



55 xenobiotic organic compounds in Tripoli landfill-Lebanon leachate and their fluxes to the Abou Ali River and Mediterranean Sea

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Abstract Pollution generated from landfill solid wastes constitute one of the major threat to the environment. The landfill leachate contains various toxic pollutants, making it the most dangerous issue of the landfills. Monitoring the xenobiotic organic concentrations in landfill leachate is an important step to evaluate the environmental impacts. This work aims to characterize the seasonal variation of 55 xenobiotic organic compounds including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), phthalic acid esters (PAEs) and bisphenols (BPs) in the leachate from municipal solid waste landfill of Tripoli, Lebanon. And also, the quantity of the pollutant's flux to the Abou Ali River and the Mediterranean Sea nearby has been estimated. The organic pollutants were extracted by using the solid-phase extraction and quantified by using GC-MS/MS. The results showed high level of PAEs, BPs, PCBs, and PAHs in the leachate samples. The fluxes

of pollutants to the Abou Ali River and Mediterranean Sea have been detected at 0.23 kg, 0.01 kg, 116.85 kg, 15.93 kg, and 7.58 kg for Σ_{16} PAHs, Σ_{28} PCBs, Σ_6 PAEs, Σ_4 BPs, and 4-NP respectively.

Keywords Landfill · Leachate · Persistent organic pollutants · Xenobiotic organic compounds · PAHs · PCBs

Introduction

Landfills are the most used method in waste management in developing countries. The landfill waste is exposed to biological and physicochemical transformations leading to highly polluted effluent called leachate. The leachate characteristics are influenced by several factors like waste composition, landfill age, landfill structure, compaction technique, and weather

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conditions (a. The leachate pollutants are classified into four types: dissolved organic matter, inorganic macrocompounds, heavy metals, and xenobiotic organic compounds. These last include polychlorobiphenyls (PCB), polycyclic aromatic hydrocarbons (PAHs), bisphenols (BPs), phthalic acid esters (PAEs), pharmaceutical residues, and pesticides (Gallen et al., 2017; Kalčíková et al., 2011). PAHs can be generated from anthropogenic and pyrogenic sources, while PCBs have anthropogenic origin; they are used in many industries and commercial applications such as electrical and heat transfer equipment, dyes, and carbonless copy paper (USEPA, 2014). Municipal dumpsites constitute an important source of PAHs and PCBs into marine environment via leachate (Merhaby et al., 2019). PAHs and PCBs are listed as persistent organic pollutants (POP) in the Stockholm Convention (United Nation Environmental Programme, 2019) and as priority pollutants due to their toxicity, mutagenicity, and carcinogenicity effect (USEPA, 2014), while PAEs and BPs are endocrine disrupting chemicals, and their environmental behavior has attracted considerable attention due to their potential impact on ecosystem and on public health. PAEs are present in many materials or products including PVC and building materials, personal care products, medical devices, children's toys, and food containers (Net et al., 2015b), while BPs are used in the production of polycarbonates, epoxy resins, thermal papers, cans, and plastics industry, the automotive industry, glasses, and others. PAEs and BPs are classified as endocrine disruptors and as priority pollutants (USEPA, 2014). PAEs and BPs are ubiquitous in the environment including in fresh and marine waters, soil, sediments, manure, compost, effluents, waste dump water, and discharge percolates (Idowu et al., 2019; Net et al., 2015a, b; Paluselli et al., 2018a, b; Ben Sghaier et al., 2017a, b; Xing et al., 2022).

Open dumps and uncontrolled landfills are dominant in Lebanon especially in big cities like Beirut and Tripoli (Idowu et al., 2019). The inappropriate strategy of waste in Lebanon is characterized by the absence of waste sorting and recycling policy (Halwani et al., 2020). Due to the dumping of many hazard waste into landfills, such as unused pharmaceutical product, medical waste, personal care product, paints, pesticides, plastics, electronic materials, and nylon, the landfill leachate constitutes an important source of organic pollutants like PAHs, PCB, PAEs, and phenols (Crawford & Quinn, 2017). The landfills constitute a high risk to

the environment due to the various pollutants in leachate. Landfill's leachate is frequently rejected into the aquatic system without any treatment especially in developing countries. Thus, toxic compounds can affect fauna, flora, human health, and also aquatic system (Qi et al., 2018; Singh et al., 2016). Several studies have focused on the physicochemical characteristics of landfill leachate (Naveen et al., 2017). However, there is a lack of data on the state of xenobiotic organic compounds release from landfill into leachate and into the aquatic environment. This paper focused on the evaluation the level of 55 xenobiotic compound including 16 PAHs, 28 PCBs, 6 PAEs, 4 BPs, and 4-nonyl phenol in Tripoli landfill leachate, North of Lebanon, where no previous studies have investigated. The objective was to characterize the seasonal variation of these 55 xenobiotic organic compounds in the leachate from municipal solid waste landfill of Tripoli, Lebanon, and to assess quantity of the pollutant's flux to the Abou Ali River and the Mediterranean Sea nearby. This last could affect the marine fauna and thus human health via the food chain and touristic activities.

Materials and methods

Chemicals and materials

C-18 cartridge (200 mg/6 ml), the standards of PAEs, bisphenols, and 4-nonyl phenol were purchased from Sigma-Aldrich (USA). PAHs standard were purchased from Restek (Bellefonte, USA). Naphthalene-d₈, Acenaphthene-d₁₀, Phenanthrene-d₁₀, Pyrene-d₁₀, and Perylene-d₁₂ used as internal standard for PAHs quantification were obtained from LGC-Promochem (UK). PCBs standard was purchased from AccuStandard Inc. (USA). TCN, CB112, and OCN used as internal standards for PCBs quantification were obtained from Dr. Ehrenstorfer (Augsburg, Germany).

