

Abschlußbericht zum Forschungs- und
Entwicklungsvorhaben 298 41 252 auf dem Gebiet des
Umweltschutzes „Modellierung und Prüfung von Strategien
zur Verminderung der Belastung durch Ozon“

The development of an emission data base over Europe and further contributions of TNO-MEP

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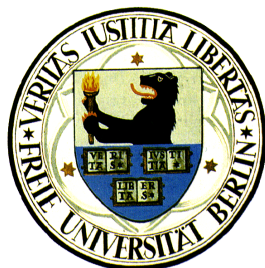
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16. Abstract The report describes the work carried out by TNO, Netherlands: a) Development of a emissions data base for dispersion modeling. Base year is 1995. The data base covers Europe with grids of 0.5 x 0.25 lat long and contains the anthropogenic emissions of SO ₂ , NO _x , NMVOC, CO, CH ₄ , NH ₃ , PM ₁₀ , PM _{2.5} .The data base uses CORINAIR and CEPMEIP information and the SNAP source group classification of CORINAIR. b) Development of the PELINDA land use data base derived from satellite information c) Review of surface resistance approaches for dry deposition d) Review of biogenic emissions parameterizations		
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The development of an emission data base over Europe and further contributions of TNO-MEP

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Summary

In this report an overview is given of the activities performed by TNO as subcontractor of the Free Univ. Berlin in the framework of the UBA-project "Modellierung und prufung von Strategien zur Verminderung der Belastung durch Ozon". A CD-ROM goes along with this report containing all data generated in this project. The activities started by Nov. 1, 1999, and ended by May 1, 2002. The activities were focussed on:

- The development of an emission data base over Europe for 1995, on grids of 0.25×0.5 lat-lon for the species SO_2 , NO_x , NMVOC, CO, CH_4 , NH_3 , PM_{10} and $\text{PM}_{2.5}$, including a land-use data base.
- Assistance in the improvement of the Atmospheric Chemistry Transport Model on the subjects aerosol modelling, biogenic emissions, boundary conditions, dry deposition, and consultancy concerning data-assimilation.

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1 General Introduction

In the project-proposal by the Institut für Meteorologie der Freie Universität Berlin dated October 1998 are the main items of the project described.

The main aim is to develop and apply a model system, an Atmospheric Chemistry Transport-ACT/ Chemistry Transport Model-CTM, for the calculation of long term effects on the eco-system due to ozone. The key parameter mentioned is AOT-40, accumulated ozone over threshold, in this case the threshold is 40 ppb. The critical level for the protection of crops is 3000 ppbh, May-July, daylight hours, and 10.000 ppbh for forests, April-September, daylight hours.

As a second aim it is mentioned that the model system should also be suited to address the EU-directives for SO₂, NO_x, Ozone and PM₁₀.

The area to be addressed is the European scale and the national scale. A link with the regional (Länder) and local (urban, street level) must be possible.

Essential is that the model-system must be able to perform model runs over long time periods of several years, in view both of the EU-directives and the AOT-40. This requirement of long term leads to a model concept of intermediate complexity. The model needs to contain all relevant processes, but a simplified way.

The two tasks of TNO in this project were the establishment of emission - and land use data, and the assistance in improving the model system.

The different elements for model improvement were partly based on a consultancy study entitled " Overview and analysis of international activities in the field of photo-oxidant modelling (Buitjes, April 1998)

The main focus in the beginning of the project was on the possibilities and limitations to model AOT-40, and to try to compare the calculated values with observations of AOT-40 (see the presentations of Nov. 1999, Febr. 2000, referenced at the end of this report).

Gradually, the focus in the project shifted from ozone to NO_x, and mainly to PM₁₀.

There are two reasons for this.

First, the concept of AOT40 became more and more a subject of debate. The discussion were whether AOT-40, without for example taking relative humidity into account, was the appropriate method to determine damage to eco-systems, or that a flux approach was more useful. Also the large sensitivity in the parameter itself, both in modelling and observations created concern. So, the attention moved away from AOT-40.

Second, there was an increasing concern about the possible exceedance of the guidelines for PM₁₀ (and also NO₂) as given in the EU-directives. The state of knowledge, both in observations and in modelling, concerning PM₁₀ is much less advanced than for ozone.

As a consequence, after the middle of 2000 most attention in the project has been given to aerosols and more specific PM_{10} .

This is also reflected in the tasks performed in the project.

During the course of the project regular contacts have been maintained with the international community. Of special importance are the contacts with EUROTRAC-2, with the subproject GLOREAM-Global and regional atmospheric modelling.

These contacts make it possible to stay in touch with the newest developments in the field, and to discuss own progress in an international forum.

At this moment-June 2002- both the REM-3 and the LOTOS model participate in two model intercomparisons which are currently underway in EUROTRAC and will be finalised by the end of 2002.

A continental scale model intercomparison for ozone focuses on the analysis of trends in ozone as observed, and modelled.

An other continental scale model intercomparison brings for the first time the few European models together which can model aerosols. The focus is on an analysis of calculated and observed aerosol-concentrations over 1995.

The following overview of the results of the tasks performed by TNO is divided in a description of the model input: anthropogenic emissions, land use data base, biogenic emissions and boundary conditions; and a description of model internal processes: aerosols, dry deposition.

Finally, the consultancy for data-assimilation is mentioned.

The report finishes with conclusions and an overview of presentations given in the frame of this project.

2 Improvement of the input to the Atmospheric Chemistry Transport (ACT)-model

2.1 European wide anthropogenic emissions

2.1.1 Introduction

The task of TNO in this project was to create an emission data base for the year 1995 for the so-called PHOXA-REM3-area, with a geographic grid system with a resolution of 0.25×0.5 latitude longitude. The database should contain annual emissions of NO_x , SO_x , NH_3 , NM-VOC, CO, CH_4 and PM_{10} and $\text{PM}_{2.5}$ for the SNAP level 1 categories 1-10 in the CORINAIR classification. For the gaseous species, the emission database should be consistent with the official information for the year 1995 in the CORINAIR data base, version 2.2, as available since October 1999 at the Topic Centre Air Emissions. PM emissions are not officially reported (yet) by the countries. The gridded emissions of PM_{10} and $\text{PM}_{2.5}$ are derived from results obtained in the CEPMEIP-project (see also subsection 2.1.5 on CEPMEIP in this Chapter).

Concerning the CORINAIR database, versions 2.2, two problems were identified. First, not all countries have delivered data for 1995. In these cases the official data for 1994 was used. Second, in CORINAIR a revision of the reporting conventions has taken place, from the so-called SNAP94 to the SNAP97 reporting convention. In SNAP94 emissions from forests are reported under category 10, whereas in SNAP97 they are reported under category 11. Some countries only reported their total emissions at SNAP level 1 and hence no information on the subcategories (i.e. at SNAP level 2) is available in these cases. In Table 2.1 an overview is given of the reporting convention per country reporting to CORINAIR. It is specified whether a country has reported under the SNAP94 or SNAP97 convention, from which base year the emission totals were taken (generally 1995, only 1994 if no 1995 data was available). It is also indicated whether the countries have provided SNAP level 2 information for category 10, the total NM-VOC emissions in category 10 and the reported biogenic VOC emissions under category 10, when applicable.

Table 2.1 Reporting convention, base year of the emissions data used for the gridding and details on the reporting of the NM-VOC emissions from managed forests.

Country code	Reporting convention	Base year of emission data used in this project	Subdivision of emissions in category 10 provided?	NM – VOC reported in category 10	Biogenic NM-VOC reported in category 10
AT	SNAP94	1995	Yes	126	124
BE	SNAP94	1994	Yes	20	19
BU	SNAP94	1995	No	34	n.a.
CH	SNAP94	1995	Yes	8	0
CZ	SNAP94	1995	n.a.	0	0
DE	SNAP94	1994	Yes	386	386
DK	SNAP97	1995	Yes	1	0
EE	SNAP94	1995	n.a.	0	0
ES	SNAP94	1994	No	79	n.a.
FI	SNAP94	1995	n.a.	0	0
FR	SNAP97	1995	Yes	20	0
GB	SNAP94	1995	Yes	40	40
GR	SNAP94	1995	Yes	893	845
HR	SNAP94	1995	n.a.	0	0
HU	SNAP94	1994	No	0	0
IE	SNAP94	1995	Yes	82	26
IS	SNAP94	1995	n.a.	0	0
IT	SNAP94	1995	Yes	163	152
LU	SNAP94	1995	Yes	1	1
MT	SNAP94	1994	n.a.	0	0
NL	SNAP97	1995	n.a.	0	0
NO	SNAP94	1995	n.a.	0	0
PL	SNAP94	1995	Yes	34	0
PT	SNAP94	1995	Yes	346	342
RO	SNAP94	1994	Yes	49	0
SE	SNAP94	1995	Yes	389	389
SI	SNAP94	1995	n.a.	0	0
SK	SNAP94	1995	n.a.	0	0

The national totals emissions per country and emitted compound are listed in Table 2.2. For the countries not reporting to CORINAIR, information from the LOTOS90 database is used.

Table 2.2 Yearly emissions totals (in kt/year) for 1995 from the TNO emissions data base. Countries not reporting to CORINAIR are in italic.

Country code	country	CEPMEIP		Totals based on CORINAIR v2.2 and the LOTOS90 database (<i>italic</i>)					
		PM ₁₀	PM ₂₅	CH ₄	CO	NH ₃	NM-VOC	NO _x	SO ₂
ALB	<i>ALBANIA</i>	8	6	66	161	31	62	52	24
ARM	<i>ARMENIA</i>	7	5	0	0	0	0	0	0
AUT	AUSTRIA	45	32	580	1147	79	283	176	60
AZE	<i>AZERBAIJAN</i>	27	18	0	0	0	0	0	0
BEL	BELGIUM	73	46	602	1515	96	316	350	252
BGR	BULGARIA	93	38	506	844	100	141	266	1497
BIH	<i>BOSNIA</i>	10	6	119	249	25	69	364	54
BLR	<i>BELORUSSIA</i>	63	39	1738	2091	73	901	467	246
CHE	SWITZERLAND	16	11	313	510	60	211	136	34
CYP	<i>CYPRUS</i>	2	1	0	0	0	0	0	0
CZE	CZECH REP.	126	57	733	874	137	286	412	1091
DEU	GERMANY	305	189	4848	6797	623	2145	2266	2998
DNK	DANMARK	29	19	428	588	99	131	250	149
ESP	SPAIN	213	147	2259	4645	344	1080	1185	2008
EST	ESTONIA	33	14	43	343	12	60	48	123
FIN	FINLAND	30	22	245	436	35	186	260	96
FRA	FRANCE	352	254	2642	8421	795	2174	1726	947
GBR	GREAT BRITAIN	229	135	3817	5478	324	2257	2295	2365
GEO	<i>GEORGIA</i>	10	7	0	0	0	0	0	0
GRC	GREECE	62	42	465	1418	110	450	356	609
HRV	CROATIA	21	14	103	479	25	72	55	63
HUN	HUNGARIA	59	33	603	767	62	148	190	883
IRL	IRELAND	22	13	813	304	124	149	115	161
ITA	ITALY	291	204	2555	7755	461	2216	1849	1322
LTU	<i>LITHUANIA</i>	20	13	625	752	26	324	115	108
LUX	LUXEMBURG	5	2	22	105	7	17	20	9
LVA	<i>LATVIA</i>	13	9	423	509	18	219	93	88
MDA	<i>MOLDOVA</i>	16	10	730	878	31	379	108	60
MKD	<i>MACEDONIA</i>	27	10	91	190	19	52	95	30
MLT	MALTA	1	1	6	24	0	6	16	16
NLD	NETHERLANDS	55	33	1171	892	147	365	498	147
NOR	NORWAY	47	41	487	739	26	366	214	34
POL	POLAND	315	127	1803	4548	379	770	1122	2276
PRT	PORTUGAL	50	36	816	1290	96	247	366	358

ROM	ROMANIA	183	90	809	2924	204	477	342	1064
RUS	RUSSIA	1429	736	20604	22173	1035	9498	2336	53
SVK	SLOWAKIA	37	20	313	400	30	41	181	238
SVN	SLOVENIA	13	7	166	91	22	42	67	119
SWE	SWEDEN	38	25	276	877	59	303	341	82
TUR	TURKEY	391	205	0	0	0	0	0	0
UKR	UKRAINE	609	282	8671	10430	362	4495	1894	1287
YUG	YUGOSLAWIA	145	49	487	1021	104	282	156	155

In Appendix A the national total emissions per SNAP level 1 categories for each of the emitted compounds are given.

2.1.2 The source categories

At the beginning of the project the emission data in use contained the source-category split-up as developed in the previous PHOXA-LOTOS projects. Since these categories are not consistent with the categories used in EMEP and CORINAIR, within this project the source category split has been modified to become consistent with CORINAIR. The CORINAIR categories and the relation with the former categories used in PHOXA/LOTOS are given in Table 2.3. The CORINAIR categories in Table 2.3 are the so-called SNAP level 1 categories. Only for traffic a further subdivision into three subcategories has been made within this project, because of the relative importance of traffic emissions. Also the VOC split (necessary input for air quality models) for the three traffic subcategories is very different (see section 2.1.7).

Table 2.3 The description of the source categories used.

CORINAIR SNAP Code	New Code	Description	Remarks
01	01	Public power, cogeneration and district heating plants	This is to a large extent the LOTOS source category No 1 (utilities point sources) and part of LOTOS source category No 2 (utility and industrial combustion)
02	02	Commercial, institutional and residential combustion	This is to a large extent the LOTOS source category No 3 (commercial and residential combustion)
03	03	Industrial combustion and processes with combustion	This is part of LOTOS source group No 2 (utility and industrial combustion)
04	04	Non-combustion production processes	More or less equal to LOTOS source category No 5 (selected production processes)
05	05	Extraction and distribution of fossil fuels	Part of LOTOS source category No 3 (commercial and residential combustion)
06	06	Solvent use	Equal to LOTOS source category No. 6 (solvent evaporation)
07		Road transport	This is a combination of LOTOS source category No 7 (road transport gasoline), No 8 (diesel) and No 9 (evaporation). This splitting is kept in the new data base, see below on 07a, 07b and 07c
07	07a	Road transport gasoline	} Subdivision of SNAP level 1, category 7 "road transport"
07	07b	Road transport diesel	
07	07c	Road transport evaporation	
08	08	Other mobile sources and machinery	Part of LOTOS source category No 3 (commercial and residential combustion), partly missing in LOTOS: air traffic and shipping
09	09	Waste treatment and disposal	Part of LOTOS source category No 3 (commercial and residential combustion)
10	10	Agriculture	Part of LOTOS source category No 3 (commercial and residential combustion)
11	11	Nature	This Corinair category will not be taken into account
	11a	Natural emissions coniferous	This is LOTOS source category No 10
	11b	Natural emissions high isoprene	This is LOTOS source category No 11
	11c	Natural emissions low isoprene	This is LOTOS source category No 12
	11d	Natural emissions nitrogen oxide	This is LOTOS source category No 13
	11e	Natural emissions PM 10	
	11f	Natural emissions PM 2.5	

Within this project only CORINAIR categories 1 – 10 are taken into account. In Table 2.2 category 11 is only listed in order to show the connection with the categorization formerly used in the PHOXA/LOTOS framework. For natural emissions a different approach has been followed, since these emissions generally depend on meteorological conditions such as temperature, wind speed, dryness of the surface and on land use. Therefore, natural emissions should be computed using the necessary input. We refer to section 2.3 for a discussion on biogenic NO_x and NM-VOC emissions.

2.1.3 The area and spatial distribution

The area as defined in the project-description was the PHOXA-REM-3 area, which covers in principle central Europe. Early in the project it was decided to create a

database covering the whole of Europe, the so-called LOTOS area. This has the advantage that in the future model calculations are possible over an extended domain comparable for example with the EMEP-area. The other advantage is that a more direct comparison with other emission databases, again like EMEP, is more straightforward.

At TNO a database is available containing point source information. This database has been set up already in the 80s and has been updated since using various sources of information such as national authorities, contacts with (local) experts, industrial interest organisations, various proprietary data bases etc. Also information on specific sources has been taken from the internet. In the framework of the CEPMEIP-project, focussing on the set-up of the first European-wide PM₁₀ and PM_{2.5} anthropogenic emissions database (see under 2.1.5), TNO has asked the responsible authorities in each country to comment on this database and to give updated information. Most countries have provided TNO with updated spatial distributions of their point sources (unfortunately, Germany was in the group of countries that did not deliver updated information). The (updated) information has been used in this project for the distribution of the emissions in category 1, 3, 4 and 5 of both the gaseous species and PM. The remaining emissions are considered as area emissions and their spatial allocation has been made using population density maps and distribution maps for agriculture. The population density map is the world-wide NOAA-CESIN data base on a resolution of 1/6x1/6 degree longitude-latitude. A more recent database is not available. For agriculture, use has been made of the Eurostat cattle distribution at nuts-3 level.

2.1.4 CORINAIR/EMEP/LOTOS

For the set-up of the emission data base for NO_x, SO_x, NH₃, NM-VOC, CO and CH₄ use has been made of the emission information as given in CORINAIR, and the official emissions data that countries have provided to EMEP. It should be noted that the CORINAIR information does not cover the whole area. So, for other areas use has been made of the original LOTOS-information and emissions generated by IIASA's RAINS v7.2 model (SO_x, NO_x, NH₃). As far as possible the country totals and per source category match the EMEP totals. In Figure 2.1 the gridded yearly emissions for a number of compounds are shown.

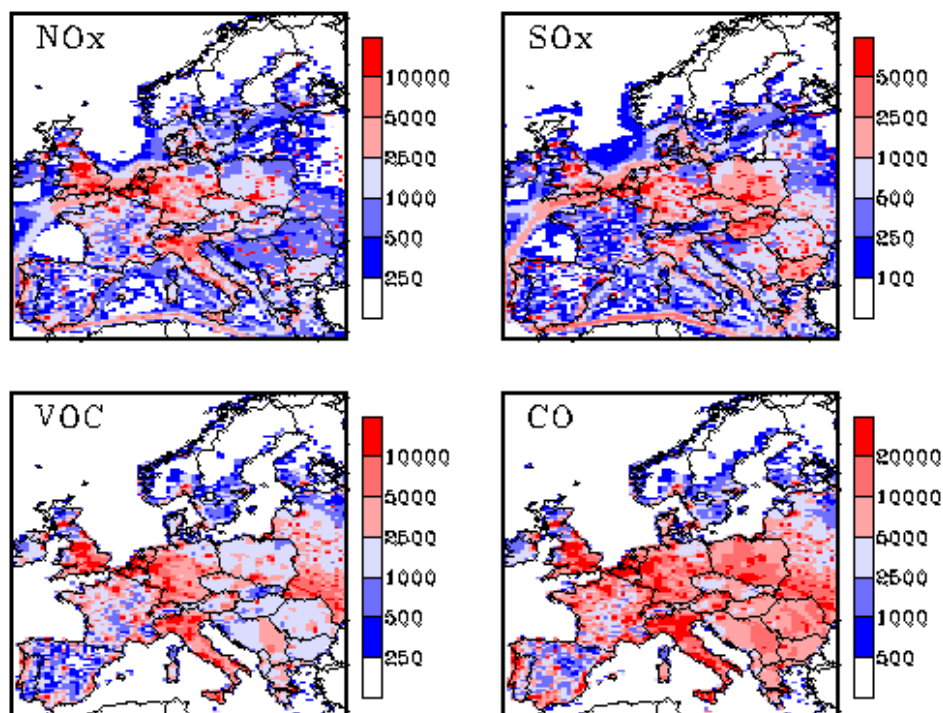


Figure 2.1 Yearly emissions of NO_x , SO_x , VOC and CO for 1995 in kg per grid cell of 0.5×0.25 degree longitude-latitude.

