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Biogenic emissions in Europe

1. Estimates and uncertainties

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Abstract. Several biogenic volatile organic compound (VOC) emission algorithms have been used, together with meteorological data from the EMEP MSC-W ozone model, to generate estimates of the emissions of isoprene from European forests and agricultural crops over several summer periods. The most up-to-date estimate combines the recently updated isoprene emission factors from the United States with available knowledge of European tree species and emission factors. In some cases these European emission factors are significantly different from their U.S. equivalents because of differences in the tree species represented within a forest classification, especially with regard to spruce genera and Mediterranean oak genera. The new estimates have resulted in an approximate factor of 2–3 increase in isoprene emissions from northern Europe but a factor of 2 decrease in isoprene estimates for southern Europe. Overall, European isoprene emissions are estimated to be about 4000 kt C yr⁻¹, approximately 50–100% greater than previous estimates. Preliminary estimates are also made of the emissions of the so-called OVOC (other VOC) from forests and of soil NO_x emissions. All of these estimates of biogenic emissions are subject to considerable uncertainty, not least because of a lack of knowledge of the species coverage in most European countries and of the appropriate emission factors which should be applied. Factors of 5–10 uncertainty are not unlikely for episodic ozone calculations. The implications of these uncertainties for the results of control strategy evaluations for rural ozone in Europe are assessed in a companion paper.

1. Introduction

In the atmospheric boundary layer over industrialized continents such as Europe and North America, ozone is formed from chemical interactions of oxides of nitrogen (NO_x = NO + NO₂) and volatile organic compounds (VOCs). This ozone formation is driven by photochemically initiated reactions and is enhanced by high temperatures, so that high ozone concentrations are typically found in high-pressure meteorological situations, with clear skies and elevated temperatures [Cox *et al.*, 1975; Guicherit and Van Dop, 1977; Scholdager, 1984].

While most NO_x emissions in Europe and North America can be ascribed to anthropogenic sources, the situation for VOC is much less clear, as natural vegetation, primarily forests, can emit copious quantities of VOC species when temperatures are high (~30°C, say). These biogenic emissions consist of a wide variety of species, although attention is usually focused on isoprene and the class of monoterpene compounds (α -pinene, β -pinene, and limonene). The remaining species consist of a number of oxygenated compounds (alcohols, aldehydes) and have proven difficult to quantify in atmospheric samples. In the following we adopt the term “other”

VOC (OVOC) for these species, as used by Guenther *et al.* [1994a]. Of the biogenic VOC compounds, isoprene is one of the most reactive and in many parts of North America isoprene seems to play a significant and often dominant role among the VOC species in ozone formation [Chameides *et al.*, 1992; Trainer *et al.*, 1987]. The role of monoterpenes is not yet clear because these hydrocarbons react rapidly with both OH and ozone itself and are expected to be rapidly lost to aerosols [e.g., Johansson and Janson, 1993; Hov *et al.*, 1983]. In any case, available investigations indicate that terpenes have only a secondary effect on ozone formation in Europe compared to that of isoprene [MacKenzie *et al.*, 1991; Simpson, 1992; Simpson and Builjtes, 1991]. The OVOC compounds have received little attention in modeling studies to date, presumably because of difficulties in quantifying specific species, their expected low reactivity in comparison to isoprene, and the fact that no reaction schemes have been developed for many of these compounds. A review of the sources and chemistry of biogenic VOCs has recently been given by Fehsenfeld *et al.* [1992].

In this paper we concentrate mainly on the generation of isoprene emission inventories for Europe. However, we also present preliminary estimates of the emissions of OVOC from vegetation and of nitrogen oxides from soils. These estimates have been obtained using meteorological data from the Norwegian Meteorological Institute's numerical weather prediction model [Grønås and Hellevik, 1982; Grønås and Midtbø, 1986; Nordeng, 1986], which provide the basis of all the EMEP MSC-W Lagrangian models, including the MSC-W oxidant model [Simpson, 1992, 1993]. Solar radiation values have been calculated on an hourly basis. Isoprene emissions have been estimated using a number of different emission-temperature-radiation formulae and both with and without the use of a

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forest canopy model. Some of these estimates are presented to set previous work on biogenic emissions in Europe in context. One of the estimates, labeled E-93 below, represents the emission estimates as implemented in the EMEP model in 1993 and is the estimate used in the companion modeling study [Simpson, this issue]. The most recent estimate, E-94 below, has been compiled using data available by January 1995 and thus represents the current best estimate of isoprene emissions in Europe. The development of this latter estimate is of particular importance because so much new information has become available during 1994. New sets of emission factors for U.S. inventories were published [Geron *et al.*, 1994; Guenther *et al.*, 1994a], which gave significantly higher isoprene emission rates than those used by previous methodologies. However, new measurement campaigns conducted in Europe under the auspices of the BIATEX, FIELDVOC, and BEMA CEC-funded projects as well as national projects suggest that isoprene emission rates for some Mediterranean vegetation might be significantly lower than assumed previously [Steinbrecher, 1994; BEMA, 1994]. Further, measurements on the widespread Norway spruce species (*Picea abies* (L.) Karst.) also show low isoprene emissions [Steinbrecher *et al.*, 1993a; Steinbrecher and Rabong, 1994]. In light of these new data, we have attempted to generate a new set of emission factors which represent an up-to-date estimate of isoprene emissions for Europe.

In the companion paper [Simpson, this issue] we will present the results of a modeling study designed to assess the importance which isoprene emissions have for ozone formation in Europe. In particular, we will consider the implications that uncertainties in the emissions of isoprene have for the results of the model's control strategy predictions. We believe that both of these studies are necessary because so little work has been done to quantify the level and importance of biogenic VOC in European areas. Unfortunately, the large amount of research conducted in the United States on the magnitude and importance of biogenic VOC cannot be simply extrapolated to Europe because of the very different climatic, demographic, and biological conditions. For example, most American studies deal with ozone formation at temperatures in excess of 30°C, but such temperatures are only rarely reached in many parts of Europe. Average daily maximum temperatures in the United Kingdom, for example, are usually less than 20°C during July–August. Even as far south as Nice or Toulouse (~ 43°N), daily mean temperatures for these months lie just above 20°C, with a daily range of ~ 10°C [Wallén, 1970]. Possibly of even more importance, the types of tree species found within given genera can be very different in Europe to those found in the United States. Consequently, emission factors derived for U.S. forest classifications can sometimes be very inappropriate for European inventories.

2. Natural VOC Emission Estimates for Europe

Following the terminology of the biogenic emissions inventory system (BEIS) developed in the United States [Pierce *et al.*, 1990; Pierce and Waldruff, 1991], we can describe VOC emissions using the equation

$$ER_i = \sum_{j=1}^m [A_j \times AEF_{ij} \times ECF_{ij}(PAR, T)] \quad (1)$$

where ER_i is the emission rate of chemical species i , m is the number of vegetation categories used, A_j is the area of vege-

Table 1. Isoprene Emission Methodologies Used in This Study

Method	Description
E-90	Emissions exponentially dependent on temperature, as given by Lubkert and Schöpp [1991]. Isoprene emissions calculated for general deciduous category. No isoprene emissions from coniferous forests. No light dependence but nighttime emissions set to zero.
E-93	Temperature and light dependence from Guenther <i>et al.</i> [1991]. Emission factors derived from BEIS system for four European forest categories (oak, other broadleaf, low-isoprene spruce, other coniferous) and crops.
E-BEIS	As E-93 but temperature and light dependence from BEIS system [Pierce, 1991], based upon the work of Tingey <i>et al.</i> [1978a, b].
E-94	Temperature and light dependence from Guenther <i>et al.</i> [1993]. Emission factors derived for this study from recent (available 1994) U.S. and European literature for five forest categories (oak, other broadleaf, Sitka spruce, other spruce, other coniferous) and crops.
E-94.CPY	As E-94 but with the BEIS forest canopy model [Pierce and Waldruff, 1991; Lamb <i>et al.</i> , 1993]

tation class j , AEF_{ij} is the area-based emission factor of chemical species i for vegetation class j (standardized for full sunlight and 30°C), and ECF_{ij} (PAR, T) is a unitless environmental correction factor representing the effects of temperature and solar radiation (photosynthetically active radiation (PAR)) on emissions.

