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## EMISSION OF NITROGEN OXIDES FROM FOSSIL FUEL COMBUSTION IN EUROPE BY A.SEMB AND E.AMBLE

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## EMISSION OF NITROGEN OXIDES FROM FOSSIL FUEL COMBUSTION IN EUROPE

#### 1 INTRODUCTION

The main man-made source of nitrogen oxides is the combination of atmospheric nitrogen and oxygen during combustion processes. The oxidation of nitrogen-containing compounds in the fuel give rise to additional nitrogen oxide formation. The conditions favouring formation of nitrogen oxides during combustion are high temperature, long residence time, and excess air.

The emission of nitrogen oxides is usually estimated from fuel consumption data and empirically determined emission factors, i.e. weight of NO<sub>x</sub> formed per unit weight of fuel. It is desirable to use existing statistical information to provide a first estimate of the total emission of NO<sub>x</sub> in Europe, and the spatial distribution of these emissions, for use in the prediction of concentration fields and deposition.

#### 2 EMISSION FACTORS

The amount of nitrogen oxides emitted per unit of fuel consumed depend on combustion conditions and nitrogen content of the fuel. Emission factors for different fuels and fuel uses reflect this, although the variation in nitrogen content for a particular fuel type is not well known and therefore not taken into account. Table 1 give som emission factors taken from the U.S. EPA compilation and various national surveys of nitrogen oxide emissions.

Fuel types and uses		Emission facto	or, g $NO_2/3$	kg fuel
	EPA	Derwent & Stewart	FRG	Levander
	(1)	(3)	(4)	(5)
Hard coal				
Power plants	9	10.5	7.2	
Industry	3-7.5	9.2	7.2	Also in
Residential	1.5	3.2		
Lignite				
Power plants	7		3.5**	
Other	3			
Residual fuel oil				
Power plants	15	11.7	11.6	15
Industry	9	12.3-15	7.8	5-10
Distillate fuel				
Residential	3	6		3
Diesel oil	52	10.6	15.5	61
Motor gasoline	≃20 <b>*</b>	16.2	20.2	29
Natural gas				
Power plants	1.2 g/	kcal		
Industry	0.2-0.4	11		
Residential	0.1-0.2	11		

Table 1: Some emission factors for nitrogen oxides from fuel combustion.

\* Pre-1972 car models

\*\* Per unit weight of lignite as consumed, with a water content of 60-60%.

N. 20

By convention, the sum of  $NO+NO_2$  is given in terms of weight equivalents of  $NO_2$  emitted per kg of fuel consumed, although typically more than 90% is emitted as NO. Oxidation of NO to  $NO_2$  with ozone and other oxidants is sufficiently rapid, however, to render a distinction between NO and  $NO_2$  emissions of limited value.

Emission factors given in terms of g NO/kg or even g NO+ g NO<sub>2</sub> have been converted to g NO<sub>2</sub> in Table 1. Emission factors for cars are usually quoted in g/vehicle km, according to certain standardized driving cycles. Conversion to g/kg fuel require some knowledge of the fuel consumption rates on which these figures were based. The EPA figures for post-1972 vehicles reflect increasingly stringent regulations and are not applicable to European conditions. A fuel consumption of 0.17 l/km has been assumed for gasoline vehicles, while the diesel emission factor (1) is an over-all figure for diesel engines. Interestingly, road tests gave a significantly lower emission factor for heavy duty diesel vehicles (2).

Whereas there is broad agreement between the various sets of emission factors for stationary sources, estimated emissions from diesel engines are highly variable. The reason for this, at least partly, is different combustion conditions for different designs of diesel, which vary from low values of  $\approx$  8 g/kg for light vehicles with indirect injection (5), and up to  $\approx$  90 g/kg fuel for some turbocharged, high efficiency engines (5).

Levander (5) estimated the total  $NO_x$  emission for Sweden using emission factors of 1.9-2.3 g/km for light vehicles and 13.2-22 g/km for heavy diesel powered vehicles. The factors in Table 1 were derived from his estimate of the total  $NO_x$ -emissions from gasoline and diesel, respectively, divided by the actual consumption of these fuels. The result is not inconsistent with the emission factors in g/km, and realistic fuel consumption figures.

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Burner and combustion chamber designs also have significant impacts on NO<sub>x</sub> emissions from stationary sources. Thus, for cyclone-fed coal combustion in large power plants, an emission factor of 27.5 g/kg is given in (1). The general emission factor for thermal power plant is in the range 7-9 g/kg coal, however.

