Working Paper

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WP-96-30
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ABSTRACT

This paper presents the results of an analysis on the total load of cadmium, zinc and lead to agricultural soils during the period 1955-1994. Total heavy metal load will serve as input for the soil modeling part of the wider IIASA study on 'Regional Material Balance Approaches to Long Term Environmental Planning. The project area (fig. 1) embraces the northwestern part of the Czech Republic (Bohemia and Morawia), southwestern Poland (Upper and Lower Silesia), and the south of the former G.D.R. (Sachsen, Sachsen-Anhalt, Brandenburg, Thüringen). Agricultural soils receive heavy metal via atmospheric deposition and via certain agricultural practices, the most important ones are: P-fertilizer application and manuring. Atmospheric deposition loads were derived from computations within the atmospheric modeling part of the IIASA IND Project. On the basis of a literature search focusing on the countries of the project area heavy metal concentration factors for P-fertilizer and manure were established. The fertilizer and manure application during the study period was derived from diverse statistical sources.

The analysis shows the importance of regional differences and of the changes in time. This refers to both, the total load of heavy metals to the soils and the share of agricultural or atmospheric load in total load. The atmospheric load is highest in the 60s or 70s and then shows a donward trend. The highest P-fertilizer and manure application rates are in the 70s or 80s (and consequently the heavy metal load due to these practices is high). After the economic changes in 1989 there is a sharp decline in fertilizer application. The agricultural share in total load is very low in the case of lead, amounting to less than 10% during the whole period (fig. 9). Agricultural share in total load of cadmium and zinc varies considerable over time and shows high regional differences. For cadmium the agricultural share in total load ranges between 10 and 60 percent, in the case of zinc between 30 and 80 percent (fig. 5-8). A general feature here is, the higher the total load, the higher the share in atmospheric deposition.

A preliminary mass balance for cadmium and lead in soils shows possible implications for long-term build up of heavy metals in soils. The release of Cadmium from soils via erosion and leaching contributes as so called diffuse load to total Cadmium load to rivers. The mass balance gives estimates for this diffuse load.

Finally sources of uncertainties are discussed. They refer in particular to spatial variations that can not be traced in this type of analysis. Close to major heavy metal emittents or in areas where uncontrolled sewage sludge application took place, the heavy metal load may be significantly higher than estimated in this study.

Chapter 1. INTRODUCTION

Environmental pollution with persistent toxic metals is a phenomenon which requires analysis in a long-term perspective. Agricultural soils, that are the focus of this study, may be subject to long-term accumulation of heavy metals. They receive a heavy metal load via atmospheric deposition and certain agricultural practices like fertilizing, manuring or sewage sludge application.

The analysis presented in this study is part of a broader IIASA project on "Regional Material Balance Approaches to Long Term Environmental Policy Planning". This project seeks to trace the flow of three heavy metals (cadmium, zinc and lead) from industrial processes and other anthropogenic sources to the environment with particular focus on agricultural soils. Depending on agricultural practices, soil characteristics and certain environmental circumstances, like acid deposition, heavy metals may either accumulate in soils or they may be in a mobile form. In the latter case they are taken up by plants and leach out to ground- and surface water. Through edible plants or drinking water they enter the food chain. lead has a variety of impacts on the human nervous and circulatory system, while cadmium is considered a probable human carcinogen (WHO, 1987). Although zinc is a plant nutrient, at high concentration levels, it has been found to be moderately toxic to plants through disturbing the nutrient cycle (Rühling et al., 1987).

The objective of this study is to analyse the inputs of cadmium, zinc and lead to farmland over the period 1955-1994 for the project area due to agricultural practices. These data will be compared with the load due to atmospheric deposition derived from computations within the atmospheric modeling part of the IIASA Project. Only for cadmium the results are compared with a similar study in the Rhine Basin (Stigliani et al., 1993). This includes the adaptation of a simple methodology for a mass balance of cadmium in the soils.

The project area includes highly industrialized and severely polluted areas of the former centrally planned economies (see fig. 1). It includes the northwestern part of the Czech Republic (Bohemia and Morawia), southwestern Poland (Upper and Lower Silesia), and the south of the former G.D.R. (Sachsen, Sachsen-Anhalt, Brandenburg, Thüringen). The area around the common border of Poland, the Czech Republic and Germany are commonly referred to as "Black Triangle". The region is highly industrialized and densely populated. Half of the area is used for agricultural production. Extensive atmospheric impact of pollutants from coal burning and other industrial activities resulted in chemical deterioration of soils. Measurements show that some regions within the project area have elevated levels of heavy metal soil concentration (Kucharski et al., 1983-1994).

Fig. 1 Project Area of the IIASA study 'Regional Material Balance Approaches to Long Term Environmental Planning'

Chapter 2. METHODOLOGY OF ESTIMATES

2.1 Load of heavy metals due to agriculture

In general, the sources of heavy metals to the soils due to agricultural activities are:

- mineral fertilization
- manuring
- sewage, sewage sludge and wastes application
- pesticides application

These products applied to agricultural soils contain heavy metals as impurities. The estimates of our database account for phosphate-fertilizers and manure as sources of cadmium, zinc and lead. Other mineral fertilizers (nitrogen and potassium) are not included since their contribution to the total agricultural load of these metals to the soils would be very low and can therefore be neglected. The data available for heavy metals in lime fertilizers are too uncertain to be included in the database. The metal loads with sewage, sewage sludge, wastes and pesticides applications were also not included both for the lack of the data and for their much lower contribution to the total load. The chapter about uncertainties will discuss this in more detail later in the paper.

P-fertilization

The assessment of heavy metal load due to phosphate fertilizers is based on statistical data on P-fertilizers consumption for particular administrative units in the countries within the project area and on the literature data on heavy metal content in P-fertilizers. The source of the statistical data on P- fertilizers consumption are:

- Poland - Statistical Yearbooks published by GUS (The Main Statistical Office), Warsaw in the period 1955-1994 and FAO fertilizer yearbook,Vol.30, 1981, vol.42, 1992
- Czech Republic - Statistical Yearbooks (1955-1994), Czech Statistical Office, Prague
- Germany- Statistical Yearbooks (1955-1990) of G.D.R., Statistisches Bundesamt, Berlin; and Statistical Yearbooks (1990-1994) of Germany, Statist. Bundesamt, Berlin

Statistical data on the share of different types of P-fertilizers were available only for Poland. According to FAO Statistical Yearbooks the percentage share of particular P-fertilizers used in Poland on average during the period 1977-1990 was Single Superphosphate (48 %), Triple superphosphate (14 %), Ammonium phosphate (20 %), other P-fertilizers (5 %) and other complex fertilizers (13 %). For the whole project area we finally assumed a share of 50% Single Superphosphate; 25% Triple Superphosphate and 25% for Other and Complex Fertilizers.

The content of heavy metals in P-fertilizers varies and depends on kind of fertilizer and phosphate rocks used for their production. The sources of data on heavy metals in P-fertilizers are listed in Table 1. They were the basis for our estimates as described below.

Table 1. Cadmium, lead and zinc content in P-fertilizers in Poland and the Czech Republic (in mg kg^{-1} P).