Targeted compounds

Four families of micro-pollutants including 16 PAHs, 28 PCBs, 6 PAEs, and 5 phenolic compounds were studied. (i) 16 PAHs are as follows: Naphthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phen), Anthracene (Ant), Pyrene (Pyr), Fluoranthene (Flt), Benzo[a]

anthracene (B(a)A), Chrysene (Cry), Benzo[b]fluoranthene (B(b)F), Benzo[k]fluoranthene (B(k)F), Benzo[a]Pyrene (B(a)P), Benzo[ghi]perylene (B(g,h,i)P), Dibenzo[ah]anthracene (D(ah)A), and Indeno[1,2,3-cd]pyrene (IP). (ii) 28 PCBs are as follows: CB8, CB18, CB28, CB44, CB52, CB66, CB101, CB77, CB81, CB114, CB105, CB123, CB128, CB126, CB138, CB187, CB118, CB167, CB157, CB156, CB169, CB170, CB180, CB153, CB189, CB195, CB206, and CB209. (iii) 6 PAEs are as follows: Dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DNBP), butyl benzyl phthalate (BBP), bis-2-ethylhexyl phthalate (DEHP), and di-n-octyl phthalate (DNOP). (iv) 5 BPs are as follows: bisphenol C (BPC), bisphenol E (BPE), bisphenol F (BPF), bisphenol G (BPG), and 4-nonylphenol (4-NP). Chemical formula, the abbreviation, qualification and quantification ions, retention time (RT), internal standard for the quantification of each compound, and number of chlorinated atom or cycle of PCBs, PAHs, PAEs, and BPs are presented in the Table S1, S2, and S3 in supplemental information.

Study area

The Tripoli landfill is located in Tripoli, on the coastal area of the Mediterranean Sea and at the level of the Abu Ali estuary with the coordinates 34° 27' 19.49" N and 35° 50' 26.91" E. Tripoli is the largest city in northern Lebanon with 450000–550000 inhabitants and is characterized by a Mediterranean climate, cold and wet during the winter. Tripoli landfill, located in Tripoli along the coastline of the Mediterranean Sea and adjacent to the Abou Ali River, covers an area of 60000 m². Tripoli landfill was used as an uncontrolled dumpsite for over 20 years until 1999 when the site was rehabilitated and operated as a controlled landfill receiving the waste from Al Fayhaa Union municipalities, with an average of 14000 tons/month. Tripoli landfill received various types of wastes without sorting process where the organic wastes represents 64% of the total volume of wastes, followed by papers and cardboards (15%), plastics (10%), glass (5%), and metal wastes (2%). Collected wastes are landfilled by spreading and compression in layers of 50 cm and then covered by inert soil. The landfill body features a heterogeneous physical structure, which is constituted of two sections, the lower section (old—unit A), and the upper section

(new—unit B). These units present different conditions like height and degree of compaction. Age of waste in unit A is about 30 years and lower than 10 years in unit B. The flow rates of leachate reach 3 m³/d in unit A and 40 m³/d in unit B. The generated leachate is drained out by a pipes system and periphery ditches and then passes into the estuary of Abou Ali River, which leads to the Mediterranean Sea. The continuous accumulation of rubbish has transformed the site into a mountain of trash with about 45 m levels making Tripoli's dumpsite one of the highest landfills in Lebanon. Figure 1 presents the sampling points.

Sample collection

The samples were collected in duplicate from the output leachate pipe of both sections every 2 months from September 2017 to September 2018. Six sampling campaigns were performed in the new section of landfill, and four sampling campaigns were performed in the old section due to the drying of leachate during summer. The drying contribute to the sorting of organic matter from landfilled waste and/or the deviation of leachate flow rate caused by the damage of periphery wall in section A (Halwani et al., 2020). Samples collected in pre-cleaned glass bottles were directly transported to the laboratory. The coordinates of the two sampling points of leachate are 34° 27' 21.26" N and 35° 50' 29.6" E for point A and 34° 27' 19.22" N and 35° 50' 25.46" N for point B, and they are depicted in the Fig. 1. The leachate was generated by Tripoli landfill draining directly to the Abou Ali River by pipes and then reached the Mediterranean Sea without any treatment.

Sample extraction

The samples were directly filtered with pre-calcinated 0.47 µm Whatman GMF. The target compounds were extracted by using solid-phase extraction (SPE) with C-18 cartridge. Firstly, each sample was spiked with the internal standards. Briefly, the cartridges were conditioned by 9 ml of acetonitrile, 9 ml of 2-propanol, and then 12 ml of a mixture of Milli-Q/2-propanol (85/15, v/v) acidified to pH 2.5 with sulfuric acid. After conditioning step, 100 ml of leachate samples were passed through the cartridges with a flow rate of 1 drop/second. The cartridges were washed with 30 ml of a mixture of Milli-Q/2-propanol (85/15,

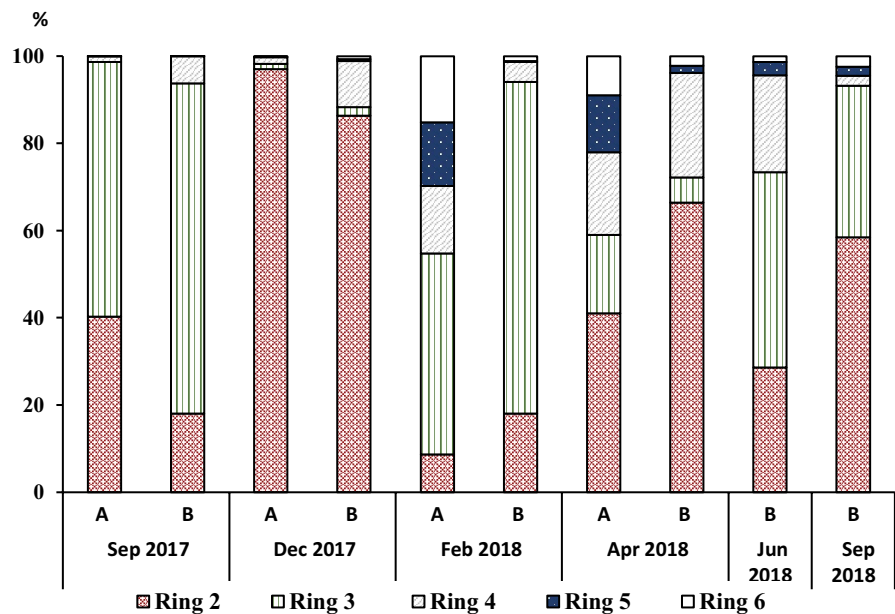


Fig. 1 Photo of sampling site and sampling points from September 2017 to September 2018, for every 2 months

v/v) acidified to pH 2.5 and then dried with N₂. The targeted compound was eluted with a 12 ml of a mixture of hexane/acetone/2-propanol (90/5/5, v/v/v)

followed by 3 ml of dichloromethane. The extract was then concentrated to a volume of 200 μl by N₂ and kept at -18 °C until GC-MS analysis (Fig. 2).

