Working Paper

Heavy Metals Contamination in Eastern Europe: Background Load from the Atmosphere

László Bozó
Joseph Alcamo
Jerzy Bartnicki
Krzysztof Olendrzyński

WP-91-46
November 1991

IIASA International Institute for Applied Systems Analysis □ A-2361 Laxenburg □ Austria
Telephone: +43 2236 715210 □ Telex: 079 137 iiasa a □ Telefax: +43 2236 71313

Heavy Metals Contamination in Eastern Europe: Background Load from the Atmosphere

László Bozó
Joseph Alcamo
Jerzy Bartnicki
Krzysztof Olendrzyński

WP-91-46
November 1991

International Institute for Applied Systems Analysis □ A-2361 Laxenburg □ Austria
Telephone: +43 2236 715210 □ Telex: 079 137 iiasa a □ Telefax: +43 2236 71313

Forword

Since the revolutions of 1989 the public throughout Europe and elsewhere has been awakened to the scope of environmental problems in Central and Eastern Europe. Unfortunately, it has also become clear that economic and political problems in the region will delay any kind of massive effort to solve these problems. Instead, the amount of funds and resources available for environmental protection will be rather modest in the coming years. It is therefore crucial to carefully analyze environmental problems in the region so that sensible priorities can be set for pollution control.

As part of these analyses it is also important to keep in mind that these problems have an important regional and international aspect in that air pollutants travel long distances and water pollutants are often discharged into international waterways. Therefore, these analyses should have both a local and "regional" perspective. The IIASA project on "Toxic Pollution and the European Environment" focuses on this regional perspective in its analysis of problems in Europe. This Working Paper is one of the project's efforts to analyze the extent of heavy metals contamination from the atmosphere in the Central and Eastern European region. The authors take exception to the conventional view that heavy metals contamination is only a "local", urban problem which occurs in the vicinity of a smelter or other pollutant source. Using computer modeling as a tool, they point out that heavy metals contamination also has a regional and rural aspect, and belongs to the class of problems having a transboundary nature. These findings should be taken into account when setting priorities for environmental protection in Eastern Europe.

Joseph Alcamo
Leader, Toxic Pollution and the European Environment

Acknowledgements

The authors wish to thank F. Axenfeld, J. Münch, and J. Pacyna for providing emissions' data needed for these calculations, and W. Schöpp and E. Taffel for their computer assistance in drawing the maps. They also are grateful to E. Meszaros for reviewing this manuscript.

Abstract

In recent years there has been an increased interest in trace metals in the atmosphere and the environmental effects of their deposition. This is to large extent because heavy metals can accumulate in the biosphere and may be toxic to living systems. On the basis of IIASA's TRACE model, the total (wet plus dry) deposition of As, Cd, Pb and Zn has been estimated for Eastern Europe. These are annual averages for rural areas, and relate to the situation in Europe in the mid-1980s. The maximum deposition value is 3.5 mg m^{-2} yr^{-1} for As, 1.5 mg m^{-2} yr^{-1} for Cd and 50 mg m^{-2} yr^{-1} for Zn. All these maxima occur in Southern Poland. The highest total deposition of Pb (15.0–20.0 mg m^{-2} yr^{-1}) has been computed for western Czechoslovakia and also for southern Poland. Deposition levels throughout most of Eastern Europe are at least one or two orders of magnitude greater than observed in remote parts of the world.

The annual average concentration of metals in some rural areas are lower, but within a factor of two of drinking water guidelines. This is cause for concern because some short-term concentrations are almost assuredly much higher than the annual average.

Because of long-range transport, there is a very significant transboundary exchange of heavy metals within Eastern Europe. As with acid-causing pollutants, the problem of heavy metals contamination in the region depends on the reduction of this transboundary pollution.

Contents

Heavy Metals Contamination in Eastern Europe: Background Load from the Atmosphere

László Bozó
Joseph Alcamo
Jerzy Bartnicki
Krzysztof Olendrzyński

1 Introduction

Since the middle of this century, energy generation, industrial production and transportation have caused serious environmental contamination by trace elements including heavy metals. The rate of contamination can vary from place to place as a function of source densities and intensities of heavy metals flux as well as meteorological conditions. Aerosols containing heavy metals can also be transported far away from their sources by advection before being deposited.

The TRACE model developed at IIASA (Alcamo, et al., 1991, Bartnicki and Alcamo, 1991) focuses on European-scale emission, transport and deposition of four heavy metals: As, Cd, Pb and Zn. These metals are particularly important because of their toxicity and/or ability to accumulate in the natural environment. On the basis of TRACE model calculations, this paper presents calculations of total deposition, concentration in precipitation and budgets of heavy metals in Eastern Europe. An attempt has also been made to estimate the effects of the most intensive regional sources in Eastern Europe on the deposition of As, Cd and Zn in the surrounding countries.

2 Modeling Deposition Processes of Heavy Metals

The main sources of As, Cd, and Zn in the lower atmosphere over Eastern Europe are power plants, metallurgical plants and other large industrial facilities. For Pb, a main source is also motor vehicles. Pacyna and Münch (1988). These metals are emitted to the atmosphere either as volatile gases or very fine particles and usually disperse and mix fairly quickly into the lower atmosphere. Within a few minutes or hours the emitted gases condense into or adhere onto fine particles, in the size range of 0.1 to 10 microns in diameter. Particles of this size are too fine to effectively settle, and too coarse to be deposited by diffusion. Hence, they can be transported hundreds or more kilometers from their sources before gradually being removed from the atmosphere by dry or wet deposition. Particles are continuously deposited onto soil, vegetation, lakes, and other surfaces by gravitational settling and diffusion. This is called "dry deposition". Particles containing heavy metals are also swept from the air by precipitation as it falls to earth or are incorporated into cloud droplets and later fall to earth as these cloud droplets are

collected by precipitation. This process is called "wet deposition". The TRACE model represents these processes in a simple way for all Europe.

The model relies on long-term average meteorological data as input, and computes the long-term average levels of heavy metals in the atmosphere. Details are presented in Alcamo, et al., (1991); here we only briefly review its main features.

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

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

Where c = air concentration at the receptor due to a single emission source; (x_r, y_r)= receptor position; (x_e, y_e)= emission source position; E = amount of emissions at the source; R = distance between source and receptor, i.e. $R = \sqrt{(x_r - x_e)^2 + (y_r - y_e)^2}$; α = local deposition coefficient; k_d and k_w = first order loss coefficients, in units of inverse time, which reflect the loss of mass from the air parcel by dry and wet deposition, respectively; and t^* = time of travel between sources and receptors. The factor β is derived by assuming mass conservation and is given by:

$$\beta = [2\pi hk \cdot A]^{-1} \tag{2}$$

where h = mixing height, k = $k_d + k_w$, and A is given by

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

in which $y = x + \frac{b}{2a}$. The variable $\bar{x}$ is equal to $\frac{\delta x}{\sqrt{\pi}}$ in which δx is the model grid size. The constants a and b in this equation are estimated from a relationship between geographic distance and transport time described in Alcamo, et al., (1991).

The total concentration, $c(x_r, y_r)$, at the receptor is computed from the sum of contributions coming from all emission sources, weighted according to the frequency of backward trajectories, $F(s)$, coming from a particular sector, s:

$$c(X_r, Y_r) = \sum_{s=1}^{8} F(s) c_s(x_r, y_r) \tag{3}$$

In the second step of the calculation, wet and dry deposition of the pollutant at the receptor is computed from the air concentration. Wet deposition, d_w, is computed with a scavenging ratio:

$$d_w = c(x_r, y_r) \cdot W_q \cdot P \tag{4}$$

where W_q is the scavenging ratio, i.e. the ratio of the concentration of heavy metals in precipitation to their concentration in air. Despite the simplicity of this approach, it nevertheless takes into account the amount of precipitation at a particular location, which accounts for much of the spatial variability of wet deposition.

Dry deposition is computed from:

$$d_d = c(x_r, y_r) \cdot v_d \tag{5}$$

The dry deposition velocity v_d is computed with the semi-empirical model of Sehmel (1980). This deposition model is based on wind-tunnel experiments and theoretical removal rates via brownian diffusion and gravitational settling. Dry deposition velocities are computed as a function of particle size (D_p), surface roughness (z_o) and friction velocity (u_*). The advantage of using this model over assigning deposition velocities *a priori* is that it provides an independent basis for estimating the spatial variation of dry deposition velocity in Europe as a function of "local" meteorological conditions (as indicated by u_* and z_o). Also, the necessary friction velocity and surface roughness data are available on a European grid.

Disadvantages of this model are that it is based partly on empirical wind tunnel measurements and that it requires as input the characteristic size of metal particles in the atmosphere, which is obviously difficult to estimate for all Europe.

We use an assumed particle size distribution for heavy metals to compute a size-weighted v_d:

$$v_d = \sum_{i=1}^{n} v_d(D_{p_i}, u_*, z_o) f(D_{p_i}) \tag{6}$$

where $v_d(D_{p_i}, u_*, z_o)$ is taken from the curves presented in Sehmel (1980). Data for u_* and z_o were obtained on a European grid with a spatial resolution of 150×150 km^2 from the EMEP Synthesizing Center West (J. Saltbones, personal communication).

The variable f(D_{p_i}) is the fraction of mass with diameter D_{p_i}. For these data we have provisionally used the Mediterranean measurements by Dulac, et al., (1989) to represent Southern European conditions. To represent Northern European conditions, we use particle data measured in Norway by Cornille, et al., (1991). These data were used because (1) they cover relatively long periods of measurement (3 to 12 months) rather than only short field campaigns; (2) they were collected at several sites and represent a wide geographic area rather than a single station; (3) the measurement sites were probably not significantly affected by local sources but nevertheless were influenced by distant anthropogenic sources. These characteristics are consistent with the assumptions of the TRACE model.

The model was tested with air concentrations and wet deposition data measured at several sites in Europe between 1978 and 1985 (Alcamo, et al., 1991). Because of uncertainty of data from Eastern Europe, only measurements from Western Europe (with one exception) were used for testing. This is an unfortunate situation since it would be obviously more desirable to test the model's accuracy in Eastern Europe by comparing it with data from this region. On the other hand, it is known from other studies that emissions of heavy metals from Eastern Europe significantly affect measured levels of heavy metals in Northern and Western Europe (see, e.g. Pacyna, et al., 1984). Hence, the agreement of model calculations with measurements in these areas provide some validation of calculations in Eastern Europe itself. As to the results of model testing— in general, it was found that model calculations agree fairly well with As and Pb data, but underestimate Cd and Zn data. After sensitivity analysis of the TRACE model it was concluded that this underestimation may be due to either underestimated emissions or measurement contamination before 1985 which could have resulted in extreme overestimation of measurements (Alcamo, et al., 1991).

