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(Citation)
Reviews in Environmental Science and Bio/Technology,20(3):751-779

(Issue Date)
2021-09

(Resource Type)
journal article

(Version)
Accepted Manuscript

How does the Cucurbitaceae family take up organic pollutants (POPs, PAHs, and PPCPs)?

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Abstract

Crop contamination with organic pollutants is an important food safety concern. Since organic pollutants are highly toxic, human consumption of contaminated crops can harm human health. Thus, understanding the mechanisms of how organic pollutants are accumulated in crop plants can aid the development of strategies for safer crop production. It is well known that the Cucurbitaceae family accumulates organic pollutants in its fruits at high concentrations. Previous studies have described the organic pollutant-uptake mechanisms of the Cucurbitaceae family. However, an integrated understanding of organic pollutant uptake by Cucurbitaceae is still lacking. In this review, we discuss the uptake mechanisms from the perspective of plant molecular biology. We clearly show that major latex-like proteins identified from the Cucurbitaceae family play a crucial role in the uptake of organic pollutants. This is the first review to describe the mechanisms underlying the accumulation of organic pollutants in the Cucurbitaceae family across the entire uptake pathway.

KEYWORDS: crop contamination, Cucurbitaceae family, major latex-like proteins, organic pollutants

1. Introduction

Organic pollutants are widely distributed in the world and contaminate the environment (Wania and MacKay 1996; Shen et al. 2013). Organic pollutants remain in the environment without degradation because they have long half-lives (Mattina et al. 1999; Sarma et al. 2019). In agriculture, crop contamination with organic pollutants is a severe problem. If maximum residue limits are exceeded in crops, farmers must discard all of the crops cultivated on the same agricultural land. Thus, the economic loss for farmers is enormous. Furthermore, organic pollutants show toxicity, such as carcinogenicity (Abdur Rehman et al. 2017) and neurotoxicity (Pessah et al. 2019; Shi et al. 2019) in human beings, and the intake of contaminated crops can lead to severe diseases. Therefore, crop contamination is an urgent problem in recent crop production. In this review, we introduce three groups of organic pollutants involved in crop contamination: persistent organic pollutants (POPs), polyaromatic hydrocarbons (PAHs), and pharmaceuticals and personal care products (PPCPs).

POPs include organochlorine insecticides (chlordane, dichlorodiphenyltrichloroethanes [DDTs], drins [aldrin, endrin, and dieldrin], and hexachlorocyclohexane [HCHs]); industrial materials (polychlorinated biphenyls [PCBs], polybrominated diphenyl ethers (PBDEs), and per- and polyfluoroalkyl substances [PFASs]); and unintentional products (polychlorinated dibenzo-*p*-dioxins [PCDDs] and polychlorinated dibenzofurans [PCDFs]). The use and production of POPs were strictly prohibited in 181 countries in 2020. In particular, large amount of organochlorine insecticides have been applied in agricultural land before they were banned from being use. For example, Japan produced or imported about 44,467 t of DDT, 683 t of dieldrin, and 315,000 t of HCH (Namiki et al. 2018).

PAHs, unlike POPs, are not intentionally produced and released to agricultural land. PAHs are mainly generated from the biomass combustion (Shen et al. 2021). They are produced by incomplete straw burning and biochar treatment used to promote plant growth in agricultural lands (Jenkins et al. 1996; Fabbri et al. 2013). Since they are not easily degraded, the amount of PAHs in agricultural field is increasing unknowingly.

PPCPs are new and emerging pollutants. They are released into wastewater after their use. After the treatment, the reclaimed wastewaters are irrigated to agricultural lands in countries where freshwater supply is limited (Pedersen et al. 2003; Miller et al. 2016). Some PPCPs are detected in irrigated water and agricultural runoff and lands (Pedersen et al. 2003, 2005; Xu et al. 2009; Bourdat-Deschamps et al. 2017). Consequently, carbamazepine, one of the PPCPs, is detected in the urine of people who eat crops treated with the irrigated water (Paltiel et al. 2016).

Many crops with edible aerial parts (fruits, leaves, and stems) do not accumulate organic pollutants in their aerial parts. However, high concentrations of organic pollutants are detected in the aerial parts of the Cucurbitaceae family, such as cucumber (*Cucumis sativus*), melon (*C. melo*), pumpkin (*Cucurbita maxima*), squash (*C. pepo*), and zucchini (*C. pepo*) (Otani et al. 2007).

The purpose of this review is to discuss the uptake mechanisms of organic pollutants in the Cucurbitaceae family in four steps: (1) solubilization, (2) absorption, (3) translocation, and (4) transport, and show the crucial steps in their uptake. Since the amount of organic pollutants in the aerial parts of the Cucurbitaceae family shows that they are not or less metabolized in the plants, we mainly discuss the uptake of organic pollutants.

(1) Organic pollutants in the soil bind to soil organic matters (SOMs), and their bioavailability is low. However, root exudates, especially organic acids, disrupt the linkage between SOMs and organic pollutants and then solubilize organic pollutants.

(2) Solubilized organic pollutants are adsorbed to the roots, absorbed into the root cells, taken up into the root tissues, and localized in the root tissues depending on their physicochemical properties.

(3) Organic pollutants are translocated from the roots (inside the Casparian strip) into xylem vessels.

(4) The organic pollutants translocated into xylem vessels are transported to aerial parts by the cohesion force of water (Pockman et al. 1995; Chunfang et al. 1999).

We show that the translocation of organic pollutants from the roots to xylem vessels plays a crucial role in their uptake in the Cucurbitaceae family. Major latex-like proteins (MLPs) produced in the root cells bind to organic pollutants through their internal hydrophobic cavity and increase their solubility (Inui et al. 2013; Goto et al. 2019; Fujita et al. 2020b; Iwabuchi et al. 2020). MLP-organic pollutant complexes are translocated into xylem vessels, and organic pollutants are transported to the aerial parts (Goto et al. 2019). Consequently, contamination with organic pollutants occurs in the Cucurbitaceae family. We discuss the organic pollutants contaminating agricultural land and their accumulation in the Cucurbitaceae family. We show that the different accumulation levels of organic pollutants in their aerial parts depend on their physicochemical properties, such as hydrophobicity and chemical structure. Furthermore, we introduce a cultivation method for safer crop production. Finally, we conclude by suggesting the need for necessary future research for a comprehensive understanding of the uptake of organic pollutants in the Cucurbitaceae family. This is the first review to focus on organic pollutant uptake from the perspective of plant molecular biology. This review leads to the development of new techniques to produce safer crops and provide them in our daily diet.

2. Uptake mechanisms

2.1. Solubilization

The first step in the uptake of organic pollutants is their solubilization in the rhizosphere and the increase in their bioavailability for plants. The bioconcentration factor (BCF) of PCDD/Fs in the aerial parts of *C. pepo* in soil culture is much lower than that in hydroponic culture (Inui et al. 2008, 2011). This suggests that root exudates are necessary to release PCDD/Fs from the soil components. The roots exude sugars, organic acids, amino acids, secondary metabolites, carbohydrates, and proteins for the uptake of nutrients and signal transduction between the roots (Walker et al. 2003; Preece and Peñuelas 2020; Wang et al. 2020). Particularly, organic acids contribute to the uptake of metal ions by dissolving metal ions through chelation (Yang et al. 2001; Dakora and Phillips 2002; White and Kottler 2002; White et al. 2003a, 2006; Luo et al. 2006; Chen et al. 2017). However, they have a secondary influence on the rhizosphere. Organic acids disrupt the soil structure and humic organic compounds-metal ions-mineral linkages (Yang et al. 2001; White and Kottler 2002; White et al. 2003a, 2006; Luo et al. 2006). Bioavailable *p,p′*-DDT has a negative correlation with the amount of SOM (Luo et al. 2006). DDX includes dichlorodiphenyldichloroethane (DDD) (*o,p′*-DDD and *p,p′*-DDD), dichlorodiphenyldichloroethylene (DDE) (*o,p′*-DDE and *p,p′*-DDE), and DDT (*o,p′*-DDT and *p,p′*-DDT) (Meijer et al. 2001). Since soil organic carbon (SOC) binds to DDX in the rhizosphere, it decreases the mobility of DDT (Tao et al. 2004). Organic acids (acetic acid, succinic acid, l-aspartic acid, malic acid, L-glutamic acid, salicylic acid, shikimic acid, isocitric acid, chorismic acid, sinapic acid, caffeic acid, *p*-hydroxybenzoic acid, gallic acid, tartaric acid, ferulic acid, protocatechuic acid, *p*-coumaric acid, mugineic acid, oxalic acid, citric acid, and piscidic acid) release organic pollutants from the linkages, and then, organic pollutants are solubilized and taken up into the roots (Badri and Vivanco 2009).

Citric and oxalic acids were the candidates that promoted the bioavailability of organic pollutants in the rhizosphere through solubilization. These acids desorb PAHs (Subramaniam et al. 2004; Yoshihara et al. 2014), *p,p′*-DDE (White et al. 2003a), and *p,p′*-DDT (Luo et al. 2006). However, oxalic acid was not detected in the rhizosphere of *C. sativus* and *C. pepo*. Therefore, citric acid is a possible promoter of the bioavailability of organic pollutants in the root exudates of the Cucurbitaceae family (Wang et al. 2004). Citric acid promotes the accumulation of perylene (Yoshihara et al. 2014), PCBs (White et al. 2006), and *p*,*p*′-DDE (White et al. 2003a) in the aerial parts of the Cucurbitaceae family. The amount of citric acid in the rhizosphere of *C. sativus* was higher than that of *C. pepo*, and the root concentration factor (RCF) of *p*,*p*′-DDE of *C. sativus* was higher than that of *C. pepo* (Wang et al. 2004). This clearly shows that the amount of citric acid correlates with the root uptake of organic pollutants. However, citric acid does not play a crucial role in the uptake of organic pollutants to the aerial parts because the BCF of *p*,*p*′-DDE in the aerial parts of *C. pepo* is much higher than that of *C. sativus* (Wang et al. 2004). Moreover, citric acid promotes the root uptake of *p*,*p*′-DDE in the non-Cucurbitaceae family (White and Kottler 2002). These studies suggest that citric acid plays an important role in the solubilization of POPs and then uptake into roots, but the released amount from the Cucurbitaceae family tends to be lower than that from the non-Cucurbitaceae family. *C. pepo* root exudates increase the root uptake of PCBs in weeds (beggar's tick [*Bidens cernua*], lamb's quarter [*Chenopodium album*], wild carrot [*Daucus carota*], broad-leaved plantain [*Plantago major*], and curly dock [*Rumex crispus*]), and *C. pepo* (Ficko et al. 2011). Therefore, citric acid promotes the bioavailability and root uptake of organic pollutants from the rhizosphere, but it does not influence the accumulation of organic pollutants in the aerial parts of plants in the Cucurbitaceae family.

