



Analysis of heavy metal content (Pb, Cu, Cd, and Hg) on refilled drinking water in Malang City based on atomic absorption spectroscopy using the PCA method

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ABSTRACT

Refillable drinking water was chosen as an alternative to bottled drinking water because the price is cheaper. Drinking water has benefits for the body, but if the levels of heavy metals contained exceed the Ministry of Health's standards, it will have a negative impact on body health. This research was conducted with the aim of determining the quality of refillable drinking water in terms of heavy metal levels. Atomic absorption spectroscopy was used to test the levels of heavy metals (Pb, Cu, Cd, and Hg) in the samples. Research data shows that refill drinking water samples A1, A2, A3, A4 and A5 contain heavy metals at levels that exceed the Ministry of Health's standards. PC1 and PC2 have the highest eigenvalues, with a proportion of PC1 of 60.86% and PC2 of 22.69%. The two PCs have a cumulative proportion of 83.55% and are considered capable of representing the entire data. PCA method was used to identify patterns and group samples based on heavy metal content, with PC1 and PC2 reflecting 83.55% of the data variability. Pb and Cd are the variables that have the longest resultant lines, which shows that these two variables have a large contribution to the formation of new variables. The three secondary data samples, namely A6, A7, and A8, are control samples because their quality is good. Refill drinking water samples that are almost close to control are samples A1, A4, and A5, while samples A2 and A3 are far from control. The further the sample is from the control, the lower the quality. The results of this study highlight the need for strict supervision of refillable drinking water depots and the implementation of more effective purification methods to reduce heavy metal content exceeding health standards.

Keywords: Atomic absorption spectroscopy, heavy metal, PCA, refill drinking water

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INTRODUCTION

Water is one of the most important substances in the life of living creatures after air (Białecka-Dębek & Pietruszka, 2019). Water has many benefits, one of which is that it is consumed as drinking water, because water is very important for the human body. Many people consume refillable drinking water as a substitute for drinking water. Water is very important for the human body (Cook et al., 2019; Muthmainnah & Mulyono, 2022). Many people consume refillable drinking water as a substitute for bottled drinking water (Liska et al., 2019). The price of refillable drinking water is 1/3 cheaper than bottled drinking water, but not all depots are managed well, especially regarding the quality of drinking water (Azteria & Rosya, 2023). Refill drinking water is water that can be drunk directly without boiling it first (Hasanuddin, 2022; Nakamura & Kondo, 2021). Water that is safe for consumption is water that meets the standard parameters set by the Minister of Health. Based on Minister of Health regulations, these parameters include physical, chemical, microbiological and radioactive parameters (Onyango et al., 2018).

Heavy metals are one of the parameters in the Minister of Health's standards that must be considered in the content of drinking water. At low levels heavy metals are needed by the body, but if the levels are excessive they will harm the body. Copper metal is a metal that is needed by the body in very low levels (Westgard et al., 2021; Yang et al., 2021). Lead (Pb), cadmium (Cd), and Mercury (Hg) are heavy metals that fall into the category of dangerous and toxic metals and is a metal that has no function in the body (Agrawal et al., 2020). To analyze the content of heavy metals lead and cadmium in refillable drinking water, atomic absorption spectroscopy can be used (Dorji & Chopel, 2023; Valerko et al., 2022). The results of the research showed that metal levels in repackaged drinking water samples showed lead levels of 0.11-1.87 ppm and cadmium levels of 0.44-0.54 ppm (Tian et al., 2019).

The manganese metal contained in several drinking water samples exceeds the specified standards (Yang et al., 2021; Rezaei et al., 2022). Several other researchers have researched the content of heavy metal levels in refillable drinking water. They stated that the samples studied still contained iron, lead, and chrome at levels that exceeded the Minister of Health's standards (Rajeshkumar et al., 2018). Tazi carried out research using the PCA method in electronic tongue research which was used to measure the development of milk taste from fresh to stale. The response of the electronic tongue was evaluated using the PCA method. Based on the PCA results, the two milk samples showed a pattern of change over time (Tazi et al., 2018).

Other research related to the PCA method was also carried out on citrus fruit samples using an electronic tongue. Based on PCA analysis, the electronic tongue can classify all patterns from four bases, with PC1 and PC2 values of 95.1%. Previous research on refill drinking water samples used 2 or 3 heavy metal parameters and did not use PCA as an analysis method (Bhatia et al., 2023). Therefore, it is necessary to carry out studies related to the analysis of heavy metal content using more parameters and additional analysis methods using PCA.