Briefly, the BEIS approach calculates hourly biogenic emissions from forest canopies and other vegetation as a function of temperature, sunlight (cloud cover), and the coverage of each vegetation category (e.g., oak, other broadleaf). It also includes an eight-layer canopy model for estimating profiles of temperature and sunlight within forest canopies [Lamb *et al.*, 1993; Pierce and Waldruff, 1991], taking into account also relative humidity and wind speed.

In the following work we compare a number of isoprene emission estimates derived using different assumptions concerning emission factors, temperature and radiation influences, and land use data. The different algorithms are summarized in Table 1, in historical order of development. The emission factors used in these algorithms are discussed in section 2.2.

Three of these methodologies represent different implementations of isoprene emission algorithms in the EMEP ozone model. Estimate E-90 has been used in most previous applications of the EMEP model [Simpson, 1992, 1993]. The emission-temperature relationship, as given by Lubkert and Schöpp [1989], was originally developed by V. Aalst (unpublished report, 1984) for the PHOXA modeling program [Buitjes *et al.*, 1988]. The forest cover database used for E-90 was also largely based upon that of Lubkert and Schöpp [1989].

Estimate E-93 was developed at the start of this biogenic emissions study and forms the basis of the companion modeling study [Simpson, this issue]. The emission factors were largely based upon the BEIS emission factors but taking into account the forest cover and biomass data compiled by Veldt [1989, 1991]. The temperature and sunlight corrections to emission rates were based upon the Guenther *et al.* [1991] formulae.

For comparison with other European biogenic emission estimates, E-BEIS was developed using the same emission fac-

tors and land use data as E-93 but with the BEIS ECF algorithms rather than the Guenther et al. relationship. For example, E-BEIS follows roughly the methodology proposed for the European Commission's CORINAIR database estimates [e.g., Ortiz and Dory, 1990; Veldt, 1988]. The LOTOS modeling work on biogenic emissions has also used similar emissions factors [Builjtes et al., 1993].

The E-94 estimates have been developed specifically for this study using the most recent knowledge. They are thus thought to represent the current best estimates of isoprene emissions in Europe. The new estimates represent a significant increase in complexity over the E-90 methodology used in previous EMEP reports [Simpson, 1992, 1993]. Although there are great uncertainties in even the more sophisticated approaches, the newer methodologies have several advantages:

1. The increased number of vegetation categories allows us to take account of the significant national differences in tree species. Thus the amount of oak (*Quercus* spp.), which is probably the most important isoprene emitter among European broadleaf species, is specified explicitly. As another example, the coniferous forests in most countries are not expected to be major isoprene emitters, but in the United Kingdom, the dominant coniferous species is the isoprene-emitting Sitka spruce (*Picea sitchensis* (Bong.) Carr.) [Hewitt and Street, 1992], and inclusion of this species will be shown to have a major effect on the isoprene emission estimates for the United Kingdom.

2. The high-temperature behavior of the isoprene emissions obtained from the Guenther et al. [1991, 1993] algorithms, and in the original BEIS algorithm, is almost certainly better than that of the earlier approach which featured an exponentially increasing isoprene emission with temperature. The Guenther et al. algorithms, in particular, have temperature dependencies which are related to biological processes and seem to represent a sound basis for predicting isoprene emissions over a wide temperature range.

3. Both the Guenther et al. and the BEIS approaches explicitly correct for the effect of solar radiation on isoprene emissions. This dependence on solar radiation has not been taken into account in previous estimates of European biogenic VOC emissions.

4. The forest canopy model developed within BEIS allows a reasonable first-order estimate of the importance of sunlight and temperature changes within a forest canopy. However, it should be noted that in particular, the estimates of temperature variation within a canopy are very uncertain and that in the latest Geron et al. [1994] inventory of U.S. emissions a canopy correction is only applied for solar radiation variations. Nevertheless, in section 3 we present estimates of European isoprene emissions both with and without the BEIS forest canopy model, in order to illustrate its importance. Each of the factors contributing to equation (1) is discussed separately below.

2.1. Vegetation Cover in the EMEP grid (A_j , Equation (1))

Forests. For each country, forest cover data have been generated using the data compilation of Veldt [1989]. This compilation gives areas of total forest per country, together with percentages of *Quercus* (oak), *Fagus*, *Betula* plus *Populus*, and other broadleaf, *Picea* (spruce), *Pinus*, *Abies* plus *Larix*, and "other" coniferous tree species. For this study, these classifications were further grouped into (1) oak, (2) other broadleaf, (3) spruce, and (4) other coniferous forest.

The area covered by these forest types are given for each

country in Table 2. The uncertainties associated with these figures are discussed in section 4.

The four categories were chosen in order to classify species in terms of their isoprene emission characteristics. Unfortunately, even with the species disaggregations given above, this assignment is not straightforward because of the very large variations in isoprene emissions even within trees of the same genus and because species mixtures vary considerably across Europe. The E-93 and E-94 inventories treat these categories in different ways. In the E-93 inventory, uniform isoprene emission factors are applied to each of these forest species regardless of country, except in the case of spruce (see below). The E-94 inventories are more complex, in that the isoprene emission factors for a category vary between countries also for the oak category. The reason is that in northern Europe the oak category is known to be dominated by the high-isoprene emitting species such as *Quercus robur* L., whereas in southern Europe, it is the very low isoprene-emitting species such as *Q. ilex* L. which dominate. In fact, Spanish data from Ortiz and Dory [1990] suggest that only ~ 10% of oaks are *Q. robur*.

In E-90 all coniferous species were assumed to be nonisoprene emitters. However, the later inventories take account of the fact that some spruce (*Picea*) species are known to be isoprene emitters. Sitka spruce (*Picea sitchensis* (Bong.) Carr.) is one such species, and it is estimated to constitute 81% of spruce forests in the United Kingdom [Hewitt and Street, 1992]. In the Netherlands and Denmark this species constitutes ~ 13 and 20%, respectively, of total spruce [Veldt, 1991; L. Mortensen, Danmarks Miljøundersøgelser, Roskilde, Denmark, personal communication, 1995]. However, this imported species is restricted to only a few, mainly coastal areas of northwest Europe. In Finland, Germany, Norway, and Spain this species is known to be present in only small or negligible amounts [BBELF., 1987, 1990; Ortiz and Dory, 1990; L. Kärenlampi, University of Kuopio, Finland, personal communication, 1994; S. Tomter, Norwegian Institute of Land Inventory, Ås, Norway, personal communication, 1995]. Other isoprene emitters include Engelmann spruce, and black, red, and white spruce [Evans et al., 1982; Evans, 1985; Novak and Reagan, 1986], but few data are available on the coverage of these species in Europe. More importantly, isoprene has recently been identified in the VOC emission pattern of Norway spruce, one of the most widespread species in the boreal forests [Steinbrecher et al., 1993a; Steinbrecher, 1994]. The assumptions made with regard to isoprene emissions from spruce are detailed in section 2.2. Within each country the national totals of each forest category, as derived above, were spatially distributed across the EMEP grid using the gridded database for the deciduous and coniferous forest of Lubkert and Schöpp [1989].

Agricultural crops. Areas of agricultural crops within each country, given in Table 2, were generally taken from the United Nations Economic Commission for Europe (UNECE) [1992], modified for the Mediterranean countries as described below. These national totals have been spatially disaggregated using the land use database of Chadwick and Kuylenstierna [1990].

The UNECE crop areas appear to match quite well independent data for the United Kingdom [Anastasi et al., 1991] and Denmark [Runge and Asman, 1989]. However, the UNECE figures are much greater than other estimates for the Netherlands [Veldt, 1988] and Spain [Ortiz and Dory, 1990]. The reason for this seems to be differences in the definitions of agricultural crops, grassland, and some types of forest. For

