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Biogenic volatile organic compound (VOC) emissions from forests in Finland

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We present model estimates of biogenic volatile organic compound (VOC) emissions from the forests in Finland. The emissions were calculated for the years 1995–1997 using the measured isoprene and monoterpene emission factors of boreal tree species together with detailed satellite land cover information and meteorological data. The three-year average emission is 319 kilotonnes per annum, which is significantly higher than the estimated annual anthropogenic VOC emissions of 193 kilotonnes. The biogenic emissions of the Finnish forests are dominated by monoterpenes, which contribute approximately 45% of the annual total. The main isoprene emitter is the Norway spruce (*Picea abies*) due to its high foliar biomass density. Compared to the monoterpenes, however, the total isoprene emissions are very low, contributing only about 7% of the annual forest VOC emissions. The isoprene emissions are more sensitive to the meteorological conditions than the monoterpene emissions, but the progress of the thermal growing season is clearly reflected in all biogenic emission fluxes. The biogenic emission densities in northern Finland are approximately half of the emissions in the southern parts of the country.

Introduction

The gas-phase chemistry of the troposphere is driven by the formation and destruction of ozone and other oxidants in photochemical reaction cycles involving volatile organic compounds (VOCs) and the oxides of nitrogen (NOx). The main sources of nitrogen emissions are traffic and stationary combustion, whereas VOCs are emitted to the atmosphere from various natural and anthropogenic sources.

At a global level, the anthropogenic VOC emissions are far outweighed by the biogenic emissions: according to Müller (1992) the total anthropogenic non-methane VOC emissions are about 150 Tg yr^{-1}, while Guenther *et al.* (1995) estimated the global natural VOC emissions to be of the order of 1 200 Tg(C) yr^{-1}. At the regional level, there is more variability in the relative emission rates. In the United States of America, the biogenic VOC emissions equal or exceed the anthropogenic emissions (Lamb *et al.* 1987, 1993), but in Europe the national emissions can be dominated by either category, depending on the land

use characteristics of different countries (Simpson *et al.* 1995, 1999). In the densely forested Nordic areas, especially in Finland, the biogenic VOC emissions are generally higher than the anthropogenic emissions (Simpson *et al.* 1999, Lindfors *et al.* 1995), and they can thus play an important role in the oxidant budget of these regions.

Due to the continuously increasing anthropogenic NOx and VOC emissions, photochemical ozone has become a common air quality problem in the summertime atmosphere of densely populated areas of Central Europe and the United States. It is estimated that e.g. in Europe 330 million people — almost half of the population — can annually be exposed to at least one incident of ozone concentration levels higher than the limit value set for human health protection, and that elevated ozone concentrations may cause several hundred hospital admissions per year (EEA 1998).

While the anthropogenic emissions of ozone precursors in Europe are known to a reasonable accuracy, especially in the countries with the largest emissions, there are huge uncertainties connected with the estimation of biogenic emissions (e.g. Simpson *et al.* 1997). The uncertainty of the best known biogenic VOC species, isoprene, can be as large as 500%, introducing a factor of five to ten uncertainty in model calculations of episodic ozone (Simpson 1995, Simpson *et al.* 1995, 1999), which are commonly used in the development of emission reduction scenarios. The uncertainties in biogenic emission estimates are partly due to an incomplete understanding of the emission processes themselves and the limited availability of experimental emission data, and partly due to the lack of explicit land cover information, which is essential in determining the geographical distribution and the total biomasses of the emitting vegetation types.

The emission characteristics of different plant species are currently the object of intensive research efforts. In Europe, two large measurement campaigns have been carried out in recent years: BEMA (Biogenic emissions in the Mediterranean area), which focused on biogenic emissions and their influence on tropospheric photochemistry in the Mediterranean region (e.g. Seufert *et al.* 1997), and BIPHOREP (Biogenic VOC emissions and photochemistry in the boreal regions of Europe), aimed at quantifying the biogenic VOC emissions from boreal forests (e.g. Laurila *et al.* 1997). However, there are still large gaps in the emission factor data available for detailed emission inventories, especially in remote ecosystems where the species distribution may differ considerably from the typical American or Central European vegetation.

In this work, we combine the most current information on the emission factors of boreal tree species with detailed land cover data, based on satellite measurements, to construct a forest VOC emission inventory for Finland. The results are discussed in the context of measured ambient VOC and ozone concentrations, and compared to available direct emission flux measurements. This work is an extension of the earlier emission calculations of Lindfors *et al.* (1995), with updated emission algorithms, emission factor data, and land use classification.

The role of biogenic VOCs in tropospheric chemistry

With respect to tropospheric photochemistry and ozone formation, the highly reactive monoterpenes ($C_{10}H_{16}$) and isoprene (C_5H_8) are considered to be the most important VOCs emitted in biogenic processes. Deciduous trees, such as oak, willow and aspen, are the main isoprene emitters, but there is now a growing amount of experimental data on isoprene emissions by certain spruce species (e.g. Steinbrecher and Rabong 1994, Kempf *et al.* 1996). Among them is the Norway spruce (*Picea abies)*, which is prevalent in the Finnish forests. The isoprene emissions are under enzymatic control, and they are strongly dependent on leaf temperature and light intensity (e.g. Fall 1999, Monson *et al.* 1995). A wide spectrum of terpenes is emitted by conifers (Tingey *et al.* 1991), and some Nordic birch species are also high monoterpene emitters (Hakola *et al.* 1998). The most common monoterpenes are α-pinene, β-pinene, Δ^3-carene and limonene. Terpene emissions are mainly regulated by leaf temperature (Guenther *et al.* 1993, Schuh *et al.* 1997, Hauff *et al.* 1999). Thus, unlike isoprene whose emissions are practically nonexistent at night, monoterpenes can also be emitted in the dark. In addition to the terpenoids, biogenic emissions contain numerous

other VOCs (OVOCs) such as alcohols, esters, ethers, aldehydes, ketones, alkenes and alkanes. Emissions from different tree species have characteristic VOC composition and strength. However, the emissions are also influenced by environmental factors and tree phenology, and there may be great variability in emission patterns between individual trees of the same genus and species, depending on their growing conditions.

Once emitted into the atmosphere, the biogenic VOCs react with ozone and hydroxyl and nitrate radicals with a timescale of minutes to hours (Atkinson, 1994). Depending on the NOx concentration, they may contribute to the formation of ozone on the regional scale, and influence the photochemical oxidant formation processes. They also produce organic acids, contributing to the deposition of acidity in rural and remote continental areas, and are involved in the generation of organic nitrates and the formation of organic aerosols. The main atmospheric oxidation pathways of isoprene are presently quite well understood (e.g. Carter 1996), and isoprene oxidation is included in several tropospheric chemistry models. Many aspects of terpene oxidation, however, are still unclear, although considerable progress has recently been made in characterizing their reaction products and aerosol formation potential (e.g. Calogirou *et al.* 1999, Griffin *et al.* 1999a, 1999b). The ozone forming ability of different hydrocarbons varies considerably, depending on their reactivities, but for example isoprene and α-pinene are known to be effective ozone precursors (e.g. Seinfeld and Pandis 1998).

