Working Paper

Assessment of long term impacts of cadmium and lead load to agricultural soils in the upper Elbe and Oder River Basins

Sylvia Prieler and Stefan Anderberg

WP-96-143 November 1996

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Abstract

This report investigates effects of long term load of two heavy metals, cadmium and lead to agricultural soils for a project area in Central Europe. The time frame for the historic analysis is 1955 to 1994. The major source of lead is atmospheric deposition. In the case of cadmium, besides atmospheric deposition, agricultural activities, such as P-fertilizer and manuring, are additional sources of heavy metal input to agricultural soils. Extremely high depositions that were measured in a "hot spot" region in the project area are included in the analysis. A soil model is used to perform a quantitative analysis of potential accumulation or release of cumulative heavy metal loads. A GIS database enables us to undertake a regional analysis. Potential future risks are addressed in a scenario approach covering the time frame 1995 to 2050.

For the majority of the project area there was no significant increase in cadmium and particularly lead soil concentration compared to background values and guidelines. The parts of the project area which had the highest cumulative cadmium deposition historic cadmium accumulation may be of concern. However these assessments related to long term suitability for agricultural food production depend on environmental criteria and the time frames taken into account. Locally, in hot spot areas, atmospheric deposition were and still are much higher and soil guideline values may be exceeded within 10 to 50 years.

The cadmium mass balance for the project area covering the period 1955 to 1994 suggests that from the cumulative load of 3062 t of cadmium (about two thirds from atmospheric deposition and one third from agricultural sources), one third is lost (836 t) and two thirds are accumulated in the soil (2226 t).

Estimates of future atmospheric cadmium and lead deposition are low compared to historic depositions. Cumulative cadmium deposition during 1991 and 2010 is only 10 to 40% of the cumulative deposition during 1970 and 1990. The average lead deposition in 2010 is only 10% of the average deposition in the 70s or 80s.

Due to declining pH-value, triggered by the abandonment of agricultural land and/or a conversion into forest in scenario 1, major releases of cadmium are expected. Even the maximum assumed deposition is not high enough to compensate cadmium loss due to declining pH-value. The extent of decrease depends mainly on the assumed initial concentration in 1994 because cumulative future atmospheric deposition is very low compared to the cadmium already stored in the soils at present.

Scenario 2 assumes intensive agricultural production until 2050. Agricultural activities are now the major source of cadmium load to the soils. In most cases a study state will be reached, the maximum delta increase over the future 55 years is 0.08 mg/kg.

In both scenarios lead soil concentrations are likely to decrease slightly in the future due to losses via erosion, which exceed atmospheric deposition.

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Chapter 1. Introduction

1.1 Background, study area and objectives

Environmental pollution from 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 loads of heavy metals via atmospheric deposition and certain agricultural practices like fertilizing, manuring or sewage sludge application.

Depending on agricultural practices, soil characteristics and certain environmental circumstances, such as 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 groundwater and surface water. Through edible plants or drinking water they enter the food chain. Continuous accumulation of heavy metals in soils may lead to exceedence of guidelines in a long term perspective. Lead has a variety of impacts on the human nervous and circulatory system, while cadmium is considered a probable human carcinogen (WHO, 1987). Guidelines for cadmium and lead soil concentration have been established by various countries and institutions.

The analysis presented in this report is part of a broader IIASA project on "Regional Material Balance Approaches to Long Term Environmental Policy Planning" (or for short IND project for Industrial metabolism¹). 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. This report deals only with two heavy metals, cadmium and lead. Zinc is closely related to cadmium in terms of geographical pattern of deposition and chemical behavior in soils. Therefore many of the conclusions for cadmium may also be true for zinc.

The project area is located in Central Europe (fig. 1) comprising the Upper Elbe and Oder River Basins. It embraces the northwestern part of the Czech Republic (Bohemia and Morawia), southwestern Poland (Upper and Lower Silesia), and the most of the former G.D.R. (Sachsen, Sachsen-Anhalt, Brandenburg, Thüringen). In total it covers an area of 180,000 km². Parts of this area are highly industrialized and densely populated. It includes sites with average heavy metal load in Europe, as well as severely polluted areas with the highest historical excessive cadmium and lead depositions (Olendrzynski, 1996). Half of the area is used for agricultural production, one third is forestry and the remaining 15% are mainly used for urban and infrastructure purposes.

This study attempts to assess environmental impacts and their potential risks related to long term cadmium and lead loads to agricultural soils. The major objective is to identify the key factors that determine heavy metal soil contamination and subsequent uptake by food products. They shall make it possible to draw policy relevant conclusions for land use practices.

¹Industrial Metabolism refers to a concept that embodies a systems approach to minimizing emissions of toxic chemicals by considering all sources of the chemicals, the pathways by which they flow through the industrial economy, and mechanisms by which they are transformed into outputs that must be absorbed and processed by the environment (Stigliani, 1993).

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



1.2 Methodology

Agricultural soils are complex environmental systems. They are influenced by both, natural conditions and anthropogenic activities, most importantly, land management practices, like fertilizing or ploughing, but also unintended effects, like acid deposition. The behavior of heavy metals in soils depends on a complex set of interacting soil characteristics. The most important are pH-value, organic matter content and texture (soil particle size distribution). They are influenced by soil type, land management and to a certain extent by acid deposition. This, combined with regional differences in heavy metal load, demands an incorporation of a *spatial dimension* into the analysis.

Heavy metals are trace elements. Their yearly load to soils is small compared to the soil volume and to heavy metals already present in the soil. Effects of continuous heavy metal loads may only be observed after decades. Therefore the analysis requires a *long term perspective*.

Major uncertainties are involved for this type of assessment. Besides scientific problems, like the development of a heavy metal soil model, uncertainties stem also from missing or incorrect input databases (e.g. heavy metal loads, spatial resolution of soil types). A methodology is required that is able to identify key elements of environmental change with a limited amount of available data. Furthermore the methodology shall make it possible to draw policy relevant conclusions. Data derived from a geographic information system (GIS) enable us to account for regional differences in environmental conditions and heavy metal loads. One possible response to uncertainties related to input data is to analyze the range of possible conditions. By this, we are able to eliminate certain risks and to identify areas of possible concern, which may also require further investigation. Besides estimates for heavy metal loads to the soils in our project area on a regional scale, extremely high depositions only occurring in so called "hot spot" areas are analyzed. A long term perspective is introduced by analyzing effects of the cumulative load of heavy metals over the time period 1950 to 1994. Future potential risks are assessed by analyzing effects of plausible scenarios for future heavy metal loads and land use changes. The combination of the identification of current risks and a scenario analysis shall provide the means to derive policy relevant recommendations for decision makers.

Therefore the core elements of the methodology applied in this study include:

- a historic database for the period 1955 to 1994 of heavy metal atmospheric deposition and heavy metal load via certain agricultural practices,
- measurements of cadmium and lead load in a "hot spot" region within the project area from 1992,
- a heavy metal soil model,
- a GIS database for the project area, and
- a scenario framework for designing future heavy metal loads and environmental conditions.

These elements are described in more detail below. Atmospheric deposition and agricultural sources of heavy metal loads for the period 1955 to 1994 are derived from two databases that were completed within the IIASA IND project. They are described in detail in Olendrzynski et al (1995) and Prieler et al (1996a). Atmospheric deposition was computed using the long-range transport model HMET (Heavy Metal Eulerian Transport) based on a database on historic atmospheric emissions of cadmium and lead. The spatial resolution of the cumulative deposition is a 150 x 150 km grid (Olendrzynski et al, 1995). For 1992, computations for a 50 x 50 km grid are also available (Bartnicki and Olendrzynski, 1996).

The most important agricultural sources of heavy metals are phosphate fertilizer application and manuring. 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 for each of 17 administrative units in the project area. In general lead load stems to more than 90% from atmospheric deposition. Cadmium on the other hand is loaded via both, atmospheric deposition and agricultural sources. The share of each varies with time and region, with a general feature that the higher the total cadmium load, the higher the share of atmospheric deposition compared to agricultural sources (Prieler, 1996a).

The Katowice district in southern Poland is a region with heavy industry and extensive mining. Excessive loads of diverse atmospheric pollutants have been deposited over the past decades. The local sanitary board records measurements of heavy metal deposition in Katowice (SANEPID, 1994). Here, measurements from 1992 are analyzed to study effects of maximum deposition loads. They shall represent conditions in "hot spot" areas.

