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Absorption and translocation of polybrominated diphenyl ethers (PBDEs) by plants from contaminated sewage sludge

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ABSTRACT

Polybrominated diphenyl ethers (PBDEs) are used as additive flame retardants. PBDEs are persistent, bioaccumulative and toxic compounds. They are often detected in sewage sludge which is applied on agricultural soils as fertilizer. The objective of this study was to find out whether plants are able to accumulate and translocate PBDEs. Tobacco (*Nicotiana tabacum*) and nightshade (*Solanum nigrum*) were planted in pots containing contaminated sewage sludge and uncontaminated substrate. After 6 months of plant cultivation in sewage sludge up to 15.4 ng g^{-1} dw and 76.6 ng g^{-1} dw of PBDE congeners – BDE 47, BDE 99 and BDE 100 were accumulated in the nightshade and tobacco tissue, respectively. Corresponding values in plants vegetated in the control garden substrate were 10 times lower. The bioconcentration factors (BCFs) of accumulated congeners were calculated. Tobacco exhibited higher BCFs values and for both plants BCFs values of BDE 47, BDE 99, BDE 100 and BDE 209 negatively correlated with their octanol–water partition coefficients (log K_{ow}). The exception was decaBDE (BDE 209) which was accumulated only in tobacco tissue in the concentration of 116.8 ng g^{-1} dw. The majority of PBDEs was detected in aboveground plant biomass indicating that both plants have the ability to translocate PBDEs. To our knowledge this is one of the first studies reporting the accumulation of both lower PBDEs and BDE 209 in plants. Our results suggest that absorption, accumulation and translocation of PBDEs by plants and their transfer to the food chain could represent another possible risk for human exposure.

1. Introduction

Polybrominated diphenyl ethers (PBDEs) have been widely used as flame retardants in many countries throughout the world for more than 30 years, although they are known to present health and environmental risks. They are lipophilic with bioaccumulative properties (Rahman et al., 2001) and despite their low acute toxicity the low-brominated congeners act as endocrine disruptors, carcinogens and/or neurodevelopment toxicants (Hardy, 2002; McDonald, 2002). Three technical mixtures of PBDEs are industrially manufactured: Deca-, Octa- and PentaBDE. PBDEs are used as additive flame retardants in plastics products, electrical components, textiles, building materials and other products (Rahman et al., 2001; McDonald, 2002). They enter the environment more easily than the reactive BFRs and therefore entrance into the food chain is facilitated (Alaee and Wennig, 2002; McDonald, 2002). The main sources of PBDEs in the environment are effluents from factories producing BFRs and flame-retardant polymers, flame-resistant products and the waste that contains PBDEs. PBDEs are released into the environment in various of forms: associated with particles, by leaching, and by volatilization from flame-resistant products during their use and waste disposal such during incineration of municipal waste (Watanabe and Sakai, 2003; Hites, 2004).

PBDEs have been found in both abiotic samples and biota. These compounds have been detected in the air, sewage sludge, sediments, soils, water, aquatic organisms (marine mammals – e.g. whales, dolphins, seals and fishes), birds that feed on fish (e.g. ospreys, cormorants and gulls) (Hites, 2004; Hajšlová et al., 2007; Knoth et al., 2007) and also birds of prey family (kestrel, sparrow hawk and owl) (Chen et al., 2007). Studies have also confirmed the presence of PBDEs in human blood, adipose tissue and breast milk. There is proof that PBDEs concentrations are rising in human tissues and in biota (Hites, 2004; Pulkrabová et al., 2009). The primary route of human exposure to PBDEs is *via* ingestion of food with the high content of fat (e.g. fatty fish, meat and diary products) (Darnerud et al., 2001; Vonderheide et al., 2008; Frederiksen et al., 2009). The other significant source of PBDEs in human tissues

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is inhalation of compounds in the gas phase or dust particles and direct dermal exposure to flame-resistant product (Hites, 2004; Frederiksen et al., 2009). Inhalation of house dust presents unwanted source of PBDEs during all life stages (Jones-Otazo et al., 2005; Allen et al., 2008).