Biogenic emission modeling

In Finland, the dominant tree species are pine (*Pinus sylvestris*, 64.5% of forest land area), spruce (*Picea abies*, 25.7% of forest land area), and birch (*Betula pubescens* and *Betula pendula*, 6.2% and 1.3% of forest land area, respectively), with minor contributions of other coniferous species, and deciduous trees such as aspen (*Populus tremula*), and alder (*Alnus* sp.) (FFRI 1997). In the northern parts of the country, the Norway spruce (*P. abies*) is partly substituted by Siberian spruce (*Picea abies* ssp. *obovata*) (Hämet-Ahti *et al.* 1992).

The relatively narrow tree species distribution simplifies the construction of the Finnish forest VOC emission inventory; however, it is only recently that comprehensive emission factor information has become available for these trees in the North European environmental conditions. Earlier emission assessments for Finland (Simpson *et al.* 1995, Lindfors *et al.* 1995), therefore, relied on emission factor databases (e.g. Geron *et al.* 1994, Guenther *et al.* 1994) compiled from measurements made in the United States and Central Europe, and often representing species and subspecies not found in the North European coniferous forests.

We based the estimation of the VOC emissions from forest foliage on the method described by Guenther (1997). The emission flux F per ground area (in $\mu g\ m^{-2}\ h^{-1}$) is given by

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

Here ε is the emission potential in $\mu g\ g(\text{dry weight})^{-1}\ h^{-1}$, D is the foliar biomass density in $g(\text{dry weight})\ m^{-2}$, and γ is a nondimensional environmental correction factor, which includes the effect of temperature and light conditions. The emission potential and the foliar biomass density are species specific properties, and they should be assessed individually for each tree genera and subspecies, in conditions representative of those in the actual ecosystems.

Biogenic emission potential

The emission potentials applied in this study are summarized in Table 1. Whenever practical, we adopted the recommended emissions for European ecosystems compiled by Simpson *et al.* (1999). In this work we also followed their recommendation that most tree species with no documented isoprene emissions should be assigned a minimum emission rate, because their isoprene emissions may be so low that they are not quantifiable, and to include the emissions of unaccounted-for vegetation within the forest area. Thus, the isoprene emission potentials of *Pinus sylvestris* and the *Betula* species were assumed to be 0.1 $\mu g\ g(\text{leaf biomass})^{-1}\ h^{-1}$. Since pine is the dominant tree species in Finland, this choice of emission factors is important for the emission calculations, and calls

for a closer inspection. According to comprehensive literature surveys carried out to compile the existing emission factor databases (e.g. Guenther *et al.* 1994, Kesselmeier and Staudt 1999, Simpson *et al.* 1999), no earlier studies reported significant isoprene emissions for Scots pine or the birches. This is confirmed by the new results of the BIPHOREP campaign, where emission factors of the main boreal trees were measured at forest sites in Finland and in Sweden. Hakola *et al.* (1998, 1999) consistently found the *Betula* species in southern Finland to be low or insignificant isoprene emitters. Steinbrecher *et al.* (1999) detected isoprene emissions from *Pinus sylvestris* both in central and northern Finland, but they were too low (detection limit ~1.6 ng g^{-1} h^{-1}) to be quantified. Janson and DeServes (1999) also observed isoprene emissions from *Pinus sylvestris* at all BIPHOREP measurement sites in Finland and in Sweden, but the emissions were always lower than 25 ng(C) g^{-1} h^{-1}. Thus, our use of the low default emission factors for *Pinus sylvestris* as well as *Betula pendula* and *Betula pubescens* appears to be well justified. The results of Hakola *et al.* (1999) and Steinbrecher *et al.* (1999) together with some other recent work (Kempf *et al.* 1996, and references therein, Janson 1993) were also extensively utilized to obtain the isoprene emission potentials of the known isoprene emitters (*Populus*, *Salix* and *Picea* species) and the monoterpene emission potentials of the Finnish trees given in Table 1. The emission potentials of OVOCs are highly uncertain (e.g. Kesselmeier and Staudt 1999), pending further experimental data, and therefore the default value 1.5 μg g(leaf biomass)$^{-1}$ h^{-1} (Guenther *et al.* 1995, Simpson *et al.* 1999) has been used for all species.

Foliar biomass density

According to Simpson *et al.* (1999), the default foliar biomass density of European deciduous trees is 320 g m^{-2}. The foliar biomass densities of *Pinus sylvestris* and the *Picea* species are highly variable with latitude. The values recommended to be used in areas north of 60°N are 500 g m^{-2} and 800 g m^{-2} for *Pinus* and *Picea* species, respectively (Simpson *et al.* 1999). However, the forest areas of Finland cover a wide range of latitudes (from 60°N to 70°N), and in this inventory we have taken into account the biomass variability within this region. From an analysis of the forest biomass data base created for the BIPHOREP project (Kellomäki 1999), it was found that the foliar biomass density of *Pinus sylvestris* varies between 400 g m^{-2} and 200 g m^{-2}, and that of *Picea abies* between 1250 g m^{-2} and 750 g m^{-2}, when going from southern to northern Finland (Lindfors *et al.* 1999b). This database was derived from the results of the permanent sample units of the 8th Finnish National Forest Inventory, which was carried out in the years 1986–1994. The permanent units are located systematically throughout the country, with a south–north and west–east distance of 16 km in southern Finland (from 60°N to 64°N) and 32 km in northern Finland (from 64°N to 70°N) (FFRI 1997, Kellomäki 1999). Each permanent unit consists of a cluster of 14 sample plots arranged along the sides of 1 500 m (S–N) by 1200 m (W–E) rectangle. For the BIPHOREP forest biomass database, only the units situated on upland mineral soils were selected, resulting in a total of 1 256 units of which 1 009 were in southern Finland and 247 in northern Finland

Table 1. The emission potentials [μg g(leaf biomass)$^{-1}$ h^{-1}] of dominant boreal tree species.

	Isoprene	Monoterpenes	OVOC
Deciduous trees			
Betula pendula and *Betula pubescens*	0.1[a)]	1.0[b)]	1.5[a)]
Populus tremula	43[b)]	1.0[b)]	1.5[a)]
Salix sp.	34[a)b)]	0.3[b)]	1.5[a)]
Coniferous trees			
Pinus sylvestris	0.1[a)]	1.5[a)]	1.5[a)]
Picea abies	1.0[c)d)]	1.5[c)d)e)]	1.5[a)]
Picea a. ssp. *obovata*	0.1[c)]	1.5[c)d)]	1.5[a)]

[a)]Simpson *et al.* 1999, [b)]Hakola *et al.* 1998, 1999, [c)]Steinbrecher *et al.* 1999, [d)]Kempf *et al.* 1996, [e)]Janson 1993.

(Kellomäki 1999).

In this work, we used a 10×10 km^2 grid analysis of LANDSAT satellite data to obtain the regional forest area coverage in different parts of Finland. The analysis was based on the LANDSAT TM image interpretation comprising over 50 land-use categories for 25×25 m^2 pixels. The LANDSAT database has been created mainly by the Finnish National Board of Survey and the Environment Data Centre of Finland (later part of the Finnish Environment Institute) in collaboration with several other institutes. The TM image interpretation was done using supervised classification based on linear discriminant function pairvise classifier over land surfaces and parallelepiped classification over water bodies (Vuorela 1992, 1997, 1999).

