30–270 Tg particles at global scale annually. BNMVOC suppress concentrations of the hydroxyl radical (OH), enhance the production of peroxy (HO_2 and RO_2) radicals and generate organic nitrates that can sequester NO_x and allow long-range transport of reactive N (Fehsenfeld et al., 1992). Since the surface fluxes of these compounds are critical in controlling the OH concentration in the troposphere, they determine the growth rate of atmospheric methane and CO concentrations and play a key role in the global climate and global carbon cycle (Poisson et al., 2000; Roelofs and Lelieveld, 2000; Guenther et al., 1995).

Total global BNMVOC emissions are estimated to range from 700 to 1150 TgC per year and represent about 90% of total NMVOC emissions (including anthropogenic sources) (Lathière et al., 2005; Guenther et al., 1995). However, at regional scale, the mass ratio of biogenic versus anthropogenic NMVOC emissions may change significantly. In Europe, for example, anthropogenic and biogenic

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NMVOC emissions are estimated to have comparable magnitudes (Simpson et al., 1999), while in the Mediterranean area, NMVOC emissions are dominated by emissions from vegetation during summertime (Symeonidis et al., 2008; Simpson et al., 1999).

Due to their importance in atmospheric chemistry, BNMVOC emissions must be considered in numerical Chemistry Transport Models (CTMs) simulations. The modeling of BNMVOC emissions is rather complicated because of their great sensitivity to environmental parameters (mainly temperature and radiation), vegetation type and leaf area (Guenther et al., 1995). BNMVOC emissions were first included as inputs to oxidant models in mid 80's; by the 90's they were routinely included in CTMs, but typically as static emission inventories of usually low spatial and temporal resolution. Such emission inventories have been compiled at global (Guenther et al., 1995), continental (Simpson et al., 1999, 1995; Lamb et al., 1993) and regional scales (Parra et al., 2004; Simeonidis et al., 1999; Benjamin et al., 1997) with various degrees of sophistication and reliability. However, BNMVOC emission models, which can be integrated into regional and global CTMs, are required (Smiatek, 2008; Yarwood et al., 2007; Guenther et al., 2006; Smiatek and Steinbrecher, 2006; SMOKE 2.6, 2009 (Biogenic Emissions Inventory System (versions 2, 3.09 and 3.14))). The use of such models facilitates the studies of the earth system interactions and feedbacks, and ensures the consistency between land-use and weather variables used for atmospheric process models (Guenther et al., 2006). In addition, ozone and particle biogenic precursors originate from diffuse and highly complex sources (e.g. forests, grassland) and as a result BNMVOC emission inventories should require detailed input data of land-use with a high spatial resolution (Steinbrecher et al., 2009; Arneth et al., 2008a). In this context, the BNMVOC emission models are also useful since they can provide emissions estimates at high spatial and temporal resolutions.

The aim of this paper is to present a grid-oriented emission model for the estimation of BNMVOC surface fluxes in spatial and temporal resolutions defined by the user. Gridded emissions are calculated for any domain covering part or whole of the European continent and can support air-quality modeling studies. In Section 2, the model methodology, input/output data, structure and procedures are described in detail. Hourly isoprene, monoterpene and OVOC emission fluxes were calculated in Europe and adjacent countries/areas with 30-km resolution for the year 2003. In Section 3, the spatial distribution and temporal variation of these emissions are discussed. In Section 4, the monthly isoprene and monoterpene emissions for January and July 2003 are compared with the estimates of the model MEGAN (Guenther et al., 2006) and the differences are discussed. A comparison between the observed and the CAMx model simulated isoprene concentrations is also shown. The conclusions are drawn in the final section of the paper.

2. Model description

The grid-oriented Biogenic Emission Model (BEM) consists of four parts: 1) a geospatially referenced land-use database, 2) a land-use and chemical species-specific emission potentials and land-use specific foliar biomass densities database, 3) temperature and solar radiation data provided by the meteorological model MM5 (version 3) and 4) a Fortran90 code developed to process all input data and perform the calculation of emissions (Giannaros, 2007). BEM is available to the modeling community upon request from the developers (Giannaros T., Poupkou A., Melas D.). Fig. 1 shows the flow chart of the BEM procedures.

The model uses the methodology described in Guenther et al. (1995) for the calculation of isoprene "synthesis" emissions (depending on both temperature and light) and monoterpene and Other Volatile Organic Compound (OVOC) "pool" emissions (depending on temperature only). The model also accounts for the light dependency of monoterpene emissions from some vegetation species. According to this methodology, for a given land-use i and chemical species j, the emission $E(i,j)$ (μg h^{-1}) is estimated as:

$$E(i,j) = A(i)\ \varepsilon(i,j)\ D(i)\ \gamma(j) \tag{1}$$

where $A(i)$ (m^2) is the area of the emitting land-use, $\varepsilon(i,j)$ (μg g-dry weight foliage^{-1} h^{-1}) is an emission potential at a standard temperature (=303 K) and Photosynthetically Active Radiation (PAR) (=1000 μmoles-photons (400–700 nm) m^{-2} s^{-1}), $D(i)$ (g-dry weight foliage m^{-2}) is the foliar biomass density and $\gamma(j)$ is a unitless environmental correction factor, used in order to account for the effect of leaf temperature and radiation variations on emissions. In BEM, a non-canopy approach is adopted. This approach assumes that leaf temperature and PAR flux within the canopy are identical to the ambient levels and that the use of branch-level emission potentials, which are typically a factor of 1.75 smaller than the leaf-level values (Guenther et al., 1994), accounts for the shading effects (Simpson et al., 1999).

The emission model can be operated in a grid mode. For each grid cell of the modeling area, the grid cell area A, the grid cell average emission potentials ε, the grid cell average foliar biomass density D and the meteorological data on temperature and light intensity are required. A more comprehensive description of the model's input/output data, architecture and procedures is given in the following sections.

2.1. Input data

2.1.1. Land-use

The main source of the land-use data is the Eurasia Land Cover Characteristics database (version 2) developed from satellite observations and freely distributed by the U.S. Geological Survey (USGS) (http://edc2.usgs.gov/glcc/eadoc2_0.php, last accessed 20 Apr 2010). The data have 1-km spatial resolution. The main dataset used is the "Seasonal Land Cover Regions" (SLCR) Legend that assigns each 1-km^2 area of Eurasia's surface to one of 253 different land-use classes The European continent (and adjacent countries/areas) is covered by 196 SLCR land-use classes (vegetation species and/or ecosystem types) that emit BNMVOC. It should be noted that originally, the SLCR dataset does not include a land-use class for the urban areas. For this reason the "International Geosphere Biosphere Programme" (IGBP) dataset, which includes the urban land-use class, is also used. Both the SLCR and IGBP datasets come in flat, headerless, raster format (binary) and share the same projection (Lambert Azimuthal Equal Area). The model combines the SLCR and IGBP datasets and creates a new hybrid SLCR dataset with the addition of a new land-use class for the urban areas.

