

Assessment of the pollution concentration of phthalate ester (PAEs) affecting the water quality of Ho Tay Lake

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Abstract: The study presents the evaluation results of typical PAEs (6 PAE indicators) of the PAE family in water samples from Ho Tay Lake, Hanoi. The quality of lake water in general and in Hanoi in particular is influenced by many factors. The sources of treatment in lake water such as restaurant businesses and residential areas nearby, can degrade lake water quality. In order to assess water quality, three methods are still implemented: survey method, data collection, sampling and sample analysis method, and risk assessment method. DEP with the largest value in May 2022 and November 2022 is 0.048 and 0.0846 respectively, in the very low to low-risk range.

Keywords: PAEs; Risk assessment; Composition; Pollution.

1. Introduction

In Vietnam, assessment of the residues of hazardous chemicals in the environment is of great concern, including phthalate esters (PAEs). PAE is commonly used as a plasticizer and additive in a wide variety of products such as PVC resins, some flooring materials and wall coverings. PAE is also used in plastic toys, adhesives, and in plastic films for food packaging [1]. Around the world, PAE pollution in rivers has been studied extensively. For example, there are some published reports on the concentration of PAEs in the Seine River, showed the spread of this group of substances in the environment [2]. PAE is a carcinogenic and teratogenic toxic chemical that is bioaccumulated widely in the environment, which includes: Pseudarthrobacter defluvii E5 and soil agricultural products showing degradability. and mineral PAE for DBP, DEHP. Further research shows that P. defluvii E5 has a promising broad application in microplastic contaminated environments [3]. PAEs have been identified as endocrine disrupting chemicals (EDCs) due to their estrogen and endocrine disrupting properties causing adverse health effects including reproductive system malformations, atherosclerosis [4]. In addition, PAE is teratogenic and carcinogenic, which is metabolized by organisms in the sediment [5], used as a moisture repellent, in filter papers and paper towels, as a substrate in bioactive paper [6], as a popular product of the e-waste recycling area, PAE is released into the environment more aggressively and exposure is higher [7]. PAE is widely used in industrial and household plastic products. The global annual consumption of PAEs was around 6–8 million tons in 2015, while more than a third was in China. In China, about 2.46 million tons of plastic films were used in agriculture in 2018 [8].

PAE was also studied in the accumulation and transport model of 6 PAEs in dicotyledonous vegetables under hydroponic conditions. The PAEs tested included DBP, DEP, DAP, DiBP, DMP, BBP, which resulted in 6 PAEs being absorbed by vegetables from solution, although their accumulation and distribution varied among PAEs [9].

In the world, the occurrence of partitions and six components and partitions of six phthalate esters (PAEs) include: DMP, DEP, DiBP, DBP, BBP, DEHP [10].

The different relationships between flow and low- and high-carbon PAEs may suggest their different sources. DnBPs present a much greater ecological risk than other studied PAE congeners as indicated by its likelihood factor (PAF) and margin of safety (MOS10). The studied PAE congeners pose little health risk to neighboring species [11].

This 5-month study consisted of two parts: (1) monitoring the levels of 11 phthalate ester metabolites (PAEMs) and two beta agonists in human urine samples collected from a small group of people agreed to be tested. Each type of human urine and drinking water contained 183 individual samples. The results of the analysis showed that nine PAEMs were detected in human urine and eight PAEs were detected in drinking water samples [12].

Baseline microplastic pollution and the occurrence, spatial distribution, and ecological risks of phthalate ester (PAE) absorbent microplastics in coastal sandbar sediments of the Gulf of Guinea were investigated. Land-based anthropogenic activities are the major source of MP, while the area's oceanographic characteristics form the main distribution driver [13].

In addition, PAEs with their interactions and with the environment and aquatic animals are still controversial. In this study, PAE is present in many parts of the developing world, and total PAE concentrations even exceed 200 µ/l, exposure to PAE also induces oxidative stress, disorders, and immunosuppression [14].

PAE accounted for 65% of global plasticizer consumption in 2017. DEHP was the most used in 2016 with plasticizers consuming up to 3.1 million tons. PAE is found in plastic and PCP products at concentrations up to several percent by volume [15].

