Inventory of natural emissions in Switzerland

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Abstract

Biogenic species are emitted mainly from vegetation (volatile organic compounds, e.g. isoprene, monoterpenes) and from soils (nitrogen compounds). The emission rates per unit area from forests are known to be correlated to leaf temperature, photosynthetically active radiation and biomass density leading to space and time dependent emissions. In this study, a biogenic emission inventory for Switzerland was established, the grid size being 5 *km* x 5 *km.* The spatial vegetation distribution was taken from the Swiss land-use inventory. Hourly values of the air temperature and the solar radiation above the canopy were calculated using their dependence on altitude. For forests, these quantities were converted to the corresponding values within the canopy using a forest canopy model. The hourly biogenic emission inventory was prepared for three consecutive days in summer 1993 as well as for monthly averages over ten years.

1 Introduction

Chemical processes in the atmosphere are not only influenced by anthropogenic emissions of nitrogen oxides, carbon monoxide, volatile organic compounds (VOC), etc., but also by organic trace gases from plants or by nitrogen compounds emitted from soils. Neglecting these natural emissions in air pollution models may cause inaccurate results leading to ineffective abatement strategies for adverse species such as nitrogen oxides or ozone. Among the variety of organic species, only isoprene and monoterpenes from forests, crops and pasture have been studied in detail [1-3] because of their high reactivity which controls the OH and NO_3 mixing ratio. Isoprene and monoterpene emissions have recently been estimated for Switzerland [4,5]. About 97% of the emissions are monoterpenes, most of them released by Norway spruce and fir. Isoprene is emitted mainly by oak trees and pasture. In this work, an isoprene, monoterpene and nitric oxide emission inventory for Switzerland was established with a grid cell size of $5\,km \times 5\,km$. Hourly emission rates were calculated for three consecutive days in summer 1993 as well as for monthly averages over ten years.

2 Methodology

Emission rates The emission rates of monoterpenes are usually given as increasing functions of leaf temperature T_{leaf}. For Norway spruce the correlation $E = f(T_{leaf})$ was taken from Schürmann [2]. The rates of emission from fir, Scots pine and larch were computed according to the exponential correlation of Tingey [1].

Isoprene emission is both temperature and radiation dependent. For Norway spruce, the emission rate was assumed to amount to 10% of the α-pinene emission rate during daytime [3]. The rate of isoprene emission from oak trees were split into two factors, C_T and C_L, which depend on leaf temperature and on photosynthetically active radiation (PAR, radiation within the spectral range 400 - 700 nm), respectively [6]:

$$E = E_s\, C_T(T_{leaf})\, C_L(PAR) \tag{1}$$

E_s is the reference emission rate at $T_{leaf} = 30\ ^oC$ and $PAR = 1000\ \mu E\, m^{-2}\, s^{-1}$ which was calculated according to [1].

The isoprene and monoterpene emissions from corn, wheat and pasture were calculated on the basis of the method given by Lamb et. al. [7] where emission fluxes at $T_{leaf} = 30\ ^oC$ and $PAR = 400\ \mu E\, m^{-2}\, s^{-1}$ are reported. The dependence of the emissions on leaf temperature and PAR was taken into account using Tingey's correlation [1].

Emissions of nitric oxide from soils due to microbial activity were calculated for corn, wheat, pasture and forest soils. The dependence of NO emission on soil temperature was taken from Williams et al. [8].

Meteorology Hourly values of air temperature T_a and solar global irradiance G are available from the meteorological monitoring network (ANETZ, 72 stations) of the Swiss Meteorological Institute (SMI). The air temperature can, in general, be represented by a polynomial decreasing with altitude. For clear sky conditions the global irradiance increases with altitude. However, there is still a large scatter due to local dust and cloud effects. In order to compute the PAR values within the canopy, G was split into the direct and the diffuse proportion by simulating the radiation transfer in the atmosphere using the radiation transfer code LOWTRAN 7. The coefficients to convert global, direct and diffuse irradiance [$W\, m^{-2}$] to PAR [$\mu E\, m^{-2}\, s^{-1}$] amount to 2.1, 1.9 and 2.7 $\mu E\ W^{-1}\, s^{-1}$, respectively.

