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Assessment of Total Mercury in Public Drinking Water: A Case Study of Sana'a City, Yemen

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Abstract

Mercury (Hg) contamination in drinking water poses a significant health risk, as acknowledged by the World Health Organization (WHO). However, research on Hg contamination in potable water sources in Yemen is lacking. This study assessed Hg levels in potable water wells across different regions of Sana'a City, aiming to provide insights for future monitoring and policy development. Water samples from 58 wells in Sana'a City were analyzed using the validated DMA-80 method. Hg concentrations in all samples were below the WHO and Yemeni Organization for Standards Metrology and Quality Control threshold level (1 µg/L). Notably, the investigated wells exhibited some of the lowest levels of Hg contamination compared to reported values in literature from other regions globally. Moreover, no significant correlation was found between well depth and Hg concentration in the dataset. These findings establish a crucial baseline for monitoring efforts and policy development to mitigate Hg contamination in Sana'a's drinking water supply. The dataset from this study offers valuable information for policymakers, aiding their efforts to combat Hg contamination, ensure safe drinking water, support effective health measures, and enhance understanding of Hg contamination in Sana'a City, Yemen.

Keywords: Total Mercury, DMA, Public drinking water, Environmental health, Wells water contamination.

Introduction

Mercury (Hg) naturally occurs on Earth and poses a significant global threat as it is highly toxic, persists in the environment, and accumulates in living organisms. Hg pollution in drinking water occurs through both natural releases, which contribute approximately 5.2 thousand tons of emissions into the atmosphere annually, and various human activities are responsible for roughly 2.3 thousand tons of emissions each year [1]. These human activities include waste incineration, mining, metallurgy, chlorine alkali industries, coal combustion, metal refining, dental amalgam restoration, and unauthorized mercury storage and release. Among these activities, mining and stationary coal combustion are the major contributors, accounting for 37.5% and 21% of total emissions, respectively [2].

The developed countries have implemented stricter regulations to control Hg emissions. However, developing and underdeveloped countries often lack the necessary infrastructure and resources for effective Hg management and the risk of exposure is higher in these countries [3]. Hg exists in different forms, and while all forms are poisonous, organic Hg is particularly hazardous [4]. Dimethyl mercury (Me_2Hg) and methyl mercury (MeHg) are the more volatile and bio-accumulative forms that can result from the conversion of any form of Hg [5]. MeHg constitutes 70 – 80% of blood Hg in the U.S. population [6]. Exposure to Hg can lead to damage in various organs and systems, including the brain, liver, kidneys, and reproductive organs, with potential harm to the fetus during pregnancy. Therefore, accurate assessment of Hg contamination is crucial for its control. In response to the global threat posed by Hg to the environment and human health, the Minamata Convention was established under the United Nations Environmental Program. This international agreement aims to protect human health and the environment by addressing anthropogenic emissions and releases of Hg and its compounds [7]. The level of Hg in groundwater varies globally and is influenced by factors such as its oxidation state and interactions with contaminated surroundings like soil, sediments, and water flow paths [1, 8, 9].

Reports from different regions have indicated varying levels of Hg contamination in drinking water. For example, studies have reported Hg contamination in drinking water from China (4.5-3200 µg/L) [10], Colombia (0.0025-1000 µg/L) [11], Baghdad/Iraq (5.1 µg/L) [12], Sudan (0.26 µg/L) [13], Tinga Ghana (1-259 µg/L) [14], and Makassal, Indonesia (2050 µg/L) [15]. The World Health Organization (WHO) guidelines state that the acceptable level of Hg in drinking water is 1 µg/L or less [16], which requires analytical methods with high sensitivity and selectivity. Several methods, including cold vapor atomic absorption spectrometry (CV-AAS), inductively coupled plasma mass spectrometry (ICP-MS), X-ray fluorescence (XRF), gas chromatography cold vapor atomic fluorescence spectrometry (GC-CV-AFS), and direct mercury analyzers (DMA), have been utilized for Hg detection in water [17-19]. Although the complexity and lack of selectivity of conventional analytical processes are significant constraints, other parameters, such as the sample matrix's characteristics, sample preparation steps, the quantity of mercury contamination, the length of the analysis time, operating cost, reproducibility, sensitivity, recovery, and precision, must be taken into account [17]. This presents a difficult challenge in the selection of the analytical method for Hg determination in water samples.

