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Determination of benzene derivatives in geothermal waters of Lake Bogoria hot springs

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ABSTRACT

Lake Bogoria geothermal springs are a cultural heritage site in Kenya, offering benefits such as tourism, therapeutic use of hot spring water and recreational activities. However, the hot spring water contains dissolved minerals and potentially harmful organic compounds that can pose risks to humans and the environment, including flamingos. The benzene derivatives present in the water may undergo transformation and bioaccumulation, leading to long-term ecological impacts. This study focuses on the determination of benzene derivatives in Lake Bogoria geothermal springs using gas chromatography - mass spectrometry (GC-MS). Quantitative analysis revealed a total mean concentration of 0.368±0.29 ppm for all identified benzene derivatives. Concentrations were found to be below the recommended limits set by the United States Environmental Protection Agency (US EPA). Among the sampling points, SP4 exhibited the highest total mean concentration (0.120±0.08 ppm). 1,2,4,5-Tetramethylbenzene showed the highest relative abundance (19.97%), while 2-ethyl-1,3-dimethylbenzene had the lowest contribution (1.78%).

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1. Introduction

Studies on geothermal fluid chemistry over the years have mainly focused on inorganic species in order to assess the fluid mineral assemblage and possible environmental health impacts. However, we need to understand the origin, composition, transport and fate of organic matter in geothermal reservoirs [1]. In order to get a proper understanding of fluid chemistry and reservoir conditions, information on the presence and occurrence of dissolved organic matter is essential. It can also lead to the attainment of optimal operating conditions for geothermal power and the development of a better environmental impact assessment [2]. Organic geo-chemistry improves our scientific knowledge of the fluid dynamics and circulation that occur in hydrothermal systems [3]. It is essential to note that there is a significant influence on the composition of geothermal water that results from hydrochemical processes during its ascent to the surface and can lead to environmental degradation [4]. The presence of organic compounds in hydrothermal systems can be attributed to biogenic, abiogenic or thermogenic origins, depending on the process of their formation [5,6]. The leaching of sedimentary organic matter into groundwater in deep subsurface aquifers, coupled with increased biodegradation due to high temperatures, can result in a substantive presence of dissolved organic matter in geothermal reservoirs [7,8]. Organic compounds can also be synthesized abiotically from hydrogen and carbon dioxide in the presence of inorganic rock minerals in reactions similar to the Fischer-Tropsch type of experiments [9].

Experimental studies have shown that, at elevated temperatures, aqueous solutions can enhance the reactivity of monocyclic aromatic compounds, which can lead to various functional group transformations. This can be cascaded to our understanding of the role that these reactions can play in controlling the relative amounts of various organic compounds in geological settings, such as hydrocarbon reservoirs and geothermal systems [10,11]. Increased microbial activity in geothermal power plants resulting from organic compound nutrients can lead to microbial-induced corrosion, scale formation, and the occurrence of biofilms [12]. These phenomena can affect the overall efficiency of geothermal power production [13]. Several studies in geothermal fields around the world have shown a significant presence of various organic compounds that contaminate the environment, including simple hydrocarbons, carboxylic acids, and aromatic compounds [14]. Carboxylic acids have the ability to act as metal complex ligands, which can improve their bioavailability in aquatic environments [15,16].