The LANDSAT forests are classified into pure pine, pure spruce, pure deciduous, and several mixed coniferous and mixed coniferous and deciduous forest types. According to the LANDSAT analysis, a majority (66%) of the forests in Finland are mixed, with the relative shares of pure pine, pure spruce, and pure deciduous forests 11%, 15%, and 8% of the total forest area, respectively. Besides the forest type, the LANDSAT forests are also assorted into five growing stock volume classes, ranging from < 50 m^3 ha^{-1} to >200 m^3 ha^{-1}. We used these volume classes to obtain an estimate for the foliar biomass in pure pine, pure spruce, and pure deciduous forest categories in different parts of the country. To convert the growing stock into total tree biomasses, we used the average densities 420, 380, and 480 g l^{-1} for pine, spruce, and deciduous trees, respectively (Kauppi *et al.* 1995). The average share of foliage of pine, spruce, and deciduous trees is 5%, 15%, and 4% of the stemwood mass, respectively (Kauppi *et al.* 1995), which gives us an estimate of their foliar biomass distribution over Finland. The average foliar biomass densities resulting from this analysis are given in Table 2. For pine and spruce these values are in good agreement with the biomass densities obtained from the analysis of the BIPHOREP biomass database (Lindfors *et al.* 1999b). The average spruce biomass density also agrees with the value recommended by Simpson *et al.* (1999), whereas the pine biomass densities in Finland appear to be significantly lower than the European average value of 500 g m^{-2}. The deciduous foliar biomass density obtained by the analysis of the LANDSAT data is of the same order of magnitude, but somewhat higher, than the average European value of 320 g m^{-2} recommended by Simpson *et al.* (1999).

In this emission inventory calculation we used the values given in Table 2 as representative of the foliar biomass densities in southern and northern Finland. The average values have been used in the central parts of the country (for the classification of the model regions into southern (South boreal), central (Middle boreal) and northern (North boreal) *see* Table 3).

Light and temperature correction

The environmental correction factor γ in Eq. 1 describes the diurnal variation of the biogenic VOC emissions. Several numerical algorithms have been developed to simulate the effect of light and temperature on isoprene and monoterpene emissions (e.g. Lamb *et al.* 1987, 1993, Guenther *et al.* 1991, 1993). In this work, we adopted the algorithms proposed by Guenther *et al.* (1993), which have been shown to perform extremely well when applied to different vegetation types and environmental conditions (e.g. Guenther *et al.* 1993, Guenther 1997, Simpson *et al.* 1999). According to this approach, the terpene emissions are controlled by the volatilization of hydrocarbons from storage pools inside the leaf (temperature control), while isoprene is emitted directly after it has been synthesized in the plant (light and temperature control).

The environmental correction factor for isoprene emissions is thus

$$\gamma_{ISO} = C_T \times C_L \qquad (2)$$

Table 2. The average foliar biomass densities (g m^{-2}) of the main Finnish tree species according to the LANDSAT growing stock analysis of the 'pure pine', 'pure spruce', and 'pure deciduous' forest categories (*see* text).

	Pine	Spruce	Deciduous
Southern Finland	300	900	400
Northern Finland	200	750	350
Country average	300	900	400

where C_T is the temperature correction and C_L is the light correction.

The light correction has the form

$$C_L = \frac{\alpha C_{L1} L}{\sqrt{1+\alpha^2 L^2}} \quad (3)$$

where L is the photosynthetically active photon flux density (PPFD, μmol photons m^{-2} s^{-1}), and α (= 0.0027) and C_{L1} (= 1.066) are empirical coefficients (Guenther 1997).

The temperature correction is given by

$$C_T = \frac{\exp\left(\frac{C_{T1}(T-T_s)}{RT_sT}\right)}{C_{T3}+\exp\left(\frac{C_{T2}(T-T_M)}{RT_sT}\right)} \quad (4)$$

Here T (K) is the leaf temperature, T_S is the leaf temperature at standard conditions (= 303.15 K), R is the universal gas constant, and C_{T1} (= 95 000 J mol^{-1}), C_{T2} (= 230 000 J mol^{-1}), C_{T3} (= 0.961), and T_M (= 314 K) are empirical coefficients (Guenther 1997). In this version of the emission model, the leaf temperature is assumed to be equal to the ambient temperature.

The environmental correction for terpene emissions is

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

where β (= 0.09 C^{-1}) is an empirical coefficient and T_S is the standard temperature given above. This correction factor is generally also used for OVOCs, because experimental data on the OVOC emissions is still too scarce to facilitate the development of specific emission algorithms (Guenther *et al.* 1994, Simpson *et al.* 1999).

Recently, it has been shown that the terpene emissions of some oak species are also light and temperature controlled (e.g. Steinbrecher and Hauff 1996, Seufert *et al.* 1997). In addition, Steinbrecher (1994) and Steinbrecher *et al.* (1999) suggest a light dependence in the terpene emissions of Norway spruce, indicative of both storage emissions and *de novo* synthesis. Oak is not an important tree in the Finnish forests, and since the above mentioned emission flux measurements of spruce are also well described by the pool model (Steinbrecher *et al.* 1999), we retained the simple idea

Table 3. Regional land cover information of Finland, based on the LANDSAT satellite data analysis. The geographical coordinates of the synoptic station (*see* text) are given for each N.U.T.S. area, and the boreal zone classification of the regions is indicated in parenthesis (S = South boreal, M = Middle boreal, N = North boreal, C = coastal).

NUTS Level 3 Area code	Synoptic station		Forest area, 1000 km^2		
	Lat. (°N)	Long. (°E)	Pine	Spruce	Deciduous
1 (S)	60.32	24.95	2.1	2.3	0.8
2 (S)	60.51	22.27	2.5	1.8	0.6
3 (S/C)	60.15	19.88	0.4	0.1	0.1
4 (S)	61.47	21.80	2.2	2.5	0.6
5 (S)	60.82	23.50	1.2	1.6	0.6
6 (S)	61.42	23.42	4.5	4.1	1.0
7 (S)	60.97	25.63	1.3	1.6	0.5
8 (S)	60.90	26.93	1.2	1.8	0.5
9 (S)	61.73	27.30	1.2	2.1	0.6
10 (S)	61.73	27.30	4.0	5.4	2.0
11 (S)	63.02	27.80	3.6	5.6	3.8
12 (M)	62.67	30.93	5.0	5.2	2.2
13 (S)	62.40	25.68	5.4	6.0	2.3
14 (M)	63.10	23.03	3.6	4.0	1.3
15 (M/C)	63.10	23.03	1.8	2.1	0.9
16 (M)	63.10	23.03	1.6	1.2	0.6
17 (M)	65.37	27.02	10.1	9.4	8.1
18 (M)	64.28	27.67	7.0	7.1	3.4
19(N)	67.37	26.65	27.3	21.9	15.6

of representing all terpene emissions by the γ_{TERP} algorithm in Eq. 5. Given the large overall uncertainties in both the emission factors and the land cover information, this simplification is not likely to introduce any discernible bias in the emission estimates.