Fertilizer	Cd	Zn	Pb	Reference
POLAND				
Single superphosphate ($18\% \ P_2O_5$)	2.2-2.6 2.3 - 9.6	130-180 152 150 	30-32 31 - 	Malysowa & Patorczyk -Pytlik (1986) Czuba & Siuta (1976) Górecki (1990)
Triple superphosphate ($46\% \ P_2O_5$)	17.6 44.2	372 -	37 -	Malysowa & Patorczyk-Pytlik (1986) Górecki (1990)
Complex fertilizers: NP($46\% P_2O_5$) NPK($24\% P_2O_5$)	 8.4 4.0			 Górecki (1990)
Phosphorus fertilizers - in general	0.1-170	50-1500	7-225	Kabata-Pendias & Piotrowska (1987)
CZECH REPUBLIC				References*
Superphosphate	8.8 8.4 17.0	172 - 184	3.7 2.0 -	Bican (1988) Tomkova(1983) Benes (1984)
Superphosphate granulated	10.2 15.4	170 -	3.25 2.0	Bican(1988) Tomkova(1983)
Triple superphosphate	0.12 0.45 19.2	0.68 20 -	- - <4	Bican (1988) Benes (1984) Benes (1994)
NPK-1	0.08	-	0.2	Tomkova (1983)
NPK-2	0.61	13.3	-	Bican (1988)

* all references after Benes (1994)

The concentration of *cadmium* in single superphosphate in Poland was on average (based on our literature sources) 5.9 mg/kg phosphor, which is equivalent to 32 mg/kg P_2O_5. In triple superphosphate the Cd content was on average 30.9 mg/kg phosphor (66 mg/kg P_2O_5). Complex fertilizers (NP, NPK) contain on average 17 mg Cd per kg P_2O_5. Since no data were available for Cd content in ammonium phosphate we assumed the same content as in complex fertilizers. Taking into account the percentage distribution of P-fertilizers in Poland we've got an input value of 32 Cd mg/kg P_2O_5. The average content of Cd in single superphosphates used in Czech Republic is assumed to be 12.1 mg/kg, in triple superphosphate of 7.0 Cd mg/kg and for complex fertilizers 0.2 Cd mg/kg. Following the same procedure as for Poland we got the input value of about 37.0 Cd mg/kg P_2O_5. Since the data we've got on the Cd content in P-fertilizers in Germany were too uncertain, Czech and Polish data were applied. Finally one value of 35 mg of Cd input per 1 kg of P_2O_5 applied was assumed for the whole project area.

For the *zinc and lead* content in P-fertilizers in Poland data by Malysowa & Patorczyk-Pytlik (1986) for superphosphates were used. Since no data on zinc and lead in other P-fertilizers were available it was assumed that the contribution of those fertilizers in zinc and lead input was very small compared to those in superphosphates and therefore it was neglected. Thus, following the way as for Cd it was calculated that in Poland the input of zinc was of about 500 mg/kg P_2O_5 and for lead it was equal to 100 mg/kg P_2O_5.

In the Czech Republic single superphosphates contained an average of 175 mg/kg zinc and 3 mg/kg lead. In triple superphosphate zinc was 10.3 and in complex fertilizers 13.3 mg/kg. For lead the content in triple and complex fertilizers was the same as for single superphosphates. Again, following the way as for Cd it was calculated that zinc input to the soils with P-fertilizers in Czech Republic was of approximately 504 mg/kg of P_2O_5 and for lead it was about 17 mg/ kg P_2O_5. Finally for zinc the value of 500 mg/kg P_2O_5 was accepted and for lead the value of 60 mg/kg P_2O_5 (as an average from the Polish and Czech data).

Estimates of Cadmium, zinc, lead load with manure

Manure production in the whole project area was derived from the number of animals per 1ha of agricultural land. Three major groups of animals have been distinguished: cattle, pigs and sheep. For Poland until 1982 horses were included as a fourth group. The sources of the data were the same Statistical Yearbooks as used for mineral fertilizers. According to Fotyma and Mercik (1992) the annual manure production by one so called "manure head" of particular animals (refers to the head in statistical yearbook) is for cattle 10 tones per year, for pig 1.5 tones, for sheep 0.8 tones and for horse 7 tones per year. Further it was assumed that the produced manure was evenly applied on agriculturally used land.

Data on cadmium and lead content in manure are scarce (Table 2). For Poland it has been decided to take an average value from the lower range value given by Piotrowska (1989) and Kabata -Pendias and Piotrowska (1987). Thus, the cadmium content is 0.3 mg/kg dry weight, and for lead it is 6 mg/kg dry weight. Assuming 25% of dry matter on average (Fotyma and Mercik, 1992) in manure it equals to 0.075 g of cadmium per 1 ton of the fresh manure applied and 1.5 g per ton for lead. These averages and Benes's data for cadmium and lead in manure in the Czech Republic resulted in a value of 0.055 mg/kg fresh manure for cadmium and of 1.0 mg/kg for lead. These values were used for calculations of cadmium and lead input with manure in the whole project area. For the zinc content in manure, the data by Andruszczak et al. (1988) are taken as they seemed to be the most representative. The authors studied 5258 samples of mixed farmyard manure taken from 826 farms located all over Poland in the period 1976-1982. They stated that the content of zinc in manure amounted , on average, to 40 mg/kg of fresh manure with a standard deviation of 21.07 and variability coefficient of 52.5. This is comparable with the range values given by Benes (20-90 mg). Thus, the assumed input of zinc in the whole region is 40g/ton of fresh manure.

Table 2. The content of Cd, Zn and Pb in manure

Fertilizer	Cd [mg/kg d.w.]	Pb [mg/kg d.w.]	Zn	Reference
POLAND				
slurry or manure	0.3-0.8.	5-7	15-340 mg/kg d.w.	Piotrowska (1989)
			17-42 ppm	Czuba 1979
manure	0.3-0.8	6.5-15	15-250 mg/kg d.w.	Kabata-Pendias & Piotrowska (1987)
mixed manure n=5258	-	-	avg. 40 mg/kg of fresh manure	Andruszczak et al. (1988)
manure n=58	-	-	27-343 mg/kg d.w. avg 174	Mazur (1972)
manure n=40			avg 18 mg/kg fresh manure	Koter&Krauze (1972)
CZECH REP.				
manure	0.035	0.4	3 mg/kg fresh manure	Benes (1994)
			20-90 mg/kg fresh manure	Benes (1994)

In summary the following concentration factors were used in this study for the estimates of Cd, Zn, Pb loads to the soils due to agricultural activity:

Cadmium: **0.035 g/kg P_2O_5 and** **0.055 g/1 ton fresh manure**
Zinc: **0.5 g/kg P_2O_5 and** **40 g/1 ton fresh manure**
Lead: **0.06 g/kg P_2O_5 and** **1 g/1 ton fresh manure**

2.2 Atmospheric deposition of heavy metals

The database on atmospheric deposition is derived from computations within the atmospheric modeling part of the IIASA IND Project. Atmospheric emissions of cadmium, lead and zinc in Europe during the period 1955-1987 were derived at IIASA and recently published in IIASA Working Paper (Olendrzynski et al., 1995). The corresponding atmospheric depositions were computed using IIASA climatological TRACE (TRace toxic Air Concentrations in Europe) model.

Recently, emission data for the period 1989-1992 became available as a result of collaboration with Jozef Pacyna from the Norwegian Institute for Air Research (NILU). These emission data together with the earlier data for the 1955-1987 period were fed into the long-range transport model HMET (Heavy Metal Eulerian Transport) developed by Jerzy Bartnicki form the Norwegian Meteorological Institute (DNMI). Consequently estimates on the atmospheric cumulative depositions during the period 1955-1994 were calculated for cadmium, lead and zinc using a more sophisticated Eulerian transport model (HMET) and an extensive meteorological data set provided by DNMI. The emissions beyond 1992 were assumed to be constant. For the years 1955-1989 a linear interpolation of country total emissions was applied to translate the cumulative deposition into yearly depositions.

In order to compare atmospheric deposition with the agricultural load some processing was necessary. Atmospheric deposition is estimated on a grid level (150x150 km) while agricultural input on an administrative unit level. Consequently a weighting procedure was performed on a Geographic Information System which assigned atmospheric deposition values to the administrative units. For those maps that demonstrate the total cumulative load of heavy metals (i.e. agricultural and atmospheric load over the period 1955-1994) an overlay of the atmospheric deposition grid and the administrative units polygons has been performed and their particulate heavy metals load were summed up.

Chapter 3. RESULTS AND DISCUSSION

Heavy metal load due to phosphate fertilizer and manure application depends on the intensity of agriculture in the project area. Application rates vary with time and region. The *phosphorus fertilizer application* in the years 1955-1994 ranged from 6.4 to 98.2 P_2O_5 kg/ha/a in eight voivodeships studied in Poland, from 5.7 to 97.0 P_2O_5 kg/ha/a in five districts of Czech Republic, and from 5.0 to 80.1 P_2O_5 kg/ha/a in four districts in Germany. Fig. 2 shows the time series for average P_2O_5 application. It can be seen that the P-fertilizer consumption constantly rose during the first 20 years. Then in the late seventies and 80s the application rates remained at a relative high level in the Czech and Polish part of the project area. In the German part (former G.D.R.) a slow downward trend can be observed. This reflects the general supply crisis of the G.D.R. economy in the eighties. Since about 1988 there is a significant drop in P-fertilizer use down to the application rates of the 50s. This is a consequence of the restructuring of the former centrally planned economies.

