

ORIGINAL RESEARCH ARTICLE

Mapping and identifying heavy metals in water use as chemicals of potential concerns in upper watershed

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ABSTRACT

BACKGROUND AND OBJECTIVES: Excessive presence of heavy metals in water sources can reduce water quality and harm human health. However, research on heavy metals from water sources for sanitation and hygiene purposes and drinking water in the Upper Citarum Watershed remains limited. This study focuses on the distribution of heavy metals and chemicals that have potential health risks.

METHODS: Ten heavy metals, namely, lead, cadmium, chromium, copper, cobalt, iron, mercury, manganese, arsenic, and zinc, were analyzed. Groundwater samples were collected from 160 locations, and drinking water samples (for respondents who do not drink groundwater) were collected from 98 locations. Heavy metal concentrations were detected using inductively coupled plasma optical emission spectrometry.

FINDINGS: The levels of arsenic, cadmium, cobalt, iron, mercury, manganese, and lead exceeded the quality standards for drinking water, while those of arsenic and cobalt did not exceed the quality standards for water hygiene and sanitation. Arsenic and cobalt quality standards were more stringent for drinking water compared with those for water hygiene and sanitation. Lead–cadmium and iron–manganese in groundwater showed a positive Spearman correlation ($p < 0.05$) and may originate from the same source. Copper and zinc did not exceed the quality standard in 100% of drinking water samples. Iron and zinc in groundwater differed significantly due to variations in topography and soil type ($p < 0.05$). This study reveals that 6 out of 10 heavy metals are chemicals of potential concern and are sorted based on potential risks to health, that is, arsenic > mercury > lead > cobalt > manganese > cadmium. Ingestion is the main pathway for potential risk, and children are more likely to be at risk than adults.

CONCLUSION: Stakeholders and decision makers must immediately implement sustainable actions to protect public health. Evaluation of water sources, technology, maintenance processes, and water quality should be conducted before and after technology use from Refill Drinking Water Depots to ensure that raw and processing water meets the quality standards.

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INTRODUCTION

The Citarum River is one of the rivers that support the lives of most people in West Java (Salami and Pradita, 2020). However, it has been a polluted river due to the impact of human activities. In the Action Plan for Control of Pollution and Damage to the Citarum River Basin for 2019–2025 published by the Regional Government of West Java Province, the status of the Citarum River is polluted, where domestic and industrial activities within the area cause contamination. Based on data from the Coordinating Ministry for Maritime Affairs in Kusuma et al. (2018), in at least 3,236 textile industries, 90% do not have waste water treatment plants (WWTPs) and 280 tons of chemical wastes per day are discharged into the Citarum River. The Upper Citarum watershed is an area that has a high industrial distribution. The high number of industries in Bandung Regency will lead to a high potential for heavy metal pollution levels around the area. Various types of heavy metals in the environment originate from the textile industry (Van Ginkel, 2015; Bielak and Marcinkowska, 2022; Sumantri and Rahmandi, 2020; Bhardj et al., 2014; Thanki and Bhattacharya, 2022; Baek et al., 2020). Coal fuel is used for steam generators (boilers) in the textile industry in Bandung Regency, and burning coal increases exhaust gas emissions in the form of particles containing heavy metals (Febrion and Falah, 2018). Heavy metal concentrations in fly ash after coal combustion are high in small particles (<1 m) (Czech et al., 2020). Heavy metal pollution is a serious concern in Bandung Regency, especially in groundwater. The groundwater system is related to air, soil, and river water, so the potential for heavy metal pollution in groundwater is very large. Heavy metal contamination in soil occurs because wastewater effluent containing high concentrations of heavy metals is released into the drainage system and has a negative impact on community soil and groundwater. Groundwater can be polluted due to the infiltration of water containing contaminants into an aquifer (Van Ginkel, 2015; Hartmann et al., 2021; Araiza-Aguilar et al., 2020). Heavy metal pollution in rivers can occur by releasing wastewater effluent directly into receiving river water bodies and/or through drainage systems. The content of heavy metals in the Upper Citarum watershed will also affect the quality of groundwater. The interaction between river water and groundwater can occur with groundwater filling river water (gaining stream) through streambed and/or river water filling