In fact, PAE is a group of flexible, malleable, and elastic compounds of plastics that simplify plastic production [16].

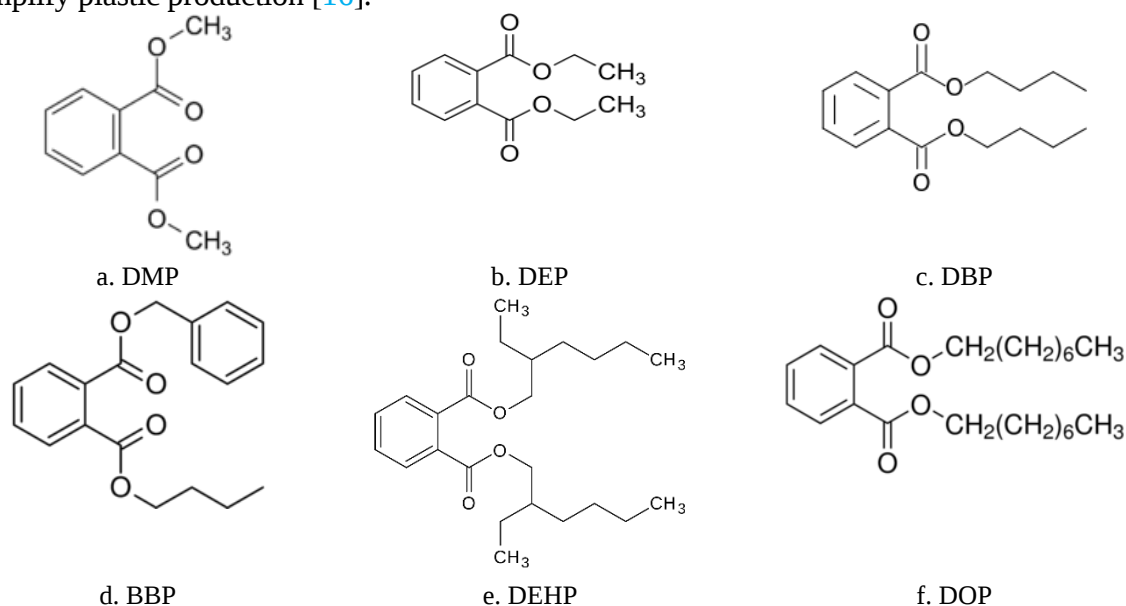


Figure 1. Structure of 6 PAE.

The above studies have contributed to confirm the penetration of PAE into environmental components in some cities in Vietnam and need to be inherited for further assessment of PAE pollution.

Table 1. Some physicochemical properties of PAE.

Symbol	Molecular Formula	S (mg/l)	lg K _{ow}
DMP	C ₁₀ H ₁₀ O ₄	4000	1.6
DEP	C ₁₂ H ₁₄ O ₄	1000	2.5

Symbol	Molecular Formula	S (mg/l)	lg K _{ow}
DBP	C ₁₆ H ₂₂ O ₄	13	4.5
BBP	C ₁₉ H ₂₀ O ₄	0,7	4.9
DEHP	C ₂₄ H ₃₈ O ₄	0,3	7.5
DOP	C ₂₄ H ₃₈ O ₄	0,27	8.1

2. Materials and Methods

2.1. Description of study site

Ho Tay is the largest lake in Hanoi city with an area of 500 ha, circumference of 14.8 km, located in the northwest of Hanoi center, with an area of 500 ha. Ho Tay Lake is a place to drain water when flooded, a place for aquaculture, sightseeing and entertainment. Ho Tay Lake is an area with many beautiful natural landscapes with famous historical and cultural relics such as Tran Quoc Pagoda, Quan Thanh Temple, Tay Ho Palace,... socio-economic development, cultural tourism, as well as an important part of ecological balance and environmental protection of Hanoi Capital. Adjacent to the lake, there are many residential households and agencies, tourist facilities, and services for exploiting the lake's surface. Around the lake, there are 12 main sluices and a drainage system for wastewater entering the lake from surrounding households.

