

# Occurrence of Chlorobenzenes in Nine United Kingdom Retail Vegetables

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An analytical method for the measurement of chlorobenzenes (CBs) in vegetables was developed. Vegetables were ground, dewatered, Soxhlet extracted, and column cleaned and CBs determined by capillary gas chromatography. The effects of different drying techniques and extraction times on the recovery of CBs were examined. Nine U.K. retail vegetables were collected from supermarkets and analyzed. All of the CBs were detected in at least some of the commercial vegetables. Hexachlorobenzene (HCB) and 1,4-dichlorobenzene were the most frequently occurring compounds (in 89% and 61% of samples, respectively) and had the highest average concentrations (0.55 and 0.26  $\mu\text{g kg}^{-1}$  of fresh weight, respectively). However, monochlorobenzene was detected in cabbages at high concentrations (average of 207  $\mu\text{g kg}^{-1}$  of fresh weight). Peels of the root crops had higher CB concentrations than their cores. CB contents of the leaf vegetables were higher in the outer parts than in the inner parts. However, CB concentrations in the seeds of peas and beans were higher than in their pods. Human exposure to CBs by consumption of these vegetables is estimated, using typical U.K. intake rates, although the study was not designed to be statistically representative for the United Kingdom as a whole. Potatoes were clearly the most important source among the vegetables analyzed. HCB accounted for a large proportion of the calculated total average intake of CBs by the U.K. population.

**Keywords:** Chlorobenzenes; vegetables; gas chromatography; human exposure

## INTRODUCTION

CBs have been found in human adipose tissue (LeBel and Williams, 1986; Williams et al., 1988; Tanabe et al., 1993), serum, blood cells, liver (Mussalo-Rauhamaa, 1991), and milk (Jan, 1983), indicating that the general population is exposed to CBs. The ubiquitous occurrence of many CBs at trace levels in the environment may lead to general human exposure, with vegetation likely to be important. Uptake of CBs by carrots from soil has been investigated (Wang and Jones, 1994c), showing that bioaccumulation of CBs can occur in plant-soil systems. It is useful to estimate the potential human exposure to CBs through plant-based foods. However, little information is available on this to date. This paper presents the results obtained from a preliminary study on selected U.K. vegetable crops. An analytical method for CBs in vegetables by Soxhlet extraction, column cleanup, and capillary gas chromatography was developed. Nine important U.K. vegetables were chosen for this study, each of which accounts for more than 10% of the "green vegetables" or "other vegetables" in the U.K. diet (MAFF, 1988). These vegetables included edible roots, leaves, stem and flower, fruit, and seeds. The samples were purchased from two supermarkets and analyzed for their CB content. This information was then used to make a preliminary estimation of the significance of vegetables as a source of CBs in the U.K. diet.

## MATERIALS AND METHODS

The vegetables were all produced in Britain and obtained fresh. After purchase from major supermarket chains, the vegetation samples were bagged and kept at 4 °C in a refrigerator prior to analysis. Carrots and potatoes were separated into peel (<1 mm) and core with a normal kitchen peeler. The peeler only removed the endodermis from the root,