The only observations from Eastern Europe used to test the model were Cd and Pb data measured at the EMEP station K-Puszta in Central Hungary (46° 58′ N, 19° 35′ E) (Table 1). Model results at this station were similar to results at Western European

Table 1: Model calculations versus measurements at K-puszta in Hungary.

Constituent	years	Calculated		Measured	
		Mean	Range	Mean	Range
Pb Air Concentration (μg m^{-3})	1981–85	28.95	23.40–35.23	56.6	45.10–71.90
Pb Wet Deposition (mg m^{-2} a^{-1})	1983–85	5.44	4.11– 6.32	6.68	5.03– 7.69
Cd Air Concentration (μg m^{-3})	1981–85	0.31	0.28– 0.36	1.21	1.03– 1.50
Cd Wet Deposition (mg m^{-2} a^{-1})	1984–85	0.095	0.09– 0.10	0.63	0.55– 0.71

stations in that model calculations were within a factor of two of Pb measurements, and underestimated Cd measurements.

3 Long-term Deposition of Heavy Metals

The annual total deposition of heavy metals is an important indicator of their long-term impact on the environment because many metals are known to gradually accumulate in lake and river sediments, in forest soils and in vegetation (Ottar, et al., 1989). Despite the fact that heavy metals contamination can lead to many environmental impacts, not much work has been devoted to quantifying a threshold deposition level above which the natural environment is adversely affected. To this point, only damage threshold levels for sulfur and nitrogen deposition have been recommended in Europe (see, e.g. Sverdrup, et al., 1990).

Figures 1 (a) through (d) present the computed total deposition (wet plus dry) of heavy metals for Eastern Europe. These maps are based on 1985 meteorological data such as precipitation and wind direction because reliable meteorological data are available for this year for all Europe. Emissions data are from 1982 for As, Cd, and Zn, and 1985 for Pb because these were the most recent data available for gridded emissions covering all of Europe. Hence, these maps present the situation in Europe as of the mid-1980s.

The As deposition pattern shows two peaks—one in the Southern part of Poland and one in Eastern Yugoslavia (Figure 1 (a)). The maximum deposition value is around 3.5 mg m^{-2} yr^{-1} in Poland. The local maximum is over 2.0 mg m^{-2} yr^{-1} in Yugoslavia. Moving towards the edge of the continent total deposition of around 0.1 mg m^{-2} yr^{-1} is reached. It can be seen from this figure that total arsenic deposition remains relatively high (2.0–3.0 mg m^{-2} yr^{-1}) even at a distance of more than 100 km from the peak areas. The lowest As total deposition values can be seen in Hungary and Romania.

Although, as noted above, the threshold levels of As and other metals are not known, we can at least compare their computed magnitude in Europe with their levels in remote parts of the world. (Antarctica, South Pacific, and similarly remote areas.) Arsenic deposition, for example, ranges from 0.01 to 0.20 mg m^{-2} yr^{-1} in remote areas (Lahmann, et al., 1986). This is exceeded by one or two magnitudes throughout most of Europe. (Figure 1 (a)).

The Cd pattern shows one high and several lower peaks in Eastern Europe (Figure 1 (b). As for arsenic, the high peak occurs in South Poland and the lower peaks in the central part of Bulgaria and South-eastern part of Yugoslavia. The maximum of total Cd deposition is around 1.5 mg m^{-2} yr^{-1} in Poland and 0.4–0.5 mg m^{-2} yr^{-1} in Yugoslavia and Bulgaria. These levels may be compared to the range of 0.006 to 0.229 mg m^{-2} yr^{-1} for remote areas (Lahmann, et al., 1986).

The pattern of Pb (Figure 1 (c)). is different from the other metals because Pb has a different emission profile. Sources of Pb are relatively well distributed motor vehicles, while As, Cd and Zn come mostly from point sources in industrialized areas. It is also to be taken into consideration that the removal rate of Pb is less than for the other metals so Pb on-the-average travels further and can be deposited over a wider area with lower maxima. Highest total deposition of Pb has been computed for the western part of Czechoslovakia and South Poland (15.0–20.0) mg m^{-2} yr^{-1}. It can be assumed—regarding the prevailing wind directions over this part of Europe—that this relatively high level of deposition is caused by the higher emission densities in the Western European countries. Deposition levels of Pb in remote areas of the world range from 0.026 to 3.65 m^{-2} yr^{-1} (Lahmann, et al., 1986).

The deposition pattern of Zn (Figure 1 (d)) is very similar to that of As. This is not surprising taking into consideration that a major source of As is zinc smelting. Differences are only in the absolute values of total deposition. In the Southern part of Poland it reaches 50 mg m^{-2} yr^{-1}. In Bulgaria and Yugoslavia two peaks can be detected by the TRACE model calculations. Over these areas the total deposition of Zn is 10.0–15.0 mg m^{-2} yr^{-1}. On the other part of Eastern Europe the value of total Zn deposition is 2.0–7.0 mg m^{-2} yr^{-1}.

4 Concentration of heavy metals in precipitation

Because rainwater is used directly or indirectly as a source of drinking water in Eastern European rural areas, the concentration of metals in precipitation is important to public health. Since the monitoring of these elements in precipitation is very scattered in Eastern Europe, the only possibility to estimate their concentration for the whole area of Eastern Europe is through model computation. Using the TRACE model, the concentrations of As, Cd, Pb and Zn have been calculated on the basis of wet deposition calculations (also by TRACE model) and annual precipitation data over the EMEP grid system. Results of these computations are presented in Figure 2. For reference, we present some recommended international guidelines for these substances in drinking water (Table 2.).

In general, the patterns of concentration in precipitation (Figure 2) are similar to total deposition patterns (Figure 1). However, deposition patterns have somewhat stronger spatial gradients because "local" deposition is quite high in the vicinity of strong sources.

In the case of As (Figure 2(a)), two large areas with local maxima can be spotted. The first one is in South Poland—North Czechoslovakia with a maximum over 2.0 μg l^{-1}. In the surroundings of this area a circle with a radius of around 300–400 km can be detected where the As concentration in precipitation is 1.0–2.0 μg l^{-1}. This area covers practically the whole territory of Czechoslovakia and the Southern and Central part of Poland. A lower local maximum is situated around the Bulgarian-Romanian-Yugoslavian border. A peak of near 2.0 μg l^{-1} occurs in Yugoslavia. Towards the edge of the continent the As concentration in precipitation decreases to around 0.1 μg l^{-1}.

The Cd pattern (Figure 2(b)) shows one considerable peak in Poland (over 0.7 μg l^{-1})

Table 2: Examples of guidelines for heavy metals in drinking water.

Metal	μg/l	Institution
As	50	World Health Organization[a]
	50	European Community[b]
	10	Rhine Basin Association—Category "A" [c]
	50	Rhine Basin Association—Category "B" [c]
Cd	5	World Health Organization[a]
	5	European Community[b]
	1	Rhine Basin Association—Category "A" [c]
	5	Rhine Basin Association—Category "B" [c]
Pb	50	World Health Organization[a]
	50	European Community[b]
	30	Rhine Basin Association—Category "A" [c]
	50	Rhine Basin Association—Category "B" [c]
Zn	500	Rhine Basin Association—Category "A" [c]
	1000	Rhine Basin Association—Category "B" [c]

[a]WHO (1984) Guidelines for Drinking- Water Quality. Vol. 1. Recommendations. WHO: Geneva.

[b]EC Directive 80/778/EEC. (1980).

[c]Internationale Arbeitsgemeinschaft der Wasserwerke im Rheineinzugsgebiet. (1987) Rheinbericht '86/'87. Water supplies in Category "A" do not require special treatment before being used for drinking water; Category "B" requires physical/chemical treatment.

and two others in Bulgaria and Yugoslavia (0.3–0.4 μg l^{-1}). In the central part of Eastern Europe (Hungary, North-western part of Romania) the Cd concentration in precipitation is 0.1–0.2 μg l^{-1}. On the Northern and Adriatic coast of Yugoslavia the Cd concentration is below 0.1 μg l^{-1}. In North Czechoslovakia the Cd concentration is 0.2–0.4 μg l^{-1}1 partly due to the effect of South Polish sources.

The pattern of Pb—as it was indicated when presenting the total deposition maps—is different from the other metals. Pb concentration in precipitation over 15.0 μg l^{-1} can be seen in certain parts of Poland. In other regions of Eastern Europe it varies between 2.5 and 15.0 μg l^{-1}.

The maximum Zn concentration (Figure 2(d)) in precipitation has been computed also for South Poland. Its value is over 20.0 μg l^{-1}. A smaller maximum in Bulgaria can also be detected (around 10.0 μg l^{-1}). For the remainder of Eastern Europe Zn concentration of 2.0–5.0 μg l^{-1} has been calculated.

On one hand it can be seen that computed levels of all elements are below the drinking water guidelines presented in Table 2. On the other hand, the concentration of these elements are within an order of magnitude of these guidelines in the maxima regions, and in some case within a factor of 2 of these guidelines. This is of importance because over short periods of time these concentrations can be much higher than the annual average.

Table 3: Arsenic—annual emissions (1982) and deposition (mid-1980s).

Country	Emitted ($t\ yr^{-1}$)	Deposited ($t\ yr^{-1}$)	Average Flux ($kg\ km^{-2}\ yr^{-1}$)
Albania	17.2	22.2	0.77
Bulgaria	146.9	73.7	0.66
Czechoslovakia	93.7	160.0	1.25
Hungary	16.1	52.1	0.56
Poland	591.3	281.0	0.90
Romania	116.2	113.5	0.48
Yugoslavia	272.0	206.3	0.81

Table 4: Cadmium—annual emissions (1982) and deposition (mid-1980s).

Country	Emitted ($t\ yr^{-1}$)	Deposited ($t\ yr^{-1}$)	Average Flux ($kg\ km^{-2}\ yr^{-1}$)
Albania	0.70	7.03	0.24
Bulgaria	65.50	29.24	0.26
Czechoslovakia	21.60	43.19	0.34
Hungary	4.40	16.15	0.17
Poland	180.40	89.31	0.28
Romania	43.40	39.53	0.17
Yugoslavia	85.80	60.49	0.23

5 Atmospheric budgets of heavy metals for Eastern European countries

The emission densities of heavy metals vary considerably in Eastern Europe. As a result of this, total deposition of heavy metals in a country with relatively low emission density can be highly affected by other countries with a higher emission density. It is possible to estimate the relative contribution of different regions in Europe to the total deposition of each Eastern European country only on the basis of model computations.

The results of these computations—separately for each country and heavy metal—are presented in Tables 3 through 6. It was computed that both Bulgaria and Poland emit twice as much arsenic as they receive in the form of deposition. Yugoslavia also "exports" more arsenic than it receives.

