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Uptake, translocation, bioaccumulation, and bioavailability of organophosphate esters in rice paddy and maize fields

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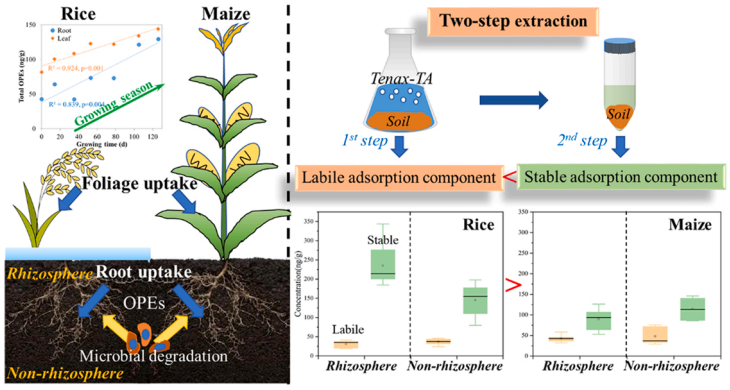
HIGHLIGHTS

- We used two-step extractions to obtain the labile and stable adsorbed OPEs in soil.
- OPEs in rice tissues increased linearly with the growing time.
- Rice root exudates increased the bioavailability of OPEs in soil during its growth.
- *Sphingomonas* and *Geobacter* associated with OPE degradation enriched in rhizosphere.

GRAPHICAL ABSTRACT



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ABSTRACT

Rice and maize are two main crops with different growth habits in Northeast China. To investigate the uptake, translocation, and accumulation of organophosphate esters (OPEs) in those two crops, we measured the OPE concentrations in their agricultural soil-crop systems during different growing seasons. OPE concentrations were higher in paddy (221 ± 62.0 ng/g) than in maize (149 ± 31.6 ng/g) soil, with higher OPE levels in the rhizosphere than in bulk soil for rice, and the opposite in maize. Two-step extractions were used to obtain the labile and stable adsorption components of OPEs. The stable-adsorbed OPEs were activated to be more bioavailable by root exudates as rice grew. OPEs in rice increased linearly with the growing period. The uptake and translocation processes of OPEs by crops were not well-explained by logK_{ow} alone, indicating other processes such as growth dilution are significant for understanding OPE levels in plant. The translocation factors of OPEs from nutritive to reproductive organs indicated that OPEs in rice seeds may follow the translocation from root to leaf and then transfer to grains. Two genera, *Sphingomonas* and *Geobacter*, associated with degradation of organophosphorus compounds were enriched in rhizosphere soils, indicating enhanced OPE degradation.

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1. Introduction

Following the global prohibition of brominated flame retardants, organophosphate esters (OPEs) have been increasingly used as alternatives due to their excellent ability to meet flammability standards in products. In 2015 global OPE consumption was ~680,000 tons, and it continues to increase annually[43]. OPEs in most products are used as "additive" flame retardants and plasticizers, and are therefore easily released into the environment through volatilization and abrasion[54]. This has led to high observed OPE levels in many environmental media [43,47]. Alongside these high concentrations, toxicological and epidemiological evidence has shown that exposure to OPEs may cause harmful health effects, including endocrine disruption, reproductive failure, and immune toxic effects[28,59].

Dietary ingestion is believed to be the dominant exposure pathway for many OPEs, making it essential that we understand the dynamics of OPE uptake in plants, especially in crops. OPEs can be absorbed either by plant roots from soil and transported to shoots[31] or by plant leaves from air via gaseous uptake and particulate deposition[5]. OPE levels in soil can be altered by plant through biodegradation or bioaccumulation [31], while their bioavailabilities in soil can be affected by root exudates or deposits [21]. Gaps remain in our understanding of OPE uptake dynamics, as many of these studies have focused on hydroponically grown crops or have not evaluated plant uptake across the entire growing season in field settings.

Rice (*Oryza sativa* L.) and maize (*Zea mays* L.) are two of the most widely-grown staple-crops worldwide, with rice responsible for feeding over half of the world's population[10], while maize provides almost half of the calories to populations in Africa and America[38]. These two crops are also the main food crops in Northeast China. The different species and growing conditions of these two crops may lead to dissimilar environmental fates of organic pollutants in their fields. OPEs have been frequently detected in Chinese agriculture soils[17], including rice paddies[58]. Rice consumption is considered as a major pathway for human exposure to OPEs in China[56]. Maize plants can also uptake OPEs via hydroponic cultures[2], although lower levels have been observed in Chinese agricultural soils than in rice paddies[49], presumably due to differing horticultural practices including less irrigation to maize fields than rice paddies.

Generally, soil can act either as a reservoir or a source for organic pollutants. After being released into soil, a pollutant's bioavailability and environmental behaviors can be significantly influenced by aging (or weathering) processes[1]. According to sorption and desorption processes, pollutants in soil can be divided into: dissolved, "rapidly desorbed" (surface-adsorbed), "slowly desorbed", and "very-slowly desorbed" (strongly bounded or micropore-adsorbed) components[4, 33]. Only the dissolved and surface-adsorbed fractions are considered bioavailable to organisms[7]. Therefore, it is crucial to furnish detailed information on the fractions, bioavailability, and transformation of organic pollutants in soils. An extraction method using Tenax-TA, a polymeric sorbent synthesized from 2,6-diphenyl-p-phenylene oxide, has been developed and confirmed to be a promising technique for measuring the bioavailable fraction of virous organic pollutants in soil [4,42].