2.2. Absorption

Organic pollutants are transported from the root periphery to the vascular bundles. Figure 1 shows the typical dicot cross root structure. The outer surface is covered with epidermis with root hair cells. From the outer surface, the root is mainly divided into the epidermis, cortex, endodermis, pericycle, and vascular bundles (phloem and xylem vessels) (Banda et al. 2019). Compounds are transported to vascular bundles via any of these three pathways: apoplastic, symplastic, or transcellular (Ramakrishna and Barberon 2019). In the apoplastic pathway, compounds are transported by passive diffusion through the extracellular space in the cell wall, and their transport is blocked by the Casparian strip formed by lignin (Naseer et al. 2012). Since the plasma membrane at the Casparian strip attaches to intercellular walls, it works as a diffusion barrier in endodermis (Roppolo et al. 2011). In the symplastic pathway, compounds are transported from cells to cells through plasmodesmata, which comprises cytoplasmic connections between adjacent cells from the epidermis to pericycle (Ma and Peterson 2001). They depend on polarized influx and efflux or diffusion gradients through the plasma membrane, and their transport is blocked by suberin lamella (Barberon et al. 2016; Doblas et al. 2017). In the transcellular pathway, compounds are transported by crossing membranes of neighboring cells (Peterson and Enstone 1996; Li et al. 2017).

Perylene, a hydrophobic organic pollutant, is passively diffused into the plasma membrane and transported intracellularly via plasmodesmata (Yamazaki et al. 2015). Surprisingly, the transport of perylene is not blocked by the Casparian strip and localized in the plasma membrane of the endodermis and pericycle. Thus, perylene is transported by the symplastic pathway through the plasma membrane. In addition, it has been suggested that non-ionic compounds such as hydrophobic organic pollutants are passively transported to vascular bundles through the cell membrane (Collins et al. 2006). However, the adsorption and absorption of organic pollutants in the root tissue are not different in *C. pepo* ssp. *ovifera* (low accumulator) and *C. pepo* ssp. *pepo* (high accumulator) (Yamazaki et al. 2015). Therefore, the absorption step does not influence the uptake of organic pollutants in the Cucurbitaceae family.

2.3. Translocation

The crucial factor for the uptake of organic pollutants in the Cucurbitaceae family is the translocation of organic pollutants into xylem vessels. Since perylene is localized in the endodermis and pericycle of both *C. pepo* ssp. *ovifera* and *C. pepo* ssp. *pepo*, their specific translocation mechanisms would underlie the uptake of organic pollutants in the Cucurbitaceae family (Yamazaki et al. 2015).

Chlordane and heptachlor *exo*-epoxide (HEPX) are transported from roots to aerial parts through xylem vessels in *C. sativus* and *C. pepo* (Mattina et al. 2004). Xylem sap consists of water from the roots, and it is difficult to solubilize hydrophobic organic pollutants. Since the solubilization ability of xylem sap by protein deactivators is decreased, xylem sap from the Cucurbitaceae family is thought to contain protein that has the ability to solubilize organic pollutants (Murano et al. 2010; Inui et al. 2013). This suggests that the protein in xylem sap binds to and solubilizes organic pollutants.

The 17 kDa protein in xylem sap from several *C. pepo* cultivars has a positive correlation with the BCF of PCBs in aerial parts and is identified as a major latex-like protein (MLP) (Inui et al. 2013). They were first identified in opium poppy (*Papaver somniferum*) in 1985 (Nessler et al. 1985) and have been identified in the Cucurbitaceae family (*C. sativus* (Iwabuchi et al. 2020), *C. moschata* (Iwabuchi et al. 2020), fig leaf squash [*Cucurbita ficifolia*] (Iwabuchi et al. 2020), loofah [*Luffa cylindrica*] (Iwabuchi et al. 2020), *C. melo* (Aggelis et al. 1997), white-flowered gourd [*Lagenaria siceraria*] (Iwabuchi et al. 2020), and *C. pepo* (Inui et al. 2013; Goto et al. 2019)) and non-Cucurbitaceae family (*Arabidopsis thaliana* (Wu et al. 2008), cotton [*Gossypium hirsutum*] (Chen and Dai 2010), ginseng [*Panax ginseng*] (Choi et al. 2015), grapevine [*Vitis vinifera*] (Zhang et al. 2018), peach [*Prunus persica*] (Ruperti et al. 2002), and soybean [*Glycine max*] (Strömvik et al. 1999)). MLPs have various biological functions, which include: disease resistance (Yang et al. 2015; Gai et al. 2018; Song et al. 2020), salt stress tolerance (Wang et al. 2016), leaf formation (Litholdo et al. 2016), enzymatic activity (Lichman et al. 2020), and plant hormone response (Ruperti et al. 2002; Sun et al. 2010; Li et al. 2013; Zhang et al. 2018). The most striking structural feature of MLPs is the internal hydrophobic cavity, which enables them to bind to hydrophobic compounds (Lytle et al. 2009; Fernandes et al. 2013; Choi et al. 2015). Recombinant MLPs from the Cucurbitaceae family bind to organic pollutants, such as 17β-estradiol (Goto et al. 2019), 4-hydroxy-2′,3,3′,4′,5′-pentachlorobiphenyl (4OH-PeCB106) (Inui et al. 2013), 4-*t*-octylphenol (Goto et al. 2019), dieldrin

(Goto et al. 2019; Fujita et al. 2020b), and pyrene (Fujita et al. 2020b). Therefore, organic pollutants are solubilized in xylem sap through the binding of MLPs. Since *MLP* genes are mainly expressed in the root (Goto et al. 2019), MLPs bind to organic pollutants in the plasma membrane of the endodermis and pericycle. Finally, MLP-organic pollutant complexes are translocated into xylem vessels (Figure 2).

2.4. Transport

Translocated organic pollutants into xylem vessels are transported to aerial parts by water cohesion force (Pockman et al. 1995; Chunfang et al. 1999). Phloem vessels can also be the route. Xylem and phloem sap are acidic and basic, respectively. Since MLPs bind to organic pollutants under an acidic condition, xylem vessels are an important route (Inui et al. 2013; Goto et al. 2019; Fujita et al. 2020b; Iwabuchi et al. 2020). Transported organic pollutants to aerial parts cause physiological changes in plants. *p*,*p*′-DDE induces genes related to signal transduction, environmental stress, and photosynthesis in *C. pepo* (Chhikara et al. 2010). This suggests that *p*,*p*′-DDE in the soil induces stress in plants and inhibits physiological functions such as photosynthesis.

There is a difference in the uptake ability of the Cucurbitaceae family. The concentrations of dieldrin and endrin in the shoots of *C. pepo* were two and three times higher than those in the shoots of *L. cylindrica*, respectively (Otani et al. 2007). The binding affinity of MLPs can explain this difference. MLP from *L. cylindrica* does not bind to PeCB106, but MLP from *C. pepo* does (Inui et al. 2013; Iwabuchi et al. 2020). This suggests that the cavity of MLP from *L. cylindrica* is less hydrophobic, and the binding affinity of MLP to hydrophobic organic pollutants is relatively low. Hence, the uptake amount of organic pollutants in *L. cylindrica* is low.

Previous studies have shown that cross-breeding influences the uptake amount of organic pollutants (White 2010; Isleyen et al. 2013; Sugiyama et al. 2013, 2016). The concentrations of DDX and chlordane in F1 hybrids (*C. pepo* ssp. *pepo* × *C. pepo* ssp. *ovifera*) were lower than those of *C. pepo* ssp. *pepo* parents, and those in F1 back-cross plants are in the middle between parents and F1 plants (White 2010; Isleyen et al. 2013; Sugiyama et al. 2013, 2016). These results suggest that a single gene or locus controls the uptake of organic pollutants following Mendelian segregation. This suggests that the gene expression level made a difference in their uptake ability. In contrast, a recent study proposes two hypotheses regarding the inheritance of uptake ability trait, and they are two or three different dominant gene models (Sugiyama et al. 2016). The candidate genes are *MLP* and *zinc finger protein* (*ZFP*) genes. ZFPs, like MLPs, play a crucial role in the uptake of organic pollutants in the Cucurbitaceae family (Inui et al. 2015). ZFPs function as transcription factors through the binding of the zinc finger motif to DNA. The expression level of *ZFP* genes in *C. pepo* ssp. *pepo* is higher than that in *C. pepo* ssp. *ovifera*, and the expression of *CpZFP* genes promotes the accumulation of 3,3′,4,4′,5-pentachlorobiphenyl (PeCB126) in the aerial parts of transgenic tobacco (*Nicotiana tabacum*) plants. These results suggest that CpZFPs bind to the promoter region of *NtMLP* genes and induce the expression of *NtMLP* genes. However, only a single *MLP* gene, *FB7-4,* has ever been identified in *N. tabacum* (Neale et al. 1990). Thus, it is thought that ZFPs induce other genes responsible for the uptake of organic pollutants other than *MLP* genes.

3. Organic pollutants taken up by the Cucurbitaceae family

3.1. DDX (Table 1.A)

DDT was applied to control agricultural pests until it was prohibited owing to its toxicity (Lunney et al. 2004). However, DDT is still used to control malaria in several countries in Africa and Asia (Van Den Berg 2009). DDT is transformed to DDE by exposure to weathering, and DDE is the most abundant compound in DDX in the environment (Aislabie et al. 1997; Megharaj et al. 1997). DDT is directly transformed into DDD under aerobic condition (Corona-Cruz et al. 1999). In 2001, it was first confirmed that *C. pepo* accumulated a higher concentration of *p,p'*-DDE in their fruits compared with other plant families (White 2001; Lunney et al. 2004). In the Cucurbitaceae family, the genus *Cucurbita* accumulates DDX more than the genera *Cucumis* and *Citrullus* (White 2002; Gent et al. 2007; Isleyen and Sevim 2012; Isleyen et al. 2012a; Namiki et al. 2013). Furthermore, *C. pepo* has a different uptake ability in their subspecies: *C. pepo* ssp. *pepo* accumulates higher concentrations of DDX in their aerial parts than *C. pepo* ssp. *ovifera* and *texana* (White 2002, 2010; White et al. 2003b, 2005; Isleyen et al. 2012b, 2013). The BCF value of *C. pepo* ssp. *pepo* in the stem is 11 times higher than that of *C. pepo* ssp. *texana* (White et al. 2003b). Translocation factor from the roots to the stems of *C. pepo* ssp. *pepo* is 4.9 times higher than that of *C. pepo* ssp. *ovifera* (White et al. 2005). Thus, *C. pepo* ssp. *pepo* has efficient translocation mechanisms of DDX from roots to xylem vessels.