In Romania the emission and total deposition of As are approximately balanced. In

Table 5: Lead—annual emissions (1982) and deposition (mid-1980s).

Country	Emitted ($t\ yr^{-1}$)	Deposited ($t\ yr^{-1}$)	Average Flux ($kg\ km^{-2}\ yr^{-1}$)
Albania	136.3	201.5	7.00
Bulgaria	1569.2	674.8	6.08
Czechoslovakia	1151.0	1752.6	13.70
Hungary	596.9	727.1	7.81
Poland	2956.3	2508.5	8.02
Romania	1154.6	1382.4	5.83
Yugoslavia	1961.9	2281.0	8.92

Table 6: Zinc—annual emissions (1982) and deposition (mid-1980s).

Country	Emitted (t yr^{-1})	Deposited (t yr^{-1})	Average Flux (kg km^{-2} yr^{-1})
Albania	36.8	183.3	6.38
Bulgaria	1760.8	760.8	6.86
Czechoslovakia	755.9	1189.8	9.30
Hungary	199.3	451.7	4.86
Poland	4040.1	2233.2	7.14
Romania	716.6	818.3	3.45
Yugoslavia	1958.8	1412.6	5.52

Hungary, Czechoslovakia and Albania the deposition exceeds the emission, i.e., they are net As importers. The effects of the high emission densities in Poland and Bulgaria can be detected as high contribution from Eastern Europe to the total deposition over Czechoslovakia, Hungary and Romania, Albania, respectively.

The situation for Cd is similar in some ways to As. Poland and Bulgaria are the most important "net Cd exporters" in Eastern Europe. The Cd emissions of Yugoslavia are also very high compared to its total deposition. Over Hungary, Czechoslovakia and Albania, the Cd deposition is far below Cd emissions. The contribution of Hungarian sources to the total deposition over Hungary has also been estimated by a simple trajectory model (Bozó and Horvath, 1991). There is a very good agreement between the two different calculations since the relative contribution estimated is around 10 % for both cases.

Pb budgets for the eastern European countries are much more balanced than they are for As and Cd. Poland, and Bulgaria have turned out to be "net Pb exporters". In the case of Bulgaria the ratio of Pb emission/Pb deposition is around 2. This is the highest value in Eastern Europe.

As expected, the Zn pattern is very similar to that of As. The most important "net exporters" are Poland and Bulgaria. Total deposition of Zn over Czechoslovakia, Hungary and Albania is much lower than their Zn emission.

If we divide the total deposition to each country by the area of the country we obtain the average annual flux of these metals (last column in Tables 3 through 6). These values are much more similar between countries than the values of total deposition. Whereas total deposition varies by up to a factor of ten between countries, flux differs only by a factor of two. Czechoslovakia receives the largest flux of all metals, and Poland has the second largest flux of all metals except Pb.

In Figure 3 we present the estimated origin of the heavy metals flux to each country broken down according to contributions from (1) the country itself, (2) Western Europe, (3) other Eastern European countries. The contribution from other Eastern countries is significant for As, Cd and Zn flux, ranging from 12 to 83% of the total flux to different countries (Figure 3(a)(b)(d)). The contribution of emissions from Western Europe to these metals is relatively minor (20% or less of the total flux). The situation for Pb is different from the other metals. Because of the the high Pb emissions in Western Europe in the mid 1980s, the West contributed significantly to the Pb flux to Eastern European countries (Figure 3(c). In the case of Poland and Czechoslovakia, the West contributed more to Pb flux to these countries than was contributed by their Eastern European neighbors.

6 Effects of sources with high emission densities to the deposition in surrounding countries

Since the particles containing heavy metals can travel long distances after being emitted, the deposition of heavy metals in rural areas is greatly affected by emissions hundreds of kilometers away. On the basis of TRACE model calculations, it was also possible to estimate the effects of these sources on the deposition heavy metals in the surrounding countries. As an example, we compute the effects of southern Polish sources on wet deposition of As, Cd and Zn (Figure 4(a)(b)(c)). It is not surprising that near the sources—approximately in 150–200 km distance—the contribution of the emitters to the wet deposition of As, Cd and Zn is over 80 %. Over the northern part of Czechoslovakia the contribution of these sources to the wet deposition pattern of As, Cd and Zn is also very high. Moving towards Hungary and Yugoslavia, the contribution decreases to 30–40%. It can also be seen in Figure 4 that the prevailing western winds in Europe cause a higher contribution to wet deposition moving eastward from the sources.

7 Summary and Conclusions

This paper presents estimates of the background contamination of rural areas in Eastern Europe due to the atmospheric load of heavy metals (As, Cd, Pb, and Zn). This contamination occurs because particles containing heavy metals travel long distances from their sources in industrial areas. The TRACE model quantifies this long range transport, and has been used in this paper to estimate annual average levels of heavy metals in Eastern Europe in the mid-1980s (the only period with reliable emissions data).

We compute that peak levels of heavy metals deposition in Eastern Europe occur in southern Poland and eastern Yugoslavia. Deposition levels of heavy metals throughout most of rural Eastern Europe exceed levels in remote areas of the world by one or two orders of magnitude. The long term consequences of this load on ecosystems in Eastern Europe should be examined.

High concentrations of heavy metals in precipitation occur, as expected, where the highest deposition occurs. However, the spatial pattern of concentration in precipitation in Eastern Europe is not quite the same as the pattern of deposition—spatial gradients of deposition are stronger because "local" deposition is very high where emissions are strong.

Although the computed concentrations of As, Cd, Pb and Zn in precipitation are below drinking water guidelines, they approach within a factor of two of these guidelines in some areas. This is of concern because it is likely that short-term concentrations are much higher than the computed annual averages.

Some countries are "net exporters" of As, Cd, Zn (Bulgaria, Poland, and Yugoslavia), whereas other countries are "net importers" (Albania, Czechoslovakia, and Hungary). The situation is different for Pb, because lead originates largely from well-distributed vehicles, whereas the other metals originate primarily from concentrated industrial sources. Hence, the emissions and deposition of Pb are more closely balanced for Eastern European countries, with the exception of Bulgaria and Poland who are net exporters of Pb.

The origin of the load of heavy metals to Eastern European countries was also investigated. Western Europe contributes 20% or less to the loads of arsenic, cadmium and zinc, but was a significant contributor to the load of Pb in the mid-1980s. Now, however, Western Europe's adoption of lead-free petrol has resulted in lower emissions in Western

Europe and a smaller absolute contribution to the Pb load in Eastern Europe.

Other Eastern European countries contribute a substantial percentage of the atmospheric load to their Eastern European neighbors. In the case of arsenic, this contribution ranges from 12% for Poland to 83% for Hungary. There is also a significant transboundary exchange of Cd, Pb, and Zn within Eastern Europe.

Summing up, although the deposition of heavy metals is very severe in industrial areas near to sources, a great amount of these heavy metals travel beyond borders and contribute to air pollution problems in neighboring countries. As with acid-causing pollutants, the problem of heavy metals contamination in Eastern Europe depends on a reduction of transboundary pollution in the region.

References

Alcamo, J., Bartnicki, J. and Olendrzyński, K. (1991). Computing heavy metals in Europe's atmosphere: Part I: Model development and testing. *Atmospheric Environment.* Submitted.

Bartnicki, J., and Alcamo, J. (1991). Computing heavy metals in Europe's atmosphere: part II. Maps of air concentration and deposition. *Atmospheric Environment.* Submitted.

Bozó, L. and Horvath, Z. (1991). Atmospheric concentration and deposition of lead and cadmium over Hungary. *Ambio* (in press).

Cornille, P., Maenhart, W. and Pacyna, J. (1991). PIXE analysis of size-fractionated aerosol samples collected at Birkenes during spring 1987. *Nuclear Instrumentation and Methods in Physics Research* (in press).

Dulac, F., Buat-Menard, P., Ezat, U., Melki, S. and Bergametti, G. (1989). Atmospheric input of trace metals to the western Mediterranean: Uncertainties in modelling dry deposition from cascade impactor data. *Tellus*, **41B**, 362-378.

Lahmann, E., Munari, S., Amicarelli, V., Abbaticchio, P. (1986). Heavy metals: identification of air quality and environmental problems in the European Community. Vol. 1 EUR 10678. CEC: Brussels.

Ottar, B, Lindberg, S.E., Voldner, E., Lindquist, O., Meyer, R., Semb, A., Steiness, E., and Watt, J. (1989). Special topics concerning interactions of heavy metals with the environment in: *Control and Fate of Atmospheric Trace Metals.* 365–372. J.M. Pacyna and B. Ottar, ed., Kluwer Academic Publishers.

Pacyna, J. and Münch, J. (1988). Atmospheric emissions of arsenic, cadmium, mercury and zinc in Europe in 1982. Norwegian Institute for Air Research, NILU OR: 17/88.

Pacyna, J., Semb, A., Hanssen, J. E. (1984). Emission and long-range transport of trace elements in Europe. *Tellus*: **36B:** 163–178.

Sehmel, G. A. (1980). Particle and gas dry deposition: a review. *Atmospheric Environment,* **14**, 983–1011.

Sverdrup, H., de Vries, W., Henriksen, A. (1990) Mapping critical loads. Nordic Council. NORD 1990:98.

TA-Luft (Technische Anleitung zur Reinhaltung der Luft). (1983). Allgemeine Verwaltungsvorschrift zur Änderung der Ersten Allgemeinen Verwaltungsvorschrift zum Bundesimmissionsschutzgesetz. *Gemeinsames Ministerialblatt,* 34–94.

