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Inventorying emissions from nature in Europe

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Abstract. As part of the work of the Economic Commission for Europe of the United Nations Task Force on Emission Inventories, a new set of guidelines has been developed for assessing the emissions of sulphur, nitrogen oxides, NH_3 , CH_4 , and nonmethane volatile organic compounds (NMVOC) from biogenic and other natural sources in Europe. This paper gives the background to these guidelines, describes the sources, and gives our recommended methodologies for estimating emissions. We have assembled land use and other statistics from European or national compilations and present emission estimates for the various natural/biogenic source categories based on these. Total emissions from nature derived here amount to $\sim 1.1 \text{ Tg S yr}^{-1}$, $6\text{--}8 \text{ Tg CH}_4 \text{ yr}^{-1}$, $70 \text{ Gg NH}_3 \text{ (as N) yr}^{-1}$, and $13 \text{ Tg NMVOC yr}^{-1}$. Estimates of biogenic NO_x emissions cover a wide range, from 140 to $1500 \text{ Gg NO}_x \text{ (as N) yr}^{-1}$. In terms of relative contribution to total European emissions for different pollutants, then NMVOC from forests and vegetation are clearly the most important emissions source. Biogenic NO_x emissions (although heavily influenced by nitrogen inputs from anthropogenic activities) are very important if the higher estimates are reliable. CH_4 from wetlands and sulphur from volcanoes are also significant emissions in the European budgets. On a global scale, European biogenic emissions are not significant, a consequence of the climate and size (7% of global land area) of Europe and of the destruction of natural ecosystems since prehistoric times. However, for assessing local budgets and for photochemical oxidant modeling, natural/biogenic emissions can play an important role. The most important contributor in this regard is undoubtedly forest VOC emissions, although this paper also indicates that NMVOC emissions from nonforested areas also need to be further evaluated. This paper was originally conceived as a contribution to the collection of papers arising as a result of the Workshop on Biogenic Hydrocarbons in the Atmospheric Boundary Layer, August 24–27, 1997. (Several papers arising from this workshop have been published in *Journal of Geophysical Research*, 103(D19) 1998.)

1. Introduction

Europe is a continent covering <7% of the world's land area, inhabited by 680 million people (13% of the world's

population). It accounts for $\sim 30\%$ of the world's CO_2 emissions, over 20% of SO_x , NO_x , and VOC emissions, $\sim 15\%$ of CH_4 emissions and 10% of N_2O and CO emissions [Stanners and Bourdeau, 1995, and references therein].

Most of these emissions in Europe come from the combustion and processing of fossil fuels, rather than from biogenic or other natural sources. Europe is unique in this respect; in other continents and globally biogenic emissions clearly outweigh anthropogenic. However, even in Europe, biogenic and natural emissions can be important, especially within individual countries. For example, sulphur emissions in Italy are dominated by volcanic sources, VOC emissions in the Mediterranean area are dominated by emissions from forests during summertime, and methane emissions from Scandinavia by wetlands.

There are several reasons for preparing an inventory of emissions from nature. One is that emissions from natural sources take part in the background chemistry of the atmosphere and are thus worthy of study in themselves. Additionally, emissions from nature can interact with man-made emissions with deleterious effects, a notable example being ozone formation brought about by the mixing of NO_x -rich plumes from urban areas with VOC emissions from surrounding forests [e.g., Chameides, 1988]. A more accurate assessment of these emissions also becomes more important as emission con-

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trols of combustion sources become ever more stringent. Indeed, policies under consideration by both Economic Commission for Europe of the United Nations (UN-ECE) and the European Union (EU) are based upon meeting so-called critical thresholds, levels of deposition or concentration above which damage is believed to occur. The amount of emission reduction required to meet these targets will in some cases depend very heavily on assumptions made about the level of ambient concentrations from "natural" emissions.

It is thus important that European natural and biogenic emissions are assessed to the fullest extent possible and areas where emissions may be considerable highlighted. Unfortunately, the inventorying of any compound in Europe has a number of problems not found in, for example, the United States or Canada. Not least, over 40 countries are involved, and emission inventory developers cannot be assumed to speak the same language, let alone have access to similar levels of resources or statistical data. For many countries, only one national expert will be responsible for the emission inventory, so this expert has to cover the details of all anthropogenic sources (combustion sources, solvent, etc.) as well as natural emissions. Reporting of emissions has to take place to several international organizations, notably the IPCC, the UN-ECE European Monitoring and Evaluation Programme (EMEP), and for many countries to the European Union's CORINAIR system.

Despite these difficulties, the inventorying of anthropogenic emissions in Europe has attained a high standard of consistency and detail within the last few years. This has largely been achieved through the use of a joint nomenclature agreed between the EMEP and CORINAIR programmes and the joint EMEP/CORINAIR Atmospheric Emission Inventory Guidebook [McInnes, 1996]. This Guidebook, the European equivalent of America's AP42 document [U.S. Environmental Protection Agency, 1997], aims to provide an up-to-date and comprehensive summary of emission inventory methodology and emission factors for each of the pollutants and sources to be quantified. Unfortunately, the first edition of this Guidebook was restricted to anthropogenic emissions; biogenic emissions were only considered for agricultural activities, especially for NH_3 .

In order to correct this situation, a so-called "Nature Panel" was set up to write the chapters of the Guidebook dealing with emissions from biogenic and natural sources. This paper presents a summary of the work of this panel. The sources covered are described, recommended methodologies are outlined, and emissions are calculated on the basis of these methodologies. It should be noted that these methodologies were primarily designed for incorporation into the Guidebook and as such are intended to capture the most important factors required for emission estimates, rather than to be detailed research methodologies. In any case, it is our experience that the limiting factor in estimating emissions in Europe is the lack of underlying statistical data (e.g., land use), so complex methodologies are not useful. In many ways the most important outcome of the Guidebook, and hopefully this paper, will be to stimulate further activities to refine the database on natural emissions in Europe.

1.1. Emissions Included

The main pollutants to be covered are sulphur (SO_x and reduced compounds), NO_x , NH_3 , nonmethane volatile organic compounds (NMVOC), and CH_4 as these are the main pol-

lutants for which new methodologies have been devised by the Nature Panel. For all pollutants except methane we consider only emissions, in accordance with normal EMEP/CORINAIR practice. However, for methane, both emissions and consumption are considered, in accordance with the practice of IPCC where this pollutant is important.

It should be noted that several important emissions are not addressed as part of this study. This includes aerosols, CO_2 , N_2O , and heavy metals. They have been excluded either because the recommended methodologies are identical to those of IPCC or because no methodology has been accepted yet as part of the Guidebook. However, future activities will seek to introduce also these pollutants into the Guidebook.

Deciding which source groups should be considered "natural" proved extremely problematic. Almost every piece of land in Europe is affected by man's activities to some degree, either by direct intervention such as forestry practices or indirectly through changes brought about by acid deposition. The importance and difficulties of this question is tackled by Winiwarter *et al.* (On the boundary between man-made and natural emissions: Problems in defining European ecosystems, submitted, 1998) who attempt to give a natural scientist's view of possible boundaries between natural and anthropogenic emissions. For this paper the following sources are included: (1) Forest (foliar emissions), even if heavily managed, (2) natural grassland and other low vegetation, including lands used for grazing, (3) forest fires, (4) soils, (5) wetlands, (6) waters, (7) wild animals and humans, (8) lightning, (9) volcanoes, and (10) gas seeps. (The order shown above is kept within the sections on sources, methodology, input data, and uncertainties which follow.)

Excluded from any detailed consideration are (1) agricultural crops and farm animals (except possibly deer, etc.), (2) biomass burning for fuel, agricultural biomass burning (straw, etc.), and (3) emissions arising from leaching of agricultural fertilizers/nutrients into natural ecosystems such as rivers.

In this paper, we will firstly give a general overview of Europe and its geography (section 2). The emission sources are described in section 3, the methodology for estimating emissions in section 4, and the land use and other input data in section 5. The resulting emission estimates are presented in section 6, followed by a discussion of the uncertainties in section 7.

2. Europe, Land Use and Climate

Table 1 lists the European countries considered in this work, and gives the areas of total land, forest and meadow/pasture. Note that here and in many tables throughout this paper, we keep some newly independent European states grouped together with their former state (Czechoslovakia, Yugoslavia). This is a purely practical matter as often statistics are available for the former political units but not the new states.

Plate 1 shows the land use distribution over much of Europe as mapped by the Stockholm Environment Institute (SEI) at York, University of York, York, England. On average, ~33% of Europe's land area consists of forests, 43% is used for agriculture, with the remaining 24% consisting of rocks, tundra, wetlands, etc. [van de Velde, 1994]. Some countries, such as the United Kingdom, are seen to be almost devoid of forests, whereas Sweden and Finland are extensively covered (the United Kingdom has 10% forest cover, while Finland has 76%).

Relative to other continents such as the Americas, Europe is

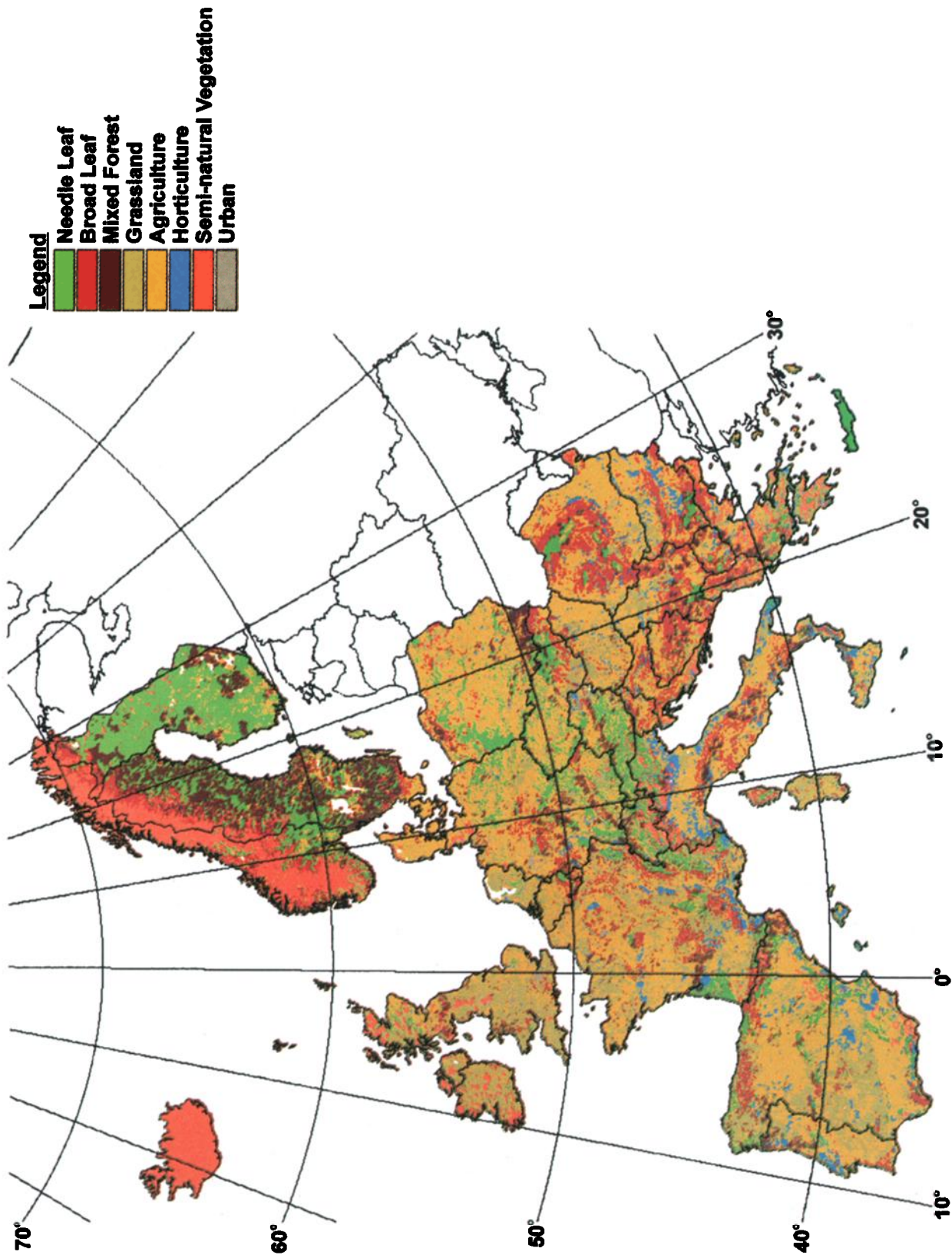


Plate 1. Land cover of (Western) Europe: major land use types.

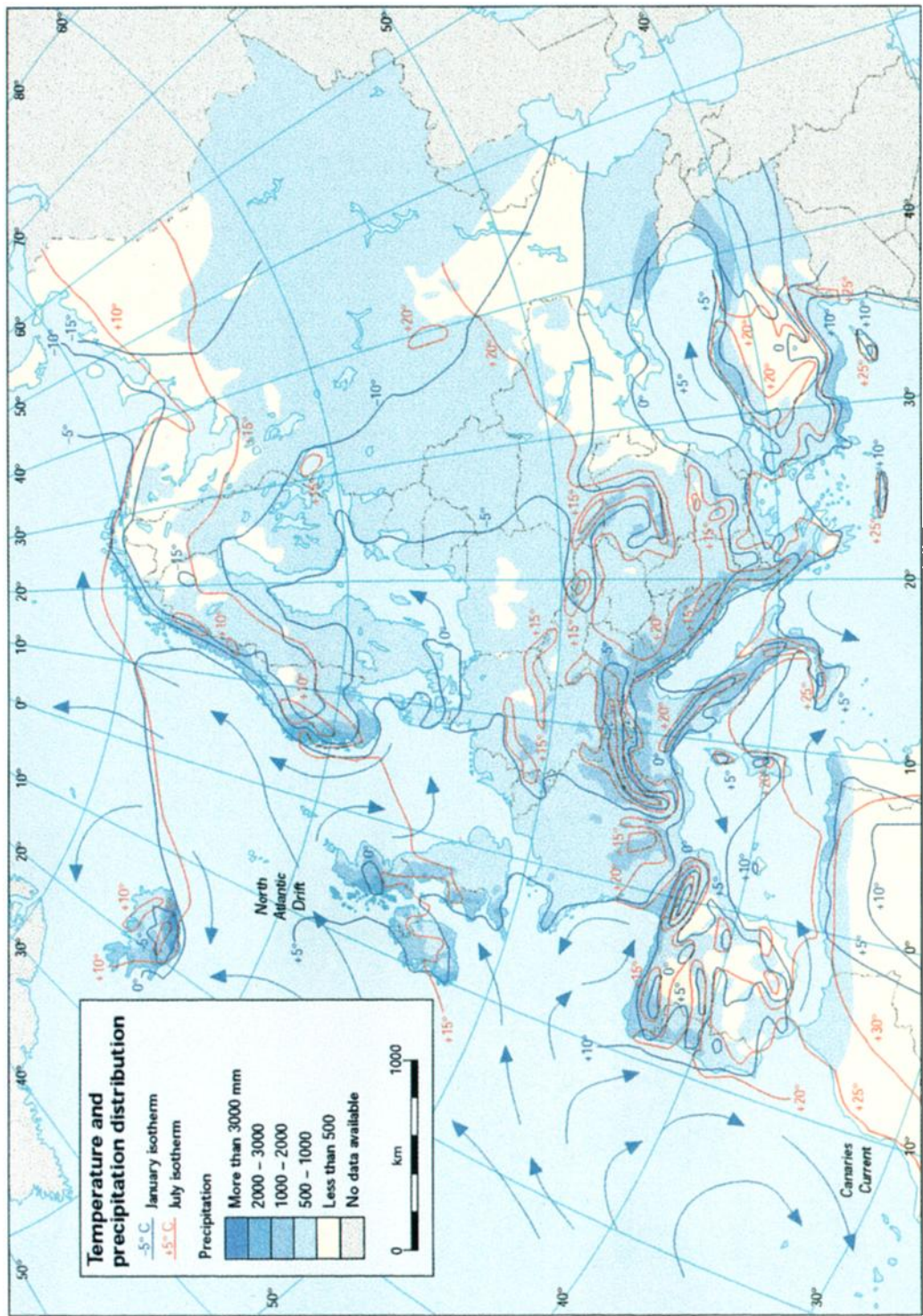


Plate 2. Temperature and precipitation distribution in Europe, based on *De Agostini* [1992]. © Instituto Geografico De Agostini, Novara.

very poor in flora and fauna in both diversity and numbers. For example, just six tree species have been found to cover two thirds of the forest area, namely *Pinus sylvestris* (24%), *Picea abies* (23%), *Fagus sylvatica* (9%), *Pinus pinaster* (4%), *Quercus robur* (4%), and *Quercus ilex* (3%) [DGV1, 1996]. Most diversity in Europe is found in one region: the Mediterranean, which has at least 100 species of trees. In the rest of Europe, only ~30 species of trees are found in any abundance [le Houérou, 1981].

One reason for this is that during the last two ice ages the extensive forests of central and northern Europe were destroyed. With the retreat of the ice, the forests returned, but fewer species recolonized the landscape. The fact that the Mediterranean escaped the ravages of the glaciers, along with its naturally favorable and stable climate, explains the large diversity of species that built up in this area and that still prevails today, despite 10,000 years of degradation brought about by man's activities.

In fact, human influence has undoubtedly been the biggest factor in shaping today's landscape. Without man's influence, ~90% of Europe's land would be forested. Clearing of the forest began in the Mediterranean in prehistorical times. By the 5th century B.C., Plato [Critias, 111 B.C.] lamented the destruction around Attica. Today the landscape consists in large part of scrubland and eroded mountains. In central and northern Europe the forests were cleared through the middle ages [Darby, 1956; le Houérou, 1981]. However, the importance of forestry as an industry seems to have stabilized forest coverage in recent times, with total coverage the same today as reported by Dietrich [1928]. In many countries, forests have actually been increasing in recent years, though the increase tends to be in productive and often exotic conifer forests rather than in naturally indigenous species.

Wetlands have been significantly reduced in area through drainage or other factors, with for example a loss of nearly 94% of wetlands in Italy since Roman times, or more recently a loss of 40% of the coastal wetlands of Brittany, France, since 1960 [Moser, 1992; Baldock, 1984].

Europe encompasses many different types of climate. The mean temperature distributions of Europe in July and January, together with precipitation amounts, are illustrated in Plate 2. The climate types are illustrated in Figure 1. In the Arctic climates of northern Scandinavia and Russia, temperatures rarely exceed 10°C. Snow covers the ground for many months, and growing seasons for vegetation can be very short, often <3 months, in contrast to the more typical 6 months of central Europe. At the other extreme the Mediterranean is characterized by a warm temperate climate, with temperatures above 6°C all year. Only the Mediterranean experiences temperatures that can be compared to those common in the United States. In fact, in terms of both vegetation and climate, the Mediterranean probably has more in common with southern California than with its more northerly neighbors in Europe [Di Castri, 1981].

3. Sources

3.1. Forests (Foliar Emissions)

This section treats NMVOC emissions from the foliage of forests. Fluxes of methane and NO_x from forest soils are treated in section 3.4, and fluxes of NMVOC from wet peat areas of the forest floor are treated in section 3.5. We do not have enough information to attempt a calculation of fluxes of

Table 1. Size and Land Cover of European Countries and Land Code

Country/Unit	Code	Total Land Area, km ²	Total Forest Area, km ²	Meadows/Pastures, km ²
Albania	AL	27,400	10,460	4030
Austria	AT	82,730	32,270	19,950
Belarus	BY	207,480	73,834	31,568
Belgium	BE	32,820	6170	5564
Bulgaria	BG	110,550	38,710	19,990
Czechoslovakia, ex ^a	CS	125,360	46,190	16,730
Czech Republic	CZ	78,864 ^b	26,421 ^b	11,343 ^b
Slovak Republic	SK	49,036 ^b	19,400 ^b	5387 ^b
Denmark	DK	42,390	4930	2120
Estonia	EE	42,270	18,692	2693
Finland	FI	304,610	232,220	1230
France	FR	550,100	148,500	111,980
Germany	DE	349,310	104,030	53,290
Greece	GR	128,900	26,200	52,550
Hungary	HU	92,340	17,010	11,730
Iceland	IS	100,250	1200	22,740
Ireland	IE	68,890	3450	46,940
Italy	IT	294,060	67,520	48,800
Latvia	LV	62,050	28,032	8438
Lithuania	LT	64,800	19,677	11,721
Luxembourg	LU	2576	886	686
Moldova, Republic of	MD	32,970	3570	3000
Netherlands	NL	33,920	3000	10,800
Norway	NO	306,830	83,300	1130
Poland	PL	304,420	87,810	40,380
Portugal	PT	91,950	29,680	8380
Romania	RO	230,340	66,900	47,780
Russian Federation ^c	RU	3,815,000 ^c	1,446,420 ^c	590,000 ^c
Spain ^d	ES	499,440	158,580	103,000
Sweden	SE	411,620	280,200	5540
Switzerland	CH	39,770	10,520	16,090
Ukraine	UA	603,550	92,930 ^e	70,000
United Kingdom	UK	241,600	24,100	111,800
Yugoslavia, ex ^e	YUG	255,400	91,200	63,520
Bosnia Herzegovina	BA	51,000	23,724 ^b	14,000
Croatia	HR	55,920	20,776	15,620
Macedonia, FYR of	MK	25,430	10,790 ^b	2506 ^b
Serbia Montenegro	YU	100,990	41,196 ^b	18,863 ^b
Slovenia	SI	20,120	10,140	5600

From EUROSTAT [1995] and Food and Agricultural Organizations [1997].

^aSome former states are also given, often the only source of statistics.

^bDerived areas for some new states from EUROSTAT [1995] and gridded land use, see section 5.1.

^cEuropean part.

^dExcluding Canary Islands.

^eEuropean Forest Institute (<http://www.efi.fi/kupka/spec.htm>)

^fTotal land area (including lakes, etc.).

NMVOC from the forest floor, but available investigations suggest that emissions are probably much less than from the forest canopy [Janson et al., 1998; Steinbrecher et al., 1993b].

