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Genotypic variation in the uptake, accumulation, and translocation of di-(2-ethylhexyl) phthalate by twenty cultivars of rice (*Oryza sativa* L.)

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ABSTRACT

Agricultural soil in China contains high levels of di-(2-ethylhexyl) phthalate (DEHP), especially in paddy-field soil of Guangdong province of China, but the accumulation and translocation of DEHP by rice (*Oryza sativa* L.) remains unknown. In the present study, twenty rice cultivars were cultivated in paddy soil spiked with DEHP, and variations in DEHP accumulation and translocation among various cultivars were investigated. Our results showed that DEHP concentrations in roots and shoots of different rice cultivars at four growth stages (i.e., ripening, tillering, jointing, and flowering stages) varied greatly from 0.26 to 11.8 mg/kg (dry weight, dw) and 0.40 to 7.58 mg/kg (dw), respectively. No obvious change over time was observed. The greatest variation in DEHP concentrations among the rice cultivars occurred at ripening stage, whereas the lowest variation at flowering stage. During ripening stage, the largest variation in DEHP concentrations among cultivars were observed in stems (varying from 0.35 to 13.2 mg/kg), whereas the least one was observed in roots (ranging from 1.01 to 5.72 mg/kg). Significant differences in DEHP concentrations in the roots, stems, leaves and grains of most rice cultivars were found. The translocation factors of DEHP from roots to stems or stems to leaves were higher than those from shoots to grains. Overall, cultivars Tianfengyou 316, Wuyou 308, and Peizataifeng, which contained low levels of DEHP in grains but high levels in shoots, were ideal cultivars for simultaneous production of safe food and phytoremediation of contaminated soil.

1. Introduction

Phthalic acid esters (PAEs) are commonly used as plasticizers in industrial, medical, and consumer products (Guo et al., 2011a). The global annual production of phthalates was approximately 5.2 million tons in 2005 (Mackintosh et al., 2006). The widespread use of PAE-containing products has caused frequent occurrence of PAEs in environmental compartments such as dust, soil, and water (Yang et al., 2007; Cai et al., 2008a; Zeng et al., 2008; Guo and Kannan, 2011b; Shi et al., 2012; Benning et al., 2013). The wide occurrence of PAEs in the environment can result in human exposure to PAEs via inhalation, dermal absorption, and dietary intake, as confirmed by the high detection frequency of phthalate metabolites in human urine samples from multiple countries (Guo et al., 2011a; C.F. Wu et al., 2013a, Z. Wu et al., 2013b; Kranich et al., 2014). Some PAE compounds, such as di(2-ethylhexyl) phthalate (DEHP) and di-*n*-butyl phthalate (DBP), are suspected to be endocrine-disrupting chemicals that exert various adverse effects, such as anti-androgenic effect, or teratogenic, mutagenic, and carcinogenic toxicity (Staples et al., 1997; Rusyn and Corton, 2012; Kranich et al., 2014). DEHP is listed as a priority pollutant by the United States Environmental Protection Agency (US EPA) and considered as a strongly accumulative priority substance listed in Part A of Annex II of the New Directive 2008/105/EC (European Commission). Thus, DEHP is a compound of great concern to both the scientific community and general public.

DEHP is one of the most frequently detected PAE compounds in agricultural soils and vegetables, and its concentrations are higher than most other individual PAE compounds (excluding DBP) (Vikelsoe et al., 2002; Yang et al., 2007; Cai et al., 2008a; Zeng et al., 2008; Mo et al., 2009; Rhind et al., 2013; Wang et al., 2013; Niu et al., 2014). In China, Guangdong, Fujian, and Xinjiang provinces

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showed the highest levels of total PAEs in agricultural soils (Niu et al., 2014). The average concentrations of DEHP in agricultural soils of Guangdong Province decreased in the order paddy-field soil > banana soil > vegetable soil > sugarcane soil > orchard soil (Yang et al., 2007). Elevated levels of DEHP were observed in vegetable fields located adjacent to the urban districts or in intensive agricultural soils that use plastic film (Zeng et al., 2008; Wang et al., 2013). DEHP can be taken up and accumulated by vegetables and other plants (Cai et al., 2008b; Mo et al., 2009; Fu and Du, 2011; Z. Wu et al., 2013b; Li et al., 2014; Lü et al., 2014), and high concentrations were detected in vegetables from plastic film greenhouses (Fu and Du, 2011), which cause contamination of vegetables by DEHP and pose a potential threat to human health. However, no study has reported DEHP uptake and accumulation by rice (*Oryza sativa* L.).

Rice planting is one of the most prevalent land use types throughout South and East Asia, where rice provides the dominant staple food. Rice root tissue can absorb heavy metals from soil and transport them to the shoot via xylem, and then to grains via xylem-to-phloem transfer and post-phloem transport (Fujimaki et al., 2010; Ueno et al., 2010). Recently, numerous studies have reported the accumulation of heavy metals (e.g., cadmium, lead, and arsenic) by rice, indicating that heavy metal accumulation in rice grains is genotype-dependent or cultivar-specific (Ueno et al., 2010; Norton et al., 2012; Tripathi et al., 2012; Liu et al., 2013). For example, significant genotypic variation was found in rice grain arsenic accumulation and speciation (Norton et al., 2012; Ye et al., 2012). Rice plants can also accumulate polycyclic aromatic hydrocarbons (PAHs) in various tissues (Tao et al., 2006; Du et al., 2011). The concentration distribution of heavy metals and PAHs in various tissues of rice generally followed the order roots > leaves > stems > grains (Tao et al., 2006; Du et al., 2011; Liu et al., 2013; Zheng et al., 2013). As discussed above, the average concentration of DEHP in paddy-field soil was higher than in vegetable soil (Yang et al., 2007), and exceeded the allowable soil concentration recommended by New York State of US (Cai et al., 2008a; Wang et al., 2013). On the other hand, grains from New York State contained significantly higher average concentrations of DEHP than fruits/vegetables, meats, and milk from the same region (Schecter et al., 2013). Cereals (including rice) and vegetables from China also contained DEHP (Mo et al., 2009; Guo et al., 2012; Sui et al., 2014), indicating that plant products are indeed contaminated by PAEs. Dietary intake of cereals in China contributed to the primary food sources of DEHP (44.57% for adults and 39.44% for children) (Sui et al., 2014). Nevertheless, the primary source of DEHP in rice grains remains unknown. Whether DEHP in soil can be taken up by rice root and transported to the shoot and grains also remains unclear.