This research is entitled "Analysis of Heavy Metal Content (Pb, Cu, Cd, and Hg) in several Drinking Water Refills in Malang City Based on Atomic Absorption Spectroscopy Using the PCA Method". Atomic absorption spectroscopy is a quantitative analysis method

whose measurement is based on the amount of radiation absorbed by the atom. In this research, four heavy metal parameters were used, namely Pb, Cu, Cd, and Hg, with the hope that the samples tested were truly suitable for consumption (Du et al., 2019; Misra & Paunikar, 2023).

The aim of this research is to determine the content and levels of heavy metals in samples, determine the quality of refillable drinking water in terms of the levels of heavy metals contained, and analyze test result data using the PCA method. Data analysis using the PCA method aims to differentiate data patterns from test results without having to analyze them one by one. In Malang City, a study conducted by Melinda in 2017 reported that 25% of refill drinking water depots did not meet one or more quality standards set by the Ministry of Health (Melinda et al., 2017). This alarming data underscores the importance of continuous monitoring and advanced analysis methods such as PCA to ensure public health safety in Malang. PCA was chosen as the analysis method because it is a powerful multivariate statistical technique that helps to simplify complex datasets with multiple variables by reducing dimensionality. It allows for the identification of underlying patterns and correlations among heavy metal concentrations, which is crucial for understanding the overall water quality. Furthermore, PCA provides a more comprehensive view of the data by capturing the maximum variability in fewer components, thus offering a more efficient analysis of multiple heavy metal parameters simultaneously. This study contributes to existing knowledge by demonstrating the use of PCA to uncover patterns and relationships among multiple heavy metal variables simultaneously, an approach that differs from previous studies focusing on fewer parameters and lacking multivariate techniques. The integration of PCA with atomic absorption spectroscopy provides a comprehensive framework to evaluate and improve water quality assessment, particularly in urban areas with diverse contamination sources.

RESEARCH METHODS

This experimental-based research aims to determine the heavy metal content in the samples. The research was carried out in 2023 at the Analysis and Measurement Unit, FMIPA, Brawijaya University, and at the Sensor Laboratory, Physics Department, Faculty of Science and Technology, Maulana Malik Ibrahim State Islamic University, Malang. The tools and materials used in this research include atomic absorption spectroscopy (AAS), beakers, measuring flasks, measuring pipettes, Erlenmeyer flasks, stir sticks, watch glasses, filter paper, refillable drinking water, distilled water, and concentrated nitric acid (HNO₃).

The sample selection process was conducted with specific criteria to ensure comprehensive and representative data. The depots selected for the study were required to meet the following criteria: (1) depots should be located in diverse geographical areas within Malang City, including the northern, southern, eastern, western, and central regions, to capture variations in water quality across the city; (2) depots with a high consumer turnover were prioritized, as they represent a larger consumer base and thus, more accurately reflect the quality of drinking water that is commonly consumed; (3) depots from both small-scale and large-scale businesses were included to account for potential differences in management and water quality practices; (4) depots with a known history of managing refillable drinking water were also selected, to ensure that the quality of water was of public interest. Within each area, depots

were selected randomly to adhere to these criteria and avoid bias.

The sample collection began with a preliminary visit to depots to inform owners about the study and obtain permission for sample collection. Samples were collected using sterilized 1-liter glass containers, ensuring water was allowed to flow for 2–3 minutes before collection to obtain a representative sample of running water. The samples were sealed immediately to prevent contamination and transported to the laboratory in a cool box to maintain their quality. Upon arrival at the laboratory, the samples were stored at 4°C and analyzed within 24 hours to ensure reliability.

Sample preparation involved several steps to standardize the testing process. Each sample was filtered using filter paper to remove particulate matter, and HNO₃ was added at a concentration of 1% to stabilize the metals and prevent precipitation during storage. The filtered and acidified samples were then adjusted to a consistent volume in a measuring flask. The testing process began with calibrating the AAS using standard solutions of Pb, Cu, Cd, and Hg at known concentrations, and a blank sample of distilled water with HNO₃ was used to ensure baseline accuracy.

