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CASE REPORT: HIGH DIETARY EXPOSURE TO HEXACHLOROBENZENE IN A HEAVILY CONTAMINATED AREA IN CARINTHIA (SOUTHERN AUSTRIA)

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Introduction

Hexachlorobenzene (HCB) belongs to the group of polychlorinated benzenes. HCB is a persistent organic pollutant and was formerly used in many ways for example as a plant protection agent for combating fungal diseases and in the plastics industry as plasticiser and flame retardant. However, HCB is also produced as a by-product or impurity in the manufacture of chlorinated solvents and other chlorinated compounds. Therefore it is ubiquitously distributed in the environment. Due to its high lipid solubility HCB accumulates in the food chain. Regulation (EC) No. 396/2005[1] sets maximum residue levels for HCB in food and feed of plant and animal origin.

A high intake of HCB can cause toxic effects on the endocrine and nervous system, blood formation, skin and liver in humans[2]. ATSDR[3] has established various Minimal Risk Levels (MRLs) which are estimates of the daily human exposure that are likely to be safe over a certain period of exposure. For acute-duration oral exposure (14 days or less) to HCB an MRL of 8 µg/kg/d has been derived. For intermediate-duration oral exposure (15– 364 days) an MRL of 0.1 µg/kg/d, and for chronic-duration oral exposure (365 days or more) an MRL of 0.07 µg/kg/d has been set.

In a cement plant in Carinthia HCB-contaminated lime was burned at too low temperature and HCB was released into the environment. In the present study, food samples from the affected region were analysed for the presence of HCB and dietary exposure of different population groups was estimated. Estimated intakes were then compared to the respective MRL.

Materials and methods

HCB analysis in food of animal origin was performed by the QuEChERS GC/MS-MS method published by Lichtmannegger et al.[4]. For food of plant origin, except for those with high fat content, the QuEChERS GC/MS-MS method EN 15662-2008 was applied[5]. HCB analysis in oils and fats was performed according to the GC/MS-MS method published by the European Union Reference Laboratory for Pesticides in Fruits and Vegetables[6].

For dietary intake assessment, left-censored data were treated according to the lower bound (LB) – upper bound (UB) concept. LB concentrations were calculated by setting analytical results below LOD or LOQ at zero. For the medium bound (MB) values below LOD or LOQ equal half of the respective limit. For the UB concentrations it was assumed that analytical results below LOD or LOQ are equal to the respective limit[7]. Dietary intakes were estimated using national food consumption data from a survey conducted within the scope of the Austrian Nutrition Report 2008[8]. Food consumption data of different population groups were used: females and men aged 19-65 years and children aged 6-15 years. Mean body weights (bw) were 63.6 kg for women, 81.5 kg for men and 39.7 kg for children. A deterministic approach[9] was chosen to estimate dietary intake to HCB. Point estimates were calculated by multiplying the mean concentrations of food by the mean amount of food consumed by the respective population groups.

Results and discussion

Levels of HCB in food

A total of 771 samples of food originating from the contaminated area were analysed for HCB. All details related to the occurrence of HCB in various food samples are provided in Table 1. HCB was frequently quantified in foods of animal origin. Especially in high-fat containing food groups such as fats, oils and butter, meat, processed meat, milk and milk products measurable HCB concentrations were found. The highest content was found in beef with a maximum of 625 µg/kg. But also plants such as pumpkins in the oil-rich pumpkin seeds and various herbs with large leaf surface, e.g. peppermint and thyme, may accumulate HCB. In vegetable oils the highest concentration was 111 µg/kg.

In consideration of a measurement uncertainty of 50%, 1 sample of beef, 19 samples of milk and milk products and 1 sample of butter exceeded the respective maximum residue levels.