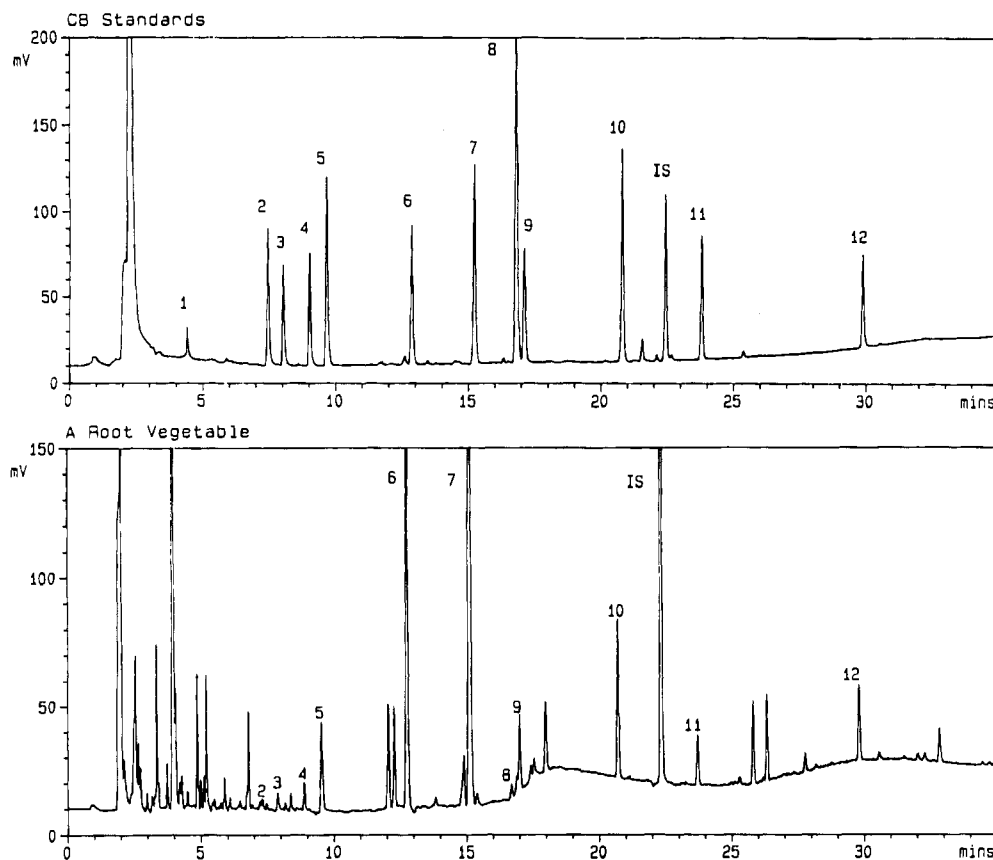
**Table 1. Vegetable Samples Used in This Study**

vegetation (Chittenden, 1965)	part 1	part 2	DMC <sup>a</sup>
carrot ( <i>Daucus carota</i> )	cores	peels	7.6
potato ( <i>Solanum tuberosum</i> )	cores	peels	22.1
cabbage ( <i>Brassica oleracea capitata</i> )	inner	outer	6.2
cauliflower ( <i>Brassica oleracea botrytis cauliflora</i> )	stems	flowers	9.9
lettuce ( <i>Lactuca sativa</i> )	inner	outer	3.2
onion ( <i>Allium cepa</i> )	inner	outer	10.9
broad bean ( <i>Vicia faba</i> )	seeds	peels	31.0
pea ( <i>Pisum sativum</i> )	seeds	peels	18.3
tomato ( <i>Lycopersicon esculentum</i> )	flesh	peels	5.0

<sup>a</sup> Dry matter content (%) for the whole vegetable.

and the rest was classified as core. The peels and cores were measured separately. Cabbages, lettuces, and onions were divided into inner and outer parts and analyzed. Cauliflowers were divided into stems and flowers for separate analyses. Beans and peas were analyzed as seeds and pods, respectively. Tomatoes were also peeled and measured as peels and flesh separately. Except for the bean and pea pods, all of the samples analyzed were edible. All of the vegetable parts were weighed carefully to calculate the CB concentrations in the whole vegetables. For each vegetable, a composite sample of many individuals (e.g., 8 cauliflowers, 40 carrots, 56 tomatoes, hundreds of pea pods) was used. All of the composite samples were analyzed in duplicate. The vegetation samples used for this study are listed in Table 1. Dry matter content of the vegetation was measured by heating the samples at 100 °C for 24 h. The average dry matter content of the whole vegetables was 12.7%. Whole carrots were used as the samples for the method development.

Fresh samples were washed with tap water and deionized water (root samples were washed in an ultrasonic generator), divided into two parts (as described above), carefully weighed, chopped into small sections, and homogenized in a blender mill; 65–85 g (depending on both the dry matter content and the type of sample) of the homogenized sample was mixed with 130–170 g (2–3 times the weight of the sample, depending



**Figure 1.** (A, top) Chromatogram of chlorobenzene mixture in hexane on DB-Wax column. Compound numbers and concentrations ( $\mu\text{g mL}^{-1}$ ): 1, MCB (75.9); 2, 1,3-DCB (0.40); 3, 1,4-DCB (0.64); 4, 1,2-DCB (0.41); 5, 1,3,5-TCB (0.088); 6, 1,2,4-TCB (0.096); 7, 1,2,3-TCB (0.070); 8, 1,2,3,5-TeCB (0.085); 9, 1,2,4,5-TeCB (0.049); 10, 1,2,3,4-TeCB (0.049); 11, PeCB (0.019); 12, HCB (0.0097); IS, TBB (0.18). (B, bottom) Chromatogram of a root vegetable sample on DB-Wax column. Compound numbers as in (A).