2.1.2. Foliar biomass densities and emission potentials

Foliar biomass densities (g-dry weight foliage m^{-2}) and isoprene (synthesis), monoterpene (pool and synthesis) and OVOC (pool) emission potentials (μg g-dry weight foliage^{-1} h^{-1}) have been assigned for every month of the year to each of the hybrid SLCR land-use classes that emit BNMVOC. The methodology adopted for assigning foliar biomass densities and emission potentials values by land-use class is presented and discussed in detail in Symeonidis et al. (2008). The database of foliar biomass densities and emission potentials presented in Symeonidis et al. (2008) was used, having been updated and extended in order to include values for additional land-use classes covering the European continent and the adjacent countries/areas. The foliar biomass densities and emission potentials values were selected from the list of references given in Symeonidis et al. (2008) and from additional

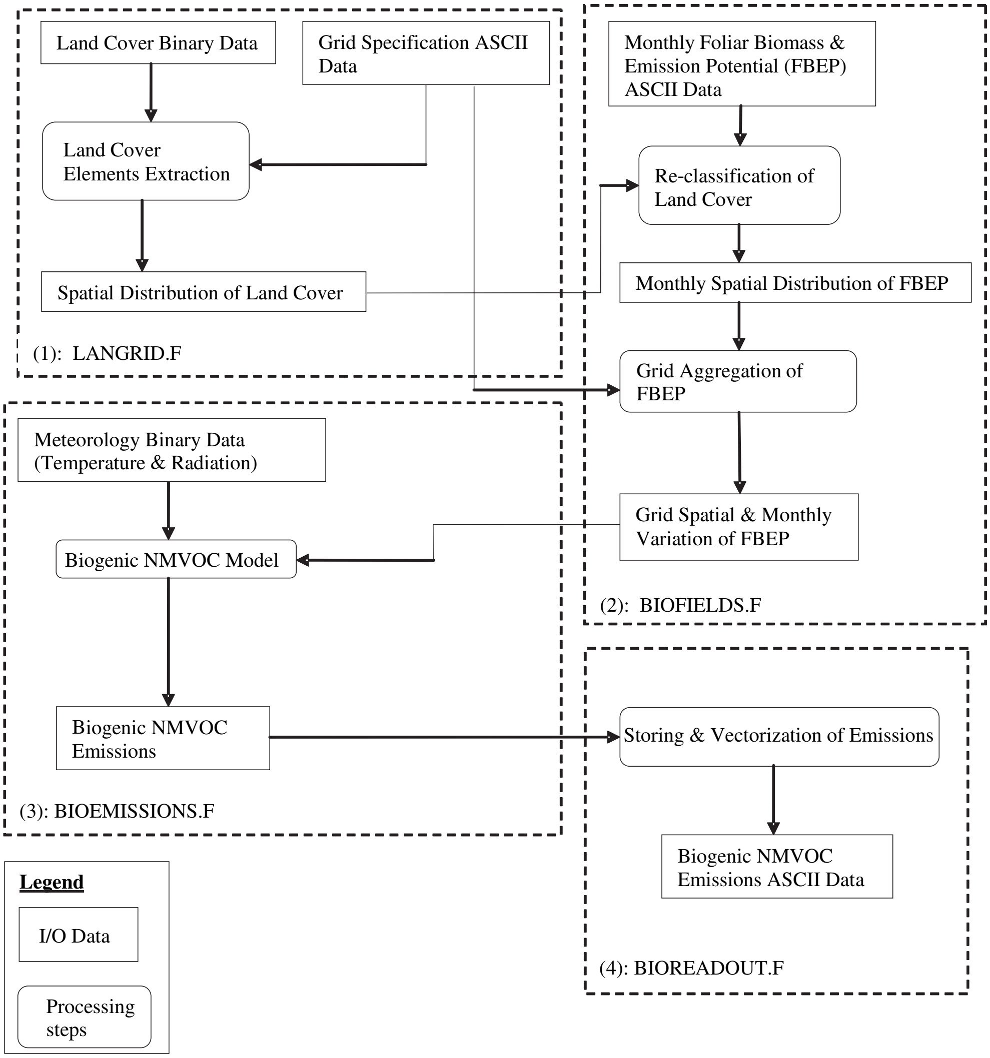

Fig. 1. Flow chart of the BEM procedures.

research studies (Tao and Jain, 2005; SMOKE 2.2, 2005; Stewart et al., 2003; Klinger et al., 2002; Simpson et al., 1999; Benjamin et al., 1996). The model reads the land-use specific foliar biomass densities and emission potentials from twelve ASCII files (one for every month of the year).

2.1.3. Temperature and solar radiation data

The Fifth Generation NCAR/Penn State Mesoscale Model (MM5 version 3) (Dudhia, 1993) is used in order to provide BEM with estimates of air temperature and surface downward shortwave radiation, which is correlated with PAR (http://www.mmm.ucar.edu/mm5/mm5v3.html, last accessed: 20 Apr 2010). The application of a numerical weather prediction model instead of reanalysis data is necessary in order to obtain meteorological data of high temporal (usually hourly) and spatial resolution. In the emission model, *PAR* flux (μmoles-photons m^{-2} s^{-1}) is estimated from the MM5 surface downward shortwave radiation flux (*SWDOWN*) (W m^{-2}) using the formula:

$$PAR = c_f \; c_u \; SWDOWN \qquad (2)$$

where $c_f = 0.45$ represents an estimate of the fraction of *PAR* in *SWDOWN* and $c_u = 4.6$ is the conversion factor to convert W m^{-2} to photon units. The MM5 output files include data in binary format and big-header and sub-header flags that make the read-in procedure a rather simple task.

2.2. Model structure and procedures

The calculation of the biogenic emissions is performed in four new developed subroutines, totally coded in Fortran90, that have been integrated in the interface (MM5–CAMx program code version 4.6) between the mesoscale meteorological model MM5 (version 3) and the photochemical Comprehensive Air Quality Model with extensions (CAMx version 4.3 and later) (ENVIRON, 2006) (http://www.camx.com/, last accessed 20 Apr 2010). The model can be executed on a LINUX PC in order to calculate gridded and temporally-resolved BNMVOC emissions and generate CAMx meteorological input files from MM5 output files.

In order to run BEM, a simple job script is required, where user-control variables must be defined. The most important variables for the calculation of the biogenic emissions include:

a) the output domain grid configuration, i.e., the projection (Universal Transverse Mercator (UTM) or Latitude/Longitude (Lat/Lon) or Lambert Conic Conformal (LCC)), the origin, the grid size (horizontal rows and columns, and vertical layers) and horizontal resolution. It should be noticed that the output grid must fit within the confines of the MM5 grid but can have different configuration (with some limitations existing only for the vertical structure),

b) the start and end date/hour for the calculations (spanning from a single hour to several days), which should be consistent with the MM5 simulation period, and the temporal resolution of the MM5 output fields being the same with that of the calculated emissions,

c) the MM5 files to be processed.

Once the user-control variables have been defined, the main model procedures used for the calculation of the biogenic emissions are:

1. Read the binary land-use datasets and create the hybrid SLCR land-use field based upon the output domain grid configuration.
2. Read the tabular input data (monthly land-use specific foliar biomass densities and emission potentials) based on the date.
3. Calculate average foliar biomass densities and emission potentials for each grid cell of the output grid. The BEM identifies the hybrid SLCR land-use elements (1-km resolution) within each grid cell area defined by the coordinates of the grid cell corners. Each land-use class has already been associated with a foliar biomass density and emission potentials (step 2). The model calculates average foliar biomass densities and emission potentials for each grid cell applying the following formulas:

$$D(i,j) = \frac{\sum_{k=1}^{N} D_k}{N} \tag{3}$$

$$\varepsilon(i,j) = \frac{\sum_{k=1}^{N}\left(\frac{\varepsilon_k \cdot D_k}{N}\right)}{D(i,j)} \tag{4}$$

where $D(i, j)$ and $\varepsilon(i, j)$ are respectively the average foliar biomass density and emission potential (either for isoprene or monoterpenes or OVOC) values for the grid cell (i, j), respectively D_k and ε_k are the foliar biomass density and emission potential (either for isoprene or monoterpenes or OVOC) values for each hybrid SLCR land-use element within the grid cell and N is the total number of land-use elements within the grid cell.