Land cover and meteorological data

The 10×10 km^2 grid analysis of LANDSAT satellite data, obtained from the Finnish Environmental Information Center, as described above, was used to construct the Finnish land cover database for the emission model. For the database, all the LANDSAT forest categories were reallocated into pine, spruce, or deciduous model forest classes, with the allocation of mixed forests based on the assumption that mixed coniferous forests comprise of 54% pine and 46% spruce, and mixed coniferous & deciduous forests of 18% deciduous, 44% pine, and 38% spruce (Kauppi *et al.* 1995, FFRI 1997). The regional grouping was done on the basis of the N.U.T.S. (the *Nomenclature des Unités Territoriales Statistiques* of the European Union) Level 3 area classification of Finland (Fig. 1). Since average emission potentials and broad averages of foliar biomasses were used in this study, the geographical areas were chosen to be relatively large in order to smooth over any small-scale variability introduced by specific vegetation types or other local phenomena.

The regional forest coverage, calculated from the LANDSAT data as described above, is given in Table 3. The total analyzed LANDSAT forest area is about 218 000 km^2. According to the Finnish Statistical Yearbook of Forestry (FFRI 1997), the total forest land area of the country is 200 320 km^2 but this estimate does not include areas classified as scrub lands or waste lands, some of which may have been classified as different forest categories of the LANDSAT data set. The relative distribution of the model forest classes in Table 3 should not be directly compared with the forest area dominance percentages cited previously, as those refer to the dominant tree species in individual forest stands, thus showing the prevalence of the species but neglecting the contribution of the non-dominant trees which, however, is important for the emission calculations.

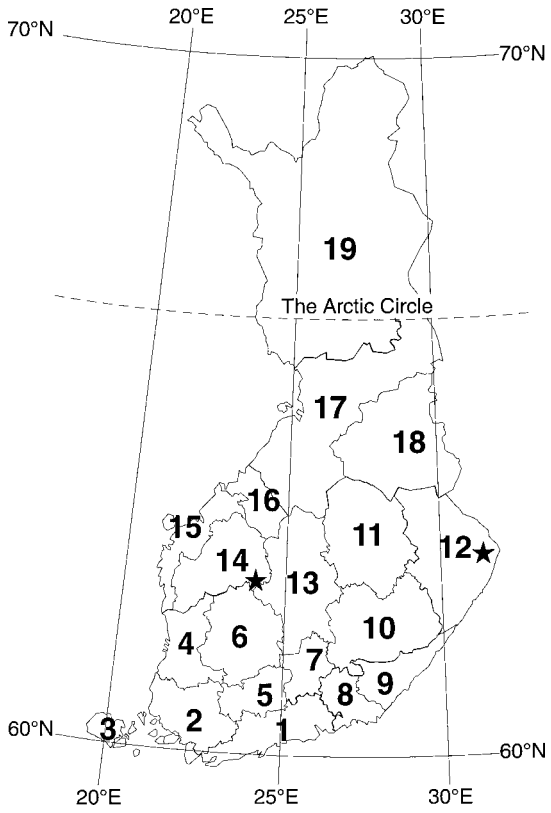

Fig. 1. The N.U.T.S. Level 3 area classification in Finland used in this work. The locations of the air quality monitoring station Ähtäri at the border of Areas 13 and 14, and the BIPHOREP campaign site at Mekrijärvi in Area 12 (*see* text) are indicated with stars.

A representative synoptic station was selected for each geographical area, and meteorological data was obtained for the modeling period (1 April–30 September) in 1995–1997 from the database of the Finnish Meteorological Institute. The three-hourly values of temperature, relative humidity, cloudiness, and wind speed were interpolated linearly to construct continuous time series of hourly meteorological data for each area. The geographical coordinates of the synoptic stations are also given in Table 3. The 19 regions were classified into the South, Middle and North boreal zones (assumed to represent the forests in southern central and northern Finland, respectively) according to Solantie (1990) and Ahti *et al.* (1968). The classification is indicated in Table 3.

Calculation of hourly emissions

The calculation of the hourly emissions from each region was done with the FMI/BEIS emission model. The model is based on the updated version of the Biogenic Emissions Inventory System (BEIS) developed at the Environmental Protection Agency (EPA) of the U.S.A. (Birth and Geron 1995, Pierce 1996, Pierce *et al.* 1998). For application with the Finnish emission inventory, the model code was rewritten to treat only the dominant boreal forest types, with two coniferous forest classes and one deciduous class. To facilitate the use of the LANDSAT land cover database, where the average fraction of allocated pure pine, spruce, or deciduous forests is less than 50% of the total forest area, we have developed species profiles for each forest type. The profiles take into account the mixing of species within coniferous and deciduous forests, according to the Finnish forest statistics (FFRI 1997). In the absence of more specific information about the relative abundances of the main *Picea* species in different parts of the country, we treated all spruce as *Picea abies*, except for the northernmost province (Lapland, area 19), where the spruce foliar biomass is assumed to be divided equally between *Picea abies* and *Picea abies* ssp. *obovata* (Hämet-Ahti *et al.* 1992). The deciduous tree types were divided into three classes, based on their isoprene emission potential, to reflect the relative share of high isoprene emitters (e.g. *Populus* and *Salix* sp.), low isoprene emitters (e.g. *Betula* sp.) and non-isoprene emitters (e.g. *Alnus* sp.). Deciduous forest was assumed to contain a small fraction of coniferous trees, and coniferous forests to have a share of deciduous species. The ensuing distribution of the total forest biomass among the forest classes and between the tree species are given in Table 4.

The emission fluxes of isoprene, total monoterpenes and OVOCs were calculated for each forest type according to Eq. 1

$$F_{\mathrm{ISO,TERP,OVOC}} = \varepsilon_{\mathrm{ISO,TERP,OVOC}} \times D_{\mathrm{ISO,TERP,OVOC}} \times \gamma_{\mathrm{ISO,TERP,OVOC}}(T,L)$$

using the environmental correction factors given in Eqs. 2 (isoprene) and 5 (monoterpenes and OVOCs). The FMI/BEIS model includes the original EPA/BEIS program code for the calculation of the photosynthetically active photon flux density (L, PPFD) from the cloudiness information of the hourly meteorological time series (Birth and Geron 1995). Measured PPFD can also be used as model input when available. A simple canopy model with five vertical levels is used to adjust the PPFD within the forest canopy (Geron *et al.* 1994).

Using the land cover information, we calculated the emissions for each of the 19 areas on hourly and daily basis over the model period (1 April to 30 September). Total annual emissions were obtained by summing the daily totals over the calculation period.

Results and discussion

Calculated emissions in 1997

The calculated total annual biogenic VOC emissions from forests in Finland in 1997 were 347 kilotonnes (Table 5). This is considerably higher than the average annual anthropogenic VOC emissions of 193 kilotonnes (Mroueh 1994). Isoprene, monoterpenes and OVOCs contributed 25, 154, and 167 kilotonnes, i.e. 7%, 45%, and 48% of the

Table 4. Contribution of deciduous and coniferous species in the model forest types, based on the results of the Finnish forest inventory (FFRI 1997). The deciduous species are classified into high isoprene, low isoprene, and non-isoprene emitters, as explained in the text.

