ENVIRONMENTAL VERSUS HUMAN EXPOSOMICS IN GERMAN SPECIMEN BANKING

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Introduction

Strong associations were reported between the environmental exposures and diseases[1]. According to systematically searched electronic database the publications defining the environment in the narrow sense (air, water, food, and soil pollutants) tended to have smaller attributable risk estimates, whereas publications referring to the environment in the broadest sense (including lifestyle factors, occupational exposures, and pollutants) had consistently larger risk estimates[2]. The term "exposome" of real organisms refers to the measure of all the exposures of an individual in a lifetime and how those exposures relate to disease[3]. Exposomics is the study of the exposome of humans as well as the related environments of people and circumpasses also aspects of genomics, metabonomics, lipidomics, transcriptomics, proteomics etc. Since vast majority of toxic chemicals affect critical targets inside the body, the relevant "environment" is the body's internal chemical environment[4]. However, linking endogenous and exogenous information about biomarkers of exposure will disclose relations to biomarkers of effect more properly. In addition, the exposure of humans can be estimated from environmental figures vice versa.

Especially persistent organic pollutants (POPs) have properties to estimate exposure to them more accurately. POPs once marketed are expected to remain in the environment and biota for a long time and might peak even years after their use differently in the environmental compartments and in the following generations of individuals.

The objective of the present study was to investigate the principle relationships between concentrations in human and environmental matrices of polychlorinated biphenyls (PCB) in short distance comparable areas within Germany by employing the data of the German Environmental Specimen Banking (ESB).

Materials and methods

Investigated blood plasma and pine shoots samples

Human data that encompassed blood plasma of students from the University of Halle sampled between 1995 and 2008 with an even number of female and male at the age of 20 to 29 were selected[5]. Furthermore, different environmental samples were studied such as (1) pine shoots from the nature park Dübener Heide sampled during 1993, 1995, 1997, 1999-2004; (2) and (3) egg matter of city pigeons from the residential use areas in Halle and Leipzig sampled during 1997, 2000-2008, and 2000-2008, respectively; (4) and (5) earthworm (Lumbricus terrestris) from park areas of Halle and Leipzig sampled during 2000-2008 and 1999-2008, respectively; (6) roe deer liver from nature park Dübener Heide sampled during 1995 and 1999-2008. These human and environmental data were selected due to the short distance between collected samples of approximately 60 and 40 km within nature park Dübener Heide-Halle and Leipzig - Halle, respectively. The data was given from the German Environmental Specimen Bank (ESB) (http://www.umweltprobenbank.de/en/documents)[6]. Analysis of Hexachlorobenzene (HCB) and PCB (PCB138, PCB 153, and PCB 180) were performed and the results were presented in µg/L fw, ng/g dw, ng/g fw, ng/g fw and ng/g fw for blood plasma, pine shoots, egg matter of city pigeons, earthworm and roe deer liver, respectively.

Investigated roe deer liver and spruce shoot samples

The collected in spring one-year-old spruce (*Picea abies*) shoots presents a comprehensive picture of the winter and summer pollution of the last year in the environment. These ESB spruce shoots data sampled in 1989, 1991, 1995, 1997, 1999, and 2001 was included in the study. The roe deer is the larger herbivores (first orderconsumer) found in the wild in Europe known as a bioindicator[6]. Consequently, one-year old roe deer (*Capreolus capreolus*) liver was sampled in the same years such as 1989, 1991, 1995, 1997, 1999, and 2001. All samples were collected from Warndt which is a forest ecosystem between the industrial regions of the Saarland and Lorraine. Analysis of PCB (PCB138, PCB 153, and PCB 180) was performed[6] and the results were presented in ng/g dw and ng/g fw for spruce shoots and roe deer liver, respectively.

Evaluation of the data
The pair of human and environmental data as well as spruce shoots and roe deer liver data within sampled year and compound was identified. The arithmetic mean pair data within investigated years and within compound were divided such as human/environmental concentration and spruce shoots/roe deer liver, named as bioconcentration ratios. The mean and standard deviation of resulted ratios among compounds were calculated. In order to find a relationship between these ratios the physicochemical properties for certain compounds were identified[7]. The investigated physicochemical properties were the molecular weight (MW): 284.8, 360.9, 360.9, 395 g/mol; the octanol-water partition coefficient (log K_{ow}): 5.5, 6.7, 6.8, 7.2; the Henry's law constant (K_H):139, 69, 13.4, 102 Pa m³ mol-1 at 298K, and the sorption partition coefficient (log K_{oc}): 5.8, 7.6, 7.2, 7.30 for HCB, PCB 138, PCB 153 and PCB 180, respectively. Finally, the linear correlation and slopes and y-intercepts were studied.

Results and discussion
The PCB results of blood plasma and pine shoots sampled from Halle and nature park Dübener Heide during 1995-2008, respectively are shown on Figure 1 (A).

