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RESEARCH ARTICLE

# Variation in accumulation and translocation of di-*n*-butyl phthalate (DBP) among rice (*Oryza sativa* L.) genotypes and selection of cultivars for low DBP exposure

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**Abstract** Di-*n*-butyl phthalate (DBP) is a typical endocrine-disrupting chemical with higher detection frequency and concentration in agricultural soil (particularly in paddy-field soil of Guangdong Province) of China. In this study, a greenhouse experiment was conducted to investigate variation in uptake and accumulation of DBP by 20 rice cultivars and to screen low DBP-accumulating cultivars. DBP concentrations in plants varied greatly with rice cultivars, growth stages, and tissues. The highest DBP concentrations in both roots and shoots were observed at the ripening stage, with concentrations 2–100-fold higher than those at tillering, jointing, and flowering stages. At the ripening stage, DBP concentrations decreased in the order of leaf > root > stem > grain, and significant differences of DBP concentrations were observed among various rice cultivars. Moreover, the magnitude of variation in DBP concentrations among various cultivars was greater in stems and grains than in roots and leaves. The translocation factors of DBP from roots to stems and from shoots to grains were <1.0, and those from stems to leaves were almost >1.0. Overall, cultivars Yuxiangyouzhan, Jinnongsimiao, Tianyou 122, and Wuyou 380 accumulated relatively lower DBP in grains, resulting in lower DBP exposure. The DBP uptake and translocation pathways in rice require further investigation.

**Keywords** Di-*n*-butyl phthalate · Rice · Cultivar · Accumulation · Translocation · Variation

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## Introduction

Phthalic acid esters (PAEs) are widely used as plasticizers and solvents in household and industrial products, including food packaging, vinyl flooring, building materials, and polyvinyl chloride (Guo et al. 2011; He et al. 2015). The global annually production of PAEs is approximately 6.0 million t (Xie et al. 2007), and China is one of the largest consumers of PAEs (He et al. 2015). As a result of their large-scale production and widespread application, PAEs have been released into the environment (Kong et al. 2012), and many studies have revealed the ubiquitous nature of PAEs in various environmental media (Yang et al. 2007; Cai et al. 2005, 2008a; Zeng et al. 2008; Guo et al. 2011; Shi et al. 2012; Benning et al. 2013; Kim et al. 2013 Domínguez-Morueco et al. 2014; Niu et al. 2014; He et al. 2015; Jambeck et al. 2015; Net et al. 2015). Human exposure to PAEs can occur via inhalation, dermal absorption, and dietary intake (Guo et al. 2011, 2012; Frederiksen et al. 2013; Kranich et al. 2014; Schecter et al. 2013; Wu et al. 2013a; Das et al. 2014). For example, a major incident of phthalate-contaminated foodstuffs occurred in Taiwan

between April and July 2011 (Wu et al. 2013a). Intake doses of di(2-ethylhexyl) phthalate (DEHP) and di-*n*-butyl phthalate (DBP) in the urban population of Delhi, India via different exposure pathways (e.g., food, air, dust, drinking water) reached or exceeded the established exposure limits; food was the major exposure source, contributing 67–74% of the estimated daily intake (Das et al. 2014). PAEs in the environment and entering food chain are receiving great concerns.

DBP, one of the most abundant PAEs, has been listed as a priority pollutant by the US Environmental Protection Agency (USEPA). In vivo studies demonstrated that DBP had adverse effects including antiandrogenic effects and even mutagenic toxicity (Aoki et al. 2011; Rusyn and Corton 2012; Kranich et al. 2014). DBP has been frequently detected in agricultural soils and crops at higher concentrations than most other individual PAE compounds (except for DEHP) (Vikelsøe et al. 2002; Yang et al. 2007; Cai et al. 2005, 2008a; Zeng et al. 2008; Mo et al. 2009; Rhind et al. 2013; Wang et al. 2013; He et al. 2015; Lu et al. 2016). Elevated concentrations of DBP were detected in agricultural soils adjacent to urban districts or associated with the use of plastic film (Zeng et al. 2008; Wang et al. 2013; He et al. 2015). The average DBP concentration in the paddy soils of Guangdong Province, China was higher than that in other land use types in the same region (Yang et al. 2007). DBP in soil can be taken up by crops, from which it enters the food chain (Cai et al. 2008b; Mo et al. 2009; Fu and Du 2011; Wu et al. 2013b; Li et al. 2014; Lü et al. 2014; Sun et al. 2015; Zhao et al. 2015; Lin et al. 2016; Wang et al. 2016). Some studies have indicated that vegetables and rice were contaminated by PAEs, including DBP (Mo et al. 2009; Fu and Du 2011; Guo et al. 2012; Lü et al. 2014; Lu et al. 2016), which posed a potential threat to human health. Nevertheless, to date, no study has reported on DBP uptake and accumulation by rice (*Oryza sativa* L.).

To reduce and eliminate PAEs from contaminated soils, remediation techniques including physical, chemical, and biological treatments have been conducted (He et al. 2015). However, many of these techniques are expensive, time-consuming, or impractical for use in large-scale soils contaminated by PAEs. In recent years, a novel alternative strategy of selecting or breeding low accumulation cultivars of contaminants has been proposed to reduce human exposure to contaminants through the food chain. Some scientists have reported low-accumulation crop cultivars of heavy metals (e.g., cadmium, arsenic) including rice (Ueno et al. 2010; Norton et al. 2012; Wu et al. 2016), wheat (Stolt et al. 2006), soybean (Fang et al. 2013), barley (Cao et al. 2014), water spinach (He et al. 2015), watercress (Wang et al. 2015), etc. and key genes that limit Cd accumulation in rice and barley have been identified (Ueno et al. 2010; Cao et al. 2014). Screening low-accumulation crop cultivars of contaminants and planting them in agricultural soils polluted by corresponding contaminants are both cost-effective and feasible. Nevertheless, study on the variations in uptake and accumulation of organic contaminants among crop cultivars is still scarce (Cai et al. 2008b; Zhao et al. 2015).