Table 2. Forest and Crop Cover (1000 km²) Used in the Calculations

	Total Forest	Oak	Other Broadleaf ^a	Spruce	Other Coniferous ^b	Crop
AL, Albania	13	4.9	7.2	0.0	0.8	5.5
AT, Austria	38	1.1	6.8	22.4	7.6	30.2
BE, Belgium	6.8	2.5	1.0	2.1	1.2	15.3
BG, Bulgaria	38	14.1	13.7	1.5	8.7	61.6
CS, former Czechoslovakia	46	4.1	12.4	20.2	9.2	67.8
DK, Denmark	4.8	0.4	1.4	2.1	1.0	29.2
FI, Finland	230	0.0	34.5	71.3	124.2	27.1
FR, France	150	57.0	42.0	7.5	43.5	319.5
DD, East Germany	26	1.6	4.4	5.5	14.6	62.3
DE, West Germany	72	6.5	18.0	26.6	20.9	133.6
GR, Greece	25	13.8	3.8	0.0	7.5	46.0
HU, Hungary	16	6.7	7.5	0.2	1.4	64.7
IE, Ireland	3.8	0.2	0.7	1.4	1.6	51.3
IT, Italy	80	26.4	36.8	5.6	11.2	97.5
LU, Luxembourg	0.8	0.1	0.5	0.1	0.1	1.3
NL, Netherlands	3.5	0.7	0.7	0.2	2.0	24.0
NO, Norway	87	0.9	23.5	38.3	24.4	11.1
PL, Poland	87	3.5	13.9	6.1	63.5	188.4
PO, Portugal	30	13.5	3.0	0.0	13.5	15.3
RO, Romania	65	14.3	33.8	13.0	3.9	151.1
ES, Spain	125	53.8	12.5	0.0	58.8	135.1
SE, Sweden	278	2.8	69.5	111.2	94.5	36.7
CH, Switzerland	11	0.2	3.1	3.9	3.9	18.9
TU, Turkey	201	80.4	10.1	40.2	70.3	248.1
SU, former USSR ^c	1570	94.2	486.7	533.8	455.3	897.7
GB, United Kingdom	22	1.1	8.1	5.1	7.7	161.0
YU, former Yugoslavia	105	19.9	67.2	8.4	9.4	71.0

All political units are defined after those existing in 1989.

^aIncludes *Fagus*, *betula* plus *populus*, and other broadleaf from Veldt [1989].

^bIncludes *Pinus*, *Abies* plus *Larix*, and other coniferous from Veldt [1989].

^cEuropean part of former USSR.

example, in Spain the UNECE gives a crop coverage of 271,000 km², whereas Ortiz and Dory give 113,000 km². It seems likely that areas of Monte hueco, low vegetation, olive trees, etc., which are included by Ortiz under forests are classified as agricultural areas in the UNECE document. As this seems likely to be a general feature of Mediterranean countries, we have accordingly reduced the UNECE estimates of these areas by a factor of 2, regarding the remaining area as being nonisoprene emitting.

2.2. Emission Factors for Biogenic VOC Emissions (AEF_{ij}, Equation (1))

For forested areas the area-based emissions factors (in $\mu\text{g C m}^{-2} \text{ h}^{-1}$) are derived as the product of biomass density (in g dry matter m⁻²) and standardized mass-based emission factors (in $\mu\text{g C g}^{-1} \text{ h}^{-1}$ at 30°C, full sunlight = 1000 $\mu\text{mole photons photosynthetically active radiation (PAR) m}^{-2} \text{ s}^{-1}$). The values of these parameters adopted for this study are summarized in Table 3 and described in more detail below.

Biomass densities. Estimation of the biomass densities which should be used in the model is rather difficult because of a lack of representative data for European conditions and major variations both between and within species, as a result of age, climate, etc. Veldt [1989] has reviewed the available U.S. and European data and arrived at default biomass densities for the major tree species. He suggests 320 g m⁻² for oak and 300 g m⁻² for other deciduous species. For comparison, Ortiz and Dory [1990] suggest a mean biomass density of 290 g m⁻² for Spain (although we should note that this mean value is derived from a mixture of oak species with 400 g m⁻² and other forest types, e.g., Monte hueco, with biomass densities as low as 100

g m⁻²). For coniferous species the corresponding values are much more variable. Veldt suggests densities of 700–1400 g m⁻² for different species < 60°N latitude, whereas Ortiz and Dory use 400 g m⁻².

The values chosen are given in Table 3. The choice of most of these figures has been rather pragmatic, using Veldt's data as a guide, but not attempting to distinguish much between different species or latitudes. Given the uncertainties in the available data, and more importantly the lack of data for most of the countries and tree species in Europe, this approach seems justified.

Table 3. Isoprene Emission Rates Adopted in this Study

Land Use Type	Biomass Density, g m ⁻²	Emission Rate, ^a $\mu\text{g C g}^{-1} \text{ h}^{-1}$		
		E-93 and E-BEIS	E-94.CPY	E-94
Northern Europe oak	300	14.69	70	40
Mediterranean oak	300	14.69	7	4
Other broadleaf	300	0.0	0	0
LI/Sitka spruce ^b	1400	3.0	10	6
Other Spruce	1400	0.0	1.75	1
Other coniferous	1000	0.0	0	0
Nondominant vegetation ^c	3–15% ^c	0.0	14	8

^aAt 30°C, full sunlight = 1000 $\mu\text{E m}^{-2} \text{ h}^{-1}$.

^bLI, low-isoprene spruce used in E-93 system, Sitka used in E-94; see text.

^cNondominant vegetation assumed to be 15% of broadleaf biomass, 3% of spruce biomass, and 5% of other coniferous biomass; see text.

Mass-based emission factors. The mass-based emissions factors for isoprene for the E-93/E-BEIS and E-94 inventories are given in Table 3. The emission factors vary substantially between the two inventory types, reflecting recent improvements in knowledge about the source strength of isoprene from different vegetation types.

The emission factors for isoprene for the E-93/E-BEIS inventories were largely based upon the BEIS values [Pierce, 1991] for oak, other broadleaf, and all coniferous forests except for some spruce species. In E-93 we defined a low-isoprene-emitting Spruce category, with a mass-based emission factor for isoprene of $3.0 \mu\text{g g}^{-1} \text{h}^{-1}$, consistent with the data of Evans *et al.* [1982] for Sitka spruce. Low-isoprene-emitting spruce (consisting of Sitka and other isoprene emitters) were assumed to make up 80% of U.K. spruce and 20% of northern European spruce. All other spruce (including all Mediterranean spruce) were assigned zero isoprene emissions.

In generating the new E-94 data sets, we have taken into account the upward revision in U.S. emission factors as given by Guenther *et al.* [1994a] and Geron *et al.* [1994]. However, we have modified the genera level emission factors from these studies to take into account the different distribution of species in European countries and to reflect recent European measurements from spruce (see below) and from the Mediterranean where low-isoprene emission rates were found for the oak species prevalent in southern Europe [BEMA, 1994].

As a result of these European measurements, we have developed separate emission factors for spruce, and for *Quercus* species in the Mediterranean area (here defined as Portugal, Spain, Italy, Albania, Greece, former Yugoslavia, Turkey, and southern France $<45^\circ\text{N}$). Other European areas will hereafter be referred to as northern European areas.

Emission factors for northern European oak in E-94 have been taken directly from Guenther *et al.* [1994a], as it is the deciduous species such as *Quercus robur* which seem to dominate this category in both the United States and northern Europe. Following the notation of Guenther *et al.*, this emission is expressed as $70(40) \mu\text{g g}^{-1} \text{h}^{-1}$, where the first number gives the emission rate for a fully illuminated leaf (to be used in connection with a canopy model, i.e., in E-94.CPY) and the number in parentheses gives the branch level emissions, which are used where no canopy model is available, i.e., in E-94. European measurements are very few but seem comparable, especially given the wide range of emissions expected between different measurements. For example, the branch-level emission rate of $40 \mu\text{g g}^{-1} \text{h}^{-1}$ above is the median of a range of 20–60. For 3-year-old trees of *Q. robur* branch standard emission rates of $40 \mu\text{g g}^{-1} \text{h}^{-1}$ were calculated on the basis of climate chamber experiments [Steinbrecher *et al.*, 1993b]. Isidorov *et al.*'s [1985] measurements on a 10-year-old *Q. robur* in Russia gave two values in sunny conditions: $24.3 \mu\text{g g}^{-1} \text{h}^{-1}$ at 19°C and $32.6 \mu\text{g g}^{-1} \text{h}^{-1}$ at 25.5°C (fresh weight). Assuming the temperature correction of Guenther *et al.* [1993], we obtain standard (30°C) emission factors of $\sim 60\text{--}100 \mu\text{g g}^{-1} \text{h}^{-1}$ (fresh weight).

For the Mediterranean countries it is assumed that 10% of the oaks are “high” emitters (70,40) and 90% do not emit isoprene [cf. Ortiz and Dory, 1990], giving an emission factor for Mediterranean oaks of $7(4) \mu\text{g g}^{-1} \text{h}^{-1}$. This reflects the prevalence of such oak species as *Q. ilex* or *Q. suber* which are known to be extremely low emitters of isoprene [Steinbrecher, 1994; BEMA, 1994].

