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WP-95-35
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Acknowledgments

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1. Abstract

This paper presents a preliminary estimate of atmospheric emissions of cadmium, lead and zinc in Europe during the period 1955-1987. The emission data are used as input to the IIASA's atmospheric transport model, TRACE (TRace toxic Air Concentrations in Europe), to compute cumulative deposition loads of heavy metals onto European soils during the investigated time period. To the authors' knowledge, this is the first attempt of this kind in the open literature. The computed with the TRACE model maxima of cumulative depositions for the three metals are approximately: 60 mg/m^2 for Cd, 1450 mg/m^2 for Pb and 2600 mg/m^2 in the case of zinc. The results presented should be considered first-order approximations.

Major uncertainties embedded in such calculations are discussed. Heavy metals enter the soils from atmospheric load and with the application of fertilizers and sewage sludge. Once in the soil the metals can be mobilized, leading to plant and ground water contamination. This threat is particularly valid for heavily polluted regions in Central Europe. The results of this study can be applied in assessing environmental and health effects of heavy metals, and therefore are important for scientists as well as policy makers.

2. Introduction

In recent years there has been a growing interest in obtaining information on the flow of toxic heavy metals through the environment. Heavy metals such as cadmium, lead and zinc, are emitted into the atmosphere during various industrial processes. Combustion of fuels in stationary and mobile furnaces (engines) is among the most important processes. Once in the atmosphere metal aerosols can travel hundreds to thousands of kilometers and deposit far from their emission sources. Even relatively low fluxes across the earth's surface result in accumulation of metals in soils. From the soil the metals can be taken up by plants and may leach out to ground water. Through ground waters, that can serve as sources of drinking water, and through edible plants, heavy metals enter the food chain. Lead (Pb) has a variety of impacts on the human nervous and circulatory system, while cadmium (Cd) is considered a probable human carcinogen (WHO, 1987). Zinc (Zn), at high concentration levels, has been found to be moderately toxic to plants through disturbing the nutrient cycle (Rühling et al., 1987). Cadmium, lead and zinc are among the most abundant and most toxic metals in the environment (Calow, 1993).

In the IIASA Project on Regional Material Balance Approaches to Long-Term Environmental Policy Planning, we concentrate on the impact of heavy metals on agricultural soils within the so called Black Triangle and Upper Silesia (BTUS) region covering the southern part of the former German Democratic Republic, south-western Poland and northern part of Bohemia (Czech Republic). Heavy metals enter agricultural soils through atmospheric deposition, phosphate fertilizers, sewage sludge and manure. For forest soils atmospheric deposition is essentially the only source of heavy metals. In this paper we deal with the atmospheric emission, transport and deposition of Cd, Pb and Zn. Because heavy metals deposited to the BTUS region can be brought through long-range transport from sources far away from the receptor, we deal with all of Europe to account for the transboundary fluxes. We present a preliminary emission inventory of anthropogenic atmospheric emissions of cadmium,

lead and zinc for thirteen major emitter countries in Europe. The database covers the period 1955-1987. The emissions in other European countries are estimated by assuming similar trends (temporal variations of emissions) as for selected countries included in the database and by applying data from other existing databases (Pacyna and Münch, 1988). The emission data are used to compute atmospheric concentrations and depositions in Europe by applying IIASA's TRACE transport model, developed specifically for the needs of a former IIASA Project on the Rhine Basin (Stigliani et al., 1993; Anderberg and Stigliani, 1994).

An important outcome of the study is the computation of the cumulative atmospheric deposition loads onto soils in Europe for the 1955-1987 period. This information, in turn, can be used as input for soil and ground water models, and in general is important for estimating the long-term environmental impacts of heavy metals.

Both the emission inventories and the atmospheric transport model are currently under development and therefore the results of this study should be treated as preliminary. In the future, we plan to use updated emissions for the same or extended time period, and a new more sophisticated transport model HMET (Bartnicki, 1994) to obtain more accurate results. Nevertheless, rough deposition calculations shown in this paper provide first estimates presented in the open literature and therefore are important for both scientists and decision makers.

3. Emission inventories

The emission inventories for cadmium, lead and zinc were developed using various production and consumption statistics from thirteen European countries. Then the emission factors developed by Pacyna (1991) were applied to estimate contributions from various sectors of economy (coal burning, traffic, etc.). The detailed description of the methodology will be published elsewhere (Anderberg and Stigliani, 1995). Here we give only a brief overview. The preliminary versions of the inventories were prepared in early 1992 and were presented by Stigliani and Anderberg at the "Industrial Ecology and Global Change" Workshop in Snowmass, Colorado in July 1992. In the Fall of 1993 some corrections were made particularly concerning coal burning in earlier periods. It should be stressed here that the estimates given below should be considered as a first-order estimation of country total emissions. These data are now revised in connection with the UN Economic Commission for Europe (ECE) Task Force on Heavy Metal Emissions (e.g. Pacyna, 1994) and compared with national estimates available for some countries.

3.1 Cadmium

The inventory includes atmospheric emission estimates for: Germany, France, The Netherlands, Switzerland, Luxemburg, the former German Democratic Republic (GDR), Poland, the former Czechoslovakia (CSSR), the European part of the former Soviet Union (USSR), Belgium, Great Britain, Italy and Spain. The time interval is 1955-1987. The country totals for seven time cuts (reference years) are presented in Table 1.

Table 1. Atmospheric emissions of cadmium from thirteen European countries during the period 1955-1987. Units: tons/year.

	1955	1960	1965	1970	1975	1982	1987
Germany	494.2	522.6	474.9	306.8	185.6	87.6	59.4
France	157.3	189.8	255.0	112.5	72.5	45.3	25.1
Netherlands	58.5	70.7	82.2	88.3	12.7	7.4	4.2
Switzerland	4.4	5.0	5.8	7.1	5.0	4.4	2.4
Luxemburg	2.5	3.0	3.4	4.3	2.7	0.9	0.6
GDR	70.8	71.5	79.4	73.9	76.4	66.6	59.1
Poland	229.1	268.6	226.3	200.2	305.7	200.5	160.0
CSSR	28.0	32.4	39.1	41.7	43.3	32.9	29.2
USSR[1]	310.5	375.4	440.7	532.5	614.7	478.3	360.8
Belgium	300.6	344.6	316.2	185.6	78.0	20.0	18.3
Great Britain	345.4	303.3	186.3	152.9	85.7	50.8	39.2
Italy	39.8	49.9	50.4	90.8	79.5	47.4	39.9
Spain	49.8	75.9	77.9	115.9	53.9	31.8	16.2
SUM	**2090.9**	**2312.7**	**2237.6,**	**1912.5**	**1615.7**	**1073.9**	**814.4**

Table 1 includes the largest emitter countries in Europe (see Pacyna and Münch, 1988) except Bulgaria and Romania. It can be seen from Table 1 that in the late 1950s and early 1960s the largest emissions originated in Germany, Great Britain, the former Soviet Union and Belgium. The maximum value for Germany was 522.6 tons in 1960. Since the late 1960s the emissions in the West European countries had been generally decreasing, while in Eastern Europe the peak emissions occurred in the mid 1970s. The decreasing emissions in Western Europe have been a result of the gradual introduction and development of more and more efficient control techniques. The introduction of new process technologies in non-ferrous metallurgy and steel production has also played a significant role. Increasing energy demands and industrial production with no program for emission control in Eastern Europe, inevitably led to emission increases in the region. In the 1970's emission control equipment began to be installed also in Eastern Europe, resulting in decreasing heavy metals emissions after 1975. The maximum in the Soviet Union was 614.7 tons in 1975. The temporal course of cadmium emissions, with the division between Eastern and Western Europe, is shown in Figure 1.

[1]European part of the Soviet Union covered by the EMEP grid

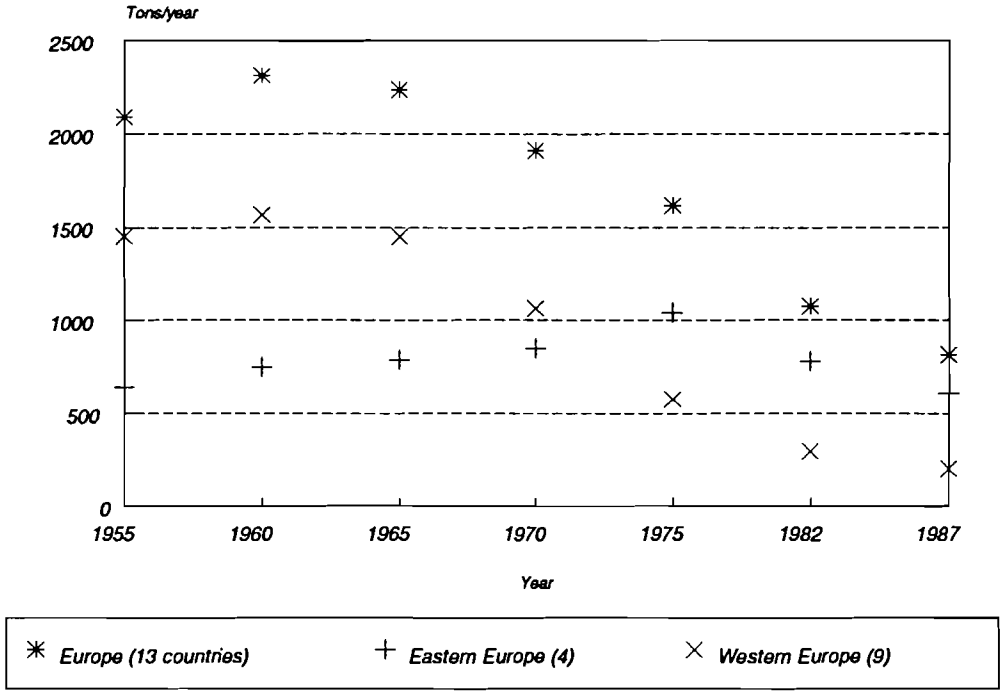

Figure 1. Atmospheric emissions of cadmium from thirteen European countries in the period 1955-1987. See Table 1. for details.

The emissions from the thirteen European countries reached the maximum of over 2300 tons around 1960 and then decreased. By 1987 the total for the thirteen countries listed, decreased by some 60% to a figure just above 800 tons. Until 1970 the total emissions from the nine West European countries exceeded those from the four East European counterparts. In the 1970s and 1980s the situation reversed. In 1987 some 75% of European emissions originated in East European countries.

3.1.1 Emission estimates for other European countries

Simple assumptions have been made to obtain first-order estimates for the remaining European countries. It should be noted that the thirteen countries that are treated directly are responsible for more than 80% of total European emissions (see Table 2 below). As a rule we make use of the temporal variations (trends) from selected countries and apply them to other countries with similar economies. As a scaling factor we use the 1982 estimates given by Pacyna and Münch (1988) and Axenfeld et al. (1992) here termed as the 1982 Pacyna data. For particular countries we applied the following approach.