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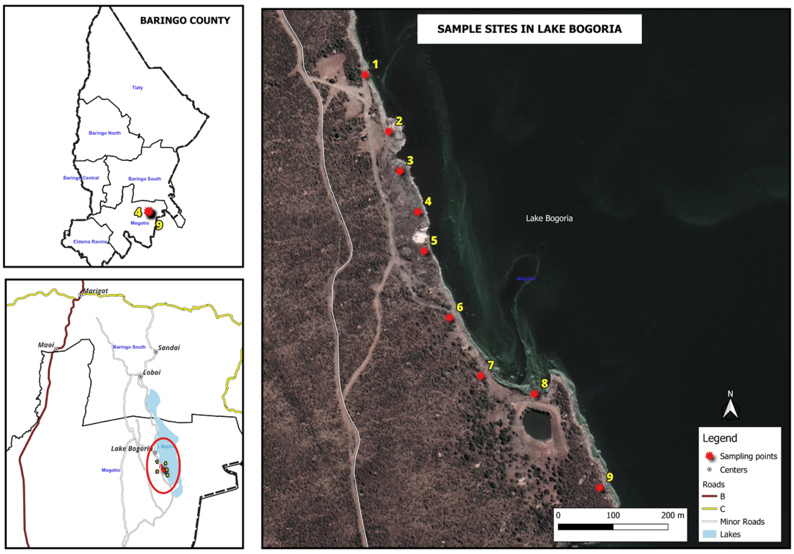

Figure 1. Map of the section of Lake Bogoria where water samples were collected.

On the other hand, aromatic compounds are known to exhibit enhanced stability as a result of resonance. This makes the benzene concentration in most hydrothermal systems relatively high over a wide range of temperature conditions [17]. The presence of these organic compounds in the geothermal fluid matrices can affect the optimal efficiency of geothermal power plants and can as well impact on the terrestrial and aquatic environment. The presence of aromatic compounds in geothermal water can lead to their persistence and bioaccumulation in the food chain [18]. Toxic organometallic complexes can also be produced under natural conditions as a result of interactions between organic compounds and metals. A case in reference is the biotransformation of mercury through the action of microorganisms that leads to the formation of methylated mercury, which is lipid-soluble, extremely poisonous and bio-accumulative in the aquatic ecosystem [19,20]. It should be noted that hydrothermal gases have been found to contain relatively high amounts of benzene that is capable of undergoing various functional group interconversions to form other aromatic derivatives that can be of health concern [21]. The long-term presence of organic contaminants and the lack of mitigation strategies can affect the flora and fauna of the surrounding ecosystem [22,23].

The Lake Bogoria hot springs are located within the saline alkaline basin of the lakes of the Great Rift Valley [24]. This region is considered to be a rich carbon cycling sink, and some studies have shown the presence of unique carbon reservoirs in the lakes, consisting of aromatic and saturated hydrocarbons derived from microbial activity and photodegradation [25]. The origin and formation of dissolved organic matter is a complex process that produces a mixture of diverse molecules whose transport and bioavailability is quite vast and varied [26]. The Lake Bogoria geothermal regime has been designated for future geothermal power production and currently serves as a tourist attraction center since it is a sanctuary for flamingos and other rare bird species. In addition, it is a conservation game reserve that is home to endangered animal species [27]. The influence of hot spring water on the lake water quality is of paramount importance, since it can affect the flora and fauna of the immediate ecosystem [28]. In the recent past, there have been unusual flamingo deaths whose cause is yet to be established. Such long-term occurrences can affect the hospitality industry. Coupled with this is the safety of the water used in recreational activities at the Lake Bogoria hotel [29,30]. Organic contaminants in water are a growing concern due to their high persistence and toxicity and hence the need to monitor their sources and presence in water ecosystems [31].

The objective of the present study was to explore the presence of alkylbenzenes in selected hot springs in Lake Bogoria geothermal springs. These compounds are toxic, carcinogenic and mutagenic even at low concentrations. Alkylated compounds can be converted to phenols and catechols, which are harmful ecotoxins whose toxicity is related to their hydrophobicity and their ability to generate organic radicals and reactive oxygen species [32]. Their peroxidative capacity can adversely affect physiological processes in living organisms, including digestion and the central nervous systems. Benzene derivatives are endocrine disrupters and can cause a change in bird species breeding patterns, as well as cause morbidity and mortality [33,34]. Furthermore, they are known to bioaccumulate and bioconcentrate in food chains at higher trophic levels with disastrous effects on humans and natural ecosystems [35]. Limited studies have been carried out to determine the contribution of hot springs to the dissolved organic matter in East African saline lakes and hence the need for this study. This study focused primarily on exploring the presence of aromatic hydrocarbons in geothermal springs located in the geothermal prospect of Lake Bogoria. The results of the study of the levels of benzene derivatives in geothermal waters in Lake Bogoria are expected to provide information on the presence of these toxicants and the possible fate of the contaminants in the potential geothermal resource and hence create awareness about the hidden organic hazards of hydrothermal water. The methodology applied in this study is reliable and can be replicated in similar geological settings for comparative studies.