Considering that studies on the uptake of OPEs by crops during the entire growing period are still very limited, especially in the field environment, the objectives of this study were to: (1) investigate the concentration, distribution, and bioaccumulation of OPEs in rice and maize and their corresponding soils during different growing seasons; (2) discover the influence of crops on the variations of bioavailability of OPEs in soil; (3) explore the uptake and translocation of OPEs in soil-crop systems; and (4) seek the potential effects of soil microbial communities on the degradation of OPEs. The result will help us to have a better understanding of uptake and accumulation of OPEs by crops in the fields.

2. Materials and methods

2.1. Sample collection

A rice (*Oryza sativa* L. subsp. *Japonica*) paddy and a nearby maize (*Zea mays* L.) field from a rural area of Benxi City (N41°18'52", E125°15'57"), Liaoning Province of China, were selected as the study area. Twenty-eight surface soil samples (0–10 cm depth) and 41 crop samples (23 rice and 18 maize samples) were collected from May to October, 2020. Soil and crop samples were collected during 8 different growing seasons of rice: seeding, regreening, tillering, jointing, heading, filling, mature, and idle (after harvest) periods. Crop samples were separated into root, stem, leaf, and seed (if any, only rice seeds were collected), and then washed with deionized water. Samples of soil from both the rhizosphere (distance to root <0.5 cm) and non-rhizosphere (distance to root >10 cm) zones were collected from both rice (7 RS and 8 NRS) and maize (6 RS and 7 NRS) fields. Each sample is a composite of 5 subsamples randomly collected from 5 different plants in the same field. All the samples were immediately transported to the laboratory, freeze-dried, ground, and stored at − 20 °C until further analysis. Soil organic matter content (SOM) was measured by the loss-on-ignition method at 500ºC for 3 h.

2.2. Sample extraction, purification, analysis & quality assurance/quality control

Briefly, soil samples were extracted in a two-step process based on a previous study[4] in order to obtain the labile adsorption component (dissolved & rapidly desorbed) and stable adsorption component (slowly desorbed) of OPEs. The bound-residue component (non-extractable) of OPEs cannot be measured using alkaline hydrolysis, since OPEs can also be hydrolyzed by alkaline under heating condition. Plant sample was extracted by an Accelerated Solvent Extractor (Dionex Inc.) at 100 °C for 2 cycles.

After being purified, eight typical OPEs, tri-*n*-butyl phosphate (TNBP), tri(2-chloroethyl) phosphate (TCEP), tris (1-chloro-2-propyl) phosphate (TCIPP), tris (1, 3-dichloro-isopropyl) phosphate (TDCIPP), triphenyl phosphate (TPHP), 2-ethylhexyl diphenyl phosphate (EHDPP), triphenyl-phosphine oxide (TPPO), and tricresyl phosphate (TMPP), (Table S1 in the Supporting Information, SI) were analyzed using a GCMS-QP2020 in electron ionization (EI) mode.

At least one procedure blank and one parallel sample were carried out for every 10 samples. The method detection limits (MDLs) were 0.03–2.35 ng/g for soil and 0.02–1.28 ng/g for crop (Table S2, SI). Reported concentrations are blank and surrogate-recovery corrected. Details of sample extraction, purification, determination, QA/QC, and data analysis and statistics are shown in S1-S3, SI.

2.3. Bioaccumulation and translocation assessments

Root concentration factors (*RCFs*) and translocation factors (*TFs*) were used to evaluate the bioaccumulation and translocation of OPEs in the plants, respectively. *RCFs* and *TFs* were calculated as [6,11]:

$$\mathrm{RCF} = \frac{C_{root}}{C_{soil}} \quad (1)$$

$$\mathrm{TF} = \frac{C_{shoot}}{C_{root}} \quad (2)$$

The root, shoot (stem or leaf), and soil concentrations (all ng/g) are denoted as C_{roots}, C_{shoot}, and C_{soil}, respectively.

2.4. Microbial community analysis

Rhizosphere and non-rhizosphere soils of rice and maize from rice tillering period were collected and shipped immediately with refrigerant

to the Sangon Biotech (Shanghai) Co., Ltd. of China for DNA extraction and microbial community analysis. The microbial community was analyzed by high-throughput 16 S rRNA pyrosequencing. Details are shown in S4, SI.

3. Results and discussion

3.1. OPEs in soils

The labile adsorption, stable adsorption, and total extraction (labile + stable adsorption) concentrations of OPEs in the rhizosphere (RS) and non-rhizosphere soils (NRS) of rice and maize during different growing seasons were measured (Fig. 1a). The total extraction concentration of Σ_8OPEs in paddy soil ranged from 125 to 364 ng/g with a mean value of 221 ± 62.0 ng/g, while the concentration in maize soil ranged from 98.5 to 197 ng/g with a mean of 149 ± 31.6 ng/g (Fig. 1a and Table S3). OPE concentrations in the paddy soils were significantly higher than those in the maize soil, especially for TNBP, suggesting that rice may be exposed to more OPEs than maize, probably mainly due to its horticultural practices of frequent irrigation or flooding (soil moisture content of rice: 18.4–40%, maize: 9.7–14.4%). Meanwhile, different plant dilution and transformation effects in rice and maize may also influence their OPE concentrations. OPEs in agricultural soil may come from irrigation water, sewage sludge application, and air deposition;