DMA offers advantages in terms of sensitivity, speed, simplicity, low operational cost, high analytical throughput, and reliable results [15, 18, 19]. This method utilizes thermal sample decomposition, catalytic Hg reduction, and gold system fusion to trap and pre-concentrate Hg vapor, followed by desorption and measurement by atomic absorption spectrometry [20]. However, the success of DMA analysis depends on the sample matrix, necessitating rigorous method validation for each sample type. While certified standard materials are typically used for calibration curves, it is recommended to include an external calibration procedure using an aqueous standard solution and adhere to a quality control process for method validation or verification [5].

Limited research has been conducted for the assessment of Hg concentrations in public drinking water in Yemen, a country that has ratified the Minamata Convention [21, 22]. Specifically, no available data exists on Hg contamination in the drinking water of Yemen's capital city, Sana'a. The Dhahaban District in Sana'a is considered a water sanctuary supplying local wells. However, due to urbanization, the use of soak pits for sewage, lax factory regulation, and potential exposure to pollution, the water sanctuary may be at risk of Hg contamination in its drinking water wells. Therefore, this study aims to investigate the levels of Hg in the drinking water of Sana'a, Yemen, specifically focusing on the Dhahaban District. The DMA is employed as the analytical method for detecting and quantifying Hg concentrations in drinking water samples. Method validation parameters will be evaluated to ensure the reliability and reproducibility of the method. The obtained results are compared against the WHO and the Yemen Standardization and Metrology Organization (YSMO) guidelines. The findings from this study offer valuable insights into the presence of Hg in the drinking water of Sana'a City, Yemen. In addition, these results can aid in evaluating the potential risks to human health and the environment, as well as guide the implementation of suitable measures to reduce Hg pollution and safeguard the well-being of the local population.

Materials and Methods

Chemicals and Instruments

The Hg standard solution 1000 mg/L in 10% HNO_3 was purchased from Perkin Elmer, USA. Hydrochloric acid ACS reagent, 37% Honeywell Fluka, Spain was used. Deionized water was produced in the lab with a resistivity of 18.2 µΩ.cm using Direct Q3-Millipore - USA. The Dual-cell Direct Mercury Analyzer DMA-80 evo (Milestone, Waltham, Sorisole, Italy) was used to measure the total Hg in water wells.

Procedure for Hg Analysis

In general, DMA-80 direct Hg analysis involves the application of the principles of atomic absorption spectrophotometry, thermal decomposition, catalytic conversion, and amalgamation. By employing controlled heating phases, a sample introduced into a quartz tube was initially dried and subsequently thermally decomposed. The decomposition products were conveyed via an uninterrupted stream of oxygen across a high-temperature catalyst bed, which ensnares halogens, nitrogen, and sulfur oxides. Following the reduction of every Hg species to Hg^0, the Hg was transported to a gold amalgamator along with reaction vapors, where it was selectively captured. The system evacuates all non-Hg vapors and byproducts of decomposition through the consistent circulation of gas. Following this, all Hg that was entrapped was discharged into the single beam, fixed wavelength atomic absorption spectrophotometer via heating the amalgamator. The absorbance was determined noted at 253.7 nm, concerning mercury concentration [18, 19, 23].

For Hg analysis in water samples, 0.5 g of water was directly taken quartz boat using an analytical balance (Electronic scale, Germany) and mixed well to achieve homogeneity. The sample was dried inside the DMA-80 for 90 s at 200 °C. The furnace temperature was then raised to 650 °C for 120 s to allow the sample degradation. All forms of Hg were trapped quantitatively as vapor in the gold amalgamator, delivered by the carrier gas (flow rate 120 mL/min) to the catalyst tube, and converted to Hg^0. The absorption was measured using atomic absorption at a wavelength of 253.65 nm. After the resulting Hg absorption was integrated by the Easy Doc software (Milestone Inc., Bergamo, Italy), the total Hg content was then given in (µg/L). The DMA-80 was flushed through three consecutive times before each batch of sample analysis. The average of three consecutive studies of empty boats was used to gauge the effectiveness of quartz boat cleaning [18, 22, 23].