Due to the hydrophobic character of PBDEs, these chemicals are strongly bound to solid particles such as soil, sediments and sewage sludge (Hites, 2004; Hale et al., 2006). With regards to abiotic samples, such as atmosphere and aqueous media, the occurrence of pentaBDE congeners is higher than decaBDE due to their physico-chemical properties (Vonderheide et al., 2008).

In the biotic samples low-brominated congeners (BDE 47, BDE 99, BDE 100, BDE 153 and BDE 154) appear to be the most predominant congeners because of their high bioavailability (de Wit, 2002; Watanabe and Sakai, 2003; Mariottini et al., 2008). This predominance could be caused by their higher potential for bioaccumulation and according the study Wan et al. (2008) the predominance of BDE 47 in high trophic level animals could be also contributed by the debromination of higher brominated PBDEs and its relatively high biomagnification potential in food webs.

Some studies suggest that PBDEs concentrate at different trophic levels within aquatic ecosystems and have a biomagnification potential in the food chain (Hajšlová et al., 2007). In contrast, much less is known about terrestrial systems and there are only two reports describing the plant uptake of PBDEs from soil and their translocation to above-ground part of plant. Mueller et al. (2006) showed that plants such as radish and zucchini have the ability to take up and accumulate pentaBDEs from contaminated soil. Huang et al. (2010) described the uptake, translocation and metabolism of BDE 209 in six plant species. Plants could therefore play an important role for the transfer of PBDEs into the food chain. Pirard and De Pauw (2007) found that PBDEs can be accumulated in the liver and the abdominal fat of chickens, and consequently they can be transferred to eggs. Kierkegaard et al. (2007) have described that there is an accumulation of BDE 209 in the body fat and meat of cows and its further metabolic debromination leads to formation of low-brominated congeners nonaBDE (BDE 207), octaBDEs (BDE 196 and BDE 197) and heptaBDE (BDE 182). People may be thus exposed to PBDEs by consuming of herbivore meat.

The main source of PBDEs in soil is application of enormous volumes of contaminated sewage sludge to agricultural areas (Law et al., 2006; Eljarrat et al., 2008; Vonderheide et al., 2008; US EPA, 2009). Based on the median concentrations of PBDEs analyzed in 15 sewage sludge samples in the Czech Republic in 2006 it was calculated that 29.2 kg of Penta- and OctaBDE mixtures and 67.6 kg of DecaBDE mixture were land-applied per year (Stiborová et al., 2009). A similar study shows that in Germany 150 kg per year of Penta- and OctaBDE mixtures and 350 kg per year of DecaBDE were released into the environment *via* sewage sludge land-application in 2001 (Knoth et al., 2007). There are no guidelines for the content of organic pollutants such as PBDEs in sewage sludge used in agriculture (Eljarrat et al., 2008).

The aim of the present study was to investigate the uptake of PBDEs from sewage sludge into plants and their translocation to above-ground tissues. The application of sewage sludge in the fields used for the growth of the plants for human consumption represent a worst case scenario and many more studies are needed to display enough evidence to ban that on European level. We have chosen *Nicotiana tabacum* and *Solanum nigrum* for our experiment because these plants are used as the model plants in many studies (Evangelou et al., 2007; Gichner et al., 2007; Rezek et al., 2007; Wei et al., 2010) and for their known ability to accumulate PCBs (Mackova et al., 2009) which are structurally similar to PBDEs.

