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Review

Organophosphate esters in agro-foods: Occurrence, sources and emerging challenges

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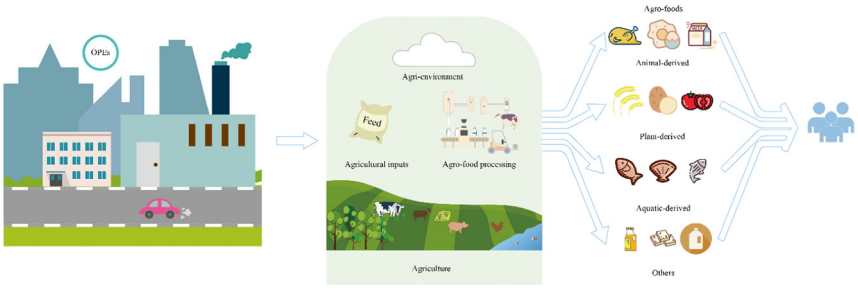
HIGHLIGHTS

- OPEs are globally distributed and ubiquitous in various agro-foods.
- Three potential sources of OPEs contamination in agro-foods are suggested.
- Plants and animals can absorb OPEs via the contaminated agri-environment.
- OPEs could transfer from feed to food via animal breeding.
- Packaging and processing could be potential sources of OPEs in agro-foods.

GRAPHICAL ABSTRACT



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ABSTRACT

Safety and sustainable agro-food production is important for food and nutrition security. Agro-foods safety is challenged by various emerging environmental contaminants. Organophosphate esters (OPEs) have been reported to occur in various agro-food items worldwide, which has resulted in increasing concerns for effects on health of humans and wildlife, including through agriculture. However, information on presence, sources and transfer routes of OPEs in agro-foods, and consequent health risks remains scant. This review critically evaluates available information on concentrations of OPEs in various agro-foods, and discusses potential sources of OPEs in agro-foods, which are closely related to the ambient agri-environment, agricultural inputs, and agro-foods processing. Some directions for future research are suggested. First, since food is an important exposure pathway to OPEs, systematic monitoring of concentrations of OPEs in various categories of agro-foods is recommended. Second, surveillance of concentrations and characteristics of OPEs in agro-foods and ambient agri-environments, agricultural inputs or processing in the agro-food chain is needed to obtain a more complete description of exposure and transmission behavior of OPEs in agro-foods. Third, future comprehensive studies of transmission, metabolism and accumulation of OPEs in animals or plants, are required. Finally, measures to control emissions of OPEs as sources to agriculture should be taken.

Contents

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1. Introduction

Organophosphate esters (OPEs), which are widely used as flame retardants (FRs) and plasticizers in furniture, textiles, plastics and electronics, have been increasingly produced worldwide in recent decades, since restrictions on uses of polybrominated diphenyl ethers (PBDEs) (Chen and Ma, 2021). OPEs can be divided into four main categories, including chlorinated (Cl)-OPEs, alkyl-OPEs, aryl-OPEs, and brominated (Br)-OPEs (van der Veen and de Boer, 2012). The global consumption of OPEs increased from 100,000 tons in 1992 to 680,000 tons in 2015 (Hou et al., 2016). In 2007, a total consumption of OPEs in Europe was estimated to be 85,000 tons, while more than 70,000 tons of OPEs were produced in China (Wang et al., 2015; Wang et al., 2010).

OPEs are frequently used as additives, rather than being chemically bonded to various materials (Chen and Ma, 2021). The continuous production and usage of OPEs have caused their globally distributed and ubiquitous in the environment, and thus resulted in their pervasive and increasing concentrations in abiotic and biotic environment matrices. As a class of emerging environmental contaminants, OPEs have been detected in diverse environmental matrices, such as air (Kim et al., 2019), dust (Yin et al., 2019; Zhao et al., 2020a), water (Bollmann et al., 2012; Pantelaki and Voutsa, 2019; Regnery and Püttmann, 2010), sediment (Giulivo et al., 2017; Hu et al., 2017), soil (Wang et al., 2018) and biota (Hallanger et al., 2015) samples. Moreover, due to increasing usage, concentrations of OPEs in environments are likely to continue increasing (Ye et al., 2021). The ubiquitous occurrence of OPEs in various environments might pose a threat to health of humans and wildlife. Presence of OPEs in human tissues, including hair (Liu et al., 2015), serum (Hou et al., 2020), urine (He et al., 2018a; Li et al., 2019b) and breast milk (Chen et al., 2021b; Sundkvist et al., 2010) have been reported. Human exposure to OPEs can occur via inhalation of indoor air and dust, but dietary exposure via food and drinking water is now recognized as another important pathway (Gbadamosi et al., 2021; Hou et al., 2016).

Agro-foods, including various edible plants, livestock and aquatic products, and the corresponding products of primary processing, are the main sources of food for human consumption (Li et al., 2015). OPEs have been reported to occur in various agro-food items, including cereals like rice and wheat, vegetables, fruits, meat, eggs, aquatic food and dairy products (Ding et al., 2018; Poma et al., 2018; Zhang et al., 2016; Zhao et al., 2019). Environmental pollution in agro-food production chain, such as agri-environments, agricultural inputs and agro-foods processing have greatly threatened agro-food safety. Considering the increasing production and complexity environmental behavior of OPEs and potential for bioaccumulation, it is likely that OPEs occur widely in agri-environments (e.g. agricultural lands, water), agricultural inputs (e.g. feed or bioresource materials), as well as in agro-foods processing (e.g. packaging), which might impose detrimental effects on human consumers through agro-food chain transmission. Therefore, risks of exposure to OPEs and potential adverse effects on health of humans who consume agricultural products should be assessed.

Several reviews of OPEs have been published in recent years. These reviews are mainly focused on analytical methodology for identifying and quantifying OPEs in the environment, biodegradation, metabolism, toxicity, and exposure to humans and wildlife (Liu et al., 2021; Pantelaki and Voutsa, 2019, 2020; van der Veen and de Boer, 2012; Waaijers and Parsons, 2016; Wang et al., 2020; Wei et al., 2015; Yao et al., 2021). Concentrations and distributions of typical OPEs in foodstuffs as well as human dietary exposure were reviewed (Li et al., 2019a). Another review assessed exposure of humans to OPEs with a focus on dietary intake (Gbadamosi et al., 2021). Exposure of humans to OPEs through informal e-waste handling activities, was reviewed and transfer of OPEs between various environmental media and associated human exposure pathways was elucidated (Ma et al., 2021). Because plants can accumulate OPEs from soil, water, and other environmental matrices, OPEs transformation OPEs in plants were summarized (Zhang et al., 2021b). But, a review focused specifically on OPEs in agriculture was still lacking. Given this, we gathered relevant studies and review articles published up to 2021 from the Web of Science database using “organophosphate flame retardants” (or “OPFRs”), “organophosphate esters” (or “OPEs”), and “food”, “agricultural” as the search items. The following inclusion criteria were considered: 1) the studies must be published in a scientific journal, 2) they must refer to edible agro-foods, and the occurrence of OPEs in ambient agri-environments, agricultural inputs and agro-foods processing. A total of 991 articles from the online database were found. In this article, we provided a synthesis and critical evaluation of the current state of knowledge of OPEs occurring in agro-foods, and proposed potential sources of OPEs in agriculture. Research gaps and directions for future studies were also proposed.

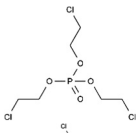
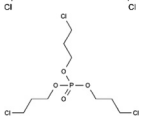
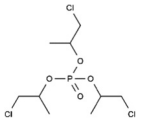
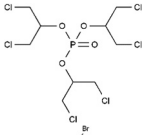
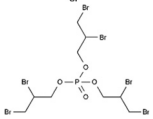
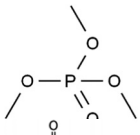
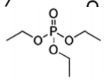
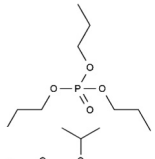
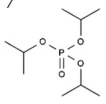
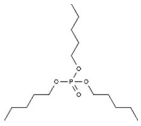
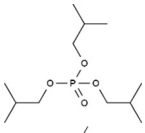
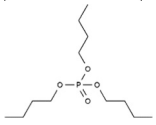
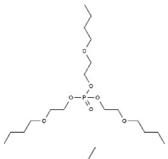
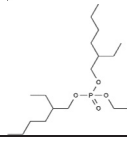
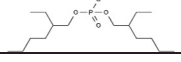
2. OPEs contamination in agro-foods

OPEs contaminations have been reported in a variety of agro-foods around the world. Here we have summarized names, structures, abbreviations, CAS numbers and properties of the 17 most prevalent OPEs reported in agro-foods (Table 1). Published data on OPEs were classified into four groups: plant-derived agro-foods, including cereals, vegetables and fruits, animal-derived agro-foods, including meats, dairy products and eggs, aquatic-derived foods and other agro-foods.