Sampling Sites, Sample Collection, and Preparation Procedure for Hg Analysis

As shown in Fig. 1, 58 wells are located throughout the city of Sana'a. The samples were collected according to established protocols for evaluating water and wastewater from December 2021 to June 2022 [24]. Before collecting the water sample in pre-cleaned polyethylene bottles, each well was purged for a minimum of 10 min. The bottles were then placed in a refrigeration box until they were transported to the laboratory following standard operating procedures for analysis. After filtration through a 0.45 µm Nylon membrane filter (Sterlitech, Auburn, Washington, USA), all samples were stored in a refrigerator set at 4 °C until DMA analysis. A series of dilution processes was carried out to prepare (0.2, 0.5, 1, 2, 3, 4, 5, 6, 10, and 20 µg/L) Hg working solutions from 1000 µg/L Hg stock solution. Deionized water containing (2% v/v) HCl was used for dilution as a stabilizer.

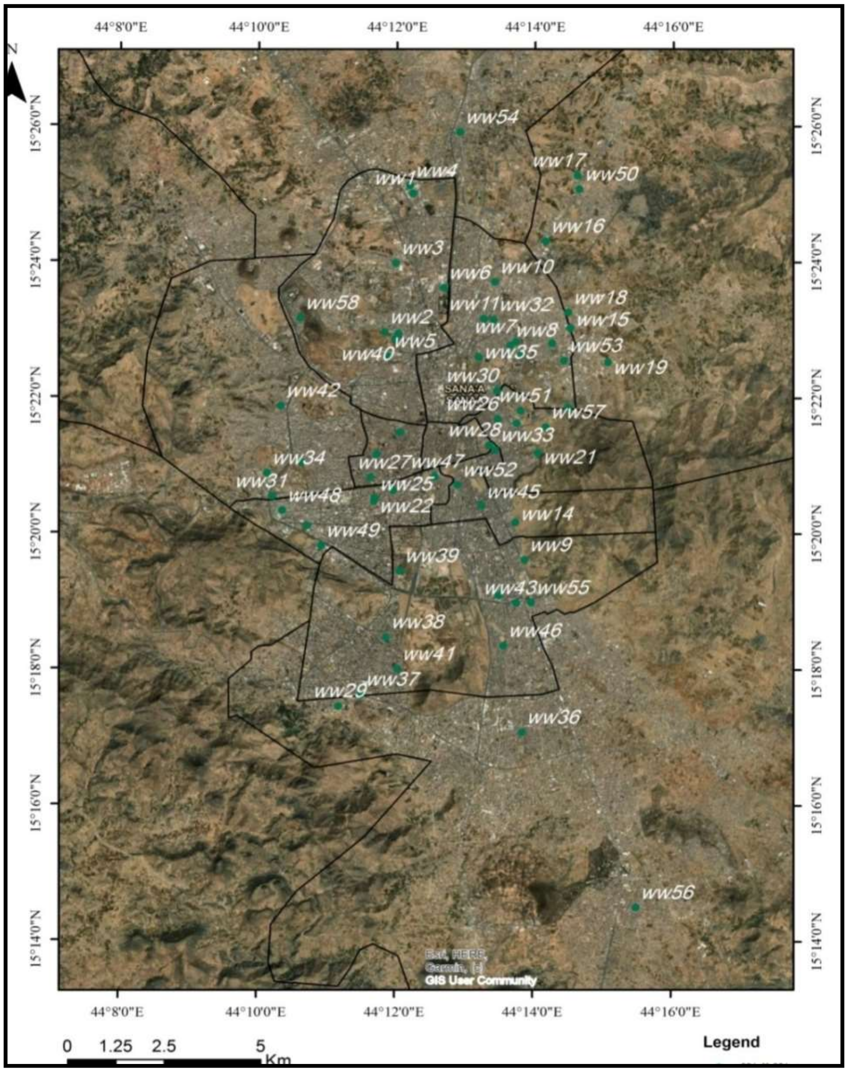

Figure 1. **Study area map showing the locations of the tested wells**

Analytical Method Validation

The accuracy of the method was assessed by standard addition method, so 0.5 g of real samples were put into the sample boat, spiked with 50, 100, and 150 µL of a standard solution containing 10 µg/L of Hg, and then subjected to DMA analysis to assess. The recovery (Hg) was determined using the difference in concentration between spiked and un-spiked samples. After injecting adequate quantities of Hg into blank water samples, all validation tests were conducted. The blank water sample was utilized in fortification experiments, calibration studies, and recovery investigations after being first examined for any presence of Hg. The limits of detection and quantification, as well as precision, accuracy, and linearity, were assessed as technique validation parameters.