Rice is the main staple food for over half of the world's population, especially in developing Asian countries (Tesio et al. 2014). Numerous studies have reported that rice is able to accumulate heavy metals (e.g., cadmium, lead, arsenic, etc.) (Juang et al. 2012; Ueno et al. 2010; Wu et al. 2016) as well as other organic pollutants (e.g., polycyclic aromatic hydrocarbons (PAHs), where the accumulation in the grains is species-specific or cultivar-specific (Tao et al. 2006; Zhang et al. 2009; Ueno et al. 2010; Du et al. 2011; Liu et al. 2013; Wu et al. 2016). For example, significant genotypic variation was observed for arsenic accumulation and speciation in rice grains (Norton et al. 2012; Wu et al. 2016). Many countries have established limits for maximum concentrations of heavy metals in rice grains and their tolerable daily (or weekly) intakes or daily dietary exposure (Sommella et al. 2013). As mentioned above, the average PAE concentrations in agricultural soil of Guangdong Province were higher than other province in China (except Fujian and Xinjiang Province) (Cai et al. 2008a; Niu et al. 2014), and the average DBP and DEHP concentrations were higher in paddy soils of Guangdong Province than in other land use types and even exceeded the allowable soil concentration suggested by New York State of the USA (Yang et al. 2007). Moreover, the average concentrations of PAEs (including DBP) in the soil of China were much higher than those of other organic contaminants (e.g., PAHs) (Cai et al. 2008a). On the other hand, the average concentration of DBP in grains in New York (USA) was significantly higher than that in fruits/vegetables, meats, milk, etc. (Schecter et al. 2013). PAEs were also detected in cereals (including rice) and vegetables from China (Mo et al. 2009; Guo et al. 2012; Sui et al. 2014; Lu et al. 2016), implying that plant agricultural products are indeed contaminated by PAEs. A study showed that the main dietary food sources responsible for PAE intake in China were cereals (accounting for >39.44% of the total intake) (Sui et al. 2014). Nevertheless, whether DBP can be taken up by rice and translocated to the shoots and grains remain unknown.

The major objectives of this study were (1) to investigate the variation in the accumulation of DBP by different genotypic cultivars of rice (including seven normal cultivars and thirteen hybrid cultivars), (2) to investigate the distribution of DBP in different plant tissues (e.g., root, stem, leaf, and grain) and at different growth stages (e.g., tillering, jointing, flowering, and ripening), and (3) to identify low-DBP-accumulating rice cultivars.

## Materials and methods

### Soil preparation and spiking

DBP (CAS number 84-74-2) was used as a PAE model compound throughout this trial. DBP solution (Analytical grade,

>98.5% purity) for the pot experiment was purchased from Tianjin Chemical Reagent Factory, China. Paddy soil used in this study was collected from an uncontaminated experimental field (0–20 cm depth) of a university in Guangzhou. The soil was air-dried and passed through a 2-mm sieve. The soil contained 30.2 g/kg organic matter, 1.26 g/kg total N, 1.79 g/kg total P, 18.0 g/kg total K, 7.67 cmol/kg cation-exchange capacity, 33.5% sand, 18.5% silt, and 48% clay. The background concentration of DBP in soil was 0.19 mg/kg.

A portion of soil (10% of the total quantity of soil) was spiked with a DBP solution in acetone and mixed thoroughly. The spiked soil was transferred to a wide stainless steel pot, placed under a fume hood, and mixed every half hour to evaporate any traces of acetone and ensure to be homogenized. When the acetone had evaporated (48 h), the spiked soil (500 g) was mixed progressively with uncontaminated soil (4.5 kg) and homogenized for each pot. The final concentration of DBP in the treated soils, set according to the general concentrations observed in contaminated soils of the Pearl River Delta area, was 20 mg/kg (on a dry weight basis). The treated soils were packed into ceramic pots (23 cm inner diameter, 18 cm height; 5 kg dry weight soil per pot) and submerged in 2–3 cm water for 2 weeks before the rice seedlings were transplanted. Prior to cultivation, the soil of each pot was fertilized using 0.20 g/kg N, 0.15 g/kg P, and 0.15 g/kg K with urea, superphosphate, and potassium chloride, respectively, and mixed thoroughly again.

## Pot experiment

Twenty rice cultivars, including seven normal cultivars and 13 hybrid cultivars (Table 1) were used in this study. These cultivars are widely grown in Guangdong Province, China. The rice seeds were obtained from Guangdong Academy of Agricultural Science and South China Agricultural University, China. The seeds were surface sterilized in 0.5% $HgCl_2$ and soaked in water for 24 h at room temperature. The seeds were germinated under moist conditions in a light incubator (GXZ-280B, China) at 36 °C for 24 h. The germinated seeds were grown in a mixed substrate of uncontaminated soil and organic fertilizer. After 20 days, 15 uniform seedlings (clustered as three plants per pot) were selected and transplanted into the pots packed with soil. Each treatment was with four replicates. Immediately after transplanted, the pots were laid out in a completely randomized block design in a glass greenhouse. The pot soil was maintained under flooded conditions (with 2–3 cm of water above the soil surface) during the experiment. For tillering, jointing, and flowering stages, samples of rice tissues were collected from four replicates and combined, divided into shoots and roots. For ripening stage, rice plant samples were taken from each pot, washed orderly with tap and deionized water, and divided into roots, shoots, leaves, and grains. After freeze-dried (Thermo Heto PowerDry LL3000, Thermo Fisher Scientific, USA), rice tissues were cut using a stainless scissors into fragments 1–2 cm, mixed thoroughly. Then, the fragments of rice tissues were ground to a fine power using a pre-cleaned mortar and mixed again before analysis.

