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Temporal and spatial variation of forest VOC emissions in Germany in the decade 1994–2003

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Abstract

Biogenic emissions are strongly governed by temperature and light. One key issue for air quality modelling is to quantify the emission source strength variation of biogenic volatile organic compounds (BVOC) on different temporal and spatial scales. In Germany, this variation has been investigated for forests (the major source of BVOC) at country, state, and local scales over 10 years. Hourly emission rates for each vegetation period (April to October) from 1994 until 2003 were calculated in 10 km resolution. A database was compiled containing land cover, forest distribution, foliar biomass density, leaf area index (LAI) and plant specific emission potential. Meteorological input parameters were obtained using the non-hydrostatic meteorological mesoscale model MM5. The semi-empirical BVOC model (seBVOC) uses these parameters as input. SeBVOC considers in addition seasonality of the emission potentials, light extinction within the canopy as well as leaf temperature, air humidity, wind speed, and solar angle. The results show an average BVOC emission of 366 Gg, with 40 Gg isoprene, 188 Gg monoterpenes, and 138 Gg other VOC (OVOC), for the period 1994–2003. The dominating emitter type is Norway spruce (*Picea abies*) contributing approximately 40% to the total BVOC. In the years 1994–2003, the annual BVOC emissions varied in the range of ±20%. However, in specific episodes and at specific locations the variation is as high as 150% compared to average emissions. Also the BVOC composition emitted is location specific. Using a sensitivity analysis, the contribution of uncertainties in estimating foliar biomass, emission potential, as well as temperature to the overall uncertainty of calculated potential surface BVOC emission rates is addressed.
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Keywords: Isoprene; Monoterpene; BVOC; Inventory; Emission modelling

1. Introduction

Volatile organic compounds (VOC) play an important role in the chemistry of the atmosphere. Being a driving factor of the photochemical cycle (Atkinson, 2000) also influencing the particle formation (Hoffmann et al., 1997; Fuentes et al., 2001) they must be considered in any numerical chemistry-transport-model (CTM). VOC are emitted from anthropogenic as well as from biogenic sources. Biogenic volatile organic compounds (BVOC) are much more reactive (Atkinson, 2000) and therefore also important in air chemistry in industrialised areas where emissions from anthropogenic sources (AVOC) dominate. As input in CTMs, however,

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often average country-wide annual values of isoprene, monoterpenes and other VOC (OVOC) (Simpson et al., 1999) are used to account for the effect of BVOC on air pollutant distributions. Using annual averages, spatial and temporal disaggregations as well as hypothetic VOC splits lead to significant errors in regional and local BVOC estimates. For sound air quality prognoses CTMs require inventories at high spatial and temporal resolution using for the case of BVOC available plant specific VOC speciation according to the employed chemical mechanism.

This article presents such inventories for Germany as well as for state and local areas. Hourly BVOC emission fluxes from forests for each vegetation period (April–October) from 1994 until 2003 were calculated in 10 km resolution for 28 chemical compounds. The spatial and temporal variation of the emissions is discussed. Finally, results of sensitivity analyses considering the impact of uncertainties in estimations of the foliar biomass, emission potentials as well temperature and light on calculated emission rates are presented.

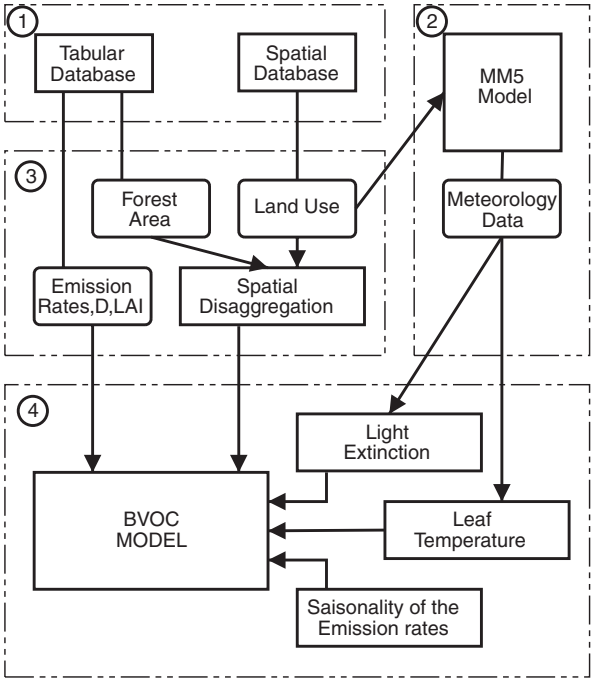

Fig. 1. Flow chart of the BVOC emission modelling procedure.

2. Model description

The BVOC modelling system is integrated in a geographical information system (GIS) and a relational data base management (RDBMS) environment of input data processing, model running, and visualization. The principal information flow is shown in Fig. 1. It consist of four parts: (1) a geo-spatial and tabular database, (2) a meteorology model used for providing meteorology input data, (3) a system of reusable classes, written in object-oriented Perl allowing fast input data processing and visualization, and (4) a semi-empirical BVOC emission model (seBVOC) written in Fortran 90.