In this study, twenty rice cultivars (including seven normal and thirteen hybrid cultivars) were planted in DEHP-contaminated soil. The aims of this study were to investigate variations in DEHP accumulation in different genotypic rice cultivars and the DEHP distributions in tissues (e.g., root, stem, leaf, and grain) and growth stages (e.g., tillering, jointing, flowering and ripening stages) of rice plants to identify rice cultivars that accumulate low DEHP levels.

2. Materials and methods

2.1. Chemicals and materials

DEHP solution of analytical grade (> 98.5% purity) for the pot experiment was purchased from Tianjin Chemical Reagent Factory, China. A composite stock standard solution containing DEHP (1000 μg/mL in dichloromethane, 99.8% purity) for instrument analysis was purchased from o2si Smart Solution (USA). The organic solvents, including dichloromethane and *n*-hexane, were of HPLC grade. All other chemicals used in the study were of analytical grade.

Rice cultivars, including seven common and thirteen hybrid cultivars (Table 1), were selected from those widely cultivated in Guangdong province, China. Rice seeds were obtained from Guangdong Academy of Agricultural Science and South China Agricultural University, China.

2.2. Experimental design

The pot experiment was conducted in a glasshouse in South China Agricultural University, Guangzhou, China. The upper layer of soil (0–20-cm depth, in which rice is grown in the long-term) was taken from an uncontaminated experimental field of South China Agricultural University. The soil was air-dried, mixed thoroughly, and passed through a 5-mm sieve to remove gravel, plant debris, and other inert materials. Then, a portion of the soil above was passed through a 2-mm sieve for DEHP spiking. The soil had the following initial characteristics: 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.

An aliquot of soils (pass through a 2-mm sieve, 10% total quantity of soil) was spiked with a DEHP solution in acetone, mixed thoroughly, and then left to stand for 48 h in stainless steel containers under the fume hood to evaporate any traces of acetone. The spiked soil was thoroughly mixed with uncontaminated soil and homogenized, and then passed through a 5-mm sieve again to ensure homogeneity. Five kilograms of the mixed soil above were packed into ceramic pots (21.4-cm inner diameter and 20.6-cm height). Pots were randomly arranged in a greenhouse,

Table 1
Cultivars of rice from different genetic background.

Name	Source[a]	Type	Name	Source	Type
Hemeizhan	Fengmeizhan/Hesizhan	N[b]	Tianyou 998	Tianfeng A/Guanghui 998	H[c]
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

[a] Source refers to female/male parents; Data was from China Rice Data Center http://www.ricedata.cn/variety/.
[b] N: Normal cultivar.
[c] H: Hybrid cultivar.

and the soil in each pot was submerged in water (about 2-cm depth) for two weeks to equilibrate; the rice seedlings were then transplanted. Prior to cultivation, the soil was fertilized with 0.20 g/kg N, 0.15 g/kg P, and 0.15 g/kg K (using urea, superphosphate, and potassium chloride, respectively). The background concentration of DEHP in the soil was 0.65 ± 0.11 mg/kg. The initial concentration of DEHP in the treated soils, set according to the concentrations detected in contaminated soil of Guangdong province, was 20 mg/kg (on a dry weight basis), and the measured value by gas chromatography coupled with mass spectrometry (GC/MS) (more analytical details provided in Section 2.3) was 19.68 ± 0.23 mg/kg (n=3, each sample being a composite of five subsamples).

Rice seeds were germinated in a substrate mixed with uncontaminated soil and organic fertilizer. After 20 days, the uniform seedlings were selected and transplanted into the pots (15 seedlings clustered as three plants per pot). All treatments were in four replicates. The soil in pots was submerged beneath 2–3 cm of water for the entire growth period. Rice plant samples were collected at the tillering, jointing, flowering, and ripening stages, respectively. The rice plants were washed successively with tap water and deionized water. The plants were divided into roots, shoots, leaves, and grains (at the ripening stage), and freeze-dried using a freeze drier (Thermo Heto Power Dry, LL3000, USA), and then ground (1-mm sieve).

2.3. Analytical procedure, quality assurance, and quality control

DEHP was extracted ultrasonically from plant samples following USEPA method 3540C. Briefly, 2.0 g of rice sample were extracted in triplicate using a sonicator (KH-250E, Kunshan, China) with 20-mL dichloromethane for 10 min, and centrifuged for 5 min at 4000 rpm. Then, the supernatant was combined and concentrated to about 1.0 mL using a rotary evaporator (52-A, Yarong, China). The concentrated extract was cleaned-up using a combined glass chromatography column of silica gel and anhydrous sodium sulfate with 50 mL of dichloromethane (Cai et al., 2007a). The obtained elute was concentrated successively using a vacuum rotary evaporator and under a nitrogen steam.

DEHP in the extract was analyzed using gas chromatography coupled with mass spectrometry (Shimadzu, GC-MSQP2010). The separation was performed on a DB-5MS capillary column (length 30 m, i.d. 0.25 mm, film 0.25 µm, Agilent). Helium (99.999% purity) was used as a carrier gas at a constant flow of 0.70 mL/min. The injector temperature was maintained at 250 °C and 1 µL of extract was injected in splitless mode. The temperature program for DEHP analysis was 100 °C for 2 min, heating to 110 °C at 35 °C/min, increasing to 129 °C at 15.0 °C/min, and heating to 280 °C at 40.0 °C/min (hold for 4 min). The temperatures for the transfer line and ion source were 220 °C and 250 °C, respectively.