The GIS database for the project area includes a 1:1,000.000 soil map (Joint Research Centre, 1995) and a land use map on a scale of approximately $1:3,000.000^2$. Precipitation and topography are derived from a grid (approx. $15 \times 15 \text{ km}$). The databases about atmospheric heavy metal deposition and heavy metal loaded to the soils from agricultural activities have been transfered into the GIS. The computations for atmospheric deposition were performed on a grid used by EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe) and has a spatial resolution of about 150x150 km. The agricultural database was established for each of the 17 administrative units in the project area.

Once loaded to agricultural soils and entering the upper soil layers, lead is very immobile and therefore most of the load accumulates in the upper soil horizon. In contrast, the behavior of cadmium strongly depends on soil chemistry, most importantly the pH-value and the organic matter content. A *heavy metal soil model* developed within the IIASA IND project (Schimada et al, 1996a) has been used to determine, which fraction of the loaded metals remains in the soil horizon and which is available in a mobile form that may leach to deeper soil layers or be taken up by the plants. The input variables into the model include the heavy metal load, pH-value, landuse, organic carbon, texture, slope, topography, precipitation and crop information. The model simulates the behavior of the heavy metal soil concentration in the upper 20 cm, cumulative heavy metal loss over the studied time period via erosion, plant removal and leaching, heavy metal concentration in the edible part of the plant to name the most important ones.

Besides studying effects of historic heavy metal loads, there is a need to look closer into what types of future development could be risky and how these can be avoided. For this, a scenario approach is chosen as a tool to stimulate analysis of possible problems and risks in an uncertain future. The ambition is to identify possible effects of future heavy metal loads and future changes in the environmental conditions. The rational for two chosen scenarios is based on former studies completed within the IIASA IND project. The first developed scenarios for land use change (Prieler et al, 1996b). The second estimated future heavy metal emissions until the year 2010 (Pacyna, 1996 and De Bruyn, 1996).

The databases described above provide input variables for the heavy metal soil model. Three different runs were performed. The first for the time frame 1955 to 1994 (chapter 2). The second and third runs are future scenarios that will be described in chapter 3. Scenario 1 foresees the abandonment of agricultural land and/or conversion to forest. The basic feature is a pH-decline between 1995 and 2050. Scenario 2 is based on the assumption that agriculture will be widespread and intensive until 2050. For each run different pH-values and organic matter contents are tested as well as different initial heavy metal soil concentrations in the base years (1955 for historic loads and 1995 for future scenarios).

 $^{^{2}}$ A 1:1,000.000 scale means that the smallest spatial unit is about 10 x 10 km representing a 1x1cm square on the map.

Chapter 2. Historic Cadmium and Lead Load

The first run of the heavy metal soil model investigates the period 1955 to 1994. Effects of five heavy metal loads are studied. They are 10, 5, 22, 100 and 300 g/ha/a for cadmium and 200, 100, 300, 1500 and 7000 g/ha/a for lead. These loads are based on the following rationale:

- 1. An average load (over time and space) for the project area has been 10 g/ha/a for cadmium and 200 g/ha/a for lead.
- 2. The lower range of our estimated load for the project area has been 5 g/ha/a for cadmium and 100 g/ha/a for lead, with the minimum amounting to 2 g/ha/a (cadmium) and 70 g/ha/a (lead).
- 3. The maximum cumulative load in the project area has been 900 g/ha for lead and 13 kg/ha for lead. The yearly equivalent assuming equal yearly loads is 22 g/ha/a (cadmium) and 300 g/ha/a (lead).
- 4. Measurements for the Katowice district in southern Poland show for the year 1992 approximately one third of the area 100 g/ha/a cadmium load and 1500 g/ha/a lead load (SANEPID, 1996).
- 5. In the same measurements the maximum depositions are located in areas where cadmium amounts to 300 and 700 g/ha/a and lead deposition is 4000 and 7000 g/ha/a.

The first three loads represent typical loads on the regional scale, while the latter two loads are examples for "hot spot" areas in the region.

The behavior of each load is investigated at four different pH-values ranging from 4 to 7 and at four different organic matter contents ranging from 0.8 to 6%. They are kept constant over time. Precipitation, topography and texture are assumed to be constant at average values (500 mm precipitation, 300 m topography and medium texture).

Depending on the type of crop grown a soil pH-value between 5 and 7 is required for optimal yields. The pH-value of agricultural soils is artificially managed. Lime application shall counteract acidifying processes caused by nitrogen fertilizer application, crop removal and in certain areas acid deposition. All these processes are strongly influenced by the soil type. In order to determine the required lime application rates, agricultural soils are regularly monitored.

The main characteristics of regions with deposition high heavy metal are concentrated heavy industry and high population densities. These characteristics commonly trigger release of other kind of pollutants including acid deposition. Parts of the project area have therefore not only received the highest heavy metal depositions in Europe, but also very high acid depositions (Krutzfield, 1996). Despite liming efforts, in certain regions the pH of agricultural soils was acid. In the Czech Republic, for

Table 1. Czech Republic: Percentage ofarable land for a pH-values class indifferent time periods

| pH value | < 5.5 | 5.6- | > 6.5 |
|----------|-------|------|-------|
| | | 6.5 | |
| 1961-65 | 28 | 32 | 40 |
| 1966-70 | 32 | 27 | 41 |
| 1971-75 | 34 | 27 | 39 |
| 1976-80 | 32 | 30 | 38 |
| 1981-83 | 30 | 34 | 36 |
| 1984-86 | 25 | 36 | 39 |
| 1987-89 | 21 | 36 | 43 |

example, between the 70s and the mid-80s, 30% of arable land was below a pH of 5.5 (Environmental Yearbook of the Czech Republic, 1993) (Tab. 1). In Poland about half of the agricultural areas have acid or very acid soils (Kern, 1987). There, this may however be mainly due to natural conditions. Krutzfield (1996) estimated the pH-values for the soils in the project area over the period 1955 to 1994 based on regional information on N-fertilizer, manure and lime application and acid deposition. He showed that in parts of the region there is a high probability that lime application was not sufficient to avoid pH-declines of about 0.5 to 1 unit over the 40 year period.

Though difficult to quantify, there is some evidence that during the past 40 years parts of the agricultural land in the project area has had acid soil pH-values, which were under the optimal range for agricultural production.

2.1 Cadmium

Tables 2 and 3 present selected results from the first model run. These results show the importance of the pH-value and organic matter content. In general the higher the pH-value and organic matter content, the more of the loaded cadmium is accumulated in the soil. The maximum load estimated for our project area causes an increase of up to 0.3 mg/kg over the 40 year period. The lower range of load results in increases of less than 0.1 mg/kg even at high pH-values and high organic matter contents. An approximate average for cadmium accumulation in the upper 20 cm soil layer, based on an average cadmium load and an average agricultural pH-value of 6 to 7, is about 0.1 mg/kg over the studied 40 years period.

Table 2. Increase in cadmium soil concentration over the period 1955 to 1994 for different cadmium loads, pH-values and organic matter contents

| load [g/ha/a] | 101 | | | 52 | | | | 22 ³ | | | | |
|----------------------------|-------|------|------|------|-------|------|------|-----------------|------|-----|-----|-----|
| pH-value | 4 | 4 | 7 | 7 | 4 | 4 | 7 | 7 | 4 | 4 | 7 | 7 |
| organic matter content [%] | 1 | 5 | 1 | 5 | 1 | 5 | 1 | 5 | 1 | 5 | 1 | 5 |
| delta increase* [mg/kg] | -0.01 | 0.08 | 0.07 | 0.14 | -0.03 | 0.02 | 0.06 | 0.07 | 0.02 | 0.2 | 0.3 | 0.3 |

*1955 to 1994;

1 = average load project area, 2 = low load project area, 3 = max. load project area,

Background soil concentrations of cadmium in unpolluted areas are less than 0.5 mg/kg (Schachtschabel et al, 1992)³. Guidelines for cadmium soil concentration differ from country to country and they are commonly related to the soil type. Dutch reference values for good quality soil depend on the organic matter and clay content of the soil. Depending on soil type they range approximately between 0.5 to 1.2 mg/kg cadmium soil concentration. The target value for a standard soil (10% organic matter and 25% clay), for example, is 0.8 mg/kg (VROM, 1991, Vegter, 1995). In addition, The Netherlands have, so called "signal values", for agricultural soils, which are between 0.5 and 3 mg/kg depending on soil characteristics. At concentrations higher than this value, the quality standards for food might be exceeded (Vegter, 1995).