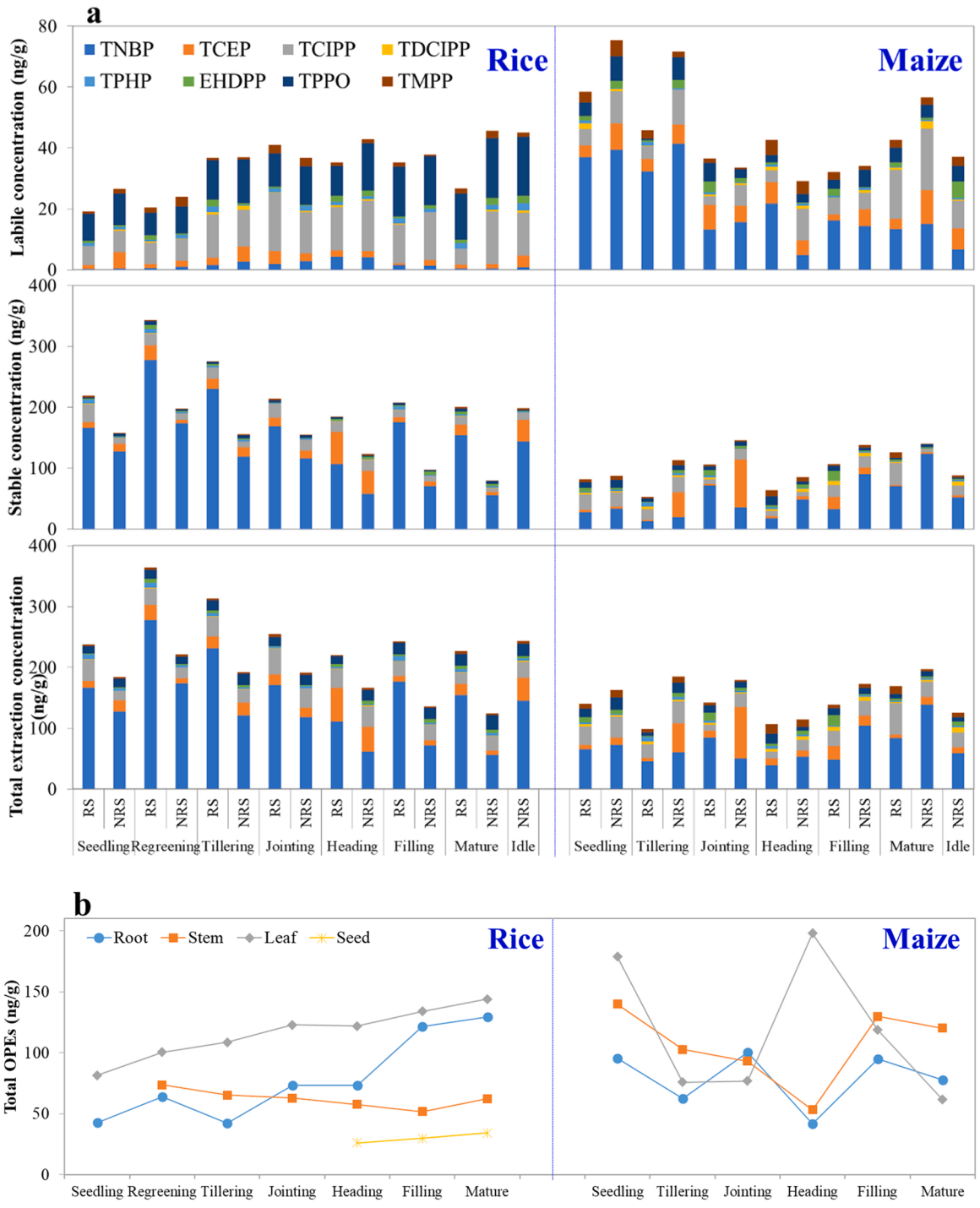

Fig. 1. Concentrations of OPEs in the rhizosphere and non-rhizosphere soils (a) and plant tissues (b) of rice and maize during different growing seasons.

oxidation derivatives of organophosphite antioxidants used in plastic mulch films are an additional potential source[13]. The OPE concentrations in this area were comparable with those in soil (4.50–430 ng/g, median 36.6 ng/g) collected across mainland China[48], slightly higher than those in farmland soil (0.54–54.9 ng/g) from Beijing-Tianjin-Hebei core area, Northern China[17], but much lower than those in soil (38–1250 ng/g) from a plastic waste treatment site[45].

In the two-step extraction, the labile fraction (1st step) is usually considered to determine the bioavailability of organic contaminants [35]. For paddy soil, the labile OPE concentrations ranged from 19.1 to 45.6 ng/g with a mean of 34.0 ± 8.6 ng/g, while the less bioavailable stable-adsorbed OPE concentration (2nd step) ranged from 79.4 to 344 ng/g (mean: 187 ± 66.3 ng/g, Table S3). For maize soil, the labile concentrations ranged from 29.1 to 75.4 ng/g (mean: 45.9 ± 15.2 ng/g), while the stable-adsorbed concentrations ranged from 52.7 to 146 ng/g (mean: 103 ± 29.5 ng/g). The stable adsorbed OPE concentrations were much higher than the labile OPEs in both soils, suggesting that more OPEs were stable adsorbed or bounded on soil particles than those dissolved in soil pore water or rapid adsorbed/desorbed in soil[4], especially for the OPEs with low octanol-water partition coefficient (K_{ow}).