2.1. Plant-derived agro-foods

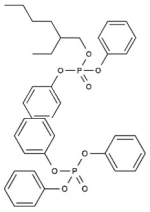
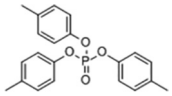
Plant-derived agro-foods, such as cereals, vegetables and fruits, are an indispensable part of human nutritional health, and are consumed by billions of people around the world. Moreover, epidemiological studies have

Table 1
List of major OPEs cited in this paper and their physicochemical properties.

Name	Abbreviation	CAS no.	Chemical structure	Molecular formula	Molecular weight (g/mol)	$LogK_{ow}$	BCF
Tris (2-chloroethyl) phosphate	TCEP	115-96-8		$C_6H_{12}Cl_3O_4P$	285.49	1.44	0.425
Tris (3-chloropropyl) phosphate	TCPP	1067-98-7		$C_9H_{18}Cl_3O_4P$	327.57	3.11	49.14
Tris (2-chloroisopropyl) phosphate	TClPP	13674-84-5		$C_9H_{18}Cl_3O_4P$	327.57	2.89	3.27
Tris (1,3-dichloro-2-propyl) phosphate	TDClPP	13674-87-8		$C_9H_{15}Cl_6O_4P$	430.90	3.65	21.4
Tris (2,3-dibromopropyl) phosphate	TDBPP	126-72-7		$C_9H_{15}Br_6O_4P$	697.61	4.29	47.06
Trimethyl phosphate	TMP	512-56-1		$C_3H_9O_4P$	140.07	−0.6	3.162
Triethyl phosphate	TEP	78-40-0		$C_6H_{15}O_4P$	182.15	0.87	3.162
Tripropyl phosphate	TPP	513-08-6		$C_9H_{21}O_4P$	224.24	2.35	1.911
Tri-isopropyl phosphate	TiPP	513-02-0		$C_9H_{21}O_4P$	224.23	2.12	24.0
Tripentyl phosphate	TPEP	2528-38-3		$C_{15}H_{33}O_4P$	308.39	5.29	268.2
Tri (isobutyl) phosphate	TIBP	126-71-6		$C_{12}H_{27}O_4P$	266.31	3.6	19.51
Tri-*n*-butyl phosphate	TNBP	126-73-8		$C_{12}H_{27}O_4P$	266.31	4.00	39.81
Tris (2-butoxyethyl) phosphate	TBOEP	78-51-3		$C_{18}H_{39}O_7P$	398.47	3.00	25.6
Tris (2-ethylhexyl) phosphate	TEHP	78-42-2		$C_{24}H_{51}O_4P$	434.63	9.49	3.162
2-Ethylhexyl diphenyl phosphate	EHDPP	1241-94-7		$C_{20}H_{27}O_4P$	362.40	5.73	855.3

(continued on next page)

Table 1 (*continued*)

Name	Abbreviation	CAS no.	Chemical structure	Molecular formula	Molecular weight (g/mol)	$LogK_{ow}$	BCF
Triphenyl phosphate	TPHP	115-86-6		$C_{18}H_{15}O_4P$	326.28	4.59	113.3
Tris (methylphenyl) phosphate	TMPP	1330-78-5		$C_{21}H_{21}O_4P$	368.36	6.34	163.6

Note: K_{OW} = Octanol-water partition coefficient, BCF = Bioaccumulation factor.

indicated that plant-derived agro-foods might contain greater concentrations of OPEs than do other products (He et al., 2018b; Zhang et al., 2016).

Cereals, including rice, wheat, maize, potato, are important economic and nutritious staple food consumed by people in daily life. Contaminations of OPEs in cereals have been reported in Sweden, China, United States, Australia, Belgium and Saudi Arabia (Fig. 1, Table S1). TCEP, TCIPP, TDCIPP, TBOEP, TEHP, TNBP, EHDPP, TPHP and TMPP were the most predominant OPEs compounds reported in cereals. Mean concentration of ΣOPEs ranged from 8.08 to 802 ng/g dry mass (dm) and 1.52 to 26.8 ng/g wet mass (wm), respectively. Concentrations of OPEs in cereals varied among regions and items, with concentrations in rice generally greater than those in other kinds of cereals, like maize or in other sources of starch such as potatoes (He et al., 2018b; Poma et al., 2017; Poma et al., 2018; Wang and Kannan, 2018; Wang et al., 2021b; Zhang et al., 2016).

Among the three countries, China, United States and Australia, rice from China had the greatest concentrations of ΣOPEs, ranging from 8.68 to 802 ng/g dm. While the mean concentration of ΣOPEs in rice in the United States was 1.94 ng/g wm (0.60–93.1 ng/g wm), and 3.80 ng/g wm (2.227–8.784 ng/g wm) in Australia. Mean concentrations of OPEs in rice seeds, including kernel and husk, from rural areas of Dalian, China (298 ng/g dm) were greater than in rice (0.38–287 ng/g dm, mean:69.9 ng/g dm) collected from four other representative regions of China, including Hubei, Chongqing, Sichuan and Guangxi provinces (Wang et al., 2021b; Zhang et al., 2016). TCIPP (mean: 174.7 ng/g dm), TCEP (mean: 92.2 ng/g dm), TBOEP (mean: 50.3 ng/g dm) were the three predominant OPEs detected in rice from Dalian, China. Meanwhile, in rice plants, concentrations of OPEs and ΣOPEs in leaves were greater than those in roots (Wang et al., 2021b). In another study, conducted in China, rice, maize and other cereals collected near factories manufacturing OPEs or industrial areas including Hubei, Sichuan, and Chongqing provinces, exhibited greater concentrations of ΣOPEs compared to rural or agricultural areas of Guangxi province (Zhang et al., 2016). The greatest contribution to mean dietary intakes of ΣOPEs for adult males and females was from rice, accounting for approximately 60% of the total intake (Zhang et al., 2016). Since rice are important staple foods in Eastern and Southeast Asia countries, the relatively high contaminations of OPEs in rice seeds may pose potential risks to people via daily dietary intakes in these regions.

OPEs were also detected in maize, potatoes, cereals and grains with different contamination pattern. For instance, TNBP and TEHP were detectable in potato from Belgium (Poma et al., 2018), but not in potatoes from Sweden (Poma et al., 2017). Data from two important urban area of Saudi Arabia (Riyadh and Al-Jubail) indicated that, the median concentration of ΣOPEs in crops for human consumption was 45.6 ng/g, in farm crops of 13.4 ng/g, and in natural vegetation of 51.7 ng/g, TCIPP was detected to be the predominant compounds in all these matrices (Picó et al., 2021). Cereals from the United States exhibited relatively lesser concentrations of OPEs, with most median concentrations less than method detection limits (MDLs) (Wang and Kannan, 2018). Collectively, the major OPE compounds detected in cereals were TCIPP, TPHP, EHDPP, TBOEP for rice from China, grains and potatoes from Belgium, or maize from China, respectively.

Several studies have reported that OPEs were contaminated in vegetables, such as tomato, cabbage, rape, carrot, broccoli, onion and celery (Fig. 1, Table S2). Limited information on this category of samples exhibited comparable concentrations of OPEs. When compared with cereals, the mean concentration of ΣOPEs in vegetables ranged from 9.4 to 31.4 ng/g dm and 1.07–135.29 ng/g wm, respectively. Various vegetables exhibited different relative proportions of OPEs. TEHP (mean: 12.7 ng/g dm), TIBP (mean: 1.69 ng/g wm), TEP (mean: 0.69 ng/g wm) and TNBP (mean: 9.95 ng/g dm) were the major compounds in observed during four independent surveys from Tianjin (Zhang et al., 2016), Shenyang (Luo et al., 2018), Nanjing (Zhao et al., 2019), and Chongqing (He et al., 2019) of China. For vegetable from Belgium and Australia the predominant OPEs were TPHP (mean: 2.01 ng/g wm) and TCIPP (mean: 1.91 ng/g wm), respectively (He et al., 2018b; Poma et al., 2018). In Saudi Arabia, TCIPP was the most abundant type of OPE, with a relative proportion 45%, and greatest concentration (40.2 ng/g) than other compounds in natural vegetation (Picó et al., 2021). Recently, a considerably greater concentration of OPEs in cabbage from rural areas of Dalian (leaf: 2128 ng/g dm, root: 554 ng/g dm) was reported (Wang et al., 2021b), than those detected in other leafy vegetables from Shenyang or Nanjing of China (Luo et al., 2018; Zhao et al., 2019) or from Southeast Queensland State in Australia (He et al., 2018b). Seven OPEs, TCIPP, TCEP, TBOEP, TNBP, TPHP, TMPP, and EHDPP, were detected, of which the mean concentration of TCIPP (leaf: 1314.2 ng/g dm, root: 118.5 ng/g dm) were much greater than those of others. Leaf samples exhibited greater concentrations of OPEs compared with root. Other leafy vegetables, including crown daisy chrysanthemum, pakchoi cabbage, and Chinese chives, had greater concentrations of OPEs than did stems or roots (Zhang et al., 2016). This result is consistent with leafy vegetables, due to the larger waxy and cuticle surface area, being more vulnerable to deposition of organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs) (Bansal and Kim, 2015). Greater concentrations of OPEs in roots of cabbage might be due to active intervention in vegetable gardens, like fertilization and irrigation (Kong et al., 2012). Greater evaluation of risks posed by OPEs in leafy vegetables is needed.

