Present and future surface emissions of atmospheric compounds

EVK2-1999-00011

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Citation of this report: Olivier, J., J. Peters, C. Granier, G. Pétron, J.F. Müller, and S. Wallens, Present and future surface emissions of atmospheric compounds, POET Report #2, EU project EVK2-1999-00011, 2003.

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1. Introduction

Surface fluxes of chemical species are difficult to quantify on a global scale. They are difficult to measure directly, due to their large spatial and temporal variability. Over the past few years, high resolution global inventories have been developed within national, European or international projects. The more recent available inventory is characteristic of 1990 statistics. Within the project, emission distributions for the 1990-2000 period have been developed, that have been used in POET models for different activities, the model intercomparison (see the "Intercomparison of chemistry-transport models" report), the inverse modelling (see the "Optimisation of surface emissions by inverse modelling" report), the simulation of changes in the tropospheric composition over the past decade (see the "Variation of the atmospheric composition over the 1990-2000 period" report), the quantification of the global production of ozone (see the "Global ozone production" report).

Scenarios describing future emissions have been developed in the past, more particularly within IPCC, which describe the evolution of surface emissions of tropospheric constituents such as CO_2 and CH_4. Within POET, scenarios of the evolution of ozone precursors such as NO_x, CO and hydrocarbons, which are not included in the IPCC studies, have been developed**.** These scenarios take into account emission reduction policies which are consistent with European or international agreements.

This report describes the main characteristics of these inventories. The details on the development of the anthropogenic emission inventories for 1990-1995 are given in Olivier and Berdowski (2001) and Olivier *et al.* (2001). The documentation on the emissions files used in the models will be published in a MPI technical note in preparation, entitled "The POET 1990-2000 emissions database".

2. Anthropogenic emissions during the 1990-2000 period

2.1 Inventory for the 1990, 1995 and 1997 years

Version 3 of the EDGAR inventory for anthropogenic emissions has been developed at RIVM, which are typical of years 1990 and 1995. The methodology has been described in Olivier et al. (2001). Basically, activity data from international statistics are used, with data gaps filled by interpolation and extrapolation and emission factors for various source categories to calculation the annual emissions on a per country or per region basis. Next, national total emissions are, per source category, distributed on a 1x1 degree grid using appropriate grid maps for these sources. The EDGAR information system enables to extract the data at various aggregation levels. The inventories, for 1990, 1995 and 1997, by world regions have been first developed, and gridded inventories have been completed next, based on previous spatial distributions. The 1997 inventory has been compiled by combining the inventory for 1995 with regional trend data for various sources for 1995-1997 based on either activity data or reporting emission trends. Details on this approach are provided in Peters and Olivier (2003). Compared to the previous version 2 of the EDGAR inventory, the amounts of

agricultural waste burning have been revised substantially, based on recent insights, as shown in Figure 1.

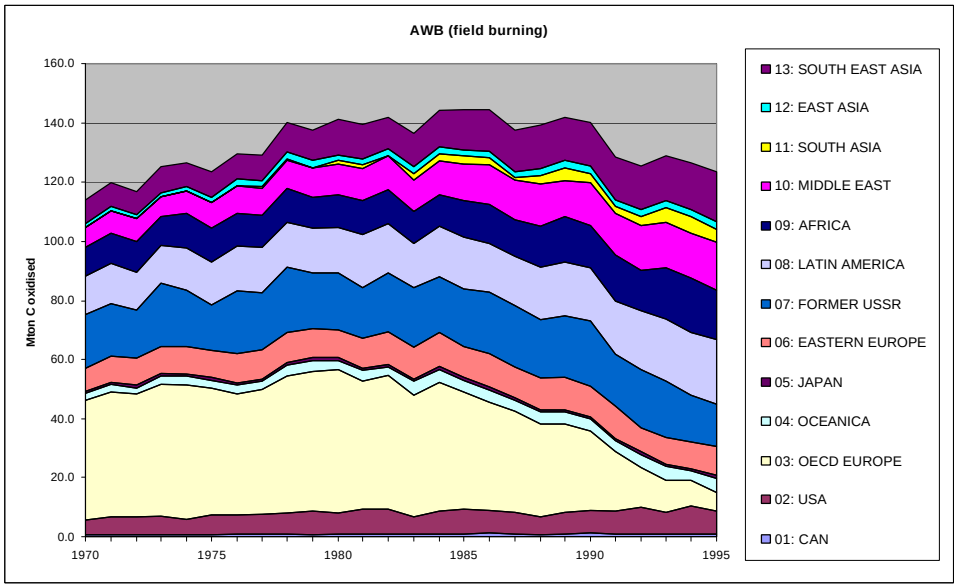


Figure 1. Changes in agricultural waste burning since 1975 for different areas of the world.

Vegetation fires in temperate climate regions are now also added in the inventory. Figure 2 shows the large variability of the vegetation fires in temperate regions.

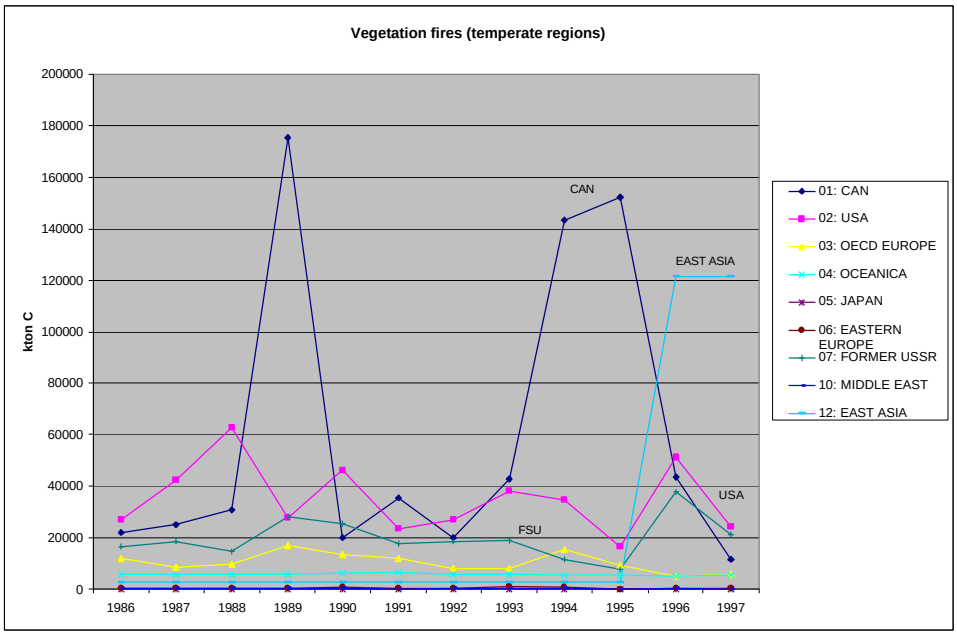


Figure 2. Trend in vegetation fires in temperate climate regions.

Globally the largest anthropogenic sources of CO are residential biofuel use and road transport (both 25%), savannah burning (20%) and tropical forest fires (10%). For NO_x the largest sources are road transport (25%), electric power generation (20%), industrial fossil fuel combustion and international shipping (both 10%), savannah burning (almost 10%), and residential biofuel use and cement production processes (both about 5%). For

total non-methane volatile organic carbon emissions (NMVOC) largest emissions were found to be road transport (about 20%), oil production/handling, residential biofuel use and solvent use (each about 15%), whereas savannah burning and temperate vegetation fires contribute another 5% each. However, these sources are not distributed equally over world regions, let alone individual countries. In OECD and Economies-In-Transition (former USSR, etc.) fossil-fuel use and industrial processes tend to dominate, while in other regions biomass burning, both large-scale as well as biofuel use, often is the largest source. Moreover, some of the largest sources tend to be concentrated in urban areas, e.g. road transport, fuel combustion by industry and power generation, others are most found in the rural area, as can be illustrated when looking at the emissions on a 1x1 degree grid. The shares of regions in the global total and those that determine the trend in the 1990s are given in Table 1, where the four world regions are used that were defined for the SRES scenarios of IPCC (see Section 4).

Table 1. Regional anthropogenic emissions in 1995 in EDGAR 3.2 and 1995-1997 trend (in Tg molecular mass; NO_x as NO_2).

Species	Emissions in 1995					Trend 1995-1997				
	OECD	EIT	ASIA	ALM	**Total**	OECD	EIT	ASIA	ALM	**Total**
CO	226	47	251	329	**854**	5%	-12%	1%	1%	**1.1%**
NO_x	46	12	30	27	**114**	2%	-6%	3%	7%	**2.7%**
NMVOC	55	18	37	51	**161**	2%	-13%	3%	3%	**0.8%**
CH_4	70	41	117	73	**301**	-2%	-8%	1%	2%	**-0.5%**

Note: Excluding natural sources; trend without tropical forest fires.
Definition of regions/correspondence with EDGAR 3 regions: OECD: OECD'90 (USA, CAN, W EUR, JPN, AUS, NZL); EIT: Economies-In-Transition (Reform regions) (former USSR and Eastern Europe); ASIA: South Asia, East Asia, Southeast Asia; ALM: Africa, Latin America, Middle East.

The reference year for the model work and intercomparison is 1997. The total emitted for the chemical species for 1997 included in the POET models are given in Table 2. Here, the emissions for biomass burning have been modified according to recent data compiled by Andreae and Merlet (2001) (see Section 2.2.2).

2.2 Uncertainty

It should be stressed that all emissions have an inherent uncertainty, since most are based on models rather than being measured for the largest sources. It was not feasible to go beyond the uncertainty tables compiled for EDGAR 2.0 (Table 3). However, in addition some conclusions on uncertainty could be drawn from the comparison of Version 2.0 with other datasets as part of the validation. The approach of interpreting uncertainty indications in terms of order-of-magnitude estimate of the uncertainty has been applied for methane and proved to be quite useful for application at global levels. The largest uncertainties in anthropogenic emissions are estimated for large-scale biomass burning, which may also vary substantially between years, biofuel use and for natural sources. Moreover, larger uncertainties appear in case of economies or technologies in transition (e.g. EIT, catalytic converter in cars); changes in emissions

can be large in those regions, even in a few years. In these cases accurate monitoring of recent trends proves to be difficult.