The emissions from Denmark and Austria were scaled on the basis of temporal emission changes in Germany. For example for Denmark:

$D_i = D_p \cdot (G_i / G_{1982})$ where:

D_i - emission from Denmark for the year i

D_p - emission from Denmark (1982 Pacyna data)

G_i - emission from Germany for the year i (this inventory)

G_{1982} - emission from Germany (1982 this inventory)

Here, Germany was selected due to its history of coal use which is similar to that in Denmark and Austria. The Nordic countries: Finland, Norway and Sweden have never been coal users to the extent as Germany. Therefore, their emissions were calculated according to those in Switzerland. Emissions from East European countries: Albania, Bulgaria, Hungary, Romania and former Yugoslavia were approximated in a similar way by scaling the 1982 Pacyna data, on the basis of temporal emission changes in the former CSSR, former GDR, Poland and the former Soviet Union. In this case the average of the four countries was used as the scaling factor. The emissions for Ireland were scaled with those for Great Britain and emissions for Portugal and Greece were scaled using Italy's emissions. No emissions were assumed for Iceland. The results are shown in Table 2. The 1982 Pacyna data are also included in Table 2.

Table 2. Atmospheric emissions of cadmium in Europe during the period 1955-1987. Units: tons/year.

	1955	1960	1965	1970	1975	1982	1982[2] Pacyna	1987
Albania	0.6	0.7	0.7	0.8	0.9	0.7	0.7	0.5
Austria	29.7	31.4	28.6	18.5	11.2	5.3	5.3	3.6
Belgium	300.6	344.6	316.2	185.6	78.0	20.0	12.2	18.3
Bulgaria	53.7	62.9	66.1	71.4	87.5	65.5	65.5	51.3
CSSR	28.0	32.4	39.1	41.7	43.3	32.9	21.6	29.2
Denmark	35.5	37.6	34.2	22.1	13.3	6.3	6.3	4.3
Finland	8.0	9.0	10.5	12.8	9.0	8.0	8.0	4.3
France	157.3	189.8	255.0	112.5	72.5	45.3	31.8	25.1
GDR	70.8	71.5	79.4	73.9	76.4	66.6	37.1	59.1
Germany	494.2	522.6	474.9	306.8	185.6	87.6	81.1	59.4
Greece	2.7	3.4	3.4	6.1	5.4	3.2	3.2	2.7
Hungary	3.6	4.2	4.4	4.8	5.9	4.4	4.4	3.4
Ireland	5.4	4.8	2.9	2.4	1.3	0.8	0.8	0.6
Italy	39.8	49.9	50.4	90.8	79.5	47.4	35.7	39.9
Luxemburg	2.5	3.0	3.4	4.3	2.7	0.9	0.6	0.6
Netherlands	58.5	70.7	82.2	88.3	12.7	7.4	5.5	4.2
Norway	2.1	2.4	2.8	3.4	2.4	2.1	2.1	1.1
Poland	229.1	268.6	226.3	200.2	305.7	200.5	180.4	160.0
Portugal	2.0	2.5	2.6	4.6	4.0	2.4	2.4	2.0
Romania	35.6	41.7	43.8	47.3	58.0	43.4	43.4	34.0
Spain	49.8	75.9	77.9	115.9	53.9	31.8	133.1	16.2
Sweden	16.4	18.6	21.6	26.5	18.6	16.4	16.4	8.9
Switzerland	4.4	5.0	5.8	7.1	5.0	4.4	0.9	2.4
USSR[3]	310.5	375.4	440.7	532.5	614.7	478.3	308.6	360.8
Great Britain	345.4	303.3	186.3	152.9	85.7	50.8	30.7	39.2
Yugoslavia	70.4	82.4	86.6	93.5	114.7	85.8	85.8	67.1
SUM	**2356.7**	**2614.5**	**2545.7**	**2226.6**	**1948.0**	**1318.1**	**1123.4**	**998.4**

There is fairly good agreement between estimates for 1982 in this inventory and in Pacyna inventory. However there are some differences that need to be explained. Our calculations for 1982 are not fully compatible with the 1982 Pacyna estimates. For East European countries we applied Pacyna emission factors (EF) but with a time

[2]Pacyna and Münch (1988), Axenfeld et al. (1992)

[3]European part of the Soviet Union covered by the EMEP grid

delay of 5-10 years. For example for 1987 we used EF referring to 1980, for 1982 we used EF for 1975 (midpoint 1970-1980) etc. Also the energy statistics and the information on the metal smelters were different. This resulted in higher emission figures notably for the former Soviet Union, the former GDR and Poland. The emissions for the five East European countries which were not treated directly (Albania, Bulgaria, Hungary, Romania and Yugoslavia) are probably overestimated in the 1955-70 period. The production figures in the 1950s and 1960s in those countries were relatively lower than for the other four countries in this part of Europe. On the other hand in Spain the most important smelters are all electrolytic, producing much smaller atmospheric emissions (Roskill, 1978). This fact explains the largest qualitative difference between the two inventories for 1982 (133.1 vs. 31.8 tons).

To sum up the comparison for 1982 we notice that this inventory gives a figure that is 17 percent higher than the one in Pacyna inventory (1318.1 vs. 1123.4 tons). The temporal course of European emissions in the period 1955-1987 is qualitatively similar to that in Figure 1 and Table 1.

3.2 Lead

As in the case of cadmium this inventory includes atmospheric emissions from thirteen countries in the period 1955-1987. They are listed in Table 3.

Table 3. Atmospheric emissions of lead from thirteen European countries during the period 1955-1987. Units: tons/year.

	1955	1960	1965	1970	1975	1982	1987
Germany	9349.1	13717.2	13583.8	16949.5	12936.3	5547.3	3887.9
France	7377.6	9329.4	12968.9	16850.2	18862.5	10230.0	8510.2
Netherlands	1109.6	1505.7	1956.2	2719.6	2263.7	1719.7	983.5
Switzerland	423.0	637.9	941.9	1338.4	1358.1	514.3	406.5
Luxemburg	315.4	391.3	457.4	543.8	416.9	193.8	165.5
GDR	875.4	1080.9	1386.3	2810.6	3509.2	2433.8	2390.9
Poland	2442.1	3555.0	4225.8	5817.5	6981.9	4999.1	4117.9
CSSR	941.9	1288.7	1525.8	2311.3	2914.3	1832.7	1694.5
USSR[4]	14558.0	22836.6	31150.5	40696.2	54131.5	32600.4	28082.0
Belgium	3603.1	4136.0	4789.1	5003.6	4236.1	1663.0	1489.2
Great Britain	8841.7	9439.5	10528.9	12827.3	10427.4	8797.0	3884.6
Italy	2383.9	3914.3	7897.4	13219.5	13882.4	10318.7	5851.5
Spain	1659.8	2144.9	2705.9	4702.7	6325.0	4184.9	3144.7
SUM	**53880.6**	**73977.4**	**94117.9**	**125790.2**	**138245.3**	**85034.7**	**64608.9**

Table 3 includes the largest emitter countries in Europe (Pacyna and Münch, 1988). The noticeable exceptions are the former Yugoslavia, Bulgaria and Greece. Europe's largest emitter is the Soviet Union (the European part covered by the EMEP grid). Its emissions rose from 14 500 tons in 1955 to a peak of 54 000 tons in 1975. By 1987 the Soviet Union emissions decreased by some 48% to 28 000 tons. Germany, France and Italy were the other big emitters. Germany systematically reduced its emissions after 1970 mainly by introducing lead-free gasoline. The other West European countries also reduced their emissions in the late 1970s and in the 1980s, but not so

[4]European part of the Soviet Union covered by the EMEP grid

drastically as Germany. The temporal course of lead emissions, with the division between Eastern and Western Europe, is shown in Figure 2.

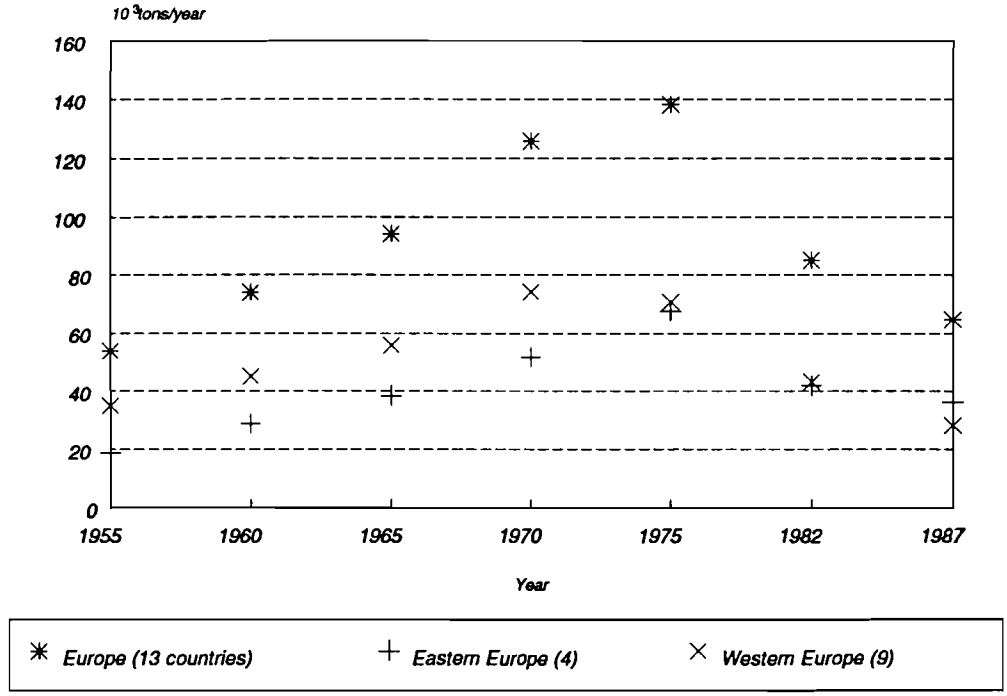

Figure 2. Atmospheric emissions of lead from thirteen European countries in the period 1955-1987. See Table 3 for details.

During the investigated time period the lead emissions from the thirteen European countries reached the maximum of 138 000 tons around 1975, some fifteen years later than in the case of cadmium. After 1975 emissions decreased significantly and in 1987 were reduced by 54% to reach the level of the late 1950s (64 000 tons). The national emission totals from the nine West European countries exceeded or were equal to those from the four East European counterparts for the entire period except in the year 1987. This temporal course follows closely the statistics for gasoline consumption (OECD, 1987) - the major emission source especially after 1975.

3.2.1 Emission estimates for other European countries

As in the case of cadmium emissions, simple assumptions were made to give first-order estimates for the remaining European countries. It should be noted that the thirteen countries that are treated directly make up more than 86% of total European emissions (see the 1982 Pacyna data in Table 4. below). As above we used the temporal variations from certain countries, and apply them to other countries with similar economies. As scaling factors we use the 1982 estimates given by Pacyna and Münch (1988) and Axenfeld et al. (1992), here, once again, termed as the 1982 Pacyna data. For particular countries we applied the same scaling as for cadmium. For example the emissions from Denmark and Austria were scaled on the basis of temporal emission changes in Germany using the 1982 Pacyna data. See section 3.1.1 for details. The results are shown in Table 4.

Table 4. Atmospheric emissions of lead in Europe during the period 1955-1987. Units: tons/year.