Results and Discussion

Analytical Performance and Method Validation

The method's linearity was investigated by analyzing blank water samples containing various amounts of Hg (0.2, 0.5, 1, 2, 3, 4, 5, 6, 10, and 20 µg/L) and examined by DMA to find the method's linearity. Fig. 2 displays the data from the calibration curve and shows that within the range of 0.2 to 20 µg/L, the validated DMA-80 method demonstrated good linearity between the analytical signal and the Hg content.

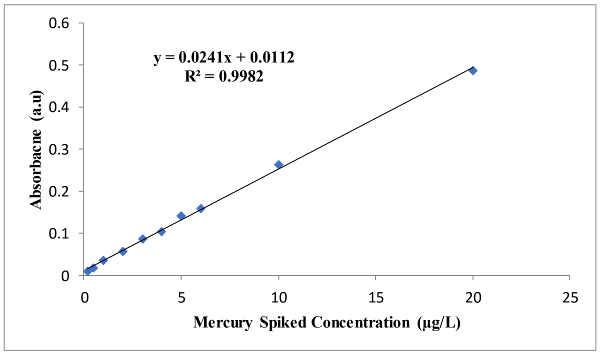

Figure 2. **Calibration curve of mercury spiked concentrations**

The linearity of the verified method is similar to that which Ahmed et al. [13] reported, higher than values obtained using a similar DMA technique [25] and marginally lower than the values reported by Sanchez et al. [26].

The precession of the method was measured by the variation in intraday recovery of Hg in spiked water samples. Independent water samples (blank) were spiked with Hg at ten different concentrations. Each spiked sample was analyzed in triplicate with DMA-80. Table 1 displays the repeatability results as %RSD. The average detector response consistently increases with rising Hg concentrations, indicating a linear relationship between analyte concentration and instrument signal. The method's precession, as reflected by the %RSD values, demonstrates a general trend of improvement with increasing Hg concentration. At lower concentrations (0.2-1 µg/L), the RSD values were relatively high, ranging from 7.18% to 13.13%, suggesting higher variability in measurements near the method's lower limit of quantification. However, as the concentration increases, the %RSD values generally decrease, with most measurements above 2 µg/L showing RSD values below 5%. It is important to note that excellent precision was achieved at 20 µg/L, with an RSD of 1.73%, which suggests good reproducibility at higher concentrations. The data indicates when Hg concentrations exceed 2 µg/L the DMA method yields become consistent. It also maintains acceptable precision at lower concentrations that satisfy the AOAC's Peer-Verified Methods criteria [27].

Where SD and S represent the standard deviation and slope of the calibration curve, respectively. The LOD and LOQ values were obtained at 0.091 and 0.303 µg/L, respectively, and these values are below the WHO-mandated acceptable limit of 1 µg/L for Hg in drinking water, indicating that the developed method is suitable for measuring Hg in drinking water samples [14]. The observed value of LOD of the DMA-80 is lower than the LOD values reported previously using different techniques such as UV-Vis molecular absorption (240 µg/L) [28], Molecular fluorimetry chemosensor (151 µg/L) [29], and Molecular fluorescence ion imprinted polymer (4 µg/L) [30]. But still higher than that of the more complex and highly sophisticated technique HPLC-ICP-MS (0.004 µg/L) [30].

Table 1. **Precision results of Hg analysis using DMA.**

Hg spiked Conc. (µg/L)	Average of detector Response (A)	SD	RSD (%)
0.2	0.0099	0.0013	13.13
0.5	0.0181	0.0013	7.180
1.0	0.0346	0.0029	8.380
2.0	0.0575	0.0038	6.600
3.0	0.0862	0.0021	2.440
4.0	0.1045	0.0038	3.640
5.0	0.1409	0.0034	2.410
6.0	0.1596	0.0077	4.910
10.0	0.2617	0.0057	2.630
20.0	0.4851	0.0084	1.730

The limit of detection (LOD) and limit of quantitation (LOQ) of the method were calculated using the following equations:

LOD = 3* SD/S LOQ = 10 * SD/S

The accuracy of the DMA-80 method for analysis of Hg in spiked real samples was evaluated using the %R (percentage recovery) and %Bias values as presented in Table 2. The %R values, calculated as (measured concentration / spiked concentration) × 100%, range from 92.23% to 108.90%. These values indicate reasonable accuracy, with %R values close to 100% suggesting a high level of accuracy where measured concentrations closely match spiked concentrations.