on the water content of the sample) of previously heated anhydrous sodium sulfate and extracted with a mixed solvent (hexane/acetone 2:1) for 24 h in a pre-extracted cellulose thimble in a full glass Soxhlet extractor. The extract was then concentrated to  $\sim 2$  mL in a Buchi RE 121 Rotovapor (Switzerland) at  $70^\circ\text{C}$  without using the vacuum system. Cleanup of this extract was accomplished by eluting the concentrated extract with hexane from a hexane prewashed column (i.d., 7 cm) packed with Florisil, silica gel/ $\text{H}_2\text{SO}_4$ ,  $\text{NaHCO}_3/\text{Na}_2\text{SO}_4$ , and Florisil (5, 2.5, 1.5, and 1 cm, respectively, from top to bottom), and collecting the first 10–15 mL in a glass vial. The clean extract was evaporated to 500  $\mu\text{L}$  under the gentle flow of nitrogen and dosed with 1  $\mu\text{L}$  of 88  $\text{mg mL}^{-1}$  of 1,3,5-tribromobenzene (TBB) solution in hexane. The dosed extracts were analyzed by gas chromatography (GC) with an electron capture detector (ECD) on DB-Wax and Ultra 2 columns. Details of the GC analysis have been reported previously (Wang and Jones, 1991). For illustrative purposes, Figure 1A shows a chromatogram of the chlorobenzene standard mix separated on the DB-Wax column and Figure 1B shows a chromatogram of a root vegetable extract. In addition, five samples of different vegetables were screened on a HP5970B mass selective detector to confirm the identity of the CBs with the DB-Wax column. For each pair of the duplicated samples, a blank sample, namely the same amount of  $\text{Na}_2\text{SO}_4$  (without vegetables) as used for the samples, was treated and analyzed using exactly the same procedures for the vegetable samples described here to prevent contamination during the treatment operation. When the samples were determined on GC-ECD, one injection of a mixture of CB standards was always made before every pair of vegetable samples, to correct for any possible variation in the compound responses.

## RESULTS AND DISCUSSION

**Extraction Time.** Because of their high lipid content, carrots were chosen as a sample to optimize the

**Table 2. Results from a Carrot Sample Extracted for Different Times (Micrograms per Kilogram of Fresh Weight,  $n = 2$ )<sup>a</sup>**

compd	6 h	SD	24 h	SD	36 h	SD
MCB						
1,3-DCB						
1,4-DCB	0.244	0.0261	0.249	0.0130	0.247	0.027
1,2-DCB						
1,3,5-TCB	0.0614	0.0052	0.0596	0.0133	0.0595	0.0063
1,2,4-TCB						
1,2,3-TCB						
1,2,3,5-TeCB						
1,2,4,5-TeCB						
1,2,3,4-TeCB	0.0064	0.0012	0.0079	0.0033	0.0077	0.0003
PeCB	0.0077	0.0009	0.0078	0.0006	0.0079	0.0008
HCB	0.0074	0.0005	0.0085	0.0011	0.0084	0.0015
$\Sigma\text{CBs}$	0.327	0.106	0.333	0.025	0.330	0.101

<sup>a</sup> A blank space indicates below detection limit.

extraction time for vegetables. Whole carrots were washed, chopped, homogenized, mixed with anhydrous sodium sulfate, and divided into aliquots and extracted for 6, 24, and 36 h, respectively. The extracts were then concentrated, cleaned up, and analyzed as described under Materials and Methods. The results listed in Table 2 show there was no significant difference with the different times. However, 24 h of extraction gave a slightly higher value and was therefore chosen for use in the study.

**Sample Dewatering Technique.** A sample of commercially available carrots was washed, chopped, homogenized, and divided into aliquots and treated by two

**Table 3. Results from a Carrot Sample Pretreated Using Different Techniques (Micrograms per Kilogram of Fresh Weight,  $n = 2$ )<sup>a</sup>**

compd	FD	SD	FS	SD
MCB			238	9.80
1,3-DCB				
1,4-DCB				
1,2-DCB				
1,3,5-TCB	0.0144	0.0005	0.184	0.0059
1,2,4-TCB				
1,2,3-TCB			0.0313	0.0026
1,2,3,5-TeCB			0.0080	0.0004
1,2,4,5-TeCB				
1,2,3,4-TeCB			0.0077	0.0025
PeCB			0.0033	0.0002
HCB	0.0032	0.0002	0.0066	0.0005
ΣCBs	0.0176	0.0013	239	9.81

<sup>a</sup> A blank space indicates below detection limit.