Forest type	Deciduous species			Coniferous species	
	high-iso	low-iso	non-iso	pine	spruce
Pine	1.0%	16.0%	1.0%	82.0%	0.0%
Spruce	0.5%	10.0%	0.5%	0.0%	89.0%
Deciduous	3.5%	64.0%	3.5%	16.0%	13.0%

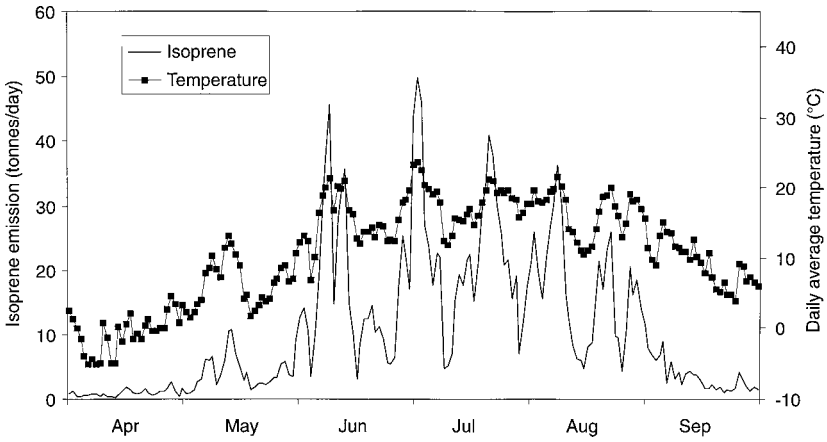

Fig. 2. Daily isoprene emission (left axis) and daily average temperature (right axis) in Central Finland (Area 13) during the modeling period in 1997.

total emissions, respectively. Coniferous forests appear to be the most important emitters, even with respect to isoprene, due to the large biomass of the low emitting spruce species. On average, the coniferous forests emit 80%–90% of the total biogenic VOCs in Finland (Table 5).

In Central Finland (Area 13), for example, the seasonal variation of isoprene emissions followed closely the temperature variability, with the warm spells during the summer clearly evident as emission maxima in the time series (Fig. 2). The sharper variability of the daily emissions reflects the great sensitivity of the isoprene emission mechanism to the environmental conditions (Eqs. 3 and 4). In the beginning and at the end of the model period, both the greater variability of the temperature (from below zero values at night to high afternoon values) and the reduced availability of solar

Table 5. Calculated biogenic emissions (kilotonnes per annum) from coniferous and deciduous forests in Finland in 1997.

Area	Isoprene		Monoterpene		Other VOC		Total VOC
	conif.	decid.	conif.	decid.	conif.	decid.	
1	0.7	0.2	4.4	0.4	4.6	0.6	11
2	0.6	0.1	4.0	0.3	4.2	0.4	10
3	0.1	0.0	0.4	0.0	0.4	0.1	1
4	0.8	0.1	4.8	0.3	5.0	0.4	11
5	0.4	0.1	2.8	0.3	3.0	0.4	7
6	1.2	0.2	8.0	0.5	8.4	0.6	19
7	0.5	0.1	2.8	0.2	3.0	0.3	7
8	0.5	0.1	3.1	0.2	3.3	0.3	8
9	0.5	0.1	3.3	0.3	3.5	0.4	8
10	1.4	0.3	8.9	0.9	9.3	1.2	22
11	1.5	0.6	9.6	1.8	10.1	2.5	26
12	1.3	0.3	8.6	1.0	9.0	1.3	21
13	1.6	0.4	10.4	1.0	10.8	1.4	26
14	1.1	0.2	7.0	0.6	7.3	0.8	17
15	0.6	0.2	3.6	0.4	3.7	0.6	9
16	0.4	0.1	2.3	0.3	2.4	0.4	6
17	2.0	1.0	14.9	3.3	15.6	4.4	41
18	1.5	0.5	11.3	1.4	11.9	1.9	29
19	2.2	1.5	25.6	5.0	26.8	6.7	68
Country total	18.8	6.2	136.0	18.4	142.4	24.7	347

radiation limited the daily emissions, even though the average temperatures were close to the mid-summer values. The average isoprene emission in Central Finland in 1997 was 11 tonnes per day, which corresponds to a flux of approximately 0.8 kg isoprene per km^{-2} of forest area per day. During the warmest days, however, the maximum emissions could be as high as 50 tonnes of isoprene per day, i.e. more than 3.6 kg per km^{-2} of forest area per day.

The N.U.T.S. regions with the largest forest areas were expectedly the largest contributors to the national total biogenic VOC emissions. Generally they are also the regions with the highest total land area. In order to compare the forest emissions in different parts of the country, we calculated the annual emission fluxes for each of the 19 regions (Table 6). The highest isoprene emission densities were found in the southern and central parts of the country. Emissions were somewhat lower in the coastal regions and in the easternmost parts of the country. In the northernmost areas the cooler climate restricted the emissions, with the isoprene emission densities approximately half and monoterpene and OVOC emission densities two thirds of the country average values.

When considering the latitudinal dependence of the emission densities, we found that between southern and northern Finland both isoprene and monoterpene emissions were reduced by approximately 50% (Fig. 3). The emission reduction as a function of latitude is almost linear, with the exception of Area 3 (latitude 60.32°N), which is an island in the Baltic Sea and differs from the other areas with respect to both climate conditions and vegetation type.

Interannual variability of biogenic emissions

A great interannual variability is introduced in the biogenic emission fluxes by changes in the meteorological conditions during the growing season. In Finland, these changes are accentuated by the considerable latitudinal span of the country (from 60°N to 70°N) (Table 7).

In 1995, the early spring was warm but there was a severe cold spell in May. The last week of May and all of June were warmer than average, but in July there was an exceptionally cold pe-

Table 6. Calculated annual (1 April–30 September) biogenic emission fluxes [tonnes km^{-2}(forest land)] in different parts of Finland in 1997.

Area	Isoprene	Monoterpenes	OVOC	Total VOC
1	0.17	0.92	0.99	2.08
2	0.15	0.87	0.93	1.95
3	0.10	0.65	0.70	1.46
4	0.16	0.96	1.02	2.14
5	0.16	0.90	0.97	2.03
6	0.15	0.88	0.94	1.97
7	0.16	0.92	0.98	2.06
8	0.18	0.97	1.03	2.18
9	0.16	0.93	0.99	2.09
10	0.16	0.86	0.93	1.95
11	0.16	0.88	0.96	2.00
12	0.13	0.76	0.82	1.72
13	0.14	0.83	0.89	1.87
14	0.15	0.85	0.91	1.91
15	0.15	0.84	0.90	1.89
16	0.13	0.76	0.82	1.72
17	0.11	0.66	0.72	1.49
18	0.12	0.73	0.79	1.63
19	0.06	0.47	0.52	1.05
Country average	0.12	0.71	0.77	1.59