groundwater (losing stream) through streambed and saturation zone (Safaeq and Fares, 2016), so rivers can be the main source of groundwater recharge (Gao et al., 2022). Air pollution can occur due to exhaust gas emissions from industries in the form of particulates in air containing heavy metals and can experience deposition in soil/groundwater (Jarsjo et al., 2020; Adnan et al., 2022; Masum and Pal, 2020; Karbassi et al., 2015). Air pollution in the industrial environment can affect people living within the vicinity through the absorption of heavy metals into the human body (Manisalidis et al., 2020; Zhao et al., 2022). About 55.86% of the population in Bandung Regency still use groundwater (shallow well water and pumping wells) as a source of clean water; the use of groundwater containing heavy metals in daily life has negative impacts. Groundwater is used by the community for sanitation and hygiene, and some people still use it for drinking and other consumption purposes. Communities also use other sources of water for drinking, and the quality of water is unknown. Heavy metal contaminants in groundwater can be a source of danger to the surrounding community. Heavy metals in environmental media are very dangerous because they are toxic, persistent in the environment, and bioaccumulative, so they pose a potential threat to public health (Ali et al., 2019; Fahimah et al., 2020; Mitra et al., 2022). Heavy metal exposure pathways from groundwater to the community can occur through ingestion (oral, usually from exposure to the food chain) and dermal absorption (usually from the use of water) (Olawoyin, 2018; Briffa et al., 2020). Heavy metals are cytotoxic, hepatotoxic, nephrotoxic, neurotoxic, and carcinogenic (Anyanwu et al., 2018). Heavy metals contribute to many cases of noncommunicable disease (NCDs), including neurobehavioral disorders (lead, mercury), cardiovascular diseases (lead, cadmium), kidney diseases (lead, cadmium), and some cancers (arsenic, chromium) (Nordberg et al., 2015). Heavy metals can also affect the formation of clefts in the lips and palate (Pi et al., 2018; Takeuchi et al., 2022). The incidence of cleft lip and palate is caused by 22% genetic factors and 78% environmental factors (Loho, 2013). Evidence indicates the association between maternal and paternal exposure to environmental pollutants and the risk of cleft palate in offspring (Hao et al., 2015). According to Martin and Griswold (2009), chronic lead exposure can cause autism, kidney damage, brain damage, birth defects, and

even death. Several researchers have also found a relationship between stunting and an increase in heavy metals in the body (Gardner *et al.*, 2013; Gleason *et al.*, 2017). The Upper Citarum watershed has 10 heavy metals, namely, lead (Pb), cobalt (Co), chromium (Cr), iron (Fe), manganese (Mn), cadmium (Cd), copper (Cu), zinc (Zn), mercury (Hg), and arsenic (As) (Sukarjo *et al.*, 2021; Sumantri and Rahmani, 2020; Utami, 2009; Fadhilah *et al.*, 2018; BRES, 2018; Komarawidjaja, 2017; Septiono and Roosmini, 2015; Budiman *et al.*, 2012; Salam *et al.*, 2019). Despite the presence of various heavy metals in the Upper Citarum Watershed, only few reports are available regarding the determination of which heavy metals have the potential to pose a health risk (Septiono and Roosmini, 2015; Thufailah, 2020; Tamang *et al.*, 2020). Bandung Regency Health Profile Data show that the incidence of several types of NCDs increased from 2015 to 2020. Therefore, scholars should study the concentrations of various types of heavy metals in groundwater and determine the chemicals of potential concern (COPC). Analysis of heavy metals is conducted in groundwater prior to its use for sanitation and hygiene; analysis of other water sources is also important to assess the potential cumulative risk that society accepts and other water sources still indicate local pollution. This study is a preliminary work prior to comprehensive examination through epidemiological studies of heavy metals. Results can be used by other researchers who are interested in environmental epidemiology issues that have not been carried out in this study area. In this context, this study aims to determine the concentration and distribution of Pb, Co, Cr, Fe, Mn, Cd, Cu, Zn, Hg, and As in groundwater and other water sources (for people who do not use groundwater for drinking); compare concentration data with the quality of water used for sanitary hygiene and consumption; and determine heavy metals that have high risks to public health (or referred to as COPCs) in adults and children. COPCs were determined based on two exposure pathways, i.e., oral and dermal absorption. The study was conducted in seven sub-districts located in the Upper Citarum River Watershed, Bandung Regency in Indonesia, in 2022.