Considering the %Bias values, which represent the percentage difference between measured and spiked concentrations, a range of -7.77% to 8.90% was observed. Negative %Bias values indicate underestimation, while positive values indicate overestimation. The %Bias values in Table 2 show slight deviations from the expected values but generally fall within an acceptable range. Overall, based on the %R and %Bias values, the DMA-80 method demonstrates good accuracy performance for Hg analysis in real samples which are comparable to previous results published by Cobbina et al. [14] and Suvi et al. [17].

Table 2. **Recoveries % R and % Bias for Mercury in Spiked Real Samples.**

Hg Conc. (µg/L)	Detector Response (A)			% R	% Bias
	Standard (n=3)	Real sample	Spiked real sample (n=3)		
2	0.0528	ND	0.0575	108.90	8.90
4	0.1133	ND	0.1045	92.23	-7.77
6	0.1668	ND	0.1596	95.68	-4.32
10	0.2739	ND	0.2617	95.55	-4.45
20	0.506	ND	0.4851	95.87	-4.13

ND: non-detectable

Further investigation on the repeatability of the DMA-80 validated method using real well-water samples was carried out and depicted in Table 3. The %RSD was calculated as (%Standard deviation/mean) × 100. From the provided data, it can be observed that the RSD values for actual Hg concentration vary from 0.08% to 2.99% across different well-water samples. The higher RSD value of 2.99% observed only in a sample of only one well could be attributed to various factors such as sample inhomogeneity, instrumental limitations, or measurement errors. Overall, based on the %RSD values presented in Table 3, the DMA-80 validated method demonstrates good repeatability and precision for Hg determination in most of the well-water samples.

Table 3. **The %RSD of Hg concentration as a measure of method precision.**

Sample	Average Hg Conc.(µg/L), n = 4	SD	RSD (%)
Ww1	0.1217	0.0004	0.39
Ww2	0.1329	0.0001	0.08
Ww12	0.1331	0.0002	0.15
Ww13	0.1556	0.0047	2.99
Ww21	0.1071	0.0007	0.65
Ww38	0.1469	0.0005	0.34
Ww44	0.1739	0.0003	0.17
Ww54	0.0947	0.0004	0.49

Ww: well water

Real Samples Analysis

According to the results presented in Table 4, Hg concentrations in the water samples of only eight of the 58 wells tested ranged from 0.0947 to 0.1738 µg/L. This is above the method detection limit but remains below the WHO [31] and YSMO [32] specified permissible value of 1 µg/L. The Hg concentration in the remaining samples was below the method's detection limit. Notably, the data indicate that the samples obtained from the ancient city of Sana'a contained the highest Hg concentrations. The eight wells that had elevated levels of Hg were dispersed throughout various districts of the ancient city of Sana'a, and situated close to commercial establishments like dental clinics, hospitals, and small factories, which could potentially contribute to the contamination of wells with Hg.