Fig. 2 PAHs distribution profile for the leachate of units A and B



GC–MS analysis

Each family of organic compound was analyzed separately, using an Agilent 7890B GC, equipped with ZB-XLB-HT a Zebron capillary column (30 m×0.25 mm i.d.×0.25- μ m film thickness), and coupled with a TQ Agilent series 7000/7010 MS. Helium was used as carrier gas at a flow rate of 1 ml/min. Samples were analyzed in the splitless mode at 280 °C. The oven temperature was programmed as follows: from 48 °C (1.5 min) to 170 °C at 13 °C/min, then ramped at 4 °C/min to 230 °C, and then at 3 °C/min (32 min) to 280 °C.

Quantitative analysis and quality control

The concentration of each compound was determined according to 6-point internal calibration methods. Internal standards were added for each calibration point in order to better fit to the properties of each targeted compounds. To minimize the error of quantification, the procedural blank for the entire analytical procedure was performed in triplicate together with each environmental sample batch. Our results showed that the procedural blanks were for PCBs, PAHs, and BPs and were free from any targeted PCBs, PAHs, and BPs. However signification level higher than LOQ of PAEs was sometimes detected in procedural blanks, especially for DNBP and DEHP. The concentration of DNBP and DEHP measured in blanks could reach up to 4% of the concentration measured in leachate sample. Consequently, the results of DNBP and DEHP were corrected by subtracting the values found in procedural blanks as recommended by Net et al. (2015c). The precision and accuracy of the method were confirmed by duplicate analysis of the samples. The precision of the technique, specified by the standard deviation, was found between 1 and 5%.

Results and discussion

PAHs concentration and composition profile

Among the 16 PAHs, 12 of them were detected, and their concentrations are presented in Table 1. High heterogeneity of the number of detected compound and the concentration level of each sampling point and each campaign were found. In unit A, Nap

showed the highest level and followed by Acy and Pyr. In unit B, similarly, Nap was detected dominant, and it followed by Phen and Flu (Fig. 3). Our results showed the dominant of low molecular weight PAH with 2 and 3 aromatic rings (Nap, Ace, Phen, and Flu). High molecular weights with 5–6 aromatic ring including B(b)F, B(a)P, D(ah)A, and B(ghi)P were bellow LOQ in both units. However, the Σ_{16} PAHs in leachate from unit B were found to be 5 times higher than which found in unit A. The Σ_{16} PAHs ranged from 0.1 to 5.9 $\mu\text{g l}^{-1}$, average of 1.8 $\mu\text{g l}^{-1}$ in unit A, and from 0.8 to 29.6 $\mu\text{g l}^{-1}$ (average of 9.2 $\mu\text{g l}^{-1}$) in unit B (Table 1).

The differences between these two units can be attributed to the type, age, degradation state of waste, and leachate flow rate. In comparison, our results were much higher than which found by Borjac et al. (2019) in south Lebanon (0.19–1.1 $\mu\text{g l}^{-1}$) and Kalmykova et al. (2013) in leachate of Sweden at 0.6 $\mu\text{g l}^{-1}$. However, our results were much lower than which reported by Jiries et al. (2005) in Jordan where the Σ_{16} PAH ranged at 7.1–12.6 mg l^{-1} with an average of 9.1 mg l^{-1} . 8 PAHs are classified as probably (B(a)A, B(a)P, DB(ah) A) or possibly carcinogenic (Nap, Chry, B(b)F, B(k)F, I(cd)P) to humans by the (USEPA, 1993; IARC, 2002). The highest Σ_8 PAHs carcinogenic were determined in unit B at 4.4 $\mu\text{g l}^{-1}$ of average (0.2–13.8 $\mu\text{g.l}^{-1}$), while the average in unit A were determined at 1.6 $\mu\text{g l}^{-1}$, and Fig. 2 shows the composition profile of PAHs.

PAHs source distribution

The PAHs source can be identify by various ratios. Among the ratios, B(a)A/B(a)A + Chry (BaA/228) ratio is commonly used. The ratio <0.20 indicates petroleum origin and >0.35 indicates combustion source (Yunker et al., 2002; Christensen & Bzdusek, 2005). From Table 1, the B(a)A/(B(a)A + Chry) ratio was calculated. The ratio shows the mixture of combustion and petrogenic source of PAHs pollution. Moreover, Phe/Ant ratio can be used to distinguish the petrogenic and combustion sources. When Phe/Ant ratio <10 indicates the pyrolytic sources and >10 indicates the petrogenic sources (Maliszewska-Kordybach et al., 2008). Our results showed the ratio of 0–13.6, where 75% of Phe/Ant ratio is <10 and 25% >10. Thus, the combustion is the dominant source in the unit A. Similarly for unit B, the Phe/Ant ratio was

