

PHTHALATES AND HBCD STEREOISOMERS IN U.S. FOOD

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Introduction

Phthalates are diesters of phthalic acids, a class of industrial chemicals extensively used since the early 20th century, which have been found in many personal care and industrial products. Phthalates are ubiquitous synthetic compounds and therefore difficult to measure; they have not previously been reported in food purchased in the United States¹. Phthalates have been associated with reproductive pathology², endocrine disruption^{3,4} and developmental alteration in male rodents⁵ and male infants^{6,7}.

Our objectives for this study were to report concentrations of 9 phthalates in 72 individual food samples purchased in Albany, New York, compare these with findings with those from other countries, and calculate average dietary intake. Additionally, we are reporting on HBCD total levels and HBCD stereoisomer levels in U.S. food.

Materials and methods

Convenience samples of commonly consumed foods (N=72) were purchased from supermarkets in Albany, New York, U.S.A. Methods were developed to analyze these foods for 9 phthalates, correcting for background exposure using gas chromatography-mass spectroscopy. Seven of the 72 foods were measured, but not included in any of the eight food categories analyzed because they were mixtures and difficult to place in food categories. Dietary intakes of phthalates were calculated as the product of a food contact rate and concentration of phthalates in that food⁸. Sampled foods included baby food, dairy, meat products and fish.

Results and Discussion

The range of detection frequency of individual phthalates was 6% for dicyclohexyl phthalate (DCHP) to 74% for di-2-ethylhexyl phthalate (DEHP). Results of note were also found for BBzP, DBP, and DnBP. The frequencies of detection for these three phthalates were 54, 31, and 55%, respectively. Mean concentrations were always higher than median concentrations. Interestingly, out of three vegetable oil samples collected, the highest phthalate concentrations were found in the virgin olive oil in the glass jar instead of being found in the two oil samples in

plastic containers as would have been expected. Condiments (i.e., tomato ketchup, pancake syrup, barbecue sauce) had the lowest consumption rate of 0.2 g/kg/day in our sample categories, while infant foods had the highest consumption rate of 56 g/kg/day. The intakes of phthalates are comparable between adults and infants on a body weight basis, with the exception of DEHP, where the infant baby food intake is more than twice that of adults at 4.2 µg/kg-day. DEHP had highest levels detected in all food types except beef, ranging from an average of 4 ng/mL for beverages to 300 ng/g for pork. DCHP was the least frequently found phthalate, quantified in only four samples, with three of these positives near or less than 1 ng/g. Adult intake ranged from 0.004 µg/kg-d for dimethyl phthalate (DMP) to 0.673 µg/kg-d for DEHP.

Table 1 reports the frequency of detection of the nine phthalates by food group. Table 2 presents the food group mean and median concentrations, calculated at ND = ½ DL and ND = 0.

Phthalates are widely present in U.S. foods. While calculated intakes for individual phthalates were an order of magnitude lower than established EPA Reference Doses (RfDs), cumulative exposure to phthalates and exposure to mixtures of other toxic chemicals found in food is of concern. A more representative survey of U.S. foods is indicated.

Total dietary intake of HBCD, with samples below the LOD estimated to zero, was 15.4 ng/day. The largest contribution to intake was from meat (12.5 ng/day), largely from pork (4.2 ng/day), and chicken (4.2 ng/day). HBCD intake from fish, vegetable products, and dairy and eggs was 0.9 ng/day, 2.0 ng/day and 0 ng/day, respectively. Of 36 individual food samples, fifteen (42%) had at least one detectable HBCD stereoisomer. α-HBCD was present in 13 samples (36%); β-HBCD was present in 3 samples (8%); and γ-HBCD was present in 8 samples (22%). Median (range) of α, β and γ- HBCD concentrations were 0.003 (<0.005 – 1.307), 0.003 (<0.005 – 0.019) and 0.005 (<0.010 – 0.143) ng/g ww, respectively.

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Table 1. Detection frequencies of phthalate esters by food group from Albany, New York, USA

Food	Number of samples	Detection Frequency N (%)								
		DMP	DEP	DIBP	DBP	DNHP	BBzP	DCHP	DEHP	DNOP
Beverages	8	2 (25%)	0	3 (38%)	0	0	0	0	1 (13%)	0
Milk	2	0	1 (50%)	1 (50%)	1 (50%)	0	1 (50%)	0	2 (100%)	1 (50%)
Other dairy	9	4 (44%)	6 (67%)	7 (78%)	5 (56%)	3 (33%)	6 (67%)	1 (11%)	9 (100%)	1 (11%)
Fish	5	2 (40%)	3 (60%)	2 (40%)	2 (40%)	1 (20%)	2 (40%)	0	4 (80%)	0
Fruits/Vegetables	5	0	1 (20%)	4 (80%)	0	0	2 (40%)	0	2 (40%)	0
Grain	7	5 (71%)	7 (100%)	6 (86%)	6 (86%)	3 (43%)	7 (100%)	0	7 (100%)	0
Beef	2	1 (50%)	2 (100%)	0	0	1 (50%)	1 (50%)	0	0	1 (50%)
Pork	4	2 (50%)	4 (100%)	1 (25%)	0	0	1 (25%)	0	3 (75%)	1 (25%)
Poultry	6	3 (50%)	5 (83%)	0	0	1 (17%)	2 (33%)	0	5 (83%)	0
Meat and meat products	13	7 (54%)	12 (92%)	1 (8%)	0	2 (15%)	4 (31%)	0	9 (69%)	2 (15%)
Vegetable oils	3	1 (33%)	0	2 (67%)	1 (33%)	1 (33%)	3 (100%)	1 (33%)	2 (67%)	1 (33%)
Condiments	6	3 (50%)	3 (50%)	5 (83%)	3 (50%)	0	4 (67%)	1 (17%)	5 (83%)	1 (17%)
Infant food	7	0	4 (57%)	5 (71%)	2 (29%)	0	6 (86%)	1 (14%)	7 (100%)	2 (29%)
Total	65	24 (37%)	37 (57%)	36 (55%)	20 (31%)	10 (15%)	35 (54%)	4 (6%)	48 (74%)	8 (12%)