It thus appears possible to obtain a reasonable estimate of regional nitrogen emissions from available statistical data on fuel consumption, particularly where a sufficiently detailed breakdown in fuel types and consumption sectors is available, such as in the OECD Energy Statistics (6).

### 3 ESTIMATED NO EMISSIONS FOR INDIVIDUAL COUNTRIES

The emission factors in Table 2 have been chosen to conform with the statistical breakdown. Because of the variability of emissions per unit fuel within each group of combustion sources, for which no specific or detailed information is available, the choice of emission factors is to some extent arbitrary. However, the results show that within OECD Europe the major emissions are associated with transport and electric power production, estimated at 43 and 28 percent, respectively, of the total emissions.

More specific and detailed information on the emission factors for these groups of sources will be essential if the emission estimates are to be improved.

Calculated emissions of  $NO_x$  for individual countries are presented in Appendix 1, and summarized in Table 3. It is interesting to compare the calculated national emissions with the total fuel consumption in energy equivalents as given by the UN Statistics (7). It has been assumed that coal and coke used in metallurgical processes does not contribute appreciably to the  $NO_x$  emissions, this amount has therefore been subtracted from the energy consumption given by (7).

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Figure 1 shows that, although the nitrogen oxide emissions depend on fuel consumption pattern as well as on the total amounts consumed, the ratio of  $NO_x$  total emission to energy throughput is remarkably constant within the region. Obviously, as a first estimate, this information may also be used to approximate the emissions of  $NO_x$  in countries outside the OECD region.

Because of the uncertainty involved in the application of this emission factor outside the economic region for which it has been deduced, a "conservative" emission factor of 5 kg NO<sub>2</sub>/tonne for coal equivalents have been used, and the amounts of coal used in iron and steel production has been estimated from pig iron production figures.

The calculated national emissions are given in Table 4.

#### 4 SPATIAL DISTRIBUTION

Surveys of spatial distribution within countries are already available in some cases (3,4) and are based on location and capacity of power plants, fuel consumption by districts, and distribution of road traffic. Because of the intercorrelation with population density, it is not surprising that there is high correlation between sulphur dioxide and nitrogen oxides emission for grid squares within one country (Figure 2).

In the EMEP emission grid (9), excessive  $SO_2$  emissions occur in certain grid squares due to metal smelters (grid no. 15,26; 17,26; 27,37; 28,37), burning of local fuel with high sulphur contents (22,17 and 23,18) and processing of natural gas with high sulphur content (21,8). The  $SO_2$  emissions in these squares have been reduced by subjective assessment, and the remaining  $SO_2$  emissions have been used to distribute the national emissions of  $NO_x$ . The resulting  $NO_x$  emission grid is given in Figure 3.

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#### 5 IMPROVING THE EMISSION SURVEY

Many of the assumptions used in estimating the national emissions and their spatial distribution may well be improved. This would require further information to be collected from the countries particularly with respect to emissions from internal combustion engines and from thermal power plants. This information should satisfy certain requirements with respect to consistency between countries, and compatibility with available statistical data. Differences in emission factors between countries should be explainable in terms of different technologies or consumption pattern.

In this emission survey, identical emission factors have been used together with available statistical data to provide an estimated emission field corresponding to the EMEP emission survey for sulphur oxides. Comparison between dispersion model estimates should be carried out to test if the estimated NO<sub>x</sub> emissions are consistent with measured air concentrations of NO<sub>2</sub> and HNO<sub>3</sub>, and with nitrate in precipitation.

	Emission factor g/kg	Fuel consumption (Tg)	NO emission x Tg NO <sub>2</sub>
Hard coal			
Power plants	9	133	1.2
Industry	6	22	0.1
Other	2	24	0.05
Brown coal			
Power plants	4	137	0.5
Residual fuel o	il		
Power plants	12	69	0.8
Refineries	8	19	0.15
Industry	8	95	0.75
Other	6	27	0.16
Gas diesel oil			
Industry	8	24	0.2
Other	4	121	0.5
Transport	36	46	1.7
Motor gas			
Transport	25	90	2.2
Natural gas	g/kcal:	Tcal:	
Power plants	1	336	0.3
Industry	0.3	642	0.2
Other	0.2	554	0.1
			9.0