Table 2. Global annual emissions for 1997 (first version of EDGAR 3 and natural sources)

Global emissions from first version of Edgar v3 for year 1997						
	Total emission resulting from					
Species	anthropo-genic activities	forest burning	savanna burning	agriculture waste burning + fuelwood use	biogenics / soils	oceans
NOx (Tg N)	28.2	1.3	2.7	2.6	8	
CO (Tg CO)	336.4	119.2	150.6	247.9	160	20
CH4 (Tg CH4)	205.2	4.9	4.5	11.0		
C2H6 (Tg C)	3.1	2.3	0.7	2.5	0.8	0.8
C3H8 (Tg C)	4.9	0.4	0.2	1.0	1.6	1.0
C2H4 (Tg C)	2.1	4.0	1.9	4.8	4.2	1.2
C3H6 (Tg C)	0.9	1.3	0.6	2.3	0.9	1.3
isoprene (Tg C)					500	
terpenes (Tg C)					126.7	
acetone (Tg C)	0.6	1.0	1.1	0.1	15.0	
CH2O (Tg C)	0.3	1.8	0.4	0.4		
Other HCs (Tg C)	43.5	20.2	12.9	13.2		6.8

Table 3. Indication of uncertainty estimate for ozone precursors.

Main source	Sub-category	Activity	Emission factors			Global total and regional Emissions		
		data	CO	NO_x	NMVOC	CO	NO_x	NMVOC
Fossil fuel use	Fossil fuel combustion	S	M	M	M	M	M	M
	Fossil fuel production	S	-	-	M	-	-	M
Biofuel	Biofuel combustion	L	M	M	M	L	L	L
Industry/	Iron & steel production	S	M	M	L	M	M	L
solvent use	Non-ferro production	S	M	M	L	M	M	L
	Chemicals production	S	M	M	L	M	M	L
	Cement production	S	-	-	-	-	M	-
	Solvent use	M	-	-	M	-	-	M
	Miscellaneous	V			V			V
Landuse/	Agriculture	S	-	-	-	-	-	-
waste	Animals (excreta; ruminants)	S	-	-	-	-	-	-
treatment	Biomass burning	L	M	L	L	L	L	L
	Landfills	L	-	-	-	-	-	-
	Agricultural waste burning	L	L	L	L	L	L	L
	Uncontrolled waste burning	L	-	-	L	-	-	V
Natural	Natural soils	M	-	L	-	-	L	-
sources	Natural vegetation	M	M	-	L	M	-	L
	Oceans/wetlands	M	L	-	-	L	-	-
	Lightning	S	-	L	-	-	L	-
			CO	NO_x	NMVOC	CO	NO_x	NMVOC
All sources		-	-	-	-	M	M	L

Notes: Expert judgement of uncertainty ranges, which were assigned with the following classification in terms of order of magnitude of the uncertainty in mind: S = small (10%); M = medium (50%); L = large (100%); V = very large (>100%).
"-" Indicates that the compound is not applicable for this source or that emissions are negligible.

2.3 Temporal variation

For test-run purposes with several models a set of temporal (seasonal and diurnal) factors has been constructed for other anthropogenic sources (Figure 3a). This set is primarily based on (sometimes selected) Western European data. Therefore the representativity for other European countries and even more so for other world regions is highly uncertain. This provisional dataset is primarily meant for sensitivity assessments in atmospheric chemistry and climate models, awaiting a more comprehensive compilation and assessment of the relative importance for models. An example of have NOx emissions may be affected by seasonal variation of emissions is provided in Figure 3b, where the profiles for fossil fuel combustion sources have been aggregated to regional variation in total combustion emissions. Apparently, in the Northern Hemisphere regional emissions may vary up to 15% from a uniform distribution.

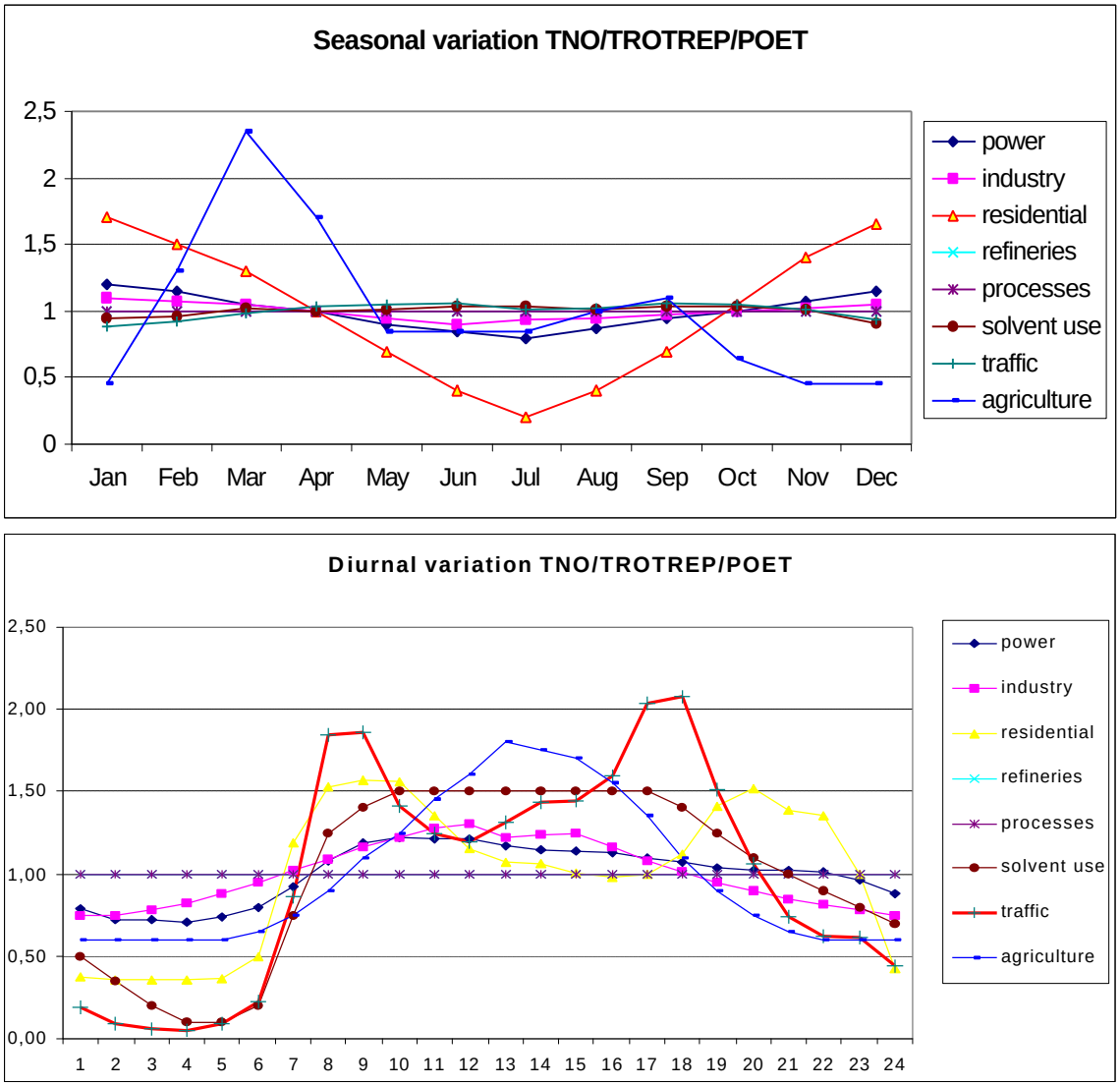


Figure 3a. Temporal profiles for different sources developed for test purposes: season (top) and diurnal (bottom) variation in TNO-TROTREP/POET profiles.

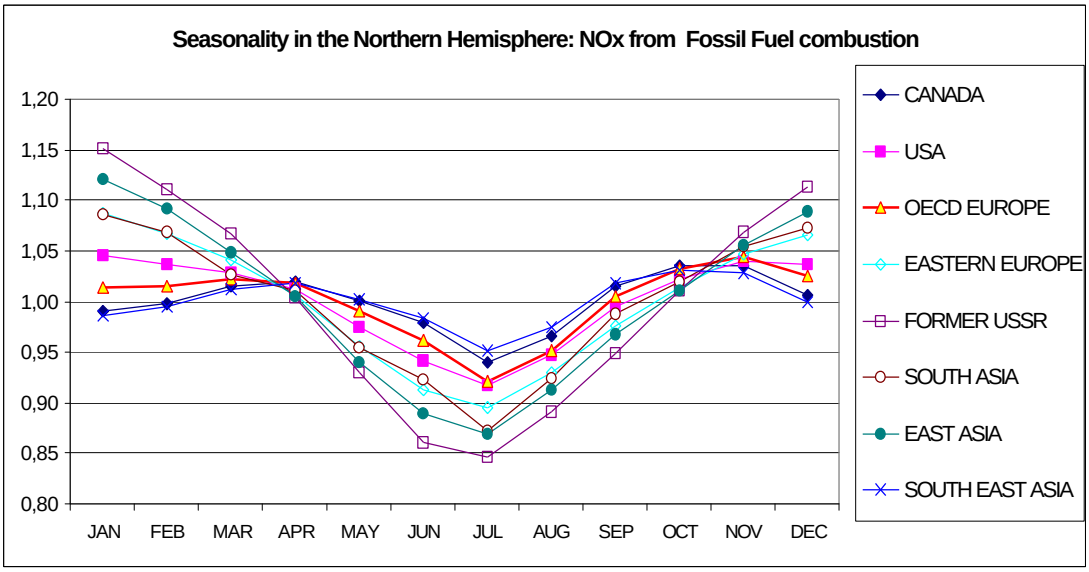


Figure 3b. Seasonality of regional NO_x emissions from fossil fuel combustion based on TNO-TROTREP/POET profiles.