	1955	1960	1965	1970	1975	1982	1982[5] Pacyna	1987
Albania	61.2	93.6	124.6	168.1	219.8	136.3	136.3	118.1
Austria	1892.5	2776.7	2749.7	3431.0	2618.6	1122.9	1122.9	787.0
Belgium	3603.1	4136.0	4789.1	5003.6	4236.1	1663.0	2097.2	1489.2
Bulgaria	705.3	1078.0	1435.1	1935.4	2531.4	1569.2	1569.2	1360.0
CSSR	941.9	1288.7	1525.8	2311.3	2914.3	1832.7	1151.0	1694.5
Denmark	1101.0	1615.5	1599.8	1996.1	1523.5	653.3	653.3	457.9
Finland	923.4	1392.5	2056.1	2921.7	2964.7	1122.7	1122.7	887.4
France	7377.6	9329.4	12968.9	16850.2	18862.5	10230.0	8682.8	8510.2
GDR	875.4	1080.9	1386.3	2810.6	3509.2	2433.8	1749.5	2390.9
Germany	9349.1	13717.2	13583.8	16949.5	12936.3	5547.3	5561.8	3887.9
Greece	322.0	528.6	1066.6	1785.4	1874.9	1393.6	1393.6	790.3
Hungary	268.3	410.1	545.9	736.2	962.9	596.9	596.9	517.3
Iceland	0.0	0.0	0.0	0.0	0.0	0.0	39.2	0.0
Ireland	439.3	469.0	523.2	637.4	518.1	437.1	437.1	193.0
Italy	2383.9	3914.3	7897.4	13219.5	13882.4	10318.7	8591.9	5851.5
Luxemburg	315.4	391.3	457.4	543.8	416.9	193.8	165.2	165.5
Netherlands	1109.6	1505.7	1956.2	2719.6	2263.7	1719.7	2205.8	983.5
Norway	598.3	902.2	1332.2	1893.0	1920.8	727.4	727.4	574.9
Poland	2442.1	3555.0	4225.8	5817.5	6981.9	4999.1	2956.3	4117.9
Portugal	88.0	144.5	291.6	488.1	512.6	381.0	381.0	216.1
Romania	519.0	793.2	1056.0	1424.1	1862.6	1154.6	1154.6	1000.7
Spain	1659.8	2144.9	2705.9	4702.7	6325.0	4184.9	4227.7	3144.7
Sweden	851.0	1283.4	1895.0	2692.7	2732.3	1034.7	1034.7	817.8
Switzerland	423.0	637.9	941.9	1338.4	1358.1	514.3	450.7	406.5
USSR[6]	14558.0	22836.6	31150.5	40696.2	54131.5	32600.4	30924.4	28082.0
Great Britain	8841.7	9439.5	10528.9	12827.3	10427.4	8797.0	8615.3	3884.6
Yugoslavia	881.8	1347.8	1794.2	2419.7	3164.9	1961.9	1961.9	1700.4
SUM	**62531.7**	**86812.5**	**110587.8**	**148318.9**	**161652.4**	**97326.3**	**89710.4**	**74029.8**

3.3 Zinc

As in the case of cadmium and lead, this inventory includes atmospheric emissions from thirteen countries in the period 1955-1987. They are listed in Table 5. As in the case of cadmium and lead thirteen countries are listed. Four are from Eastern Europe and nine are from the Western Europe. The list includes the largest emitter countries in Europe (Pacyna and Münch, 1988) with the exception of the former Yugoslavia and Bulgaria. Until 1965 the largest emissions came from Germany with a maximum of 20 000 tons per year. Since 1965 the Soviet Union has become the largest emitter with a maximum of 23 000 tons in 1975. During the 1955-1975 period the other major emitters were Great Britain, Belgium and France. After 1970 the contributions from the East European countries had been increasing with Poland and the former GDR being the major emitters. The similar explanation as for cadmium can be given for the changes of zinc emissions over time. The temporal course of zinc emissions, with the division between Eastern and Western Europe, is given in Figure 3.

[5]Pacyna and Münch (1988), Axenfeld et al. (1992)

[6]European part of the Soviet Union covered by the EMEP grid

Table 5. Atmospheric emissions of zinc from thirteen European countries during the period 1955-1987. Units: tons/year.

	1955	1960	1965	1970	1975	1982	1987
Germany	17731.0	20028.6	20119.4	16034.5	9106.7	4231.5	3074.6
France	7557.4	9503.5	12389.7	9045.6	5475.0	2366.2	1668.6
Netherlands	1971.0	2524.0	3122.2	3652.7	1012.8	391.8	321.2
Switzerland	302.3	367.3	472.6	567.4	354.4	232.6	195.4
Luxemburg	670.1	813.5	903.8	1060.2	556.7	158.4	93.7
GDR	2608.7	2747.5	3096.3	3019.3	3337.0	2681.8	2334.8
Poland	7742.6	9152.6	8006.7	6590.2	8144.1	4255.7	3785.1
CSSR	1640.4	2143.3	2569.0	3044.3	3511.7	2400.6	2065.1
USSR[7]	10402.8	13478.9	16159.9	19487.3	23317.5	15561.8	14123.5
Belgium	10632.4	12292.7	11827.3	8370.1	3950.2	1152.5	932.9
Great Britain	13164.1	12381.1	8963.9	8902.8	4464.9	1817.2	1482.8
Italy	2359.9	3130.9	3893.4	6697.4	5563.6	2592.8	1898.5
Spain	1868.9	3072.3	3119.6	5081.0	3165.6	1444.4	797.1
SUM	**78651.6**	**91636.2**	**94643.8**	**91552.8**	**71960.2**	**39287.3**	**32773.3**

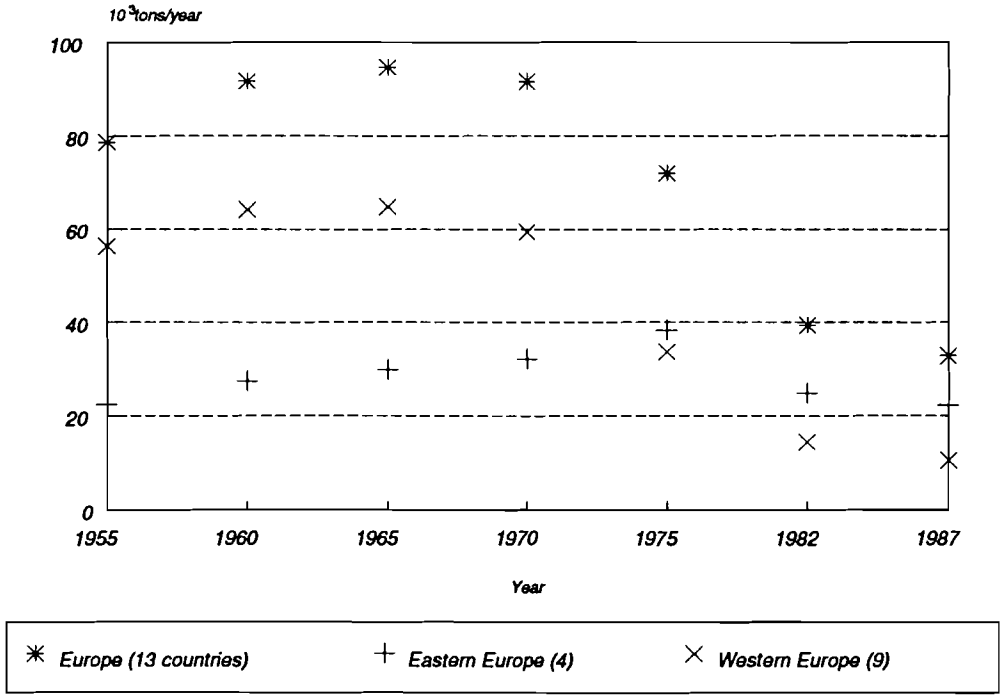

Figure 3. Atmospheric emissions of zinc from thirteen European countries in the period 1955-1987. See Table 5 for details.

During the investigated time period zinc emissions from the thirteen European countries reached a maximum of 94 000 tons around 1965. During the 1960s the emissions were stabilized at the level of some 90 000 tons. After 1970 there was a sharp decrease with a roughly two third reduction from the peak by 1987 (to 32 000 tons). In 1975 the contributions from East European countries exceeded those from the Western Europe. The absolute emissions for Eastern Europe did not vary significantly during the entire period. The figure for 1987 (22 300 tons) is almost

[7]European part of the Soviet Union covered by the EMEP grid

identical with the one for 1955. In West European countries the emissions decreased from 64 000 tons in 1965 to 10 000 tons in 1987.

3.3.1 Emission estimates for other European countries

As in the case of cadmium and lead emissions, we applied simple assumptions to give first-order estimates for the remaining European countries. It should be noted that the thirteen countries that are treated by us directly contribute more than 85% of total European emissions (see the 1982 Pacyna data in Table 6 below). Once again the temporal variations from selected countries were applied to other countries with similar economies. As a scaling factor we use the 1982 estimates given by Pacyna and Münch (1988) and Axenfeld et al. (1992), here termed as the 1982 Pacyna data. For individual countries we applied the same scaling as for cadmium. For example the emissions from Denmark and Austria were scaled on the basis of temporal emission changes in Germany using the 1982 Pacyna data. See section 3.1.1 for details. The results are shown in Table 6.

Table 6. Atmospheric emissions of zinc in Europe during the period 1955-1987. Units: tons/year.

	1955	1960	1965	1970	1975	1982	1982[8] Pacyna	1987
Albania	34.9	42.9	46.5	50.1	59.7	38.8	38.8	34.8
Austria	985.1	1112.8	1117.8	890.9	506.0	235.1	235.1	170.8
Belgium	10632.4	12292.7	11827.3	8370.1	3950.2	1152.5	694.5	932.9
Bulgaria	1583.6	1946.2	2109.6	2272.9	2709.1	1760.8	1760.8	1577.5
CSSR	1640.4	2143.3	2569.0	3044.3	3511.7	2400.6	755.9	2065.1
Denmark	539.7	609.6	612.4	488.1	277.2	128.8	128.8	93.6
Finland	281.5	342.0	440.1	528.4	330.0	216.6	216.6	182.0
France	7557.4	9503.5	12389.7	9045.6	5475.0	2366.2	3636.9	1668.6
GDR	2608.7	2747.5	3096.3	3019.3	3337.0	2681.8	818.9	2334.8
Germany	17731.0	20028.6	20119.4	16034.5	9106.7	4231.5	3699.4	3074.6
Greece	177.1	235.0	292.2	502.7	417.6	194.6	194.6	142.5
Hungary	179.2	220.3	238.8	257.3	306.6	199.3	199.3	178.6
Iceland	0.0	0.0	0.0	0.0	0.0	0.0	1.4	0.0
Ireland	312.2	293.7	212.6	211.2	105.9	43.1	43.1	35.2
Italy	2359.9	3130.9	3893.4	6697.4	5563.6	2592.8	1997.7	1898.5
Luxemburg	670.1	813.5	903.8	1060.2	556.7	158.4	93.4	93.7
Netherlands	1971.0	2524.0	3122.2	3652.7	1012.8	391.8	294.3	321.2
Norway	152.2	184.9	237.9	285.7	178.4	117.1	117.1	98.4
Poland	7742.6	9152.6	8006.7	6590.2	8144.1	4255.7	4040.1	3785.1
Portugal	89.4	118.6	147.5	253.7	210.7	98.2	98.2	71.9
Romania	644.5	792.0	858.5	925.0	1102.5	716.6	716.6	642.0
Spain	1868.9	3072.3	3119.6	5081.0	3165.6	1444.4	3918.0	797.1
Sweden	553.1	672.1	864.7	1038.2	648.5	425.6	425.6	357.5
Switzerland	302.3	367.3	472.6	567.4	354.4	232.6	61.8	195.4
USSR[9]	10402.8	13478.9	16159.9	19487.3	23317.5	15561.8	13160.0	14123.5
Great Britain	13164.1	12381.1	8963.9	8902.8	4464.9	1817.2	2298.8	1482.8
Yugoslavia	1761.7	2165.1	2346.8	2528.4	3013.8	1958.8	1958.8	1754.9
SUM	**85945.9**	**100371.4**	**104169.2**	**101785.1**	**81826.2**	**45420.7**	**41604.5**	**38113.0**