2. Experimental

2.1. Sample collection

The sampling site was located at an altitude of 1,094 m above sea level with coordinates (0°13'42.2" N, 36°05'35.3" E) and (0°13'39.174" N, 36°05'35.38" E). Nine water/steam samples (500 mL each) were collected in triplicate from randomly identified spring sites (SP1 to SP9) near the shores of Lake Bogoria at intervals of 2 hours between 8:00 a.m. and 4:00 p.m., as illustrated in Figure 1. The samples were transferred into well-labeled polythene containers and kept in cold boxes at

4 °C prior to transportation to the Chemistry Laboratory of Egerton University for further treatment and analysis.

2.2. Liquid-liquid extraction of benzene derivatives

Organic compounds in the water samples were extracted using a liquid-liquid extraction method [36]. Briefly, 150 mL of each water sample was transferred into a separatory funnel and extracted with 75 mL of dichloromethane. The mixture was shaken vigorously and allowed to settle for phase separation. The organic phase was collected and the extraction was repeated to ensure maximum recovery. The combined organic extracts were dried over anhydrous sodium sulfate and concentrated using a rotary evaporator. The extracts were then transferred into amber vials and stored prior to GC-MS analysis.

2.3. GC-MS analysis of benzene derivatives

GC-MS analysis of organic extracts was performed using a Gas Chromatography Mass Spectrometry system (Agilent 5977A GC/MSD) [37]. Separation was achieved on an RTX-5 fused silica capillary column (5% phenyl, 95% dimethyl polysiloxane). The samples were introduced into the GC–MS using an autosampler. Helium was used as the carrier gas at a constant flow rate of 1 mL/min. The analysis was conducted in total ion current (TIC) mode, scanning over an m/z range of 15-140 with a scan rate of 0.2 s. The oven temperature program was set as follows: initial temperature of 40 °C held for 2 min, increased at 10 °C/min to 80 °C, then ramped at 15 °C/min to 150 °C and held for 10 min. Alkyl benzene compounds were identified based on their mass spectra and retention indices (Kovats index) [38] by comparison with the NIST library database [39]. Relative concentrations were estimated from the corresponding peak areas.

2.4. Multivariate analysis of benzene derivatives in water samples

Multivariate analysis of benzene derivative compounds was carried out using principal component analysis (PCA) and hierarchical cluster analysis (HCA) [40]. Statistical evaluation of their distribution at different geothermal hot spring sampling points was performed using SPSS statistical software (version 25.0) [41]. All GC-MS measurements were conducted in triplicate and the results were expressed as mean ± standard deviation. These analyses were performed using quantified concentrations of benzene derivatives obtained from geothermal hot spring water samples.

3. Results and discussion

3.1. Benzene derivatives in Lake Bogoria geothermal hot springs waters

In this work, the water samples obtained from the nine sampling points were analyzed for the concentration profiles of poly-substituted alkylbenzenes as reported in Table 1. The results show that a total of 13 benzene derivatives were analyzed, although they were not present in all water samples. The methyl poly-substituted compounds showed higher abundance values compared to ethyl substituted compounds as shown in Table 1, which can be attributed to steric factors, the relative stability of a methyl substituent compared to an ethyl group, electronic interactions and the energy of formation [42]. This suggests that ethylbenzenes are thermodynamically less stable and are also easily converted to other compounds [43,44], resulting in their relatively low abundance. In this study, 1,2,4,5-tetramethylbenzene with methyl substituents only, had the highest mean concentration of 0.119±0.08 ppm (19.97% abundance), which can be attributed to steric factors, energetics, and resonance stabilization by substituents in symmetric positions [45,46]. In contrast, 2-ethyl-1,3-dimethyl benzene was the least detected with a mean concentration of 0.028±0.01 ppm accounting for 1.78% of the alkyl benzenes analyzed.