Analyte ionization was performed using electron ionization (70 eV), and signal acquisition was performed in selected ion-monitoring (SIM) mode. The identity of the DEHP peak was confirmed by comparison of the retention time of DEHP standard and the characteristic ion (the primary characteristic ion of DEHP being 149) in the mass spectra. Quantitative analysis was performed using the external calibration method based on a five-point calibration curve (0–4.0 µg/mL). The instruments were calibrated daily using calibration standards. Method blank (solvent), spiked blank (standard spiked into solvent), matrix spiked duplicates, and sample duplicates were routinely analyzed together with samples. The DEHP recoveries in plant samples ranged from 87.4% to 107.2%. The detection limit of DEHP in samples, based on threefold of the signal-to-noise ratio, was 2.5 µg/kg, and the limit of quantification was 8.3 µg/kg.

2.4. Statistical analysis

Concentrations of DEHP were expressed on a dry weight basis. Data was subjected to analysis with one-way ANOVA and least significant differences ($P < 0.05$) for comparison of treatment means. All data were processed using the statistics analysis system (SAS, version 8.0) for Windows software package.

3. Results and discussion

3.1. DEHP concentrations in rice plants at different growth stages

The DEHP concentrations at the four growth stages (tillering, jointing, flowering and ripening) were combined for direct comparisons among the tissues. DEHP concentrations in rice roots ranged from 0.26 to 11.8 mg/kg (Table 2). Both the lowest (0.26 mg/kg) and highest concentrations (11.8 mg/kg) were observed at jointing stage. Obviously, DEHP accumulation in roots of various rice cultivars was considerable different (Table 2). At both the tillering and jointing stages, cultivar Jinnongsimiao showed the highest DEHP concentrations (4.71 and 11.8 mg/kg), which were higher by 50% and 25-fold than the lowest ones, 1.88 mg/kg for cultivar Tianyou 2168 at the tillering stage and 0.26 mg/kg for cultivar Fengyou 428 at the jointing stage, respectively. At both the flowering and ripening stages, the highest DEHP concentrations were detected in cultivar Hemeizhan, and the lowest in cultivars Tianyou 390 and Wuyou 308. Although the highest DEHP concentrations were greater than 5 mg/kg and reached 11.8 mg/kg, DEHP concentrations in 90% or more of root samples were between 1.0 and 4.0 mg/kg. Additionally, for the majority of individual cultivars, the highest concentration of DEHP was observed at the tillering or jointing stage (excluding cultivars Hemeizhan and Tianyou 2168), indicating the higher uptake and accumulation abilities of rice at these two stages. Regarding rice types, no significant difference in DEHP concentrations was observed between normal and hybrid rice cultivars (Fig. 1a).

Concerning rice shoots (including stems and leaves), DEHP concentrations varied from 0.40 to 7.58 mg/kg, and the maximum concentration was found at the ripening stage (Table 2). For most of individual cultivars, DEHP concentrations were higher at the tillering or ripening stage compared with the other two stages. Similar differences were observed for lead concentrations in rice shoots (Liu et al., 2013). This may be related to the rapid increase in shoot biomass during the jointing and flowering stages, and DEHP may be diluted during transport from roots to shoots. Shoot biomass did not increase further during the ripening stage, while the DEHP transported from roots to shoots likely accumulated in rice shoots. Uraguchi et al. (2009) reported that root-to-shoot cadmium translocation via the xylem is the major and common physiological process determining the cadmium accumulation levels in shoots. Xylem and phloem play important roles in the transportation of arsenic from rice roots to shoots (Carey et al., 2011). Gao and Collins (2009) reported that the transfer of PAHs from White clover roots to shoots and their accumulation therein was driven by the transpiration stream flux. However, Su and Zhu (2008) reported that the transport of PAHs—such as phenanthrene—from roots to shoots through xylem contributed little to PAH accumulation in shoots. PAH compounds were transported very short distances from roots (only up to 1500 µm over a 56-day period) (Wild et al., 2005). A significant fraction of shoot PAH accumulation resulted from aerial deposition derived from volatilized PAHs (Su and Zhu, 2008; Gao and Collins, 2009). However, the structure and physico-chemical properties of DEHP (e.g., water solubility: 2.49 µg/L; lg K_{ow}: 7.0–7.8; average organic carbon normalized partition coefficients (K_{oc}): 482,000; Staples et al., 1997)

Table 2
DEHP concentrations (mg/kg) of rice roots and shoots at different growth stages of various cultivars.

Rice type	Cultivars	Tillering	Jointing	Flowering	Ripening
Roots					
Normal	Hemeizhan	2.27	3.28	6.13	5.72 ± 1.70a[a])
	Guinongzhan	2.33	1.97	1.73	1.50 ± 0.50bc
	Hefengzhan	3.17	1.67	1.46	1.33 ± 0.38bc
	Fengmeizhan	2.57	1.64	1.68	1.41 ± 0.11bc
	Yuxiangyouzhan	3.38	1.55	1.72	1.11 ± 0.16c
	jinnongsimiao	4.71	11.8	1.74	1.23 ± 0.20c
	Huahang 31	2.96	2.22	1.86	2.09 ± 0.22bc
Hybrid	Tianyou 103	2.64	2.44	2.18	2.37 ± 0.60b
	Tianyou 122	2.71	3.50	2.52	1.46 ± 0.12bc
	Tianyou 2168	1.88	1.99	5.51	1.50 ± 0.25bc
	Tianyou 998	3.21	3.17	2.65	1.44 ± 0.30bc
	Tianyou 390	2.66	2.82	0.98	2.01 ± 0.60bc
	Tianfengyou 316	3.07	0.40	2.02	1.25 ± 0.13c
	Fengyousimiao	3.57	2.06	2.77	1.67 ± 0.33bc
	Fengyou 428	2.52	0.26	1.34	1.57 ± 0.13bc
	Wufengyou 128	2.49	2.75	2.38	1.36 ± 0.16bc
	Wufengyou 2168	3.52	1.96	2.24	1.40 ± 0.16bc
	Wuyou 308	1.89	3.75	2.38	1.01 ± 0.08c
	Yueza 889	2.69	10.61	4.90	1.21 ± 0.32c
	Peizataifeng	3.41	2.76	1.98	2.08 ± 0.27bc
Shoots					
Normal	Hemeizhan	1.90	1.78	1.91	1.20 ± 0.13cd
	Guinongzhan	1.76	0.67	1.40	1.25 ± 0.07cd
	Hefengzhan	1.74	1.69	1.43	0.95 ± 0.14d
	Fengmeizhan	2.47	0.94	0.94	1.10 ± 0.09cd
	Yuxiangyouzhan	1.95	0.80	1.18	1.85 ± 0.40bcd
	Jinnongsimiao	1.93	0.73	1.37	1.06 ± 0.12cd
	Huahang 31	2.04	0.84	0.60	2.18 ± 0.87bcd
Hybrid	Tianyou 103	1.65	0.75	0.68	0.93 ± 0.10d
	Tianyou 122	1.47	1.03	1.72	1.37 ± 0.28cd
	Tianyou 2168	1.56	0.79	0.95	1.51 ± 0.26bcd
	Tianyou 998	1.76	1.10	0.91	3.12 ± 2.34bcd
	Tianyou 390	2.97	0.40	0.70	2.23 ± 1.09bcd
	Tianfengyou 316	1.31	1.20	0.68	4.85 ± 0.14abc
	Fengyousimiao	1.07	0.67	1.06	1.18 ± 0.23cd
	Fengyou 428	1.28	0.71	0.81	4.41 ± 2.42abcd
	Wufengyou 128	2.19	0.93	1.15	1.00 ± 0.04d
	Wufengyou 2168	1.55	1.19	0.68	1.73 ± 0.41bcd
	Wuyou 308	1.60	1.56	0.85	7.58 ± 0.93a
	Yueza 889	3.32	1.11	1.08	1.03 ± 0.17cd
	Peizataifeng	1.51	1.21	0.87	5.22 ± 1.90ab