³For Central & Eastern Europe cadmium concentration in unpolluted agricultural soils are in the order of 0.5 mg/kg (Kabata-Pendias et al, 1993). In Poland the average background value is 0.35 mg/kg (Smal et al, 1995). Alloway (1990) estimated the cadmium content in agricultural soils at 0.2 - 1 mg/kg for the countries of the European Union.

A general problem of long term soil studies is that they can not be verified against measurements unless archived soil samples are available. This is in particular true for cadratium since the delta increase over the 40 year period is low compared to the range of soil background concentrations in unpolluted areas (< 0.1 to 1 mg/kg).

The agricultural experimental station at Rothamsted (England) has a unique dataset of archived soil samples dating back to 1840. A comparison of today's cadmium soil concentration with the concentration in archived samples shows that the concentration of cadmium in the topsoil has increased by about 50% since 1840, even where no fertilizers have been applied (IACR, 1993). Thus atmospheric deposition must have been the only source of cadmium. On these untreated plots the increase in cadmium soil concentration ranges between 0.1 and 0.25 mg/kg for 100 or 130 year periods. Their estimated yearly average increase is 1.2 μ g/kg (Johnston and Jones, 1992). For 40 years this would be comparable to 0.05 mg/kg. This is comparable with our estimated average increase of 0.1 mg/kg for the 40 year period, considering that we've also accounted for cadmium load from fertilizers and manure and that our project area includes sites with the highest cumulative atmospheric depositions in Europe.

Hot spot areas

Cadmium loads, measured in 1992 in "hot spot" areas result in significant increases in cadmium soil concentration (tab. 3). Calculations for cadmium loads amounting to 100 g/ha/a show that depending on environmental characteristics of the soil only 5 to 10 years are needed for an increase of 0.2 mg/kg (tab. 4). For the significant increase of 0.5 mg/kg only 26 years are needed to accumulate 0.5 mg/kg assuming 50% of the load accumulating.

| Table 3 | . Increase | in | cadmium | soil | concentration | over | the | period | 1955 | to | 1994 | for |
|---------|-------------|------|--------------|-------|---------------|------|-----|--------|------|----|------|-----|
| cadmiur | n loads typ | oica | l for "hot s | pot'' | areas | | | | | | | |

| load [g/ha/a] ¹ | | 10 | 0 | | 300 | | | |
|---|-----|-----|-----|-----|-----|-----|-----|-----|
| pH-value | 4 | 4 | 7 | 7 | 4 | 4 | 7 | 7 |
| organic matter content [%] | 0.9 | 5 | 0.9 | 5 | 0.9 | 5 | 0.9 | 5 |
| delta increase over 40y. period [mg/kg] | 0.4 | 1.1 | 1.4 | 1.4 | 1.3 | 3.5 | 4.3 | 4.4 |

1... Measurements for the Katowice district in southern Poland show for the year 1992 approximately for one third of the area a cadmium load of 100 g/ha/a (SANEPID, 1996). There are two local areas with the maximum cadmium depositions of 300 and 700 g/ha/a.

Depending on cadmium already stored in the soil and environmental conditions, a forty year increase of 0.1 or 0.2 mg/kg may be considered of concern for the long term potential of a soil to grow food products. Table 4 reproduces the number of years it takes to accumulate either 0.2 or 0.5 mg/kg cadmium in the soil for different cadmium loads and different environmental characteristics. In soils with high pH-values and high organic matter contents the average historic cadmium load accumulates an additional 0.2 mg/kg after 52 years. Accordingly the accumulation rate for conditions where only 50% of the load remains in the soil is about 100 years. This is approximately the case when there is a pH-value of 6 or a pH-value of 5 combined with high organic matter content (>3%).

From table 4 it may be concluded that a time frame of 50 to 100 years must be considered to identify critical increases in cadmium soil concentration. It is obvious that this time frame strongly depends on the definition of a critical cadmium increase and that there are major regional differences for accumulation rates.

Table 4. Number of years until 0.2 or 0.5 mg/kg Cadmium are accumulated in the upper 20 cm of the soil layer for two different environmental conditions*

| load [g/ha/a] | 10 | 5 | 22 | 100 | 300 |
|---|-----|-----|-----|-----|-----|
| | | | | | |
| 100% of load accumulates in soil ¹ | | | | | |
| number of years until increase of 0.2 mg/kg | 52 | 104 | 23 | 5 | 2 |
| number of years until increase of 0.5 mg/kg | 130 | 260 | 59 | 13 | 4 |
| 50% of load accumulates in soil ² | | | | | |
| number of years until increase of 0.2 mg/kg | 104 | 208 | 46 | 10 | 4 |
| number of years until increase of 0.5 mg/kg | 260 | 520 | 118 | 26 | 8 |

*... The conversion is done for the upper 20 cm soil layer and a bulk density of 1.3

1... approximately at high pH-values of 7 or pH-values of 6 combined with organic matter content greater than 3% more than 90% of the load remain in the soil

2... approximately at pH-values of 6 and pH-values of 5 combined with organic matter content greater than 3% half of the cadmium load remains in the soil and half is leaching out

Low cadmium soil accumulation rates mean higher amounts of cadmium available in a mobile form. It is difficult to assess risks related to leaching cadmium or cadmium uptake by plants. The soil model used in this study calculated no critical cadmium concentrations in the edible portion of the plants except for extreme "hot spot" areas. The model also estimates that the majority of the losses of mobile cadmium occur via leaching. If the risk of groundwater pollution is of high concern, concepts for risks related to groundwater pollution are required. A combination of high groundwater table and low binding capacity of the soil for cadmium (at low pH-values and low organic matter contents) is an indication of potential groundwater pollution. However, it will be difficult to prove these risks, since a translation of leaching into cadmium groundwater concentration is very difficult to estimate. An example for a leaching rate for the average cadmium load is 20 mg/m² assuming that 50% of the load is leaching.

Regional analysis

The GIS database enabled to undertake a regional explicit analysis of cadmium accumulation or release from the agricultural soils. For the project area the estimated heavy metal loads for the period 1955 to 1994 was overlaid with a soil map. To each soil type a pH-value and organic matter content typical for agricultural soils was assigned (Appendix 1). The pH-values range between 5 and 7. The organic matter contents range between 1% and 6%, except for Histosols, which have by definition a high organic matter content of 30% or more. As a result of the overlay procedure each polygon has information on the heavy metal load, precipitation, topography, pH-value, organic matter content and land use. For the whole project area medium texture and level slope was assumed. The initial soil concentration in the upper 20 cm soil layer is 0.05 mg/kg. Based on this, the heavy metal soil model calculated cadmium accumulation and release over the period 1955 to 1994 for each polygon.

Figures 2, 3 and 4 show regional differences for selected variables of the fate and transport of loaded cadmium to agricultural soils in the project area during the period 1955 to 1994. The

majority of the area has delta increases in cadmium soil concentration over the 40 year period of less than 0.1 mg/kg (fig. 2). The pattern reflects the particular importance of atmospheric deposition. In the southeastern grid (Katowice and Opole voivodship) cumulative atmospheric deposition was highest within the project area. There the delta increases in cadmium soil concentration are above 0.2 mg/kg with the maximum of 0.31 mg/kg. The remaining regional differences are due to a combination of soil type and total cumulative loads. Fertile soils with high pH-values in the German part of the project area (Sachsen-Anhalt and Sachsen) are the main reason for the higher cadmium accumulation in this part of the project area falling into the class of 0.1 to 0.2 mg/kg delta increases.

Figure 2. Delta increase in cadmium soil concentration of the upper 20 cm soil layer over the period 1955 to 1994



If a danger of groundwater cadmium contamination is of concern, like in areas with high groundwater tables, the rate of cadmium leaching is of interest (fig. 3). In about half of the area cadmium leaching is less than 5 mg/m^2 , for the other half it is 5 to 10 mg/m^2 . In approximately 10% of the agricultural land leaching is above 10 mg/m^2 with a maximum of 28 mg/m^2 . The map shows that these areas are situated in the mountainous parts of the region, where precipitation is high and in the region with the highest cadmium load. The uneven distribution of the pattern in this map reflects the importance of soil types on the one hand the importance of interactions of all the variables in the model on the other hand.