	Estimated NO emission X Gg NO <sub>2</sub>	Energy consumption 10 <sup>6</sup> tonnes coal equiv.	Average emission- factor g NO <sub>2</sub> /kg coal equiv.
Austria	150	24	6.3
Belgium	290	50	5.8
Denmark	180	27	6.7
Finland	150	20	7.5
France	1300	190	6.8
FRG	2200	315	7.0
Greece	150	18	8.3
Iceland	10	1	10.0
Ireland	60	10	6.0
Italy	1000	160	6.3
Luxembourg	20	4	5.0
Netherlands	400	77	5.2
Norway	100	9	11.1
Portugal	76	7	10.0
Spain	560	69	8.1
Sweden	250	40	8.2
Switzerland	125	19	6.3
Turkey	175	23	7.4
United Kingdom	1900	280	6.8
	9095	1343	

Table 3: Estimated NO emission from fossil fuel combustion and NO producing energy consumption for individual OECD countries in Europe.

		-producing rgy* 10 <sup>3</sup> t SKE	er	stimated NO x nissions ) <sup>3</sup> t
Albania	1	733		9
Bulgaria	40	035		200
Czechoslovakia	101	145		500
Germany, Dem.Rep.	113	614		570
Hungary	36	785		185
Poland	167	066		840
Romania	77	393		390
USSR	1 354	524	6	800
Yugoslavia	41	208		190
			9	684

Table 4: Energy consumption and estimated  ${\rm NO}_x$  emissions for countries outside OECD.

\* Less hydro and nuclear energy and corrected for coal used in primary iron production. A factor of 0.4 x the pig iron production has been used to estimate the amount of coke consumed in primary iron production.

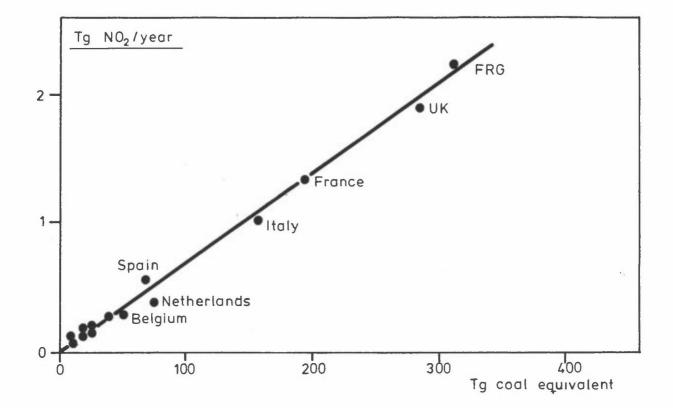


Figure 1: Fossil fuel consumption and estimated NO  $_{x}$  emission for countries within OECD Europe.

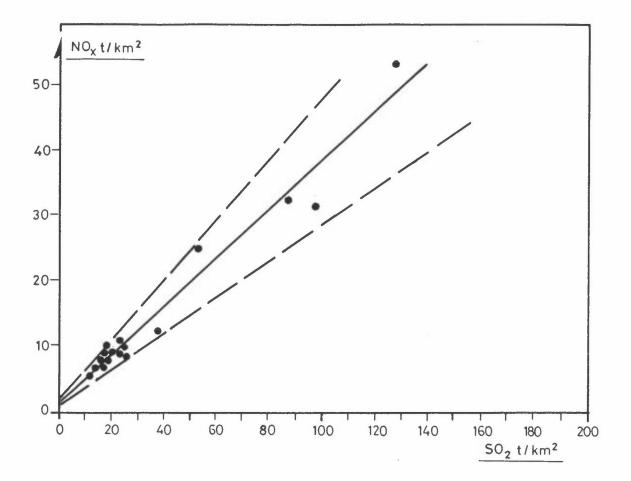


Figure 2: SO<sub>2</sub> and NO emissions in individual grid squares within the<sup>x</sup>Federal Republic of Germany, Based on data from reference (4).

1	2	3	4	5	6	7	8	9	10	11	12	U	14	15	16	17	13	19	20	21	22	23	24	25	26	27	28	29	-	31	_	33	34	35	36	37	38	39
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-		-	6	E.				1.5	-	10				1	2	3	19	15	14	1	2	7	T	80	15	6	3	6	3	60				2	-	2	-	
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			32					1	100	12.5		2	10	1	40	15	Ð	1	/53	10	19(	32.			68	32	55	45	11	2	2					38	Halt	400
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Figure 3: Estimated NO $_x$  emission field (10 $^3$  t NO $_2/a$ ).