[1] Mean±S.D. (n=3) followed by the same lower case letters within a column for shoots or roots were not significantly different ($P > 0.05$).

differed from those of heavy metals and PAHs. Future studies should examine the pathway of DEHP uptake and translocation in rice.

In the various cultivars, the greatest variation in DEHP concentrations in shoots was found at ripening stage, and the lowest at the jointing and flowering stages (Table 2). For example, at the ripening stage, the highest DEHP concentration (7.58 mg/kg in cultivar Wuyou 308) was significantly higher than those of cultivars Tianyou 103, Wufengyou 128, Yeza 889, Hefengzhan, Fengmeizhan, and Jinnongsimiao, (~1.0 mg/kg), while the lowest DEHP concentration (0.93 mg/kg) was detected in cultivar Tianyou 103. At the tillering stage, cultivars Yueza 889 and Fengyousimiao showed the highest (3.32 mg/kg) and lowest (1.07 mg/kg) DEHP concentrations, respectively, whereas at both the jointing and flowering stages, cultivar Hemeizhan showed the highest DEHP concentrations. These results indicated that the accumulation of DEHP in rice shoots is somewhat cultivar-specific. Metal (zinc, copper, or cadmium) accumulation by rice also differed between shoots and roots and between the *japonica* and *indica* genotypes

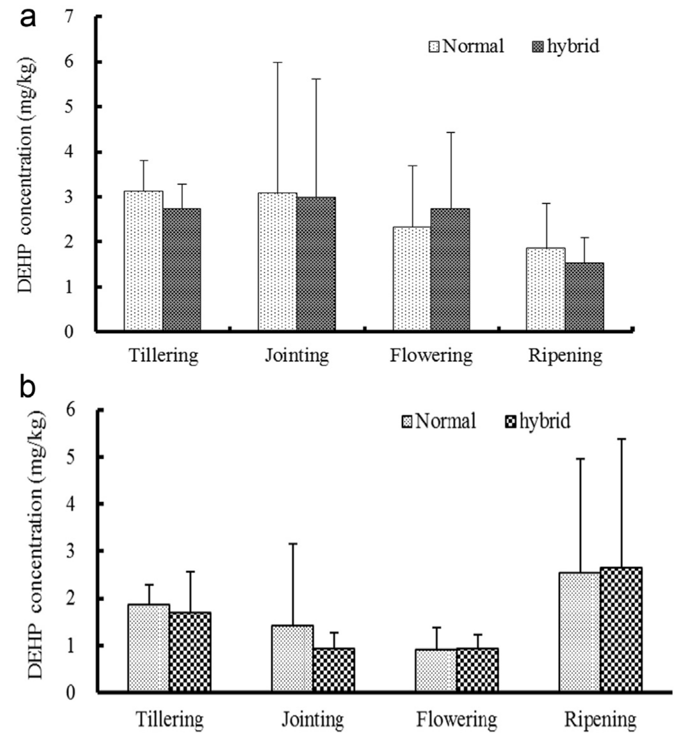

Fig. 1. DEHP concentrations in roots (a) and shoots (b) of normal rice and hybrid rice at different growth stages.

(Yoshihara et al., 2010). Concerning the rice types, the average DEHP concentrations of normal cultivars were comparable to those of hybrid cultivars (except at the jointing stage) (Fig. 2b). Similarly, Liu et al. (2013) reported that lead concentrations in the shoots of various rice types showed no significant difference.

3.2. Genotypic variation in DEHP accumulation among rice tissues

In this study, the concentrations of DEHP in roots, shoots, leaves, and grains at the ripening stage were measured separately (Table 3). DEHP concentrations varied from 0.12 to 13.2 mg/kg. Both the highest DEHP concentration and the largest variation in DEHP concentrations were observed in stems. Moreover, DEHP concentrations in stems of three cultivars (e.g., Tianfengyou 316, Wuyou 308, and Peizataifeng) were greater than 10.0 mg/kg, whereas DEHP concentrations in roots, leaves, and grains were less than 5.0 mg/kg (excluding leaves of cultivar Tianfengyou 316). The levels of DEHP in rice tissues were comparable with those in *Ipomoea aquatica* cultivars (Cai et al., 2008b) and vegetables from Pearl River Delta area (Mo et al., 2009).