4. Read the raw MM5 data and convert them to output grid (horizontal interpolation, vertical aggregation, units conversion etc).
5. Calculate isoprene (synthesis), monoterpene (pool and synthesis) and OVOC (pool) emissions in each grid cell using air temperature data for the first model layer and SWDOWN data that have been converted to PAR.
6. Sum up monoterpene pool and monoterpene synthesis emissions in each grid cell.
7. Iterate steps 4–6 (or 1–6 in case that the calculation time period varies between different months) for all required time steps.
8. Write the gridded and temporally-resolved BNMVOC emissions.

2.3. Output data

Gridded and temporally-resolved isoprene, monoterpene and OVOC emissions (in g/grid cell/time) are calculated for any user-defined domain covering part or whole of the European continent. The emissions are written in ASCII files that can be easily read and processed in order to support the implementation of CTMs. The BEM outputs a separate file for each day of the user-defined simulation period. Originally, the MM5–CAMx program horizontally interpolates and vertically aggregates the MM5 data to the output grid and produces the binary CAMx meteorological input files (temperature, wind, vertical diffusivity, water vapor, cloud/rain, height/pressure and land-use). Additionally, the new program makes use of the MM5–CAMx program interpolation options in order to output the surface downward shortwave radiation flux in binary format, as it is one of the variables that drive biogenic emissions.

3. Model application and results

BNMVOC emissions were calculated for Europe and neighboring countries/areas in 30-km spatial resolution and hourly temporal analysis for the year 2003. The output grid consisted of 164 columns by 151 rows and 15 layers (up to about 7-km agl), used the Lambert Conic Conformal projection and was centered at 13°E and 55°N. The first layer had a thickness of about 20 m.

The fifth generation NCAR/Penn State University Mesoscale Model MM5 version 3.6 was implemented in order to provide the emission model with hourly estimates of air temperature and solar radiation. The domain used covered Europe and part of the adjacent countries/areas with a horizontal resolution that was equal to 30-km. The grid used a Lambert Conformal projection and was centered at 13°E and 55°N. The modeling domain of MM5 consisted of 199 × 175 grid cells. The atmosphere was divided in 29 layers from the surface to 100 hPa. The yearly simulation was performed using 3-day periods. Every simulated period was initialized at 12 UTC on the day before in order to have a 12-h spin-up period. Initial and boundary conditions were developed using the European Centre for Medium-Range Weather Forecast (ECMWF) 6-hourly operational analyses for the year 2003. Standard MM5 parameterizations were used. MM5 was implemented on a PC/LINUX cluster with a total of 16-CPUs and 8 GB of available memory. The execution of the model and interfaces was fully automated.

The total annual BNMVOC emissions in the study area are estimated to be 39.6 Tg, composed of 33.3% isoprene, 24.3% monoterpenes and 42.4% OVOC. The 2003 annual European total BNMVOC emissions are about 29.6 Tg with 33.0% isoprene, 25.5% monoterpenes and 41.5% OVOC. Simpson et al. (1999) reported that, in Europe, BNMVOC emissions are 13.4 Tg per year with composition similar to that of the present study. According to the results of the European Commission project “Improving and applying methods for the calculation of natural and biogenic emissions and assessment of impacts to the air quality” (NATAIR, 2007), in the EU27(+Switzerland + Norway) area, the 2003 total annual BNMVOC emissions are 13.0 Tg in very good agreement with the estimation of the present study being 14.4 Tg. In a European domain extending from 15°W to 35°E and from 35°N to 70°N, the 2003 annual emissions for isoprene and terpenes, according to Curci et al. (2009), are 3.5 Tg and 6.0 Tg, respectively, when calculated with a model described by Steinbrecher et al. (2009). According to the present study, for the same domain, the 2003 annual emissions for isoprene and monoterpenes are estimated to be 7.7 Tg and 5.2 Tg, respectively. According to Curci et al. (2009), the 2003 annual European emissions for isoprene and terpenes (excluding emissions from Russia and Ukraine) are 6.7 Tg and 6.6 Tg, respectively, when calculated as described in Derognat et al. (2003) using the emission algorithms of Guenther et al. (1995) and emission potentials and foliar biomass densities from Simpson et al. (1999). In the present study and for a similar domain (not including emissions from Russia and Ukraine), the 2003 annual emissions for isoprene and monoterpenes are estimated to be 6.4 Tg and 4.3 Tg, respectively. The annual emissions of the present study compare within a factor of 2.2 with the results from the above mentioned studies suggesting that the results of the emission model are in good agreement with previous estimations especially when taking into consideration that the level of uncertainty in the calculation of biogenic emissions in Europe is a factor of 4 (NATAIR, 2007).

The spatial distribution of annual isoprene and monoterpene emissions in the study area is shown in Fig. 2. Maximum isoprene emissions, in the order of 7 tn km^{-2}, can be found in the northern Balkan Peninsula and along the coasts of the Black Sea. These emissions are mainly associated with the presence of deciduous broadleaf forests, which are considered to be a significant source of isoprene. Another isoprene emissions spot can be found in Belarus and western Russia where croplands and woodlands form a mixed mosaic that contributes to the release of significant amounts of isoprene. With regards to monoterpenes, maximum emissions (over 10 tn km^{-2}) are modeled in the Mediterranean countries (Portugal, Spain, Italy and Greece), where monoterpene emissions are dominated by evergreen oak woodland emissions. High monoterpene emissions are found also in the Scandinavian countries and in northern Russia and are associated with the presence of boreal forests, which are dominated by needle leaf tree species (high monoterpene emitters).

Fig. 3 shows the annual variation of the BNMVOC emissions. Isoprene emissions range from 6.2 Gg in December to 3728.2 Gg in July. Monoterpene emissions are minimum in February (122.6 Gg) and increase by a factor of about 20 in July (2292.2 Gg). OVOC emissions are maximum in July (4188.0 Gg) and take the minimum value of 122.5 Gg in February. Approximately, 95% of the annual isoprene emissions are produced during May–September. This percentage is a bit lower for monoterpenes and OVOC (81% and 85%, respectively). In winter months, the total BNMVOC emissions are composed of 2.3% isoprene, 48.3% monoterpenes and 49.4% OVOC. In summer months, the respective composition is 36.7%, 22.4% and 40.9%. The annual cycle of the total biogenic emissions shows that the highest emission values occur in summer months because of high air–temperature and solar radiation values. Total biogenic emissions range between 252.8 Gg in February and 10,208.4 Gg in July.

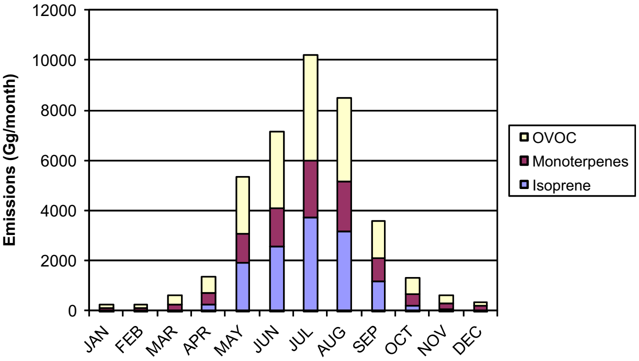

Fig. 3. Monthly BNMVOC emissions in the study area (Europe and neighboring areas).