The other broadleaf species encompass the categories Fa-

gus, betula plus populus, and “other broadleaf” given by Veldt [1989]. The Fagus and other broadleaf categories can be assumed to be nonisoprene emitters consistent with data in the work of Guenther *et al.* [1994a]. The grouping betula plus populus has been given the same zero emission rate as the other broadleaf genera, based upon data for the United Kingdom, Finland, and Norway which show that the nonisoprene emitting betula genera are much more extensive than the isoprene emitting populus genera. Except in northern Europe the coverage of this category is less than that of oak, so in any case, national isoprene emissions will not be very sensitive to any errors in the assumed betula/populus distribution. The practical result then is that all nonoak broadleaf categories are treated identically as nonisoprene emitters.

Recent data also suggested a revision of the spruce emission factors for E-94. For U.S. landscapes, Guenther *et al.* [1994a] assign an emission rate of $14(8) \mu\text{g g}^{-1} \text{h}^{-1}$ for all spruce species. Such high emissions do not seem appropriate for the majority of European spruce. Of particular importance, it is the species Norway spruce which dominates the landscape of Scandinavia, Germany, and other northern European countries. Recent measurements in the field on Norway spruce have shown that this spruce species is a low-isoprene emitter with an emission factor of $1 \mu\text{g C m}^{-2} \text{h}^{-1}$ (range 0.46 to 1.54) [Steinbrecher, 1994]. For the majority of Spruce forests in Europe then we have adopted an emission factor of $1.75(1) \mu\text{g g}^{-1} \text{h}^{-1}$.

However, recent measurements in Sitka Spruce plantations in north west England and Scotland, and in greenhouse experiments, suggest standard emission factors of $2\text{--}7.5 \mu\text{g g}^{-1} \text{h}^{-1}$ [Street, 1995; Boissard *et al.*, 1995], similar to those found by Evans *et al.* [1982]. As discussed in section 2.1, this species is very important in the United Kingdom and is also found in Denmark and the Netherlands. We have therefore treated Sitka spruce as a separate category in E-94 and assigned a “moderate” emission factor of $10(6) \mu\text{g g}^{-1} \text{h}^{-1}$ to this category. Coverage of Sitka spruce is based upon the data discussed in section 2.1: 81% for the United Kingdom, 20% for Denmark, and 13% for the Netherlands.

For the “other coniferous” forest categories we have adopted a default emission rate of $0 \mu\text{g g}^{-1} \text{h}^{-1}$ in all emission algorithms, consistent with results from Steinbrecher *et al.* [1993b] and Guenther *et al.* [1994a].

It should also be recognized that there are many species which are isoprene emitters but which are not accounted for in the above scheme. For example, eucalyptus is a high-isoprene emitter [Guenther *et al.*, 1991] which is important in Portugal and Spain [Nunes *et al.*, 1995; Ortiz and Dory, 1990]. However, incorporation of the emissions of such species on a European scale will require a much more extensive data set than that available to the authors. Also, in addition to the dominant trees in each class there will be other trees and shrub species, some of which are isoprene emitters. There is little information on European forests on the extent and importance of such nondominant vegetation, although it is more likely to be more important in broadleaf than in coniferous forest. Following Guenther *et al.* [1994a], we assume that nondominant vegetation has an emission rate of $14(8) \mu\text{g g}^{-1} \text{h}^{-1}$ and represents 15% of the total biomass for broadleaf forests, 3% for spruce forests, and 5% for other coniferous forest. Per forest category we thus obtain emission factors of: broadleaf, $2.1(1.2) \mu\text{g g}^{-1} \text{h}^{-1}$; spruce, $0.42(0.24) \mu\text{g g}^{-1} \text{h}^{-1}$; and other coniferous, $0.7(0.4) \mu\text{g g}^{-1} \text{h}^{-1}$. It should be noted that in a screening study of nondominant plants in northern European forests

in a climate chamber, *Steinbrecher et al.* [1993b] found no significant isoprene emission, so at least in northern Europe this assumed emission factor may represent the upper limit of emission factors for nondominating woody species.

Very little reliable experimental data on the emissions of OVOCs are available, and consequently, *Guenther et al.* [1994a] recommended the use of a uniform emission rate of $1.5 \mu\text{g g}^{-1} \text{h}^{-1}$ for all tree species. To make a preliminary, first-order estimate of OVOC emissions in Europe, we have applied the same emission rate to all European trees.

Emissions from crops are, as suggested by *Veldt* [1991], with standardized area-based emission factors of $8 \mu\text{Cg m}^{-2} \text{h}^{-1}$ for isoprene and $12 \mu\text{Cg m}^{-2} \text{h}^{-1}$ for OVOC. It may be that the area-based emission factor for isoprene is an upper limit for crops since *Evans et al.* [1982] and *Steinbrecher et al.* [1993b] report only low emissions. As other nonforested areas are not thought to make a significant contribution to isoprene emissions, we have not included these in our estimates.

2.3. Environmental Correction Factors (ECF_{ij} (PAR, T), Equation (1))

The environmental correction factor describes the relationship between the emission rate of biogenic VOC as a function of meteorological parameters: ambient temperature, sunlight (actually PAR), and possibly other conditions. A major complication in this relationship is that biogenic emissions are actually driven by the temperature and sunlight conditions appropriate to each leaf, rather than by the values of temperature and sunlight external to the forest canopy.

In the simplest approach we can assume that temperatures and sunlight levels within a canopy are identical to ambient levels. This is referred to below as the noncanopy approach and is used in E-90, E-BEIS, E-93, and E-94. It is worth noting that the emission factors given in Table 3 for these algorithms are “branch”-level factors, so do include the effects of shading to some extent.

In the more complex “canopy” approach of E-94.CPY, we have made use of the forest canopy model developed within BEIS [*Pierce and Waldruff*, 1991; *Lamb et al.*, 1993]. This approach uses an eight-layer forest canopy model to estimate leaf temperature and sunlight levels throughout the forest canopy, as a function of ambient temperatures, sunlight, wind speed, and relative humidity outside of the forest. This model has been adapted to run with input temperature and radiation data from the EMEP system, allowing the calculation of emissions from each EMEP grid square throughout the summer. (In this work we have set fixed values of wind speed (5 m s^{-1}) and relative humidity (60%), as experiments have clearly shown that these parameters have only a secondary influence on isoprene emissions compared to those of temperature and radiation.)

Regardless of whether we use a noncanopy or canopy approach to obtain temperature and sunlight levels, there are also a number of alternative formulae available for predicting isoprene emissions as a function of leaf temperature and sunlight. The formulae adopted for the E-94 estimates is that of *Guenther et al.* [1993] in which isoprene emissions are expressed as a function of light and temperature through

$$\text{ECF}_{\text{isoprene}} = C_L C_T \quad (2)$$

where the light dependence C_L is defined by

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

Figure 1. Environmental correction factors (ECF): normalized relationships between isoprene emissions and temperature according to the formulae used in the E-90, E-BEIS, E-93, and E-94 algorithms. The canopy model results for E-94.CPY represent values obtained by running the forest canopy model with different combinations of latitude (40° – 60°N in steps of 5°), wind speeds (1 – 15 m s^{-1} in steps of 4 m s^{-1}), relative humidities (30 – 90 , in steps of 10), hourly from 0900 to 1500 UT, and averaging. It was found that all of the above combinations give similar ECF values.

and temperature dependence C_T is described by

$$C_T = \frac{\exp \frac{c_{T1}(T - T_0)}{RT_0 T}}{1 + \exp \frac{c_{T2}(T - T_M)}{RT_0 T}} \quad (4)$$

where L is the PAR flux in $\mu\text{mole m}^{-2} \text{s}^{-1}$, R is the gas constant ($=8.314 \text{ J K}^{-1} \text{mole}^{-1}$), and α ($=0.0027$), c_{L1} ($=1.066$), c_{T1} ($=95000 \text{ J mole}^{-1}$), c_{T2} ($=230000 \text{ J mole}^{-1}$), and T_M ($=314 \text{ K}$) are empirical coefficients. T_0 ($=303 \text{ K}$) is the standard temperature at which emission factors are specified.

Figure 1 illustrates the emission temperature relationships (environmental correction factor (ECF) values) used in the various inventories. For the E-94.CPY case we have calculated a typical canopy modification, accounting for both light and temperature variations, by running the canopy model over a range of meteorological conditions and for a range of latitudes (see figure text).