Forests are the main source of biogenic NMVOC. Globally, emissions from forests are estimated to contribute ~820 Tg yr⁻¹ to a total biogenic emission of 1150 Tg yr⁻¹ [Guenther et al., 1995]. Global anthropogenic NMVOC emissions amount to only 10% of this, ~100 Tg yr⁻¹ [Müller, 1992; Piccot et al., 1992]. In contrast, European anthropogenic and biogenic NMVOC have comparable magnitudes; biogenic NMVOC emissions are estimated at ~14 Tg yr⁻¹ (this study), compared to man-made emissions of around 24 Tg yr⁻¹.

Thus European forest emissions are only a small part of the

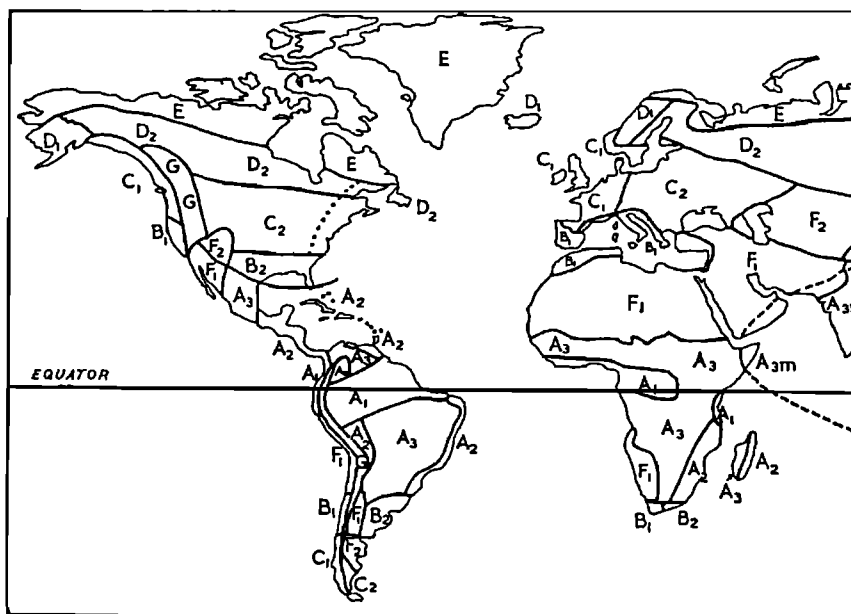


Figure 1. Climates of Europe and other regions, adapted from *Monkhouse* [1981] with permission. Key: A, hot; B, warm temperate; C, cool temperate (C1, marine; C2, continental); D, cold temperate (C1, marine; C2, continental); E, Arctic.

global total, but even on an annual basis they are significant in the European NMVOC inventory. Further, these emissions are strongly dependent on season, temperature and light intensity [e.g., *Guenther*, 1997; *Schnitzler et al.*, 1997] so that in many countries of Europe they may exceed anthropogenic NMVOC emissions during warm summer days, often those days most conducive to ozone formation.

Biogenic NMVOC (BVOC) include a wide range of chemical compounds, including hydrocarbons, oxygenated compounds, sulphur-, and nitrogen-containing substances. A qualitative summary of worldwide investigated plants with respect to BVOC emission can be found in the work of *Hewitt et al.* [1997, available at <http://www.es.lans.ac.uk/es/people/pg/pas/download.html>]. Emission factor estimates by chemical compound for common European plant species have been summarized by *Steinbrecher* [1997].

The hydrocarbons isoprene and several monoterpenes (α -pinene, β -pinene, limonene, etc.) are generally considered the most important compounds for regional ozone formation in Europe [*Simpson*, 1995; *Stockwell et al.*, 1997].

The remaining NMVOC species ("other VOC" or OVOC) include some reactive species such as alkenes [*Goldstein et al.*, 1996] but are generally dominated by oxygenated compounds. Little is known about the source strength and chemistry of these compounds, and a large number of these OVOC, especially oxygenated compounds, have proven difficult to quantify in air samples [*Puxbaum*, 1997]. Emissions may be large, however, especially during the flowering and harvesting seasons [*Arey et al.*, 1991a, b].

Previous efforts to estimate biogenic NMVOC emissions for Europe have been faced with very limited databases of vegetation distributions and biogenic NMVOC emission rate factors [*Simpson et al.*, 1995]. The large uncertainties associated with these emission rates are mainly due to ill-defined land use classifications, emission factors, and emission algorithms. Recent investigations in Europe have resulted in an improved

understanding of biogenic NMVOC emissions from parts of the Mediterranean area [*Seufert et al.*, 1997], although many species and areas still need to be studied.

Here we present a revised BVOC emission calculation for Europe, which improves on previous efforts in several important respects: (1) emissions of isoprene, monoterpenes and OVOC are separately estimated and in a consistent fashion; (2) a large amount of new information has been collected on the different forest species in each country; (3) detailed land use maps have been utilized that enable a better estimate of the species distributions within a country; and (4) new emission rates have become available for many important European species, for example, for terpene emissions from evergreen oaks.

3.2. Natural Grassland and Other Low Vegetation

This section deals with NMVOC emissions from all types of vegetation (natural, seminatural and in some cases cultivated) that do not fit easily into the forest classification. We do not consider emissions of other species, for example, of NH_3 from pastures (due to animal droppings) and meadows (in particular when fertilized with manure), although these may be significant, as these are dealt with under Agricultural emissions in the UN-ECE/CORINAIR system. Some of the main vegetation types are the following:

1. Grasslands are areas dominated by grassy plants, usually also containing other herbs. There are mainly two families of grassy plants: *poaceae* ("sweet grasses") and *cyperaceae* ("acidic grasses"), the first of the two being most frequent in European grasslands.
2. Maquis are comprised of evergreen shrubs and small trees, typically olive (*Olea oleaster*), carob (*Ceratonia siliqua*), dwarf *Quercus ilex* and *Erica multiflora*. (Also known as matorral denso, espinal, chaparral, macchia alta.)
3. Garrique are comprised of midheight shrubs, 0.6–2 m high on calcareous soils, typically *Pistachia lentiscus*, *Arbutus*

unedo, *Myrtus communis*, and *Ulex* sp. (also known as matorral claro, scrub, macchia bassa).

4. Monte-hueco, which means vacuous forest, consists of pastures with scattered and degraded oaks (*Q. ilex*, *Q. suber*).

Names and definitions of Mediterranean landscape classes vary from country-to-country and from author-to-author [Di Castrì *et al.*, 1981; Veldt, 1989]. For example, garrique is sometimes used for vegetation <0.6 m high also, in which case “lande,” “tomillar,” “gairriga,” and “phyrgana” are alternative names. However, the above are in common usage.

Most of the grasslands in middle and northern Europe are seminatural, in the sense that they are agriculturally used for either harvesting hay (meadows) or for grazing (pastures). Natural grasslands can be found in alpine regions above the timberline (alpine Tundra), at lower elevation northward of the timberline (boreal Tundra), in dry climatic regions (Steppe), or on saltflats and on moorland.

Other low vegetation is widespread across Europe. For example, in the United Kingdom, bracken and dense shrub heath cover 12% of the land surface (R. Bunce, ITE, Merlewood, personal communication, 1997) with *Erica* sp., *Ulex* sp., *Calluna* sp., *Pteridium* sp., and similar species being common. In the Mediterranean region maquis, garrique, and jaral are characteristic landscapes.

Only a small number of screening studies have surveyed biogenic VOC emissions from nonforest vegetation. Hewitt and Street [1992] tested the 21 most abundant grass and herbaceous species in the United Kingdom with a qualitative method. Only purple moor grass (*Molinia caerulea*), bracken (*Pteridium aquilinum*), and common gorse (*Ulex europaeus*) were found to emit isoprene, and only ivy (*Hedera helix*) and cocksfoot grass (*Dactylis glomerata*) were found to emit monoterpenes. König *et al.* [1995] tested VOC emissions from agricultural plants such as wheat, rye, rape, grape, and three types of grassland in East Austria. They used the Arey *et al.* [1991b] approach to include also specified OVOC emissions. Wheat, rye, oilseed rape, grape, and two of the grass plots examined were found to emit primarily OVOC. For one of the examined grass plots, terpene and OVOC emissions were of equal importance. After mowing of one of the grass plots the emissions of terpenes and OVOCs increased roughly by a factor of 3, so presumably herbivory has a similar effect.

3.3. Forest Fires

On a global scale, biomass burning has been estimated to account for over 40% of global CO and CO₂ emissions and over 30% of NO_x emissions. However, most burning takes place in the tropical and subtropical regions [Andreae *et al.*, 1988], so emissions from European fires have received very little attention.

Fires have always been a feature of forest ecosystems. However, although natural forest fires may be initiated by lightning, recent estimates indicate that in Europe the vast majority of fires are associated with man's presence, despite the fact that slash-and-burn agricultural practices, or prescribed burns (upon which most emission-factor measurements are based), are much less frequent than in the Tropics or America. Korovin [1996] reports that nearly 70% of fires in Russia occur within 5 km of a road, and 60% of fires occur within 10 km of a populated area. Conrad and Ivanova [1997] report that lightning-induced fires account for over 50% in remote regions of the Asian part of Russia, but only 3% in the European part. Stanners and Bourdeau [1995] suggest that in Germany and

Italy <1% of fires are caused by lightning, with 3% and 8% cited for Finland and Spain, respectively. The single biggest cause of fires in Europe is stated to be arson, followed by accidents.

The major products of biomass burning are CO₂ and water vapor [Andreae *et al.*, 1988]. However, a large number of aerosol- and gas-phase species are produced, including the products of incomplete combustion (CO, NMHCs) and nitrogen and sulphur species. These arise partly from nitrogen and sulphur contained in the vegetation and organic matter in the surface soils. Additionally, emissions can arise from the volatilization of substances that have been deposited [Hegg *et al.*, 1990].

3.4. Soils

3.4.1. Soil NO_x emissions. Nitric oxide (as well as N₂ and N₂O) is produced in intermediate steps in microbial nitrification and denitrification processes. As emissions depend on the amounts of nitrogen going through these processes, agricultural soils, subject to direct fertilization and manure, are responsible for the great majority of emissions, and some ecosystems may have NO_x fluxes approaching those of anthropogenic sources [E. Williams *et al.*, 1992]. The quantity of NO_x emissions from agricultural land is dependent on the rate of fertilizer application and the subsequent microbial nitrogen processing in the soil, together with a multitude of other environmental factors. A large number of studies have been discussed in relation to possible controlling factors by Davidson and Kingerlee [1997], Skiba *et al.* [1997], and Veldkamp and Keller [1997]. Some of the major problems in parameterizing soil NO_x emissions are further discussed by Hutchinson *et al.* [1997].

Bakwin *et al.* [1990] pointed out that some of the NO emitted from soils is quickly converted to NO₂. This gas is deposited within a vegetation canopy, so reducing the NO_x flux to the atmosphere. Indeed, the study of Yienger and Levy [1995] gave a global emission estimate of 10.2 Tg N yr⁻¹ without this “canopy-reduction” effect but only 5.5 Tg N yr⁻¹ when it was included. However, most NO fluxes from agricultural land are measured by cover box techniques, enclosing crop and soil. In this case, canopy reduction factors do not need to be considered. Canopy reduction should probably be considered for forests, but in general these are expected to contribute little to overall emissions.

Although the magnitude of soil NO_x emissions may be small in overall comparison to anthropogenic NO_x emissions, there is considerable uncertainty in the estimates, as will be illustrated in section 6. Further, soil NO emissions occur in low-NO_x regions where ozone formation is most sensitive to NO_x availability, and the highest fluxes of NO occur in the warmer months of the year, times when photochemical smog is of concern.

Emissions of NO_x from soils are estimated to be between 10 and 20% of the global budget of NO_x in the troposphere [Lee *et al.*, 1997; Levy *et al.*, 1996]. The contribution of soil NO emissions from agricultural lands has previously been estimated to be 15% of the total European NO_x emissions inventory [Simpson *et al.*, 1995].

3.4.2. Soil CH₄ consumption. Methane production has been observed to occur in forest soils, especially in the upper organic horizons of soil profiles [e.g., Adamsen and King, 1993; Bender and Conrad, 1994], but not at rates exceeding the consumption at deeper horizons. Thus the net methane exchange

between soils and the atmosphere is negative, that is, forest soils consume atmospheric methane through a biologically mediated process [e.g., *Crill*, 1991]. With the exception of tundra soils [*Whalen and Reeburgh*, 1990], oxidation zones seem to form a few centimeters below the humus layer in all forest soils [*Whalen et al.*, 1992; *Adamsen and King*, 1993; *Koschorreck and Conrad*, 1993; *Yavitt et al.*, 1993; *Bender and Conrad*, 1994; *Tyler et al.*, 1994; *Czepiel et al.*, 1995; *Priem et al.*, 1996; *Roslev et al.*, 1997]. The reason for this depth variation has not yet been clarified, neither have any organisms responsible for the process been isolated [*Hanson and Hanson*, 1996]. Nonforested soils exhibit a wide range of behaviors, but again seem to act as a sink of methane, although with lower consumption rates than forest soils.

Both temperature and soil moisture affect methane consumption rates. The presence of water in soil pores reduces the diffusion rate of methane and oxygen, which are the methanotrophic substrates. Soil moisture, negatively correlated with methane uptake, was found to be the dominating factor in most studies [*Crill*, 1991; *Nesbit and Breitenbeck*, 1992; *Koschorreck and Conrad*, 1993; *Lessard et al.*, 1994; *Castro et al.*, 1994b, 1995; *Dobbie and Smith*, 1996]. Soil particle size also affects porosity and diffusion rates, so that a soil with large particles can promote high methane oxidation rates [*Dörr et al.*, 1993; *Bender and Conrad*, 1994]. In a cluster analysis made on a large data set, *Ball et al.* [1997] found that parameters related to soil porosity were the most important. In other studies, soil temperature was found to be the dominating factor [*Macdonald et al.*, 1997; *Priem and Christensen*, 1997], thus strengthening a seasonal pattern with highest uptake rates in summer.

Another factor is the age of the standing vegetation, with oxidation rates being generally higher in environments with older stands [*Kruse and Iversen*, 1995; *Priem et al.*, 1997].

Among nutrients, the effects of nitrogen has been most intensively studied, but the results are ambiguous. In some cases, nitrogen fertilization resulted in a negative impact on methane oxidation rates [e.g., *Steudler et al.*, 1989; *Castro et al.*, 1994a; *Schnell and King*, 1994; *Sitaula et al.*, 1995], while other field experiments with N fertilization showed no effect at all [*Whalen et al.*, 1991; *Dunfield et al.*, 1995] or possible recovery within one year [*Neff et al.*, 1994; *Börjesson and Nohrstedt*, 1998]. Further, both negative [*Dobbie and Smith*, 1996; *Macdonald et al.*, 1997] and positive [*Goldman et al.*, 1995] correlation with respect to ammonium have been reported. Other observations [*Castro et al.*, 1995; *Kruse and Iversen*, 1995] indicate that there is a positive relationship between soil fertility and consumption of atmospheric methane. Since no long-term negative effects of N-fertilization in forest soils have been shown, and as a normal fertilizer intensity is expected at one application per life cycle, the added nitrogen lowers methane consumption by <1% and is therefore left out of our forest methane budgets.

A low pH promoted methane oxidation in an experiment with acidic irrigation conducted by *Sitaula et al.* [1995], but other pollutants with possible inhibitory effect remain to be investigated.

3.5. Wetlands

For the purposes of this work "wetland" is used as an overall term for any area of permanently or seasonally flooded soils, where soils are saturated long enough for the soil to become reduced, and a methanogenic population established. The types of wetlands are differentiated by soil characteristics, their

vegetation, which affects the amount and type of organic substrate available and transport of CH₄, and by season of flooding or thawing. The distribution of these wetlands throughout Europe is very uneven, concentrated in northern Europe. An extreme example is Estonia, where >20% of the country is wetland.

Wetlands emit CH₄, biogenic sulphur gases, some VOC, and small amounts of N₂O. As discussed below, CH₄ is the only gas emitted in quantities that are globally significant. However, it should be noted that peatlands are large terrestrial carbon stores (circa 412 Gt of C is stored in peatlands worldwide [*Woodwell et al.*, 1995]) and thus play a significant role in the global carbon cycle [*Oechel*, 1993].

Wetlands are here divided into six classes: bogs, fens, swamps, marshes, flood plains, and shallow lakes. This classification is derived from *Zoltai and Pollet* [1983] and was also used by *Aselmann and Crutzen* [1989]. A similar scheme was used by *Matthews and Fung* [1987].

Agricultural wetlands such as rice fields are not covered, though the biogeochemical processes resulting in emissions are the same. (See *Schütz et al.* [1989] for fluxes from Italian rice fields.) Wetland areas are affected by human management when drained for agriculture, peat harvest or construction; maintained for wildlife habitat or water treatment; or built and/or converted for water storage and transport such as farm ponds, or canals.

3.5.1. Methane. Methane is produced by anaerobic bacteria (methanogens) in the soil, diffused through soil water and transported to the atmosphere by plants, ebullition, or diffusion. Ground water-table depth, type of vegetation, soil characteristics, available substrates, and local climate are all important factors regulating methane emissions [*Gujer and Zehnder*, 1983; *Conrad*, 1996, and references therein].

Emissions vary seasonally, usually following soil temperature, plant growing season or saturation season, though exceptions may be found [*Svensson and Rosswall*, 1984; *Whalen and Reeburgh*, 1992; *Westermann*, 1993]. For example, in high northern latitudes, wetlands are usually classified as bogs, forested bogs, and fens with maximum emissions from June to September. Methane emissions increase when soil temperature increases above 0°C but have been measured at very low levels from frozen soil. Seasonal wetlands such as flood plains will only emit methane during the wet season, and methane emissions vary within wetlands along moisture gradients [*Svensson*, 1976; *Moore et al.*, 1990]. Dry, aerated soils are usually sinks of methane; though drought or other changes in the water table may cause a source area to become a sink [*Harriss et al.*, 1982; *Whalen et al.*, 1991].

Cao et al. [1996] and *Christensen et al.* [1996] have modeled the carbon system and methane emissions from wetlands. This type of model is rather complicated but allows modeling changes of methane emissions due to changes in climate. However, these modeled flux estimates cannot yet be regarded as a replacement for other global estimates, as they have mainly been intended to duplicate earlier results based on measurements [*Matthews and Fung*, 1987; *Aselmann and Crutzen*, 1989; *Bartlett and Harris*, 1993].

Wetlands are estimated to produce ~20% of the annual global methane emissions. Recent global estimates have been 100–110 Tg per year, with a range of ~50–150 Tg [*Matthews*, 1993].

3.5.2. Biogenic sulphur gases. Sulphur gases emitted from wetlands are estimated to be a small percentage of the

total sulphur budget [Warneck, 1988; Andreae, 1984], although estimates vary widely. Bates *et al.* [1992] suggest a conservative annual estimate of $0.008 \text{ Tg S yr}^{-1}$ from wetlands, whereas Andreae [1990] suggest $1\text{--}2 \text{ Tg S yr}^{-1}$ from saline marshes. Recent estimates of total sulphur emission are around $100\text{--}200 \text{ Tg}$ [Benkovitz *et al.*, 1995; Bates *et al.*, 1992; Bluth *et al.*, 1993]. Early studies which indicated a much larger source of biogenic sulphur gases from wetlands were either not reproduced or may have been an artifact of the sampling process (see Chin and Davis [1993] for further discussion). Sulphur emissions from wetlands are thus not significant compared to other sources and are thus not considered further here.

3.5.3. NMHC. Recent work has shown that Sphagnum wetlands emit isoprene, some monoterpenes, and a few light hydrocarbons, notably ethene and propene [Klinger *et al.*, 1994; Janson and De Serves, 1998]. The latter found that isoprene clearly dominated the flux from Sphagnum fens in southern Sweden and Finland, with fluxes as high as $1800 \mu\text{g C m}^{-2} \text{ h}^{-1}$ from the wet flark environments, but only up to $70 \mu\text{g C m}^{-2} \text{ h}^{-1}$ from the drier hummock environments. Nighttime emissions were reported to be zero or near zero, probably due to the absence of light as is the case with foliar emissions. The emission season starts after the spring thaw and ends with the onset of the autumn frost. June emissions appear to be lower than August emissions. Other details of the seasonal variation are unclear, but moss and soil temperature, and PAR are expected to be important controlling factors.

3.6. Waters

Emissions of reduced sulphur compounds and NH_3 from European seas and lakes are considered for this inventory. Emissions of other compounds such as VOCs have been measured from oceans [Rudolph and Ehhalt, 1981; Bonsang *et al.*, 1988; Plass-Dülmer *et al.*, 1993; Guenther *et al.*, 1995], but these are not expected to be major contributors in the European area. Emissions of CH_4 from shallow lakes have been discussed in section 3.5.

The biological activity of marine and freshwater algae is considered to be responsible for the production of reduced sulphur gases, especially dimethylsulphide (DMS), carbon disulphide (CS_2), methylmercaptan (CH_3SH), and dimethyldisulphide (CH_3SSCH_3). Carbonyl sulfide (OCS) and hydrogen sulfide (H_2S) are photochemically produced, in particular OCS in seawater can result from the photosensitized reaction of organosulphur compounds. Of all these components, DMS is by far the most abundant and contributes $\sim 90\%$ of the total reduced sulphur in surface waters [Cline and Bates, 1983; Leck and Rodhe, 1991].

Current global estimates of DMS fluxes to the atmosphere vary from 16 Tg S yr^{-1} [Bates *et al.*, 1992] to 39 Tg S yr^{-1} [Andreae and Raemdonck, 1983]. There is general agreement on considering the global flux of OCS and CS_2 to be $1\text{--}2\%$ of the DMS flux, while the global flux of H_2S can possibly be as high as 10% of the DMS flux [Saltzman and Cooper, 1988]. On a global scale, the contribution of reduced sulphur from surface waters represents between 15 and 25% of the total sulphur emissions to the atmosphere.

