E-WASTE IN LANDFILLS - SOURCE OF PERSISTENT ORGANIC POLLUTANTS

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Abstract

Soil pollution can have lasting socioeconomic consequences and an impact on the environment. Pollution can seriously affect the ability of soil to carry out some of its essential functions, and it can affect human health by direct contact or by involvement in the food chain. In order to prevent this, it is necessary to get more information about contaminated sites and the pollutants that are found there. Polybrominated diphenyl ethers are one of the new emerging pollutants found in the environment due to using the equipment containing flame retardants and as a result of irresponsible and illegal disposal and treatment of electronic waste.

The aim of this study was to examine the impact of e-waste improperly disposed of in landfills and some other non-landfill sites. The method for determining PBDE's in soil has been optimized, using chemometric model developed for optimization of extraction conditions and instrumental analysis of PBDE's. Optimal duration of extraction, the best ratio of solvents used for extraction and instrumental conditions that give the best recovery of extraction were defined. The optimized conditions were applied on soil samples taken from the surface layer of soil from 24 locations (near landfills, ex marshalling yard, dump of secondary raw materials and automotive waste) in Autonomous Province of Vojvodina. Recovery values of PBDE's congeners ranged between 80% and 115%. Total PBDE's concentrations ranged from 4.4 to 729 $\mu g \cdot kg^{-1}$ of absolutely dry soil. Highest contribution to total sum have BDE 99, BDE 47 and BDE 209.

Keywords: *POPs, PBDE, landfills, electronic waste, soil*

1. INTRODUCTION

Electronic waste, or e-waste, is a general term used for various electrical and electronic equipment that have ceased to benefit their users. E-waste is represented by all electrical appliances such as large household appliances (refrigerators, freezers, dishwashers, stoves, dryers, microwave ovens, heaters, TA ovens, electric radiators, electric fans, air conditioners, water heaters), small home appliances (vacuum cleaners, irons, toasters, fryers, coffee machines, hair clippers, hairdryers, shavers, watches, watches), IT and telecommunications equipment (computers, printing units, copiers, calculators, fax machines, telephones), radio and TV sets, and a lighting fixture. Given that the information and technological revolution has exponentially increased the need and use of new electronic equipment, the increase in discarded electrical devices is a reality both in the world and in our country, and it is of concern to the entire world public. According to United Nations estimates, global electronic waste increases by about 50 million tons each year. At the same time, it is a devastating fact that only some 15% of discarded electronic components are recycled, and the remaining 85% end up in landfills.

Toxic waste has an adverse socio-economic impact, negatively affecting public health and the environment. Toxic substances present in e-waste are released into the environment and stay there for a long time. E-waste is a potential source of lead, cadmium, mercury, hexavalent chromium, barium, beryllium, phosphorus, arsenic, selenium, trichloroethylene, and PVC. Additionally, persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) can stem from e-waste.

1.1. Polybrominated diphenyl ethers (PBDEs).

PBDEs are compounds used as flame retardants (FR). These are chemicals that are most commonly used in electronic equipment, textiles, furniture, and insulation materials to reduce the flammability of these materials. There are several groups of FR depending on their chemical composition: brominated, phosphorus, nitrogen, chlorinated and inorganic.

Brominated flame retardants (BFR) prevent fire as well as its spread. They do not change the properties of materials, so they are used in various sectors: in plastic items, textiles, floor coverings, computers, televisions, vehicles (cars and trains), and construction materials. Nevertheless, because they do not chemically bind to materials, they are mobile in the environment [2], [4]. They are persistent and have excellent bioaccumulation potential, posing a potential danger to the environment [5]. In this group of compounds, the most significant danger to human health and the environment are PBDE, hexabromocyclododecane (HBCD), tetrabromobisphenol A (TBBPA), polybrominated biphenyls (PBBs) [12]. Soil is the primary medium where PBDEs are located and stay longer. They reach there through atmospheric precipitation, landfills, and sewage water containing wastewater from various sources (such as households, offices, and industrial plants) that treat wastewater differently or do not treat it. The PBDE's concentration in the soil depends on the soil type, the proximity of the urban or agricultural zone, and the type of landfill. Liu and his associates [7] detected the presence of 19 PBDEs (from mono to deca-BDE) in the soil taken from the e-landfill area with the highest concentration of 789 ng·g-1. Similar results (0.26 to 824 ng · g-1) were obtained by Dongli Wang and his associates [13], who sampled soil near plastic-burning landfills, where printer toners (cartridges) are disposed of, and in river sediment which passes through Guiyu - a city in China known as the largest e-landfill in the world.