Figure 4 also shows the total cumulative loss of cadmium from the upper 20 cm soil layer. This includes losses via leaching, erosion and plant uptake with subsequent harvest removal. The pattern again is highly disperse and would require detailed analysis of each polygon to identify the reasons for high or low losses. The majority of the area has losses of less than 10 mg/m^2 , for about 1/3 the losses amount to more than 10 mg/m² with a max. of 34 mg/m².

Fig. 3. Cumulative leaching of cadmium from the upper 20 cm soil layer over the period 1955 to 1994



Fig. 4. Cumulative total loss (=leaching+erosion removal + plant uptake and subsequent harvest removal) of cadmium from the upper 20 cm soil layer over the period 1955-1994



Table 5 shows for some of the model parameters the range and weighted average of values. The average delta increase in cadmium soil concentration in the upper 20 cm soil layer of agricultural soils is 0.08 mg/kg. The maximum amounts to 0.3 mg/kg. The importance of these 40 years increases have been discussed above. The model calculates a small range of cadmium concentrations in the edible portion of the plants. The average here is 0.1 mg/kg. These values are below guideline values.

| | | weighted average | min. | max. |
|--|-------------------|------------------|------|------|
| plant conc. in 1994 | mg/kg d.w. | 0.11 | 0.05 | 0.16 |
| delta increase in soil conc. 1955-1994 | mg/kg d.w. | 0.08 | 0.01 | 0.31 |
| | | | | |
| total loss from upper 20 cm soil layer | . 2 | 8.4 | 0.01 | 14.6 |
| for 1955 to 1994 | mg/m ² | | | |
| loss via erosion | mg/m ² | 0.7 | 0.03 | 2.9 |
| loss via leaching | mg/m ² | 4.2 | 0.07 | 27 |
| plant uptake and subsequent removal | mg/m ² | 3.5 | 1.4 | 5.8 |
| | | | | |
| cumulative load 1955 to 1994 | mg/m ² | 31 | 17 | 87 |

Mass balance

The regional analysis also enables to calculate a mass balance of cadmium accumulation and release from the upper 20 cm soil layer over the period 1955 to 1994 (table 6). Due to the resolution of the GIS database, the area of agricultural land is overestimated. In the GIS land use database agricultural area amounts to 13 mio ha while statistical sources for 1992 show an area of 10 mio. ha utilized for agricultural production. Therefore the mass balance derived from the GIS database has been reduced by 20% (or a factor of 0.8).

From the cumulative load of 3062 t of cadmium entering the soil, one third is lost (836 t) and two thirds accumulated in the soil (2226 t). About two thirds of the load stems from atmospheric deposition and one third from agricultural sources. Regional and temporal differences in this share are discussed in detail in Prieler et al (1996a). The majority of the loss occurs via leaching and plant uptake, with leaching being slightly more important than plant uptake and subsequent removal. Despite very low assumptions for erosion rates in the model, generally amounting to less than 3 t/ha/a, nearly 10% of the total loss occurs via erosion.

The amount of cadmium in the soil in 1994 is a calculated value. It is strongly dependent on the assumed initial soil concentration in 1955, which can only be roughly estimated. For this study the assumed background concentration of 0.05 mg/kg is based on results derived from a soil sampling program in the study area (Shimada and Jaffe, 1996a). On twelve sites over 50 soil samples were taken. The median soil concentration of cadmium in a depth of 40 cm was 0.05 mg/kg. This value is expected to be at the lower end of possible background concentrations in 1955. As stated above literature sources give background concentrations of less than 0.5 mg/kg (Schachtschabel et al, 1992) or 0.2 to 1 mg/kg for agricultural soils (Alloway, 1990). In addition some of the regions in the project area have had mining tradition or were industrialized long before 1955.

Table 6. Mass balance for cadmium in the upper 20 cm soil layer over the period 1955to 1994 for the agricultural land in the project area (10 mio ha)

| | tons | percentage | |
|--|------|------------|-----|
| Cadmium in the soil in 1955* | 1346 | | |
| cumulative Cd load over period 1955 to 1994 | | | |
| atmospheric deposition | 2198 | 72 | |
| agricultural sources | 864 | 28 | |
| total load | 3062 | 100 | 100 |
| cumulative Cd loss over period 1955 to 1994 | | | |
| loss via leaching | 421 | 50 | |
| loss via plant uptake and subsequent removal | 348 | 42 | |
| loss via erosion | 68 | 8 | |
| total loss | 836 | 100 | 27 |
| delta increase (load - loss) | 2226 | | 73 |
| Cadmium in the soil in 1994 | 3572 | - | |

* based on the assumption of an equal soil concentration in the whole project area of 0.05 mg/kg (= 13.5 mg/m^2 for bulk density of 1.3 and upper 20 cm soil layer)

2.2 Lead

Lead is very insensitive to pH and soil organic matter content. At pH-values over 4 and even low organic matter contents, more than 90% of the lead remains in the soil. Therefore the delta increase in soil concentration is given only for different loads (table 7). The estimated average load for the project area is about 200 g/ha/a for the past 40 years. This translates into an increase of 3 mg/kg lead soil concentration. The maximum estimated load is 13 kg/ha for the period 1955 to 1994, which is equivalent to 4 mg/kg lead in the plough layer of the soil.

Lead background soil concentrations range between 2 and 60 mg/kg (Schachtschabel et al, 1992). Background values in 'unpolluted' soils in Poland are between 10 and 25 mg/kg, for Central & Eastern Europe 35 mg/kg (Smal, 1995). Guideline values for lead soil concentration are for example in Poland 50 mg/kg for light texture 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.

A 40 year increase of 4 mg/kg in lead soil concentration is therefore marginal compared to the range of background values in 'unpolluted' sites and existing guidelines. Only much higher lead depositions, which are found locally in hot spot areas cause increases in lead soil concentration of concern (table 7). Assuming a yearly lead deposition of 1500 g/ha, as it was measured in 1992 in Katowice, it would take only 17 years until additional 10 mg/kg accumulate in the soil.

In addition to low increases on a regional scale, losses of lead via erosion may be of the same order as the lead load (except for the hot spot areas). In general erosion losses of less than

10 t/ha/a are considered to be tolerable from an erosion protective point of view (Schwertmann et al., 1987).

Table 7. Increase in lead soil concentration over the period 1955 to 1994 for different lead loads

| lead load [g/ha/a] | 2001 | 1002 | 15003 | 40004 |
|---|------|------|-------|-------|
| delta increase for a 40 year period.* [mg/kg] | 3 | 1 | 22 | 60 |

* delta increase in soil conc. in upper 20 cm soil layer, assuming a bulk density of 1.3

1...average load over time and area for project area, 2...lower range of load in project area, 3 and 4....examples for loads in "hot spot" areas, 1992 measurement in Katowice show these loads for parts of the district.

Even at low erosion rates, the lead loss via erosion may equalize the lead loaded dependent on the assumed erosion and lead soil concentration. For example, a yearly erosion of 5 t/ha/a and an assumed lead soil concentration of this eroded soil of 20 mg/kg means a yearly lead loss via erosion of 100 g/ha/a. This is half of the estimated average lead load on the soils (200 g/ha/a). Furthermore, at high erosion rates, surface runoff is likely to be higher as well, increasing the amount of metals removed from agricultural soils.

Chapter 3. Scenarios for Future Cadmium and Lead Load

3.1 Description of Scenarios

For environmental assessments which focus on large-scale, long-term interactions between development and environment, scenarios are indispensable tools. The aim is to identify possible future problems within a framework of plausible development paths. As indicated earlier, risks of heavy metals in agricultural soils, depend on a combination of heavy metal load and certain soil characteristics. The scenarios applied for this study need to incorporate atmospheric deposition scenarios and scenarios for future land use and land management. The latter to determine both, heavy metal load from agricultural practices and soil characteristics influenced by land use and land management.

Future atmospheric heavy metal emissions and deposition

Future atmospheric heavy metal emissions for Europe, until the year 2010, have been estimated by Pacyna (1996). De Bruyn (1996) designed emission scenarios especially for the project area (De Bruyn, 1996). In general the scenarios foresee a decrease in emissions over the next decades due to a restructuring of the economy as well as investments into pollution control technologies and general improvement of process technologies. The envisaged pollution control goals of the countries in the project area are ambitious. Full enforcement may proove to be more difficult than expected. Uncertainties are therefore mainly related as to when the expected decrease comes into effect.