2. Materials and methods

2.1. Chemicals

The following set of standard solutions containing PBDE congeners (concentration 50 μg mL^{-1} in nonane) were obtained from Wellington Laboratories, Canada: 2,4,4′-triBDE (BDE 28); 3,4,4′-triBDE (BDE 37); 2,2′,4,4′-tetraBDE (BDE 47); 2,2′,4,5′-tetraBDE (BDE 49); 2,3′,4,4′-tetraBDE (BDE 66); 2,2′,3,4,4′-pentaBDE (BDE 85); 2,2′,4,4′,5-pentaBDE (BDE 99); 2,2′,4,4′,6-pentaBDE (BDE 100); 2,2′,4,4′,5,5′-hexaBDE (BDE 153); 2,2,4′,4,5′,6′-hexaBDE (BDE 154); 2 2,2′,3,4,4′,5′,6-heptaBDE (BDE 183), decaBDE (BDE 209) and ^{13}C BDE 209. Standard solution of PCB 112 (10 μg mL^{-1} in isooctane) was purchased from Gr. Ehrenstorfer GmSH (Germany). The organic solvents (cyclohexane, dichloromethane, hexane, ethylacetate and isooctane) declared for organic trace analyses grade were all supplied by Merck (Germany). Anhydrous sodium sulphate supplied by Penta Chrudim (Chrudim, Czech Republic) was heated at 600 °C for 5 h and then stored in desiccator before use. Styrene–divinylbenzene gel (Bio Beads S-X3, 200–400 mesh) was purchased from Biorad Laboratories (USA).

2.2. Analytical methods

Extraction of PBDEs was performed by Soxhlet extraction using dichloromethane as solvent for sewage sludge and a mixture of hexane:dichloromethane (1:1, *v/v*) for plants. The crude extract was carefully evaporated and the sample dissolved in solvent mixture cyclohexane–ethylacetate (1:1, *v/v*) that was used as a mobile phase in gel permeation chromatography (GPC) employing Bio Beads S-X3 column for separation of interfering co-extracts.

An Agilent 6890 (Agilent, USA) gas chromatograph equipped with a single quadrupole mass analyzer Agilent 5975 XL operated in negative chemical ionization mode (GC/MS-NCI) and DB-XLB capillary was employed for routine analyses of PBDEs in purified extracts. The GC conditions were as follows (for all analyzed PBDEs with exception of BDE 209): capillary column DB-XLB column (30 m × 0.25 mm i.d. × 0.1 μm film thickness, J & W Scientific, Folsom, USA), column temperature program: from 105 °C (hold 2 min) to 300 °C at 20 °C min^{-1} (hold 5 min); carrier gas: helium (Siad, Czech Republic) with constant flow 1.5 mL min^{-1}; injection temperature: 275 °C; injection volume: 1 μL using pulsed splitless injection mode (splitless time: 2 min). The mass selective detector with quadrupole analyzer was operated in a selective ion-monitoring mode (SIM) in a negative chemical ionization (NCI). Monitored ions (*m/z*) were 79, 81, 159 and 161. Ion *m/z* 79 was used for quantification of all target analytes. Methane, which was used as a reagent gas (purity 99.995%, Siad, Czech Republic), was set at a pressure 2×10^{-4} mbar. Ion source temperature was 150 °C and quadrupole temperature 105 °C.

The presence of BDE 209 was monitored using the same GC/MS-NCI employing a shorter capillary column BD-XLB (15 m × 0.25 mm i.d. × 0.1 μm film thickness J & W Scientific, USA). The temperature program was the following: from 80 °C (hold 2 min) to 280 °C at 20 °C min^{-1} and to 320 °C at 5 °C min^{-1} (hold 5 min); carrier gas: helium with constant flow 3 mL min^{-1}; injection temperature: 285 °C; injection volume: 1 μL using pulsed splitless injection mode (splitless time: 2 min). Monitored ions (*m/z*) were 485 and 487 for BDE 209, 495 and 497 for ^{13}C labelled BDE 209 (Stiborová et al., 2008).

2.3. Quality assurance/quality control

Analysis was carried out in an accredited testing laboratory (No. 1316.2) in the Czech Republic (current standard: EN ISO/IEC

17025). The scope of accreditation covered the analyses of various halogenated POPs including BFRs in environmental, human and food samples.