The seBVOC implemented algorithm of BVOC emission is based on the approach presented by Guenther (1997) and discussed in detail by Stewart et al. (2003). For a given tree species i and chemical compound j the emission E_{ij} is estimated as

$$E_{ij} = A_i \times \varepsilon_{ij} \times D_i \times ECF_{T,\mathrm{PAR}}. \quad (1)$$

In Eq. (1) A_i is the area of the emitting plant species (in m^2), D the foliar biomass density (in $g\,m^{-2}$), ε_{ij} an average emission potential (in $\mu g\,g^{-1}h^{-1}$) standardised for 30 °C leaf temperature and 1000 $\mu mol\ m^{-2}\,s^{-1}$ photosynthetic active radiation (PAR) and $ECF_{T,\mathrm{PAR}}$ a dimensionless environmental correction factor. In the implemented algorithm two emission potentials are used (Steinbrecher et al., 1999; Shao et al., 2001): (1) synthesis emissions depending on both light and temperature and (2) pool emissions depending on temperature only. Thus, in a model run synthesis E_{synth} and pool E_{pool} emissions for each of the N land use categories and M chemical compounds are estimated (where applicable) and then summed up (Eq. 2):

$$E = \sum_{i=1}^{N}\sum_{j=1}^{M} E_{\mathrm{synth}_{ij}} + \sum_{i=1}^{N}\sum_{j=1}^{M} E_{\mathrm{pool}_{ij}}. \quad (2)$$

The BVOC emission model can be operated in a grid and in a point mode. The grid mode builds upon the mesh specification used in the Fifth Generation NCAR/Penn State Mesoscale Model (MM5) (Grell et al., 1993). Thus, meteorology data from a MM5 run can directly be used. For each grid cell of the modelling area, the area A of the emitting tree species, foliar biomass density D, leaf area index (LAI) and meteorological data on temperature and light intensity are required. The point mode which does not require an area A can be used to model BVOC emissions at a specific site with measured meteorology data.

In the model, a variety of options allow an appropriate treatment of the seasonality of emission potentials, canopy light extinction as well as the leaf temperature. In this study the sun/shade model of de Pury and Farquhar (1997) was used to simulate the light extinction within the canopy. The shaded and sunlit parts of the canopy are separated by taking into account the actual solar angle and the foliage density described by the LAI. The leaf temperature is calculated in an iterative process from the energy balance. A Quasi-Newton–Raphson method provides temperatures of the sunlit and shaded leaves (Dai et al., 2004). The seasonality of the emission potentials is simulated with a bell-shaped variation proposed by Staudt et al. (2000) and discussed in detail by Schaab et al. (2003). Using this approach, the seasonal emission potential adaptation factors for conifers increase from a winter base level of $0.2\,\mu g\,g^{-1}\,h^{-1}$ in January to the maximum of one in July. After that, they decrease again for 6 months to the January level. For deciduous plant species, potential emission is assumed to start in March with emerging leaves indicated by increasing seasonal adaptation factors from zero to the maximum in July and then again decreasing to zero in November. In winter, no leaves are present and thus the emission potential of deciduous trees is zero (seasonal adaptation factor of zero).

2.1. Modelling domain

The modelling of the BVOC emission has been performed in a grid approach with a $10\,km \times 10\,km$ grid at three stages: (1) country-wide, for the area of Germany with $357\,000\,km^2$ where 29.5% are covered by forests, (2) regional, for the federal state of Bavaria with an area of $70\,500\,km^2$ and a forest coverage of more than 35% forming almost 25% of the forest area in Germany, and (3) for an area close to Frankfurt/Main with $1600\,km^2$ and large proportion of high isoprene emitters (oak trees), and finally employing the point mode for a Norway spruce forest (Waldstein monitoring site) in the Fichtelgebirge, north-east Bavaria. At Waldstein site above canopy BVOC flux measurements have been performed within the integrated project *Biogenic emissions of reactive volatile organic compounds from forests: Process studies, modelling and validation experiments* (BEWA2000). The field data were used for model validation (Forkel et al., 2005).

Table 1
Foliar biomass densities D, leaf area index LAI and forest coverage

Name	D ($g\,m^{-2}$)	LAI	Forest area		
			Germany ($1000\,km^2$)	Bavaria	Frankfurt
Fir	1400	7.0	1.6	0.4	—
Scots pine	700	5.0	30.6	5.6	0.23
Spruce	1400	9.0	36.3	11.9	0.09
Oak	320	5.0	8.1	1.4	0.08
Beech	320	6.0	15.1	2.7	0.13
Larch	300	5.0	2.1	0.4	0.02
Douglas fir	1000	9.0	1.2	0.1	0.003
SWL[a]	300	5.0	5.6	1.1	0.04
SHL[b]	300	5.0	4.7	0.9	0.02
Sum	—	—	105.3	24.5	0.61

[a]SWL—Other soft wood deciduous: Birch, Aspen, Willow.
[b]SHL—Other hard wood deciduous: Maple, Ash, Hornbeam, Chestnut.