# APPENDIX

# ESTIMATED NO EMISSIONS FOR INDIVIDUAL COUNTRIES WITHIN OECD EUROPE

- (8) United Nations World energy supplies 1972-1976. New York 1978.
- (9) Dovland, H. Emissions of sulphur dioxide in Saltbones, J. Europe in 1978. Lillestrøm 1978. (EMEP/CCCreport 2/79.)

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(3)	Derwent, R.G. Stewart, H.N.M.	Air pollution from oxides of nitrogen in the United Kingdom. Atmos. Environ. 7, 385-401 (1973).
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(7)	OECD	Energy statistics 1974/1976. Paris 1978. Organisation for eco- nomic co-operation and development.

	Fac- tor	cons.	.0 <sup>3</sup> t Em	Spa	in	Portug	al	Turk	еу
Hard coal									
P.plants Ind. Other	9 6 2	- 114 4	0,7 0	5525 2735 247	49,7 16,4 0,5	186 48 11	1,7 0,3 0	1052 243 242	9,5 1,5 0,5
Patent fuel	2	-		127	0,3	1	0	-	-
Brown coal									-
P.plants B.K.B.	4 2	15317 90	61,3 0,2	3110	12,4	-	-	2391 20	9,6
Res. fuel oil									
P. plants Ref. Ind. Other	12 8 8 6	1395 319 1781 322	16,7 2,6 14,2 1,9	7540 1583 10382 866	90,5 12,7 83,1 5,2	774 265 1447 53	9,3 2,1 11,6 0,3	1182 554 2084 218	14,2 4,4 16,7 1,3
Gas/Diesel oil									
Ind. Other Trans.	8 4 36	154 1402 567	1,2 5,6 20,4	150 2305 4518	1,2 9,2 162,6	100 123 805	0,8 0,5 29,0	130 1108 1791	1,0 4,4 64,5
Motor gas	25	948	23,7	4532	113,3	795	19,9	1927	48,2
Natural gas		10 <sup>9</sup> k	cal						
P.plants Ind. Other	1* 0,3* 0,2*	-		4284 5627 744	4,3 1,7 0,1				
Σ			149		563		76		176

		10	<sup>3</sup> t						
	Fac- tor	Cons Em Germany		Nethe	rlands	Belg	ium	United	Kingdom
Hard coal									
P. Plants Ind. Other	9 6 2	31584 2869 2746	284.3 17.2 5.5	98 54 160	0.9 0.3 0.3	2646 298 2068	23.8 1.8 4.1	9684	673.0 58.1 27.0
Patent fuel	2	1426	2.9	8	-	385	0.8	1287	2.6
Brown coal									
P. Plants B.K.B	4 2	109764 5889	439,1 11.8	- 14	-	- 23	-	-	-
Res. fuel oil									
P. Plants Ref. Ind. Other	12 8 8 6	4684 3921 16375 1483	56.2 31.4 131.0 8.9	757 1646 1209 669	9.1 13.2 9.7 4.0	3482 255 1948 1344	41.8 2.0 15.6 8.1	3342 14139	151.1 26.7 113.1 20.6
Gas/diesel oil									
Ind. Other Trans.	8 4 36	6143 41206 8517	49.1 164.8 306,6	514 3214 2443	4.1 12.9 87,9	916 5880 1459	7.3 23.5 52,5	5734	39.6 22.9 264,5
Motor gas	25	19999	500.0	3476	86.9	2795	69.9	15783	394.6
Natural gas				109	kcal				
P. Plants Ind. Other	1* 0.3* 0.2*	123980 162729 82894	48.8	109583 90544 149610	109.6 27.2 29.9	19824 45923 21949		21621 125595 166646	21.6 37.7 33.3
Σ			2198		396		290		1886