Together with each extraction batch (consisting from five samples), one procedure blank was processed. The results were corrected for blank interferences and for recovery (PCB 112 was added as surrogate before GPC clean up). For recovery testing of overall analytical method, sewage sludge/plants were spiked at level 10 ng g^{-1} (of each analyte). Real-life samples were also analyzed, to get background levels of analytes. PBDEs recoveries in sewage sludge and plants ranged between 89–105% and 81–107%, respectively. Precision of analytical method (repeatability) was also determined from analyses of six samples of spiked sewage sludge/plant; it was in range 4–12% (expressed as relative standard deviation). Limits of quantification (LOQs) of overall analytical method were in range 0.1–1.2 ng g^{-1} dw for sewage sludge and 0.05–0.5 ng g^{-1} dw for plants.

The trueness of generated data was controlled by the simultaneous analysis of certified reference materials Heavily Contaminated Sediment (EDF-5184) from CIL, USA.

We wish to emphasize that the method we use is fully validated. To document the trueness of generated data, the laboratory participates every year in several inter-laboratory studies organized by the Institute for Reference Measurements and Materials (IRRM, Geel, Belgium) and/or in Food Analysis Performance Assessment Scheme (FAPAS®) coordinated by the Food and Environment Research Agency (CSL, York, UK).

2.4. Growth of plants

Seedlings of *N. tabacum* and *S. nigrum* were obtained from the Institute of Organic Chemistry and Biochemistry of the Academy of Sciences of the Czech Republic, v.v.i. (IOCB ASCR, v.v.i.). Due to better homogeneity and the differences between the longer industrially contaminant burden and artificially contaminated substrate we have decided to use industrially contaminated sewage sludge which was not subsequently mixed with uncontaminated soil to prevent dilution of PBDEs. The contaminated sewage sludge was obtained from WWTP (wastewater treatment plant) in Hradec Králové, Czech Republic. (The amount of cleaned wastewater: 16 million m^3 in 2008; the total length of sewerage net: 496 km; the number of sewer connection: 16 775.)

Five different treatments were established. Five pots were filled with sewage sludge and were left unplanted; five pots were filled with sewage sludge and were planted with seedlings of *N. tabacum*; five pots were filled with sewage sludge and were planted with seedlings of *S. nigrum*. Six pots using a control substrate for garden plants (composition: peat, high-quality ripe bark humus, treated pH, enriched by the nutrients sufficient for 6 weeks. AGRO CS a.s., Česká Skalice, CZ; no PBDEs detected) were planted with seedlings of *N. tabacum* (three pots) or *S. nigrum* (three pots). The volume of plastic pots was 200 mL and they were lined with aluminium foil to prevent sorption of PBDEs from sewage sludge on pots and to prevent outflow of water from pots. Pots were maintained at 25 °C and were regularly watered for 6 months. The plants in the control substrate were fertilized periodically after 6 weeks. Dry leaves were removed from the plant and stored at −20 °C till the end of the experiment.

2.5. Harvesting

Plants were harvested after 6 months of growth. They were gently removed from the substrate and separated into root, stem, leaf and fruit fractions. Dry fractions (drying at 50 °C for 48–72 h) from the same treatment pots were used to calculate average mass values of root, stem, leaf and fruit biomass and then the PBDEs were extracted for GC/MS-NCI analysis.

The sludge and control substrates from all pots of the same treatment were mixed thoroughly and stored at 4 °C before PBDEs extraction and GC/MS-NCI analysis.