The emissions of SO_x and NO_x in Figure 2.1 include estimates for ship emissions. These estimates are not part of the TNO inventories, but are taken from EMEP. Especially for NO_x and SO_x emissions from ships contribute significantly to the total emissions in some parts of Europe. EMEP information on ship emissions of VOC and CO is also available, but their contributions to the total emissions are relatively small. This explains why the ship routes are not visible in the VOC and CO panels in Figure 2.1.

2.1.5 CEPMEIP

In September 2000 TNO received a contract to create an emission database for anthropogenic PM_{10} and $\text{PM}_{2.5}$ for EMEP. This CEPMEIP-data base is meant to provide guidance to the countries in their obligation to provide EMEP with PM_{10} and $\text{PM}_{2.5}$ emissions for the year 2000. Use has been made in that study of previous work as described in *Berdowski et.al.*(1997)

In CEPMEIP an update of the spatial distribution of point sources has been taken place. In view of that, it was decided to use this new CEPMEIP point source information in the current project not only for PM_{10} and $\text{PM}_{2.5}$, but also as the basis for the other species.

Although the decision to use the CEPMEIP-information has led to a substantial time-delay in the current project, the advantage is that the new emission database made for 1995 is now consistent with CEPMEIP, and is of good quality.

For the current project a gridding has been performed in order to obtain an emission database with a resolution of 0.25×0.5 degrees in geographical coordinates (lon-lat). In order to make a geographical distribution of the reported national emissions per category use is made of various databases containing spatial information, such as population density, land use etc. These databases usually have a very fine resolution of e.g. a few kilometres and can thus safely be used to construct the spatial distribution of the emissions on the resolution required in this project.

The final database contains per grid the amount of surface-emissions (area-sources) and the amount of higher-level emissions. No specific information on the height of the point source emissions is given. Also, no explicit information of the location of point sources is included. This is in accordance with the modelling approach in which point sources are not treated explicitly, but emit in the grid-volume at a specific height. For modelling purposes one may assume the point source emission in category 1 and 3 to take place at a height of 150m and 50m, respectively. The point source emission in category 4 and 5 are assumed to take place near ground level.

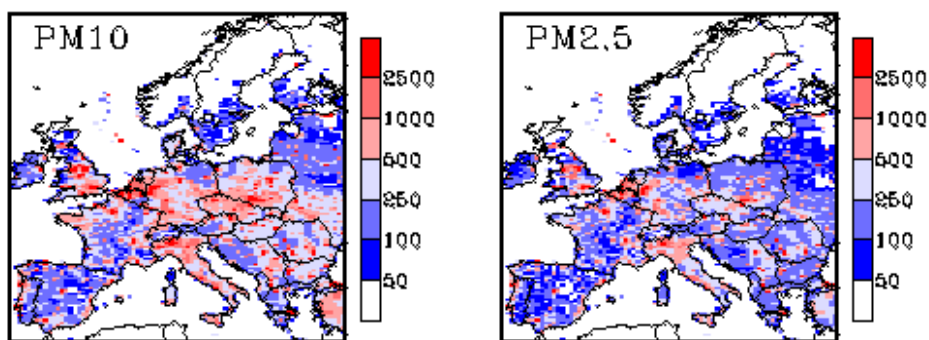


Figure 2.2 Total PM_{10} and $PM_{2.5}$ emissions from the CEPMEIP database in kg per grid cell of 0.25×0.5 degree

Concerning CEPMEIP two remarks should be made. The emission factor for non-tailpipe traffic related PM-emissions contains the emissions of particles from tires, surface and breaks. However, particle emissions from resuspension are not included. Second, the original emission factor for diesel cars has been lowered by a factor two, based on official Auto-oil data.

2.1.6 Time- and temperature factors

The basic information, which is also the input data for the chemistry-transport-model (CTM), is the gridded yearly averaged anthropogenic emission database.

However in reality emissions of specific source categories, as for example road transport, fluctuate in time, and/or with temperature, as road transport evaporation. The time and temperature factors that were in use in the PHOXA/LOTOS system, have been critically reviewed. This review has taken place in cooperation with the project TROTEP, a 5th FP EU-project (coordinator P. Monks, University of Leicester, UK) on Tropospheric Ozone, its trends, budgets and policy). TNO-MEP is a partner in the TROTEP project that will be finalized early 2003. The revised time factors from TROTEP are given in Table 2.4 to Table 2.7.

category	jan	feb	mar	apr	may	jun	jul	aug	sep	oct	nov	dec
1 Power generation	1.20	1.15	1.05	1.00	0.90	0.85	0.80	0.87	0.95	1.00	1.08	1.15
2 Residential, commercial and other combustion	1.70	1.50	1.30	1.00	0.70	0.40	0.20	0.40	0.70	1.05	1.40	1.65
3 Industrial combustion	1.10	1.08	1.05	1.00	0.95	0.90	0.93	0.95	0.97	1.00	1.02	1.05
4 Industrial processes	1.02	1.02	1.02	1.02	1.02	1.02	1.00	0.84	1.02	1.02	1.02	0.90
5 Extraction distribution of fossil fuels	1.20	1.20	1.20	0.80	0.80	0.80	0.80	0.80	0.80	1.20	1.20	1.20
6 Solvent use	0.95	0.96	1.02	1.00	1.01	1.03	1.03	1.01	1.04	1.03	1.01	0.91
7a Road transport gasoline	0.88	0.92	0.98	1.03	1.05	1.06	1.01	1.02	1.06	1.05	1.01	0.93
7b Road transport diesel	0.88	0.92	0.98	1.03	1.05	1.06	1.01	1.02	1.06	1.05	1.01	0.93
7c Road transport evaporation	0.88	0.92	0.98	1.03	1.05	1.06	1.01	1.02	1.06	1.05	1.01	0.93
8 Other mobile sources and machinery	0.88	0.92	0.98	1.03	1.05	1.06	1.01	1.02	1.06	1.05	1.01	0.93
9 Waste treatment and disposal	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
10 Agriculture	0.45	1.30	2.35	1.70	0.85	0.85	0.85	1.00	1.10	0.65	0.45	0.45

Table 2.4 Monthly emissions factors for the SNAP level 1 categories

category	Mon	Tue	Wed	Thu	Fri	Sat	Sun
1 Power generation	1.06	1.06	1.06	1.06	1.06	0.85	0.85
2 Residential, commercial and other combustion	1.08	1.08	1.08	1.08	1.08	0.8	0.8
3 Industrial combustion	1.08	1.08	1.08	1.08	1.08	0.8	0.8
4 Industrial processes	1.02	1.02	1.02	1.02	1.02	1.02	1
5 Extraction distribution of fossil fuels	1	1	1	1	1	1	1
6 Solvent use	1.2	1.2	1.2	1.2	1.2	0.5	0.5
7a Road transport gasoline	1.02	1.06	1.08	1.1	1.14	0.81	0.79
7b Road transport diesel	1.02	1.06	1.08	1.1	1.14	0.81	0.79
7c Road transport evaporation	1.02	1.06	1.08	1.1	1.14	0.81	0.79
8 Other mobile sources and machinery	1	1	1	1	1	1	1
9 Waste treatment and disposal	1	1	1	1	1	1	1
10 Agriculture	1	1	1	1	1	1	1

Table 2.5 Emission factors for the day of the week for the SNAP level 1 categories

category	Hour of day											
	1	2	3	4	5	6	7	8	9	10	11	12
1 Power generation	0.79	0.72	0.72	0.71	0.74	0.80	0.92	1.08	1.19	1.22	1.21	1.21
2 Residential, commercial and other combustion	0.38	0.36	0.36	0.36	0.37	0.50	1.19	1.53	1.57	1.56	1.35	1.16
3 Industrial combustion	0.75	0.75	0.78	0.82	0.88	0.95	1.02	1.09	1.16	1.22	1.28	1.30
4 Industrial processes	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
5 Extraction distribution of fossil fuels	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
6 Solvent use	0.50	0.35	0.20	0.10	0.10	0.20	0.75	1.25	1.40	1.50	1.50	1.50
7a Road transport gasoline	0.19	0.09	0.06	0.05	0.09	0.22	0.86	1.84	1.86	1.41	1.24	1.20
7b Road transport diesel	0.19	0.09	0.06	0.05	0.09	0.22	0.86	1.84	1.86	1.41	1.24	1.20
7c Road transport evaporation	0.19	0.09	0.06	0.05	0.09	0.22	0.86	1.84	1.86	1.41	1.24	1.20
8 Other mobile sources and machinery	0.19	0.09	0.06	0.05	0.09	0.22	0.86	1.84	1.86	1.41	1.24	1.20
9 Waste treatment and disposal	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
10 Agriculture	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Table 2.6 Emission factors for the hour of day (1:00-12:00) for the SNAP level 1 categories

category	Hour of day											
	13	14	15	16	17	18	19	20	21	22	23	24
1 Power generation	1.17	1.15	1.14	1.13	1.10	1.07	1.04	1.02	1.02	1.01	0.96	0.88
2 Residential, commercial and other combustion	1.07	1.06	1.00	0.98	0.99	1.12	1.41	1.52	1.39	1.35	1.00	0.42
3 Industrial combustion	1.22	1.24	1.25	1.16	1.08	1.01	0.95	0.90	0.85	0.81	0.78	0.75
4 Industrial processes	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
5 Extraction distribution of fossil fuels	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
6 Solvent use	1.50	1.50	1.50	1.50	1.50	1.40	1.25	1.10	1.00	0.90	0.80	0.70
7a Road transport gasoline	1.32	1.44	1.45	1.59	2.03	2.08	1.51	1.06	0.74	0.62	0.61	0.44
7b Road transport diesel	1.32	1.44	1.45	1.59	2.03	2.08	1.51	1.06	0.74	0.62	0.61	0.44
7c Road transport evaporation	1.32	1.44	1.45	1.59	2.03	2.08	1.51	1.06	0.74	0.62	0.61	0.44
8 Other mobile sources and machinery	1.32	1.44	1.45	1.59	2.03	2.08	1.51	1.06	0.74	0.62	0.61	0.44
9 Waste treatment and disposal	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
10 Agriculture	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00

Table 2.7 Emission factors for the hour of day (13:00-24:00) for the SNAP level 1 categories

The hour of day is local time, hence information over the deviation from GMT is needed for each country. The following time-zones are incorporated:

- GMT+0 UK, Ireland, Iceland and Portugal
- GMT+1 all other European countries except those listed with GMT+2:
- GMT+2 for Finland, Estonia, Latvia, Belarus, Ukrain, Moldavia, Romania, Bulgaria, Greece and Turkey
- GMT+3 Azerbaidjan, Armenia, Georgia, Russia untill the Oeral.

Currently it is assumed that all countries have the shift from summer to wintertime and vice versa at the same days, i.e. the last Sunday of October and March, respectively.

In addition to the time factors specified above in Table 2.4 to Table 2.7 a temperature factor for road transport, categories 7a and 7b, is applied for the emissions of VOC and CO. Their emissions are assumed to decrease linearly with temperature, as shown in Figure 2.3

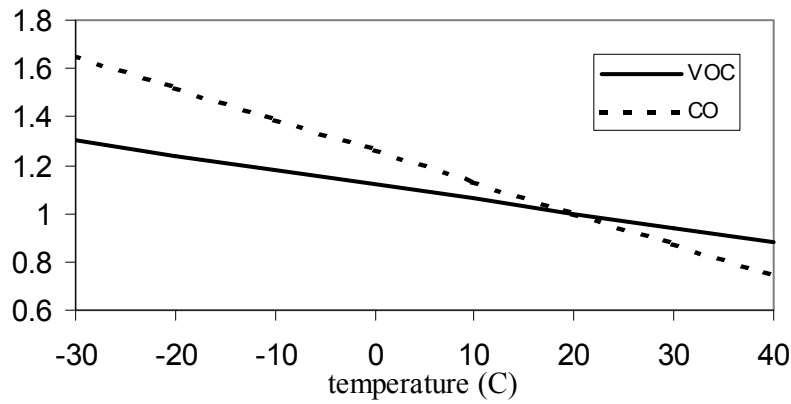


Figure 2.3 Temperature factors to be applied for VOC and CO from road transport category 7a and 7b (gasoline and diesel)

The higher emissions for VOC and CO at lower temperatures are due to the so-called “cold start”.

2.1.7 NMVOC-speciation

An update has taken place of the NMVOC-profiles for the relevant anthropogenic source categories. In Table 2.8 as an example the VOC speciation is given for The Netherlands. For each country a similar table is available, see Appendix B.

Sector name	acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & alkynes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
1	0.0	0.0	2.2	17.0	0.0	0.0	5.3	3.9	0.0	0.1	17.0	0.0	0.0	0.5	0.0	0.4	9.6	21.3	17.1	1.1	2.6	0.0	1.7
2	0.4	0.2	9.0	19.7	0.0	0.0	0.1	0.2	0.1	0.1	4.0	0.0	17.8	0.1	0.1	0.0	0.0	34.5	8.9	0.1	5.0	0.0	0.0
3	2.0	1.0	0.5	0.0	0.0	0.0	0.5	0.9	0.4	0.3	0.1	0.1	0.2	0.5	0.5	0.1	192.0	0.1	0.1	0.4	0.2	0.0	0.1
4	0.0	0.0	0.1	0.6	0.0	0.0	1.6	8.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	87.8	0.3	1.0	0.0	0.0	0.0	0.0
5	0.0	0.0	0.2	26.9	0.0	0.0	34.9	0.0	0.0	0.0	8.8	0.0	0.0	1.3	0.0	0.0	0.0	10.4	17.3	0.0	0.1	0.0	0.1
6	0.0	9.4	0.0	0.0	10.2	11.7	0.0	0.0	4.6	0.0	30.8	10.9	0.0	0.0	0.0	1.7	4.0	0.0	0.0	0.0	8.3	0.0	8.3
7a	0.0	0.0	4.5	4.3	0.0	0.0	1.4	7.2	0.0	4.5	22.8	0.1	1.7	6.9	1.2	11.1	0.0	6.4	0.1	3.8	12.0	3.9	8.1
7b	0.0	0.0	2.0	2.0	0.0	0.0	1.0	12.0	0.0	4.0	30.0	1.5	6.0	5.0	6.5	20.5	0.0	2.0	1.0	3.0	1.5	0.0	2.0
7c	0.0	0.0	1.0	32.0	0.0	0.0	0.0	0.0	0.0	0.0	15.0	0.0	0.0	9.5	0.0	0.0	0.0	40.0	1.0	0.0	1.0	0.0	0.5
8	0.0	0.0	2.0	2.0	0.0	0.0	1.0	12.1	0.0	4.0	29.6	1.5	6.2	5.1	6.7	20.2	0.2	2.0	1.0	3.0	1.5	0.0	2.0
9	11.0	6.1	3.6	7.7	1.2	1.3	5.1	7.4	2.4	2.4	12.6	1.6	1.2	4.0	2.8	2.3	6.5	6.9	4.2	2.9	3.8	0.5	2.5
10	See text																						

Table 2.8 VOC speciation per source category in percent of the total VOC emission in The Netherlands.

For agriculture (SNAP level 1 category 10) a VOC speciation in terms of the compounds listed in Table 2.8 is not available. However, a speciation in terms of the compounds of the CBM-IV mechanism is available and is given in Table 2.9.

Table 2.9 NM-VOC speciation for source category Agriculture in terms of the compounds of the CBM-IV mechanism in mol C_x/kg

ole	par	tol	xyl	form	ald	ket	acet	eth	unreactive
2.13	22.4	0.37	0.09	1.95	1.66	0.1	0.05	4.5	10.67

2.1.8 Update factors for other years

The emission database is valid, both in spatial distribution and in totals per country per source category for the year 1995.

To be able to make model runs for the years 1996-2001, the following procedure has been followed to estimate update factors:

- The spatial distribution has been left unchanged.
- The emission data for PM₁₀ and PM_{2.5} have not been updated, in view of the large uncertainty in the values for the base year 1995.
- The other species have been updated per country and per source category based on available EMEP/CORINAIR-data (see http://www.emep.int/emis_tables/stab1.html).

An example of such an update table for one component and one SNAP level 1 category is given in Table 2.10. The tables with update factors for all species (except PM, see above) have been delivered electronically and can also be found on the CD-ROM accompanying this report.

Table 2.10 NM-VOC Emission update factors for traffic

Country code	Change 1996 - 1995	Change 1997 - 1996	Change 1998 - 1997	Change 1999 - 1998	Change 2000 - 1999	Change 2001 - 2000
alb	1.00	1.00	1.00	1.00	1.00	1.00
aut	0.88	0.91	0.90	0.90	1.00	1.00
bel	0.92	0.98	0.97	0.97	1.00	1.00
bgr	1.00	1.00	1.27	1.27	1.00	1.00
bih	1.00	1.00	1.00	1.00	1.00	1.00
blr	1.00	1.00	1.00	1.00	1.00	1.00
che	1.00	1.00	1.00	1.00	1.00	1.00
cze	2.79	1.19	0.91	0.91	1.00	1.00
deu	0.89	0.87	0.88	0.88	1.00	1.00
dnk	0.89	0.88	0.97	0.97	1.00	1.00
esp	1.05	0.97	0.99	0.99	1.00	1.00
est	1.00	1.00	1.30	1.30	1.00	1.00

fin	1.00	1.00	1.00	1.00	1.00	1.00
fra	0.91	0.92	0.93	0.93	1.00	1.00
gbr	0.93	0.90	0.91	0.91	1.00	1.00
grc	1.05	1.02	1.04	1.04	1.00	1.00
hrv	1.00	1.00	1.00	1.00	1.00	1.00
hun	1.00	1.00	1.00	1.00	1.00	1.00
irl	1.10	1.02	1.00	1.00	1.00	1.00
ita	0.95	0.99	1.00	1.00	1.00	1.00
ltu	1.09	1.00	1.24	1.24	1.00	1.00
lux	1.00	1.00	1.00	1.00	1.00	1.00
lva	0.89	1.47	0.92	0.92	1.00	1.00
mda	1.00	1.00	1.00	1.00	1.00	1.00
mkd	1.00	1.00	1.00	1.00	1.00	1.00
nld	0.88	0.94	0.94	0.94	1.00	1.00
nor	1.00	1.00	1.00	1.00	1.00	1.00
pol	1.00	1.05	0.93	0.93	1.00	1.00
prt	1.02	0.99	0.98	0.98	1.00	1.00
rom	1.00	1.00	1.00	1.00	1.00	1.00
rus	1.00	1.00	1.00	1.00	1.00	1.00
svk	1.00	1.00	1.00	1.00	1.00	1.00
svn	1.00	1.00	1.00	1.00	1.00	1.00
swe	0.89	0.91	0.94	0.94	1.00	1.00
ukr	1.00	1.00	1.00	1.00	1.00	1.00
yug	1.00	1.00	1.00	1.00	1.00	1.00

It should be noted that in most cases only two reference years are available: 1995 and 2000. In these cases linear update factors have been derived. This explains why in many cases the numbers in Table 2.10 are row-wise constant.

