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DISTRIBUTION OF CHLORINATED HYDROCARBONS IN DIFFERENT ECOSYSTEMS IN GERMANY*

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The German Environmental Specimen Bank (GESB) was established in **1985** as a permanent institution for the systematic collection, processing, characterization and storage of representative environmental samples from different ecosystems throughout Germany. Immediately after the reunification a special sampling campaign was performed in representative areas of the former GDR focusing on the assessment of the environmental pollution situation. These samples can be regarded as a basis for monitoring the development of spatial and temporal trends in Eastern Germany. Selected results about the determination of chlorinated hydrocarbons (CHC) in various bioindicators (earthworms, pigeon eggs, poplar leaves, pine shoots, breams) are presented. Particular emphasis is given to the comparsion of the CHC burden in East and West Germany with respect to different contamination sources.

Keywords: Environmental specimen banking; chlorinated hydrocarbons; accumulation indicator; biomonitoring; East Germany

INTRODUCTION

The German Environmental Specimen Bank (GESB) is a systematic established archive of frequently collected representative environmental specimens and human organ samples. Because of extreme low storage temperature $(T = < -150^{\circ}C)$ it is guaranteed that the samples are not subject to chemical

^{*} **A** contribution to the Federal Environmental Specimen Bank

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changes during the long-term storage. It has been successfully established as a permanent environmental surveillance tool in Germany $[1,2]$. The banking activities are focused on the preparation, characterization and storage of representative samples from different ecosystems in Germany. The GESB program started in 1985. Since that time extended sampling campaigns have been performed for a number of species in all areas selected for the GESB. Based on reliable and well-documented analytical procedures the obtained monitoring results offer the opportunity for long-term control of environmental pollution by spatial differences and time-dependent trends. **A** number of organic and inorganic compounds are regularly determined in the samples. The further discussion is restricted to chlorinated hydrocarbons (CHC).

Numerous chlorinated insecticides and industrial CHC *(e.g.* polychlorinated biphenyls (PCB) or polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDDR)) are extremely resistant to degradation in the environment. Residues of these xenobiotics have been identified throughout the world although most of them have been banned since the 70s. Because of their toxicological properties and accumulation effects, long-term studies on their residue levels are essential to understand the environmental contamination in the past and to predict future trends.

The former German Democratic Republic (GDR) was one of the major producers of elemental chlorine in East Europe. Due to economical reasons the production and application of low volatile CHC played a more important role in the chemical pest control in agriculture and forestry of East Germany than in the Western countries $\left[3\right]$. Under the pressure of worldwide application restrictions the production has ceased since the early 70s (DDT) or late **80s** (lindane, toxaphene) respectively. Thus, the different economic and technical development of the two German states led to partly different patterns of environmental pollution.

Reliable data of the pollution situation, especially of the instrustrialized areas, in the former GDR were not available on account of the strict political restriction regime. In order to detect the efficiency of legislative regulations for the environmental redevelopment, which were initiated directly after the reunification, and for the assessment of the environmental pollution a special sampling campaign was performed in representative areas of the former GDR in 1990/91^[4]. These samples can be regarded as a basis for monitoring the development of spatial and temporal trends in Eastern Germany. The aim of the present work was the characterization of the CHC burden of the former GDR in various selected environmental matrices representing the terrestrial, aquatic, and atmospheric ecosystems according to the quality assurance system of the GESB. The results obtained within this study provide comparable baseline data that can be used to plan and manage further banking activities **[51.** This report describes local and regional variations of the CHC contamination in East and West Germany and **an** attempt to evaluate possible sources.

MATERIALS AND METHODS

Chemicals

All solvents (residue analysis grade) were purchased from Promochem (Wesel, Germany). All standards (purity > 98 %) were obtained from **Dr.** Ehrenstorfer (Augsburg, Germany). Sodium sulfate (p.a.) and seasand were from Merck (Darmstadt, Germany). BIO-Beads **S-X 8** (200-400 mesh) was supplied from Bio-Rad Labs (Richmond, USA), and Hypersil silicagel from Bischoff Chromatography (Leonberg, Germany). The individual compounds investigated and their abbreviations are listed in Table I.