2.4 Emission changes over the 1990-2000 period

2.4.1 Anthropogenic emissions

The emission dataset has been extended and formatted so that it can be used in a user-friendly manner by the POET models. Emissions for the 1990-2000 period have also been developed, using a linear extrapolation. This extrapolation has been performed for each type of emissions for each species. The changes in the emissions reflect the 1990-195and 1995-1997 changes, which are summarized in Table 4.

Table 4: Anthropogenic emission trends 1990-1995 and 1995-1997.

Period	CH_4 emissions	CO emissions	NO_x emissions
1990-1995	Global: 0% • Agriculture -0% • Waste handling +6% • Fossil fuel -4%	Global: -0% • Biomass burning +3% • Biofuel +7% • Fossil fuel -8%	Global: +2% • Fossil fuel -0% • Biofuel +9% • Biomass burning +4%
1995-1997	Global: +1% • former USSR (fSU): -9% • S Asia: +4%	Global: +1% • fSU and E Europe: -10% • OECD Europe: -7% • USA: +15%	Global: +3% • fSU: -7% • S Asia +16% • SE Asia: +9%

Period	NMVOC emissions
1990-1995	Global: +4% • Biofuel +7% • Biomass burning 10%; without TVF*: -5% • Fossil fuel +3%
1995-1997	Global: +0% • former USSR (fSU): -13% • Middle East +6% • SE Asia: +8%

* TVF = Temperate Vegetation Fires.

The totals emitted for different types of emissions are given in Table 5 for each year during the 1990-1999 period for carbon monoxide (5.a) and nitrogen oxides (5.b). The emissions have been grouped into two emission categories, technological and biofuel emissions.

Table 5a. Global anthropogenic CO emissions in 1990-1999 (Tg CO/year)

Source category	**1990**	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**
Fuel - industrial	4.136	4.140	4.144	4.147	4.151	4.155	4.032	3.909	3.885	3.908
Fuel - power	1.332	1.368	1.405	1.441	1.477	1.514	1.540	1.566	1.600	1.636
Fuel – other transf	0.269	0.260	0.251	0.242	0.233	0.224	0.230	0.237	0.233	0.230
Fuel – residential etc	52.53	49.53	46.53	43.53	40.53	37.53	35.28	33.02	30.56	28.62
Road transport	199.5	198.7	198.0	197.2	196.5	195.7	195.7	195.7	195.3	197.5
Non-road transport	20.29	19.44	18.58	17.73	16.87	16.02	14.75	13.48	12.58	12.42
International ships	0.091	0.093	0.095	0.098	0.100	0.102	0.012	0.102	0.104	0.106
Oil production	8.517	8.631	8.744	8.858	8.971	9.085	9.311	9.537	9.705	9.879
Coal fires	16.12	16.12	16.12	16.12	16.12	16.12	16.12	16.12	16.12	16.12
Iron and steel proc.	34.22	33.59	32.96	32.33	31.70	31.07	31.14	31.20	30.80	30.41
Aluminium proc.	0.072	0.073	0.075	0.076	0.078	0.079	0.080	0.081	0.082	0.084
Pulp and paper proc.	0.532	0.553	0.573	0.594	0.615	0.635	0.601	0.566	0.571	0.576
Waste incineration	3.630	3.670	3.710	3.750	3.790	3.830	3.818	3.806	3.831	3.856
Sum technological	**341.2**	**336.2**	**331.2**	**326.1**	**321.1**	**316.1**	**312.7**	**309.3**	**305.4**	**305.3**
Biofuel industrial	4.483	4.553	4.623	4.694	4.764	4.835	4.986	5.136	5.230	5.323
Biofuel power	0.239	0.248	0.258	0.267	0.277	0.287	0.287	0.288	0.295	0.302
Biofuel other transf	6.567	6.686	6.806	6.926	7.045	7.165	7.165	7.165	7.251	7.337
Biofuel residential	204.4	207.2	210.1	213.0	215.9	218.8	221.4	224.0	226.9	229.7
Biofuel transport	0.284	0.294	0.305	0.315	0.325	0.336	0.337	0.338	0.345	0.353
Sum biofuel	**215.9**	**219.0**	**222.1**	**225.2**	**228.3**	**231.4**	**234.2**	**237.0**	**240.0**	**243.0**

Table 5.b. Global anthropogenic NO_x emissions in 1990-1999 (Unit: Tg N/year).

Source category	**1990**	**1991**	**1992**	**1993**	**1994**	**1995**	**1996**	**1997**	**1998**	**1999**
Fuel - industrial	3.328	3.333	3.338	3.343	3.348	3.353	3.301	3.249	3.252	3.256
Fuel - power	7.127	7.121	7.115	7.109	7.103	7.097	7.219	7.341	7.372	7.414
Fuel – other transf	0.835	0.798	0.761	0.725	0.688	0.651	0.661	0.670	0.675	0.681
Fuel – residential etc	1.019	1.010	1.000	0.990	0.980	0.970	0.946	0.921	0.908	0.897
Road transport	8.413	8.403	8.393	8.383	8.373	8.363	8.636	8.909	8.982	9.063
Non-road transport	1.548	1.513	1.479	1.444	1.409	1.374	1.335	1.295	1.259	1.236
International ships	2.607	2.671	2.734	2.798	2.862	2.925	2.925	2.925	2.970	3.015
Oil production	0.063	0.075	0.087	0.098	0.110	0.122	0.125	0.128	0.138	0.147
Coal fires	0.084	0.084	0.084	0.084	0.084	0.084	0.084	0.084	0.084	0.084
Iron and steel proc.	0.292	0.290	0.288	0.286	0.284	0.283	0.286	0.289	0.289	0.288
Chemistry proc.	0.230	0.228	0.225	0.223	0.221	0.218	0.215	0.211	0.208	0.207
Pulp and paper proc.	0.043	0.045	0.047	0.048	0.050	0.052	0.052	0.052	0.053	0.054
Cement proc.	1.043	1.096	1.150	1.203	1.257	1.310	1.359	1.408	1.460	1.516
Waste incineration	0.041	0.042	0.042	0.043	0.043	0.044	0.044	0.045	0.045	0.046
Sum technological	**26.67**	**26.71**	**26.74**	**26.78**	**26.81**	**26.84**	**27.18**	**27.52**	**27.79**	**27.90**
Biofuel industrial	0.142	0.145	0.147	0.150	0.153	0.156	0.161	0.166	0.169	0.172
Biofuel power	0.113	0.120	0.128	0.136	0.143	0.151	0.151	0.152	0.157	0.163
Biofuel other transf	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003	0.003
Biofuel residential	1.887	1.914	1.941	1.968	1.995	2.022	2.046	2.071	2.097	2.123
Biofuel transport	0.014	0.014	0.015	0.015	0.016	0.016	0.016	0.016	0.017	0.017
Sum biofuel	**2.16**	**2.20**	**2.23**	**2.27**	**2.31**	**2.35**	**2.38**	**2.41**	**2.443**	**2.478**

2.4.2 Biomass burning emissions over the 1997-2001 period

Satellite fire products, which give the location of fires at a high spatial resolution, have the potential to construct inter-annual time series of fire activity, but the quantification of biomass burning emissions using the satellite products remains challenging. We have built an inventory of emissions resulting from tropical forest and savanna fires using the fire counts from the Advanced Thermal Scanning Radiometer (ATSR) satellite. The redistribution of the emissions from the distribution of fire pixels was done in several steps. First, an annual integral of CO_2 emissions for forest and savanna fires using the Hao and Liu (1994) climatological biomass burning emissions is computed. Then, the annual average of fire counts from ATSR using the full 1997-2001 record is computed for forest and savannas. A monthly average of CO_2 emissions over each 1x1 degree grid point is calculated, using the monthly integrated fire counts over the grid point and the corresponding annual average. The CO_2 emission for each month over the observation period as a result of forest and savanna fires is shown in Figure 4, together with a comparison of the value provided by EDGAR 3.