Computations within the atmospheric modeling part of the IIASA IND Project translated future emissions of De Bruyn (1996) into future atmospheric heavy metal deposition in the project area. Pacyna's (1996) emission estimates for Europe

| Table 8. Range of atmospheric cadmium and lead |
|--|
| leposition in the project area in different time |
| periods [in g/ha/a] |

| | 1955 (| to 1994 per | Year 2010 | | |
|---------|----------|-------------|-----------|------|---------|
| | max. | average | min. | max. | average |
| Cadmium | 30* | 5** | 1 | 3.7 | 0.7 |
| Lead | 400* 160 | | 42 | 29 | 14 |

*1975 in Katowice, **average over area and time

were used to account for inflow of heavy metals from the neighbouring countries of the project area. From 2010 to 2050 the deposition is assumed to remain constant at the level of 2010. In general future atmospheric depositions are significantly lower than historic depositions (Table 8). Between 1995 and 2010 there is a constant decrease of deposition over the whole project area. In 2010 the maximum cadmium deposition is only slightly higher than the minimum deposition over the period 1955 to 1994. The average cadmium deposition in the project area in 2010 is less than one third of the average deposition during the past 40 years. For lead the declining trend is even more extreme.

Figure 5 compares the cumulative atmospheric cadmium and lead deposition in the project area for two time periods, 1970 to 1990 and 1990 to 2010. The 70s and 80s (and also the 60s) were characterized by extensive industrial acitivity and little concern about environmental considerations. The downward trend in atmospheric deposition started in the mid or late eighties and accelerated in the beginning of the 90s due to the economic and political changes in Poland, the Czech Republic and former G.D.R.

The expected cumulative cadmium deposition during 1990 to 2010 is only a fourth to a third of the deposition during 1970 to 1990. The respective decrease for lead is even stronger amounting to more than 80% over the whole area. In general the future shows a more evenly distribution of deposition. Some of the grids with very high deposition in the past do not have important deposition in the future, such as the grids in eastern Germany. The highest historic cadmium deposition was in southwestern Poland (Katowice voivodship). This grid still shows the maximum values for both, cadmium and lead, but only at a fourth or a fifth of the deposition in the past.

Fig. 5 Cumulative atmospheric cadmium and lead deposition -Comparison between the periods 1970 - 1990 and 1991 - 2010

CADMIUM



1970 - 1990

1991 - 2010

LEAD



Future land use

A framework for land use change in the project area was developed by Prieler et al (1996b). Three scenarios describe possible developments of agriculture and land use in the European context. The time frame is present to 2050.

The first Scenario, "Large Scale Increase of Wooded Area" anticipates the introduction of a free market economy to the agricultural sector. No subsidies are required any more for agriculture. Large areas of marginal farmland have to be taken out of production. The price of land decreases. Farmers will either afforest their farmland or try to sell it for non-agricultural uses like urban development, recreation or nature conservation.

Scenario 2, "Alternative Agricultural Products", assumes a shift from food production to nonfood products, mainly biofuel and incentives for an extensification of agricultural production. Subsidies, that are still required for the agricultural sector, will be kept. The overall policy aims at keeping the land open, avoiding uncontrolled spreading of urban development and providing prospect for development and employment in rural areas.

The main characteristic of Scenario 3, "Europe as Food Exporter", is an increase in the demand for agricultural products, which by approximately 2010 triggers an increase in the world market price for food products. Reasons for this are population and wealth increase, especially in China an southeast Asia, combined with environmental constraints like water or fertile land scarcity and erosion. As a result agricultural production in Europe becomes prosperous in the frame of a free market environment. No further subsidies are required for the sector. After 2010 all available farmland is used for food production.

With respect to cadmium and lead problems in agricultural soils, two issues are of particular importance for the discussion of the long-term effects of cumulated toxic pollution. The first is a continued load of cadmium and lead over the next decades. The second is the potential release of already accumulated heavy metals in soils triggered by pH-changes.

For this study two scenarios are considered to be sufficient to identify possible future problems and risks related to heavy metals in soils. The chosen time frame is present until 2050. The first scenario described above means an abandonment or conversions of agricultural land to forestry. A pH-decline will follow these land use changes. Scenario 1 of this study simulates the abandonment of agricultural land and/or conversion to forest. The basic feature is a pH-decline between 1995 and 2050. Atmospheric deposition is the only cadmium and lead load to the soils.

The land use change scenarios 2 and 3 foresee a continuation of agricultural production in different economic environments. The major difference is the intensity in agricultural production and the types of products grown. In order to evaluate future risks related to cadmium and lead in soils it is sufficient to discuss one scenario, which simulates intensive agricultural production until the year 2050. This is the basic rational for Scenario 2 of this study. Cadmium and lead loads stem from atmospheric deposition and agricultural sources. Different pH-values and organic matter contents are tested as well as different initial heavy metal soil concentrations in 1995.

3.2 Scenario 1 - Abandonment of agricultural production and/or convertion to forest (the pH-decline scenario)

After abandoning farmland, the extent of pH-decline depends most importantly on soil type, former management practices and in certain areas on acid deposition. The growing of forest on former agricultural soils tends to acidify soils additionally. Coniferous forests are commonly more acidifying than deciduous forests.

The area with the highest atmospheric acid deposition in the project area is at the border between eastern Germany and the Czech Republic. Figure 6 shows the future cumulative acid deposition (NOx+SOx+NHx) derived from the RAINS 7.0 model⁴. The future distribution of acid deposition shows a similar pattern than the historic one (see Krutzfield, 1996 for historic acid deposition). A comparison of the period 1970 to 1990 with the period 1990 to 2010 shows a decrease in cumulative acid deposition between





10 and 40% depending on the grid. The highest reductions occured in the grids with the highest acid deposition during the last 20 years. Therefore the estimates foresee a more even distribution of acid deposition for the future.

At Rothamsted, the same long-term experimental station that has already been mentioned above (p.7), the pH-change of an arable field on which in the course of 100 years deciduous woodland became established was measured. As an arable field the site must have received heavy dressings of lime in the 18th and 19th century which raised soil pH to over 7. One hundred years later, by 1983 the pH of the upper 20 cm soil layer has fallen by 2.9 units down to a pH of 4.2 (Johnston, 1986).

The basic feature of Scenario 1 is a pH-value decline between 1995 and 2050 and atmospheric deposition as the only source of cadmium load. Three different rates of pH-decline are tested, one minor decline from 6 to 5 and two strong declines, one from 7 to 5, the other from 6 to 4.

Effects of two different heavy metal loads (high load and average load in the project area, see table 8), three different cases of pH-decline and four different organic matter soil contents ranging from 1 to 8% are tested with the heavy metal soil model. Texture, precipitation, topography and slope are assumed to be constant at average values over the examined time period (texture is medium, precipitation is 600 mm/a, topography is 300 m, slope is level). In addition there are two different initial cadmium and lead soil concentrations in 1995. The first

⁴ The RAINS model has been developed at IIASA and calculates acid atmospheric deposition for Europe (Alcamo et al, 1990). It is under constant development and improvement. The latest version 7.0 will soon be released.

assumes a rather high, but not unlikely heavy metal soil concentration in 1995 amounting to 0.6 mg/kg for cadmium and 50 mg/kg for lead. The second assumes lower initial heavy metal soil concentrations, based on median values of soil sampling measurements completed within the IIASA IND project (Shimada and Jaffe, 1996a). These are 0.2 mg/kg for cadmium and 20 mg/kg for lead. Though difficult to prove, the latter concentrations may represent average cadmium and lead concentrations in the majority of the project area.

In all tested cases this scenario shows a decrease in heavy metal soil concentration by 2050. The major reason for this is the low cadmium and lead load to the soils. Even the maximum assumed deposition is not high enough to compensate cadmium loss due to declining pH. Because of this low cumulative load, the concentration in 2050 depends fully on the assumed 1995 soil concentration and the environmental conditions. The marginal importance of the heavy metal load becomes obvious by comparing the amount of heavy metals in soil in 1995 with the estimated cumulative load over the period 1995 to 2050 (table 9). Our initial cadmium soil concentrations in the upper 20 cm of the soil layer are 150 mg/m² for the high assumption and 60 mg/m² for the lower assumption. The higher cumulative load until 2050 is estimated to be only 13 mg/m². For lead this relation is even more extreme.