Occurrences of common OPEs in fruits, such as apple, banana, orange, pear, peach, mandarin, strawberry and grape have been assessed in China, Sweden and Australia (Fig. 1, Table S2). Greatest concentrations of OPEs were found in fruit from supermarkets in Tianjin, China, with a mean concentration of 9.74 ng ΣOPEs/g dm (range: 2.64–27.4 ng/g dm). Chlorinated OPEs, including TDCIPP, TCIPP and TCEP were the main contributors to the total OPEs in fruits with concentrations of Σ_{Cl}OPEs of 1.03–26.2 ng/g, and TDCIPP (5.05 ng/g dm) was the major OPE detected

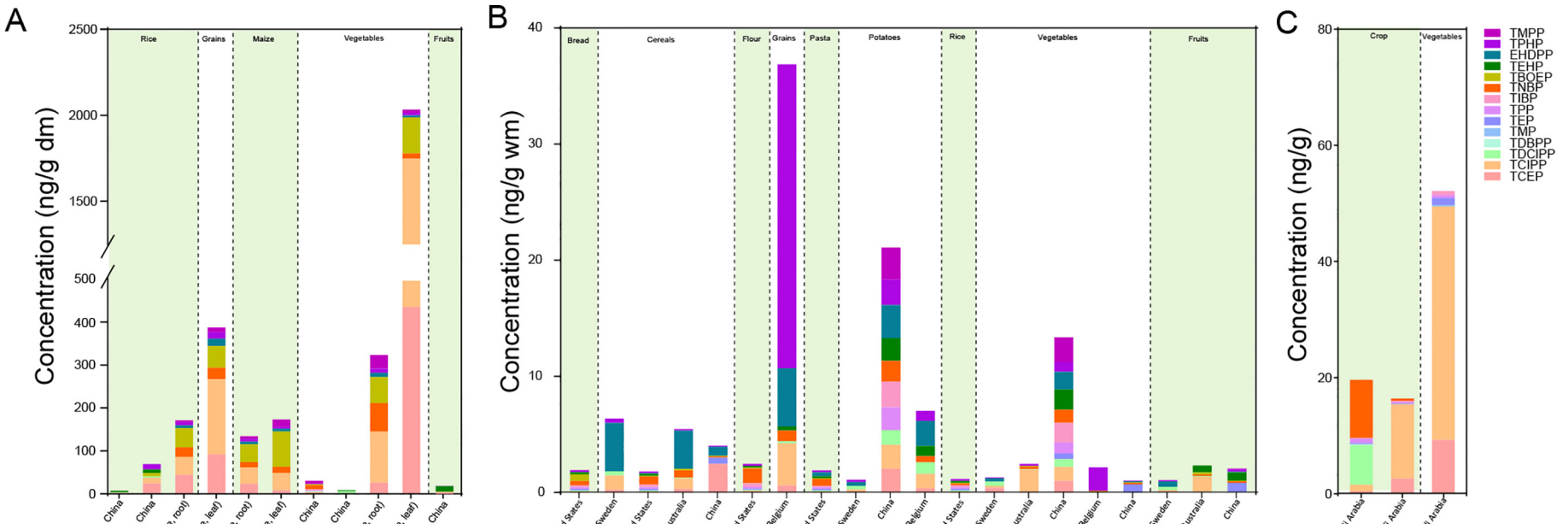


Fig. 1. Mean concentrations and compositions of OPEs in plant-derived agro-foods in different countries. The concentration units are expressed as ng/g dm (A), ng/g wm (B), or ng/g (C) depending on the specific literatures. Detailed data are presented in Table S1 and S2 of supporting information.

(Zhang et al., 2016). TCIPP (mean: 1.29 ng/g wm) and EHDPP (0.47 ng/g wm) were the predominant compounds in fruits from Southeast Queensland State, Australia (He et al., 2018b) and Uppsala region of Sweden (Poma et al., 2017), respectively. EHDPP had the greatest median concentration in most samples of cereals, vegetables and meats from Sweden, which was possibly due to uses of EHDPP in food packaging materials (Poma et al., 2017).

2.2. Animal-derived agro-foods

As the main sources of human protein supply, large amounts of animal-derived products, including meat, milk, eggs and their derivatives are produced and consumed around the world. To date, OPEs have frequently been detected in various animal-derived foods in various countries. Different bioaccumulative properties and metabolic patterns of OPE compounds, as well as the contamination of ambient agri-environments and agricultural inputs can result in distributions of OPEs in foods derived from animal tissues.

OPEs have been investigated in various meats from cattle, chicken, turkey, pig, lamb and goat, in various countries including Belgium, Sweden, United States, China and Australia (Fig. 2 and Table S3). The median concentration of ΣOPEs reported in the United States was 6.76 (range: 1.43–46.1) ng/g wm, which was significantly greater than concentrations of OPEs in dairy or cereal products. TBOEP accounted for 55.2–90.3% of concentrations of ΣOPEs in meat, with a median concentration of 5.72 ng/g wm (Wang and Kannan, 2018). In a similar study conducted in the USA, comparable concentrations of ΣOPEs (median: 6.2 ng/g wm) were reported (Han et al., 2019). TCIPP was the constituent most frequently detected (28% frequency of detection) and occurred at the greatest concentrations (1.3–15.8 ng/g wm). Similar concentrations of OPEs have been reported for China (Ding et al., 2018; Zhang et al., 2016) in meats including beef, chicken and pork, with concentrations of 4.13 ng ΣOPEs/g dm and 3.95 ng ΣOPEs/g wm, respectively. As with cereals and vegetables, the greatest mean concentrations (0.64 ng EHDPP/g wm) was observed in meat from Sweden, which was possibly the result of contamination from food packaging materials (Poma et al., 2017). TCIPP and TNBP were the most abundant compounds in meat from Australia, with mean concentrations of 0.36 and 0.27 ng/g wm, respectively (He et al., 2018b). Recently, relatively great concentrations of OPEs were observed in domestic livestock, including pork, beef and chicken, from an agricultural area of Chongqing, China (He et al., 2019). Greatest concentrations of ΣOPEs were found in chicken (676 ng/g lipid mass (lm)), followed by beef (545 ng/g lm) and pork (535 ng/g lm), the major components were TNBP in chicken (mean: 243 ng/g lm) and beef (mean: 168 ng/g lm), while TMPP in pork (mean: 115 ng/g lm). Concentrations of OPEs were inconsistent with the trends in amounts of lipid, which indicated a lipid-independent accumulation of OPEs in biota.

There is growing concern over the presence of OPEs in dairy products. Concentrations of OPEs in milk, yogurt, butter, cheese, and milk powder have been reported for Belgium, Sweden, United States, China and Australia (Fig. 2 and Table S4). Concentrations of ΣOPEs were ranged from 0.60 to 71.4 ng/g wm. Concentrations of EHDPP in dairy samples from Sweden (Poma et al., 2017) and samples of desserts from Belgium (Poma et al., 2018) were observed to be increasing. TPHP was the predominant OPE in milk and cheese from Belgium (Poma et al., 2018). TNBP (0.95 ng/g wm) and TCIPP (0.89 ng/g wm) were the major OPEs detected in Australia (He et al., 2018b). Similar results were observed in the United States, where TNBP and TCIPP were the dominant OPE compounds found in dairy products (TNBP: 20.3–53.6%, median: 0.21 ng/g wm; TCIPP: 5.23–42.5%, median: 0.23 ng/g wm) (Wang and Kannan, 2018). Chlorinated OPEs were the main contributors to the total OPEs in dairy products with concentrations of Σ_{Cl}OPEs of 18.7–2312 ng/L (Zhang et al., 2016). The greatest concentrations of ΣOPEs in dairy were 3065 ng/L in yogurt. TDCIPP was the predominant OPE with a mean concentration of 296 ng/L. In China, TCEP (0.32 ng/mL) exhibited the greatest median concentration in cow milk from the city of Shouguang in Shandong province, which is an area where flame retardants are produced (Chen et al., 2021a). It has been concluded that solid dairy products exhibited greater concentrations of OPEs compared to fluid dairy products, and emphasized that human exposure to OPEs from dairy products cannot be ignored (Li et al., 2019a).