3. Results and discussion

3.1. Physiology of plants

In our experiment the growth and physiology of model plants were influenced only by the substrate qualities such as contaminated sludge and the control substrate. Dry fractions of plant material from the same treatment pots were used to calculate the average weight of various plant tissue classes, namely roots, stems, leaves and fruits. At the end of the experiment the weight of plant shoot biomass cultivated in the sewage sludge was higher than that of plants cultivated in control substrate without PBDEs (in the case of tobacco the value increased by 4-fold) (Table 1). Plants also exhibited visually better growth. These observations correspond with the fact that the sewage sludge is used for fertilization of agricultural and forest soil, for recultivation or for production of compost because it contains a high proportion of organic matter and it is rich in organic and inorganic nutrients. Its application improves the physico-chemical and biological properties of soil (Singh and Agrawal, 2008). Our experiments confirmed that use of sewage sludge as a growth substrate resulted in more robust plants with higher amount of biomass.

3.2. PBDEs recovery and accumulation by plants

BDE 209 was the predominant congener (400.3 ng g^{-1}) in the original sewage sludge, followed by lower brominated congeners (334.4 ng g^{-1}) – BDE 47 (139.4 ng g^{-1}), BDE 99 (166.3 ng g^{-1}) and BDE 100 (28.7 ng g^{-1}). The sum of other congeners – BDE 28 (1.1 ng g^{-1}) BDE 49 (9.9 ng g^{-1}), BDE 66 (3.3 ng g^{-1}), BDE 85 (6.6 ng g^{-1}), BDE 153 (9.1 ng g^{-1}), BDE 154 (8.9 ng g^{-1}) and BDE 183 (3.2 ng g^{-1}) – was 42.1 ng g^{-1}. The recovery of total PBDEs concentration did not significantly change after 6 months of cultivation in any of our three experimental systems (both in the unplanted sewage sludge and in the sewage sludge planted with tobacco or nightshade) (Fig. 1).

There are several reports showing that plants enhance the degradation of POPs, for example PCBs (Rezek et al., 2007) and DDT (Suresh et al., 2005), but we have not detected any significant decrease in any of PBDEs congener concentrations. These results are not surprising due to their persistence and slow degradation rates (Gerecke et al., 2006; Tokarz et al., 2008).

Organic, as well as inorganic pollutants can be accumulated by plants from contaminated soil (Cunningham and Berti, 1993). Their accumulation and magnification are determined by their physico-chemical properties (molecular weight, water solubility, vapor pressure and octanol–water partition coefficient (K_{ow})) and also by environmental conditions (e.g. temperature, humidity, soil properties) (Paterson and Mackay, 1994; Alkorta and Garbisu, 2001). Our results demonstrate that plants have the ability to take up, transfer and accumulate some of PBDEs (Table 2). The accumulation of BDE 47, BDE 99 and BDE 100 in nightshade and tobacco was up to 15.4 and 76.6 ng g^{-1} dw plant tissue respectively, whereas the accumulation of BDE 47, BDE 99 and BDE 100 in zucchini and radish published by Mueller et al. (2006) was in the range of <5 ng g^{-1} dw plant tissues. The difference could be caused not only by different plant species but also by the higher initial concentration of these PBDEs in the sludge in our experiment (334.4 ng g^{-1} dw of BDE 47, BDE 99 and BDE 100 compared to

Table 1
Comparison of plants biomass grown in pots with contaminated sludge, and control plants grown in pots with control substrate.

	Biomass (g)	*A*: biomass of plant grown in sludge[a] (g)	*B*: biomass of plant grown in control substrate[a] (g)	Rate *A*/*B*
Nightshade[b,c]	Total	1.75	1.10	1.59
	Root	0.02 ± 0.00	0.09 ± 0.01	0.22
	Stem	0.23 ± 0.02	0.17 ± 0.01	1.35
	Leaf	0.57 ± 0.04	0.17 ± 0.01	3.35
	Fruits	0.93 ± 0.03	0.67 ± 0.02	1.39
Tobacco[d,e]	Total	3.79	1.08	3.51
	Root	0.17 ± 0.02	0.29 ± 0.03	0.59
	Stem	0.59 ± 0.03	0.12 ± 0.01	4.92
	Leaf	3.03 ± 0.07	0.67 ± 0.02	4.52

n: number of plant replicates grown in the same substrate.
[a] Values are mean ± standard deviation.
[b] $n = 4$ for nightshade grown in the sewage sludge (one plant died at the beginning of our experiment).
[c] $n = 3$ for nightshade grown in the control substrate.
[d] $n = 5$ for tobacco grown in the sewage sludge.
[e] $n = 3$ for tobacco grown in the control substrate.