Due to lipophilicity, PBDE's have the ability to bioaccumulate and can be found in human and animal tissues in measurable concentrations. The presence of PBDE's in the tissues of living beings is associated with the effects on their health. Although the level of a single compound may not lead to an adverse effect, the cumulative effect of these compounds may be of toxicological significance. [1]. No directive defines the maximum permitted amounts of PBDE's in food or products for everyday use. The RoHS Directive [3] on electrical and electronic equipment defines a maximum permissible concentration of hazardous substances (including PBDE's) of 0.1% by weight in homogeneous systems. Observing the impact of PBDE's on humans, animals, or both, they can be primarily classified as endocrine disruptors. Their thyrotoxicity [14], estrogenicity [9], and potential carcinogenicity are known.

1.2. Legislation

Due to the potential danger arising from the properties of POPs, the world is increasingly paying attention to the system of control and management of toxic substances and waste. International conventions, regulations, and protocols have been adopted and implemented, regulating specific details directly or indirectly related to POPs chemicals. One, arguably the most important, is the Stockholm Convention, which became effective in May 2004. It was signed by 152 countries and ratified by 142, including Serbia (2009). The main goal of the Stockholm Convention on POPs is to protect human health and the environment from the harmful effects of POPs chemicals. States that have signed the Convention should establish, prohibit or restrict the production, foreign trade, and use of POPs chemicals and an obligation to reduce or eliminate the emission of POPs compounds. By adopting the Law on Ratification of the Stockholm Convention, Serbia has committed itself to comply with all prescribed provisions. National legislation must comply with the Convention through a single act and all laws related to POPs. In this particular case, it would refer to the laws that regulate the issues: management of chemicals and plant protection products, quality of food, waste, air, water, and others [8].

Given the need to harmonize domestic legislation with commitments, decisions involving the regulation of the production, import, and use of products containing specific PBDE congeners have been made. Thus, in the Ordinance on restrictions and prohibitions on the production, placing on the market, and use of chemicals that pose an unacceptable risk to human health and the environment [10] in Annex 1, Part 1, the list of restrictions and prohibitions includes two substances belonging to the PBDE group: penta BDE and octa BDE.

Improper management of these chemicals during the life cycle of products containing them, especially in waste management, can lead to the emission of POPs and their accumulation in the environment. A particular problem is an e-waste, which is a potential source of PBDE in soil [11]. Therefore, the first steps in dealing with these POPs are to establish regular monitoring, identify sources and present their current state in the environment.

1.3. PBDE's in Serbia

In Serbia, only a few researchers investigated the presence of PBDE's and other POP's in the soil. Therefore, there are no data on the presence of PBDE in the environment. Due to the potential danger to the environment and the obligation arising from the signing of the Stockholm Convention, the subject of this research is PBDE congeners, with particular emphasis on eight (out of 209 possible) congeners with different degrees of bromine substitution, which are indicators of the presence of these compounds in the environment.

2. MATERIALS AND METHODS

2.1. Materials

Hexane (catalog number: 363242.1612) and dichloromethane (catalog number: 361254.1612) for UV, IR, HPLC, ACS were purchased from Panreac (AppliChem, Germany). Silica gel 60 (Davisil grade 636, 35-60 mesh, catalog number 236802) and aluminum (activated, neutral, Brockmann I, 58 Å, catalog number 199974) from Sigma Aldrich, and SPE columns (catalog number: 54222-U) from the manufacturer Supelco were used to purify the extract. Silica gel was activated at 190 °C for 24 hours. Acid silica gel was prepared by combining concentrated sulfuric acid (30 ml) with activated neutral silica gel (50 g). PBDE standard solution (8 congeners of primary interest) was purchased from Accu Standard. Agilent 7890 B gas chromatograph, with CTCPAL sampler for liquid and gaseous samples, EC detector, capillary column HP-5 was used (Agilent J&W GC Columns, catalog number 19091J-413, length 30 m x 0,30 mm x 0,25 μm).