**Table 4. Recoveries of Chlorobenzenes from Spiked Carrot Samples**

compd	low concn ( $n = 3$ )			very low concn ( $n = 4$ )		
	concn ( $\mu\text{g kg}^{-1}$ of fresh wt)	recovery (%)	CV (%)	concn ( $\mu\text{g kg}^{-1}$ of fresh wt)	recovery (%)	CV (%)
MCB	749	39.7	4.66			
1,3-DCB	3.07	66.1	2.28	0.326	72.1	2.25
1,4-DCB	4.86	73.8	5.33	0.516	52.3	31.7
1,2-DCB	3.12	78.6	5.22	0.332	62.3	3.93
1,3,5-TCB	0.666	80.2	2.44	0.0707	77.0	1.65
1,2,4-TCB	0.733	85.5	1.54	0.0777	76.0	9.76
1,2,3-TCB	0.531	76.5	11.3	0.0563	125	11.4
1,2,3,5-TeCB	0.695	83.0	1.49	0.0737	90.4	1.36
1,2,4,5-TeCB	0.375	79.0	1.34	0.0398	81.0	6.62
1,2,3,4-TeCB	0.374	89.5	1.92	0.0397	102	4.01
PeCB	0.142	91.6	1.36	0.015	108	5.36
HCB	0.0737	92.7	0.06	0.0078	108	10.8

techniques: freeze-drying (FD) and freshly mixed with preheated anhydrous sodium sulfate (FS). The samples were then treated according to the procedures described under Materials and Methods. The results listed in Table 3 show that FS yielded much higher concentrations of CBs in the carrots than FD, which resulted in some loss of CBs in the samples. FS was therefore chosen for the treatment of vegetable samples.

**Recoveries and Detection Limits.** Recoveries of the CBs from vegetables by these methods were tested by analyzing carrot samples spiked at two levels and corresponding samples. The CB standards were spiked into the samples after the homogenization step. All of the other treatments followed the procedures described under Materials and Methods. The inherent concentrations of CBs in the samples were determined and subtracted from the results of spiked samples. The compound recoveries are listed in Table 4. Recoveries from both treatments were similar. MCB is more volatile than the other CBs, and it was lost significantly during the pretreatment procedures (mainly during the concentration operation). The detection limits (fresh weight) of this method are estimated as 0.0015 (HCB), 0.002 (pentachlorobenzene, PeCB), 0.004–0.007 (tetrachlorobenzenes, TeCBs), 0.007–0.01 (trichlorobenzenes,

TCBs), 0.08–0.1 (dichlorobenzenes, DCBs), and 200  $\mu\text{g kg}^{-1}$  (MCB).

**Concentrations of CBs in Vegetables.** The concentrations of CBs in the vegetable parts are presented in Table 5 on a fresh weight basis. This does not include the CB concentrations in the whole carrot samples used for the method development tests (Tables 2 and 3). All of the CBs were detected in at least some of the commercial vegetables purchased from supermarkets. Because vegetables are normally consumed fresh, most of the discussion here will be based on the results expressed on a fresh weight basis. CB concentrations based on dry weight can be calculated using the dry matter contents of the samples given in the table.

MCB was only detected in the cabbage samples, except the carrot samples used for the dewatering technique test (see the corresponding section and Table 3). Compared with the other CBs, MCB had a very high detection limit and low recovery by the method used here, which made the quantitative precision for MCB much lower than for the other CBs. Therefore, the MCB concentrations should not be considered as reliable as the other CBs, although they are also reported here. The total concentrations minus MCB ( $\Sigma\text{CBs} - \text{MCB}$ ) are therefore used for the general discussion.

Potato samples had the highest  $\Sigma\text{CBs} - \text{MCB}$  content (fresh weight), i.e., 3.96  $\mu\text{g kg}^{-1}$  for the cores and 8.72  $\mu\text{g kg}^{-1}$  for the peels, in which HCB accounted for 75% (cores) and 70% (peels) of the total. Cabbage samples gave the lowest  $\Sigma\text{CBs} - \text{MCB}$  concentrations (0.029  $\mu\text{g kg}^{-1}$  for the inner part and 0.13  $\mu\text{g kg}^{-1}$  for the outer part). However, if MCB is included, cabbage samples were the most contaminated (364  $\mu\text{g kg}^{-1}$  for the inner part).