The greatest mean concentrations of ΣOPEs (6.35 ng/g wm) in eggs were observed in a local supermarket in China (Zhao et al., 2019) (Fig. 2 and Table S5). The primary OPE was EHDPP which was detected in 64.0% of samples with a mean concentration of 4.06 ng/g wm. In China, concentrations of OPE exhibited relatively greater median concentrations with a range of 1.62 to 2.59 ng/g wm in free-range chicken eggs collected from e-waste recycling area in Qingyuan (Guangdong province). TPHP, EHDPP, TCEP, TDCIPP were detected in eggs, but only TCEP were detected in 100% of the samples. Chlorinated OPEs, like TCEP, TCIPP and TDCIPP, were observed in eggs greater than 50% of samples, probably due to the fact that chlorinated OPEs are more persistent or bioaccumulative in eggs than non-chlorinated OPEs (Zheng et al., 2016). Similar findings have been reported for TCIPP (0.43 ng/g wm) and TCEP (0.19 ng/g wm), both of which had greater concentrations in eggs from Australia (He et al., 2018b). There was no significant difference between concentrations of ΣOPEs in eggs from e-waste sites and the control site, e-waste recycling activities seem to have little influence on concentrations of OPE in eggs

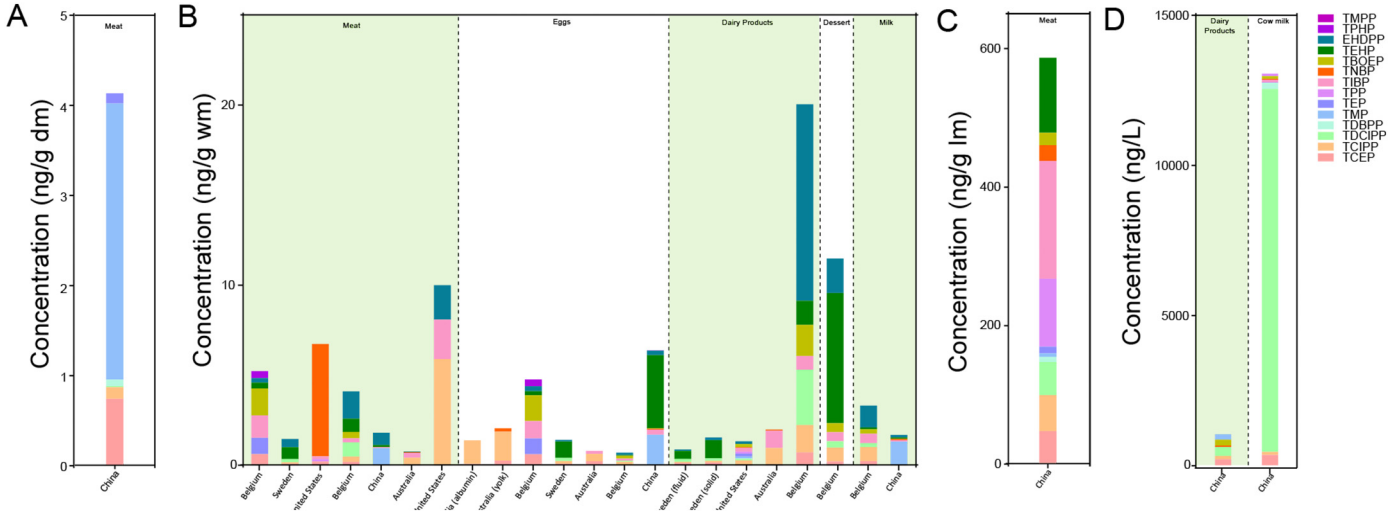


Fig. 2. Mean concentrations and compositions of OPEs in animal-derived agro-foods in different countries. The concentration units are expressed as ng/g dm (A), ng/g wm (B), ng/g (C), or ng/L (D) depending on the specific literatures. Detailed data are presented in Table S3-S5 of supporting information.

(Zheng et al., 2016). EHDPP was the major component (0.91 ng/g wm) in fresh eggs from Sweden (Poma et al., 2017). While in Belgium, TPHP (0.27 ng/g wm) was the major OPE in eggs collected in 2013 (Xu et al., 2015), TCIPP (0.15 ng/g wm) and TEHP (0.15 ng/g wm) were the major OPEs in eggs collected between 2015 and 2016 (Poma et al., 2018). In Australia, no significant spatial differences were observed for concentrations of OPEs in eggs from various locations. Also, there was no difference in concentrations of OPEs between free range and caged eggs. However, frequencies of detection varied among OPEs in yolk and albumin. Concentrations of TCEP and TBOEP were greater in yolk, while concentrations of TCIPP, TDCIPP and TPHP were greater in albumin. These results indicated that logK_{ow} might not be an effective determinant for the distribution of OPEs between fractions of eggs. Concentrations of ΣOPEs were comparable in egg yolk (2.2–6.8 ng/w wm) and albumin (2.3–6.6 ng/w wm), and TCIPP (1.8 ng/g wm, median) was the predominant OPE in either yolk or in albumin (Li et al., 2020b).

Some nontraditional animal-derived agro-foods, like insect-based foods have been reported to contaminated by OPEs (Fig. 3 and Table S7). OPEs contamination was reported in Japan, which ranged from 2.3 to 240 ng/g wm in edible insects and from 2.4 to 106 ng/g wm in insect-based food. TCIPP was the most detected (detection frequency: 81%) OPE compound in both edible insects and insect-based food, accounting for 56% and 37% of the total load, respectively, which was followed by EHDPHP (12%) and TPHP (9%) in edible insects, and TDCIPP (30%) and TPHP (8%) in insect-based food (Poma et al., 2021). Similar levels of OPEs in edible insects were also reported from Europe and Asia, ranged from 2.3 to 185 ng/g wm (Poma et al., 2019).

2.3. Aquatic-derived foods

OPEs are continuously discharged into aquatic environments from various sources, therefore aquatic foods, like fish, shrimp, mussels, are expected to be exposed to and accumulate OPEs. Concentrations of OPEs in aquatic foods have been reported from major water bodies, including both lakes and rivers as well as marine environments among various regions, but primarily from Belgium, Sweden, Spain, Italy, Australia, United States and China (Fig. 4 and Table S6).

Concentrations of OPEs in freshwater fishes have been reported for perch, carps, yellow eels, barbell and redfin culter (*Cultrichthys erythropterus*) from Sweden (Sundkvist et al., 2010), Belgium

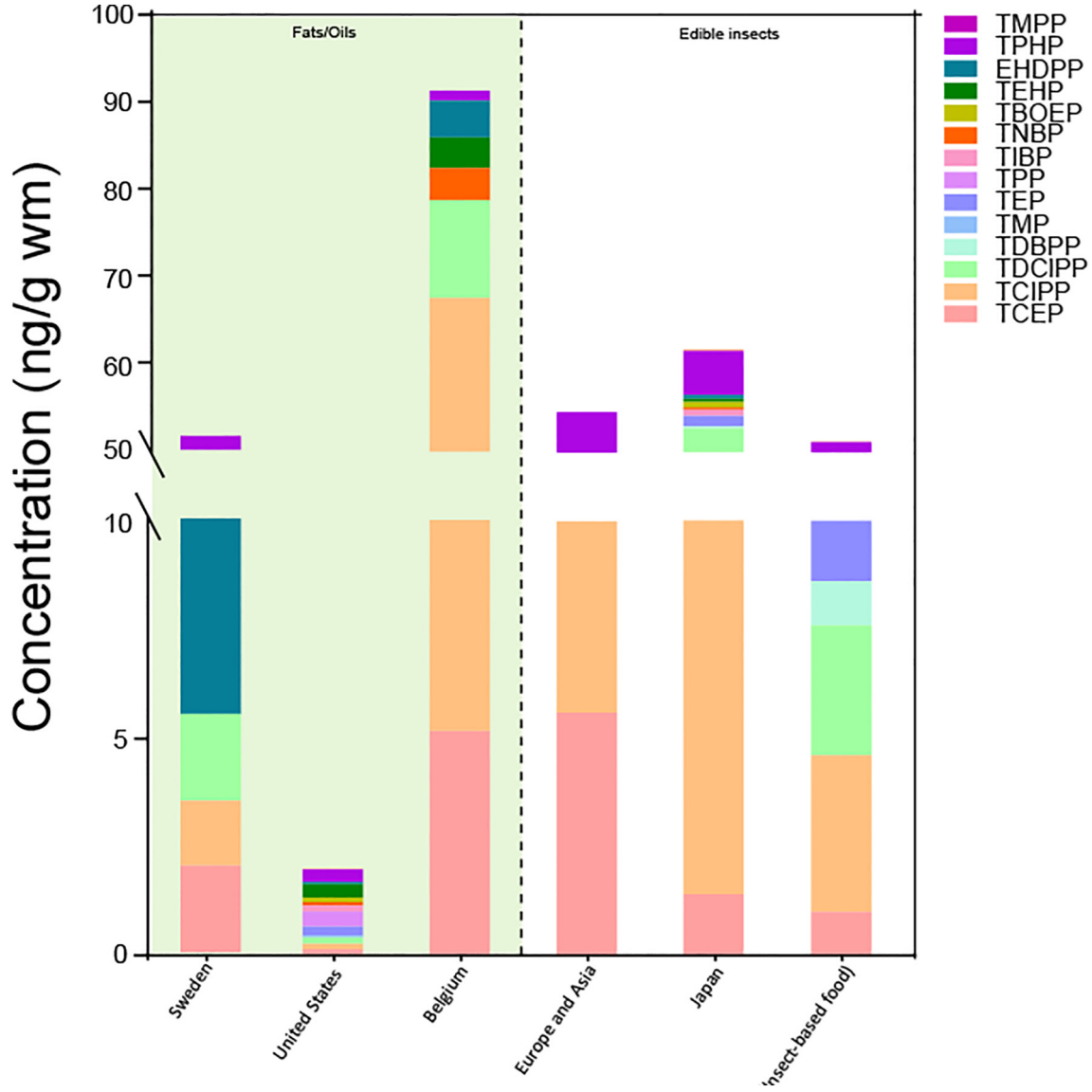


Fig. 3. Mean concentrations and compositions of OPEs in fats, oils, and edible insects in different countries. The concentration units are expressed as ng/g wm. Detailed data are presented in Table S7 of supporting information.