2.2. Methods

2.2.1. Extraction, cleanup and GC analysis

Based on literature data, it has been determined that landfill's are a potential source of PBDE's. Therefore, 20 landfill's on Vojvodina territory were selected, and soil samples were taken along the landfill's perimeter (Figure 1). Based on the terrain analysis, it was decided to sample at four more locations: ex marshaling yard, secondary raw materials dump, car waste, and an illegal landfill where e-waste was noticed. Sampling was performed with a shovel to a depth of 15-20 cm. Samples were taken from 5 to 10 individual sites depending on the landfill's size, homogenized in a separate container, and packed in canvas bags for transport to the laboratory.

Samples were transported in a hand-held refrigerator to the laboratory. They were stored in a freezer at -20°C until analysis.

Prior to analysis, the samples were dried at room temperature, cleaned of stones, glass, and twigs, mixed, crushed with a mortar and pestle, and passed through a 250 μm sieve to obtain a homogeneous matrix.



Fig. 1. Map of Vojvodina with marked sampling sites

Ten grams of blank soil matrix were spiked for recovery testing with 50 µl PBDE standard solution with a 2 µg/ml concentration. USEPA Soxhlet extraction method (Method 1614, EPA 2007) was used for PBDEs extraction from spiked soil. In order to determine the best ratio of solvents and optimum extraction time, 9 extraction experiments were conducted using three different solvent ratios (hexane/dichloromethane 1:1, 2:1, and 3:1) and three-time periods (12, 18, 24 h) of extraction. Each experiment was repeated twice.

After the extraction, the volume of the extract was reduced down to 2 ml by rotary evaporation in the presence of steam of nitrogen at a temperature of 45 °C. Purification was carried out in two columns using the procedure explained in the literature (Covaci et al., 2002). The first column was loaded with acidic silica gel and the second one (multi-layered) contained activated aluminum oxide, a neutral silica gel, and acidic silica gel. Both columns were first washed with hexane. After applying extract, elution was performed in two steps, with hexane and hexane/dichloromethane (1:1), respectively. The total eluate was concentrated to 1 ml using a rotary evaporator at a temperature of 45 °C. The final eluate was transferred to a vial.

The final determination was carried out with gas chromatography with an electron capture detector (GC-ECD) (Agilent 7890B) equipped with a capillary column DB5 (30 m x 0.32 mm i.d., 0.25 µm, Agilent J&W GC Columns). Working conditions of the GC system were set as follows: injector at 300 °C, a detector at 310 °C, initial oven temperature at 90 °C, for 2.0 min, heating to 220 C °at rate 50 °C/min, then to 300 °C at rate 5 °C/min, with a final 20 min hold. Splitless injection with a flow of 60 ml/min was used. Fig. 2 shows a chromatogram obtained from GC-ECD analysis.