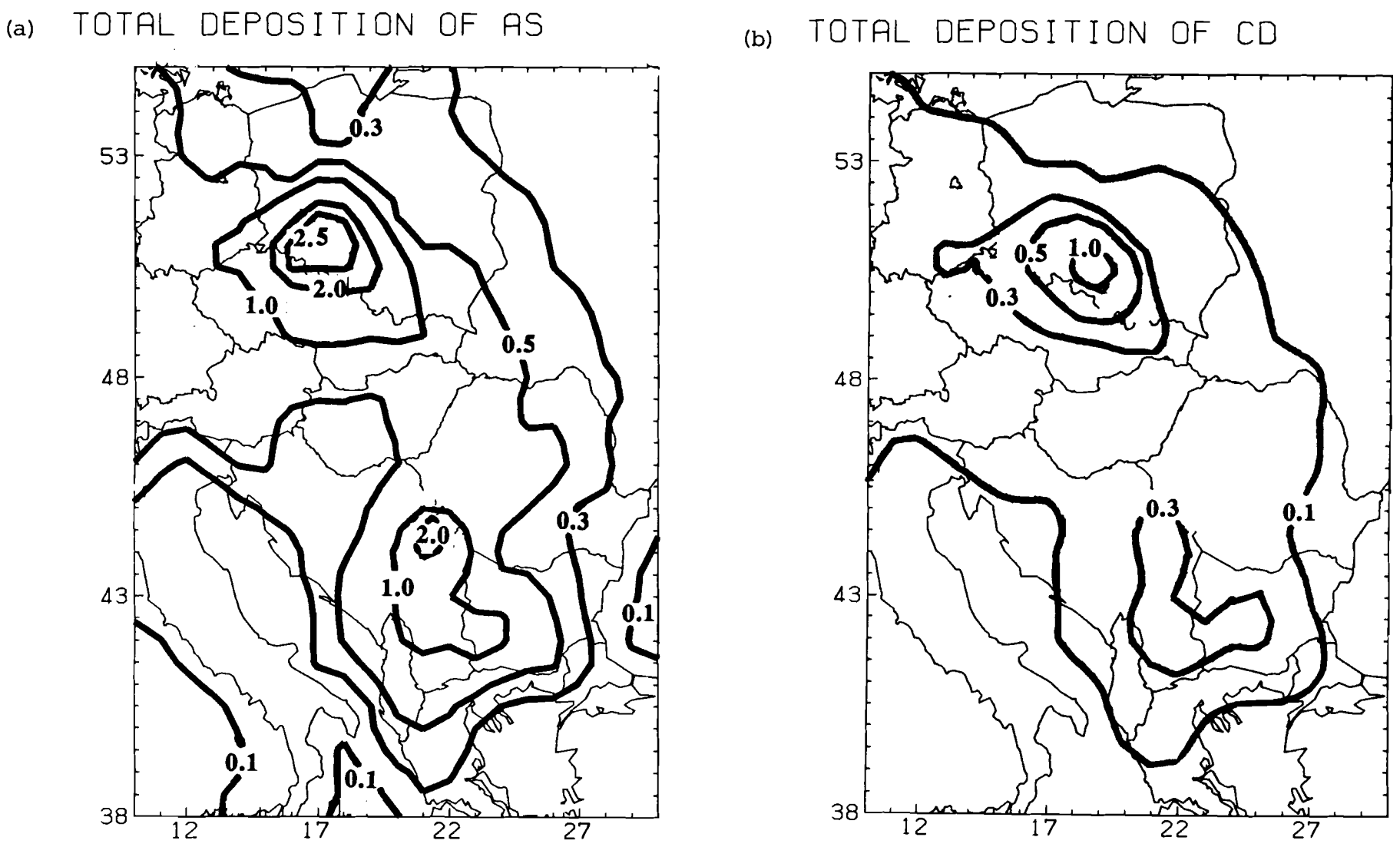
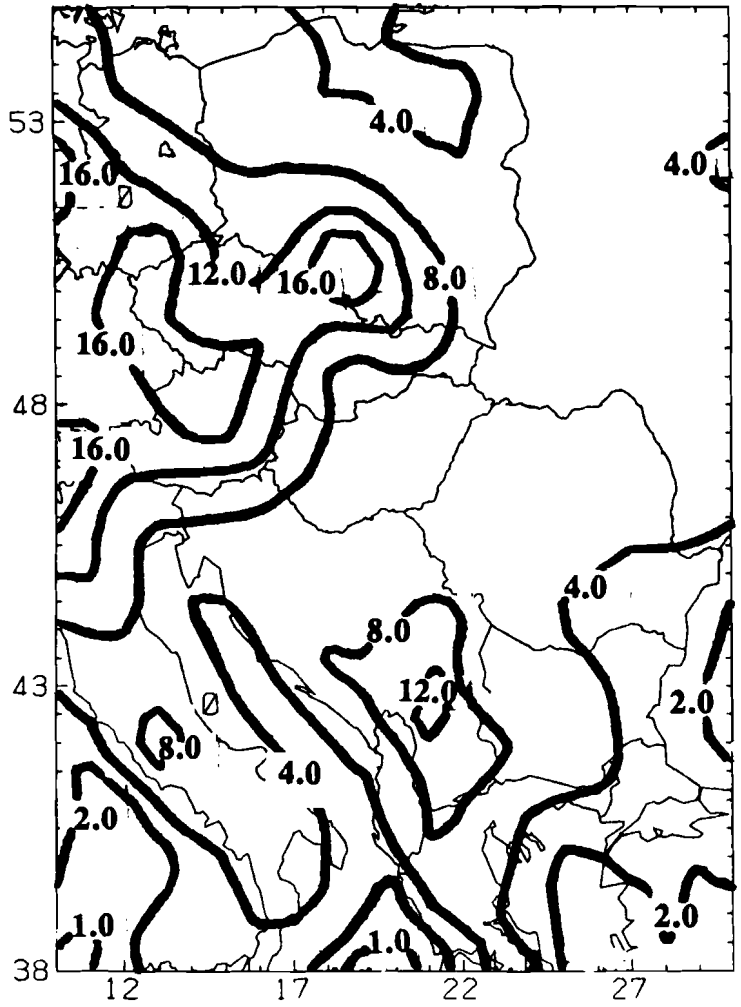


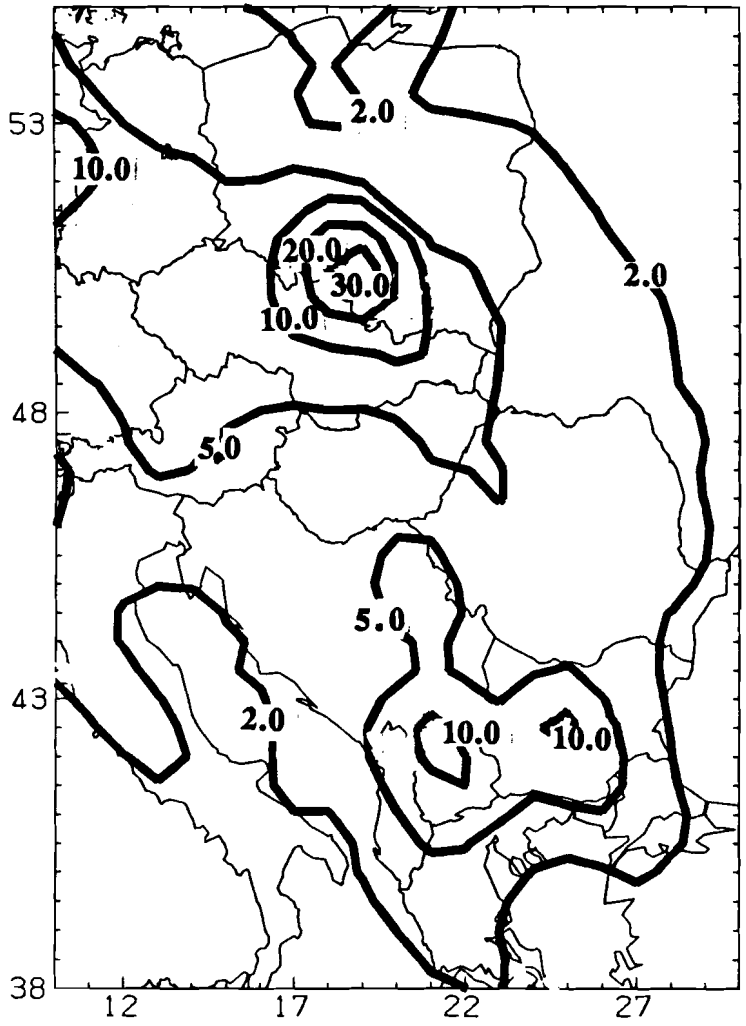
Figure 1. Total (wet plus dry) annual deposition of heavy metals, mid 1980s: (a) Arsenic, (b) Cadmium, (c) Lead, (d) Zinc.