Table 4. **Mercury concentrations in the real groundwater samples.**

Sample code	Depth (m)	Location	Amount of Hg (ng) (n = 3)	Hg Conc. (µg/L) (n = 3)
Ww1	300	Althaorah	0.0610±0.0002	0.1219 ± 0.0004
Ww2	432	Althaorah	0.0665±0.0001	0.1329±0.0001
Ww3	418		BDL	BDL
Ww4	345		BDL	BDL
Ww5	430		BDL	BDL
Ww6	400		BDL	BDL
Ww7	389		BDL	BDL
Ww8	470		BDL	BDL
Ww9	400		BDL	BDL
Ww10	380		BDL	BDL
Ww11	340		BDL	BDL
Ww12	394	Shaub	0.0666± 0.0001	0.1331±0.0002
Ww13	385	Shaub	0.0778± 0.0024	0.1556 ± 0.0047
Ww14	380		BDL	BDL
Ww15	447		BDL	BDL
Ww16	408		BDL	BDL
Ww17	362		BDL	BDL
Ww18	400		BDL	BDL
Ww19	440		BDL	BDL
Ww20	428		BDL	BDL
Ww21	408	Al tahriar	0.0532± 0.0004	0.1065 ± 0.0007
Ww22	425		BDL	BDL
Ww23	680		BDL	BDL
Ww24	630		BDL	BDL
Ww25	580		BDL	BDL
Ww26	402		BDL	BDL
Ww27	1010		BDL	BDL
Ww28	482		BDL	BDL
Ww29	386		BDL	BDL
Ww30	385		BDL	BDL
Ww31	380		BDL	BDL
Ww32	382		BDL	BDL
Ww33	915		BDL	BDL
Ww34	900		BDL	BDL
Ww35	900		BDL	BDL
Ww36	486		BDL	BDL
Ww37	884		BDL	BDL
Ww38	565	Maain	0.0734± 0.0002	0.1468 ± 0.0005
Ww39	823		BDL	BDL
Ww40	760		BDL	BDL
Ww41	300		BDL	BDL
Ww42	860		BDL	BDL
Ww43	648		BDL	BDL
Ww44	363	Old Sana'a City	0.0869± 0.0001	0.1738± 0.0003
Ww45	603		BDL	BDL
Ww46	620		BDL	BDL
Ww47	680		BDL	BDL
Ww48	600		BDL	BDL
Ww49	803		BDL	BDL
Ww50	740		BDL	BDL
Ww51	630		BDL	BDL
Ww52	408		BDL	BDL
Ww53	480		BDL	BDL
Ww54	295	Bani Alharith	0.0473± 0.0002	0.0947 ± 0.0004
Ww55	876		BDL	BDL
Ww56	950		BDL	BDL
Ww57	1000		BDL	BDL
Ww58	454		BDL	BDL

BDL = below detection limits

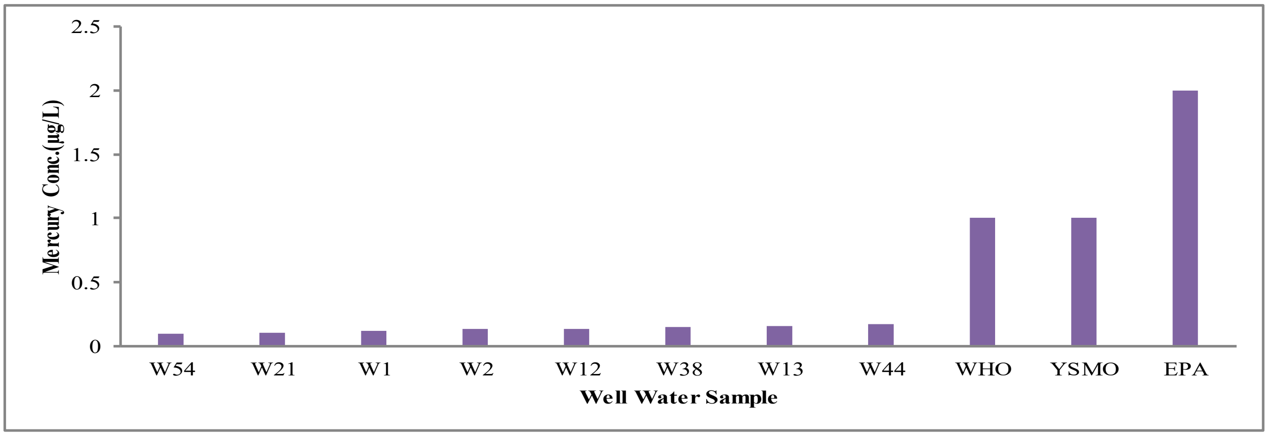

Figure 3. **Comparison of Hg concentrations in study samples to regulatory agencies' utmost allowable levels**

To gain a more comprehensive understanding of the Hg contamination in Sana'a drinking water, further investigations are necessary. These investigations may include detailed geological studies, mapping potential pollution sources, and analyzing the correlation between land use practices and Hg levels. It is crucial to monitor and assess Hg levels regularly to ensure the safety and well-being of the population relying on these water sources. A comparison is made between the Hg concentrations in these eight wells and the maximum allowable Hg levels established by local and international regulatory agencies, which is illustrated in the histogram in Fig. 3. This research also investigated the correlation between Hg concentration and well depth. The well's depth exhibited a range of 292-1010 meters. There is no correlation between the Hg concentration and the depth of wells, as shown in Table 4. The literature survey reveals the inconsistency of the effect of depth water on the level of Hg. For instance, Khattak et al. [33], reported that shallow water is more contaminated with Hg than deeper water, while Romero and co-workers suggested that Hg concentrations tend to increase as the profundity of the water column increases [34].