Manure production varied from 3.0 to 10.4 tons per year per ha of agriculturally used land in Poland, from 4.2 to 10.9 t/ha/a in Czech Republic and from 5.0 to 15.7 t/ha/a in Germany. Generally it was higher in Germany than in Poland and Czech Republic indicating the higher intensity of livestock production in the German part of the project area (fig. 3). Like for P-fertilizer application a sharp drop in manure application can be observed after 1988.

Fig. 2. Average P_2O_5 Consumption in the Project Area from 1955-1994

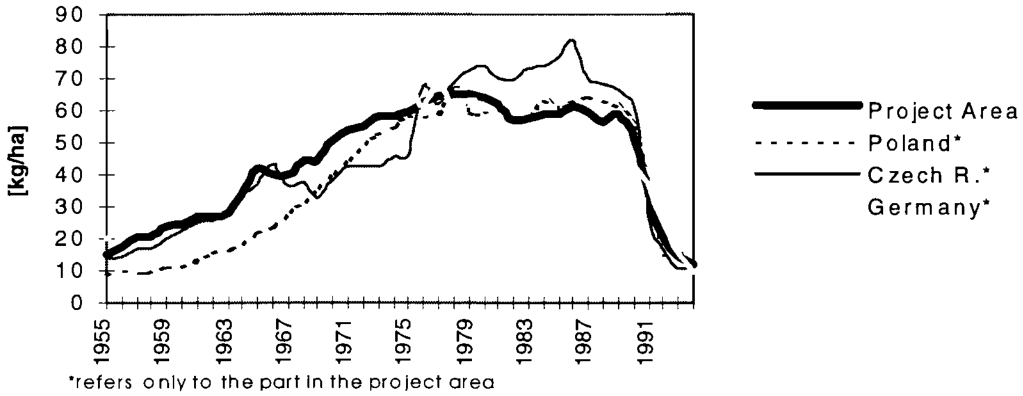
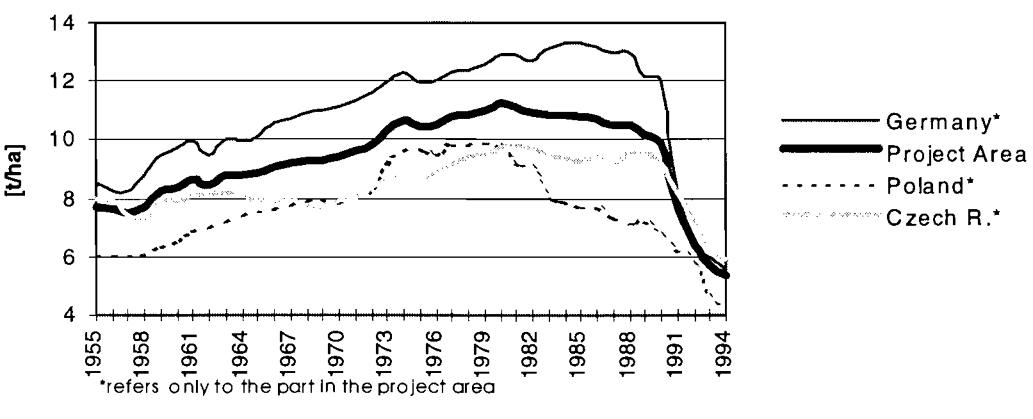


Fig. 3. Average Manure application from 1955-1994 (fresh weight in t/ha)



Chapter 3.1 CADMIUM

Table 3 presents a summary for the cadmium load to agricultural soils. The load with agriculture is derived from P_2O_5 consumption and manure production. Summing up agricultural and atmospheric load gives the total cadmium load per year for an administrative unit. In addition an average for the whole project area is given. It is based on the weighted contributions of each administrative unit.

The minimum *cadmium load via P-fertilizers* over time and in the project area was 0.2 g/ha/a, the maximum 3.4 g/ha/a. The average load for the different spatial units was from 1.1 to 1.5 g/ha/a in Poland, from 1.5 to 1.6 g/ha/a in the Czech Republic and from 1.5 to 1.8 g/ha/a in Germany. These values are lower in comparison with estimates done for the EU countries. In the mid 1970s average cadmium input with P-fertilizers was reported to be, in general, of 5.4 g/ha/a (Behrendt,1993). For the time around 1985 Fraters et al. (1993) estimate cadmium load via fertilizer on arable land 4.7 g/ha/a as an average for the EC12 countries (maximum in Germany, Belgium and Luxembourg 5.1 g/ha/a). The reason for this refers to both the generally higher fertilizer application rate in the EU compared to the Eastern European countries and a higher cadmium content in the P-fertilizers applied. Fraters study estimates twice as high cadmium concentration in P-fertilizer (78 mg/kg P_2O_5) than our estimates of 35 mg/kg P_2O_5.

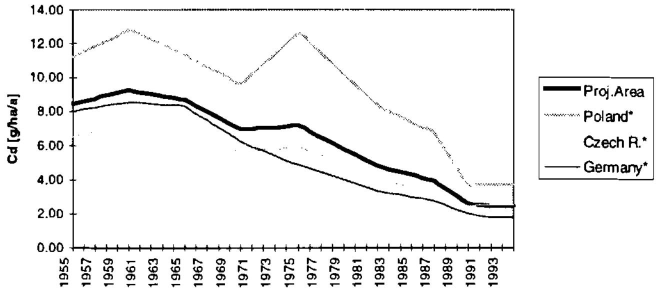
Cadmium load due to manure varied less and was lower compared to the load due to P-fertilizers. It ranged from 0.2 to 0.9 g/ha/a over the time period and over the project area with an average of 0.4 g/ha/a. Similarly to the cadmium load with P-fertilizers, the load with manure increased in 1970s and 1980s and decreased after 1990. However, the changes were not as high as important.

Table 3. Sources of cadmium loads to agricultural soils.
Minimum, maximum and average values (in g/ha/a).

Region	Cd	Cd	Cd	Cd	Cd
	via P-fertiliser	via manure	total agriculture	via atmosphere	total load
Polish part of project area					
minimum	0.2	0.2	0.4	1.6	2.1
maximum	3.4	0.6	4.0	23.2	25
avg(55-94) for adm. units	1.1-1.5	0.4-0.5	1.6-2.0	4-13	5.8-18
Czech part of the project area					
minimum	0.2	0.2	0.4	1.7	2.2
maximum	3.4	0.6	4.0	9.6	11.4
avg(55-94) for adm.units	1.5-1.6	0.4-0.5	1.9-2.1	3-6	6.1-9.1
German part of the project area					
minimum	0.2	0.3	0.6	1.3	1.9
maximum	2.8	0.9	3.7	10	12
avg(55-94) for adm. units	1.5-1.8	0.5-0.7	2.0-2.5	4-6	6.6-8.5
avg(55-94) for project area	1.4	0.4	1.9	5.9	9

Total agricultural load (i.e. P-fertilizer and manure load) amounted from 0.4 to 4.0 g/ha/a with an average of 1.9 g/ha/a. These figures are about half of the average cadmium load in the 12 European Community countries around 1985 where it was estimated to be equal to 5.4 g/ha/a on arable land and to 4.2 g/ha/a on grassland (Fraters et al., 1993). Assuming our results P-fertilizer contribute about 4/5th and manure 1/5th of total agricultural load.