There is a wide variation in the concentration of alkylbenzenes in the sampled water from all the collection points (Table 1). For example, sample SP4 showed the highest mean concentration of 0.120±0.08 ppm while SP9 exhibited the lowest mean concentration of 0.033±0.02 ppm. This can be attributed to the nature of origin of each spring, the chemical interactions of geothermal fluids and the reservoir temperatures [47,48]. Samples SP4, SP5, and SP8 were characterized by relatively higher mean concentrations (≥ 0.100 ppm), in contrast to the other sampling points which displayed lower values. Evaporation and volatility of alkyl benzenes could also have contributed to these variations as the low molecular compounds are easily lost at high boiling temperatures of the thermal waters [49] According to this study, the mean concentration value for the sampled points was 0.368±0.29 ppm, underscoring the presence of alkyl benzenes.

Table 1 shows that 1,2,3-trimethylbenzene was found to have a concentration of 0.134±0.05 ppm in SP4, which was much higher than those reported in any other sample. On the other hand, 1-ethyl-2-methylbenzene was highest in SP2 with a concentration of 0.056±0.02 ppm. It is also observed that the concentration of 1, 2, 4-trimethylbenzene was highest in SP4 (0.202±0.04 ppm) contrasting other samples. 1,2,4,5-tetra methylbenzene was found to have the highest concentration in sample SP8, with a mean concentration of 0.233±0.06 ppm compared to other sampled points. 1,2,3,5-tetramethyl benzene and 1-ethyl-3,5-dimethylbenzene gave significant concentrations of 0.246±0.08, 0.246±0.05, respectively, in SP5.

Although the concentrations of naphthalene in samples SP4 (0.03±0.01 ppm), SP6 (0.022±0.01 ppm) and SP8 (0.046±0.01 ppm) were low, it can bioaccumulate in the human body, aquatic animal, and flamingos, which are in constant interaction with the lake and geothermal waters, which may cause serious effects such as adverse toxicities and cancer cases [50] Sample 7 contains 1,2,3,4-tetramethylbenzene at a concentration of 0.188±0.04 ppm, representing a significant amount in the sample. 1,2,3,5-tetramethylbenzene is predominant in SP3 and SP5 samples at concentrations of 0.213±0.06 and 0.246±0.08 ppm, respectively. Sample 8 contained the highest amount of naphthalene at 0.046±0.01 ppm. In general, all the sites sampled showed the presence of benzene derivatives, with sample SP4 being highly contaminated at 0.120±0.08 ppm while sample SP9 is least contaminated at 0.033±0.02 ppm. Variations in outcomes can be attributed to various factors, including the origin of geothermal water, temperature and chemical interactions of the water with other matrices [51].

Alkylated benzenes have a characteristic feature of higher relative concentrations in comparison to the poly aromatic hydrocarbons such as naphthalene whose occurrence was quite low as shown in Table 1. Alkylbenzenes are usually precursors for the formation of naphthalene and the results show that there is a low conversion rate. This is due to the complex pathways involved, kinetics and thermodynamic factors [52].

3.2. Environmental implications of aromatic hydrocarbons and their occurrence

The concentrations of organic compounds in the samples were generally low, as shown in Table 1. However, the cumulative effect of these compounds, even at low concentrations, could potentially pose an environmental concern over time. For example, alkylbenzenes are bioaccumulative in the ecosystem and can be transformed into more potent derivatives, such as halogenated alkyl benzenes, which are more toxic to the environment [53].

Table 1. Concentration profiles of benzene derivatives in the Lake Bogoria geothermal system.