Units: mg m^{-2} yr^{-1}

(c) TOTAL DEPOSITION OF PB



(d) TOTAL DEPOSITION OF ZN



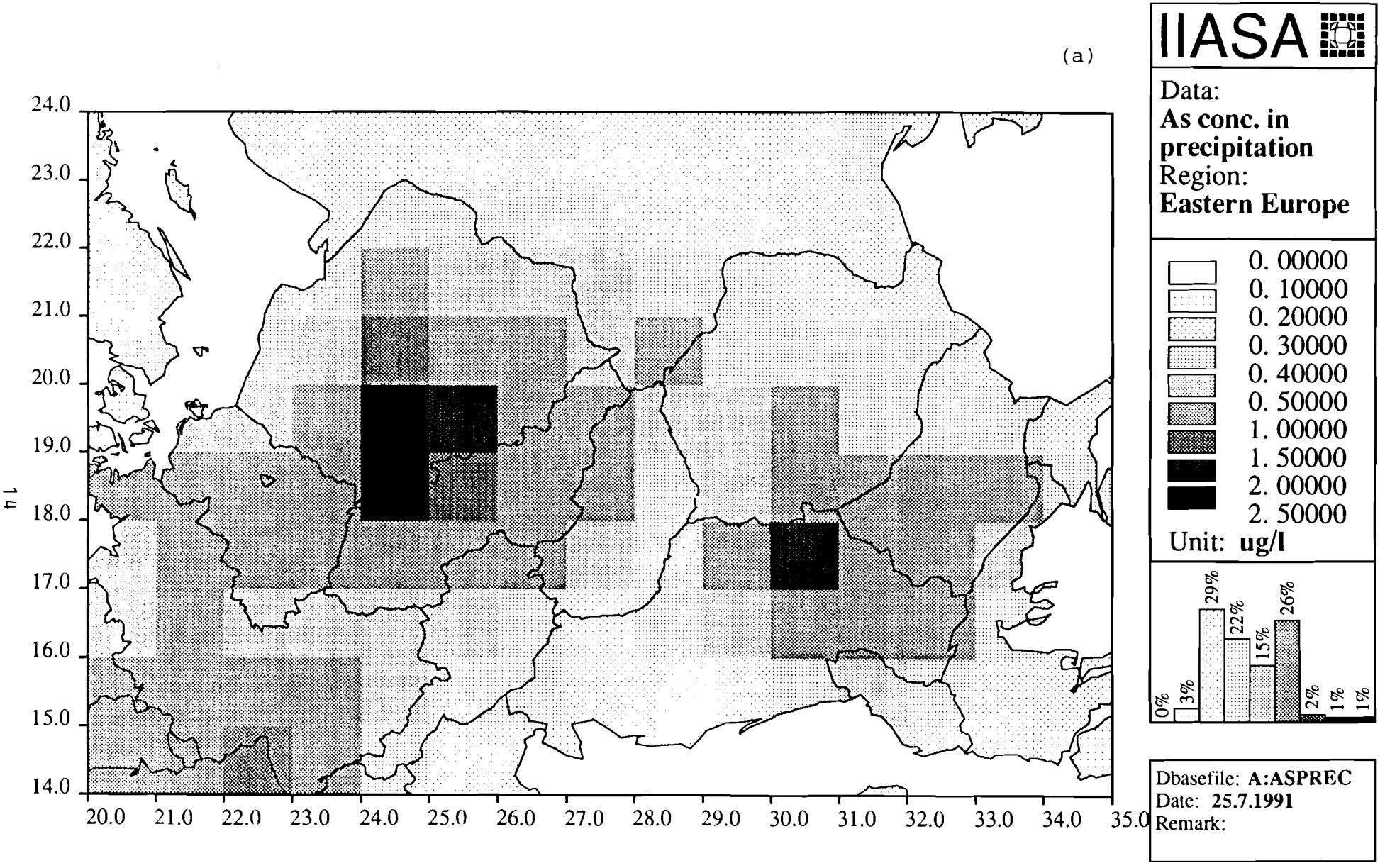
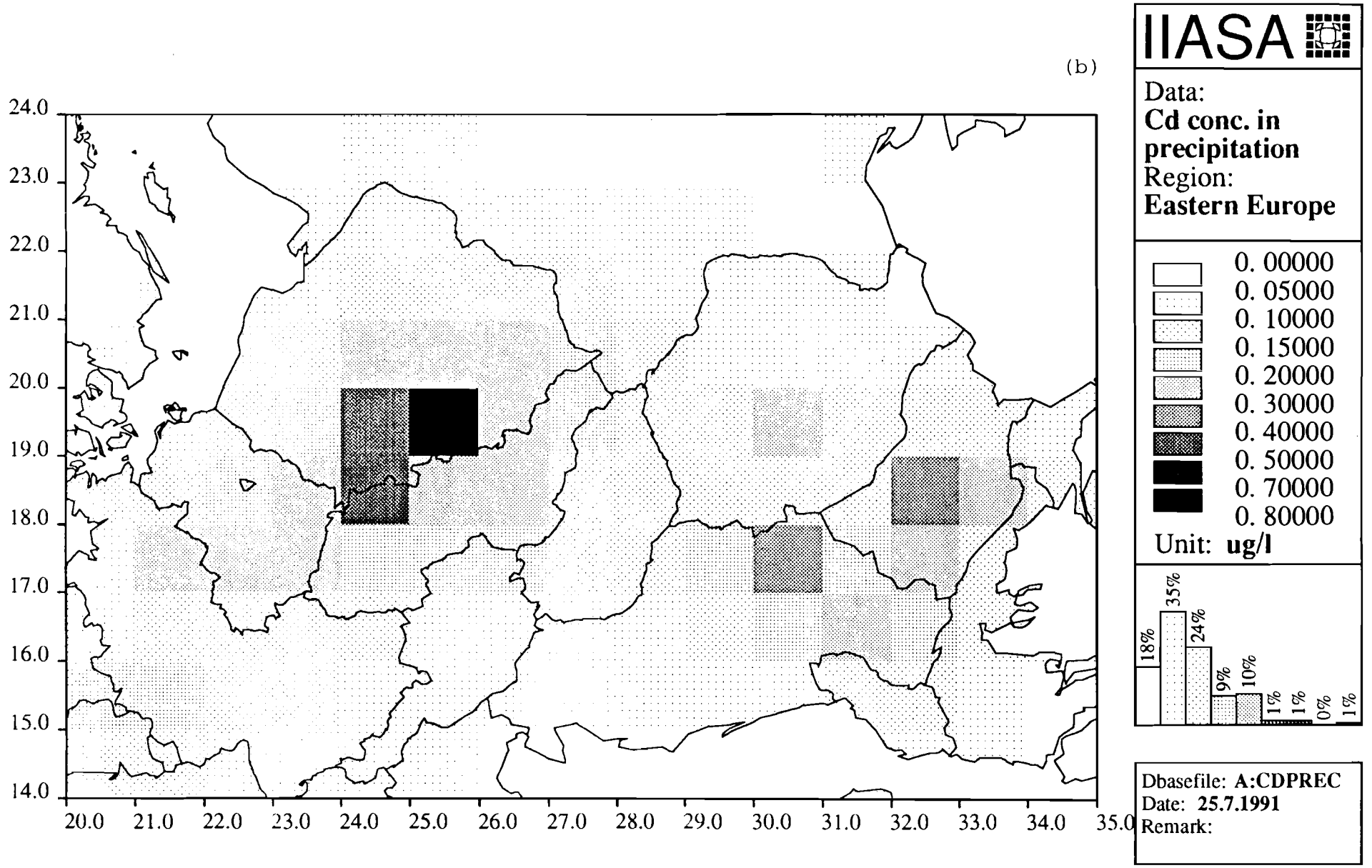
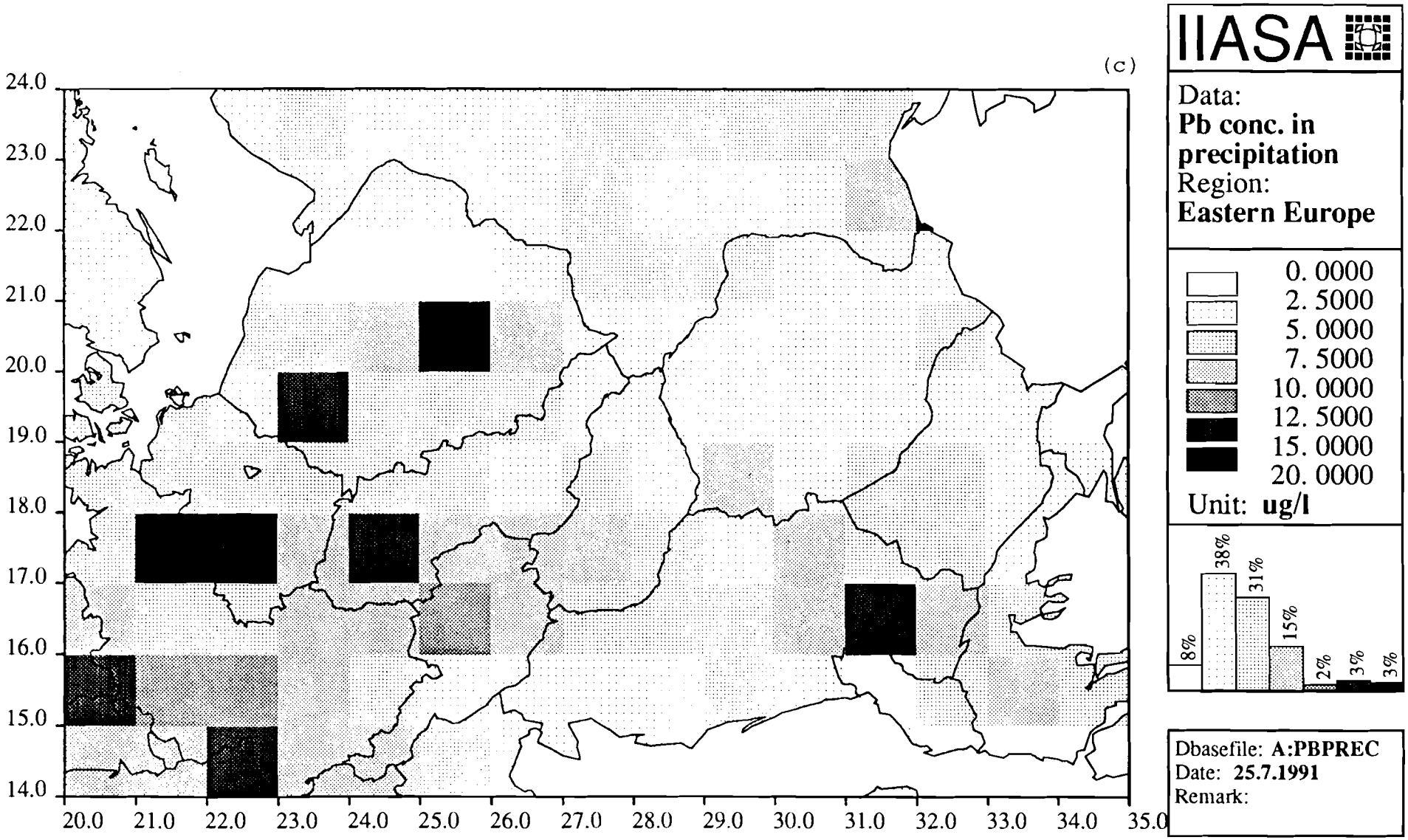


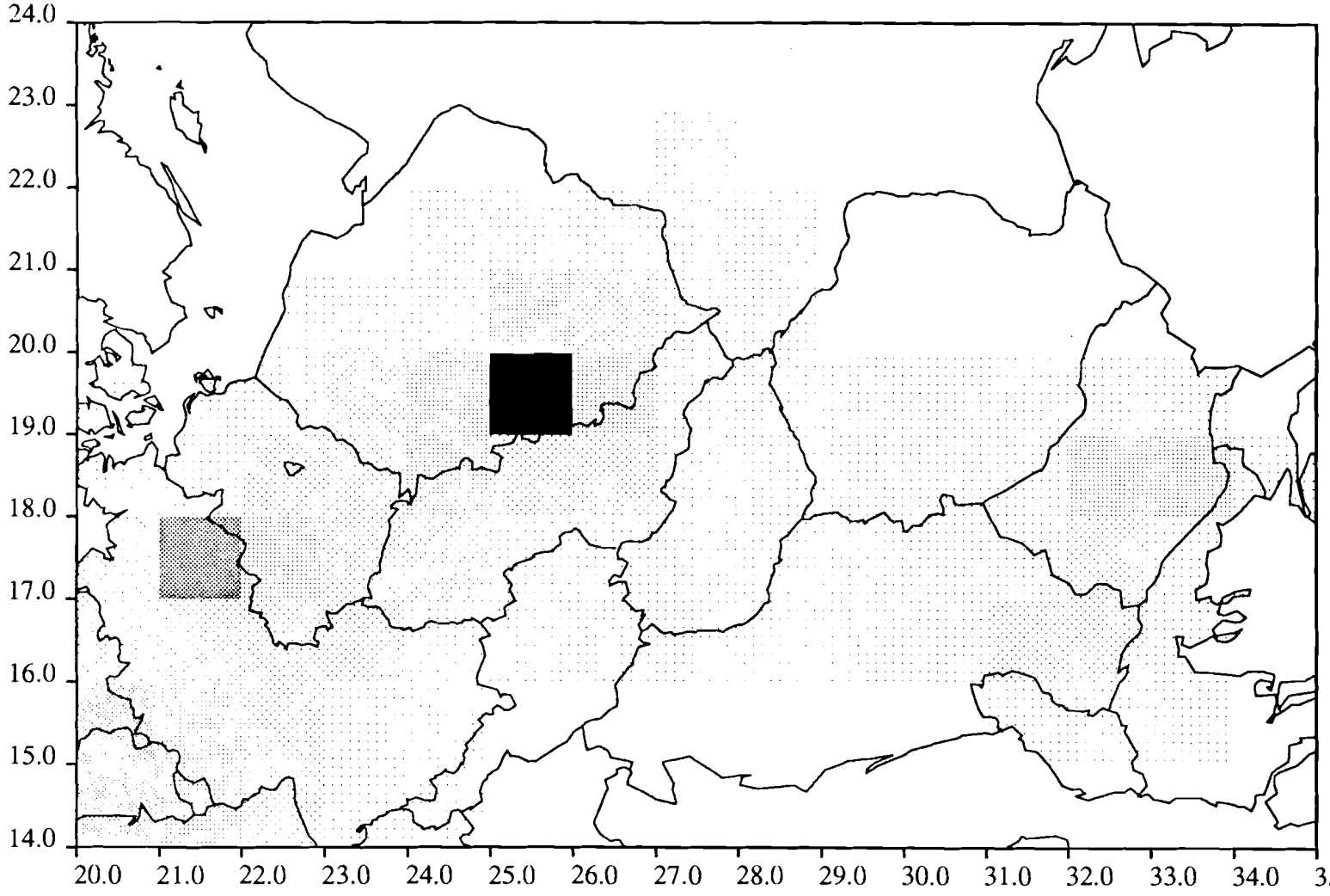
Figure 2. Annual average concentration of heavy metals in precipitation, mid 1980s: (a) Arsenic, (b) Cadmium, (c) Lead, (d) Zinc.