For paddy soil, TPPO (39%) and TCIPP (36%) dominated in the labile fraction (Fig. S1, SI), while TNBP (45%) dominated in the stable-adsorbed fraction, followed by TCIPP and TCEP. By contrast, TNBP dominated both the labile (76%) and stable-adsorbed (48%) fractions (Fig. S1) in maize soil. As mentioned above, the relatively high compositions of TPPO and TCIPP in the labile fraction of paddy soil may due to the frequent irrigation or flooding of rice paddy, especially for these OPEs with relatively low K_{ow} (TCIPP and TPPO). Moreover, different microorganisms in paddy and maize fields may also lead to a different OPE composition[32].

The rhizosphere is a narrow region of soil where physical, chemical, and biological parameters of soil are significantly influenced by root secretions and associated microorganisms. The average concentrations of total extracted OPEs (labile + stable) in the rhizosphere soil (RS) and non-rhizosphere soil (NRS) were 266 ± 53.0 and 174 ± 34.0 ng/g for rice, and 133 ± 25.9 and 168 ± 28.8 ng/g for maize, respectively (Fig. 1a). For rice, almost all the OPE congeners were higher in the stable-absorbed component and the total components of RS than those in the NRS, but lower in the labile component of RS (except for TPHP and EHDPP). Furthermore, the labile fraction OPEs were negatively correlated with OPEs in the root ($p = 0.001$) and leaf ($p = 0.046$), indicating that plant tissues and labile soil shared a fixed mass of OPEs. These results suggest that rice roots primarily promote the absorption or degradation of labile OPEs in the rhizosphere, with the "activation" of non-extractable OPEs via root exudates playing a smaller but still noticeable role. For maize, the regular pattern of higher levels in the stable than the labile fraction was not consistent for all OPE congeners. Generally, OPEs with relatively low logK_{ow} (≤4) were lower in the RS than in the NRS, especially in the labile fraction and the total fractions. The results suggest that maize roots may also absorb labile OPEs and leave stable-absorbed OPEs behind, and thus uptake of OPEs by maize may be K_{ow}-depended. This can be proved by the significant correlations between the RS/NRS ratio of labile OPEs for maize and their logK_{ow} (R^2 =0.746, $p = 0.006$), but it was not the case for rice ($p = 0.201$). Plant uptake of OPEs from soil occurs either by depleting the mass in the labile fraction, or by "activating" the stable fraction to make it labile, due to the dissipative effect of root exudates on organic pollutants[12]. Besides the influence of root exudates, the different rhizosphere phenomenon between rice and maize may also be due to their different plant species or growth patterns. Significant positive correlations were found between the OPE concentrations in rhizosphere soils and their corresponding non-rhizosphere soils for both rice ($p \leq 0.018$) and maize ($p \leq 0.027$, except for the Jointing stage), suggesting that OPEs in the rhizosphere and non-rhizosphere originated from same sources.

The percentage of labile OPEs to the sum of labile and stable-adsorbed OPEs in soil are shown in Fig. 2. The labile fractions varied widely with plant species, growing period, and chemical properties. Generally, the labile fractions of most OPE congeners in the paddy soil were slightly higher than those in the maize soil, except for TNBP and TCEP. The relatively high labile component of OPEs in rice paddies vs maize fields may be due to the more frequent irrigation of rice paddies, which could replenish labile OPEs or transfer more root exudates from rhizosphere to bulk soil resulting in the activation of adsorbed OPEs. Meanwhile, the labile fraction in soil generally increased with the crop growing period, especially during the tillering, jointing, and heading periods, and then decreased as the crop matured. This pattern suggests that more stable-adsorbed OPEs were activated to be more bioavailable by root exudates with the rapid growth of crops. However, as the plant matured, the reduction of root exudates coupled with continued or increased plant uptake caused the decrease of labile OPEs in soil. The activation can continue to promote the plant accumulation of OPEs, which may lead to an increased potential health risks through food chain. The labile fractions of OPEs in the RS were generally lower than these in the NRS for both rice and maize, especially during their early life stage, indicating that crop root preferentially absorb or adsorb labile over stable OPEs. We found no significant correlations ($p > 0.05$) between SOM (Table S3, SI) and labile OPE percentages in soil, suggesting that SOM did not influence OPE bioavailability, unlike some other chemicals such as PCBs[19].

We found no correlation between logK_{ow} and the median concentrations of labile fractions of OPEs in paddy soil, but a weak positive correlation (Fig. S2a, R^2 =0.553, $p = 0.034$) with logK_{oa}, suggesting the bioavailability of OPEs might be related to their K_{oa} or volatilities. During growth, rice roots undergo alternating aerobic and anaerobic processes accompanied with flooding and drainage practices, which may influence the soil redox potential (Eh) and lead to a different environmental fate of OPEs in paddy field compared with dry land. By contrast, the labile OPE percentages in maize soil were negatively, but not significantly, correlated with logK_{ow} (Fig. S2b, R^2 =0.435, $p = 0.075$). This negative correlation suggests that labile OPEs in maize soil may come from water irrigation or precipitation.

3.2. OPEs in crop tissues

Generally, OPE concentrations in maize tissues were slightly higher than those in rice tissues despite the higher OPE concentrations observed in paddy soil. The total OPE concentrations in the rice tissues ranged from 26.1 to 144 ng/g (mean: 79.2 ± 36.2 ng/g); vs 41.6–198 ng/g (mean: 101 ± 41.5 ng/g) for maize, as shown in Fig. 1b and Table S4, SI. The uptake, accumulation, and translocation of OPEs by plants depends on several plant-, compound-, and location-specific processes, and therefore the observed differences between rice and maize could be driven by compound-specific processes at each site, species differences between two crops, or cropping patterns. Σ_8OPEs in the rice and maize tissues (26.1–198 ng/g) in this study were slightly higher than those in wheat (9–51 ng/g) from farmlands near the plastic treatment areas[45] and grains (mean: 36.9 ng/g ww) from Belgium [30]; but slightly lower than those in rice seeds (including kernel and husk, mean: 298 ng/g) from Dalian[49] and rice (0.38–287 ng/g) from Hubei, Chongqing, Sichuan, and Guangxi of China[56].