The calibration of the AAS was performed by preparing a set of standard solutions with known concentrations of Pb, Cu, Cd, and Hg. These solutions were carefully prepared by accurately weighing the appropriate amounts of each metal's standard salt and dissolving them in distilled water. The AAS was then calibrated by measuring the absorbance of these standard solutions at the respective wavelengths for each metal. Calibration curves were generated by plotting the absorbance values against the known concentrations. To ensure the validity and accuracy of the measurements, the calibration curve was checked regularly throughout the analysis. In addition, the performance of the AAS was verified by measuring the blank sample and ensuring that it gave an absorbance close to zero. If discrepancies were detected, adjustments were made before proceeding with the sample analysis (Nasution et al., 2022). Each sample was analyzed for heavy metal content using AAS, specifically measuring lead (Pb), copper (Cu), cadmium (Cd), and mercury (Hg). To ensure consistency and accuracy, each measurement was repeated three times.

The obtained test results were recorded and converted into concentration units (ppm) based on the AAS calibration curve. The data was further analyzed using Principal Component Analysis (PCA) to identify patterns and groupings among the samples based on their heavy metal content. This comprehensive approach ensures the reliability and robustness of the analysis in determining the quality of refillable drinking water in Malang City.

RESULTS AND DISCUSSION

The test results of refillable drinking water showed that there were heavy metals lead (Pb), copper (Cu), cadmium (Cd), and mercury (Hg) in refillable drinking water samples A1, A2, A3, A4, and A5. Samples A6, A7, and A8 are standard data obtained from research conducted by Chopel (Dorji & Chopel, 2023). Refill drinking water samples contained heavy metals at levels that exceeded the standards set by the Minister of Health regarding drinking water requirements (see Table 1). The maximum permitted levels of heavy metals are: Pb 0.01 mg/L, Cu 2 mg/L, Cd 0.003 mg/L, and Hg 0.001 mg/L. Secondary data shows that the sample

contains very little levels of the heavy metal copper (Cu) and does not exceed the Ministry of Health's standards. Samples from secondary data do not contain the heavy metals lead (Pb), cadmium (Cd), and mercury (Hg).

Table 1. Heavy Metal Levels in Refillable Drinking Water Samples and Minister of Health Standards.

Heavy metal	Concentration (mg/L)								Standard Minister of Health (mg/L)
	A1	A2	A3	A4	A5	A6	A7	A8	
Pb	0.15	0.21	0.18	0	0.12	0	0	0	0.01
Cu	0.003	0.005	0.011	0.006	0.003	0.005	0.003	0.005	2
CD	0.17	0.52	0.48	0.32	0.15	0	0	0	0.003
Hg	0	0.14	0	0	0	0	0	0	0.001

Data analysis using the PCA method aims to reduce data from large dimensional variables to smaller dimensional variables (Rwoo et al., 2017). There are several ways to determine the number of Principal Components (PC) taken to reduce variable dimensions, one of which is a scree plot (Zolkipli et al., 2018).

Table 2. Sequence of eigenvalues.

No	Eigenvalues	Variance (%)	Cumulative
1	2.43435	60.86	60.86
2	0.90773	22.69	83.55
3	0.4409	11.02	94.57
4	0.21702	5.43	100.00

A scree plot is a plot that shows the relationship between PC and eigenvalues. This score plot only takes 2 dimensions with reference to the order of the first 2 largest eigenvalues (see Table 2). Based on Figure 1, PC1 and PC2 have the highest eigenvalues compared to the eigenvalues of other PCs. Figure 1 shows a scree plot of heavy metal levels in the samples.

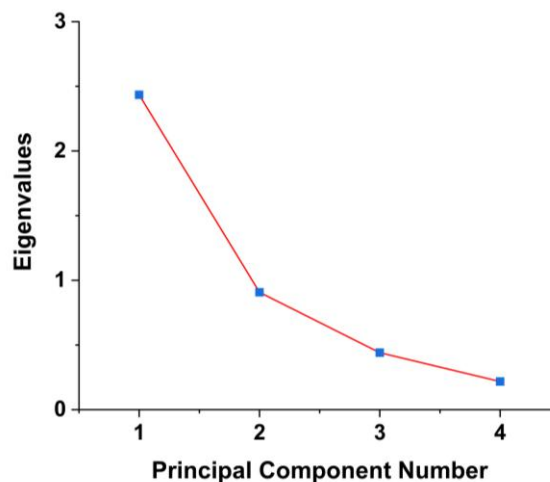


Figure 1. Scree plot heavy metal levels in samples

Data from PC1 and PC2 can be used to determine the contribution of each variable to data

grouping through loading plot analysis. Loading plot shows the correlation between the original variable and the new variable. The loading plot provides an indication of which original variables have a big influence on the formation of new variables, which is indicated by the loading plot value and the long resultant line. The longer the resultant line, the greater the contribution of the variable in forming new variables.