## Analytical procedures, quality assurance, and quality control

DBP in samples was extracted by ultrasonic-assisted extraction following USEPA method 3540C. Briefly, 2 g of plant samples was extracted in triplicate with 20 mL dichloromethane (HPLC grade) in an ultrasonic bath (KH-250E, Kunshan, China) for 10 min. The extracts were centrifuged at 4000 rpm for 5 min and then combined and concentrated to about 1.0 mL using a vacuum rotary evaporator (52-A, Yarong,

**Table 1** Cultivars of rice from different genetic background

| Cultivar name | Source[a] | Type | Cultivar name | Source | Type |
|---|---|---|---|---|---|
| Hemeizhan | Fengmeizhan/Hesizhan | N | Tianyou 998 | Tianfeng A/Guanghui 998 | H |
| Guinongzhan | Guangnongzhan × Xinaozhan/Guinongzhan | N | Tianyou 390 | Tianfeng A/Guanghui 390 | H |
| Hefengzhan | Fengmeizhan/Guanghezhan | N | Tianfengyou 316 | Tianfeng A/Shanhui 316 | H |
| Fengmeizhan | Xinguangmei/Zhongerzhan | N | Fengyousimiao | Yuefeng A/Guanghui 998 | H |
| Yuxiangyouzhan | TY36 × IR100/IR100 | N | Fengyou 428 | Yuefeng A/Guanghui 428 | H |
| Jinnongsimiao | Jinhuaruanzhan/Guinongzhan | N | Wufengyou 128 | Wufeng A/Guanghui 128 | H |
| Huahang 31 | Tehuazhan/H-31 × Huahang131 | N | Wufengyou 2168 | Wufeng A/Guanghui 2168 | H |
| Tianyou 103 | Tianfeng A/Jinhui 103 | H | Wuyou 308 | Wufeng A/Guanghui 308 | H |
| Tianyou 122 | Tianfeng A/Guanghui 122 | H | Yueza 889 | GD-1S/R889 | H |
| Tianyou 2168 | Tianfeng A/Guanghui 2168 | H | Peizataifeng | Aipei 64S/Taifengzhan | H |

*N* normal cultivar, *H* hybrid cultivar

[a] Source refers to female/male parents. Data was from China Rice Data Center http://www.ricedata.cn/variety/

China). A glass chromatography column packed with silica gel (10 g, 100 mesh) and anhydrous sodium sulfate (3 g) was used for purification of the extracts, and DBP was eluted with 50 mL dichloromethane (Cai et al. 2007; Schecter et al. 2013; Zhao et al. 2015). The final elute was concentrated again using a vacuum rotary evaporator and under a gentle stream of nitrogen.

DBP in the extract was measured using gas chromatography coupled with mass spectrometry (Shimadzu, GC-MSQP2010) in the selective ion monitoring mode. A fused-silica capillary column (DB-5MS; 30 m × 0.25 mm i.d.; 0.25 μm film thickness) was used for separation. The carrier gas was helium at a constant flow of 0.70 mL/min. The temperature of both injector and ion source was 250 °C. The oven temperature settings were as follows: 100 °C (hold for 2 min), increased to 110 °C at 35 °C/min, increased to 129 °C at 15.0 °C/min, and finally increased to 280 °C at 40.0 °C/min (hold for 4 min). Sample extract (1 μL) was injected in the splitless mode.

Analyte ionization was performed by electron ionization (70 eV), and signal acquisition was realized in the selected ion monitoring mode. A composite stock standard solution containing DBP (1000 μg/mL in dichloromethane, 99.8% purity) for instrumental analysis was purchased from O2si Smart Solutions (Charleston, SC, USA). Identification of DBP was based on matching the retention time with the standard DBP solution and characteristic ions (the primary characteristic ion of DBP being 149). The quantitative analysis was based on a five-point calibration curve (ranging from 0 to 4.0 μg/mL).

Care was taken to eradicate any potential contamination so as not to influence the results. No plastic material was used during the experiment. All glassware was soaked in a solution of $K_2CrO_4$–$H_2SO_4$–$H_2O$ (20 g:360 mL:20 mL) for 8 h or more and washed with tap and redistilled water before baking at 300 °C for 8 h. Glassware was wrapped in aluminum foil until use and pre-cleaned with acetone before use.

The instruments were calibrated daily using calibration standards. Peak area responses were used against concentrations of DBP and an internal standard to calculate the response factors. Additionally, a solvent blank, procedural blank, spiked blank (a standard spiked into the solvent), matrix spiked duplicates, and sample duplicates were routinely analyzed with each batch of 20 samples. The detection limit of DBP was 2.5 μg/kg. The DBP recoveries in the plant samples ranged from 80.8 to 90.3%.

### Statistical analysis

The results are expressed on a dry weight basis. Analysis of variance (ANOVA) was performed using Statistical Analysis System (SAS) software version 8.2. Differences among the means were evaluated using Duncan's test.