3.2. Drins (Table 1.B)

Drins consist of organochlorine insecticides aldrin, dieldrin, and endrin, and are registered as POPs (Jorgenson 2001). Dieldrin is produced through the epoxidation of aldrin by soil bacteria and is a stereoisomer of endrin (Good and Ware 1969; Ferguson and Korte 1977). In the United States, aldrin and dieldrin were the second most applied pesticides in the 1960s, and the amount of aldrin applied in Iowa between 1961 and 1965 reached 5-6.6 million pounds (Jorgenson 2001). Since dieldrin had been applied to control agricultural pests, such as spotted cucumber beetle (*Diabrotica undecimpunctata*), before it was banned, a large amount of dieldrin remains in agricultural lands (Gladstone and Wong 1977). Dieldrin has been detected in agricultural soils in Japan (Hashimoto 2005), Portugal (Gonçalves and Alpendurada 2005), the United States (Harner et al. 1999), and Switzerland (Hilber et al. 2008). Since 1965, the Cucurbitaceae family has accumulated dieldrin in their fruits (Lichtenstein and Schulz 1965). Dieldrin and endrin were found to be accumulated at much higher concentrations in the Cucurbitaceae family than in other plant families (Alliaceae, Amaranthaceae, Apiaceae, Asteraceae, Brassicaceae, Chenopodiaceae, Euphorbiaceae, Fabaceae, Lamiaceae, Linaceae, Malvaceae, Pedaliaceae, Poaceae, Polygonaceae, Solanaceae, Tiliaceae) (Otani et al. 2007; Murano et al. 2010; Saito et al. 2012; Namiki et al. 2013, 2018). Contamination with dieldrin in the Cucurbitaceae family is a severe problem in crop production and often occurs in Japan. The agricultural land in Tokyo is contaminated with dieldrin, and in *C. sativus,* maximum residue limit has been exceeded (Hashimoto 2005). One of the reasons for the accumulation is the low maximum residue limit of dieldrin (0.02 ppm) compared with that of other POP-organochlorine insecticides (HCHs [0.2 ppm] and DDX [0.2 ppm]). In addition, the chemical properties of dieldrin can be a factor for contamination: the half-life of dieldrin is longer than that of HCHs, and the logK_{ow} value of dieldrin is lower than that of DDX (Namiki et al. 2013). Thus, dieldrin is not readily degraded in soil and is not retained in the soil. Hence, the amount of dieldrin taken up into the roots is more than that of HCHs and DDX. Another reason is the binding affinity of MLPs to dieldrin. The BCF of bulky PCBs in the aerial parts of *C. pepo* is higher than that of planer PCBs (Matsuo et al. 2011). This suggests that compounds with a bulky structure fit into the hydrophobic cavity of MLPs. Since dieldrin is a bulky structure, the 3D structure of dieldrin may fit into the cavity of MLPs, and thus, the amount of dieldrin in the aerial parts of the Cucurbitaceae family is large. Consequently, maximum residue limit of dieldrin is often exceeded in crops.

3.3. Other POPs and their related organochlorine insecticides (Table 1.C)

HCH has been used as an insecticide, and commercial HCH is mainly a mixture of α-, β-, and γ-HCH isomers (Vijgen et al. 2011). Only γ-HCH, called Lindane, shows insecticidal activity, and 450,000 t of γ-HCH have been applied to agricultural land between 1950 and 2000 in the world (Vijgen et al. 2011). In particular, β-HCH is highly toxic to humans through the activation of estrogenic action (Steinmetz et al. 1996). Hence, HCH isomers (α, β, and γ) are registered as POPs (Vijgen et al. 2011).

HCHs are also taken up by the Cucurbitaceae family, but their uptake, which is different with isomer, has not been investigated (Namiki et al. 2013, 2015, 2018).

Technical chlordane is a mixture of more than 140 compounds and has been applied as an insecticide and herbicide in the United States for agricultural and residential purpose (Dearth and Hites 1991; Mattina et al. 1999). The major components of technical chlordane include chlordane isomers (*cis*-chlordane and *trans*-chlordane), *trans*-nonachlor, and heptachlor (Dearth and Hites 1991). *Cis*-chlordane and *trans*-chlordane contain enantiomers: (-) *cis*-chlordane and (+) *cis*-chlordane, (-) *trans*-chlordane, and (+) *trans*-chlordane (Mattina et al. 2002). It is known that chlordane and HEPX are detected in the *C. pepo* and *C. melo* cultivated in the United States (Mattina et al. 2000). *C. pepo* accumulated the highest concentrations of chlordane in the edible aerial parts of all tested crops cultivated in the soil where chlordane was applied 38 years ago (Mattina et al. 2000). The amount of *cis*-chlordane taken up into the roots of *C. pepo* was higher than in *trans*-chlordane (Mattina et al. 2002). Enantiomer fraction (EF) is used to understand enantioselectivity and is defined as the ratio of (+) enantiomers in the sum of (-) and (+) enantiomers. EFs of chlordane isomers in the fruits of *C. pepo* were higher than those in the roots (Mattina et al. 2002; White et al. 2002). In contrast, EFs in the fruits of *C. sativus* were lower than those in the roots (Mattina et al. 2002). From the roots to the fruits of *C. pepo*, EF of *cis*-chlordane increased, but that of *trans*-chlordane decreased (White et al. 2002). *Cis*-chlordane is the dominant component in the roots, stems, and fruits of *C. pepo* ssp. *pepo* and *ovifera*. From the roots to the fruits of *C. pepo* ssp. *pepo* and *ovifera*, EF of *trans*-chlordane increased, while that of *cis*-chlordane decreased; (+) *trans*-chlordane and (-) *cis*-chlordane in the fruits are more dominant than those in the roots (Isleyen et al. 2013). These results suggest that chlordane is taken up and accumulates isomer- and enantiomer- selectively. These enantiomer selectivities can be explained by the binding selectivity of MLPs because chlordane isomers are hydrophobic compounds.

3.4. PCBs and PCDD/Fs (Table 1.D)

Polychlorinated organic pollutants, such as PCBs (Manz et al. 2001; Armitage et al. 2006; Zhang et al. 2007) and PCDD/Fs (Jou et al. 2007; Shen et al. 2009; Deng et al. 2011), are detected in agricultural lands via waste emission and volatilization. The uptake of PCDD/Fs in the Cucurbitaceae family was investigated in 1994 (Hülster et al. 1994). The study showed, for the first time, that *Cucurbita* genus accumulated organic pollutants from the roots to the fruits through the stem and denied the established theory that the evaporation of organic pollutants from the soil contaminated the fruits in the Cucurbitaceae family. The Cucurbitaceae family, especially the *Cucurbita* genus, shows a higher transpiration stream concentration factor of PCDD/Fs than other plant species, although RCF is equivalent (Zhang et al. 2009). In addition, it is shown that PCBs are also accumulated from the roots via xylem vessels (Whitfield Åslund et al. 2008; Greenwood et al. 2011).

It is notable that the hydrophobicity of PCB congeners does not correlate with their BCF, although the hydrophobicity of PCDD/Fs congeners has a negative correlation with their BCF in *C. pepo* (Matsuo et al. 2011). Thus, the uptake of PCBs depends on factors other than hydrophobicity (Inui et al. 2011). PeCB congeners with chlorines at *ortho*-positions tend to be highly taken up (Matsuo et al. 2011). Furthermore, in the four PeCB congeners accumulated in *C. pepo*, there were three PCB congeners with chlorines at *ortho*-positions (2,2',3,5,6-PeCB [PeCB93], 2,2',3,5',6-PeCB [PeCB95], and 2,3,3',4,4'-PeCB [PeCB105]) (Whitfield Åslund et al. 2007). These results clearly show that the uptake of PCBs in *C. pepo* shows congener selectivity (Matsuo et al. 2011; Goto et al. 2019). The volume of PCB congeners with chlorines at *ortho*-positions is bulky because they prevent the rotation of the C-C bond between the rings (Fujita et al. 2020b). In contrast, the accumulation of tri-, tetra-, and hexachlorinated biphenyl (HxCB) congeners is not influenced by their bulkiness but by the number of chlorines, hydrophobicity, and molecular weight (Greenwood et al. 2011). However, PeCB and HxCB congeners with chlorines at *ortho*-positions are highly accumulated in *C. pepo* ssp. *pepo* (Matsuo et al. 2011). Thus, further research is needed to clarify the accumulation mechanisms of PCB congeners.

3.5. Others (Table 1.E)

PAHs show high hydrophobicity and are also accumulated in the aerial parts of the Cucurbitaceae family (Mattina et al. 2006; Parrish et al. 2006). However, a comparison of the uptake ability of PAHs between the Cucurbitaceae family and the non-Cucurbitaceae family has never been investigated. To date, the accumulation level of PAHs is thought to be equivalent to that of POPs. Pyrene, a PAH, show high hydrophobicity (logK_{ow}, 5.2) (Miller et al. 1985) and is accumulated in *C. pepo* (Fujita et al. 2020b). The same hypothesis that the BCF of POPs accumulated in *C. pepo* ssp. *pepo* is more than that in *C. pepo* ssp. *ovifera* and *C. sativus* is confirmed in case of PAHs such as anthracene, fluoranthene, and phenanthrene (Mattina et al. 2006). Therefore, the Cucurbitaceae family would accumulate PAHs like POPs at higher concentrations than other plant families.