For the root vegetables, all of the peels were more contaminated than the cores. The ratios of the peel  $\Sigma\text{CBs}$  concentration to those in the core were 2.6 and 2.2 times for the carrots and potatoes respectively, implying the CBs associated with the root crops were probably derived from root uptake and that penetration through the peels was hindered. This is in line with the results from an experiment on uptake of CBs by carrots (Wang and Jones, 1994c). All of the outer parts of cabbages, lettuces, and onions had higher  $\Sigma\text{CBs} - \text{MCB}$  contents than their inner parts; the ratios of the concentrations of the outer to the inner were 4.5, 1.4, and 1.2 separately. This phenomenon was possibly due to foliar uptake—the older outer parts may have been exposed to the compounds more than the younger, protected inner portions. For cauliflowers, CB concentrations in the flower were higher than those in the stems, most probably also due to uptake from the atmosphere as well. Tomato peel contained much higher CB concentrations than the flesh, presumably also caused by uptake from the atmosphere. It is interesting that the seeds of beans and peas all had higher concentrations of  $\Sigma\text{CBs}$  than their pods, probably caused by the ability of the seeds to sorb compounds and/or receive internally transferred compound or by a different uptake mechanism. However, the HCB concentrations in the pods were higher than those in the seeds, presumably implying a greater penetration of DCBs and TCBs through the pods than that for HCB. This explanation would be consistent with the observation on carrots previously (Wang and Jones, 1994c).

It is clear that the peeling of root vegetables and the removal of the outer part portion of leafy crops will substantially reduce ingestion of these compounds.

Table 5. Chlorobenzene Concentrations in Vegetable Parts (Micrograms per Kilogram of Fresh Weight, n = 2)<sup>a</sup>

compd	carrots			potatoes			cabbages			cauliflowers			lettuces					
	core	SD	peel	SD	peel	SD	inner	SD	outer	SD	stem	SD	flower	SD	inner	SD	outer	SD
MCB	(7.4) <sup>b</sup>		(8.6)		(13.9)		(6.0)		(6.4)		(8.9)		(10.6)		(3.3)		(2.9)	
1,3-DCB	0.198	0.055	0.416	0.383	0.224	0.166	0.0240	0.0025	0.0205	0.0090	0.214	0.043	0.529	0.104	0.237	0.208	0.118	0.048
1,4-DCB		0.328	0.064															
1,2-DCB		0.0090	0.0030		0.0320	0.0130	0.0240	0.0025	0.0159	0.0025	0.0104	0.0053	0.0469	0.0087			0.0079	0.0011
1,3,5-TCB		0.0550	0.0080														0.0070	0.0009
1,2,4-TCB																		
1,2,3-TCB																		
1,2,3,5-TeCB		0.0297	0.0198		0.0216	0.0005											0.0022	0.0006
1,2,4,5-TeCB		0.486	0.0180		2.19	0.0262											0.0120	0.0032
1,2,3,4-TeCB		0.0218	0.0128		0.0726	0.0005											0.0041	0.0016
PeCB		0.0034	0.0010		0.0734	0.0033												
HCB	0.0204	0.0042	0.0896	0.0024	6.09	0.103	0.0046	0.0005	0.0913	0.0120	0.0150	0.0009	0.1180	0.0307	0.0289	0.0005	0.234	0.0195
ΣCBs	0.219	0.079	0.561	0.414	8.72	0.232	364	135	0.128	0.0185	0.240	0.049	0.700	0.144	0.266	0.209	0.385	0.063
ΣCBs - MCB	0.219	0.079	0.561	0.414	8.72	0.232	0.0286	0.0091	0.128	0.0185	0.240	0.049	0.700	0.144	0.266	0.209	0.385	0.063
compd	onions			beans			peas			tomatoes								
	inner	SD	outer	SD	seeds	SD	pod	SD	seeds	SD	pod	SD	flesh	SD	peel	SD		
MCB	(10.9)		(10.9)		(25.3)		(33.9)		(26.9)		(13.5)		(4.8)		(9.0)			
1,3-DCB	0.0304	0.0006	0.0488	0.0138	0.717	0.030	0.117	0.065	0.087	0.023					0.619	0.190		
1,4-DCB									1.31	0.263								
1,2-DCB									0.112	0.100								
1,3,5-TCB									0.0391	0.0189								
1,2,4-TCB									0.0676	0.0062								
1,2,3-TCB													0.0467	0.0005				
1,2,3,5-TeCB																		
1,2,4,5-TeCB																		
1,2,3,4-TeCB																		
PeCB							0.0086	0.0037										
HCB	0.0021	0.0003			0.0032	0.0000	0.0137	0.0011	0.0033	0.0007					0.0100	0.0022		
ΣCBs	0.0823	0.0437	0.0964	0.0038	0.720	0.030	0.139	0.070	1.61	0.474			0.0467	0.0005	0.997	0.224		
ΣCBs - MCB	0.0823	0.0437	0.0964	0.0038	0.720	0.030	0.139	0.070	1.61	0.474			0.0467	0.0005	0.997	0.224		

<sup>a</sup> A blank space indicates below detection limit. <sup>b</sup> The values in parentheses are the dry matter contents (%) of the samples.