2.2 European land use database

The task of TNO in this project was to develop, to recommend, a state-of-the-art land use database, which is suited for the requirements of the CTMs in use.

The land-use database has to fill the following tasks:

- Input data for dry deposition parameters: roughness length z_0 and surface resistance r_c .
- Input data for the biogenic emissions of NMVOC and NO.

In recent studies it has become clear that the accuracy of the amount of biogenic emissions, see also under 2.3, depends to a large extent upon the choice of the land use data base. In other words, the land use database is the key element in the determination of biogenic emissions.

At the start of the project the land use data base which was used by REM3/LOTOS was based on the previous work by Veldt (1989), the so-called PHOXA/LOTOS land use data base, see also Meeuwsen et al. (1994). This database was based on previous work by Henderson-Sellers.

The database contained 11 categories.

Modern landuse data bases use satellite data as a starting point. The CORINE land cover database is based on images from Landsat TM and SPOT and uses visual interpretation, and consultation of ancillary data. In total 44 classes are distinguished. At the start of the project, the CORINE database covered only part of Europe. This was the main reason to start analysing a land cover data base at that moment under development in the INDAVOR-project (Instrument for Detecting Land Cover Change for Europe). As reflected in the title, the aim of INDAVOR was to set-up a land-cover database which could be easily updated to other years, de Boer, 1999. In INDAVOR use was made of the NOAA AVHRR images. The NDVI (normalised differenced vegetation index) was used to analyse different vegetated areas. The finest resolution is 1.1 x 1.1 km².

For our modelling purposes the choice has been made to use 15 land cover categories, as shown in Table 2.11. The table gives also the relation with the previous LOTOS/PHOXA classification.

Table 2.11 Land cover categories used for the classification and required for the LOTOS model

Nr.	Classified Land cover types	LOTOS land cover types
1	Urban area	*
2	Arable land	*
3	Irrigated arable land	
4	Permanent crops	*
5	Pastures	*
6	Natural grassland	
7	Shrubs and herbs	
8	Coniferous forest	*
9	Mixed forest	*
10	Deciduous forest	*
11	Bare soil	*
12	Permanent ice and snow	*
13	Wetlands	*
14	Inland water	
15	Sea	*

The overall result, the so-called PELINDA, Pan European Land Cover Database for the year 1997 is presented in Figure 2.4.

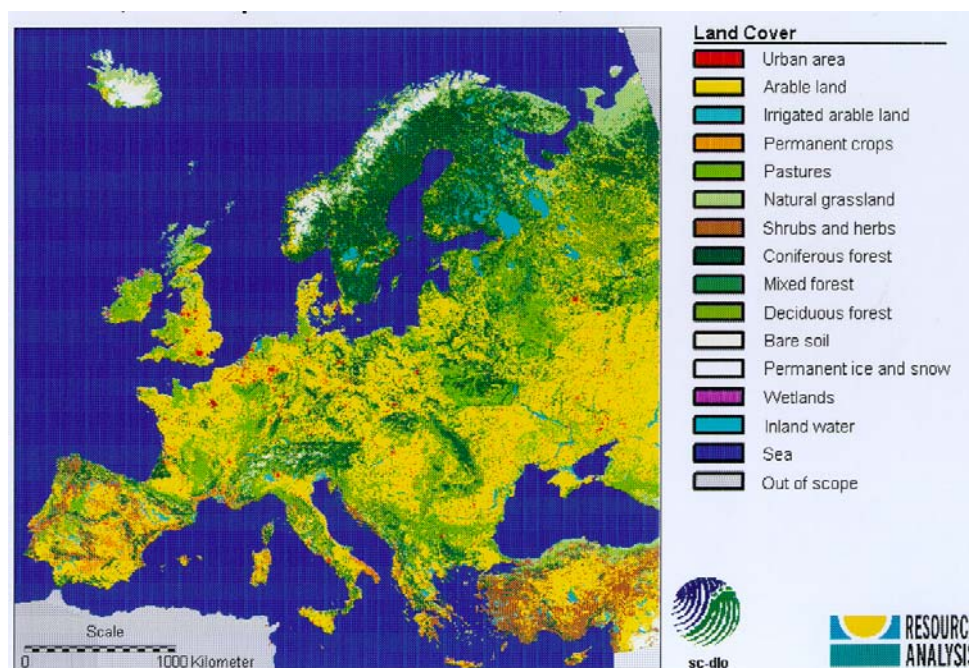


Figure 2.4 Pelinda, Pan European Land Cover database for 1997

The data on $1 \times 1 \text{ km}^2$ have been integrated to the grid size for REM-3/LOTOS of 0.25×0.5 lat-lon with a percentage of each category per grid.

For a more detailed description of the land use data base PELINDA and the related parameters, see Nijenhuis and Groten, 1999, and de Boer et al., 2000.

2.3 European wide biogenic emissions

Starting point for the determination of biogenic emissions is the choice of the land cover database. Here we use the PELINDA database with the 15 categories as described in 2.2. As has been mentioned, this choice determines to a large extent the calculated total biogenic emissions.

A description will be given of biogenic NO-emissions and biogenic NMVOC emissions.

2.3.1 Biogenic NO-emissions

First a remark should be made about the nature of these emissions. The current NO-emissions from the soil cannot be considered as the 'natural' or 'pre-industrial' soil emissions. These emissions are a function of the deposited N from the air, or as for arable land, from the N-fertilization rate.

The best definition for these NO-emissions is that they reflect the current, uncontrollable, emissions from the soil.

In the previous LOTOS/PHOXA study a first estimate, based on at that moment existing information, was made that Europe wide the biogenic NO-emissions amounted to 4 % of the anthropogenic NO-emissions. Model calculations have been performed with these data, using at that moment the anthropogenic spatial distribution also for the biogenic NO.

In Simpson et al., 1995, a rather simple method is given to calculate biogenic NO-emissions. They are a function of the soil temperature, and of the soiltype. Simpson only distinguished between forest areas (category 8,9,10 of PELINDA) and grassland/pastures which he uses for all no forested land areas.

In this study TNO has performed a literature survey, which is given in Appendix C "NO-emissions through soil".

Based on this analysis the method of Yienger and Levy, 1995, is selected as the most appropriate method at the moment. The method is also used in Global models as MOZART.

For this method the PELINDA land use categories coniferous and deciduous forest are used, and also arable land/grassland, wetlands, crops.

The method needs soil temperature and soil humidity, wet and dry conditions, LAI and SAI (stomata area index), canopy reduction and pulsing.

Pulsing is the effect that there is an increase in NO-emissions due to precipitation on a dry soil. For a detailed description, see Appendix C.

From the description in Appendix C it is clear that the method of Yienger and Levy is more complicated than the method of Simpson, and requires more input data.

Currently, a detailed analysis which method performs best relative to reality is still lacking.

2.3.2 Biogenic NMVOC-emissions

In the LOTOS/PHOXA studies the biogenic NMVOC-emissions from forests were given by a method developed by Veldt, 1991. Apart from the difference between deciduous, coniferous and mixed forest, the only other parameter was ambient temperature.

Extensive studies by Guenther showed that next to ambient temperature also the Photosynthetic Active Radiation(PAR) is important, see Guenther, 1994. These findings by Guenther have been applied to Europe by Simpson, Newitt and Steinbrecher, see Simpson, 1995.

Although many uncertainties still exist, the method by Simpson is the most suited at the moment. This method distinguishes in more detailed forests types as currently available in PELINDA. Next to coniferous also northern Europe and Mediterranean oak, other broadleaf, LI/Sitka spruce and other spruce should be treated separately. Although these data can in principle be made available, they are not contained in PELINDA yet. So although the percentages of these species are

known per country, only a first estimate using PELINDA can be done, because the location within a country is currently lacking.

Needed for the calculation of the isoprene emissions is per tree-type the biomass density and the mass-based emission factors, see Simpson et al. 1995. The ambient temperature is given in the meteorological database.

The PAR-flux in microEinstein/m²s can be derived from the global radiation.

The global radiation in W/m² multiplied by 2 gives the PAR-flux in microEinstein/m²s.

(The conversion is first from global, direct+diffuse radiation, to short wave by multiplication by a factor between 0.45 - 0.55, and then to microEinstein by 4.6, leading to a factor of about 2.3. Common practice in Europe is the use of a factor 2, Steinbrecher, 2002, see also Langholz und Häckel, 1985).

It should be noted that estimates of the overall uncertainty of the total European wide biogenic NMVOC are as high as 500 %, Simpson, 1995.

2.4 Determination of boundary conditions

2.4.1 Introduction

The task of TNO in the project was to give advice concerning, and to produce, adequate boundary and initial conditions for the REM-3/Calgrid model. Because models like REM-3 are run for long time periods of a year or more, the initial conditions are of minor influence, so the discussion here will be focussed on boundary conditions.

Boundary conditions are needed for all models with a spatial coverage smaller than the total globe. So, continental models like REM-3 need boundary conditions as a prerequisite to perform model runs.

In principle, two types of boundary conditions (BC) exist:

- I) BC based on observations.
- II) BC based on global model runs.

Some models use observations for the BC, like the EMEP model, some models like LOTOS make use of global model runs for the BC. The advantage of using observations is that these data are close to 'reality'. The disadvantage of using observations is that they might be inconsistent and might contain errors, and that they give a fixed value valid for a specific time-period. The advantage of using BC from models is that they are consistent, and that by running scenario-calculations for future years they can also give modified BC. The disadvantage is that these BC might deviate from observations, limiting the model performance, and that producing and using these BC in continental model runs is time- and computer-consuming.

Recent model studies, like Langmann et al. (2002) using the GEOS-CHEM with REMO-model, show the advantage of using nested model runs. The nested model

gives clearly a better model performance than the model runs with BC from observations.

2.4.2 Global models

At the beginning of the project the BC for the LOTOS-model, a model in operation at TNO which has many similarities with REM3, were derived from the 2-D global Isaksen-TNO model, Roemer et al.(1991). Although this model is in reasonable agreement with observations concerning ozone, it was considered out-of-date in view of existing 3-D global models.

In principle, three types of 3-D global models exists:

- 3-D models which use a 20-year climatology as meteorological input. An example is the MOGUNTIA-model developed by Zimmermann and Crutzen (1989), see for recent results Dentener and Raes (2002).
- 3-D models which use a diagnostic meteorology as input, so-called off line models. An example is the TM-3 model, originally developed by Heimann (1989).
- 3-D models which use a prognostic meteorology, and calculate atmospheric chemistry and meteorology in one model, so-called on line models. An example is the ECHAM-model, developed at the Max-Planck Institute in Hamburg, Roeckner (1996), see also Roelofs et al. (1997).

Model output of MOGUNTIA is available. Both TM-3 and ECHAM are in operation at the Institute of Marine and Atmospheric Research, Univ. Utrecht, IMAU. TNO has access to the model results.

At the moment, the TM-3 model has as chemical module CBM-4, as has LOTOS and REM3. Also TM-3 has been run for several years to study the behaviour and trend in CH₄-concentrations. Because also LOTOS and REM-3 operate with diagnostic meteorological input, it has been decided to use TM-3 to provide BC for the continental scale model runs of REM-3.

2.4.3 Results of TM-3

The TM-3 model, Transport Model -3, is in operation at the moment at IMAU (Dr. Maarten Krol), KNMI (Dr. Michiel van Weele c.s.), JRC-Ispra (Dr. Frank Dentener) and TNO (Dr. Michiel Roemer c.s). Numerous studies are being performed on both ozone, methane and aerosols.

A detailed model run has been performed for 1997, and the results can directly be used for BC. The model output is 6-hourly, and at several vertical layers up to about 10 km. An example of model output for ozone is given in Figure 2.5 and Figure 2.6 for August 15, 1997 at 12.00 AM.

In principle, these model results can only be used for REM-3 runs for 1997. The BC for other years will be different, and would require TM-3 model runs and

analysis for these years, say for 1995-2001. Also the use of TM-3 results for 1997 is only 'correct' in case the meteorology of TM-3 and LOTOS or REM-3 is -nearly-identical.

Further more, it should be noted that the BC of TM-3 are used without interaction with REM-3 or LOTOS, the method of one-way nesting is used.

This might lead to the problem of 'double counting'; the TM-3 model runs do already contain European emissions. However these impacts will be minor, provided the meteo is consistent (the wind direction in TM-3 and the continental models should be similar)

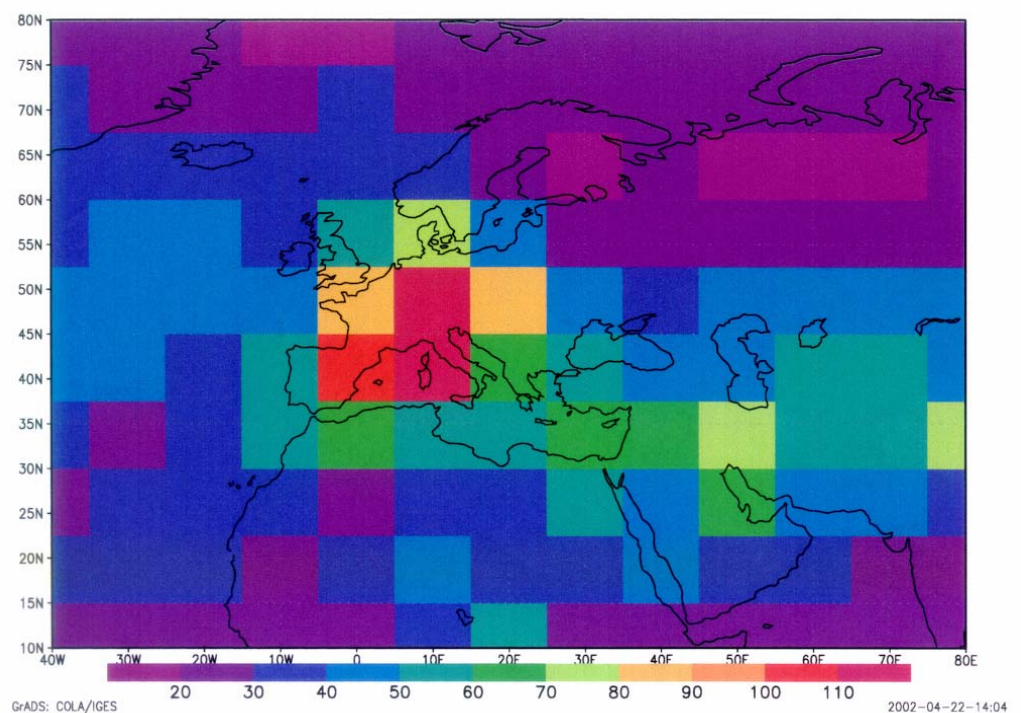


Figure 2.5 TM-3 August 15, 1997, surface layer.

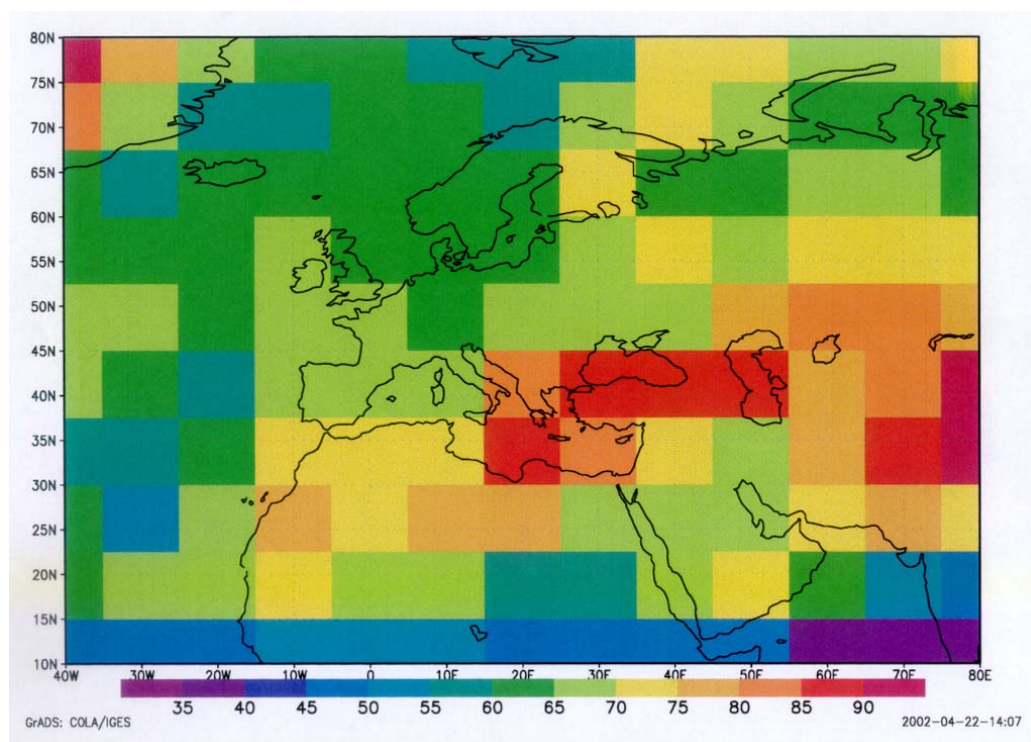


Figure 2.6 TM-3 August 15, 1997, 4 km

2.4.4 Discussion

Two items should be mentioned here, the influence of the description of vertical-transport and - exchange, and the hemi-spherical ozone budget.

– *Vertical transport and exchange.*

BC have an impact on the horizontal boundaries of the domain, so outside Europe, but also in the vertical. Especially with models like REM-3 and LOTOS, with a limited extension of about 2-3 km (chosen to make long term runs possible), the impact of the BC at the height of 2-3 km can be substantial. Both REM-3 and LOTOS use diagnostic horizontal wind fields based on observations as model input. The vertical velocities are calculated subsequently from the mass-balance, convergence and divergence. In case these vertical velocities are too high or too low, relative to the real world, the impact of the BC is too large, or too little. This aspect deserves further attention.

– *The hemi-spherical ozone budget.*

There is currently much debate concerning the impact of Asian and American emissions on the ozone over Europe. Most global models show a rather small impact at ground level of only a few ppb of ozone due to US-emissions, and a negligible impact of Asian emissions, see Li (2002). The question is whether the model

runs cover a sufficient long time scale, of more than 10-20 years, to describe adequately the impact of methane.

3 Improvement of the internal processes in the ACT-model

3.1 Development and testing of an aerosol-module

The task of TNO in this project was to analyse and propose an aerosol module, which can be used in REM-3/LOTOS. The aim was to combine such an aerosol module with the gas phase mechanism, which is used, the CBM-4 mechanism. The final goal is to use these combined gas phase/aerosol model to calculate both PM₁₀ and PM_{2.5} concentrations, including their chemical composition to be able to define the contributions of the different source categories to the calculated concentrations.

The PM_{2.5} concentration consists of inorganic aerosols (SO₄, NO₃, and NH₄ forming ammonium sulphate, ammonium(bi)sulphate and ammoniumnitrate), organic aerosols, both anthropogenic and biogenic (the largest organic fraction) and also primary anthropogenic aerosols. The PM_{2.5} fraction could also contain some sea-salt and wind-blown dust.

The PM₁₀ concentrations consist of everything contained in PM_{2.5}, and the coarse fraction between 2.5 and 10 which mainly contains primary emitted anthropogenic aerosols, windblown dust and sea salt, and -maybe- particles from tires and the street surface.