Canopy model Leaf temperature and PAR in forests substantially vary within the canopy. This effect was taken into account by applying the canopy model given in Baldocchi et al. [9], supplemented by experiments carried out at the Hartheim Forest (Federal Republic of Germany) and in Central Switzerland [10]. The attenuation of *PAR* incident at the zenith angle θ is given by

$$PAR\ (z)\ = PAR\ (z = 1)\ \exp\ [-\frac{g}{\cos\theta}\int_{1}^{z}(\ LAD\ (z') + BAD\ (z')\ )\ dz'] \qquad (2)$$

where z is the height normalized to the canopy height and g is the average cosine of the angle of incidence relative to the leaf surface (0.5 for a spherical leaf orientation). *LAD* and *BAD* are the leaf area and the branch area densities at the height respectively. Integration of these two quantities from top to bottom yields the leaf area index (LAI) and the branch area index (BAI) which were set to 6 $m^2\ m^{-2}$ and 2 $m^2\ m^{-2}$, respectively [10].

The difference between the air temperature above and the temperature within the canopy was found to vary with height and with time of the day [10]. Hence, a correction term was added to the value estimated from the ANETZ data. Finally, the leaf temperature is reported to exceed the air temperature at high irradiance levels and to lie below at low levels due to transpiration (e.g. [7]). As the difference between air and leaf temperature amounts to a few degrees only, this difference was approximated by a linear function of the global irradiance at the level z

Biomass density and land-use The conversion from the dry leaf weight related emission rates to the rates based on surface area was performed using the biomass density data typical for the Swiss forests [11]. For crops and pasture the data given by Lamb [7] was used. The land-use data for forests were taken from the Swiss national forest inventory where the coverage of Norway spruce, fir, scots pine, larch and oak were taken into account [12]. The spatial distributions of corn, wheat and pasture were estimated on the basis of the land use inventories [13,14].

3 Results

In Figure 1 the air temperature and the global irradiance measured by the ANETZ stations are depicted as a function of the altitude. As an example, the data of July 29, 1993, 12:00 UTC is given.

The LAD profile together with those of leaf temperature and PAR are given in Figure 2, the correlations being valid for $PAR\ = 1600\ \mu E\ m^{-2}\ s^{-1}$ and $T_a = 20\ ^{o}C$ above the canopy. It is evident from these figures that PAR strongly decreases with decreasing height whereas the leaf temperature is subjected to changes of a few degrees only. The isoprene emission rate per unit leaf area (not given in Figure 2) shows a similar behaviour as PAR. Weighting this emission rate with the LAD yields the rate per unit surface area.

The emission rates of isoprene from oak trees and monoterpenes from Norway spruce are shown in Figure 3. Results are given for calculations with and without canopy model. For isoprene, a decrease of 50% to 85% was found when the canopy model is applied. The canopy effect for monoterpenes, however, is only a few percent.

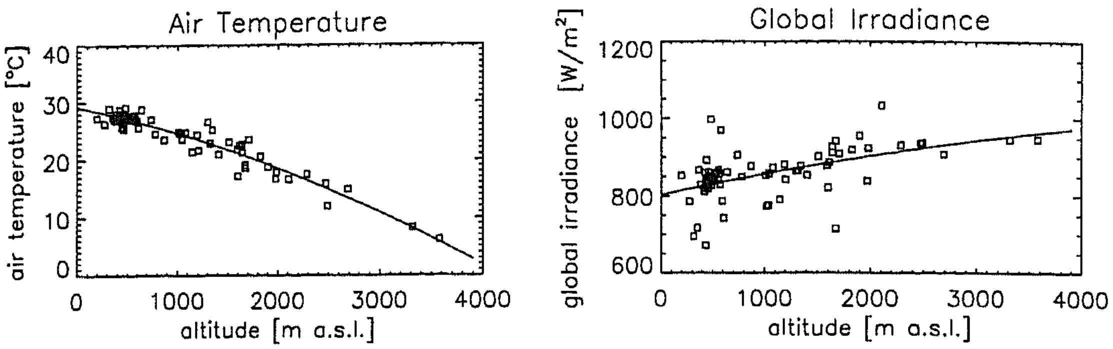

Figure 1: Air temperature and global irradiance of the ANETZ stations, together with the least square fit (solid). July 29, 1993, 12:00 UTC