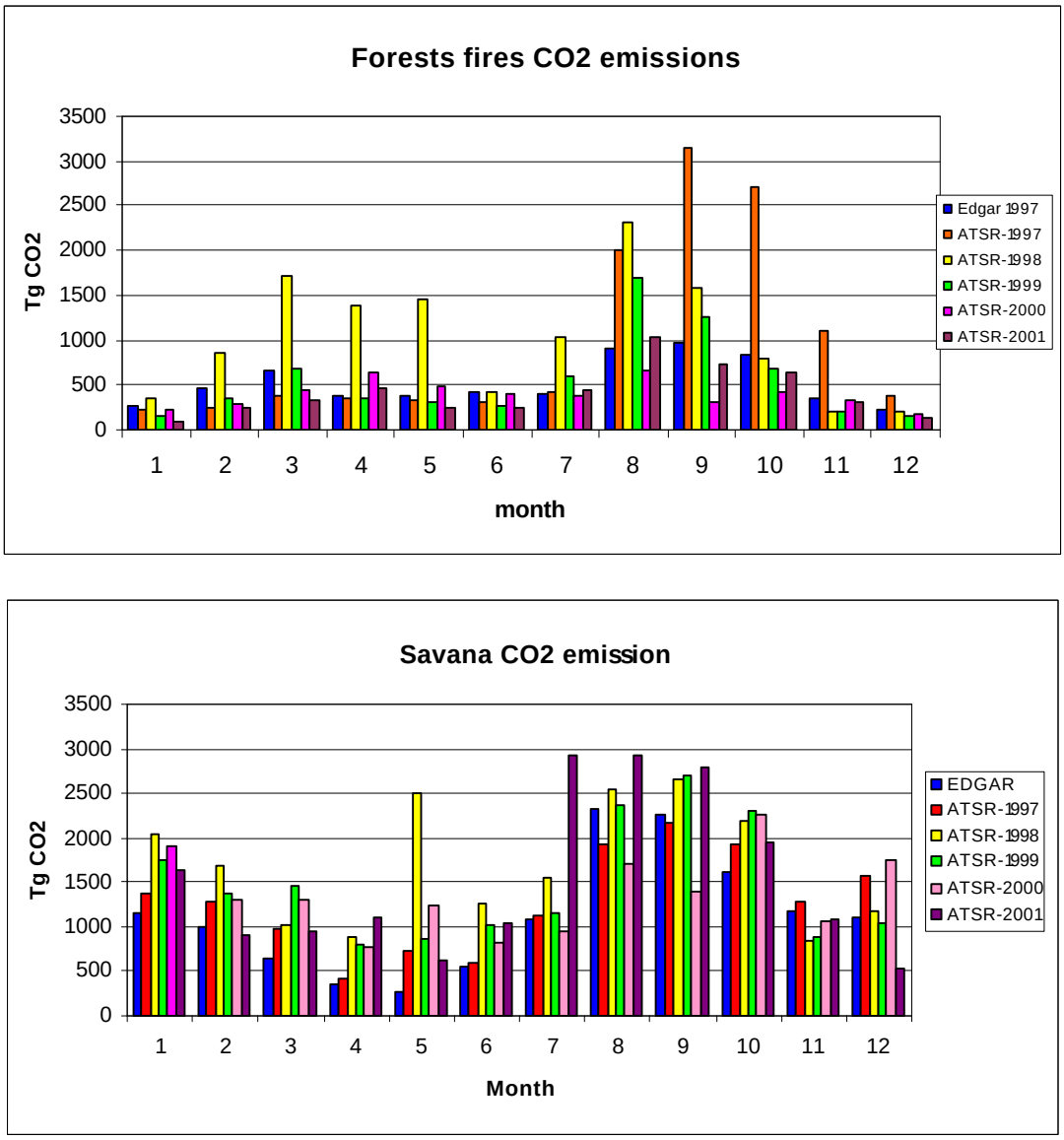


Figure 4: CO_2 emissions from tropical forest (top) and savanna (bottom) fires base on ATSR fire counts and comparison with the EDGAR 3 dataset.

The emissions of each chemical species considered in the models is then computed using the emission ratios provided by Andreae and Merlet (2001), and listed in Table 6.

Table 6: Emission ratios to CO_2 expressed as molecule per molecule of CO2/C for different types of burning.

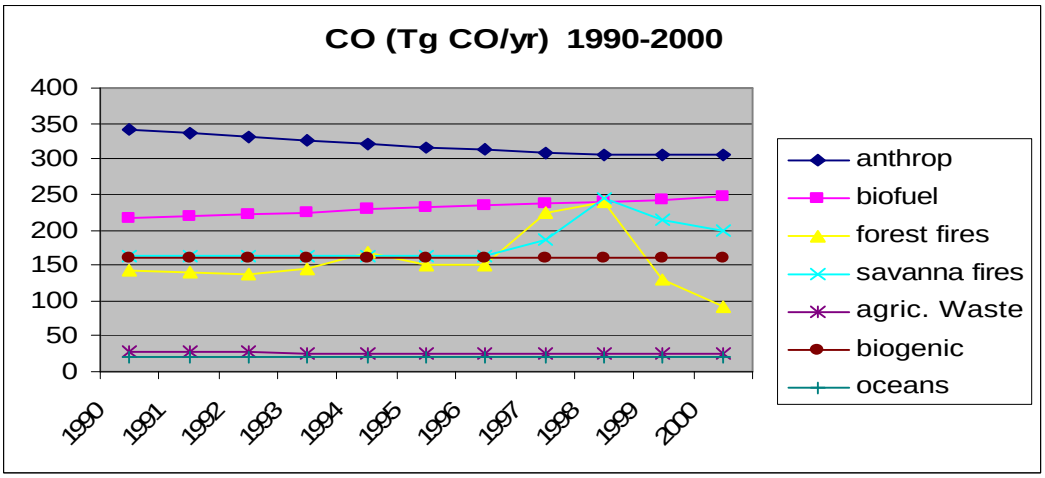
	Deforestation	Savanna burning	Agricultural waste	Non-tropical fires
	molec./molec(C)	**molec./molec(C)**	**molec./molec(C)**	**molec./molec(C)**
CH_4	1.13E-2	3.82E-3	4.5E-3	7.8E-3
CO	0.099	0.0619	0.088	0.102
NO_x (as NO_2)	1.41E-3	3.47E-3	2.22E-3	2.66E-3
C_2H_6	1.07E-3	2.84E-4	8.64E-4	5.32E-4
C_3H_8	9.0E-5	5.45E-5	3.16E-4	1.15E-4
C_2H_4	1.86E-3	7.54E-4	1.33E-3	1.07E-3
C_3H_6	3.49E-4	1.66E-4	6.34E-4	3.74E-4
CH_2O	8.89E-4	2.23E-4	8.39E-4	1.4E-3
Acetone	2.85E-4	2.03E-4	2.9E-4	2.52E-4
Other HC[(1)]	6.25E-3	3.75E-3	4.5E-3	8.75E-3
NH_3	2.04E-3	1.64E-3	2.04E-3	2.19E-3
SO_2	2.38E-4	1.46E-4	1.67E-4	4.16E-4

[(1)]IMAGES/MOZART species

[a] Obtained by assuming that TNMVOC = the sum of all NMVOCs listed in Table 1 of Andreae and Merlet (2001).

2.4.3 Summary

The full emission dataset has been extended and formatted so that it can be used in a user-friendly manner by the POET models. The changes over the 1990-2000 period of the global emissions for CO, nitrogen oxides and one hydrocarbon, ethane (C_2H_6) are shown in Figure 5.



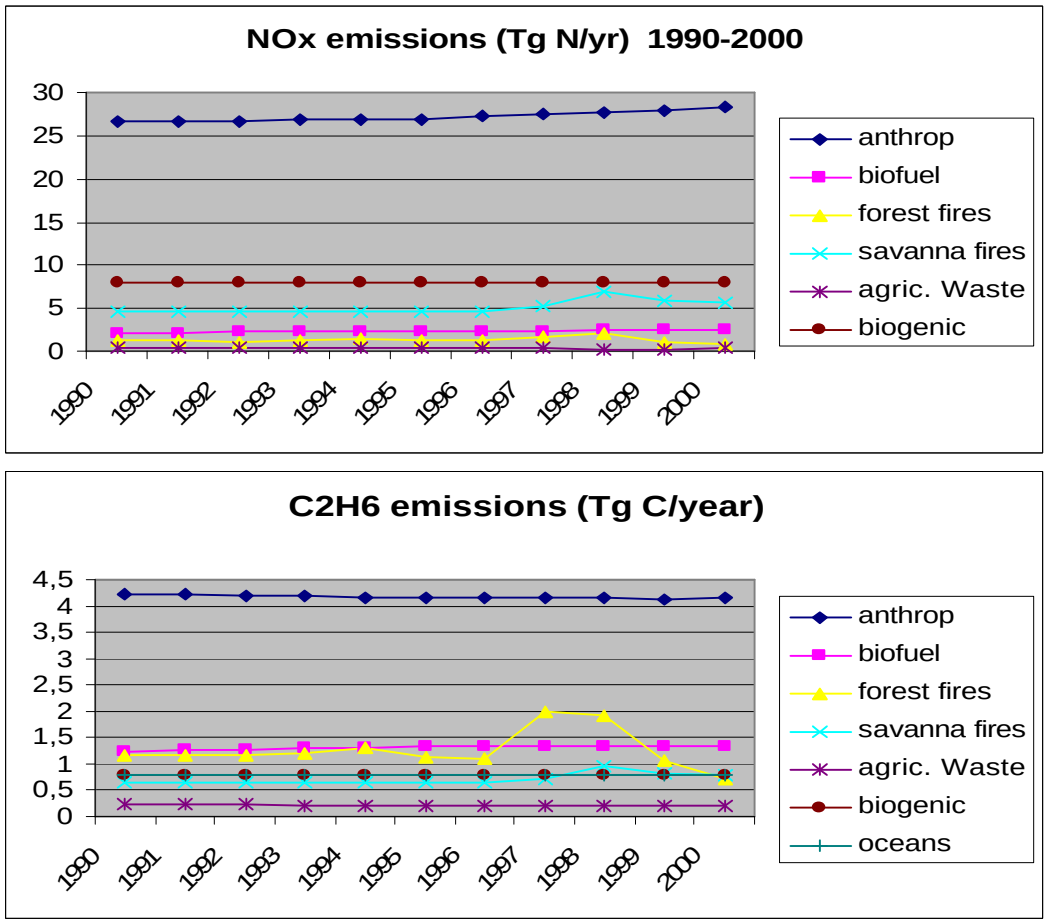


Figure 4: CO_2 emissions from tropical forest (top) and savanna (bottom) fires base on ATSR fire counts and comparison with the EDGAR 3 dataset.

3. Natural emissions

The geographical distribution and seasonal variation of the biogenic emissions are adapted from the GEIA inventory (for nitrogen oxides, isoprene and terpenes) and from Müller and Brasseur (1995) (for carbon monoxide).

A detailed vegetation canopy model has been developed at IASB for the calculation of isoprene and terpenes emissions. This multi-layer model calculates the transfer of visible and near infra-red radiation in the canopy as well as the temperature of the leaves. It has been used in order to evaluate the uncertainties in the emission estimates by comparisons of the model predictions with campaign flux measurements at a number of sites: a mixed forest in the North-eastern US, two savanna sites in Southern Africa, and a forest in Amazonia. The model/data biases are typically a factor of two, except at the Amazonian site, where the model overpredicts the isoprene flux measurements by a factor between 5 and 10. This work is highlighted in the poster displayed below, which was presented at the EU symposium in Torino in September 2001.