[8]Pacyna and Münch (1988), Axenfeld et al. (1992)
[9]European part of the Soviet Union covered by the EMEP grid

4. Atmospheric Transport and Deposition

4.1 TRACE Model

The TRACE model (TRace toxic Air Concentrations in Europe) was developed at IIASA for use in the Rhine Basin Project (Stigliani et al., 1993). The model computes the air concentrations and depositions of four heavy metals (As, Cd, Pb, Zn) on a European scale. The description of the model and its verification against observations is given in Alcamo et. al, 1992, and Alcamo et al., 1991. Here, for the convenience of the reader, we repeat only the most important features. TRACE is an improved climatological-type model in that (1) travel time is computed from an empirical function rather than from an assumed constant velocity, (2) the model tends to conserve mass, (3) the regularity of spatial deposition patterns is captured, and (4) parameters are objectively determined. The dry velocity is spatially varying, and is computed with a dry deposition model of Sehmel (1980) as a function of "local" friction velocity u_*, surface roughness z_0, together with an assumed characteristic particle size distribution. The TRACE model was found to agree with available observations of arsenic (not used in this study) and lead within a factor of two. Model calculations underestimate Cd observations but are correlated and within a factor of two of observations. The Zn calculations are even greater underestimated but are still correlated to measurements. It is suggested that Cd and Zn emissions are in general underestimated.

The calculation procedure of the model is divided into two steps: first the loss of pollutant from a parcel of air as it travels from a source to a receptor is represented by a simple loss term. This equation gives the air concentration of a pollutant at a receptor located x distance downwind from a source (see the Appendix):

$$c(x_r, y_r, x_e, y_e) = \beta \frac{E(x_e, y_e)}{x}(1-\alpha)e^{-(k_d+k_w)t^*}, \qquad (1)$$

where c=air concentration at the receptor due to a single emission source [in kg/m^3]; (x_r,y_r)=receptor position; (x_e,y_e)=emission source position; E=emission at the source [kg/s]; x=distance between source and receptor, i.e. $x = \sqrt{(x_r - x_e)^2 + (y_r - y_e)^2}$ [m]; α=local deposition coefficient [1]; k_d and k_w=first order loss coefficients, in units of inverse time, which reflect the loss of mass from the air parcel by dry and wet deposition, respectively; and t^*=time of travel between sources and receptors [s]. The factor β is derived assuming mass conservation [s/m^2] (see Appendix). The total concentration, $c(x_r,y_r)$, at the receptor is computed from the sum of contributions coming from all emission sources, weighted according to the frequency of backward trajectories, F(s), arriving from a particular wind rose sector, s:

$$c(x_r, y_r) = \sum_{s=1}^{8} F(s) c_s(x_r, y_r) \qquad (2)$$

In the second step of the calculation, wet and dry deposition of the pollutant at the receptor is computed from the air concentration. Wet deposition, d_w [kg/m^2/year] is computed with a scavenging ratio:

$$d_w(x_r, y_r) = c(x_r, y_r) W_q P(x_r, y_r) \tag{3}$$

where P is annual precipitation amount [m/year] and W_q is the scavenging ratio, i.e. the ratio of the concentration of heavy metals in precipitation to their concentration in air. Dry deposition, d_d [kg/m^2/year] is computed from:

$$d_d(x_r, y_r) = c(x_r, y_r) v_d(x_r, y_r) \tag{4}$$

The dry velocity v_d varies in space but is constant in time, because the long-term climatologic value of v_d is used for each location in the model computations.

4.2 Computational procedure

In order to compute cumulative deposition for the 1955-1987 period we applied the following computational procedure. The data in Tables 2, 4 and 6 represent country totals for each time cut. In order to calculate air concentrations and depositions it was necessary to create a spatial distribution of emissions for each European country in a grid system. It was done in the following way. First we used the emission inventory developed by Axenfeld et al. (1992) with 1985 data for lead (Pb), and 1982 data for cadmium and zinc (Cd, Zn). This emission inventory is in the form of a European grid in the polar stereographic projection with a spatial resolution of 150 x 150 km at 60^0 N (the so-called EMEP grid system). The grid consists of 39 x 37 grid cells. As an example of the emission spatial distribution in Europe, in Figure 4 we present emission inventory for lead for the year 1985. The corresponding figures for Cd and Zn can be found in Bartnicki et al. (1993). Emission data, in the form presented in Figure 4, served as input to the TRACE atmospheric transport model. Neither emissions from natural sources nor the background concentrations were taken into account.

We applied 1985 meteorological data, namely precipitation, mixing height and frequency of backward trajectories arriving at 99 measurement stations in Europe, to come up with the air concentration and deposition maps for each metal in 1985 (Figures 5-10).

Then the so called country-to-grid source-receptor (sr) matrices (Alcamo et al., 1985; Alcamo and Bartnicki, 1990) for 1985 were computed which relate country total emissions with depositions in each cell of the EMEP grid system. Use of sr matrices assumes that there is a linear relationship between pollutant emission and deposition, which is justified by the TRACE model assumptions (see e.g. Alcamo et al., 1987).

Using these sr matrices and emission inventories presented in Section 3, cumulative 1955-1987 total deposition patterns were computed for each metal separately (Figures 11-13). For years in between the time cuts in Tables 2, 4 and 6, linear interpolation was used to compute emissions for each individual country. The total deposition was computed as a sum of dry, wet and local deposition.

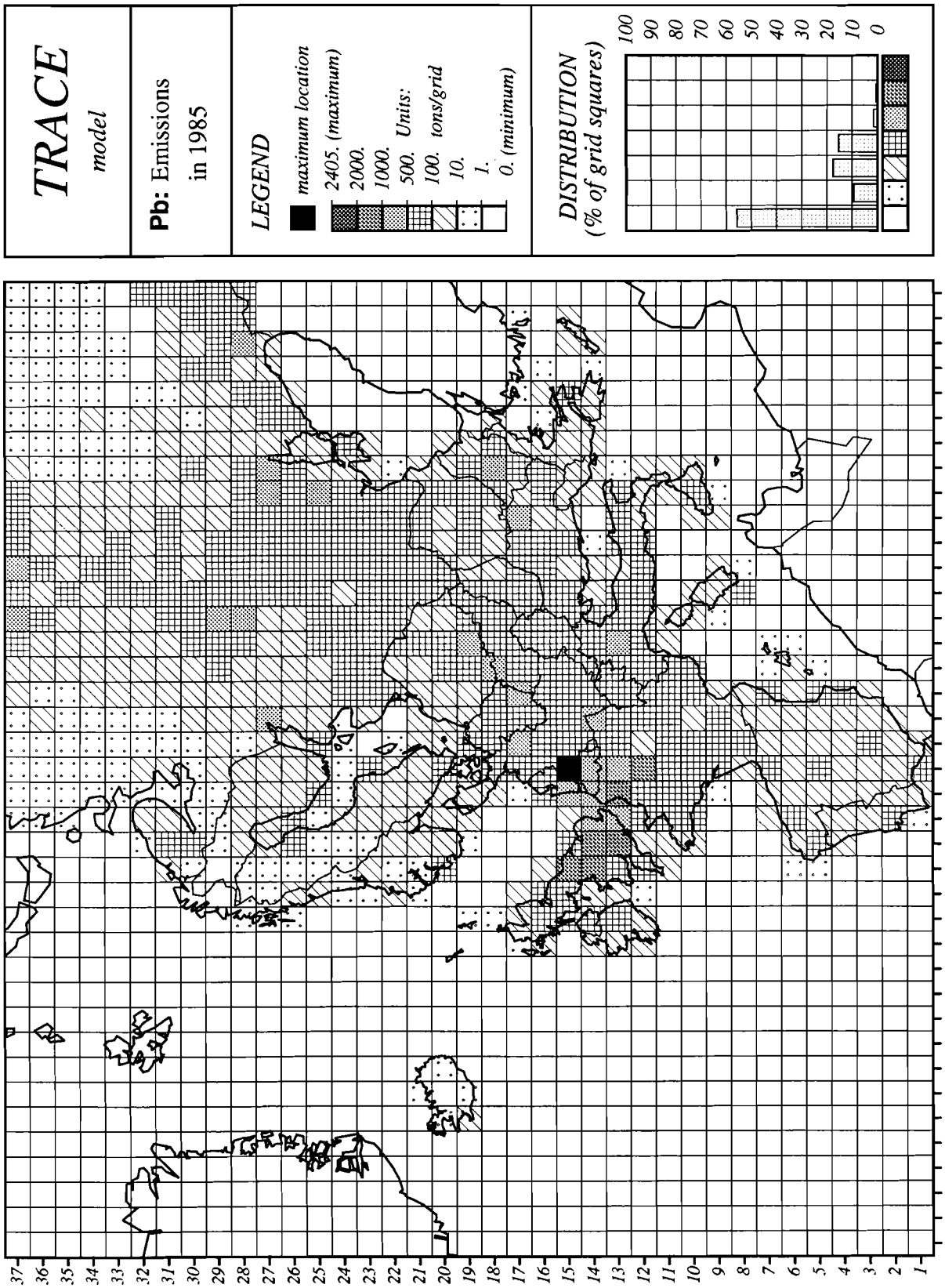

Figure 4. Atmospheric emissions of lead in Europe in 1985. Units: tons/grid.

4.3 Concentrations and depositions in 1985

The computed concentration maps are presented in Figures 5-7[10]. The patterns for cadmium (Figure 5) and zinc (Figure 6) are very similar. This could be expected because both metals are emitted into the atmosphere mostly during the same technological processes (e.g. coal burning). Therefore spatial patterns of atmospheric emissions of Cd and Zn, not shown here, are very similar. The largest Cd and Zn concentrations in Europe are found within the BTUS region, in the Balkans, northern Spain, and in the former Soviet Union. In the case of zinc, high concentrations exceeding 20 ng/m^3 can be also found in northern France and the Benelux countries. Qualitatively the computed concentrations agree well with the results obtained by Bartnicki et al. (1993) with the help of the Heavy Metal Eulerian Transport Model for Europe (HMET). However, the maximum values computed by the TRACE model are approximately 50% lower than the corresponding figures from the HMET model (1.5 versus 2.97 ng/m^3, for Cd and 42.77 versus 96.64 ng/m^3 for Zn).

The spatial pattern for lead concentrations in 1985 (Figure 7) is more uniform than in the case of cadmium and zinc. This is mainly because of more uniform distribution of lead sources (traffic). In large parts of Western and Central Europe the concentrations exceeded 25 ng/m^3. The highest concentrations exceeding 50 ng/m^3 are found in Germany, the Benelux countries, northern France and Great Britain. The maximum value computed by the TRACE model, 75.4 ng/m^3, is lower by some 25% than the corresponding figure obtained with the HMET model (102.3 ng/m^3).