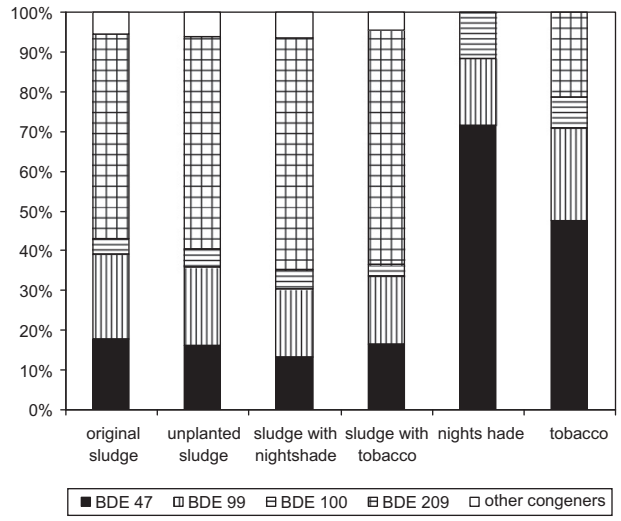

Fig. 1. Percentage of the major congeners in the sludge and plants after 6 months (including BDE 209).

75 ng g^{-1} dw of commercial PentaBDE mixture) and longer time of cultivation in our set up (6 months compared to 10 weeks).

Extremely hydrophobic compounds ($\log K_{ow} > 6$) are less absorbed from soil and transported within plant compared to hydrophilic ones (Cunningham and Berti, 1993). However several studies have described the accumulation and translocation of such compounds (Hulster et al., 1994; Inui et al., 2008). Our study shows that PBDEs were absorbed by plants directly from contaminated substrate compared to the controls. The PBDE levels detected in controls grown in PBDE-free substrate were approximately 10 times lower than those detected in plants grown in contaminated substrate (Table 2). Due to the propensity of PBDEs to accumulate in dust (Allen et al., 2008; Vonderheide et al., 2008,), the low amount of PBDEs detected in controls probably arose from this source.

In our experiments the concentration of individual congeners (BDE 47, BDE 99 and BDE 100) accumulated in tobacco is nearly 10 times higher than in nightshade (Table 2). The level of PBDEs uptake can thus be interpreted as plant species dependent. The mechanisms controlling the accumulation in plant species are still unresolved, however it is known that the uptake of xenobiotics varies among plants (White, 2002) even among different subspecies of zucchini plants (Inui et al., 2008).

3.3. Translocation of PBDEs, bioconcentration factors (BCFs) and correlation between BCFs and octanol–water partition coefficients ($\log K_{ow}$)

Our results demonstrate that plants have the ability not only to take up xenobiotics to roots but they also translocate PBDEs to above-ground parts of plants. The PBDEs were detected in shoots and also in fruits in the case of nightshade (Table 3). Nightshade accumulated less PBDEs and also the degree of translocation was lower compared to tobacco. In both plants the shoot systems be-

Table 2
The total concentration of the major PBDE congeners in plant tissue [ng g^{-1} dw].