Estimated dietary exposure to HCB
Table 2 shows the mean estimated dietary intake to HCB for different Austrian population groups. For children, the mean estimated dietary exposure of HCB ranged from 0.201 to 0.295 µg/kg bw/d (LB – UB). Women`s average intake was between 0.165 (LB) and 0.260 µg/kg bw/d (UB). Mean dietary intakes for men were in the range of 0.171 and 0.244 µg/kg bw/d (LB – UB).
Considering a high consumption for children, a dietary exposure between 0.402 und 0.511 µg/kg bw/d was estimated. For women the HCB intake in a high consumption scenario was between 0.468 and 0.580 µg/kg bw/d. High intakes for men ranged from 0.433 to 0.522 µg/kg bw/d (LB – UB) (data not shown).

Characterisation of health risks associated with the dietary exposure to HCB
Comparing the estimated dietary intakes to the MRLs revealed that intakes were below the acute MRL and above the intermediate and chronic MRL for all population groups in the average intake scenario (Figure 1). The estimated scenario reflected the situation in the contaminated region in the year 2014 assuming that the people only ate food from the contaminated area. Hence the dietary exposure to HCB may be an overestimation.
The high concentrations in food and exposure levels exceeding the health based guidance values were an alarming signal for the competent authorities taking effective steps to remedy the contamination source. The cement plant stopped the burning of lime, contaminated roughage was exchanged and contaminated food was no longer sold nor recommended to be eaten. In 2015, the environmental HCB concentration was declining again. HCB concentrations in food of animal origin from the area are now below the European residue levels for HCB.

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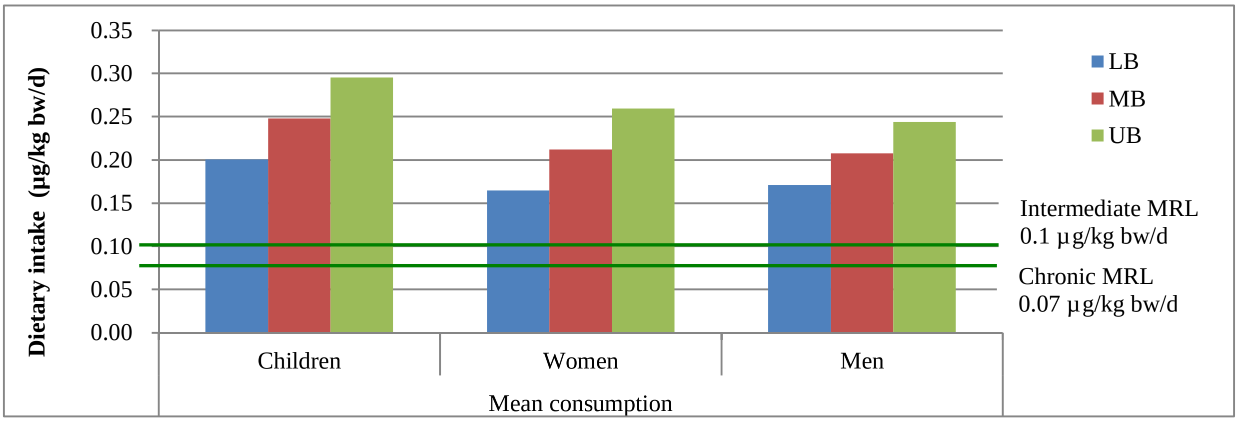
Table 1: Mean and maximum concentrations of HCB in different food groups expressed in µg/kg