**Table 6. Occurrence, Range, Median, Mean, Standard Deviation, and Abundance Ranking of CBs in Vegetable Parts (Micrograms per Kilogram)**

compd	occurrence (%)	DL <sup>a</sup> (fresh wt)	fresh wt				av abund rnkng	dry wt				av abund rnkng
			max	median	mean <sup>b</sup>	SD		max	median	mean <sup>b</sup>	SD	
MCB	5.6	200	364	bd <sup>c</sup>	20.2	85.7	1	6020	bd	334	1420	1
1,3-DCB	11.1	0.08	0.096	bd	0.010	0.030	10	0.41	bd	0.04	0.12	11
1,4-DCB	61.1	0.1	1.31	0.158	0.261	0.347	3	7.11	3.38	2.37	2.54	3
1,2-DCB	11.1	0.09	0.328	bd	0.024	0.080	5	1.42	bd	0.10	0.34	8
1,3,5-TCB	50.0	0.008	0.160	0.0039	0.0196	0.0381	6	1.18	0.136	0.177	0.295	5
1,2,4-TCB	38.9	0.01	0.0675	bd	0.0108	0.0193	9	0.443	bd	0.092	0.135	9
1,2,3-TCB	22.2	0.007	0.0549	bd	0.0110	0.0213	8	0.973	bd	0.117	0.261	7
1,2,3,5-TeCB	11.1	0.005	0.0297	bd	0.0028	0.0084	12	0.346	bd	0.0278	0.0873	12
1,2,4,5-TeCB	22.2	0.007	2.19	bd	0.150	0.521	4	15.7	bd	1.02	3.71	4
1,2,3,4-TeCB	22.2	0.004	0.0725	bd	0.0060	0.0174	11	0.522	bd	0.0571	0.135	10
PeCB	33.3	0.002	0.127	bd	0.0119	0.0334	7	1.41	bd	0.118	0.346	6
HCB	88.9	0.0015	6.09	0.0177	0.552	1.55	2	43.8	0.222	4.03	10.5	2
ΣCBs	100		364	0.473	21.3	85.5		6020	6.26	343	1420	
ΣCBs - MCB	100		8.72	0.473	1.06	2.13		62.7	6.26	8.15	14.5	

<sup>a</sup> DL, detection limit. <sup>b</sup> Values below detection limits were treated as zero. <sup>c</sup> bd, below detection limit.

The concentration range, mean, median, and congener abundance ranking are presented in Table 6. HCB had the highest occurrence (88.9%) among the CBs, showing the widespread nature of this persistent compound. 1,4-DCB had the second highest occurrence, caused by its very broad use for both domestic and industrial purposes (Wang and Jones, 1994a,b; Wang et al., 1992, 1993). Apart from MCB, these two compounds were also the most abundant compounds in the vegetables in this study. HCB is a probable carcinogen in humans (EPA, 1985).

**Potential Exposure.** On the basis of the weight of each part of the crops, the CB concentrations in the whole vegetables (fresh weight) were calculated and are given in Table 7. Again, concentrations on a dry weight basis can be calculated using the dry matter contents of the samples listed in the same table. As a whole, potatoes had the highest concentrations of ΣCBs - MCB, i.e., 4.52 μg kg<sup>-1</sup>, while the other vegetables were lower than 1 μg kg<sup>-1</sup>. Peas and cauliflowers had the second and third highest concentrations of ΣCBs - MCB, 0.69 and 0.5 μg kg<sup>-1</sup>, respectively. Potential human exposure to the CBs through vegetation in the United Kingdom was estimated on the basis of "typical" or average consumption of the edible parts (MAFF, 1988), and the results are listed in Table 8. It should be noted that these calculations assume that the foods were eaten raw. In reality, of course, many vegetables are cooked, which may substantially affect the final CB content of ingested vegetables. Among the vegetables, consumption of potatoes by people was the highest (50.6 kg person<sup>-1</sup> year<sup>-1</sup>). Therefore, potatoes are likely to be the most important source of exposure to CBs (229 μg person<sup>-1</sup> year<sup>-1</sup>) of the crops studied. From Table 8, for example, potatoes would account for 96% of the total CBs and 99% of the HCB that people may consume from these vegetable crops. According to Table 8, peas, cauliflowers, and carrots were the second, third, and fourth most important sources, all individually resulting in potential exposure to more than 1 μg person<sup>-1</sup> year<sup>-1</sup>.