For oxidic freshwater lakes, DMS is again the major reduced sulphur compound emitted [Turner and Liss, 1985] and its production depends on the type of aquatic ecosystem. Although observations of volatile reduced sulphur emissions in hypersaline evaporitic environments show DMS concentrations of the same order of magnitude as those observed in

marine environments (32 to 128 ng S L^{-1} in South Catalonia, Spain), the concentrations in neighboring sulphate-rich karstic lakes can be considerably smaller [Simó *et al.*, 1993]. Large variations were also reported by Nriagu and Holdway [1989] in the Great Lakes, United States, with DMS concentrations ranging during summer from 3.2 to 32 ng S L^{-1} . Measurements of DMS in lakes are sparse and highly variable, but since the volume of lakes in Europe, and globally, are at least 2 orders of magnitude smaller than the volume of salt water, we can expect the freshwater contribution to global (or European) values to be in the same proportion.

The emission of ammonia from ocean waters is also biologically determined, as it depends on the ammonium contents in seawater. Oceanic ammonium is produced from zooplankton grazing and from the decay of organic material. It is a nutrient for both phytoplankton and bacteria. Ammonia can either be emitted from or deposited to ocean waters; the direction of the flux depends upon a number of physical and chemical parameters including the atmospheric concentration of ammonia [Asman *et al.*, 1994].

Estimates of the oceanic emission of ammonia vary from the global flux of 13 to 7 Tg N yr^{-1} . The higher estimate was extrapolated by Schlesinger and Hartley [1992] from observed averaged fluxes of $98 \text{ mg N m}^{-2} \text{ d}^{-1}$ reported by Quinn *et al.* [1990]. The lower estimate was derived by Dentener and Crutzen [1994] by assuming equal molar emissions of NH_3 and DMS as proposed by Liss and Galloway [1993]. More recently, Bouwman *et al.* [1997] propose a global emission of 8 Tg N yr^{-1} based on the model calculations of Six and F. J. Dentener (manuscript in preparation, 1998). According to this estimate, the oceanic source would represent $\sim 15\%$ of the total flux of ammonia to the atmosphere, although there remain uncertainties because the existence of nonzero air concentrations of NH_3 has not been considered in these calculations.

3.7. Wild Animals and Humans

The major global sources of methane are mammals (primarily ruminants and rodents) and termites, although the latter are not considered important in Europe. Metabolic processes in the intestines of animals, and to a lesser extent in their excretions are responsible for gas formation. One important pathway is the anaerobic degradation of cellulose plant material by symbiotic microflora leading to the formation of methane [Crutzen *et al.*, 1986]. The other known pathway causing ammonia emissions (and also N_2O) derives from the decay of urea and uric acid in animal manure [Buijsman *et al.*, 1987; Bouwman *et al.*, 1997]. This latter process is much more pronounced for domestic animals than for wild animals (as farm manure is collected and kept liquid for longer periods of time) or for other sites where animals live in very dense populations (e.g., bird breeding colonies on small islands [see Sutton *et al.*, 1995]). Other emissions include VOCs such as isoprene; however, this source is negligible compared to emissions from plants.

Within this source group, emissions from human evaporations (sweat, breath) are considered, but in agreement with the EMEP/CORINAIR methodology not the emissions from sewage treatment.

3.8. Lightning

The electrical discharge of lightning creates plasma channels in the atmosphere characterized by strong ionization and high temperatures. Major compounds of the atmosphere, notably

nitrogen, oxygen, and water, may be ionized and then undergo chemical transformation [Sisterson and Liaw, 1990; Gallardo and Cooray, 1996]. The major species of interest is nitric oxide (NO), but other compounds containing nitrogen, oxygen, and hydrogen atoms are also formed. The high temperature during the flash (up to 30000 K), and the subsequent rapid cooling below 1500 K, are crucial for the formation and subsequent stabilization of NO.

While lightning exhibits different characteristics depending on whether it is cloud-to-ground (CG), cloud-to-cloud or within-cloud (intercloud and intracloud, IC), emission estimation techniques have not been resolved to this level of detail. It has been reported that IC discharges may be up to 10 times less efficient in producing NO than the CG discharges [Sisterson and Liaw, 1990]. However, more recent information suggests that these discharges may be nearly equal [Levy *et al.*, 1996; Gallardo and Cooray, 1996]. The amount and distribution of NO produced is believed to depend on the energy and the frequency of lightning strokes, which in turn depend on cloud temperatures and cloud heights. IC lightning is known to be more frequent than CG lightning. The ratio has been correlated to the cold cloud thickness (cold cloud, below freezing temperature), representing the size of the electric field involved that may determine the number of IC flashes [Price and Rind, 1993]. Despite generally large variations in this ratio, a dependence on geographical latitude has been found using cold cloud thickness as a parameter. While virtually all of the oxidation product is originally NO, ~25% of this may be transformed to NO₂ very quickly [Franzblau and Popp, 1989].

3.9. Volcanoes

This source category, important in Italy and Iceland, includes emissions from geothermal activities, both eruptive and noneruptive. Sources include fumaroles, geysers, metamorphic degassing or other activities related to molten magma from the Earth's crust, but volcanoes themselves are by far the dominant source of emissions. The gases emitted consist of sulphur dioxide, carbon dioxide, hydrogen sulphide, mercury, and chlorine, among others. Noneruptive volcanoes that outgas at relatively constant rates seem to be more important than sporadic eruptions, both for CO₂ [Gerlach, 1991] and SO₂ [Andres and Kasgnoc, 1998]. However, the sporadic emissions are much more difficult to assess.

Differentiation can also be made among eruptive emissions: eruptions in an arc tectonic regime tend to be the most violent but seem to have a more predictable pattern of explosivity strength versus SO₂ emissions [Bluth *et al.*, 1993]. In general, the volcanic sources in Europe are well known and documented.

3.10. Gas Seeps

Bubbles of natural gas rising from the seafloor have been detected at several different sites in the world. Such emissions have been attributed to seepage from underground storage, but very little information is available. Obviously, seepage has been observed primarily in lakes and coastal areas (in Europe primarily around Denmark), but also further out, in particular in the North Sea. Depending on the bubble size and on the water depth, some or even all of the methane emerging from the sea floor may be dissolved in water and readily oxidized by bacteria. Therefore atmospheric emissions are limited to the continental shelf.

Seepage usually occurs at storage sites of relatively recent

times, such as from the glaciation periods. Natural gas reservoirs may start seeping gas after seismic activities (earthquakes) or in response to changes in the outside temperature and pressure. Observed gas flow takes place primarily during summer and autumn, and during low tide [Martens and Klump, 1980], often taking the form of activity outbursts. Such variable methane emissions are assumed to be produced by on-going processes from buried organic material, with reservoirs being emptied periodically. Carbon 14 depletion in the resulting methane emission suggests that this material is from fossil origin [Hovland *et al.*, 1993]. In addition to bacterial production, thermogenic production is assumed, with the associated emissions being accompanied by oil seeps. Emissions are typically composed of ~75% methane, 7% propane, 7% n-butane, and 6% ethane (by weight [Radian Company, 1996]).

4. Emissions and Methodology

4.1. Forests

For forests, grasslands and other vegetation, an appropriate system describing the emissions flux F (in $\mu\text{g m}^{-2} \text{h}^{-1}$) of NMVOC species, is based on Guenther [1997]:

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

The emission potential ε is the emission rate expected for a particular plant species at a reference temperature of 30°C and PAR of 1000 $\mu\text{mol photons (400–700 nm) m}^{-2} \text{s}^{-1}$. "D" is the foliar biomass density ($\text{g dry weight (DW) m}^{-2}$), γ is a dimensionless environmental correction factor representing the effects of temperature, and in some cases solar radiation, on emissions.

The emission estimates to be presented here have been made using the meteorological data from the EMEP ozone model, solving (1) as described by Simpson *et al.* [1995]. In order to obtain a representative estimate, 5 years of meteorology have been used.

Two major alternatives for using (1) to calculate emission inventories are (1) to perform these calculations at a genus or preferably species specific level (requiring for example separate statistics for Norway spruce, Douglas fir, etc.); (2) to perform the calculations for different ecosystem types. In this method, each ecosystem is assumed to consist of a number of species, and the assigned emission rates attempt to give the average emissions from this category.

The first approach, using genus and species level data, is the one we have adopted for European forest estimates. This approach requires considerable effort but is justified because of the difficulties in assigning ecosystem-specific emission factors for Europe. For example, isoprene emission potentials for a deciduous forest in Germany range from almost zero to 40 $\mu\text{g g}^{-1} \text{h}^{-1}$ depending on the relative amounts of European oak and beech in this ecosystem type.

The very low species diversity in Europe (see section 2 above) also encourages a species-specific approach. In Norway, Sweden, Finland, Germany, and several other countries it is a very good approximation to say that all spruce is *Picea abies* and all pine is *Pinus sylvestris*. An extensive botanical textbook on Norwegian and Swedish flora mentions only two oak species, *Q. robur* L. and *Q. petraea* (Matt.) Liebl. [Lid, 1974]. Other species are present in these countries of course but often for ornamental purposes in very limited numbers.

The situation is more complex in the Mediterranean area

Table 2. Biomass Densities D and Emission Potentials ϵ for Isoprene and Both Light-Dependent and Temperature-Only-Dependent Emissions of Monoterpenes

Common Name (Example)	Latin Name	D , g/m ²	Emission Potentials			Additional References	
			Isoprene (1)	Terpenes (2)		Isoprene	Terpenes
				Light	Store		
Fir	<i>Abies</i>	1400	0.1	...	3.0		
Maple/sycamore ^a	<i>Acer</i>	320	0.1	...	3.00	S93	S93
Alder	<i>Alnus</i>	320	0.1	...	1.5	S93	S93
Birch	<i>Betula</i>	320	0.1	...	0.2	K, HLR	P, K, HLR
Hornbeam	<i>Carpinus</i>	320	0.1	...	0.65	K	K
Chestnut	<i>Castanea</i>	320	0.1		
Orange	<i>Citrus</i>	300	0.1	...	1.5		
Cypress	<i>Cupressus</i>	700	0.1	...	0.65		
Blue gum*	<i>Eucalyptus</i>	400	20.0	...	3.0	Str97b	H
Beech	<i>Fagus</i>	320	0.1	...	0.65	P, S93, K, Sh	K, Sh
Ash	<i>Fraxinus</i>	320	0.1	S93	S93
Juniper	<i>Juniperus</i>	700	0.1	...	0.65	O	
Larch	<i>Larix</i>	300	0.1	...	1.5	S93	S93
Olive	<i>Olea</i>	200	0.1		
Spruce*	<i>Picea</i> sp.	varies (3)	1.0	1.5	1.5	as <i>P. abies</i>	as for <i>P. abies</i>
Norway spruce*	<i>Picea abies</i>	varies	1.0	1.5	1.5	J98, S94, Ke	S94, SS, Ke, J93, 98, LP
Sitka spruce*	<i>Picea sitchensis</i>	varies	6.0	...	3.0	Str96, 97b, Sm	Str96, 97b
Pines ^b	<i>Pinus</i> sp.	700	0.1	...	3.0		...
Aleppo pine*	<i>Pinus halepensis</i>	700	0.1	...	0.65		H
Umbrella pine*	<i>Pinus pinea</i>	700	6.0	Sf, Ks97, Std, Str97a	Sf, Ks97, Std, Str97a
Maritime pine*	<i>Pinus pinaster</i>	700	0.1	...	0.2		Si
Scots pine*	<i>Pinus sylvestris</i>	varies (4)	0.1	...	1.5		J93
Pistachio	<i>Pistacia</i>	320	0.1	...	3	H, Ha	H, Ha
Plane ^c	<i>Platanus</i>	320	34.0		
Poplar	<i>Populus</i>	320	60.0	H, HLR	HLR
Cherry	<i>Prunus</i>	300	0.1		
Douglas fir	<i>Pseudotsuga</i>	1000	0.1	...	1.5		D
Oaks							
Default deciduous ^d *	...	320	60.0	...	0.2	Sf	Sf
Default evergreen*	...	500	0.1	20.0	...	Sf	Sf
Turkey oak*	<i>Q. cerris</i>	320	0.1	...	1.0	S97	S97
Holm oak	<i>Q. ilex</i>	500	0.1	20.0	...	Ks96, Be, Str97a	Ks96, 97, Be, Str97a
Sessile oak	<i>Q. petraea</i>	320	60.0	...	0.2	S97, K, Str97b	K, S97
Downy oak	<i>Q. pubescens</i>	320	60.0	...	0.2	S97	S97
European oak	<i>Q. robur</i>	320	60.0	...	0.2	S93, I	S93
Cork oak*	<i>Q. suber</i>	500	0.1	...	0.2	S97	S97
Locust	<i>Robinia</i>	320	34.0
Willow	<i>Salix</i>	150	34.0	Op, HLR	S93, HLR
Elm	<i>Ulmus</i>	320	0.1	...	0.2		

(1) Minimum isoprene rate set to 0.1 for all species, see text. Branch-level rates given; (2) Terpene emission potentials given as "LIGHT" use MTL algorithm, "STORE" use MTS algorithm, see text; (3) For spruce, $D = 800$ g/m² north of 60°N, 1400 g/m² between 55°–60°N, and 1600 g/m² < 55°N; (4) For Scots pine, $D = 500$ g/m² north of 60°N, 700 g/m² elsewhere. Emission potentials given in $\mu\text{g g}^{-1}$ DW h⁻¹. Additional references: Be, *Bertin et al.* [1997]; D, *Duyzer* [1993]; H, C. N. Hewitt and S. Owen, personal communication, 1997; HLR, *Hakola et al.* [1998]; I, *Isidorov* [1985]; J93, 98, *Janson* [1993]; *Janson et al.* [1998]; K, *Koenig et al.* [1995]; Ke, *Kempf et al.* [1996]; Ks96, 97, *Kesselmeier et al.* [1996, 1997]; LP, *Lindskog and Potter* [1995]; Ha, *Hanson et al.* [1997]; O, *Owen et al.* [1997]; Op, S. Owen (personal communication, 1998); P, *Puxbaum* [1997]; Sh, *Schuh et al.* [1997]; Si, *Simon et al.* [1994]; Sf, *Seufert et al.* [1997]; Sm, *Simpson et al.* [1995]; Std, *Staudt et al.* [1997]; S94, *Steinbrecher* [1994]; S93, 97, *Steinbrecher et al.* [1993b, 1997a]; Str96, -97a, -97ba, *Street et al.* [1996], [1997a, b]. Asterisk indicates ϵ values not derived from *Guenther et al.* [1994, 1997] and *Geron et al.* [1994] for genus level.

^aEuropean sycamore, *Acer pseudoplatanus*.

^bPine default rates include *P. brutia*, *cembra*, *contorta*, *laricio*, *mughus*, *nigra*, *radiata*, *strobus*.

^cAlso called American sycamore in the United States.

^dIncludes *Q. Faginea*, *Q. borealis*, *Q. Lusitanica*, and *Q. pubescens*.

where many more species are present. Emission characteristics even within the same genus are quite variable, for example, common Mediterranean oaks may emit either isoprene or monoterpenes or none of these compounds [*Seufert et al.*, 1997; *Steinbrecher et al.*, 1997a]. However, the increasing prevalence of commercially grown species such as eucalyptus is reducing the complexity of this region. Indeed, reasonable statistics can often be found on the main species because of their economic importance, so a species-specific approach again seems reasonable.

4.1.1. Emission potentials ϵ . Isoprene and total monoterpene emission factors for common European forest species are listed in Table 2. In order to include the dependence of some terpene emissions on light, two separate emission potentials are given for terpenes; denoted "LIGHT" (actually light-and-temperature dependent) and "STORE" (only temperature dependence). The appropriate environmental correction factors to be applied with these potentials are discussed in section 4.1.3.

In general, emission factors have been specified for a genus

Table 3. Ecosystem-Default Emission Potentials ϵ and Biomass Densities D

Ecosystem	Emission Potential				Main Reference
	D , g m ⁻² DW	Isoprene $\mu\text{g g}^{-1}$ h ⁻¹	Terpene		
			Light	Store, $\mu\text{g g}^{-1}$ h ⁻¹	
Grass	400	0.1	...	0.1	K
Pasture	400	0.1	...	0.1	K
Maquis	400	8	...	0.65	O, G95
Garrique	200	8	...	0.65	O, G95
Moorland/heathland	200	8	...	0.65	C
Mediterranean Shrub	200	8	...	0.65	O
Monte-hueco ^a	100	1	10	...	
Crops	1000	0	...	0.1	PK

Notes: Terpene emission potentials given as light-dependent (Light) and temperature-only-dependent (Store); C, Cao *et al.* [1997]; O, Owen *et al.* [1997]; summertime rates considered; K, König *et al.* [1995]; G95, Guenther *et al.* [1995]; PK, default estimate for crops based on Puxbaum *et al.* [1997]; König *et al.* [1995].

^aBiomass from Ortiz and Dory [1990], emission potentials assume 50% *Q. ilex*, 50% *Q. suber*.

or species. Isoprene emission potentials are set in classes at one of 0.1, 1, 6, 10, 20, 34, or 60 $\mu\text{g g}^{-1} \text{h}^{-1}$, for “branch-level” conditions (see section 4.1.3). This class system, adapted from Guenther *et al.* [1994], has the disadvantage that emissions factors for a particular species are not usually set at the best available estimate. On the other hand, it avoids an artificial sense of accuracy, which is not warranted because of all the uncertainties associated with these measurements. Indeed, as shown by Guenther *et al.* [1996], measurements of individual species at specific locations may easily fall outside the range of the default class for that species.

Given the wide variety of emissions found for important European oak, spruce, and pine species, species-level descriptions were preferred over genus-level descriptions wherever possible for these genera, although emission potentials were still assigned to classes. Norway spruce has received somewhat special treatment as it so widespread over the continent, and a number of independent studies are available characterizing its emissions [Janson, 1993; Janson *et al.*, 1998; Steinbrecher, 1993b, 1994, 1997b; Kempf *et al.*, 1996]. Steinbrecher [1994, 1997b] has demonstrated a light dependency of the terpene emissions, so as a first approximation we have assumed that 50% of the terpene emissions are light dependent.

Note that a minimum isoprene emission potential of 0.1 $\mu\text{g g}^{-1} \text{h}^{-1}$ has been set for all species. This accounts partly for the fact that isoprene emissions are often detected but at unquantifiable levels in many species, and partly for emissions from unaccounted-for vegetation within the forest area. However, as we have attempted to map both dominant and nondominant forests onto our land use maps, as discussed in section 5.1, no extra accounting for nondominant emissions from trees is included.

For the other VOC (OVOC) very few reliable experimental data are available, especially for polar compounds with less than four carbon atoms (e.g., methanol, HCHO, etc.). However, it has been shown that plants can emit methanol [MacDonald and Fall, 1993], low molecular weight aldehydes [Kotzias *et al.*, 1997; Steinbrecher *et al.*, 1993a] and low molecular weight organic acids [Bode *et al.*, 1997] in significant quantities.

In the BEMA study Kesselmeier *et al.* [1997] report emission potentials from *Quercus ilex* of 0.22–0.44 $\mu\text{g g}^{-1} \text{h}^{-1}$ for formic acid, 0.037–0.29 $\mu\text{g g}^{-1} \text{h}^{-1}$ for acetic acid, 0.69 $\mu\text{g g}^{-1} \text{h}^{-1}$ for acetaldehyde, and 0.38–0.52 $\mu\text{g g}^{-1} \text{h}^{-1}$ for formaldehyde. Emission potentials for *Pinus pinea* were similar. Janson *et al.* [1998] found carbonyl (acetone and acetaldehyde) emissions of 0.3–4.6 from Norway spruce. Other measurements on European trees have generally shown emissions of >C4 compounds of 0–0.3 $\mu\text{g g}^{-1} \text{h}^{-1}$ [König *et al.*, 1995; Puxbaum, 1997], somewhat lower than values found in America of about 0.6 $\mu\text{g g}^{-1} \text{h}^{-1}$ [Arey *et al.*, 1991b; Winer *et al.*, 1992].

Until future studies reduce the large uncertainties associated with existing OVOC emission factors, we have decided to retain the previous default OVOC factor of 1.5 $\mu\text{g g}^{-1} \text{h}^{-1}$.

4.1.2. Foliar density D . For deciduous trees, foliar densities vary markedly over the year, and this can be straightforwardly incorporated into calculations if data were available. However, as pointed out by Isidorov *et al.* [1993], the relative weight of foliar biomass compared to total biomass increases in harsher conditions, and is also strongly depends on the age distribution of the forests. Such factors vary considerably over European forests; the information required to attempt an assessment of this in any detail is simply not available.

Instead, we make use of the seasonal average foliar biomass densities (Table 3), derived mainly from Veldt [1989]. These suggestions appear to fit quite well a wide range of measurements, but the variability of Mediterranean vegetation may introduce large uncertainties. For coniferous forests, Veldt suggests densities of 700–1400 g m^{-2} for different species south of 60°N, whereas Ortiz and Dory [1990] use 400 g m^{-2} . However, variations also occur in non-Mediterranean Europe. For example, Veldt [1989] suggests a default oak biomass of 320 g m^{-2} , whereas Andreani-Aksoyoglu and Keller [1995] quote 530 g m^{-2} for oaks in Switzerland.

4.1.3. Environmental correction factor γ . For isoprene emissions, γ is a function of light and temperature. We have used the so-called ISOG algorithm of Guenther *et al.* [1993], which has proved remarkably successful across a range of vegetation types ($\gamma_{iso} = C_{T_{iso}} \times C_L$ [see Guenther *et al.*, 1993; Guenther, 1997]).

Emissions of terpenes from most types of vegetation result from the volatilization of these compounds from stores contained within the plant tissue. Such emissions are assumed to be controlled only by temperature. Emissions from these stored terpenes are denoted “MTS” in this study and have been found to be well described by the algorithm of Guenther *et al.* [1993]:

$$\gamma_{mts} = C_{T_{mts}} = \exp [\beta(T - T_s)] \quad (2)$$

where ($\beta = 0.09 \text{ K}^{-1}$) is an empirical coefficient based on nonlinear regression analysis of numerous measurements present in the literature, and T_s is the standard temperature 303 K.

Past studies have used the above MTS algorithm to model all monoterpene emissions. However, one major outcome of the emission measurements on typical Mediterranean plant species was the detection of monoterpene emitting oaks, notably Holm oak (*Quercus ilex*) and Kermes oak (*Quercus coccifera*) [Seufert *et al.*, 1997; Steinbrecher and Hauff, 1996]. Unlike coniferous terpene emitters, these plants have no storage tissues for monoterpenes. This monoterpene emission is under strict temperature and light control similar to the isoprene emission from green tissue, indicating a de novo synthesis in

the leaves. It has also been demonstrated by *Steinbrecher* [1994a, 1997b] that Norway spruce (*Picea abies* L. Karst.) shows temperature and light dependent emission of monoterpenes. Similar findings for other plants containing monoterpene reservoir have also been reported by *Schuh et al.* [1997] and *Staudt et al.* [1997].