The main culverts are Tau Bay sluice, Cay Si sluice (connecting with Truc Bach), Nhat Tan sluice That is the basic and main cause of environmental pollution in Ho Tay Lake, sampling locations are shown in Figure 2.

Table 2. Location of sampling sites in the Ho Tay Lake.

Numerical order	Location Description	Longitude	Latitude
1.	NM1: Opposite house number 117 Trich Sai street.	105°48'52E	21°02'53N
2.	NM2: Opposite Ly Tu Trong flower garden. No. 5 Nguyen Dinh Thi Street	105°50'05E	21°02'37N
3.	NM3: Tran Quoc Pagoda	105°50'14E	21°02'53N
4.	NM4: Gate of Thang Loi Hotel	105°50'02E	21°03'22N
5.	NM5: Opposite Sheraton Hanoi Hotel, corner of road intersects with Xuan Dieu Street	105°49'53E	21°03'39N
6.	NM6: Opposite house number 35 Quang An street, near Tay Ho district	105°49'11E	21°03'16N
7.	NM7: Opposite Quang Ba guesthouse	105°49'07E	21°58'01N
8.	NM8: Cong Cai – Next to Ho Tay Lake water park	105°49'06E	21°04'22N
9.	NM9: Ve Ho Street, where 2 stone dragons are located, the intersection with Xuan La and Xuan Dinh streets	105°48'45E	21°04'08N
10.	NM10: At the end of Ve Ho Street, it intersects with Lac Long Quan Street. Opposite house number 447 Lac Long Quan	105°48'32E	21°03'34N



Figure 2. Map of sampling locations in Ho Tay.

2.2. Data collection

Using the method to investigate information on the main waste sources of Ho Tay Lake. At the same time, collect information of existing researches in the country and in the world on research subjects.

Sampling locations were selected to represent the space and waste sources of the study area. Based on field survey, nine sampling locations were selected (symbols from M1 to M10), from locations near the beginning of To Ngoc Van Street to the end of Lac Long Quan Street. At each location, a surface water sample was taken in May 2022 (sample symbols from NM1 to NM10). November 2022, repeat 10 samples at 10 locations above (symbols from NM1' to NM10').

2.3. Sampling method and sample analysis

Treat the sample by liquid extraction according to EPA method 3510C and clean the sample according to EPA method 3630C. Samples after extraction and cleaning were injected into a gas chromatograph mass spectrometer (GC–MS).

Take 500 ml of water sample with a measuring cup, pour into a separating funnel, add 60 ml of dichloromethane solvent, cover the separating funnel, unlock the separating funnel to allow the excess vapor in the separating funnel to escape. Lock the separating funnel, shake for 3–5 minutes, stop shaking and let stand for 3–5 minutes for layering, collect the lower separating solvent. Repeat the above process 2 more times. The solvent obtained after shaking 3 times was filtered through anhydrous Na_2SO_4 to remove water. Add 30 ml of Dichloromethane solvent to rinse the separating funnel flask and pass through anhydrous Na_2SO_4 . The solution obtained after passing through anhydrous Na_2SO_4 was evaporated at 60°C until almost dry, changed the n–hexane solvent by adding 30 ml of n–hexane solvent to the flask, shook well and returned to 1–2 ml. The solution after changing the n–hexane solvent and evaporating is aspirated into the cleaning column prepared as follows: Using a locking glass cleaning column, lining the bottom is a layer of glass wool, followed by a layer of Florisin. activated (about 5–7 cm thick), and topped with a thin layer of anhydrous

Na₂SO₄, rinse the column with 50 ml of n-hexane. Aspirate the solution obtained after evaporating in the above step into the cleaning column, rinse the flask with 20 ml of n-hexane solvent and pass it through the cleaning column, using an additional 30 ml of n-hexane solvent to rinse the cleaning column. The cleaning solution was collected into a flask, returned to about 1 ml, aspirated into a graduated cylinder, rinsed the flask with about 2 ml of n-hexane solvent, and collected it completely. Using nitrogen gas to blow gently, concentrate the solution to exactly 1ml, put it into a 1.5 ml bottle with a tight lid, and analyze it by GCMS.