2.2. Forest data

The forest coverage data set has been compiled by Werner (2003) from results of the German Forest Inventory (Polley, 1993) as well as from data provided by various forest authorities. It depicts the percentage of nine forest categories (see Table 1) reported as polygon coverage. For using the data in the BVOC model, they have been disaggregated into grid cells of 10 km by 10 km. A general land cover data base from the CORINE programme (StaBuA, 1996) was employed to exclude nonforested areas. Finally, the forest area has been scaled (making up approximately 5%) to meet the latest forest coverage data available from the Federal Statistical Office, Germany (StaBuA, 2001). A Web Services-based system exposing GIS/RDBMS and data processing functionalities within a heterogeneous network is used to provide tailored input data for the BVOC model. This approach is discussed in detail by Smiatek (2005). Table 1 depicts the tree species distribution as well as associated foliar biomass densities and LAI values. In the presented BVOC inventories, country-wide best estimates of the foliar biomass density D and LAI for each tree species were used as more sophisticated data are not available yet. The LAI values used represent medians of ranges compiled by Breuer et al. (2003) and ORNL DAAC (2001), foliar biomass densities are from Simpson et al. (1999) and EEA (2003).

2.3. Emission potentials

Plant and chemical species specific potential emission factors used in this modelling exercise are based on public available databases (Kesselmeier and Staudt, 1999; Steinbrecher et al., 1999; Mannschreck et al., 2002; Wiedinmyer et al., 2004) as well as based on up-dates due to recent findings. In Table 2 plant and chemical compound specific emission factors are listed representing the forest-forming types in Germany. The emission factors for the forest classes soft wood (SWL) and hard wood (SHL) are calculated as mean values of the reported tree species and weighted according to the abundance of the tree species. Default values are used if there is an indication that a chemical compound is emitted but an emission factor is not available (e.g. Fall, 2003). This is mainly the case for chemical compounds other than the isoprenoids. Estimated default values may represent the lower limit of a

Table 2
BVOC standard emission potentials (emission factors) in $\mu g\,g^{-1}$ dry weight h^{-1}, normalised to 30 °C leaf temperature and $1000\,\mu mol\,m^{-2}\,s^{-1}$ photosynthetic active radiation (PAR) used in the modelling studies

Compound	Model	Fir	Pine	Spruce	Oak	Beech	Larch	Dgl.Fir	SWL[a]	SHL[b]
Ethene	pool	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
Ethane	pool	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02	0.02
Propene	pool	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
1-Butene	pool	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03	0.03
Isoprene	synth.	0.10	—	0.87	60.00	0.10	0.10	0.70	15.00	0.10
α-Pinene	pool	3.00	0.49	0.90	0.10	—	1.50	0.30	0.10	0.10
	synth.	—	0.22	0.90	—	—	—	—	—	—
β-Pinene	pool	—	0.22	0.07	0.10	—	—	0.60	—	—
	synth.	—	0.08	0.79	—	—	—	—	—	—
Limonene	pool	—	0.07	0.19	0.10	1.00	—	0.30	—	—
γ-Terpinene	pool	—	0.10	—	—	1.00	—	—	—	—
Formic acid	pool	0.01	0.01	0.02	0.01	0.03	0.01	0.01	0.03	0.03
Acetic acid	pool	0.02	0.02	0.02	0.02	0.03	0.02	0.02	0.03	0.03
Formaldehyde	pool	0.03	0.03	0.03	0.03	—	0.03	0.03	0.03	0.03
Acetaldehyde	pool	0.05	0.05	0.14	0.05	—	0.05	0.05	0.05	0.05
Propanone	pool	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Camphene	pool	—	0.06	0.04	—	—	—	—	—	—
	synth.	—	0.04	0.05	—	—	—	—	—	—
Δ^3-Carene	pool	—	0.46	0.05	—	—	—	0.30	—	—
	synth.	—	0.08	0.05	—	—	—	—	—	—
p-Cymene	pool	—	0.02	—	—	—	—	—	—	—
	synth.	—	0.20	—	—	—	—	—	—	—
Myrcene	pool	—	0.09	0.06	—	—	—	—	—	—
	synth.	—	0.16	0.12	—	—	—	—	—	—
β-Phellandrene	pool	—	0.10	0.08	—	1.00	—	—	—	—
Sabinene	pool	—	0.10	0.01	—	12.00	—	—	—	—
	synth.	—	—	0.09	—	—	—	—	—	—
Methanol	pool	1.00	1.00	0.10	1.00	10.00	1.00	1.00	1.00	1.00
Ethanol	pool	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
1,8 Cineol	pool	—	—	0.30	—	—	—	—	—	—
Hexenylacetate	pool	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
Hexenal	pool	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
Hexenol	pool	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
Hexanal	pool	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
Acetone	pool	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10	0.10
Isoprene	synth.	0.10	—	0.87	60.00	0.10	0.10	0.70	15.00	0.10
Sum Monoterpenes	pool	3.00	1.60	1.64	0.30	15.00	1.50	1.50	0.10	0.10
	synth.	—	0.42	1.88	—	—	—	—	—	—
Sum OVOC	pool	1.86	1.88	1.06	1.86	10.81	1.86	1.86	1.89	1.89

[a]SWL—Other soft wood deciduous: Birch, Aspen, Willow.
[b]SHL—Other hard wood deciduous: Maple, Ash, Hornbeam, Chestnut.

likely emission which has to be quantified by future measurements. For beech *Fagus sylvatica* L. latest experimental findings on the source strength of VOC indicate that this tree species is a strong sabinene (Holzke et al., 2006) as well as methanol emitter (Wildt, 2005). To account for these new results the respective emission factors for beech have been increased by a factor of 10 in a first approach. If needed, the chemical compound specific emission potential of a grid cell can be grouped according to reaction classes required in air chemistry models, e.g. reaction classes for the regional air chemistry model RACM. In this paper the potential BVOC emission is split into isoprene, sum of monoterpenes and other VOC emission.