## Results and discussion

### Variation in DBP concentrations in rice plants at different growth stages

In this study, rice growth stages were classed into tillering, jointing, flowering, and ripening stages. DBP concentrations varied with growth stages and rice cultivars (Table S1, Fig. 1). DBP concentrations in rice shoots at tillering, jointing, and flowering stages were <0.5 mg/kg, and ~60% of them were <0.1 mg/kg; concentrations at the ripening stage ranged from 1.35 to 6.28 mg/kg, 10–60-fold higher than those at other stages. DBP concentrations in rice roots at three stages (except ripening stage) varied from 0.03 to 8.87 mg/kg with 90% <1.0 mg/kg; at the ripening stage, they were >1.0 mg/kg (2.21–8.87 mg/kg), indicating that the highest DBP concentrations in both roots and shoots were mostly observed at ripening stage. This differed from the uptake trend for heavy metals (e.g., copper, zinc, lead, arsenic) by rice roots, where the highest concentrations occurred at tillering stage and the lowest at ripening stage (Mo et al. 2002; Liu et al. 2013). Liu et al. (2013) reported the highest uptake and accumulation of lead by rice at tillering stage, with relative low accumulation at the other stages. This may be due to the inhibition of the metabolic and uptake abilities after the roots have accumulated the heavy metals (Mo et al. 2002).

In this study, the average DBP concentration in both roots and shoots of ripening stage increased dramatically compared

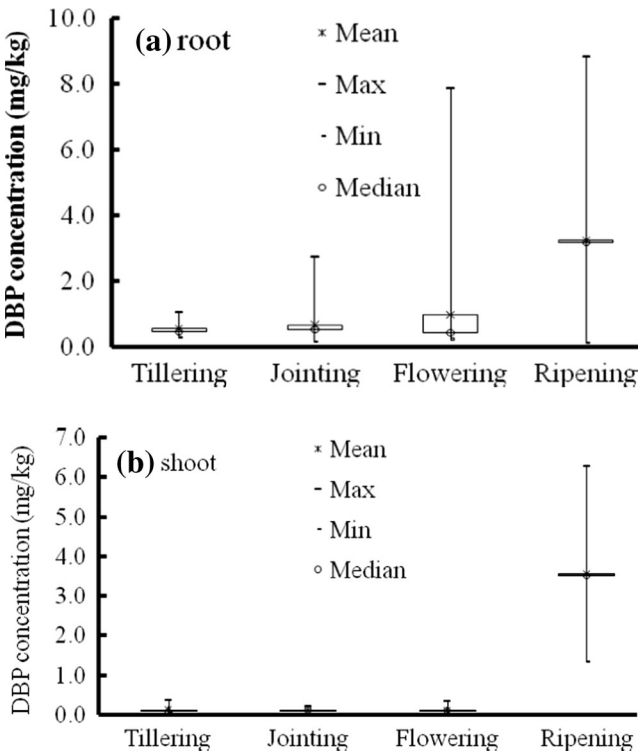


**Fig. 1** DBP concentration distribution of 20 rice cultivars at different growth stages

with the other stages (Fig. 1). This could be related to the relatively lower DBP concentrations in the soil and accumulating in roots at the three stages (except ripening stage), which did not influence root growth and DBP uptake abilities and then resulted in DBP uptake by the roots, followed by transportation to the shoots and finally accumulation in the plants. Our previous studies found that DBP could be taken up by the roots of water spinach (*Ipomoea aquatica*) and Chinese flowering cabbage (*Brassica parachinensis*) and translocated to the shoots (Zeng et al. 2005a, b; Cai et al. 2008b; Zhao et al. 2015). However, due to the rapid increase in plant biomass during tillering and flowering stages, DBP in the plants might be diluted to some degree, resulting in relatively lower DBP concentrations in the plants at jointing and flowering stages, while the increase in plant biomass between flowering and ripening stages was relatively small, and DBP concentrations in the plants were therefore higher. On the other hand, plant metabolism of organic contaminants might occur (Gao et al. 2013; Sun et al. 2013; Yu et al. 2013; Sun et al. 2015). For example, some scientists reported in vivo metabolism of PAHs by tall fescue (*Festuca arundinacea* Schreb.) (Gao et al. 2013), polybrominated diphenyl ethers by pumpkin plant (*Cucurbita maxima* × *Cucurbita moschata*) (Sun et al. 2013; Yu et al. 2013), DBP and DEHP by lettuce (*Lactuca sativa* L.), Quinault strawberry (*Fragaria* × *ananassa*.), and carrot (*Daucus carota* var. *sativus*) (Sun et al. 2015). Nevertheless, no study has reported the metabolism of PAEs in plants. Whether in vivo metabolism of DBP in rice plant will influence its accumulation variation among cultivar needs further research. Concerning the difference between roots and shoots, DBP concentrations at tillering, jointing, and flowering stages were higher in the roots, but at ripening stage, about 70% of cultivars DBP concentrations in the shoots were higher than in the roots, implying that DBP taken up by the roots during flowering and ripening stages was preferentially transported to the shoots. Additionally, some of the DBP in the shoots may have resulted from shoot uptake of DBP that evaporated from the soil. The detailed uptake pathways and affecting factors require further investigation.

Significant differences were present in the DBP concentrations in the shoots (or roots) among the various cultivars of rice. DBP variation among the cultivars was related to the different growth stages, and the observed trend was complicated. DBP concentrations of some cultivars were higher at certain growth stages, but lower at other growth stages, and vice versa (Table 2). For example, DBP concentrations in the shoots at four different stages were relatively lower in cultivars Guinongzhan and Wufengyou 2168, whereas those in the roots were relatively lower in cultivars Hemeizhan, Guinongzhan, Fengyou 428, and Wufengyou128. DBP concentrations in both shoots and roots of cultivar Guinongzhan were low at all growth stages. During the ripening stage, DBP concentrations in both shoots and roots were relatively low in the cultivars Hemeizhan, Guinongzhan, Hefengzhan, Fengyousimiao, Fengyou 428, and Yeza 889, but high in Fengmeizhan. DBP concentrations in the roots were higher in cultivars Peizataifeng and Tianyou 998 but were lower in the shoots. Overall, because inter-cultivar variation in DBP concentrations was quite high, no significant difference was observed for DBP concentrations in both roots and shoots between normal rice cultivars and hybrid rice cultivars (Table 1S, Figs. 2 and 3).