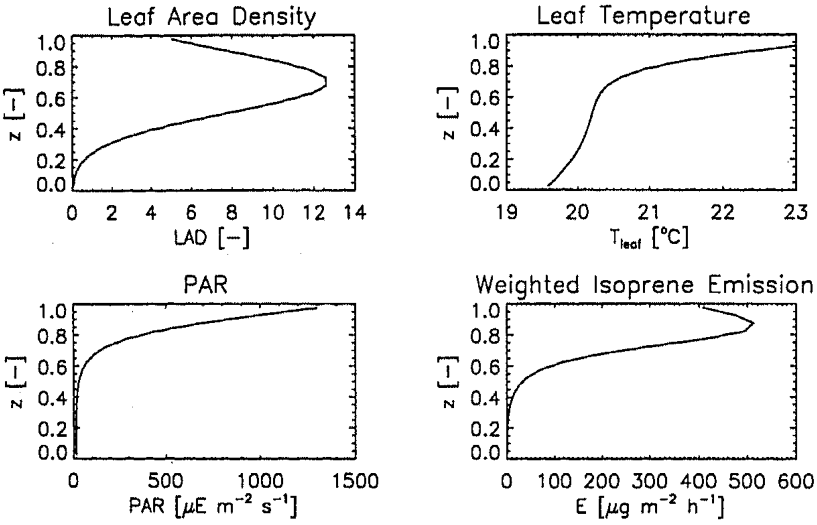

Figure 2: Leaf area density *LAD*, leaf temperature T_{leaf}, photosynthetically active radiation PAR, and isoprene emission rate weighted with *LAD* as functions of the normalized height *z*.

The spatial distributions of the emission rates of isoprene, monoterpenes and nitric oxide were estimated for July 28 -30, 1993, a summer smog episode where a field experiment was performed. Figure 4 shows the emission pattern in the early afternoon. Isoprene is emitted mainly by oak forests and pasture which are located preferentially in the northern part of Switzerland, in the Rhone valley and in the areas south of the Alps. Monoterpenes are released by Norway spruce and, to a minor extend, by fir trees which are found all over

Switzerland. Nitric oxide is emitted by soils of crops and pasture which grow in the Jura, the Swiss Plateau, the Pre-Alpine region and the northern part of the Alps. The main sources of uncertainties are the limited accuracy of the emission model parameters (20 to 50% for isoprene and monoterpenes, up to 200% for nitric oxide), the transfer of known emission data to plant species where no experimental data are available (up to 50%), the individual deviation of air temperature and radiation from the average altitude dependence (10 to 30%), the uncertainty of the land use (20 to 30%) and the year to year variation (about 20%). The total uncertainties were estimated to be about 80% for isoprene, 110% for monoterpenes and more than 200% for nitric oxide.

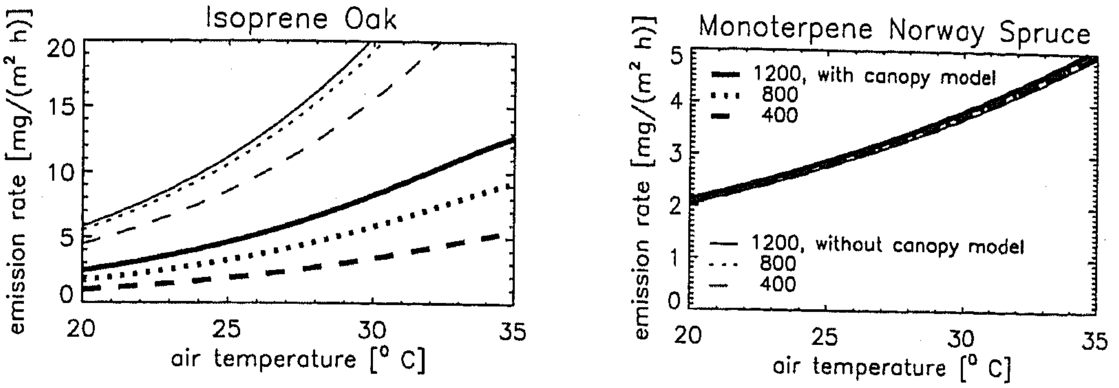

Fig. 3: Emission rates of isoprene from oak trees and monoterpenes from Norway spruce. *PAR = 400, 800* and *1200* $\mu E\ m^{-2}\ s^{-1}$. With (thick lines) and without canopy model (thin lines).