The BEIS formulae for emission-temperature-sunlight relationships have been based upon the work of *Tingey et al.* [1978a, b] and in various forms have been widely used in inventory development and modeling work [e.g., *Builtjes et al.*, 1993; *Roselle et al.*, 1991; *Veldt*, 1991]. The formula used in E-93 was developed by *Guenther et al.* [1991] and is quite similar to the more recent *Guenther et al.* [1993] formula used in E-94. Hereinafter, we refer to these formulae as “G91” and “G93.”

Comparing the formulae in Figure 1, one notable feature is the much larger isoprene emissions predicted by the E-90 formula at high temperatures, compared to those of the BEIS/*Guenther et al.* methodologies. Examination of the original data upon which all these factors were based [e.g., *Veldt*, 1988] does not help in resolving these discrepancies, as the scatter in the measurement data does indeed cover an order of magni-

tude. However, more recent measurements suggest that at temperatures in excess of $\sim 40^\circ\text{C}$, isoprene emissions begin to decline, as described by the G91, G93 formulae (see *Guenther et al.* [1991, 1993] for references). This behavior corresponds also to that expected from biological considerations, as enzymatic activity in the plant is hindered at high temperatures. At temperatures below 20°C the G93/G91 formulae leads to a significant reduction in isoprene emissions with decreasing temperature. This behavior fits measured emissions at low temperature [e.g., *Evans*, 1985; *Street*, 1995] better than Tingey's, but this is not surprising when we consider that 20°C was the lowest limit used in Tingey's experiments. As noted earlier, mean summertime temperatures in much of Europe lie below or around 20°C , so we can expect European emission estimates to be much more sensitive to the behavior of the emission curves at low (sub- 20°C) temperatures than corresponding American studies.

Considering the effects of the forest canopy model on isoprene emissions for the E-94 estimates, we see that emissions are significantly reduced when temperatures are high ($>25^\circ\text{C}$), caused by the reduction in radiation within the canopy. However, at more moderate temperatures ($<25^\circ\text{C}$) both E-94 and E-94.CPY have similar ECF values. This effect occurs when the calculated temperature increase inside the upper portion of the canopy outweighs the effect of reduced radiation in the lower portion.

For the preliminary estimate of OVOC emissions the formulae recommended by *Guenther et al.* [1993, 1994a, 1995] and *Geron et al.* [1994] is used:

$$\text{ECF}_{\text{OVOC}} = \exp[\beta(T - T_s)] \quad (5)$$

where β ($= 0.09$) is an empirical coefficient derived from several data sets.

2.4. Biogenic NO_x Emissions

Emissions of NO from soils might make an important contribution to NO_x levels in remote areas [*Skiba et al.*, 1992; *Simpson et al.*, 1990; *Stocker et al.*, 1993; *Williams and Fehsenfeld*, 1991]. For this preliminary estimate of European emissions the emission formulae of *Pierce and Novak* [1991], based upon the work of *Williams and Fehsenfeld* [1991] and *Williams et al.* [1992], is used:

$$\text{flux (ng N m}^{-2} \text{ s}^{-1}) = C \exp(0.071T_s) \quad (6)$$

where C is an experimentally derived coefficient for each land use category and T_s is soil temperature ($^\circ\text{C}$). In the work of *Pierce and Novak*, C varies from 9.0 as an upper bound for agricultural areas to 0.004 for wetlands, and in addition, the dependence of soil temperature T_s on ambient temperature varies with land categories. For grassland and pasture, *Pierce and Novak* suggest $C = 0.9$, and the formula $T_s = 0.67 T_a + 8.8$ (T_a = ambient temperature in $^\circ\text{C}$), valid for $15 < T_s < 35$, $T_a > 15$ ($^\circ\text{C}$).

For simplicity we have adopted the grassland/pasture values for all nonforested land areas. For forest areas we have used the *Pierce and Novak* values directly: $C = 0.07$, $T_s = 0.84 T_a + 3.6$. Uncertainties are discussed in section 4.

3. Results, Biogenic Emission Estimates for Europe

The data presented here have been generated from the emission algorithms listed in Table 1, applied together with

meteorological data from the EMEP MSC-W modeling systems. The data used consist of surface (2 m) temperature and cloudiness for each $150 \times 150 \text{ km}^2$ grid square of the EMEP grid, with a 6 hourly resolution. Solar radiation fields, based upon spatial location, time of year, and the above meteorological data, are calculated every hour for these estimates.

Isoprene. Table 4 gives the calculated isoprene emissions for April–September 1989, generated using the five different methodologies. Man-made VOC emissions for 1989 are also shown for comparison.

Comparing first the E-93 with the E-94 inventories, it is clear that the revision in emission factors has resulted in large changes in estimated emissions. For northern European countries the E-94 estimates are considerably higher than the E-93 values, typically by a factor of about 2.5. On the other hand, emissions from Mediterranean countries are typically a factor of 2 lower in E-94 than in E-93. The differences in these estimates follow almost entirely from the different emission factors used (Table 3). Overall, the revised E-94 procedure has resulted in approximately 50% more isoprene emission than E-93 and about 100% more isoprene than the E-BEIS estimate.

Comparing the E-90, E-BEIS, and E-93 methodologies, we see that in most cases the different estimates are within a factor of 2. The biggest difference occurs for the United Kingdom, and this can be simply ascribed to the inclusion of some spruce trees as low-isoprene emitters in the E-93/E-BEIS methodology.

On a yearly basis we see that isoprene emissions are approximately 20% of man-made emissions across Europe as a whole, totaling $\sim 4000 \text{ kt yr}^{-1}$. Half of these isoprene emissions come from the forests of the former USSR, although the E-94 estimates clearly represent a large fraction of reactive VOC emissions in many countries. As shown in previous EMEP work [*Simpson*, 1992, 1993], isoprene emissions are much more important during the warmest months. Figures 2a and 2b present an updated version of the monthly mean emissions presented by *Simpson* [1992, 1993], now including estimates from all five biogenic emission algorithms.

Comparing the canopy versus noncanopy predictions in Table 4, we see that in most cases the use of the forest canopy model does not give dramatically different results from simple application of the branch-level emission factors and G93 formulae directly. In a few cases the differences are about 30%. In most cases the canopy model gives higher emissions than the noncanopy calculation, with the exceptions being the Nordic countries. The exact relationship between the canopy and the noncanopy model results obviously depends on the climate, as the forest canopy has two main opposing effects on emissions. First, the canopy shields part of the foliage from sunlight, tending to reduce emissions. On the other hand, the leaf temperatures inside the canopy tend to be higher than ambient temperatures, leading to an increase in emissions. For most countries the combination of the higher emission factors used in E-94.CPY compared to E-94 (Table 3) and this temperature effect is enough to offset the reduced PAR levels within the canopy.

The calculated emissions of isoprene obtained from method E-94 are given by vegetation category in Table 5. It is clear that for many countries a major part of the estimated isoprene emissions comes from the assumption of a nondominant vegetation component. This assumption is consistent with *Guenther et al.* [1994a] (although we have used lower biomass frac-

Table 4. Comparison of Isoprene Emission Estimates, 1989

Country ^a	E-90 ^b	E-BEIS	E-93	E-94		Man-made VOC
				CPY	N-CPY	
Albania	16.6	11.0	14.9	9.6	6.2	33
Austria	11.2	13.1	15.3	36.5	32.0	408
Belgium	6.6	10.1	12.6	39.1	30.4	335
Bulgaria	49.0	40.5	54.4	202.5	135.2	167
Former Czechoslovakia	25.0	23.6	30.9	88.3	70.9	294
Denmark	0.9	1.7	1.5	4.9	4.4	176
Finland	30.7	30.1	31.8	72.1	82.5	208
France	127.0	209.0	265.6	640.9	481.8	1972
East Germany	13.4	7.8	9.7	30.6	25.8	1050
West Germany	41.5	32.0	40.3	117.9	96.0	2536
Greece	46.0	38.1	58.3	31.0	21.3	185
Hungary	33.0	25.2	32.9	113.3	81.9	205
Ireland	0.3	1.1	1.1	2.6	2.2	90
Italy	96.3	82.5	104.7	71.8	53.2	1642
Luxembourg	0.9	0.6	0.7	2.4	1.8	13
Netherlands	3.2	2.2	2.8	9.3	7.3	442
Norway	7.3	17.6	12.9	27.1	28.7	216
Poland	34.6	14.9	19.3	76.9	64.3	982
Portugal	57.1	73.9	86.9	46.1	36.4	149
Romania	67.0	48.6	63.7	219.1	154.8	386
Spain	371.0	282.1	322.3	175.2	137.8	897
Sweden	24.7	53.2	47.0	103.6	108.2	460
Switzerland	4.3	2.1	2.3	6.6	5.5	305
Turkey	204.8	260.3	400.4	282.7	215.1	263
Former USSR ^c	1461.4	671.9	860.4	2256.1	2012.5	8156
United Kingdom	1.9	11.1	10.6	24.2	22.7	1986
Former Yugoslavia	152.3	52.3	67.0	65.5	47.2	290
Sum	2888	2016	2570	4756	3966	23854

Emissions (kt a⁻¹) have been calculated using the methodologies summarized in Table 1. Estimates made using the forest canopy model are labeled CPY. Estimates without the canopy are N-CPY.