Thus we define a class of light- and temperature-dependent monoterpene emissions “MTL,” where

$$\gamma_{mtl} = \gamma_{iso}$$

Also, γ_{iso} is as given by *Guenther* [1997].

These algorithms will likely need to be revised in the future when a better biological understanding of the biosynthesis and emission of terpenoids is available, as there remains an uncertainty in the resulting emission estimates that is at least a factor of 3 or more. This variation is mainly due to the (1) differences in the emissions from branch to branch and from tree to tree, (2) variation with season, (3) nutrient condition of the plant, (4) stress, and (5) experimental errors. Recent work by *Schuh et al.* [1997] has suggested a possible alternative algorithm based upon studies of beech and sunflower, but such new algorithms will require extensive testing before they could be applied to inventory studies of all European species.

For OVOC, the relationship between environmental conditions and emissions is even less understood than for isoprene and monoterpenes. Emissions of some of these compounds, including a group of C6 unsaturates, are strongly influenced by external factors other than light and temperature, such as plant wounding by microbes, insects or mechanical stress. Given the lack of other information regarding the factors controlling oxygenated hydrocarbon emission, the use of (2) for the parameterization of OVOC emissions is recommended [*Guenther et al.*, 1994].

4.1.4. Other factors. When calculating the BVOC emission from a forest canopy the importance of lower layers to the total canopy emission has to be considered. Forest canopy environment models can be used to estimate the temperature and radiation environment at different heights within a canopy [e.g., *Pierce and Waldruff*, 1991; *Lamb et al.*, 1993]. These environmental conditions can then be used to calculate the VOC emission from each layer. Such canopy approaches should be used with “leaf-level” emission potentials (given, for example, by *Guenther et al.* [1994]).

A simpler noncanopy approach assumes that ambient temperature is similar to leaf temperature and that the use of “branch-level” emission potentials (as given in Table 2), which are typically a factor 1.75 smaller than leaf-level values [*Guenther et al.*, 1994], accounts for the shading effect. Tests in European conditions have suggested differences in total emissions between the two methodologies of ~20% [*Simpson et al.*, 1995]. Given the many uncertainties introduced by the forest-canopy model itself (e.g., in temperature and light profiles within the canopy), and the lack of evaluation of such models under European conditions, a canopy model is not used in the present European inventory.

A host of other, often longer-term, factors influence emissions, including leaf-onset, nutrients, water status or other stresses [*Guenther et al.*, 1995, 1997]. Possible algorithms to account for these longer-term effects have been proposed by *Guenther et al.* [1997], and for *Quercus robur* in Europe by *Schnitzler et al.* [1997]. These types of algorithms will undoubtedly improve emission estimates in future, but further evalua-

Table 4. Emission Factors for Biomass Fires

	Moles \times per 1000 Mole CO ₂ Emitted			g \times (kg C) ⁻¹ Emitted (“Best Guess”)
	Field Measurements	Laboratory Studies	“Best Guess”	
CO	6.5–140	59–105	100	230
CH ₄	6.2–16	11–16	11	15
NMHCs	6.6–11	3.4–6.8	7	21*
NO _x	2–8	0.7–1.6	2.1	8 (as NO ₂)
NH ₃	0.9–1.9	0.08–2.5	1.3	1.8
SO _x	0.1–0.34	...	0.3	1.6

After *Andreae* [1991].

*Average mass of NMC assumed to be 37, derived from *Radke et al.* [1991].

tion is required, and they have therefore not been adopted for the current inventory.

4.2. Natural Grasslands and Other Low Vegetation

The methodology for calculating emissions from grasslands and other low vegetation is almost identical to that for forest vegetation, except that we must use landscape emission potentials rather than species-specific factors in most cases. The justification for this is that grass-, shrub-, moor- and heathlands, and other low vegetation ecosystems consist generally of plant communities (except for crops which are usually monocultures), even though often only a few species dominate the community. The data required to resolve these communities into constituent species simply do not exist.

We have not found a comprehensive discussion of biomass densities for low vegetation. However, there is information on the annual net primary production for grasslands [*Ruimy et al.*, 1994; *Lieth and Whittaker*, 1975], from which we have derived default biomass factors (H. Puxbaum et al., manuscript in preparation, 1998). For moorland/heathland we have used values appropriate to Gorse (*Ulex*), assuming 50% ground cover. For the Mediterranean we have used values from *Veldt* [1989].

Emissions potentials and biomass factors assumed for these ecosystems are given in Table 3.

4.3. Forest Fires

Emissions are obtained in a two-step process: (1) Estimate the emissions of carbon from the burned land and (2) Estimate the emissions of other trace gases using emission ratios with respect to carbon. The basic calculation of the mass of carbon emitted, $M(C)$, follows the methodology of *Seiler and Crutzen* [1980]:

$$M(C) = 0.45 \times A \times B \times \alpha \times \beta \quad (3)$$

where 0.45 is the average fraction of carbon in fuel wood, A is the area burnt (m²), B is the average total biomass of fuel material per unit area (kg/m²), α is the fraction of the average aboveground biomass relative to the total average biomass B , β is the burning efficiency (fraction burnt) of the aboveground biomass.

Emission factors of trace gases relative to carbon emitted by burning are based on the recommendations of *Andreae* [1991] and are given in Table 4.

Values of B , α , and β are given for relevant biomes in Table 5. The biomass data B are derived from statistics on European forests and are about a factor of 2 lower than those recommended by *Seiler and Crutzen*. The main reason for this is

Table 5. Biome Characteristics and Emission Factors for Forest Fires

Biome	Biomass B , kg/m ²	Above Ground Biomass Fraction, α	Burning Efficiency, β	Derived Emission Factors, kg/ha, Assuming Default B , α , β values					
				CO	CH ₄	NMVOC	NO _x	NH ₃	SO _x
Boreal forest	10	0.75	0.2	1600	100	140	54	12	12
Temperate forest	20	0.75	0.2	3100	200	280	110	24	24
Mediterranean forest	4	0.75	0.25	780	51	71	27	6	6
Shrubland	2	0.64	0.5 ^a	660	43	60	23	5	5
Grassland (steppe)	2	0.36	0.5 ^a	370	24	34	13	3	2.9

NO_x as NO₂; The biomass data B are derived from sources given in Appendix A for boreal and temperate forests, and for Mediterranean forest derived from the Spanish CORINAIR 1990–1993 inventories [see also *Rodriguez Murillo*, 1994]; α and β coefficients from *Seiler and Crutzen* [1980].

^aAssuming burning efficiency of European grass/shrublands is less than the data on tropical biomes for which Seiler and Crutzen suggested 0.8.

probably that the Seiler and Crutzen data, and indeed similar data used by other workers [e.g., *Kasischke et al.*, 1995], are based heavily on North American ecosystems, where forests are generally much older and have a much larger fraction of biomass in the ground layer (litter, dead trees, etc.) than their European counterparts. For example, *Kasischke et al.* [1995] illustrate the biomass characteristics of Alaskan forests for stand ages of up to 600 years, and state that fire-return cycles limit the age of trees. In Europe, tree age is determined almost entirely by forestry practices and is usually <100 years. In the United Kingdom the average age of planted trees is just 29 years.

Some estimates of emissions from biomass burning distinguish between different phases of burning. In the “smoldering” phase emissions tend to be higher than in the burning phase [*Cofer et al.*, 1991], as it is the most easily combustible material which burns in the early phases. During the smoldering phase the less oxidized products (CO, HCs, etc.) are produced in higher proportions [*Cofer et al.*, 1989, 1991]. However, all phases of burning display a mixture of complete and incomplete combustion. Given the lack of data on typical European fires, and the lack of significant emissions, such refinements are not considered here.

An additional complication may be that forest fires can generate large convective elements (e.g., up to 5 km [*FIRESCAN*, 1996]), so that many of the emitted species are ejected into the free troposphere above the boundary layer. Such emissions should not be reported within the EMEP/CORINAIR system, but so far we are unable to say how large a fraction of the emissions are affected in this way.

4.4. Soils

4.4.1. Soil NO_x emission. A number of methodologies are available for estimating soil NO_x emissions. The most widely used so far by the atmospheric modelling community is that of *Novak and Pierce* [1993], commonly known as the second version of the Biogenic Emissions Inventory System (BEIS-2). BEIS-2 estimates NO emissions for forests, agricultural crops, urban trees, and grasslands as an exponential function of temperature, as determined by *E. Williams et al.* [1992]. This method has been applied previously in Europe by *Simpson et al.* [1995] and *Stohl et al.* [1996].

A more detailed method has been presented by *Yienger and Levy* [1995] for global soil NO_x emissions. In this approach, the variation in soil NO emissions is associated with biomass burning, rainfall events (pulsing), temperature, soil moisture, vegetation cover type (biome), canopy reductions, and fertilization

rate. As mentioned in section 3.4.1, this approach also included a canopy-reduction factor to allow for the uptake of NO_x within a vegetation canopy.

Both the above methods require quite detailed meteorological data. Alternative approaches include assigning annual flux estimates to different land use classes. For example, *Davidson and Kingerlee* [1997] assigned mean flux rates of 3.6 kg N ha⁻¹ yr⁻¹ to temperate cultivated grassland, 1.2 kg N ha⁻¹ yr⁻¹ to temperate grass/woodland, zero emission to boreal forest, 0.1 kg N ha⁻¹ yr⁻¹ to temperate forest, and 2.7 kg ha⁻¹ yr⁻¹ to “N-affected” temperate forest. The high flux rate for this latter biome is based upon measured fluxes at two German forests, so it is somewhat speculative to apply these rates to other countries. In any case, the emission rates obtained in this study are at the upper end of current emission estimates, possibly because extrapolating from short measuring periods to yearly fluxes overestimates the latter (most measurements are short term and usually at times when fluxes are expected to be high, after fertilizer applications, in summer and perhaps in the tropics at the onset of the wet season).

Skiba et al. [1997] and *Veldkamp and Keller* [1997] attempted to derive soil NO emissions as functions of fertilizer N inputs. Skiba et al. found that, as a geometric mean, 0.3% of applied fertilizer-N was released as NO. Veldkamp and Keller applied a regression analysis to a carefully selected set of long-term data studies, and found that on average 0.5% of applied fertilizer N was released. (We can note that Yienger and Levy assumed that 2.5% of applied-N was released as NO_x. As the *Skiba et al.* [1997] and *Veldkamp and Keller* [1997] figures are based upon a larger literature than Yienger and Levy, and as they include many European measurements, they are probably a better estimate for European inventories. However, the range illustrates well the uncertainties associated with this emission source.)

For the EMEP/CORINAIR Guidebook a methodology based on *Skiba et al.* [1997] was chosen. Two additional assumptions were made in order to account for nonagricultural areas, and also to allow for the effect of atmospheric N deposition (which is substantial in many parts of Europe). A background emission of 0.1 ng NO-N m⁻² s⁻¹ was assumed, and the fraction of applied N released as NO_x was applied to all N inputs: fertilizer, manure, animal excretions and atmospheric deposition. No canopy effect is considered (see section 3.4). This approach was adopted as only statistics of annual fertilizer consumption, and of atmospheric N deposition, are required, and these are usually quite well known. Further, despite the simplicity of this approach, emissions are explicitly related to

Table 6. Methane Consumption in Temperate Forest Soils, Measured in Situ

Site	Latitude/Longitude	Biotope	Methane Consumption		Reference
			Mean (s.d.)	Range	
Heidelberg, Germany	49°N, 9°E	mixed forest		0.09–1.3	<i>Born et al.</i> [1990]
Denmark		beech 25 years	0.049 (0.049)		<i>Ambus and Christensen</i> [1995]
Denmark		spruce 30 years	0.12 (0.10)		<i>Ambus and Christensen</i> [1995]
Strødam, Denmark	56°N, 12°E	Norway spruce 32 years	0.34		<i>Priem and Christensen</i> [1997]
Dunslair, Scotland		sitka spruce 12 years	0.13		<i>Macdonald et al.</i> [1997]
380 m a.s.l.					
Dunslair, Scotland		sitka spruce 12 years	0.07		<i>Macdonald et al.</i> [1997]
615 m a.s.l.					
Gårdsjön, Sweden	58°N, 12°E	mixed conifers, 100 years	0.14 (0.08)		<i>Klemetsson and Klemetsson</i> [1997]
Mean of above			0.14	0.07–0.34	

Consumption measured in $\text{g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$

one of the main drivers of soil- NO_x formation, the applied N amounts.

In section 6.2 we will present result from BEIS-2, Yienger and Levy, Davidson and Kingerlee, and this Skiba et al. approach in order to illustrate the range of estimates obtained.

4.4.2. Soil CH_4 consumption. In the scientific literature, different approaches to model methane oxidation in soils have been used. *Dörr et al.* [1993] used the FAO Soil Map of the World to make their estimate with soil texture classes as the main parameter. A model based on soil diffusivity was also used by *Potter et al.* [1996], although in this case seasonality was added together with an adjustment for nitrogen amendments. *Dobbie et al.* [1996] used field data from three countries and extrapolated these in order to estimate fluxes from Europe.

For our estimates, a simpler approach is adopted. Methane fluxes to forest and grassland soils are simply estimated by

$$W_{\text{CH}_4} = A_i \times F_i \quad (4)$$

where A_i is the area (ha) of forests or grassland and F_i is the seasonal average flux ($\text{kg CH}_4 \text{ ha}^{-1} \text{ yr}^{-1}$) for these ecosystems. For a national inventory, the model should ideally include a seasonality factor, based on local temperatures and precipitation, but as the substance of this factor is currently both qualitatively and quantitatively uncertain it has been left out at this stage.

The flux factor F_i for forest soils is derived from annual estimates of methane fluxes reported for a number of sites in Europe, Table 6. Evidently, there is a bias in Table 6 towards the northwest of Europe. Despite this, it should be noted that flux rates are fairly uniform (in comparison with fluxes of other gases from natural systems) across the different sites. From Table 6 we calculate a mean consumption rate in forest soils of

$0.14 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$; this is adopted for the calculations presented here.

Grasslands have been reported to consume methane at half the rates of corresponding forest soils in the same areas [*Kruse and Iversen*, 1995; *Boeckx et al.*, 1997]. Contradictory results were presented by *MacDonald et al.* [1997], who found that moorland consumed more CH_4 than did forest sites in Scotland, while *Kruse and Iversen* [1995] reported an almost zero methane uptake in Danish heathland. *Ambus and Christensen* [1996] reported low net emissions of CH_4 from coastal grasslands in Denmark ($22 \text{ mg CH}_4\text{-C m}^{-2} \text{ yr}^{-1}$), while abandoned farmland was consuming CH_4 at rates comparable to the spruce forest of their site (80 and $91 \text{ mg CH}_4\text{-C m}^{-2} \text{ yr}^{-1}$, respectively). *Van den Pol-van den Dasselaar et al.* [1997] reported a consumption of $0.08\text{--}0.31 \text{ kg CH}_4 \text{ ha}^{-1} \text{ yr}^{-1}$ in peat soil in the Netherlands. These rates were lower than other temperate grasslands, but similar to those of other drained soils reviewed by these authors.

Within a large range of uncertainty we may estimate the nonforest group (including pastures and meadows) at half of the consumption rates in forest soils, i.e., $0.07 \text{ g CH}_4 \text{ m}^{-2} \text{ yr}^{-1}$.

4.5. Wetlands

4.5.1. Methane. Emissions from wetlands (W_{CH_4} , in mass units) are estimated by

$$W_{\text{CH}_4} = \sum_i (A_i \times F_i \times S_i \times \text{c.f.}) \quad (5)$$

where $i = 1 \dots 6$ for the 6 wetland types; A_i is the area in each wetland type; F_i is the seasonal average flux (in mass/area/time units, usually $\text{mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$); S_i is the length of the season of methane emission. The season is the time the soil is thawed for boreal and northern temperate wetlands, and the length of time the soil is inundated for flood plains and seasonal marshes and swamps; "c.f." is the appropriate units conversion factor.

Emission factors (Table 7) are based on *Bartlett and Harriss* [1993], who did a thorough review of flux measurements from wetlands and shallow lakes for the purpose of making global estimates.

Measured fluxes from England at 55°N latitude [*Clymo and Reddaway*, 1971], and Sweden at 68°N latitude [*Svensson*, 1976; *Svensson and Rosswall*, 1984] are similar to the fluxes in Table 7. Measurements made in Scotland at 58°N latitude [*Gallagher et al.*, 1994] are lower but were not seasonally averaged and were made at the end of a 2-year drought. The climate zones of Table 7 apply best to the American continents, as most of

Table 7. Emissions Factors for Methane From Wetlands

Climate Zone ^a	Flux by Wetlands Type, $\text{mg m}^{-2} \text{ d}^{-1}$					Shallow Lakes ^b
	Bogs	Fens	Marsh	Swamp	Floodplain	
Arctic	96	96				
Boreal	87	87	87	87		35
Temperate	135	135	70	75	48	60
Tropical	199	199	233	165	182	148

^aThe default climate zones are arctic: $60^\circ\text{--}90^\circ$ latitude; boreal: $45^\circ\text{--}60^\circ$ latitude; temperate: $20^\circ\text{--}45^\circ$ latitude; tropical: $0^\circ\text{--}20^\circ$ latitude.

^bLess than 2 m deep.

the northern hemisphere studies are from Canada and the United States, and most southern hemisphere studies were done in Brazil. They are probably less appropriate to at least Scandinavia, where the Gulf stream ensures a warmer climate than at similar latitudes in America. Thus the default season lengths have been adjusted for the estimates presented in section 6. Since the seasonal methane flux is usually lowest at the beginning and end of the emission season, the Table 7 fluxes may overestimate regional emissions with longer seasons, but we do not have enough information to correct for this.

4.5.2. NMHC. The factors controlling emissions of isoprene from peatlands are still unclear. More importantly, we do not have an estimate of the coverage of wet Sphagnum moss within European forests on which to base a good emission estimate. However, in view of the large fluxes observed from wetlands we present a very tentative estimate of isoprene fluxes, in order to illustrate their possible magnitude. The methodology is identical to that followed for isoprene emissions from forests and other vegetation.

Janson and De Serves [1998] derived a mean temperature- and light-dependent emission potential (ϵ , see section 4.1) of $617 \pm 345 \mu\text{g C m}^{-2} \text{ h}^{-1}$ for the wet flark environment. For the area studied we can estimate that $\sim 50\%$ of the peatland surface emits isoprene. However, for other forests and countries this figure could vary very much. For the purposes of this calculation we use an emission potential $\epsilon = 600 \mu\text{g C m}^{-2} \text{ h}^{-1}$ and assume that between 10 and 50% of the area within a peatland emits isoprene.

4.6. Waters

The flux of a compound across the sea-air interface is generally estimated by

$$F = K_w \Delta C \quad (6)$$

where K_w is an exchange coefficient and ΔC is the concentration difference across the interface driving the flux.

For the case of DMS, the concentrations in seawater are at least 2 orders of magnitude higher than the concentration at equilibrium with atmospheric concentrations [Arends *et al.*, 1995; Bürgermeister *et al.*, 1990; Andreae *et al.*, 1985; Cline and Bates, 1983]. This implies that the sea surface is supersaturated with DMS, resulting in a net flux of DMS to the atmosphere. Consequently, the flux can be estimated as the product of the exchange coefficient and the observed concentrations in seawater.

As DMS has intermediate solubility in water, sea-to-air exchange is limited by the resistance in the liquid phase. Other relevant factors determining the sea-air exchange are sea roughness, seawater temperature and wind speed. Liss and Merlivat's [1986] parameterization distinguishes three different roughness regimes and establishes a nonlinear dependence of the sea-air exchange with the wind fields. The dependence with surface temperature is determined by the change of DMS molecular diffusivity as parameterized by Saltzman *et al.* [1993]. It should be noted here that exchange coefficients derived from the Liss and Merlivat formulation are generally about 30% smaller than those derived from other methods, in particular, exchange coefficients derived from the ^{222}Rn deficit technique.

Current estimates of the spatial distribution of DMS concentrations in seawater are based on area averages of observations [Bates *et al.*, 1992, 1987; Andreae, 1986]. This simple

approach is recommended until the testing of different ancillary data for extrapolating DMS production in seawater gives more conclusive results. More sophisticated extrapolations, derived from satellite data on chlorophyll content from the coastal zone color scanner (CZCS), have up to now shown limited success because DMS production is species dependent [Holligan *et al.*, 1987; Turner *et al.*, 1988; Keller *et al.*, 1989].

The measurements indicate a significant seasonal variation with maximum DMS concentrations in spring/summer that are up to 2 orders of magnitude larger than during the winter minimum [Davison and Hewitt, 1992; Leck and Rodhe, 1991; Turner *et al.*, 1989; Bates *et al.*, 1987]. By collating observed DMS concentrations over different biogeographical regions, a monthly cycle of DMS concentrations in seawater has been derived and recommended values and ranges are given in Table 13.

The North Sea extrapolation is based on observations by Turner *et al.* [1996]. For the Baltic, the Mediterranean and the Black Seas, DMS production in seawater is better represented by the extrapolation of "Coast and Shelf Regions." "Open Sea" values are recommended over the Mid-Atlantic Ocean. These values are based on a collation of observations compiled over world oceans. Still, the recommended averages and ranges for "coast and shelf regions" correspond fairly well with observations over the Baltic Sea ($82 \pm 50 \text{ ng S L}^{-1}$ in July, reported by Leck and Rodhe [1991] and the Mediterranean Sea ($93 \pm 50 \text{ ng S L}^{-1}$ reported by Simó *et al.* [1997]; 140 ng S L^{-1} in summer, reported by Belviso *et al.* [1993]; and 518 ng S L^{-1} in spring reported by Boniforti *et al.* [1993]).