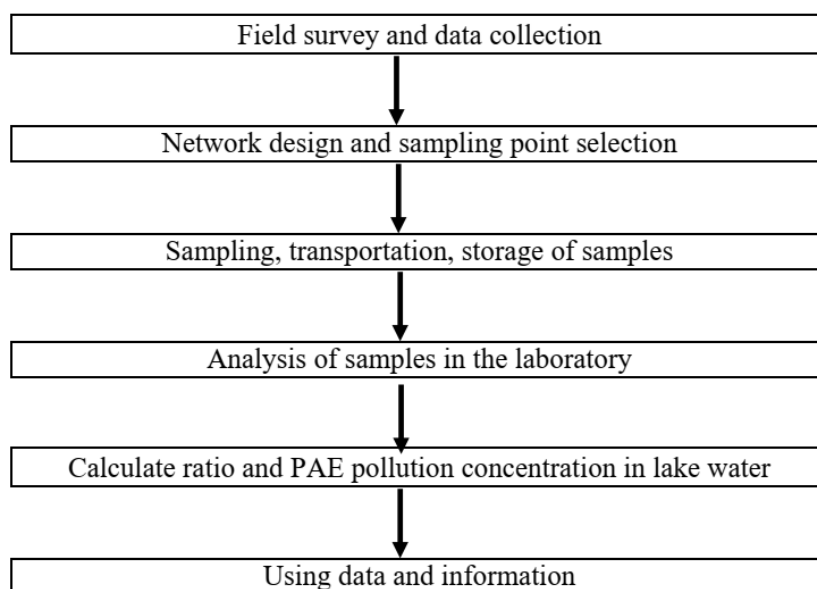


Figure 1. Research structure diagram.

2.4. Methods of risk assessment

Environmental risks due to PAE in Ho Tay Lake surface water were assessed using the risk quotient (Risk quotient, RQ). RQ is calculated according to the formula:

$$RQ = C / MAC \quad (1)$$

where C is the concentration of PAEs in water samples; MAC is the maximum allowable concentration for PAEs in surface water of river. The following qualitative ranking of cancer risk estimates was used to rank the risk as: very low ($RQ \leq 0.01$), low ($0.01 < RQ \leq 0.1$), moderate ($0.1 < RQ < 1$), high ($RQ \geq 1$) [17].

3. Results and discussion

3.1. Assessment of PAE pollution in Ho Tay Lake water

The analysis results showed that all 6 selected PAEs were detected in the lake water samples (Table 3).

Table 3. PAE concentration (µg/l) in Ho Tay Lake water.

Compound	May 2022	November 2022
DMP	0.15–0.87 (0.49±0.22)	0.12–0.78 (0.42±0.26)
DEP	5.68–11.26 (7.65±1.81)	5.69–15.69 (10.38±3.03)
DBP	2.84–2689.00 (272.92±848.92)	3.98–5.99 (4.91±0.66)
BBP	0.02–0.09 (0.06±0.02)	0.01–0.06 (0.02±0.02)
DEHP	13.98–21.63 (20.13±5.19)	15.69–32.10 (21.77±5.01)
DOP	1.36–2.85 (1.90±0.54)	1.21–1.86 (1.49±0.20)
Σ ₆ PAE	24.03–2725.7 (303.15±856.7)	26.7–56.48 (38.99±9.16)

Analysis results in May 2022 obtained a concentration of $\Sigma 6\text{PAE}$ in Ho Tay Lake water.