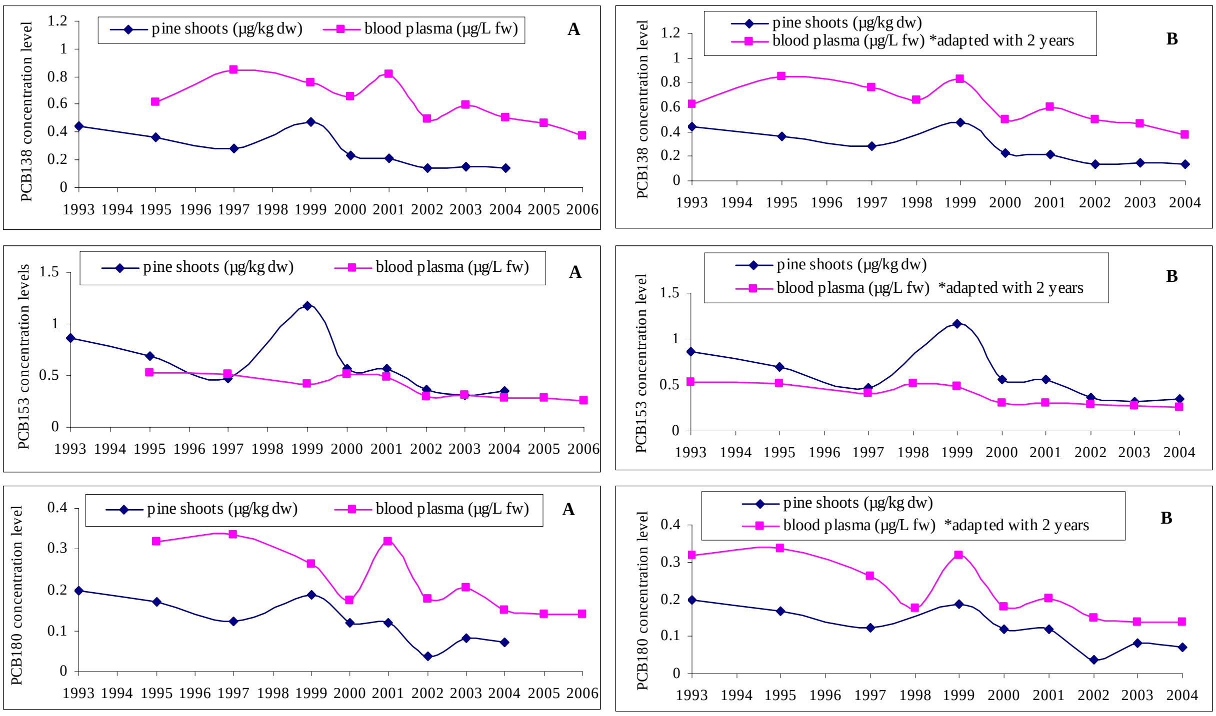

Fig. 1: PCB138, PCB 153 and PCB 180 level in human blood plasma during 1995-2006 and pine shoots during 1993-2004 sampled from Halle and nature park Dübener Heide, respectively (A). Human data adapted by shifting by two years back (B).

Similar pattern was found for both type samples during investigated years. Additionally, the figure revealed that the human data followed the corresponding environmental levels with some delay. In order to visualize it, the human data curve was additionally adapted by shifting it by two years back as shown on Figure 1 (B).

Consequently, fairly good relationship was found for both blood plasma and pine shoots samples in regard to PCB 138 and PCB 180. However, PCB 153 that was the prevailing congener did not confirm such dependence.
The correlation of PCB data of the roe deer liver and spruce shoot samples in different years is presented on Figure 2.

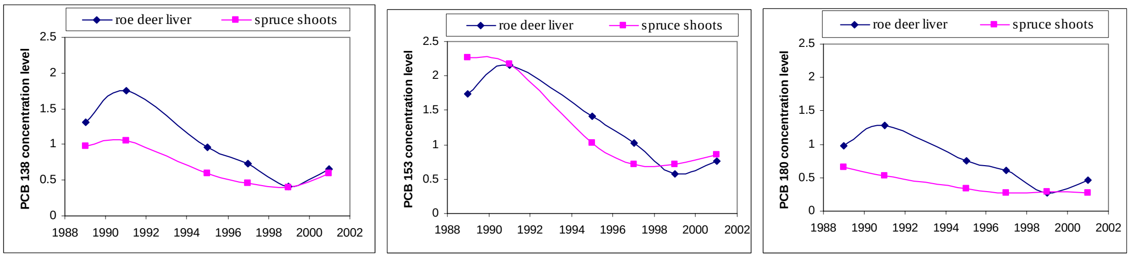

Fig. 2: PCB138, PCB 153 and PCB 180 level in roe deer liver and spruce shoots during 1989, 1991, 1995, 1997, 1999, and 2001 sampled from Warndt, respectively.