2.4. Meteorology data

The massive parallel version of the Fifth Generation NCAR/Penn State Mesoscale Model (MM5) version 3.6 was used to model hourly estimates of temperature, humidity, wind speed and global surface radiation, correlated with PAR. In the model setup, meteorology data as well as radiosonde observations from NCAR Mass Storage System (MSS) and a multi-layer soil model were used. However, the land cover data have been replaced by an improved land cover map for Europe (LUE2.5; Smiatek, 2004). The monthly runs for the vegetation period ranging from April to end of October for the years 1994 until 2003 have been run on a NEC Linux cluster computer. Due to large demand of computational power, the runs were limited to the vegetation period. To show the influence of not considering the winter season in the yearly sum of BVOC, monthly emission rates for the whole year 2000 are presented.

3. Results and discussion

3.1. Emission inventories

The average BVOC emission from forest of the years 1994–2003 in Germany is in range of 366 Gg as calculated by the model. Compared to the anthropogenic VOC emissions of 1602 Gg (Federal Environmental Agency, 2003) in the year 2000, BVOC contribute to only 22.5% to the total annual VOC load. However, this contribution is likely to increase in the future as national efforts in reducing anthropogenic VOC emission are aiming at a value of 995 Gg in the year 2010 (European Parliament and Council, 2001). For the year 2000, the hourly emissions have been modelled for the whole year including winter months January to March, November, and December. The results show that only 0.1% of isoprene, 2.5% of monoterpene and 3.3% of other VOC (OVOC) emissions occur in the winter season. Thus, the emission of the vegetation period is viewed here as annual value.

The annual average isoprene emission in Germany was estimated to be 40 Gg contributing 11% to the total BVOC emission. This is only one third of the amount estimated in a previous study by Simpson et al. (1999). This difference is explained by the application of a light extinction module, necessary for accounting for forest canopy effects on the light distribution. It provides a more realistic PAR estimates at lower levels of the forest canopy (Geron et al., 1994). On the other hand, the sums of monoterpenes and other VOC are higher in the order of +20% with 188 Gg and 138 Gg, respectively. The higher emission factors for beech used here are the main reason for this increase as will be shown later by the sensitivity study. In Table 3, the 10 years annual mean values for Germany and each state are presented. In Germany, each square kilometre is emitting roughly one ton of BVOC per year. About 50% of that are monoterpenes. The state with the highest BVOC emission is Bavaria with about 90 Gg per year, followed by Baden–Württemberg with 59 Gg per year and Hesse with 36 Gg per year.

The spatial distributions of 10 years of annual average of isoprene, monoterpenes, other VOC and total BVOC emissions in Germany are shown in Fig. 2. In the warmer western parts of Germany where a high share of oak forests exists isoprene emissions dominate, whereas in the central part monoterpene emissions prevail as a result of conifer forests. The highest emissions occur in the forested mountainous central and southern parts of the country with peak emissions of over 2000 $kg\,km^{-2}yr^{-1}$. All mountainous areas in the central, southern and eastern parts of the country are strong monoterpene emitters with peak emissions over 1200 $kg\,km^{-2}yr^{-1}$. In some of these areas also emissions of other BVOC are observed. In total, the mountain areas in Germany emit the main part of BVOC in Germany. Based on these findings, it is clear that due to transport and photochemistry, in densely populated areas in the flat land such as the industrial areas Dortmund, Düsseldorf, Köln, Rhine-Main (Frankfurt) as well as Stuttgart, NO_x

Table 3
Estimated 10 years average VOC emission from forest in Germany in the period 1994–2003

State	Area ($1000\,km^2$)	Forests	Isoprene	Monoterpenes (1000 t)	Other VOC	Sum
Schleswig–Holstein	15.8	1.5	0.5	1.7	1.4	3.6
Mecklenburg-Western Pomerania	23.2	4.9	1.2	3.5	3.9	8.6
Lower Saxony	47.6	10.0	3.6	13.8	11.3	28.7
Hamburg	0.08	0.03	0.01	0.03	0.03	0.07
Brandenburg	29.5	10.3	0.9	13.0	13.7	27.6
Bremen	0.4	0.001	0	0	0	0.01
Saxony-Anhalt	20.4	4.4	1.0	6.2	5.1	12.3
Berlin	0.9	0.2	0.1	0.1	0.1	0.3
North Rhine-Westphalia	34.1	8.4	4.6	14.2	9.8	28.8
Saxonia	18.4	4.9	1.2	6.9	5.0	13.1
Hesse	21.1	8.4	4.5	18.4	13.4	36.3
Thuringia	16.2	5.2	1.4	12.1	7.8	21.3
Rhineland-Palatinate	19.8	8.1	6.1	14.6	11.2	31.9
Bavaria	70.6	24.5	9.0	48.8	32.4	90.2
Baden-Württemberg	35.8	13.6	5.4	33.0	21.4	59.8
Saarland	2.6	0.9	0.7	1.4	1.1	3.2
Germany	356.5	105.3	40.2	187.7	137.6	365.5

traffic emissions and BVOC may react producing high pollutant concentrations under certain meteorological conditions.