Table 5 provides a summary of the Hg concentration in the groundwater as documented in this study in comparison to values reported in the literature [14, 16, 35-45]. According to the data, the Hg contamination level in the potable groundwater of Sana'a City, Yemen remains among the lowest in the world. Furthermore, these comparisons highlight the variability in Hg concentrations across different regions and emphasize the importance of local environmental factors and anthropogenic activities in determining the Hg levels in water samples. It is crucial to consider these variations when interpreting and comparing Hg concentration data from different studies.

Overall, the results of the current study in Sana'a City, Yemen, indicate the presence of detectable levels of Hg in some well-water samples, albeit at relatively lower concentrations compared to some other regions. Further research and monitoring are necessary to gain a comprehensive understanding of Hg contamination in water sources and its potential implications for human and environmental health in Yemen. Implementing appropriate water treatment technologies and establishing consistent monitoring programs are essential steps to take. Public awareness campaigns play a significant role in educating communities and empowering them to make informed choices.

Table 5. **Mercury level comparison between values reported in this work and literature.**

Year	Authors	Location	Hg Conc. (µg/L)	Ref
2002	Kabir et al.	Bhopal, India	1 - 24	[35]
2006	Kowalski et al.	Poznan, Poland	0.8 – 4.1	[36]
2013	Ochiba	Ongata Rongai, Kenya	0.2 to 1.9	[37]
2015	Cobbina et al.	Nangodi and Tinga, Northern Ghana	0.38 - 0.64	[14]
2016	Ali et al.	Thar Coalfield, Pakistan	4.43	[38]
2016	Johari et al.	Sekotong Barat, Pelangan, Batu Putih, West Nusa Tenggara Province, Indonesia	<60	[39]
2016	Navarro et al.	Girona, Spain	0.08 - 1	[40]
2019	Nevondo et al.	Pretoria, South Africa	2.07	[41]
2020	Harahap and Simatupang	Batu Bola Village, Indonesia	0.4	[16]
2020	Samaniego et al.	Puerto Princesa, Philippines	0.01 - 0.204	[42]
2020	Khattak et al.	District Swabi, Pakistan	0.08 - 084	[33]
2021	Aristide and Ernest	Ivory Coast Divo, Abidjan	0.0087, 1.05	[43]
2023	Aflah et al.	Aceh Jaya, Indonesia	9.7	[44]
2023	Protano et al.	Southern Tuscany, Italy	0.52 - 10.30	[45]
2024	Current study	Sana'a City, Yemen	BDL - 0.1739	Current work

Developing and enforcing policies are vital for effectively managing the use and disposal of Hg. Exploring alternative water sources and employing remediation strategies are crucial to prevent further contamination. Collaborating with research institutions enhances our understanding of the issue and facilitates the development of effective mitigation strategies. By taking these collective actions, we can ensure the safety of the water resources for the population.

Conclusion

The initial nationally representative baseline data set for Hg in well water from Sana'a city has been produced through this study. Water samples were collected from 58 wells in Sana'a City. Hg concentration was ascertained utilizing a Direct Mercury Analyzer (DMA-80). This method is rapid, uncomplicated, and direct, and it exhibited exceptional linearity, sensitivity, and accuracy. As the Hg concentration in each of the 58 actual well samples was found to be below the threshold set by both local and international regulatory agencies, this indicates that the wells under examination are presently devoid of any Hg contamination. The 1 parts per billion (µg/L) threshold level of Hg in potable water, as established by the Yemeni Organization for Standards Metrology and Quality Control YSMO and the World Health Organization, was not exceeded in any of the 58 well water analyses. The research findings enable the decision-maker to formulate strategies aimed at mitigating the origins of Hg contamination and ensuring the safety of potable water for human consumption.

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Conflict of Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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