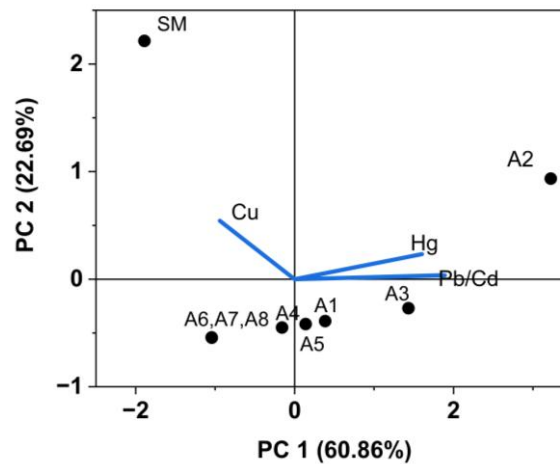


Figure 2. Loading plot of heavy metal levels in samples

In Figure 2, the variables Pb and Cd have the highest loading plot values. These two variables make a positive contribution as the original variables that have the most influence on the formation of new variables, especially on PC1.

Based on the screen plot, the proportion of each Principal Component (PC) can be seen, which is obtained from dividing the eigenvalues of each PC by the sum of the eigenvalues of all PCs (Khanniri et al., 2023). This proportion can be calculated using Equation (1):

$$\text{kth PC} = \frac{\lambda_k}{\lambda_1 + \lambda_2 + \lambda_3 + \dots + \lambda_p} \times 100\% \quad (1)$$

The results of data reduction are visualized in the form of a two-dimensional score plot (Figure 3). Score plot is a visualization of metal levels in a sample. The PC1 proportion value is 60.86% and the PC2 proportion is 22.69%, so the proportion of the two PCs is 83.55%, which means that the two PCs present the diversity of the data and are considered sufficient to represent the entire data.

In this analysis, samples A6, A7, and A8 serve as controls, where the three samples contain very low levels of heavy metals (Cu) and are below the Ministry of Health's standards, so that the water quality can be concluded to be good and suitable for consumption. Based on the score plot, the refill drinking water samples closest to the control are A1, A4, and A5. Sample A3 is the sample that is far from control, and sample A2 is the sample that is farthest from control. The three samples A1, A4, and A5 are more suitable for consumption compared to samples A3 and A2. The further the sample is from the control, the lower the quality.

Some of the advantages of using the PCA method are that it can eliminate correlations cleanly (correlation = 0), can be applied to various types of research and data conditions, can be used without reducing the number of original variables, and provides more accurate conclusions than other methods (Tian et al., 2019). Heavy metals that enter the water, either directly or indirectly, will chemically reduce the quality of the water. Water has the ability to

purify itself, especially groundwater, through filtration of soil pores and plant roots. However, if the heavy metal is in large volumes or high doses, it can exceed the carrying capacity of the water. According to Ahmed the presence of heavy metals in refilled drinking water can be caused by several factors, including raw water, production machines and production equipment, production processes, and management of the refilled drinking water depot itself (Ahmed et al., 2023).