Monthly 10 year average isoprene, monoterpenes and other VOC emissions are higher in summer months when temperatures, light intensities, emission factors and emitting plant biomass are highest (Fig. 3). The variation with a span of approximately 80% from the 10 year mean value is observed in July, reflecting very different weather situations in the years from 1994 to 2003. The other months do not show such a pronounced variation. The monthly differences in the calculated emission rates do not only reflect the changed meteorological situation but also the seasonal increase and decrease in the emission factors and biomass (e.g. Staudt et al., 2000; Lehning et al., 2001). During summer, the monoterpenes are the dominating compound class emitted. At the beginning and the end of the vegetation period other VOC emissions and monoterpene emissions are almost equal. This changed ratio is primarily a result of the very small contribution of the monoterpene synthesis term to the pool emission of the overall emission of monoterpenes during April and October as a result of the small synthesis emission factor, low temperature and low radiation.

In the period 1994–2003 the variation in total VOC emission was studied on different levels of aggregation (see Fig. 4). Based on the 10 year average the observed variation for all three levels of aggregation, country, state, and region, is not greater than 20%. Cold years such as the years from 1996 to 2001 lead to lower emissions, hot years such as the year 2003 lead to higher emissions than the average. On small spatial scale, the observed variation of BVOC emissions is higher in general as on the country or state level. Surprisingly, the variation on state level is in several years less than on country level. A likely explanation for this feature is that Bavaria is a state with large forested areas (see Table 3). Forested areas may buffer somewhat the extremes in temperature and thus reduce the variation in the BVOC emission rates.

The year 2003 was a hot year, compared to the 10 year average. In this year, the period from 1 August to 10 August was extraordinary hot in Germany with temperatures reaching more than 40 °C at some locations. In Fig. 5 the effect of this extreme weather episode on the hourly BVOC emission is compared to the hourly 10 years average at the different levels of spatial aggregation. In this period BVOC were almost doubled compared to the mean values in Germany. In the industrialised area of Frankfurt, BVOC emissions were up to 140% larger than average. On the state level, the exceedance above normal was similar to the country level. Surprisingly, the amplitude of the diurnal exceedance in emission, with high values around noon and low values during the night was greater on country and