### Variation in DBP accumulation by different rice tissues

Rice tissues at ripening stage were divided into root, stem, leaf, and grain, and their DBP concentrations varied with different cultivars. Overall, DBP concentrations in the different tissues of various cultivars decreased in the order of leaf > root > stem > grain (Fig. 3). This distribution pattern was considerably different from that of heavy metals and PAHs accumulated by rice (Tao et al. 2006; Du et al. 2011; Liu et al. 2013). Several studies reported that after uptake, most heavy metals (e.g., cadmium, zinc, copper, lead) were retained in the roots, with only a small amount transported to the shoots; heavy metal concentrations in different tissues decreased in the order of root > shoot > ear (or grain) (Mo et al. 2002; Liu et al. 2013) or straw > husk > brown rice in the case of arsenic (Hu et al. 2013). Tao et al. (2006) reported that the distribution of PAHs in various tissues of rice plants decreased in the order of root > hull > leaf > internode > seed, and Du et al. (2011) reported that $^{14}$C-phenanthrene in rice tissues decreased in the order of root > leaf > shell > stem > grain. These results indicated that heavy metals and organic pollutants, including DBP, were unevenly distributed in different rice tissues.

In this study, the highest concentrations of DBP were observed in the leaves of most of cultivars (except Hefengzhan, Huahang 31, Tianyou 998, Fengyou 428, and Peizataifeng) (Fig. 3c). Moreover, DBP concentrations in the leaves of cultivars Fengmeizhan, Jinnongsimiao, Tianyou 103, Tianyou 122, Tianyou 390, and Wufengyou 128 were approximately twice as high as those in other tissues. Plants could take up organic contaminants such as PAHs and transport them to the aboveground parts by transpiration stream flux and even translocate them among the different tissues (Gao and Collins 2009). The transportation or translocation of contaminants in plants was somewhat cultivar specific. For example, Ueno et al. (2010) and Satoh-Nagasawa et al. (2012) found that the *OsHMA3* gene limited Cd translocation from rice roots to the aboveground tissues by selectively sequestrating Cd into the root vacuoles. Overexpression of *OsHMA3* from the low-Cd-accumulating cultivar selectively decreased the accumulation of Cd in rice, but *OsHMA3* from the high-Cd-accumulating cultivar had lost its function, probably because of the single amino acid mutation (Ueno et al. 2010). These results imply that accumulation and translocation of contaminants by plants was genotype-dependent.

**Table 2** DBP translocation factors of different tissues at ripening stage of rice

| | Cultivars | Root-stem | Stem-leaf | Shoot-grain |
|---|---|---|---|---|
| Normal | Hemeizhan | 0.96 ± 0.25a | 1.20 ± 0.47cd | 0.22 ± 0.09cd |
| | Guinongzhan | 0.98 ± 0.30a | 0.90 ± 0.84d | 0.21 ± 0.01cd |
| | Hefengzhan | 0.51 ± 0.10abc | 1.09 ± 0.23cd | 0.46 ± 0.27bcd |
| | Fengmeizhan | 0.43 ± 0.10bc | 5.41 ± 1.39ab | 0.17 ± 0.22cd |
| | Yuxiangyouzhan | 0.65 ± 0.28ab | 3.31 ± 1.71bc | 0.07 ± 0.05d |
| | Jinnongsimiao | 0.68 ± 0.21ab | 3.04 ± 0.76bc | 0.05 ± 0.04d |
| | Huahang 31 | 0.83 ± 0.22ab | 2.84 ± 0.98bc | 0.24 ± 0.10cd |
| Hybrid | Tianyou 103 | 0.61 ± 0.28ab | 3.42 ± 0.88bc | 0.12 ± 0.07d |
| | Tianyou 122 | 0.39 ± 0.23bc | 8.88 ± 4.11a | 0.05 ± 0.02d |
| | Tianyou 2168 | 0.54 ± 0.28abc | 2.38 ± 1.07bcd | 0.19 ± 0.14cd |
| | Tianyou 998 | 0.40 ± 0.23bc | 2.18 ± 1.47bcd | 0.44 ± 0.09bcd |
| | Tianyou 390 | 0.51 ± 0.19abc | 4.68 ± 1.57ab | 0.74 ± 0.08b |
| | Tianfengyou 316 | 0.56 ± 0.31abc | 2.76 ± 0.26bcd | 1.46 ± 0.79a |
| | Fengyousimiao | 0.58 ± 0.17ab | 2.12 ± 0.40bcd | 0.39 ± 0.07bcd |
| | Fengyou 428 | 0.72 ± 0.20ab | 3.53 ± 1.16bc | 1.58 ± 0.56a |
| | Wufengyou 128 | 0.11 ± 0.07c | – | 0.27 ± 0.06bcd |
| | Wufengyou 2168 | 0.57 ± 0.20ab | 1.98 ± 0.62cd | 0.25 ± 0.08cd |
| | Wuyou 308 | 0.68 ± 0.10ab | 2.35 ± 0.81bcd | 0.11 ± 0.08d |
| | Yueza 889 | 0.72 ± 0.28ab | 2.62 ± 0.88bc | 0.64 ± 0.40bc |
| | Peizataifeng | 0.37 ± 0.15bc | 1.12 ± 0.34cd | 0.23 ± 0.02cd |

Mean ± S.D. ($n$ = 3) followed by the same letters within a column were not significantly different ($P > 0.05$)

