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Occurrence and Dietary Exposure of Adult Population to Phthalates in Hong Kong

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Abstract

This study aimed (1) to determine the levels of seven phthalates including Di-Ethyl Phthalate (DEP), Di-n-Butyl Phthalate (DnBP), Butyl Benzyl Phthalate (BBzP), Di-(2-Ethylhexyl) Phthalate (DEHP), Di-n-Octyl Phthalate (DnOP), Di-isononyl Phthalate (DiNP) and Di-isodecyl Phthalate (DiDP) in selected foods that are commonly consumed in Hong Kong as well as foods those are reported to be adulterated with phthalates; (2) to estimate the dietary exposure to phthalates of the Hong Kong adult populations at territory-wide scale; and (3) to assess the health risk associated with the exposure. Total Diet Study (TDS) approach was used to assess the associated health risk to the local people. Among the seven phthalates examined, DEHP was the most commonly detected phthalate, followed by DiNP, DnBP, BBzP, DiDP, DEP and DnOP. The maximum detected levels were found to be 23, 43, 93, 560, 3,500, 3,800 and 7,900 µg kg^{-1} for DnOP, DEP, BBzP, DnBP, DEHP, DiDP and DiNP respectively. It is believed that elevated levels of phthalates detected in isolated samples were more related to chemical nature of the food substrates. Food contact materials used in food manufacturing and packaging may also explain the situation. This study estimated that the dietary exposures to seven phthalates analyzed in the average adult consumer population ranged from a low of 0.098 µgkg-bw^{-1} day^{-1} for DnOP (upper bound) to a high of 4.8 µgkg-bw^{-1} day^{-1} in the case of DiNP. The exposure to both average and high consumers (95^{th} percentile, or "P95") of the adult populations were well within the corresponding Health-Based Guidance Values (HBGVs) for individual phthalate (maximum 13% of HBGVs). Furthermore, no age-sex population sub-group exceeded their respective HBGVs. The findings indicate that dietary exposures to seven phthalates analyzed in this study were unlikely to pose an unacceptable health risk to the Hong Kong population. The food group "cereal and its products" was the major contributor for DnBP, BBzP, DEHP, DnOP and DiNP dietary exposure, while non-alcoholic drinks and poultry were the major contributors for DEP's and DiDP's, respectively.

Keywords: Phthalates, Di-ethyl phthalate, Di-n-butyl phthalate, Butyl benzyl phthalate, Di-(2-ethylhexyl) phthalate, Di-n-octyl phthalate, Diisononyl phthalate, Diisodecyl phthalate, Dietary exposure.

Abbreviations: PVC-Poly Vinyl Chloride, NRC-National Research Council, NICNAS-National Industrial Chemicals Notification and Assessment Scheme, CDC-Centers for Disease Control and Prevention, HBGVs-Health-Based Guidance Values, TDS-Total Diet Study, DEP-Diethyl phthalate, DnBP-Di-N-Butyl Phthalate, BBzP-Butyl Benzyl Phthalate, DEHP-Di-(2-Ethylhexyl) Phthalate, DnOP-Di-N-Octyl Phthalate, DiNP-Di-isononyl Phthalate, DiDP-Di-isodecyl Phthalate, FCS-Food Consumption Survey, FEHD-Food and Environmental Hygiene Department, USDA-United States Department of Agriculture, NHANES-National Health and Nutrition Examination Survey, GC-MS/MS-Gas Chromatography-Tandem Mass Spectroscopy, UPLC-MS/MS-Ultra-Performance Liquid Chromatography-Tandem Mass Spectroscopy, LODs-Limits of Detection, LOQs-Limits of Quantification, LB-Lower Bound, EASY-Exposure Assessment System.