Significant differences in DEHP concentrations were found among tissues of the majority of rice cultivars (excluding Guinongzhan, Hefengzhan, Huahang 31, Tianyou 2168, and wufengyou 2168). The highest DEHP concentrations were detected in stems for seven cultivars (Huahang 31, Tianyou 390, Tianfengyou 316, Fengyou 428, Wuyou 308, and Peizataifeng), which were significantly higher than those in other tissues (excluding cultivar Wufengyou 2168). The highest DEHP concentrations occurred in leaves for eight cultivars (e.g., Guinongzhan, Fengmeizhan, Tianyou 122, Fengyousimiao, etc.) and in roots and grains for only two cultivars. These results indicated that stems and leaves accumulated higher DEHP levels compared with roots and grains. This distribution pattern differed from that of PAHs or ^{14}C-phenanthrene in rice plants (Tao et al., 2006; Du et al., 2011). Tao et al. (2006) collected rice plants from a PAH-contaminated

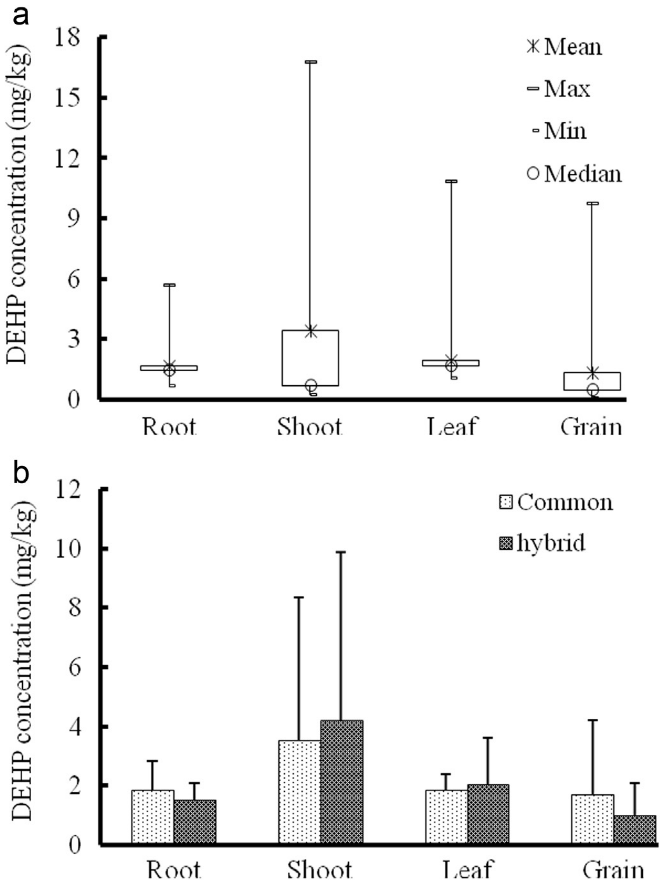

Fig. 2. DEHP concentrations in rice different tissues at ripening stage. (a) Concentration distribution and (b) variation of normal rice and hybrid rice.

site in Tianjin, Northern China, and reported that PAH levels decreased in the order roots > hulls > leaves > internodes > seeds. The concentrations of ^{14}C-phenanthrene in rice tissues decreased in the order roots > leaves > shells > stems > grains (Du et al., 2011). These results suggested that DEHP and PAHs were unevenly distributed among the tissues of rice plants. This may be associated with DEHP translocation among the tissues of various cultivars. For example, scientists revealed that *OsHMA3* is a useful gene for limiting cadmium translocation from rice roots to the above-ground tissues by selectively sequestrating cadmium into the root vacuoles, and OsHMA2 is a major transporter of zinc and cadmium from rice roots to shoots (Ueno et al., 2010; Satoh-Nagasawa et al., 2012). Overexpression of the functional gene (*OsHMA3*) from a low cadmium-accumulating cultivar selectively decreased cadmium accumulation by rice, but the *OsHMA3* transporter from a high-cadmium cultivar had lost its function probably due to a single amino acid mutation (Ueno et al., 2010). In this study, DEHP concentrations were higher in stems or leaves of several cultivars, as mentioned above. However, the mechanism underlying this accumulation and translocation requires further study.

DEHP concentrations in stems and leaves of some cultivars were higher or comparable with those in the corresponding roots (Table 2). This result may be attributable in part to the shoot DEHP accumulation following aerial deposition. For example, it has been shown that direct aerial deposition from volatilization contributed greatly to plant accumulation of PAHs (such as phenanthrene and pyrene) (Tao et al., 2006; Gao and Collins, 2009). The concentrations of phenanthrene and pyrene in rice shoots showed no significant differences with various PAH level treatments and were not correlated with concentrations in rice roots (Su and Zhu, 2008). In the present study, Henry's law constant (H) of DEHP ($H_c = 1.71 \times 10^{-5}$) (Staples et al., 1997) was comparable with those of phenanthrene and pyrene ($H_c = 2.56 \times 10^{-5}$ and 1.14×10^{-5}, respectively) (Toronto Public Health, 1998), indicating that DEHP could volatilize to the air to some extent. Our previous study demonstrated that the DEHP concentrations in leaves of Chinese flowering cabbage (*Brassica parachinensis*) cultivated in DEHP-contaminated soil were greater than those grown in the same contaminated soil covered with uncontaminated soil in a glasshouse, indicating that plant leaves could take up DEHP volatilized from contaminated soil (Zeng et al., 2005). Further studies should explore the effect of aerial deposition on DEHP accumulation by rice plant.

Rice grains (including about 73.9% polished rice, 8.7% hull, and

Table 3
DEHP concentrations (mg/kg) of rice different tissues at ripening stage of various rice cultivars.