Higher DBP concentrations in the leaves of most of the cultivars may be partly attributed to uptake by the roots and transport to the different tissues and partly to leaf uptake and accumulation of DBP from the air. Recently, plant uptake of organic pollutants such as PAHs and PAEs has attracted considerable attention (Wild et al. 2006; Cai et al. 2008b; Gao and Collins 2009; Li and Chen 2014; Sun et al. 2015). For example, one study reported that the uptake of organic contaminants such as PAHs by plants from the soil to the roots was a major pathway, a significant fraction of shoot contamination resulted from aerial deposition of PAHs volatilized (Gao and Collins 2009). In this study, the Henry's law constant ($H$) of DBP ($Hc = 8.83 \times 10^{-7}$) (Staples et al. 1997) was lower than those of phenanthrene and pyrene ($Hc = 2.56 \times 10^{-5}$ and $1.14 \times 10^{-5}$, respectively) (Toronto Public Health 1998), implying that DBP could volatilize from the soil to some degree. Our previous study showed a positive correlation between leaf area and DBP concentrations in *I. aquatica* (Cai et al. 2004, 2008b), demonstrating that leaves could take up and accumulate DBP from the air. Nevertheless, our previous study indicated that DBP and DEHP in the shoots of *B. parachinensis* derived dominantly from root uptake, while leaf uptake of DBP and DEHP volatilizing from polluted soil was minor (Zeng et al. 2005a, b). The leaf areas of rice were relatively smaller than those of vegetables. Hence, the direct aerial deposition of DBP volatilized from contaminated soils might contribute slightly to the concentrations of DBP in leaves.

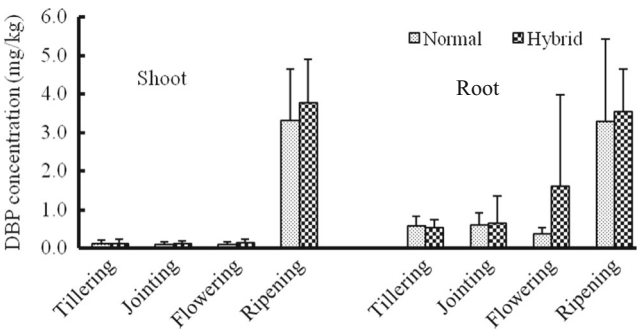


**Fig. 2** Average DBP concentrations in normal and hybrid rice cultivars

DBP concentrations differed greatly not only among plant tissues but also among cultivars (Fig. 3). For example, cultivar Peizataifeng showed a significantly higher DBP concentration in the roots compared with the other cultivars. DBP concentrations in leaves of cultivars Fengmeizhan and Tianyou 122 were significantly higher than those in most other cultivars. DBP concentrations in the roots and stems of cultivars Fengyousimiao, Wufengyou 128, and Yueza 889 were relatively lower compared with most other cultivars. These results indicated that DBP uptake and accumulation by rice was cultivar-specific. Furthermore, the magnitude of variation in DBP concentrations of stems and grains among the various cultivars was greater than those in the roots and leaves. This is different from the distribution of lead in the tissues of various rice cultivars, in which case the magnitude of variation in the