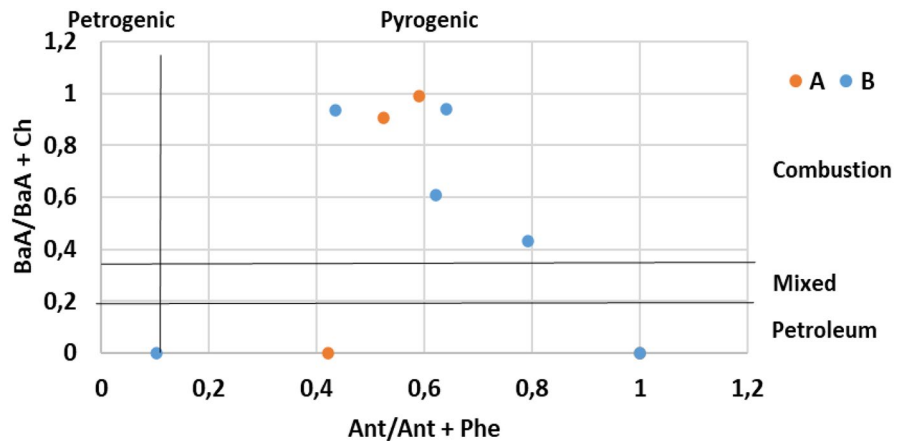
Table 1 Individual concentration of PAHs in units A and B from September 2017 to September 2018

Sampling date	Sep 2017		Dec 2017		Feb 2018		Apr 2018		Jun 2018		Sep 2018	
	A	B	A	B	A	B	A	B	A	B	A	B
PAHs $ng\ l^{-1}$												
Nap*	260±10	153±7	5751±12	12,221±14	8±0.1	5345±11	154±7	1867±11	754±12	2946±12		
Acy	357±13	503±14	-	-	9±0.3	358±10	19±2	22±1	845±2	18±1		
Ace	-	-	13±1	38±2	5±0.1	172±6	3±0.3	8±1	246±4	50±2		
Flu	3±0.1	11±1	5±0.4	67±3	8±0.4	2720±9	21±2	48±3	84±1	203±3		
Phen	16±1.6	125±10	-	-	15±2	18,843±8	-	-	352±5	1111±10		
Ant	1±0.01	3±0.2	59±6	169±6	5±0.1	432±4	24±2	84±4	414±7	370±9		
Pyr	-	-	81±7	1488±13	3±0.1	263±2	22±1	108±4	232±4	98±1		
Flt	9±0.7	53±4	-	-	-	-	-	-	-	-		
B(a)A*	-	-	-	-	11±1	670±6	44±3	527±6	334±6	8±1		
Chry*	-	-	5±0.3	12±1	0.01	429±5	5±0.4	37±1	21±1	10±1		
B(b)F*	-	-	-	-	-	-	-	-	-	-		
B(k)F*	-	-	10±1	54±4	13±1	40±1	49±3	49±1	80±2	107±2		
B(a)P*	-	-	-	-	-	-	-	-	-	-		
DB(ab)A*	-	-	-	-	-	-	-	-	-	-		
B(g,h,i)P	-	-	-	-	-	-	-	-	-	-		
I(cd)P*	-	-	-	101±6	14±1	349±3	34±2	61±3	34±1	121±3		
Min	-	-	-	-	-	-	-	-	-	-		
Max	357	503	5751	12221	15	18843	154	1867	754	2946		
Σ_{16} PAHs	647	849	5931	14150	90	29620	375	2810	2636	5041		
Σ_8 Carcinogenic PAHs	261	153	5774	12389	45	6833	286	2542	1224	3192		
BaA/BaA + Chry	-	-	-	-	1	1	1	1	1	1		
Phe/Ant	14	39	-	-	3	44	-	-	1	3		
Ant/Ant + Phe	-	-	1	1	-	-	1	1	-	-		

“-”: < LOQ; n = 4 replicate

* Human carcinogen compounds IARC (2002)

Fig. 3 Cross plot of Ant/ Ant+Phe versus BaA/ BaA+Ch for leachate samples during the study period



from 0 to 43.6, where 67% is < 10 and 33% > 10. The results indicate the combustion as dominant source of PAHs pollution. Besides, Ant/Ant+Phe ratio < 0.1 indicates a petrogenic source and > 0.1 indicates a pyrogenic source (Bucheli et al., 2004). From the data in Table 1, the Ant/Ant+Phe ratio can be calculated where the values ranged from 0 to 1 for both unit. For the unit A, 25% of Ant/Ant+Phe ratio is < 0.1 and 75% > 0.1, while in the unit B, 33% < 0.1 and 67% of Ant/Ant+Phe ratio > 0.1. Ant/Ant+Phe ratio confirms the combustion process as the major origin of PAHs in our leachate. The cross plots of BaA/BaA+Ch and Ant/Ant+Phe in the two units indicated that PAHs originated mainly from combustion sources (Fig. 3). Our results show that the PAHs in leachate samples of Tripoli landfill were predominantly pyrogenic sources. These results may be due to the historical state of waste incineration, and it is compatible with research established by Borjac et al. (2019) in leachate in south Lebanon (Borjac et al., 2019; Halwani et al., 2020; Zhang et al., 2013). In addition, the atmospheric deposition can be a source of PAHs from the incineration of tire and electric cable near the landfill and the incineration of fossil fuel in the power plants and road traffic (Lebanese Ministry of Environment, 2006).

PCBs composition profile

Among the 28 PCBs, 15 were detected, and the level of Σ_{28} PCBs was in the range of 30–400 ng l⁻¹ with an average of 200 ng l⁻¹ in unit A and 90–2000 ng l⁻¹ with an average of 500 ng l⁻¹ in unit B. The individual concentration of PCB is presented in Table 2.

Our results are in the same range of which found by Herbert et al. (2006) in leachate of Portugal landfill (700–2090 ng l⁻¹). However, our results were much lower than which founded by Yusoff et al. (2013) in leachate of Malaysians landfill where 8.9 mg l⁻¹ of Σ_{28} PCBs were found. PCB 28, 52, 101, 118, 138, 153, and 180 were classified as PCB-indicators (PCBi) which are frequently detected in the environment. Also 12 PCBs were classified as PCBs-dioxin-like (PCBs-DL: 77, 81, 105, 114, 118, 123, 126, 156, 157, 167, 169, and 189) known to be toxic to humans and persistent in the environment (Kimbrough et al., 2010; Tanabe & Minh, 2010; Merhaby et al., 2019). The composition profile of PCB in leachate of unit A and unit B is presented in Fig. 4.