Rice type	Cultivars	Root	Stem	Leaf	Grain
Normal	Hemeizhan	5.72 ± 1.70aA[a]	0.98 ± 0.15deB	1.42 ± 0.18bB	4.39 ± 0.82aA
	Guinongzhan	1.50 ± 0.50bcdA	0.78 ± 0.03deA	1.72 ± 0.11bA	1.16 ± 1.03cdA
	Hefengzhan	1.33 ± 0.38cdA	0.40 ± 0.07eA	1.50 ± 0.21bA	3.29 ± 2.22abA
	Fengmeizhan	1.41 ± 0.04bcdA	0.54 ± 0.02eB	1.67 ± 0.17bA	0.18 ± 0.04d C
	Yuxiangyouzhan	1.11 ± 0.09cdAB	1.51 ± 0.10deA	2.18 ± 0.56bA	0.12 ± 0.02dB
	Jinnongsimiao	1.23 ± 0.20cdA	0.57 ± 0.08deB	1.56 ± 0.11bA	0.20 ± 0.02dC
	Huahang 31	2.09 ± 0.22bcA	2.95 ± 0.74cdA	2.02 ± 0.31bA	2.86 ± 0.60abcA
Hybrid	Tianyou 103	2.37 ± 0.60bA	0.35 ± 0.07eC	1.51 ± 0.10bAB	0.55 ± 0.13dBC
	Tianyou 122	1.46 ± 0.07bcdB	0.48 ± 0.06eC	2.25 ± 0.27bA	0.27 ± 0.08dC
	Tianyou 2168	1.50 ± 0.25bcdA	1.27 ± 0.32deA	1.76 ± 0.23bA	1.16 ± 0.86cdA
	Tianyou 998	1.44 ± 0.30bcdB	0.42 ± 0.09eB	1.40 ± 0.04bB	3.86 ± 0.43abA
	Tianyou 390	2.01 ± 0.60bcdB	3.93 ± 1.64cA	1.87 ± 0.15bAB	0.65 ± 0.17dB
	Tianfengyou 316	1.25 ± 0.13cdC	13.1 ± 0.63aA	6.04 ± 2.46aB	0.77 ± 0.26cdC
	Fengyousimiao	1.67 ± 0.33bcdA	0.44 ± 0.04eB	1.92 ± 0.42bA	0.33 ± 0.05dB
	Fengyou 428	1.57 ± 0.13bcdB	6.00 ± 0.08bA	1.59 ± 0.38bB	0.51 ± 0.11dC
	Wufengyou 128	1.36 ± 0.16cdA	0.62 ± 0.09deB	1.37 ± 0.09bA	0.57 ± 0.21dB
	Wufengyou 2168	1.40 ± 0.16bcdA	1.78 ± 0.73cdeA	1.68 ± 0.09bA	1.05 ± 0.44cdA
	Wuyou 308	1.01 ± 0.05dB	13.2 ± 2.1aA	1.99 ± 0.24bB	0.28 ± 0.05dB
	Yueza 889	1.21 ± 0.32cdAB	0.42 ± 0.09eB	1.65 ± 0.11bA	1.90 ± 0.45bcdA
	Peizataifeng	2.08 ± 0.37bcB	12.4 ± 1.1aA	2.01 ± 0.14bB	0.53 ± 0.06dB

[a] Mean ± S.D. ($n=3$) followed by the same lowercase letters within a column or by the same capital letters within a row were not significantly different, respectively ($P > 0.05$).

17.4% chaff; Endo et al., 2013) are the edible part, and their DEHP concentration directly influences the safety of agricultural products. In this study, DEHP concentrations in grains ranged from 0.12 to 4.39 mg/kg, and were less than 1.0 mg/kg for 60% of cultivars (Table 3). In particular, DEHP concentrations were less than 0.5 mg/kg in the grains of cultivars Fengmeizhan, Yuxiangyouzhan, Jinnongsimiao, Tianyou 122, Fenyousimiao, and Wuyou 308. Moreover, in cultivars with grain DEHP concentrations of less than 1.0 mg/kg, the DEHP levels in grains were significantly lower than those in one or more tissues. In three cultivars (i.e., Hemeizhan, Tianyou 998, and Yeza 889), DEHP concentrations in grains were significantly greater than those in other tissues. Generally, grains develop later for long time than the other tissues (roots, stems or leaves) of rice, and uptake of contaminants by rice grains occurs mainly during the post-flowering stage (Rodda et al., 2011). However, rice could redistribute carbon or contaminants accumulated in the plant body (shoots and roots) to the grains (Rodda et al., 2011). For example, it was estimated that 60% of the final grain cadmium contents was remobilized from that accumulated by the plant prior to flowering, while the remaining 40% originated from uptake during grain maturation (Rodda et al., 2011). In this study, DEHP in other rice tissues was likely translocated and redistributed to grains. The details of DEHP redistribution in various rice tissues should be investigated in future. In addition, Tao et al. (2006) reported that PAH concentrations in hulls were significantly higher than those in seeds. Grain, especially polished rice, was the edible component. The DEHP concentrations and distributions in rice hull, polished rice, etc., must be clarified to enable selection of cultivars that accumulate lower levels of DEHP.

The variation in DEHP accumulation occurred not only among rice tissues but also among cultivars (Table 3). For example, cultivar Hemeizhan showed the highest DEHP concentrations in both root and grain tissues, while cultivars Wuyou 308 and Yuxiangyouzhan showed the lowest concentrations in roots and grains, respectively. For stem tissue, cultivars Wuyou 308 and Tianyou 103 showed the highest (13.2 mg/kg) and lowest DEHP concentrations (0.35 mg/kg), respectively; the former being ~37-fold higher than the latter. Regarding leaf tissue, the highest DEHP concentration (6.04 mg/kg) was observed in cultivar Tianfengyou 316, and the lowest (1.37 mg/kg) in cultivar Wufengyou 128. These results demonstrated that DEHP uptake and accumulation by rice were cultivar-specific. Furthermore, the greatest difference magnitudes in DEHP concentrations among cultivars were found in stems, followed by leaves and grains, and the least in roots (Table 3). This observation differs from lead distribution patterns in the tissues of various rice cultivars (Liu et al., 2013). Liu et al. (2013) reported that the variation magnitudes in lead concentrations among six rice cultivars were greater in grains than in shoots and roots. The variation in contaminant accumulation in rice tissues was related not only to the cultivars but also to translocation of contaminants among tissues, as well as to the soil contamination levels. For example, Yu et al. (2006) reported that, under low cadmium level exposure, the grain cadmium concentrations in normal rice cultivars were significantly higher than in hybrid cultivars, while the opposite trend was observed under high cadmium level exposure. In the present study, the initial concentration of DEHP in the soil was set at 20 mg/kg, but the residual concentrations of DEHP in the soils grown different cultivars varied greatly at flowering and ripening stages (Table 4). This might be attributed to the variation in uptake by different rice cultivars and biodegradation in soils. Generally, the bioavailability of DEHP in the soils grown various cultivars might be different with root exudates, soil properties and microbial communities (Jelali et al., 2010; de Graaff et al., 2013). It should be noted that the variation in DEHP concentrations in the soils of different treatments were less than those in plant tissues. This might be related to the variation in uptake pathway and metabolism of DEHP among different cultivars as well as rhizospheric degradation. The detailed reasons need to be investigated by other rigid experiment.