Table 2. Mean and median food group concentrations (ng/g whole weight) of phthalate esters displayed as ND = ½ DL / ND = 0, from Albany, New York, USA

Food	Statistic	DMP	DEP	DIBP	DBP	DNHP	BBzP	DCHP	DEHP	DNOP
Beverages	Mean	0.13/0.06	0.1/0	0.29/0.23	0.7/0	0.1/0	0.1/0	0.1/0	3.89/2.28	0.5/0
	Median	0.1/0	0.1/0	0.1/0	0.7/0	0.1/0	0.1/0	0.1/0	1.85/0	0.5/0
Milk	Mean	0.1/0	0.17/0.12	0.2/0.15	1.5/1.15	0.1/0	0.55/0.5	0.1/0	48.55/48.55	1.51/1.26
	Median	0.1/0	0.17/0.12	0.2/0.15	1.5/1.15	0.1/0	0.55/0.5	0.1/0	48.55/48.55	1.51/1.26
Other dairy	Mean	0.48/0.42	1.37/1.34	1.91/1.89	104.7/104.39	1.25/1.18	4.22/4.19	0.3/0.21	143.78/143.78	2.76/2.31
	Median	0.1/0	0.66/0.66	0.79/0.79	4.77/4.77	0.1/0	1.2/1.2	0.1/0	92.8/92.8	0.5/0
Fish	Mean	0.21/0.15	0.6/0.56	1/0.94	11.02/10.6	0.13/0.05	1.61/1.55	0.1/0	31.72/31.35	0.5/0
	Median	0.1/0	0.86/0.86	0.1/0	0.7/0	0.1/0	0.1/0	0.1/0	39.6/39.6	0.5/0
Fruit/ Vegetables	Mean	0.1/0	0.12/0.04	0.55/0.53	0.7/0	0.1/0	0.67/0.61	0.1/0	6.2/5.09	0.5/0
	Median	0.1/0	0.1/0	0.48/0.48	0.7/0	0.1/0	0.1/0	0.1/0	1.85/0	0.5/0
Grain	Mean	0.3/0.27	12.55/12.55	3.54/3.52	15.87/15.77	0.23/0.17	5.92/5.92	0.1/0	61.6/61.6	0.5/0
	Median	0.34/0.34	1.17/1.17	1.64/1.64	5.14/5.14	0.1/0	4.65/4.65	0.1/0	50.6/50.6	0.5/0
Beef	Mean	0.18/0.13	0.64/0.64	0.1/0	0.7/0	2.47/2.42	0.61/0.56	0.1/0	1.85/0	3.57/3.32
	Median	0.18/0.13	0.64/0.64	0.1/0	0.7/0	2.47/2.42	0.61/0.56	0.1/0	1.85/0	3.57/3.32
Pork	Mean	0.33/0.28	0.55/0.55	6.25/6.18	0.7/0	0.1/0	0.23/0.15	0.1/0	300.28/299.82	2.86/2.49
	Median	0.16/0.11	0.59/0.59	0.1/0	0.7/0	0.1/0	0.1/0	0.1/0	20.63/20.63	0.5/0
Poultry	Mean	0.15/0.1	0.41/0.4	0.1/0	0.7/0	0.21/0.12	0.66/0.6	0.1/0	18.64/18.33	0.5/0
	Median	0.15/0.1	0.33/0.33	0.1/0	0.7/0	0.1/0	0.1/0	0.1/0	14.8/14.8	0.5/0
Meat & meat products	Mean	0.22/0.17	0.49/0.48	1.99/1.9	0.7/0	0.51/0.43	0.48/0.41	0.1/0	101.77/101.2	1.7/1.28
	Median	0.2/0.2	0.45/0.45	0.1/0	0.7/0	0.1/0	0.1/0	0.1/0	7/7	0.5/0
Vegetable oils	Mean	1.2/1.14	0.1/0	3.2/3.17	3.53/3.07	0.19/0.12	153.85/153.85	14.27/14.2	116.92/116.3	0.84/0.5
	Median	0.1/0	0.1/0	0.25/0.25	0.7/0	0.1/0	2.2/2.2	0.1/0	48.9/48.9	0.5/0
Condiments	Mean	0.33/0.28	0.77/0.72	1/0.98	15.36/15.01	0.1/0	1.99/1.96	0.13/0.05	30.42/30.11	1.19/0.77
	Median	0.2/0.15	0.16/0.11	0.81/0.81	1.6/1.25	0.1/0	1.33/1.33	0.1/0	20.6/20.6	0.5/0
Infant food	Mean	0.1/0	0.35/0.31	0.77/0.74	1.14/0.64	0.1/0	3.36/3.35	0.18/0.1	75.06/75.06	2.5/2.14
	Median	0.1/0	0.28/0.28	0.22/0.22	0.7/0	0.1/0	2.37/2.37	0.1/0	29.4/29.4	0.5/0

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