In order to study the daily variation of emissions, the study area was divided in three spatial zones of 12° width each: South (34.5°N to 46.5°N), Central (46.5°N to 58.5°N) and North (58.5°N to 70.5°N) zone representing warm, cool and cold climates, respectively. Fig. 4 shows the daily emissions in the three spatial zones of the study area. In the southern part of the domain, the peak emission season is summer. During this period, the daily isoprene emissions, although a bit higher, they are comparable with OVOC emissions, while monoterpene daily emissions are half of the isoprene ones. In the central and northern part of the domain, the peak emission season is less extended. It starts in the beginning of July and finishes in mid August. During this season, in the central part of the study area, the daily monoterpene emissions represent about one third of the isoprene emissions being comparable with the OVOC ones. The results for the northern part of the domain show that the daily monoterpene emissions are greater than the OVOC ones and exceed isoprene emissions by a factor of 2.5. The above results indicate the abundance in isoprene emissions in the south and central zones due to the presence of high isoprene emitting vegetation species (e.g., deciduous oaks) and the abundance in monoterpene emissions in the north domain zone because of the high land coverage of monoterpene-emitting vegetation species (e.g., needle leaf trees).

The difference of the daily isoprene, monoterpene and OVOC emissions from the mean monthly values in the southern, central and northern part of the study area is presented in Fig. 5. The

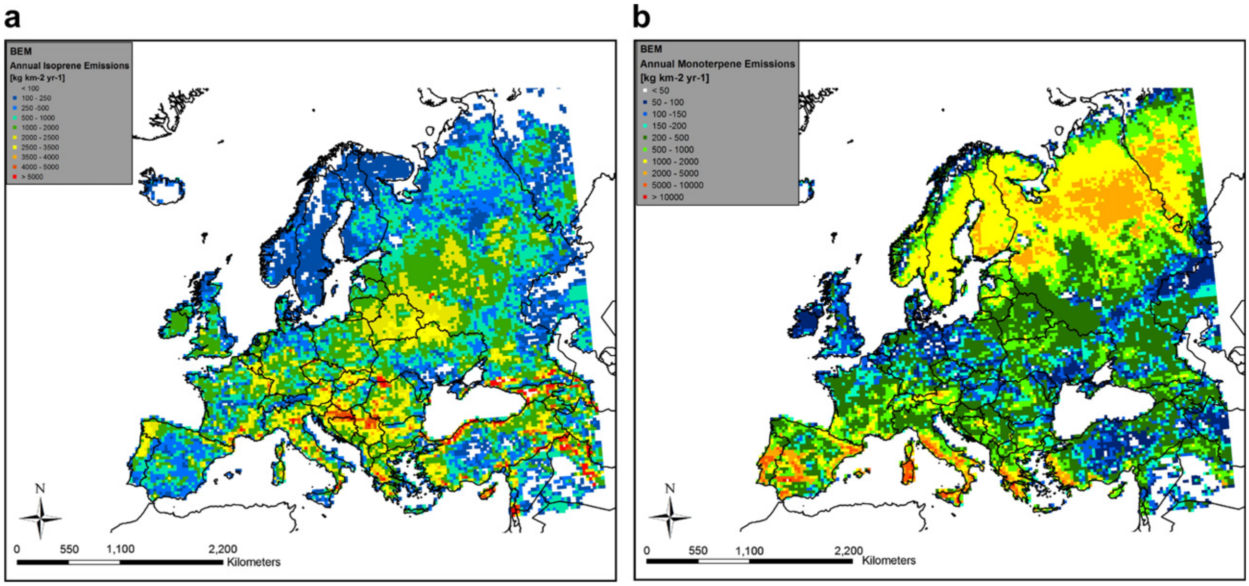

Fig. 2. Spatial distribution of the annual isoprene (a) and monoterpene (b) emissions for 2003.

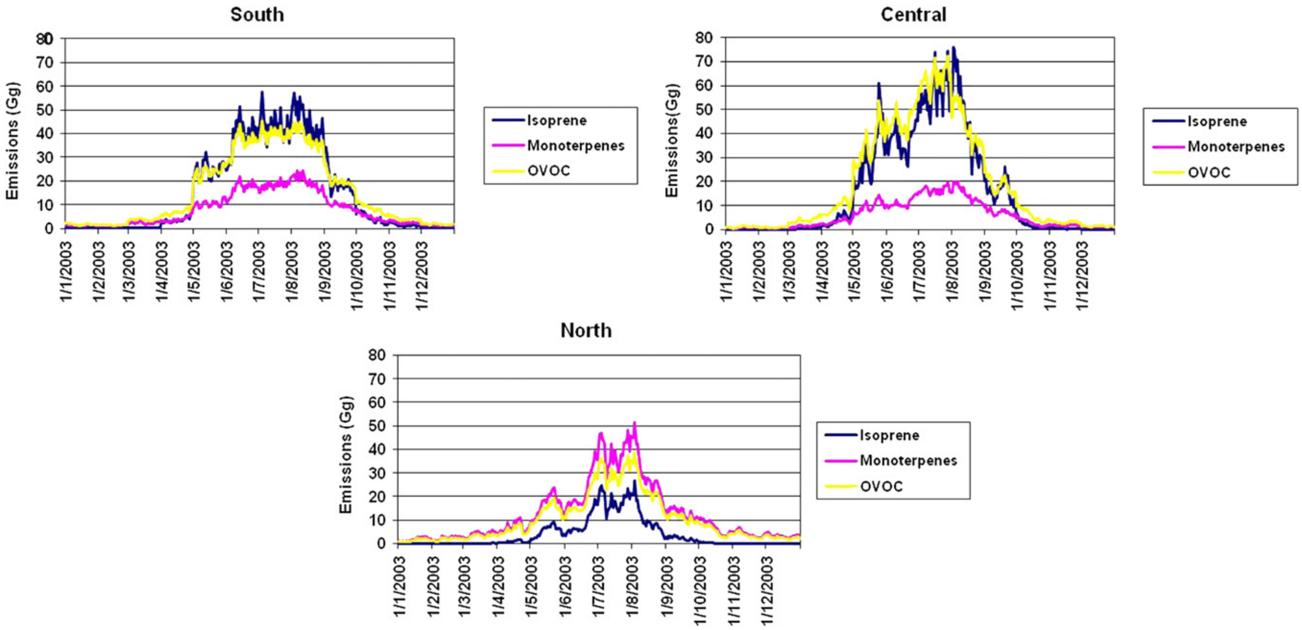

Fig. 4. Daily BNMVOC emissions (Gg) in the three spatial zones of the study area.

difference is defined from the range of the percentage differences between the monthly emission mean and the 5th and the 95th percentiles of the daily emissions within a month. The range of variation of the isoprene emissions is larger than that of monoterpene and OVOC emissions for every month of the year 2003 in the central and northern zones and for most months of the year 2003 in the southern zone of the domain. Throughout the year, the range of variation of monoterpene emissions in the south of the domain is larger compared to that of the OVOC ones. In the central and north zones, the monoterpenes and OVOC emissions have almost the same variation range since the vegetation species in these parts of the domain are mainly pool monoterpene emitters. During the colder months of the year (October–April) and for all chemical species emitted, daily emissions present lower variability in the southern part of the domain and higher variability mainly in the north. During the summer months, the greatest variability in emissions exists in the northern part of the study area. During the same period, the variability of the daily emissions in the central zone, when it is not higher, it is comparable with that in the south. The above results are in accordance with the simulated