The OPE concentration in rice roots, leaves, and seeds increased linearly across the entire growing season, with the concentrations peaking in the mature stage (root: R^2 =0.839, $p = 0.004$; leaf: R^2 =0.924, $p = 0.001$; seed: R^2 =0.987, $p = 0.072$; Fig. S3, SI), while concentrations in the stem tended to decline (decline, R^2 =0.974, $p = 0.002$, without the Mature stage). However, OPEs showed no obvious bioaccumulation in maize tissues across the growing period. This may be attributed to a relatively higher elimination of OPEs or growing rate vs their accumulation rates in maize. OPEs can be accumulated by plant via root uptake or foliage absorption, and their concentrations may decrease due to biotransformation, growth dilution, or

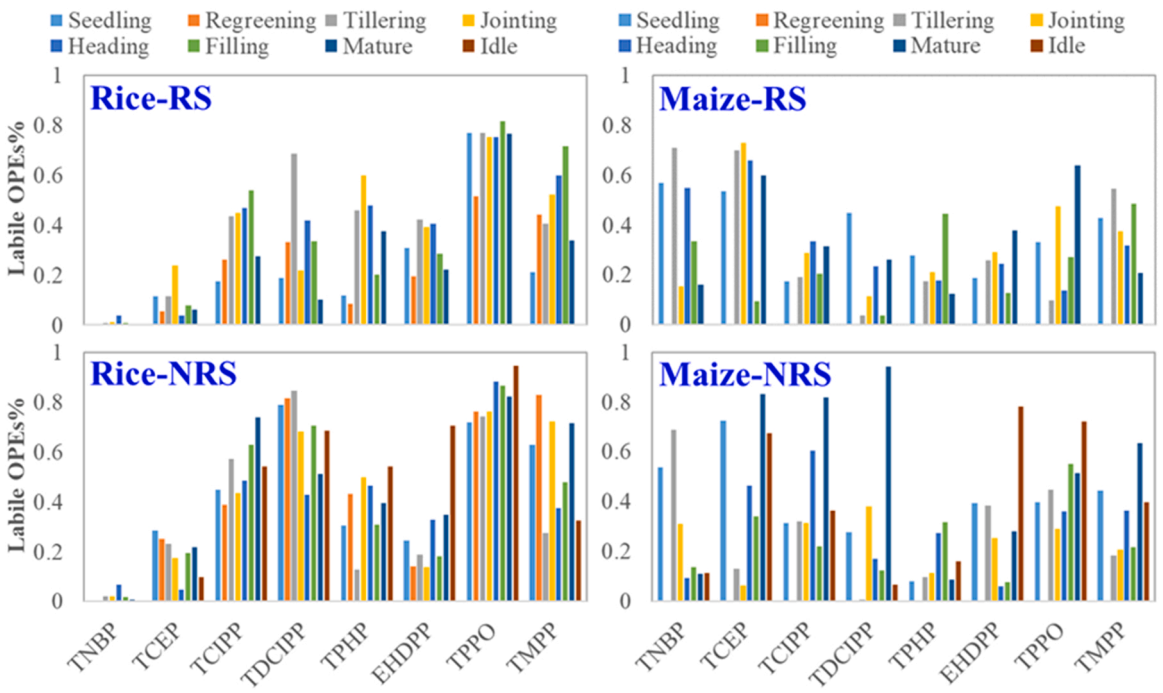

Fig. 2. Percentage of labile OPEs to the sum of labile and stable-adsorbed OPEs in the rhizosphere and non-rhizosphere soils of rice and maize.

translocation to other parts of the plant. For the root uptake pathway, OPEs in soil (or water) first adsorb to the plant root epidermis [51,55], and then transport into root interior through either the symplastic pathway (through plasmodesmata) or the apoplastic pathway (through intercellular space between cell wall and membrane)[27]. A rapid equilibrium between plant root and hydroponic solution indicates that OPEs may mainly enter roots via passive diffusion[40] followed by apoplastic transfer to the vascular bundle, and accumulate in plant leaves[23]. The labile component of OPEs in RS may be the major contributor to the OPEs in rice, given the significant negative correlations between rice tissues and their labile fraction in the corresponding RS (Spearman; root vs RS, $p = 0.001$; leaf vs RS, $p = 0.046$) and their labile fraction in the corresponding RS. However, this was also not the case for maize.

OPE concentrations also varied with crop tissues OPEs in rice followed the order of leaf > root > stem > seed, while OPEs in maize followed the order of leaf > stem > root (Table S4). Surprisingly, OPEs in crop leaves were much higher than these in their roots. Besides root uptake, airborne OPEs released from sources or re-suspended from soil can also enter the aboveground part of plant in the form of gas and particle-bound via exchange or deposition[20,61,8]. The relatively high OPE concentrations in crop leaves suggests that foliage uptake may also be an important pathway for OPEs to enter crops. A previous study also illustrated that accumulation of airborne organic pollutants is important for crops especially after the seedling stage[55]. A chamber study with different exposures of PBDE-contaminated air, soil, and dust found that foliar uptake of wheat from the air and particle contributed 81.3%–99.6% of PBDEs accumulated in leaves[61]. The occurrence of OPEs in plant tissues was the combined result of uptake in root and foliage pathways, translocation and metabolism in a long-term dynamic equilibrium[23,44]. Thus, the relatively low concentrations in rice seeds may be due to its short growing period.