Food group	N	N<LOD	N<LOQ	N>LOQ	Mean LB	Mean MB	Mean UB	Max
Milk and milk products (excluding butter)	**453**	**86**	**160**	**207**	**9.28**	**10.07**	**10.86**	**137**
Cheese	71	11	31	29	17.23	17.53	17.83	137
Milk and curdled milk	341	75	91	175	8.35	9.23	10.11	66,75
Milk products	41	0	38	3	3.23	4.14	5.06	19
Non-alcoholic beverages	**65**	**3**	**58**	**4**	**1**	**2.38**	**3.77**	**24**
Fruit juices	18	3	15	0	-	-	-	<LOQ
Tap water	41	0	40	1	0	0.01	0.02	20
Syrup	2	0	2	0	-	-	-	<LOQ
Tea and tea beverages	4	0	1	3	-	-	-	24
Processed meat	**58**	**0**	**40**	**18**	**22.22**	**23.94**	**25.66**	**218**
Cured meat	36	0	24	12	28.01	30.24	32.46	218
Sausages	22	0	16	6	12.73	13.64	14.55	96
Vegetables	**43**	**8**	**33**	**2**	**0.95**	**4.72**	**8.49**	**13**
Leafy vegetables	22	5	17	0	-	-	-	<LOQ
Fruiting vegetables	1	1	0	0	-	-	-	<LOD
Mixed vegetables	2	0	2	0	-	-	-	<LOQ
Legumes	1	0	1	0	-	-	-	<LOQ
Herbs	4	0	2	2	-	-	-	13
Sprouts and leek vegetables	4	1	3	0	-	-	-	<LOQ
Root and tuber vegetables	9	1	8	0	-	-	-	<LOQ
Fats, Oils, Butter	**38**	**10**	**10**	**18**	**49.11**	**50.55**	**52**	**556**
Butter	26	10	5	11	53.13	54.29	55.44	556
Vegetable oils	10	0	4	6	-	-	-	111
Animal fats (pork)	2	0	1	1	-	-	-	30.5
Eggs	**34**	**13**	**10**	**11**	**8.8**	**10.11**	**11.42**	**50**
Honey	**32**	**1**	**31**	**0**	**0**	**4.89**	**9.78**	**<LOQ**
Meat, poultry, game, offal	**28**	**3**	**8**	**17**	**78.57**	**79.81**	**81.04**	**625**
Meat (beef, lamb, pork)	21	3	3	15	94.17	94.86	95.55	625
Poultry	3	0	3	0	-	-	-	<LOQ
Kidney (pork)	1	0	1	0	-	-	-	<LOQ
Game (deer)	3	0	1	2	-	-	-	205
Cereals	**20**	**1**	**19**	**0**	**0.35**	**4.43**	**8.5**	**<LOQ**
Total	**771**	**125**	**369**	**277**	**-**	**-**	**-**	**625**

N=number of samples, LOD=limit of detection, LOQ=limit of quantification, LB=lower bound, MB=medium bound, UB=upper bound

Table 2: Mean dietary intake of HCB for average consumption of contaminated foods (in µg/kg bw/d)

	Consumption (g/d)			Exposure (µg/kg bw/d)								
Food group	Children	Women	Men	Children			Women			Men		
				LB	MB	UB	LB	MB	UB	LB	MB	UB
Milk and milk products (excluding butter)	214.4	193.4	166.0	0.05	0.054	0.059	0.028	0.031	0.033	0.019	0.020	0.022
Non-alcoholic beverages	852.8	1314.7	1261.7	0.021	0.051	0.081	0.021	0.049	0.078	0.015	0.037	0.058
Processed meat	33.8	36.2	84.3	0.019	0.020	0.022	0.013	0.014	0.015	0.023	0.025	0.027
Vegetables	78.6	190.3	164.2	0.002	0.009	0.017	0.003	0.014	0.025	0.002	0.010	0.017
Fats, Oils, Butter	18.9	32.3	35.0	0.023	0.024	0.025	0.025	0.026	0.026	0.021	0.022	0.022
Eggs	17.3	16.8	17.2	0.004	0.004	0.005	0.002	0.003	0.003	0.002	0.002	0.002
Honey	1.0	2.5	2.0	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000
Meat, poultry, game, offal	40.9	59.1	92.1	0.081	0.082	0.084	0.073	0.074	0.075	0.089	0.090	0.092
Cereals	16.6	26.4	30.3	0.000	0.002	0.004	0.000	0.002	0.004	0.000	0.002	0.003
Total	1274.3	1871.7	1852.8	0.201	0.248	0.295	0.165	0.212	0.260	0.171	0.208	0.244



LB=lower bound, MB=medium bound, UB=upper bound

Figure 1: Comparison of the dietary HCB intake to the intermediate and chronic minimal risk level