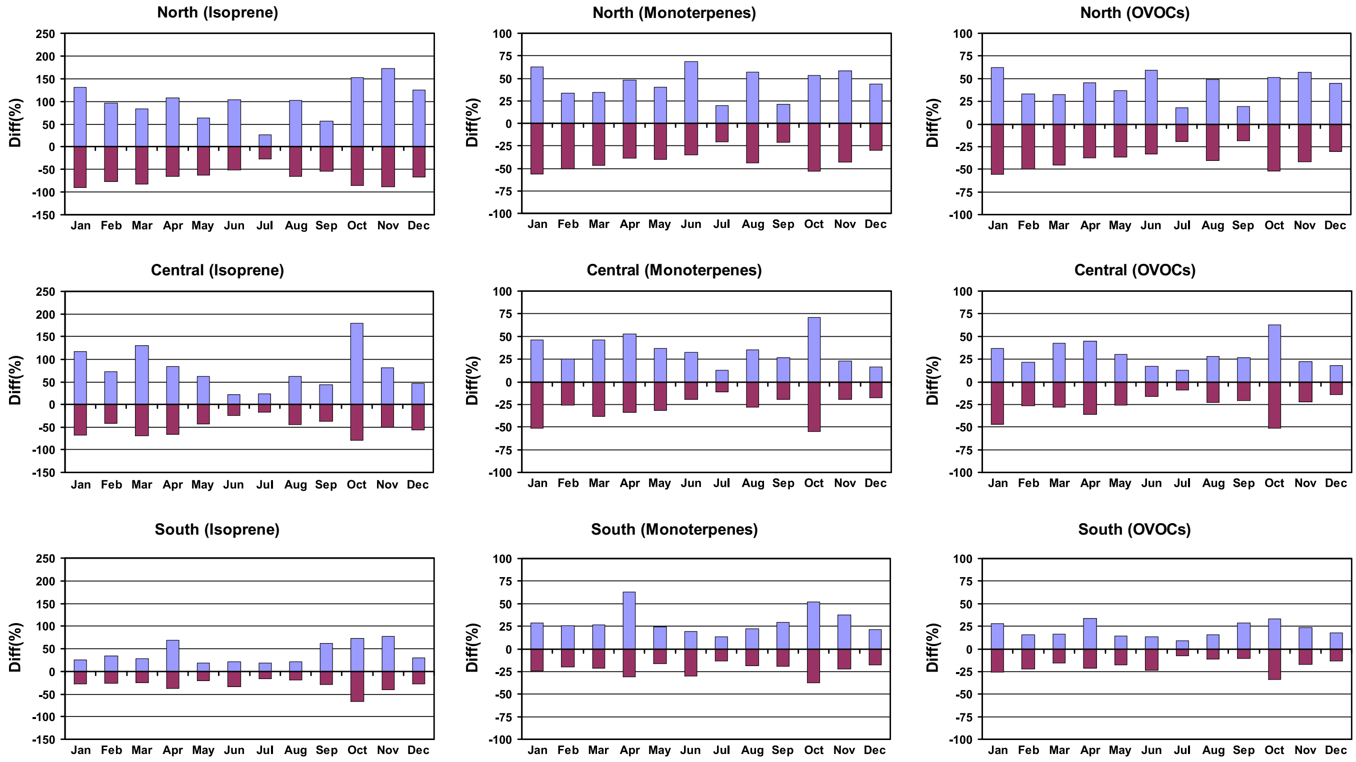

Fig. 5. Difference of the daily BNMVOC emissions from the mean monthly values in the southern, central and northern parts of the study area (reference year 2003).

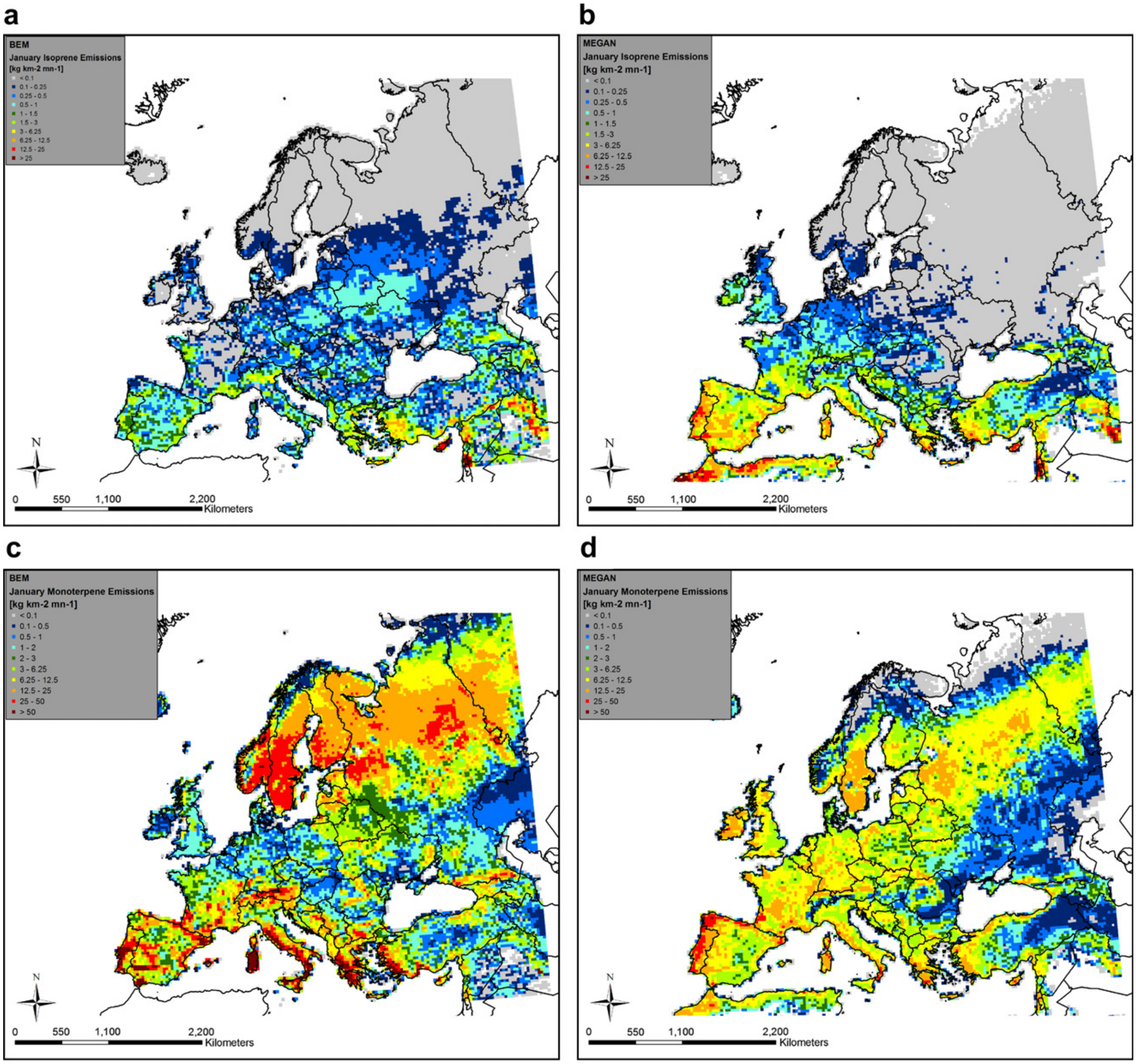

Fig. 6. Spatial distribution of isoprene and monoterpene emissions calculated with BEM and MEGAN for January 2003.

temperature and PAR data showing greater variation range in the southern part compared to the northern part of the domain (mainly for the temperature data).