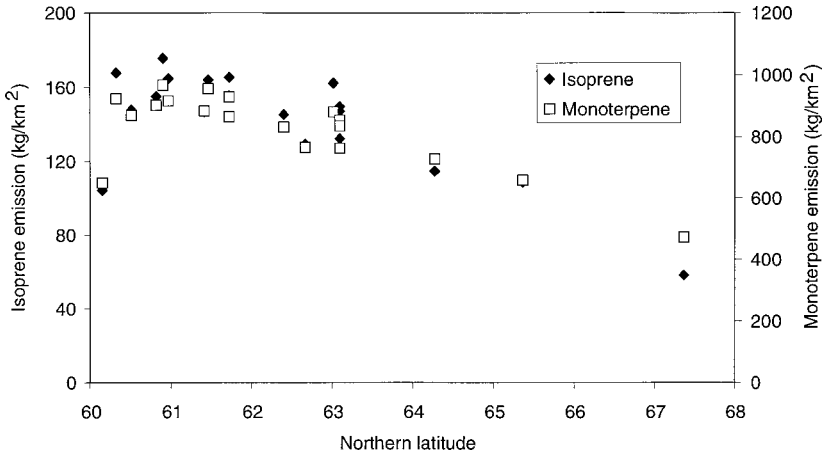

Fig. 3. Annual isoprene (left axis) and monoterpene (right axis) emission fluxes (emitted hydrocarbon mass per km^2 of forest land) in Finland in 1997 as a function of the latitude (°N) of the synoptic station used in the model calculations.

riod, which lasted for two weeks. The late summer was again warmer, as was the autumn, and in the southern parts of the country the thermal growing season extended all the way to October. The summer of 1996 was cold, rainy, and short. In the southernmost parts of the country, the thermal growing season progressed close to normal in spring, while being about a week late and two weeks late in the central and northern parts of the country, respectively. July was exceptionally cold in the whole country, while August was warmer than normal. In 1997, spring was late and the thermal growing season progressed very slowly due to severe cold spells in May. However, the whole summer was warmer than normal, with exceptionally high temperatures during the nights. The ther-

Table 7. Calculated annual (1 April–30 September) average biogenic emission fluxes [tonnes km^{-2}(forest land)] in Finland in 1995–1997.

	Southern Finland	Central Finland	Northern Finland	Country average
1995				
Isoprene	0.14	0.10	0.04	0.09
Monoterpene	0.86	0.69	0.44	0.66
OVOC	0.92	0.75	0.48	0.72
Total VOC	1.92	1.54	0.96	1.47
1996				
Isoprene	0.11	0.09	0.04	0.08
Monoterpene	0.76	0.64	0.40	0.60
OVOC	0.82	0.69	0.44	0.65
Total VOC	1.69	1.43	0.88	1.34
1997				
Isoprene	0.16	0.13	0.06	0.12
Monoterpene	0.90	0.75	0.47	0.71
OVOC	0.97	0.81	0.52	0.77
Total VOC	2.03	1.69	1.05	1.59
3-year average fluxes				
Isoprene	0.14	0.11	0.05	0.10
Monoterpene	0.84	0.70	0.44	0.66
OVOC	0.90	0.75	0.48	0.71
Total VOC	1.88	1.55	0.96	1.47

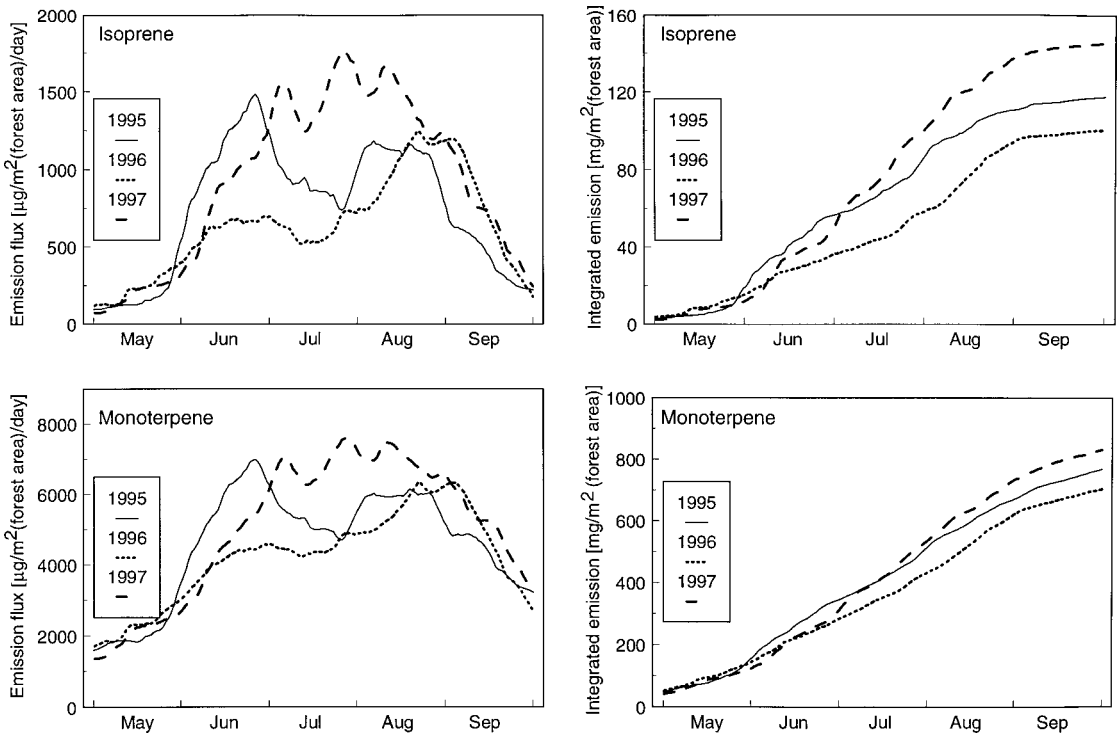

Fig. 4. Comparison of the 30-day running average emission fluxes (left-hand panels) and the integrated emissions (right-hand panels) of isoprene (upper panels) and monoterpenes (lower panels) in Central Finland in May–September 1995–1997.

mal growing season lasted until the third week of September in the north, and to the beginning of October in the central and southern parts of the country (statistics of the Finnish Meteorological Institute).

The total emission fluxes were the highest in 1997 and the lowest in 1996 in all parts of the country (Table 7). In Central Finland (Area 13), for example, the seasonal patterns of the emission fluxes and the accumulation of the seasonal isoprene and monoterpene emissions in 1995–97 reflected the meteorological variability (Fig. 4). The onset of emissions in the cold springtime conditions of 1995 was slow, followed by a strong emission period when the temperature increased in the end of May. The emissions were low during the cold summer of 1996, and there were intense emission peaks during the exceptionally warm summer of 1997 (Fig. 4). The integrated emission fluxes in Central Finland (Area 13) over the modeling period were 118, 100, and 145 mg of isoprene, and 768, 704, and 831 mg of monoterpenes per m^2 of forest area in 1995, 1996, and 1997, respectively. The corresponding accumulated temperatures (T > 5 °C) were 1 174, 1 034, and 1 273 degree-days in 1995, 1996, and 1997, respectively. Isoprene emissions appear to be more sensitive to the meteorological conditions than the monoterpene emissions: in 1997 the integrated isoprene emission flux in Central Finland was 45% higher than in 1996, while the difference in the integrated monoterpene fluxes between the warmest and the coldest year was only 18%.