PFASs have recently received attention owing to their persistence and toxicity (Brambilla et al. 2015). The Cucurbitaceae family accumulates PFASs, such as polyfluoroalkyl phosphate diesters (Lee et al. 2014), perfluorocarboxylic acids (Lee et al. 2014), and 6:2 fluorotelomer sulfonic acid (Zhao et al. 2019), but does not accumulate them at high concentrations compared with other plant families (Lechner and Knapp 2011; Felizeter et al. 2014; Zhao et al. 2018). The logK_{ow} values of PFASs, such as perfluorooctane sulfonate and perfluorooctanoic acid, are 5.26 and 4.59, respectively, and nearly equivalent to that of dieldrin (logK_{ow}, 5.2) (Namiki et al. 2013; Milinovic et al. 2016). Since their translocation is not blocked by the Casparian strip, they are localized in the endodermis and pericycle (Yamazaki et al. 2015). Therefore, PFASs are targeted by MLPs for binding. However, PFASs have an alkyl chain, unlike other POPs.

The structures of PFASs are different from those of DDX, drins, and PCBs, which contain two benzene rings (DDX and PCBs) or a naphthalene ring (drins) as a basic structure (Chakraborty and Das 2016). However, PFASs contain a long alkyl chain (Ghisi et al. 2019). Compounds binding MLPs from the Cucurbitaceae family usually contain ring structures, and compounds with a long chain have never been identified as MLP-binding compounds. Therefore, MLPs are thought to not bind PFASs. Consequently, PFASs are not translocated into xylem vessels as MLP-PFAS complexes.

PPCPs include acetaminophen, caffeine, and carbamazepine. Carbamazepine is detected in the aerial parts of *C. sativus* and *C. pepo* (Shenker et al. 2011; Knight et al. 2018). Sixteen PPCPs were tested in 17 PPCPs accumulated in the shoots of *C. sativus* (Sun et al. 2018). However, the Cucurbitaceae family does not accumulate PPCPs at high concentrations compared with other plant families (Wu et al. 2013; Garvin et al. 2015). The uptake level (BCF and TSCF) of PPCPs was not higher in the Cucurbitaceae family than in the non-Cucurbitaceae family, probably owing to the low hydrophobicity of PPCPs, except for triclocarban, which shows a high hydrophobicity (Wu et al. 2013; Garvin et al. 2015). For example, the $\log K_{\mathrm{ow}}$ values of carbamazepine and caffeine are 2.45 and -0.07, respectively (Shenker et al. 2011; Garvin et al. 2015). Therefore, PPCPs do not enter the inside of the Casparian strip, and consequently, MLPs do not bind PPCPs at a high affinity.

4. Approaches for safer crop production

Agricultural lands in the world are contaminated with organic pollutants, and farmers have to produce crops for food supply. It is essential to produce safer crops even in agricultural lands that are contaminated with organic pollutants. Recently, novel approaches to reduce crop contamination through the regulation of MLPs have been proposed for the treatment of agrochemicals: suppression of the expression of *MLP* genes and inhibition of the binding of MLPs to organic pollutants. Since the expression of *MLP* genes is influenced by environmental factors, such as temperature (Sun et al. 2010;

Zhang et al. 2018; Inui et al. 2020), light period (Neale et al. 1990; Inui et al. 2020), drought (Sun et al. 2010; Wang et al. 2016; Ma et al. 2017; Zhang and Shi 2018; Lv et al. 2020), and flood (Mustafa et al. 2015), it suggests that the application of agrochemicals can control the expression of *MLP* genes. The fungicide Daconil suppresses the expression of *MLP* genes in the roots and reduces the uptake of dieldrin and pyrene in *C. pepo* (Fujita et al. 2020a). In contrast, MLPs, which are identified in several plants, bind various hydrophobic compounds (Lytle et al. 2009; Choi et al. 2015). Thus, agrochemicals that bind MLPs can inhibit the binding of MLPs to organic pollutants. The insecticide Colt containing the MLP-binding compound, pyrifluquinazon, as an active ingredient, inhibits the binding of MLPs to dieldrin and pyrene, and its application reduces the uptake of dieldrin and pyrene into *C. pepo* (Fujita et al. 2020b). These studies contribute to safer crop production in agricultural lands contaminated with organic pollutants.

Transgenic plants are powerful tools for the reduction of the uptake of organic pollutants in the Cucurbitaceae family. Since recent studies have developed an efficient transformation method using hairy root culture, the transformation of the Cucurbitaceae family is easier than ever (Nanasato et al. 2013). LinA from *Sphingomobium japonicum* UT26 was identified as a dehydrogenase responsible for the degradation of γ-HCH (Imai et al. 1991). *C. moschata* expressing *LinA* accumulates and degrades γ-HCH (Nanasato et al. 2016). To date, genes responsible for the degradation of organic pollutants, such as PCBs (Kimbara et al. 1989) and PCDD/Fs (Habe et al. 2001; Miyauchi et al. 2008), have been identified in microorganisms. Therefore, the Cucurbitaceae family that expresses these genes can help in the remediation of agricultural lands.

The soil type affects the accumulation of organic pollutants in the Cucurbitaceae family. The cultivation in allophanic soils suppressed the accumulation of chlordecone in the fruits of *C. sativus* because the clay fractal structure of the allophane traps chlordecone in the soil (Woignier et al. 2012; Clostre et al. 2014). To date, the application of adsorbents was attempted to trap organic pollutants.

The wood chip charcoal decreased the concentration of dieldrin in the fruits of *C. sativus* (Saito et al. 2011). In addition, the active carbon showed the higher suppression effects on the accumulation of dieldrin in the fruits of *C. sativus* (Hashimoto 2007; Saito et al. 2011) and decreased HEPX concentrations in the fruits of *C. maxima* (Murano et al. 2009).

Grafting also controls the uptake of organic pollutants. Since grafting decreases the concentration of dieldrin in *C. sativus* fruits on low accumulator rootstocks (Otani and Seike 2007), the concentrations of organic pollutants in the scion (aerial parts) depend on the uptake ability in the rootstock (Otani and Seike 2006, 2007; Hashimoto 2007; Isleyen and Sevim 2012). In Japan, 80% of *C. sativus* is cultivated by grafting on the *Cucurbita* rootstock (Otani and Seike 2006). Thus, the selection of rootstock of low-accumulator species is crucial for safer crop production.

5. Concluding remarks and future perspectives

In this review, we discussed the specific uptake mechanisms of organic pollutants in the Cucurbitaceae family. Their uptake in plants occurs in four steps: (1) solubilization, (2) absorption, (3) translocation, and (4) transport. We showed that the translocation of organic pollutants plays a crucial role in their uptake in the Cucurbitaceae family. MLPs bind organic pollutants on the plasma membrane of the endodermis and pericycle in the roots, and MLP-organic pollutant complexes are translocated into xylem vessels. The solubilization of organic pollutants is promoted through the binding of MLPs, and solubilized organic pollutants in xylem sap are transported to the fruits (Figure 3). Therefore, the Cucurbitaceae family accumulates hydrophobic organic pollutants such as organochlorine insecticides, PCBs, and PCDD/Fs. It is thought that MLPs tend to bind hydrophobic organic pollutants with a high affinity because MLPs have an internal hydrophobic cavity as their binding site. However, hydrophilic organic pollutants, such as PFASs with a short alkyl chain and PPCPs, are not accumulated at high concentrations in the Cucurbitaceae family compared with other plant families.

MLPs are a key factor for the uptake of organic pollutants in the Cucurbitaceae family. However, there are still gaps in understanding their accumulation and biological functions. Thus, further research is recommended owing to the following:

(1) MLPs are distributed in dicots and monocots. *MLP* genes in *G. hirsutum* (Yang et al. 2015), *N. benthamiana* (Song et al. 2020), and sugar beet (*Beta vulgaris*) (Kloos et al. 2002; Oltmanns et al. 2006) are highly expressed in the roots like *C. pepo* (Goto et al. 2019). However, only MLPs from the Cucurbitaceae family translocate organic pollutants from the roots into xylem vessels.

(2) Factors responsible for different uptake abilities in these subspecies have not been clarified. Since the localization of organic pollutants in the root tissues is not different among the subspecies, that of MLPs can be different. For example, MLPs in *C. pepo* ssp. *ovifera* are not secreted from the endodermis and pericycle, but those in *C. pepo* ssp. *pepo* are released and readily translocate into xylem vessels. Thus, the investigation of the localization of MLPs in root tissues may lead to the understanding of different uptake mechanisms in these subspecies.

(3) The translocation mechanisms of MLP-organic pollutant complexes from the roots to the xylem vessel remain unclear. Thus, further research is necessary to clarify the translocation mechanisms of MLPs. One possibility is the interaction of MLPs with other proteins. Several proteins have been identified as interaction partners of MLPs, and it is possible that these proteins help to translocate MLP-organic pollutant complexes into xylem vessels (Yang et al. 2015; Litholdo et al. 2016; Wang et al. 2016; Lv et al. 2020).

(4) Organic pollutants accumulated in the aerial parts are metabolized during uptake. However, few studies focusing on metabolism are published (Chhikara et al. 2010; Zhai et al. 2011). As far as we know, only the metabolites of heptachlor (Hayashi et al. 2018) and PFASs (Zhao et al. 2018, 2019) have been investigated in the Cucurbitaceae family. Since PeCB126 is metabolized by mammalian cytochrome P450 species, it is essential to identify the metabolites in plants because they can show

higher toxicity than parent compounds (Mise et al. 2016). This suggests that the current contamination assessment in crops underestimates the toxicity effects.

This review clearly shows the uptake mechanism of organic pollutants in the Cucurbitaceae family. It is well known since the 1960s that the Cucurbitaceae family accumulates organic pollutants in their fruits. Subsequently, many studies have been performed to understand the mechanisms involved in their solubilization, absorption, translocation, and transport. In the last decade, MLPs have been identified as transporting factors for organic pollutants, and the crucial role of MLPs in crop contamination by hydrophobic pollutants has been recognized. Clarification of the mechanisms involved in the translocation of organic pollutants by MLPs would lead to new approaches for safer crop production.

Conflicts of interest

We declare that we have no known competing financial interests or personal relationships that could influence the work reported in this review.

Acknowledgments

We would like to express our deepest gratitude to Dr. Shigeo Takumi (Graduate School of Agricultural Science, Kobe University) and Dr. Shinji Sugiura (Graduate School of Agricultural Science, Kobe University) for their fruitful discussion that helped in the writing of the manuscript.