Table 9. Comparison of heavy metal soil concentration in 1995 with the estimated cumulative atmospheric deposition 1995 to 2050

| | | Cadmium [mg/m ²] | Lead [mg/m ²] |
|------------------------------|--------------|------------------------------|---------------------------|
| 1995 soil concentration | assumpt. 1 | 150 (=0.6 mg/kg) | 13625 (=52 mg/kg) |
| | assumpt. 2* | 60 (= 0.2 mg/kg) | 5300 (=20 mg/kg) |
| cum. atmosph. dep. 1995-2050 | high load | 13 | 224 |
| | average load | 5 | 122 |

* The values in assumption 2 are derived from a sampling program of soils in the project area (Shimada and Jaffe, 1996b)

Table 10 presents selected results for cadmium soil concentrations in 2050 based on average cadmium loads. It demonstrates the vital importance of the organic matter soil content and the extent of pH-decline for the cadmium lost. For a pH-decline from 6 to 4 combined with low organic matter content, the soil concentration in 2050 will be about the same, no matter whether the initial 1995 concentration was 0.6 or 0.2 mg/kg. The difference is only in the amount of cadmium lost from the soil over this period. The loss is 0.5 mg/kg (or 1300 g/ha assuming bulk density of 1.3 and 20 cm soil layer) versus 0.2 mg/kg (or 520 g/ha). On the other end, even a strong pH-decline but at higher pH-values, i.e. from 7 to 5 shows no change in soil concentration for an initial concentration of 0.23 mg/kg.

Table 10. Scenario 1 - Cadmium soil concentrations in 2050 (in mg/kg) for different pHdeclines, organic matter contents and initial 1995 soil concentration (based on an average cumulative deposition of 50 g/ha over the 55 year period)

| pH-decline | strong (6 to 4) | | strong | (7 to 5) | small (6 to 5) | | |
|---------------------------|-----------------|------|--------|----------|----------------|------|--|
| organic matter [%] | 1 | 5 | 1 | 5 | 1 | 5 | |
| 0.6 mg/kg in 1995 | 0.10 | 0.39 | 0.27 | 0.47 | 0.19 | 0.44 | |
| loss during 1995 to 2050 | 0.5 | 0.21 | 0.33 | 0.13 | 0.41 | 0.16 | |
| 0.23 mg/kg in 1995 | 0.04 | 0.15 | 0.11 | 0.22 | 0.08 | 0.18 | |
| loss during 1995 to 2050 | 0.19 | 0.08 | 0.12 | 0.01 | 0.15 | 0.05 | |

Abandoning agricultural land on soils which have a low natural binding capacity for cadmium are likely to show more dramatic effects of cadmium release than soils with a high cadmium binding capacity. This is mainly an effect of pH-value and organic matter, which are to a certain extent kept at artificially high values on farmland. In general a low pH-value, a low organic matter content and a coarse texture of a soil entails a low cadmium binding capacity. Blume and Bruemmer (1991) developed a methodology that allows to assess the relative binding capacity for a heavy metal depending on pH-value, organic matter content and texture. For each heavy metal they assign a "relative binding strenght" derived from the sum of the binding strength for a particular pH-value, organic matter content and texture. This methodology was used and applied to the available soil map. In the project area there are 125 different "units". Each gives the percentage distribution of the four most important soil types. For the major soil type per unit (covering at least 70% of the unit) or for the two most important soil types per unit (covering together at least 70%) the relative binding capacity for cadmium was determined based on a know how basis of ph-value, organic matter content and texture of this soil type. The soil units also give information on texture, which has been used in some cases to determine the relative binding capacity of the soil.

Figure 7 shows the relative binding capacity of soils for camdmium. These maps enable us to identify areas which are particularly in danger to release cadmium in case of the ceasing of agricultural production (i.e. soils with a low capacity to bind cadmium). It shall be noted that most of the soils with a low binding capacity are also poor soils in terms of yield and types of crops grown. Therefore depending on some other factors like slope, accessibility or water regime these soils are those that are most likely to be abandoned in case of a general decrease of farmland. This type of analysis would be especially interesting in "hot spot" regions, where more heavy metals have been accumulated in the soils. Then higher resolution soil maps and, if possible, also maps showing the depth of the groundwater table are required.



Fig. 7 Relative capacity of soils to bind cadmium

Lead

For lead the model is insensitive towards environmental conditions. There is a small loss over the 50 years period. The soil concentration in 2050 depends on the assumed initial concentration in 1995. The model shows a loss of 4 mg/kg from 52 mg/kg in 1995 down to 48 mg/kg in 2050 at all tested environmental conditions. An initial concentration of 21 mg/kg in 1995 gives a concentration of 19 mg/kg in 2050. Most of the lead is lost via erosion. Even though the amount of eroded soil is very low, because of the high 1995 lead soil concentration, lead lost via erosion is higher than lead loaded to the soil. This results in an overall decrease in lead soil concentration.

Due to the linear pH-decline over time, the loss of cadmium and lead from soils is regularly distributed over time. This may not reflect real world conditions, when ph-changes may be more volatile. In general, when the pH drops below 5 the release of cadmium increases substantially.

Hot spot areas

For both cadmium and lead the effects described here would be accelerated in "hot spot" areas. Locally in urban areas, close to emittents or in densely populated areas in general (like Katowice voivodship in our project area) heavy metals already accumulated in the soils may be much higher. Measurements in the late 80s on arable soils in Katowice show for cadmium a soil concentration of 0.7 - 143 mg/kg d.w. with a geometric mean of 3.2 mg/kg and respectively for lead 4 - 8200 mg/kg with a geometric mean of 102 mg/kg (n=2270) (Dudka and Sajda, 1992). Sampling program of private allotment gardens, which are mainly located close to cities, show even higher heavy metal soils concentrations. Gzyl (1990) sampled 126 allotments in Upper Silesia and found for cadmium soil concentration a range of 1.2 to 51 mg/kg d.w. (mean 8.3 mg/kg) and respectively for lead 17 - 1640 (mean 221 mg/kg).

It is obvious that a mobilization of these high soil concentrations is much more dramatic than the one described above for the regional scale. In particular for allotment gardens it has already been shown that vegetables grown on these soils exceed guideline values (Marchwinska, 1984; Kucharski, 1989). Marchwinska et al. (1989) concluded from the heavy metal soil surveys that in Katowice voivodship 17% of the studied allotments and cultivated soils were unsuitable for any crop production and that agricultural activities should be forbidden.

In addition in some hot spot areas atmospheric deposition may remain at high levels, even though this seems only likely in the case of non-compliance with the envisaged environmental standards. *Welch* (1996) shows severe implementation deficits of environmental policy (and the reasons for these) for Katowice voivodship in southern Poland. It seems not impossible that some of these will remain at least during the near future.

In hot spot areas therefore problems with soils contaminated with heavy metals are likely to remain into the next decades. Since the future heavy metal load is expected to decrease problems will mainly be related to releases of accumulated heavy metals in soils, particularly for cadmium. The political changes in the project area may raise environmental concern in general. This could lead to increased abandonment of already contaminated soils in the region. Therefore, the land use changes described in Scenario 1 may be especially likely for hot spot areas.

3.3. Scenario 2 - Intensive agricultural production until 2050

This scenario simulates a continued cadmium and lead load from both, agricultural practices (P-fertilizing and manuring) and atmospheric deposition. The range of future atmospheric deposition has already been described in the former section. As opposed to historic heavy metal loads, agricultural load has now become the major source of cadmium. Even for lead, for which during the past 40 years more than 90% of the load was attributed to atmospheric deposition, for the future about half of the load will be from agricultural sources. In scenario 2 an average and maximum cadmium and lead load from agricultural sources is analyzed. Table 11 summarizes the assumptions we used for scenario 2.

The average load in our project area during the 1980s, when agricultural production was intensive, was 3 g/ha/a for cadmium and 12 g/ha/a for lead. 5 g/ha/a for cadmium reflects the maximum estimate for our project area during the period 1955 to 1994. However, a study on cadmium accumulation in the EC (Fraters and vanBeurden, 1993) estimates for 1985 Belgium, Germany and the Netherlands a cadmium load of 5 g/ha/a, the average for the EC12-countries is 4.7 g/ha/a. The reason is a combination of both, in the 1980s agricultural production in the EC-12 was more intensive than in our project area and the P-fertilizers used contained a higher amount of cadmium as a trace element. For lead 18 g/ha/a was the maximum load in our project area during the past 40 years.

| Table 11. | Cadmium and lead load from agricultural sources and atmospheric deposition |
|-----------|--|
| | that were used in scenario 2 (in g/ha/a) |

| | agricultural | load 1994 to 2050 | atmospheric deposition 2010 to 2050* | | |
|---------|--------------|-------------------|--------------------------------------|---------|--|
| | average | maximum | average | maximum | |
| Cadmium | 3 | 5 | 0.7 | 1.2 | |
| Lead | 12 | 18 | 13 | 28 | |

* between 1994 and 2010 there was a continuous decrease in deposition as defined by De Bruyns scenarios (see section 3.1).