In unit A, the Σ_7 PCBi were 4–324 ng l⁻¹ with an average of 126 ng l⁻¹ and represents 45% of the Σ_{28} PCBs. For unit B, the average Σ_7 PCBi was 47 ng l⁻¹ (8–148 ng.l⁻¹) and represents 12% of Σ_{28} PCBs. The Σ_7 PCBi detected in this study are much lower than those founded in the leachate of Portuguese landfills at 713–2098 ng l⁻¹ (Herbert et al., 2006) and Malaysian landfill at 770,000 ng l⁻¹ (Yusoff et al., 2013), while the Σ_{12} PCB-DL was quantified at 18.8 ng l⁻¹ of average (3.7–38 ng l⁻¹) and represents 14% of Σ_{28} PCBs in unit A. In unit B, Σ_{12} PCB-DL was 240 ng l⁻¹ of average (25–1105 ng.l⁻¹) and represents 37%. These finding is similar to which found by Herbert et al. (2006) and Yusoff et al. (2013). The Σ_7 PCBi and Σ_{12} PCB-DL represent 59% and 50% of Σ_{28} PCBs for units A and B respectively. The present of PCBs in Tripoli landfill leachate might be related to the dumping of PCB-containing waste like e-waste, ink, capacitors, and transformers;

Table 2 The concentration of PCBs in units A and B leachate from September 2017 to September 2018

Sampling date PCBs ng l ⁻¹	Sep 2017		Dec 2017		Feb 2018		Apr 2018		Jun 2018	Sep 2018
	A	B	A	B	A	B	A	B	B	B
CB8	40±3	40±1.4	60±2.8	163±7.1	5±0.3	80±4.2	15±1.4	116±7.2	840±11.3	44±3.5
CB18	-	-	-	-	-	-	-	-	-	-
CB28	374±11	18±1.1	28±0.7	15±1.4	3±0.2	2±0.1	1±0.1	14±0.9	28±1.4	6±0.6
CB44	4±0.4	2±0.1	2±0.1	6±0.6	7±0.4	6±0.5	6±0.4	7±0.6	38±1.8	6±0.6
CB52	-	-	-	-	-	-	-	-	-	-
CB66	-	-	-	-	-	-	-	-	-	-
CB101	-	-	63±2.5	18±1.6	20±1.4	16±1	-	-	3±0.3	-
CB77	5±0.4	N.A	19±1.3	61±3.5	3±0.1	40±1.4	1±0.03	42±1.7	44±1.4	43±3
CB81	2±0.1	2±0.1	1±0.1	2±0.2	-	1±0.04	1±0.03	6±0.3	926±11	4±0.4
CB114	-	-	-	-	-	-	-	-	-	-
CB105	-	-	-	-	-	-	-	-	-	-
CB123	-	-	6±0.2	4±0.3	-	-	-	3±0.1	9±0.7	-
CB128	1±0.1	1±0.1	1±0.1	1±0.1	-	1±0.03	-	1±0.01	2±0.1	-
CB126	1±0.03	2±0.2	2±0.1	4±0.4	-	2±0.1	-	13±1.3	3±0.3	2±0.1
CB138	-	-	-	-	-	-	-	-	-	-
CB187	7±0.7	9±0.9	9±0.8	12±1.1	4±0.3	3±0.2	2±0.1	7±0.4	3±0.2	8±0.7
CB118	N.A	N.A	9±0.6	5±0.2	1±0.01	1±0.1	2±0.03	35±1.4	117±7.1	1±0.1
CB167	-	-	-	-	-	-	-	-	-	-
CB157	2±0.1	1±0.04	1±0.1	1±0.03	-	-	-	2±0.1	1±0.04	-
CB156	-	-	-	-	-	-	-	-	-	-
CB169	12±1.1	17±1.4	1±0.1	1±0.1	8±0.8	25±1.4	-	17±1.1	4±0.03	-
CB170	-	-	-	-	-	-	-	-	-	-
CB180	-	1±0.1	-	-	-	1±0.1	-	1±0.03	-	-
CB153	-	-	-	-	-	-	-	-	-	-
CB189	-	-	-	-	-	-	-	-	-	-
CB195	1±0.05	1±0.1	-	-	-	2±0.1	1±0.03	1±0.04	2±0.1	-
CB206	-	-	-	-	-	-	-	-	-	-
CB209	-	-	-	-	-	-	-	-	-	-
Min	-	-	-	-	-	-	-	-	-	-
Max	374	39	63	163	20	79	15	116	926	44
Σ ₇ PCBi	374	19	101	38	25	19	4	49	148	8
Σ ₁₂ PCB-DL	21	25	38	77	13	69	4	117	1105	50
Σ ₂₈ PCB	446	96	202	293	52	178	30	261	2020	115

“-” : < LOQ; N.A. not analyzed

atmospheric deposition from the incineration of the tire and electric cable in the study area; and from the power plants. The PCBs in unit A lower than in unit B can be due to the age and the type of waste.

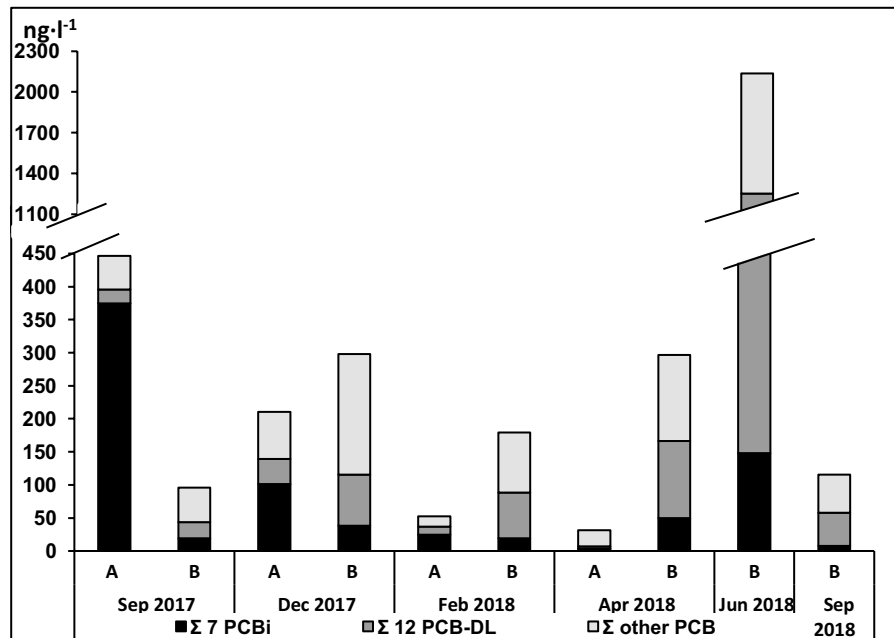
PAEs concentration profile

Five PAEs among the six were detected, and the result is shown in Fig. 5. The individual concentration