Fig. 4 Average atmospheric cadmium deposition during 1955 and 1994

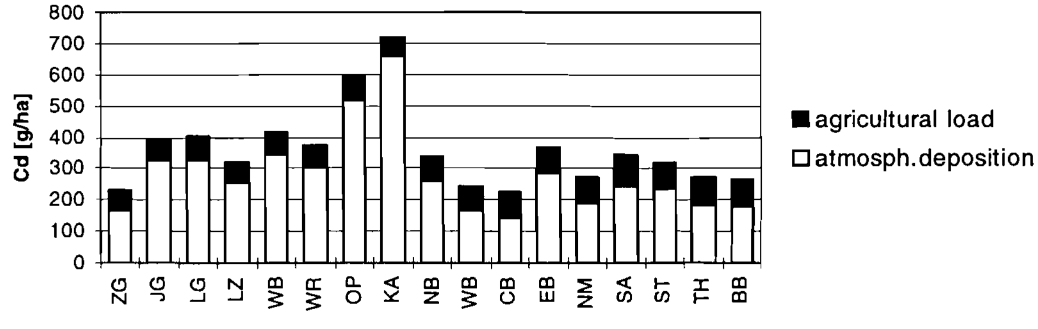


*refers only to the part of the project area

Compared to the rest of Europe the project area has high *atmospheric cadmium deposition* including the grid with the highest cumulative deposition in Europe. However the whole project area shows considerable differences in cadmium deposition ranging from 1.6 g/ha/a to 18.3 g/ha/a. Averages over time for the different spatial units are shown in figure 4. The general trend is a peak of atmospheric deposition in the 60s. Since then there was a decrease in the Czech Republic and Germany. Poland had another peak in 1978. Finally there is a dramatic decrease since 1988, which reflects the political and economic changes.

The *total cumulative cadmium load* is the sum of the agricultural and atmospheric load over the period 1955 to 1994. Figure 5 shows these loads for each administrative unit differentiated by their source. It ranges from 190 g/ha to 605 g/ha with an average of 298 g/ha. Map 1 shows the geographic distribution of the total cumulative cadmium load to the soils. It has been derived by overlaying the atmospheric deposition grid with the administrative units and summing up their particulate load of heavy metals. In addition a land use map screens out non-agricultural land. The highest total cumulative load is in the Polish voivodships of Opole and Katowice. The total cumulative load to the project area (100,000 km^2 of agricultural land) is 2880 t, comprising of 2003 t from atmospheric deposition and 829 t from agricultural sources.

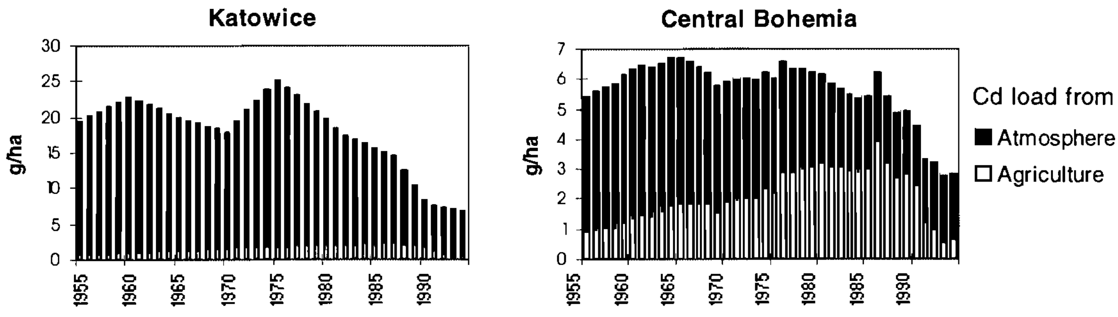
Fig. 5 Cumulative cadmium load to agricultural soils from 1955-1994 for the administrative units in the project area



POLAND: ZG=Zielona Gora,JG=Jelenia Gora, LG=Legnica, LZ=Lezno,WB=Walbrzych, WR=Wroclaw,OP=Opole,KA=Katowice; CZECH REPUBLIC: NB=North.Bohemia, WB=WesternBohemia, CB=Central Bohemia, EB=Eastern Bohemia, NM=Norhtern Morawia; GERMANY: SA=Sachsen, ST=SachsenAnhalt, TH=Thüringen, BB=Brandenburg

The *percentage share of agricultural or atmospheric load* in total load varies considerable over the project area and slightly over time. In figure 6 the development is shown for two administrative units. They represent the lower and upper limit of the possible range. In general, the higher the total load of cadmium the higher the share of atmospheric load in total load. In Katowice, which has a high total load of about 12-20 g/ha/a, the major contribution of cadmium stems from the atmosphere. In Central Bohemia the overall load is about 3 times lower than in Katowice. Here the share of agriculture in total load goes up to 60%. Map 2 shows the geographical distribution of atmospheric share in total cumulative cadmium load. It can be noticed that for about a third of the project area the atmospheric share in total load is more than 70%, for another third it ranges between 60 and 70% and for the remaining third, agricultural soils have received about half the load from agriculture and half from the atmosphere. Compared to the Western European Countries, in our project area the share of atmospheric deposition is higher. Alloway (1990) has estimated the relative contributions of cadmium in western countries to be: phosphoric fertilizers 54-58%, atmospheric deposition 39-41%, and sewage sludge 2-5%.

Fig. 6. Total cadmium load to agricultural land for two districts in the project area



Attempts to a mass balance

From a more policy oriented point of view, an important question, to be determined, is to estimate potential impacts resulting from a cumulative cadmium load. Mass balances of heavy metals in soils are a tool to get some insight in this problem. In general, the output flows for heavy metals are: removal via erosion; crop uptake and subsequent removal via harvest; and leaching from the plough layer to deeper layers. In the latter case heavy metals will finally reach ground or surface-water. This transport is dependent on soil characteristics, in particular soil type, pH-value and organic matter. The difference between input and output is the net accumulation in the plough layer of the soil. It is obvious that an accurate mass balance is a difficult and uncertain task considering the complexity of the processes in soils and the variability of soil characteristics and land use in a spatial unit.

A similar study like ours including a preliminary mass balance has been undertaken for the Rhine River Basin (Stigliani et al., 1993). The Rhine Basin study and our project area are well comparable because they cover about the same area of utilized agricultural land (100,000 km^2) and nearly the same time period has been studied (Rhine study: 1950-1988, our study: 1955-1994). The Rhine Basin study assumed that crop uptake, surface runoff and erosion are sources of cadmium losses. No leaching is considered because due to liming, a high fraction of cadmium is sorbed in the soil (Table 4). The estimated input in the Rhine basin is higher (4000 t) than the input in our study area (3000 t). The major reason is the higher cadmium-input via P-fertilizer in the Rhine Basin. Applying the output rate of the Rhine study to our project area (1.3 g/ha/a), the net gain in the soils in our project area is 248 g/ha. The equivalent in soil concentration is a net increase in a 40 year period of 0.08 mg/kg (0.1 mg/kg in the Rhine Basin). For comparison Benes (1994) estimated for the Czech Republic a total output rate of cadmium from soils of 5 g/ha/a and Kabata-Pendias (1989,1993) estimated for Poland an output of 6 g/ha/a. However, no sources for these estimates are given.

**Table 4. Mass Balance for cadmium on agricultural land.
Comparison with the Rhine Basin Study**

	Rhine Basin Study*		Project area of this Study	
study period	1955-1980		1955-1994	
input	4000 t	400 g/ha	3000 t	300 g/ha
output (1.3 g/ha/a)	500 t	50 g/ha	520 t	52 g/ha
net gain over study period	3500 t	350 g/ha (0.1 mg/kg soil concentr.)	2480 t	248 g/ha (0.08 mg/kg soil concentr.)

** Source: Stigliani et al., 1993,*

Though these calculations are very rough and average and obviously uncertain, they point towards the slow increase in cadmium soil concentration and the low increase in relation to existing guideline values. cadmium concentration in unpolluted agricultural soils are in the order of 0.5 mg/kg for Central & Eastern Europe (Kabata-Pendias,1993). For Poland the average background value is 0.35 mg/kg (Smal, 1995). Alloway (1990) estimated the cadmium content in agricultural soils at 0.2-1.0 mg/kg for the countries of the European Union. The legal standards for cadmium content in soils are 3 mg/kg in Poland. In the Netherlands a so called "signal value" for agricultural soils is between 0.5 - 3 mg/kg depending on soil characteristics. At concentrations higher than this value, the quality standards for food might be exceeded (Vegter, 1995).