TABLE I Compounds investigated

Sampling and sample preparation

In 1990/91, immediately after the reunification, various specimens were taken in the former GDR according to the guidelines of GESB **[4,61.** Sampling was performed in the lake area of Mecklenburg representing agricultural ecosystems, in the urban instrustrialized region of the Diibener Heath (an area between Leipzig/Halle/Bitterfeld) representing terrestrial ecosystems and in the River Elbe as an example of a limnic ecosystem in East Germany (Figure 1).

The other results reported in this paper were obtained within the GESB routine program $[7-9]$. Directly after collection the samples were prepared and deep-frozen on site in the gaseous phase over liquid nitrogen. After that the cyrogenic chain (T<-150 $^{\circ}$ C) is not interrupted including the final storage at the GESB facility in Jiilich. Most of the material was ground, homogenized, divided into powder subsamples (10 g) and stored in the bank until processing. For the CHC determination either the frozen single sample or the powder subsample were mixed with anhydrous sodium sulfate/seasand to form a free-flowing product which was extracted with n -hexane/acetone in an extraction column. The majority of the lipid and biogenic material was removed from the raw extract using size exclusion chromatography (column of variable length, ID: 127 mm, stationary phase: BIO-Beads **S-X** 8, mobile phase: cyclohexane/ethylacetate (v/v; 1: 1) at 2 mL/min). Afterwards the collected pesticide fraction was precleaned by a HPLC step (column length: 24 cm, ID: 8 mm, stationary phase: Hypersil silicalgel, 5 μ m, mobile phase: 0.1 % propanol-2 in *n*-hexane at 4 mL/min) to eliminate the major part of polar impurities that might be present. Determination was carried out by high resolution gas chromatography (HRGC) with electron capture detection using two columns of different polarity (DB-5 or DB-1701, respectively, column length: 30 m , ID:0.32 mm, carrier gas: H_2 , injection temperature: 230 °C, splitess injection, temperature programm: $60-120$ °C at 8 °C/min, then 120-260 °C at 4 °C/min and further at 10 °C/min to 280 °C, 10 min isothermal). The quantification evaluation was performed with two internal standards (pentachlorotoluene and decachlorobiphenyl). More information about the analytical method can be found elsewhere **[Io1.** For pine needles a different analytical procedure was applied due to the high amount **of** co-extracted impurities. The n-hexane/acetone extract was purified by chromatography on florisil (4 % water content, length: 30 cm, ID: 1.5 cm, height: 20 cm) eluating with n-hexane. CHC analysis was performed by HRGC equipped with a mass spectrometric detector (DB-5, column length: 30 m, ID: 0.32 mm, carrier gas: He, injection temperature: 230 °C, splitess injection, temperature program: 60-140 °C at 12 °C/min, then 140–210 °C at 5 °C/min and further at 8 °C/min to 250 °C). CHC concentrations were calculated by isotope dilution with 13 C labeled CHC standards. More details about the applied method are given in ^[11].

FIGURE 1 **Sampling areas of the special GESB sampling campaign in 1990/91**

RESULTS AND DISCUSSION

The DDT, HCH and PCB group were the major organochlorine contaminants present and therefore the further discussion is predominately focused on these pollutant groups.

Earthworms

Earthworms are virtually ubiquitous and ecologically important soil organisms. **As** destruents they are intensively involved in the nutrient cycle and exposed to pollutants [12,131. Deeply digging earthworms *(Lumbricus terrestris, Aporrectodea longa)* represent the only bioindicator of the GESB, which reflect the biologically available part of soil contamination in terrestrial environments.

A comparison of the 4,4'-DDT/DDE data of earthworms *(Lumbricus terrestris)* from typical agricultural and urban industrialized ecosystems in East and West Germany in 1990/91 is presented in Figure 2.