3.3. Bioaccumulation and translocation of OPEs in crops

Bulk soil concentrations are typically used when calculating *RCF*, but this introduces some uncertainty as rice and maize roots only have access to the soil directly around plant roots in the rhizosphere. To address this, we measured the OPE concentration in the rhizosphere and used that along with the root concentration to calculate *RCF*. *RCFs* of rice ranged from 0.018 to 5.03, while *RCFs* of maize ranged from 0.004 to 22.8 (Table S5). The *RCFs* calculated in this study were comparable with those in the previous studies (10^{-2}-$10^{3.5}$)[23,44,46]. Only the median *RCFs* of TDCIPP and EHDPP were larger than 1 in rice, and those of TCIPP, TDCIPP, and TPHP in maize. The result suggests that the intake of most OPEs by crop roots was relatively low.

Our results showed no significant correlation between the measured log*RCFs* and logK_{ow} or logK_{oa} ($p > 0.05$) in this study, suggesting that factors beyond a compound's physicochemical properties dominate OPE uptake in field conditions. The uptake, translocation, and accumulation of organic pollutants by plants is influenced by chemical's physicochemical properties and by plant physiology[3], and by factors such as soil texture, moisture, and organic content; or plant species, lipid content, carbohydrate content, fiber content, and leaf morphology[9,55]. Previous hydroponic studies have shown a positive correlation between log*RCFs* and logK_{ow} in hydroponic systems[44], but weaker to less significant negative correlations have been found in soil culture systems [15,57]. Together, this indicates that using plant–soil partition coefficients to characterize plant accumulation of these compounds appears unreliable, due potentially to the many processes involved in the plant uptake of chemicals[34].

The median *TF* values of OPEs based on their concentrations in root, stem, and leaf were 0.59–4.21 for rice and 0.44–3.51 for maize, respectively; while the median *TF* values of OPEs for seed/root, seed/stem, and seed/leaf for rice were in the range of 0.07–1.28 (Fig. 3 and Table S6). *TFs* varied widely with crop species, tissues, and growing periods. Generally, *TFs* of leaf/stem were much higher than these of stem/root (except for TPPO in rice, TCIPP and TMPP in maize). The higher concentrations in leaf may be due to that either OPEs are more likely to be accumulated in leaves than stems, or OPEs in leaves also include the contribution of foliage uptake. Moreover, the detected concentrations in roots or leaves included both surface adsorbed and absorbed sections, which are difficult to be distinguished and can also affect the *TF* values.

Similar to *RCFs*, the relationships between *TFs* and logK_{ow} were explored, and we found that traditional relationships based on simple

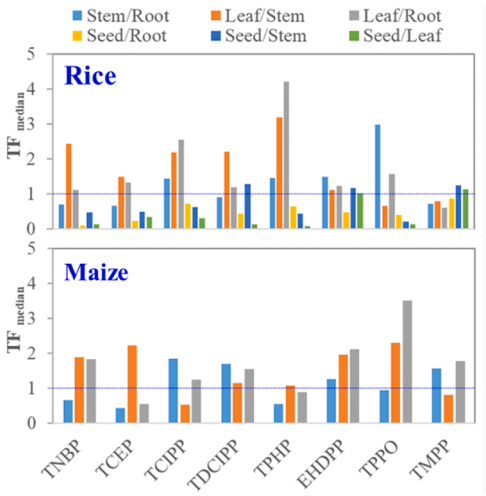

Fig. 3. Median *TF* values of OPEs in rice and maize calculated by their concentrations in different tissues.

partitioning again did not apply. No correlations were found between logK_{ow} of the 8 target OPEs and *TFs* calculated from different tissues. However, a significant negative correlation ($p = 0.03$) was found between leaf/root *TFs* and logK_{ow} of 5 OPEs in rice (without TCEP, EHDPP, and TPHP), whereas a significant positive correlation ($p = 0.004$) was found between leaf/root *TFs* and logK_{ow} of 6 OPEs in maize (without TPPO and TPHP) (Fig. 4a). Usually, highly polar and water-soluble compounds are more likely to be translocated within plant tissues via the transpiration stream, which leads to a negative correlation with logK_{ow}. However, a positive correlation was also found between root-to-stem translocation and logK_{ow} (6.31–9.45) of PBDEs in rice[57], because the higher hydrophobic compounds are more likely to bind to major latex-like proteins in xylem sap[16]. Here, the unusual positive correlation between leaf/root *TFs* and logK_{ow} in maize may be attributed to the distinct contribution of foliage uptake of OPEs. Zhang et al. [57] also found that root-to-stem *TFs* of PCBs in rice tended to increase with the increasing of logK_{ow}, but with no statistical significance. However, even compounds with similar K_{ow} values can also show drastically different partitioning and accumulation[25].