Validation of the BVOC emission fluxes - Development of a Canopy Environment Model

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Introduction

Guenther et al., 1995 estimated that vegetation is the source of over 90% (1150 Tg C yr-1) of all non-methane hydrocarbons (NMHC) in the global atmosphere. These estimates, however, are still very crude due to uncertainties and simplifications in the emission algorithms and vegetation information used in the model calculations. The forest canopy is a complex environment which controls the flux of the volatile organic compounds to and from the atmosphere. NMHC emission rates from vegetation are species specific and landscape emissions are therefore dependent on the species composition and abundance in a landscape.

In addition, the biogenic volatile organic compounds (BVOC) are emitted from trees at rates dependent upon the leaf level microclimate, in particular on leaf temperature and Photosynthetically Active Radiation (PAR) (Guenther et al., 1993, 1995). For this reason, accurate emission estimates require detailed modelling of the forest canopy influence.

Our specific aims are 1) to provide a new model with an accurate description of the canopy microclimate, in particular, the processes that determine the radiation fluxes and leaf temperature at different heights in the canopy and 2) to test, and possibly improve, the emission algorithms in globally important biomes by the means of comparisons with campaign measurements.

We evaluate the performance of this model using above canopy flux measurements from tropical savannas (Maun and Skukuza sites in Southern Africa from the SAFARI2000 campaigns), tropical rainforests (Tapajos in Brazil from the LBA campaign) and temperate forests (Harvard Forest, Massachusetts). We present model estimates of isoprene and/or monoterpenes fluxes for these different vegetation types and compare with relevant above canopy measurements from field campaigns.

Model description

BVOCs fluxes estimates are

- either light and temperature dependent (as isoprene, MBO, sometimes monoterpenes) (Guenther *et al.*, 1993, 1997)
  "isoprene algorithm" (G97)
- or only temperature dependent (as usually monoterpenes) (Guenther *et al.*, 1993)
  "monoterpene algorithm" (G93)

BVOCs fluxes are calculated using a multiple-level canopy model. In each layer, PAR and leaf temperature are determined. A detailed description of the solar radiation transfer (Goudriaan and van Laar, 1994) has been implemented. PAR and NIR are determined using adequate extinction coefficients (κ).

The diffuse fraction of the solar radiation in the canopy takes into account the radiation transfer in the atmosphere (including cloud effects and Rayleigh diffusion), as well as the scattering by the leaves.

The effect of canopy shading is included: the total leaf area is divided into sunlit and shaded fractions. The solar radiation calculated for each layer allows the estimation of leaf temperature by solving the energy balance equation at the leaf level, taking into account the leaf - environment exchange of latent and sensible heat fluxes as well as thermal radiation.

Fields campaign description

We present here the model results using above canopy flux measurements from 3 different biomes:

- Temperate forest (Harvard Forest, 1995 campaign in Massachusetts) (Goldstein et al., 1998). The main tree species in this forest is oak, considered as high-isoprene emitter, while other plant genera present in this ecosystem are low-isoprene emitters. Isoprene fluxes are computed on the June 15- September 15 period.
- Tropical savannas (Skukuza and Maun sites in Southern Africa from the SAFARI2000 campaigns). The Skukuza site is an acacia and combretum savanna. Isoprene fluxes for this site are relatively low and hence are close to the detection limit.
  The Maun site is described as Mopane woodland. The Mopane woodland forms large areas of monospecific stands of *Colophospermum mopane* (90% of the vegetation). Note that the monoterpene (α-pinene) emissions from Maun are light and temperature dependent.
  Biogenic hydrocarbons emissions were conducted during the wet season in 2001
- Tropical rainforest (Tapajos 2000 in Brazil from LBA campaign). The site is situated on the upper plateau, planalto. The forest at the site is primary terra firme rainforest, with closed canopy structure and emergent trees. The measurements of isoprene, α-pinene and β-pinene fluxes were conducted between 6 and 8 July 2000.

For all campaigns, the meteorological conditions measured at the canopy top are used as input to the model.

Results and discussion

Harvard Forest (Massachusetts):

In Fig.1, modeled isoprene fluxes are seen to be systematically underestimated compared with measurements when the standard emission rate in G97 is deduced from vegetation composition and standard emission factors from the literature. We attribute this difference to an underestimation of either the standard emission factors used for oaks or the proportion of oaks (or other high-isoprene emitters) in the surrounding vegetation. It is also calculated that using air temperature (instead of leaf temperature) in equation G97 results in ~ 5 % higher emission estimates (average over the growing period). Sensitivity tests were performed showing that the isoprene emission estimates are not much influenced by possible errors in physical parameters such as the extinction coefficient, the leaf scattering coefficient, the fraction of diffuse PAR.

Fig.2 shows a comparison of our modeled emissions to a simple 8-level canopy model that uses air temperature in G97 and a single PAR extinction coefficient in the following equation:

$$PAR_z = PAR_{top}\ EXP(-\kappa\ LAI_{cum})$$

Where PAR_z is PAR at the height z in the canopy
PAR_{top} is PAR above the canopy
κ is the extinction coefficient for the radiation
LAI_{cum} is the cumulative LAI from the canopy top to the height z

The different values of κ for the simple model are:
κ = 0.55 deduced from the in-canopy PAR measurements and a LAI = 5.0
κ = 0.42 from the literature
κ = 0.79 deduced from the in-canopy PAR measurements and a LAI = 3.5 obtained from measurements.

We have a good agreement between the micro-climate canopy model (using LAI = 3.5) and the simple 8-level model using a κ of 0.79 consistent with the measurements. This result indicates that the micro-climate canopy model reproduces correctly the extinction of the radiation inside the canopy observed at this site.

The long-term measurements dataset obtained at Harvard Forest allows to test a new isoprene algorithm (Guenther *et al.*, 1999) (G99).
The changes appear in: the light factor which varies with the canopy depth
the temperature factor which depends on the temperature that the leaf has been exposed to during the past 15 days.

From a first analysis of the model results at Harvard Forest, it was not possible to conclude whether the new isoprene algorithm G99 is better than G97. Further campaigns will be necessary to get more reliable conclusions.

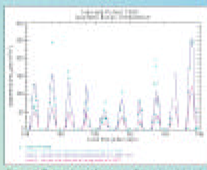

Figure 1: Comparison of isoprene fluxes (model and measurements) at Harvard Forest. Isoprene fluxes are estimated using either leaf or air temperature in G97.

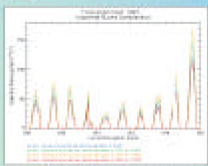

Figure 2: Comparison of isoprene fluxes using 2 different models at Harvard Forest.

Skukuza (Kruger National Park - Republic of South Africa):

Fig.3 shows the diurnal variation of the isoprene emissions using G97 and environmental conditions such as air temperature and PAR. The isoprene fluxes are seen to increase with air temperature and PAR, in agreement with the model. Nevertheless, the measured emissions seem to rise more abruptly and also later in the morning. Despite the low values of the isoprene flux measurements, the model and the measurements have comparable orders of magnitude. Further data would be required to provide a more reliable interpretation of the experimental results.

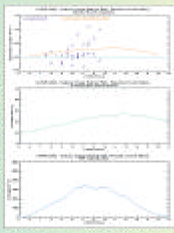

Figure 3: Diurnal variation of isoprene fluxes and environmental parameters at Skukuza site.

Maun (Botswana):

Fig.4 shows the diurnal variation of the α-pinene fluxes (model and measurements) and environmental parameters. The model uses:
1) either leaf or air temperature in the algorithms, and
2) either the standard isoprene algorithm (G97) or a modified version of this algorithm (G97 adapted) using parameters values determined from field and laboratory studies.

In all cases, the model agrees reasonably well with the observations.

The fig.5 and fig.6 shows the comparison of the light (C_L) and temperature (C_T) factors in the standard (G97) and modified (G97 adapted) isoprene algorithms. The most pronounced change appears in the C_T factor (fig.6). As the air temperature at the Maun site falls into the range of 25°-35°C, the new C_T results in a mean diurnal curve more symmetrical around noon, in better agreement with the observations.

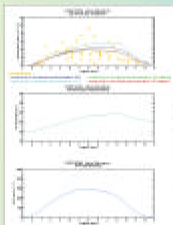

Figure 4: Diurnal variation of α-pinene fluxes and environmental parameters at the Maun site.

Figure 5: Light factor (C_L) in the isoprene algorithm G97 and G97 adapted to the Maun site.

Figure 6: Temperature factor (C_T) in the isoprene algorithm G97 and G97 adapted to the Maun site.

Tapajos (Brazil):

Fig.7 shows the isoprene fluxes (measurements and model). The measurements are obtained using 2 different techniques (FIS and DEA: see Rinne *et al.*, 2002 for the details) and the model used the isoprene G97 algorithm with leaf temperature. Although the diurnal cycle seems to be reproduced, the model overestimates the emissions fluxes by a factor 5.5 (in comparison with FIS data) to 10 (in comparison with DEA data). The standard emission rate is therefore probably too high at the Tapajos site.

Fig.8 and fig.9 show the monoterpenes fluxes (measurements and model) for α-pinene (50% of the total observed monoterpenes flux) and β-pinene (17.5%), respectively. The fluxes are measured using the DEA technique and the model used either the isoprene (G97) or the monoterpene (G93) algorithm as suggested by the observations. The fluxes are seen to be dependent either on light and temperature or on temperature only. For both α-pinene and β-pinene, the G97 model underestimates the observed fluxes by about a factor of 2, but the diurnal cycle seems to be roughly reproduced. The G93 model fluxes are higher than those obtained with G97 and the diurnal cycle is wrong.