The total deposition patterns for the three metals are shown in Figures 8-10. Qualitatively all three maps agree well with those obtained with the HMET model (their Figures: 24-26). Irregularities associated with the variability of precipitation fields in Europe are captured. Also dry deposition contributes to those irregularities as dry deposition velocities depend on the friction velocity u_*, which varies significantly throughout Europe. The maxima of Cd total deposition (Figure 8) are located in Southern Poland (1.47 mg/m^2), the former Czechoslovakia and Spain. These are the regions of high emission and concentration levels. High deposition values can also be found in the former Soviet Union, Germany and Southern Europe (former Yugoslavia, Bulgaria). The region of our interest (BTUS), especially Lower and Upper Silesia in Poland and Northern Bohemia received Cd deposition exceeding 0.5 mg/m^2.

The pattern is similar in the case of zinc (Figure 9.) The grid maximum, 43.6 mg/m^2, is located in Southern Poland (EMEP grid cell [25,19]). Other local maxima are in Spain, France and Germany with deposition exceeding 20 mg/m^2. As in the case of cadmium the deposition maxima follow the emission and concentration peaks.

In the case of lead (Figure 10) the grid maximum is located near the German-Dutch-Belgian border (EMEP grid cell [20,15]). Other local maxima are located in France,

[10]Graphics routines developed by J.Bartnicki for the HMET model were used for presenting emission, concentration and deposition fields in this paper.

Figure 5. Air concentration of cadmium in Europe in 1985 computed by the TRACE model. Units: ng/m^3.

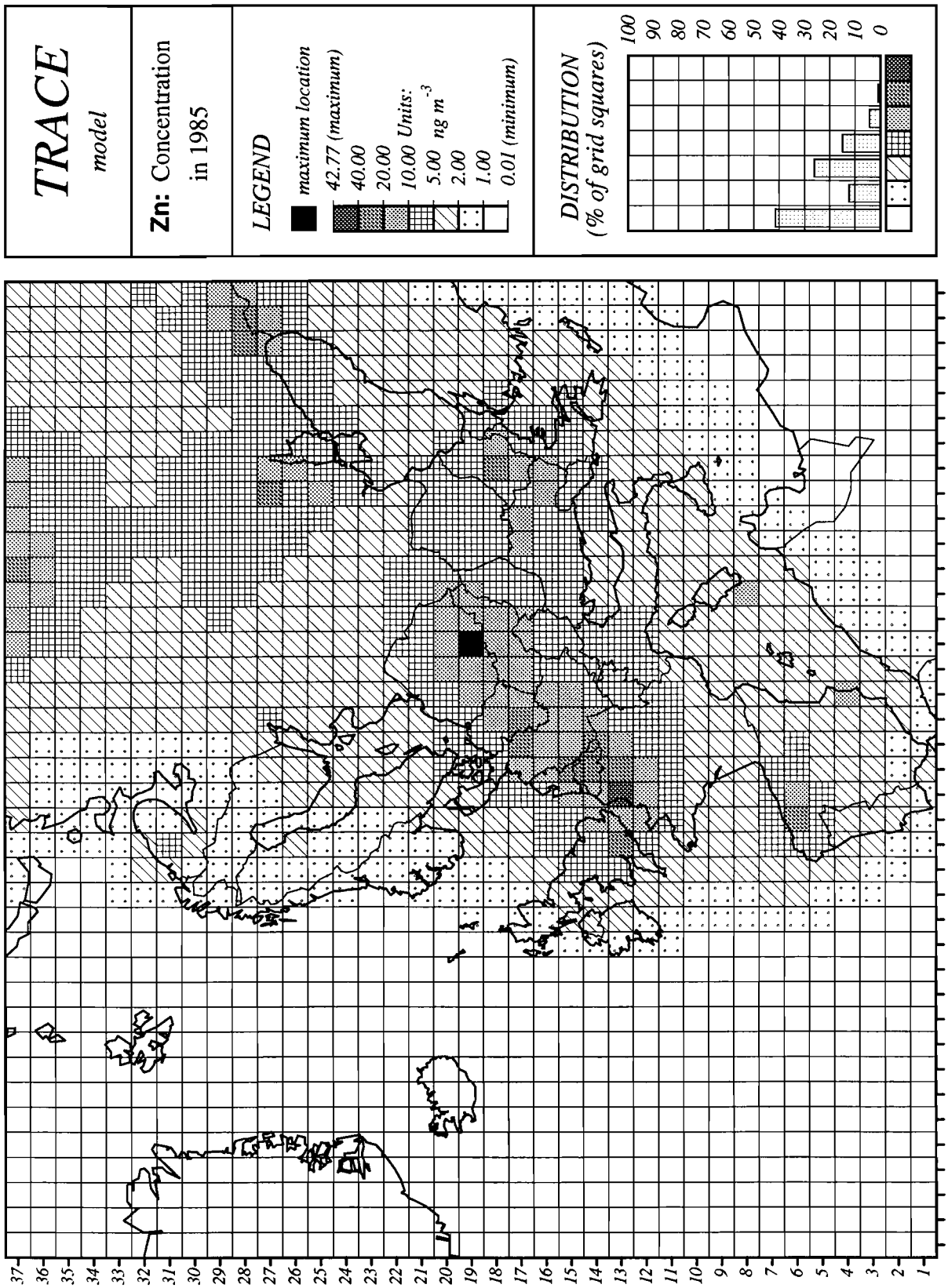

Figure 6. Air concentration of zinc in Europe in 1985 computed by the TRACE model. Units: ng/m³.

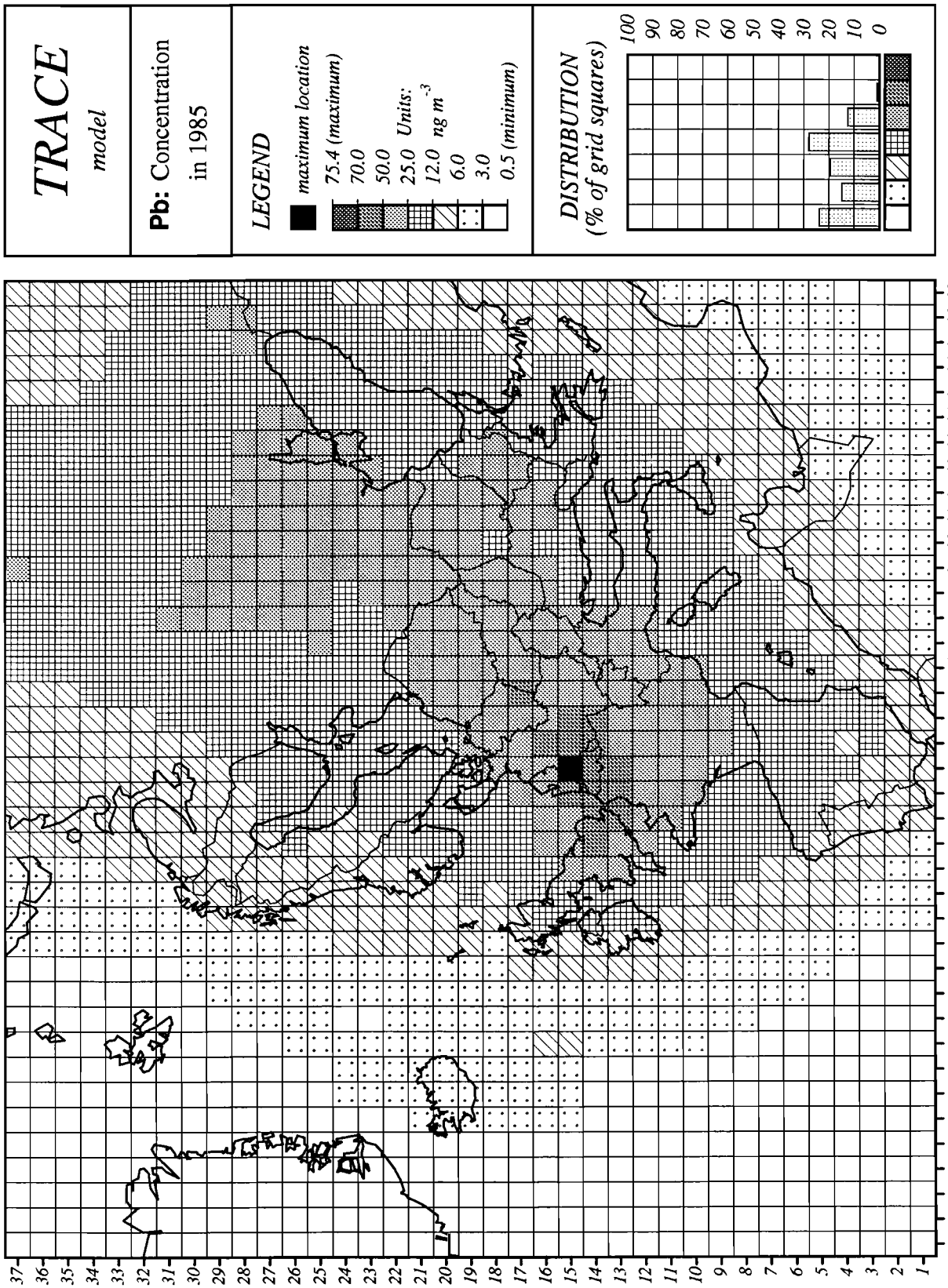

Figure 7. Air concentration of lead in Europe in 1985 computed by the TRACE model. Units: ng/m³.