Funding

The contamination of the Cucurbitaceae family in the laboratory of the authors was supported by a Grant-in-Aid for Scientific Research A from the Ministry of Education, Culture, Sports, and Technology of Japan (Nos. 17208029 and 23241028) to HI, Grant-in-Aid (Hazardous chemicals) from the Ministry of Agriculture, Forestry and Fishries of Japan (HC-06-2311-1) to HI, Pesticide Science Society of Japan in 2014 to HI, the Japan Crop Protection Association in 2016 to HI, and a Sasakawa Scientific Research Grant from The Japan Science Society (No. 2019–5004) to KF.

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Figure legends

Figure 1. Localization of organic pollutants in root cells.

Organic pollutants bind to soil organic matter in the rhizosphere. Organic acids are released from the roots as root exudates and disrupt the linkage between soil organic matter and organic pollutants. Desorbed organic pollutants are solubilized, and their bioavailability is increased. Organic pollutants absorbed into the roots are diffused in the plasma membrane of the root cells and transported to the endodermis and pericycle through the plasmodesmata.

Figure 2. Translocation mechanisms of organic pollutants into xylem vessels through binding to major latex-like proteins.

MLPs produced in the root cells bind to organic pollutants in the plasma membrane of the endodermis and pericycle. MLP-organic pollutant complexes are translocated into xylem vessels and transported to the aerial parts. As a result, contamination with organic pollutants occurs in the Cucurbitaceae family. Xv, xylem vessel Pe, pericycle; En, endodermis; Co, cortex; Ep. Epidermis.

Figure 3. Accumulation steps in the Cucurbitaceae family.

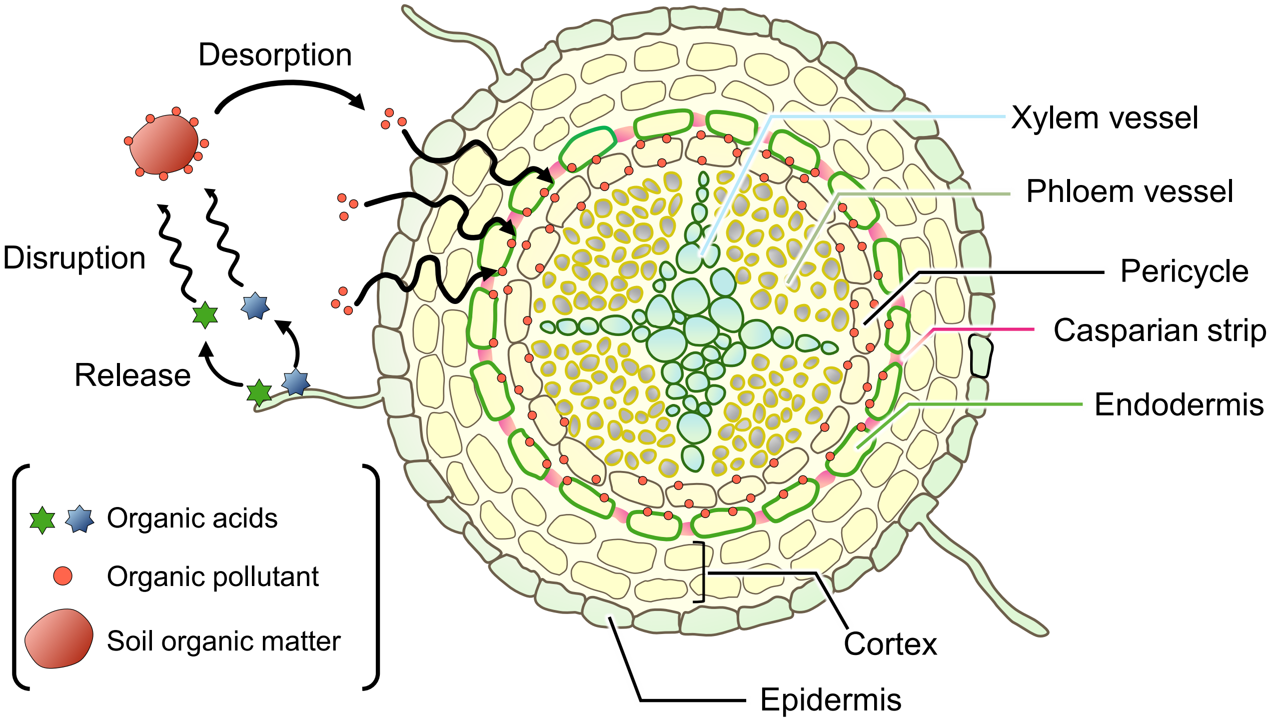

Desorption
Disruption
Release
Xylem vessel
Phloem vessel
Pericycle
Casparian strip
Endodermis
Cortex
Epidermis
Organic acids
Organic pollutant
Soil organic matter

Figure 1

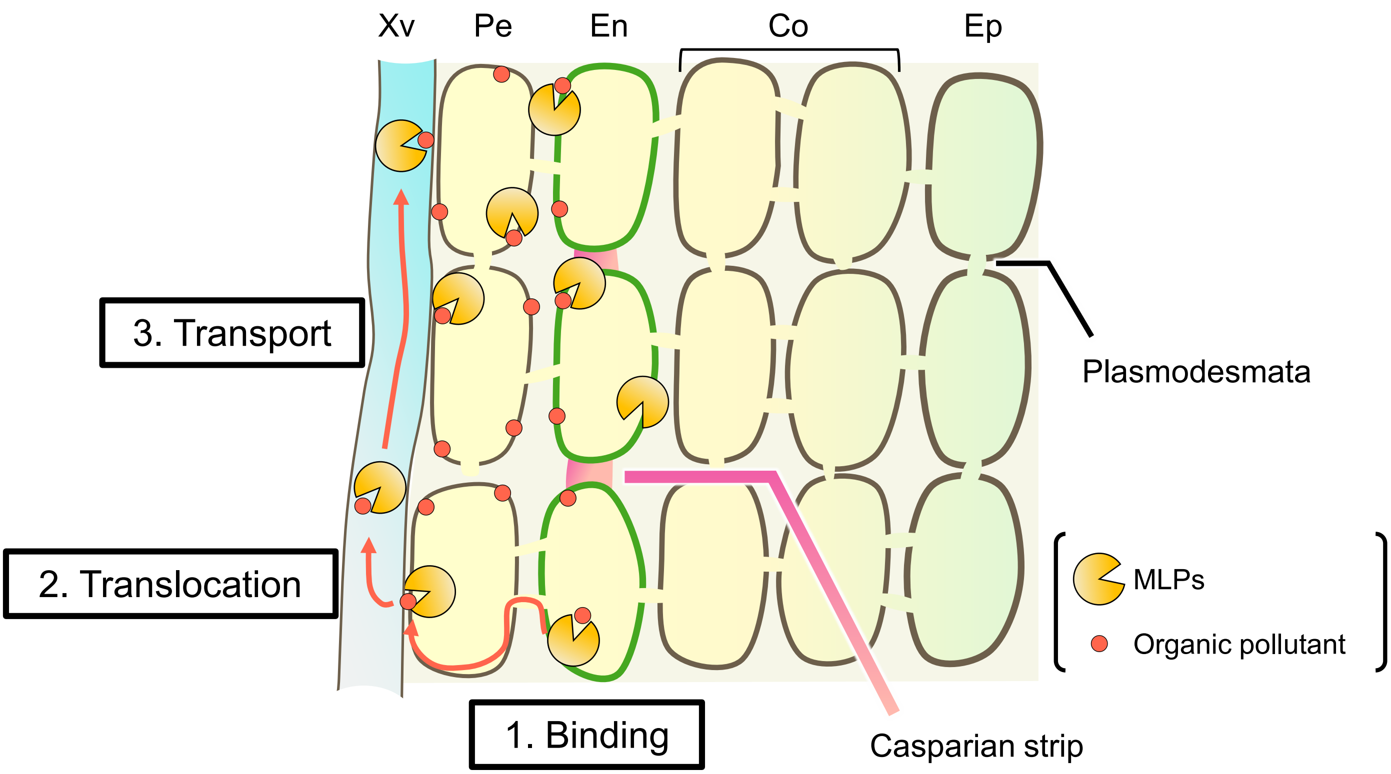

Xv
Pe
En
Co
Ep
Plasmodesmata
3. Transport
2. Translocation
1. Binding
Casparian strip
MLPs
Organic pollutant

Figure 2

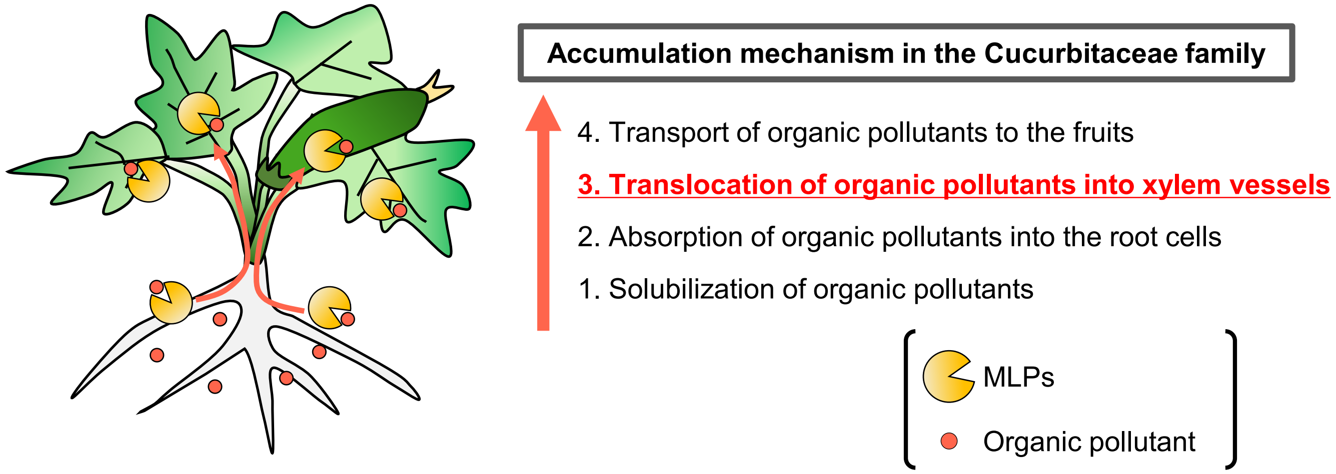
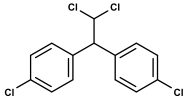
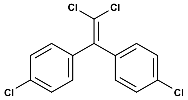
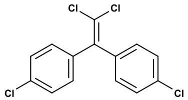