								1	
	Fac- tor	Aust	ria	Denma	ark	Finl	and	Frar	ice
Hard coal									
P. Plants Ind. Other	9 6 2	33 47 197	0.3 0.3 0.4	2632 488 47	23.7 2.9 0.1	1840 882 73	16.6 5.3 0.1	3329	108.1 20.0 6.9
Patent fuel	2	31	0.1	-	-	-	-	2772	5.5
Brown coal									
P. Plants B.K.B.	4. 2	2627 331	10,5 <sup>·</sup> 0.7	_ 20	-	- -	-	2597 180	10,4
Res. fuel oil									
P. Plants Ref. Ind. Other	12 8 8 6	727 40 2051 1656	8.7 0.3 16.4 9.9	2920 124 1222 1857	35.0 1.0 9.8 11.1	910 - 2260 1174	10.9 - 18.1 7.0	10688 3656 15767 2358	128.3 29.2 126.1 14.1
Gas/diesel oil									
Ind. Other Trans.	8 4 . 36	77 1509 725	0.6 6.0 26,1	565 4209 1021	4.5 16.8 36,7	453 2772 1012	3.6 11.1	24874 7757	41.2 99.5 279,3
Motor Gas	25	2140	53.5	1623	40.6	1367	34.2	15261	381.5
Natural gas				10 <sup>9</sup> ka	cal				
P. Plants Ind. Other	1* 0.3* 0.2*	8389 21461 4648	8.4 6.4 0.9		-	2341 4547 -	2.3 1.4 -	28877 70134 62219	28.9 21.0 12.4
			149		182		148		1313

	Fac- tor	Irela	and	Luxembo	ourg	Norwa	чy	Swed	len
Hard coal									
P. Plants Ind. Other	9 6 2	48 - 550	0.4 _ 1.1	7 509 8	0.1 3.1 -	23 384 35	0.2 2.3 0.1	- 279 30	- 1.7 0.1
Patent fuel	2	-	-	1	-	-	-	-	-
Brown coal									-
P. Plants B.K.B.	4 2	-	-	- 40	-0.1	-	-	- 15	-
Res. fuel oil									
P. Plants Ref. Ind. Other	12 8 8 6	1212 11 1023 59	14.5 0.1 8.2 0.4	62 - 445 8	0.7 - 3.6 -	4 _ 1275 190	- 10.2 1.1	1627 243 4906 4349	19.5 1.9 39.2 26.1
Gas/Diesel oil									
Ind. Other Trans.	8 4 36	179 569 277	1.4 2.3 10,0	64 330 95	0.5 1.3 3,4	609 1011 1288	4.9 4.0 46,4	852 6042 1333	6.8 24.2 48,0
Motor Gas	25	799	20.0	180	4.5	1148	28.7	3248	81.2
Natural gas				10 <sup>9</sup> k	cal				
P. Plants Ind. Other	1* 0.3* 0.2*	-	-	789 2397 627	0.8 0.7 0.1		-		-
			59		· 19		98		249 ·

	Fac <del>n</del> tor	Switze:	rland	Icela	nd	Ital	У	
Hard coal P. Plants	9	_	_	_	_	1009	9.1	
Ind. Other	6 2	97 19	0.6	-	-	217 196	1,3 0,4	
Patent fuel Brown coal	2	17	-	-	-	41	0,1	
P. Plants B.K.B	4 2	47	0.1	-	-	1362 35	5,4 0,1	
Res. fuel oil								
P. Plants Ref. Ind. Other	12 8 8 6	412 57 984 241	4.9 0.5 7.9 1.4	25 - 56 -	0.3 _ 0.4 _		219,3 23,9 123,7 32,3	
Gas/Diesel oil								
Ind. Other Trans.	8 4 36	1276 5004 461	10.2 20.0 16,6	- 144 185	0.6 6,7		13,7 52,5 173,4	
Motor Gas	25	2444	61.1	86	2.2	11208	280,2	
Natural gas		10 <sup>9</sup> ko	cal					
P. Plants Ind. Other	1* 0.3* 0.2*	2339 2345	0.7 0.5	-	-	18968 110468 62231	19,0 33,1 12,4	
			125		10		1000	



## NORSK INSTITUTT FOR LUFTFORSKNING

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TITTEL Emission of nitrogen oxides from fossil fuel combustion in Europe		PROSJEKTLEDER A. Semb NILU PROSJEKT NR 00578
FORFATTER(E)		TILGJENGELIGHET **
A. Semb E. Amble		A OPPDRAGSGIVERS REF.
OPPDRAGSGIVER		L
3 STIKKORD (á m	aks.20 anslag)	
Emission factors	fuel consumption	grid squares
	300 anslag, 5-10 linje:	, banancharag
TITLE Emission of nitrogen oxides from fossil fuel combustion in Europe.		
ABSTRACT (max. 300 characters, 5-10 lines)		
Yearly emissions of nitrogen oxides in 150x150 km grid squares have been estimated on the basis of fuel consump- tion data and emission factors.		
**Kategorier: Åpen - kan bestilles fra NILU A		

\*Kategorier: Åpen - kan bestilles fra NILU A Må bestilles gjennom oppdragsgiver B Kan ikke utleveres C

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