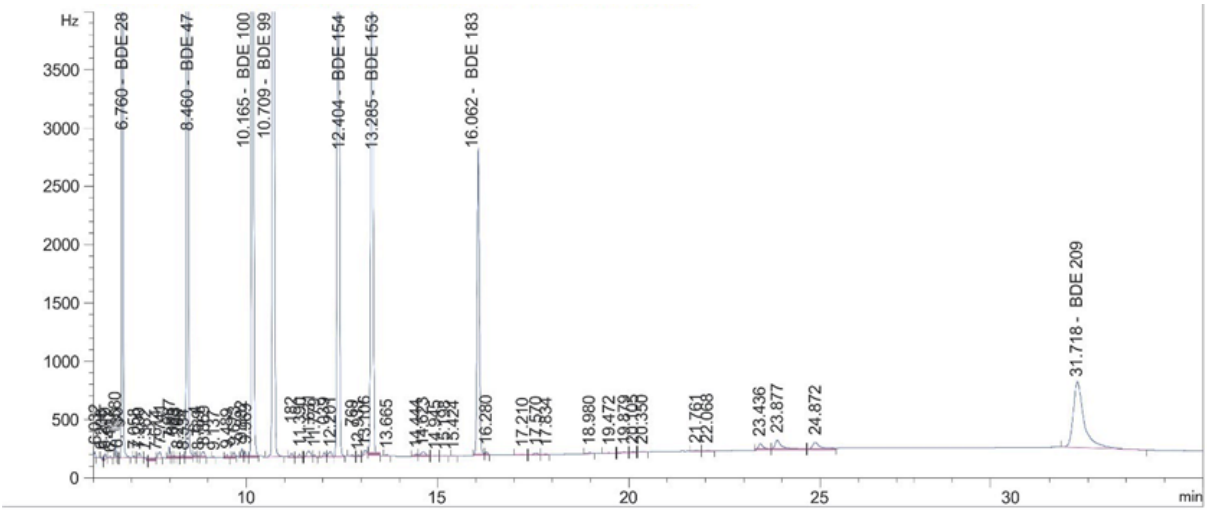


Fig. 2. Chromatogram from the GC-ECD analysis of the PBDE spiked soil blank matrix

2.2.2. Design of experiment

The obtained average recovery values for individual BDE congeners have been used for examination of the influence of the (1) extraction time and (2) hexane/dichloromethane (hexane/DCM) volume ratio by 32 factorial design of experiments (DOE) (Montgomery, 2009). Factorial DOE is a methodology used to systematically evaluate the influence of different factors (variables) on the studied system. At the same time, the remaining factors which might influence the system are kept as constant as possible. In this work, a three-level factorial design with two factors was used to examine the influence of the extraction time and hexane/dichloromethane volume ratio on the recovery. The number of required experiments can be determined by the formula $n = 3^2$. Therefore, for the factorial design with two factors, using 3^2 factorial design, a total number of nine experiments are required. During this study, two replicates of the experiments were carried out. The schematic representation of this factorial design is presented in Table 1.

Experiment	Factors		Variables	
	X_1	X_2	Hexan:DCM	Time (h)
1	-1	-1	1:1	12
2	0	-1	2:1	12
3	1	-1	3:1	12
4	-1	0	1:1	18
5	0	0	2:1	18
6	1	0	3:1	18
7	-1	1	1:1	24
8	0	1	2:1	24
9	1	1	3:1	24

Table 1. A schematic representation of 3^2 factorial design

3. RESULTS AND DISCUSSION

3.1. Analysis of a factorial design

As previously stated, the influence of the analyzed factors on the recovery of PBDEs (Table 2) from soil was performed using 3^2 factorial design. Based on literature data, it was decided to change the factors of interest in the following intervals:

- Volume ratio of Hexane / DCM 1:1, 2:1 and 3:1
- Extraction time: 12 h, 18 h, and 24 h

BDE congener	**28**	**47**	**100**	**99**	**154**	**153**	**183**	**209**
Recovery average, %	80.36	86.14	85.90	92.04	93.37	102.93	115.12	103.59

Table 2. The average recovery values of eight BDE congeners

The modeling function used for this examination of the influence of these two variables on the recovery is:

$$Y = b_0 + b_1 \cdot X_1 + b_2 \cdot X_2 + b_{12} \cdot X_1 \cdot X_2 \quad (1)$$

In this equation, X_1 and X_2 represent the coded variables (Table 1) for the two factors (columns two and three in Table 1), b0 represents the intercept from the axes of the dependent variable (in our case, recovery, labeled as Y) and b_1 represents the influence of the solvent ratio on the recovery. The coefficient b_{12} represents the interaction between the two factors.

Experiments were repeated twice. As previously stated, the volume ratio of solvents (X_1) and time of extraction (X_2) were replaced with coded values from Table 1. The estimated values for the coefficients for eight congeners are presented in Table 3.

Together with the estimated model parameters (b_0, b_1, b_2 and b_{12}) the confidence interval of these parameters (at the significance level of α=0.05) is presented in Table 3. The confidence interval is essential to recognize which of the model parameters has a significant influence on the recovery. A detailed analysis of estimated values for the main effects showed that coefficients b_1 and b_2 significantly influence most congeners' results. Both factors (X_1 and X_2) have a negative impact on the recovery of extraction. That means a larger quantity of hexane in a mixture of solvents and a longer extraction duration reduce the reaction's recovery values. Therefore, the ratio of solvents of 1:1 and extraction time duration of 12 hours was the best conditions for PBDEs extractions from the soil.