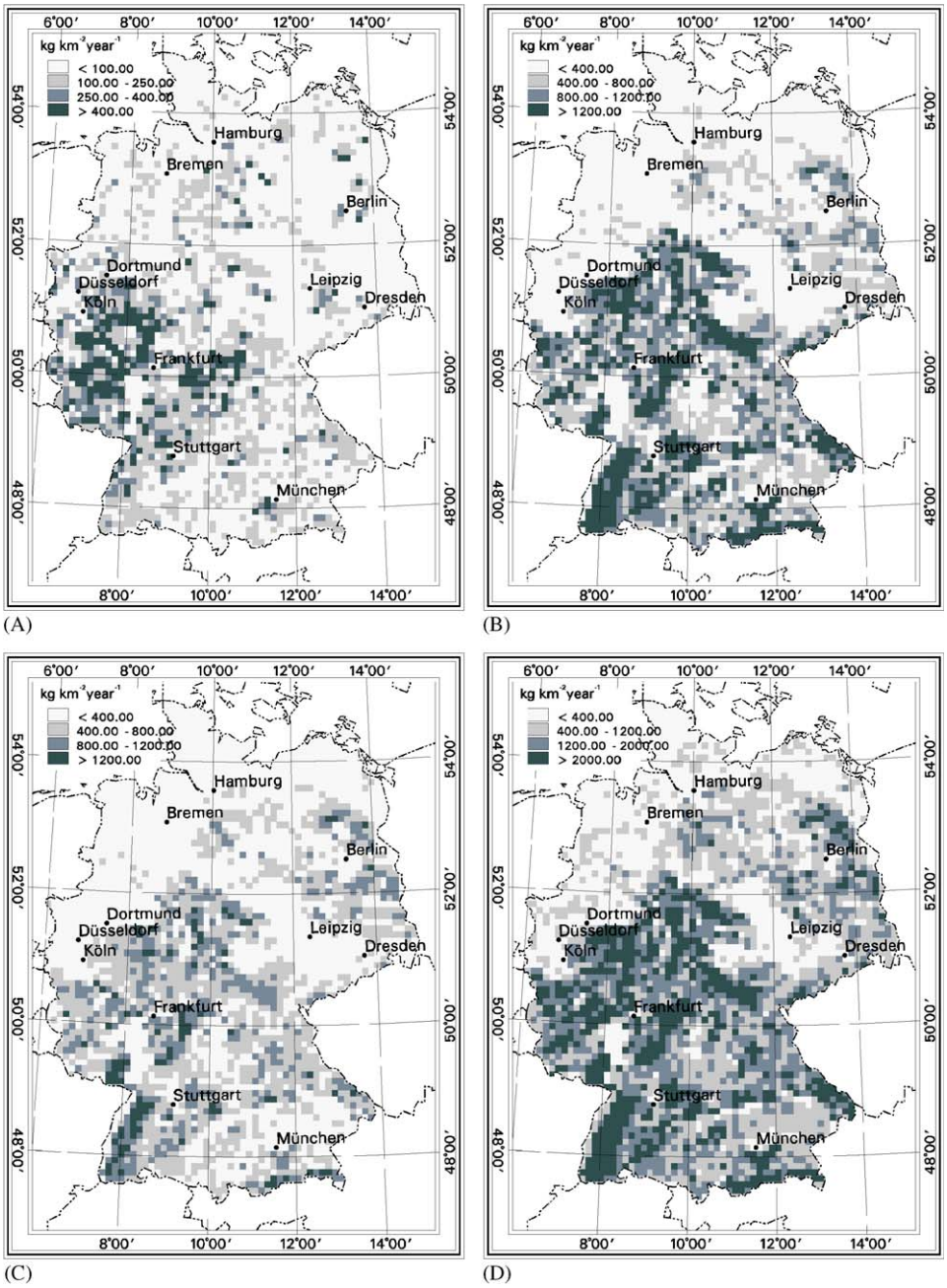

Fig. 2. Spatial distribution of the 10 years averages (1994–2003) for isoprene (A), monoterpenes (B), other VOC (C) and total BVOC (D) emission for Germany.

regional level as on state level. This finding may again be explained by the temperature buffering capacity of forested areas and thus reduced extremes in the daily temperature with the consequence of lower amplitudes in the daily VOC exceedances. As shown, regionally different weather conditions result in significantly different VOC emissions source strengths at different spatial and temporal scales. As expected, local scale phenomena are not reflected by country or state wide inventories.

3.2. Validation

The point mode of the seBVOC model has been applied to a site in north-east Bavaria, Germany,

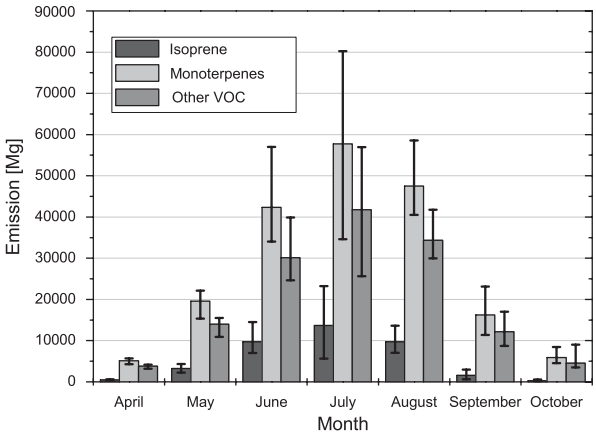

Fig. 3. Average, minimum and maximum monthly BVOC emissions from forests in period 1994–2003 in Germany.

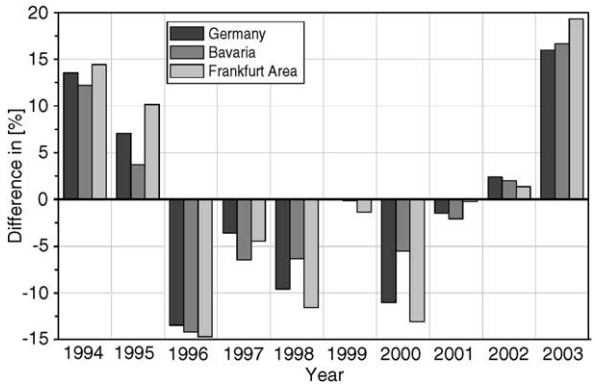

Fig. 4. Variability of the total annual BVOC emissions from forests from the 10 years mean (1994–2003).

known as the Waldstein monitoring site. This site is dominated by Norway spruce (*Picea abies* L. [Karst]), which is the tree species contributing to 40% and 55% to BVOC emission in Germany and Bavaria, respectively. Isoprene and monoterpene fluxes were modelled with measured temperature and PAR data. The forest at the Waldstein has a canopy height of approximately 18 m. The modelling results were compared with above canopy REA-PTR-MS measurements from Forkel et al. (2005) (see Fig. 6). The model catches well the measured diurnal cycles. If the model is run in the canopy mode considering light extinction through the canopy and leaf temperatures, the agreement between measured and modelled values is substantially improved. Taking into account that primary emitted BVOC undergo oxidation as well as deposition in the forest atmosphere reducing the potential canopy emission by about 20% (Forkel et al., 2005) measured and modelled values agree within a factor of less than two for isoprene and a factor of three for monoterpenes. This agreement range is well within the range of other down-/upscaling experiments (e.g. Guenther et al., 1999). It should be noted that the measured emission potentials of Norway spruce at the Waldstein site (Grabmer et al., 2005) are at the lower end of the values reported in literature ranging from 0.5 to 4.5 μg g^{-1} dry weight h^{-1} for monoterpenes and 0.4 to 1.5 μg g^{-1} dry weight h^{-1} for isoprene (Kesselmeier and Staudt, 1999;