4. Discussion

4.1. Comparison with MEGAN results

The uncertainties in biogenic emission estimates are large. Simpson et al. (1999) reported an uncertainty of a factor of 3–5 for isoprene and monoterpene emissions and a factor greater than 5 for the OVOC emissions from vegetation in Europe. An uncertainty factor of 4 has been reported for the annual biogenic emissions in European scale by NATAIR (2007). Large uncertainties characterize also the estimates in regional scale (e.g., a factor of 4 for the Great Britain biogenic emissions according to Stewart et al. (2003)). Smiatek and Steinbrecher (2006) have shown that, based on comparisons with above-canopy aircraft measurements, biogenic emissions can be estimated within a factor of 2 for isoprene and a factor of 3 for monoterpenes in regions where accurate model inputs are available. Differences of ±600 kg km^{-2} in July 2003 biogenic emissions calculated with different modeling approaches represent the current uncertainty in estimating spatially-resolved biogenic VOC emissions in Europe (Steinbrecher et al., 2009). These high uncertainties in biogenic VOC emission estimates result from insufficient knowledge on the plant species-specific emission potentials, on the land-use regarding the species composition and the associated biomass, on the producing mechanisms inside the plants and on the various climatic effects and/or chemical processes that determine the emissions of VOC in the canopy (Steinbrecher, 2006).

In this section, isoprene and monoterpene emissions calculated with BEM for January and July 2003 in 30-km spatial resolution are compared with those calculated with the MEGAN emission model (Guenther et al., 2006) (http://acd.ucar.edu/~guenther/MEGAN/MEGAN.htm, last accessed 20 Apr 2010). MEGAN is a widely used global model of biogenic emissions at 1-km spatial resolution. In the version employed here (MEGAN model version 2.04), isoprene and monoterpene environmental correction factors (those depending on temperature and light) were calculated in a similar way as in BEM. Land-use parameters (e.g., vegetation cover) were derived from MODIS satellite observations, while emission potentials at standard conditions were averaged over plant functional types (e.g., needle leaf or broadleaf trees) present in each model grid cell. For further details the reader is referred to Bessagnet et al. (2008). MEGAN emissions were calculated using the BEM grid configuration presented in Section 3. Both models were implemented using the same MM5 input data.

The total amounts of isoprene and monoterpenes emitted in the modeling domain in January 2003 are estimated to be 6.9 Gg and 125.4 Gg, respectively, with BEM and 10.0 Gg and 61.7 Gg with MEGAN (excluding emissions from the African part of the domain). The results agree within a factor of 1.4 for isoprene and 2.0 for monoterpenes.

Fig. 6 shows the spatial distributions of BEM and MEGAN isoprene and monoterpene emissions for January 2003. The spatial pattern of isoprene emissions calculated with both BEM and MEGAN presents emissions that are lower in central and northern Europe and higher in the Mediterranean countries. For most countries of the study domain, the spatially-resolved isoprene emission values of BEM are similar to those of MEGAN. However, in Portugal, Spain and Italy, MEGAN isoprene emissions are generally higher than BEM ones. The spatial distribution of BEM monoterpene emissions shows that the emissions are higher in the Mediterranean countries, in the Scandinavian countries and northern Russia. However, these areas are not so distinct in the figure that presents the spatial distribution of MEGAN monoterpene emissions. The differences in monoterpene estimates from both models are greater mainly over the Scandinavian countries and over parts of the Mediterranean countries (e.g., western Italy and Greece), where BEM monoterpene emissions are higher than the MEGAN ones, and over parts of central and western European countries (e.g., Germany, Ireland), where MEGAN monoterpene emissions are higher than the BEM ones.

The total amount of isoprene emitted in the modeling domain in July 2003 is calculated to be 3728 Gg with BEM and 3021 Gg with MEGAN (excluding emissions from the African part of the domain). Both results are in very good agreement presenting a low difference by a factor of 1.2. The total isoprene emissions in a European area extending from 15°W to 28°E and from 35°N to 58°N (including a small part of north Africa) in July 2003 are 1124 Gg (calculated according to Steinbrecher et al. (2009)), 1228 Gg (calculated according to Simpson et al. (1999)) and 1446 Gg (calculated with MEGAN version 2.04, Oct 2007), with a mean of 1266 ± 164.3 Gg (std) (Steinbrecher et al., 2009). BEM isoprene emissions for a similar domain and the same time period are estimated to be

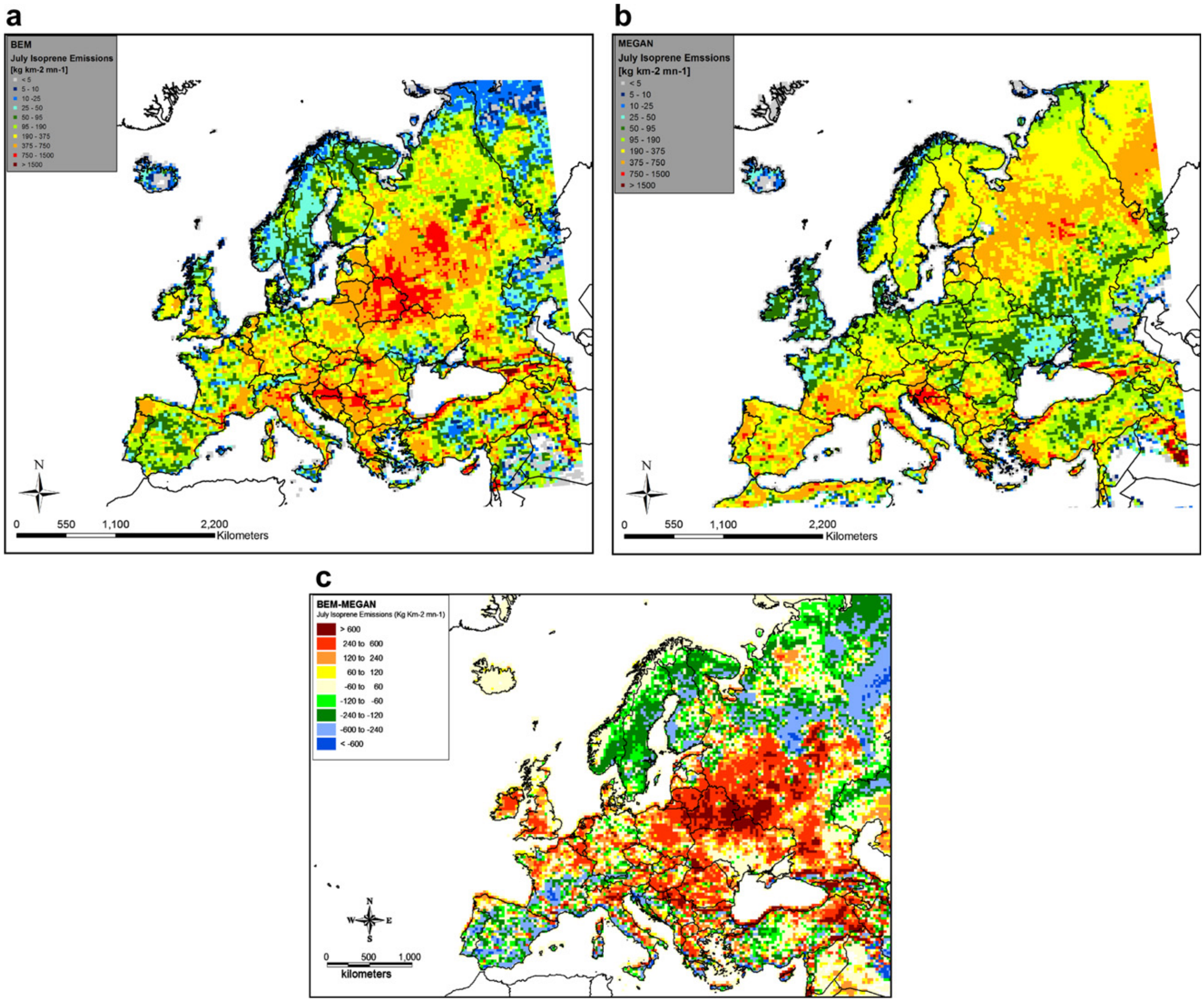

Fig. 7. Spatial distribution of isoprene emissions calculated with BEM (a) and MEGAN (b) and spatial distribution of the differences in the results from both models (c) for July 2003.