Relatively low concentrations were detected in samples from the Bornhoveder lake area and the Saarland, both situated in West Germany. The corresponding 4,4'-DDT/DDE levels in earthworms from Mecklenburg (agricultural ecosystem) and the Halle/Leipzig region (urban industrialized ecosystem) were more than a factor of 10 higher. This observation clearly demonstrates the importance of DDT in the former GDR. DDT was the most heavily used agricultural insecticide in East Germany due to its low production cost and high efficiency $[3]$. In 1971 the DDT application was gradually reduced, but it was still used in the forest pest control until the mid 80s **[14].** In contrast to the DDT burden the PCB 138 levels of earthworms *(Aporrectodea longa)* from Saarland was generally twice as high as those found in samples from Leipzig/Halle (Figure 3).

The earthworm species *Aporrectodea longa* is known to accumulate PCB at higher concentrations compared to the *Lumbricus terrestris* species and hence give a better indication of PCB contamination in their surroundings. In contrast to many Western countries the PCB consumption of East Germany was relatively small (20 000 t between 1955-1985) resulting from the dependence on PCB imports from the former CSFR since 1964 **[I5].**

Pigeon eggs

Pigeons *(Columba livia f. domestica)* have a widespread distribution in urban areas of Europe. Due to their herbivorous feeding habits, they represent the higher trophic level in the food chain and accumulate pollutants in higher

FIGURE 2 Comparison of the 4.4'-DDT and DDE mean concentrations in earthworms hornogenates *(Lurnbricus krrestris)* from different ecosystems of East and West Germany in **1990/91** (WW: wet weight)

amounts without being killed or rendered incapable of reproduction. For these reasons pigeon eggs may be a suitable bioindicator for real-time and long-term monitoring of chemical substances. The egg contents of a complete breeding season are processed to a blended bulk sample.

FIGURE 3 Comparison of the **FCB 138** mean concentrations in earthworms homogenates *(Aporrect*odea longa) from different ecosystems of East and West Germany in 1990/91 (WW: wet weight) (Abbr.: cf. Fig. 2, V-HO: Volklingen-Hostenbach)

y-HCH, DDE and PCB 138 concentrations in the pigeon eggs from Leipzig and the urban area of Saarland are shown in Figure **4.**

The analytical data demonstrate a pollutant pattern similar to that found in earthworms. Elevated DDE levels were found in eggs from Leipzig compared to the concentrations in samples from Saarland. The opposite trend was observed for PCB contamination. The significantly higher concentrations of DDE (mean: 192 ng/g wet weight) and y-HCH (mean: **11** ng/g wet weight) in pigeon eggs from the sampling site Leipzig-Stadthaus could probably be explained by the local application of DDT containing Lindane preparations e.g. in the loft. Both active substances were formulated together and traded on the market as wood preservatives (e.g. Hylotox) **[I6].** The city archive of Leipzig is located in the Stadthaus.

Poplar leaves and pine shoots

Among conifers, Scots pine *(Pinus sylvestris)* is a species which occurs in Central and North Europe with poor habitat requirements. For more than 100 years pines have been useful sensitive indicators for both emission and effect control

FIGURE 4 Comparison of the mean concentrations of selected CHC in pigeon eggs homogenates *(Columba livia f: domestica)* **from urban industrialized ecosystems** of **East and West Germany in 1991 (WW: wet weight)**

[17]. The residue levels of organic pollutants in pine needles reflect predominantly the gaseous pollutant load, particle-bound dry deposition and the uptake of lipophilic compounds by roots play only a minor role **[lgl.**

Poplars *(Populus nigru "lrulica* ") are sampled as typical representative of deciduous trees in urban industrialized areas. Because of their genetic uniformity and resistance to pollution inputs they permit extended and long-term biomonitoring studies. Pine shoots as well as poplar leaves have been proven to be appropriate passive sampling systems for the regional atmospheric pollution by chemical substances. One-year-old pine shoots were collected from the upper part of the trees during a defined period (early spring). The poplar leaves were sampled from crown sections in the late summer.