No	Benzene derivative	Rt (min)	Concentration (ppm) *									Total mean conc. (ppm)	%
			SP1	SP2	SP3	SP4	SP5	SP6	SP7	SP8	SP9		
1	1,2,3-Trimethylbenzene	5.76	0.052 ±0.02	0.075 ±0.03	0.046 ±0.02	0.134 ±0.05	0.077 ±0.03	0.073 ±0.01	0.060 ±0.03	-	-	0.074 ±0.03	10.81
2	1-Ethyl-2-methylbenzene	6.39	0.036 ±0.01	0.056 ±0.02	-	0.013 ±0.01	-	-	-	-	0.020 ±0.01	0.032 ±0.02	2.62
3	1,2,3,4-Tetramethylbenzene	8.88	0.025 ±0.01	0.094 ±0.04	-	0.269 ±0.06	0.096 ±0.03	0.062 ±0.03	0.188 ±0.04	0.084±0.03	0.064 ±0.02	0.110 ±0.08	18.46
4	2-Ethyl-1,4-dimethylbenzene	7.74	-	0.041 ±0.01	-	0.107 0.03	0.030 ±0.02	0.025 ±0.01	0.033 ±0.02	0.051±0.02	-	0.048 ±0.03	6.00
5	2-Ethyl-1,3-dimethylbenzene	7.80	-	-	-	-	0.037 ±0.01	-	0.024±0.01	-	0.024 ±0.01	0.028 ±0.01	1.78
6	1,2,4-Trimethylbenzene	5.82	-	-	0.057 ±0.02	0.202 ±0.04	-	0.095±0.04	-	0.089 ±0.03	0.024 ±0.02	0.093 ±0.07	9.77
7	4-Ethyl-1,2-dimethyl benzene	8.01	0.044 ±0.01	-	0.019 ±0.01	0.115 ±0.04	-	-	-	-	-	0.059 ±0.05	3.72
8	1-Ethyl-2,3-dimethylbenzene	7.94	-	-	0.013 0.01	-	0.086 ±0.03	0.051 ±0.03	0.049 ±0.01	-	-	0.050 ±0.03	4.16
9	1-Ethyl-2,4-dimethylbenzene	7.83	-	0.034 ±0.01	0.024 ±0.02	0.054 ±0.02	-	0.026 ±0.01	-	-	-	0.035 ±0.01	2.89
10	1,2,4,5-Tetramethylbenzene	8.82	0.048 ±0.02	-	0.063 ±0.03	0.183 ±0.06	0.096 ±0.04	0.221 ±0.05	0.079 ±0.03	0.233 ±0.06	0.032 ±0.01	0.119 ±0.08	19.97
11	1,2,3,5-Tetramethylbenzene	8.97	-	-	0.213 ±0.06	0.092 ±0.02	0.246 ±0.08	-	-	-	-	0.184 ±0.08	11.52
12	Naphthalene	10.76	-	-	-	0.030 ±0.01	-	0.022 ±0.01	-	0.046±0.01	-	0.033 ±0.01	2.05
13	1-Ethyl-3,5-dimethylbenzene	7.98	-	-	-	-	0.246 ±0.05	0.053 ±0.01	-	-	-	0.150 ±0.14	6.25
	Total mean conc. (ppm)		0.041 ±0.01	0.060 ±0.03	0.062 ±0.04	0.120 ±0.08	0.114 ±0.08	0.070 ±0.06	0.072 ±0.06	0.101 ±0.07	0.033 ±0.02	0.368 ±0.29	100.00

*** '-':** not detected, RT: retention time.

Table 2. Varimax-rotated loadings for benzene derivatives in geothermal water.

Water sample	Principal components		
	PC1 (46.614%)	PC2 (18.446%)	PC3 (12.179%)
SP1	0.261	0.432	-0.240
SP2	0.850 *	-0.071	-0.244
SP3	-0.235	0.163	0.819 *
SP4	0.482	0.740 *	0.114
SP5	0.140	-0.173	0.896 *
SP6	0.834 *	0.471	0.086
SP7	0.865 *	0.391	0.039
SP8	0.041	0.931 *	0.038
SP9	0.659 *	0.565 *	-0.057
Initial eigenvalues	4.195	1.660	1.096

* The loads with an asterisk indicate a greater influence on the principal components.