1356 Gg. This estimation falls within the 1266 ± 164.3 Gg emission range and presents small variations from the results of the above mentioned approaches that take values from +17.0% to −6.6%.

Fig. 7 shows the spatial distribution of isoprene emissions calculated with BEM and MEGAN for July 2003 and the spatial distribution of the differences between the results from both models. On a regional basis, BEM results in higher isoprene emissions in the western (e.g., Belgium, Netherlands) and central European countries (e.g., Hungary, Poland), as well as in the eastern and south-eastern European countries (Belarus, Lithuania, western part of Russia, Romania). BEM isoprene emissions are lower compared to MEGAN emissions mainly in the Scandinavian countries, in northern Russia and in the south-western European countries (Portugal, Spain, southern France). However, in the greater part of the study area, the regional differences are below the uncertainty limit of ±600 kg km^{-2} month^{-1} for the estimation of spatially-resolved biogenic VOC emission in Europe as discussed in Steinbrecher et al. (2009). The above uncertainty limit differences between the two approaches in the emissions in Belarus and the western part of Russia could be explained by the difference in the land-use characterization between BEM and MEGAN. In these regions, the dominant land cover according to BEM is a mixed mosaic of croplands and mixed forests while it is a mixture of needle leaf forests and croplands according to MEGAN. Needle leaf forests and croplands are low isoprene emitters. However, the presence of mixed forests in this region according to the BEM land-use database contributes to isoprene emissions that are much higher than those of MEGAN.

In July 2003, BEM monoterpene emissions totals in the study area (2292 Gg) are about 2 times higher than those calculated with MEGAN (1216 Gg excluding emissions from the African part of the domain). The existence of variations between emission models' estimates that are larger for monoterpenes than for isoprene is an issue that has also been found and discussed in previous studies (Steinbrecher et al., 2009; Arneth et al., 2008b). For example, according to Steinbrecher et al. (2009), in a less extended European domain (15°W–28°E and 35°N–58°N including a small part of north Africa), the July 2003 total emissions of terpenes vary between 338 Gg (calculated with MEGAN version 2.04), 789 Gg (calculated according to Steinbrecher et al. (2009)) and 1112 Gg (calculated according to Simpson et al. (1999)). BEM monoterpene emissions for a similar area and the same time period are 602 Gg, an estimation that falls within the emission range of the other models.

Fig. 8 shows the comparison between the spatial distributions of BEM and MEGAN monoterpene emissions for July 2003; it reveals that for almost all central, western and eastern parts of Europe the results from both models are similar. BEM estimates higher emissions than MEGAN in parts of the southern European countries (Portugal, Spain, western and southern Italy, western Greece), in

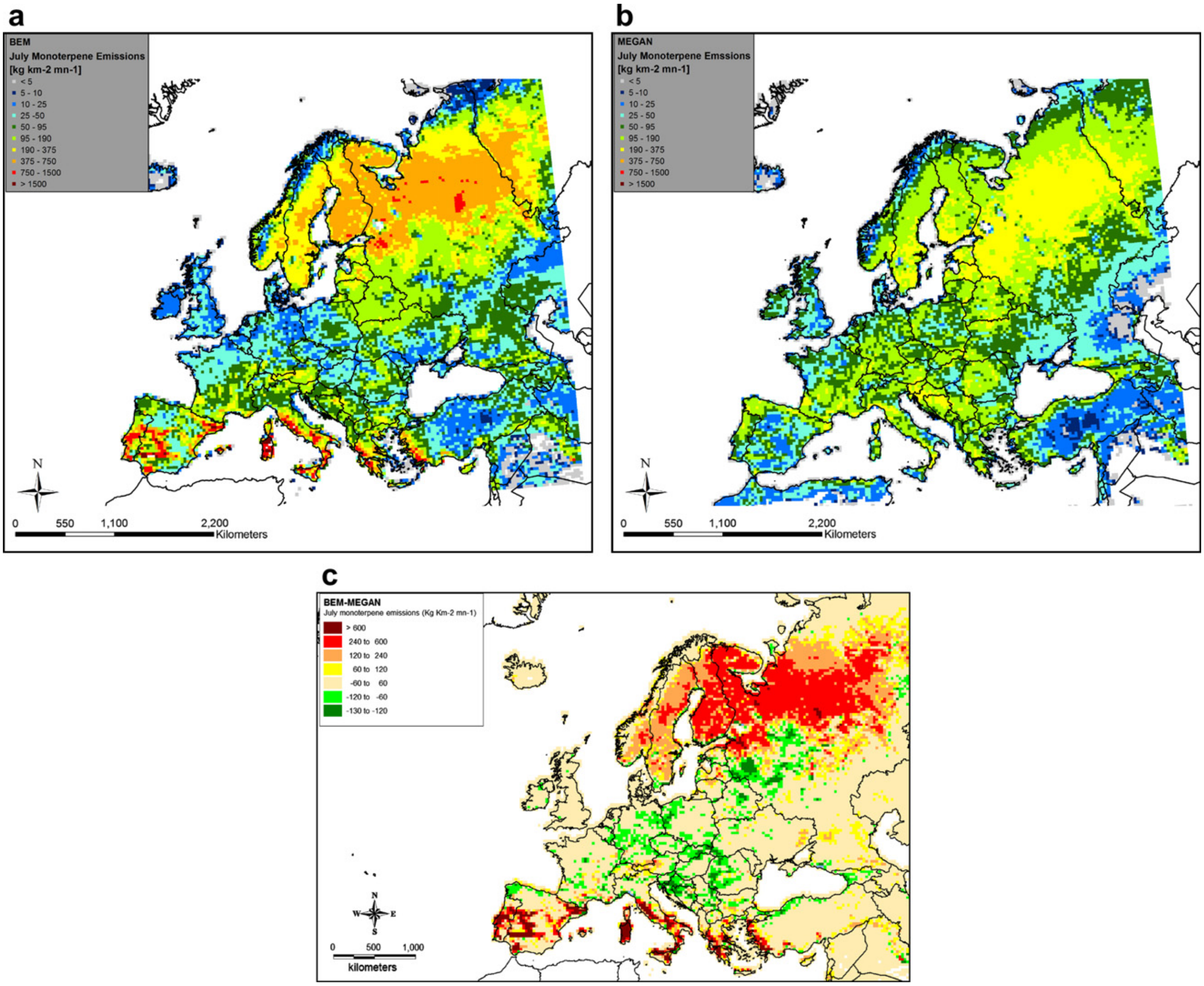

Fig. 8. As in Fig. 7, but for monoterpene emissions.

Table 1
Comparison between observed and simulated isoprene concentrations.