Plant	Part of the plant	BDE 47		BDE 99		BDE 100 [ng g^{-1} dw]		BDE 209		ΣPBDE	
		Sludge	Control	Sludge	Control	Sludge	Control	Sludge	Control	Sludge	Control
Nightshade	Root	10.4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	10.4	n.d.
	Stem	13.2	0.4	0.7	0.3	1.5	n.d.	n.d.	n.d.	15.4	0.7
	Leaf	1.1	0.4	1.0	0.2	0.4	0.1	n.d.	n.d.	2.5	0.7
	Fruit	2.1	2.4	0.7	n.d.	1.4	n.d.	n.d.	n.d.	4.2	2.4
	Total[a]	3.4	1.6	0.8	0.1	0.6	0.0	n.d.	n.d.	4.8	1.7
Tobacco	Root	0.5	1.6	n.d.	2.0	n.d.	n.d.	n.d.	n.d.	0.5	3.6
	Stem	61.6	2.1	10.5	n.d.	4.5	n.d.	116.8	n.d.	76.6	2.1
	Leaf	38.5	6.3	22.6	1.8	7.3	0.8	n.d.	n.d.	68.4	8.9
	Total[a]	40.6	5.4	19.7	1.5	6.5	0.1	18.3	n.d.	66.8	7.0

n.d.: not detected (it means zero value).
ΣPBDE: sum of lower brominated congeners BDE 47, BDE 99 and BDE 100.
[a] Total BDE congener plant concentration were calculated as the sum of the amount of BDE congener in individual plant parts divided by total amount of dry plant biomass.

Table 3
Total mass of PBDE congeners in sewage sludge and plants (ng).

		BDE47	BDE99	BDE100 (ng)[a]	BDE209
Original SS		12052.8	14372.6	2479.5	34603.8
SS with nightshade		10492.2	13391.6	3838.3	45402.3
Nightshade	Root	0.2	n.d.	n.d.	n.d.
	Stem	3.0	0.2	0.4	n.d.
	Leaves	0.6	0.6	0.2	n.d.
	Fruits	2.0	0.6	0.3	n.d.
	Plant	5.8	1.3	0.9	n.d.
SS with tobacco		12984.9	13468.7	2361.3	46259.6
Tobacco	Root	0.1	n.d.	n.d.	n.d.
	Stem	10.5	0.8	0.8	19.9
	Leaves	6.6	1.2	1.2	n.d.
	Plant	17.1	5.6	2.0	19.9

SS: sewage sludge.
n.d.: not detected (it means zero value).
In case of PBDE congeners 28, 49, 66, 85, 153, 154, 183 no accumulation was detected.
[a] Mass of PBDE congeners represents the whole content of PBDE in pot, eventually in the individual plant parts.

came the major location of PBDEs. Nightshade contained about 61% and tobacco 92% of total PBDEs in above-ground biomass (stems, leaves). In both plants PBDEs only congeners BDE 47, BDE 99, BDE 100 and BDE 209 were accumulated. In case of PBDE congeners 28, 49, 66, 85, 153, 154, 183 no accumulation was detected. Similar results were published by Mueller et al. (2006) where the pentaBDEs concentrations were higher in shoots compared to the root system especially in zucchini plant. The concentration of PBDEs in root system is open question, especially in roots of controls, which probably should be solved on the basis of more studies, with more different plants and various kinds of xenobiotics.

Plants also exhibit different congener-specific accumulation and translocation. Zucchini preferentially translocated BDE 100, whereas in radish the major detected congener in shoot was BDE 99. Our results also show different congener-specific accumulation and translocation. Total translocation was higher in tobacco, and this plant showed the ability to take up and translocate congener BDE 209. The uptake and translocation of BDE 209 was discussed also by Huang et al. (2010) who showed that root lipid content is positively correlated with BDE 209 uptake, while to the translocation to above-ground tissues has irreversible effect. Fig. 1 shows differences in concentration ratio of accumulated congeners in nightshade and tobacco and their changes comparing to ratio in the original sewage sludge.