Accumulation mechanism in the Cucurbitaceae family
4. Transport of organic pollutants to the fruits
3. Translocation of organic pollutants into xylem vessels
2. Absorption of organic pollutants into the root cells
1. Solubilization of organic pollutants
MLPs
Organic pollutant

Figure 3

Table 1. Organic pollutants taken up by the Cucurbitaceae family

(A) DDX

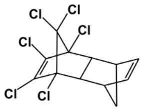
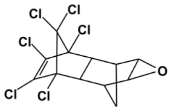
Organic pollutant	$\log K_{ow}$	Plant species	Concentration		Plant organ
			Soil	Plant	
DDD 	5.5	Cucumber (*Cucumis sativus*)	0.003 µg/g	6.46 µg/g (Shoot)	Shoot (Namiki et al. 2013)
	(Namiki et al. 2013)	Pumpkin (*Cucurbita maxima*)	0.003 µg/g	9.15 µg/g (Shoot)	
		Pumpkin (*Cucurbita moschata*)	0.003 µg/g	8.15 µg/g (Shoot)	
		Zucchini (*Cucurbita pepo*)	0.003 µg/g	7.88 µg/g (Shoot)	
DDE 	5.7	Cucumber (*Cucumis sativus*)	0.003 µg/g	18.81 µg/g (Shoot)	Shoot (Namiki et al. 2013)
	(Namiki et al. 2013)	Pumpkin (*Cucurbita maxima*)	0.003 µg/g	24.42 µg/g (Shoot)	
		Pumpkin (*Cucurbita moschata*)	0.003 µg/g	21.53 µg/g (Shoot)	
		Zucchini (*Cucurbita pepo*)	0.003 µg/g	21.40 µg/g (Shoot)	
p,p′-DDE 	6.96	Cucumber (*Cucumis sativus*)	140 µg/g	120 ng/g (Fruit)	Leaf (White 2002; Wang et al. 2004; Gent et al. 2007)
	(Sabljić et al. 1995)			(Gent et al. 2007)	Fruit (White 2002; Wang et al. 2004; Gent et al. 2007)
					Petiole(Gent et al. 2007)
					Stem (White 2002; Wang et al. 2004; Gent et al. 2007)
		Melon (*Cucumis melo*)	150–1200 ng/g	10 ng/g (Fruit)	Leaf (White 2002)
					Fruit (White 2002)
					Stem (White 2002)
		Squash (*Cucurbita pepo*)	180 ng/g	6.1 ng/g (Fruit)	Leaf (White 2002, 2010; Chhikara et al. 2010)
				(Chhikara et al. 2010)	Fruit (White 2002, 2010; Chhikara et al. 2010)
					Stem (White 2002, 2010; Chhikara et al. 2010)

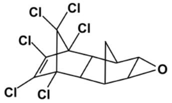
	Pumpkin (*Cucurbita maxima*)	150–1200 ng/g	46 ng/g (Fruit) (White 2002)	Fruit (White 2001, 2002) Leaf (White 2001, 2002; Peters et al. 2007) Stem (White 2001, 2002; Peters et al. 2007)
	Pumpkin (*Cucurbita pepo*)	478.4 ng/g	400 ng/g (Leaf)	Leaf (Kelsey et al. 2006) Stem (Kelsey et al. 2006)
	Watermelon (*Citrullus lanatus*)	610.21 ng/g	0.49 µg/L (Xylem sap)	Xylem sap (Isleyen and Sevim 2012)
	Zucchini (*Cucurbita pepo*)	180 ng/g	160 ng/g (Fruit) (Chhikara et al. 2010)	Fruit (White 2001, 2010; White et al. 2003a, b, 2005, 2007; Wang et al. 2004; Gent et al. 2007; Chhikara et al. 2010; Eevers et al. 2018) Leaf (White 2001, 2010; White et al. 2003b, 2005, 2007; Wang et al. 2004; Gent et al. 2007; Peters et al. 2007; Chhikara et al. 2010; Eevers et al. 2018) Petiole (Gent et al. 2007) Shoot (Namiki et al. 2013) Stem (White 2001, 2010; White et al. 2003b, 2005, 2007; Wang et al. 2004; Gent et al. 2007; Peters et al. 2007; Chhikara et al. 2010; Eevers et al. 2018) Xylem sap (Isleyen and Sevim 2012)
DDX	Pumpkin (*Cucurbita maxima*)	3,700 ng/g	4,262 ng/g (Shoot) (Lunney et al. 2004)	Shoot (Lunney et al. 2004) Stem (Denyes et al. 2016)
	Squash (*Cucurbita pepo*)	1,480 ng/g	> 4 ng/g (Leaf) (Isleyen et al. 2012a)	Fruit (Isleyen et al. 2012a) Leaf (Isleyen et al. 2012b, a) Stem (Isleyen et al. 2012b, a, 2013)

				Xylem sap (Isleyen et al. 2012b, a)
	Watermelon (*Citrullus lanatus*)	1,670 ng/g	> 2 ng/g (Leaf)	Fruit (Isleyen et al. 2012a) Leaf (Isleyen et al. 2012a) Stem (Isleyen et al. 2012a) Xylem sap (Isleyen et al. 2012a)
	Zucchini (*Cucurbita pepo*)	3,700 ng/g	2,991 ng/g (Shoot) (Lunney et al. 2004)	Aerial parts (Mattina et al. 2006) Leaf (Isleyen et al. 2012b) Shoot (Lunney et al. 2004) Stem (Isleyen et al. 2012b, 2013) Xylem sap (Mattina et al. 2006; Isleyen et al. 2012b)

DDD, dichlorodiphenyldichloroethane; DDE, dichlorodiphenyldichloroethylene; DDX, DDD, DDE, and DDT

(B) Drins

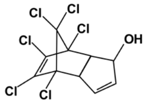
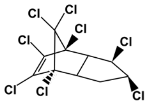
Organic pollutant	logK_{ow}	Plant species	Concentration		Plant organ
			Soil	Plant	
Aldrin 	6.5 (Shen and Wania 2005)	Cucumber (*Cucumis sativus*)	-	-	Fruit (Lichtenstein and Schulz 1965)
Dieldrin 	5.2 (Namiki et al. 2013)	Balsam pear (*Momordica charantia*)	594 µg/kg	> 100 µg/kg (Shoot)	Shoot (Otani et al. 2007)
		Figleaf squash (*Cucurbita ficifolia*)	594 µg/kg	> 1000 µg/kg (Shoot)	
		Loofah (*Luffa cylindrica*)	594 µg/kg	> 500 µg/kg (Shoot)	
		White-flowered gourd (*Lagenaria siceraria*)	594 µg/kg	> 200 µg/kg (Shoot)	
		White gourd (*Benincasa hispida*)	594 µg/kg	> 200 µg/kg (Shoot)	
		Winter squash (*Cucurbita maxima*)	594 µg/kg	> 900 µg/kg (Shoot)	
		Cucumber (*Cucumis sativus*)	0.004 µg/g	11.42 µg/g (Shoot) (Namiki et al. 2013)	Fruit (Lichtenstein and Schulz 1965; Hilber et al. 2008, 2009; Saito et al. 2011, 2012; Seike et al. 2012) Leaf (Saito et al. 2012) Shoot (Otani et al. 2007; Sakai et al. 2009; Murano et al. 2010; Namiki et al. 2013) Not mentioned (Hashimoto 2005)
		Melon (*Cucumis melo*)	594 µg/kg	> 200 µg/kg (Shoot) (Otani et al. 2007)	Fruit (Hashimoto 2007; Saito et al. 2012) Shoot(Otani et al. 2007)
		Pumpkin (*Cucurbita maxima*)	0.004 µg/g	14.64 µg/g (Shoot)	Fruit (Hashimoto 2007; Saito et al. 2012)

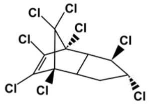
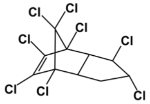
				(Namiki et al. 2013)	Leaf (Saito et al. 2012) Shoot (Otani et al. 2007; Namiki et al. 2013)
		Pumpkin (*Cucurbita moschata*)	0.004 μg/g	15.00 μg/g (Shoot)	Shoot (Namiki et al. 2013)
		Watermelon (*Citrullus lanatus*)	594 μg/kg	> 500 μg/kg (Shoot) (Otani et al. 2007)	Fruit (Saito et al. 2012) Shoot (Otani et al. 2007)
		Zucchini (*Cucurbita pepo*)	12.5 μmol/kg	1.65 μM (Xylem sap) (Fujita et al. 2020a)	Fruit (Hashimoto 2007; Saito et al. 2012) Leaf (Saito et al. 2012) Shoot (Otani et al. 2007; Murano et al. 2010; Namiki et al. 2013, 2015) Xylem sap (Murano et al. 2010; Fujita et al. 2020a, b)
Endrin 	5.2 (Namiki et al. 2013)	Balsam pear (*Momordica charantia*)	58 μg/kg	> 5 μg/kg (Shoot)	Shoot (Otani et al. 2007)
		Figleaf squash (*Cucurbita ficifolia*)	58 μg/kg	> 30 μg/kg (Shoot)	
		Loofah (*Luffa cylindrica*)	58 μg/kg	> 20 μg/kg (Shoot)	
		Melon (*Cucumis melo*)	58 μg/kg	> 10 μg/kg (Shoot)	
		Watermelon (*Citrullus lanatus*)	58 μg/kg	> 50 μg/kg (Shoot)	
		White-flowered gourd (*Lagenaria siceraria*)	58 μg/kg	> 20 μg/kg (Shoot)	
		White gourd (*Benincasa hispida*)	58 μg/kg	> 20 μg/kg (Shoot)	
		Winter squash (*Cucurbita maxima*)	58 μg/kg	> 70 μg/kg (Shoot)	
		Cucumber (*Cucumis sativus*)	0.009 μg/g	9.93 μg/g (Shoot)	Shoot (Otani et al. 2007; Namiki et al. 2013)
		Pumpkin (*Cucurbita maxima*)	0.009 μg/g	13.84 μg/g (Shoot)	
		Zucchini (*Cucurbita pepo*)	0.009 μg/g	14.57 μg/g (Shoot)	

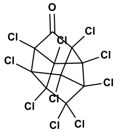
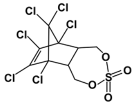
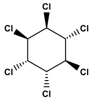
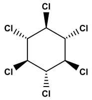
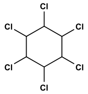
			(Namiki et al. 2013)	
	Pumpkin (*Cucurbita moschata*)	0.009 µg/g	12.28 µg/g (Shoot) (Namiki et al. 2013)	Shoot (Namiki et al. 2013)

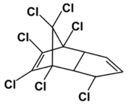
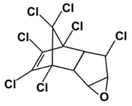
-, not mentioned

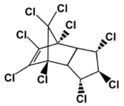
(C) Other POPs and their related organochlorine insecticides.