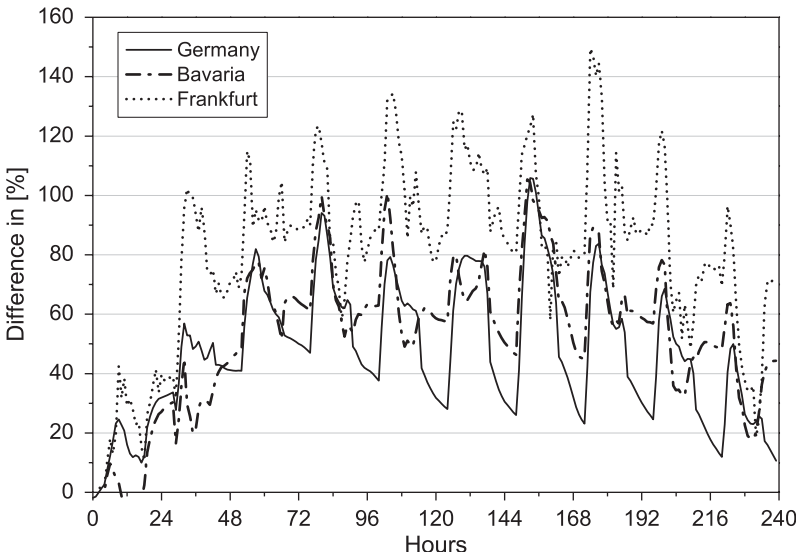

Fig. 5. Difference between the hourly 1993–2003 total BVOC emission mean and the hourly emission values in the episode 01.08.2003–10.08.2003.

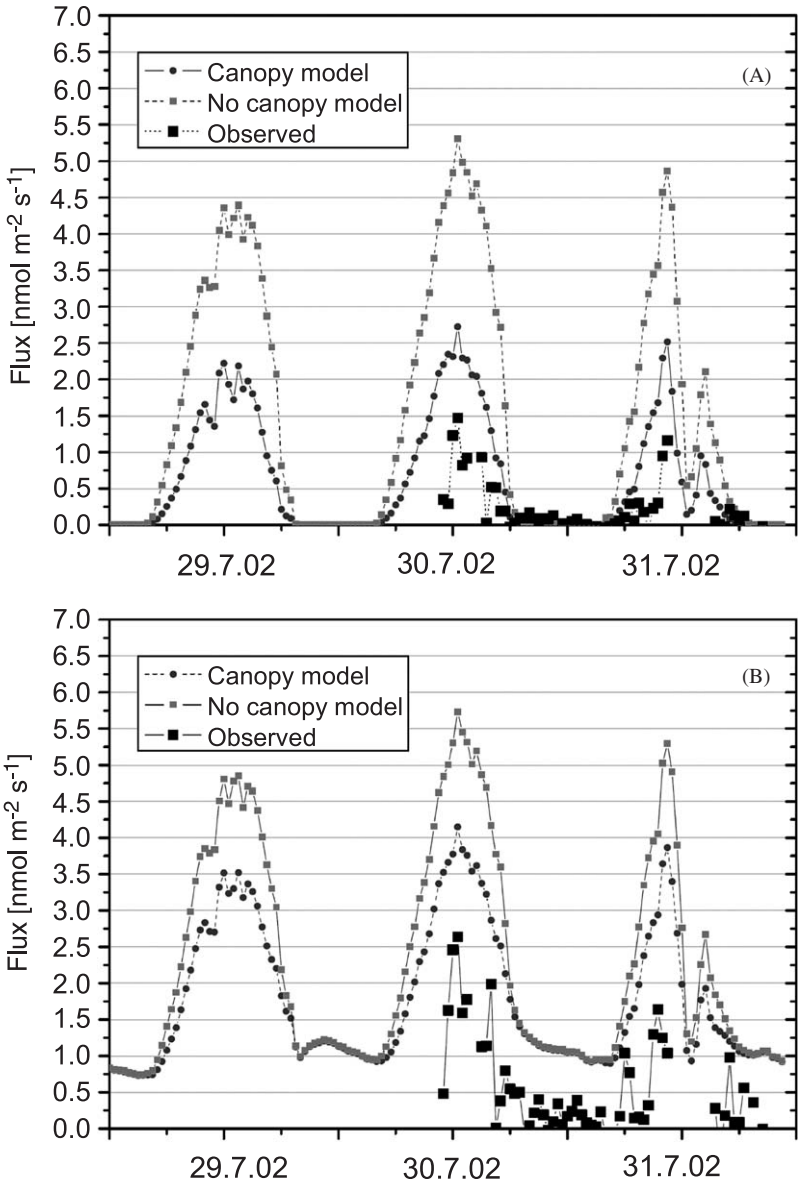

Fig. 6. Measured vs. modeled isoprene (A) and monoterpenes (B) fluxes at the Waldstein site, Fichtelgebirge, Germany.

Mannschreck et al., 2002). The model uses here mean emission potentials for Norway spruce being still higher than the site specific emission factors.

Therefore, the difference between the measurements and the modelled results at this site reflect more the natural variability of the BVOC emission from Norway spruce rather than deficits in the emission model. It also highlights the fact that a sound emission modelling at local scales needs to account for natural variation in emission potentials.