The reverse side of a slow accumulation are the difficulties to remove cadmium from the soils once it has been accumulated. The only practically feasible solution then is a change in land use, i.e. limitations of food production in areas of high accumulation, which may induce social and political problems. A part of our project area, namely Katowice voivodship, faces this problem today. An extensive soil sampling program on arable soils in the 80s showed that the cadmium content ranged from 0.7 - 143 mg/kg with a geometric mean of 3.2 mg/kg (Smal, 1995). Performing the above mass balance here, it seems not possible to explain the very high concentrations in the soils. A major problem is that it is impossible to know the cadmium soil content in 1955. However, even if we assume a high cadmium soil content of 1 mg/kg, this means we had an accumulation of 2 mg/kg. The above estimates give an accumulation over 50 years of only about 0.1 mg/kg. There may be many reasons for this. One explanation could be is that we've underestimated atmospheric deposition, at least for a certain period. Another possibility is that the cadmium soil concentration in 1955 was already very high due to mining, smelting and other industrial activities before 1955. Industrialization grew already important in this region in the second half of the 19th century.

It may for example be that a some of the farmland is located on old mine tailings. Or natural background values of heavy metals in this region may be especially high in this region.

Another result that may be derived from a mass balance for cadmium in soils is the load of cadmium from agricultural land to rivers. If we assume that half of the yearly output is taken up by plants and the other half finally ends in the rivers, we have an average yearly load to the rivers of 7 tons. Since about two thirds of the project area drain into the river Elbe, we can estimate an average yearly load of 5 tons to the upper basin of the river Elbe. In 1991 1.7 t of cadmium was released to the river Elbe from industrial aqueous point sources (Simon, 1994). The river Rhine received 13 t of cadmium via aqueous point sources in 1988. These very approximate figures indicate the importance of diffuse pollution loads into rivers and the necessity to include the analysis of diffuse sources into a program for the clean-up of a river.

Chapter 3.2 ZINC

Table 5 presents a summary of the sources of zinc load to agricultural soils. zinc loads due to agriculture are generally higher (commonly two thirds from agriculture) than the load from atmospheric deposition with the exceptions of Opole and Katowice, which have a very high atmospheric deposition (figure 7). The major agricultural zinc source is manure application caused by the high zinc concentration in manure. Average zinc loads over time reflect the intensity of livestock production in the three countries of the project area, ranging from the most intensive in Germany over the Czech Republic to Poland which had the least intensive agricultural production.

Load of zinc due to P-fertilizer is at least 10 times, during certain periods up to 70 times lower than the load due to manure application. Zinc load via manure ranged from 118 to 627 g/ha/a, while zinc load via P-fertilizer ranged from 3 to 49 g/ha/a. Our estimates for zinc load via P-fertilizer are lower than those reported for EU countries. There, the zinc load with P-fertilizer was estimated to be from 40-60 g/ha/a (Feernstr & van Baal,1989; after Behrendt 1993).

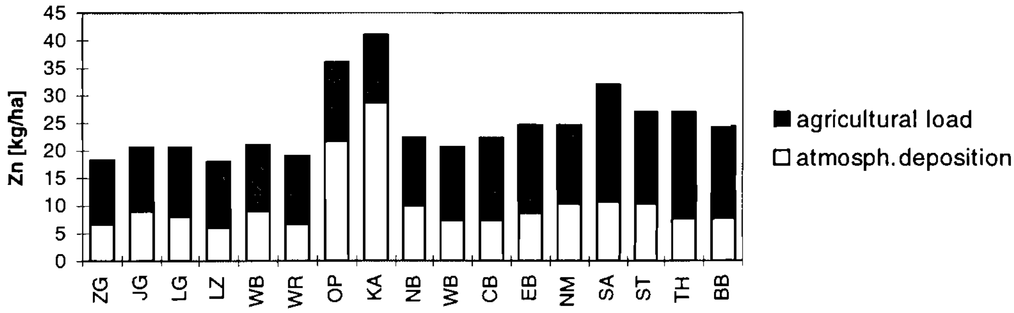
Table 5. Sources of zinc loads to agricultural soils
Minimum, maximum and average range values (in g/ha/a)

Area	via P-fertiliser	via manure	total agriculture	atmosph. depos.	total load
Polish part of project area					
minimum	3.2	118	121	48	173
maximum	49.1	416	452	879	1328
avg.(55-94) for adm. units	16.4-21.7	273-339	292-360	126-608	446-1027
Czech part of the project area					
minimum	2.9	166	169	63	258
maximum	48.5	436	476	295	711
avg.(55-94) for adm. units	21-22.8	285-374	306-397	142-204	516-621
German part of project area					
minimum	2.5	200	205	47	265
maximum	40	627	667	379	934
avg.(55-94) for adm. units	21-25	396-502	417-528	154-220	612-796

Atmospheric zinc deposition ranged from 47 to 879 g/ha/a with the highest values occurring in the Polish part of our project area. Finally the sum of agricultural and atmospheric load ranges from yearly loads of 173 to 1328 g/ha. A time series of the total load for three administrative units is given in figure 8. A significant decrease can be observed during the last 10 to 20 years.

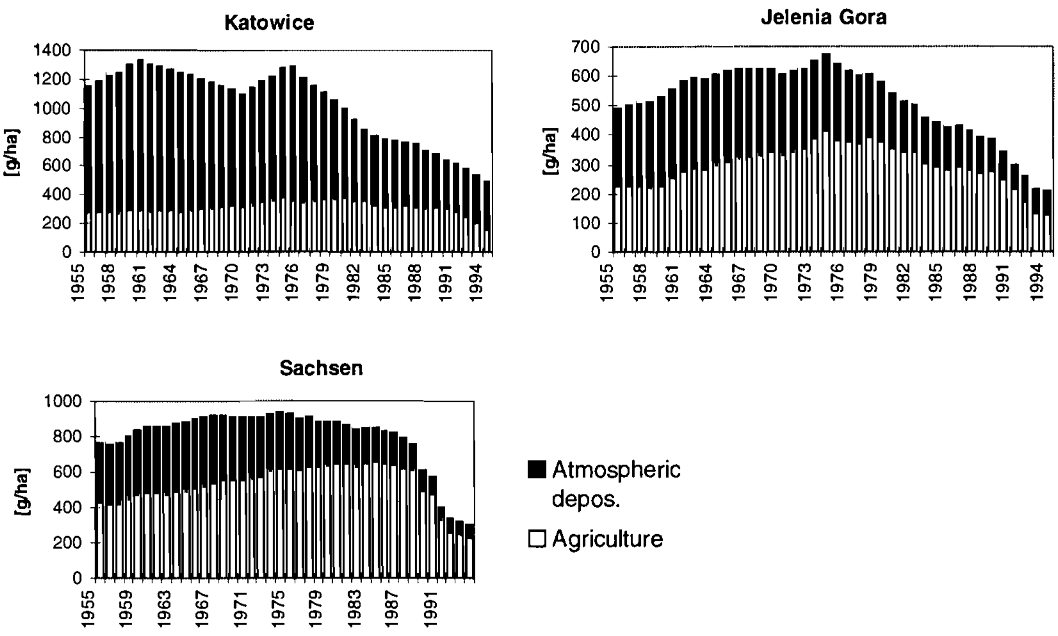
The *total cumulative zinc load* over the period from 1955 - 1994 ranged from 18 kg/ha to 42 kg/ha (fig. 7). Map 3 and figure 8 illustrate regional differences within the project area. As in the case of cadmium, the highest load occurs in Katowice and Opole, two districts in Southern Poland.