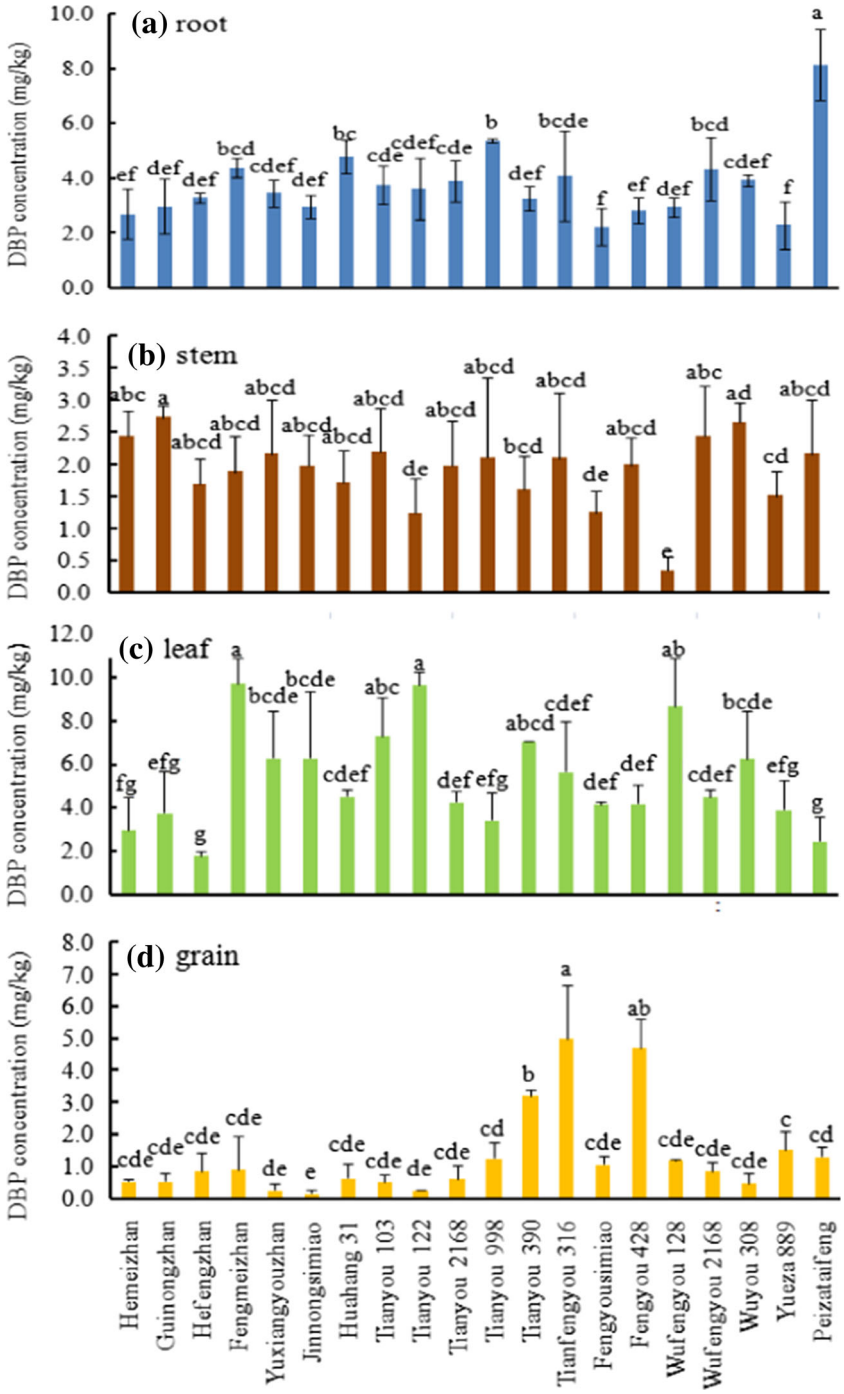


**Fig. 3** DBP concentrations of rice different tissues at ripening stage of various rice cultivars ($n = 3$)

concentrations in the ears and grains was greater than that in the shoots and roots (Liu et al. 2013). The variation in the accumulation of contaminants by rice tissues was related not only to the genotypes of the cultivars but also to the transportation of the contaminants to the tissues. For example, genotypic cultivars with higher radial oxygen loss had a stronger ability to increase their tolerance to arsenic by reducing arsenic mobilization in the roots and reduce arsenic accumulation in the shoots by limiting its translocation from the roots (Mei et al. 2012). It should be noted that significant differences in DBP concentrations in various tissues were observed among both normal (except in the grains) and hybrid rice cultivars (Fig. 3). The detailed mechanisms underlying the accumulation variation among cultivars and tissues need to be investigated through further experiments.

### Translocation of DBP to different rice tissues

To clearly describe DBP transportation and accumulation in different tissues, translocation factors (TFs; defined as the

DBP concentration ratio of stems to roots, leaves to stems, or grains to shoots) were used. The greater the TF value is, the stronger is the transportation ability. Our previous studies had shown that root uptake and transportation constituted the main source of DBP in the shoots of *B. parachinensis* and radish (*B. parachinensis*) (Cai et al. 2004, 2008b; Zeng et al. 2005a, b; Zhao et al. 2015). Here, it was assumed that DBP volatilized from the soil into the air, and therefore, uptake by rice shoots would be negligible. Rice TFs for DBP in the different tissues at ripening stage are shown in Table 2. TFs of DBP for the different cultivars and tissues differed significantly. The TFs of DBP from the roots to the stem were <1.0 (0.11–0.98), with the highest TFs occurring in cultivars Hemeizhan and Guinongzhan and the lowest in cultivars Wufengyou 128 and Tianyou 122. The TFs from the stems to the leaves were >1.0, and most of them were between 1.0 and 5.0, with cultivars Hemeizhan, Guinongzhan, Hefengzhan, and Peizataifeng having the lower TFs, and cultivars Wufengyou 128 (TF = 46.2) and Tianyou 122 (TF = 8.88) having the higher ones. The TFs from the roots to the stems were the highest in cultivars Hemeizhan and Guinongzhan, but their TFs from the stems to the leaves were the lowest; cultivars Tianyou 122 and Wufengyou 128 showed the opposite results (Table 2). For the latter two, it would be beneficial to reduce DBP transportation from the stems to the grains to decrease DBP concentrations in the edible parts. The TFs of DBP from the shoots to the grains for cultivars Tianyou 122 (TF = 0.05) and Wufengyou 128 (TF = 0.27) were relatively low, and the highest occurred in cultivars Fengyou 428 and Tianfengyou 316, reaching a value of 1.5, which is not considered safe for consumption. The TFs of DBP from the shoots to the grains were <0.50, indicating relatively poor transportation ability. Significant differences were observed in the TFs from the roots to the stems, the stems to the leaves, and the shoots to the grains among some of the cultivars, demonstrating obvious differences in DBP transportation ability among the various rice cultivars.

The transport pathway and ability of contaminants in plants can affect their accumulation in different issues. Fujimaki et al. (2010) reported that the processes of cadmium transfer from the soil to the rice grains included absorption from the soil to the roots, efflux to the xylem and then transportation to the shoots, xylem-to-phloem transfer in the nodes, phloem transport to the grains, and post-phloem transport and accumulation in the grains. Root-to-shoot cadmium translocation via the xylem was a major and common physiological process controlling cadmium accumulation levels in the shoots and grains of rice (Uraguchi et al. 2009). However, transportation of contaminants varied by rice cultivars (Ueno et al. 2010; Yu et al. 2012; Liu et al. 2013). As mentioned above, the *OsHMA3* gene of a low-Cd-accumulating cultivar was responsible for selectively decreased accumulation of Cd by rice, but a high-Cd-accumulating cultivar had lost the *OsHMA3* function (Ueno et al. 2010). The TFs of DBP of various *I. aquatica* and *B. parachinensis* cultivars differed considerably (Zeng et al. 2005a, b; Cai et al. 2008b; Zhao et al. 2015). The results of the present study also showed that DBP uptake, accumulation, and transportation differed significantly among cultivars. However, TF differences between normal and hybrid rice were not significant ($P > 0.05$), being similar to the TFs of lead from the roots to the shoots in various rice cultivars (Liu et al. 2013). The DBP concentrations in cultivars Fengmeizhan, Tianyou 122, Wufengyou 128, and Peizataifeng were relatively higher in the non-edible parts (roots, stems, and leaves), but relatively lower in the grains, enabling these cultivars to be grown in low-level DBP-contaminated soils, whereas DBP concentrations in cultivars Hemeizhan, Hefengzhan, and Fengyousimiao were relatively lower in both the non-edible parts and the grains, and these cultivars would therefore be suitable for planting in low- to mid-level DBP-contaminated soils. The suitability, differences in uptake, and transportation abilities of the various cultivars, as well as the effects of their root exudates and