DDT was the major contaminant in poplar leaves from the Leipzig/Halle/Bitterfeld region. In contrast to the animal samples the DDT values in poplar leaves were higher than the corresponding DDE levels (cf. Figure 5).

Burghausen, located in an agricultural area, exhibited the highest DDT levels (median: **10** ng/g wet weight) suggesting that the DDT source may be application of DDT remainders e.g. in allotment garden areas. The HCH inputs were espe-

TK EB KA DL LO EH BW BH Rz zs Tierklinik Elsterbecken Kanena Dolan Lochau Espenhain BitterfeldNolfen Burghausen Roitzsch Zschernitz

FIGURE *5* **Comparison of the 4,4'-DDT and DDE median concentrations in single samples of poplar leaves** *(Populus nigra "lralica")* **from the LeipzigRlallelBitterfeId area in ¹⁹⁹¹**(W **wet weight)**

cially high at the sampling site BitterfeldNolfen as can be seen in Figure *6,* thus showing the strong influence of the former chemical plant VEB Chemie-Kombinat Bitterfeld, where technical HCH and lindane were produced until 1982. In 1991 the α -HCH concentrations of Bitterfeld/Wolfen appeared to be a factor of 10 higher than those found in leaves from the other stations. High proportions of the *a-* and P-HCH indicates contamination resulting from HCH production **or** waste dumps **[31.**

FIGURE **6 Comparison** of the **HCH median concentrations** in **single samples** of **poplar leaves** *(Populus nigra "lralica")* **from the Leipzig/Halle/Bitterfeld area in 1991** (WW: **wet weight) (Abbr.: cf.** Figure **5)**

In contrast to HCH the former DDT production in Bitterfeld/Wolfen (until 1973) did not contribute to a higher atmospheric pollution with CHC (Figure 5).

A comparison of data from 1991-1995 obtained from the analysis of pine shoots from the Tornau district located in the Diibener Heath exhibits a significant decline of y-HCH concentration from 3.5 ng/g wet weight in 1991 to about **1.3** ng/g in 1995, i.e. a decrease by a factor of 2.5 (Figure 7). **A** similar trend was also detected for HCB, β -HCH and the DDT compounds.

Breams

Breams *(Abrumis brumu)* are freshwater fish with **a** small migration radius. They are an ideal organism for monitoring freshwater and sediment contamination because their feeding habits are characterized by permanent direct contact with the sediments [19].

One aim of the sampling campaign was the characterization of the CHC burden along the German part of the River Elbe and to monitor the expected recovery of the river. For the past 20 years the River Elbe has been one of the most polluted

FIGURE 7 Time course of **selected CHC median concentrations in single samples of pine shoots** *(Pinus sylvestris)* from the Tornau district (area of Leipzig/Halle/Bitterfeld) during 1991-92/94-95 (WW: wet weight)

rivers in Europe **[20,211.** Due to political changes in the Eastern part of Europe and especially in East Germany, various industries, who discharged their waste water into the River Elbe, were either closed or reduced their productive activities. In order to characterize local variations of the CHC burden seven stations were selected with regard to various types and magnitudes **of** industrial pollution (cf. Figure **8) [221.**

In addition, bream samples were collected in 9 lakes of Mecklenburg which were mainly influenced by agriculture, livestock fattening and forest industry. Figures 9-11 present a comparison of the main contaminants in bream muscle homogenates from the River Elbe and the lake area of Mecklenburg.

As shown in Figures 9 and 10 fish caught at the upper Elbe (Prossen and Löschwitz) exhibited the highest concentrations of DDE, OCS and PCB. This observation is probably a result of the considerable pollution of the River Elbe from industrialized areas (e.g. Pardubice, Neratovice, Usti) of the former CSFR **~31**

A significant decrease of these contaminants was found between stations Löschwitz and Vockerode. Between Vockerode and Heinrichsberg the levels remained nearly constant. The analytical data obtained for HCB concentrations

FIGURE 8 Location of the sampling stations of breams *(Abrumis bruma)* **from** the River Elbe and sites of chemical plants

in breams indicate a declining trend downstream of Barby. The high HCB contamination is considered to be an effect of the discharges from chemical plants in the former CSFR as well as of the influxes from the River Mulde (upstream from Aken) and River Saale (upstream from Barby). The increase of the HCH content in fish from these stations is also linked to the influence of both tributaries of the River Elbe (Figure 11).