Fig. 7 Cumulative zinc load to agricultural soils from 1955-1994 for the administrative units in the project area



POLAND: ZG=Zielona Gora,JG=Jelenia Gora, LG=Legnica, LZ=Lezno,WB=Walbrzych, WR=Wroclaw,OP=Opole,KA=Katowice; CZECH REPUBLIC: NB=North.Bohemia, WB=WesternBohemia, CB=Central Bohemia, EB=Eastern Bohemia, NM=Norhtern Morawia; GERMANY: SA=Sachsen, ST=SachsenAnhalt, TH=Thüringen, BB=Brandenburg

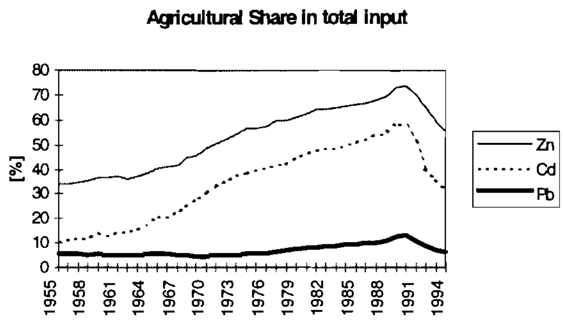
Fig 8. Atmospheric and agricultural Zinc load to agricultural soils for three provinces in the project area



Chapter 3.3 LEAD

In contrast to cadmium and zinc nearly the whole load of lead to agricultural soils is due to atmospheric deposition. In almost all cases the share of agricultural load in total load is smaller than 10% (fig. 9). The total load ranges between 70 and 480 g/ha/a. There was a constant increase in lead load until the peak in the early seventies. Then the load decreased down to the level of 1955 in 1988. This trend was observed in all three countries. The load in Poland was 10-20% higher than in the Germany and the Czech part of the project area. The cumulative total load over the period 1955 to 1994 ranges from 5 kg/ha to 13 kg/ha. Since the share of atmospheric deposition is very high the geographic distribution of the cumulative load correlates with the grid from the atmospheric deposition modeling (map 4).

Fig. 9 Agricultural Share in total load of Zn, Cd and Pb based on average values for the project area



Once lead has entered agricultural soils it is very likely to accumulate. This is because the binding strength of lead is strong above a pH-value of 4 and very strong above a pH-value of 5 (Blume and Brümmer, 1991). In order to obtain optimal yields, the pH-value of farmland soils is maintained between 6 and 7 by means of liming. Even if we assume poor liming practice or high acid deposition, the pH-value of agricultural soils is unlikely to fall below 5. Consequently we can assume that most of the load of lead accumulates in agricultural soils. Some losses occur via surface runoff and erosion.

A cumulative load of 5 to 13 kg/ha translates into a soil concentration increase of 1.6 to 4.3 mg/kg in the plough layer (upper 20 cm of the soil). Background values in 'unpolluted' soils in Poland are between 10 and 25 mg/kg, for Central & Eastern Europe 35 mg/kg (given in Smal, 1995). Depending on the background reference value, increase of 1.6 to 4 mg/kg over a 30 year period is rather small. Compared to existing guideline standards this increase is very small. Guideline standards for lead concentration in Poland amount to 54 mg/kg for light textured soils and 100 mg/kg for clayic soils. In the Netherlands the target value for lead concentration standard soil (10% organic matter and 25% clay) is 85 mg/kg. Guideline values are high compared to background values because lead strongly binds to soil particles and plant-uptake therefore only occurs at high lead concentrations in soils. With respect to danger of lead entering the food chain, soils have a high capacity to accumulate lead. On the other hand the residence time of lead in soils is also very high, amounting to 700 - 6000 years.

Like in the case of cadmium our estimates of lead load to soils can not explain measured high lead concentration in Katowice voivodship, which has the highest lead deposition within the project area. An extensive sampling program on arable soils (n=2270) in the 80s shows lead soil concentrations from 4 to 8200 mg/kg with a geometric mean of 100 mg/kg. Again, there may be several reasons why our estimates can not explain these high measured concentrations. First, we have underestimated atmospheric deposition at least during a certain period. Our estimated maximum load in Katowice is 480 g/ha/a in the 70s.

Measurements of atmospheric deposition in Katowice give for rural areas for 1981 commonly 500, 700 but sometimes 1000 or 2000 g/ha/a, for 1992, 200 to 400 g/ha/a (SANEPID, 1981, 1992). Measurements in urban areas are much higher amounting to 2000, 7000, 10000 and up to a maximum of 70000 g/ha/a. This shows that our estimates for rural areas are reasonable. In some areas however, we may have underestimated the load. Assuming a high load in rural areas of 2000 g/ha/a over a 20 year period we get a lead accumulation in soils of about 10 mg/kg. A second reason may be that before 1955 there has already been a 50 to 100 year history of lead load to the soils. A third reason may be that on those soils where high lead concentrations were measured certain practices like contaminated sewage sludge application or use of fertilizer as by-product from industrial processes took place.

From a risk assessment point of view, these back of the envelope calculations point towards the importance of the local scale. Accumulation of lead in soils is slow. However, once it took place only restrictions on land use may eliminate risks for lead entering the food chain.

Chapter 4. UNCERTAINTIES

These type of analysis is subject to several uncertainties. First there are difficulties in obtaining data for the project area (e.g. origin and source of heavy metals in fertilizers). Further spatial discontinuities can not be traced, since our assumption on agricultural practices (mineral fertilizer and manure application) are based on statistical data for administrative units. We have to assume an even application of e.g. manure production or consumed fertilizers on all agricultural area within an administrative unit. This does not reflect the real practices and the load may therefore be under- or overestimated in certain areas. However, these types of uncertainties are considered to be rather small in the framework of a large project area and the long study period.

The major sources of uncertainties due to agricultural activity refer to those sources of metals that have not been included in our assessment, like lime fertilizers and in particular sewage sludge application. As a consequence this study must have underestimated the heavy metal load, at least on a local scale.

Lime fertilizers such as ground lime in the Czech Republic are low in cadmium (0.04 mg/kg), lead (2.0 mg/kg), zinc (80 mg/kg) contents (Benes, 1994) and their contribution to the total load can be neglected. According to Dutch data (Fraters and Beurden, 1993) cadmium load due to application of lime is 0.4 g/ha/a on arable land and 0.06 g/ha/a on grassland. A load of 0.4 g/ha/a is equal to the lower ranges of load of cadmium via P-fertilizer. However, quite considerable amounts of heavy metals could have been loaded with lime fertilizers being by-products of industrial processes. Available Polish data indicate that waste limes could contain from 0.3-9.5 mg/kg cadmium, from 5-4000 mg/kg of lead and from 16-2500 mg/kg zinc (Kabata-Pendias and Piotrowska, 1987). Hence, locally, where such kind of limes were applied the heavy metal load could have been substantial.

The contribution of *sewage sludge application* to the total load of cadmium, zinc and lead in the project area was difficult to estimate. In Poland, in general, sewage sludge application was low because only a small percentage of waste water has been treated. Between 1972 and 1993 on average only on 1.2% of total farmland sewage sludge has been applied (GUS, 1990). About the same share applies to our project area. However, since sewage sludge application was not controlled for those small areas which received sewage sludge the heavy metal load may have been high.

Table 6 shows results of preliminary calculations on heavy metal load via sewage based on data from Czycyk F. (1994) who gives ranges for the content of cadmium, zinc and lead in municipal sewage for Wroclaw, Legnica, Kluczbork and Koscian. These towns are located in our project area. Heavy metals concentration in sewage varies considerable between towns and amount to 0.0003 - 0.026 mg/dm^3 for cadmium, 0.005 - 0.052 mg/dm^3 for lead and 0.031 - 1.63 mg/dm^3 for zinc. The calculations below are based on average values for the towns (Cd: 0.008 mg/dm^3; Pb: 0.041 mg/dm^3; Zn: 0.785 mg/dm^3).

The heavy metal load via sewage is approximately comparable to or a bit higher than the load via P-fertilizer and manure. The exception is Wroclaw voivodship, where intensive sewage sludge application took place on a small area (about 0.6 % of total farmland) during the last 15 years. There the loads amount to 60 g/ha/a for cadmium, 309 g/ha/a for lead and 9614 g/ha/a for zinc. In some areas in Wroclaw voivodeship municipal sewage from the town Wroclaw has been applied since 1881 (Czyczyk, 1994).

Table 6. Heavy metal load via sewage application. Average during 1978-1993 for five voivodships in the project area

	Jel. Gora	Legnica	Opole	Wroclaw	Ziel. Gora
Sewage application in ths m3	106	480	1939	16023	249
Area with sewage application [ha]	636	1361	5959	2123	8767
Area with sewage application [% of total arable land]	0.5	0.7	1.3	0.6	3.2
Cd load [g/ha /a]*	1,3	2,8	2,6	60,3	0,2
Pb load [g/ha/a]	6,8	14,2	13,3	309,4	1,2
Zn load [g/ha/a]	212	443	197	9614	36

* per ha of the area with sewage application

In the former G.D.R. application of sewage sludge on agricultural fields was a common but uncontrolled practice. About 65% of the 1.1 mio. t of sludge annually produced by the 1.100 public treatment plants was used in agriculture. Additionally it is known that soil improvement measures were also carried out by spreading heavy metal-enriched power plant filter ashes on agricultural land (Reuther, 1995).