In Figure 5 monthly totals of the above species summed over whole Switzerland are given. The most abundant organic species are the monoterpenes. Isoprene which is released in about equal amounts by oak trees and pasture plays a minor role and vanishes outside the vegetation period (April to October). Finally, nitric oxide is released by crop soils during the growing season (April to August) and by pasture during the vegetation period. The contribution of the forests was found to be negligible.

The annual emission rates of biogenic and anthropogenic sources are given in Table 1. The difference between this work and the previously estimated data in [4,5,17] is due to several reasons. On the one hand, a very coarse forest inventory as well as average temperature and radiation data were used. On the other hand, the correction of the canopy effect was not yet done. The extent of the canopy effect is similar to that reported by Lamb [7] who found a reduction of the annual value of about 50%. These simplifications lead to an overestimation of isoprene, monoterpene and NO emissions by 113%, 16%, and 19%, respectively. The isoprene values given in the EMEP report [15] agree quite well with our data, whereas NO_2 shows larger discrepancies due to different land use data. Relative to the antropogenic VOC, isoprene and monoterpenes amount to about 30%. The corresponding proportion of NO_2, after conversion of biogenic NO to NO_2, is only 2%.

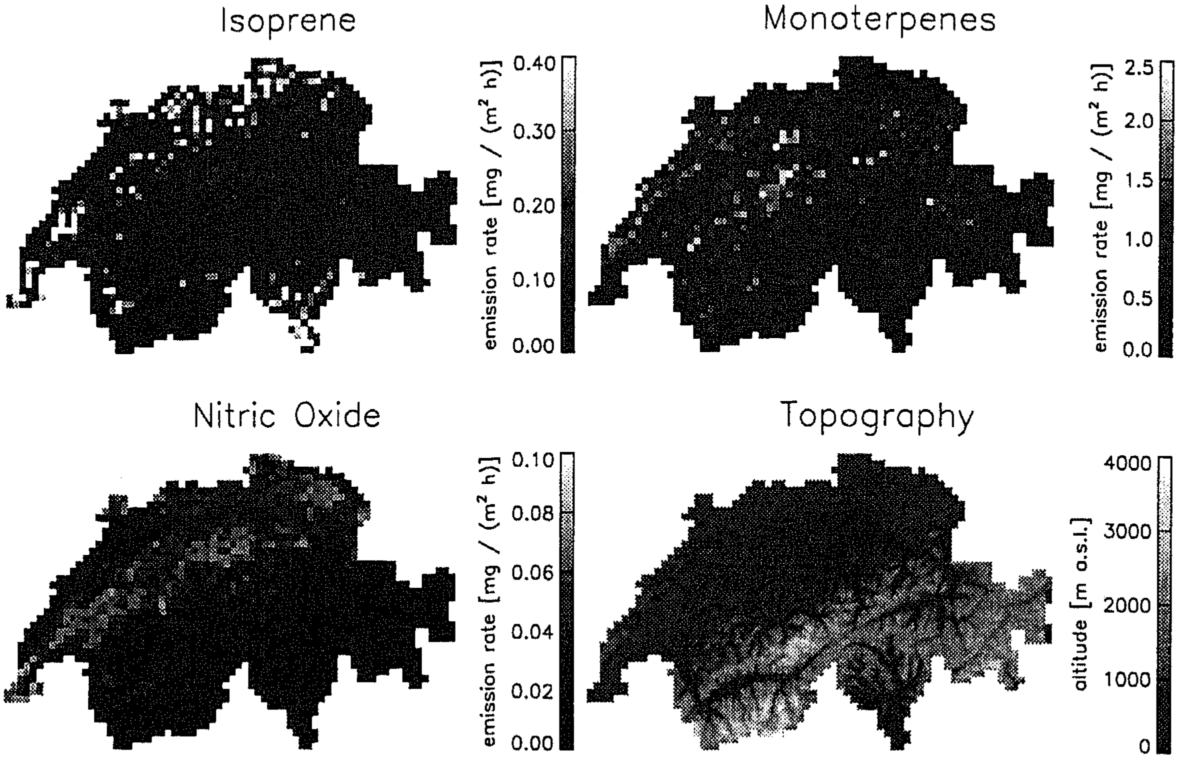

Fig. 4: Spatial distribution of the emission rate of isoprene, monoterpenes and nitric oxide. July 29, 1993, 12:00 - 13:00 UTC.