MATERIALS AND METHODS

Site description and sampling

The research was conducted in the Upper Citarum

Watershed, Bandung Regency, precisely in seven sub-districts, namely, Pangalengan, Rancaekek, Ciparay, Pacet, Baleendah, Majalaya, and Soreang. The number of sampling points in seven sub-districts is 160 points spread over 24 points in Ciparay, 27 points in Majalaya, 25 points in Rancaekek, 24 points in Baleendah, 22 points in Soreang, 20 points in Pacet, and 18 points in Pangalengan. The number of samples is not evenly distributed in several sub-districts due to lack of people who use groundwater for daily life in areas with high topography, such as Pacet and Pangalengan Sub-districts, and people are more likely to use mountain springs. From 160 sampling points, 160 groundwater samples were collected at all points and 98 other water sources for consumption were also taken from respondents who use water sources other than groundwater for consumption. The study area covers seven sub-districts located at 6°57' 30" S–7° 17' 30" S; 107° 30' 30" E–170° 50' 0" E (Fig. 1). Overall, the study area is 47,119.98 Ha. Pangalengan has an area of 19,540.93 Ha (41.47% of the total area of the study area), Pacet is 9,193,965 Ha (19.51%), Ciparay is 4,617.57 Ha (9.80%), Rancaekek is 4,524.83 Ha (9.60%), Baleendah is 4,155.54 Ha (8.82%), Soreang is 2,550,679 Ha (5.41%), and Majalaya is 2,536.46 Ha (5.38%).

From 22 March 2022 to 10 April 2022, 160 groundwater samples and 98 consumption water samples (taken from respondents who use water sources other than groundwater for consumption) were collected in the study area. The latitude and longitude at each sampling site were recorded. The study was conducted during the dry season by considering the higher concentrations of heavy metals due to the dilution of rainwater. High rainfall can cause the dilution of heavy metals in groundwater (Long *et al.*, 2021; Aithani *et al.*, 2020), leading to higher heavy metal concentrations in the dry season than in the rainy season. Written surveys and sampling of groundwater and drinking water were carried out simultaneously in seven sub-districts. The survey forms were distributed at the sampling locations, precisely at 160 sampling points to ask questions related to water source use and its designation. Each respondent signed an informed consent form after receiving a thorough explanation of the survey. The percentages of respondents who use dug wells as water for sanitation and hygiene (SH) in Ciparay, Majalaya, Rancaekek, Soreang, Pacet,