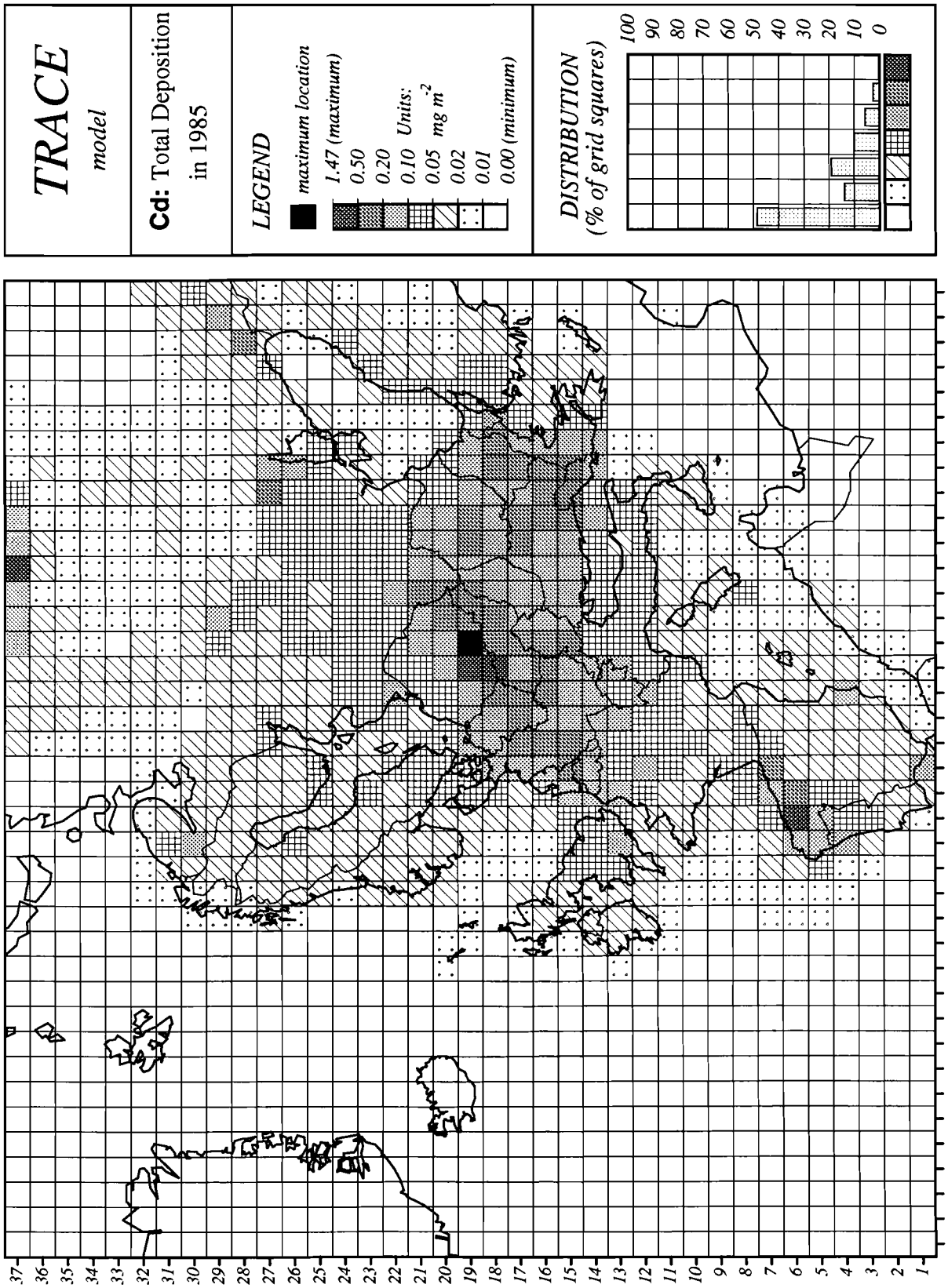

Figure 8. Total deposition of cadmium in Europe in 1985 computed by TRACE model. Units: mg/m².

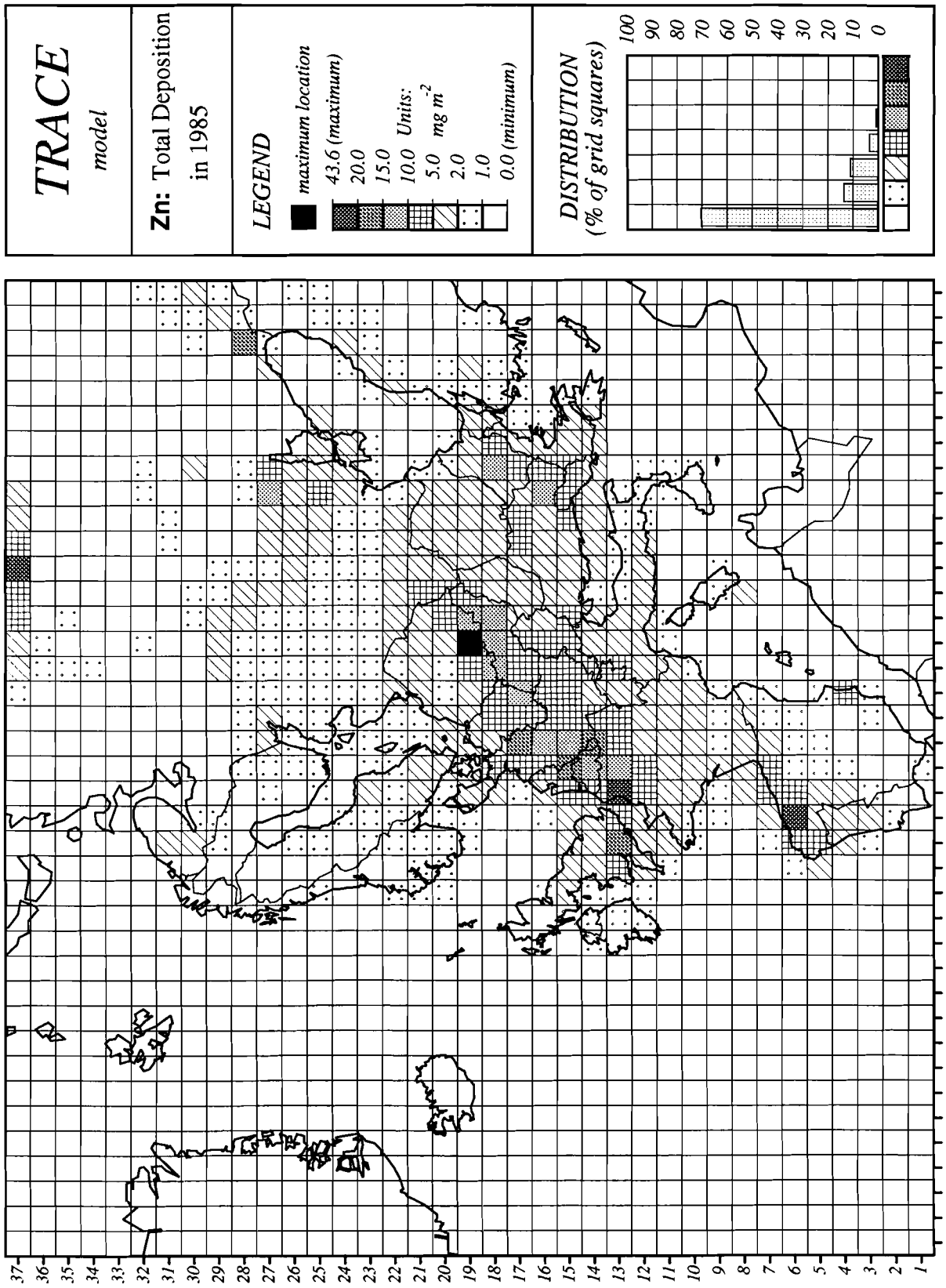

Figure 9. Total deposition of zinc in Europe in 1985 computed by the TRACE model. Units: mg/m².

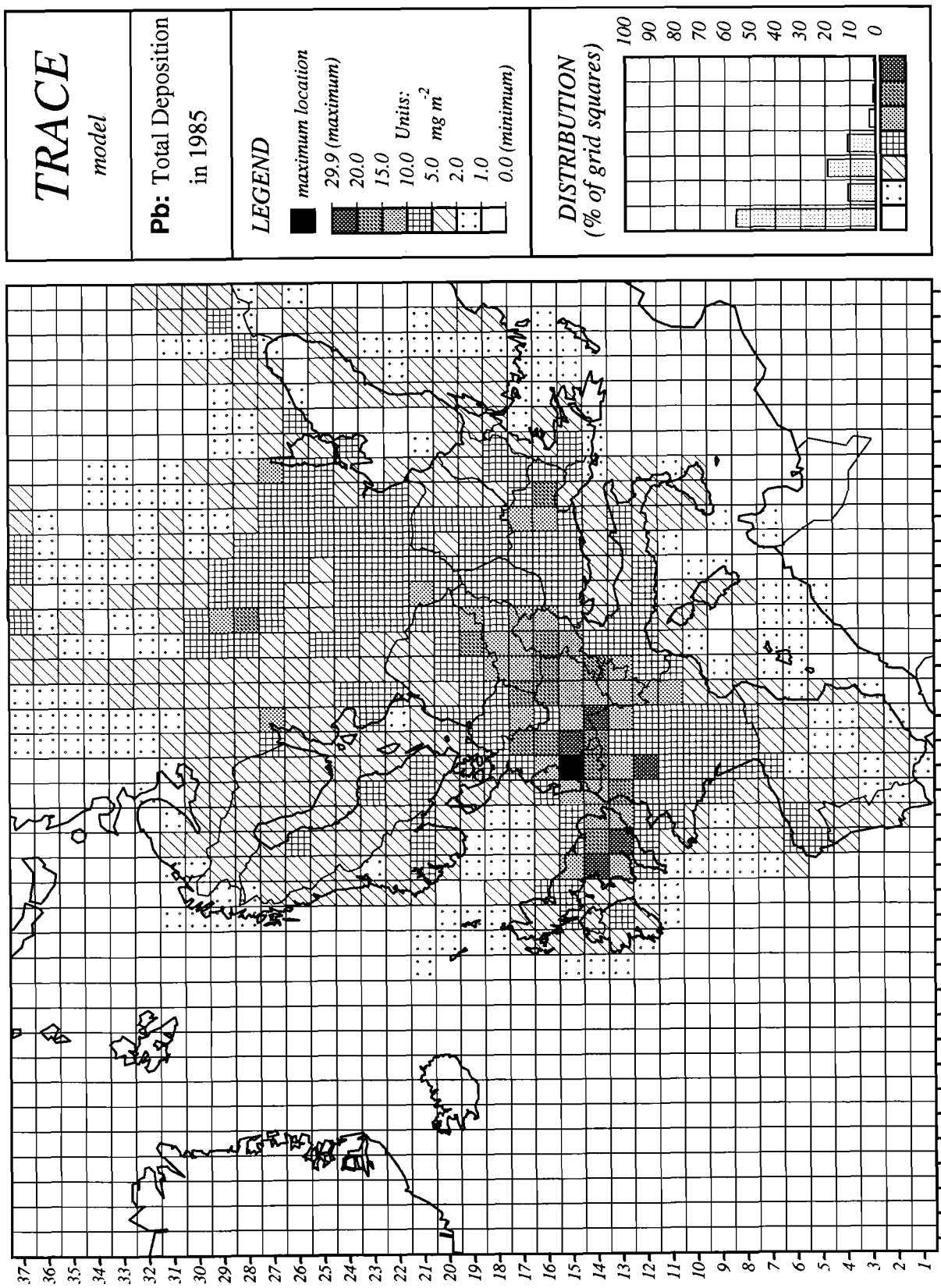

Figure 10. Total deposition of lead in Europe in 1985 computed by the TRACE model. Units: mg/m².

Great Britain and Germany with deposition figures exceeding 20 mg/m^2. These areas coincide with areas of heavy road traffic and gasoline use.

The peak computed depositions for Cd and Zn (1.47 and 43.6 mg/m^2) are some 50% higher than the corresponding results from the HMET calculations (0.91 and 29.23 mg/m^2, respectively). For lead the respective peak figures are 29.7 mg/m^2 (TRACE) and 35.11 mg/m^2 (HMET). These differences come from the differences in model structure and applied meteorological data. On the one hand, TRACE is an improved climatological type transport model which uses as input annual meteorological averages. On the other hand, HMET is a more sophisticated Eulerian type model with more detailed, real time meteorological input (see Section 4.5 for more details).

4.4 Cumulative depositions 1955-1987

The cumulative total deposition patterns for all three metals are shown in Figures 11-13. The maxima of total Cd depositions (Figure 11.) are located in Southern Poland, and the border region between Germany, The Netherlands and Belgium. In those areas the computed cumulative depositions exceed 40 mg/m^2. High levels exceeding 20 mg/m^2 are also in the former Czechoslovakia and Germany. These are the regions of high cumulative emission levels. It should be noted that in large parts of Central and Southern Europe the computed cumulative deposition figures during the period 1955-1987 exceeded 10 mg/m^2.