For the modelling of inorganic aerosols, the MADE-module has been chosen (Modal Aerosol Dynamics Model for Europe), see Ackermann et al., 1998. This model has been obtained in cooperation with FORD-Aachen. It has been tested extensively, and has been incorporated in both LOTOS and REM-3. For details see the Appendix D "Aerosols in LOTOS", which is provided separately.

The MADE-module has been tested, combined with CBM-4, in the LOTOS model for August 1997, and currently for the whole year 1995. Calculated and observed concentrations of sulphate, nitrate and ammonium show reasonable agreement, van Loon, 2001'. Model results have also be compared with satellite observations of Aerosol Optical Depth, again for August 1997, Builtjes et al., 2001.

A proper model description of aerosol-concentrations requires next to the treatment of dry deposition also wet deposition. For this, rain intensity per hour and per grid has been made available in the meteorological input data set.

3.2 Improvement of the dry deposition module

All state-of-the-art descriptions for dry deposition use the concept of resistances.

The atmospheric resistance is given by R_a (a function of the atmospheric stability only), the resistance of the viscous sub layer is given by R_b (a function of the deposited species controlled by molecular diffusion) and the surface resistance R_c . The descriptions for R_a and R_b have been developed about a decade ago, and are used in a similar fashion by all modellers, see for example Ganzeveld, 2001, Duyzer, 1995 and Erisman and Draayers, 1995. Some debate is going on concerning the reference height for the description of these parameters in models, a value between 30 and 50 m is often used, reflecting the part of the atmosphere of the constant flux layer.

Most research is focussed on the determination of R_c . In the old model approach in REM3 and LOTOS a fixed value in time was used for R_c , only dependent on the land use type and the species itself. Modern research has shown that R_c is a function of the land use category and the species, but also of the canopy temperature, of the surface wetness, of the Photosynthetic Active Radiation (PAR). This makes R_c a parameter, which fluctuates in time.

Also the type of vegetation as given by the land use category is important, because R_c depends on the Leaf Area Index, the single-side total area of leaves/needles per surface area. For a detailed and up-to-date description see Ganzeveld, 2001.

3.2.1 The module DEPAC

In the current LOTOS model, the dry deposition is parametrised by the most recent version of DEPAC (Erisman et al., 1995, see also Wesely, 1989; for a recent description and application see also Gauger et al. 2002). Apart from the parameters mentioned above DEPAC also takes into account parameters indicating the snow cover, the surface wetness and the ratio between SO_2 and NH_3 (co-deposition). DEPAC uses 9 land use classes, which is less than number of the categories in the Pelinda land use data set, presented in Section 2.2. However, the land use classes from the Pelinda database are mapped onto the classes used by DEPAC in a straightforward way. The land use classes employed by DEPAC are listed in Table 3.1.

Table 3.1 Land use classes used in DEPAC

Class no.	description
1	grass
2	arable
3	permanent crops
4	coniferous forest
5	deciduous forest
6	water
7	urban
8	other i.e. short grassy area
9	desert

For each cell the deposition velocity is calculated for every land use class and a weighted average is computed, based on the surface fractions i.e. the percentage of the area covered by each land use type. The surface wetness and snow cover have a large effect on the deposition velocities for a number of species, for instance SO₂. If not given in the meteorological input, the surface wetness is a very difficult parameter to determine. From a number of other meteorological parameters the dew point temperature could be determined and used as a criterion for whether the surface is wet by dew. In addition, an obvious assumption is to consider the surface as wet in case of precipitation. The remaining difficulty is how to determine when the surface is still wet after a dew or precipitation event. If snow cover is not part of the meteorological data a similar way of determining the snow coverage could be followed. After a snow event, (increase of the) snow coverage is assumed and the thickness of the snow deck is assumed to stay constant as long as the temperature remains below zero °C. If the temperature rises (significantly) above freezing level, the difficulty is how to determine the removal rate by melting. In LOTOS at this moment we assume that the snow cover decreases with 0.1cm/degree above the freezing point per hour, but it should be realized that this is just a crude guess and that a number of other meteorological conditions influence the process of melting of snow.

The original version of DEPAC delivers deposition velocities for the gas phase species SO₂, NO₂, NO, NH₃ and HNO₃ and for particulate SO₄, NO₃, NH₄ and BC (Na,Mg,Ca,K). The module DEPAC as delivered to FUB has been extended with O₃ as this is one of the key species in photo-oxidant models. It has also slightly been modified to give R_c-values as separate output since chemistry-transport models usually have their own parametrisations for R_a and also R_b.

4 Consultancy concerning data-assimilation

Data-assimilation is a numerical technique which makes it possible to combine in an objective way model results and observations.

In the previous PHOXA/LOTOS projects observations were used to evaluate the model performance, so for model validation. In case there was a large discrepancy between model results and observations, the method used was to perform model sensitivity runs, by modifying certain model-parameters within their bounds of uncertainty.

In fact, data-assimilation is doing the above in a more systematic and consistent way. A data-assimilation shell is build around the ACT-model (the so-called Kalman Filter method), or a special model version is build (the so-called ad joint model version-4Dvar). This data-assimilation method makes it possible to use observations as input to the model system. Subsequently noise values are given to specific model parameters or model input data like emissions, and the assimilated model system will perform calculations to minimise the difference between model results and observations (this description holds for Kalman Filtering, the procedure for 4D-var is slightly different).

The task of TNO was to give advice concerning the best practical data-assimilation method to be used in connection to REM3. Based on previous experience gathered in the 5 FP EU-project RIFTOZ (Regional Differences in Tropospheric Ozone), the choice was made to use Kalman Filtering, and not 4D-var. The main reason is that Kalman Filtering is independent of the ACT-model, and that the analysis of the results is more straightforward.

Experience has been obtained by TNO in combining and applying Kalman Filtering with LOTOS, see for example Loon, M. van, 2001.

Kalman filtering with LOTOS is operational at the moment, and has been applied to ozone observations, as well as observations of NO₂ and aerosols and satellite data of Aerosol Optical Depth.

The results by applying this method are the creation of a concentration field based on model results and observations combined. Furthermore, it makes an analysis possible of the spatial representative ness, or correctness, of observations. Also, the resulting noise factors add in the process of model validation, and model input validation as well.

Data-assimilation is a very powerful and promising method, which can be used to increase our knowledge concerning air pollution. The major disadvantage at the moment is that for one data-assimilation run, about a factor 20 more computer time is needed than for a single model run.

In the project "Entwicklung eines Modellsystems für das Zusammenspiel von Messung und Rechnung für bundeseinheitliche Umsetzung der EU-Rahmenrichtlinie Luftqualität", TNO is preparing a Kalman-Filter data-assimilation system around REM3/CALGRID.

5 Conclusions and recommendations

A new, complete and consistent emission data base for the base year 1995 has been made covering Europe with grids of 0.5 x 0.25 latlong containing the anthropogenic emissions of SO₂, NO_x, NMVOC, CO, CH₄, NH₃, PM₁₀, PM_{2.5}. A distinction has been made between area sources and point sources.

It turned out still to be problematic to combine in a straightforward manner, and in a flexible way, different sources of information and different databases. The main reason is that often these databases are not fully consistent with each other, leading still to many manual manipulations.

The new emission database is transparent, and can be applied in a flexible way. It turned out that the best way to check the emission database on errors made is to apply the data in an ACT-model and to analyse the results. It is recommended to test emission databases in this way before they are sent to other customers.

Although there is a great demand for independent information concerning the uncertainty of emission data, no systematic method is yet available. It is recommended to give attention to this aspect, in which data-assimilation could play a central role.

For the determination of biogenic NO and NMVOC-emissions, as well as for dry deposition calculations the land cover database chosen is of crucial importance. In the current project the PELINDA database has been used. It is recommended to investigate the sensitivity of the results by applying also other land cover data bases like CORINE, including a finer specification in different tree-species.

The REM3/CALGRID model has been improved in the course of this study by incorporating new findings and parametrisations. Testing new parametrisations, like the aerosol modelling, both in REM3/CALGRID and in LOTOS has the advantage that the new method is tested in two different models, which avoids major mistakes.

The challenge in aiming at models of intermediate complexity which are operational and can easily be run for many years, is to find the balance between incorporating new findings, and keeping it 'simple'.

So, it has to be investigated which processes are essential to incorporate and which can be left out. The attempt should be to include the necessary processes in a first order, parametrised way.

It is recommended that such parametrisations should be checked against more complete and complex models. It is also recommended that key model calculations, as for example to study the effectivity of proposed abatement strategies or the setting of blame-matrices, are carried out by more than one, and independent models. In this way the bandwidth of model results can be used in the further process.

ATC-models should continuously be updated using new findings, and a maintenance program should be set-up for that. A vital aspect of that is model validation- in which data-assimilation can play a role- and model intercomparison studies.

ATC-models have reached a certain stage of maturity. This means that the models can be used now to address and answer a broad range of both scientific and applied questions. It is a major challenge for the future to exploit and make full use of all the capabilities of the available models and model experience.

6 Presentations related to the project

Peter Bultjes and Rainer Stern.

Modelling and measurements of long-term values and critical loads for ozone:

AOT 40 and AOT 60

Conf. Atmospheric Environmental Research in Change; Where does air pollution control go ??, nov 15-17, 1999, Berlin, Aktuelle Reihe 6/2000

Peter Bultjes and Rainer Stern

Modellierung und Messung von Langfristwerten und kritischen Belastungsgrossen für Ozon (AOT40 und AOT60)

Poster-presentation VDI-symposium Tropospharisches Ozon, Braunschweig, February 8-10, 2000

Three presentations in the Eumetnet Workshop on groundlevel ozone forecasting. February. 21-23, 2000

1. *Peter Bultjes*
Modelling-an overview in the light of groundlevel ozone forecasting
2. *Tinus Pulles*
Availability and quality of European emission inventories
3. *Maarten van Loon*
Ozone data assimilation using a special Kalman Filter technique

Two presentations at the ITM on air pollution modelling and its application, Boulder, USA, May 2000

1. *Peter Bultjes*
Major twentieth century milestones in air pollution modelling and its application
2. *Maarten van Loon, Peter Bultjes and Arjo Segers*
Modelling and data assimilation of ozone

Peter Bultjes

Weiträumiger bis hemisphärischer Transport von Ozon und Vorläufersubstanzen. UBA Fachgespräch "Bodennahe Ozon", April 2002

Deliverables

Apart from this report, the emission database and the land cover data base PELINDA have been delivered on CD-Rom.

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Appendix A. Annual average anthropogenic emission totals per SNAP level 1 category per country

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
ALB	PM10	0	4	1	1	0	0	0	0	0	0	0	1	8
ARM	PM10	0	3	1	1	0	0	0	0	0	0	0	1	7
AUT	PM10	1	19	2	9	0	2	0	6	2	1	1	3	45
AZE	PM10	6	7	1	2	1	1	0	3	1	1	1	3	27
BEL	PM10	7	6	14	21	0	3	1	10	2	5	0	5	73
BGR	PM10	52	10	11	11	2	1	1	0	0	2	0	3	93
BIH	PM10	4	3	1	1	0	0	0	0	0	0	0	1	10
BLR	PM10	9	13	12	7	0	1	1	1	0	5	0	12	63
CHE	PM10	0	3	2	2	0	2	1	2	1	1	0	2	16
CYP	PM10	0	0	1	0	0	0	0	0	0	0	0	1	2
CZE	PM10	42	21	24	17	4	2	1	1	1	5	1	8	126
DEU	PM10	28	46	17	82	7	19	4	50	15	16	2	18	305
DNK	PM10	2	3	1	4	0	1	1	4	1	5	0	8	29
ESP	PM10	28	26	32	35	1	9	4	32	6	19	1	21	213
EST	PM10	21	4	4	1	1	0	1	0	0	0	0	1	33
FIN	PM10	1	8	3	6	0	1	0	3	1	3	0	2	30
FRA	PM10	14	113	45	50	1	14	5	46	10	17	3	35	352
GBR	PM10	48	14	30	54	5	12	5	26	9	12	1	14	229
GEO	PM10	0	5	0	2	0	1	0	0	0	0	0	2	10
GRC	PM10	11	9	10	6	1	3	2	6	2	7	1	5	62
HRV	PM10	4	4	4	2	0	1	0	1	0	2	0	3	21
HUN	PM10	18	13	4	7	1	1	1	1	0	2	1	10	59
IRL	PM10	3	4	3	2	0	1	0	2	1	2	0	4	22
ITA	PM10	26	67	37	56	0	11	9	38	10	17	3	16	291
LTU	PM10	3	4	4	2	0	0	1	0	0	1	0	3	20
LUX	PM10	0	0	0	3	0	0	0	0	0	0	0	1	5
LVA	PM10	2	3	1	2	0	0	0	0	0	1	0	2	13
MDA	PM10	4	5	0	2	0	0	0	0	0	1	0	3	16
MKD	PM10	20	2	3	1	0	0	0	0	0	1	0	1	27
MLT	PM10	0	0	0	0	0	0	0	0	0	0	0	0	1
NLD	PM10	4	2	2	16	0	5	1	9	2	5	0	9	55
NOR	PM10	0	3	1	6	26	1	0	2	1	6	0	1	47
POL	PM10	131	43	44	45	10	5	2	2	1	14	2	15	315
PRT	PM10	7	14	8	4	0	2	1	6	1	3	1	4	50
ROM	PM10	70	10	39	31	2	3	1	1	0	5	2	20	183
RUS	PM10	528	229	140	284	17	14	8	8	3	98	4	96	1429
SVK	PM10	7	6	7	9	0	1	0	1	0	1	0	4	37
SVN	PM10	5	2	1	1	0	0	0	1	0	0	0	1	13
SWE	PM10	1	7	4	11	0	2	1	4	2	3	0	2	37
TUR	PM10	102	69	110	44	3	13	3	9	3	18	3	14	391
UKR	PM10	156	47	184	132	4	5	4	0	1	31	2	44	609
YUG	PM10	100	12	16	4	2	1	0	0	0	0	1	8	145
ALB	PM2.5	0	4	0	0	0	0	0	0	0	0	0	1	6
ARM	PM2.5	0	3	0	0	0	0	0	0	0	0	0	0	5
AUT	PM2.5	1	17	1	3	0	1	0	6	1	0	1	1	32
AZE	PM2.5	2	7	1	1	1	1	0	3	0	1	1	1	18
BEL	PM2.5	4	5	8	10	0	2	1	10	1	4	0	1	46
BGR	PM2.5	13	8	6	5	0	1	1	0	0	2	0	1	38
BIH	PM2.5	1	3	0	0	0	0	0	0	0	0	0	0	6
BLR	PM2.5	3	11	8	3	0	1	1	1	0	5	0	6	39
CHE	PM2.5	0	2	1	1	0	1	1	2	0	1	0	0	11

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
CYP	PM2.5	0	0	0	0	0	0	0	0	0	0	0	0	1
CZE	PM2.5	11	15	11	6	0	1	1	1	0	4	1	5	57
DEU	PM2.5	21	36	10	27	2	12	4	50	6	15	2	4	189
DNK	PM2.5	2	3	1	1	0	1	1	4	0	4	0	3	19
ESP	PM2.5	17	25	19	13	0	6	4	32	2	18	1	10	147
EST	PM2.5	5	3	2	0	0	0	1	0	0	0	0	0	14
FIN	PM2.5	1	7	2	2	0	1	0	3	0	3	0	1	22
FRA	PM2.5	9	105	30	19	1	9	5	46	4	16	3	8	254
GBR	PM2.5	26	10	19	19	4	8	5	26	3	11	1	3	135
GEO	PM2.5	0	4	0	1	0	0	0	0	0	0	0	1	7
GRC	PM2.5	6	8	5	2	0	2	2	6	1	7	1	3	42
HRV	PM2.5	2	4	2	1	0	1	0	1	0	1	0	1	14
HUN	PM2.5	5	11	2	2	0	1	1	1	0	2	1	6	33
IRL	PM2.5	2	3	2	1	0	1	0	2	0	2	0	1	13
ITA	PM2.5	16	63	21	22	0	8	9	38	4	17	3	4	204
LTU	PM2.5	1	4	3	0	0	0	1	0	0	1	0	2	13
LUX	PM2.5	0	0	0	1	0	0	0	0	0	0	0	0	2
LVA	PM2.5	1	3	1	1	0	0	0	0	0	1	0	1	9
MDA	PM2.5	1	4	0	1	0	0	0	0	0	1	0	2	10
MKD	PM2.5	5	2	1	0	0	0	0	0	0	1	0	0	10
MLT	PM2.5	0	0	0	0	0	0	0	0	0	0	0	0	1
NLD	PM2.5	4	1	1	5	0	4	1	9	1	4	0	2	33
NOR	PM2.5	0	3	1	2	26	1	0	2	0	6	0	0	41
POL	PM2.5	34	28	22	14	1	4	2	2	0	13	2	4	127
PRT	PM2.5	4	13	5	1	0	1	1	6	1	3	1	1	36
ROM	PM2.5	19	9	26	14	0	3	1	1	0	5	2	11	90
RUS	PM2.5	149	185	78	141	9	11	8	8	1	92	4	52	736
SVK	PM2.5	2	4	4	4	0	1	0	1	0	1	0	2	20
SVN	PM2.5	1	2	1	0	0	0	0	1	0	0	0	0	7
SWE	PM2.5	1	7	3	3	0	1	1	4	1	3	0	0	25
TUR	PM2.5	29	59	52	18	0	9	3	9	1	17	3	4	205
UKR	PM2.5	36	28	86	66	1	4	4	0	0	29	2	25	282
YUG	PM2.5	24	10	7	1	0	1	0	0	0	0	1	4	49
ALB	CH4	0	0	0	0	3	0	0	0	0	0	0	64	66
ARM	CH4	0	0	0	0	0	0	0	0	0	0	0	0	0
AUT	CH4	0	15	0	0	5	0	3	0	0	0	220	336	580
AZE	CH4	0	0	0	0	0	0	0	0	0	0	0	0	0
BEL	CH4	0	4	1	3	45	0	8	1	0	0	185	354	602
BGR	CH4	1	17	1	2	322	0	1	0	0	0	36	125	506
BIH	CH4	0	4	0	2	19	0	0	0	0	0	0	93	119
BLR	CH4	2	39	3	65	1017	0	2	0	0	2	0	608	1738
CHE	CH4	0	3	0	1	13	0	3	0	0	1	70	222	313
CYP	CH4	0	0	0	0	0	0	0	0	0	0	0	0	0
CZE	CH4	5	25	2	12	405	0	1	0	0	0	144	139	733
DEU	CH4	9	60	12	9	1161	0	33	3	0	2	1900	1660	4848
DNK	CH4	2	7	1	1	16	0	2	0	0	1	75	323	428
ESP	CH4	9	40	7	4	629	0	11	1	0	2	628	928	2259
EST	CH4	0	4	0	0	8	0	0	0	0	0	0	30	43
FIN	CH4	2	6	3	4	0	0	2	0	0	1	137	90	245
FRA	CH4	2	135	8	5	307	0	18	2	0	0	622	1544	2642
GBR	CH4	13	38	6	0	843	0	22	2	0	3	1786	1104	3817
GEO	CH4	0	0	0	0	0	0	0	0	0	0	0	0	0
GRC	CH4	1	9	2	0	49	0	4	0	0	0	115	285	465
HRV	CH4	0	5	0	1	3	0	0	0	0	0	29	65	103