Based on our model calculations, the total biogenic VOC emissions from the forests in Finland were 320, 291 and 347 kilotonnes in 1995, 1996 and 1997, respectively. The 3-year average emission was 319 kilotonnes per annum, of which approximately 7% was isoprene, 45% monoterpenes, and 48% other VOCs. These results can be compared with the national emissions given by Simpson *et al.* (1999), which are based on model calculations in the European scale. They estimate the annual biogenic VOC emissions from the Finn-

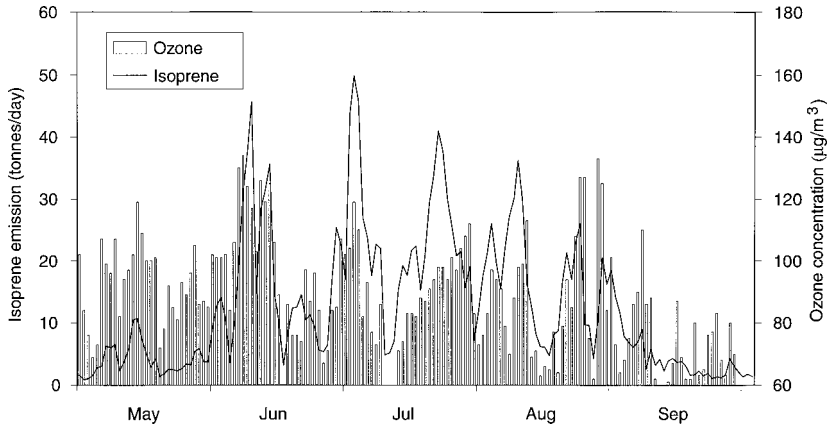

Fig. 5. Calculated isoprene emissions (left axis, solid line) in Central Finland (Area 13) together with the daily maximum ozone concentration (right axis, bars) measured at the background monitoring station in Ähtäri in May–September 1997.

ish forests to be 341 kilotonnes, consisting of 11% isoprene (39 kilotonnes), 48% monoterpenes (162 kilotonnes) and 41% other VOCs (140 kilotonnes). We obtained a slightly smaller proportion of isoprene, resulting in higher relative amounts of monoterpenes and OVOCs.

The main differences between these two modeling approaches are the treatment of foliar biomasses and the emission factors and emission algorithms applied for the spruce species. In our model the latitudinal variability of the coniferous biomass is included in closer detail as compared with the larger scale modeling of Simpson *et al.* (1999). In addition, we use the actual emission factors obtained in measurements in the Finnish environmental conditions and our isoprene emission model takes into account the distribution of spruce biomass between the low-emitting *Picea abies* and its practically non-emitting subspecies *P. abies* ssp. *obovata* in the northernmost parts of the country. Simpson *et al.* (1999) also apply the combined pool/*de novo* synthesis emission algorithm for the monoterpene emissions of *Picea abies*, which is likely to result in higher emission values.

Biogenic emissions and tropospheric ozone

When comparing the anthropogenic and biogenic emissions, one must take into account, not only the annual totals, but also the seasonal variation of the emissions. Biogenic emissions generally occur only during the growing season, while the anthropogenic emissions are distributed almost evenly throughout the year. Thus, the biogenic emissions, while practically nonexistent in winter, may exceed the anthropogenic emissions by as much as 400%–500% during the warm summer months. This enhances the importance of these highly reactive compounds in connection with the episodic high surface ozone concentrations, which are a ubiquitous phenomenon in the summertime atmosphere in Europe.

In Central Finland (Area 13), for example, the modeled isoprene emission peaks in 1997 occurred simultaneously with the elevated ozone episodes observed at Ähtäri, which is a background air quality monitoring station representative of the surface ozone levels in the region (Fig. 5). The elevated ozone concentrations are the product of complex photochemical processes in the atmosphere, often during transport over thousands of kilometers, and therefore it is not possible to connect the ozone episodes with the biogenic emissions without detailed photochemical modeling. However, it has been shown (e.g. Kuhn *et al.* 1999, Lindfors *et al.* 1999a) that substantial production of ozone and other photo-oxidants is possible when anthropogenic NOx emissions are introduced into the background of biogenic VOCs, and that under purely background conditions ozone is depleted by the biogenic compounds. As the biogenic emissions are highest under sunny high pressure conditions, which are also favourable for enhanced photochemical activity, it is possible that the high ozone forming potential of the biogenic VOCs could be realised also in the boreal zone.

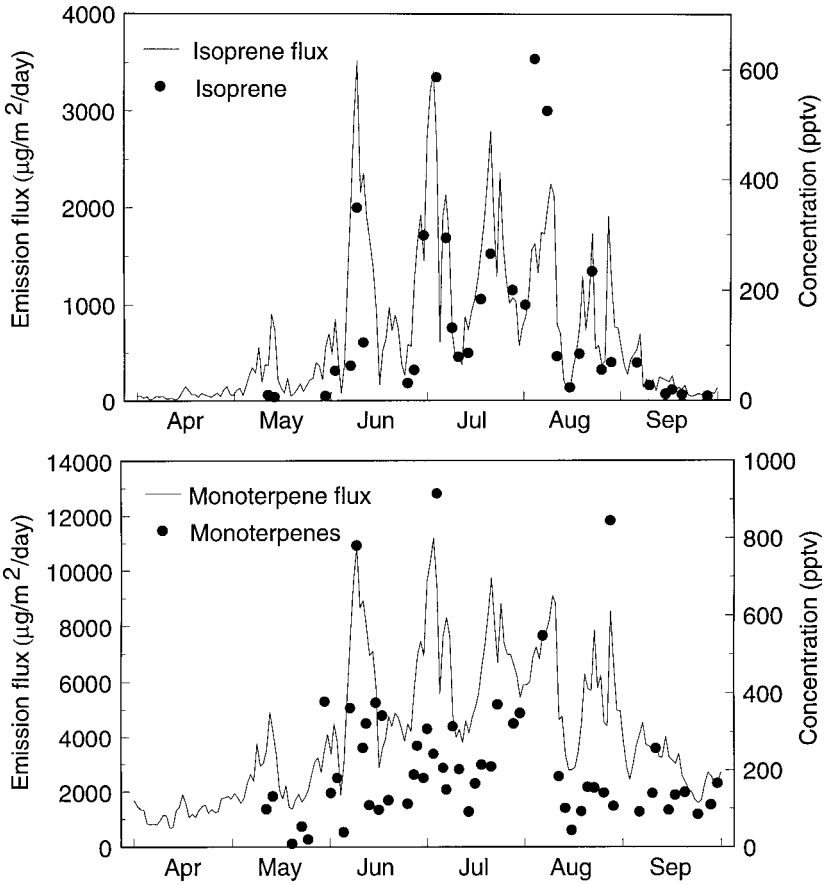

Fig. 6. Modeled isoprene (upper frame) and monoterpene (lower frame) emission fluxes (left axis, solid line) together with the respective measured concentrations (right axis, black dots) in North Karelia (Area 12) in May–September 1997.

Emission model validation and uncertainties

The validation of the emission model estimates is very difficult due to the scarcity of direct biogenic VOC flux measurements representative of the modeled time periods and the different geographical locations. Indirectly one can compare the variability of the modeled fluxes with that of observed biogenic VOC concentrations in background areas, where the local emission patterns are the main factor affecting the concentration fluctuations.