Introduction

Phthalate esters, or simply known as phthalates, are chemically known as benzene dicarboxylic di-esters. Nowadays, phthalates are found in many consumer products because these chemicals, especially those with longer side chains (e.g. C4 to C10), can impart flexibility to many types of otherwise rigid plastics products. One of the most common examples is products made with polymers of Poly Vinyl Chloride (PVC). Phthalates compounds do so by embedding themselves between long plastic polymer chains, thus increases the spacing between polymers and renders them with increased physical flexibility. In contrast, phthalates with shorter side chains (e.g. C1, C2, C4) are usually used as solvents and are detectable in plastics, cosmetics and personal care products (Human Biomonitoring Commission) [1]. For these reasons, phthalates can be recovered from a wide variety of consumer products, including but not limited to cling film, plastic sheets, containers, adhesives, detergents, lubricating oils, vinyl floorings, pharmaceuticals, personal care products, and hoses, inflatable and flexible toys. With such extensive uses, human exposures to phthalates are common [2].

Indeed, owing to the versatile nature of phthalates, their extensive use in plastics products and their ubiquitous presence in the environment, human could be exposed to phthalates through various means including ingestion, direct skin contact (e.g. personal care products, vinyl flooring, toys) and inhalation (e.g. indoor air, house dust). However, food remains as a main source of adult exposure to phthalates. It is believed that most of these exposures are a result of phthalates leached out from food contact materials used in packaging materials, food processing machineries, contaminated food and drinking water [3-5]. So far, Health-Based Guidance Values (HBGVs) are only available to the seven more commonly used phthalates compounds. As HBGVs are the basis of quantitative risk assessment, this study is meant to be a study of these seven specific phthalates in food. Availability of HBGVs is a key element

in quantitative dietary exposure. It is concluded that HBGVs are clearly established on seven phthalates compound. When multiple HBGVs are available for the same phthalates, we would accord priority to international standards over regional standards and/or the more updated standard for this risk assessment study.

Even though Bradley, et al., [6] showed low levels of phthalic monoesters, mono-n-butyl phthalate and mono-ethylhyexyl phthalate, were detected in several of the Total Diet Study (TDS) animal-based food groups, they didn't contribute significantly to dietary exposure. Therefore, metabolites of phthalates with short mono-alkyl chain were excluded from this study. Despite overseas studies demonstrated phthalates as contaminants generally pose low health risk to the public, there has been confusion and ongoing concern from public in relationship to its possible developmental effect on male reproductive system in animal studies [2]. Therefore, this study aimed to:

- Determine the levels of seven phthalates (i.e. Di-Ethyl Phthalate (DEP), Di-N-Butyl Phthalate (DnBP), Butyl Benzyl Phthalate (BBzP), Di-(2-Ethylhexyl) Phthalate (DEHP), Di-N-Octyl Phthalate (DnOP), Di-isononyl Phthalate (DiNP) and Di-isodecyl Phthalate (DiDP)) in selected foods that are commonly consumed in Hong Kong as well as foods that are reported to be adulterated with phthalates before, either through overseas studies or local data so as to provide baseline situation.
- To estimate the dietary exposure to phthalates of the Hong Kong adult populations at territory-wide scale.
- To assess the health risk associated with the exposure, if any.

Materials and Methods

Food Consumption Data

The food consumption data were taken from the Hong Kong population-based Food Consumption Survey (FCS) conducted by the CFS in 2005-2007 [7]. Through a quota sampling by gender and age groups, 5008 Hong Kong adults aged 20-84 years were invited to complete two non-consecutive 24-h dietary intake (24-h recall) questionnaires. Two separate recalls were obtained from each respondent, the first in person and the second by telephone [7]. This method has been shown in the United States Department of Agriculture (USDA) National Health and Nutrition Examination Survey (NHANES) to be feasible and valid [8]. During each of these interviews, the interviewer asked the respondent to recall in detail all the food and beverages consumed during the 24-h period of the interview day. The body weight of each respondent was weighted by the interviewer with a calibrated balance. To elicit the required detail and limit underreporting, a multiple pass interview method was used involving asking the respondent to review his/her food intake several times with clarifying probes about ingredients, preparation and amounts.

Standard bowl, plate, cup and spoon, as well as photographs of utensils in other sizes, were shown to the respondent to help him/her estimate the amount of food taken. The respondent was also required to have the Food Photo Booklet at hand for the second 24-h recall interview by telephone. The two interviews were arranged on non-consecutive days of the week and from 3 to 11 days apart, but not on the same day of the week, so that the foods consumed on those days would be more likely to be independent than if consumed on consecutive days or on the same day of the week. The survey results revealed that over 1,400 food items were being consumed by the Hong Kong people. The results were age-gender-weighted and they represent a population of about 5.394 million Hong Kong residents aged 20-84 years [7,9]. In FCS, 5,008 Hong Kong adults aged 20-84 years were successfully invited through a quota sampling by gender and age groups and completed two non-consecutive 24-h dietary intake questionnaires. Food records were subsequently coded into 1,400 food items.