Parameters	**BDE 28**	**BDE 47**	**BDE 100**	**BDE 99**	**BDE 154**	**BDE 153**	**BDE 183**	**BDE 209**
b_0 - intercept	78.47	88.15	86.62	91.36	92.13	95.54	106.20	82.96
b_1 - solvents ratio	-3.19	-8.39	-6.10	-5.75	-4.88	-4.72	-3.60	-10.12
b_2 - time	-1.58	-8.07	-4.95	-4.90	-2.90	-4.54	-7.28	-8.65
b_{12} - interaction	5.83	6.06	6.26	5.03	4.56	4.92	6.32	-12.51
Confidence interval	3.51	6.11	4.43	5.60	5.11	5.67	7.32	11.65

Table 3. Calculated effects for the examined factors for the 32 factorial design on the recovery for all analyzed substances

3.2. PBDEs content

The PBDEs content in the 24 soil samples are shown in Table 4. All samples contained PBDEs, and the total amount for eight congeners varied between 4.4 and 723 µg/kg with a median concentration of 11.48 µg/kg. The samples taken from landfills ranged from 4.4 to 67.4 µg/kg, with a median concentration of 7.97 µg/kg. The landfill sample with the most significant value of total PBDEs was sample number 1 taken at the receiving zone of the landfill in Novi Sad city (500 m far from the residential area). It is the oldest part of the landfill, used about 30 years ago. There are two assumptions about the presence of PBDE in that part of the landfill. The first assumption about the presence of PBDE is due to their persistence. The second is the constant disposal of unselected waste in the receiving area from where it is further transported depending on the type of waste. High concentrations of total PBDE were also detected in samples from the landfill in Subotica (34,1 $\mu g \cdot kg^{-1}$) and Bačka Topola (32,40 $\mu g \cdot kg^{-1}$), (samples 34 and 32).

The highest values of PBDEs concentrations were detected in a sample taken at the outer edge of the land plot used for the purchase and storage of recyclable waste materials (sample 24). The company that owns the mentioned land plot deals with purchasing an iron, batteries, aluminum, stainless steel, brass, copper, and electronic waste. The value of total PBDEs in sample 24 taken from the mentioned plot was 723 µg/kg. High concentrations of total PBDEs were also found in a sample taken from an ex marshaling yard located in a residential part of Novi Sad (the capital of Vojvodina province). Today this location is used as a repair yard for trains. Two samples were taken from this location. Sample 4 was taken close to the railway line where, since 2006 are, parked old unusable rolling stocks (manufactured in the seventies in the last century). Railway wagons, which were in the vicinity of the second sample (sample 5), were placed there in 2012.

Congener / Sample	BDE 28	BDE 47	BDE 101	BDE 100	BDE 155	BDE 154	BDE 184	BDE 209	total
1 landfill	0.48	0.01	0.05	0.15	0.21	0.28	0.46	65.79	67.43
2 landfill	0.03	0,09	0.00	0,09	0.07	0.10	0.09	7.50	7.97
3 landfill	0.07	0.06	< LOD	0.07	0.06	0.07	0.04	6.59	6.96
4 ex marshalling yard	0.32	0.78	< LOD	61.59	0.35	0.06	0.03	15.72	78.84
5 ex marshalling yard	3.04	0.38	< LOD	0.84	0.07	0.04	0.51	11.59	16.48
6 landfill	0.47	0.35	< LOD	0.16	0.03	0.03	0.06	6.18	7.28
7 car waste	0.28	1.20	0.84	3.71	0.52	0.58	0.22	13.32	20.69
8 landfill	1.06	0.42	0.07	0.25	0.02	0.11	0.13	4.53	6.59
9 landfill	6.82	1.19	0.36	0.33	0.29	0.20	0.48	7.42	17.08
10 landfill	1.00	0.38	0.00	0.07	0.04	0.05	0.19	5.36	7.09
11 landfill	0.16	0.15	< LOD	0.20	0.03	0.04	0.06	5.77	6.41
12 landfill	3.86	0.57	0.10	0.10	0.05	0.04	0.13	1.65	6.51
13 landfill	1.44	2.09	0.75	0.23	0.16	0.09	0.55	6.18	11.50
14 landfill	0.64	24.7	< LOD	1.21	0.04	0.05	< LOD	5.77	32.40
15 landfill	2.69	2.42	0.44	0.06	0.24	0.16	0.39	7.01	13.40
16 landfill	5.09	6.41	1.72	6.04	0.26	0.59	1.00	12.90	34.1
17 landfill	0.44	0.14	0.01	0.04	0.05	0.07	0.08	7.63	8.46
18 illegal landfill	0.52	0.40	0.06	0.04	0.03	0.10	0.15	3.71	5.02