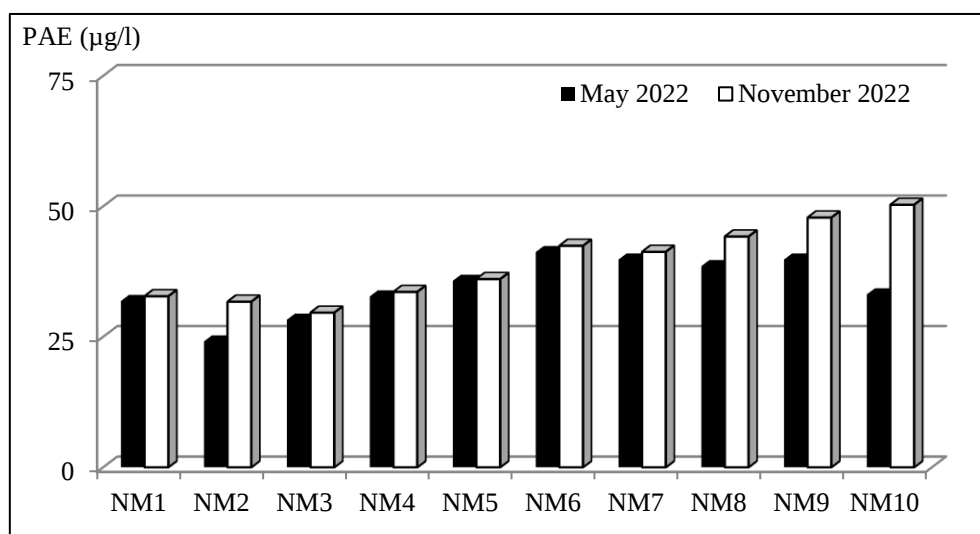


Figure 4. Month-to-month variation of concentrations in Ho Tay Lake water, Hanoi.

Analytical results in May 2022 obtained concentrations of $\Sigma 6\text{PAE}$ in Ho Tay Lake water ranging from 31.86 to 51.23 $\mu\text{g/l}$. Overall value from Table 3 shows that the concentration of $\Sigma 6\text{PAE}$ and its components tended to decrease gradually according to the positions from $\text{NM6} > \text{NM7} > \text{NM9} > \text{NM5} > \text{NM8} > \text{NM4} > \text{NM10} > \text{NM1} > \text{NM3} > \text{NM2}$. The concentration of $\Sigma 6\text{PAE}$ did not decrease gradually along the length of the studied lake, but reached the highest value at NM6 location, which is near Trich Sai road. This may be due to this location, the source of PAE emissions arising from the neighboring houses and residential areas. From these sources, PAE enters wastewater, affecting river water quality at point NM6.

When compared with world studies, the concentration of $\Sigma 6\text{PAE}$ in lake water is also higher than that of Selangor River, Malaysia (0.069–0.688 $\mu\text{g/l}$), Yellow River, China (0.358–59,474 $\mu\text{g/l}$) and river Tama, Japan (< 0.001–4.542 $\mu\text{g/l}$) [18].

Comparing the analysis results in May 2022 and November 2022, it shows that the concentration of $\Sigma 6\text{PAE}$ at each location fluctuates with a slight downward trend. Sampling time in November was selected on a day without rain. The water level in Ho Tay Lake at the two sampling times did not have large fluctuations. In addition, the possibility of PAE entering the lake is mainly from dispersed waste sources around the lake and there is no data to assess the PAE load into the lake. at two sampling times. Therefore, the pollution level and evolution trend of 6 PAE in lake water as well as potential sources of PAE discharge around the lake need to be further studied and evaluated in the coming time, thereby serving as a premise for the assessment. more detail.

3.2. Evaluation of PAE composition in Ho Tay Lake water

PAE composition in lake water is related to the variation and physicochemical properties of each PAE. The mean percentages of PAEs in the lake water samples are presented in Figures 5 and 6.