Our results also suggested that the translocation of OPEs from the nutritive organ to the reproductive organ, such as fruits or seeds, was limited, with *TF* values of OPEs from nutritive organs to rice seed generally lower than 1 (except TMPP and EHDPP). This may be due to the strong capacity of OPEs with high logK_{ow} to partition to root or leaf epidermal lipids[50] and the blockage of Casparian strips[60]. Moreover, the shorter growing period of reproductive organs than nutritive organs may also contribute to the lower *TFs* of seed/other tissue. A significant negative correlation (R^2 =0.825, $p = 0.012$) was found between seed/leaf *TFs* and logK_{ow} of 6 OPEs in rice (except for EHDPP and TMPP), whereas a significant positive correlation (R^2 =0.867, $p = 0.021$) was found between seed/root *TFs* and logK_{ow} of 5 OPEs in rice (except for TNBP, TCIPP, and EHDPP) (Fig. 4b). The accumulation of OPEs in rice seeds was suggested to follow the translocation from root to leaf blades via xylem driven by transpiration, remobilization from leaf blades via phloem, and finally transfer into grains[41,52]. The relationship between seed-based TFs and logK_{ow} seems to be consistent with the explanation, but the hypothesis still needs further experimental validation.

3.4. Microbial community analysis

Rhizosphere and non-rhizosphere soils of rice and maize from the tillering period were analyzed via high-throughput 16 S rRNA pyrosequencing. The microbial richness and diversity are shown in Table S7, SI. For bacteria, the RS and NRS samples of maize showed similar community richness (Chao1 index) but different α-diversity (Shannon or Simpson index) with the diversity higher in the RS; whereas the community richness and α-diversity in the RS of rice were both significantly higher than those in the NRS. For archaea, only the paddy soils were analyzed, and the RS and NRS showed similar community richness but different α-diversity with the diversity also higher in the RS. The significantly higher richness or diversity of bacteria and archaea in the RS may due to the influence of crop root exudates and deposits.

We detected 42 bacterial genera from 11 phyla and 8 archaeal genera from 4 phyla (Fig. 5). *Proteobacteria* (32.2–38.3%), *Actinobacteria* (9.7–31.3%), *Acidobacteria* (7.3–11.9%), *Cyanobacteria* (0.35–20.9%), *Bacteroidetes* (3.6–8.3%), *Chloroflexi* (2.2–4.4%), *Planctomycetes* (1.9–3.5%), *Gemmatimonadetes* (2.0–2.4%), and *Verrucomicrobia* (0.69–4.2%) were the most prevalent phyla (Fig. 5a), which were similar to those in the soil from a watershed [53] and a wetland of China[24]. *Proteobacteria*, documented to consist of many key denitrifiers[26], was the most dominant phylum in both rice and maize soils. *Proteobacteria* may be significantly increased by the exposure of OPEs[24]. Meanwhile, *Acidobacteria* phylum, enriched in the rice rhizosphere, was also suggested to be increased by the presence of OPEs[24]. Two genera, *Sphingomonas* and *Geobacter* (Fig. 5c), have been reported to be associated with the degradation of organophosphorus compounds[14,29]. *Sphingomonas* (rice: 3.4–3.5%, maize: 5.4–7.6%) can also utilize TNBP as the sole carbon source[22] and degrade TCEP and TDCIPP by hydrolyzing their phosphotriester bonds[37]. *Geobacter* (1.5–2.2% in paddy soil), one of the commonly studied electrogenic bacteria using acetate as electron donors, was found to be sensitive to the exposure of OPEs[14].

Thaumarchaeota, *Euryarchaeota*, and *Woesearchaeota* were the 3 most

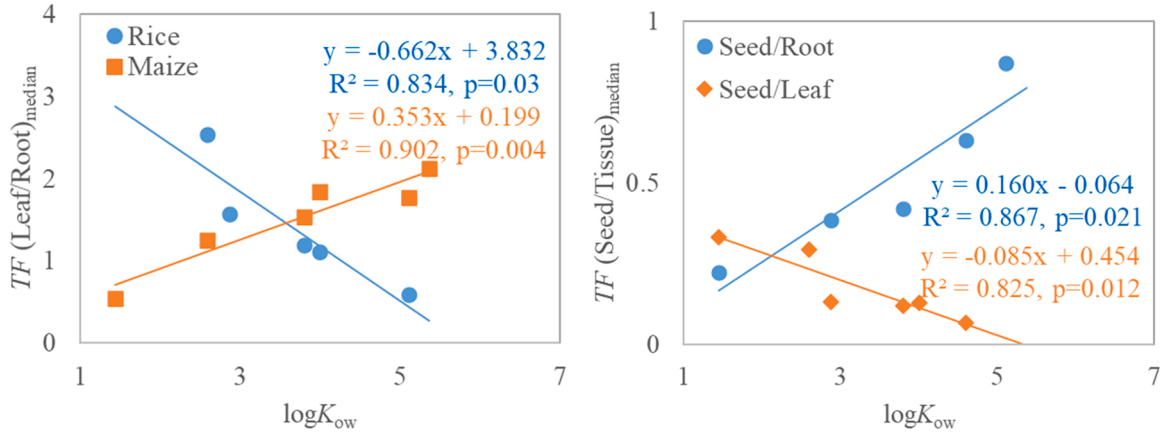

Fig. 4. Correlations between *TF* values of OPEs in rice and maize and the logK_{ow}.