For ammonia, the calculation is more complicated as NH_3 may be either deposited to or emitted from seawater [Asman *et al.*, 1994]. This bidirectional exchange is similar in concept to the so-called compensation point exhibited by some plant surfaces [Farquhar *et al.*, 1980], whereby if atmospheric concentrations fall below the compensation point, the surface may emit. The concentration gradient driving the flux is the difference between the NH_3 concentration in air which would be in equilibrium with the NH_x concentration in seawater, C_q , and the atmospheric concentration of NH_3 . C_q is calculated using the methodology of Asman *et al.* [1994].

The derived concentrations are then included in (6), using $\Delta C = C_{\text{air}} - C_{eq}$ to calculate the fluxes. The exchange velocities in this case are the deposition velocities of ammonia, calculated using the resistance approach [Asman *et al.*, 1994].

For the concentrations of NH_3 in seawater, Quinn *et al.* [1996] empirically derive an extrapolation of observed data according to different biogeographical sea areas, including monthly variations. The C_q thus calculated have been introduced by Barrett [1998] in the EMEP acid deposition model in order to evaluate fluxes from European seas, taking into account the atmospheric concentrations of NH_3 .

4.7. Wild Animals and Humans

Emissions were obtained by multiplying the number of animals with weight-dependent emission factors. These emission factors (Table 8) for wild animals are necessarily derived from factors obtained for domestic animals.

However, even taking into account the differences in animal weights, wild, and domestic animals have too many differences for these estimates to be regarded as reliable. Domestic animals are generally kept more densely, such that manure management is needed and the manure has to be stored for a longer period of time. Chemical processes in the manure (decay of urea to ammonia) are different and may be much less

Table 8. Emission Factors for Emissions From Wild Animals and Humans

	Assumed Life-Weight, kg	Emission Factor		References
		CH ₄	NH ₃	
Moose	350	50	2.2	<i>Bouwman et al.</i> [1997]
Deer ^a	100	25	1.1	derived from <i>van der Hoek</i> [1996] and <i>Sutton et al.</i> [1995]
Roe deer	15	4	0.2	derived from red deer
Boar		1.5	1	<i>Crutzen et al.</i> [1986], <i>van der Hoek</i> [1996], and <i>Sutton et al.</i> [1995]
Birds	0.8	...	0.12	<i>Sutton et al.</i> [1995]
Large birds	2.4	...	0.36	<i>Sutton et al.</i> [1995]
Humans		0.1	0.05	<i>Crutzen et al.</i> [1986] and <i>Sutton et al.</i> [1995]

In kilograms per animal/person per year.

^aRed deer, reindeer.

relevant for natural animals. Further, the metabolism (aimed at milk/meat production for domestic animals) and the diet are known to be quite different. Nevertheless, in the absence of better data, emission factors derived from domestic animals form the best basis for our estimates.

For methane, data presented for enteric fermentation of domestic animals were used [*van der Hoek*, 1996]. These emission factors are about 50% larger than those suggested previously [*Crutzen et al.*, 1986]. However, as methane emissions from manure (an additional 10–50% according to *van der Hoek*, 1996) are not included in any of the data given, the emission factors proposed here still should not be considered upper limits.

As weights for different game species vary considerably, we further scaled the emissions linearly by the animal weight from the red deer/reindeer emission factors. A more complex scaling, proportional to the 3/4 power of weight, has been suggested [*Crutzen et al.*, 1986], which may describe the food intake more closely, but other parameters also contribute to methane emissions so such refinements are not adopted here. The average weights of mammal species have been simplified from much more detailed literature data [*Niethammer and Krapp*, 1986]. We use red deer and reindeer 100 kg, fallow deer and white-tailed deer 90 kg, roe deer 15 kg, chamois 35 kg, ibex 70 kg, and mufion 25 kg. Moose emissions were assumed to be twice those of reindeer, consistent with estimates of nitrogen excretion given by *Bouwman et al.* [1997]. Methane emissions from humans, mainly in human breath, have been assessed from measured values. The resulting emission factor of 0.07 kg/person, and year is notably lower than that of pigs (1.5 kg/animal), which may have a comparable metabolism. Considering the food uptake of humans, which is about one third of that of pigs [*Jonas*, 1997], an emission factor of 0.5 kg/person would be expected. Much of this discrepancy may be due to the diet but no full explanation is possible at this time. We thus applied an emission factor of 0.1 kg/person-year.

Linear scaling of the given emission factors by weight for even smaller animals (rodents) further increases the uncertainty and probably underestimates the metabolic activity of small animals. However, *Crutzen et al.* [1986] suggest that the methane yield (fraction of food energy content that is emitted as methane) is much higher for ruminants (7–9%) than for other herbivores (1–3%). Thus linear scaling by mass may still tend to overestimate methane emission factors rather than to underestimate them, justifying this rather simple approach.

The choice of emission factors for ammonia has been discussed in detail by *Sutton et al.* [1995]. For scaling of emissions, we employed identical methods as for methane. Compared to

other literature, *Sutton et al.* suggest a very low emission factor for humans, but only these authors exclude emission from pets and sewage processing from the “human” category. This approach agrees with that of the UN-ECE/CORINAIR methodology taken here. Comparable NH₃ emission rates have been reported independently for red deer (0.9 kg NH₃/individual per year [*Sutton et al.*, 1995]) and for reindeer (1 kg N per individual per year [*Bouwman et al.*, 1997]). The weights of birds have been given by *Bauer and Glutz von Blotzheim* [1987]. As with small mammals, linear scaling using their weights is quite problematic. The weights differ up to two orders of magnitudes from those for which the emission factors have been derived, but no further data seem to be available. For birds, only ammonia emission were considered.

4.8. Lightning

A large variety of emission factors are given in the literature from laboratory as well as field investigations. According to *Biazar and McNider* [1995], the low, median, and high end of these estimates may be given at 0.36×10^{26} , 4×10^{26} , and 30×10^{26} molecules NO per flash. As indicated by *Biazar and McNider* [1995] and also discussed by *Novak and Pierce* [1993], the highest of these figures [from *Franzblau and Popp*, 1989] is not supported by studies modeling nitrate deposition. *Levy et al.* [1996] estimates global NO_x production from a best fit approximation between a global model and observations from regions where lightning is expected to be a major source. This study yields results close to the lowest of the three factors given, thus we have adopted a cloud-to-ground emission factor of 0.36×10^{26} molecules NO (0.84 kg N) for each flash of lightning. In view of the uncertainties surrounding the contribution of the IC flashes only cloud-to-ground flashes are considered here.

For the purpose of the joint EMEP/CORINAIR inventory only emissions within the mixing layer, defined for this purpose as 1000 m, are required. Hence we assume that lightning flashes take place over a depth of typically 7 km and that 20% of this amount (i.e., 0.17 kg N/flash) is assumed to be emitted below 1 km altitude.

4.9. Volcanoes

The primary source of geothermal emissions are active volcanoes. These volcanoes are well known and geologically described. Methods do exist to derive emission estimates from geological information on volcano type, explosivity of eruption, etc., but for the important European volcanoes specific emissions are known from measurements [*Andres and Kasgnoc*, 1998]. Emission measurements employed primarily correlation spectrometric (COSPEC) data [*Stoiber et al.*, 1983], but also

data from total ozone mapping spectrometer (TOMS) aboard NASA satellite Nimbus 7, which allows evaluation of SO₂ emissions [Bluth *et al.*, 1993]. Measured SO₂ data may to some extent contain H₂S emissions already converted to SO₂ but do not include SO₂ already converted to sulfate at the point of measurement.

4.10. Gas Seeps

With the sizes of seeping areas taken from literature information, emissions are calculated for each of these areas by simple multiplication with the flux per m². In addition, an assumption needs to be made on the amount of methane lost during bubbling to the surface—for the North Sea, we assumed that due to ~100 m of water depth only about one half of the released methane actually reaches the surface.

Emission factors are highly uncertain due to the irregularity of the emissions taking place. Short-term emission rates of 10 L h⁻¹ from one single seep hole have been reported near the California South Coast [Radian Company, 1996], and an area-based value of 1 L h⁻¹ m⁻² found in the Danish coastal waters [Fenger *et al.*, 1990]. These values need to be considered local and temporal maxima, occurring over short periods (low tide) at very definite sites. Measurements have shown the diurnal average to be at least a factor of 20 lower than the peak emission rate [Hovland *et al.*, 1993; Martens and Klump, 1980]. Hovland *et al.* [1993] have compiled information on all known source areas, and take into account also the seasonal and the spatial variability. The highest overall seepage rate is given for the California South Coast (Santa Barbara Channel) at 400 g yr⁻¹ m⁻². As a typical emission factor, we used the numbers given for the Gulf of Mexico, the North Carolina coast and the Danish Kattegat and Skagerrak: 50 g yr⁻¹ m⁻². These factors refer to active areas only.

5. Data Used

5.1. Forests (Foliar Emissions)

For calculating emission rates the vegetation cover (km²) and foliar biomass densities (g m⁻²) are needed, in as much detail as possible for individual forest species.

For this study, a database of national forest species distributions has been built up from either published sources or direct contact with national authorities or forestry institutes. The resulting database varies significantly from country to country in terms of complexity, but it should represent a great improvement in detail over sources used previously, for example, by Veldt [1989] and Simpson *et al.* [1995]. The largest problems occur for Mediterranean countries where large areas are occupied by degraded and often scattered trees, so the definitions of “forest” and area covered are obscure. The tree species distributions are given in Table 9.

Typically, there are large differences in the species distributions from one part of Europe to another. Scandinavia is dominated by Norway spruce and Scots pine, with little deciduous cover except for birch. Oaks in most of northern Europe are essentially all deciduous (*Q. robur*, *Q. petraea*). In the Mediterranean, many more species are present, and both deciduous and evergreen oaks are important.

The gridding of these national species distributions to the EMEP grid was done utilizing the land use map of the Stockholm Environment Institute (SEI) for most of the European countries, or with the map of RIVM [van de Velde *et al.*, 1994] for Russia, Ukraine, Belarus, Turkey, and the Baltic states.

The SEI map is the most detailed, consisting of over 50 land use classes and distinguishing between forest species such as Holm oak, Cork oak, Scots pine, as well as wet and dry soil grasslands, alpine meadow, Mediterranean scrub, etc. This database is also hierarchical, so that separate classifications are given for species (e.g., Scots pine) within mainly coniferous forests, mixed forests, or mainly deciduous forests). Some of the classes are mixed, for example “dwarf conifers.” Data sources consist of the ~1 km resolution remote sensing forest map by ESA, plus a printed land use map from the 1970s, and statistics of dominant crops (NUTS-3 level for EC countries, country level from FAO statistics for the rest of Europe). An illustration of the output of this database is given in Plate 1.

This high-resolution database was used to calculate the percentage of each land-cover type within the EMEP 150 × 150 km² grid for which meteorological data were available.

The RIVM database consists of two forest land use classes, “coniferous + mixed forest” and “deciduous” forest, as well as grasslands, arable lands, permanent crops, urban, water, and “other” and was available on the 50 × 50 km² EMEP grid.

In order to generate the database actually used for the emission calculations, the national species distributions given in Tables 9 and 10 were mapped onto the EMEP grid using the SEI or RIVM databases as keys for each species. This process was somewhat subjective but ensures for example that most *Quercus ilex* is assigned to the SEI-determined *Q. ilex* areas, but also to a lesser extent to other deciduous forests, other mixed forests, and coniferous forests in descending order of priority.

5.2. Grassland and Other Low Vegetation

The areas of meadows and pastures have been given in Table 1. Additional data were obtained from some countries on other types of vegetation coverage as summarized in Table 10. Although such data were only found for seven countries, these fortunately include several Mediterranean countries where the emissions from *maquis* is of interest, and data for the United Kingdom where emission measurements of Gorse have recently become available [Cao *et al.*, 1997].

5.3. Forest Fires

The areas of forest burnt in most European countries are reported by EUROSTAT [1995], and summarized in Table 11. The ecosystem-dependent biomass and burning parameters *B*, *α*, and *β* should ideally be estimated from local data, but for this work the values given in Table 5 provide a default.

5.4. Soil NO_x

Estimates using the Skiba methodology (see section 4.4.1) require total land area, which is given in Table 1, and the various nitrogen inputs. Fertilizer application was taken from EUROSTAT [1995]. Nitrogen deposition in each country was taken from EMEP model calculations as presented by Tsyro [1997]. N excretions of wild animals have been derived from the data used in section 5.7.

5.5. Wetlands

Estimates of wetland area for global estimates have been typically derived from large-scale data such as Aselmann and Crutzen [1989], who give percent wetland area in 2.5° latitude by 5° longitude cells. Areas derived from this database are compared with nationally derived data for total peatland, mainly from Maltby and Immirizi [1992] in Table 12.

In many cases there is quite good agreement between these

data sets, allowing for unavoidable interpolation errors. Some differences may be due to definition—for example, much of the Austrian peatland is associated with the large and shallow (mean depth 1 m) Lake Neusiedl, which may be classified in other statistics as shallow water rather than bog or fen. The biggest discrepancy occurs for Sweden, where the national estimate of *Hånell* [1989] may be assumed to be the most reliable.

No comprehensive data were found on the coverage of shallow lakes (<2 m), although these may be substantial in many countries. Some data on the largest lakes are given by *EUROSTAT* [1995]. More problematic are the large number of very small lakes which are more likely to be shallow. *Stanners and Bourdeau* [1995] estimate that Europe has 150 lakes of larger than 100 km², 2000 larger than 10 km², but 400,000–600,000 between 0.01 and 0.1 km² in area.

5.6. Waters

Given the sea-specific emission factors derived and presented in section 4.6 and Table 13, the only further statistics required are the areas of the European seas.

5.7. Wild Animals and Humans

The number of deer by species has been compiled by *Myrberget* [1990] for the European countries with the exception of the former Soviet Union. Numbers are based on hunting statistics and refer to the winter population. Assuming a typical population increase of 20–30% during four months in summer before the hunting season, the annual average is ~8% higher than the winter population. Deer counts for the former Soviet Union were estimated by comparing forest statistics from Finland and the European part of the Soviet Union. The number of large mammals in Europe is presented in Table 14. The number of humans was taken from population statistics.

For the population of moose and reindeer, an independent estimate is available [*Bouwman et al.*, 1997]. These numbers agree well for moose. For reindeer, Bouwman et al. included reindeer kept as semiwild herds, so their population numbers are much higher than ours. *Crutzen et al.* [1986] presented numbers for wild ruminants in the temperate northern region in a global emission assessment, but these are lower than even the European estimate given here for roe deer, and not much higher for moose. We consider our data, being compiled from reasonable national data, to be the most reliable.

No consistent information was available for rodents. Especially for Northern European countries, very high population numbers have been occasionally reported, but with extreme population fluctuations. The most abundant mammals at the peak of their population are lemmings, which peak every 4 years [*Chitty*, 1996]. At a weight of 40 g during summertime (the wintertime weight near 80 g is probably not relevant for the high numbers or the metabolic rate [*Nagy and Gower*, 1999]), the total biomass of lemmings in Sweden would be a little more than half of that of all moose, if reported numbers of 2×10^9 are correct. This will only be the case for peak years though, and not regular years.

Population numbers of hares as available from hunting statistics suggest that their number is consistently smaller than that of deer (Austrian Statistical Office, Statistical Office of the Czech Republic), and thus their total biomass is considerably lower (at 5 kg weight). For at least one country however (France) the number of rabbits reported is a factor of 20 larger than any of the larger deer species (M. Ritter, personal infor-

mation from the CORINAIR database, 1998). Thus, although the number of small mammals currently is highly speculative, it seems that at least their total biomass does not significantly exceed that of wild ruminants. Emissions from rodents therefore have not been specifically estimated for the current undertaking.

Numbers of European breeding birds have been systematically collected and published recently [*Hagemeijer and Blair*, 1997]. For each European native species, the number of breeding pairs is given on a per country basis; however, numbers are only given for up to ten countries per species, for those countries in which the species is most abundant in absolute numbers. Therefore useful numbers are only available for those European countries that have large bird abundancies and for the European total. It needs to be stressed that these numbers are very uncertain, especially for European Russia where for the most abundant species a range of a factor of ten is given (with the geometric average presented as best guess). For our study, no annual population cycle is considered, only the breeding pairs are included, and it is assumed that their emission rate is constant year round.

5.8. Lightning

Ground-based estimation of lightning flash densities for different European countries have been compiled by *Turman and Edgar* [1982]. These estimates, ranging from 2 to 10 cloud-to-ground flashes km⁻² yr⁻¹ for different European countries, seem difficult to interpret, however, as regions of similar climatic and orographic conditions exhibited larger differences than countries under totally different conditions.

While no European lightning climatology is available, data on lightning detection networks (for Central Europe: ALDIS-network: <http://www.aldis.at>) but also from satellite observation (NASA optical transient detector: <http://www.ghcc.msfc.nasa.gov/otd.html>) and from convective events simulated in a global circulation model [*Levy et al.*, 1996] indicate a typical density of 2 flashes km⁻² yr⁻¹, with ~4 in southern Europe, decreasing to 1 in northern Europe. Because of the lack of detailed data, for the emission estimation a mean density of 2 flashes per km² yr⁻¹ was assumed.

5.9. Volcanoes

The larger European volcanoes are under complete surveillance, and continuous emissions are available specifically for each of the volcanoes Etna, Stromboli, and Volcano, in Italy, and Kverkfjöll in Iceland. These volcanoes are considered responsible for nearly all of the continuous emissions. However, large atmospheric injections also occur from sporadic eruptions, some of which are not observed with COSPEC, but derived from TOMS data: Hekla (Iceland) with one eruption totalling 250 Gg [*Andres and Kasgnoc*, 1998]. Because of the irregular pattern of such emissions, this was not included in the emission totals.

5.10. Gas Seeps

At the Danish coasts, in Skagerrak and Kattegat, ~15,000 km² of the sea shelf is potentially active seeping area [*Hovland et al.*, 1993]. *Fenger et al.* [1990] estimate the active area to be “at least 20–40 km².” As a conservative estimate, we have used 40 km² for our emission estimates, but it should be noted that this source may be underestimated by orders of magnitude. Further seepage areas have been observed in the North Sea (potentially the area is as much as 100,000 km², according to

Table 9. Percentage Distribution of Forest Species Used in the Calculations

Species	AL	AT	BY	BE	BA	BG	HR	CS	DK	EE	FI	FR	GDR	FRG	GR	HU	IE	IT	LU
Forest cover, km ²	14420	31697	39000	6960	18268	24230	9666	26420	4115	2016	197171	133383	24680	73880	19480	18000	3940	61997	863
Abies	...	7	8	4	...	2	17	0	...
Acer	2	0
Alnus	4	0	1
Betula	30	7	1
Carpinus	1
Castanea	4	1	4	...
Fagus	22	10	...	11	25	17	25	...	17	10	7	17	11	7	3	4	31
Fraxinus	2	3
Larix	...	5	1	2	...
Olea	0
Picea unspecified	1	63	16	28	10	6	10	24	45	2	31
Picea abies	54	33	...	26	5	21	39
Picea sitchensis	9	0
Pinus unspecified	14	3	79	44	4	36	4	...	7	38	20	7	...	23	6	3
Pinus brutia
Pinus halepensis	2	18
Pinus nigra	2	7
Pinus pinaster	10
Pinus sylvestris	66	8	51	8
Populus	...	2	...	4	0	0	...	0	2	...
Populus tremula	2	0	0
Pseudotsuga	2
Quercus unspecified	27	2	5	...	25	20	25	...	7	0	4	8	...	35	...	2	10
Quercus cerris	0	1	...
Quercus robur	17	0	...
Quercus ilex	2
Quercus petraea	14
Quercus pubescens	7
Quercus suber	0	2	...
Quercus decid	7	38	...	3
Robinia	1
Salix	...	2	0
Other conifers	7	1	0	5	8	5	8	25	9	...	0	0	3	5	1	7	23	13	...
Other broadleaf	29	5	1	2	29	15	29	21	6	2	0	0	13	8	...	43	5	4	2
Other trees	0	58	21

Table 9. (continued)

Species	EE	LT	LI	NL	NO	PL	PO	RO	RU	SK	SI	ES	SE	CH	FYM	UA	UK	YU
Forest cover, km ²	2016	27781	19	2125	86761	79425	35265	49960	1108676	19186	4799	132185	279462	10845	8312	92391	19219	31721
Abies	0	...	3	5	...	0	...	14	...	3
Acer	1	2
Alnus	4	8	11	0	3	1
Betula	30	29	21	3	22	6	0	13	5	...
Carpinus	0	...	0	6	2
Castanea	1	1
Citrus	1	2
Eucalyptus	11	2
Fagus	3	32	...	30	25	2	1	16	25	13	5	25
Fraxinus	2	2	5	...
Larix	8	0	2	5	9	...
Olea	10	16
Picea unspecified	24	...	26	...	40	24	45	...	10	10	10
Picea abies	...	21	...	6	...	7	28	39	48	...	16	7	...
Picea sitchensis	1	0	40	...	4	4	...	32	...
Pinus	38	...	37	0	27	4	4
Pinus cembra
Pinus contorta	0	9	8	...
Pinus halepensis	4
Pinus laricio
Pinus nigra	8	2	...
Pinus pinaster	1	35	10
Pinus sylvestris	...	40	...	46	...	76	7	...	6	41	4	...	35	15	...
Populus	7	2	0	2	...	0
Populus tremula	2	3	5	7	2
Pseudotsuga	7
Quercus unspecified	2	20	3	14	25	...	1	...	25	22	...	25
Quercus robur	1	...	2
Quercus Faginea	4
Quercus Lusitanica	2
Quercus Ilex	13	12
Quercus suber	19	1
Quercus decid	4	1	7	10	...
Salix	0	...	0	2	...	1
Other conifers	0	3	8	8	8
Other broadleaf	2	0	4	...	3	16	4	5	29	16	3	3	29	9	...	29

For country codes, see Table 1, except FRG, Former Federal Republic Germany; GDR, former German Democratic Republic. Zeros mean <0.5%. See Appendix A for sources of distributions.

Table 10. Documented Area Coverage of Low Vegetation Categories

Biome	Austria	France ^a	Greece	Italy	Portugal	Spain	United Kingdom
Monte hueco						14,790	
Mediterranean low vegetation						48,634	
Mediterranean shrub and herb					16,914		
Temperate shrub/bush	587						
Maquis		4089	4780	2258			
Arbutus undedo		3					
Heathland		28,029					30,000
Grassland	9811					14,620	

Area of pastures given in Table 1. For sources, see Appendix A. Data not available for most countries. Coverage in km².