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
HUN	CH4	3	20	3	1	235	0	1	0	0	0	0	340	603
IRL	CH4	0	2	0	0	11	0	2	0	0	0	135	663	813
ITA	CH4	40	24	7	9	351	0	36	5	0	3	1029	1052	2555
LTU	CH4	1	14	1	23	366	0	1	0	0	1	0	219	625
LUX	CH4	0	0	0	0	2	0	0	0	0	0	3	17	22
LVA	CH4	1	9	1	16	248	0	1	0	0	1	0	148	423
MDA	CH4	1	16	1	27	427	0	1	0	0	1	0	255	730
MKD	CH4	0	3	0	2	15	0	0	0	0	0	0	71	91
MLT	CH4	0	0	0	0	0	0	0	0	0	0	6	0	6
NLD	CH4	4	21	5	5	172	0	5	1	0	1	482	476	1171
NOR	CH4	3	13	0	1	28	0	2	0	0	1	330	108	487
POL	CH4	1	9	2	0	694	0	7	2	0	1	428	660	1803
PRT	CH4	0	8	3	1	0	0	2	0	0	1	605	195	816
ROM	CH4	1	12	4	7	255	0	1	0	0	0	127	403	809
RUS	CH4	29	405	40	680	10681	0	25	3	0	24	0	8717	20605
SVK	CH4	14	0	0	6	107	0	1	0	0	0	63	122	313
SVN	CH4	0	1	0	0	44	0	0	0	0	0	82	39	166
SWE	CH4	0	0	1	0	0	0	15	2	0	2	61	194	276
TUR	CH4	0	0	0	0	0	0	0	0	0	0	0	0	0
UKR	CH4	10	193	14	323	5074	0	12	1	0	11	0	3032	8671
YUG	CH4	1	15	1	10	78	0	2	1	0	0	0	381	487
ALB	CO	0	100	0	0	0	0	56	5	0	0	0	0	161
ARM	CO	0	0	0	0	0	0	0	0	0	0	0	0	0
AUT	CO	1	460	7	320	0	0	327	12	0	14	4	2	1147
AZE	CO	0	0	0	0	0	0	0	0	0	0	0	0	0
BEL	CO	4	99	321	153	0	0	827	88	0	8	17	0	1515
BGR	CO	5	287	6	151	0	0	337	19	0	10	28	2	844
BIH	CO	2	58	1	20	0	0	152	17	0	0	0	0	249
BLR	CO	28	572	31	319	0	0	1055	51	0	36	0	0	2091
CHE	CO	1	62	15	11	0	0	303	5	0	102	11	0	510
CYP	CO	0	0	0	0	0	0	0	0	0	0	0	0	0
CZE	CO	47	288	269	3	0	0	226	21	0	19	0	0	874
DEU	CO	142	1176	676	595	13	0	3742	211	0	242	0	0	6797
DNK	CO	9	96	3	0	44	0	344	28	0	62	1	0	588
ESP	CO	18	835	394	225	0	0	2434	205	0	109	304	122	4645
EST	CO	13	88	4	0	0	0	202	10	0	25	0	0	343
FIN	CO	8	67	43	10	0	0	277	27	0	3	0	0	436
FRA	CO	13	1677	546	603	0	0	4638	297	0	414	232	0	8421
GBR	CO	244	233	49	6	48	0	3969	143	0	783	3	0	5478
GEO	CO	0	0	0	0	0	0	0	0	0	0	0	0	0
GRC	CO	7	156	15	25	0	0	893	62	0	109	24	127	1418
HRV	CO	1	115	1	9	0	0	298	32	0	23	0	0	479
HUN	CO	5	150	56	79	0	0	406	72	0	0	0	0	767
IRL	CO	4	56	2	0	0	0	224	12	0	6	1	0	304
ITA	CO	30	346	506	299	0	0	5260	465	0	543	281	25	7755
LTU	CO	10	206	11	115	0	0	379	18	0	13	0	0	752
LUX	CO	0	8	44	9	0	0	39	2	0	2	0	0	105
LVA	CO	7	139	8	78	0	0	257	12	0	9	0	0	509
MDA	CO	12	240	13	134	0	0	443	21	0	15	0	0	878
MKD	CO	1	44	1	15	0	0	116	13	0	0	0	0	190
MLT	CO	0	0	0	0	0	0	19	2	0	2	0	0	24
NLD	CO	19	106	129	94	0	0	470	38	0	36	2	0	892
NOR	CO	6	146	7	49	0	0	451	17	0	63	1	0	739
POL	CO	50	2391	21	37	0	0	1065	79	0	125	782	0	4548

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
PRT	CO	3	266	31	54	0	0	815	105	0	14	2	0	1290
ROM	CO	9	423	574	36	0	0	568	64	0	38	1211	0	2924
RUS	CO	395	6005	436	3348	0	0	11075	535	0	379	0	0	22173
SVK	CO	7	54	159	0	0	0	164	16	0	0	0	0	400
SVN	CO	1	4	1	0	0	0	75	8	0	2	0	0	91
SWE	CO	1	0	5	12	0	0	710	27	0	108	13	0	877
TUR	CO	0	0	0	0	0	0	0	0	0	0	0	0	0
UKR	CO	137	2853	154	1590	0	0	5261	254	0	180	0	0	10430
YUG	CO	8	237	3	82	0	0	622	68	0	1	0	0	1021
ALB	NH3	0	0	0	0	0	0	0	0	0	0	0	31	31
ARM	NH3	0	0	0	0	0	0	0	0	0	0	0	0	0
AUT	NH3	0	1	0	0	0	0	4	0	0	0	0	74	79
AZE	NH3	0	0	0	0	0	0	0	0	0	0	0	0	0
BEL	NH3	0	0	0	3	0	0	0	0	0	0	0	92	96
BGR	NH3	0	0	0	24	0	0	0	0	0	0	28	48	100
BIH	NH3	0	0	0	0	0	0	0	0	0	0	0	25	25
BLR	NH3	0	0	0	2	0	0	0	0	0	0	0	71	73
CHE	NH3	0	0	0	0	0	0	3	0	0	0	4	53	60
CYP	NH3	0	0	0	0	0	0	0	0	0	0	0	0	0
CZE	NH3	0	0	0	1	0	0	0	0	0	0	0	136	137
DEU	NH3	3	0	1	8	0	1	18	0	0	0	0	591	623
DNK	NH3	0	0	0	0	0	0	1	0	0	0	0	98	99
ESP	NH3	0	0	0	10	0	0	1	0	0	0	0	333	344
EST	NH3	0	0	0	0	0	0	0	0	0	0	0	12	12
FIN	NH3	0	0	0	1	0	0	0	0	0	0	0	34	35
FRA	NH3	0	0	0	27	0	0	4	1	0	0	4	759	794
GBR	NH3	4	0	0	0	0	0	9	1	0	0	10	299	324
GEO	NH3	0	0	0	0	0	0	0	0	0	0	0	0	0
GRC	NH3	0	0	0	4	0	0	0	0	0	0	0	106	110
HRV	NH3	0	0	0	2	0	0	0	0	0	0	0	23	25
HUN	NH3	0	0	0	4	0	0	0	0	0	0	0	57	62
IRL	NH3	0	0	0	0	0	0	0	0	0	0	0	124	124
ITA	NH3	0	0	0	11	0	0	3	1	0	0	18	427	461
LTU	NH3	0	0	0	1	0	0	0	0	0	0	0	26	26
LUX	NH3	0	0	0	2	0	0	0	0	0	0	0	5	7
LVA	NH3	0	0	0	0	0	0	0	0	0	0	0	17	18
MDA	NH3	0	0	0	1	0	0	0	0	0	0	0	30	31
MKD	NH3	0	0	0	0	0	0	0	0	0	0	0	19	19
MLT	NH3	0	0	0	0	0	0	0	0	0	0	0	0	0
NLD	NH3	0	0	0	4	0	1	0	0	0	0	0	141	147
NOR	NH3	0	0	0	0	0	0	1	0	0	0	0	25	26
POL	NH3	0	0	0	6	0	0	0	0	0	0	16	356	379
PRT	NH3	0	0	0	7	0	0	0	0	0	0	11	78	96
ROM	NH3	0	0	0	3	0	0	0	0	0	0	0	201	204
RUS	NH3	0	0	0	17	0	0	0	0	0	0	0	1018	1035
SVK	NH3	0	0	0	0	0	0	0	0	0	0	0	30	30
SVN	NH3	0	0	0	0	0	0	0	0	0	0	0	22	22
SWE	NH3	0	0	0	0	0	0	3	0	0	0	0	55	59
TUR	NH3	0	0	0	0	0	0	0	0	0	0	0	0	0
UKR	NH3	0	0	0	8	0	0	0	0	0	0	0	354	362
YUG	NH3	0	0	0	1	0	0	0	0	0	0	0	104	104
ALB	VOC	0	10	0	19	0	21	7	2	4	0	0	0	62
ARM	VOC	0	0	0	0	0	0	0	0	0	0	0	0	0
AUT	VOC	0	44	1	26	6	132	35	4	27	5	1	2	283

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
AZE	VOC	0	0	0	0	0	0	0	0	0	0	0	0	0
BEL	VOC	4	8	2	30	15	87	124	26	12	6	0	1	316
BGR	VOC	1	17	2	22	2	18	56	6	10	4	4	0	141
BIH	VOC	1	4	0	30	0	13	13	5	5	0	0	0	69
BLR	VOC	5	31	4	666	0	75	90	8	20	2	0	0	901
CHE	VOC	0	2	0	14	6	113	26	1	21	14	4	8	211
CYP	VOC	0	0	0	0	0	0	0	0	0	0	0	0	0
CZE	VOC	5	37	12	28	4	125	41	7	11	16	0	0	286
DEU	VOC	9	60	11	136	88	1090	310	65	302	74	0	0	2145
DNK	VOC	2	10	1	10	7	21	35	6	27	13	1	0	131
ESP	VOC	10	55	12	74	58	303	352	45	84	39	49	0	1080
EST	VOC	1	5	0	5	1	5	29	3	7	2	2	0	60
FIN	VOC	0	33	0	15	7	35	39	11	31	14	2	0	186
FRA	VOC	4	195	10	87	102	615	588	79	311	140	28	15	2174
GBR	VOC	98	32	9	292	334	656	395	29	266	121	26	0	2257
GEO	VOC	0	0	0	0	0	0	0	0	0	0	0	0	0
GRC	VOC	3	13	2	9	16	82	160	23	63	23	9	47	450
HRV	VOC	0	7	0	8	5	21	17	6	6	1	0	0	72
HUN	VOC	1	19	4	4	5	36	61	9	10	0	0	0	148
IRL	VOC	0	5	0	1	4	20	29	2	27	2	0	59	149
ITA	VOC	7	29	9	94	128	629	770	124	222	175	27	2	2216
LTU	VOC	2	11	2	239	0	27	32	3	7	1	0	0	324
LUX	VOC	0	1	0	1	2	4	6	1	1	1	0	0	17
LVA	VOC	1	8	1	162	0	18	22	2	5	0	0	0	219
MDA	VOC	2	13	2	280	0	31	38	4	9	1	0	0	379
MKD	VOC	0	3	0	23	0	10	10	4	4	0	0	0	52
MLT	VOC	0	0	0	0	0	2	2	0	1	0	0	0	6
NLD	VOC	4	11	6	71	35	82	67	19	54	12	2	0	365
NOR	VOC	2	10	1	21	206	44	49	4	11	18	1	0	366
POL	VOC	13	157	11	70	44	129	226	31	11	45	1	34	770
PRT	VOC	1	27	8	63	0	0	89	16	26	6	8	4	247
ROM	VOC	1	16	7	45	73	135	44	9	19	11	68	49	477
RUS	VOC	71	324	63	6989	0	786	945	88	215	16	0	0	9498
SVK	VOC	0	0	0	0	0	0	28	5	7	1	0	0	41
SVN	VOC	2	1	0	4	3	9	13	5	5	0	0	0	42
SWE	VOC	0	0	4	33	0	98	102	8	22	34	0	2	303
TUR	VOC	0	0	0	0	0	0	0	0	0	0	0	0	0
UKR	VOC	25	154	22	3320	0	373	449	42	102	7	0	0	4495
YUG	VOC	2	15	1	121	0	52	52	19	19	0	0	0	282
ALB	NOx	21	5	20	2	0	0	1	2	0	1	0	0	52
ARM	NOx	0	0	0	0	0	0	0	0	0	0	0	0	0
AUT	NOx	7	19	14	20	0	0	40	52	0	17	0	7	176
AZE	NOx	0	0	0	0	0	0	0	0	0	0	0	0	0
BEL	NOx	66	15	58	11	0	0	102	89	0	5	4	0	350
BGR	NOx	74	4	29	43	0	0	33	53	0	23	2	4	266
BIH	NOx	333	6	7	15	0	0	1	2	0	1	0	0	364
BLR	NOx	365	42	17	27	0	0	5	3	0	7	0	0	467
CHE	NOx	2	14	12	0	0	0	55	18	0	24	6	3	136
CYP	NOx	0	0	0	0	0	0	0	0	0	0	0	0	0
CZE	NOx	112	34	71	2	0	0	42	136	0	16	0	0	412
DEU	NOx	508	139	257	23	0	0	748	298	0	294	0	0	2266
DNK	NOx	91	7	11	1	0	0	50	32	0	57	2	0	250
ESP	NOx	243	22	168	10	0	0	257	252	0	217	15	1	1185
EST	NOx	14	1	2	0	0	0	13	8	0	9	0	0	48

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
FIN	NOx	40	7	34	8	0	0	98	73	0	0	0	0	260
FRA	NOx	117	99	151	16	0	0	536	421	0	362	23	0	1726
GBR	NOx	557	103	143	40	112	0	815	247	0	273	5	0	2295
GEO	NOx	0	0	0	0	0	0	0	0	0	0	0	0	0
GRC	NOx	79	5	22	35	0	0	57	65	0	86	2	5	356
HRV	NOx	7	3	9	2	0	0	5	17	0	11	0	0	55
HUN	NOx	33	26	22	14	0	0	20	75	0	0	0	0	190
IRL	NOx	42	8	10	0	0	0	31	14	0	9	0	0	115
ITA	NOx	376	63	205	10	0	0	425	543	0	211	15	1	1849
LTU	NOx	62	27	6	9	0	0	2	1	0	8	0	0	115
LUX	NOx	0	1	8	0	0	0	6	4	0	1	0	0	20
LVA	NOx	41	30	11	0	0	0	2	1	0	8	0	0	93
MDA	NOx	66	26	13	0	0	0	1	0	0	3	0	0	108
MKD	NOx	81	2	10	1	0	0	0	1	0	0	0	0	95
MLT	NOx	7	0	0	0	0	0	2	2	0	6	0	0	16
NLD	NOx	66	42	64	9	0	0	114	111	0	89	3	0	498
NOR	NOx	26	3	9	8	0	0	51	16	0	96	6	0	214
POL	NOx	413	116	161	17	0	0	139	180	0	95	0	0	1122
PRT	NOx	82	19	27	5	0	0	89	100	0	43	1	0	366
ROM	NOx	143	18	44	9	0	0	17	39	0	47	20	5	342
RUS	NOx	1168	338	396	309	0	0	49	31	0	46	0	0	2336
SVK	NOx	95	10	23	0	0	0	11	36	0	6	0	0	181
SVN	NOx	17	2	3	0	0	0	10	33	0	2	0	0	67
SWE	NOx	17	10	22	16	0	0	95	33	0	149	0	0	341
TUR	NOx	0	0	0	0	0	0	0	0	0	0	0	0	0
UKR	NOx	1164	374	162	123	0	0	28	17	0	25	0	0	1894
YUG	NOx	81	2	5	9	0	0	14	45	0	0	0	0	156
ALB	SOx	3	1	4	1	0	0	2	14	0	0	0	0	24
ARM	SOx	0	0	0	0	0	0	0	0	0	0	0	0	0
AUT	SOx	5	19	12	12	2	0	1	7	0	2	0	0	60
AZE	SOx	0	0	0	0	0	0	0	0	0	0	0	0	0
BEL	SOx	108	34	54	36	0	0	2	15	0	1	3	0	252
BGR	SOx	1258	85	77	32	0	0	1	5	0	19	21	0	1497
BIH	SOx	30	1	1	4	0	0	2	17	0	0	0	0	54
BLR	SOx	115	12	13	15	0	0	9	83	0	0	0	0	246
CHE	SOx	2	15	9	4	0	0	0	2	0	1	3	0	34
CYP	SOx	0	0	0	0	0	0	0	0	0	0	0	0	0
CZE	SOx	798	142	140	3	0	0	1	6	0	1	0	0	1091
DEU	SOx	2066	365	415	67	17	0	9	42	0	16	0	0	2998
DNK	SOx	107	10	19	3	0	0	0	2	0	8	0	0	149
ESP	SOx	1245	74	483	57	0	0	12	54	0	53	31	0	2008
EST	SOx	96	3	16	0	0	0	1	5	0	3	0	0	123
FIN	SOx	39	8	26	20	0	0	0	2	0	2	0	0	96
FRA	SOx	359	87	266	73	15	0	34	83	0	17	15	0	947
GBR	SOx	1798	130	282	31	2	0	18	33	0	68	4	0	2365
GEO	SOx	0	0	0	0	0	0	0	0	0	0	0	0	0
GRC	SOx	400	93	65	10	0	0	2	20	0	19	0	0	609
HRV	SOx	25	3	27	0	0	0	0	4	0	3	0	0	63
HUN	SOx	421	289	106	9	0	0	6	52	0	0	0	0	883
IRL	SOx	93	28	33	0	0	0	1	4	0	2	0	0	161
ITA	SOx	823	44	241	76	0	0	6	96	0	22	14	0	1322
LTU	SOx	20	5	3	12	0	0	7	61	0	0	0	0	108
LUX	SOx	0	1	6	0	0	0	0	1	0	0	0	0	9
LVA	SOx	18	7	3	3	0	0	6	52	0	0	0	0	88

country	species	1. Power generation	2. Residential, commercial and other combustion	3. Industrial combustion	4. Industrial processes	5. Extraction distribution of fossil fuels	6. Solvent use	7a. Road transport gasoline	7b. Road transport diesel	7c. Road transport evaporation	8. Other mobile sources and machinery	9. Waste treatment and disposal	10. agriculture	total
MDA	SOx	31	4	3	0	0	0	2	19	0	0	0	0	60
MKD	SOx	17	1	3	0	0	0	1	9	0	0	0	0	30
MLT	SOx	12	0	0	0	0	0	0	0	0	3	0	0	16
NLD	SOx	38	5	49	23	0	0	1	13	0	17	1	0	147
NOR	SOx	1	2	5	22	0	0	0	1	0	3	0	0	34
POL	SOx	1325	509	421	0	0	0	2	19	0	0	0	0	2276
PRT	SOx	222	8	81	26	0	0	1	11	0	7	1	0	358
ROM	SOx	823	128	67	21	0	0	1	13	0	10	0	0	1064
RUS	SOx	31	3	3	0	0	0	2	14	0	0	0	0	53
SVK	SOx	164	48	24	0	0	0	0	2	0	0	0	0	238
SVN	SOx	100	9	8	0	0	0	0	1	0	0	0	0	119
SWE	SOx	17	7	20	14	0	0	0	1	0	23	0	0	82
TUR	SOx	0	0	0	0	0	0	0	0	0	0	0	0	0
UKR	SOx	567	74	117	52	0	0	48	431	0	0	0	0	1287
YUG	SOx	84	2	5	6	0	0	6	52	0	0	0	0	155