^a1989 political units.

^bAs presented by Simpson [1993].

^cEuropean part of the former USSR.

tions for European coniferous forests) but remains speculative. It is not clear how much isoprene-emitting vegetation is associated with forests in Europe. Another notable point from this table is that for a few countries the assumptions regarding isoprene emissions from spruce trees have a rather significant effect on the total isoprene emissions. Most obviously, in the United Kingdom, where we have assumed that 81% of spruce are Sitka, the isoprene emissions from this forest category make up over 60% of total isoprene emissions.

For all countries, emissions of isoprene from crops are rather small compared to those from forests. This is in agreement with previous estimates for Europe [Veldt, 1988], and the global estimate of Guenther *et al.* [1995].

The spatial distribution of summertime isoprene emissions for estimates E-93 (as used in the companion paper of Simpson [this issue]) and E-94 are illustrated in Figure 3.

OVOC, other VOC. The calculated emissions of OVOC obtained from method E-94 are also given in Table 5. For many countries the estimated emissions of OVOC species shown in Table 5 are comparable to those of isoprene. This estimate suggests a higher proportion of OVOC to isoprene in Europe than in recent U.S. studies [Guenther *et al.*, 1994a; Geron *et al.*, 1994]. This is not surprising given the lower-isoprene emission factors adopted for E-94 and the generally colder and cloudier climate in Europe. OVOC emissions, as specified by (5), fall off slower with decreasing temperature than isoprene emissions and are not affected at all by radiation differences.

Nitrogen oxides. Table 6 gives the estimated emissions of nitrogen oxides obtained from soils using the adapted BEIS methodology described in section 2.4. In most cases, soil emissions make up only 5–10% of total NO_x emissions, but for some countries, this percentage is much higher. At least for the United Kingdom the calculated fluxes of NO_x (2.8% of man-made) are in line with estimates derived from other methodologies: Simpson *et al.* [1990] obtained 8%; Skiba *et al.* [1992] obtained 2–6%. Little can be said about the reliability of the estimates for any country at this stage because the emission factors are only crudely known, but these figures suggest that more work needs to be done to quantify these emissions in Europe.

Year-to-year variability. Table 7 shows the estimated biogenic isoprene emissions for 5 years, as calculated from E-94. It is clear that for some countries, year-to-year variability can be significant. For example, isoprene emissions in Finland are predicted to vary from under 60 kt yr⁻¹ in 1990 and 1991 to over 100 kt yr⁻¹ in 1988, due to differing ambient conditions from year to year. Another cause of year-to-year variability which we are unable to take into account in the present inventory results from changes in species composition over time. For example, in Portugal the coverage of the economically important high-isoprene emitter eucalyptus has increased dramatically in the last 30 years, to about 15% of total forest cover in recent years [Nunes *et al.*, 1995].

Figure 2. Calculated monthly isoprene emissions for (a) Spain and (b) Germany calculated using different methods, April–September 1989. Monthly man-made volatile organic compound (VOC) emissions (assumed as 1/12 of annual VOC) are indicated for comparison.

4. Uncertainties

Assessment of the uncertainties inherent in the calculations of biogenic VOC emissions in Europe is rather difficult. As a starting point, early estimates of the accuracy of the U.S. inventory have suggested $\sim 300\%$ in the emission-temperature relationship [Lamb *et al.*, 1987]. Further, even though much progress has been made in emission temperature algorithms [Guenther *et al.*, 1993], awareness has grown of the large uncertainties associated with specifying land cover for particular species. Therefore despite some probable improvement in the isoprene emission estimates represented by the E-94 procedure over the earlier E-93 and E-90 estimates, the level of uncertainties for European emissions are still very high. Even in the United States, where land use databases exist over the whole country in consistent format, uncertainties associated with specifying forest coverage are still significant [Guenther *et al.*, 1994a]. In Europe, such uncertainties are very much greater because such coherent land use data sets have not yet become available. We discuss some of the important factors

contributing to the total uncertainty of the European emission estimates below.

Emission factors. Little is known about the relationship between factors derived in the United States and those applicable to Europe, and unfortunately, emission factors for native European species are very few. In some specific areas of Europe, planted, nonnative species such as Sitka spruce in the United Kingdom and eucalyptus in Portugal may be significant isoprene emitters. It is clear that many more measurements are required before emissions in Europe can be described with any confidence.

Emission-temperature relationships. From the emission curves presented in Figure 1, we see that in the important temperature range from $\sim 20^\circ$ to 35°C the emission factors are rather sensitive to temperature. Emission calculations using a temperature 3° higher than that given by the EMEP meteorological data give increased isoprene emissions of between 20 and 50% in different countries over the period April–September 1989. Such a temperature error is well within the range of meteorological uncertainties on a day-to-day basis, especially when we consider that emissions are controlled by leaf temperature and that this can often be many degrees different from ambient temperatures (even as much as 20°C) [Friedland *et al.*, 1992; Hadley and Smith, 1987].

Although the BEIS canopy model appears to reproduce in a qualitative way the effects of forest shading and heating, some limited evaluations show mixed success. Comparisons between canopy model temperature predictions and measured temperatures show differences of $\sim 1^\circ\text{C}$ in one study [Martin *et al.*, 1991; Lamb *et al.*, 1993] but as much as 10°C in another study [Fuentes *et al.*, 1993]. To compound these difficulties, it is obvious that the $150 \times 150 \text{ km}^2$ spatial resolution of the EMEP model's surface temperature fields cannot represent the ambient temperatures for all forests within a grid square. For example, temperature gradients within and between valleys can be substantial, and as an added complication, the microclimate of a Sun-facing slope can be very different to that of a forest on a plain or on a shaded slope.

Land use data. The most basic land use data for our study are the forest coverages given in Table 2. The total forest areas given there compare quite well to those reported in UNECE [1992] for most countries, with a mean difference of only 8%. However, differences for individual countries can be significant: 25% for Spain and 37% for Norway. Comparing these same total forest areas with those of IIASA (as reported by Lubkert and Schöpp [1989]) shows mean differences of 25%, with the largest differences for Ireland (77%), Denmark (50%), and Hungary (41%). Of more relevance for this study are the areas obtained for broadleaf forests. On average, the Veldt [1989] data give 26% more broadleaf forest than the IIASA database. These differences are as great as a factor of 3 in the cases of France and the United Kingdom.

These differences are not too hard to understand, given that the focus of most forest statistics appears to be the area of productive, coniferous forest, rather than the categories of most interest for isoprene inventories. Even for the coniferous forest, category definitions vary greatly; 1 km^2 of coniferous forest appears to mean that 50% of the stem volume is coniferous in Finland and Norway, 70% in Sweden, 80% in Ireland, and 100% in the United Kingdom [UNECE, 1985].