^aMaquis in France includes wooded garrique also.

Hovland *et al.* [1993]). A similar ratio between potentially active and active area was assumed for the North Sea as for the Danish coast, suggesting 300 km² of seeping area. Again, this number has to be considered speculative. Other active areas in Europe may exist, for example, in the Stockholm archipelago and in the Black Sea, and off the shores of Bulgaria and Greece, but no quantification has been given.

6. Results

In contrast to the previous chapters, the results are organized by compound rather than by source group, in order to allow a relative assessment of the importance of the different sources with respect to each other and with respect to man-made emissions.

6.1. Methane

Methane fluxes by country estimated using the above methods are presented in Table 15. Also shown are previous national estimates of emissions from wetlands and waters, and from anthropogenic sources.

Considering the estimates of this study first, then wetland emissions clearly dominate the biogenic emission categories, especially on the European scale. For the Nordic countries (except Denmark), and also Russia, wetland emissions exceed those from anthropogenic sources, or are comparable. Of the other natural sectors considered here, the forest CH₄ oxidation sink is clearly important to the methane budget over Europe. Forest fires and animal emissions do not contribute much to the national budgets.

Gas seeps are estimated to contribute emissions of 2 Gg yr⁻¹ for the Danish coast, and 7.5 Gg yr⁻¹ for the North Sea, based on our best estimate of the active seepage area. As noted in section 5.10, the area of potential emissions is much larger than this best estimate area, and would give a speculative upper estimate of 750 Gg yr⁻¹ for the Danish coast and 2500 Gg yr⁻¹ for the North Sea. While in our best guess estimate, the emissions are rather small, the higher estimates would be a significant percentage of Europe's estimated CH₄ emissions.

Some of the differences in wetland emissions between our estimate and previously reported (CORINAIR-94) estimates as shown in Table 16 are disturbing. In most cases the methodology underlying the national estimates are unknown, but for Denmark the procedures are reported by Fenger *et al.* [1990]. The emission factors used by Fenger *et al.* are a factor of 10 higher than ours, and in addition a very large area (120,000 ha) of "wet meadows" is included in the Fenger *et al.* calculation, but which is not in our statistics. Some of these discrepancies may reflect emissions from highly productive ag-

ricultural wetlands, but clearly the uncertainty is very large in this case. For Austria the discrepancy between CORINAIR and our estimates seems to be due to different definitions of wetlands and waters. In our study, emissions from peatland are reported under the wetlands category. As noted in section 5.5, much of the Austrian peatland is associated with the large and shallow Lake Neusiedl, so it is likely that these emissions are reported under waters rather than wetlands in the CORINAIR reporting.

Emissions from large mammals by species are given in Table 16. Emissions from agriculture, which are dominated by enteric fermentation by farm animals, are also shown. It is clear that in comparison to these agricultural emissions, wild animals and humans represent a negligible source of methane.

Table 11. Areas of Forest Burnt per Year, Average and Range From 1985 to 1992

Country	Biome	N _y	Area Burnt, ha yr ⁻¹		
			Minimum	Maximum	Mean
Albania	m	7	90	417	240
Austria	t	7	52	200	90
Belarus	t	5	319	23,822	5412
Belgium	t	6	20	687	158
Bulgaria	t	8	223	5243	1226
Denmark	t	4	6	277	139
Estonia	b	3	58	1746	666
Finland	b	8	153	1082	413
France	m	8	6565	57,368	33,314
Germany	t	7	708	947	802
Greece	m	8	24,514	110,501	60,311
Hungary	t	2	770	1810	1290
Ireland	t	2	356	713	535
Italy	m	8	49,434	189,898	120,774
Luxembourg	t	6	2	9	4
Netherlands	t	8	34	414	177
Norway	b	8	87	1970	621
Poland	t	8	1454	33,822	6951
Portugal	m	8	22,435	182,486	105,563
Romania	t	5	15	709	312
Russian Federation	b	1	200,000
Spain	m	8	100,487	486,327	249,197
Sweden	b	1	5808	5808	5808
Switzerland	t	7	63	1102	319
Ukraine	t	3	1781	4252	2822
United Kingdom	t	7	61	568	255
Yugoslavia, ex.	m	6	6394	42,791	18,756
Croatia	m	6	170	976	463

From Eurostat [1995], except Russia from Dixon and Krankina [1993]; N_y gives the number of years for which data were available. Biome type assumed for calculations denoted: b, boreal; m, Mediterranean; t, temperate.

Table 12. Estimates of Wetland Area and Period of Emissions

Country	Area Estimate, 10 ⁶ ha					Season, days
	<i>Maltby and Immirizi</i> [1992] ^a	<i>Aselmann and Crutzen</i> [1989] ^b				
	Undrained Peatlands	Bogs and Fens	Swamp	Marsh	Floodplains	
Finland	5.4	5.48				150 ^c
Norway	2.75	3.73				150 ^c
Sweden	5.84	1.87				150 ^c
Denmark	0.06	0.07				150
Latvia	0.267					150 ^c
Iceland	0.90	0.99				100
United Kingdom	0.89	1.34				150
Ireland	0.58	1.13				150
Netherlands	0.009					150
Belgium	0.0025					150
France	0.065					150
Germany	1.29					150
Russia ^d	29.3	29.3	0.24	0.94		111
Ukraine		1.30	1.67	1.30		150
Poland	0.27	0.40	0.31	0.31		150
Austria	0.02					150
Switzerland	0.0125					150
Czechoslovakia	0.0054					150
Spain	0.0045					150
Italy	0.03					150
Greece	0.001					150
Other ^e		1.01	0.23	0.03	0.07	150
Sum	47.7	46.6	2.5	2.6	0.07	

^aExcept Sweden, from *Hånell* [1989] and Latvia, from CORINAIR-90 database (M. Ritter, UBA-Vienna, personal communication, 1997).

^bInterpolated to national areas.

^cDefault seasons for these latitudes are 100 days, but NW Europe's mild climate gives much longer seasons, for example, from 144 days in northern Sweden to 170 days in southern Sweden; 150 days used as representative.

^dWest of 60°E longitude (approximately Ural Mountains).

^eOther fens and bogs are principally in Estonia, Latvia and Lithuania; no country estimates are possible from the Aselmann and Crutzen data set. Floodplain is between Romania, Bulgaria, and Yugoslavia. Marshes and swamps are mostly from central East Europe.

6.2. Nitrogen Oxides

Table 17 gives the emissions of NO from soils, lightning, forest fires, and man-made sources. The soil NO_x calculations are presented for a number of the methodologies discussed in

Table 13. Recommended Monthly Variation of DMS Concentrations in European Seawater

	North Sea		Coast and Shelf Regions		Open Sea	
January	10.	(5–15)	10.	(7–15)	7.	(3–12)
February	20.	(15–25)	20.	(10–30)	15.	(7–15)
March	50.	(40–60)	40.	(30–60)	48.	(10–65)
April	230.	(170–290)	220.	(100–870)	85.	(15–135)
May	350.	(280–420)	250.	(80–950)	110.	(25–280)
June	240.	(190–290)	190.	(90–380)	120.	(35–250)
July	190.	(140–240)	100.	(80–170)	95.	(40–290)
August	130.	(100–160)	80.	(70–180)	75.	(40–160)
September	50.	(40–60)	60.	(50–150)	70.	(40–140)
October	35.	(30–40)	50.	(40–100)	60.	(35–85)
November	10.	(5–15)	20.	(15–45)	35.	(30–50)
December	5.	(2–8)	10.	(5–15)	10.	(3–15)

Adapted from *Tarrasón et al.* [1995]. Averages are given, with uncertainty ranges from the extrapolation of observed data in parentheses. Units are in ng S L⁻¹.

section 4.4.1. In implementing the Davidson and Kingerlee methodology for temperate forest emissions, we have applied both the default emission rate (0.1 Kg N ha⁻¹ yr⁻¹) and the N-affected emission rate (2.7 kg ha⁻¹ yr⁻¹), giving these numbers as a range. (Our definition of temperate versus boreal forest for these calculations is taken from Table 11.) Which-ever methodology is used, soil NO_x emissions dominate the biogenic NO_x budget, but the estimates of soil NO_x vary by more than a factor of 10.

The extent to which these soil NO emissions are natural is debatable. However, taking the background soil NO emissions (although this is also calculated for agricultural land) from the Skiba-methodology, and total (0–7 km) lightning, Table 17 suggests that natural (nonfertilizer or N-deposition-induced) emissions amount to <1% of the European budget. Taking the mean of the Davidson and Kingerlee estimates suggests that biogenic sources may contribute up to 20% of the European budget.

6.3. NMVOC

Table 18 gives the emissions of NMVOC from forests, other vegetation, wetlands, and fires.

The foliar emissions are clearly the dominant source of VOC from nature in Europe and indeed are comparable to man-made emissions. During summertime it is clear that the VOC

Table 14. Number of Wild Animals in Europe (Winter Population)

Counts	Red Deer	Roe Deer	Fallow Deer	Sika Deer	White-Tailed Deer	Moose	Reindeer	Chamois	Ibex	Mufflon	Boar
Austria	105,000	1,000,000	370	750				112,000	1900	5060	5200
Belgium	5000	22,000	150							500	9000
Bulgaria	18,500	142,000	2800					1600		2000	32,000
Czechoslovakia	50,000	289,000	12,700	2000	430	20		1800		7700	40,000
Denmark	5500	150,000	10,000	1000							
Finland		5000	300		35,000	91,800	600				
France	45,000	318,000	600	50				53,500		4900	7000
GDR ^b	55,000	350,000	30,000							7000	65,000
FRG ^c	85,000	1,700,000	30,000	1500				10,000		7000	40,000
Great Britain	300,000	60,000	50,000	4000							
Greece	300		200								
Hungary	47,500	219,600	9800	300						7100	31,000
Iceland							3500				
Ireland	6000		10,000	5000							
Italy	8900	103,000	6200					62,560	3500		
Liechtenstein	400	600						700			
Luxembourg	500	13,000								1150	1200
Netherlands	1200	25,000	500							300	
Norway	55,000	55,000	40			85,000	40,000				
Poland	73,000	482,000	4000	150		4600					56,000
Portugal	400		200								
Romania	45,000	287,000	10,000			20					40,000
Russia (Estimated) ^a	...	35,000	2100	...	245,000	642,600	4200
Spain	100,000										28,000
Sweden	2000	250,000	4500			311,000					
Switzerland	22,200	111,000		70				67,000	11,600		640
Yugoslavia ^d	30,000	310,000	2900		250			24,000	360	5300	56,000
Sum	1,061,400	5,927,200	187,360	14,820	280,680	1,135,040	48,300	333,160	17,360	48,010	411,040

From Myrberget [1990].

^aNumbers for Russia (actually former Soviet Union, European part) estimated from forest area, assuming same animal density as Finland.

^bFormer East Germany.

^cFormer West Germany.

^dFormer Yugoslavia.

budget over Europe will be dominated by BVOC, although country-to-country differences are large. National emission estimates are in general similar to those calculated previously by Simpson *et al.* [1995] for isoprene, OVOC, and by Guenther *et al.* [1995] for terpenes. For example, Simpson *et al.* [1995] derived an emission of 110–140 Gg yr⁻¹ isoprene from Spain, whereas Table 18 gives 177 Gg yr⁻¹. Larger differences are found in Greece, 19–28 Gg yr⁻¹ previously and now 103 Gg yr⁻¹. The difference arises because Simpson *et al.* [1995] applied one emission rate to all Mediterranean oaks, on the assumption that only 10% of these oaks were isoprene emitters. This assumption derived from detailed land use data for Spain (speciated data for other countries were not then known), but the new data used in this work (Table 9) suggests that almost all oak forest in Greece is deciduous and hence treated as isoprene emitting.

Table 19 gives the contributions of different vegetation species to the total NMVOC emissions. It is clear that only a few vegetation species dominate the emissions, especially realizing that many of the genera which are “unspecified” will probably be dominated by one species in most countries, for example, most unspecified oak in northern Europe will be *Q. robur* or *Q. petraea*. These rankings vary greatly between countries of course. In Spain, for example, the three largest isoprene emitters are Monte hueco, *Q. faginea* and *Q. lusitanica*, none of which have well-defined emission potentials. The terpene emission estimate for Spain is dominated by one species, *Q. ilex*.

The estimates of emissions from crops were based on a crude default parameterization as this vegetation lies outside

the scope of this natural emissions study. However, the relatively large emission from crops seen in Table 19 suggests more work is needed on this category in future studies.

Table 19 also suggests that emissions from the low-vegetation categories (e.g., heathland, Mediterranean scrub, etc.) are not significant. However, this is misleading as the land use data needed for these estimates were only available for a few countries. The United Kingdom was one country for which good estimates of low-vegetation cover were available, and the estimated VOC emissions for the UK are shown in Table 20. Heathland, pastures, and crops all make significant contributions to the emission totals, in addition to the forest contribution (*Picea sitchensis* and deciduous oaks). These results are a cause for concern because (1) the emission potentials for these species or ecosystems are based on very few specific measurements, and (2) the land cover and biomass data for such ecosystems are often not reported in national statistics.

Despite the very tentative nature of the calculations concerning isoprene emissions from wetlands Table 18 suggests that these are unlikely to contribute significantly to total budgets. The main reasons are their small area in relation to forests, moderate emission rate (comparable to Norway spruce for isoprene, for example), and often their location in relatively cool climates. However, on a local level, such emissions should not be ignored.

6.4. NH₃

Table 21 gives the results for NH₃. Emissions from natural sources amount to little more than 1% of the anthropogenic

Table 15. Estimated Methane Fluxes, From Natural/Biogenic Sources and Man-Made Sources

	Soils		Fires	Wetlands		Waters C94	Wild Animals	Humans	Gas Seeps	Anthropogenic ^a		References
	Forests	Grass/Other		This Study ^b	C94					Agriculture	Other	
Albania	-1.5	-0.3	0.01					0.32				
Austria	-4.5	-1.4	0.02	2.6	0.1	4.0	8.2	0.76		463	112	C94
Belarus	-10.	-2.2	1.1					1.0			430	EMEP
Belgium	-0.9	-0.4	0.05	0.3	3.8		0.25	0.99		272	149	C94
Bulgaria	-5.4	-1.4	0.24				1.22	0.90		236	351	C90
Czechoslovakia				0.7			3.1	...				
Czech Republic	-3.7	-0.8					...	1.0		508	949	C90
Slovakia	-2.7	-0.4					...	0.53		125	198	C90
Denmark	-0.7	-0.1	0.04	8	200	32	1.0	0.51	2 ^c	327	104	C94
Estonia	-2.6	-0.2	0.07	10				0.16		78	3	C90
Finland	-33.	-0.1	0.04	780	ne	ne	5.8	0.50		95	142	C94
France	-21.	-7.8	1.7	8.5	56	23	3.1	5.6		1626	1216	C94
Germany	-15.	-3.7	0.16	170	ne	ne	14.	8.0		1660	3188	C94
Greece	-3.7	-3.7	3.1	0.2	10.7	11	0.01	1.0		280	169	C94
Hungary	-2.4	-0.8	0.26				2.6	1.1		324	248	C90
Iceland	-0.2	-1.6		87-96			0.10	0.03		11	10	C94
Ireland	-0.5	-3.3	0.11	76-150	37	0	0.53	0.35		657	151	C94
Italy	-9.5	-3.4	6.2	6.1			1.5	5.8		1898	2234	C94
Latvia	-3.9	-0.6		35				0.26		110	50	C90
Lithuania	-2.8	-0.8						0.36		199	29	C90
Luxembourg	-0.1	-0.0			0	2	0.08	0.04		0.8	21	C94
Moldova	-0.4	-1.2						0.45				
Netherlands	-0.4	-0.8	0.04	1.2	38	50	0.15	1.5		563	515	C94
Norway	-12.	-0.1	0.06	400-540	ne	ne	7.4	0.42		97	370	C94
Poland	-12.	-2.8	1.4	35-130			4.4	3.8		1861	3855	C90
Portugal	-4.2	-0.6	5.4		ne	ne	0.02	1.0		204	49	C94
Romania	-9.4	-3.3	0.08				2.7	2.3		594	1270	C90
Russia	-200.	-41.	20	3200			41.	9.0			3400	EMEP
Spain	-22.	-7.2	13	0.9	ne	ne	2.8	3.9		928	1382	C94
Sweden	-39.	-0.4	0.6	840-270	1220	440	18.	0.85		216	58	C94
Switzerland	-1.5	-1.1	0.06	1.6	1.2	0	1.9	0.65		223	93	C94
Ukraine	-13.	-4.9	0.57	560				5.2				
United Kingdom	-3.4	-7.8	0.05	120-180	ne	ne	9.8	5.7		1116	2732	C94
Yugoslavia, ex.			0.96				2.5	...				
Bosnia Herz.	-3.3	-1.0					...	0.44				
Croatia	-2.9	-1.1	0.02				...	0.48		63	75	C94
Macedonia	-1.5	-0.2					...	0.22				
Serbia-Mont.	-5.8	-1.3					...	1.1				
Slovenia	-1.4	-0.4					...	0.20		42	82	C90
Other				0-130 ^d					7.5 ^e			
Sum	-460	-110	55	6300-6100			130	67	9.5	15,000	24,000	

Sources: C94, CORINAIR-94 [European Environmental Agency, 1997]; C90, CORINAIR-90 [McInnes *et al.*, 1996]; EMEP, national data submitted to EMEP, 1997. Methane fluxes are in Gg yr⁻¹.

^aWhere possible, agricultural emissions have been derived from the detailed emissions reported in C94, excluding forests. "Other" emissions are from other anthropogenic activities (combustion, etc.). For CH₄, natural emissions are mainly wetlands, agricultural mainly enteric fermentation. For emissions from EMEP, no breakdown has been possible, so the numbers given may include natural emissions.

^bRanges given derived only from different area-statistics from Table 12, Maltby and Immirizi given first.

^cDanish coastal waters.

^dApplies to estimate derived from Aselmann and Crutzen data, includes fens and bogs from principally Estonia, Latvia, and Lithuania, the floodplain between Romania, Bulgaria, and Yugoslavia, and marshes and swamps, mostly from central East Europe.

^eNorth Sea.

emissions in Europe as a whole, although with somewhat greater contributions in a few countries (e.g., 5%, Finland). Humans and birds are the greatest contributors. The relative contribution of the larger mammals to these totals have been presented in Table 16. A much larger number of species contribute to the bird totals, but the top nine contributors account for about 30% of the total emission (Table 22).

The emissions from waters given in Table 23 are adapted from Barrett [1998] and reflect the influence of terrestrial ammonia sources on European marine areas. As already indicated by Asman *et al.* [1994], the southern bight of the North Sea acts mainly as a sink for ammonia because of the intensity and proximity of terrestrial sources. In areas where observed

seawater NH₄ levels are high and adjacent terrestrial emissions are weak, European seas can be (weak) net emitters. The strongest emission fluxes are calculated for the German bight in the northern North Sea, with 126 μg N m⁻² d⁻¹, followed by 21 μg N m⁻² d⁻¹ in the northern Baltic Sea and 7 μg N m⁻² d⁻¹ in the southeastern part of in the Mediterranean Sea. Over the north Atlantic ocean along the west coast of Europe, and over much of the Mediterranean, NH₃ is on average deposited.

6.5. Sulphur Emissions

The natural sulphur fluxes are compared with anthropogenic emissions in Table 24. On an annual basis volcanoes dominate the natural emissions, and indeed account for ~10% of total

Table 16. Emissions From Wild Animals by Species

	CH ₄ , Gg/yr	NH ₃ , Gg N/yr
Moose	61.	2.2
Red deer	29.	1.1
Roe deer	24.	.89
White-tailed deer	6.8	.24
Fallow deer	4.5	.16
Chamois	3.1	.11
Reindeer	1.3	.048
Boar	.66	.36
Sika deer	.36	.013
Ibex	.33	.012
Mufflon	.32	.012
Total	132	5.1

emissions. Total DMS emissions from the seas amount to <2% of total European sulphur emissions (14 Tg [Olendrzyński, 1997]). However, emissions are comparable with emissions from smaller European countries, especially in the spring season. Emissions from fires are negligible.

The volcanic emissions derive from Allard [1991] and Andres and Kasgnoc [1998]: 0.872 Tg S yr⁻¹ for Italy (Etna, 0.73; Stromboli, 0.14; Volcano, 0.008) and 0.45 Gg S yr⁻¹ for Iceland (Kverkfjöll). The Italian estimate, derived from a 10-year data set for 1975–1985, compares rather well with the data submitted to EMEP for the years 1985–1992: 0.8–1.4 Tg S yr⁻¹ [Olendrzyński, 1997]; other estimates are considered in section 7.9.

DMS fluxes to the atmosphere were calculated for the meteorological conditions of 1988, with the resulting yearly fluxes from different sea regions presented in Table 25. The average contributions for May 1988 are also included as this is the period of maximum flux for most sea regions considered. The calculated values correspond well with other reported estimates of DMS fluxes. Leck and Rodhe [1991] estimate summer fluxes in the Baltic Sea to be 120 to 170 mg S m⁻² d⁻¹. In the North Sea, Leck and Rodhe estimate summer fluxes between 120 and 690 mg S m⁻² d⁻¹, while Malin *et al.* [1993] report June–July values from 320 to 640 mg S m⁻² d⁻¹. For the Mediterranean Sea, Simó *et al.* [1997] report an averaged flux from April to July of 175 mg S m⁻² d⁻¹.

We have not presented any methodology for sulphur emissions from soils or vegetation, as the assessment of Bates *et al.* [1992] is probably still valid. This study indicated a flux from terrestrial surfaces at European latitudes of between 0 and 0.19 μmol S m⁻² d⁻¹. Scaling for the area of Europe suggests an annual emission of ~8 Gg S yr⁻¹. This is comparable to forest fire emissions, although still negligible in comparison to other natural and anthropogenic sources.

7. Uncertainties

Uncertainty estimates from nearly all of the natural source categories share a number of common features:

1. The availability of emission factors are usually very limited. With the exception of volcanoes, none of the source sectors considered has an emission factor determined to better than a factor of 2, even for an overall estimate.
2. Definitions of the emission sectors and land use statistics are usually difficult and vary between countries. For example, 1 km² of forest or wooded land may encompass either dense

forest (2000 g m⁻² biomass, say) or scattered trees (e.g., 100 g m⁻²). Fens, bogs, mires, etc., are similarly poorly defined and are often arbitrary representations of complex ecosystems.