For zinc (Figure 12.) the maximum in Europe is located in Southern Poland (EMEP grid [25,19]). The computed cumulative deposition is greater than 2600 mg/m^2. Depositions exceeding the level of 1500 mg/m^2 are found in Northern Germany, Belgium, The Netherlands and Great Britain. In the entire BTUS the computed depositions exceed 500 mg/m^2. Loads exceeding 100 mg/m^2 are noted in the southern Europe (Italy, the Balkans), Central Europe (Hungary, Romania, Austria), southern France and southern Scandinavia.

In the case of lead (Figure 13.) the maximum figures are computed for locations in Western Europe: Germany, The Netherlands, Belgium, France and Great Britain. In Central Europe and the BTUS the cumulative depositions exceed 500 mg/m^2. In Southern Europe and former Soviet Union computed depositions exceed 200 mg/m^2.

4.5 Uncertainties

Uncertainties of the model calculations discussed here depend strongly on the uncertainties of emission estimates. It is difficult to assess precisely the accuracy of emission estimates. This accuracy depends mainly on the accuracy of emission factor estimates and statistical data. Concerning the preliminary character of the presented data, it can be suggested that emission data for lead are more accurately estimated than those for cadmium and zinc. Recently Pacyna (1994) has suggested that reliability of emission data for Europe is in the following order: Pb > Hg and Cd > remaining heavy metals. An accuracy of <25% was suggested for the Pb emission estimates, 50% or less for Cd and Hg, and 100% for the rest of the metals, including Zn. It was concluded that a rather extensive body of information on Pb emissions from various sources in European countries is the major reason for the above presented order of accuracy for heavy metals. In addition, Pb is mostly emitted from

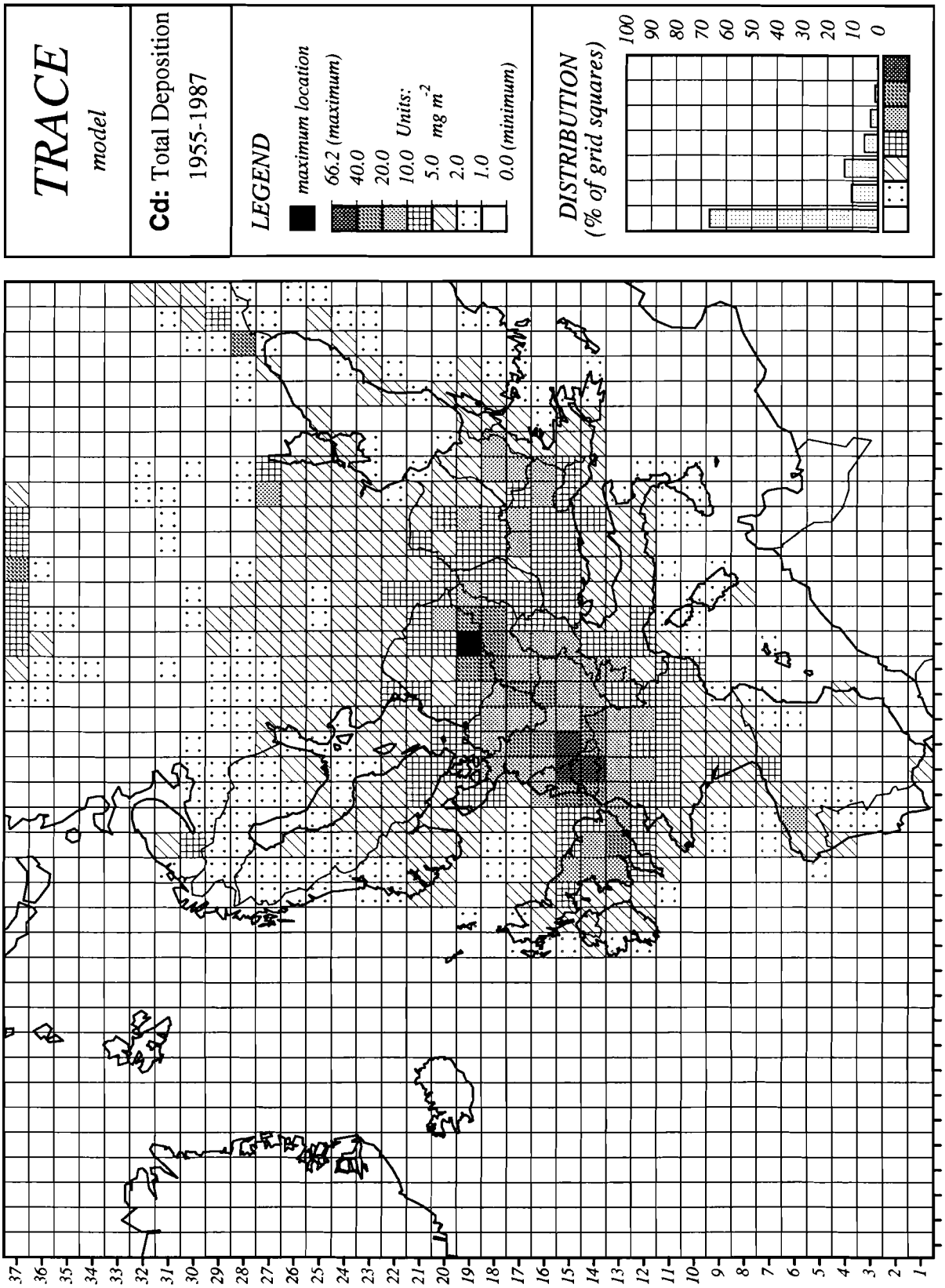

Figure 11. Cumulative deposition of cadmium in Europe during 1955-1987 computed by the TRACE model. Units: mg/m².

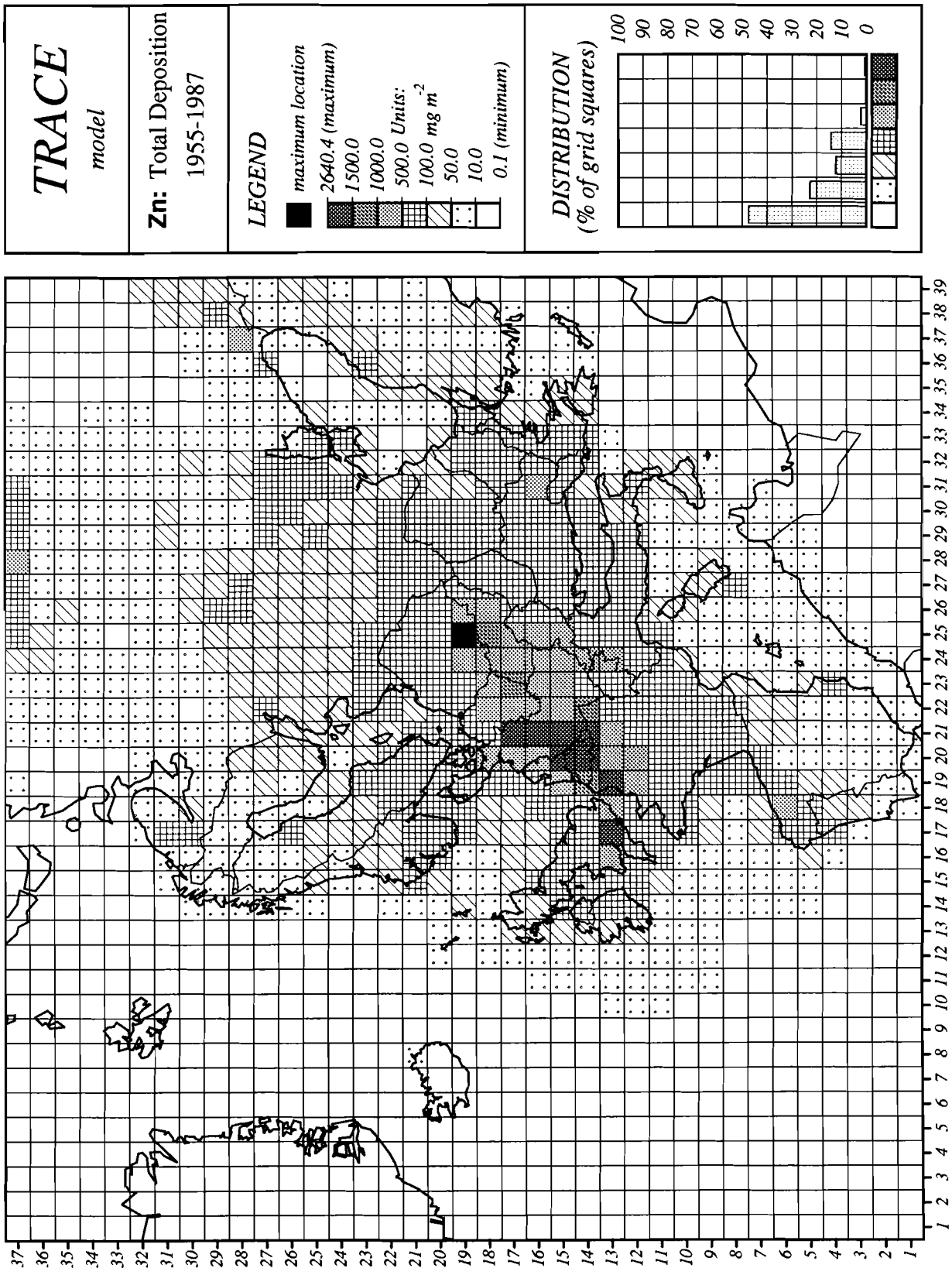

Figure 12. Cumulative deposition of zinc in Europe during 1955-1987 computed by the TRACE model. Units: mg/m².