Organic pollutant	logK_{ow}	Plant species	Concentration		Plant organ
			Soil	Plant	
1-Hydroxychlordene 	3.3 (Hayashi et al. 2018)	Zucchini (*Cucurbita pepo*)	-	-	Shoot (Hayashi et al. 2018)
cis-Chlordane 	6.1 (Shen and Wania 2005)	Cucumber (*Cucumis sativus*)	(−) 670–1,160 ng/g (+) 782–1,406 ng/g	27 ng/g (Leaf) 29 ng/g (Leaf) (Mattina et al. 2002)	Fruit (Mattina et al. 2002, 2004; Hilber et al. 2008) Leaf (Mattina et al. 2002, 2004) Stem (Mattina et al. 2002, 2004) Xylem sap (Mattina et al. 2004)
		Pumpkin (*Cucurbita maxima*)	(−) 670–1,160 ng/g (+) 782–1,406 ng/g	29 ng/g (Leaf) 34 ng/g (Leaf) (Mattina et al. 2002)	Fruit (Mattina et al. 2002) Leaf (Mattina et al. 2002) Stem (Mattina et al. 2002)
		Squash (*Cucurbita pepo*)	-	-	Fruit (Isleyen et al. 2013) Stem (Isleyen et al. 2013)
		Zucchini (*Cucurbita pepo*)	2,440 ng/g	2,940 ng/g (Aerial parts) (Mattina et al. 2006)	Aerial parts (Mattina et al. 2006) Fruit (Mattina et al. 2002, 2004; White et al. 2002; Isleyen et al. 2013) Leaf (Mattina et al. 2002, 2004; White et al. 2002), Peel (White et al. 2002) Stem (Mattina et al. 2002, 2004; White et al. 2002; Isleyen et al. 2013)

					Xylem sap (Mattina et al. 2004, 2006)
trans-Chlordane 	6.22 (Shen and Wania 2005)	Cucumber (*Cucumis sativus*)	(−) 665–1,207 ng/g (+) 557–980 ng/g	24 ng/g (Leaf) 14 ng/g (Leaf) (Mattina et al. 2002)	Fruit (Mattina et al. 2002, 2004) Leaf (Mattina et al. 2002, 2004) Stem (Mattina et al. 2002, 2004) Xylem sap (Mattina et al. 2004)
		Pumpkin (*Cucurbita maxima*)	(−) 665–1,207 ng/g (+) 557–980 ng/g	26 ng/g (Leaf) 23 ng/g (Leaf) (Mattina et al. 2002)	Fruit (Mattina et al. 2002) Leaf (Mattina et al. 2002) Stem (Mattina et al. 2002)
		Squash (*Cucurbita pepo*)	-	-	Fruit (Isleyen et al. 2013) Stem (Isleyen et al. 2013)
		Zucchini (*Cucurbita pepo*)	2,150 ng/g	2,037 ng/g (Aerial parts) (Mattina et al. 2006)	Aerial parts (Mattina et al. 2006) Fruit (Mattina et al. 2002, 2004; White et al. 2002; Isleyen et al. 2013) Leaf (Mattina et al. 2002, 2004; White et al. 2002), Peel (White et al. 2002) Stem (Mattina et al. 2002, 2004; White et al. 2002; Isleyen et al. 2013) Xylem sap (Mattina et al. 2004, 2006)
Chlordane 	6.06 (Rodan et al. 1999)	Cucumber (*Cucumis sativus*)	-	-	Leaf (Mattina et al. 2003)
		Pumpkin (*Cucurbita maxima*)	-	-	
		Zucchini (*Cucurbita pepo*)	0.327 μg/g	0.612 μg/g (Leaf) (Mattina et al. 2000)	Fruit (Mattina et al. 2000) Leaf (Mattina et al. 2000, 2003) Stem (Mattina et al. 2000)

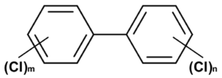
Chlordecone  	4.5 (Clostre et al. 2014)	Christophine (*Sechium edule*)	> 5,000 mg/kg	< 1,000 µg/kg (Fruit)	Fruit (Clostre et al. 2014)
		Cucumber (*Cucumis sativus*)	> 5,000 mg/kg	> 10,000 µg/kg (Fruit)	Fruit (Clostre et al. 2014) Leaf (Clostre et al. 2014) Stem (Clostre et al. 2014)
		Pumpkin (*Cucurbita moschata*)	> 5,000 mg/kg	> 30,000 µg/kg (Fruit)	Fruit (Clostre et al. 2014)
		Zucchini (*Cucurbita pepo*)	-	-	Leaf (Clostre et al. 2014) Stem (Clostre et al. 2014)
Endosulfan  	4.8 (Garvin et al. 2015)	Zucchini (*Cucurbita pepo*)	-	-	Stem (Garvin et al. 2015)
α-HCH  	3.81 (Namiki et al. 2013)	Cucumber (*Cucumis sativus*)	0.008 µg/g	1.87 µg/g (Shoot)	Shoot (Namiki et al. 2013)
		Pumpkin (*Cucurbita maxima*)	0.008 µg/g	2.72 µg/g (Shoot)	
		Pumpkin (*Cucurbita moschata*)	0.008 µg/g	2.49 µg/g (Shoot)	
		Zucchini (*Cucurbita pepo*)	0.008 µg/g	3.30 µg/g (Shoot)	
β-HCH  	3.8 (Namiki et al. 2013)	Cucumber (*Cucumis sativus*)	0.008 µg/g	1.66 µg/g (Shoot)	Shoot (Namiki et al. 2013)
		Pumpkin (*Cucurbita maxima*)	0.008 µg/g	1.98 µg/g (Shoot)	
		Pumpkin (*Cucurbita moschata*)	0.008 µg/g	2.23 µg/g (Shoot)	
		Zucchini (*Cucurbita pepo*)	0.008 µg/g	3.22 µg/g (Shoot) (Namiki et al. 2013)	Shoot (Namiki et al. 2013, 2015)
γ-HCH  	3.7 (Namiki et al. 2013)	Cucumber (*Cucumis sativus*)	0.009 µg/g	0.56 µg/g (Shoot)	Shoot (Namiki et al. 2013)
		Pumpkin (*Cucurbita maxima*)	0.009 µg/g	0.86 µg/g (Shoot)	
		Pumpkin (*Cucurbita moschata*)	0.009 µg/g	0.76 µg/g (Shoot)	

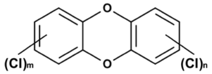
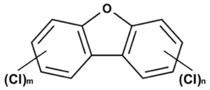
		Zucchini (*Cucurbita pepo*)	0.009 µg/g	1.00 µg/g (Shoot)	
Heptachlor 	4.62 (Hayashi et al. 2018)	Cucumber (*Cucumis sativus*)	19.8 mg/l (Hydroponics)	> 0.05 µg/g (Shoot) (Hayashi et al. 2010)	Fruit (Lichtenstein and Schulz 1965) Shoot (Hayashi et al. 2010)
		Pumpkin (*Cucurbita maxima*)	19.8 mg/l (Hydroponics)	0.014 µg/g (Shoot) (Hayashi et al. 2010)	Shoot (Hayashi et al. 2010)
		Zucchini (*Cucurbita pepo*)	19.8 mg/l (Hydroponics)	0.091 µg/g (Shoot) (Hayashi et al. 2010)	Shoot (Hayashi et al. 2010, 2018)
HEPX 	5 (Namiki et al. 2013)	Cucumber (*Cucumis sativus*)	0.001 µg/g	10.06 µg/g (Shoot) (Namiki et al. 2013)	Fruit (Lichtenstein and Schulz 1965; Hilber et al. 2008) Shoot (Namiki et al. 2013) Xylem sap (Mattina et al. 2004)
		Pumpkin (*Cucurbita maxima*)	0.001µg/g	13.29 µg/g (Shoot) (Namiki et al. 2013)	Shoot (Murano et al. 2009; Namiki et al. 2013)
		Pumpkin (*Cucurbita moschata*)	0.001µg/g	12.99 µg/g	Shoot (Namiki et al. 2013)
		Squash (*Cucurbita pepo*)	57.5 µg/kg	0.550 mg/kg (Shoot) (Sugiyama et al. 2013)	Fruit (Sugiyama et al. 2013) Shoot (Sugiyama et al. 2013, 2016)
		White-flowered gourd (*Lagenaria siceraria*)	> 0.14µg/g	> 1 µg/g	Vine (Campbell et al. 2009)
		Zucchini (*Cucurbita pepo*)	0.001µg/g	14.44 µg/g (Shoot) (Namiki et al. 2013)	Aerial parts (Mattina et al. 2006) Shoot (Namiki et al. 2013; Hayashi et al. 2018) Xylem sap (Mattina et al. 2004, 2006)
	6.35 (Maruya et al. 2009)	Cucumber (*Cucumis sativus*)	638–1,175 ng/g	23 ng/g (Leaf) (Mattina et al. 2002)	Fruit (Mattina et al. 2002) Leaf (Mattina et al. 2002) Stem (Mattina et al. 2002)

trans-Nonachlor 				Xylem sap (Mattina et al. 2004)
	Pumpkin (*Cucurbita maxima*)	638–1,175 ng/g	18 ng/g (Leaf)	Fruit (Mattina et al. 2002) Leaf (Mattina et al. 2002) Stem (Mattina et al. 2002)
	Squash (*Cucurbita pepo*)	-	-	Fruit (Isleyen et al. 2013) Stem (Isleyen et al. 2013)
	Zucchini (*Cucurbita pepo*)	1,080 ng/g	515 ng/g (Aerial parts) (Mattina et al. 2006)	Aerial parts (Mattina et al. 2006) Fruit (Mattina et al. 2002, 2004; White et al. 2002; Isleyen et al. 2013) Leaf (Mattina et al. 2002, 2004; White et al. 2002) Peel (White et al. 2002) Stem (Mattina et al. 2002, 2004; White et al. 2002; Isleyen et al. 2013) Xylem sap (Mattina et al. 2004, 2006)