No information was found about the practice of sewage sludge application in the Czech Republic. It may be assumed that practices were similar than in Poland and in the former G.D.R. Today both, the Czech Republic and Germany invest extensively in new sewage treatment plants. As part of the program on the "International Commission for the Protection of the river Elbe" 60 sewage treatment plants of a capacity of more than 20 TEGW and 78 sewage treatment plants of a capacity of more than 50 TEGW are planned for both countries. In 1994 approximately half of them were completed or are under construction (Simon, 1994). In the "New Countries" of Germany (former G.D.R.) in 1991, 20% of total sewage sludge production was applied on agricultural land. Germany today has regulations restricting the heavy metal content in sewage sludge that is applied to agricultural land.

Uncertainties in *atmospheric deposition* estimates have been discussed in detail in Olendrzynski et al (1995). They refer on the one hand to uncertainties of emission estimates.

An accuracy of <25% was suggested for the Pb emission estimates, 50% or less for Cd and 100% for Zn. The other uncertainties refer to the model structure and its parameters. The TRACE model is a long-range transport model and consequently assumes depositions homogeneously mixed within each grid cell (150x150 km). Since this study is of a regional scale covering only 18 grid cells, variations within a grid cell are of relevance. Olendrzynski et al. suggest that especially near strong sources, in some parts of the grid cell, the actual concentrations may be significantly larger (one or two orders of magnitude) then the average computed by the model for the entire grid cell.

Summarizing the most important uncertainties are considered to be an underestimation of the heavy metal load on a local scale, in particular close to major heavy metal emittents and on those areas where uncontrolled sewage sludge application took place.

Chapter 5. CONCLUSIONS

This paper presents estimates of total Cd, Zn and Pb loads to agricultural land during the period 1955-1994 for a project area (Fig.1) located in the coexistent borders of Poland, the Czech Republic and East Germany (former G.D.R.). The sources of heavy metal load are atmospheric deposition and certain agricultural practices like P-fertilizer and manure application, both contain heavy metals as impurities.

Agricultural and atmospheric load changed over the studied time period. Depending on the region the difference between minimum and maximum load was for cadmium a factor of 5 - 10, for zinc and lead a factor of 3-5. In most cases the total load increased slightly at the begin of the study period with a peek in the seventies or early eighties. Since about 1988 there is a sharp decrease in heavy metal load in the entire project area. This reflects the political and economic changes in the former centrally planned economies. The economic changes induced emission and consequently atmospheric deposition decreases and a sharp decrease in fertilizer application (Fig. 2 and 3).

There are considerable regional differences in total heavy metal loads within the project area, mainly due to differences in atmospheric deposition (Map 1,2 and 3). Areas with the highest cumulative Cd load receive three times more than those areas with the lowest load, for Zn and Pb two times respectively.

The agricultural share in total load is less than 10% for Pb during the entire time period. For Zn and Cd the agricultural share varies with time and region. For the average project area it increased with time reaching a peak in the late 1980s. In general the higher the total deposition the smaller the contribution of agricultural load compared to atmospheric deposition (Fig. 6 and 8).

On the local scale an underestimation of our heavy metals load is likely because potential heavy metal sources like sewage sludge were not included or the higher deposition rates in the vicinity of the emittents were not accounted for in the resolution of the atmospheric transport model.

Preliminary attempts to a mass balance for cadmium and lead point towards the necessity to study heavy metals in agricultural soils in a long-term perspective due to the slow accumulation rates. Once accumulation took place, it is practically impossible to remove the heavy metals from the soils. Only via certain management practices like liming or no food production on contaminated soils this problem can realistically be approached. The so called diffuse load of cadmium to rivers stemming from surface runoff, erosion and leaching from soils seems to be an important share in total cadmium load to rivers.

Even though the heavy metal load for the Katowice voivodship in Upper Sileasia is the highest within our project area, it seems not possible to explain the high measured heavy metal soil concentrations by our estimated input loads. There may be several reasons for this: First we've underestimated the atmospheric deposition (or better the atmospheric emissions) in this region at least during a period of 10 to 20 years, most probably in the 70s and early 80s. Second the region has experienced a significant heavy metal load before our studying period, i.e. before 1955. Since the region has a long tradition of mining and metal refinement these soils must have been exposed to heavy metal contamination for centuries. Background concentrations may also be hihger in this region compared to common background values.