The biggest problem highlighted by this study has been the need for forest data at a species specific level. Thus we need to know not just the coverage of oak but rather the coverage of

Table 5. Emissions of Isoprene and OVOC by Vegetation Category, April–September 1989

	Isoprene Emissions				Total Isoprene	OVOC Emissions			Total OVOC	Sum Isoprene Plus OVOC
	Oak	Spruce	N-D vege ^a	Crops		Broadleaf	Conif	Crops		
AL	3.4	0.0	2.7	0.0	6.2	7.4	1.9	0.1	9.4	15.6
AT	7.4	17.1	7.3	0.1	32.0	4.5	73.4	0.5	78.4	110.3
BE	25.7	2.6	2.1	0.1	30.4	2.7	10.2	0.3	13.1	43.6
BG	122.9	1.6	10.3	0.4	135.2	18.9	25.3	1.2	45.4	180.6
CS	38.3	20.1	12.0	0.4	70.9	11.6	83.5	1.2	96.4	167.3
DK	1.6	2.1	0.6	0.1	4.4	0.8	6.0	0.4	7.2	11.6
FI	0.0	44.2	38.2	0.1	82.5	15.8	338.3	0.4	354.4	436.9
FR	420.0	9.5	50.0	2.4	481.8	76.6	139.7	6.7	223.1	704.9
DD	13.2	5.4	6.9	0.4	25.8	3.9	48.7	1.1	53.8	79.6
DE	53.8	24.2	17.3	0.7	96.0	15.9	121.2	2.3	139.4	235.4
GR	13.4	0.0	7.6	0.3	21.3	14.1	20.8	1.0	35.9	57.2
HU	75.9	0.2	5.4	0.5	81.9	11.5	4.2	1.4	17.1	99.1
IE	0.8	0.7	0.5	0.2	2.2	0.4	5.5	0.7	6.6	8.8
IT	24.7	5.6	22.2	0.7	53.2	46.0	42.9	2.0	90.9	144.0
LU	1.4	0.2	0.2	0.0	1.8	0.4	0.6	0.0	1.1	2.9
NL	5.9	0.3	1.0	0.1	7.3	0.8	4.9	0.4	6.2	13.5
NO	3.1	16.1	9.4	0.0	28.7	8.9	95.4	0.1	104.5	133.2
PL	31.4	6.4	25.3	1.2	64.3	11.9	164.1	3.5	179.5	243.8
PO	21.3	0.0	15.0	0.2	36.4	16.5	44.9	0.4	61.8	98.2
RO	124.1	13.0	16.8	0.9	154.8	32.7	49.8	2.9	85.5	240.3
ES	79.1	0.0	57.5	1.3	137.8	63.1	184.6	3.3	251.0	388.8
SE	12.4	58.0	37.7	0.1	108.2	31.0	357.6	0.5	389.0	497.2
CH	1.2	2.4	1.8	0.1	5.5	1.6	14.9	0.2	16.7	22.2
TR	90.4	53.1	69.8	1.7	215.1	80.1	379.6	5.5	465.3	680.4
SU	961.7	583.7	460.2	6.8	2012.5	432.3	2765.1	19.6	3217.0	5229.5
GB	5.2	13.8	3.1	0.6	22.7	4.3	22.9	2.2	29.4	52.1
YU	15.5	7.1	24.2	0.4	47.2	55.0	43.1	1.3	99.5	146.7
Sum	2153	888	905	20	3966	969	5049	59	6078	10044

Countries are identified by the two-letter codes given in Table 2. Emission unit, kt a^{-1} . N-D. vege^a refers to the nondominant vegetation. Broadleaf means all broadleaf (oak plus others), and conif means all coniferous species (spruce plus others).

Q. robur and *Q. ilex*. Sitka spruce and, indeed, other isoprene-emitting spruce species provide another example. Such detailed data have only been obtained from a few countries, and these data show very large national differences. This lack of information must be one of the largest contributions to the uncertainty of the emission estimates.

Biomass data. Veldt [1989] recommended biomass densities for Norway spruce of $800\text{--}1600 \text{ g m}^{-2}$, with lower values at high latitudes. For other spruce and other coniferous, Veldt recommended 1400 and 700 g m^{-2} , respectively, regardless of latitude. In contrast, Ortiz and Dory [1990] used a density of only 400 g m^{-2} for coniferous forest in Spain, and information from Portugal would suggest $500\text{--}800 \text{ g m}^{-2}$ for *Pinus* species (C. Pio, Departamento de Ambiente e Ordenamento, Universidade de Aviero, Portugal, personal communication, 1995). In view of the limited information available the biomass data adopted in Table 3 were deliberately kept simple, with no changes in biomass with respect to latitude. However, in some regions, differences of a factor of 2 between biomass data used here and actual conditions might clearly be expected.

The biomass of nondominant vegetation is assumed to make up 3–15% of forest biomass, and emissions from this category can be significant (Table 5). Thus uncertainties in the forest biomass itself, the 3–15% nondominant values, and the assumed emission rate from this source quickly translate into a significant uncertainty in the isoprene estimate.

Comparisons with other studies. Table 8 compares the E-93 and E-94 isoprene emission estimates with a number of other studies. The most recent of these is the global biogenic isoprene estimate of Guenther et al. [1995], which calculates isoprene emissions over a spatial grid of $0.5^\circ \times 0.5^\circ$ latitude/

longitude. The emission factors used for isoprene are similar to those developed for the United States by Guenther et al. [1994a] and thus are comparable to those used in this study for northern European oaks and nondominant vegetation emissions but much higher for spruce and Mediterranean oak species. These different emission factors account for the greater emissions assigned to Germany, Sweden, Spain, and the United Kingdom by the global model, although estimates for Hungary are similar (probably because Hungary's coniferous forest consists mainly of nonspruce species, so emission factors in the two studies are then similar).

In the case of Denmark, the Netherlands, and Spain the available national estimates match the E-93 figures best. However, this apparent agreement mainly reflects the fact that all these estimates have been based around the BEIS emission factors, although assumptions vary in each study. The newer emission estimates obtained in E-94 are substantially greater than these earlier estimates for Denmark and the Netherlands but substantially lower for Spain.

The greatest discrepancy between estimates are found for the United Kingdom. The E-93 estimate is clearly lowest, with assumptions concerning Sitka spruce emission factors accounting for the differences with E-94. The Anastasi et al. estimates are similar to those of the global model, both being around 5 times greater than that of E-94. However, all models are built around quite different assumptions. For Spruce trees the isoprene emission formulae used by Anastasi seems to give an emission factor of $\sim 30 \mu\text{g g}^{-1} \text{ h}^{-1}$, twice that of Guenther et al. [1995], $14 \mu\text{g g}^{-1} \text{ h}^{-1}$, and 5 times higher than the E-94 figure, $6 \mu\text{g g}^{-1} \text{ h}^{-1}$. The biomass density for spruce used in Anastasi et al. is 1973 g m^{-2} , i.e., 41% greater than the EMEP

Figure 3. Spatial distribution of isoprene emissions on the EMEP MSC-W grid for April–September 1989. Estimates are given from two methods: (a) E-94 and (b) E-93. April–September 1989. Units, kt yr^{-1} .

value. For agricultural areas, Anastasi et al. use emission factors of $162 \mu\text{g C m}^{-2} \text{h}^{-1}$. This value, derived from measurements at 30°C is a factor of 20 greater than the value of $8 \mu\text{g m}^{-2} \text{h}^{-1}$ suggested by Veldt [1991] and used in the E-93/E-94 models. (We can also note that Lamb et al. [1993] suggest $29 \mu\text{g m}^{-2} \text{h}^{-1}$ at $\text{PAR} = 400 \mu\text{E m}^{-2} \text{s}^{-1}$ as a mean value over the United States). Further, the Anastasi value is applied throughout the summer period without any temperature correction, whereas the EMEP and global estimates use both solar radiation and temperature corrections as for the forest species.

The discrepancies between estimates for the former USSR is almost as great as for the United Kingdom, although as details

of the methodology used by Tsibulsky et al. [1993] are not known, further discussion of this is not warranted.

OVOC and soil NO emissions. The estimates of OVOC and soil NO have mainly been presented to illustrate their possible magnitude in Europe compared to man-made emissions. The problems noted above with regard to estimates of isoprene emissions apply even more to OVOC. Guenther et al. [1993] noted that the recommended emission rate of $1.5 \mu\text{g g}^{-1} \text{h}^{-1}$ is associated with a 10-fold range ($0.5\text{--}5 \mu\text{g g}^{-1} \text{h}^{-1}$) in possible emissions and that even this may underestimate some emissions.

The uncertainty associated with the soil NO emissions is very

Figure 3. (continued)

large. Williams *et al.* [1992] suggested a factor of 3 uncertainty for the temperature algorithms, but this is likely to be a lower limit for the total uncertainty of emission estimates. There is also evidence that measurements of soil flux emissions may overestimate the flux of NO to the atmosphere, as not all of the emitted NO may leave the plant canopy above the soil [Jacob and Bakwin, 1991].

Final remarks. It has been recognized that the minimum level of uncertainty in global biogenic emission estimates is a factor of 3 [Guenther *et al.*, 1995], but this is likely to represent a lower limit for the accuracy of European emission estimates. Further, this figure relates to estimates of annual emissions. Uncertainties for episodic calculations must obviously be substantially greater. In the modeling work presented in the com-

panion paper [Simpson, this issue], we have therefore chosen to consider an uncertainty of a factor of 5 in biogenic isoprene emissions.