In conclusion, the model using G97 seems appropriate to reproduce the monoterpenes fluxes, but the standard emission rate is too low.

In order to provide a more definitive assessment of the algorithm performance at the Tapajos site, the measurements conducted in April 2001 will be analysed in our future work.

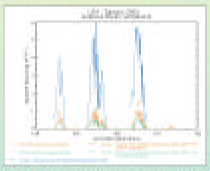

Figure 7: Isoprene fluxes comparison (measurements (FIS and DEA techniques) and model) at the Tapajos site.

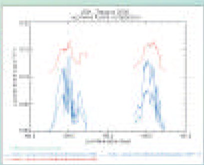

Figure 8: α-pinene fluxes comparison (measurements (DEA technique) and model) at the Tapajos site. The modelled fluxes used either G97 algorithm or G93 algorithm.

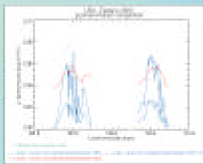

Figure 9: β-pinene fluxes comparison (measurements (DEA technique) and model) at the Tapajos site. The modelled fluxes used either G97 algorithm or G93 algorithm.

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Acknowledgements

The authors are grateful to:
A. Goldstein, B. Munger, D. Parker, G. Pilgerwald, F. Wigneron and K. Moore for their help in the analysis of Harvard Forest data.
L. Otter for site descriptions and meteorological data from Skukuza, J. Greenberg for the analysis of the VOC data and Elmar Veenendaal for meteorological data from Maun.
J. Rinne for his help concerning the Tapajos site.

4. Future emissions for the period up to 2020.

Different scenarios have been evaluated, based on scenarios prepared for the IPCC Special Report on Emission Scenarios (SRES) (IPCC, 2000). These scenarios include four groups of scenarios, all quite different in their regional and global assumptions with respect to economic development, cooperation and environmental and technological changes. The scenarios result in a highly different development of future anthropogenic emissions in different regions (i.e., large emission increases in low latitude developing countries, decreases in emissions in northern industrialized areas). Natural emissions have been assumed to stay constant.

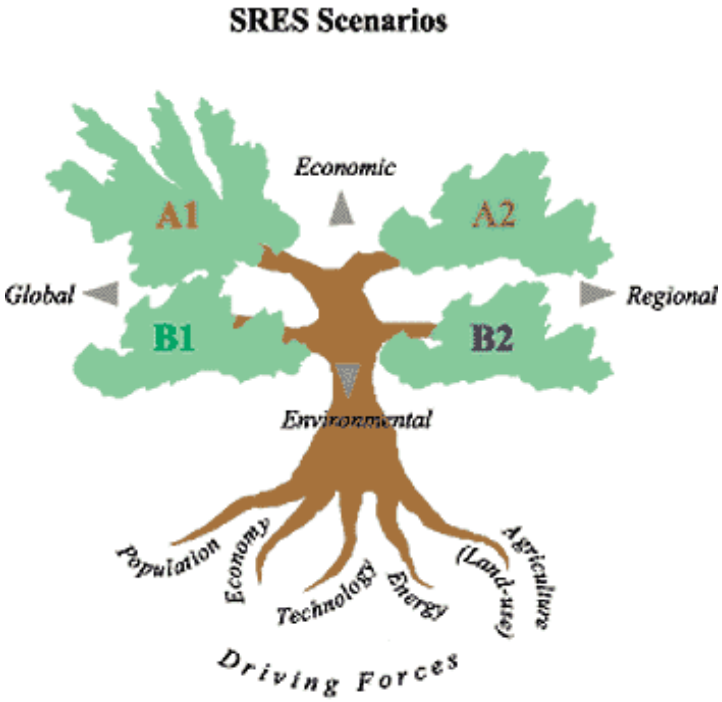

Figure 6 : Driving forces for the IPCC/SRES scenarios

Different scenarios can be envisaged from the SRES package. These four scenarios can be described as follows (see Figure 6):

- A1: very rapid economy growth, introduction of new technology, low population growth;
- A2: very heterogeneous, high population growth, fragmented and slower technological changes;
- B1: convergent world, changes towards sustainability, same low population growth as A1;

- B2: changes towards local sustainability, moderate population- growth, less rapid economical and technological changes than in A1 and B1.

The different assumptions on the main driving forces are summarized in Figure 7, whereas the different impacts of these scenarios on global total and regional anthropogenic emissions of ozone precursors are presented in Figures 8 to 10. As illustrated in Figure 7, apart from changes in growth rates of population and income, the main changes between the #1 and #2 scenarios that determine the trend in emissions are the introduction of new (control) technologies. As indicated by Figures 9 and 10 the use of clean technology is assumed to occur in all regions, but to different degrees in different scenarios.

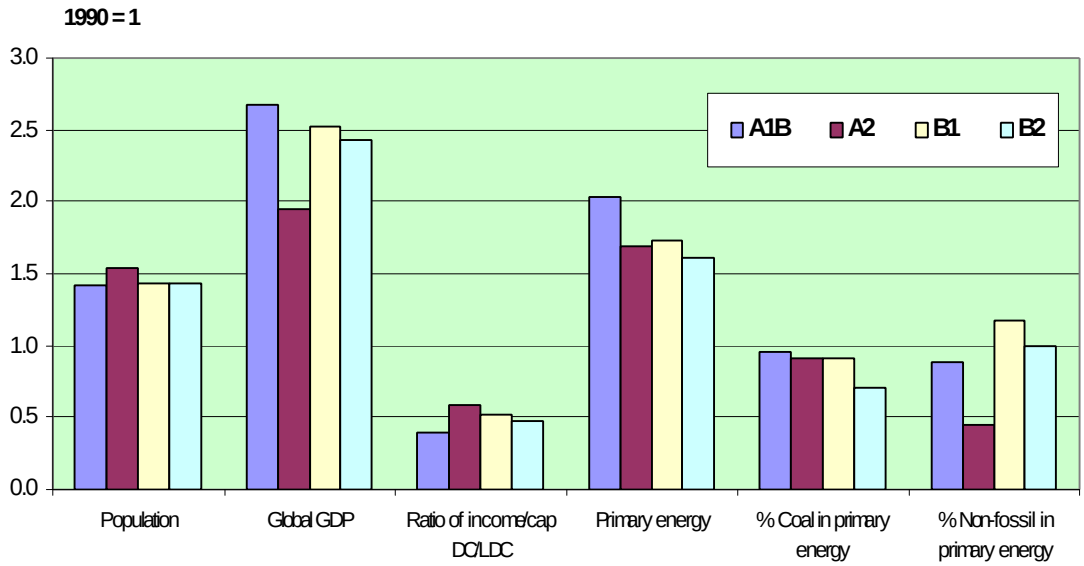

Figure 7 : Main driving forces in the four marker scenarios: index of 2020 values

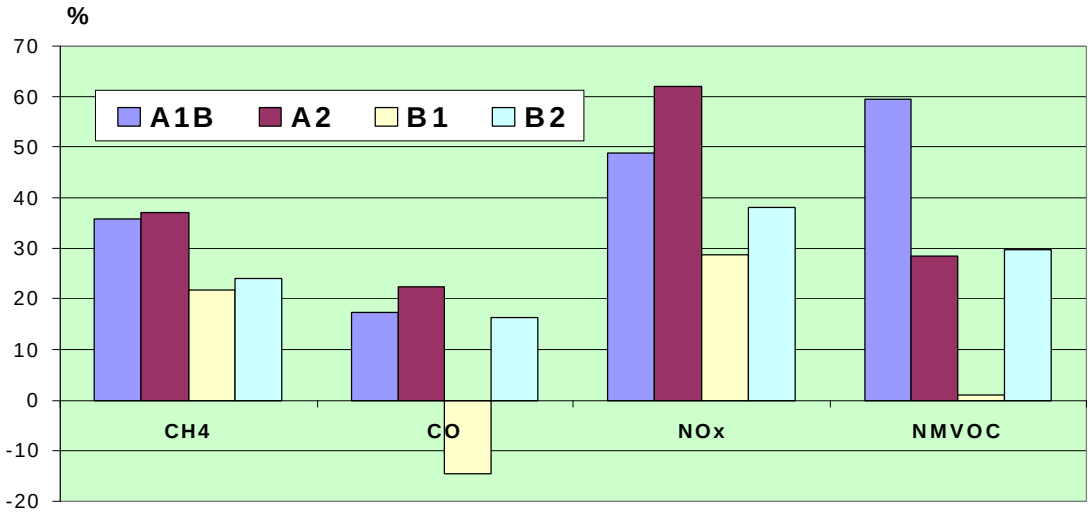

Figure 8: Global changes of anthropogenic emissions during the 1990-2020 period (in %) for the four SRES scenarios.

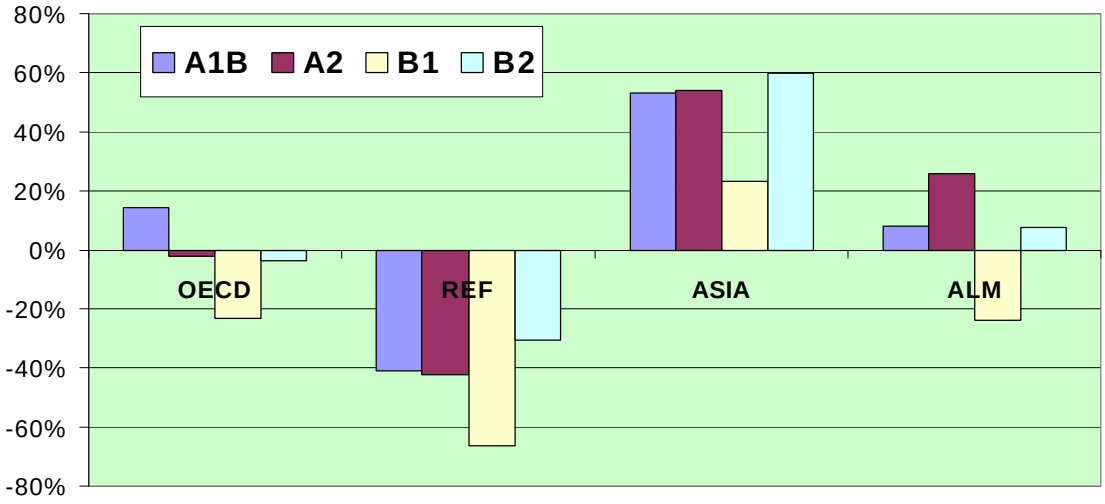

Figure 9 : Anthropogenic CO emission changes (in %) between 1990 and 2020 for 4 scenarios and 4 different regions of the world.