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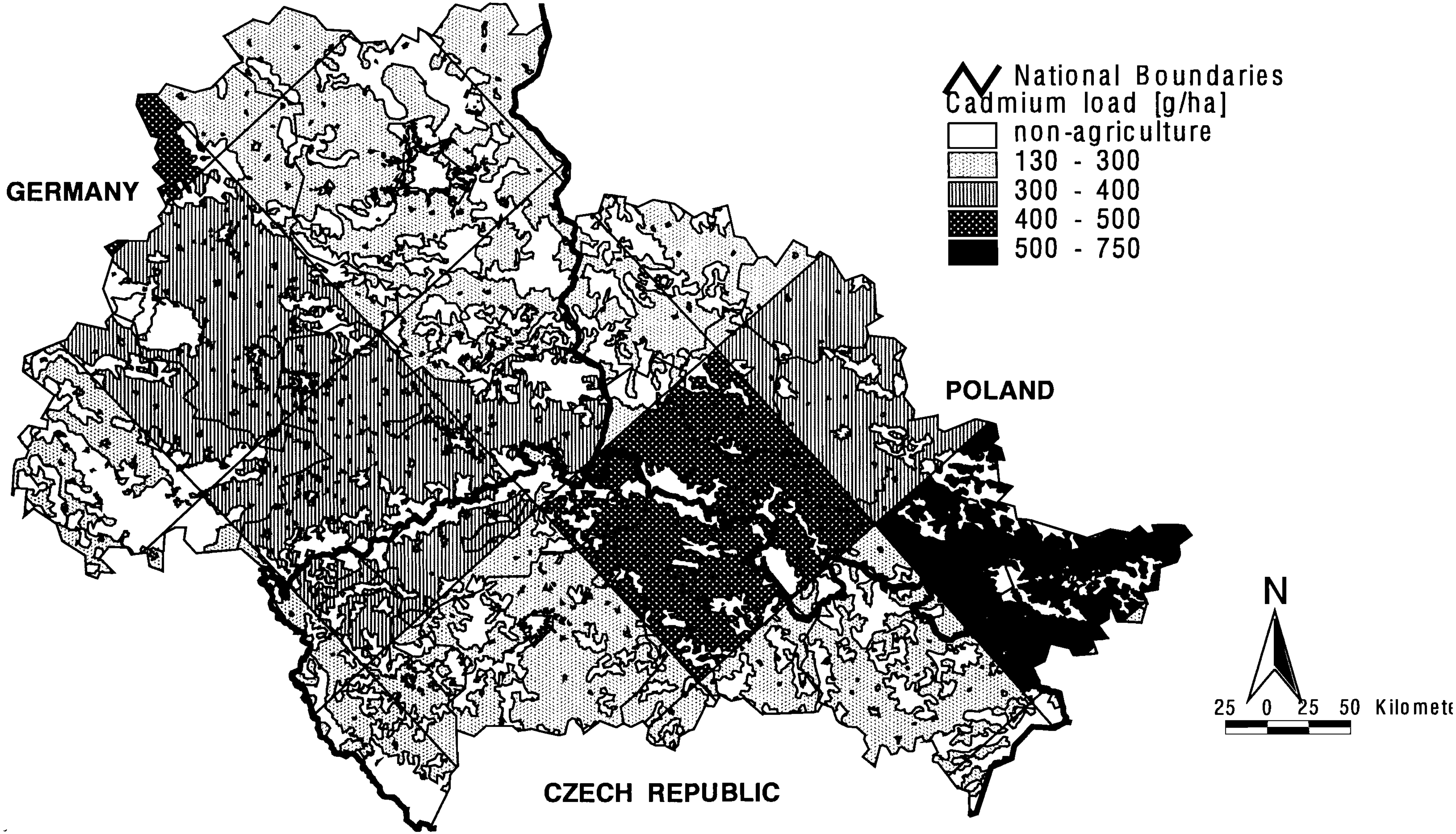
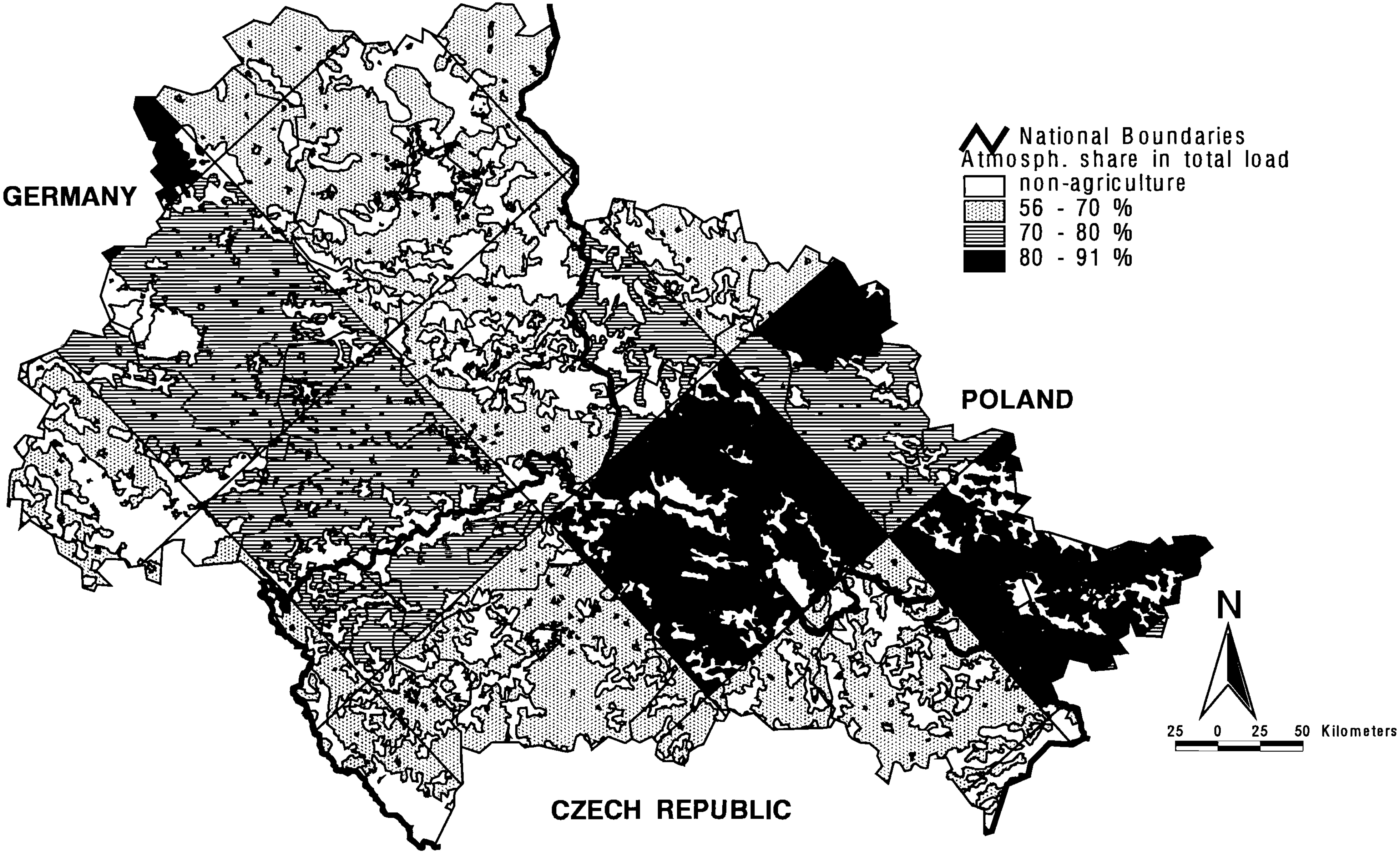
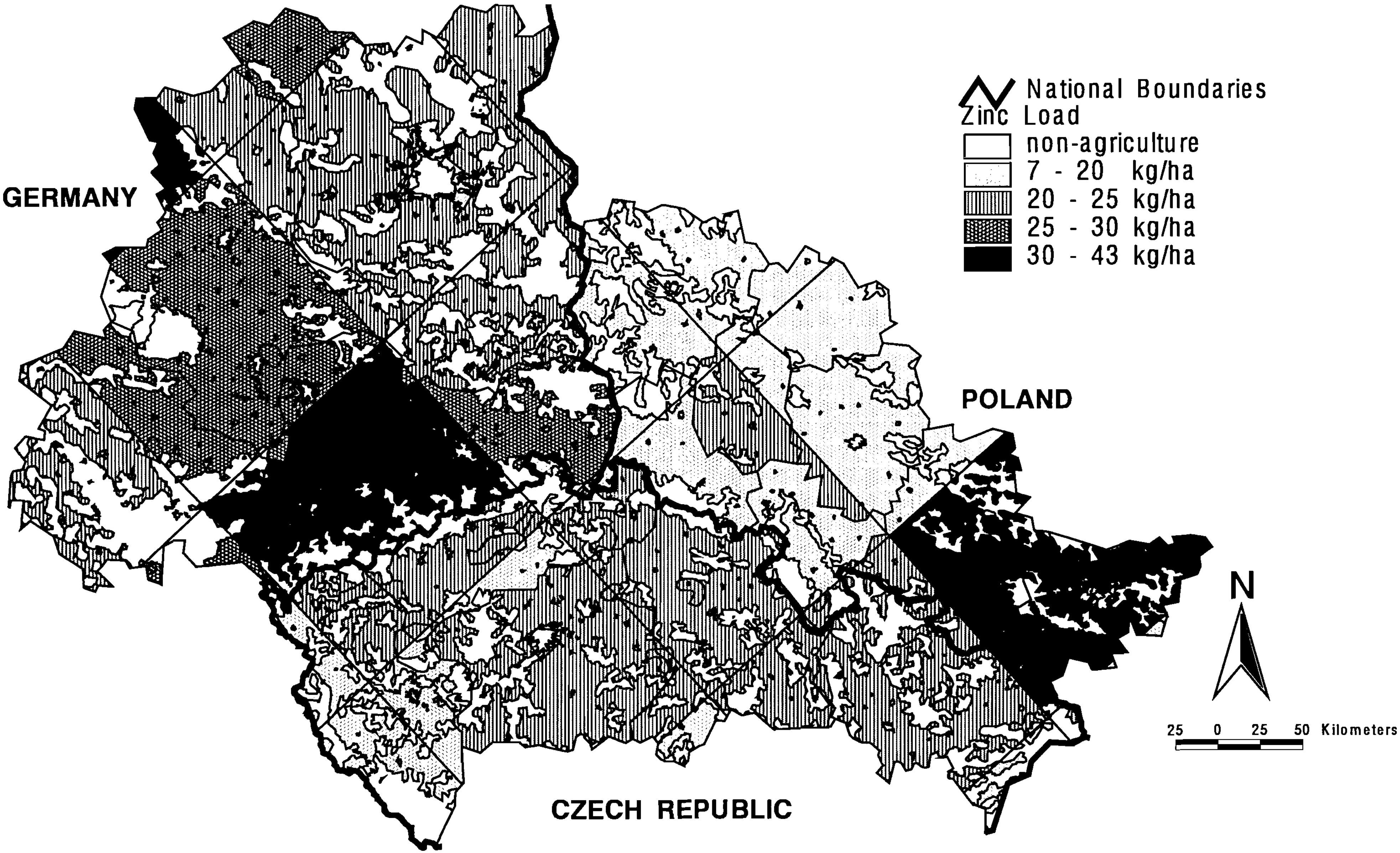
Cumulative total Cadmium load to agricultural land (atmospheric deposition and agricultural load) during the period 1955 - 1994



Cadmium - Atmospheric Share in total cumulative load



Cumulative total Zinc load to agricultural land (atmospheric deposition and agricultural load) during the period 1955 - 1994



Cumulative total Lead load to agricultural land (atmospheric deposition and agricultural load) during the period 1955 - 1994