of PAEs is presented in Table 3. PAEs were detected at the highest level compared to PAHs and PCBs in the leachate obtained from points A and B. The Σ₆PAEs varied from 571 and which can be up to 8031 μg l⁻¹ with an average of 2673 μg l⁻¹ in unit A. Similar value was found in unit B with the Σ₆PAEs of 399–4892 μg l⁻¹ and average of 2891 μg l⁻¹.

PAEs in the leachate of Tripoli landfill is higher than which found in China at 50–62 μg l⁻¹

Fig. 4 PCB composition profile in leachate of unit A and unit B from September 2017 to September 2018 in ng l^{-1}



(He et al., 2009) and in Sweden landfill leachate ($1.2\text{--}46.8 \mu\text{g l}^{-1}$) (Kalmykova et al., 2013; Öman & Junestedt, 2008). High levels of Σ_6 PAEs might be attributed to the abundance of plastics material and the absence of sorting of recyclable materials and the landfilling of various hazardous wastes containing

PAEs. Indeed, PAEs have been used in a very broad range of applications, and their content can be up to 10–60% by weight of final product (Net et al., 2015b). PAEs have been used in many materials or products including PVC products, building materials including paint, adhesive, wall covering, personal

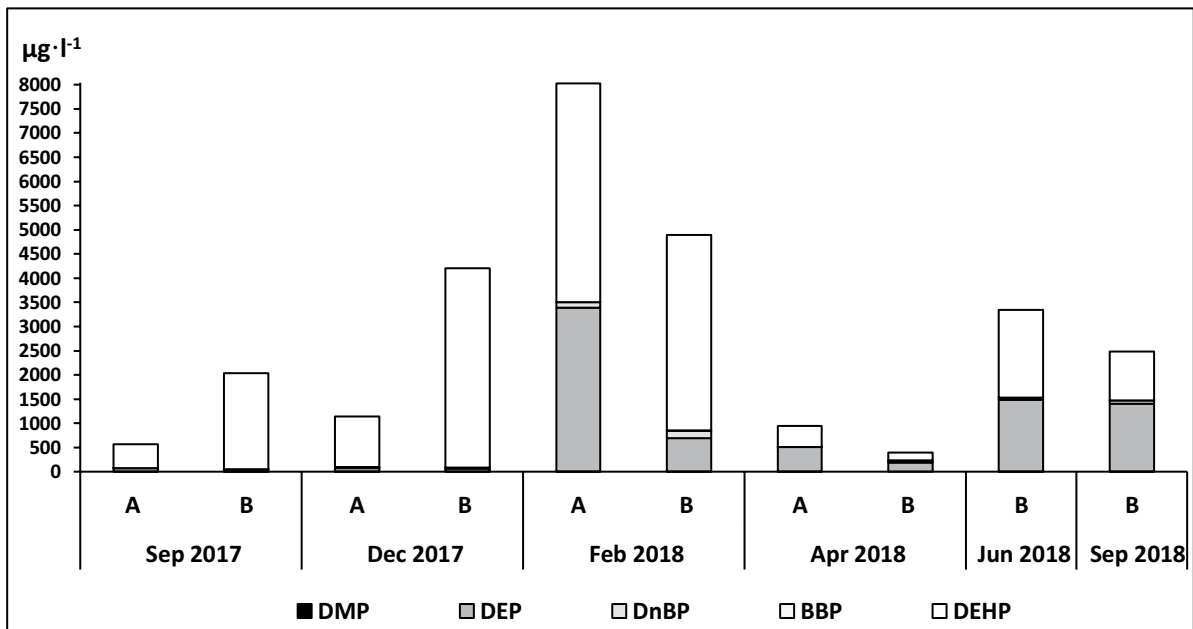


Fig. 5 PAEs composition profile in leachate of unit A and unit B from September 2017 to September 2018 in $\mu\text{g l}^{-1}$

Table 3 The concentration of PAEs and BPs in leachate from September 2017 to September 2018

Sampling date	Sep 2017		Dec 2017		Feb 2018		Apr 2018		Jun 2018		Sep 2018	
	A	B	A	B	A	B	A	B	A	B	A	B
PAEs $\mu\text{g l}^{-1}$												
DMP	0.1±0.01	0.1±0.007	1±0.1	0.1±0.01	0.5±0.02	0.3±0.01	0.1±0.01	0.001±0.0001	3±0.2	1±0.02		
DEP	71±1.8	47±1.4	70±6.5	66±2.8	3388±11.3	692±16	510±9.9	184±10.6	1477±8	1403±15.5		
DnBP	3±0.2	4±0.01	19±1.4	19±1.6	115±2.8	155±2.8	5±0.4	44±1.4	48.9±2.8	72±3		
BBP	0.2±0.01	0.03±0.001	10±0.4	5±0.4	3±0.1	3±0.1	0.1±0.01	1±0.01	1±0.1	1±0.05		
DEHP	497±14.1	1985±15.5	1043±16.9	4110±19.8	4525±17	4042±20	433±18.5	170±2.8	1809±17	1005±18.4		
DNOP	-	-	-	-	-	-	-	-	-	-		
Min	-	-	-	-	-	-	-	-	-	-		
Max	497	1985	1043	4110	4525	4042	510	184	1809	1403		
Σ_6 PAE	571	2036	1143	4200	8031	4892	948	399	3340	2481		
BPs $\mu\text{g l}^{-1}$												
BPF	199±10	268±14.1	175±11.3	170±5.6	140±3.5	132±3.5	175±8.4	154±5.7	122±4.3	156±5		
BPE	200±9.4	281±18.4	175±9.1	173±7.7	140±2.8	163±5.7	98±4.5	175±8.2	153±6.3	162±3.6		
BPC	-	-	-	-	-	-	-	-	-	-		
BPG	23±1.2	47±1.4	27±1.2	89±2.1	1±0.08	64±1.3	3±0.12	51±2	107±1.5	45±0.8		
4-NP	200±11.3	276±16.9	175±9.8	173±7	140±4.2	173±7	175±7.3	175±8.4	155±5.7	175±7.9		
Min	-	-	-	-	-	-	-	-	-	-		
Max	200	281	175	173	140	173	175	175	155	175		
Average	124	174	111	121	84	106	90	111	107	108		
Σ_4 BPs	422	596	378	433	282	359	277	380	382	363		