Food Sampling and Preparation

The 150 TDS food items from 15 food groups were mapped with 1400 food items captured by FCS in order to cover the whole diet of the Hong Kong people. The mean levels of the TDS food items were assigned to the mapped FCS food items with an application of conversion factors taking reference to the differences in water content [9]. To cite an example, cooked white rice in TDS food was mapped to cooked white rice and congee in FCS. As a result, over 99% of the food intake of the Hong Kong people was covered in the dietary exposure estimation after food mapping. Taking into account the resource limitation and the likelihood of occurrence of detectable phthalates in different foods, 98 food items from 13 food groups (excluded 2 food groups via 'legumes, nuts, and seeds' and 'sugars and confectionery' in which there was no report on their occurrences) were selected for analysis.

Table S1 (supplementary file) shows the number of samples in each food group. In order to provide risk assessment in worst case scenario, food that are more likely to have higher phthalate content (e.g. food with higher fat contents or history of phthalates abuse) or commonly consumed would be selected for testing. A total of three hundred and eight samples were collected from retailers and wholesalers between November 2016 and April 2017 in Hong Kong. A more detailed list of the types of food samples collected could be found in Table S1with number of individual samples. Most of the collected samples, like prepackaged drinks, biscuits, hamburger and pizza, were analyzed as consumed. Those raw food items, such as fish, meat, etc., were prepared by steaming, boiling or peeling, before the analysis.

Chemical Analysis of Phthalates

In this study, edible portion of 308 samples were all subjected to testing of DEP, DnBP, BBzP, DEHP, DnOP, DiNP and DiDP. The collected samples were analyzed as consumed. In brief, the phthalate levels in food samples were analyzed by Gas Chromatography-Tandem Mass Spectroscopy (GC-MS/MS) except for DiNP and DiDP by Ultra-Performance Liquid Chromatography-Tandem Mass Spectroscopy (UPLC-MS/MS) [6,10]. Deuterated analogs of 7 phthalates were fortified quantitatively into a measured amount of sample. For solid samples or oil and fat samples, extraction was done with acetonitrile by ultra-sonication (10 minutes) and then followed by orbital shaking (30 minutes). After centrifugation, the sample extract was freezed under -20˚C overnight. For liquid samples or vegetable and fruit samples, n-hexane: acetone (1:1) was used as the extraction solvent. After centrifugation, the n-hexane layer was freezed under -20˚C overnight.

If necessary, the extract was further purified with dispersive solid phase extraction of various packing materials. For acetonitrile extract, evaporate 4 mL to 2 ml. For n-hexane extract, evaporate 2 mL to dryness and then reconstitute 2 mL of acetonitrile. Then proceed to instrumental analysis. Identification was confirmed by comparing the relative retention time and the ion ratios with those of the standards. The Limits of Detection (LODs) and the Limits of Quantification (LOQs) of the seven phthalates were 5 and 15 μg kg^{-1} respectively. The LOQs were established as the lowest quantifiable concentration tested. Method blanks were performed for each sample batch to assess any background contamination. The background levels of the 7 phthalates (especially for DBP and DEHP) should be less than their LODs.

To minimize background contamination of phthalates from the environments, the precaution measures undertaken included:

- Plastic wares were not used throughout the experimental process.
- Extraction solvent like n-hexane was purified by passing through activated aluminum oxide.

- Glasswares were baked at 400°C for at least 2 hours, stored in a desiccator containing aluminum oxide and rinsed with purified n-hexane before use.
- PTFE liner septum was used for injector of gas chromatograph.
- An isolator column was added after solvent delivery system for separating background contaminants from mobile phase of liquid chromatography.