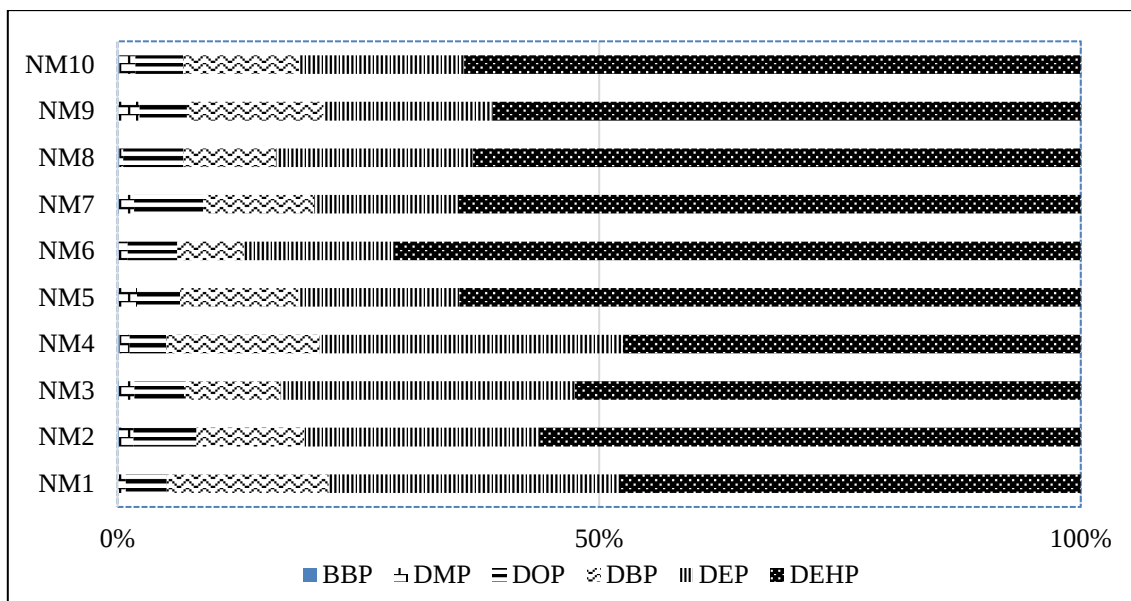


Figure 5. Mean percentage of 6 PAEs in water samples May 2022.

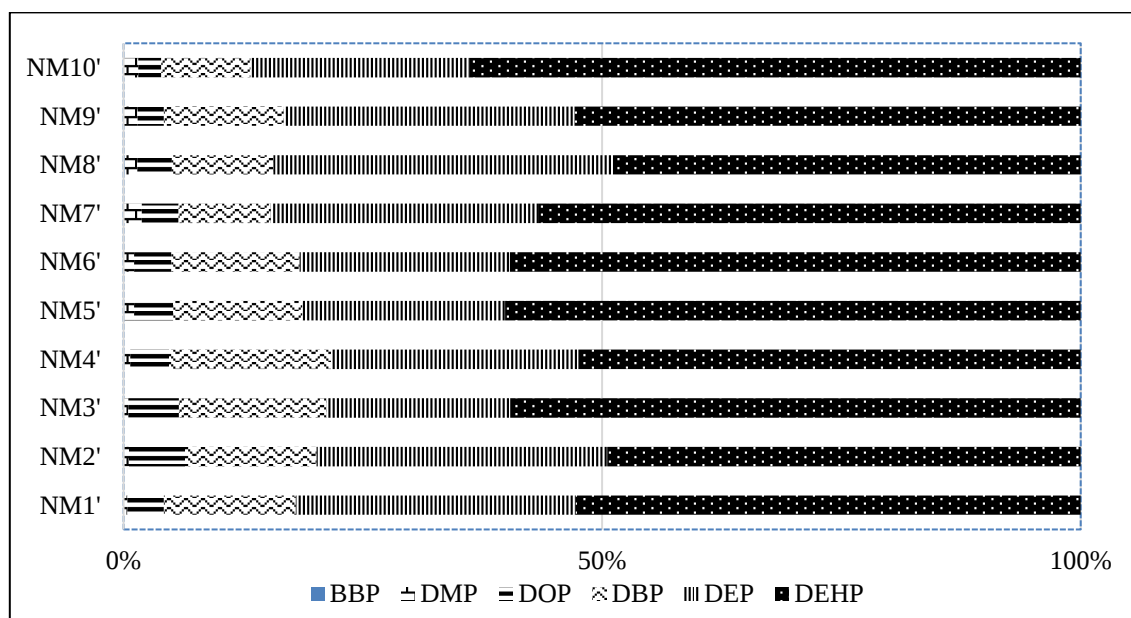


Figure 2. Mean percentage of 6 PAEs in water samples November 2022.