HCH, hexachlorocyclohexane; HEPX, Heptachlor *exo*-epoxide; -, not mentioned

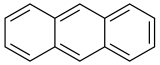
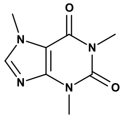
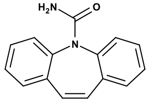
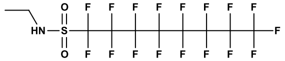
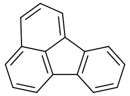
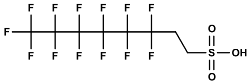
(D) PCBs, PCDDs, and PCDFs

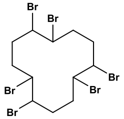
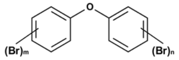
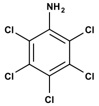
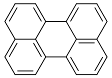
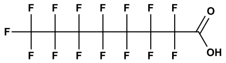
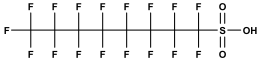
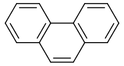
Organic pollutant	logK_{ow}	Plant species	Concentration		Plant organ
			Soil	Plant	
PCBs 	4.09-8.18 (Hawker and Connell 1988)	Cucumber (*Cucumis sativus*)	105 µg/g	> 5 µg/g (Leaf)	Fruit (White et al. 2006) Leaf (White et al. 2006) Stem (White et al. 2006)
		Pumpkin (*Cucurbita maxima*)	105 µg/g	> 5 µg/g (Leaf) (White et al. 2006)	Leaf (Whitfield Åslund et al. 2007) Leaf and petiole (Low et al. 2011) Petiole (Whitfield Åslund et al. 2007) Shoot (Ficko et al. 2011; Greenwood et al. 2011; Denyes et al. 2012) Stem (Whitfield Åslund et al. 2007, 2008; Low et al. 2011) Xylem sap (Greenwood et al. 2011)
		Zucchini (*Cucurbita pepo*)	5,100 ng - TEQ/kg	> 40 pg-TEQ/g (Aerial parts) (Inui et al. 2008)	Aerial parts (Inui et al. 2008, 2011; Matsuo et al. 2011) Fruit (White et al. 2006) Leaf (White et al. 2006; Goto et al. 2019) Stem (White et al. 2006; Goto et al. 2019) Xylem sap (Goto et al. 2019)
PCDD/Fs	3.68-8.75 (Govers et al. 1996)	Cucumber (*Cucumis sativus*)	148 ng - TEQ/kg	21.0 ng - TEQ/kg (Fruit) (Hülster et al. 1994)	Fruit (Hülster et al. 1994) Leaf (Hülster et al. 1994)

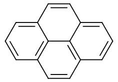
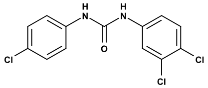
 				Shoot (Zhang et al. 2009)
	Pumpkin (*Cucurbita maxima*)	148 ng - TEQ/kg	3.1 ng - TEQ/kg (Fruit) (Hülster et al. 1994)	Fruit (Hülster et al. 1994) Leaf (Hülster et al. 1994) Shoot (Zhang et al. 2009)
	Zucchini (*Cucurbita pepo*)	5,100 ng - TEQ/kg	> 40 pg-TEQ/g (Aerial parts) (Inui et al. 2008)	Aerial parts (Inui et al. 2008, 2011; Matsuo et al. 2011) Fruit (Hülster et al. 1994) Leaf (Hülster et al. 1994) Shoot (Zhang et al. 2009)

PCBs, polychlorinated biphenyls; PCDDs, polychlorinated dibenzo-*p*-dioxins; PCDFs, polychlorinated dibenzofurans; TEQ, toxic equivalent

(E) Others

Organic pollutant	$\log K_{ow}$	Plant specie	Concentration		Plant organ
			Soil	Plant	
Anthracene 	4.54 (Miller et al. 1985)	Zucchini (*Cucurbita pepo*)	178 ng/g	15.0 ng/g (Aerial parts)	Aerial parts (Mattina et al. 2006) Xylem sap (Mattina et al. 2006)
Caffeine 	−0.07 (Garvin et al. 2015)	Zucchini (*Cucurbita pepo*)	-	-	Shoot (Garvin et al. 2015)
Carbamazepine 	2.45 (Shenker et al. 2011)	Cucumber (*Cucumis sativus*)	13.98 μg/L	25.6 μg/kg (Fruit)	Fruit (Shenker et al. 2011) Leaf (Shenker et al. 2011) Stem (Shenker et al. 2011)
		Zucchini (*Cucurbita pepo*)	20 mg/kg	> 80 mg/kg	Leaf (Knight et al. 2018)
N-EtFOSA 	6.71 (Zhao et al. 2018)	Pumpkin (*Cucurbita maxima*)	-	-	Shoot (Zhao et al. 2018)
Fluoranthene 	5.22 (Miller et al. 1985)	Zucchini (*Cucurbita pepo*)	3,970 ng/g	161 ng/g (Aerial parts)	Aerial parts (Mattina et al. 2006) Xylem sap (Mattina et al. 2006)
6:2 FTSA 	4.44 (Zhao et al. 2019)	Pumpkin (*Cucurbita maxima*)	-	-	Shoot (Zhao et al. 2019)

HBCD 	4.78 (Hou et al. 2017)	Pumpkin (*Cucurbita maxima*)	100 ng/mL (Hydroponics)	> 3 ng/g (Leaf)	Leaf (Hou et al. 2017) Stem (Hou et al. 2017)
PBDEs 	4.31-8.35 (Li et al. 2008)	Zucchini (*Cucurbita pepo*)	75 µg/kg	> 4 µg/kg	Shoot (Mueller et al. 2006)
Pentachloroaniline 	5.08 (de Wolf et al. 1994)	Cucumber (*Cucumis sativus*)	0.2 mg/kg	< 0.01 mg/kg	Fruit (Hilber et al. 2008)
Perylene 	6.5 (Miller et al. 1985)	Zucchini (*Cucurbita pepo*)	1.25 mmol/kg	3.81 nM (Xylem sap) (Fujita et al. 2020a)	Stem (Yoshihara et al. 2014) Xylem sap (Fujita et al. 2020a)
PFOA 	5.30 (Zhao et al. 2019)	Cucumber (*Cucumis sativus*)	805 µg/kg	23.8 µg/kg (Peeled edible parts)	Peeled edible parts (Lechner and Knapp 2011) Peel (Lechner and Knapp 2011)
PFOS 	6.43 (Zhao et al. 2019)	Cucumber (*Cucumis sativus*)	556 µg/kg	1.3 µg/kg (Peeled edible parts)	Peeled edible parts (Lechner and Knapp 2011) Peel (Lechner and Knapp 2011)
Phenanthrene 	4.6 (Miller et al. 1985)	Zucchini (*Cucurbita pepo*)	1,661 ng/g	193 ng/g (Aerial parts)	Aerial parts (Mattina et al. 2006) Xylem sap (Mattina et al. 2006)

Pyrene 	5.2 (Miller et al. 1985)	Zucchini (*Cucurbita pepo*)	1.25 mmol/kg	0.74 µM (Xylem sap) (Fujita et al. 2020a)	Aerial parts (Mattina et al. 2006) Xylem sap (Mattina et al. 2006; Fujita et al. 2020a, b; Inui et al. 2020)
Triclocarban 	3.5 (Snyder et al. 2010)	Zucchini (*Cucurbita pepo*)	-	-	Stem (Garvin et al. 2015)
(a)PAHs	-	Cucumber (*Cucumis sativus*)	36,300 ng/g	0.124 µg (Leaf)	Leaf (Parrish et al. 2006)
		Squash (*Cucurbita pepo*)	36,300 ng/g	0.631 µg (Leaf)	Stem (Parrish et al. 2006)
		Zucchini (*Cucurbita pepo*)	36,300 ng/g	1.13 µg (Leaf)	
PFAAs	-	(b)Pumpkin (not mentioned)	-	-	Flower (Lee et al. 2014) Fruit (Lee et al. 2014) Leaf (Lee et al. 2014) Stalk (Lee et al. 2014)
		(c)Zucchini (*Cucurbita pepo*)	-	-	Edible parts (Felizeter et al. 2014) Leaf (Felizeter et al. 2014) Stem (Felizeter et al. 2014)
PPCPs	-	Cucumber (*Cucumis sativus*)	-	-	(d)Leaf/stem (Wu et al. 2013) (e)Shoot (Sun et al. 2018)

N-EtFOSA, *N*-ethyl perfluorooctane sulfonamide; 6:2 FTSA, 6:2 fluorotelomer sulfonic acid; HBCD, hexabromocyclododecane; PBDEs, polybrominated diphenyl ethers; PFOA, perfluorooctanoic acid PFOS, perfluorooctanesulfonic acid; PAHs, polycyclic aromatic hydrocarbons; PFAAs, perfluoroalkyl acids; PPCPs, pharmaceutical and personal care products; -, not mentioned (a)PAHs include phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenz[a,h]anthracene, indeno[1,2,3-cd]pyrene, and benzo[g,h,i]perylene

(b)4:2 diPAP (polyfluoroalkyl phosphate diester), 6:2 diPAP, 8:2 diPAP, perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), and perfluoroundecanoic acid (PFUnA) were detected in flowers, fruits, leaves, and stalks. 6:2/8:2 diPAP was detected in flowers, fruits, and stalks. 10:2 diPAP was detected in fruits and stalks. Perfluoropentanoic acid (PFPeA) was detected in flowers, fruits, and leaves.

(c)PFBA (perfluorobutanoic acid), PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA (perfluoroundecanoic acid), PFDoA (perfluorododecanoic acid), PFTrA (perfluorotridecanoic acid), PFTeA (perfluorotetradecanoic acid), PFBS (perfluorobutane sulfonic acid), PFHxS (perfluorohexane sulfonic acid), and PFOS were detected.

(d)Caffeine, meprobamate, primidone, sulfamethoxazole, atenolol, trimethoprim, DEET, carbamazepine, dilantin, diuron, naproxen, diazepam, fluoxetine, atorvastatin, ibuprofen, gemfibrozil, triclosan, and triclocarban were detected.

(e)Caffeine, meprobamate, primidone, sulfamethoxazole, atenolol, trimethoprim, carbamazepine, dilantin, diazepam, atorvastatin, naproxen, ibuprofen, gemfibrozil, triclosan, diclofenac, and triclocarban were detected.