In the United States, tetrachloronitrobenzene (TCNB), in which 1,2,4,5-TeCB exists as an impurity, is used on potatoes as a sprout suppressant. In composite samples of boiled, fried, and baked potatoes, 1,2,4,5-TeCB con-

centrations were 0.095, 0.051 and 0.110 mg kg<sup>-1</sup>, respectively (Heikes et al., 1979), much higher than in this study.

HCB has been classified as a probable carcinogen in humans (EPA, 1985). Therefore, potential human exposure to this compound has received much more attention than the other CBs. According to a food survey undertaken in the United Kingdom during 1984 and 1985, the average concentration of HCB in poultry samples was 0.5 μg kg<sup>-1</sup> (MAFF, 1989). On the basis of the estimated weight of poultry eaten in Britain (6.2 kg person<sup>-1</sup> year<sup>-1</sup>; MAFF, 1988), the potential human exposure to HCB by consumption of poultry was 3.1 μg person<sup>-1</sup> year<sup>-1</sup>. This was higher than the individual annual contribution of all the vegetables analyzed in this study, except potatoes (see Table 8). The contribution made by potatoes to HCB dietary intake (based on the analytical results in this work) was much greater than that by poultry (MAFF, 1989). In the same survey (MAFF, 1989), no HCB was detected in potatoes, "green vegetables", and "other vegetables", which include all of the vegetables reported here. However, in this MAFF report, the detection limit for HCB was only mentioned as "from 1 to 20 μg kg<sup>-1</sup> depending on the compound and food examined" (MAFF, 1989). In our work, most of the HCB concentrations in the samples were less than 1 μg kg<sup>-1</sup>, which would be "not detected" by the MAFF (1989) procedure, with the probable exception of potato (if the detection limit for HCB was 1 μg kg<sup>-1</sup> in the MAFF study). Generally, HCB levels in British food did not show a consistent decline in the surveys conducted between 1969 and 1985 (MAFF, 1986, 1989). It is difficult to make any conclusive comments on the CB concentrations or the human intake amounts estimated here, since there have been no regulations imposed of the limits of CBs in vegetables (MAFF, 1986). However, the data presented here are generally more comprehensive and the result of more sensitive analytical procedures than those employed by MAFF.

An estimated value for the average human intake of HCB in the United Kingdom in 1980 was 0.14 μg day<sup>-1</sup> (WHO, 1984). If the CB concentrations detected in retail potatoes are representative of those in the United Kingdom generally, the intake of HCB by only the consumption of potatoes would be 0.46 μg day<sup>-1</sup> (as-

**Table 7. Chlorobenzene Concentrations in Whole Vegetables (Micrograms per Kilogram of Fresh Weight, n = 2)<sup>a</sup>**

compd	roots			tops			fruits											
	carrot	potato	cabbage	cauliflower	lettuce	onion	bean	pea	tomato	SD								
MCB	(7.6) <sup>b</sup>	(22.1)	(6.2)	(9.9)	(3.2)	(10.9)	(31.0)	(18.3)	(5.0)									
			207	77.2														
1,3-DCB	0.085	0.020																
1,4-DCB	0.234	0.076																
1,2-DCB	0.289	0.049																
1,3,5-TCB	0.0100	0.0093	0.0225	0.0138	0.0030	0.0363	0.0048	0.117	0.0198									
1,2,4-TCB	0.0038	0.0015	0.0068	0.0096	0.0027	0.0008		0.0342	0.0086									
1,2,3-TCB	0.0484	0.0074				0.0490	0.0335			0.0440 5.1E-4								
1,2,3,5-TeCB	0.0048	0.0032	0.0025	6.5E-5														
1,2,4,5-TeCB	0.688	0.0187			0.0046	0.0012	0.0056	0.0024										
1,2,3,4-TeCB	0.0035	0.0020	0.0086	6.3E-5	0.0016	6.1E-4				5.8E-4 1.3E-4								
PeCB	5.7E-4	1.6E-4	0.0108	0.0013	0.0035	6.1E-4	8.7E-4	2.2E-4		0.0073 6.8E-4								
HCB	0.0317	0.0199	3.35	0.199	0.0418	0.0088	0.0729	0.0176	0.108	0.0071	0.0014	0.0002	0.0101	7.6E-4	0.0039	7.8E-4	0.0139	0.0011
ΣCBs	0.274	0.031	4.52	0.350	207	77.2	0.499	0.103	0.311	0.152	0.0868	0.0306	0.337	0.102	0.690	0.202	0.102	0.012
ΣCBs - MCB	0.274	0.031	4.52	0.350	0.0711	0.0131	0.499	0.103	0.311	0.152	0.0868	0.0306	0.337	0.102	0.690	0.202	0.102	0.012

<sup>a</sup> A blank space indicates below detection limit. <sup>b</sup> The values in parentheses are the dry matter contents (%) of the samples.