Appendix B. VOC-speciation

Country code(s)	Sector name	acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & al- kynes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
ALB	1	0.0	0.0	3.0	17.5	0.0	0.0	6.8	8.3	0.0	0.2	17.5	0.0	0.0	0.6	0.0	0.4	0.0	21.9	17.7	1.5	2.8	0.0	1.8
AUT	1	0.5	0.3	2.9	17.2	0.0	0.0	6.5	7.5	0.0	0.2	17.3	0.1	0.5	0.8	0.1	0.6	0.5	21.4	17.0	1.4	2.9	0.0	2.2
BEL	1	0.2	0.0	3.5	16.0	0.0	0.0	7.5	11.6	0.0	0.4	16.1	0.0	0.3	0.8	0.0	0.6	0.4	20.1	16.0	1.7	2.8	0.0	2.1
BGR	1	0.7	0.2	2.6	17.9	0.0	0.0	5.3	3.8	0.0	0.2	18.4	0.1	0.8	1.1	0.1	1.1	0.1	22.4	17.4	1.2	3.3	0.0	3.3
CHE	1	0.0	0.0	1.9	17.8	0.0	0.0	4.4	0.9	0.0	0.0	17.6	0.0	0.3	0.4	0.0	0.4	10.8	22.4	17.6	0.9	2.7	0.0	1.8
CYP	1	2.0	2.0	1.9	18.7	0.0	0.0	4.5	0.9	0.0	0.0	18.9	0.5	1.5	0.5	0.5	0.5	0.7	23.0	18.1	0.9	3.0	0.1	1.9
CZE, SVK	1	1.0	0.2	3.9	13.5	0.0	0.0	8.3	15.3	0.0	0.6	14.9	0.0	0.4	1.6	0.0	1.4	0.1	16.5	13.4	2.0	3.0	0.0	3.9
DEU New Länder	1	1.1	0.1	2.6	13.7	0.0	0.0	5.5	6.6	0.0	0.3	15.6	0.0	0.3	1.8	0.0	1.8	11.4	16.6	13.4	1.3	3.1	0.0	4.8
DEU Old Länder	1	0.6	0.1	3.4	15.6	0.0	0.0	7.4	11.2	0.0	0.4	16.4	0.0	0.2	1.1	0.0	1.0	0.1	19.3	15.6	1.7	2.9	0.0	2.9
DNK	1	0.2	0.0	2.0	19.3	0.0	0.0	4.8	1.0	0.0	0.0	19.7	0.0	0.1	0.7	0.0	0.8	1.4	24.0	19.2	1.0	3.1	0.0	2.6
ESP	1	0.6	0.2	2.5	17.7	0.0	0.0	5.8	4.9	0.0	0.2	18.5	0.0	0.3	1.0	0.0	1.0	0.1	21.9	17.6	1.3	3.1	0.0	3.1
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	1	0.1	0.1	2.1	19.9	0.0	0.0	4.9	1.0	0.0	0.0	19.7	0.0	0.3	0.5	0.0	0.5	0.0	25.0	19.7	1.0	3.1	0.0	2.1
FIN	1	0.9	0.5	2.2	17.5	0.0	0.0	4.9	2.6	0.1	0.2	17.7	0.1	0.4	0.8	0.2	0.7	5.5	21.8	17.3	1.1	2.9	0.0	2.4
FRA	1	0.2	0.1	3.0	17.4	0.0	0.0	6.8	8.1	0.0	0.3	17.5	0.0	0.1	0.7	0.0	0.6	0.0	21.6	17.4	1.5	2.8	0.0	2.0
GBR	1	1.0	0.3	2.6	16.9	0.0	0.0	5.9	5.9	0.0	0.2	18.1	0.1	0.4	1.3	0.1	1.3	0.1	20.7	16.6	1.3	3.2	0.0	3.8
GRC	1	0.6	0.2	2.0	19.2	0.0	0.0	4.8	1.0	0.0	0.0	19.8	0.1	0.3	0.9	0.1	0.9	0.3	23.8	19.0	1.0	3.2	0.0	2.9
HUN	1	1.2	0.8	2.3	17.9	0.0	0.0	5.3	3.5	0.0	0.1	18.5	0.2	0.8	1.0	0.2	0.9	0.3	22.0	17.5	1.1	3.1	0.1	3.0
IRL	1	2.8	1.3	3.2	17.0	0.0	0.0	3.9	1.3	0.2	0.3	16.3	0.2	3.4	1.7	0.5	1.5	0.3	22.1	15.0	1.0	3.8	0.1	4.0
ITA	1	0.6	0.6	2.4	18.7	0.0	0.0	5.4	3.3	0.0	0.1	18.6	0.1	0.7	0.6	0.1	0.5	0.3	23.4	18.4	1.1	3.0	0.0	2.0
NLD	1	0.0	0.0	2.2	17.0	0.0	0.0	5.3	3.9	0.0	0.1	17.0	0.0	0.0	0.5	0.0	0.4	9.6	21.3	17.1	1.1	2.6	0.0	1.7
NOR	1	0.0	0.0	2.0	20.0	0.0	0.0	5.0	1.0	0.0	0.0	20.0	0.0	0.0	0.5	0.0	0.5	0.2	24.9	20.0	1.0	3.0	0.0	2.0
POL	1	0.9	0.0	4.6	11.8	0.0	0.0	9.5	20.0	0.0	0.8	13.4	0.0	0.2	1.7	0.0	1.5	0.0	14.3	11.8	2.3	2.9	0.0	4.1
PRT	1	0.6	0.5	2.0	19.4	0.0	0.0	4.8	1.0	0.0	0.0	19.7	0.1	0.4	0.7	0.1	0.7	0.2	24.1	19.2	1.0	3.1	0.0	2.4
ROM	1	1.9	1.5	2.9	16.2	0.0	0.0	5.8	6.8	0.0	0.3	16.6	0.4	1.7	1.1	0.4	1.0	0.6	19.9	15.3	1.3	3.1	0.1	3.0
SVN, MKD, YUG, HRV, BIH	1	1.6	0.8	2.5	16.7	0.0	0.0	5.4	4.6	0.0	0.2	18.2	0.2	0.8	1.5	0.2	1.5	0.3	20.3	16.2	1.2	3.4	0.1	4.3
SWE	1	0.2	0.1	2.5	18.2	0.0	0.0	5.8	4.8	0.0	0.1	18.3	0.0	0.1	0.6	0.0	0.5	1.6	22.7	18.2	1.2	2.9	0.0	2.0
ALB	2	2.9	1.4	8.8	17.8	0.0	0.0	0.7	1.3	0.6	0.5	3.7	0.1	16.2	0.7	0.7	0.1	0.0	31.2	8.2	0.6	4.8	0.0	0.1
AUT	2	21.5	8.8	6.4	2.5	0.0	0.0	7.5	15.2	3.4	4.3	1.2	0.9	2.2	5.4	5.0	0.5	0.3	2.5	3.9	4.8	2.6	0.0	0.9
BEL	2	13.4	3.1	5.9	7.0	0.0	0.0	9.8	20.5	0.5	4.0	1.9	1.1	3.7	4.1	2.5	0.2	1.0	6.9	8.1	3.6	2.1	0.1	0.8

Country code(s)	Sector name	Chemical categories																						
		acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & al- kenes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
BGR	2	18.7	4.1	5.6	4.0	0.0	0.0	11.5	23.4	1.3	4.7	0.8	0.7	1.0	5.1	2.6	0.2	0.4	1.2	7.3	4.7	1.7	0.0	0.9
CHE	2	1.0	12.7	6.3	6.5	0.0	0.0	0.8	8.0	0.0	3.4	7.6	6.3	5.6	3.4	12.7	0.6	6.3	11.0	3.4	0.3	2.9	0.6	0.7
CYP	2	0.0	5.1	8.0	14.9	0.0	0.0	0.0	2.5	0.0	1.3	5.5	2.5	13.4	1.3	5.1	0.3	2.5	26.1	16.7	0.0	4.2	0.3	0.3
CZE, SVK	2	17.8	1.1	5.3	5.1	0.0	0.0	13.7	27.5	0.4	4.8	0.3	0.2	0.7	4.9	0.7	0.1	0.1	1.2	9.3	4.8	1.3	0.0	1.0
DEU New Länder	2	0.0	0.0	9.0	20.0	0.0	0.0	0.0	0.0	0.0	0.0	4.0	0.0	18.0	0.0	0.0	0.0	0.0	35.0	9.0	0.0	5.0	0.0	0.0
DEU Old Länder	2	1.6	0.7	8.8	18.7	0.0	0.0	0.4	0.8	0.3	0.3	3.8	0.0	16.9	0.4	0.4	0.0	0.0	32.8	8.6	0.4	4.9	0.0	0.1
DNK	2	23.8	11.1	6.7	1.3	0.0	0.0	6.1	12.1	4.4	4.2	1.2	0.9	2.3	5.7	6.2	0.6	0.2	1.9	2.3	5.1	2.9	0.0	0.9
ESP	2	19.4	7.6	6.2	3.3	0.0	0.0	8.3	17.1	2.5	4.3	1.5	1.1	2.1	5.1	4.7	0.4	0.7	2.7	4.9	4.5	2.3	0.1	0.9
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	2	20.8	9.8	7.2	4.1	0.0	0.0	4.7	9.2	4.1	3.5	1.5	0.7	4.9	4.8	5.2	0.6	0.0	7.3	2.9	4.4	3.4	0.0	0.7
FIN	2	23.5	12.4	6.7	0.6	0.0	0.0	5.5	11.3	4.6	4.3	1.6	1.4	1.9	5.8	7.2	0.7	0.7	1.2	1.6	5.0	2.9	0.1	0.9
FRA	2	19.7	7.9	6.4	3.7	0.0	0.0	7.5	15.3	2.9	4.1	1.5	0.9	3.0	5.0	4.6	0.5	0.5	4.1	4.6	4.5	2.6	0.0	0.8
GBR	2	14.5	1.4	6.1	7.8	0.0	0.0	10.6	21.2	0.5	3.8	1.1	0.2	4.2	4.0	0.9	0.1	0.1	7.9	8.8	3.9	2.1	0.0	0.8
GRC	2	23.1	12.2	6.6	0.7	0.0	0.0	5.7	11.9	4.4	4.4	1.7	1.5	1.8	5.8	7.2	0.7	0.8	1.1	1.8	4.9	2.9	0.1	0.9
HUN	2	17.7	2.5	5.6	5.2	0.0	0.0	12.0	24.1	0.8	4.5	0.7	0.3	1.7	4.8	1.5	0.1	0.2	2.8	8.3	4.6	1.6	0.0	0.9
IRL	2	22.0	7.0	6.0	2.5	0.0	0.0	9.6	19.1	2.8	4.6	0.7	0.5	1.2	5.5	3.8	0.4	0.1	0.8	5.2	5.2	2.2	0.0	0.9
ITA	2	14.6	8.5	7.3	7.0	0.0	0.0	3.5	7.8	2.8	2.9	2.8	1.3	7.1	3.8	5.4	0.5	0.9	12.3	4.1	3.1	3.5	0.1	0.6
LUX	2	4.2	6.7	6.7	9.7	0.0	0.0	3.5	10.4	0.0	2.9	5.1	3.4	7.7	2.9	6.7	0.3	3.4	15.0	6.2	1.2	3.1	0.3	0.6
NLD	2	0.4	0.2	9.0	19.7	0.0	0.0	0.1	0.2	0.1	0.1	4.0	0.0	17.8	0.1	0.1	0.0	0.0	34.5	8.9	0.1	5.0	0.0	0.0
NOR	2	8.0	11.1	5.0	2.3	0.0	0.0	6.7	18.9	0.0	5.0	5.5	5.5	0.0	5.0	11.1	0.6	5.5	0.1	4.5	2.2	1.6	0.6	1.0
POL	2	18.0	0.6	5.1	5.0	0.0	0.0	14.3	28.6	0.2	4.9	0.2	0.1	0.4	5.0	0.4	0.0	0.0	0.6	9.6	4.9	1.2	0.0	1.0
PRT	2	24.2	12.0	6.8	0.9	0.0	0.0	5.5	11.1	4.7	4.2	1.3	1.1	2.3	5.8	6.7	0.7	0.3	1.7	1.7	5.1	3.0	0.0	0.9
ROM	2	20.3	4.7	5.5	4.5	0.0	0.0	10.6	20.9	1.4	4.2	0.7	0.4	2.0	4.6	2.1	0.2	0.1	3.1	7.1	4.8	1.8	0.0	0.8
SVN, MKD, YUG, HRV, BIH	2	18.8	3.0	5.6	4.6	0.0	0.0	12.0	24.0	1.2	4.6	0.5	0.3	1.3	5.0	1.7	0.2	0.1	1.9	7.9	4.8	1.7	0.0	0.9
SWE	2	20.4	11.5	6.3	1.3	0.0	0.0	6.1	13.4	3.5	4.4	2.3	2.1	1.6	5.6	7.5	0.6	1.5	1.2	2.5	4.5	2.6	0.1	0.9
ALB	3	25.5	12.0	6.8	0.5	0.0	0.0	5.8	11.3	5.0	4.3	1.0	0.8	2.0	5.9	6.4	0.7	0.0	1.1	1.6	5.4	3.0	0.0	0.9
AUT	3	21.2	10.1	6.1	2.1	0.0	0.0	4.7	9.0	4.0	3.5	2.3	0.8	3.2	5.2	5.2	1.1	6.7	3.2	1.9	4.3	3.2	0.0	2.1
BEL	3	9.8	3.8	4.4	8.1	0.0	0.0	1.9	2.4	0.8	1.2	10.8	0.6	7.1	5.8	1.5	5.3	3.0	10.3	3.6	1.4	5.7	0.1	12.4
BGR	3	11.7	4.4	3.8	6.1	0.0	0.0	2.4	3.0	1.1	1.5	11.8	0.6	5.2	6.8	1.8	6.2	2.4	6.3	2.7	1.7	5.9	0.1	14.5
CHE	3	10.1	4.9	3.4	4.8	0.0	0.0	1.9	2.9	1.2	1.3	6.6	0.7	5.0	4.1	2.0	3.1	27.4	6.0	2.2	1.5	3.7	0.2	7.2
CYP	3	10.5	4.1	1.8	5.4	0.0	0.0	1.6	0.8	0.0	0.8	16.8	1.0	4.7	8.1	1.0	8.7	1.4	2.5	1.8	0.8	6.9	0.3	20.9
CZE, SVK	3	14.3	7.1	6.8	8.2	0.0	0.0	3.0	5.7	2.5	2.2	3.3	0.7	8.5	3.5	3.5	0.8	3.7	13.7	4.2	2.8	3.8	0.1	1.7

Country code(s)	Sector name	Sector name																						
		acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & al- kenes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
DEU New Länder	3	1.4	0.9	1.2	3.0	0.0	0.0	0.1	0.1	0.0	0.1	1.9	0.2	2.9	0.6	0.2	0.6	78.3	4.5	1.2	0.1	1.2	0.1	1.5
DEU Old Länder	3	9.1	3.5	4.1	7.8	0.0	0.0	1.8	2.0	0.7	1.1	10.9	0.6	6.9	5.6	1.3	5.3	6.6	9.8	3.4	1.2	5.6	0.1	12.5
DNK	3	20.5	9.3	5.6	1.7	0.0	0.0	4.6	8.6	3.7	3.4	3.8	0.7	2.4	5.9	4.9	2.1	6.9	2.0	1.7	4.2	3.6	0.0	4.5
ESP	3	15.2	9.0	3.9	4.6	0.0	0.0	2.4	4.3	1.8	1.7	5.8	1.5	6.3	4.0	3.5	2.0	16.0	5.1	2.0	2.1	3.5	0.3	4.8
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	3	10.6	3.6	4.5	7.4	0.0	0.0	2.3	2.9	1.1	1.4	11.3	0.4	6.2	6.5	1.6	5.8	0.4	9.2	3.4	1.7	6.0	0.1	13.7
FIN	3	12.4	5.8	3.4	1.3	0.0	0.0	2.7	5.1	2.2	2.0	2.2	0.5	1.9	3.4	3.0	1.2	43.0	1.6	1.1	2.5	2.1	0.0	2.5
FRA	3	19.6	8.9	5.6	2.9	0.0	0.0	4.3	7.8	3.4	3.1	5.2	0.7	3.5	6.1	4.5	2.8	2.0	3.6	2.1	3.8	4.1	0.1	6.0
GBR	3	10.2	4.0	4.8	7.7	0.0	0.0	2.1	3.1	1.2	1.4	8.7	0.5	6.9	5.1	1.8	4.2	7.7	10.6	3.6	1.6	5.1	0.1	9.7
GRC	3	21.4	9.9	5.5	1.8	0.0	0.0	4.7	8.5	3.7	3.4	5.0	0.9	2.8	6.4	5.0	2.7	1.3	1.6	1.7	4.2	3.9	0.1	5.9
HUN	3	19.5	9.1	6.8	4.3	0.0	0.0	4.4	8.5	3.7	3.3	2.3	0.6	5.0	4.8	4.8	1.0	2.4	7.4	2.9	4.1	3.5	0.0	1.7
IRL	3	22.7	10.4	6.2	1.5	0.0	0.0	5.1	9.7	4.2	3.8	3.1	0.7	2.5	6.1	5.5	1.8	1.0	2.0	1.8	4.7	3.5	0.0	3.6
ITA	3	18.3	9.9	5.8	5.2	0.0	0.0	3.5	6.6	2.9	2.5	4.0	1.2	6.7	4.3	4.4	1.3	3.3	7.6	2.8	3.2	3.6	0.2	2.7
LUX	3	7.4	3.0	3.8	9.4	0.0	0.0	1.1	0.6	0.0	0.6	12.9	0.8	8.4	5.7	0.8	6.1	2.6	11.6	3.8	0.6	6.2	0.2	14.6
NLD	3	2.0	1.0	0.5	0.0	0.0	0.0	0.5	0.9	0.4	0.3	0.1	0.1	0.2	0.5	0.5	0.1	92.0	0.1	0.1	0.4	0.2	0.0	0.1
NOR	3	12.5	5.6	3.3	0.9	0.0	0.0	2.8	5.1	2.2	2.0	2.8	0.4	1.3	3.8	2.9	1.6	42.5	0.8	1.0	2.5	2.3	0.0	3.5
POL	3	13.8	4.5	4.1	4.4	0.0	0.0	3.2	4.6	1.8	2.1	11.4	0.4	3.2	7.7	2.3	6.4	0.6	3.9	2.3	2.5	5.9	0.0	14.9
PRT	3	12.4	6.0	3.1	1.1	0.0	0.0	2.6	4.9	2.1	1.9	2.4	0.6	1.8	3.4	2.9	1.2	44.4	1.1	0.9	2.4	2.1	0.1	2.6
ROM	3	11.5	5.3	7.4	10.4	0.0	0.0	2.6	4.8	2.1	1.9	3.6	0.4	10.0	3.1	2.8	1.0	1.7	17.8	5.2	2.3	4.2	0.0	2.0
SVN, MKD, YUG, HRV, BIH	3	18.2	8.4	5.8	4.2	0.0	0.0	3.9	7.1	3.0	2.8	5.3	0.8	4.8	5.6	4.2	2.5	1.6	5.8	2.6	3.5	4.2	0.1	5.6
SWE	3	12.1	5.7	3.2	0.5	0.0	0.0	2.7	5.2	2.3	2.0	1.2	0.4	1.2	3.0	3.0	0.7	50.1	0.7	0.8	2.5	1.7	0.0	1.2
AUT	4	0.0	0.0	0.1	0.8	0.0	0.0	2.1	17.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	77.7	0.4	1.3	0.0	0.0	0.0	0.0
BEL	4	0.0	2.8	0.0	0.0	0.0	5.8	0.0	6.2	23.8	0.0	0.0	0.0	0.0	0.0	3.0	0.0	58.1	0.0	0.0	0.0	0.0	0.0	0.3
BGR	4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	97.9	0.0	1.2	0.0	0.0	0.0	0.0	0.0
CHE	4	0.0	0.1	0.4	3.1	0.0	0.0	7.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	82.1	1.7	4.9	0.0	0.1	0.0	0.0
CZE, SVK	4	0.0	0.0	0.3	2.1	0.0	0.0	5.2	24.9	0.0	0.0	0.0	0.0	0.0	0.8	0.0	0.0	62.0	1.1	3.4	0.0	0.1	0.0	0.0
DEU New Länder	4	0.0	0.0	0.2	1.5	0.0	0.0	3.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	91.5	0.8	2.3	0.0	0.0	0.0	0.0
DEU Old Länder	4	0.0	0.0	0.0	0.4	0.0	0.0	0.9	5.9	0.0	0.0	0.0	0.0	0.0	2.5	0.0	0.0	89.5	0.2	0.6	0.0	0.0	0.0	0.0
ESP	4	0.0	1.2	0.0	0.4	0.0	2.6	0.7	8.6	10.5	0.0	0.0	0.0	0.0	12.5	1.3	0.0	61.3	0.1	0.5	0.0	0.0	0.0	0.1
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	4	0.0	0.0	0.3	2.1	0.0	0.0	5.1	23.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	64.7	1.1	3.3	0.0	0.1	0.0	0.0