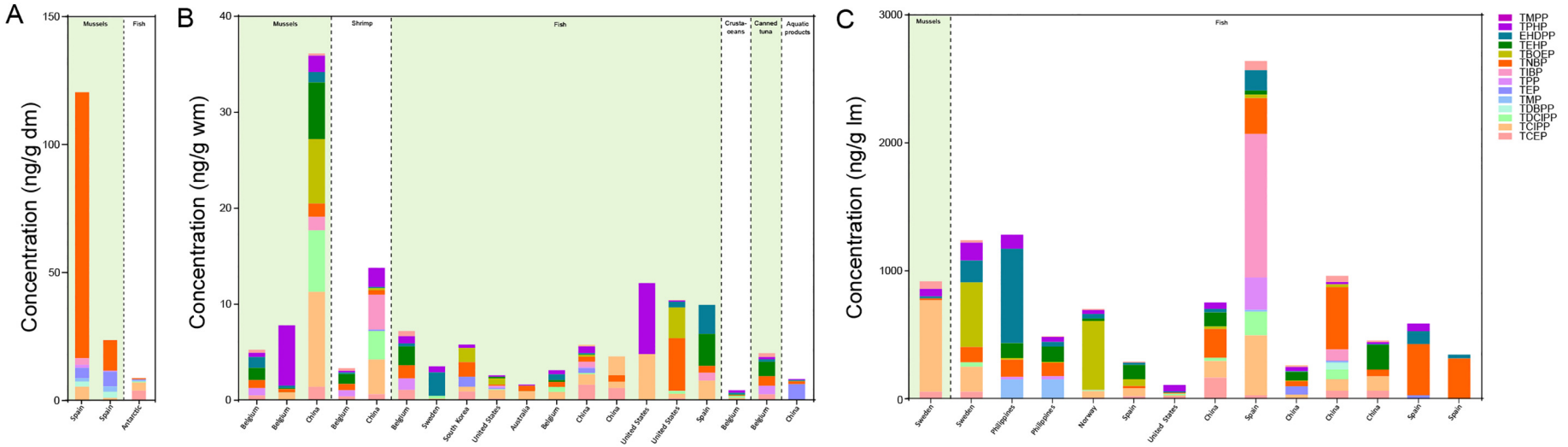


Fig. 4. Mean concentrations and compositions of OPEs in aquatic-derived foods in different countries. The concentration units are expressed as ng/g dm (A), ng/g wm (B), or ng/g lm(C) depending on the specific literatures. Detailed data are presented in Table S6 of supporting information.

(Malarvannan et al., 2015), Spain (Santin et al., 2016), United States (Zheng et al., 2020), Canada (McGoldrick et al., 2014), South Korea (Choo et al., 2018) and China (He et al., 2019; Liu et al., 2018; Liu et al., 2019b; Ma et al., 2013; Zhao et al., 2018; Zhao et al., 2019). The pattern of relative proportions of OPEs in fishes could reflect local contamination. Relatively lesser concentrations of OPE were observed in sentinel fish samples (stickleback) in Alaska, with the mean concentrations of 5.95 ng/g wm, and TNBP was the dominant OPE (mean: 5.50 ng/g wm) with a detection frequency of 88% (Zheng et al., 2020). TCEP and TBOEP were the dominant and most frequently detected compounds in freshwater fishes from Canada, but concentrations were relatively small (<10 ng/g wm) (Chu and Letcher, 2015; Guo et al., 2017; McGoldrick et al., 2014). In Sweden, fishes collected downstream of effluents from sewage treatment plants (STPs) had distinct differences in profiles of relative proportions of OPEs compared to fishes from other locations (Sundkvist et al., 2010). Fishes collected in a lake upstream of the STPs had a similar OPE profile to that of fishes from background locations. TBEP (range: 270–1000 ng/g lm) and TCIPP (range: 36–140 ng/g lm) were the major compounds in fish samples. TBEP was also detected in perch from a stream receiving surface water from an airport, which indicated a possible source of contamination. Larger perch had slightly greater concentrations of OPEs than did smaller perch, which indicated potent bioaccumulation (Sundkvist et al., 2010). OPEs were detected in all pooled yellow eels from freshwater bodies in the more populated and industrialized region of Flanders in Belgium, in decreasing order of TCIPP > TPHP > EHDPP > TBOEP > TCEP > TDCIPP. The median concentration of ΣOPEs for all locations was 44 ng/g lm (7.0–330 ng/g lm) (Malarvannan et al., 2015). Total concentrations of OPEs in fishes collected from rivers in Spain ranged from not quantified (nq) to 2423 ng/g lm, with a median value of 677 ng/g lm (Santin et al., 2016). OPEs were detected in all fishes collected from three European river basins, including Evrotas (Greece) (mean: 40.1 ng/g lm), Adige (Italy) (mean: 286 ng/g lm), and Sava (Slovenia, Croatia, Bosnia and Herzegovian and Serbia) (mean: 84 ng/g lm). In contrast, although OPE values were higher in sediments than halogenated flame retardants (HFRs), such as polybromodiphenyl ethers (PBDEs), but similar or even lower concentrations were observed for OPEs in fishes (Giulivo et al., 2017). Similar results were obtained for fishes collected from the Pearl River Delta of China (Zhang et al., 2018). When concentrations of OPEs were surveyed in China in the Pearl River Delta (Liu et al., 2018; Liu et al., 2019b), Tai Lake, Jiangsu province (Zhao et al., 2018), Beijing (Hou et al., 2017), Nanjing (Zhao et al., 2019) and Chongqing (He et al., 2019), the mean total concentrations of OPEs ranged from 141.7 to 1973.17 ng/g lm. Collectively, TCEP, TNBP, TEHP, TPHP, TEP were the predominant compounds found in fishes from China. Concentrations of OPE varied among regions. Chlorinated OPEs including TCEP, TCIPP and TDCIPP, and the aryl-OPE, TPHP, were the predominant forms of OPEs in fishes from Tai Lake (Zhao et al., 2018). Reasons for could be either consistency in spatial distribution of OPEs between fishes and sediment of Tai Lake, or to their relatively great bioaccumulation potentials, particularly for TPHP.

The marine environment acts as an ultimate sink for diverse organic pollutants, therefore, marine fishes and other biota can be exposed to OPEs. In fact, OPEs have been detected in salmon (*Salmo salar*), tuna, mullet, mojarra, coral grouper, bogue (*Boops boops*) around the world, including the Philippines (Kim et al., 2011), United States (Han et al., 2019; Wang and Kannan, 2018), Australia (He et al., 2018b), Belgium (Poma et al., 2018; Xu et al., 2015), Spain (Castro et al., 2020a; Henríquez-Hernández et al., 2017), Sweden (Poma et al., 2017), Norway (Hallanger et al., 2015). Concentrations of OPEs vary among types of fishes and regions. For example, OPEs were not detected in salmon or cod collected from markets in Sweden (Campone et al., 2010). But results from another Swedish research indicated relatively great concentrations of ΣOPEs in eelpout (*Zoarces viviparus*), with a mean of 15,000 ng/g lm, but concentrations of ΣOPEs in salmon (mean: 34 ng/g lm) and herring (*Clupeidae*) (mean: 108 ng/g lm) from this area are less (Sundkvist et al., 2010). Concentrations of OPE in bogues (*Boops boops*) from the North Western Mediterranean Sea, ranged from <LOD to 2566 ng/g lm, with TNBP and EHDPP being the major compounds. The greater values reported were for samples collected closer to the populated area (Garcia-Garin et al., 2020). Concentrations of OPEs were comparable in samples from the United States (8.7 ng/g wm, 7.11 ng/g wm) and Australia (3.32 ng/g wm) (Han et al., 2019; He et al., 2018b; Wang and Kannan, 2018). Even in same sea area, fishes could exhibit great differences. In Manila Bay, the Philippines, yellow striped goatfish (*Upeneus vittatus*) had the greatest concentration of ΣOPEs (mean: 1900 ng/g lm), followed by silver sillago (*Sillago sihama*) (mean: 1800 ng/g lm) and tripletail wrasse (*Cheilinus trilobatus*) (mean: 1500 ng/g lm). Mean concentration of ΣOPEs in fourlined terapon (*Pelates quadrilineatus*) was 100 ng/g lm (Kim et al., 2011). Demersal fishes accumulated more OPEs than did pelagic species. Meanwhile, the pattern of OPEs accumulated was different even in the same species of fish. For example, except for EHDPP, TEP and TPHP, the other nine OPEs including TEHP and TPEP, were detected in juvenile blue tail mullet (*Valamugil buchanani*) than the adult fishes (Kim et al., 2011). Growing patterns also influence concentration of OPEs in fishes. In general, aquaculture-associated bogues (*Boops boops*) contained lesser concentrations of OPEs than did wild-caught fish, especially when sums were considered (Henríquez-Hernández et al., 2017). Recently, occurrences of OPEs in fishes from the Antarctic ecosystem were reported to have a mean concentration of ΣOPEs of 2.21 ng/g wm (173 ng/g lm), which was greater than those from the Great Lake of North America, and frequency of detection of TBOEP was greater than 90% (Fu et al., 2020).