Table 4
DEHP residual concentrations (mg/kg) in the soils at flowering and ripening stages.

Rice type	Cultivars	Flowering	Ripening
Normal	Hemeizhan	4.79 ± 3.20	2.30 ± 1.36
	Guinongzhan	3.22 ± 0.12	2.29 ± 0.65
	Hefengzhan	3.31 ± 0.02	1.94 ± 1.20
	Fengmeizhan	4.15 ± 0.05	2.61 ± 0.36
	Yuxiangyouzhan	5.05 ± 2.49	2.61 ± 1.13
	Jinnongsimiao	7.03 ± 2.93	2.83 ± 2.01
	Huahang 31	5.54 ± 1.64	2.82 ± 0.20
Hybrid	Tianyou 103	3.06 ± 1.14	1.37 ± 0.15
	Tianyou 122	3.91 ± 0.73	1.26 ± 0.23
	Tianyou 2168	3.36 ± 0.10	2.69 ± 1.03
	Tianyou 998	3.27 ± 0.08	1.87 ± 0.10
	Tianyou 390	6.47 ± 1.88	1.38 ± 0.07
	Tianfengyou 316	4.43 ± 3.18	1.97 ± 1.16
	Fengyousimiao	4.96 ± 0.96	1.96 ± 0.50
	Fengyou 428	1.51 ± 0.03	1.24 ± 0.10
	Wufengyou 128	3.23 ± 0.57	1.51 ± 0.05
	Wufengyou 2168	4.69 ± 0.39	1.67 ± 0.24
	Wuyou 308	3.56 ± 0.36	1.55 ± 0.20
	Yueza 889	2.04 ± 1.77	1.73 ± 0.19
	Peizataifeng	3.20 ± 0.79	2.84 ± 0.63
Without plant		8.10 ± 4.20	4.28 ± 1.23

Additionally, genotypic cultivars with greater radial oxygen loss have strong ability to enhance arsenic tolerance by decreasing arsenic mobilization in the roots and reduce arsenic accumulation in shoots by limiting its translocation from roots (Mei et al., 2012). The reasons that some cultivars (i.e., Fengmeizhan, Fengyousimiao, and Wufengyou 128) accumulated relatively lower DEHP levels compared with cultivars Peizataifeng and Wuyou 308 should be examined in future studies.

DEHP concentrations in the same tissues varied greatly among hybrid rice cultivars or normal rice cultivars (Table 3). For example, among normal rice cultivars, DEHP concentrations in roots and grains of cultivar Hemeizhan were significantly greater than those of the other cultivars (excluding grains of cultivar Hefengzhan); among hybrid rice cultivars, DEHP concentrations in the stems and leaves of cultivar Tianfengyou 316 were significantly higher than those of the other cultivars (excluding stems of cultivars Wuyou 308 and Peizataifeng). These results confirmed that DEHP accumulation was cultivar-specific.

In Guangdong province, rice is a staple food and major source of nutrients, and adult daily intake of polished rice (rice grains including about 73.9% of polished rice; Endo et al., 2013) was about 220 g (Tang et al., 2009). Upon ingestion of the rice grains evaluated in this study, adult DEHP intake exposure would be 0.36–12.8 μg/kg-bw-day, being less than the DEHP reference dose of 20 μg/kg-bw-day proposed by the US EPA (Kang et al., 2012). In view of food safety and remediation of contaminated soil, which requires low levels of DEHP in grains but high levels in stems and leaves, cultivars Tianfengyou 316, Wuyou 308, and Peizataifeng were ideal. These three rice cultivars could be cultivated in soil contaminated with low level of DEHP to ensure the safety of rice grains, thereby achieving simultaneous production and remediation. Nevertheless, rice straw containing high DEHP level should be properly treated through composting to degrade DEHP (Cai et al., 2007b). In contrast, rice cultivars (i.e., Hemeizhan, Hefengzhan, Huahang 31, and Tianyou 998) whose grains contained high DEHP levels are unsuitable for planting in DEHP-contaminated soil.

Table 5
Translocation factors of DEHP between different tissues at ripening stage.

	Cultivars	Root–stem	Stem–leaf	Shoot–grain
Normal	Hemeizhan	1.64 ± 0.34c[a]	1.52 ± 0.53c	3.59 ± 0.48a
	Guinongzhan	0.62 ± 0.29c	2.20 ± 0.11c	0.90 ± 1.37b
	Hefengzhan	0.40 ± 0.34c	3.87 ± 0.57ab	3.73 ± 4.80a
	Fengmeizhan	0.38 ± 0.00c	3.08 ± 0.41b	0.17 ± 0.04b
	Yuxiangyouzhan	1.37 ± 0.19c	1.50 ± 0.84c	0.07 ± 0.02b
	jinnongsimiao	0.48 ± 0.18c	2.84 ± 0.69bc	0.19 ± 0.03b
	Huahang 31	14.55 ± 1.40a	0.85 ± 0.36cd	0.07 ± 0.03b
Hybrid	Tianyou 103	0.15 ± 0.02c	4.66 ± 1.42ab	0.60 ± 0.22b
	Tianyou 122	0.33 ± 0.06c	4.67 ± 0.42a	0.20 ± 0.10b
	Tianyou 2168	0.82 ± 0.15c	1.49 ± 0.38c	0.68 ± 0.74b
	Tianyou 998	6.32 ± 5.98b	3.62+0.16b	3.18 ± 2.24a
	Tianyou 390	1.36 ± 0.90c	0.42 ± 0.17d	0.48 ± 0.47b
	Tianfengyou 316	11.67 ± 1.64a	0.22 ± 0.03d	0.10 ± 0.07b
	Fengyousimiao	0.29 ± 0.14c	4.24 ± 0.89ab	0.30 ± 0.13b
	Fengyou 428	7.74 ± 3.92b	0.17 ± 0.02d	0.38 ± 0.48b
	Wufengyou 128	0.47 ± 0.17c	2.34 ± 0.88bc	0.59 ± 0.37b
	Wufengyou 2168	1.28 ± 0.99c	1.53 ± 1.35c	0.56 ± 0.19b
	Wuyou 308	13.2 ± 64.7a	0.17 ± 0.08d	0.04 ± 0.00b
	Yueza 889	0.36 ± 0.06c	4.22 ± 1.36ab	1.96 ± 1.13ab
	Peizataifeng	5.65 ± 2.39b	0.18 ± 0.12d	0.08 ± 0.03b