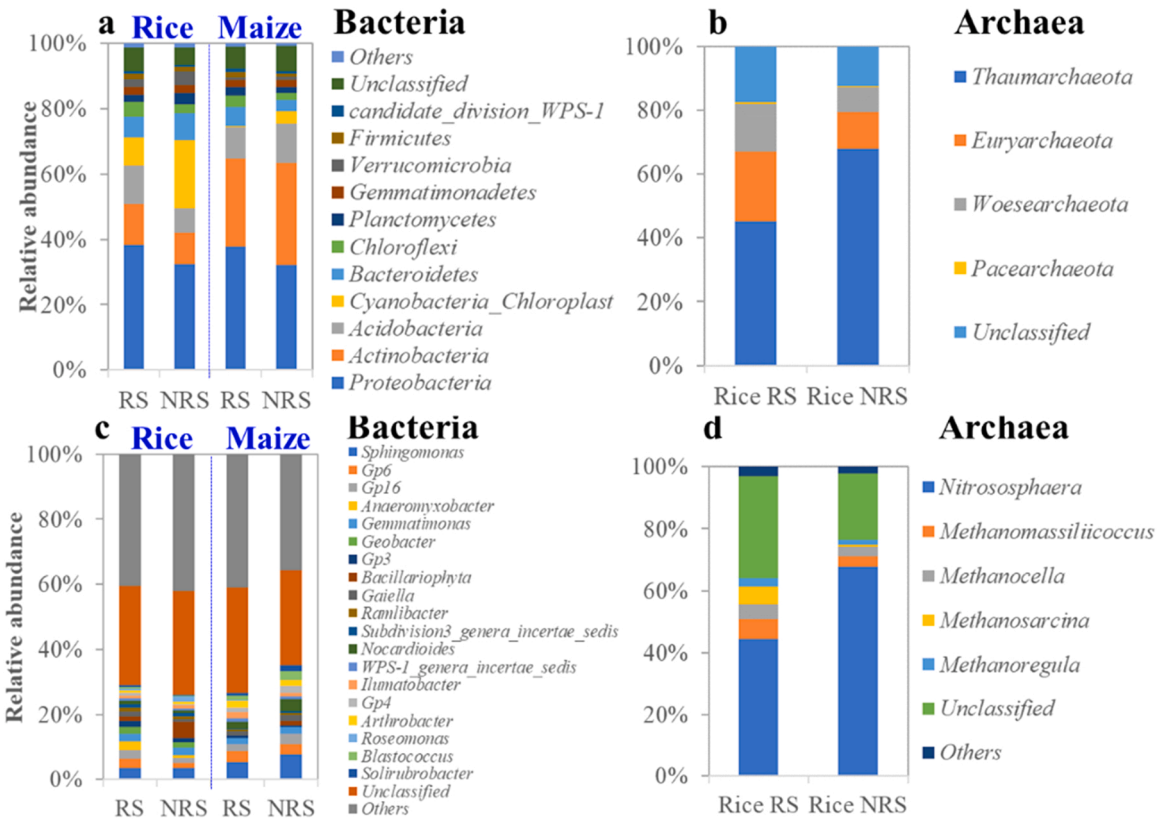

Fig. 5. Microbial community structure at the phylum (a, b) and genus (c, d) levels in the soils of rice and maize.

prevalent phyla accounting for 82.1–87.3% of all archaeal gene sequences (Fig. 5b). *Nitrososphaera* was the most predominant genus in paddy soil. It belongs to the ammonia oxidizing archaea (AOA) and can catalyze the ammonia oxidation process of ammonia oxidizing bacteria (AOB)[18]. Meanwhile, the content of Methanogens, such as *Methanomassiliicoccus*, *Methanosarcina*, *Methanocella*, and *Methanoregula*, were increased in the rhizosphere soil compared with the bulk soil due to the "rhizosphere effects"[36]. Methanogens are a group of strictly anaerobic archaea directly involved in the methane production[39]. The strictly anaerobic habitat of paddy soil may lead to a significant different transformation or degradation of OPEs compared with maize soil. The effects of microbial community diversity and functional microorganisms on the environmental fate of OPEs in rhizosphere soil deserve further investigation.

4. Conclusion

We used field studies of rice and maize as test cases to investigate the influence of crop growth on the bioavailability of OPEs in the rhizosphere, and uptake, accumulation, and translocation of OPEs within soil-crop systems. Overall, our result suggested that plant tissues and soil labile fraction may share a fixed mass of OPEs, and plant roots can promote the absorption or degradation of labile OPEs in the rhizosphere. Rice may activate the non-extractable OPEs via root exudates, but this activation plays a negligible role for maize. One upside of our results is that the accumulation of OPEs in rice grain was relatively low compared with other tissues, although the reason remained unclear. Two genera related to degradation of OPEs were elevated in the rhizosphere soil, indicating that plant roots may promote the degradation of OPEs. Our results suggest that the uptake and translocation of OPEs in crops in the "real world" is a very complicated process, and cannot be well explained by a single physicochemical parameter. Further field studies are still required.

Supplementary data

Sections S1-S4, Tables S1-S7, and Figures S1-S3 are shown in the supplementary data. Supplementary data associated with this article can be found, in the online version.

CRediT authorship contribution statement

Yan Wang: Conceptualization, Writing - review & editing, Supervision, Project administration, Resources, Funding acquisition; **Junjie Li**: Data curation, Writing - original draft; **Yue Xu**: Resources, Writing - review & editing; **Timothy F. M. Rodgers**: Methodology, Writing - review & editing; **Meijun Bao**: Investigation, Formal analysis; **Feng Tan**: Resources, Writing - review & editing.

Environmental implication

As alternatives to brominated flame retardants, organophosphate esters (OPEs) have aroused strong concerns due to their toxicities and potential threats to human health, especially via food consuming. However, information on the uptake and accumulation of OPEs by rice and maize during their whole growing seasons is very limited. The impact mechanisms of rice or maize growth on the bioavailability of OPEs in soil are still unclear. This study will help us to better understand the uptake, translocation, and bioavailability of OPEs in agricultural soil-crop systems of rice paddies and maize fields during their growing seasons.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2022.130640.

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