Occurrences of OPEs have also been reported for other aquatic-derived foods, like mussels and shrimp in The Philippines, China, Spain, Sweden, Belgium and some other European countries. Six OPEs, including TCEP, TCIPP, TNBP, EHDPP, TPHP and TMPP, were detected in blue mussels (*Mytilus edulis*) from Sweden, of which TCIPP was the predominant compound, with mean concentration of 715 ng/g, lm (range: 130–1300 ng/g, lm) (Sundkvist et al., 2010). Mussels from Belgium had lesser concentrations of OPEs, with the exception of TPHP, which was greatest in mussel (mean: 6.29 ng/g wm) (Poma et al., 2018; Xu et al., 2015). Similarly, TPHP occurred at the greatest concentration in mussel samples from the coast of Galicia of Spain, and ranged from 11 to 291 ng/g dm (Castro et al., 2020b). TCIPP, TDCIPP, TBOEP, TMPP, EHDPP and TEHP were observed in freshwater mussels from Taihu Lake in China, of which TCIPP was presented at the greatest concentration (9.9 ng/g wm) (Zhao et al., 2018). OPEs were also detected in White shrimp (*Exopalaemon modestus*) and the freshwater shrimp (*Macrobranchium nipponense*), where TCIPP, TDCIPP, TIBP were the major components found (Zhao et al., 2018).

2.4. Other agro-foods

Fats and oils from Sweden, Belgium and the United States have been reported to be contaminated with detectable quantities of OPEs (Poma et al., 2017; Poma et al., 2018; Wang and Kannan, 2018) (Fig. 3, Table S7). Limited information suggests greater concentrations of ΣOPEs in fats and oils in Belgium compared to Sweden and the United States. TCIPP (mean: 38.7 ng/g wm, range: 30.99–109.87 ng/g wm) and TDCIPP (mean: 20.18 ng/g wm, range: 16.09–57.04 ng/g wm) were the predominant compounds found in fats collected in Belgium (Poma et al., 2018). The mean concentration of TPHP (4.74 ng/g wm) was greater in fats and oils from Sweden, except for EHDPP (5.08 ng/g wm) which was the most abundant compound in these samples (Poma et al., 2017). Meanwhile, some regional agro-foods like toufu have also been reported to contain OPEs, with the concentration of 2.5 ± 0.4 ng/g fresh weight (Ding et al., 2018).

3. Sources and transfer routes of OPEs in agriculture

Because of the worldwide presence of OPEs in agro-foods, questions related to their sources, entry and transfer routes of these compounds have arisen. Information on sources of contamination and transfer routes remains scant. Collectively, OPEs can contaminate agro-foods via the following three routes (as shown in Fig. 5): (1) crops, such as rice, vegetables and

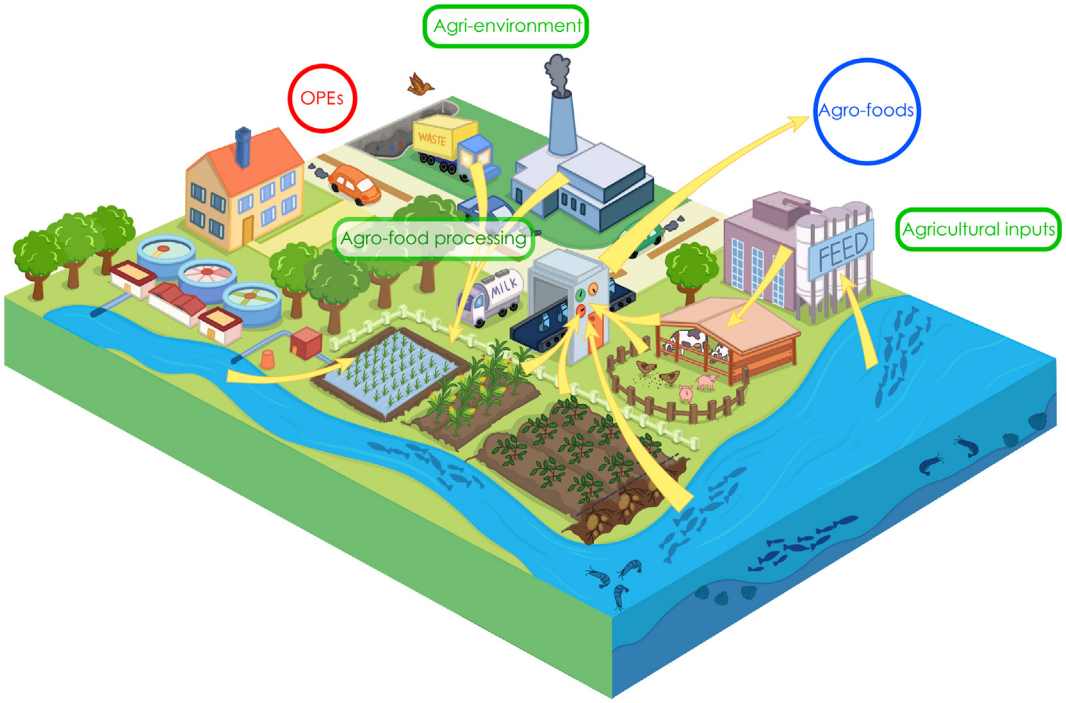


Fig. 5. Schematic illustration of the sources and transmission of OPEs in agriculture.

fruits, livestock, such as pigs, cattle, chickens and aquatic products, including fishes, shrimp and mussels that can absorb OPEs via the contaminated agri-environment, such as farmland soil, water, air, and sediments, resulting in OPEs being subsequently accumulate in the final agro-products; (2) livestock can ingest OPEs from contaminated agricultural inputs like feedstuffs, which finally present in animal-derived agro-foods, such as meat, milk and eggs; and (3) agro-foods can be contaminated by OPEs during postharvest and industrial processing, such as storage and package, due to their presence in materials used in these processes (Ding et al., 2018; Li et al., 2019a; Poma et al., 2018; Zhang et al., 2020a; Zhang et al., 2016).

3.1. Agri-environment

Agri-environments, including soil, water, air, and sediments are the most discussed potential source of OPEs in agro-foods. OPEs can escape into the environment during all stages of the OPE-containing product life cycle from manufacturing to use, disposal and recycling (Zhang et al., 2021a). Therefore, they are ubiquitous in various environmental matrices worldwide.

3.1.1. Plant

An increasing number of studies have confirmed that OPEs can be absorbed and accumulated into plants from soil, water, and air (Hyland et al., 2015; Wang et al., 2021b; Zhang et al., 2016). Root uptake from soil or water, and foliar uptake from air or from deposited particles are the two main routes through which OPEs enter plants. Accumulation of OPEs into roots of plants has been observed during field studies and pot and hydroponic experiments. Concentrations of OPEs in plant rhizosphere soils were less than those in bulk soils, which demonstrated depletion by uptake or enhanced degradation by plants (Wang et al., 2021b). Profiles of distributions and concentrations of OPEs in roots and above-ground tissues of wheat plants (*Triticum aestivum* L.) were similar to those of the corresponding farmland soils surrounding a plastic waste treatment area, which confirmed accumulation of by roots form soils (Wan et al., 2016). Uptake and translocation of OPEs by wheat (*Triticum aestivum* L.) were studied under hydroponic conditions (Gong et al., 2020; Wan et al., 2017; Wang et al., 2019a). Results indicated that OPEs with relatively greater hydrophobicity were more likely to be accumulated by roots, but OPEs with lesser hydrophobicity were more prone to be translocated from roots to foliage of wheat plants. Similar phenomena were observed in mung bean (*Vigna radiata* L. Wilczek), carrot (*Daucus carota* L. *var. sativa* Hoffm.), lettuce (*Lactuca sativa* L.), tomato (*Lycopersicon esculentum* Mill.), and soybean (*Glycine* max L.) (Hu et al., 2021; Liu et al., 2019a). Point sources of OPEs could have widespread effects on surrounding environments, including cropland soils, water and sediments, which resulted in the accumulation of TCEP and TCIPP in tree barks surrounding the manufacturing facilities (Ren et al., 2019). TCEP and TCIPP have been reported to be accumulated into lettuce (*Lactuca sativa*) and strawberry (*Fragaria ananassa*) from irrigation with contaminated water (Hyland et al., 2015). The fact that paddy fields had relatively greater concentrations of OPEs than did other farmlands, such as orchards, maize fields and vegetable gardens, suggested that frequent irrigation might promote input of OPEs to soils, and result in increased health risk associated with consuming rice (Wang et al., 2021b). Additionally, because manure and sewage sludge are used as fertilizer or soil conditioner on agricultural soils while effluent from wastewater treatment plants (WWTPs) is used for irrigation, contaminants can be transported to soil, risks of introducing OPEs into crops is therefore increased (Eggen et al., 2013). Meanwhile, the uptake of OPEs from atmosphere or volatilized from soil by foliar might also be an important pathway for chemicals with $\log K_{oa} > 6$ (Cousins and Mackay, 2001). Significant correlations of concentrations of ΣOPE concentrations between paired outdoor air and pine needle samples was observed, and the spatial distribution of OPEs in pine needles was similar to that found in air (Li et al., 2019c). It was suggested that indoor ornamental plants, such as scindapsus, can be effectively used as bio-indicators of occurrence of OPEs in the indoor environment due to the relatively long-term air-foliage partitioning (Wang et al., 2021a). In a typical research focusing on the behavior and fate of OPEs in subtropical paddy field environment of China, air-soil exchange, atmospheric deposition (rainwater plus dustfall), and irrigation water contributed to the major input fluxes of OPEs in the paddy fields. OPEs were then accumulated in rice plants but varied largely at different growing stages. In contrast, the fractions of hydrophilic OPEs, such as TCEP and TCIPP, decreased in the leaves/grains compared with the roots/stems, which was different with the model in wheats, indicated