Units: $\mu g\ l^{-1}$


(b)
IIASA
Data:
Cd conc. in precipitation
Region:
Eastern Europe
0. 00000
0. 05000
0. 10000
0. 15000
0. 20000
0. 30000
0. 40000
0. 50000
0. 70000
0. 80000
Unit: ug/l
18%
35%
24%
9%
10%
1%
1%
0%
1%
Dbasefile: A:CDPREC
Date: 25.7.1991
Remark:
24.0
23.0
22.0
21.0
20.0
19.0
18.0
17.0
16.0
15.0
14.0
20.0
21.0
22.0
23.0
24.0
25.0
26.0
27.0
28.0
29.0
30.0
31.0
32.0
33.0
34.0
35.0


(c)
24.0
23.0
22.0
21.0
20.0
19.0
18.0
17.0
16.0
15.0
14.0
20.0
21.0
22.0
23.0
24.0
25.0
26.0
27.0
28.0
29.0
30.0
31.0
32.0
33.0
34.0
35.0
IIASA
Data:
Pb conc. in precipitation
Region:
Eastern Europe
0. 0000
2. 5000
5. 0000
7. 5000
10. 0000
12. 5000
15. 0000
20. 0000
Unit: ug/l
8%
38%
31%
15%
2%
3%
3%
Dbasefile: A:PBPREC
Date: 25.7.1991
Remark:

(d)

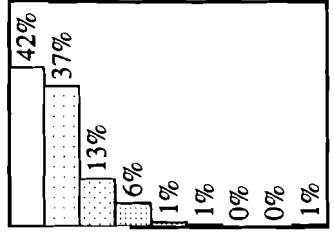


IIASA

Data:
Zn conc. in precipitation
Region:
Eastern Europe

0. 0000
2. 5000
5. 0000
7. 5000
10. 0000
12. 5000
15. 0000
17. 5000
20. 0000
22. 5000

Unit: **ug/l**



Dbasefile: **A:ZNPREC**
Date: **25.7.1991**
Remark:

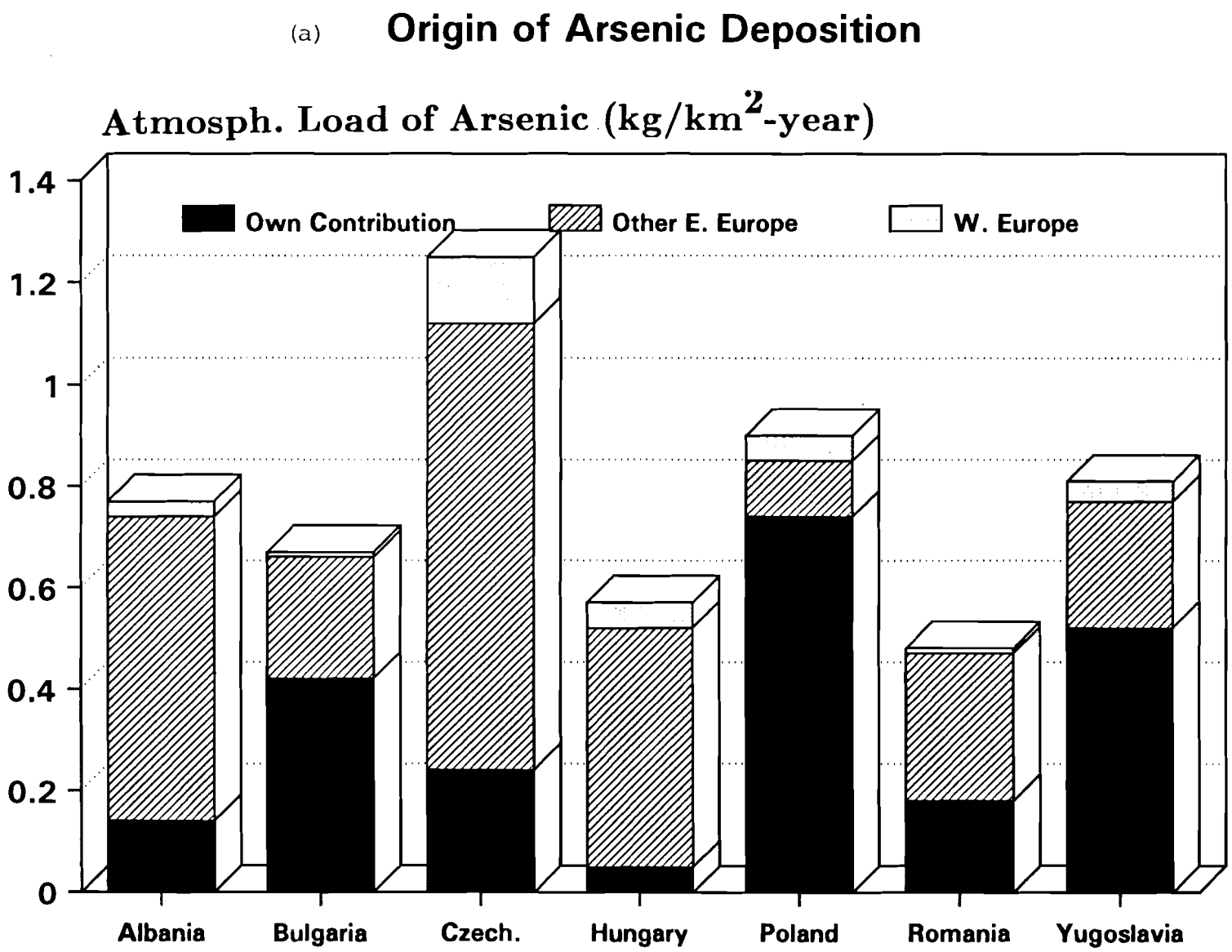
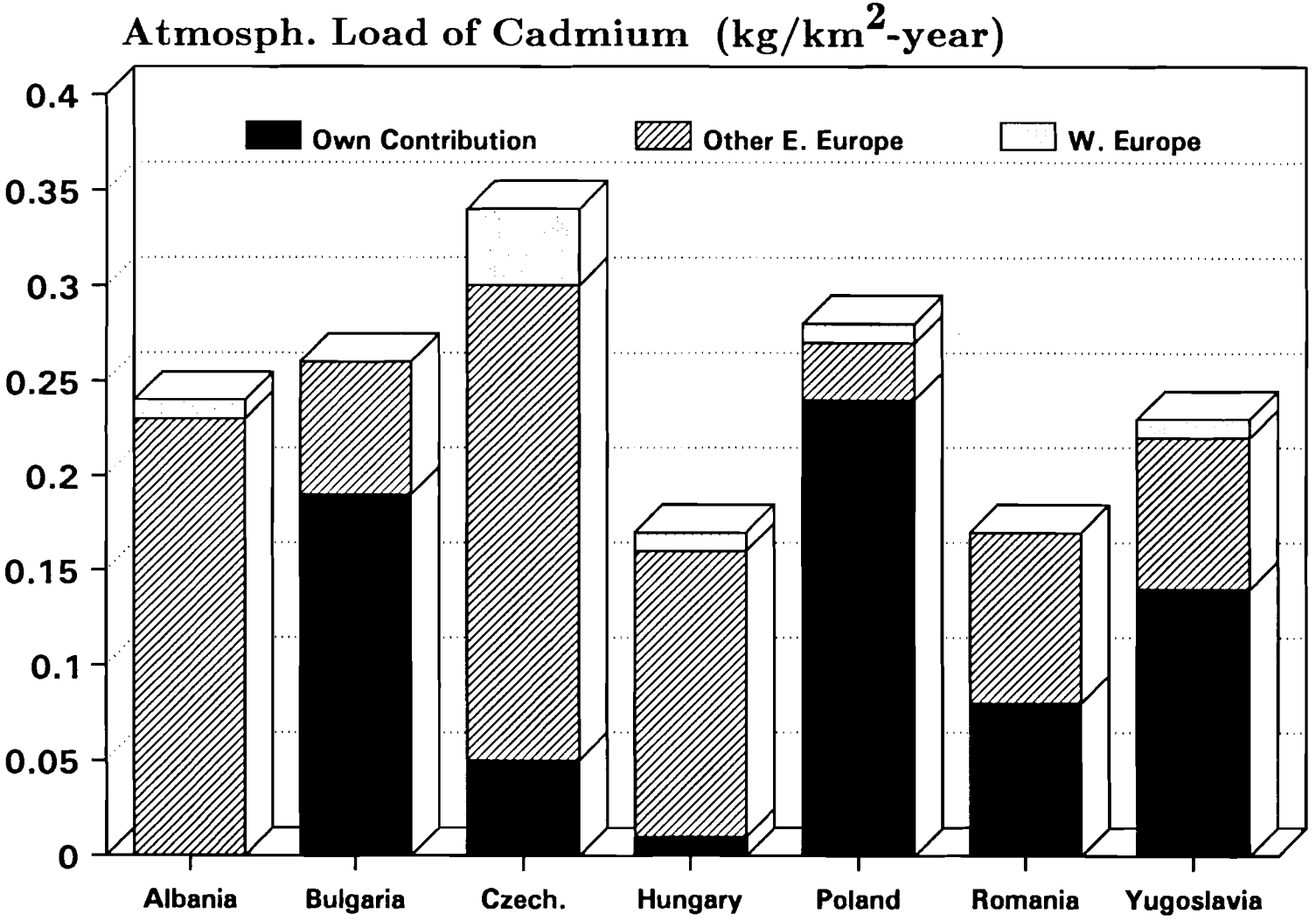
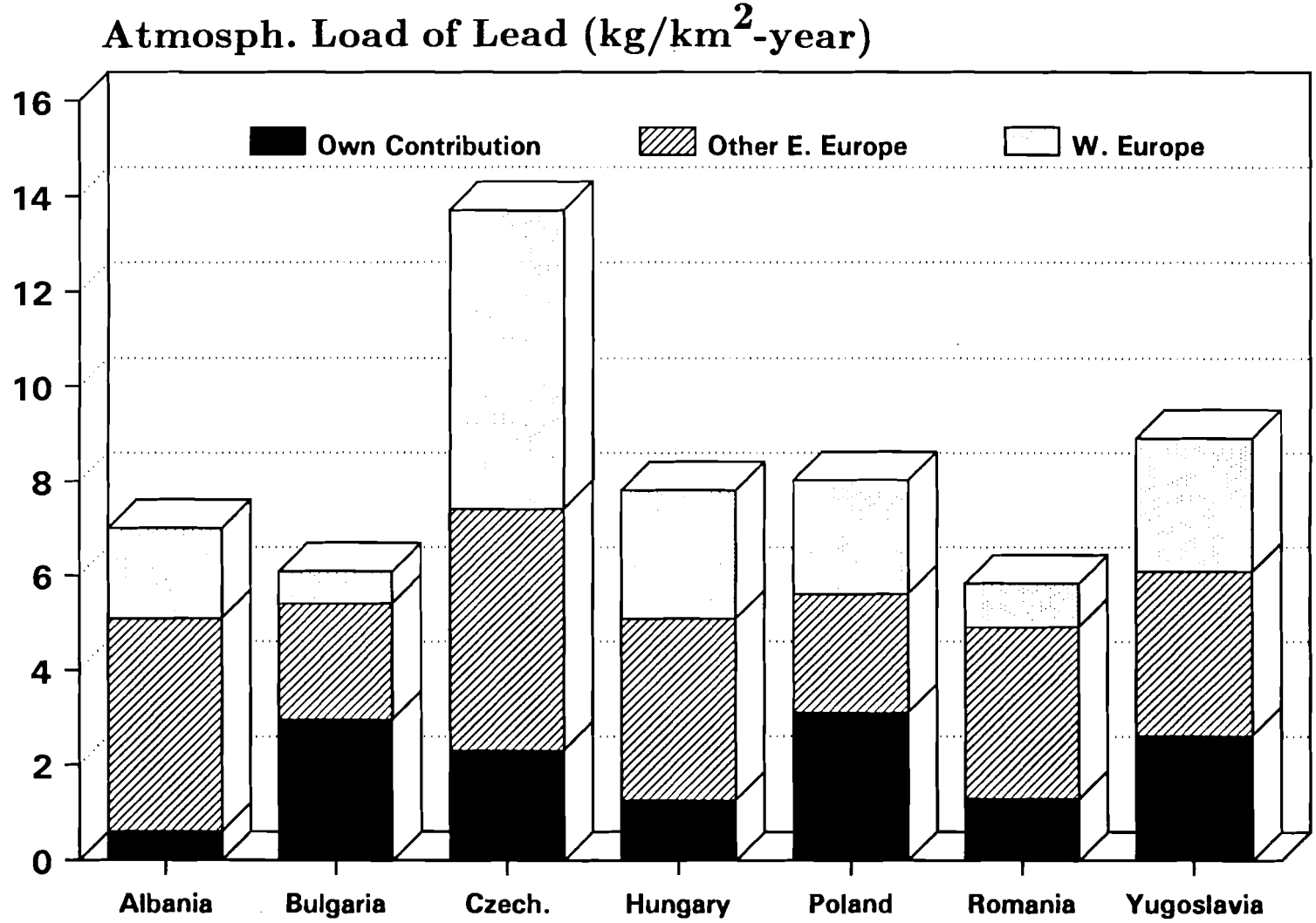