**Table 8. Estimated Consumption of CBs in the United Kingdom from Vegetables (Micrograms per Person per Year)**

compd	carrot	potato	cabbage	cauliflower	lettuce	onion	bean	pea	tomato	sum
MCB	(4.55) <sup>c</sup>	(50.6)	(4.11)	(2.58)	(1.63)	(4.16)	(1.72)	(2.18)	(4.70)	850
			850							
1,3-DCB	1.06	1.35	4.28	1.01	0.310		1.23	0.189	0.168	4.47
1,4-DCB	0.00	14.6						2.85		7.98
1,2-DCB								0.244		14.9
1,3,5-TCB	0.0220	0.510	0.0924	0.0797	0.0049	0.151		0.085	0.207	0.84
1,2,4-TCB	0.0161	0.194	0.0281		0.0044	0.204		0.147		0.453
1,2,3-TCB		2.45								2.86
1,2,3,5-TeCB		0.130								0.152
1,2,4,5-TeCB		34.8			0.0075					34.8
1,2,3,4-TeCB		0.436			0.0026					0.457
PeCB	0.0026	0.547		0.0092	0.0014			0.0345		0.595
HCB	0.144	169	0.172	0.188	0.175	0.0059	0.0055	0.0072	0.0653	170
ΣCBs	1.25	229	850	1.28	0.505	0.361	1.24	3.50	0.477	1090
ΣCBs - MCB	1.25	229	0.292	1.28	0.505	0.361	1.24	3.50	0.477	238

<sup>c</sup> The values in parentheses are the estimated vegetation consumption by the people in the United Kingdom (kilograms per person per year) (MAFF, 1988).

suming no losses during preparation for consumption), clearly higher than the "total dietary intake" estimated by WHO (1984). Excluding potatoes, the intake of HCB through consumption of the other vegetables was 2.1 ng day<sup>-1</sup>, about 1.5% of the estimated total dietary intake. In Germany, the daily intake of HCB from vegetables and potatoes was estimated as 3 ng day<sup>-1</sup> (Burton and Bennett, 1987), which was much lower than that estimated here (0.47 μg day<sup>-1</sup>). Clearly, there are continuing uncertainties over the contemporary exposure of the general population to CBs, largely resulting from detection limit constraints and a lack of reliable data.

Vegetation samples used in this study were all obtained from local commercial suppliers and are not statistically representative for the United Kingdom as a whole. However, the estimated results should be broadly indicative of intakes, given that supermarket produce was used.

#### ABBREVIATIONS USED

CB, chlorobenzene; CBs, chlorobenzenes; MCB, monochlorobenzene; 1,4-DCB, 1,4-dichlorobenzene; 1,3-DCB, 1,3-dichlorobenzene; 1,2-DCB, 1,2-dichlorobenzene; DCBs, dichlorobenzenes; 1,3,5-TCB, 1,3,5-trichlorobenzene; 1,2,4-TCB, 1,2,4-trichlorobenzene; 1,2,3-TCB, 1,2,3-trichlorobenzene; TCBs, trichlorobenzenes; 1,2,3,5-TeCB, 1,2,3,5-tetrachlorobenzene; 1,2,4,5-TeCB, 1,2,4,5-tetrachlorobenzene; 1,2,3,4-TeCB, 1,2,3,4-tetrachlorobenzene; TeCBs, tetrachlorobenzenes; PeCB, pentachlorobenzene; HCB, hexachlorobenzene; ΣCBs, total concentration of CBs; 1,3,5-TBB, 1,3,5-tribromobenzene; GC, gas chromatography; ECD, electron capture detector; FD, freeze-drying; FS, freshly mixed with sodium sulfate; DMC, dry matter content (%) for the whole vegetable; CV, coefficient of variation (%); SD, standard deviation; DL, detection limit; bd, below DL; MAFF, Ministry of Agriculture, Fisheries and Food.

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