that rice plants might have different mechanisms of uptake and or accumulation (Zhang et al., 2020b).

3.1.2. Animals

Although the ambient environment could affect concentrations and composition profiles of OPEs in biota, limited information is available on sources and transmission routes of accumulation of OPEs by domestic livestock in agri-environments. Influences of ambient environments, including air and river water, in an agricultural area on the distribution of OPEs in animals were studied (He et al., 2019). Results revealed that OPEs profiles in chickens, cattle were correlated with concentrations in outdoor air, whereas only concentrations of OPEs in cattle were strongly correlated with those in river water. Specifically, TBOEP exhibited greater biota-air accumulation factors (BAAFs), but the least biota-water accumulation factors (BWAFs) for biotic samples, which suggested that TBOEP was prone to partition from air to biota. While TMPP exhibited both great BWAFs and BAAFs values, which demonstrated that TMPP might be accumulated in biological samples both from air and river water. Bio-resource materials, including recycled waste wood, dried paper sludge, and paper sludge ash, were recycled as alternative bedding materials for livestock. Relatively great concentrations of TCIPP, TDCIPP and TCEP in these materials also potentially transferred contaminants into animal-derived foods via animal breeding (Rigby et al., 2021). OPEs contamination of edible insects might have occurred during rearing, such as the rearing substrate, as better correlations were observed between chlorinated-OPEs and their biotransformation products (Poma et al., 2019).

3.1.3. Aquatic biota

Aquatic environments are major sinks for OPEs. Process by which OPEs are transported through the atmosphere, enter water and are deposited in sediments is known to be an important source of OPEs for many aquatic organisms and sediments (Hale et al., 2003; Iqbal et al., 2017). In addition, domestic sewage and wastewaters from factories, construction sites and pavements also contribute for the accumulation of OPEs in aquatic environment (Henríquez-Hernández et al., 2017).

Bioaccumulation and biomagnification in marine and freshwater food webs are the predominant routes of transmission of OPEs and accumulation by fishes. Bioaccumulation factors (BAFs), are ratios of concentrations in biota divided by concentrations in water, are the most common factors to describe potential for bioaccumulation. BAFs of OPEs vary greatly, ranging from 0.425 for TCEP to 2534 for TMPP (Pantelaki and Voutsa, 2020). The biota sediment accumulation factor (BSAF), which defined as the ratio of the concentration of OPEs in species based on lm to the concentration of OPEs in sediment normalized to organic carbon content, is also used to assess the bioaccumulation potential of OPEs. Greater values for BSAF for TCEP and TNBP of 4.80–11 and 20.5–48.7, respectively, have been reported for crucian carp from the Nakdong River, South Korea (Choo et al., 2018). BSAF values for TNBP, TCEP and TMPP measured in fish, including trout, bullhead, grayling, chub and barbell, from the Sava and Adige river basins, Europe, were always less than 1.0 (Giulivo et al., 2017). Small values for BSAF of 0.3–0.7 have also been reported for benthic fishes (Bekele et al., 2019), as well as for a freshwater food web including river prawn, crucian carp, mud carp, catfish and snakehead (Liu et al., 2019c). In a recent study, OPE concentrations between sediment and wild fish collected from Taihu Lake, China, were analyzed. Log BCF of OPEs increased with logK_{ow} from 0.87 to 7.09 increasing, but further increasing the logK_{ow} decreased log BCF, indicated the bioaccumulation potential of OPEs with logK_{ow} < 7.09 is likely driven by its low hydrophobicity, while the high hydrophobicity of OPEs with logK_{ow} > 7.09 may not play an important role in the distribution of the compounds (Li et al., 2022). EHDPP biomagnified in the fish food web of Tai Lake (China) with a TMF (trophic magnification factor) of 3.61 as results of its relatively great hydrophobicity and lesser potential for metabolism in fish at the top of the trophic pyramid (Wang et al., 2019b). In a tropical aquatic food web, TMFs of OPEs, including TCEP, TCIPP, TDCIPP, TIBP, TNBP, TPHP, and TBOEP, were generally less than TMFs in temperate and frigid aquatic food webs (Ding et al., 2020). Due to the limited number of data sets, this trend still needs to be further elucidated. Demersal fishes have been reported to accumulate greater amounts of OPEs than do pelagic species. Lipophilic properties of OPEs make them bind well to particles and thus have a tendency to accumulate in sediments (Bekele et al., 2019; Kim et al., 2011; Sundkvist et al., 2010). Furthermore, emission from fishing boats, riverine discharge, extensive local aquaculture activities, and effluents from the nearby industrial and economic zones of wastewater treatment plants increases the opportunities of bioaccumulation and biomagnification of OPEs in biota (Bekele et al., 2019). A study conducted in Sweden showed that TBOEP detected in perch originated from a stream receiving surface water from an airport (Sundkvist et al., 2010).

Absorption through gills can be mechanism by which aquatic animals accumulate OPEs dissolved in ambient water. Significant correlations were found between fish and ambient environmental samples, suggested that water and air were the primary pathways for OPEs accumulation in fish (He et al., 2019). Results from investigations of distributions of OPEs in fish suggested that OPEs might primarily absorbed from water through the gills and epithelial tissue rather than through food web biomagnification (Kim et al., 2011; Malarvannan et al., 2015; Sundkvist et al., 2010). However, information on accumulation of OPEs by aquatic organisms through gill and epithelial tissue remains limited.

3.2. Agricultural inputs

Data on exposure to OPEs from agricultural inputs are limited. As the major food for livestock, animal feed plays important role in food safety, especially for those contaminants that might undergo food chain bioaccumulation. Residual OPEs in raw ingredients and packaging material, such as polypropylene woven bags, might be sources of the OPEs in feeds. Although OPEs tend to degrade in biota, bioaccumulation of TCIPP, TCEP and TBOEP in ecosystems have been demonstrated (Zhao et al., 2020b). Therefore, since OPEs could transfer from feed to food via animal breeding, risks posed by OPEs in feeds cannot be ignored. Results from our group's research indicated that detectable concentrations of OPEs, ranging from 12.6 to 301 ng/g dm, could be found in all animal-derived meat meal, feather meal, and blood meal, as well as plant-derived animal protein supplement feeds, and plant-derived animal protein supplement feed would be an important source of OPE exposure for livestock due to its widespread consumption (Zhao et al., 2020b). Fishmeal, which consumed one third of global annual fish production, is widely applied in the global animal farming industry, especially for agriculture (Li et al., 2018). OPEs, such as TCEP, TCIPP, TDCIPP and TBOEP, as well as their transformation products, including organophosphate diesters, were also detected in all fishmeal samples collected across the world, with a comparable average concentration (59.3 ± 92.2 ng/g dm for OPEs and 49.6 ± 27.5 ng/g dm for their metabolites) (Li et al., 2020a). Relatively great translocation of TCIPP into leaves of meadow fescue, a livestock forage species, which suggests potential accumulation of OPEs by herbivorous animals (Eggen et al., 2013). As no significant differences between free range and caged eggs were found in a survey from Australia, chicken feed, instead of soil within the farm, was suggested to be a key source for the exposure of the hens to OPEs (Li et al., 2020b). When occurrences of OPEs in mariculture farms of the Beibu Gulf, located in the northwest of the South China Sea, were investigated, concentrations of ΣOPEs in feeds (mean: 46.0 ng/g dm, range: 21.7–84.5 ng/g dm) was similar to concentrations in shrimp, crab and oyster (mean: 55.5 ng/g dm, range: 21.3–138 ng/g dm), and amounts of ΣOPEs released from feeds to the culture ponds was estimated to be 49 mg/m^2 per three month period. Chlorinated OPEs contributed the greatest proportion (97.8%) of the OPEs in feeds. Therefore, feed was recognized as an important source of OPEs in the mariculture farms (Zhang et al., 2020a).