Comparison of the modeled isoprene and total monoterpene emission fluxes in North Karelia (Area 12) in 1997, and the concentration data obtained at the Mekrijärvi Research Station during the BIPHOREP field measurement campaigns (Laurila *et al.* 1999) showed that the variability of the atmospheric concentrations is quite similar to the modeled emission variability (Fig. 6). Both the duration of the intense emission period and the strong emission maxima are reflected in the measured concentrations. Unfortunately, no measurements were available prior to 15 May, and it was not possible to judge the performance of the emission model with respect to the onset of terpene emissions. However, it is expected that the isoprene emission model somewhat overestimates the early spring emissions. This is due to the model assumption of a constant biomass throughout the modeling period, which is clearly not the case with the deciduous species in the boreal zone, where the onset of leafing generally occurs in May–July, depending on the latitude. Furthermore, in 1997 spring was late and the thermal growing season did not start until 6 or 7 May in southern and central Finland (statistics of the Finnish Meteorological Institute). It was shown by Hakola *et al.* (1998, 1999) that in our environmental conditions the deciduous isoprene emitters do not begin to synthesize the compound until about two weeks after the leaves have opened. These growth-related

processes are not yet included in the FMI/BEIS emission model.

Our modeled monoterpene emission fluxes can also be compared with the flux measurements of Rinne *et al.* (1999) during the BIPHOREP campaign at Mekrijärvi. They found a mean total monoterpene emission rate of 0.19 $\mu g\ m^{-2}\ s^{-1}$ normalized to 30 °C, on 31 July and 2 August, 1997. Our modeled monoterpene emissions during these days were approximately 6 000 $\mu g\ m^{-2}\ day^{-1}$ (Fig. 6), with the afternoon temperatures around 20 °C. This corresponds to an average emission flux of 0.17 $\mu g\ m^{-2}\ s^{-1}$, when normalized to 30°C according to Eq. 5. Even though this is only a very rough comparison, the result is encouraging, considering the fact that the monoterpene emission algorithm as well as the emission potentials used in this study were greatly simplified, and that the terpene emission mechanism itself is less well understood than that of isoprene.

As repeatedly pointed out in the previous discussion, the uncertainties connected with biogenic emission modeling are large. Possible sources of error include both the land cover and meteorological data used as input in the model, the model algorithms themselves, and, above all, the biomass and emission factor data which are based on scattered experiments and sometimes coarse categorization (especially OVOCs). More uncertainty is introduced by the model assumption that both the emitting biomass and the emission potentials of the various species remain constant throughout the modeling period, and that the emissions are only dependent on temperature and solar radiation. For example, Hakola *et al.* (1998, 1999) found that the emission profiles of the boreal deciduous trees may change considerably during the growing season. In more southern conditions, Komenda and Koppman (1999) reported seasonal variation in the monoterpene emission rates of *Pinus sylvestris*, while Schade *et al*. (1999) suggested that pine (in this case *Pinus ponderosa*) emissions are also dependent on ambient humidity.

Conclusions

A biogenic emission inventory for the Finnish forests has been created using recent experimental data on emission factors of boreal tree species together with the well-established Guenther emission algorithms and satellite land cover data. The emission fluxes and the total annual emissions have a clear South–North gradient, with the VOC emission capacity of forests in northern Finland approximately half of that in the southern parts of the country.

Because of their larger biomasses coniferous trees dominate the biogenic VOC emissions from the forests in Finland. Compared with North American and Central European forests, the isoprene emissions of the boreal coniferous forests are low. The principal isoprene emitter in Finland is the low emitting *Picea abies*. According to our model calculations, the average biogenic VOC emissions from the forests in Finland are 319 kilotonnes per annum, consisting of 7% isoprene, 45% monoterpenes and 48% OVOCs. This estimate is close to the annual biogenic emissions of 341 kilotonnes (11% isoprene, 48% monoterpenes, and 41% OVOCs), given by Simpson *et al.* (1999). In an earlier work, Simpson *et al.* (1995) estimated that the annual isoprene emissions in Finland are 74 ± 23 kilotonnes (average of the years 1985–1991) and the OVOC emissions 354 kilotonnes (year 1989). While being significantly higher than the new emission estimates of Simpson *et al.* (1999), these values also surpass our present estimates of 21 kilotonnes of isoprene and 155 kilotonnes of OVOCs per annum. These differences reflect not only the effect of our much more detailed treatment of the land use and biomass distribution in Finland as compared with the European level analysis of Simpson *et al.* (1995), but also the recent advances in knowledge of the emission factors specific to the boreal tree species.

In the North European environmental conditions, climatological factors determine the annual variability and the accumulation of the biogenic emissions. Compared with Central Europe, the vegetation period is short, and the development of the thermal growing season is immediately reflected in both isoprene and monoterpene emissions.

A comparison of the modeled isoprene and monoterpene emissions and the observed ambient concentrations in background areas indicates that the emission model developed in this work

for the Finnish forests is capable of reproducing the seasonal variability of the biogenic emissions.

It is obvious that the modeled isoprene emissions are slightly overestimated in the beginning of the modeling period because the seasonal variability of deciduous biomass is not taken into account in the present model version. However, due to the strong dominance of the coniferous species at these northern latitudes, the error thus created is probably not critical to the total inventory, given the overall uncertainties related both to the emission potentials and the land cover information used to initialize the emission model. On the other hand, the seasonal behaviour of isoprene emissions by *Picea abies* has not been established experimentally, either, and it needs to be resolved before any conclusive judgement can be made about the emission model performance.

The few data available on direct emission flux measurements in Finland, obtained during the course of the BIPHOREP campaigns, compare well with the modeled terpene emissions. Considering the fact that the emission model is based on a very general selection of emission factors, and that the model was not adjusted to the local biomass or meteorological conditions at the BIPHOREP sites, it is encouraging that the modeled and observed emission rates are within the same order of magnitude.

The logical next step in terpene emission modeling will be the development of emission algorithms for each monoterpene species emitted by the boreal trees at various phases of the growing season. This will allow the creation of compound specific emission inventories which is important for the future assessments of e.g. the aerosol producing capacity of biogenic VOCs and the role of forests as regulators of the atmospheric chemical composition.

The great uncertainty connected with the OVOC emissions is the single largest drawback of the present biogenic emission models. While it is known that this category comprises of many reactive compounds, there is only scant data available of the OVOC emission potentials of trees or other vegetation within the forests (Kesselmeier and Staudt 1999, Wilske and Kesselmeier, 1999). Janson and De Serves (1998) reported light carbonyl emissions of comparable magnitude with the monoterpene emissions for *Pinus sylvestris* and *Picea abies* from measurements made during the BIPHOREP campaigns. The light aldehydes and ketones only account for part of the emitted compounds, however, and thus the total OVOC emissions of these trees may actually be much higher. Hakola *et al.* (1999) measured considerable linalool emissions from some birch species, but other than that there is very little quantitative information of the OVOC emissions of the main deciduous trees in the Finnish forests. In this inventory, we have adopted the 'generic' OVOC emission potentials with the result that almost half of the VOCs emitted by the forests belong to this bulk emission class.

In this work, we only modeled the emissions from the forests. While wetlands are generally considered important only with respect to methane emissions (e.g. Simpson *et al.* 1999), it has recently been shown that the boreal *Sphagnum* wetlands have substantial isoprene emission potential (Janson and De Serves 1998, Janson *et al.* 2000). The highest observed fluxes can be as high as 2 000 $\mu g\ m^{-2}\ h^{-1}$ which is about five times the maximum isoprene emission flux of a typical forest in southern Finland. Wetlands may thus also contribute significantly to the reactive VOC budget of the boreal regions and they should be taken into account in future emission inventories in these areas.

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