Analytical Quality Assurance: The validation study was performed on the basis of the Eurachem guideline [11]. The LOQ was established as the lowest quantifiable concentration tested. Replicates (10) of spiked recovery experiments at level of LOQ were performed in each of sample matrices, rice, beer and pork. Recoveries and precisions were within 80-118% and <10% RSD respectively. The on-going performance were monitored by spiked recovery experiments at concentrations of 15-50 ng g^{-1} of real samples in duplicate, including beverages, dairy products, fish, meat, vegetables, fruits, cereals, edible oils and butter. The average spiking recovery percentages of the 7 phthalates ranged from 92-101% with RSD <10%.

Treatment of Non-Detected Results

In this study, data were treated with the Lower Bound (LB) and upper bound (Human Biomonitoring Commission-UBA) approach. That is, at the LB, results below the LOD were replaced by zero whilst at the UB; results below the LOD were replaced by the value reported as the LOD. This approach compares the two extreme scenarios, based on the consideration that the true value for results less than LOD may actually be any value between zero and the achieved LOD. The LB scenario assumes that the chemical is absent; therefore, to results reported as <LOD a value of zero is assigned. The UB scenario assumes that the chemical is present at the level of the LOD; thus, to results reported as <LOD a value of the corresponding LOD is assigned.

Dietary Exposure Estimates

In order to cover the whole diet of the Hong Kong population, a food mapping process was carried out by mapping the TDS food items with food items captured by FCS. The mean levels of the TDS food items were assigned to the mapped FCS food items with an application of conversion factors taking reference to the differences of water content [9]. For examples, cooked white rice was mapped to rice congee with a conversion factor of 0.5 as rice congee contains only half the amount of rice compared with cooked white rice of the same weight. The dietary exposures were then estimated individually by combining the assigned levels of mapped FCS food items with their corresponding food consumption amounts. A weighting based on the population distribution by age and gender in the 2006 Population By-census was applied to adjust for bias arising from the age-gender quotas [7,8]. The mean and 95th percentile exposure levels among the FCS respondents after weighting by age-gender were used to represent the dietary exposures of the average and the high consumer of the Hong Kong population, respectively. Dietary exposure estimation was performed with the aid of an in-house developed web-based computer system, Exposure Assessment System (EASY) that takes food mapping and weighting of data into consideration. All results greater than LOD were taken directly to dietary exposure estimation.

Results

Occurrence

Vast majority of the 308 samples analyzed (99%) had at least one phthalate detected at quantified levels and only four samples (1.3%) were free from seven phthalates analyzed. These four prepackaged samples included one konjac snack sample, lemon tea sample, juice drink sample and soda drink sample. The results tally with similar overseas studies that phthalates are widespread in food **(Figure 1)**.

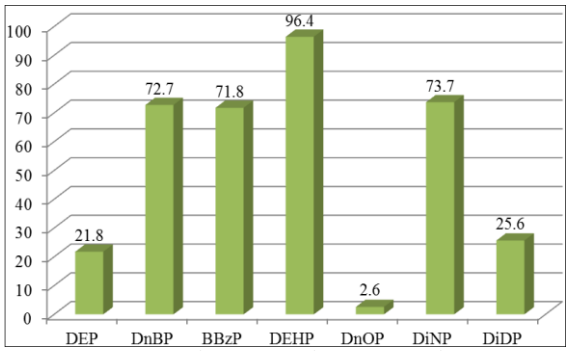

Figure 1: Detection frequencies of phthalates in food samples.

DEHP was the most commonly found phthalate in this study and was recovered from about 96% of the samples. This finding was in line with different studies [12-17]. In contrast, DnOP was only detected in about 2.6% of samples tested. The detection rate of the other five phthalates varied between around 20% and 75%. The maximum levels for the seven phthalates studied also varied widely. The maximum detected levels were 23, 43, 93, 560, 3,500, 3,800 and 7,900 µg kg^{-1} for DnOP, DEP, BBzP, DnBP, DEHP, DiDP and DiNP respectively. **Table 1a, and table 1b** shows the average phthalate levels in each food group while **Table S2** (supplementary file) shows the phthalate levels of each food item. Out of the 308 samples tested, only four samples (1.3% of the samples) were found to have phthalates at levels exceeding the Hong Kong's action levels.

Two edible oils got DEHP level higher than 1,500 µg kg^{-1}, included one peanut oil sample with 3,500 µg kg^{-1} and one olive pomace oil sample with 3,300 µg kg^{-1}. Besides, the olive pomace oil sample also got the highest DnOP level amongst all tested samples. Two Chinese white wine samples with DnBP at levels of 560 and 470 µg kg^{-1} exceeded the action level of 300 µg kg^{-1}. Migration of phthalates into these prepackaged products from plastic wares during the production process is the most possible source of contamination as they contain high level of fat or ethanol that can dissolve phthalates. Generally, the average levels of DiNP and DEHP in food were found to be much higher than the other phthalates (Table 1). The differences were even more pronounced for certain food groups like pork, oil and fats, and mixed dishes **(Figure 2)**. To certain extent, these elevated mean levels were explainable by individual samples or sub-groups of samples with high phthalates levels (e.g. 7,900 µg kg^{-1} DiNP in one minced pork sample, 3,800 µg kg^{-1} DiDP in one roasted duck sample, 3,500 µg kg^{-1} DEHP and 1,500 µg kg^{-1} DiNP in one peanut oil sample, and 3,500 and 900 µg kg^{-1} DEHP and 1,100 and 1,500 µg kg^{-1} DiNP in two olive pomace oil samples respectively).

A minced pork sample was also found to have the highest DiNP level of 7,900 µg kg^{-1} amongst the same group. Subsequently, a small scale study was conducted on 20 minced meat samples (14 minced pork, 5 minced beef and 1 minced chicken) that were packed with wrapping film. **Figure S1** (supplementary file) showed the levels of phthalates in these minced meat samples. Two (10%) minced pork samples collected from same supermarket (but at different branches) were detected with higher than average levels of DiNP (8,600 µg kg^{-1} and 7,300 µg kg^{-1} against the average of 848 µg kg^{-1} for 20 follow-up samples). Besides, DEHP of these two samples are also much higher than the rest minced meats (680 µg kg^{-1} and 360 µg kg^{-1} against their average of 103 µg kg^{-1}). Both the DiDP and DEHP levels of these 2 minced pork samples have not exceeded the Hong Kong's action levels of 9,000 and 1,500 µg kg^{-1} respectively. For a 60 kg adult has to take at least one kilogram of

these minced meat every day for prolonged period before risk of adverse health effect could not be confidently excluded. Risk of ill health from usual consumption is therefore unlikely.

Amongst these samples, only one of these wrapping films was not composed of PVC, but polyethylene. It is well know that plasticizers are added to PVC so as to soften the texture. However, it is not logical to correlate the issue with PVC wrapping film as similar PVC wrapping film was used in another 17 samples. Therefore, the source of contamination is still an unsolved mystery. Similarly, the high level of DiDP found in a roasted duck is another mystery as the package used was composed of polypropylene that contain high percentage of phthalate is unlikely. Further investigation in these areas should be conducted in future.

Food Group	Average phthalate level (μg kg^{-1})						
	DEP	DnBP	BBzP	DEHP	DnOP	DiNP	DiDP
Cereal and its products	5.0	21	32	60	5.0	55	8.8
Vegetables	5.0	5.6	9.1	59	5.0	20	15
Fruits	7.8	7.8	6.5	46	5.0	23	5.1
Meat, poultry and its products	5.5	43	15	110	5.0	310	84
Eggs	5.0	5.0	7.6	40	5.0	5.5	5.0
Fish and seafood	6.8	17	13	67	5.2	55	8.5
Dairy products	5.1	16	31	47	5.0	39	5.2
Oils and fats	5.2	8.1	6.9	350	6.4	330	170
Beverage, alcoholic	10	100	5.5	110	5.0	5.0	5.0
Beverages, non-alcoholic	8.3	13	10	18	5.0	8.9	5.0
Mixed dishes	5.0	48	10	210	6.1	750	8.4
Snacks	5.0	12	15	80	5.0	73	35
Condiments and sauces	5.0	50	6.7	210	5.0	110	18
Overall	6.0	23	15	100	5.2	130	37

Table 1a: Upper bound-Average phthalate levels by food groups.

Food Group	Average phthalate level (μg kg^{-1})						
	DEP	DnBP	BBzP	DEHP	DnOP	DiNP	DiDP
Cereal and its products	0.0	20	31	60	0.0	54	4.5
Vegetables	0.0	1.8	8.8	59	0.0	18	11
Fruits	5.4	5.0	5.4	46	0.0	21	0.40
Meat, poultry and their products	1.1	43	14	110	0.0	310	81
Eggs	0.0	0.0	7.6	40	0.0	3.9	0.0
Fish and seafood	4.3	16	12	67	0.4	54	4.3
Dairy products	1.0	13	31	48	0.0	41	0.63
Oils and fats	0.69	4.9	3.7	350	2.3	330	170
Beverage, alcoholic	8.2	100	1.4	110	0.0	0.0	0.0
Beverages, non-alcoholic	5.8	11	7.2	16	0.0	4.4	0.0
Mixed dishes	0.0	48	9.0	210	1.7	750	5.1
Snacks	0.0	10	14	79	0.0	72	32
Condiments and sauces	0.0	50	2.5	210	0.0	110	16
Overall	2.1	21	14	100	0.33	130	34

Note: *LOD = 5 μg kg^{-1}, LOQ = 15 μg kg^{-1}.

Table 1b: Lower bound-Average phthalate levels by food groups.

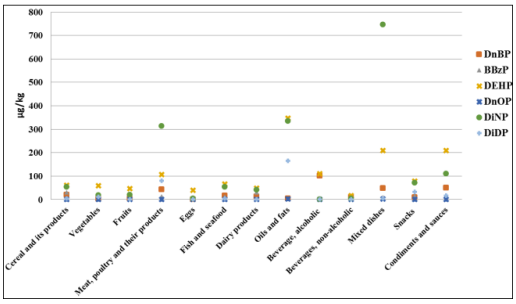

Figure 2: Average levels of phthalates by food groups (LB).

Dietary Exposure

Overall, the dietary exposures to the seven phthalates tested in average local adult consumer population were estimated from a low of 0.011-0.098 μg kg-bw^{-1} day^{-1} for DnOP (LB-UB) to a high of 4.8 μg kg-bw^{-1} day^{-1} for DiNP **(Table 2)**. As the maximum dietary exposure contributed only 13% of HBGV even for high consumers (P95), the result indicates health risk from phthalates to local adult population is quite remote from public health point of view. Actually, similar findings were seen for the dietary exposure to individual age-sex population sub-groups for both average and high consumers, meaning that there is no evidence of particular higher health risk to any age/sex subgroup of adult population from dietary exposure to phthalate **(Table S3)** (supplementary file). For the seven phthalates analyzed, the overall dietary exposures for both average and high consumers were low for adult populations in Hong Kong. This conclusion also applied to all sub-populations of different ages and sexes of local adult population. At current levels of phthalate detection, the local adult populations would not experience adverse health outcome due to exposure to the seven phthalates analyzed.

Discussion

Major Food Group Contributors

The relative contribution of each food group to overall LB phthalates exposure would be employed. LB figures would be used as they are considered a better reflection of the actual contribution to overall exposures, especially for those with a large percentage of samples below detectable levels. **Table 3** summarized the top three food group contributors of each phthalate. For DEP, the largest contributor in terms of food groups to average consumers is non-alcoholic drinks (0.0160 μg kg-bw^{-1} day^{-1} (LB), 48% of the DEP exposure), followed by fruits (0.0088 μg kg-bw^{-1} day^{-1} (LB), 26% of the DEP exposure), alcoholic drinks (0.0040 μg kg-bw^{-1} day^{-1} (LB), 12% of the DEP exposure) and fish (0.0025 μg kg-bw^{-1} day^{-1} (LB), 7.3% of the DEP exposure).

For DnBP, BBzP, DEHP, DnOP and DiNP, the largest contributor in terms of food groups for average consumers is cereal and its products, which accounts for about 73 to 97% of the dietary exposure to the five phthalates. After cereal and its products, food groups like fruits, vegetables, meat, poultry and their products were among the more prevalence contributors in terms of food groups to average consumers was meat, poultry and their products (60%). The contribution from cereal and its products was shrunk to 19%, followed by vegetables (15%). Although the food groups "cereal and its products" was a major contributor to many of the phthalates in the adult population, this can be explained by their relatively high consumption amount by local adults. Since the overall dietary exposure was way below corresponding HBGVs, the finding does not carry the inference that consumption of "cereal and its products" is hazardous. To be more exact, our assessment confirmed that there is no need to modify the dietary habit as the overall exposure to the seven phthalates is well below the relevant HBGVs.

We also noticed that relatively high phthalates levels were detected in a number of samples, citing the situation of mixed dishes, pork products and edible oil below. For mixed dishes comprising primarily of hamburgers, pizza and prepackaged lunchbox, higher levels of DiNP (2,100 and 3,800 μg kg^{-1} in two different samples) and DEHP (990 μg kg^{-1}) were more commonly found among prepackaged lunch boxes in microwavable packing. In contrast, these phthalates were much lower in pizza and hamburgers in alternative forms of product packing. In the case of pork samples, the higher mean DiNP level for was mainly contributed by a minced pork sample with a particularly high DiNP level of 7,900 μg kg^{-1}, where the other pork samples have much lower levels of DiNP which range from 7.4 to 870 μg kg^{-1} DiNP. Among all speculations, the use of PVC-based packaging films is suspected as the most

possible contributing factor. Regarding different types of edible oil sampled, higher levels of phthalates were found in samples of some peanut oil (DiNP in three peanut oil ranged from 630 to 1,500 µg kg^{-1} and one of them has a DEHP level of 3,500 µg kg^{-1}) and samples of olive oil (DiNP ranges from 350 to 1,300 µg kg^{-1}, and one olive oil sample has 1,200 µg kg^{-1} of DiDP.)

While the levels in these samples will not cause health issues, it was believed that exposure to those phthalates chemicals can be further reduced by modification in the process of products manufacture and packaging for some products. Actually, there is no cause for undue alarm even for samples with elevated levels of phthalates as highlighted above. The existing action levels were established to screen out food that had been intentionally adulterated with phthalates. As both edible oil and ethanol (spirits) are lipophilic in nature and extract phthalates readily from the plastic polymers upon direct physical contact, these results were not of surprise and do not point to intentional adulteration as in the 2011 Taiwan plasticizer incident. Risk assessment also confirmed that all these samples would not cause adverse health problem from phthalates upon usual consumption. Besides, as food contact materials used in food manufacturing and packaging process may also explain the situation, it is therefore believed that exposure to those phthalates chemicals can be further reduced by modification in the process of products manufacture and packaging.

Co-occurrence of phthalates

Amongst 308 samples tested for phthalates, co-occurrence of phthalates was observed in most of the samples. An olive oil samples was found to contain 7 targeted phthalates in detectable amount. As DEHP was the most commonly found phthalates, the co-occurrence rates of DEHP with DnBP, BBzP or DiNP were roughly the same of 71, 71 and 72% respectively. The top three food groups detected with higher sum of average levels (LB) of seven phthalates were "mixed dishes" (1,000 µg kg^{-1}, which were mainly contributed by DiNP and DEHP), "oils and fats" (860 µg kg^{-1}, which were mainly contributed by DEHP, DiNP and DiDP) and "meat, poultry and their products" (680 µg kg^{-1}, which were mainly contributed by DiNP and DEHP). Co-occurrence of phthalates can arise from phthalates are ubiquitous presence in the environment.

International Comparison

Overseas data on phthalate exposure were retrieved for comparison **(Table 4)**. Despite that sample coverage, methodology and analytical methods differ, the average adult exposure data in this study are considered at comparable levels. It was noted that the exposure of DiNP in Hong Kong is the highest amongst recent studies. It is likely due to the exceptionally high DiNP level found in 3 individual samples, via minced pork, peanut oil and olive oil, and 3 olive pomace oils. Furthermore, DiDP and DiNP are the least studied phthalates as they get lowest sensitivity amongst other phthalates.

Phthalate	HBGV (µg kg-bw^{-1} day^{-1})	Average Exposure		High Consumer (P95) Exposure	
		Exposure (LB-UB) (µg kg-bw^{-1} day^{-1})	%HBGV (LB - UB)	Exposure (LB-UB) (µg kg-bw^{-1} day^{-1})	%HBGV (LB-UB)
DEP	5,000	0.034-0.11	0.0007-0.0021	0.088-0.19	0.0018-0.0039
DnBP	10	0.37-0.39	3.7-3.9	0.73-0.75	7.3-7.5
BBzP	500	0.27-0.29	0.054-0.058	0.48-0.52	0.10-0.10
DEHP	25	1.7-1.7	6.6-6.6	3.3-3.3	13-13
DnOP	150	0.011-0.098	0.0027-0.024	0.025-0.17	0.0062-0.043
DiNP	150	4.8-4.8	3.2-3.2	11-11	7.2-7.2
DiDP	400	0.096-0.18	0.064-0.12	0.49-0.57	0.33-0.38

Table 2: The exposure of seven phthalates for average and high consumers.

	First	Second	Third	%contribution of top 3
DEP	Beverage, non-alcoholic (48%)	Fruits (26%)	Beverage, alcoholic (12%)	86
DnBP	Cereal and its products (74%)	Meat, poultry and its products (10%)	Beverage, non-alcoholic (7%)	91
BBzP	Cereal and its products (73%)	Fruits (5%)	Vegetables (5%)	83
DEHP	Cereal and its products (76%)	Vegetables (7%)	Meat, poultry and their products (7%)	90
DnOP	Cereal and its products (97%)	Meat, poultry and its products (2%)	Oils and fats (1%)	99
DiNP	Cereal and its products (91%)	Meat, poultry and its products (8%)	Fruits (1%)	100
DiDP	Meat, poultry and their products (60%)	Cereal and its products (19%)	Vegetables (15%)	94

Note: Results from lower bound scenario are listed.

Table 3: Top three of the food groups contributing most to the long-term intake of the seven phthalates.

Average exposure (LB – UB) (µg kg-bw^{-1} day^{-1})								
Places	**DEP**	**DnBP**	**BBzP**	**DEHP**	**DnOP**	**DiNP**	**DiDP**	**Reference**
Belgium	0.039	0.16	0.088	1.45	---	---	---	(Fierens, et al. 2014)
China	0.051*	0.703*	0.022*	1.60*	---	---	---	(Guo, et al. 2012)
	0.14 – 1.33	5.62 – 6.30	0.44 – 1.67	6.03 – 6.38	0.00 – 1.27	0.69 -1.73	---	(Yang, et al. 2018)
Norway*	0.0024 – 0.022	0.026 – 0.038	0.030 – 0.051	0.40 – 0.44	0.024 – 0.031	0.48 – 0.49	0.024 – 0.046	(Sakhi, et al. 2014)
United Kingdom	0.15 – 0.3	0.2 – 0.3	0.03 – 0.4	2.4 – 2.9	---	---	---	(Bradley, et al. 2013)
United States*	0.033	0.184	0.085	0.673	0.021	---	---	(Schecter, et al. 2013)
Hong Kong	0.034 – 0.11	0.37 – 0.39	0.27 – 0.29	1.7 – 1.7	0.011 – 0.098	4.8 – 4.8	0.096 – 0.18	Present study

Note: * Based on medium bound --- Not studied.

Table 4: Comparison of adult dietary exposure to seven phthalates in various countries.

Conclusion

This is the first study to provide the estimate of the average dietary exposure of the adult population in Hong Kong to phthalates. The results confirmed that the studied phthalates are ubiquitous in food. Regarding the dietary exposure assessment, the exposure to both average and high consumers (95th percentile, or "P95") of the adult populations were well within the corresponding HBGVs (maximum 13% HBGV) for individual phthalate. Furthermore, no age-sex population sub-group had exceeded the HBGVs. The food group "cereal and its products" was the major contributor for DnBP, BBzP, DEHP, DnOP and DiNP dietary exposure, while non-alcoholic drinks and poultry were the major contributors for DEP and DiDP dietary exposure, respectively. It is believed that elevated levels of phthalates detected in isolated samples were more related

to chemical nature of the food substrates. Food contact materials used in food manufacturing and packaging may also explain the situation. There was no evidence of intention adulteration of phthalates in food as occurred in 2011. All parties along the food supply chain, including the food manufacturers, distributors and retailers, should use appropriate food contact materials and minimize the occurrence of phthalates in food.

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