The major polluters of the River Mulde were the chemical plants in the Bitterfeld/Wolfen area, although the production of chlorinated pesticides has stopped

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River Elbe		Lake area of Mecklenburg	
EP	Prossen	MW	Wanzkaer See
EL	Löschwitz	MZ.	Zwirn-See
EV	Vockerode	MS	Schweingarten-See
EA	Aken	ML	Lutowsee
	Barby	MZI	Zierker See
	Heinrichsberg	MН	Haussee
EB EH ЕC	Cumosen	MR	Rödliner See
		MT	Großer See von Teterow
		MST	Stechlin-See

FIGURE 9 Comparison of the DDE and DDD mean concentrations in bream muscle homogenates (Abramis brama) from the River Elbe and the lakes of Mecklenburg in 1990/91 (WW: wet weight)

several years ago $[3]$. The CHC burden in the River Saale originated from Buna, Leuna and the pulp mill industry. Lindane and **DDT** containing wood preserving agents (e.g. Hylotox IP/Hylotox 59) were produced in Leuna. The high contamination levels of HCH in bream at the station Heinrichsberg might be explained by the influence **of** discharges from the pesticide plant Fahlberg-List, located in Magdeburg. The results are consistent with waste water analysis of the plant and river sediments in 1990, which showed that both matrices were extremely contaminated by HCH **[241.** Although the production of lindane has ceased since **198** 1, the pesticides were highly accumulated in the river sediments.

FIGURE 10 Comparison of **the HCH mean concentrations in bream muscle homogenates** *(Abramis brama)* **from the River Elbe and** the **lakes** of **Mecklenburg in** 1990/91 (WW: **wet weight) (Abbr.: cf. Fig. 9)**

Another interesting aspect can be seen from Figure **9.** In the literature DDD is known as the dominant DDT metabolite in fish from the River Elbe **[251.** It is a degradation product of 4,4'-DDT by anaerobic conversion as well as a byproduct of the DDT synthesis. In sediments of the River Elbe and the River Mulde DDD levels were generally higher than those of DDE ^[26,27]. Interestingly, the recorded DDE values from the upstream stations (Prossen and Löschwitz) were twice as high as those of DDD. From Vockerode to Cumlosen DDD/DDE ratios ranged between 1.3-1.7. This result seems to imply that the DDD burden of sediments is well reflected by breams.

With the exception of the DDT metabolites the CHC burden **in** breams from Mecklenburg was considerably lower in comparsion with fish from the River Elbe as could be expected due to the absence of chemical industries. The DDE levels were comparable to those found in fish from the River Elbe on account of the intensive agriculture in this region. Slightly higher PCB concentrations were found in breams from the Lake Stechlin-See and the Lake Zierker See. The Lake Stechlin-See is situated in a nature reserve which is influenced by the local recreation traffic from Berlin as well as by a former nuclear power station "Rheinsberg". The elevated PCB levels in the Lake Zieker See could be attributed to the long-term influx of untreated waste water from the city Neustrelitz.

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FIGW **11 Comparison of the mean concentrations of selected CHC in bream muscle homogenates** *(Abramis brama)* **from the River Elbe and the lakes of Mecklenburg in** 1990/91 (WW: **wet weight) (Abbr.: cf. Fig.** 9)

Figures **12** and **13** illustrate results on the major CHC contaminants in bream livers from *5* different German rivers, obtained within the GESB routine program in **1995** [91. Since **1994** bream samples have been periodically taken in the River Saar and since **1995** also in the River Mulde, River Saale and River Rhine (cf. Figure **12).**

Samples were also obtained from stations along the River Elbe (Prossen, Zehren, Barby, Cumlosen, Blankenese), four stations along the River Rhine (Weil am Rhein, Iffezheim, Koblenz and Bimmen), one station situated upstream of the River Saar (Gudingen) and Rehlingen located downstream under the industrialized area of Saarbriicken. The samples from the River Mulde were collected near Dessau and fish of the River Saale between Halle and Wettin.