5. Summary and Conclusions

Several biogenic VOC algorithms have been used together with meteorological data from the EMEP MSC-W ozone model to generate estimates of the emissions of isoprene from European forests and agricultural crops over summer periods.

E-94, the most up-to-date estimate, combines the recently updated isoprene emission factors from the United States with available knowledge of European tree species and emission factors. In some cases these European emission factors are

significantly different from their U.S. equivalents because of differences in the tree species represented within a forest classification, especially with regard to spruce genera and Mediterranean oak genera. The new estimates have resulted in an approximate factor of 3 increase in isoprene emissions from northern Europe but a factor of 2 decrease in isoprene estimates for southern Europe. Overall, European isoprene emissions are estimated to be about 4000 kt C yr⁻¹, approximately 50–100% greater than previous estimates.

It should be recognized that none of the biogenic emission inventories used in Europe can be compared in terms of complexity or accuracy with those generated in the United States. All European estimates, including those presented in this paper, have been severely limited by the availability of data on a European scale. Several key items are either missing or known to only a limited extent, necessitating some rather arbitrary choices. The following points require most urgent attention:

1. A species specific database is needed, preferably on a gridded basis (50 × 50 km² would be ideal for EMEP purposes). This database should provide coverage (preferably in terms of biomass) of at least those species thought to be the most important isoprene emitters. For example, details of Sitka spruce, *Q. robur*, and eucalyptus are essential for a good emission estimate. These data probably exist within each country but need to be made available for Europe as a whole and spatially disaggregated.

2. More measurement data are urgently needed for European species. So far, studies in Europe have been limited to only a few species and a few locations. The available evidence

Table 7. Estimated Isoprene Emissions in Europe, Year-to-Year Variations

Country	Calculated Isoprene Emissions, ^a kt yr ⁻¹				
	1985	1988	1989	1990	1991
AL	9.1	9.6	6.2	5.8	4.6
AT	33.1	39.9	32.0	26.7	20.3
BE	27.5	27.6	30.4	23.6	20.3
BG	149.0	181.3	135.2	121.6	97.7
CS	71.5	81.5	70.9	57.2	51.0
DK	4.2	5.0	4.4	4.1	4.2
FI	73.1	108.2	82.5	56.4	51.7
FR	412.0	466.5	481.8	393.1	348.1
DD	26.5	27.4	25.8	20.0	18.1
DE	91.4	102.0	96.0	76.2	70.2
GR	25.4	27.8	21.3	20.5	19.0
HU	76.3	99.6	81.9	69.6	66.2
IE	2.3	2.5	2.2	1.9	1.8
IT	49.7	68.8	53.2	49.8	41.9
LU	1.7	1.8	1.8	1.4	1.3
NL	6.8	6.8	7.3	5.7	5.1
NO	26.7	36.8	28.7	22.1	20.5
PL	65.3	72.8	64.3	50.0	46.9
PO	33.6	36.9	36.4	30.4	31.2
RO	163.9	195.0	154.8	132.5	114.1
ES	123.8	137.5	137.8	117.0	110.7
SE	109.8	147.7	108.2	85.7	84.3
CH	4.6	6.0	5.5	4.6	3.6
TR	220.6	252.3	215.1	198.5	185.3
SU	1631.2	2251.7	2012.5	1306.3	1477.1
UK	23.9	24.8	22.7	18.5	17.4
YU	52.7	64.1	47.2	42.2	34.1
Sum	3516	4482	3966	2942	2946

^aEmissions estimated using method E-94.

Table 6. Emissions of Man-made and Soil NO_x Emissions Used in the Calculations

Country	Emissions of NO _x (as NO ₂), kt a ⁻¹		
	Man-made	Soil	Soil/Man-made, %
AL	9.0	14.5	161.1
AT	211.0	23.3	11.0
BE	317.0	21.6	6.8
BU	156.0	29.1	18.7
CS	1122.0	44.4	4.0
DK	270.0	32.2	11.9
FI	284.0	53.2	18.7
FR	1772.0	125.2	7.1
DD	670.0	33.5	5.0
DE	2707.0	66.6	2.5
GR	746.0	74.1	9.9
HU	249.0	37.2	14.9
IE	127.0	25.2	19.8
IT	1761.0	119.0	6.8
LU	16.0	3.8	23.8
NL	551.0	17.6	3.2
NO	233.0	62.2	26.7
PL	1480.0	73.2	4.9
PO	132.0	40.9	31.0
RO	390.0	59.0	15.1
ES	367.0	126.7	34.5
SE	413.0	71.4	17.3
CH	183.0	12.4	6.8
TU	175.0	135.8	77.6
SU	4094.6	985.2	24.1
GB	2717.0	75.2	2.8
YU	430.0	55.9	13.0
Sum	21582	2418	11.3

All estimates are for 1989. Soil emissions are calculated as a function of temperature as described in the text. Countries are identified by the two-letter codes given in Table 2.

suggests that the emission factors used for U.S. genera can sometimes be applied to similar European classifications (e.g., to oak forests in northern Europe), but in other cases the U.S. data would give erroneous results (e.g., for spruce emissions, or oak emissions in the Mediterranean).

3. The applicability of U.S.-derived emission algorithms (for temperature and light dependencies) to Europe needs to be checked, as it may be that Europe's colder climate requires a modification to these algorithms.

4. As in the United States, the emissions of other VOC need to be quantified and speciated.

We can conclude that all estimates of biogenic isoprene emissions in Europe are still subject to considerable uncertainty. The factor of 3 uncertainty originally specified for the United States would seem optimistic for Europe. Factors of 5–10 uncertainty are not unlikely for some regions of Europe and especially during particular ozone episodes.

Preliminary estimates suggest that the emissions of OVOC, the so-called other VOC, from biogenic sources are of similar magnitude to those of isoprene. This suggests that more consideration should probably be given to these emissions in future measurement and modeling activities.

Preliminary estimates of NO_x emissions from soils suggest that in most countries, such emissions represent only a small percentage of man-made (combustion derived) emissions. However, for some countries these emissions could be very significant. Uncertainties in these estimates are still very large, and the need for better estimates of these emissions is evident. The implications of the uncertainties in isoprene emissions for the results of control strategy evaluations in Europe have also

Table 8. Comparison of EMEP Isoprene Estimates (kilotonnes) With Other Work

Country	Year	E-93 ^a	E-94 ^a	Other
Denmark	1985	1.5	4.2	1.56 ^b
	1989	1.5	4.4	1.79 ^b
	1990	1.5	4.1	1.76 ^b
East and West Germany	1990	40	96	370 ^c
Greece	1985	70	25	73 ^{d,e} , 77 ^{d,f}
Hungary	1990	28	70	120 ^c
Netherlands	1985	2.6	6.8	1.88 ^g
Spain	1985	300	124	239 ^h
	1990	280	117	820 ^c
Sweden	1990	37	86	370 ^c
United Kingdom	1989	11	23	115 ⁱ
	1990	8.7	19	110 ^c
Former USSR ^j		570–950 ^k	1310–2250 ^k	4302–4528 ^l

^aSee Table 1 for definition of E-93 and E-94.

^bE. H. Runge (personal communication, 1994). Calculations using land use data from Runge and Asman [1989], hourly temperature, cloud cover, and radiation data, BEIS model with forest canopy, April–September (inclusive).

^cGuenther et al. [1995] global model calculations for IGAC-GEIA archive.

^dEmissions from deciduous forests but not specified to be only isoprene.

^eHolzappel et al. [1992], emissions from deciduous trees based upon Lubkert and Scöpp [1989] methodology.

^fPistikopoulos et al. [1994], emissions from deciduous forest.

^gVeldt [1988].

^hOrtiz and Dory [1990] using Veldt and EMISNAT methodology.

ⁱAnastasi et al. [1991]. Note that unit is kt C. Figures derived from total nonmethane hydrocarbon (NMHC) emission figures using isoprene/NMHC ratios given in paper. Isoprene emissions estimated at 13.1 kt C a⁻¹ from deciduous trees, 59.8 kt C a⁻¹ from agricultural areas, and 42.5 kt C a⁻¹ from spruce trees.

^jEuropean part of former USSR.

^kEMEP estimates are maximum and minimum of values from 1988, 1989, 1990, and 1991.

^lTsibulsky et al. [1993]. Maximum and minimum values refer to coldest and warmest years from 1987 to 1991. Estimates based upon experimental data.

been assessed using the EMEP MSC-W ozone model and are presented in the companion paper of Simpson [this issue].

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