Figure 3. Country Budgets of heavy metals for various Eastern European countries. (a) Arsenic, (b) Cadmium, (c) Lead, (d) Zinc. Units: kg km^{-2} yr^{-1}

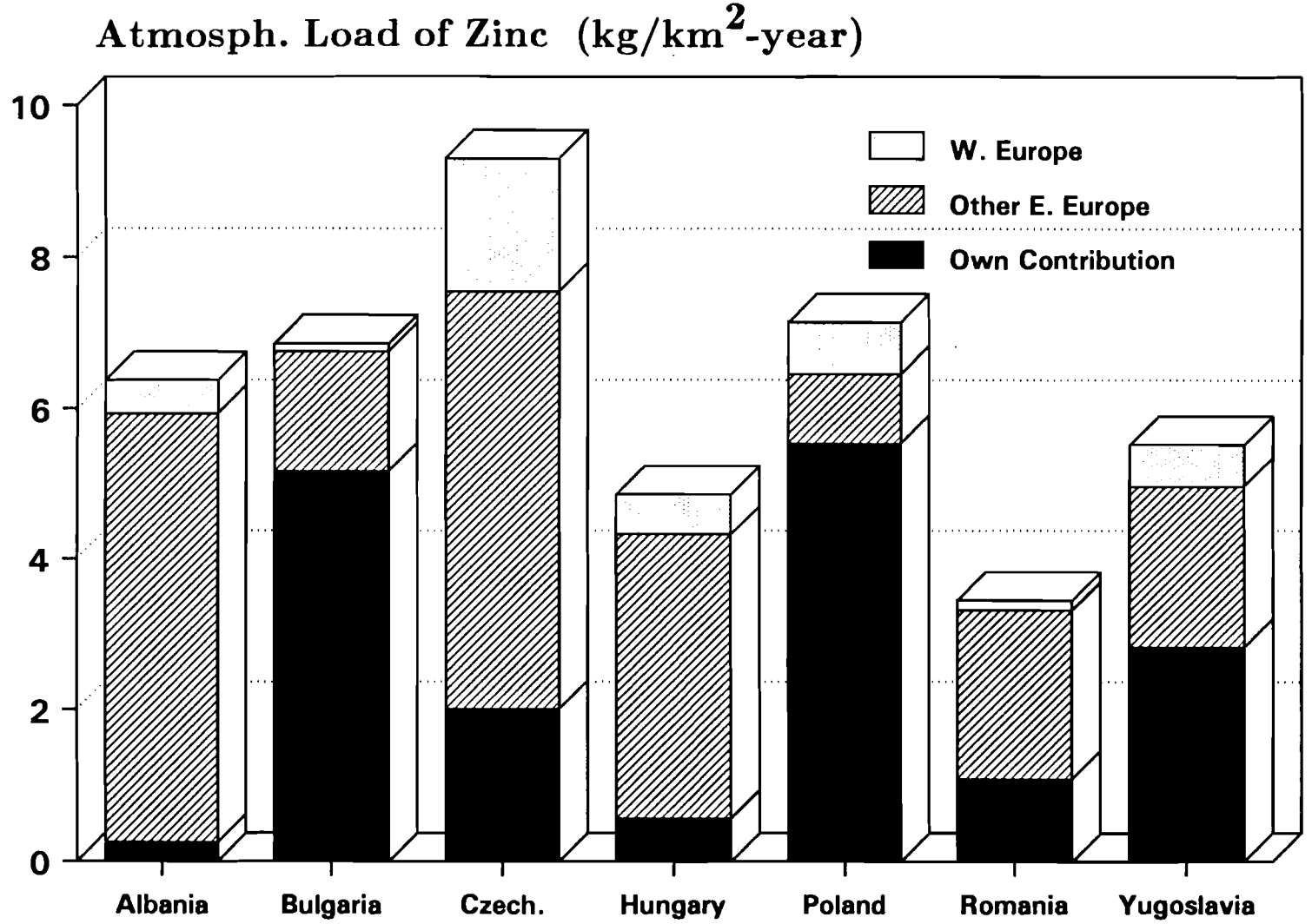
(b) **Origin of Cadmium Deposition**



(c) Origin of Lead Deposition



(d) Origin of Zn Deposition



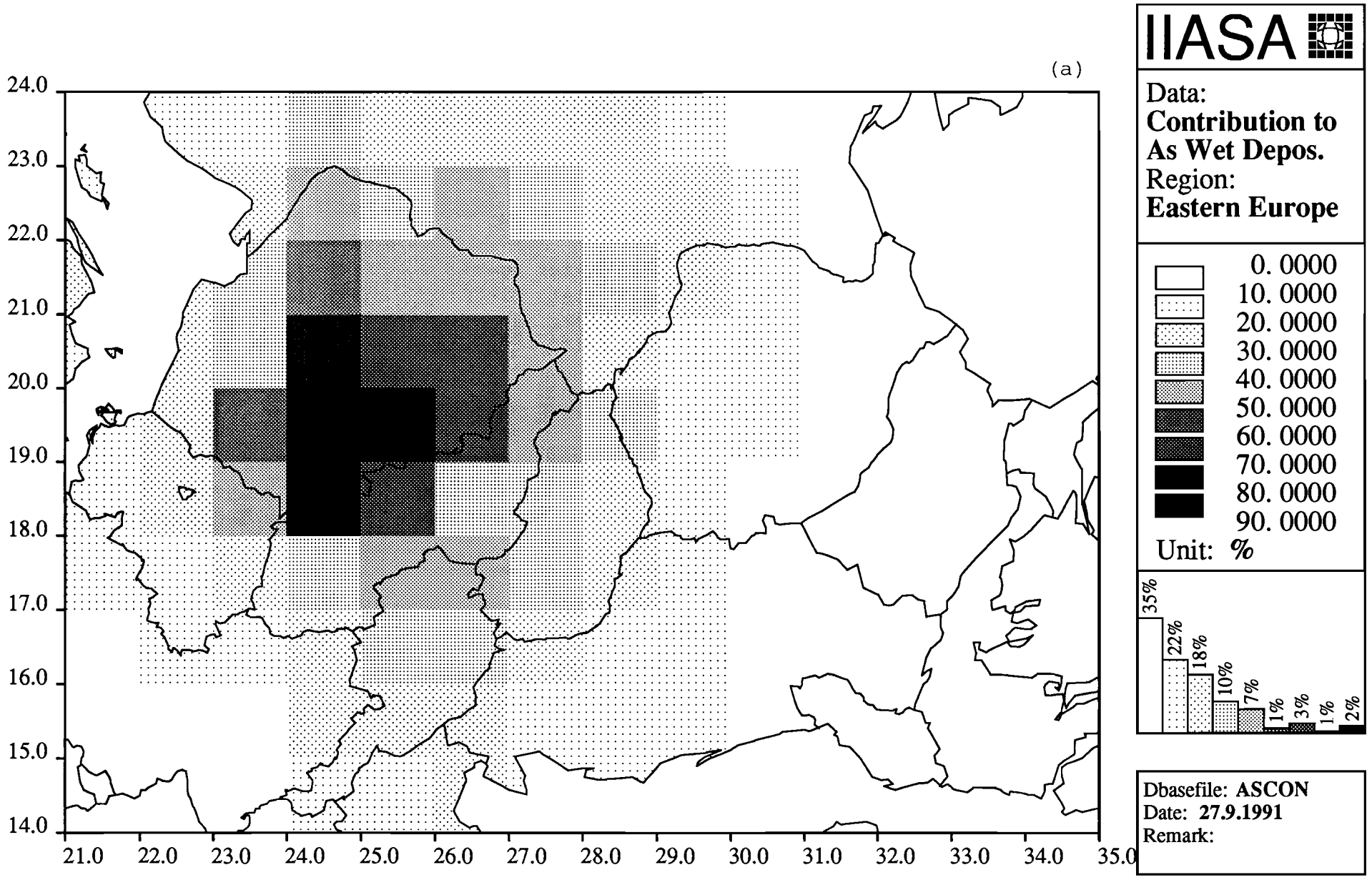
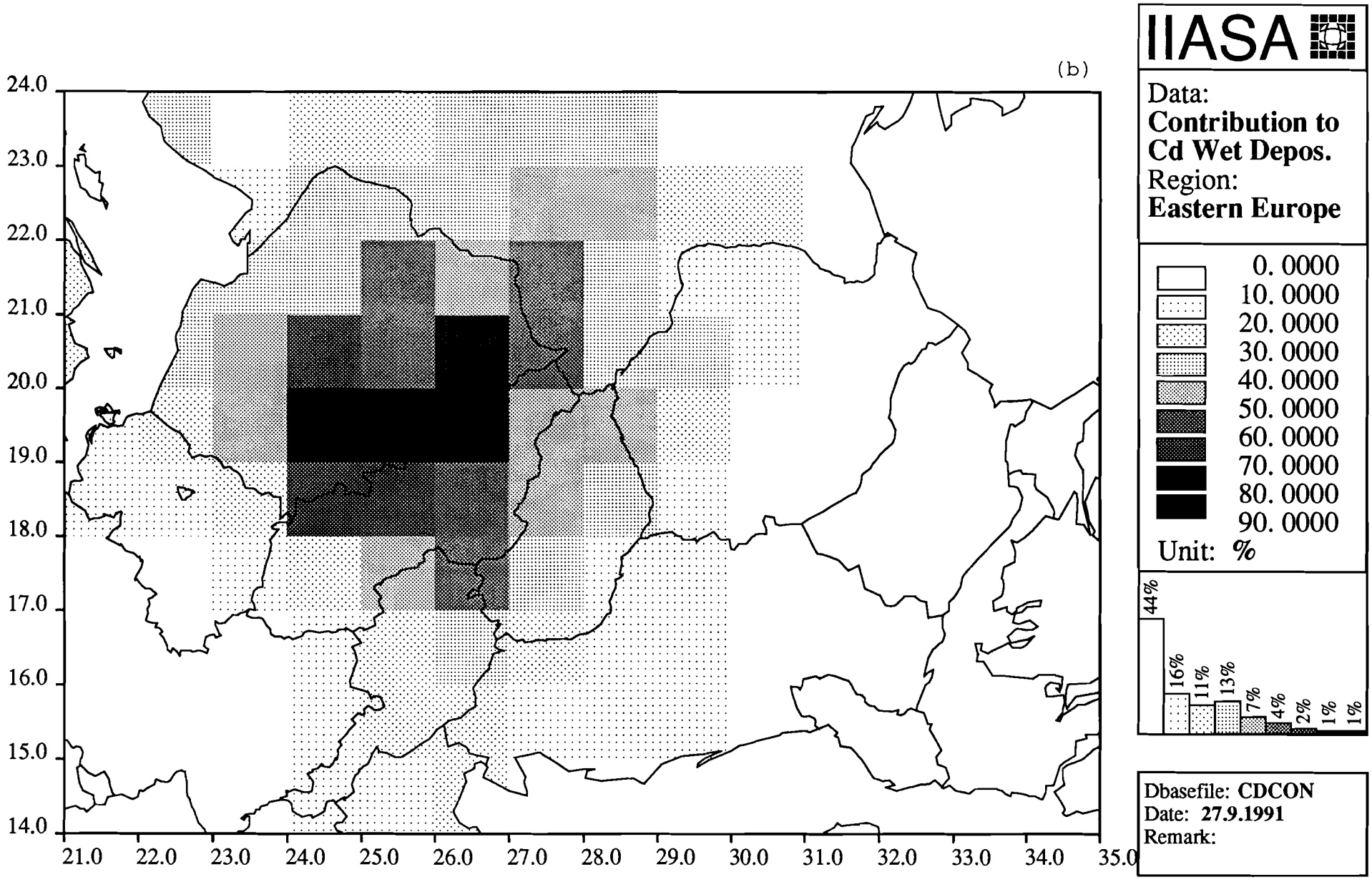
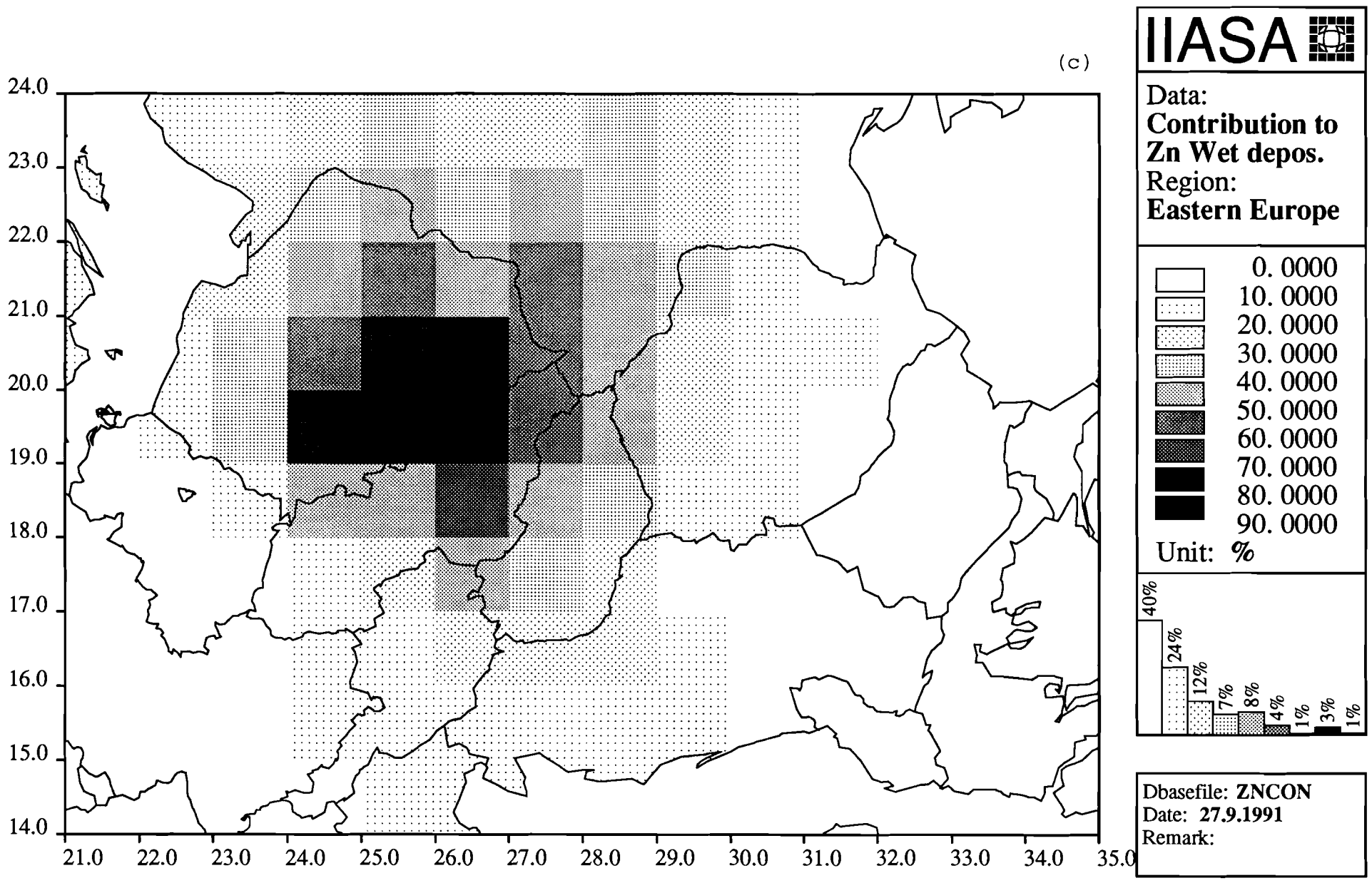


Figure 4. Contribution of Southern Polish sources to wet deposition of heavy metals in Central and Eastern Europe.
(a) Arsenic, (b) Cadmium, (c) Zinc. Units: %


(b)
IIASA
Data:
Contribution to
Cd Wet Depos.
Region:
Eastern Europe
0. 0000
10. 0000
20. 0000
30. 0000
40. 0000
50. 0000
60. 0000
70. 0000
80. 0000
90. 0000
Unit: %
44%
16%
11%
13%
7%
4%
2%
1%
1%
Dbasefile: CDCON
Date: 27.9.1991
Remark:
24.0
23.0
22.0
21.0
20.0
19.0
18.0
17.0
16.0
15.0
14.0
21.0
22.0
23.0
24.0
25.0
26.0
27.0
28.0
29.0
30.0
31.0
32.0
33.0
34.0
35.0


(c)
IIASA
Data:
Contribution to
Zn Wet depos.
Region:
Eastern Europe
0. 0000
10. 0000
20. 0000
30. 0000
40. 0000
50. 0000
60. 0000
70. 0000
80. 0000
90. 0000
Unit: %
40%
24%
12%
7%
8%
4%
1%
3%
1%
Dbasefile: ZNCON
Date: 27.9.1991
Remark:
24.0
23.0
22.0
21.0
20.0
19.0
18.0
17.0
16.0
15.0
14.0
21.0
22.0
23.0
24.0
25.0
26.0
27.0
28.0
29.0
30.0
31.0
32.0
33.0
34.0
35.0