19 landfill	0.43	0.26	< LOD	0.12	0.06	0.23	0.28	12.10	13.40
20 landfill	2.85	6.51	2.94	0.27	1.01	2.05	0.41	6.18	22.20
21 landfill	0.23	0.33	0.03	0.16	0.08	0.08	0.09	4.12	5.12
22 landfill	0.45	0.40	0.20	0.18	0.06	0.08	0.23	2.83	4.43
23 landfill	1.25	1.18	0.17	0.20	0.07	0.12	0.13	10.78	13.9
24 secondary raw materials dump	237.4	277.9	8.60	32.79	0.27	6.64	4.55	160.8	729.1
MIN	0.03	0.01	0.03	0.04	0.02	0.03	0.03	1.65	4.43
MAX	237.45	277.96	8.60	61.59	1.01	6.64	4.55	160.8	729.1
average	1.46	2.19	0.48	3.31	0.17	0.23	0.26	10.03	17.9
LOD*	0.003	0.003	0.003	0.003	0.003	0.003	0.003	1.500	

***LOD = Limit of Detection (calculated as 3 times the standard deviation of the baseline noise).**

Table 4. Concentrations of PBDEs in soil samples

Congers BDE 209, BDE 99 and BDE 47 have the most considerable contribution to the total amount of PBDE in soil samples. Concentrations of BDE 209 in landfill samples ranged from 1.65 to 65.8 $\mu g \cdot kg^{-1}$ absolutely dry soil (a.d.s.). This concentration is less than the concentration detected near the BDE 209 production plant in China [6] but in the range of detected concentrations near the e-waste recycling site in South China [13]. BDE 209 is always combined with antimony trioxide in polymers, mainly in polystyrene used in the television industry. BDE 209 has also been used to produce polypropylene draperies in upholstery fabrics and can also be found in some synthetic carpets. Due to its wide use in everyday items is an inevitable part of the everyday waste that people dispose of without prior selection. Therefore, the appearance of BDE 209 congeners in the land next to the landfill is expected. High value of BDE 209 from 160.9 µg/kg was detected in sample from secondary raw materials dump.

According to Illinois Environmental Protection Agency (EPA, 2006) BDE 209 has the ability to bioaccumulate in the environment. The most important health effects from exposure to decaBDE, lower-brominated congeners, or both, appear to be liver (Lee et al., 2010), kidney (Li et al., 2014), thyroid, reproductive/developmental (Lichtensteiger et al., 2003), and neurological effects (Goodman et al., 2009).

Pentabromodiphenyl ether (BDE 99) and Tetrabromodiphenyl ether (BDE 47) are the most abundant congeners in a commercial, technical mixture penta BDE. Penta BDE is or has been used as flame retardant almost exclusively in the manufacture of flexible polyurethane (PUR) foam for furniture and upholstery in homes and vehicles, packaging, and non-foamed PUR in casings and electronic equipment. They are also used to a certain extent in specialized applications in textiles and industry. Concentrations of BDE 99 in analysed samples were in the range from 0,02 to 61,6 µg/kg a.d.s. The biggest value of BDE 99 was detected in sample 4, sampled at ex marshaling yard. BDE 47 was detected in all samples, but the highest value (not considering sample 24) was in the sample taken from the landfill in Backa Topola (sample 14).

A graphical representation of PBDE concentrations in soil samples is shown in Figure 3 and Figure 4.