It is apparent that the stations on the upper Elbe still exhibited the highest concentrations of HCB, DDE and PCB. High values for HCB and DDE were also found in samples from the River Mulde. This observation is mainly a result of the former industrial emissions of the chlorine industry and a leaching dumpsite at Bitterfeld. Wilken et al. ^[28] reported, that there had been further pesticide emitters in the area upstream of the River Mulde in Saxony, especially with respect to DDT. In West Germany the HCB and DDE concentrations were much

FIGURE 12 **Comparison** of the **mean concentrations** of **selected CHC in bream liver homogenates** *(Abrarnis brama)* **from different German rivers in** 1995

lower in comparsion with the levels in East Germany. Breams caught near the Rhine station Iffezheim contained HCB at higher levels because of a former chemical plant located upstream, which produced pentachlorophenol **[291.** The highest DDE amount in fish from West Germany were found in the River Saar at Rehlingen, in contrast to its small HCB levels.

Elevated levels of PCB were detected in breams from the lower Rhine at Bimmen and from the lower Saar at Rehlingen. The increasing concentrations along the River Rhine are partly a result of the influx of the River Mosel (upstream of Koblenz), containing high amounts of PCB^[30]. The River Saar is strongly influenced by emissions from the mining industry situated in the Saar region **[311.**

The data shown in Figure 13 clearly indicate the HCH pollution of the River Mulde. Wilken et al. ^[28] reported that sediments and soils from River system Mulde are still highly contaminated by pesticides and PCDD/F although the industrial emission has stopped. These sediment-bound pollutants can be remobilized and may lead to a secondary contamination of the River Elbe by pollutant transport. Therefore, it remains to be seen how long the recovery of the River Elbe will take.

Generally HCH patterns of breams from East Germany differ from those found in fish from West Germany. The γ -isomer predominates in the Western samples, whereas higher levels of the α - and β -isomers are detected in the fish from the Eastern part of Germany.

FIGURE 13 Comparison of the mean HCH concentrations in bream liver homogenates *(Abmmis brama)* from different German rivers in 1995

The comparison **of** the CHC patterns in bream livers exhibits significant distinctions between the different ecosystems (cf. Figure 14). DDT metabolites and PCB contributed to one third each to the total CHC burden of bream livers from the limnic ecosystem of the River Elbe. PCB were the major organochlorine contaminants (70%) in breams of a typical industrialized area (Saarland) in West Germany in contrast to agricultural areas of East Germany, where DDT metabolites were the dominant pollutants (80%).

CONCLUSION

The collected specimens have been successfully utilized for the characterization of the environmental pollution by CHC in the former GDR.

The dominant position **of** the DDT group in nearly all Eastern samples clearly reflects the central role of DDT in the pest control of the former GDR. For this **CHLORINATED HYDROCARBONS** *241*

FIGURE 14 Comparison of the proportion of different CHC pollutant classes in bream livers *(Abrumis bramu)* **from East and West Germany (WW: wet weight)**

pollutant class significant spatial differences between East and West Germany were detected in terrestrial, atmospheric and limnic environments. The HCH concentrations in animal samples were considerably lower. With the exception of local hot spots of pollution the lindane contamination of both German parts was in the same range, whereas the concentrations of α - and β -HCH isomers, byproducts of the lindane production, appeared to be higher in samples from East Germany, especially in the vicinity of former pesticide plants. Relatively low PCB levels were observed in specimens from the East compared to West Germany due to low usage of PCB in the former GDR.

It can be concluded that the study provided useful analytical data for the successful implementation of representative areas of the former GDR into the GESB routine program. Furthermore, the obtained results may allow **us** to gauge the success of legislative regulatory measures that have been applied in East Germany directly after the reunification.

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