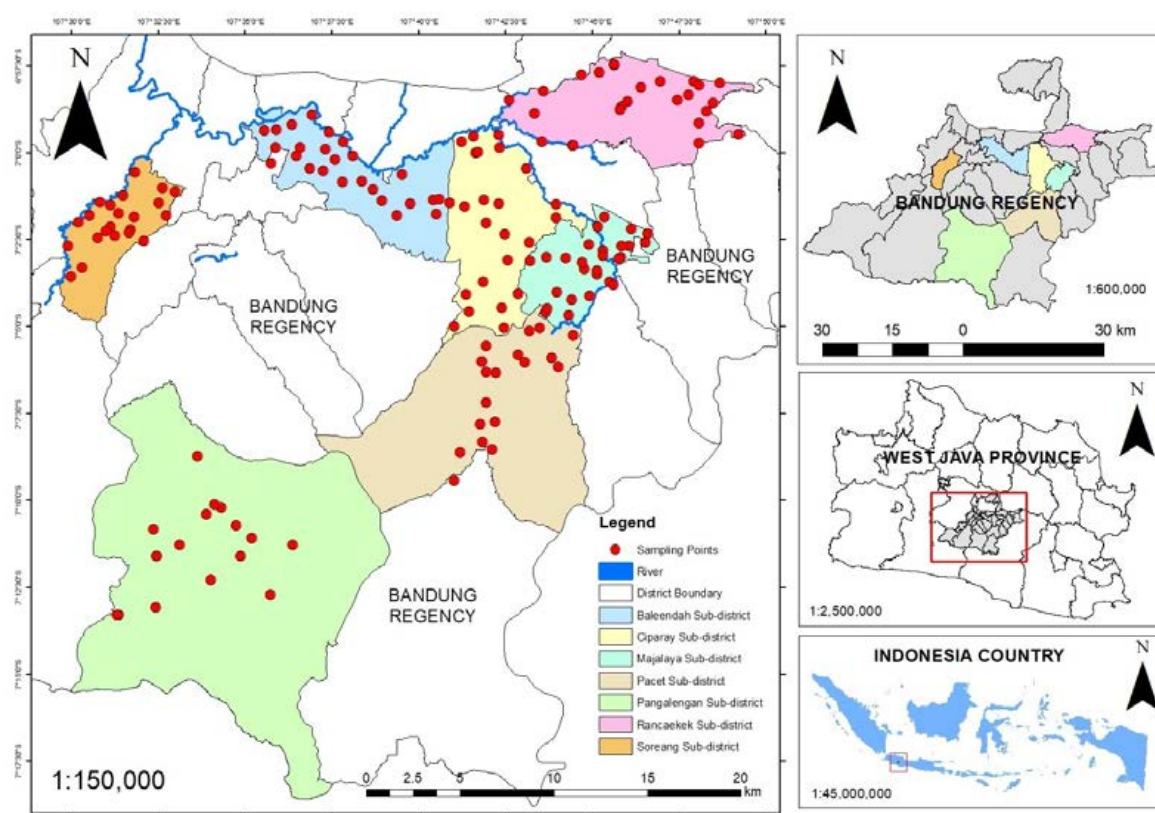


Fig. 1: Geographic location of the research area and distribution of sampling points in the Upper Citarum Watershed, Bandung Regency in Indonesia

and Pangalengan are 70.83%, 77.78%, 64%, 59.09, 60%, and 100% of the respondents, respectively. In Baleendah, the majority of respondents (56%) used bore wells for SH. People rarely use pump wells to meet their water needs for SH and they were only found in Ciparay (8.33%) and Baleendah (16%). The type of groundwater used by the respondent will affect the quality of water; for example, bore well water has better water quality than water from dug wells (Alina and Oshunrinade, 2016; Lestari *et al.*, 2021). Respondents who use groundwater for SH do not necessarily use it for consumption. In addition to groundwater, other sources of water for consumption include refilled water, spring water, and rainwater. Most respondents in Ciparay, Majalaya, Rancaekek, Baleendah, and Soreang use refilled water for consumption, with sequential percentages of 50%, 66.67%, 80%, 88%, and 50%, respectively. Some respondents use groundwater for consumption, that is, 50% in Ciparay Sub-district, 22.22% in Majalaya,

12% in Rancaekek, 12% in Baleendah, and 40.91% in Soreang. In Pacet and Pangalengan, most respondents still use groundwater for consumption (80% and 76.47%, respectively). In addition, some respondents use spring water for consumption, that is, as much as 3.70% in Majalaya. Meanwhile, about 4% of the respondents in District of Rancaekek use rainwater for consumption.

Water sampling and preservation

Groundwater sampling is conducted by taking a momentary sample (grab sample) following the Indonesian National Standard Index (SNI) 6989.58: 2008 concerning Groundwater Sampling Methods. Groundwater sampling is also carried out by SNI (2008) concerning Groundwater Sampling Methods, where a bailer is used to collect groundwater. For deep well water (from drilled wells), sample is collected by opening the well water faucet and flowing it for 1–2 minutes and is placed in a container.

Groundwater samples are collected in 250 mL high-density polyethylene (HDPE) bottles, which are rinsed with the sampled water in advance. Refilled water (gallon water) is sampled by rinsing first the HDPE bottle with sample water and then filling it with the sample 1 to 2 inches from the top (USEPA, 2016). The sampling location is recorded using global positioning system (GPS). Water samples are preserved by adding 6–7 drops of nitric acid (HNO_3) to pH < 2 and placed in a cool box at 4 °C (Standard Method for the Examination for Water and Wastewater 22nd Edition, 2012). Water samples are stored not exceeding the maximum storage time of 6 months for heavy metal analysis (SNI, 2018).