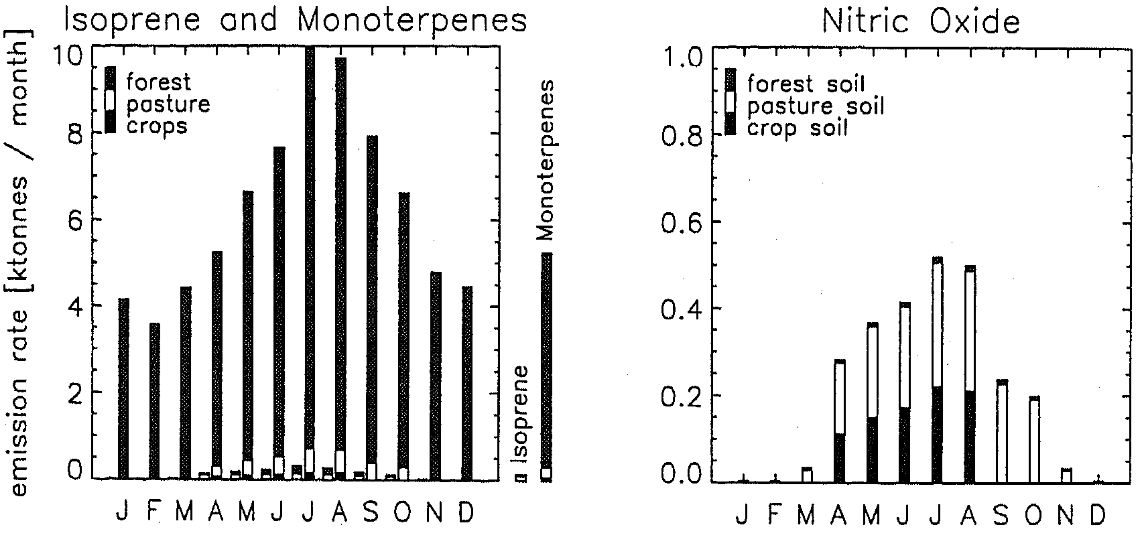

Fig. 5: Seasonal variation of the rate of isoprene, monoterpene and nitric oxide emissions by forests, pasture and crops.

4 Summary and Conclusions

The hourly emission rates of isoprene and monoterpenes from forests, crops and pasture as well as the soil emissions of nitric oxide were estimed for Switzerland, the grid cell size being 5 *km* x 5 *km*. The calculation was based on air temperature and solar radiation data which have been converted to leaf temperature and photosynthetically active radiation within the canopy. Taking into account emission factors and models given in the literature and the Swiss land-use inventory, hourly emission rates were computed for three consecutive days in summer 1993 as well as for monthly averages over ten years. The uncertainties were estimated to be 80 - 200%. Yearly totals of 1.5, 75 and 2.6 *ktonnes* y^{-1} were calculated for isoprene, monoterpenes and nitric oxide, respectively. Isoprene agrees well with the results obtained by EMEP [15] whereas a substantial discrepancy was found for nitric oxide, this probably due to different land-use data. Further investigations will elucidate the influence of biogenic species on the formation of photo-oxidants in the troposphere.

	biogenic emissions				anthrop. emissions	
	Isoprene	Monoter.	NO	NO_2 1)	VOC	NO_2
this work	1.5	75	2.6	4.0	-	-
Andreani and Keller [4,5,17]	3.2 2)	87	3.1	4.7	-	-
EMEP 5/95 [15]	1.4	-	8.1 3)	12.4 3)	305	183
BUS 75 [16]	3.0 4)	-	-	6.6	-	-

Table 1: Biogenic and anthropogenic emissions in in Switzerland [*ktonnes* y^{-1}]. 1) NO_2 emissions were converted from NO data, 2) without canopy model, 3) non-forested land areas, 4) VOC emitted by deciduous forests.

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