3.3. Sensitivity analysis

The sensitivity of the employed seBVOC model has been tested by re-calculating the hourly emissions of the year 1994–2004 for Germany using different scenarios by varying critical input data such as biomass density, temperature, and emission factors in a certain range. The results are summarised in Table 4. Scenario 1 is the base scenario. In scenarios 2 and 3 the variations of the foliar biomass density (D) have been set to $\pm 100\,\mathrm{g\,m^{-2}}$ for

Table 4
Sensitivity analysis of the emission model

Scenario	Name	Description	Isoprene	Monoterpenes	Other VOC	Sum
1	base	Parameters as described	n/a	n/a	n/a	n/a
2	minD	Foliar biomass density at minimum	−30.0	−28.8	−31.6	−29.9
3	maxD	Foliar biomass density at maximum	30.0	28.8	31.6	29.9
4	Fagus	Emission rate for sabinene and methanol	0	−25.7	−22.7	−21.9
5	$T+3\,^{\circ}$C	Air temperature increased by 3 °C	64.9	44.9	44.7	46.9
6	$T-3\,^{\circ}$C	Air temperature decreased by 3 °C	−59.2	−41.9	−36.1	−41.5
7	EIG	Emission rates for ISO, MT as given in the Emission Inventory Guidebook (EEA, 2003)	2.2	−36.2	−15.7	−25.4

The differences between the scenario and the base case are reported in (%).
n/a—not applicable.

deciduous broadleaved tree species and $\pm 300\,\mathrm{g\,m^{-2}}$ for coniferous trees following the uncertainty ranges compiled by Lenz (2001). The results show a linear decrease or increase of the emissions in range of 30%.

In scenario 4 the emission rate of sabinene for beech *Fagus sylvatica* has been set to $0.1\,\mu\mathrm{g\,g^{-1}\,h^{-1}}$ as available in the BVOC database (Mannschreck et al., 2002) from previous studies. The emission rate of methanol for the same tree species has been reduced to the default value of $1\,\mu\mathrm{g\,g^{-1}\,h^{-1}}$. Compared to the literature scenario the likely higher potential emission factors for beech result in 25% higher emissions of monoterpenes and 23% higher other VOC emissions. Taking into account this result, it becomes clear why 20% higher annual monoterpenes as well as other VOC emission estimates are reported here compared to a previous estimate by Simpson et al. (1999). Another factor of uncertainty which has not been discussed so far is the seasonality of emission factors. Methanol emissions may have a completely different seasonal cycle than other BVOC emitted. There is some indication that methanol emissions are exceptionally high during leaf development and tend to be very low when the leaf has fully developed (Nemecek-Marshall et al., 1995). This seasonal behaviour is in contrast, for example to the seasonal behaviour of the isoprene emission potential (Lehning et al., 2001). In general, knowledge about the source strength of methanol emission from plants is very limited, therefore methanol was not handled separately here. To further reduce the uncertainty in BVOC emission estimates, in particular for Germany, there is a need for sound new beech emission factors to be used in future emission calculations as well as elucidating the seasonal cycle of methanol emission from plants.

Scenarios 5 and 6 reflect the impact of the modelled temperature on the BVOC emissions. Modelled air temperature may have an uncertainty range of $\pm 3\,^{\circ}$C (Schaller, 2003). Generally, a temperature increase produces higher emission rates in a non-linear way. For example, a non-linear effect is important, particularly taking into account future climatic trends. This temperature effects are very pronounced for isoprene emission where a 3 °C increase results in a 65% increase in the emission. Monoterpenes and other VOC increase only by 45%. This different response of monoterpene and isoprene emissions to the same temperature change is due to different temperature parameterisations for isoprene and monoterpenes in the emission algorithms.

Finally, potential emission factors for isoprene, monoterpenes and other VOC have been set to values recommended by the Emission Inventory Guidebook (EEA, 2003) based on the available information for 1999. In this scenario 36% less monoterpenes and 15% less of other VOC are calculated compared to the values obtained here. This is mainly because of the increased emission factor for beech.

A still unresolved problem are causes of the natural variability of the emission factors of a tree species and how to account for this variability in an emission inventory. As an example, the effect of a possible range of emission factors for beech trees on the annual VOC emissions has been studied in detail. However, also other forest forming tree species show a variability of potential emission factors in the order of 10 (Kesselmeier and Staudt, 1999; Mannschreck et al., 2002; Komenda and Koppmann, 2002). The uncertainty in the emission potential of plant species accounts for about half of the overall uncertainty of a factor of four for e.g. an annual emission inventory of Great Britain (Stewart et al., 2003). This indicates that an emission inventory on what spatial scale ever can catch the "true emission" not better than within a range of a factor of 2. This statement is also supported by the results of the validation experiment.

4. Conclusions

A tool for estimating BVOC emissions with high temporal and spatial resolution highly related to land use type has been presented. A validation of the seBVOC model output with field data showed that the applied methodologies as well as input data sets are sufficiently accurate to provide sound BVOC emission estimates at least for isoprenoids. This latest inventory shows that annual isoprene emission was overestimated by about two thirds by previous emission inventories, but monoterpene emissions were comparable. In consequence air chemistry over Germany is less influenced by isoprene chemistry as thought before. The dominating part of BVOC chemistry in the lower troposphere is a result of emitted monoterpenes. This may lead to the hypothesis that perhaps a major fraction of secondary organic aerosol over Germany is resulting from biogenic monoterpenes.

The flexibility of this tool allows performing scenario studies with different input data set in various formats with different spatial and temporal resolutions. The 10 year model study highlights the year to year variation as well as the high spatial variation of the BVOC source strength and pattern.

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