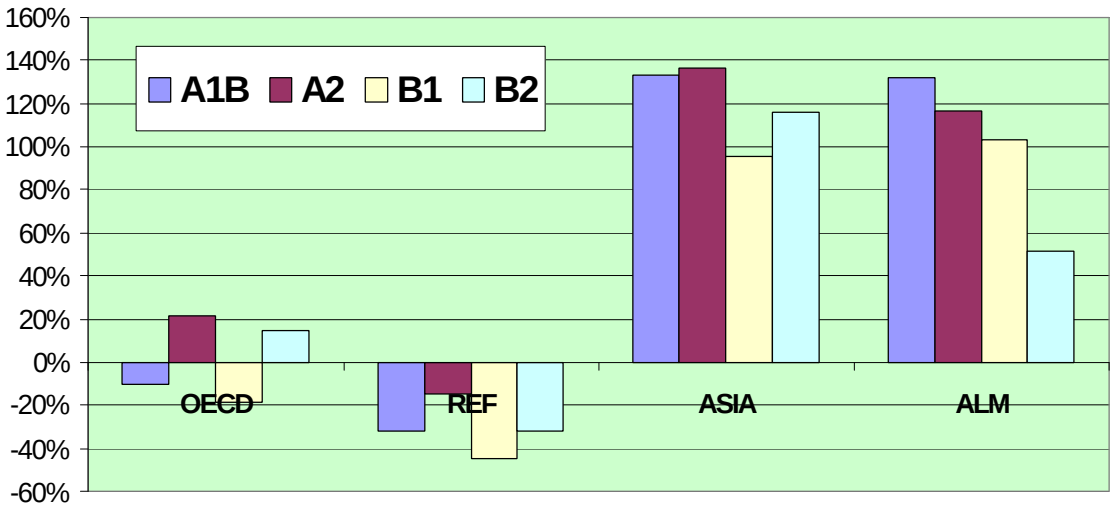

Figure 10: Anthropogenic NO_x emission changes (in %) between 1990 and 2020 for 4 scenarios and 4 different regions of the world.

The long-term relative changes in emissions of CH_4, CO, NO_x and hydrocarbons is presented in Figure 11 for different areas of the world, as indicated in Table 7.

Table 7 : List of the regions used in the EDGAR inventory and in Figure 11

Region number	Region name	Region number	Region name
1	Canada	9	Africa
2	USA	10	Middle East
3	OECD Europe	11	South Asia
4	Japan	12	East Asia
5	Oceania	13	South East Asia
6	Eastern Europe	14	Greenland
7	Former USSR	15	Antarctica
8	Latin America	16	Oceans

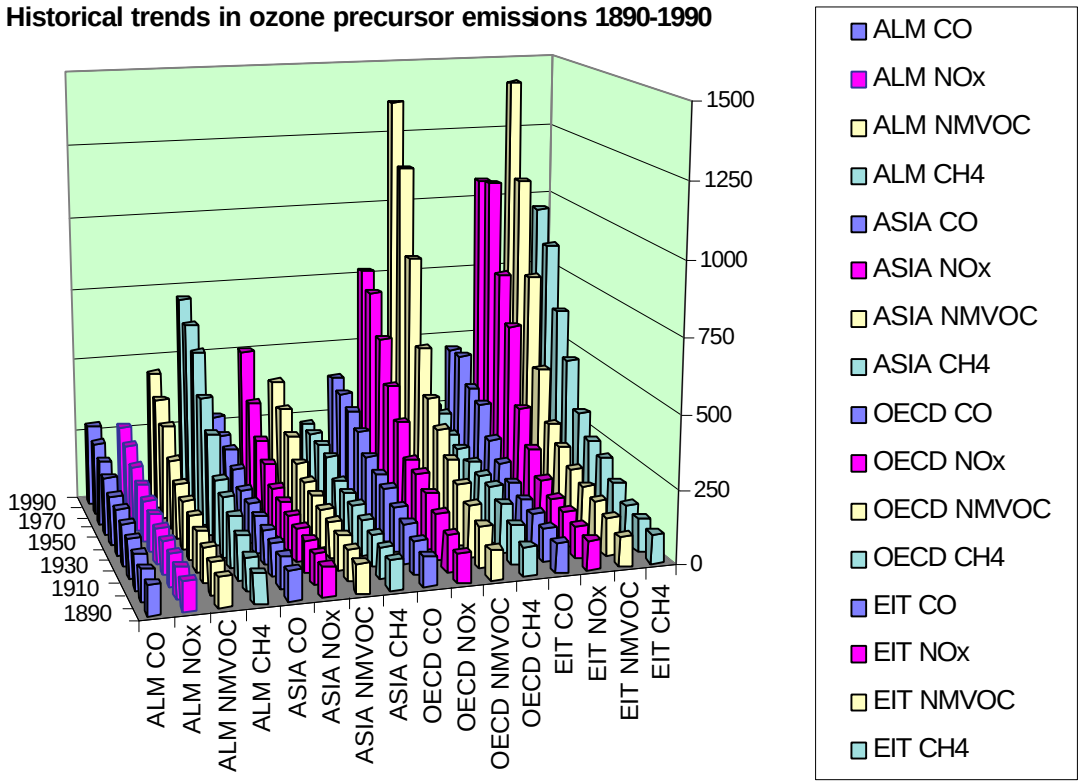

Figure 11: Long term historical trend in regional anthropogenic ozone precursor emissions 1890-1990; index 1890 = 100 (for definition of regions see Table 1) (source: Van Aardenne et al., 2001).

The methodology for creating emission scenarios on grid is very similar to the compilation of the gridded 1997 datasets (see Section 2.1). Per EDGAR standard source category and for each of the 13 continental EDGAR 3 world regions (see Table 7), the

emission trends were calculated relative to the 1995 emissions of the scenarios for each five year; for intermediate years the trend has been interpolated. Next, these region-specific and source-specific indices for 1995-2020 were applied as multipliers to the gridded 1995 emissions per source category, resulting in gridded emission patterns that have been scaled per each of the 13 world regions.

For these gridded emission scenarios, RIVM's IMAGE 2 implementation of the SRES scenarios have been used, since these contain more detailed sources and regions thereby producing a more consistent, higher-resolution scenario than would be possible when using only the four SRES regions and main SRES source categories. Moreover, a large part of the non-energy source emissions in the SRES report have been based on IMAGE model results. The IMAGE 2 model is an integrated model for assessing the effects of human activities on global change (IMAGE team, 2002).

An analysis made of the global and regional ozone precursor trends in the four so-called marker scenarios, led to the selection of the A2 and B1 scenarios as *priority scenarios* for gridding to and including 2020. This was done as they differ most from the present emission levels (A2) or provide the most contrasting trends between compounds (B1). In addition to the regional scenarios, we had to make projections for international transport since these were not explicitly considered in the SRES scenarios. The development of regional fuel consumption for international air and water transport was based on the average elasticity to GDP observed for OECD and non-OECD regions in the period 1970-2000. Table 8 shows the calculated global total trends. Assuming no substantial changes in the emission factors, these indices were then used to scale the 1995 gridded emissions with a globally uniform factor. Natural emissions have been assumed to stay constant in the next 25 year time period.

Table 8: Trends in international shipping and aviation in the A2 and B1 scenarios (1995 = 100)

Source	Scenario	1995	2000	2005	2010	2015	2020
International shipping	A2	100	105	110	114	120	126
	B1	100	104	109	114	118	123
Aviation	A2	100	112	126	138	153	168
	B1	100	112	124	136	148	161

As shown in Figure 8, global emissions of CO, NOx and NMVOC in 2020 are in the B2 scenario 30% or more lower than in the A2 scenario. For CO this results in an absolute decrease compared to 1990 for all world regions except Asia (Figure 9). For NO_x in the B1 scenario a decrease over time was calculated for OECD and EIT countries (Figure 10). Figures 12 and 13 show the changes in the emissions of major sources of CO and NO_x from 1995 to 2020 for the A2 scenario. The B1 scenario assumes continuing globalization and economic growth, and a focus on the environmental and social - immaterial - aspects of life. This in combination with regulations and financial incentives leads to a rapid decline in energy- and material-intensive economic activities as people become more open to the need for and relevance of more sustainable forms of development. Governments can effectively solve environmental problems using

regulation, ecotaxing and burden-sharing as the main policy instruments. There is a rapid diffusion of resource-efficient technology (IPCC, 2000; IMAGE team, 2002).