Bioresource materials used in agriculture for soil improvement were a possible source of OPEs in plant-derived agro-foods. Biosolids (treated sewage sludge), meat and bonemeal ash, poultry litter ash, paper sludge ash and compost-like-output are commonly recycled as soil amendments. OPEs were detected in these bioresource materials. TCIPP was the

dominant compound in biosolids and compost-like-output, TCEP was found in paper sludge ash and poultry litter ash (Rigby et al., 2021). The occurrence of such a wide range of flame retarding compounds in these soil improvements emphasized the risk to plant-derived agro-foods.

Plastic mulch film is increasingly used in modern agriculture to improve ground temperature, water retention, and grain yield, results in high resides of mulch film in farmland soils. Organophosphite antioxidants (OPAs), which are commonly used as auxiliary antioxidants in the manufacture of plastic polymers, and OPEs were detected, with the median concentrations of 2.66 ng/g (Σ_6OPAs) and 100 ng/g (Σ_5OPEs) in the film-mulching soil and 1.16 ng/g (Σ_6OPAs) and 47.9 ng/g (Σ_5OPEs) in the nonfilm-mulching soil, respectively (Gong et al., 2021). Although the information on the migration risk of typical OPA remains scant. As OPAs could be oxidized to the corresponding organophosphate esters under biotic and abiotic conditions during production, processing or in the soil environment. Consequently, OPAs could be an important source of OPEs to farmland oils.

3.3. Agro-foods processing

Agro-food packaging and industrial processing, including processing equipment and tools, could be potential sources of OPEs in agro-foods. In an investigation of OPEs in food from Australia, results exhibited lesser concentrations of OPEs in samples with inedible outer parts, such as banana, orange and egg, than those contacting directly with plastic packages or display trays, though these differences were not significant. The results suggested that package materials or other use of the food might be a potential source of OPEs in these foods (He et al., 2018b). In Belgium and China, concentrations of OPEs, especially EHDPP, which has been approved by the U.S. Food and Drug Administration (US-FDA) for packaging application, were greater in processed food, such as cheese, oil, meat, milk and candy, than in non-processed food, which suggested contamination during industrial processing and manipulation, including packaging, canning and drying, could be a key source for the contamination of these foods (Poma et al., 2018; Zhao et al., 2019). Similar results were reported in the United States, food packaging was suggested as a source of TCPP contamination in meats and fish (Han et al., 2019). The similar OPE profiles in food packaging materials and in food samples suggested that food packaging could be a source of TBOEP and TCIPP detected in foods. But ΣOPEs in food packaged in plastic and paper materials were statistically insignificant (Wang and Kannan, 2018). But, alternatively, Zhang et al. (2016) compared the distribution of total concentrations of OPEs in food with different packaging materials (cans, paper, and plastics), and the results indicated that the greatest mean concentration of total OPEs was found in non-packaged food. OPE concentrations in cereal products (such as flour, bread, et al.) and fast/oils which undergo considerable industrial processing were low, indicated that food processing did not introduce sources of OPEs (Wang and Kannan, 2018). The presence of OPEs in insect samples likely resulted from industrial processing. This was clearly the case for alkyl-OPEs, where the divergent load from their biotransformation products suggested that contamination occurred during insect seasoning and industrial processing after harvesting (Poma et al., 2019). Besides seasoning, OPEs contamination of edible insects may have also occurred as a result of cross-contamination during manufacturing of insect products, storage conditions, or migration from food packaging materials (Poma et al., 2019; Poma et al., 2021). In addition, OPEs contamination might also occur during distribution and storage process (Watanabe et al., 2019).

4. Looking forward

Here we have concluded that OPEs are ubiquitous detected in various agro-foods globally due to their extensive use, thus present potential challenge to food and nutrition security, and health hazards to humans. However, the information on the occurrence of OPEs in different agro-foods is still limited, which is currently mainly limited to several countries, i.e., China, Sweden, Belgium, the United States and Australia. As a consequence, a systematic global monitoring of OPE contamination in various categories of agro-foods (including raw and industrial processed agro-foods) in more countries or regions, are needed for better assessment of dietary exposure to OPEs, as diet is considered to be an important pathway of human exposure to OPEs. Moreover, several studies have suggested that OPE derivatives, such as organophosphate diesters and hydroxylated metabolites, could co-exist with parent OPEs in environmental samples or agro-foods (Fu et al., 2017; He et al., 2018b; Hou et al., 2017). Also, some emerging OPEs, such as tris(2,4-di-tert-butylphenyl) phosphate (TDtBPP), have been recently discovered in the environment and fish samples (Li et al., 2022; Ye et al., 2021). Studies are thus recommended to investigate the occurrence of possible OPE metabolites or emerging OPEs in agro-foods and ambient agri-environment, and agricultural inputs. Meanwhile, the current targeted ion monitoring strategy to identify OPEs in environmental and food samples can only detect a limited number of potential compounds. There is still a challenge for these targeted approaches to identify unknown or novel derivatives of OPEs. To comprehensively identify or evaluate all known and unknown OPEs in environmental and food samples, it is necessary to develop untargeted strategies tailored for screening of OPEs. Recently, a LC-Orbitrap-HRMS based non-target analytical strategies have been established, and BEHPP have been identified in environmental samples (Meng et al., 2020). More efforts should be taken to expand these strategies into agro-foods samples, and comprehensively investigate all potential OPEs in these samples.

Agri-environment, agricultural inputs and processing are suggested the three major sources of contamination in agro-foods by OPEs. However, at present, there is a lack of sufficient evidence on sources of exposure to OPEs during animal breeding, plant cultivation, or agro-foods processing, and on the route of how OPEs transmit to crops or animals from ambient sources. Therefore, more relevant research is required, not only focused on concentrations of OPE compounds in agro-foods, but also on occurrences and the characteristics of OPE compounds in ambient agri-environments such as farmland soil, irrigation water, sediment and air, agricultural inputs, such as feeds and contamination during processing or from packaging materials, in the agro-food chain, in order to obtain a full picture of exposure and transmission behavior of OPEs in agro-foods in a spatially and temporally-consistent fashion. These results will also benefit for risk analysis during crop cultivation, animal breeding, or agro-foods production.

Animals or plants can ingest or uptake OPEs from the exposure sources, and therefore residue in either animal-derived or plant-derived agro-foods via different transmission routes. The common metabolic pathways of OPEs including O-dealkylation, hydroxylation and oxidative dehalogenation, and are considered species diverse. In general, the metabolic rate between OPEs varied significantly, and non-halogenated alkyl OPEs were metabolized faster than halogenated alkyl OPEs, which suggests structural dependence of metabolic pathways of OPEs. But the data on the transmission, metabolism and accumulation of OPEs in biota, especially in food animals or plants, is still limited. In addition, many metabolites of OPEs are unclear, and their side effects in organisms, including adsorption, distribution, metabolism and excretion, remain unknown. Future comprehensive studies on the fate, bioavailability, and metabolic mechanism of OPEs in animals or plants, and the mechanism and kinetics of tissue-specific accumulation of OPEs by organisms are required. More in vivo studies can be conducted to identify more comprehensive metabolites to provide detailed view of the metabolism of OPEs in different organisms. Advanced analytical methods need to be explored to fill the gaps. Furthermore, the controversial results about biomagnification of OPEs underlines the need of more studies to clarify the behavior of these compounds along different food webs, to better understand the pathways of accumulation of OPEs.

Although estimated daily intake of total OPEs from consumption of agro-foods are less than reference dose values given in various reports (Liu et al., 2019b; Poma et al., 2018). Considering increasing consumption of OPEs, which might result in greater concentrations of these chemicals in environmental matrices and agro-foods, more severe exposure to humans around the world is expected. Measures to control emissions of OPEs to

agricultural areas and agro-products should be taken. Meanwhile, there is a need for establishing the related testing or limit standard for OPEs in agriculture. Furthermore, effective removal or degradation techniques for OPEs, such as nanoscale particle based strategies, are expected to develop to protect the agri-environment or agricultural inputs from further pollution.

CRediT authorship contribution statement

Wei Zhang: Conceptualization, Methodology, Writing - original draft, Visualization. **John P. Giesy:** Conceptualization, Writing - review & editing. **Peilong Wang:** Resources, Supervision, Project administration.

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.

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Appendix A. Supplementary data

Comparison of the concentrations of the predominant organophosphate esters (OPEs) in cereals (Table S1), vegetables and fruits (Table S2), meat (Table S3), dairy products (Table S4), eggs (Table S5), aquatic-derived foods (Table S6), fats, oils, and edible insects (Table S7). Supplementary data to this article can be found online at doi:https://doi.org/10.1016/j.scitotenv.2022.154271.

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