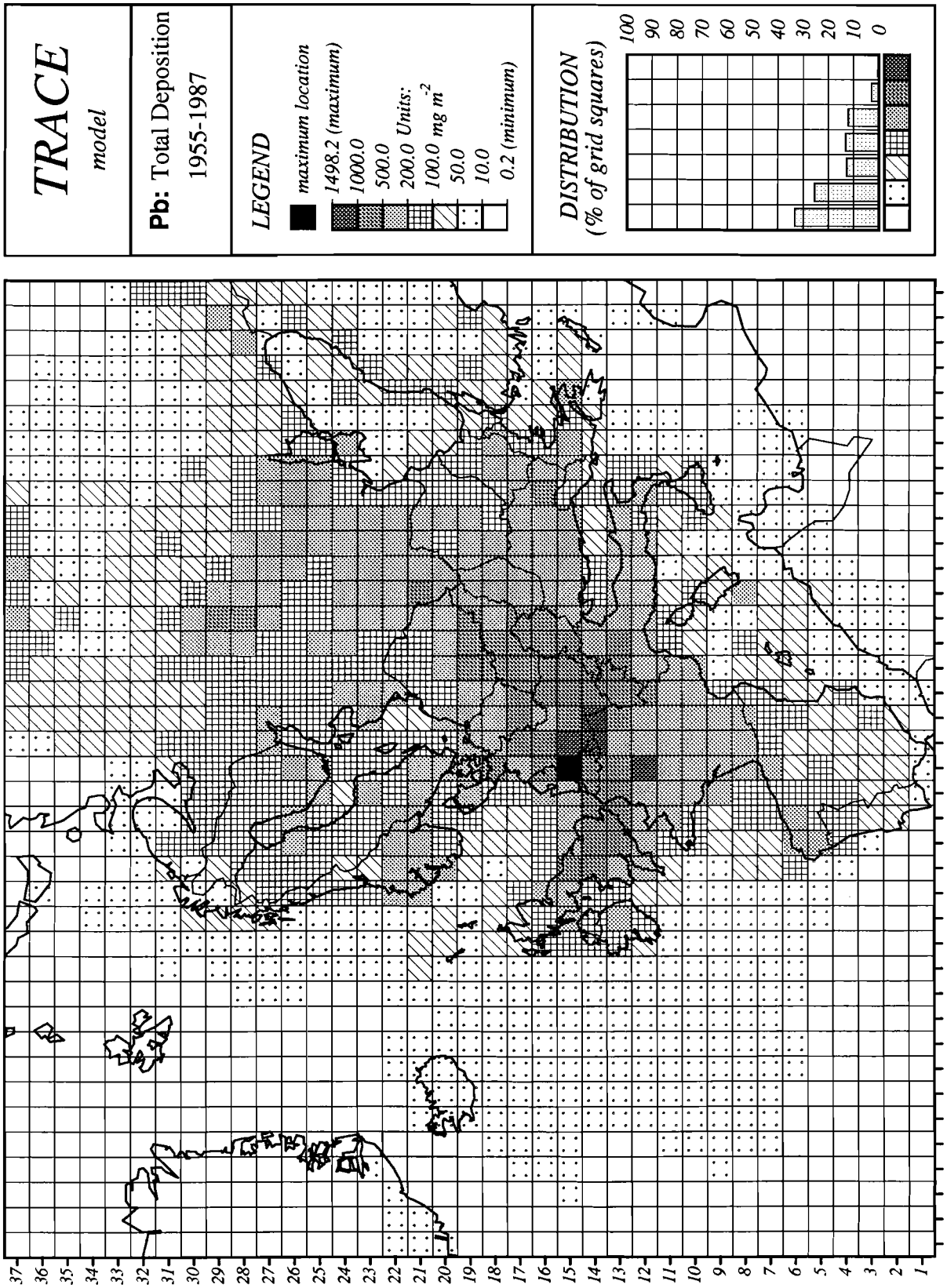

Figure 13. Cumulative deposition of lead in Europe during 1955-1987 computed by the TRACE model. Units: mg/m².

combustion of leaded or low-leaded gasoline. Fairly accurate information exists on the use of lead additives for gasoline in individual European countries, as well as on emission generation process during gasoline combustion. The Zn emission inventory is less accurate than the Cd emission inventory due to the fact that a sector list of emission sources for the latter metal is more complete than for Zn.

The other uncertainties of the results presented in this paper come from the TRACE model structure and its parameters. The approach in developing the TRACE model and the sensitivity to its key parameter values were extensively discussed by Alcamo et al. (1992). Here only the main conclusions are repeated. The improved climatological approach applied for TRACE seems to be a reasonable alternative to more sophisticated models especially regarding computations regarding past time periods (before 1990), for which there are few reliable measurements and the emission and meteorological data are either scarce or unavailable. The sensitivity analysis with regard to individual parameters (e.g. scavenging ratio, dry deposition velocity, local deposition coefficient) showed that TRACE is robust, i.e., the individual uncertainties of these parameters do not have a large effect on model computations. However this may not be the case for the combined effect of several model input parameters.

Other large sources of uncertainties include the applied computational procedure and the meteorological data. As was mentioned earlier, the spatial distributions of emissions, in form of gridded values, were available only for 1982 (Cd and Zn) and 1985 (Pb). The use of source-receptor matrices implies that the spatial distribution in each individual country varies, in time, linearly with the changes of the respective country total. This assumption does not hold true in case of rapid changes of country emissions due to opening or closures of industrial plants, or implementation of drastic emission reduction technologies. In such cases the spatial distribution of emissions varying with time within individual countries should be considered. This type of information is very difficult to obtain as most of the production and consumption statistics are on a national level.

To gather and process the required meteorological data for the entire investigated period, 1955-1987, is a formidable task. At this stage we did not attempt to complete this task. In view of the numerous uncertainties discussed above, instead of using fragmentary meteorological data from a number of years (1979-1987) that were at our disposal, we chose to apply a complete set for a single year 1985. This approach should be regarded as very preliminary. In subsequent papers we plan to apply a data series for a number of consecutive years, so that the interannual variations can be accounted for.

Finally, it should be mentioned that the TRACE model, like other long-range transport models (HMET, EMEP (Tuovinen et al., 1994)), assumes that emitted metals are homogeneously mixed both horizontally and vertically within each grid cell. This assumption is not fulfilled in reality, especially near strong sources. Therefore, in some parts of the grid cell, the actual concentrations may be significantly larger (one or two orders of magnitude) than the average computed by the model for the entire grid cell (150 x 150 km). Also the deposition levels within a single grid cell may very significantly especially in the case of high concentration levels and large variations in surface types. Then the dry deposition velocity becomes

highly variable (e.g. with respect to forests, arable land or urban areas) and the wet deposition depends strongly on the local precipitation patterns.

5. Conclusions

In this study we presented a preliminary estimation of atmospheric emissions of cadmium, lead and zink in Europe during the period 1955-1987. The emission inventory was then used as input for the TRACE model to compute the spatial patterns of cumulative atmospheric depositions. This information, although of preliminary character, is required for the assessment of environmental impacts and health effects of heavy metals. Moreover the cumulative soil inputs (target loads) can provide the basis for the development of policy recommendations for the long-term sustainable heavy metals use.

Heavy metals have been recognized as toxic chemicals causing adverse effects in the environment. Cadmium, lead and zinc has been assigned priority, considering their behavior in the environment and effects on human and environmental health (UN ECE, 1994). The above has been realized within major international organizations, such as the United Nations Economic Commission for Europe and the World Health Organization, which coordinate international activities aiming at proper management of the environment and structuring environmental policies. The UN ECE has established a Task Force on Heavy Metal Emissions with a goal to assess sources, fluxes, and behavior of heavy metals, and if necessary, to propose emission reduction scenarios. The Task Force is preparing documentation for the European-wide protocol on emission reductions for heavy metals. The results of the work described here, although preliminary, contribute significantly to our understanding of heavy metal behavior in the environment. For the first time, an approach has been made to assess cumulative deposition of Cd, Pb and Zn onto the terrestrial surface in Europe during the period of great industrial expansion. Also, the estimates of past emissions are a milestone, although somewhat incomplete and inaccurate. The results of the preliminary work discussed here are quite promising and indicative of how to obtain better accuracy and completeness of emission data and how to improve the modeling techniques. For example, information on consumption of low-leaded and unleaded gasoline should be searched individually for the European countries. The use of scaling factors should be reduced to the necessary minimum. During recent years, IIASA has been particularly active in providing the above mentioned information. One could clearly see the benefits and the potential users of this activity. The UN ECE, as well as the three major European conventions protecting the sea environment from air pollution, namely the OSPAR (the North Sea), HELCOM (the Baltic Sea), and BARCELONA (the Mediterranean Sea) conventions, should be interested in the outcomes of IIASA's modelling of cumulative depositions of Pb, Cd, and Zn in Europe, on the basis of historical emissions.

Another benefit of the discussed work is related to fulfilling air quality goals and proposing proper environmental policies in highly deteriorated regions in Europe, e.g. the BTUS Region. Clearly, lessons from the past on historical emissions concentrations and cumulative depositions should be used for the future "clean air" policies.

List of Tables and Figures

Tables:

Figures:

Appendix: Computation of air concentration in the TRACE model.

Since some typographical errors occurred in the Appendix of Alcamo et al. (1992) we give corrected equations below.

We propose that the concentration, c(x), of a non-decaying air pollutant is directly proportional to emission E, inversely proportional to distance x from source to a receptor, and decreases exponentially with time of travel from a source to a receptor $t^* = x/u$, where u is velocity:

$$c(x_r, y_r, x_e, y_e) = \beta \frac{E(x_e, y_e)}{x}(1-\alpha)e^{-(k_d+k_w)t^*}, \tag{A1}$$

where β is a proportionality constant, α is the local deposition coefficient, and k_d and k_w are the "dry" and "wet" deposition rates, respectively. The constant, β is derived by mass balance as follows. First, the upward flux of metals from emissions is set equal to the downward flux of deposition:

$$E'\tau = ck\tau\lambda h, \tag{A2}$$

where the left side gives the upward flux and the right side the downward flux, and E' is the total emissions from the model domain minus local deposition, k is the sum of k_d and k_w in Equation (A1), τ is an arbitrary time period, λ is the area of integration, and h is the average mixing height. Integrating to infinity, and substituting $2\pi x dx$ for λ gives:

$$E'\tau = \int_0^\infty c(x)k\tau 2\pi x h dx. \tag{A3}$$

Substituting (A1) in (A3), and canceling and re-arranging terms yields

$$\beta = \left(2\pi hk \int_0^\infty e^{-k(x/u)}dx\right)^{-1}. \tag{A4}$$

Assuming a constant u yields:

$$\beta = (2\pi hu)^{-1}. \tag{A5}$$

The assumption of a constant u can be avoided by using the relationship between travel time and distance:

$$t^* = x/u = ax^2 + bx, \tag{A6}$$

where a and b are empirically derived coefficients. Furthermore, for mass conservation reasons, we use two ks to solve (A4):

$$k = \begin{cases} k_1 & \text{for } (0, \bar{x}) \\ k_2 & \text{for } (\bar{x}, \infty), \end{cases} \tag{A7}$$

where $\bar{x}$ is the distance from the center to edge of the emitter grid cell. Using expressions (A6) and (A7) the integral of β in (A4) can be re-computed. Let this integral be:

$$A = k\int_0^{\infty} e^{-k(x/u)}dx. \tag{A8}$$

Then:

$$A = k_1\int_0^{\bar{x}} e^{-k_1(ax^2+bx)}dx + k_2\int_{\bar{x}}^{\infty} e^{-k_2(ax^2+bx)}dx. \tag{A9}$$

Letting y=x+b/2a and solving the integrals, yields:

$$A = k_1 e^{k_1\frac{b^2}{4a}}\int_{\frac{b}{2a}}^{\frac{b}{2a}+x} e^{-k_1 a y^2}dy + k_2 e^{k_2\frac{b^2}{4a}}\int_{\bar{x}+\frac{b}{2a}}^{\infty} e^{-k_2 a y^2}dy. \tag{A10}$$

$$A = k_1 e^{k_1\frac{b^2}{4a}}\int_{\frac{b}{2a}}^{\bar{x}+\frac{b}{2a}} e^{-k_1 a y^2}dy + k_2 e^{k_2\frac{b^2}{4a}}\left\{\int_0^{\infty} e^{-k_2 a y^2}dy - \int_0^{\bar{x}+\frac{b}{2a}} e^{-k_2 a y^2}dy\right\} \tag{A11}$$

The second integral in Equation (A11) is the Poisson-Euler integral with an analytical solution equal to $\sqrt{\pi}\Big/\left(2\sqrt{k_2 a}\right)$. The other two integrals, both on finite domains, are solved numerically.

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