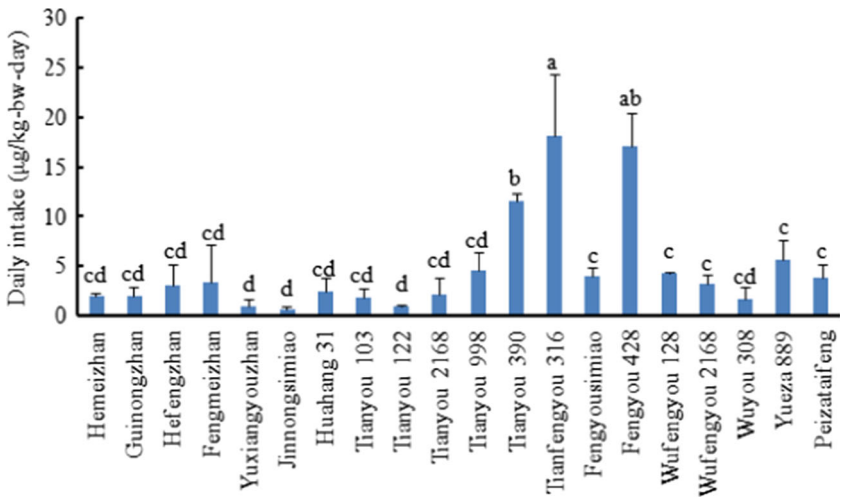


**Fig. 4** Daily intake of DBP via digestion of rice grains for Guangdong people ($n = 3$)

rhizospheric microorganism on DBP degradation, require further investigation.

### Variation in DBP concentration of grains and human exposure

Grain is the edible part of a cereal crop, and DBP concentrations in the grains are directly linked to the safety of agricultural products. Root uptake and transportation are potential sources of DBP in rice grains. Generally, uptake and accumulation of contaminants by rice grains occurred mainly at post-flowering stage (Rodda et al. 2011). However, carbon or contaminants accumulated in the shoots and roots could be redistributed to rice grains (Rodda et al. 2011; Tani et al. 2011). In the present study, DBP concentrations in rice grains varied greatly from 0.15 to 4.96 mg/kg (Fig. 3d). In normal rice cultivars, DBP concentrations in the grains were <1.0 mg/kg, with the lowest occurring in Jinnongsimiao (0.15 mg/kg). In the hybrid rice, DBP concentrations in the grains of 61.5% cultivars were >1.0 mg/kg, and they were >4.0 mg/kg in two cultivars (Tianfengyou 316 and Fengyou 428), which was significantly higher than those of almost all other cultivars (except for Tianyou 390; Fig. 3d). However, in some cultivars of the hybrid rice, DBP concentrations in grains were less than 1.0 mg/kg or even less than 0.5 mg/kg (i.e., Tianyou 122 and Wuyou308).

In Guangdong Province, the daily adult intake of polished rice (rice grains including about 73.9% polished rice, 8.7% hull, and 17.4% chaff; Endo et al. 2013) is ~220 g (Tang et al. 2009). Assuming a normal daily intake for this study, this would incur a DBP exposure ranging from 0.56 to 18.1 μg/kg body weight (bw) per day (Fig. 4), which is lower than the maximum DBP Reference Dose of 100 μg/kg bw per day set by USEPA (Kang et al. 2012). From a food safety perspective, and taking into account the remediation of contaminated soils, the following rice cultivars had low DBP concentrations in the grains but higher concentrations in shoots: normal rice cultivars Yuxiangyouzhan and Jinnongsimiao, and hybrid rice Tianyou 103, Tianyou 122, Wufengyou 128, and Wuyou 308, being all ideal cultivars. These could be planted in low-DBP-contaminated soils to ensure safe rice grains, thereby achieving the dual aims of production and soil remediation. However, the straw containing high DBP concentrations would need to be suitably treated (for example, by composting) to degrade the DBP (Cai et al. 2007). On the other hand, cultivars Tianyou 390, Fengyou 428, and Tianfengyou 316, whose grains contained high DBP concentrations, would not be suitable to grow in DBP-contaminated soils.

## Conclusion

DBP accumulation in rice plants varied greatly with cultivars, growth stages, and tissues. DBP concentrations in the roots and shoots were higher at ripening stage and lower at tillering, jointing, and flowering stages. At ripening stage, significant differences were observed not only among different tissues, with the leaves showing significantly higher DBP concentrations than the other tissues, but also among various cultivars. The translocation of DBP from the stems to the leaves was higher than those from the roots to the stems and from the shoots to the grains. No significant difference was observed in DBP accumulation or transportation between normal and hybrid rice cultivars. Cultivars Yuxiangyouzhan, Jinnongsimiao, Tianyou 122, and Wuyou 380 accumulated relatively lower amounts of DBP in grains, resulting in low DBP exposure to humans. The uptake and transportation pathways of DBP by rice and their molecular mechanisms require further investigation.

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