The results showed that the average percentage of 6 PAEs did not change much according to the sampling time of the year. At positions M1 to M5, DEHP, DEP and DBP have a larger mean percentage than the rest of the PAEs. DEHP has the largest percentage of samples. Of the 6 PAEs, DEHP is produced in larger volumes and has more applications than the other PAEs. As a result, the waste source is likely to contain more DEHP, contributing to a larger concentration and percentage of DEHP in the sample. DEP and DBP belong to PAE group with small molecular weight (Low molecular weight PAE, L-PAE), higher water solubility than PAE group with large molecular weight (High molecular weight PAE, H-PAE), so it is easy Soluble in water. H-PAE tends to accumulate in sediments due to its high bioaccumulation capacity. The results of the analysis of the percentage of PAEs in the river water sample showed that there was a match for the physicochemical properties of PAE and the ability to change PAE in the environment. Particularly at position NM6, the percentage of DEHP is smaller than DEP and DBP.

3.3. Risk assessment due to PAH pollution in Ho Tay Lake, Ha Noi

PAE has penetrated the Ho Tay Lake and from there could spread in a wide range. A risk assessment due to domestic PAE is required to consider the potential impact on the Ho Tay Lake environmental quality. The selected MAC values for some PAE compounds in lake water are presented in Table 3 [19]. RQ values at the sampling points are calculated and presented in Table 4.

The results showed the match of the BP5 gene fragment with the 16S rRNA fragment of *Bacillus megaterium* mj1212 with accession number KJ451626.1. Similarly, the gene sequence of strain BP6 matches the gene fragment of strain *Alcaligenes aquatilis* RC43 with accession number MT572474.1 (Figure 8).

Table 3. MPC value of PAEs.

PAE	MPC (mg/l)
DMP	– (*)
DEP	3.7
DBP	10
BBP	–
DEHP	0.19
DOP	–

(*) : no data

Table 4. RQ value of PAEs in water samples.

PAE	RQ May 2022		RQ November 2022	
	Min	Max	Min	Max
DEP	0.0004	0.048	0.0004	0.0846
DBP	0.0002	0.008	0.0002	0.0058
DEHP	0.0002	0.853	0.0005	0.7815

The results of calculating RQ from the results in Table 5 show that the DBP at all sampling points in the river water environment is in a very low level of risk ($RQ \leq 0.01$). DEP at sampling points ranges from very low risk to low risk ($0.01 < RQ \leq 0.1$). Particularly, DEHP at most positions is in the medium risk level ($0.1 < RQ < 1$).

4. Conclusion

Ho Tay Lake, Hanoi contains PAE contamination in a rather wide range. Six representative PAEs were found in lake water samples taken in May 2022 and November 2012. The level of PAE pollution in Ho Tay Lake water is higher than in some rivers in Vietnam and in the world. Evaluation of PAE pollution composition showed that DEHP, DEP and DBP had a higher percentage than the remaining PAEs. The level of environmental risk from PAE contamination varies from very low to moderate, depending on the individual PAE and the sampling point. The above study presents the results of water quality pollution in 2022 to assess the overall status of the water environment of Ho Tay Lake. This is a scientific basis to help managers make plans to conserve and develop organisms.

Currently, several zooplankton species living in Ho Tay Lake are not intrinsic to the lake but an alien species. The snail group is very few. The natural fish composition of Ho Tay Lake is increasingly shrinking, while the composition of fish farming is increasing day by day. Currently, there is only a very small amount of large aquatic plants left in the lake bed, the number of often difficult to quantify. The composition of birds that have been and are present in Ho Tay Lake has identified 43 species belonging to 26 them and 10 sets, decreasing compared to before. The composition of the group of reptiles – frogs in the

study area in Ho Tay Lake has many species are not many and there is no change in species composition over time [20].

Although the water pollution of Ho Tay Lake is not too serious, it has also had a significant impact on the Ho Tay Lake ecosystem. To assess the overall status of the water environment of Ho Tay Lake and come up with effective management solutions to improve water quality, it is necessary to have a plan to monitor and monitor water quality in Ho Tay Lake at many points and regularly.

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