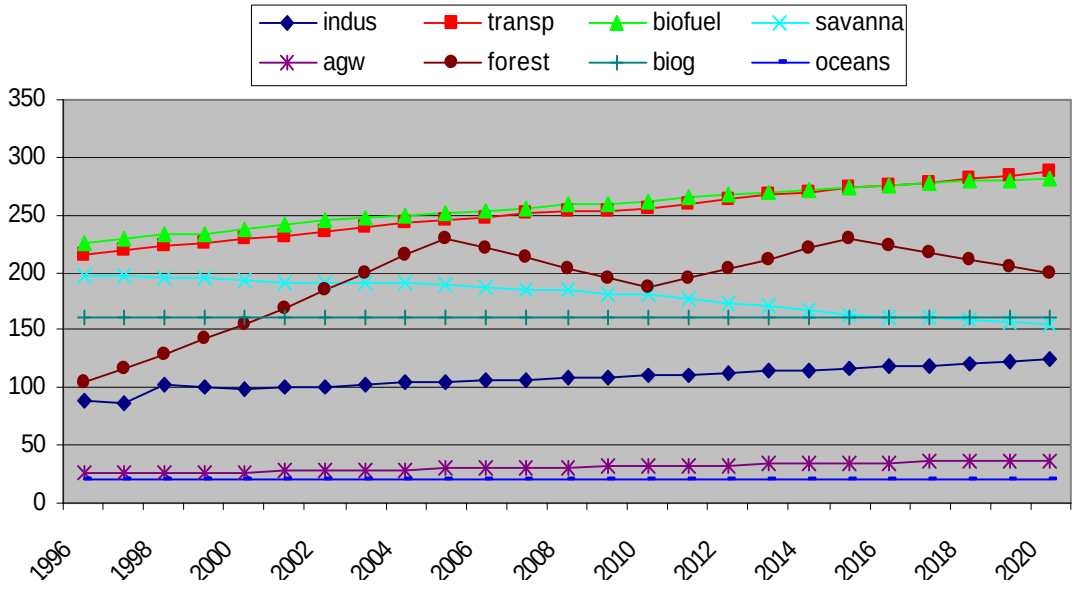

Figure 12 : Change in the CO anthropogenic surface emissions for the 1995-2020 period for the A2 scenario.

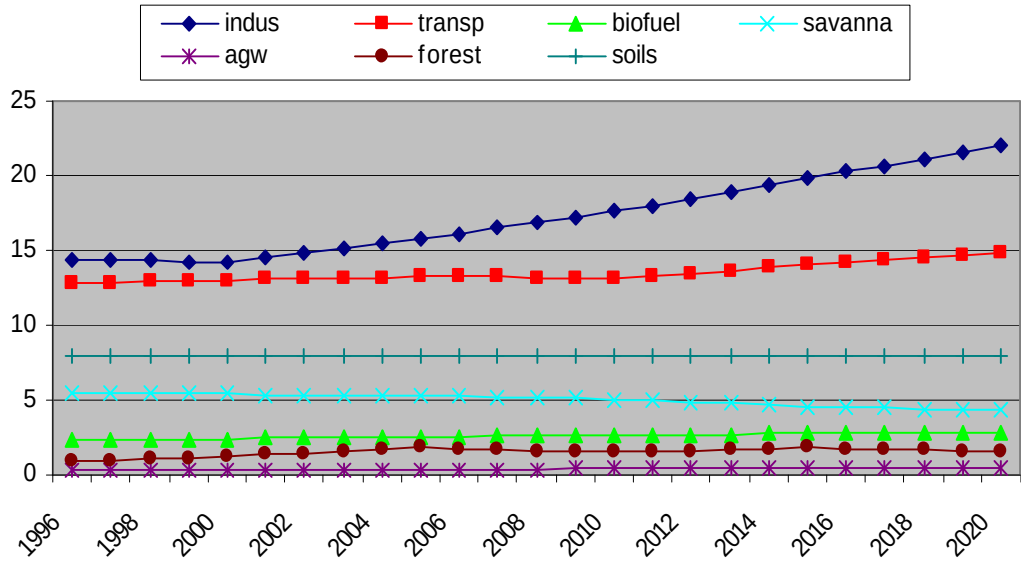

Figure 13 : Change in the NO_x anthropogenic surface emissions for the 1995-2020 period for the A2 scenario.

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Abstracts presented at international meetings

Development and Evaluation of a Canopy Environment Model for Regional and Global Predictions of Biogenic VOC Emissions, by S. Wallens, J.F. Müller and A. Guenther, International workshop on "Emissions of Chemical Species and Aerosols into the atmosphere", June 2001, Paris, France.

Biogenic VOC emissions are very sensitive to leaf temperature and light intensity. Thus an accurate description of canopy microclimate is a necessary component of a biogenic VOC emission model. Predicting emissions at regional and global scales is challenging because of the need to specify general canopy characteristics for relatively broad vegetation types. We present an improved canopy model that computes leaf temperature and light intensity for multiple vertical layers in a vegetation canopy. Leaf temperature is calculated by energy balance and a detailed radiation transfer model is employed. The model is parameterized for seven basic growth forms.
We present fluxes estimates of isoprene, sensible heat, and latent heat for the seven basic growth forms and compare these measurements with relevant above canopy measurements from previous field studies. In addition, we present a model sensitivity analysis and discuss priorities for future model refinement.

Global emissions of ozone precursors: sources, trends and uncertainties, by Jos G.J. Olivier, presented at the Workshop 'Air Pollution as a Climate Forcing', Honolulu, USA, 29 April-3 May 2002

Global anthropogenic sources of ozone precursors CO, NO_x, NMVOC and CH_4 have been estimated for the period 1990-1997 based on national activity data (e.g. kg fuelwood combustion per year), emission factors (emissions per unit of human activity, e.g. kg CO/kg fuelwood combusted), and grid maps for spatial allocation of the emissions within a country (e.g. human population density). The results of a comprehensive study to estimate global emissions and their trend will be discussed briefly. Globally the largest anthropogenic sources of CO are residential biofuel use and road transport (both 25%), savannah burning (20%) and tropical forest fires (10%). For NO_x the largest sources are road transport (25%), electric power generation (20%), industrial fossil fuel combustion and international shipping (both 10%), savannah burning (almost 10%), and residential biofuel use and cement production processes (both about 5%). For total non-methane volatile organic carbon emissions (NMVOC) largest emissions were found to be road transport (about 20%), oil production/handling, residential biofuel use and solvent use (each about 15%), whereas savannah burning and temperate vegetation fires contribute another 5% each. However, for modelling of the atmospheric chemistry its the most reactive compounds within total NMVOC that are important. Many of the key ones are being emitted by the largest sources, but not all. However, these sources are not distributed equally over world regions, let alone individual countries. In OECD and Economies-In-Transition (former SU etc.) fossil-fuel

use and industrial processes tend to dominate, while in other regions biomass burning, both large-scale as well as biofuel use, often is the largest source. Moreover, some of the largest sources tend to be concentrated in urban areas, e.g. road transport, fuel combustion by industry and power generation, others are most found in the rural area, as can be illustrated when looking at the emissions on a 1x1 degree grid. The shares of regions and sources in the global total and those that determine the trend in the 1990s will be discussed. In addition the uncertainty of the emission estimates will be discussed. The largest uncertainties are estimated for large-scale biomass burning, which may also vary substantially between years, biofuel use and for natural sources. Moreover, larger uncertainties appear in case of economies or technologies in transition (e.g. EIT, catalytic converter in cars); changes in emissions can be large in those regions, even in a few years. In these cases accurate monitoring of recent trends proves to be difficult. Subsequently, the recent regional emission trends 1990-1997 will be compared to the longer historical trend 1890-1990 as well as with four scenarios for future emissions for the period 1990-2020 developed by the IPCC. Historical trends of precursor emissions are quite different per region. It is concluded that future development of precursor emissions may show quite different characteristics in terms of size and sign of the growth but also the development could be quite different per compound.

Publications during the full duration of the project

Peer Reviewed Articles:

Authors	Date	Title	Journal	Reference
Van Aardenne, J.A., F. Dentener, J.G.J. Olivier, C.G.M. Klein Goldewijk, and J. Lelieveld	2001	A 1^{o}x1^{o} resolution dataset of historical anthropogenic trace gas emissions for the period 1890-1990	Global Biogeo-chemical Cycles	15, 909-928
Wotawa, Gerhard, P. Novelli, M. Trainer and C. Granier,	2001	Inter-annual variability of summertime CO concentrations in the Northern Hemisphere explained by boreal forest fires in North America and Russia	Geophys. Res. Lett.	28, 4575
Endresen O., J. K. Sundet, E. Soergaard, S. Dalsoeren, G. Gravir, T. F. Berglen and I. S. A. Isaksen	2003	Emission from international sea transportation and environmental impact	J. Geophys. Res	108, D17, 4560, doi:10.1029/2002JD002898

Non refereed literature:

Authors / Editors	Date	Title	Event	Reference
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Olivier, J.G.J., and J.J.M. Berdowski	2001	Global emissions and sinks	in "The Climate System", Eds. Berdowski, Guicherit and Heij , ISBN 90 5809 255 0	pp 73-78, Balkema Publishers /Swets & Zeitlinger, NL
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S. Dalrosen, I.S.A. Isaksen, J.K. Sundet and G. Myhre	Sept. 2001	The contribution of biomass burning to the global ozone budget and oxidation of chemical compo	EU Symposium: A changing atmosphere, Torino, Italy
Wallens, S., J.-F. Muller, and A. Guenther	Sept. 2001	Development of a canopy environment model for new biogenic VOCs emissions inventories	EU Symposium: A changing atmosphere, Torino, Italy
Olivier, J.G.J.	April 2002	Global emissions of ozone precursors: sources, trends and uncertainties.	Air Pollution as a Climate Forcing: A Workshop, Hawaii, USA
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J. Hoelzemann, G. P. Brasseur, C. Granier, and M.G. Schultz	Sept 2002	Quantifying global biomass burning emissions for chemistry transport modeling: a new inventory for MOZART	7th IGAC/CACGP Conference, Hersonissos, Greece
Olivier, J.G.J.	Oct 2002	Input data and uncertainties in emission inventories: Who cares? - Why and how should it be addressed?	CLRTAP International Workshop on Validation and Evaluation of Air Emission Inventories, Gothenburg, Sweden
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