Aromatic hydrocarbons, being volatile organic compounds (VOCs), can also contribute to the formation of secondary organic aerosols and peroxides [54-56], which can negatively affect air quality and cause respiratory issues in the geothermal environment.

The United States Environmental Protection Agency (US EPA) and the World Health Organization (WHO) have established limits for benzene and some of its derivatives because they are considered as priority pollutants. For example, the recommended occupational exposure limit for 1,2,3-trimethylbenzene is 25 ppm for an 8-hour period [57]. For drinking water, the limits are 5 ppb for benzene, 0.7 ppm for ethylbenzene, 1 ppm for toluene, and 10 ppm for xylenes [58]. However, many polyalkylbenzenes do not yet have established toxicity criteria, highlighting the need for further in vitro and in vivo studies to evaluate their potential hazards [59].

Gas chromatography has been widely used to characterize organic compounds from various geothermal sites worldwide. For example, steam condensate and water samples from hydrothermal systems in Kamchatka, Russia, contained 14.4% aromatic hydrocarbons and 3.7% halogenated aromatic hydrocarbons [60]. Similar observations have been reported at the Mutnovsky and Annensky geothermal sites, where aromatic compounds were present in considerable amounts [61]. Quantitative fluid inclusion analyzes from the Coso and Karaha-Telaga Bodas geothermal systems indicated that benzene concentrations and alkane/alkene ratios were directly related to hydrogen fugacity [62]. Against this background, the present investigation focused on benzene derivatives in Lake Bogoria, given their potential environmental impact.

3.3. Principal component analysis

Factor analysis was performed on the dataset of 13 benzene derivatives in geothermal hot spring water using the Varimax rotation method [63] followed by Kaiser normalization [64]. The principal components (PC) were selected based on eigenvalues greater than 1. The PCA results identified three components that collectively explained 77.239% of the total variance (Table 2), suggesting that the benzene derivatives may have come from three distinct sources.

The first principal component (PC1) accounted for 46.614% of the variance, the second component (PC2) for 18.446%, and the third component (PC3) for 12.179%. PC1 exhibited high positive loads for water samples SP2 (0.850), SP6 (0.834), SP7 (0.865), and SP9 (0.659), while SP3 (-0.235) contributed a negative load. PC2 showed significant positive loadings for SP4 (0.740), SP8 (0.931), and SP9 (0.565), with SP2 (-0.071) and SP5 (-0.173) recording low negative loadings. PC3 was characterized by high positive loadings for SP3 (0.819) and SP5 (0.896), while SP1 (-0.240), SP2 (-0.244) and SP9 (-0.057) had low or negative loadings.

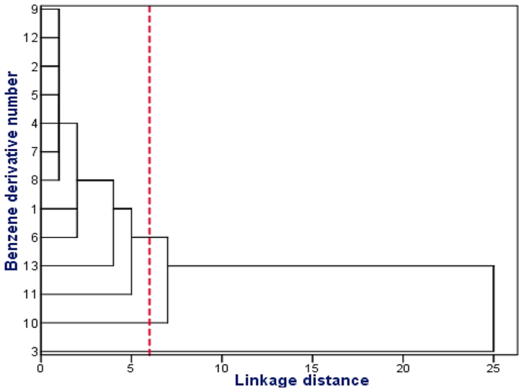

Figure 2. Dendrogram of benzene derivatives in water using centroid linkage hierarchical cluster analysis (HCA).

Table 2 presents the Varimax-rotated loadings, where bold values indicate the loadings with the greatest influence on each principal component. The corresponding initial eigenvalues were 4.195, 1.660, and 1.096 for PC1, PC2, and PC3, respectively. These results demonstrate that the distribution of benzene derivatives in geothermal water samples is influenced by three principal sources, each principal component representing a distinct pattern of variation among the sample sites.