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

3.3. *DEHP translocation among rice tissues*

The transportation of contaminants from soil to the grains of rice involved: (1) absorption from soil to root tissue; (2) efflux to xylem or sequestration in root cells; (3) xylem transport to shoot; (4) xylem-to-phloem transfer in nodes; (5) phloem transport to the grains; and (6) post-phloem transport and accumulation in grains (Fujimaki et al., 2010). To increase our understanding of the transportation of DEHP in rice tissues, the translocation factors (TFs, referred to DEHP concentration ratio of stems to roots, leaves to stems, or grains to shoots) were calculated (Table 5). The TFs of DEHP in various rice tissues varied greatly from 0.04 to 14.6. The highest TF of DEHP was from roots to stems, and those of cultivars Huahang 31, Tianfengyou 316, and Wuyou 308 were greater than 10.0, significantly higher than those of the other cultivars. Variations in the TFs of DEHP from stems to leaves were smaller, from 0.17 to 4.66. The TFs of DEHP from shoots to grains ranged from 0.04 to 3.73, and those for 80% of cultivars were less than 1.0, and less than 0.1 for cultivars Yuxiangyouzhan, Huahang 31, Wuyou 308, and Peizataifeng. In contrast, the TFs of DEHP from stems to leaves were higher than 1.0 for 70% of cultivars. Obviously, the TFs of DEHP from roots to stems and from stems to leaves were relatively higher. This trend differed from the TF values for cadmium; i.e., the TFs from rice shoots to ears (heading stage) or grains (maturity) were markedly higher than those from roots to shoots (Liu et al., 2014). As mentioned above, root-to-shoot cadmium translocation via xylem is a major physiological process controlling cadmium accumulation levels in the shoots and grains of rice (Uraguchi et al., 2009). As stated previously, rice stems and leaves could accumulate DEHP through aerial uptake, which may result in higher TFs for root-to-stem and stem-to-leaf. The specific contribution from root and aerial uptake to DEHP accumulation in rice tissues should be further investigated based on stable isotope tracing experiments.

The TFs of DEHP also differed among rice cultivars. For example, DEHP TFs from roots to stems were high, 14.6, 13.2, and 11.7 for cultivars Huahang 31,Wuyou 308, and Tianfengyou 316, respectively; while they were less than 0.5 for cultivars Tianfeng 103, Tianfeng 122, Fengyousimiao, Yeza 889, Hefengzhan, and Fengmeizhan (Table 5). The TFs of DEHP from shoots to grains for cultivars Hemeizhan (TF=3.59), Hefengzhan (TF=3.73), and Tianyou 998 (TF=3.18) were more than 30-fold higher than those of the other cultivars (TFs ≤ 0.1). Significant variation in TFs for cadmium and arsenic by various rice cultivars has also been reported (Uraguchi et al., 2009; Ye et al., 2012; Liu et al., 2014) and for DEHP by several cultivars of *Ipomoea aquatica* and *Brassica parachinensis* (Zeng et al., 2005; Cai et al., 2008b). Generally, contaminants in grains may be derived from either direct xylem transport from roots or remobilization of shoot contaminant pools through the phloem during grain filling (Norton et al., 2010). In this study, rice genotypes significantly affected the TFs of DEHP from roots to stems and shoots to grains. These results demonstrated that DEHP concentrations in grains were associated with DEHP uptake and transportation from roots and shoots to grains.

The differences in TFs between normal rice and hybrid rice were non-significant ($P > 0.05$). Similarly, Liu et al. (2013) reported that differences in the TFs of lead from roots to shoots between rice types were not significant. These results suggested that the variation in lead and DEHP uptake and translocation in rice plants was genetically mediated. Breeding of rice cultivars with low contaminant accumulation could effectively reduce the influx of contaminants into the food chain at almost no additional cost (Ye et al., 2012). In the present study, cultivars (i.e., Huahang 31, Wuyou 308, and Tianfengyou 316) with higher TFs from roots to stems showed lower TFs (TFs < 0.1) from shoots to grains, indicating that these cultivars could be used to decrease DEHP translocation from roots and shoots to grains, and thus to reduce DEHP levels in the edible components. As mentioned previously, the agricultural soils (especially paddy-field soil) of Guangdong province showed higher levels of DEHP compared with other provinces of China (Yang et al., 2007; Niu et al., 2014). Breeding rice with low concentrations of DEHP in grain (as well as low TFs from shoots to grains) could provide a means for farmers to cope with this risk and decrease the influx of contaminants into the human food chain.

4. Conclusions

Concentrations of DEHP in rice plants differed significantly among rice cultivars, growth stages, and tissues. The differences in DEHP concentrations among rice cultivars were large at the ripening stage, but relatively small at the flowering stage. At the ripening stage, the greatest variations in DEHP concentrations among cultivars were in stems, whereas the least was in roots. Significant differences in DEHP concentrations were observed among tissues for the majority of rice cultivars. The TFs of DEHP from stems to leaves or roots to stems were higher than those from shoots to grains. When the total concentration in the whole plant is taken into consideration, cultivars Tianfengyou 316, Wuyou 308, and Peizataifeng, which had lower DEHP in grains but higher levels in stems and leaves, were ideal cultivars for planting in soil with low or medium levels of DEHP. Further studies should focus on the pathway of DEHP uptake and translocation in rice and the underlying molecular mechanism.

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