The results suggest that the PCB were predominantly accumulated in roe deer liver than spruce shoots. PCB153 was present in higher concentrations than the other congeners. Overall, concerning the different sampling years seems to demonstrate the tendency to a decrease of the contamination by organics compounds in Warndt area. Also, similar PCB 138 pattern was found for both matrices whereas in regard to PCB 153 a minor delay in the accumulation of roe deer liver compare to the spruce shoots was observed.
Individual PCB congeners exhibit different physico-chemical properties that result in different profiles for environmental distribution and toxicity. The mean and standard deviations for HCB, PCB138, PCB153 and PCB180 based on bioconcentration ratios are presented in Table 1.

Bioconcentration ratios		PCB138 mean±STD	PCB153 mean±STD	PCB180 mean±STD	HCB mean±STD
blood plasma/ egg matter of city pigeons (a) and (b)	residential use areas in Halle (a)	0.5±0.2	0.3±0.1	0.2±0.1	0.8±0.4
	residential use areas in Leipzig (b)	0.6±0.2	0.3±0.1	0.2±0.1	0.8±0.4
blood plasma/ earthworm (c) and (d)	park areas in Halle (c)	1.7±1.0	1.3±0.6	1.8±0.9	0.2±0.1
	park areas in Leipzig (d)	2.2±0.7	1.3±0.5	2.1±0.8	0.8±0.7
blood plasma/ roe deer liver from Dübener Heide park (e)		0.8±0.6	0.8±0.6	0.7±0.5	0.6±0.7
spruce shoots/roe deer liver from Warndt (f)		0.7±0.2	1.0±0.2	0.6±0.2	

Table 1: Mean and standard deviation values of the bioconcentration ratios. The dimensions are shown in the Material and Methods section.

Since the nineteenth century, Halle has been an industrial city since the nineteenth century, somewhat lower bioconcentration ratios were found for this area compared to the human/environmental ratios regarding Leipzig. Due to lack of HCB data the bioconcentration ratio (f) of spruce shoots and roe deer liver was given for PCB congeners only. In addition, the linear regression coefficient and related slope and y-intercepts as a function of MW, log K_{ow}, K_H, and log K_{oc} are shown in Table 2.

Table 2: Linear regression coefficients and y-intercepts as a function of the ratios and MW, log K_{ow}, K_H, and log K_{oc} constants

BR	MW (g/mol)		log K_{ow}		K_H (Pa m³ mol^{-1})		log K_{oc}	
	R^2	y	R^2	y	R^2	y	R^2	y
a	0.89	-161x + 423	0.94	-2.6x + 7.7	0.02	-40.6x + 152	0.70	-2.4x + 8.0
b	0.86	-165x + 430	0.89	-2.6x + 7.8	0.04	-50.6x + 158	0.55	-2.3x + 8.1
c	0.93	64.9x + 270	0.91	1.0x + 5.3	0.12	-33.0x + 174	0.93	1.1x + 5.6
d	0.71	58.8x + 255	0.66	0.9x + 5.1	0.40	-63.1x + 236	0.78	1.1x + 5.2
e	0.37	300x + 128	0.42	5.0x + 2.8	0.00002	-3.3x + 136	0.77	7.4x + 1.5
f	0.55	-65.8x + 423	0.41	-0.8x + 7.5	0.99	-217x + 233	0.18	-0.4x + 7.7

Particularly, in case of blood plasma/city pigeons egg matter pairs and spruce shoots/roe deer liver negative slopes were found for the correlations with all constants. The octanol–water partition coefficient (K_{ow}) and sorption partition coefficient (log K_{oc}) as well as the Henry's constant (K_H) are used to calculate the transport and fate of chemicals in the environment or eco-system. Therefore, the results suggest that the accumulation of lipophilic compounds is more pronounced in pigeon eggs than in human blood and in roe deer liver than in spruce shoots. The finding confirms the assumption that the lipid content is the predominant factor influencing the bioconcentration of lipophilic organochlorinated compounds in various tissues. In case of blood plasma/earthworm pair the opposite behavior was exhibited, but also with strikingly similar slopes and y-intercepts for the two different locations.

Moreover, fairly good relationship was demonstrated for the dependence of the ratios concerning egg matter of city pigeons and earthworm in the park areas in Halle, respectively to the MW, log $K_{ow,}$ and log K_{oc} constants. Another study has found a significant positive correlation between the sum of PCB concentrations in blue tit eggs and great tit eggs, suggesting similar exposure pathways, mechanisms of accumulation and maternal transfer of PCBs[8]. No linear correlations were found for the blood plasma/roe deer liver and spruce shoots/roe deer liver ratios in regard to all physicochemical constants here studied except log K_{oc} (R^2=0.77) and K_H (R^2=0.99), respectively.

Although the data set is poor with respect to the number of chemicals, the study shows some interesting and also promising relationships between environmental exposome and human biomonitoring data. These examples illustrate the preference biomarkers of exposure in epidemiology. Furthermore, the usage of "omic" technologies may assist to clarify the gene-environmental interactions.

Acknowledgements

The authors would like to thank Dr. Schröter-Kermani for provided support in this study.

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