3.4. Hierarchical Cluster Analysis

Hierarchical cluster analysis (HCA) [65] was performed using the centroid clustering method [66], with the squared Euclidean distance used as the similarity measure [67]. This approach was used to identify correlated clusters of benzene derivatives, providing insight into their distribution patterns in geothermal hot spring water. The HCA results are presented in the dendrogram shown in Figure 2. At a centroid linkage distance of approximately 6, three clusters were identified, capturing most of the variation among benzene derivatives across the sampling points at different temperatures between 8:00 am and 4:00 pm on the collection day. Significant correlations were observed among the benzene derivatives within the same clusters, and these correlated patterns were consistent with the principal component analysis (PCA) results, indicating that PCA and HCA provide complementary insights in this study. As shown in Figure 2, derivatives 10 (1,2,4,5-tetramethylbenzene) and 3 (1,2,3,4-tetramethylbenzene) formed individual correlated groups, as they were present in all water samples except SP2 and SP3, respectively. This indicates that these compounds were absent at those specific sampling sites. Concentration levels of these derivatives were observed to vary throughout the day, generally increasing after midday, suggesting that their levels are influenced by fluctuations in temperature (Table 1). Other benzene derivatives formed additional correlated groups, reflecting their absence in certain samples. These results are consistent with the PCA findings regarding the number of correlated groups, suggesting that the analysed benzene derivatives may originate from three natural sources: flood events that can submerge the geothermal hot springs, contributions from surrounding soil profiles, and elevated subsurface temperatures that lead to thermogenic formation from geothermal activity, thereby contributing to their significant presence in the hot spring water.

4. Conclusions

This study demonstrates that the presence of organic contaminants in geothermal environments cannot be ignored. Monitoring of organic compounds in hydrothermal systems is essential to determine their concentrations and assess their potential threats to the ecosystem, particularly during the utilization of geothermal resources for hospitality, horticulture, and domestic purposes. The results revealed a considerable presence of aromatic organic contaminants, with a total mean concentration of 0.368±0.29 ppm for alkyl benzenes, which is below the recommended limits of the US EPA. Although this is below the regulatory limits, these compounds are toxic and potentially carcinogenic and their presence warrants regular monitoring. Due to the continuous flow of geothermal water into Lake Bogoria, bioaccumulative alkylbenzenes may impact the lake's flora and fauna over time. Therefore, strategies for degrading these contaminants must be developed, especially during large-scale exploitation of geothermal resources for energy production. Such measures would help minimize occupational health risks for geothermal plant workers and reduce potential environmental contamination. More research is recommended on organic contaminants and remediation strategies, including bioremediation using thermophilic bacteria. Additionally, the isolation and characterization of other organic compounds in geothermal water should be pursued, along with the analysis of surrounding soils and vegetation to identify potential sources of benzene derivatives. In conclusion, the development of geothermal energy as a green and sustainable resource must be accompanied by rigorous environmental monitoring and assessment to mitigate potential public health and ecological risks.

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Sample Availability: Samples of the compounds are available from the corresponding author upon reasonable request.

CRediT authorship contribution statement CR

Conceptualization: Joshua Kiprotich Kibet; Methodology: Anzelim Eliwa Sunguti, Joshua Kiprotich Kibet; Software: Joshua Kiprotich Kibet; Validation: Thomas Karanja Kinyanjui; Formal Analysis: Joshua Kiprotich Kibet; Investigation: Anzelim Eliwa Sunguti, Joshua Kiprotich Kibet,Thomas Karanja Kinyanjui; Resources: Anzelim Eliwa Sunguti; Data Curation: Joshua Kiprotich Kibet; Writing - Original Draft: Anzelim Eliwa Sunguti; Writing - Review and Editing: Joshua Kiprotich Kibet, Thomas Karanja Kinyanjui; Visualization: Joshua Kiprotich Kibet; Funding acquisition: Anzelim Eliwa

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