Country code(s)	Sector name	Chemical categories																					
		acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & al- kenes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes
FIN	4	0.0	0.0	0.2	1.6	0.0	0.0	4.0	25.7	0.0	0.0	0.0	0.0	0.0	0.0	0.0	65.0	0.9	2.6	0.0	0.1	0.0	0.0
FRA	4	0.0	0.0	0.1	0.5	0.0	0.0	1.1	6.4	0.0	0.0	0.0	0.0	4.8	0.0	0.0	86.1	0.2	0.7	0.0	0.0	0.0	0.0
GBR	4	0.0	1.4	0.1	0.4	0.0	2.9	1.0	2.3	11.9	0.0	0.0	0.0	3.2	1.5	0.0	74.4	0.2	0.7	0.0	0.0	0.0	0.1
GRC	4	0.0	0.0	0.0	0.2	0.0	0.0	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	98.6	0.1	0.4	0.0	0.0	0.0	0.0
HUN	4	0.0	0.0	0.1	0.6	0.0	0.0	1.4	9.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	87.5	0.3	0.9	0.0	0.0	0.0	0.0
ITA	4	0.0	0.3	0.0	0.3	0.0	0.5	0.6	1.7	2.2	0.0	0.0	0.0	7.3	0.3	0.0	86.0	0.1	0.5	0.0	0.0	0.0	0.0
NLD	4	0.0	0.0	0.1	0.6	0.0	0.0	1.6	8.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	87.8	0.3	1.0	0.0	0.0	0.0	0.0
NOR	4	0.0	0.1	0.4	3.2	0.0	0.0	7.9	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	81.5	1.7	5.1	0.0	0.1	0.0	0.0
POL	4	0.0	0.0	0.2	1.3	0.0	0.0	3.2	10.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	82.2	0.7	2.1	0.0	0.0	0.0	0.0
PRT	4	0.0	0.0	0.3	2.2	0.0	0.0	5.4	26.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	61.0	1.2	3.4	0.0	0.1	0.0	0.0
ROM	4	0.0	0.0	0.2	1.6	0.0	0.0	4.0	34.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	56.5	0.9	2.6	0.0	0.1	0.0	0.0
SVN, MKD, YUG, HRV, BIH	4	0.0	0.0	0.1	1.0	0.0	0.0	2.5	17.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	77.1	0.5	1.6	0.0	0.0	0.0	0.0
SWE	4	0.0	0.0	0.0	0.2	0.0	0.0	0.4	3.7	0.0	0.0	0.0	0.0	0.0	76.3	0.0	19.1	0.1	0.2	0.0	0.0	0.0	0.0
ALB	5	0.0	0.0	0.1	29.8	0.0	0.0	8.2	0.0	0.0	0.0	25.9	0.0	0.0	0.3	0.0	0.0	17.5	18.1	0.0	0.0	0.0	0.0
AUT	5	0.0	0.0	0.5	30.0	0.0	0.0	8.9	0.0	0.0	0.0	18.0	0.0	0.0	4.8	0.0	0.0	27.3	9.7	0.0	0.5	0.0	0.3
BEL	5	0.0	0.0	0.8	29.3	0.0	0.0	10.2	0.0	0.0	0.0	12.9	0.0	0.0	7.7	0.0	0.0	34.1	3.8	0.0	0.8	0.0	0.4
BGR	5	0.0	0.0	0.4	22.9	0.0	0.0	34.0	0.0	0.0	0.0	8.3	0.0	0.0	3.4	0.0	0.0	19.8	10.8	0.0	0.4	0.0	0.2
CHE	5	0.0	0.0	1.0	31.5	0.0	0.0	2.0	0.0	0.0	0.0	14.6	0.0	0.0	9.2	0.0	0.0	38.8	1.5	0.0	1.0	0.0	0.5
CYP	5	0.0	0.0	1.0	32.0	0.0	0.0	0.0	0.0	0.0	0.0	15.0	0.0	0.0	9.5	0.0	0.0	40.0	1.0	0.0	1.0	0.0	0.5
CZE, SVK	5	0.0	0.0	0.2	20.2	0.0	0.0	44.2	0.0	0.0	0.0	5.8	0.0	0.0	1.8	0.0	0.0	14.2	13.3	0.0	0.2	0.0	0.1
DEU New Länder	5	0.0	0.0	0.5	25.4	0.0	0.0	25.9	0.0	0.0	0.0	9.6	0.0	0.0	4.9	0.0	0.0	24.3	8.6	0.0	0.5	0.0	0.3
DEU Old Länder	5	0.0	0.0	0.7	29.9	0.0	0.0	9.9	0.0	0.0	0.0	14.7	0.0	0.0	6.5	0.0	0.0	30.8	6.5	0.0	0.7	0.0	0.3
DNK	5	0.0	0.0	0.2	30.2	0.0	0.0	8.2	0.0	0.0	0.0	24.3	0.0	0.0	1.2	0.0	0.0	19.1	16.7	0.0	0.1	0.0	0.1
ESP	5	0.0	0.0	0.8	31.1	0.0	0.0	4.4	0.0	0.0	0.0	15.7	0.0	0.0	7.7	0.0	0.0	34.7	4.4	0.0	0.8	0.0	0.4
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	5	0.0	0.0	0.1	27.3	0.0	0.0	21.1	0.0	0.0	0.0	19.1	0.0	0.0	0.3	0.0	0.0	14.0	18.0	0.0	0.0	0.0	0.0
FIN	5	0.0	0.0	0.9	30.8	0.0	0.0	4.4	0.0	0.0	0.0	14.1	0.0	0.0	8.7	0.0	0.0	37.4	2.2	0.0	0.9	0.0	0.5
FRA	5	0.0	0.0	0.7	30.4	0.0	0.0	6.5	0.0	0.0	0.0	16.2	0.0	0.0	6.7	0.0	0.0	32.3	6.0	0.0	0.7	0.0	0.4
GBR	5	0.0	0.0	0.2	30.1	0.0	0.0	8.5	0.0	0.0	0.0	24.1	0.0	0.0	1.2	0.0	0.0	19.0	16.6	0.0	0.1	0.0	0.1
GRC	5	0.0	0.0	0.7	31.4	0.0	0.0	2.2	0.0	0.0	0.0	19.2	0.0	0.0	6.2	0.0	0.0	32.1	7.1	0.0	0.7	0.0	0.3
HUN	5	0.0	0.0	0.2	24.7	0.0	0.0	29.4	0.0	0.0	0.0	12.9	0.0	0.0	1.7	0.0	0.0	16.3	14.5	0.0	0.2	0.0	0.1
IRL	5	0.0	0.0	0.6	27.2	0.0	0.0	23.9	0.0	0.0	0.0	9.3	0.0	0.0	5.3	0.0	0.0	24.3	8.7	0.0	0.6	0.0	0.3

Country code(s)	Sector name	Chemical compounds																						
		acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(adi)enes & al- kynes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
ITA	5	0.0	0.0	0.5	29.1	0.0	0.0	14.4	0.0	0.0	0.0	15.4	0.0	0.0	4.6	0.0	0.0	0.0	25.0	10.3	0.0	0.5	0.0	0.2
LUX	5	0.0	0.0	0.9	30.9	0.0	0.0	4.0	0.0	0.0	0.0	14.2	0.0	0.0	8.8	0.0	0.0	0.0	37.7	2.1	0.0	0.9	0.0	0.5
NLD	5	0.0	0.0	0.2	26.9	0.0	0.0	34.9	0.0	0.0	0.0	8.8	0.0	0.0	1.3	0.0	0.0	0.0	10.4	17.3	0.0	0.1	0.0	0.1
NOR	5	0.0	0.0	0.1	30.2	0.0	0.0	7.7	0.0	0.0	0.0	26.3	0.0	0.0	0.1	0.0	0.0	0.0	16.8	18.7	0.0	0.0	0.0	0.0
POL	5	0.0	0.0	0.4	23.8	0.0	0.0	32.4	0.0	0.0	0.0	8.5	0.0	0.0	3.6	0.0	0.0	0.0	20.0	10.8	0.0	0.4	0.0	0.2
PRT	5	0.0	0.0	1.0	32.0	0.0	0.0	0.0	0.0	0.0	0.0	15.0	0.0	0.0	9.5	0.0	0.0	0.0	40.0	1.0	0.0	1.0	0.0	0.5
ROM	5	0.0	0.0	0.1	24.5	0.0	0.0	32.7	0.0	0.0	0.0	12.8	0.0	0.0	0.5	0.0	0.0	0.0	12.1	17.1	0.0	0.1	0.0	0.0
SVN, MKD, YUG, HRV, BIH	5	0.0	0.0	0.3	28.9	0.0	0.0	13.1	0.0	0.0	0.0	19.8	0.0	0.0	2.4	0.0	0.0	0.0	21.0	14.0	0.0	0.3	0.0	0.1
SWE	5	0.0	0.0	1.0	31.9	0.0	0.0	0.5	0.0	0.0	0.0	14.9	0.0	0.0	9.4	0.0	0.0	0.0	39.7	1.1	0.0	1.0	0.0	0.5
ALB	6	0.0	7.8	0.0	0.0	19.1	9.0	0.0	0.0	3.9	0.0	32.3	8.6	0.0	0.0	0.0	1.3	4.5	0.0	0.0	0.0	6.7	0.0	6.7
AUT	6	0.0	9.6	0.0	0.0	11.5	11.5	0.0	0.0	4.6	0.0	30.6	10.3	0.0	0.0	0.0	1.6	3.9	0.0	0.0	0.0	8.1	0.0	8.1
BEL	6	0.0	9.2	0.0	0.0	10.8	11.2	0.0	0.0	4.6	0.0	31.5	10.9	0.0	0.0	0.0	1.6	3.9	0.0	0.0	0.0	8.2	0.0	8.2
BGR	6	0.0	8.2	0.0	0.0	15.2	9.4	0.0	0.0	4.3	0.0	33.3	9.4	0.0	0.0	0.0	1.4	4.5	0.0	0.0	0.0	7.1	0.0	7.1
CHE	6	0.0	8.8	0.0	0.0	12.8	10.9	0.0	0.0	4.5	0.0	31.3	10.4	0.0	0.0	0.0	1.6	3.8	0.0	0.0	0.0	8.0	0.0	8.0
CYP	6	0.0	9.1	0.0	0.0	13.0	11.3	0.0	0.0	4.8	0.0	30.1	9.6	0.0	0.0	0.0	1.6	4.2	0.0	0.0	0.0	8.1	0.0	8.1
CZE, SVK	6	0.0	9.3	0.0	0.0	10.5	11.1	0.0	0.0	5.2	0.0	31.7	10.3	0.0	0.0	0.0	1.6	4.0	0.0	0.0	0.0	8.2	0.0	8.2
DEU New Länder	6	0.0	9.7	0.0	0.0	8.4	12.3	0.0	0.0	5.4	0.0	30.8	10.5	0.0	0.0	0.0	1.7	3.9	0.0	0.0	0.0	8.7	0.0	8.7
DEU Old Länder	6	0.0	9.5	0.0	0.0	10.6	11.5	0.0	0.0	4.4	0.0	31.2	11.0	0.0	0.0	0.0	1.6	3.9	0.0	0.0	0.0	8.2	0.0	8.2
DNK	6	0.0	9.5	0.0	0.0	11.4	11.2	0.0	0.0	4.4	0.0	31.9	10.3	0.0	0.0	0.0	1.6	3.9	0.0	0.0	0.0	7.9	0.0	7.9
ESP	6	0.0	8.5	0.0	0.0	12.6	10.2	0.0	0.0	4.2	0.0	32.6	10.3	0.0	0.0	0.0	1.5	4.9	0.0	0.0	0.0	7.6	0.0	7.6
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	6	0.0	10.1	0.0	0.0	9.5	12.4	0.0	0.0	5.1	0.0	30.2	10.1	0.0	0.0	0.0	1.7	3.8	0.0	0.0	0.0	8.5	0.0	8.5
FIN	6	0.0	10.1	0.0	0.0	11.2	11.8	0.0	0.0	4.6	0.0	30.2	10.3	0.0	0.0	0.0	1.6	3.8	0.0	0.0	0.0	8.2	0.0	8.2
FRA	6	0.0	8.9	0.0	0.0	11.1	10.9	0.0	0.0	4.2	0.0	32.3	11.0	0.0	0.0	0.0	1.6	4.2	0.0	0.0	0.0	7.9	0.0	7.9
GBR	6	0.0	8.9	0.0	0.0	11.2	11.1	0.0	0.0	4.3	0.0	31.6	10.9	0.0	0.0	0.0	1.6	4.2	0.0	0.0	0.0	8.1	0.0	8.1
GRC	6	0.0	8.7	0.0	0.0	13.6	10.7	0.0	0.0	4.3	0.0	31.4	10.2	0.0	0.0	0.0	1.5	4.1	0.0	0.0	0.0	7.7	0.0	7.7
HUN	6	0.0	9.5	0.0	0.0	9.2	11.7	0.0	0.0	5.0	0.0	31.9	10.2	0.0	0.0	0.0	1.7	4.3	0.0	0.0	0.0	8.3	0.0	8.3
IRL	6	0.0	8.1	0.0	0.0	15.8	9.8	0.0	0.0	4.1	0.0	31.8	10.1	0.0	0.0	0.0	1.5	4.1	0.0	0.0	0.0	7.4	0.0	7.4
ITA	6	0.0	8.9	0.0	0.0	12.2	10.4	0.0	0.0	4.4	0.0	32.0	10.7	0.0	0.0	0.0	1.6	4.3	0.0	0.0	0.0	7.8	0.0	7.8
LUX	6	0.0	10.1	0.0	0.0	6.5	12.6	0.0	0.0	4.9	0.0	31.0	11.6	0.0	0.0	0.0	1.8	3.8	0.0	0.0	0.0	8.8	0.0	8.8
NLD	6	0.0	9.4	0.0	0.0	10.2	11.7	0.0	0.0	4.6	0.0	30.8	10.9	0.0	0.0	0.0	1.7	4.0	0.0	0.0	0.0	8.3	0.0	8.3
NOR	6	0.0	9.9	0.0	0.0	10.7	12.2	0.0	0.0	4.8	0.0	29.9	10.4	0.0	0.0	0.0	1.7	3.7	0.0	0.0	0.0	8.4	0.0	8.4