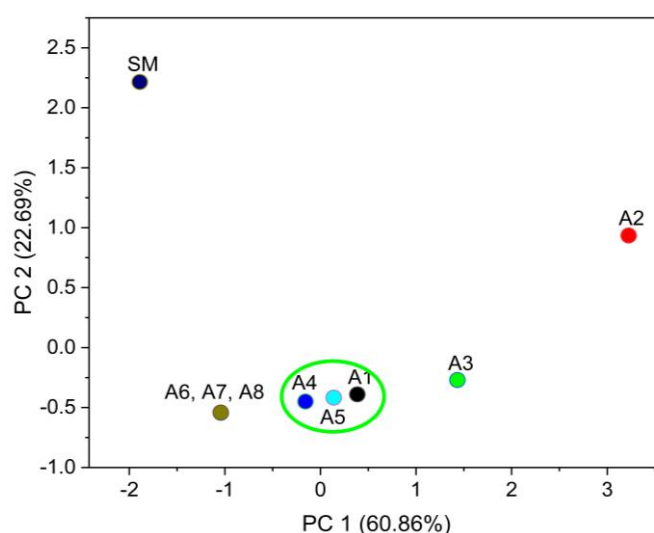


Figure 3. Score plot heavy metal levels in samples

Several factors may contribute to the heavy metal contamination of refillable drinking water, including the quality of the source water, improper handling during production, and substandard cleaning practices in refillable drinking water depots (Yadaf et al., 2023). Raw water used in the production of refillable drinking water may contain heavy metals due to contamination from industrial waste, agricultural runoff, or natural sources such as soil erosion. Furthermore, production equipment, if not properly cleaned and maintained, can contribute to contamination during the filling process (Lubal et al., 2024). Depots may also use poor-quality pipes or containers that leach metals into the water, further exacerbating the problem. Additionally, a lack of strict monitoring and quality control in refillable drinking water depots can lead to practices that do not meet health standards.

Heavy metals in the body can enter through several intermediaries, including food, drink, breathing, skin, and others (Azteria & Rosya, 2023). The presence of heavy metals in the body at high levels can have a negative impact on body health (Melinda et al., 2017; Kumar et al., 2019). Cadmium levels in five refill drinking water samples were the highest levels and exceeded the standards set by the Minister of Health. According to Raharini & Yuniarti (2023) cadmium is a byproduct of zinc production. Soil, rocks, coal, and mineral fertilizers contain some amount of cadmium (Kanellis, 2018). Water can also be contaminated when water flows through soil containing cadmium. In a journal written by Tian et al. (2019) mild cadmium poisoning can cause stomach upset, vomiting, diarrhea and kidney failure.

Pb was detected in significant concentrations in several samples, raising concerns about its long-term health effects. As a neurotoxin, lead can affect the nervous system, particularly in

children, leading to developmental delays, reduced IQ, and behavioral issues (Gupta et al., 2017). In adults, chronic exposure to lead can cause hypertension, kidney dysfunction, and cardiovascular diseases. A study by Ghaderpoori et al. (2017) reported similar findings, where lead contamination in drinking water exceeded permissible levels and was linked to industrial pollution and poor water management practices. This aligns with the results of the current study, emphasizing the need for stricter regulation and monitoring of refillable drinking water depots (Ghaderpoori et al., 2017). Hg, although detected in lower concentrations compared to cadmium and lead, poses significant health risks even at trace levels due to its bioaccumulative nature (Chowdhury et al., 2016). Hg exposure can impair the nervous system and renal function, with severe exposure leading to neurological damage and immune system suppression, corroborating findings by Alidadi et al. (2019) that highlighted Hg presence in water near industrial zones. In contrast, Cu, although an essential micronutrient, was found within acceptable limits in this study, aligning with previous research by Alfian et al. (2023); however, excessive copper intake could lead to gastrointestinal irritation, liver damage, and oxidative stress. Comparing these results with studies from other regions, such as Gupta et al. (2017), it is evident that industrial and agricultural activities contribute significantly to heavy metal contamination in water sources. These findings highlight localized challenges in Malang City while resonating with global issues of water quality management and its implications for public health, underscoring the urgency for implementing advanced monitoring systems and remediation strategies to mitigate heavy metal contamination in drinking water.

CONCLUSION

The research results showed that refill drinking water samples contained the heavy metals lead (Pb), copper (Cu), cadmium (Cd), and mercury (Hg), with some samples exceeding the maximum limits set by the Minister of Health. The Principal Component Analysis (PCA) method is used to reduce data dimensions and analyze the influence of variables on data diversity. Based on the scree plot, PC1 and PC2 represented 83.55% of the data diversity, with PC1 contributing 60.86% and PC2 22.69%. Loading plot analysis revealed that the variables Pb and Cd had a significant contribution in the formation of new variables, indicating the large influence of these two heavy metals on drinking water quality. The score plot shows that samples A1, A4, and A5 are closer to the control and have better quality compared to samples A2 and A3. The further the sample is from the control, the more its quality decreases. The PCA method is proven to be effective in reducing data dimensions and providing accurate analysis without reducing the number of original variables, offering a more comprehensive evaluation of water quality compared to previous studies that often focused on fewer parameters and lacked multivariate analysis.

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