Country	StationID	Lon/Lat	Mean observed (ppt)	Mean simulated (ppt)	Ratio Mean obs/ Mean sim	Agreement within a factor of 2 (%)	Agreement within a factor of 3 (%)	Agreement within a factor of 4 (%)
Switzerland	CH005	8.46°E/47.07°N	121.8	236.3	0.52	39.1	59.5	64.3
Czech Republic	CZ003	15.08°E/49.58°N	119.2	170.5	0.70	61.5	88.5	92.3
Germany	DE002	10.76°E/52.80°N	75.3	133.9	0.56	52.2	78.3	82.6
	DE005	13.22°E/48.82°N	556.0	475.3	1.17	63.6	95.5	100.0
	DE009	12.73°E/54.43°N	567.9	161.6	3.51	15.0	25.0	55.0
Spain	ES009	3.14°W/41.28°N	143.8	99.6	1.44	64.0	88.0	88.0
France	FR008	7.13°E/48.50°N	2006.9	1279.2	1.57	38.5	84.6	92.3
	FR015	0.75°W/46.65°N	582.4	329.2	1.77	52.0	56.0	92.0

the Scandinavian countries and in north Russia. However, in the greater part of these areas, the existing differences are below ±600 kg km^{-2} month^{-1}, a value that represents the current uncertainty limit for the estimation of spatially-resolved biogenic VOC emission in Europe as discussed in Steinbrecher et al. (2009). Differences that are above this uncertainty limit are found in some grid cells of the Mediterranean countries where BEM estimates high emissions mainly due to the presence of evergreen oaks, which are high monoterpene emitters. It should be noted that, apart from the different land cover data used, the differences between both inventories discussed in this section either for monoterpenes or for isoprene can be also attributed to the differences in the modeling approaches of BEM and MEGAN and in the emission factors and biomass densities used.

4.2. *Comparison between simulated and observed isoprene concentrations*

In this section, isoprene measurements from the EMEP measurement network (http://tarantula.nilu.no/projects/ccc/emepdata.html, last accessed 20 Apr 2010) are compared with hourly isoprene concentrations simulated with the photochemical model CAMx (version 4.40) during a summer period when the impact of BNMVOC emissions on atmospheric chemistry is more pronounced. Canister grab samples were taken at the EMEP stations twice a week and were analyzed with Gas Chromatography/Flame Ionization Detector (GC/FID). Only in Rigi-Switzerland (CH005 station of EMEP), a continuous GC monitor was operating.

CAMx was applied for a domain which was the same with that of the BEM model (see Section 3). The 2003 summer season was simulated. Annual anthropogenic emission data of gaseous (NO_x, SO_2, NMVOC, CH_4, NH_3, CO) and particulate matter (PM_{10}) pollutants provided by The Netherlands Organization (Visschedijk et al., 2007) were used. The emission inventory was prepared for the Global and Regional Earth-System Monitoring using Satellite and in-situ data project (GEMS project) (Hollingsworth et al., 2008) in order to account for the year 2003 emissions in the European territory as well as in a part of west Asia in a grid spacing of 1/8 by 1/16°. The annual emission data were temporally disaggregated (seasonal, weekly and diurnal temporal profiles) according to Friedrich (1997). More detailed regional emission inventories (e.g., for Greece (Markakis et al., 2010a,b)) and ship emission data from the EMEP database were also used. BEM BNMVOC emissions were included in model runs. CAMx simulations were driven by the MM5 meteorological data (see Section 3). The global chemistry transport model MOZART-IFS (Flemming, 2008) provided the CAMx boundary conditions (including isoprene concentrations). The chemical mechanism used in CAMx runs was the Carbon Bond Mechanism version 4 (CB-IV) with isoprene chemistry based on Carter (1996). In CB-IV mechanism, isoprene is an explicit species and its chemical pathways include reactions with OH, O_3, and NO_3.

Table 1 shows the comparison between isoprene measurements and first model layer hourly concentrations simulated at the grid cells where the EMEP stations are located. The mean observed and mean simulated isoprene values are presented in the table along with the percentages of the simulated concentrations that agree with the observations within a factor of 2, 3 and 4, respectively. Significant discrepancies are found for two out of the ten EMEP stations providing isoprene observations, for which the correspondence between the satellite-derived and the ground-based land-use characterization for the areas where the stations are located was low (stations DE008(10.77°E/50.65°N) and FR013 (0.18°E/43.62°N) not shown in the table). For all other stations, the majority of modeled values agree with the observed ones within a factor of 4 that represents an average limit of uncertainty in the calculation of isoprene emissions in Europe. In fact, for most of the stations, the agreement is even better since high percentages of simulated isoprene concentrations (ranging from 60% to 96%) agree with the observations within a factor 3. There exist several stations for which the agreement between the majority of modeled concentrations and observations is better than a factor of 2. In almost all sites, the mean observed isoprene levels agree with the mean simulated ones within a factor of less than 2.

The above results in conjunction with the comparisons and uncertainties discussed in Section 4.1 provide evidence that the developed emission model can be used to produce good BNMVOC emissions estimates at European scale.

5. Conclusions

BEM is a new, simple but efficient grid-oriented emission model for the estimation of BNMVOC emissions in Europe in high spatial and temporal resolutions and in different grid projections. The calculation of emissions is based on a methodology that is widely used by the scientific community. Also, the calculation is performed with the use of a newly developed database of land-use specific monthly average emission potentials and foliar biomass densities for Europe. The database represents the synthesis of land-use and vegetation species emissions potentials and biomass densities that were derived from different published references. The database is associated with a high resolution, coherent and detailed, satellite-derived land-use dataset distributed by the USGS, that has been optimized for Eurasia in order to contain unique elements based on the geographic aspects of the continent and in order to be used in a wide range of environmental research studies. BEM represents new software based on four Fortran90 routines that are currently incorporated in the MM5–CAMx interface. So, it can be useful to support the modeling studies based on the application of the MM5–CAMx modeling system which is widely used by the scientific community. However, the routines developed can be easily used to produce results driven also by the meteorological model Weather Research & Forecasting Model (WRF).

BEM was applied for a European domain (including the neighboring countries/areas) and hourly BNMVOC emissions were calculated in 30-km spatial resolution for the year 2003. The 2003 European total biogenic emissions were found to consist of 33.0% isoprene, 25.5% monoterpenes and 41.5% OVOC. During the 2003 peak emission season, the daily isoprene emissions of the southern and central parts of the study area were higher than the monoterpene ones while the daily monoterpene emissions of the northern part of the domain exceeded the isoprene ones by a factor of 2.5. The daily isoprene emissions varied more than the monoterpene and OVOC emissions from the mean monthly values. The daily emissions of the northern part of the domain presented greater variability than those of the southern part.

The BEM annual emission values were in reasonable agreement (within a factor of 2.2) with the results from previous studies. The differences between BEM and other emission models in isoprene total emissions for July 2003 (when biogenic emissions are maximum) were very low. The July 2003 monoterpene emissions estimation with BEM agreed within a factor of 2 with the results from other emission models. The comparison of the spatial distributions of July 2003 isoprene and monoterpene emissions calculated with BEM and MEGAN showed that, in the greater part of the study area, the existing differences were below the uncertainty limit for the estimation of the spatially-resolved biogenic VOC emissions in Europe presented in Steinbrecher et al. (2009). Differences that were above this limit were found for isoprene mainly in the eastern European countries (e.g., Belarus and western Russia) and for monoterpenes in the Mediterranean countries.

The above results in conjunction with the results from the comparison between simulated and observed isoprene concentrations suggest that BEM can provide reasonable spatially and temporally-resolved BNMVOC emissions estimates at European scale. This is an important issue given the significant role that BNMVOC play in determining tropospheric chemistry with repercussions on air quality. Future improvements of BEM include the description of the variation of BNMVOC emissions also as a function of the seasonality of the emission potentials, the estimation of chemical compound-specific BNMVOC emissions, that are needed by the modeling community to correctly assess surface ozone and particle production in the atmosphere, and further development of the model in order to support the application of additional air-quality modeling systems other than the MM5–CAMx system.

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