Country code(s)	Sector name	acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & al- kenes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
POL	6	0.0	9.7	0.0	0.0	10.9	12.0	0.0	0.0	5.0	0.0	30.3	10.0	0.0	0.0	0.0	1.7	3.8	0.0	0.0	0.0	8.3	0.0	8.3
PRT	6	0.0	8.8	0.0	0.0	12.6	10.8	0.0	0.0	4.5	0.0	31.4	10.2	0.0	0.0	0.0	1.6	4.4	0.0	0.0	0.0	7.9	0.0	7.9
ROM	6	0.0	8.8	0.0	0.0	14.1	10.5	0.0	0.0	4.6	0.0	31.6	9.6	0.0	0.0	0.0	1.5	4.0	0.0	0.0	0.0	7.6	0.0	7.6
SVN, MKD, YUG, HRV, BIH	6	0.0	9.3	0.0	0.0	12.6	10.9	0.0	0.0	4.7	0.0	31.4	9.7	0.0	0.0	0.0	1.6	4.2	0.0	0.0	0.0	7.8	0.0	7.8
SWE	6	0.0	9.6	0.0	0.0	12.0	11.5	0.0	0.0	4.5	0.0	30.7	10.3	0.0	0.0	0.0	1.6	3.8	0.0	0.0	0.0	8.0	0.0	8.0
All countries	7a	0.0	0.0	4.5	4.3	0.0	0.0	1.4	7.2	0.0	4.5	22.8	0.1	1.7	6.9	1.2	11.1	0.0	6.4	0.1	3.8	12.0	3.9	8.1
All countries	7b	0.0	0.0	2.0	2.0	0.0	0.0	1.0	12.0	0.0	4.0	30.0	1.5	6.0	5.0	6.5	20.5	0.0	2.0	1.0	3.0	1.5	0.0	2.0
All countries	7c	0.0	0.0	1.0	32.0	0.0	0.0	0.0	0.0	0.0	0.0	15.0	0.0	0.0	9.5	0.0	0.0	0.0	40.0	1.0	0.0	1.0	0.0	0.5
ALB	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.3	0.0	4.6	5.8	2.7	16.6	8.9	15.9	2.3	13.3	0.2	0.2	5.7	0.6	0.0	0.5
AUT	8	8.3	0.3	3.4	2.4	0.0	0.0	7.3	23.6	0.0	4.7	4.2	1.5	8.6	6.9	8.2	2.0	6.7	0.2	4.7	5.2	0.8	0.0	0.8
BEL	8	0.0	0.2	2.1	0.4	0.0	0.0	1.0	18.0	0.0	4.5	10.1	2.5	14.7	8.2	14.2	5.5	11.0	0.5	0.3	5.2	0.7	0.0	0.8
BGR	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.3	0.0	4.6	5.8	2.7	16.6	8.9	15.9	2.3	13.3	0.2	0.2	5.7	0.6	0.0	0.5
CHE	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.2	0.0	4.6	6.0	2.7	16.5	8.9	15.8	2.4	13.2	0.2	0.2	5.7	0.6	0.0	0.5
CYP	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.3	0.0	4.6	5.8	2.7	16.6	8.9	15.9	2.3	13.3	0.2	0.2	5.7	0.6	0.0	0.5
CZE, SVK	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.3	0.0	4.6	5.8	2.7	16.6	8.9	15.9	2.3	13.3	0.2	0.2	5.7	0.6	0.0	0.5
DEU New Länder	8	0.0	0.0	2.0	2.0	0.0	0.0	1.0	12.2	0.0	4.0	29.4	1.5	6.2	5.1	6.7	20.1	0.3	2.0	1.0	3.1	1.5	0.0	2.0
DEU Old Länder	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.3	0.0	4.6	5.8	2.7	16.6	8.9	15.9	2.3	13.3	0.2	0.2	5.7	0.6	0.0	0.5
DNK	8	3.5	3.5	1.8	2.9	0.0	0.0	0.8	9.9	0.0	3.3	26.2	2.1	7.6	4.3	6.2	16.9	1.2	2.4	1.0	2.5	1.7	0.3	1.8
ESP	8	1.3	1.5	2.0	0.9	0.0	0.0	0.9	16.3	0.0	4.2	11.8	2.6	13.9	7.5	12.9	6.5	9.7	0.9	0.4	4.7	0.9	0.1	0.9
EST, LTU, LVA, BLR, UKR, MDA, RUS, KAZ, GEO, AZE, ARM	8	9.2	1.1	3.4	3.0	0.0	0.0	7.3	21.8	0.0	4.4	7.9	1.4	6.9	6.0	6.3	4.6	4.3	0.7	4.9	4.6	1.0	0.1	1.1
FIN	8	0.9	1.1	2.1	0.5	0.0	0.0	0.9	17.7	0.0	4.4	8.4	2.7	15.5	8.2	14.5	4.0	11.7	0.6	0.3	5.2	0.8	0.1	0.7
FRA	8	0.0	0.2	2.2	0.3	0.0	0.0	1.0	18.4	0.0	4.6	8.2	2.6	15.4	8.6	14.8	4.0	11.9	0.5	0.3	5.4	0.9	0.1	0.8
GBR	8	0.2	0.3	2.1	0.3	0.0	0.0	1.1	18.4	0.0	4.5	8.7	2.6	15.2	8.4	14.6	4.4	11.6	0.5	0.4	5.3	0.7	0.0	0.7
GRC	8	2.0	2.0	2.1	1.0	0.0	0.0	1.1	16.6	0.0	4.1	9.7	2.7	14.6	7.6	13.2	4.8	10.5	0.9	0.5	4.8	0.9	0.1	0.8
HUN	8	1.4	0.2	2.3	0.8	0.0	0.0	2.1	18.7	0.0	4.5	9.9	2.3	13.3	7.9	12.9	5.5	9.8	0.5	1.1	5.1	0.8	0.0	0.8
IRL	8	0.0	0.3	2.1	0.1	0.0	0.0	1.0	19.0	0.0	4.6	6.9	2.7	16.1	8.8	15.5	3.1	12.7	0.3	0.2	5.6	0.6	0.0	0.6
ITA	8	0.0	0.2	2.2	0.3	0.0	0.0	1.0	18.4	0.0	4.6	7.9	2.5	15.4	8.6	14.8	3.7	12.0	0.5	0.2	5.5	1.0	0.1	0.9
LUX	8	0.0	0.3	2.1	0.0	0.0	0.0	1.0	19.3	0.0	4.6	5.8	2.7	16.6	8.9	15.9	2.3	13.3	0.2	0.2	5.7	0.6	0.0	0.5
NLD	8	0.0	0.0	2.0	2.0	0.0	0.0	1.0	12.1	0.0	4.0	29.6	1.5	6.2	5.1	6.7	20.2	0.2	2.0	1.0	3.0	1.5	0.0	2.0
NOR	8	1.1	1.3	2.0	1.2	0.0	0.0	0.9	15.3	0.0	4.1	15.5	2.4	12.3	6.9	11.6	9.3	7.7	1.2	0.6	4.3	1.1	0.1	1.1

Country code(s)	Sector name	acids	alcohols	benzene	butanes	chlorinated HC's	esters	ethane	ethene	ethers	ethyne	hexanes & higher alkanes	ketones	methanal	other alk(ad)enes & al- kenes	other alkanals	other aromatics	others	pentanes	propane	propene	toluene	trimethylbenzenes	xylene
POL	8	13.3	0.1	4.2	3.7	0.0	0.0	11.3	27.0	0.0	4.9	2.1	0.7	4.1	5.9	3.9	1.0	3.2	0.1	7.5	5.1	0.9	0.0	0.9
PRT	8	0.0	0.2	2.1	0.2	0.0	0.0	1.0	18.4	0.0	4.5	8.6	2.6	15.4	8.5	14.8	4.4	11.8	0.4	0.3	5.4	0.7	0.0	0.7
ROM	8	2.2	2.4	2.0	0.8	0.0	0.0	0.9	17.2	0.0	4.1	6.1	3.0	16.4	8.1	14.7	2.0	12.6	0.7	0.3	5.1	0.8	0.2	0.6
SVN, MKD, YUG, HRV, BIH	8	5.8	0.2	3.1	1.6	0.0	0.0	5.5	22.7	0.0	4.7	4.0	1.8	11.3	7.7	10.8	1.5	9.0	0.2	3.3	5.5	0.7	0.0	0.7
SWE	8	0.9	1.1	2.1	0.4	0.0	0.0	0.9	18.1	0.0	4.4	7.3	2.7	15.9	8.4	14.9	3.2	12.3	0.5	0.3	5.3	0.7	0.1	0.6
All countries	9	11.0	6.1	3.6	7.7	1.2	1.3	5.1	7.4	2.4	2.4	12.6	1.6	1.2	4.0	2.8	2.3	6.5	6.9	4.2	2.9	3.8	0.5	2.5

For category 10 the VOC speciation is not known at this level. However, a profile in terms of the components of the CBM-IV mechanism is available, see the table below.

ole	par	tol	xyl	form	ald	ket	acet	eth	unreactive
2.13	22.4	0.37	0.09	1.95	1.66	0.1	0.05	4.5	10.67

Appendix C. NO_x emissions through soil

The literature gives several methods to calculate the NO-emission from soil. From all the methods two (one by Yienger and Levy (1995) and one by Stohl et al. (1996)) are discussed below. These two methods are discussed because they are suitable with regard to this project.

B.1 NO-emissions from the soil, by Yienger and Levy (1995)

The basic formula Yienger and Levy are using is:

$$Flux = f_{w/d}(T_s, A_{w/d}(biome)) \cdot P(precipitation) \cdot CR(LAI, SAI) \quad (1)$$

Flux	: NO flux [ng N·m ⁻² ·s ⁻¹]
f _{w/d}	: function of the flux dependant on land use, soil temperature and soil humidity (w=wet, d=dry) [ng N·m ⁻² ·s ⁻¹]
T _s	: soil temperature [°C]
A _{w/d}	: coefficient per land use class for wet (w) and dry (s) conditions [ng N·m ⁻² ·s ⁻¹]
P	: ‘pulsing’, correction factor for extra emission as a result of pulsing [-]
CR	: ‘canopy reduction’, correction factor due to absorption of NO by the canopy which results is a lower NO _x -emission above the canopy [-]
LAI	: leaf area index [m ² /m ²]
SAI	: stomata area index [m ² /m ²]

The basic function above is discussed in more detail below.

B.1.a Soil temperature and land use

The influence of the soil temperature (of the top 10 cm) on the NO-emission is different for dry and wet soils. In dry soils the emission is more or less linear with the soil temperature. In a wet soil the NO-emission is linear, exponential or constant with the soil temperature. Because data of the soil temperature is generally lacking the soil temperature is calculated using the air temperature. The way to calculate the soil temperature is different for dry and wet soils. A soil is dry when in the 2 weeks preceding the precipitation is less then 1 cm.

B.1.b Wet soil

Soil temperature

In a wet soil the temperature depends on the air temperature as well on the land use. The following formula gives the relation of the soil temperature for wet soils on the air temperature and land use:

$$T_s = a \cdot T_{air} + b \quad (2)$$

T _{air}	: air temperature [°C]
a, b	: constants for each land use class [-], [°C]

The factors a and b are based on the study by Williams et al. (1992). Table B.1 gives an overview of these factors.

Table B.1 Factors for calculating the soil temperature using the air temperature.

Land use	a [-]	b [°C]
grassland	0.67	8.8
forest	0.84	3.6
wetlands	0.92	4.4
agricultural areas		
corn	0.72	5.8
cotton	1.03	2.9
wheat	1.03	2.9
soybeans	1.03	2.9

NO_x-flux

As stated before the NO-flux of wet soils depends on the soil temperature. Within a certain temperature range one can describe the influence of the soil temperature on the NO-flux with the formulas given in Table B.2.

Table B.2 Formulas to calculate the NO-emission by wet soils under influence of the soil temperature.

Temperature range	Formula	
<0°C	$f_w(T_s, A_w(\text{biome})) = 0$	(3)
0-10°C	$f_w(T_s, A_w(\text{biome})) = 0.28 \cdot A_w(\text{biome}) \cdot T_s$	(4)
10-30°C	$f_w(T_s, A_w(\text{biome})) = A_w(\text{biome}) \cdot e^{0.103 \cdot T_s}$	(5)
>30°C	$f_w(T_s, A_w(\text{biome})) = 21.97 \cdot A_w(\text{biome})$	(6)

The coefficient $A_w(\text{biome})$ is given for each land use class except rain forests and agricultural areas (see Table B.3). These coefficients based on several measuring campaigns and are strictly for wet soils.

Table B.3 Wet soil biome data ($\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$).

Land use	$A_w(\text{biome})$
grassland, savanna	0.36
coniferous/deciduous forests	0.03
drought deciduous forests	0.06
woodland*	0.17
tundra	0.05

* average of grassland and forest.

The NO-emission by arable land ($A_w(\text{agriculture})$) depends on the N-fertilizer rate. Assumed is that 2.5% loss of applied N fertilizer annually. To calculate $A_w(\text{agriculture})$ the following formula is used:

$$A_w(\text{agriculture}) = A_w(\text{grassland}) + S \cdot \text{fertrate} \cdot c \quad (7)$$

- S : loss of applied N fertilizer (2.5%) [-]
 fertrate : applied N-fertilizer [$\text{kg N} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$],
 c : factor to convert $\text{kg N} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$ to $\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ (= 38.58, assuming 30 days a month).

Assumed is that the fertilization elevates soil nitrogen t constant levels during the growing season, and that during the off-season, no-residential fertilizer remains, so that soils emit as grassland. The growing season depends on the latitude. The growing season is:

- north of 30° N: May-August,
- between 30° N en 30° S: January-December,
- south of 30° S: November-February.

B.1.c Dry soil

Soil temperature

The temperature influence on the NO_x -flux is weak. The dry soil temperature also depends different on the air temperature that a wet soil. The dry soil temperature is calculated in the following way:

$$T_s = T_{\text{air}} + 5 \quad (8)$$

T_{air} : air temperature [$^{\circ}\text{C}$]

NO_x -flux

The NO -flux depends linear on the soil temperature for temperatures below 30°C. Above the 30°C the NO -flux is assumed to be constant (this is the optimum emission), see Table B.4

Table B.4 Formulas to calculate the NO -emission by dry soils under influence of the soil temperature.

Temperature range	Formula	
<0°C	$f_d(T_s, A_d(\text{biome})) = 0$	(9)
0-30°C	$f_d(T_s, A_d(\text{biome})) = \frac{A_d(\text{biome}) \cdot T_s}{30}$	(10)
>30°C	$f_d(T_s, A_d(\text{biome})) = A_d(\text{biome})$	(11)

The coefficients $A_d(\text{biome})$ for dry soils are given in Table B.5.

Table B.5 Dry soil biome data ($\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$).

Land use	$A_d(\text{biome})$ [$\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$]
grassland, savanna	2.65
coniferous/deciduous forests	0.40
drought deciduous forests	0.22

'woodland'*	0.37
tundra	1.44

* average of grassland and forest.

calculated as the optimum flux ($T > 30^\circ\text{C}$) in wet soils divided by 3.

The NO-flux of dry agriculture soils are calculated by using formulas 3-6 where $A_d(\text{agriculture}) = A_w(\text{grassland})$.

B.1.d Pulsing

Pulsing means the extra NO-emission due to precipitation on a dry soil.

Yienger en Levy distinguish 4 'pulse' categories. Each category has his own relation with regard to precipitation and NO-emission (see Table B.6). Relations 12-15 result in a correction factor which is used in function (1).

Table B.6 Relations between precipitation and NO-emission for dry soils.

Pulse category	Rain intensity [cm/day]	Relation ¹⁾	Time scale [day]
no pulse	<0.1	$P(\text{precipitation}) = 1.0$	-
'sprinkle'	>0.1 en <0.5	$P(\text{precipitation}) = 11.19 \cdot e^{-0.805 \cdot t}$	1-3
'shower'	>0.5 en <1.5	$P(\text{precipitation}) = 14.68 \cdot e^{-0.384 \cdot t}$	1-7
'heavy rain'	>1.5	$P(\text{precipitation}) = 18.46 \cdot e^{-0.208 \cdot t}$	1-14

¹⁾ $0 > P(\text{precipitation}) \leq 1$

B.1.e Canopy reduction

The canopy absorbs part of the emitted NO_x by the soil. This leads to a lower NO_x-flux above the canopy with regard to the flux from the soil. This phenomenon is called 'canopy reduction' (CR). The calculation of the CR is quite complicated. But to take the effect of canopy reduction into account Yienger and Levy formulated a fairly simple semi-empirical relation between the stomatal area index (SAI) and the leaf area index (LAI). The correction factors are calculated during daytime (NO_x absorption by stomata) en night time (NO_x absorption by cuticle).

$$CRF_d(SAI) = e^{-(k_s \cdot SAI)} \quad (16)$$

$$CRF_n(LAI) = e^{-(k_c \cdot LAI)} \quad (17)$$

k_s : stomatal absorption constant ($8.75 \text{ m}^2/\text{m}^2$)

k_c : cuticle absorption constant ($0.24 \text{ m}^2/\text{m}^2$)

CRF_d : reduction factor vegetation during daytime [-]

CRF_n : reduction factor vegetation during night time t [-]

SAI : stomatal area index (ratio stomatal area – leaf area) [m^2/m^2]

LAI : leaf area index [m^2/m^2]

The by Yienger en Levy derived LAI, SAI and diurnal averaged CRF are given in Table B.7.

Table B.7 Canopy reduction factors.

land use	SAI (m ² /m ²)	LAI (m ² /m ²)	CRF (m ² /m ²)
grassland (yearly averaged)	0.018	3.6	0.64
coniferous forests (yearly averaged)	0.036	12	0.39
deciduous forests (fall/winter)	0.025	5	0.55
'woodland' (fall/winter)	0.020	4	0.61
tundra (yearly averaged)	0.010	2	0.77
Agricultural areas ¹⁾ (growing season)	0.032	4	0.57

¹⁾ increasing during the growing season: 1st month, CRF=0; 2nd month, CRF=1/3 CRF(growing season); 3rd month, CRF=2/3 CRF(growing season); 4th month, CRF=CRF(growing season).

The flux above the canopy is calculated by multiplying the soil emission with the correction factor (CRF) obtained by de SAI and LAI.

$$F_{\text{from canopy}} = F_{\text{from soil}} \cdot CRF(LAI, SAI) \quad (18)$$

$F_{\text{from canopy}}$: NO flux above the canopy [ng N·m⁻²·s⁻¹]

$F_{\text{from soil}}$: NO flux from soil to atmosphere [ng N·m⁻²·s⁻¹]

B.2 European inventory of soil nitric oxide emissions and the effect of these emissions on the photochemical formation of ozone (Stohl et al., 1996)

Stohl suggests to calculate the shallow slab (<5 cm) soil temperature on basis of net radiation, air temperature and the soil temperature of the lower slab. The soil temperature in the top layer peaks earlier during the day than air temperature because it responds fast to solar radiation, whereas the air temperature maximum lags the radiation by several hours. The information of the radiation, air temperature and lower slab soil temperature should be obtained from synoptic meteorological measuring stations.

$$\frac{\partial T}{\partial t} = \frac{Q}{C} + \frac{2\pi}{P} (T_m - T) - a(T - T_a) \quad (19)$$

T : soil temperature (top 5 cm)

Q : net radiation (W/m²)

C : heat capacity of the soil per m²

P : diurnal cycle (86400 s)

T_m : soil temperature lower slab

T_a : air temperature

a : conductivity between soil and air

The problem with the method above is the availability of the soil temperature in the lower slab. A lot of interpolation between the synoptic station has to be what leads to large(r) uncertainties.

Stohl doesn't take pulsing into account or makes a distinction between dry and wet soils. He uses the following basic function:

$$F = A \cdot e^{0.071 \cdot T_s} \quad (20)$$

- F : NO flux from the soil [$\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$]
 A : emission factor which depends on the land use fertilization [$\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$]
 T_s : soil temperature [$^{\circ}\text{C}$]
 k : temperature influence factor [$^{\circ}\text{C}^{-1}$]

The factors A and k differ from the factors used by Yienger and Levy. Stohl used the factors given in Table B.8.

Table B.8 Emission factors ($\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$) per land use class.

Land use	A*
grassland, savanna	0.70
permanent crops	0.70
forests	0.05
inland waters	0.06
urban areas	0.05
others	0.40

* These factors are $\pm 30\%$ lower than given by Williams et al. (1992) to correct for 'canopy reduction'.

Stohl assumes a loss of 5% of the applied N fertilization. The emission of arable soils are calculated using the following relation:

$$A_{(agriculture)} = S \cdot fertrate \cdot c \quad (21)$$

- $A_{(agriculture)}$: standard NO-flux of agricultural soils [$\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$],
 S : loss of applied N fertilizer (5%) [-]
 fertrate : applied N-fertilizer [$\text{kg N} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$],
 c : factor to convert $\text{kg N} \cdot \text{ha}^{-1} \cdot \text{month}^{-1}$ to $\text{ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$ (= 38.58, assuming 30 days a month).

Function 21 is only suitable during the growing season (May-August), outside the growing season the NO-flux of arable soils is constant, i.e. $0.5 \text{ ng N} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$.

Stohl et al. assumes that 10% of the total calculated NO_x -emission is NO_2 and 90% is NO this because the ozone in the top layer of the soil reacts with the NO_x .

Literature

Stohl A., Williams E., Wotawa G. and Kromp-Kolb, H., A European inventory of soil nitric oxide emissions and the effect of the emissions on the photochemical formation of ozone, *Atmospheric environment* vol. 30 no. 22, 3741-3755, 1996.

Williams E.J., Guenther A. and Fehsenfeld F.C., An inventory of nitric oxide emissions from soils in the United States, *J. geophys. Res.* 97, 7511-7519, 1992.

Yienger J.J. and Levy J.II, Empirical model of global soil-biogenic NO_x emissions, *J. geophys. Res.* 100, 11447-11464, 1995.

Appendix D. Aerosols in LOTOS

This appendix is provided as a separate report, with reference:

Schaap, M., "Aerosols in LOTOS, technical report TNO-MEP R 2000/405, November 2000

An electronic version of this report can be found at the CD-ROM that goes along with this report.

8 Authentication

Name and address of the principal:

Names and functions of the cooperators:

Names and establishments to which part of the research was put out to contract:

Date upon which, or period in which, the research took place:

Signature:

Approved by: