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Investigation on Risk of Phthalate Ester in Drinking Water and Marketed Foods

MEI-CHIN YIN* AND KUO-HSIUNG SU

Institute of Nutritional Science, Chungshan Medical & Dental College, Taichung, Taiwan, R.O.C.

ABSTRACT

The concentration of dibutyl phthalate and dioctyl phthalate in Taiwan's drinking water and several marketed foods were analyzed. The samples examined included 5 water sources, 3 vegetables, 4 fish and 3 meat products. While, the residue of dibutyl phthalate was found only in underground water samples, the residue of dioctyl phthalate was found in most tested food samples. The dioctyl phthalate content of fish samples was higher than that of vegetables followed by meat products. The results show that samples of Taiwan drinking water and food are contaminated with dioctyl phthalate and/or dibutyl phthalate. The contamination pathway may result from the disposal of the materials containing dibutyl phthalate and/or dioctyl phthalate leaching into soil and ground water and may reflect a potentially larger scale of environmental contamination.

Key words: Phthalate ester, food, water.

INTRODUCTION

Phthalate esters (PAEs) are widely used in industry as plasticizers, which enhance the flexibility of polymers[1]. Dibutyl phthalate (DBP) and dioctyl phthalate (DOP) are two lower-molecular-weight PAEs often used as required components of resins, cosmetics, textile and packaging materials[2]. Because of the widespread usage, industrial production of PAEs increased[3,4]. However, these chemicals are not chemically bound to the polymeric matrix and may migrate from plastics to enter the environment. The release of PAEs into water and soils may contaminate drinking water and food products[5].

The damage of PAE to human health is irreversible. PAE has been found to be toxic to red blood cells and the central nervous system, resulting in neurobehavioral disturbances[6]. The ingestion and inhalation of PAE can result in the accumulation of PAE in liver and/or kidney[7]. Animal studies have also shown that PAE accumulation in pregnant rats can act as a teratogen on the fetus and result in malformations of the cardiovascular, cerebral and skeletal systems[8,9].

The contamination of PAE in environment has been reported in USA, Canada and Japan. The residue of DBP and DOP in soil, water and foods has been found in Canada and Japan[5,10,11,12]. The industrial production and

Correspondence to: Mei-Chin Yin

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usage of DBP and DOP has annually increased in Taiwan. However, the information regarding water and food contamination by DBP and/or DOP in Taiwan remains unknown. The purpose of this study was to determine the residue of DBP and DOP in samples of drinking water and marketed food stuff in Taiwan and the implications these sources of contamination may have on human health.

MATERIALS AND METHODS

I. *Sample*

The water samples examined in this study included underground water, fountain water, mineral water, rain water, river water. Samples were collected from three different sources for each kind of water. Food samples included: milkfish, tilapia, hairtail, jumboprawn, pork, chicken, duck, white-leaf cabbage (water- and land-cultivated), water convolvulus (water- and land-cultivated) and Chinese cabbage. Food samples were randomly collected from three different local supermarkets. Land-cultivated vegetables were purchased from three different local traditional markets. 100 ml water samples or 100 g food samples taken from the edible parts were collected for DBP and DOP analyses.

II. *Analytical Procedure*

The method of Adams et al. (1995) was used to analyze the DBP and DOP residue in samples. DBP and DOP were extracted by mixing the water with 50 ml hexane and shaking vigorously. This extraction procedure was repeated three times. The hexane layers were collected and concentrated to 50 ul with a vacuum evaporator. One hundred grams food sample (edible portion) and 50 ml distilled water plus 50 ml hexane were mixed thoroughly. The mixture was filtered and the filtrate was collected by a separate funnel. Fifty ml hexane was added into the funnel followed by vigorous shaking. This extraction procedure was repeated three times. Hexane layers were collected and concentrated to 50 ul in a vacuum evaporator.

The contents of DBP and DOP in each sample was quantified by Gas Chromatography (GC) and compared to a standard. Shimadzu GC-14B gas chromatograph with an electron capture detection (ECD) was used. The GC separation was performed on a Perkin-Elmer model 3920 column: 6' x 0.25" (o.d.) x 2 mm (i.d.), column packing was 3% OV-101 on 80/100 supelcoport. The GC condition was as follow: injector temperature: 300°C; detector temperature: 300°C; initial column temperature: 160°C; rate: 10°C/min; final temperature: 290°C; KPa of H_2, N_2 and Air were: 50, 45 and 50, respectively. In this study, all chemicals were purchased from Sigma Chem. Co. (USA), the solvents were purchased from Fisher Chem. Co. (USA).

RESULTS AND DISCUSSION

The contents of DBP and DOP in water and foods are shown in Table 1. Recoveries averaged 95 and 93% for water and meat samples, respectively. In this study, DBP was found in underground water samples only. DOP was found in most food samples. No detectable level (≤ 1 ppb) of DOP or DBP were found in mineral water (commercial spring) and duck.

Both DBP and DOP were found in underground water. The DOP content of the examined water sources was found to be concentrated in the following order: river water > underground water > rain water > fountain water > mineral water. This result suggests that the quality of water in Taiwan is not safe and that river water is the most contaminated. Unfortunately, underground water and fountain water are used by many families for the daily needs. The high content of DOP in river water is evidence for DOP leakage from its original sources (industrial usage). In some areas of Taiwan, river water is used for rice, vegetable and fruit cultivation. Therefore, the presence of DOP in river water may further contaminate rice, vegetable and fruit products. The appearance of DOP in rain water suggests that DOP contamination is also airborne. The influence of the aerosol

Table 1. DBP and DOP contents of food or water (N=3)

	Item	DBP	DOP
Water	underground water	12±1ppb	18.4±1.5 ppb
	fountain water	–	14.3±1.1 ppb
	mineral water	–	–
	rain water	–	16.5±0.8 ppb
	river water	–	198.4±5.6 ppb
Vegetable	white-leaf cabbage (water-cultivated)	–	1.1±0.1 ppm
	white-leaf cabbage (land-cultivated)	–	1.3±0.1 ppm
	water convolvulus (water-cultivated)	–	1.2±0.1 ppm
	water convolvulus (land-cultivated)	–	1.4±0.3 ppm
	chinese cabbage (land-cultivated)	–	1.2±0.1 ppm
Fish	milkfish	–	1.3±0.1 ppm
	tilapia	–	1.8±0.3 ppm
	hairtail	–	1.8±0.4 ppm
	jumboprawn	–	1.3±0.2 ppm
Meat	pork	–	1.2±0.1 ppm
	chicken	–	1.1±0.1 ppm
	duck	–	-

"–" : No detectable level (≦ 1 ppb) of DBP or DOP was found.

DOP in air may include: 1) DOP inhalation by animals and accumulation in internal organs and tissues and 2) DOP contamination of water sources and subsequent food chain contamination water as well.

The three kinds of vegetable examined in this study are very popular and available throughout the whole year. DOP was found in all examined vegetables. For land-cultivated vegetables, the DOP content followed in order of concentration: water convolvulus > white-leaf cabbage > Chinese cabbage. For the same kind of vegetable, land-cultivated forms had higher DOP levels compared to water-cultivated ones. This suggests that the potential contamination of DOP in land (soil) seems more severe.

All fish samples examined had DOP residue. The content of DOP in fish samples followed in order of concentration: tilapia = hairtail > milkfish = jumboprawn. The amount of DOP found in fish samples was higher than water, vegetable and meat samples. This may result from fish living in contaminated water sources which promotes absorption and concentration of DOP in fish tissues. In this group, the hairtail was the only marine sample. The higher content of DOP in the hairtail further suggests that the ocean around Taiwan might be contaminated.

In the meat group, only lean muscle was examined for meat samples. DOP was found in pork and chicken. No detectable level of DOP was found in duck meat. The DOP may be absorbed and deposited in animal muscle tissue through food and/or water consumption. Both DOP and DBP are lipid soluble. Meat samples should provide the favored condition for PAE deposition. However, the result of this study showed that the contents of DOP in lean pork and chicken were lower than vegetable and fish groups. Further study may be necessary to examine other unknown reasons, e.g. animal feed, animal physiological conditions.

Based on the property of lipid solubility, the metabolic breakdown of PAE is very slow. These PAE materials have been shown to be deposited and concentrated in adipose tissue, liver, kidney and central nervous system subsequent to absorption[6]. The frequent consumption of foods containing PAE may result in the additive accumulation of PAE in adipose tissues and organs. Eventually, clinic signs appear. Unfortunately, acute and/or chronic toxicity have no cure.

In Canada, DBP was found in butter, margarine and diethyl phthalate esters (DEP) was found in pies[5]. In USA, PAE was also found in U.S. surface water and seafoods[12,13]. The results of this study support the thesis that improper treatment and/or disposal of PAE can result in the contamination of PAE in Taiwan's environment. This problem cannot be ignored because the presence of DBP and/or DOP in foods and water can seriously threaten the health of consumers and offspring. Further study is necessary to examine the content of PAE in other foods frequently consumed in Taiwan.

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市售食品及飲用水中含 Phthalate Ester 安全性評估

殷梅津* 蘇國雄

私立中山醫學院 營養科學研究所

摘　要

Dibutyl phthalate (DBP) 和 dioctyl phthalate (DOP) 為兩種應用廣泛的 phthalate ester (PAE)。DBP及DOP 已被大量應用於食品工業、紡織工業、汽車工業及化妝品業作塑膠可塑劑。由於工業產品不當的處理或棄置以及這類可塑劑的低安定性，使得DBP和DOP很容易滲漏出，進而造成環境的污染。

本研究以氣相層析法分析數種市售食品及飲用水中DBP及 DOP之含量。結果顯示DOP出現在大部分的檢測樣品中，而DBP 僅在地下水中被檢測出。食物及飲用水中 DBP及 DOP的殘留說明了 PAE 的滲漏已污染了土壤、水源以及食物。由於 PAE 對人體健康有極大的威脅，因此PAE的污染值得詳加探討以防止其惡化。

關鍵詞：飲用水，可塑劑，環境污染。