care products, medical devices, packaging, printing inks and coatings, pharmaceuticals and food products, and textiles. These products and materials have been frequently used in everyday life of Tripoli inhabitants and then reached the Tripoli landfill as solid waste. It is interesting to note that PAEs are not chemically but only physically bound to the polymeric matrix. Thus, they can be easily released from the waste containing PAEs to the leachate (Net et al., 2015b). DEHP was the most detected followed by DEP, DnBP, BBP, and DMP with concentration of 1624, 1010, 35, 3, and 1 $\mu\text{g l}^{-1}$ in unit A and 2187, 645, 57, 2, and 1 $\mu\text{g l}^{-1}$ in unit B, respectively. DnOP is under LOQ for both units. Our results were similar to previous studied with references herein (He et al., 2009; Kalmykova et al., 2013; Öman & Junestedt, 2008).

High level of DEHP could be due to the wide and abundant in material and products used in everyday life. DEHP could be persistence in Tripoli landfill since it was detected at high concentration even in the old unit (A) after more than 40 years of waste storage. All types of rubbish including PVC products have been dumped into Tripoli landfill without

sorting, which can be the important source of DEHP. Indeed, DEHP is abundantly used as PCV plasticizers (Kalmykova et al., 2013; Net et al., 2015b). Also, other types of waste might be the sources of PAEs in Tripoli landfill like fibers, lubricants, cosmetics, and pharmaceutical residue (Peijnenburg, 2008). Figure 5 shows the PAEs composition profile in leachate of unit A and unit B from September 2017 to September 2018.

Phenolics and its derivatives composition profile

The results of phenolic compounds are shown in Table 3 and in Fig. 6. The 4-nonylphenol and all BPs, except for BPC, were found in leachate samples in both units. Lower concentrations of the Σ_4 BPs were found in unit A at 277–422 $\mu\text{g l}^{-1}$ (340 $\mu\text{g l}^{-1}$ of average) versus 359–596 $\mu\text{g l}^{-1}$ (419 $\mu\text{g l}^{-1}$) in unit B. The Σ_4 BPs are in the same range of which found in the Japanese landfills (0.009–3600 $\mu\text{g l}^{-1}$, average of 230 $\mu\text{g l}^{-1}$) and higher than which reported for the leachate in south Lebanon (Borjac et al., 2019; Kurata et al., 2008). A minor deviation of BPs concentration was observed between the sampling period

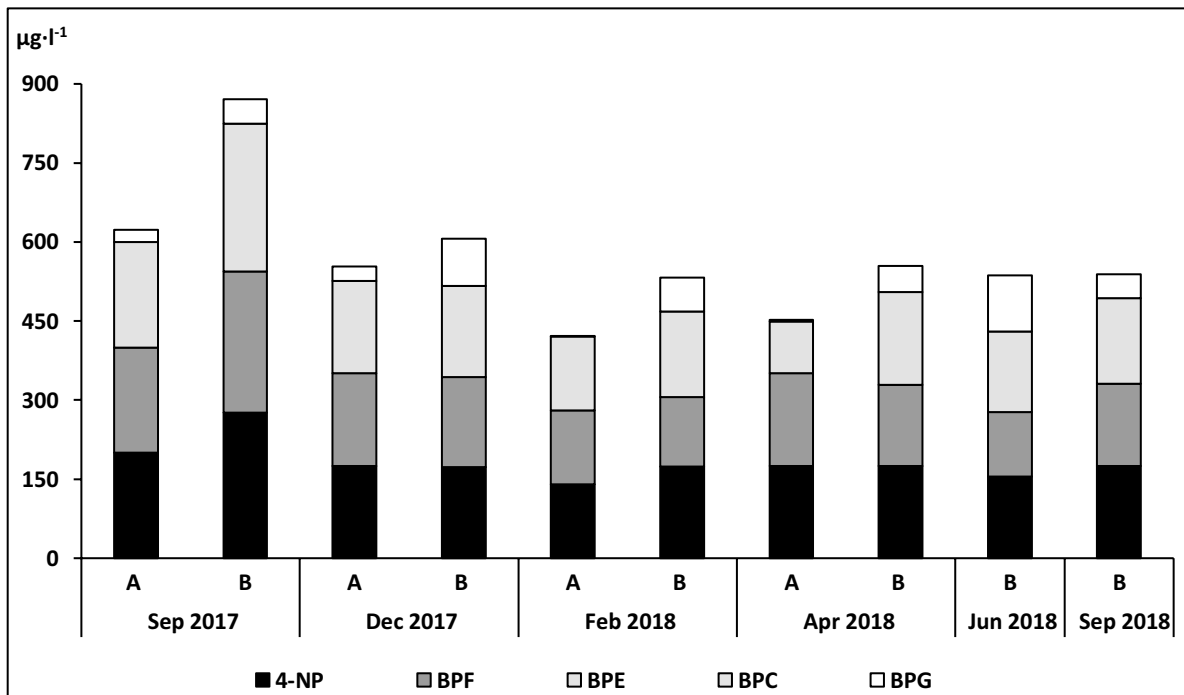


Fig. 6 Phenolic compound profile in leachate of unit A and unit B from September 2017 to September 2018 in $\mu\text{g l}^{-1}$

for both units, which might be due to the seasonal variation (Asakura et al., 2004; Urase & Miyashita, 2003). Different concentrations of BPs were observed between units A and B, which might be due to the stability of BPs with time. The levels of BPs in solid waste landfill leachates depend on the types of waste. The BPs in leachate can be due to the landfilling of many wastes containing BPs like food contact materials (Česen et al., 2016; Liao & Kannan, 2013), digital media, electronic equipment, medical instruments, pipes and toys (Chen et al., 2016), thermal paper (Skledar & Mašič, 2016), dyes, and leather tanning products (Cao et al., 2012).