3. The availability of statistical information is often very variable from country to country. This is partly due to the definition problems above but also reflects national/economic interests. For example, statistics associated with the forestry industry are usually well documented for those countries where forestry is a major activity (e.g., Scandinavia). On the other hand, statistics for unmanaged forest, seminatural vegetation, wild animals, or wetlands have little economic value and hence are not always collected.

This means that the variability presented here does not present a basis for performing error propagation calculations but rather indicates areas of data limitations, where additional research may significantly improve the reliability. Table 26 presents a subjective overview of the uncertainties associated with the nature sources categories. The justification for the assigned uncertainty codes is given below, but it should be remembered that within all classes some subsystems (e.g., particular ecosystems, countries) may be better characterized than this table suggests, whereas others are worse.

7.1. Forests

As illustrated in Table 20 isoprene emissions from Europe are expected to be dominated by a few oak species, notably *Q. robur* and *Q. petraea*, as well as by Norway spruce. The European basis for emissions potentials for these oak species is very sparse: Isidorov [1985], König *et al.* [1995], Schnitzler *et al.* [1996], and Steinbrecher *et al.* [1997a]; although results seem broadly consistent with the more numerous American results [Guenther *et al.*, 1994]. Norway spruce emissions are currently predicted to be the major coniferous source of isoprene, and Janson *et al.* [1998], Kempf *et al.* [1996], and Steinbrecher *et al.* [1997b] obtain emission potentials for this species which agree within a factor of 2. The major uncertainties in making isoprene emission estimates are in eastern and southern Europe, where emission potentials for several important species have currently to be assigned on the basis of very few or no experiments. The Mediterranean region especially has a much more varied mix of species than that found in the rest of Europe, and it will be many years before the emission characteristics of this region are adequately characterized. Improvements in the land use databases, particularly regarding species composition and associated biomass data, are clearly required for all regions, but especially for eastern and southern Europe.

In general, emission potentials calculated for terpenes are much more uncertain than for isoprene, and in many cases probably overestimated, as enclosure-type measurements will usually disturb the tree and stimulate emissions. Fortunately, for the important Norway spruce species, Janson [1993], Kempf *et al.* [1996], and Steinbrecher *et al.* [1997b] obtain monoterpene emission potentials which agree within a factor of 2. On the other hand, Seufert *et al.* [1997] notes that different studies from the BEMA campaign obtained emission potentials (ϵ values) of between 2.3 and 58 μg/g/h for *Q. ilex*, the wide range presumably caused by different conditions and behaviors of the tree/shrub varieties of this species. Under standard conditions (sunlit, exposed, etc.) a much narrower range was achieved, from 16 to 27 μg g⁻¹ h⁻¹. An 11-laboratory intercomparison exercise as part of the BEMA campaign [Larsen *et al.*, 1997] suggested that in moderate to high ozone levels (61–125 ppb) most laboratories could measure β-pinene in artificial air to

Table 17. Estimated Emissions of NO_x From Biogenic and Man-Made Sources

Soil NO _x Emissions ^a														
Skiba Methodology					D&K Methodology					Lightning				
Fertilizer	Background	N dependent	Sum	BEIS-2	Y&L	Crop	Grass	Woods	Sum	<1000 m	0–7 km	Fires	Man-Made ^b	
Albania	0.2	0.06	0.08	0.4	4	1	2.5	0.5	0.1–2.8	3.1–5.8	0.01	0.0	0.00	30
Austria	0.4	0.20	0.46	1.1	7	7	5.5	2.4	0.3–8.7	8.2–17	0.03	0.1	0.00	54
Belarus	2.0	0.49	0.77	3.3	*	7	23	3.8	0.8–20	27–46	0.07	0.3	0.18	62
Belgium	0.5	0.08	0.23	0.8	7	4	3.0	0.7	0.06–1.7	3.7–5.3	0.01	0.1	0.01	105
Bulgaria	1.3	0.26	0.48	2.1	9	12	15	2.4	0.4–11	18–29	0.04	0.2	0.04	70
Czechoslovakia				14	16									
Czech Republic	1.2	0.02	0.44	1.7		13	13	1.4	0.3–7.1	15–22	0.03	0.1		132
Slovakia	0.5	0.12	0.22	0.9		5.1	5.1	0.7	0.2–5.2	6.0–11	0.02	0.1		53
Denmark	1.2	0.10	0.25	1.5	10	3	9.2	0.2	0.05–1.3	9.5–11	0.01	0.1	0.01	83
Estonia	0.2	0.10	0.12	0.4	*	1	4.1	0.3		4.4	0.01	0.1	0.01	15
Finland	0.6	0.72	0.17	1.5	16	3	9.1	0.1		9.2	0.11	0.6	0.01	86
France	7.4	1.30	1.95	10.6	38	53	69	13.4	1.5–40	84–123	0.18	0.9	0.27	512
Germany	5.1	0.83	2.41	8.4	30	37	43	6.4	1.0–28	51–78	0.12	0.6		673
Greece	1.3	0.31	0.22	1.8	23	5	14	6.3	0.3–7.1	21–28	0.04	0.2	0.03	109
Hungary	1.1	0.22	0.40	1.7	11	10	19	1.4	0.2–4.6	21–25	0.03	0.2	0.50	57
Iceland	0.0	0.24	0.03	0.3		0	0.0	2.7		2.7	0.03	0.2	0.04	7
Ireland	1.1	0.16	0.23	1.5	8	3	3.4	5.6	0.0–0.9	9.0–9.9	0.02	0.1		36
Italy	2.6	0.70	1.10	4.4	36	16	43	5.9	0.7–18	50–67	0.10	0.5	0.02	656
Latvia	0.4	0.15	0.18	0.7	*	2	6.2	1.0		7.2	0.02	0.1	0.99	14
Lithuania	0.6	0.15	0.25	1.0	*	3	8.4	1.4		9.8	0.02	0.1		23
Luxembourg	0.1	0.01	0.02	0.1	1	0	0.2	0.1	0.01–0.2	0.3–0.5	0.00	0.0		7
Moldova	0.3	0.08	0.11	0.5	*	6	6.1	0.4	0.0	6.5	0.01	0.1		11
Netherlands	1.2	0.08	0.39	1.6	5	5	3.3	1.3	0.0–0.8	4.6–5.4	0.01	0.1	0.01	165
Norway	0.3	0.73	0.39	1.4	19	2	3.1	0.1		3.2	0.12	0.6	0.01	68
Poland	2.2	0.72	1.56	4.5	22	35	53	4.8	0.9–24	59–82	0.10	0.5	0.23	336
Portugal	0.4	0.22	0.17	0.8	12	5	11	1.0	0.3–8.0	13–20	0.03	0.2	0.87	77
Romania	2.0	0.55	0.85	3.3	18	21	36	5.7	0.7–18	43–60	0.08	0.4	0.01	97
Russia	7.4	9.03	4.55	21.0	*	216	230	71		301	1.42	7.1	3.28	607
Spain	3.2	1.18	0.84	5.2	39	35	72	12	1.6–43	86–128	0.16	0.8	2.05	372
Sweden	0.6	0.97	0.61	2.2	22	5	10	0.7		10.7	0.15	0.7	0.10	113
Switzerland	0.2	0.09	0.23	0.5	4	3	1.5	1.9	0.1–2.8	3.5–6.3	0.01	0.1	0.01	42
Ukraine	5.5	1.43	2.11	9.0	*	50	125	8.4	0.9–25	134–158	0.20	1.0	0.09	173
United Kingdom	4.5	0.57	0.90	6.0	23	17	24	13.4	0.2–6.5	37–44	0.07	0.4	0.01	737
Yugosl. ex					17	15	28						0.15	
Bosnia Herz.	0.1	0.12	0.15	0.4			4.0	1.7	0.2–6.4	5.9–12	0.02	0.1		24
Croatia	0.4	0.13	0.18	0.7			5.8	1.9	0.2–5.6	7.9–13	0.02	0.1	0.00	18
Macedonia	0.1	0.06	0.06	0.2			2.9	0.3	0.1–2.9	3.4–6.2	0.01	0.0		12
Serbia Mont.	0.4	0.24	0.7	0.7			9.9	2.3	0.4–11	13–23	0.03	0.1		16
Slovenia	0.1	0.05	0.10	0.2			1.1	0.7	0.1–2.7	1.9–4.5	0.01	0.0		20
Sum	60	24	24	110	700 ^a	600	940	190	13–350	1100–1500	3.4	17	8.9	5700

Emissions in Gg N/yr. The asterisk indicates estimates of BEIS-2 soil NO_x emissions for former Soviet Union (300 Gg N) included in sum.^aFor descriptions of Skiba, BEIS-2, Y&L (Yienger and Levy), D&K (Davidson and Kingler) methodologies see text.^bEMEP emissions for 1994 from *Olendzyski* [1997].

Table 18. Estimated Emissions of NMVOC From Biogenic (This Study) and Anthropogenic Sources

	Forests				Grasslands/Pastures				Crops ^a	Fires ^b	Wetlands ^c	Biogenic Total ^d	Man-Made ^e
	Isoprene	MT	OVOC	Sum	Isoprene	MT	OVOC	Sum					
Albania	40	7	7	54	0	0	2	2	4	0.02		60	32
Austria	28	57	40	125	1	1	9	10	6	0.03	<0.03	141	358
Byelorussia	30	77	43	150	1	1	18	20	38	1.5		209	366
Belgium	11	14	9	34	0	0	3	4	5	0.04	<0.01	43	320
Bulgaria	60	26	18	104	1	1	12	13	26	0.34		143	141
Czechoslovakia										...	<0.01		
Czech Republic	15	55	39	108	0	0	6	7	21	...		136	310
Slovakia	43	24	20	86	0	0	3	4	9	...		99	108
Denmark	5	6	4	15	0	0	1	1	9	0.04	0–0.1	25	156
Estonia	1	2	1	4	0	0	1	1	5	0.09		10	13
Finland	39	162	140	341	0	0	0	0	9	0.06	1–5	350	190
France	829	111	110	1050	44	9	79	132	127	2.4	0–0.1	1311	2308
Germany										0.22	0.4–2	474 ^f	2153
Former West	95	123	88	306	1	1	20	23	43	...		(372)	...
Former East	17	29	26	71	0	0	7	8	23	...		(102)	...
Greece	103	28	22	153	18	4	41	63	29	4.3	<0.01	249	362
Hungary	86	5	10	101	0	1	9	10	41	0.36	<0.01	152	140
Iceland	0	0	0	0	0	0	0	0	0	...	0.1–0.3	0	6
Ireland	2	7	4	13	1	1	16	18	3	0.15	0.1–0.6	34	93
Italy	36	32	46	114	8	3	28	39	70	8.6	0–0.1	232	2239
Latvia	12	24	22	59	0	0	4	4	9	...	0.1–0.4	72	24
Lithuania	0	0	0	0	0	0	6	7	13	...		20	54
Luxembourg	1	1	1	3	0	0	0	0	0	0	<0.01	3	17
Netherlands	3	2	2	7	0	0	5	6	5	0.05	<0.01	18	375
Norway	33	74	53	160	0	0	0	0	2	0.09	0.4–2	162	365
Poland	71	83	78	232	1	1	22	25	87	2.0	0.1–0.5	345	785
Portugal	43	61	36	140	24	3	12	39	27	7.5		203	227
Romania	118	42	38	197	1	2	27	31	61	0.09		289	465
Russia	1719	2057	1350	5125	16	21	315	352	362	28.	7–35	5605	2861
Spain	177	225	109	511	117	27	78	222	87	18.	<0.01	838	1120
Sweden	120	252	209	581	0	0	2	2	9	0.81	1–6	594	459
Switzerland	5	15	10	31	0	0	6	6	2	0.09	<0.01	39	218
Ukraine	278	102	94	474	2	3	45	50	237	0.79		762	429
United Kingdom	38	25	15	77	20	5	48	73	27	0.07	0.2–0	177	2354
Yugoslavia										1.3		...	
Bosnia Herz.	51	10	11	72	0	0	7	8	9	...		89	101
Croatia	33	7	7	47	1	1	13	15	14	0.03		76	73
Macedonia	21	4	4	30	0	0	1	1	4	...		35	7
Serbia Mont.	78	16	17	112	1	1	11	12	17	...		141	52
Slovenia	13	3	3	19	0	0	4	5	3	...		27	34
Sum	4300	3800	2700	11,000	260	86	860	1200	1400	77	10–50	13,500	19,300

MT, monoterpene; OVOC, other VOC emissions.

^aDefault estimate only, see text.^bMean of several years.^cRange given for assumption of 10–50% isoprene emitting area within peatland.^dExcluding wetlands.^e1994 emissions, *Olendryński* [1997].^fSum of East + West Germany.

within 50% of the expected value, but for *d*-limonene, *trans*- and *cis*- β -ocimene, and linalool factors of 10 difference were found. This study also suggested that the more reactive terpenes will hardly show up in many measurements.

Greatest uncertainty is associated with the OVOC emission potentials, which dominate the total VOC estimate. There is hardly any basis for an estimate of these emissions in Europe, except the default rate suggested by *Guenther et al.* [1994], 1.5 $\mu\text{g g}^{-1} \text{h}^{-1}$, which was based on a measured range from 0.5 to 5 $\mu\text{g g}^{-1} \text{h}^{-1}$. (See also section 4.1.1.)

Even though much progress is being made in emission algorithms [*Guenther et al.*, 1993; *Schnitzler et al.*, 1997; *Schuh et al.*, 1997], awareness has grown of the large uncertainties associated with specifying land cover for particular species. 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*, 1997].

In Europe, coherent land use data sets suitable for these emission estimates data sets are simply not available. The data used in this study were derived as far as possible from national statistics, and quality will differ greatly from land to land. Satellite data, such as that underlying the SEI database used in this study, provide a spatially comprehensive method of mapping vegetation with very high resolution. Use of such data is extremely valuable, but only if ground validation has been performed. When ground measurements are used to calibrate multitemporal satellite data, dominant plant species can often be resolved but the resulting algorithms can be applied only over a limited area. Studies in the United States demonstrate

Table 19. Estimated Contribution of Vegetation Species to Total European NMVOC Emissions, Sorted in Order of Total Contribution

Species	Isoprene	Terpenes	OVOC	Total BVOC
Picea unspecified	545	1530	1070	3150
Crops	0.	90	1350	1440
Quercus unspecified	1300	5.7	42	1350
Pinus unspecified	22	865	433	1320
Picea abies	194	557	400	1150
Populus tremula	882	0.	29	910
Pasture	41	53	790	885
Pinus sylvestris	13	275	275	564
Quercus petraea	376	1.6	12	389
Quercus robur	320	1.3	10	332
Quercus ilex	1.1	220	21	243
Quercus decid	186	0.8	6.1	193
Med low veg	113	11	26	151
Other conifers	2.4	98	49	149
Abies	2.1	86	43	130
Quercus pubesc.	113	0.5	3.5	117
Quercus Faginea	81	0.3	2.5	83
Eucalyptus	50	9.7	4.8	65
Heathland	48	5.0	12	65
Populus unspecified	60	0.0	1.9	62
Pinus pinaster	2.5	6.5	48	57
Picea sitchensis	27	20	9.8	56
Fagus	1.9	16	37	56
Other broadleaf	2.6	0.0	51	54
Maquis	34	3.7	8.5	46
Quercus Lusitan.	39	0.2	1.2	40
Pinus halepensis	1.1	9.8	23	34
Med shrub and herb	24	2.3	5.2	31
other trees	1.3	0.0	24	26
Betula	0.9	2.5	19	22
Pinus laricio	0.4	14	7.0	21
Grass	0.8	1.0	15	17
Pinus nigra	0.3	11	5.6	17
Monte hueco	1.2	12	2.2	15
Quercus suber	0.5	1.4	10	12
Salix	11	0.1	0.6	12
Pseudotsuga	0.3	5.5	5.5	11
Olea	0.5	0.0	9.1	9.6
Pinus radiata	0.2	6.2	3.1	9.5
Quercus other	8.4	0.1	0.5	8.9
Alnus	0.2	3.9	3.9	8.0
Larix	0.2	3.5	3.5	7.1
Robinia	5.5	0.2	0.5	6.3
Pinus brutia	0.1	3.6	1.8	5.5
Pinus uncinata	0.1	2.9	1.4	4.4
Noncitric fruit	0.2	0.5	3.5	4.2
Castanea	0.2	0.0	3.4	3.5
Vitis	0.1	0.2	2.9	3.2
Pinus contorta	0.1	2.1	1.0	3.2
Carpinus	0.1	0.9	2.1	3.1
Quercus borealis	2.6	0.0	0.1	2.7
Citrus	0.1	1.2	1.2	2.5
Juniperus	0.1	0.7	1.6	2.4
Fraxinus	0.1	0.0	1.7	1.8
Acer	0.0	0.6	0.3	0.9
Pinus cembra	0.0	0.5	0.3	0.8
Pinus pinea	0.0	0.6	0.1	0.7
Temp shrub bush	0.5	0.1	0.1	0.6
Quercus rubra	0.5	0.0	0.0	0.5
Prunus	0.0	0.0	0.3	0.3
Quercus cerris	0.0	0.0	0.3	0.3
Pinus strobus	0.0	0.2	0.1	0.2
Cedrus	0.0	0.0	0.2	0.2
Malus	0.0	0.1	0.1	0.2
Corylus	0.0	0.0	0.1	0.1
Platanus	0.1	0.0	0.0	0.1

Emissions given in Gg/yr.

Table 20. Estimated Biogenic VOC Emissions From the United Kingdom

Species	Area, km ²	Isoprene	Terpenes	OVOC	Total BVOC
Betula	884	0.0	0.0	0.2	0.2
Fagus	884	0.0	0.1	0.2	0.3
Fraxinus	884	0.0	0.0	0.2	0.2
Larix	1767	0.0	0.4	0.4	0.8
Picea abies	1325	0.9	2.7	2.0	5.6
Picea sitchensis	6185	22.6	16.3	8.2	47.1
Pinus contorta	1546	0.0	2.1	1.0	3.1
Pinus nigra	442	0.0	0.6	0.3	0.9
Pinus sylvestris	2872	0.1	1.9	1.9	3.9
Pseudotsuga	442	0.0	0.4	0.4	0.8
Quercus decid	1988	13.8	0.1	0.5	14.4
Heathland	30,000	18.2	2.0	4.7	24.9
Pasture	111,800	2.1	2.9	43.6	48.6
Crops	66,000	0.0	1.7	25.7	27.4
Total		57.7	31.2	89.3	178.2

Emissions given in Gg/yr.

that there are discrepancies of up to a factor of 5 between satellite-derived isoprene emissions and ground-based determinations [Lamb *et al.*, 1997]. A particular problem in Europe is that species such as spruce or scots pine are “dominant” over very large areas, masking the nondominant but important isoprene-emitting species such as oaks or poplars.

Further discussion of the uncertainties surrounding land use can be found in the works of Guenther [1997] and Simpson *et al.* [1995].

7.2. Natural Grassland and Other Low Vegetation

Few measurements are available of VOC emissions from natural grasslands, shrubs and bushes. The studies available include a limited number of intensive field campaigns held at a few locations in the northwestern Mediterranean region, as part of the BEMA project [e.g., Owen *et al.*, 1997] and in the UK [e.g., Cao *et al.*, 1997]. Additionally, a limited amount of screening work has been carried out on these ecosystems [e.g., Hewitt and Street, 1992]. More data are clearly needed on NMVOC emissions (including <C₄ oxygenated compounds) for major grassland and shrub-type biomes in Europe. For example, there is hardly any information about heather, tundra, grasslands, mountainous regions in northern Europe with ferns and other scrub, alpine pastures, or steppe.

The vegetation species found in these ecosystems are often very aromatic and may be expected to emit a very wide and complex range of volatile organic compounds. This is especially so for Mediterranean vegetation. By far the majority of efforts to date have been focused on the emissions of isoprene and monoterpenes, so it is difficult to quantify the emissions of these other VOC (including the oxygenated compounds), nitrogen or sulphur compounds.

Single herbaceous species may occur in certain areas in relatively large quantities, but little is known about most of them. One example for which measurements exist is *allium ursinum* (wild garlic) which grows in middle and northern Europe in beech and other mixed hardwood forests in spring with biomass densities up to 300 g m⁻². Although wild garlic emits no isoprene and little terpenes, the emission potential of OVOCs was found to be 2.6 µg g⁻¹ h⁻¹ [Puxbaum and König, 1997]. Although it seems unlikely that such uninventoried spe-

Table 21. Estimated NH₃ Emissions

Country	Fires ^a	Mammals (Wild)	Birds	Humans	Sum Nature	Man- Made ^b
Albania			0.00	0.13	0.13	26
Austria	0.00	0.30		0.31	0.62	77
Belarus	0.11		0.70	0.43	1.2	180
Belgium	0.00	0.02		0.41	0.43	79
Bulgaria	0.02	0.07	1.02	0.37	1.5	120
Czechoslovakia ^c		0.14			(1.1)	
Czech Republic			0.34	0.42	0.76	76
Slovakia				0.22	0.22	39
Denmark	0.00	0.04	0.40	0.21	0.65	93
Estonia	0.01		0.00	0.07	0.08	24
Finland	0.00	0.21	0.88	0.21	1.3	26
France	0.16	0.12	1.17	2.31	3.8	550
Germany	0.02	0.60		6.62	7.2	512
Greece	0.30		0.00	0.41	0.71	64
Hungary	0.03	0.12		0.44	0.6	128
Iceland			1.26	0.01	1.3	2
Ireland	0.01	0.02		0.15	0.18	101
Italy	0.60	0.05		2.37	3.0	320
Latvia			0.00		0.0	14
Lithuania			0.00		0.0	66
Luxembourg			0.00	0.02	0.02	7
Moldova			0.00	0.18	0.18	39
Netherlands	0.00	0.01		0.61	0.62	141
Norway	0.01	0.27	1.06	0.17	1.5	21
Poland	0.14	0.21	0.65	1.57	2.57	316
Portugal	0.52		0.00	0.43	0.97	76
Romania	0.01	0.13	0.47	0.95	1.56	182
Russia	1.98	1.47	7.29	3.71	14.	636
Spain	1.23	0.12		1.61	2.9	284
Sweden	0.06	0.65	0.95	0.35	2.0	48
Switzerland	0.01	0.07		0.27	0.35	49
Ukraine	0.06		0.43	2.14	2.6	600
United Kingdom	0.00	0.35	2.10	2.36	4.8	264
Yugoslavia, ex. ^c	0.09	0.14			(1.2)	
Bosnia Herz				0.18	0.18	26
Croatia	0.01			0.20	0.21	20
Macedonia				0.09	0.09	14
Serbia-Mont				0.44	0.44	74
Slovenia				0.08	0.08	22
Total	5.4	5.1	27. ^d	28.	65	5315

Emissions given in Gg N/yr.