Finally two different heavy metal loads are tested. The first is the sum of the grid with maximum atmospheric deposition and maximum agricultural load (cumulative load from 1995 to 2050 is 350 g/ha for cadmium and 3200 g/ha for lead). The second assumes average agricultural load and average atmospheric deposition (cumulative load is 220 g/ha and 190 g/ha for cadmium and lead respectively). For each load three different pH-values (5, 6 and 7) typical for agricultural soils and three organic matter contents (0.8, 3.5 and 7%) are examined. The same two initial 1995 heavy metal soil concentrations are used as in scenario 1.

Cadmium

Accumulation of cadmium again depends strongly on pH-value and organic matter concentration. For most assumptions in this scenario there is a net increase in cadmium soil concentration until 2050 (table 12). The maximum increase is 0.1 mg/kg over the 55 year period (at high ph-values, high organic matter contents combined with our estimated

maximum future cadmium load). However, at a pH-value of 5 and low organic matter soil concentration the model calculates a net leaching of cadmium. An interesting feature here is that the soil model foresees a stronger leaching at similar conditions if the initial soil concentration is higher. Figure 7, that shows the relative binding strength of a soil to accumulate cadmium may be used to assist in a regional analysis. Those areas that show a high binding strength are more likely to accumulate cadmium.

| pH-value | | 5 | | | 6 | | | 7 | |
|-----------------------------------|------|--|------|------|------|------|------|------|------|
| organic matter [%] | 0.8 | 3.5 | 7 | 0.8 | 3.5 | 7 | 0.8 | 3.5 | 7 |
| | | 1995 soil conc.: 0.23 mg/kg (=60 mg/m ²) | | | | | | | |
| max. load (35 mg/m ²) | 0.10 | 0.23 | 0.27 | 0.22 | 0.29 | 0.31 | 0.29 | 0.32 | 0.33 |
| avg. load (22 mg/m ²) | 0.07 | 0.19 | 0.22 | 0.18 | 0.25 | 0.26 | 0.25 | 0.28 | 0.28 |
| | | 1995 soil conc.: 0.6 mg/kg (=150 mg/m ²) | | | | | | | |
| max. load (35 mg/m^2) | 0.16 | 0.44 | 0.53 | 0.40 | 0.57 | 0.60 | 0.56 | 0.63 | 0.64 |
| avg. load (22 mg/m ²) | 0.13 | 0.40 | 0.48 | 0.36 | 0.52 | 0.56 | 0.51 | 0.58 | 0.59 |

Lead

With our estimated loads for lead no major changes in soil concentration are expected between 1995 and 2050. For the higher assumption of the 1995 lead soil concentration there is even a small lead loss of 1 mg/kg soil. The reason is that the lead load is the same as the lead lost via erosion. For example the average yearly load in scenario 2 is 2.5 mg/m²/a (= 25 g/ha/a; this is the sum of average agricultural input and atmsopheric deposition as shown in tables 11). Assuming 2 t/ha/a erosion and a soil concentration of 10 mg/kg, the yearly loss via erosion is 2 mg/m². The erosion and lead soil concentrations assumed here are low from a soil conservation point of view. Further, the estimated cumulative load on a regional scale is less than 5% of the lead already present in the upper soil 20 cm soil layer (table 9).

Hot spot areas

The estimates so far are valid for the regional scale. Like in the case of Scenario 1 the effects in hot spot areas could be much stronger. The future problems however are expected to result mainly from the high heavy metal soil concentrations that have already been accumulated during the last decades. It is difficult to estimate future hot spot depositions. Only an indication may be the measured deposition in 1992 in southern Poland of 100 g/ha/a (see table 3) considering that in 1992 effects of the structural changes in the region should have already become visible.

4. Conclusions

This report investigates effects of long term load of two heavy metals, cadmium and lead to agricultural soils. A soil model (Shimada and Jaffe, 1996a) is used to perform a quantitative analysis of potential accumulation or release of cumulative heavy metal loads. Historic cadmium and lead loads to agricultural soils for a project area in Central Europe (fig. 1) as well as future plausible scenarios are investigated. Estimates for historic heavy metal loads are derived from databases on atmospheric deposition and agricultural loads for the period 1955 to 1994 (Olendrzynski et al, 1995 and Prieler et al, 1996a). In addition extremely high depositions that were measured in a "hot spot" region in the project area are included in the analysis. Based on a GIS database for the project area including soil types, land use, precipitation, topography and cumulative heavy metal loads a regional analysis of historic accumulation and release of cadmium and lead during the period 1955 to 1994 was performed.

The time frame for the scenario approach is 1995 to 2050. Scenario 1 simulates the abandonment of agricultural land and/or a conversion into forest. Atmospheric deposition is the only source of heavy metal load. Effects of pH-value declines, triggered by the land use changes in this scenario, are studied. Scenario 2 assumes intensive agricultural production until 2050. Atmospheric deposition as well as agricultural activities are sources of heavy metal loads. Different pH-values and organic matter contents in soils shall reflect different environmental conditions that are typical for agricultural soils.

The methodological approach of this analysis, combining a temporal and regional perspective, suggests that the following factors and in particular their interrelationship are *key elements* in determining the mass balance of heavy metals in soils and their potential to endanger a long term use of these soils for growing agricultural food products. For lead they include the load of heavy metals, the background concentration of heavy metals in the beginning of the studied period, erosion and the time period studied. For Cadmium, in addition to these factors, the pH-value and organic matter content of the soil are of particular importance. While lead stems primarily from atmospheric deposition, both atmospheric deposition and agricultural input are of importance for cadmium.

Precipitation may also be included in this list since it determines infiltration rates and the movements for transports of heavy metals. A significant difference can however only be seen by comparing for example average precipitation rate of 500 - 600 mm per year, with the 1000 mm precipitation observed at higher altitudes in the project area. The analysis in this study used average precipitation in Central Europe.

In general a discussion on heavy metal soil problems requires a thorough distinction between the regional scale and local hot spot problems. The major difference between cadmium and lead is their sensitivity to soil properties. Cadmium accumulation or release depends highly on pH-value and organic matter content. Lead accumualtes in the soil at those pH-values and organic matter contents that are typical for agricultural soils.

Cadmium

The share of cadmium loaded to the soil that remains in the upper 20 cm soil layer strongly depends on the soil pH-value and organic matter content. Combined with variations in cadmium loads this causes significant regional differences for cadmium accumulation or release. Cadmium soil background concentration in 'unpolluted' sites are in the order of 0.05 to 1 mg/kg. Guidelines are 0.5 to 1 mg/kg depending on soil characteristics.

Historic average cadmium loads to agricultural soils in the project area were about 10 g/ha/a. Assuming a low initial cadmium soil concentration of 0.05 mg/kg in 1955 and this average load, there is a net loss of cadmium over the period 1955 to 1994 at a pH-value of 4 and an organic matter content of less than 1%. On the other end, at a pH-value of 6 and organic matter content of more than 3%, or a pH-value of 7, more than 90% of the loaded cadmium remains in the soil. An approximate average for cadmium accumulation in the upper 20 cm soil layer, based on an average cadmium load and an average agricultural pH-value of 6 to 7, is about 0.1 mg/kg over the studied 40 years period.

At high pH-values, the maximum cadmium load to the project area causes a 40 year increase of 0.3 mg/kg in Cadmium soil concentration (table 2). In other words, the higher range of historic cadmium loads combined with high pH-values and organic matter contents, will accumulate an additional 0.2 mg/kg in approximately 25 years. In a short term perspective with a similar forty year increase of 0.1 or 0.3 mg/kg seems not significant. In a long term perspective however, such as 100 year, with continued equally high deposition, these increases grow in importance and may reach guideline values depending on the cadmium soil concentration at the beginning of the studied period.

Consequently for the majority of the project area there was no significant increase in cadmium soil concentration. At the high end of cadmium loads and at average cadmium load combined with high pH-values, the increase in soil concentration is considered to be of concern in a long term perspective of about 100 years. Any risk assessment here depends on the definition of guidelines and standards, the cadmium soil concentration in the beginning of the studied period and on the time horizon taken into account.