The 4-nonylphenol were detected at $173 \mu\text{g l}^{-1}$ ($140\text{--}200 \mu\text{g l}^{-1}$) in unit A and $188 \mu\text{g l}^{-1}$ ($155\text{--}276 \mu\text{g l}^{-1}$) in unit B. The detected values are much higher than which found in Japanese landfills at $0.027\text{--}6.4 \mu\text{g l}^{-1}$ (Kurata et al., 2008), and in Sweden at $0.1\text{--}7.3 \mu\text{g l}^{-1}$ (Kalmykova et al., 2013). The high levels of 4-nonylphenol could be explained by the disposal of detergent containers, paints, personal care products, plastics, incombustible, and incineration residues (Kurata et al., 2008). The PBs in Tripoli landfill leachates are thought to be important pollutants and pose a high risk to the surrounding environment and human. For example, BPA can cause a profound effect on organism at low concentration. Indeed, BPA can impact on the endocrine and reproductive systems, on the nervous system and has genotoxicity character and many others (Xing et al., 2022).

Fluxes and potential sources to the Abou Ali River and Mediterranean Sea

The annual rainfall and the surface of landfill were determined at 700 mm/year and 60 000 m² respectively. If the total amount of rainfall on the landfill surface solubilize pollutants containing in the landfill and then, they throw in the Abou Ali River and Mediterranean Sea, the annual volume of leachate throw out is approximately 42000 m³/year. A rough estimation of the pollutant inputs of the Tripoli landfill to the Abou Ali River and then to the Mediterranean Sea can be estimated by the contaminant flux by multiplying the concentrations measured with the corresponding annual volume of leachate. The annual fluxes of xenobiotic organic pollutants thrown out from the Tripoli landfill to the Abou

Ali River and Mediterranean Sea were calculated at 0.23 kg, 0.01 kg, 116.85 kg, 15.93 kg, and 7.58 kg for $\Sigma_{16}\text{PAHs}$, $\Sigma_{28}\text{PCBs}$, $\Sigma_6\text{PAEs}$, $\Sigma_4\text{BPs}$, and 4-NP, respectively. These assessments give an image about the pollution effect of Tripoli landfill on the Abou Ali estuary and Mediterranean Sea. In view of this result, the flow of PAEs, BPs, and 4-NP were significant and can strongly impact the ecosystem functioning due to their potential impact on the endocrine system of wild flora and fauna (Net et al., 2015b; Xing et al., 2022). Moreover, a recent study conducted by our group showed high contamination level of physico-chemical parameter and metallic pollutants in Tripoli leachate. The annual fluxes of TDS, COD, chloride, BOD, TKN, ammonium, TSS, VSS, sulfates, total phosphorus, and nitrates were 711 tons, 577 tons, 253 tons, 207 tons, 137 tons, 96 tons, 58, 38 tons, 25 tons, 7 tons, and 3 tons, respectively. And, the annual fluxes of the $\Sigma_{21}\text{metals}$ achieved 217 tons (Moustafa et al., 2022).

Conclusions

Due to the absence of waste management strategy in Lebanon, environmental pollution generated from uncontrolled landfills become more complicated to manage especially in big cities like Tripoli. The concentration of fifty-five organic pollutants including 16 PAHs, 28 PCBs, 6 PAEs, 4 BPs, and 4-nonylphenol in the leachate of Tripoli landfill has been investigated. The concentration of the selected xenobiotic organic pollutants in leachate collected from old unit is lower than which collected from new unit, which may be due to the age and the transformation of waste in each unit. The concentration of carcinogenic POPs detected the high level in leachate and thus may cause high toxicity. Among the selected compounds, the highest levels were determined for PAEs and followed by phenolic compounds (PBs and 4-nonyl phenol). It is worth to note that PAEs and phenolic compounds are known as endocrine disruptor compounds. The annual fluxes of $\Sigma_6\text{PAEs}$, $\Sigma_4\text{BPs}$, and 4-NP were estimated, respectively, at 116.85 kg, 15.93, and 7.58 kg to the Abou Ali River and Mediterranean Sea. The release of these endocrine disruptor compounds may impact on the ecosystem functioning.

There are no landfills for chemical and hazardous waste in Lebanon, and the results of the present study

reflect the historical use of chemicals and provide additional insight into the nature of wastes eliminated at Tripoli landfill. The disposal of waste containing POPs in a municipal solid waste landfill harms environmental and human health. It is clear that stopping the dumping of hazardous waste into Tripoli landfill is one of the solutions to prevent the damage. The treatment of the leachate becomes the necessity to reduce the impacts on the surrounding environment and particularly the Abou Ali River and coastal area nearby. That summons attention from the national authorities to stop the landfilling of hazardous waste in a municipal solid waste landfill and to stop the drainage of leachate to the water resources and also the citizens to stop any touristic activities and reap seafood in the study area.

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Author contribution Moustafa A and Net S conducted the experiments and analyzed the data. Hamzeh, Baroudi M, and Ouddane B assisted in the experiments and discussed the results. Moustafa A and Net S wrote the manuscript and drew the graphs. All the co-authors revised and approved the final manuscript.

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Data availability The authors declare that all relevant data supporting the findings of this study are included in this article.

Declarations

Conflict of interest The authors declare no competing interests.

Disclaimer The funders had no role in the writing of the manuscript or in the decision and use of data.

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