^aMean given from multiple years.^bEMEP emissions for 1994 from Olendrzyński [1997].^cSum nature in parentheses from sum of data for new Republics.^dTotal European emissions greater than sum of country emissions; see text.

cies will change regional emission totals significantly, local emission totals might be influenced.

7.3. Forest Fires

Andreae [1991] suggests that the uncertainty of emission estimates from forest fires is ~50% for CO₂ and a factor of 2

Table 22. Species Contributing Most to NH₃ Emissions from Birds and Percent Contribution to Total

Species	Life Weight, kg	Emissions	Cumulative, %
Carrion crow	0.55	1244	5%
Woodpigeon	0.48	1215	9%
Blackbird	0.1	1063	13%
Starling	0.07	946	17%
Mallard	1.1	826	20%
Pheasant	0.8	810	23%
Rook	0.5	804	26%
Feral pigeon	0.35	728	29%
Hazel grouse	0.8	705	31%

Emissions given in Mg N/yr.

for the other trace gases. Although most measurements have been made outside Europe (e.g., South America, United States, Canada), the emission ratios obtained (ratio of trace gas to CO₂ emission) have been similar regardless of location [see Andreae, 1991, and references therein]. Thus the emission factors used here, and the uncertainties suggested by Andreae, are likely to be applicable also within Europe. However, one possible cause for concern lies in results reported by Hegg *et al.*

Table 23. Averaged Fluxes of NH₃ Between Sea and Atmosphere

	Annual Emissions	Annual Deposition
North sea	12.0	138
Baltic sea	3.6	113
Mediterranean sea	4.0	162
Black sea	0.9	40

Adapted from Barrett [1998]. Fluxes given in Gg N.

[1987], which suggested that areas which had experienced substantial N deposition could have emission ratios for NO_x an order of magnitude greater than those obtained in unpolluted rural areas. Indeed, emissions of purely man-made species such as F12 are also observed from forest fires, again the result of resuspension of previously deposited pollutants [Hegg *et al.*, 1990]. Such resuspension is very likely in many areas of Europe and suggests that our estimated NO_x emissions may be a lower limit.

Few measurements are available of emissions from natural forest fires, and all emission rates and biome factors reported here are based on studies in North America or the Amazon. For example, the burning efficiency is here set to 0.2 for forest fires, following Seiler and Crutzen [1980]. However, efficiencies of 0.76 have been reported from wild fires in Australia [Hurst *et al.*, 1996], or 0.1 for fires in Siberia [Dixon and Krankino, 1993]. More measurements in European conditions are re-

Table 24. Estimated Sulphur Emissions

Country	Fires ^a	Waters	Volcanoes ^a	Sum Nature	Man-Made ^b
Albania	0.	36
Austria	0.	37
Belarus	0.06	0.12	162
Belgium	0.	0.01	127
Bulgaria	0.01	0.03	740
Czech Republic	635
Denmark	0.	0.01	78
Estonia	0.01	0.03	71
Finland	0.01	0.01	56
France	0.10	0.19	507
Germany	0.01	0.02	1498
Greece	0.18	0.34	278
Hungary	0.02	0.03	371
Iceland	0.45	0.45	12
Ireland	0.01	0.01	89
Italy	0.36	...	870	870	719
Latvia	26
Lithuania	59
Luxembourg	7
Moldova	54
Netherlands	0.	0.01	77
Norway	0.	0.02	17
Poland	0.08	0.15	1303
Portugal	0.32	0.60	136
Romania	0.	0.01	456
Russia	1.2	3.04	1492
Slovak Republic	119
Slovenia	89
Spain	0.75	1.42	1031
Sweden	0.04	0.09	49
Switzerland	0.	0.01	16
Ukraine	0.03	0.06	858
United Kingdom	0.	0.01	1360
Yugoslavia, ex	0.06	0.10	...
Bosnia Herz.	240
Croatia	0.	0.01	45
Macedonia	53
Serbia Mont.	212
Seas					
Baltic Sea		10		10	36
Black Sea		14		14	...
Mediterranean		141		141	6
North Sea		31		31	238
Total	3.3	196	871	1074	13,400

Emissions in Gg S yr^{-1} .

^aMean and/or range given from multiple years.

^bEMEP emissions for 1994 from Olendrzynski [1997] (includes shipping).

Table 25. Averaged Fluxes of DMS to the Atmosphere

	Yearly Fluxes, $\text{mg S m}^{-2} \text{d}^{-1}$	Year Emission, Gg S	May Flux, $\text{mg S m}^{-2} \text{d}^{-1}$	May Emissions, % of annual
North Sea	150.	32	420.	24%
Baltic Sea	67.	10	180.	23%
Mediterranean Sea	154.	141	550.	30%
Black Sea	83.	14	240.	24%
Total		197		

quired to establish the validity of the methodology adopted here.

Overall, a factor of 3 uncertainty would seem a reasonable first guess for emissions of gases such as NO_x from Europe.

7.4. Soils

In developed areas of the world, such as Europe, the greatest uncertainty in total soil NO emissions is probably in the amount of NO emitted from heavily fertilized farmland, and possibly from N-affected forests if the Davidson and Kingerlee [1997] suggestions are correct. Emissions from natural grasslands are not large, but there is little information on their emission rates. [see, e.g., Davidson and Kingerlee, 1997; Skiba *et al.*, 1997; Veldkamp and Keller, 1997].

The uncertainty of soil NO emission estimates is illustrated well by Table 17, where different methods result in a factor of 10 difference in the annual emissions. Uncertainties over shorter timescales are obviously much greater. Most of the emission algorithms and experiments are associated with temperate (moist) soils, so estimates are probably also poorer for Mediterranean areas than for northern Europe.

Studies are needed to determine the fraction of nitrogen inputs that are subsequently released into the atmosphere as NO. The role of plant canopies in mitigating the flux of NO into the free atmosphere also needs to be explored, and further

Table 26. Subjective Uncertainties of Natural Source Emissions

Source	Emissions	Emission Factor	Land Use/Activity Statistics	Overall
Forests	isoprene	C-D	B-D	D-E
	terpenes	D-E	B-D	D-E
	OVOC	E	B-D	E-F
	total VOC	E	B-D	E-F
Grass/low vegetation	isoprene	C-D	D-E	D-E
	terpenes	D-E	D-E	D-E
	OVOC	E-F	D-E	E
Fires		B-C	A-B	B-C
Soils	NO_x emission	C-D	A-C	C-E
	CH_4 consumption	C-D	A-C	D-E
Wetlands	CH_4	D-E	A-C	D-E
	Isoprene	D	E	E-F
Waters	DMS	B	D	D
Wild animals		D-E	B-C	D-E
Humans		B	A	B
Lightning	NO_x	D	C	E
Volcanoes	SO_2	A-B	A	A-B
Gas seeps	CH_4	D	E	F

Estimated uncertainty factors: A, 25%; B, factor 1.5; C, factor 2; D, factor 3; E, factor 5; F, larger than factor 5.

field studies comparing atmospheric measurements of NO_x fluxes with soil emissions derived from chamber measurements are also clearly needed.

7.5. Wetlands

Wetland flux estimates are probably the greatest source of uncertainty in methane estimates. Although there are measurements in all wetland types from the principal wetland areas, fluxes may vary over several orders of magnitude at a single site, and site-to-site variation is large [e.g., *Christensen et al.*, 1996; *Roulet et al.*, 1994]. Interannual variation of seasonal averages can vary by as much as an order of magnitude. The heterogeneity of wetlands is obviously a key problem [*Öquist and Svensson*, 1996]. Most boreal and temperate zone flux measurements have been made in North America and Scandinavia, and most tropical zone measurements have been made in Central and South America. Since there are few or no other measurements of methane flux from other parts of the world, the uncertainty of using the available measurements cannot be calculated but may be large. Measurements of methane flux in Europe have fit in the range of other boreal and high temperate zone measurements, however.

The estimated areas of wetlands may differ greatly depending on the underlying vegetation databases. The differences in area estimates between *Matthews and Fung* [1987] and *Aselmann and Crutzen* [1989] are discussed at length in the latter paper and in the paper by *Bartlett and Harris* [1993]. Their total areas are very close, but their distribution differs greatly, particularly in the tropics. Their estimates of total area for the northern hemisphere temperate and boreal zones are very close, but their vegetation classes are not strictly comparable.

7.6. Waters

The extrapolation of the European DMS oceanic fluxes summarized in Table 25 to the world oceans results in a global flux of 15 Tg S yr^{-1} , which coincides with the global estimate of *Bates et al.* [1992]. The reason for the good agreement between these estimates is probably that both consider the seasonal cycle of DMS production in seawater, while the earlier estimate of *Andreae and Raemdonck* [1983] did not.

Still, the largest uncertainty is related to the spatial and temporal extrapolation of DMS and NH_x concentrations in seawater from the collected set of observations. The uncertainty of such extrapolation to different biogeographical areas has been quantified to a factor of 3, derived from the standard deviation of the observations. The choice of exchange coefficients in the parameterizations of sea-air exchange is less significant: on average, we can expect variations within 30% depending on the choice of the exchange model.

7.7. Wild Animals and Humans

The applicability of the emission factors is not well known. As noted in section 4.7, the metabolism of farm animals is generally very different from wild animals. Further, we have had to extrapolate emissions factors (emissions per kilogram) derived from farm animals (cattle) to animals weighing up to 30 times less (roe deer). Comparing direct measurements of human methane emissions with estimates scaled from measurements on pigs suggests a factor of 5 uncertainty in such methods (section 4.7); this may serve as a general guideline for this kind of derived emission factors. Human emissions, which are based on direct measurements, are expected to be correct within 50%.

While emission factors for ammonia generally seem to be better understood (based on food intake/excretion of nitrogen), the "canopy effect" has not been taken into account. Within a forest or grassland canopy, deposition of NH_3 may be so efficient that it is removed to the leaves and grass before reaching the open atmosphere. *Bouwman et al.* [1997] therefore assumed NH_3 emissions from forest animals to be negligible. We have no data on the fraction of animals actually living and excreting in a forest rather than in open areas. However, as large herbivores frequently feed on grassland, and birds are abundant above tree canopies, we assume canopy effects (and the related uncertainty) to be <50%. For small animals, the most abundant of which spend part of their time even beneath the ground (hares, voles, lemmings), emission of ammonia may be considered negligible.

With respect to deer numbers, the uncertainty of hunting statistics seems to be relatively small, within ~30%. However, the lack of data for the former Soviet Union is a major problem, as this covers ~40% of the European territory and has a large percentage of the European wild animals. Overall uncertainty in the numbers then is about a factor of 2. Additionally, the bird counts are probably correct only to a factor of 2 or more. With respect to the usual standards for emission inventories, the data quality for human population numbers is very high.

Total emissions from large animals for Europe are therefore probably in the range of $50\text{--}400 \text{ Gg yr}^{-1} \text{ CH}_4$ and $2\text{--}10 \text{ Gg yr}^{-1} \text{ NH}_3$. Ammonia emissions from birds are probably between 10 and 100 Gg yr^{-1} . The human emissions are estimated at $50\text{--}100 \text{ Gg yr}^{-1} \text{ CH}_4$ and $25\text{--}50 \text{ Gg yr}^{-1} \text{ NH}_3$.

Considering the totals of the complete sector, probably the methane emissions are underestimated due to the sources missing in the inventory. This may be small vertebrates (rodents) but also invertebrates such as insects (which are all considered irrelevant for ammonia emissions because of the canopy effect). According to calculations by *Crutzen et al.* [1986], based on food availability, an upper limit of these missing methane emissions from animals may be 6 times the emissions of large wild animals globally. For Europe, at 800 Gg yr^{-1} this is still more than an order of magnitude smaller than methane emissions from animal husbandry. No lower limit or most probable estimate can currently be given due to lack of data.

7.8. Lightning

The uncertainty in the emission factors has previously been estimated to be a factor of 3 [*Novak and Pierce*, 1993]; however, the validity of these results have to be checked with respect to those literature estimates giving results different by up to an order of magnitude [*Biazar and McNidar*, 1995]. A recent study on the contribution of cloud-to-cloud lightning [*Gallardo and Cooray*, 1996] indicates that previous numbers (including the ones used in this paper) may be too low. However cloud-to-cloud lightning is primarily relevant for upper troposphere emissions which are important on the global scale, not for the low-level emissions considered here. Compared to the emission factors, the uncertainty for flash numbers seems to be considerably smaller. Even results from ground-based assessments of flash densities [*Turman and Edgar*, 1982] typically deviate by less than a factor of 2. Altogether, a total uncertainty range of a factor of 5 seems to be indicated from available data ($1.5\text{--}8 \text{ Gg N yr}^{-1}$).

Table 27. Estimated Contribution of Natural/Biogenic Emissions to European and Global Annual Emissions

	S, Tg/yr	NO _x , Tg N/yr	CH ₄ , Tg/yr	NH ₃ , Tg N/yr	NMVOC, Tg/yr
Forests (foliar)					11
Grasslands and other vegetation					1.2
Crops ^a					1.4
Soils ^b		0.1–1.5	–0.57		
Waters	0.2				
Wetlands			6.2		<0.1
Forest fires	...	0.01	0.06	0.005	0.1
Volcanoes	0.9		...		
Wild mammals + birds			0.13	0.03	
Humans			0.07	0.03	
Gas seeps ^c			0.01–2		
Lightning ^d		0.02	...		
Sum of nature	1.1	0.14–1.5	6–8	0.06	13
Anthropogenic, Europe ^e	13	5.7	38	5.3	20
Global emissions	200 ^f	44 (23–81) ^g	500 ^h	54 ⁱ	1200 ^j

Some agricultural emissions are included for comparison.

^aDefault estimate only for comparison.

^bNO_x includes emissions from fertilizers, range given from different methodologies; CH₄ includes forest and other soils.

^cBest guess and speculative upper limit presented.

^dEmissions up to 7 km presented here.

^eOlendrzyński [1997].

^fBluth *et al.* [1993].

^gLee *et al.* [1997].

^hWatson *et al.* [1990].

ⁱBouwman *et al.* [1997].

^jGuenther *et al.* [1995].

7.9. Volcanoes

One dominant source (Mount Etna) accounts for most of the European emissions. The uncertainty in emission fluxes from this source is given as 20% by Allard *et al.* [1991], based on measurements between 1975 and 1985. However, this may be a low estimate that does not fully reflect the natural flux variation. Calabiano *et al.* [1994] obtained a “baseline” activity from measurements in the years 1987–1991, which is 20–40% higher than that given by Allard.

In addition, explosive periods and events have to be considered. During 1990, the SO₂ flux from Etna was on average more than twice the figures used here [Calabiano *et al.*, 1994]. Also, the eruption of an otherwise dormant volcano like Hekla (Iceland) in 1991 may also increase the European annual volcanic emissions by as much as 30% [Andres and Kasgnoc, 1998].

7.10. Gas Seeps

While the emission rate seems to be well established, considering the irregular behavior of this source, huge discrepancies exist in the literature on the size of the active seepage area. Using the most conservative estimates, gas seeps are an almost negligible source of methane. With the inclusion of all potential seepage areas (an area 300 times larger than the our best estimate), gas seeps would be a major contributor to European and also global emissions, as has been reported by Hovland *et al.* [1993]. The upper estimate for Europe, ~3 Tg yr⁻¹, would be of the order of 7% of total European methane emissions.

8. Summary and Conclusions

As part of the work of the UN-ECE Task Force on Emission Inventories, a so called “Nature Panel” was set up to write a chapter for the *EMEP/CORINAIR Atmospheric Emission In-*

ventory Guidebook [McInnes, 1996] dealing with emissions from biogenic and natural sources. This Guidebook is used by many European countries in reporting their national emissions to UN-ECE and the European Union. Here we have presented the applied results of the work of this Panel, a set of (practical) methodologies for estimating emissions of sulphur, nitrogen oxides, NH₃, CH₄, and nonmethane volatile organic compounds (NMVOC) from biogenic and other natural sources in Europe. The source categories covered include forests, grasslands and other low vegetation, forest fires, wetlands, waters, animals, volcanoes, lightning, and gas seeps. We have assembled land use statistics from European or national compilations and presented emission estimates based on these.

Table 27 summarizes our estimated emissions on a European scale, together with estimates of European anthropogenic and global total source strengths. In terms of contribution to total European emissions, NMVOC from forests and vegetation, CH₄ from wetlands, and sulphur from volcanoes are the most significant emissions, using the methodologies recommended for the Guidebook. However, the estimates of soil NO_x emissions cover a very large range. The lower estimate, adopted for the Guidebook, suggests that biogenic NO_x emissions are not significant. With the higher estimate, derived from Davidson and Kingerlee [1997], biogenic sources contribute more than 20% of European NO_x emissions.

On a global scale the biogenic emissions from Europe are not significant, a consequence both of the climate of Europe and of the reduced extent of natural sources such as forests or wetlands since pre-historic times. However, for assessing local budgets, and for photochemical oxidant modelling, natural/biogenic emissions can play an important role. The most important contributor in this regard is undoubtedly forest VOC emissions, although this paper also indicates that NMVOC

emissions from nonforested areas need to be further evaluated.

Other emissions may have great local or regional importance. The volcanic sulphur emissions from Italy exceed the combustion emissions of that country, and have a large effect on calculations of acid deposition in the Mediterranean. In most Nordic countries, CH₄ emissions are dominated by wetland sources. In Russia, emissions of CH₄ from wetlands are comparable to those from anthropogenic activities.

It should be noted that some important natural/biogenic emissions are not addressed as part of this study. These include aerosols, CO₂, N₂O, and heavy metals. They have so far been excluded either because the recommended methodologies are identical to those of IPCC, or because no methodology has been accepted yet as part of the Atmospheric Emission Inventory Guidebook. However, these emissions often have both local and global significance, and future work will seek to include them into the Guidebook.

The uncertainties of natural/biogenic emissions are much larger than those associated with anthropogenic sources. In part this is due to the complexity and variability of natural ecosystems, so that much basic research and extensive measurements will be needed before reliable emissions factors and algorithms are developed.

A significant contributor to the uncertainty of these estimates is also the lack of land use statistics in an appropriate form. Of course, statistics for many sources do exist in many European countries, but formats and definitions vary from country to country, and these data do not exist in any Europe-wide database. One of the most important outcomes of the work presented here will be to establish natural emissions properly into the EMEP/CORINAIR emissions reporting procedures, encouraging countries to supply land use data specifically collected for biogenic inventories to a central database.

A significant accomplishment of this work has also been to establish a framework for identifying the major sources and uncertainties in European estimates, thus enabling the priorities for future research work to be identified.

Appendix

Sources of forest data follow. For the two letter country codes used, see Table 1. Data were obtained as far as possible from national sources, sometimes combined with the following: EFI, *European Forest Institute* [1995] compilation of areas of oak, beech, pine, spruce, deciduous, and coniferous exploitable forest; SB, *Stanners and Bourdeau* [1995]; SEI, SEI land use database, see section 5.1; and EUS, *EUROSTAT* [1985].

AL, 4160 km² allocated by EFI, remaining 10,330 km² (unexploitable forest and other wooded land), split as per *Veldt* [1989]; AT, *König et al.* [1995], assuming 1.4% softwood deciduous split 50:50 between populus, salix; BE, EFI accounts for 4680 km² of 6200 km², plus 260 km² populus from EUS; BG- EFI; BY: EFI; CH, *Andreani-Aksoyoglu and Keller* [1995]; DK, Skove og plantager 1990, February 1994, ISBN 87-501-0887-5. "Pinus sp." for *Pinus mugo* and *Pinus contorta* only, other pines such as *Pinus sylvestris* included in "other conifers"; ES, *Ortiz and Dory* [1990]; FI, *Aarna* [1994]; FR, Inventaire forestier national, 1997 (<http://www.ifn.fr/index-gb.html>); DDR, Bericht des Bundesministers für Ernährung, Landwirtschaft und Forsten, Waldschäden in der Bundesrepublik Deutschland, Reihe A; Angewandte Wissenschaft, no. 349, Landwirtschaftsverlag GmbH, Münster-Hiltrup, Germany, October

1987. FRG, Waldschäden in der Bundesrepublik Deutschland, Reihe A; Angewandte Wissenschaft, no. 390, Landwirtschaftsverlag GmbH, Münster-Hiltrup, Germany, November 1990, with poplars from EUS. Meadows/arable from SB, divided using SEI land cover; GR, Forest Research Institute, Athens, Greece, other wooded land excluded, see text. HU, *Szepesi* [1997] for oak, beech, scots pine. Other coniferous, other broadleaf derived from SB; EI, EFI; IT, Ministero per le Politiche Agricole, 1997; LV, <http://www.rfl.pswfs.gov/pubs/psw-gtr-164/index.html>; NO, Norwegian Institute of Land Inventory [1995], with adjustment for Oak from *Veldt* [1989]; NL, National Reference Centre for Nature, Forests and Landscape, Wageningen, The Netherlands; PL, V. A. Isidorov et al. (Natural VOC emissions in Poland, submitted, 1997); PO, *Pio et al.* [1995]; RO, EFI; UK, Forest research, Alice Holt Lodge, Wrecclesham, UK, for forest cover, heath + moorland derived from B. Bunce (ITE, Merlewood, personal communication, 1997); RU, Total cover for European part estimated from RIVM, forests split using data from *Isidorov* [1993]; SK, *Novotny et al.* [1994]; UA, total from EFI, speciation from *Buksha et al.* [1996]; SI, HR, BA, FYM, YU, total areas for former Yugoslavia split by SEI database, speciated from *Veldt* [1989].

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