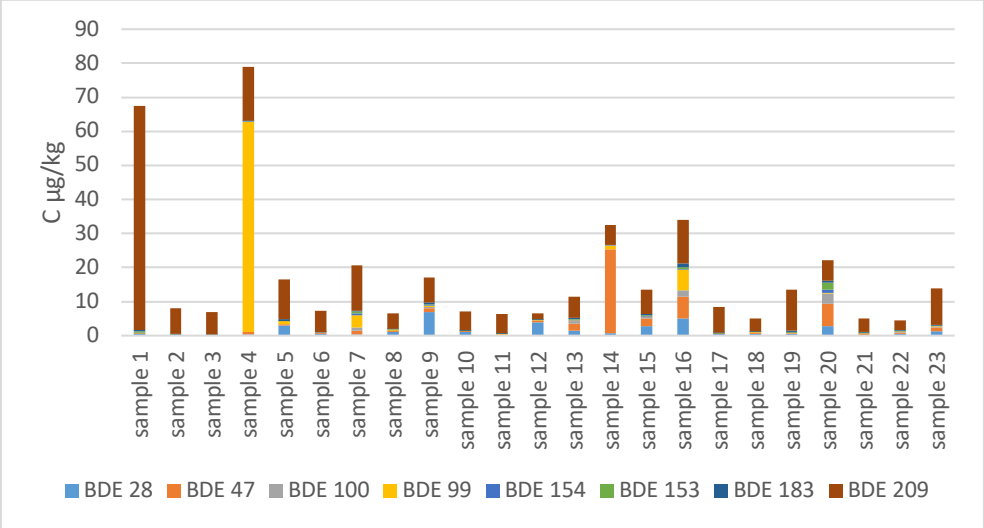


Fig. 3. Graphical representation of the concentrations of eight PBDE congeners in soil samples

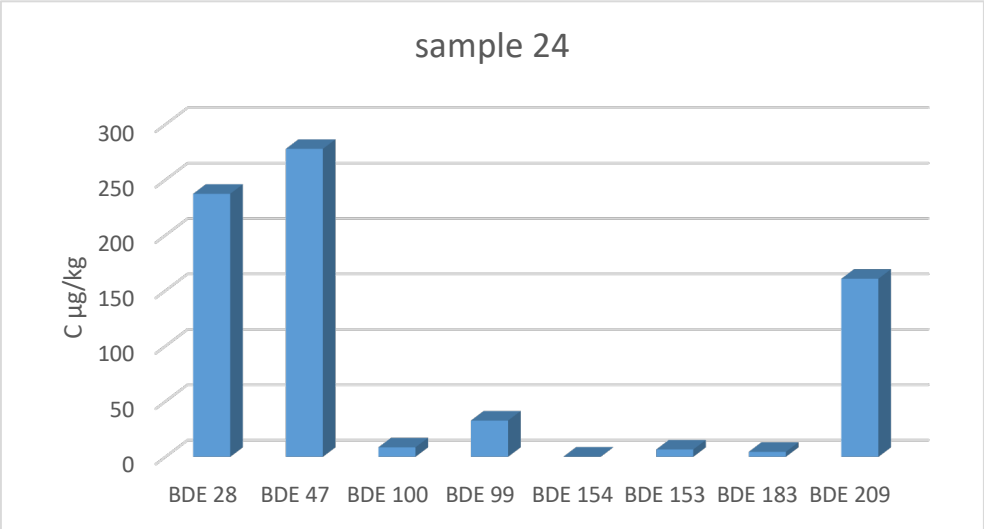


Fig. 4. Graphical representation of the concentrations of eight PBDE congeners in sample 24

4. CONCLUSIONS

On the territory of Serbia, there are over 160 landfills (44 on the territory of Vojvodina) under the jurisdiction of public utility companies and many illegal landfills, which represent a particular problem for the environment and human health. According to official data, a small amount of research has been done on the existing landfills' locations regarding the soil quality and organic and inorganic pollutant concentrations in soils.

Of particular interest are long-lasting organic pollutants, the production, and use of which are partly regulated by the Stockholm Convention on POPs. In this study, particular emphasis was placed on PBDE. The presence of PBDE in soil samples was analyzed, and their concentrations were determined using an optimized method in the laboratory of the Faculty of Environmental Protection, Educons University. As a result, the presence of all eight BDE congeners in soil samples was proven.

Furthermore, electronic waste was noted during the visit to the selected landfills. Based on that, it can be assumed that e-waste is one of the sources of these long-lasting pollutants.

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