Generally, the distribution of the major lower brominated congeners in both tested plants decreased with amount of bromine in root, stem, leaf and fruit. Their BCFs (Fig. 2) were calculated as the ratio of the total plant concentration of certain congener (dry-weight basis) and those in the sewage sludge (dry-weight basis). Tobacco showed higher BCFs for all congeners compared to nightshade and the highest difference in BCF was registered for congener BDE 99, which content was about 24-fold higher than that in nightshade. The high differences in BCFs values for different plants are consistent with the reports by White et al. (2005) discussing *p,p′*-DDE uptake and Inui et al. (2008) who reported up to 400 times higher BCFs for some PCBs for high cultivar accumulators. Fig. 2 also shows that BCFs of BDE 47, BDE 99, BDE 100 and BDE 209 correspond with their hydrophobicity and negatively correlate with their log K_{ow} (BDE 47 < BDE 100 < BDE 99 < BDE 209) for both plants.

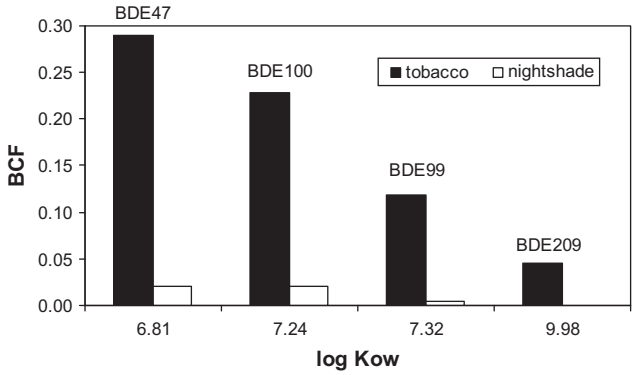

Fig. 2. Relationship between BCF of PBDE congeners in plants and their log K_{ow}.

However the exception is BDE 209 which was accumulated only in tobacco exhibiting better accumulation properties throughout the study. The similar trend was also observed by Inui et al. (2008) who studied uptake of dioxin-like compounds by zucchini (*Cucurbita pepo*) subspecies. The "hyperaccumulators" (cultivars 'Black Beauty' and 'Gold Rush') accumulated preferentially some penta-, hexa- and even heptachlorinated biphenyl congeners whereas other accumulators (cultivar 'Patty Green') accumulated preferentially tetrachlorinated congener PCB 77. Mueller et al. (2006) described also preferential uptake of more hydrophobic congener BDE 100 in zucchini. The mechanism of preferential absorption of PBDEs by plants is still unknown but it is believed to be influenced by several factors. One of the possibility controlling the sorption of contaminants by plants is its desorption from soil (Otani et al., 2007; Inui et al., 2008). Root exudates could alter the bioavailability of POPs (Mueller et al., 2006; Aslund et al., 2008) by forming a more hydrophilic complex which is better transported throughout the plant. Another possibility for the better desorption of hydrophobic compounds from soil is the presence of POP-binding proteins in plants (Inui et al., 2008). Phloem proteins (P-proteins) could be responsible for accumulation of hydrophobic compounds by facilitating their transportation. Overall plant physiology, lipid composition, wax, water content and transpiration rates could also play an important role (Paterson and Mackay, 1994; Simonich and Hites, 1995).

4. Conclusions

Our study demonstrates that contact of plants with the sewage sludge contaminated with PBDEs could be the primary route of PBDEs transport *via* the roots into plant tissues. Therefore the application of contaminated sewage sludge in agricultural practice could lead to accumulation of PBDEs in plants. This practice provides an avenue for input of PBDEs into the food chain by the following exposure pathways: contaminated

soil → plants → livestock → human or contaminated soil → plants → human. Thus, the origin of PBDEs in the body fat and meat of cows (Kierkegaard et al., 2007) could be originated from the grassland plants or the maize grown on fields contaminated by PBDEs. Because the uptake of PBDEs is influenced by plant species, more information is needed regarding PBDE accumulation, translocation and their metabolic pathways in plants to be able to establish the burden of human exposure.

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