A cadmium mass balance for the project area covering the period 1955 to 1994 suggests that from the cumulative load of 3062 t of cadmium entering the soil, one third is lost (836 t) and two thirds are accumulated in the soil (2226 t). About two thirds of the load stems from atmospheric deposition and one third from agricultural sources. The majority of the loss occurs via leaching and plant uptake, with leaching being slightly more important than plant uptake and subsequent removal. Despite very low assumptions for erosion rates in the model nearly 10% of the total loss occurs via erosion (table 6). The regional analysis shows an average increase of 0.08 mg/kg for the project area (table 5).

Locally, in *hot spot areas*, measurements of cadmium deposition are as high as 100 g/ha/a. If not already exceeded, assuming a low initial cadmium soil concentration and continued high deposition, guideline values will be exceeded within 10 to 50 years depending on pH-value and organic matter content (table 4).

Estimates of *future* cadmium atmospheric deposition are low compared to historic depositions (table 8). A comparison of the cumulative cadmium loads during the periods 1970 to 1990 and 1991 to 2010 shows a decrease in cumulative cadmium deposition between 60 and 90% (fig.5).

In scenario 1, simulating a pH-decline and atmospheric deposition as the only source of cadmium load, even the maximum assumed deposition is not high enough to compensate cadmium loss due to declining pH. Consequently by 2050 cadmium soil concentration decreased compared to 1995. The extent of decrease is dependent on the assumed initial soil concentration in 1995, the extent of pH-decline and organic matter content (table 10). Since the cumulative future atmospheric deposition is very low compared to the cadmium already stored in the soil, it has a neglegible influence on the soil concentration in 2050 (table 9).

Scenario 2 simulates intensive agricultural production until 2050. Cadmium load now stems from both, agricultural activities and atmospheric deposition. Because of the low atmospheric deposition agricultural sources become the major source of cadmium load to agricultural soils. Here, assuming an initial soil concentration of 0.2 mg/kg in 1995 and a yearly load of 4 g/ha (average), the maximum soil concentration in 2050, occurring at high pH-values and organic matter contents, is 0.28 mg/kg (i.e. a delta increase of 0.08 mg/kg over the 55 years period). At a pH-value of 5 and pH-value of 6 combined with a low organic matter content, there will be no change in soil concentration or a decrease in soil concentration (table 12).

Overall, for the future 50 years minor increases in cadmium soil concentration are expected. The main reason is the expected decrease in atmospheric deposition. Intensive agricultural production will keep the cadmium soil content at a similar level as in 1995. Abandonment of agricultural land with subsequent pH declines will trigger major releases of cadmium from the soil. The amount of cadmium leaching to deeper soil layers and subsequent to the groundwater depends most importantly on the present cadmium soil concentration. Again pH-value and organic matter content are also of importance (table 10). In general pH-values below six or a pH-value of six combined with an organic matter content of less than 2% triggers major cadmium mobilization.

Locally, in hot spot areas, the developments described in scenario 1 and 2 may be much stronger. Hot spot areas are most likely to remain in those areas where the soil pollution is already high at present and in areas where envisaged pollution control measures face implementation problems. The latter refers in particular to guidelines on atmospheric emissions and sewage sludge application.

A qualitative analysis of the binding capacity of soils for cadmium in the project area (fig. 7) was performed. It enables us to identify areas which are likely to accumulate versus areas that are likely to release cadmium. In the case of ceasing agricultural production soils with a low cadmium binding capacity are likely to release, while soils with high binding capacity will accumulate more cadmium when agriculture remains intensive.

Lead

More than 90% of lead loaded to soils accumulates in the soils. Therefore regional differences are determined by the estimated background concentration and the amount of lead loaded. At lower lead loads, lead lost via erosion is in the same range as the loaded lead, even at low erosion rates.

In the project area over the past forty years the lead loaded was small (the 40 year average increase is 3 mg/kg, based on a yearly load of 200 g/ha) compared to the range of natural background concentrations (2 - 60 mg/kg) and guidelines (50 - 100 mg/kg). Only locally high measured depositions of more than 1000 g/ha/a cause soil concentration increases over

20 mg/kg over a 40 year period (table 7). Therefore lead soil contamination is concentrated in hot spots area and has to be considered as a local problem only.

Like in the case of cadmium, the future scenarios expect a significant decrease in atmospheric lead deposition. The maximum lead deposition in 2010 in our project area is 28 g/ha/a compared to an average lead deposition between 1955 and 1994 of about 200 g/ha/a (table 8, fig. 5). At low loads the lead loss via erosion compensates the lead loaded via atmospheric deposition. The estimated cumulative future lead load is very low compared to lead already stored in the soils (table 9). For the future 50 years no significant increase in lead soil concentration is expected, expect in local hot spot areas.

To summarize, on a regional scale, there was a minor accumulation of lead during the past 40 years and for the future no increases are expected. Locally, in "hot spot" areas significant increases in lead soil concentration must have taken place in the past amounting to at least a doubling of lead soil concentration during a 50 year period.

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| Soil Type | | Area | pH-value | OM [%] |
|-----------|--------------------------|------------|----------|--------|
| <u> </u> | | [1000 km²] | | |
| Bd | Dystric Cambisol | 37.74 | 5 | 2.6 |
| Lo | Orthic Luvisol | 22.32 | 6 | 2.1 |
| Po | Orthic Podzol | 16.51 | 5 | 3.4 |
| Be | Eutric Cambisol | 13.57 | 6 | 3.1 |
| Lgs | Stagno-gleyic Luvisol | 8.68 | 6 | 2.6 |
| Gd | Dystric Gleysol | 8.67 | 5 | 3.4 |
| Je | Eutric Fluvisol | 8.54 | 6 | 5.2 |
| Dd | Dystric Podzoluvisol | 6.76 | 5 | 2.6 |
| Ch | Haplic Chernozem | 6.62 | 7 | 5.2 |
| Jc | Calcaric Fluvisol | 6.56 | 7 | 5.2 |
| Pl | Leptic Podzol | 6.36 | 5 | 3.4 |
| Bds | Spodo-dystric Cambisol | 5.09 | 5 | 8.6 |
| Oe | Eutric Histosol | 3.86 | 6 | 34.4 |
| Qc | Cambic Arenosol | 3.66 | 6 | 0.9 |
| Gm | Mollic Gleysol | 3.60 | 6 | 2.6 |
| QI | Luvic Arenosol | 3.18 | 6 | 0.9 |
| Eo | Orthic Rendzina | 2.69 | 7 | 4.3 |
| Bv | Vertic Cambisol | 2.53 | 6 | 3.4 |
| Gh | Humic Gleysol | 2.46 | 6 | 10.3 |
| Bgg | Stagno-Gleyic Cambisol | 2.04 | 6 | 3.1 |
| La | Albic Luvisol | 1.97 | 5 | 2.6 |
| Lga | Albo-gleyic Luvisol | 1.81 | 5 | 2.6 |
| Ge | Eutric Gleysol | 1.52 | 6 | 2.6 |
| Ck | Calcic Chernozem | 1.11 | 7 | 5.2 |
| Hh | Haplic Phaeozem | 0.87 | 6 | 5.2 |
| Cl | Luvic Chernozem | 0.75 | 6 | 5.2 |
| Hc | Calcaric Phaeozem | 0.69 | 7 | 5.2 |
| U | Ranker | 0.50 | 6 | 3.1 |
| Bec | Calcaric eutric Cambisol | 0.49 | 7 | 3.1 |
| Ba | Calcaric Cambisol | 0.36 | 7 | 2.6 |
| Mo | Orthic Greyzem | 0.21 | 6 | 5.2 |
| Ic | Calcaric Lithosol | 0.17 | 7 | 0.9 |
| Rd | Dystric Regosol | 0.13 | 5 | 0.9 |
| Rc | Calcaric Regosol | 0.06 | 7 | 0.9 |
| Od | Dystric Histosol | 0.04 | 5 | 34.4 |

Appendix 1. Distribution of soil types in the project area - Estimates of pH-value and organic matter content for each soil type assuming agricultural land*

*...Ph-value and organic matter content was required as input into the heavy metal soil model (results from this exercise see fig.2-4) and for the qualitative assessment of the relative binding capacity of soils for cadmium (results in fig.7).