Heavy metal concentration analysis in the laboratory

Sample is prepared at the Laboratory of Industrial Hygiene and Toxicology, Institute Technology Bandung, Indonesia. Sample (digest with HNO_3) is prepared by transferring 100 mL of homogenized water with preservative into a beaker, adding 3 mL of sensitive HNO_3 , and covering it with a beaker glass. The beaker is heated, evaporated to less than 5 mL, and then cooled. The walls of the beaker and beaker glass are rinsed with distilled water, added with 5 mL of concentrated HNO_3 , and reheated to boiling. Heating is carried out until NO_2 gas is formed (generally indicated by a color change to brownish yellow followed by the reflux process). After cooling, the sample is added with 10 mL of 1:1 HCL and 15 mL of distilled water, reheated for 15 minutes, and cooled again. Beaker glasses are washed again with distilled water and filtered. The filtrate is transferred to a 100 mL volumetric flask (SNI, 2018). The concentration of heavy metals is measured by ICP-OES at the Central Laboratory, Directorate of Research and Community Service, Padjadjaran University, Bandung City, West Java, Indonesia. Prior to measurement, a calibration blank solution and a calibration curve are prepared.

COPC analysis

COPCs are substances that are potentially hazardous to human health. The COPCs identified will be used for further evaluation in the risk assessment process. COPCs are selected from the chemical of interest (COI) by using the following general approach (Woodward-Clyde, 1998; MDEP, 2021).

§ If the concentration of heavy metals exceeds the applicable screening level (SL) or risk-based

concentration (RBC), then these heavy metals are considered as COPCs.

§ If the concentration of heavy metals is less than the applicable SL or RBC, then these heavy metals are not considered as COPCs.

RBC or SL is a risk-based concentration (benchmark) from a standard equation that combines the assumptions of exposure information with toxicity data. This RBC equation is adopted from the EPA Superfund Program Risk Assessment Guide from USEPA (2022b). RBC is a useful protector for humans (including sensitive groups). RBCs can help identify areas, types of contaminants, and conditions that require further attention. In this study, the calculated RBC is the RBC on tap water–residents. These receptors are exposed to chemicals in water that is delivered to the residence from sources such as groundwater or surface water. Swallowing drinking water is the proper pathway for all chemicals. Inhalation exposure pathway is considered for volatile compounds but is not considered in the present study. Activities such as bathing and washing contribute to dermal absorption. RBC will be calculated for cancer effects and non-cancer effects and on the pathway of exposure through ingestion (oral) and skin contact (dermal). The equation used is a special equation for inorganic compounds (not organic) because the 10 heavy metals analyzed (As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Pb, and Zn) are inorganic compounds. The equations used to calculate RBC are as follows:

A. Non-cancer effects

a. Oral (Ingestion) pathway, using Eq. 1 (USEPA, 2022b).

$$(RBC_{oral})_{NC} = \frac{THQ \times AT \times BW}{\left(\frac{1}{RfD_o}\right) \times EF \times ED \times IRW} \quad (1)$$

b. Dermal absorption pathway, using Eq. 2 (USEPA, 2022b).

$$(RBC_{dermal})_{NC} = \frac{DA_{event}}{Kp \times ET} \quad (2)$$

Where it is used as Eq. 3 (USEPA, 2022b).

$$DA_{event} = \frac{THQ \times AT \times BW}{\left(\frac{1}{RfD_o \times GIABS}\right) \times EF \times ED \times EV \times SA} \quad (3)$$

c. Non-cancer effects RBC, using Eq. 4 (USEPA, 2022b).

$$RBC_{NC} = \frac{1}{\frac{1}{(RBC_{oral})_{NC}} + \frac{1}{(RBC_{dermal})_{NC}}} \quad (4)$$

B. Cancer effects

a. Oral (Ingestion) pathway, using Eq. 5 (USEPA, 2022b).

$$(RBC_{oral})_C = \frac{TR \times AT \times LT}{CSF_o \times IFW} \quad (5)$$

b. Dermal absorption pathway, using Eq. 6 (USEPA, 2022b).

$$(RBC_{dermal})_C = \frac{DA_{event}}{Kp \times ET} \quad (6)$$

Where, it is used as Eq. 7 (USEPA, 2022b).

$$DA_{event} = \frac{TR \times AT \times LT}{\left(\frac{CSF_o}{GIABS}\right) \times DFW} \quad (7)$$

c. Cancer effects RBC, it is used as Eq. 8 (USEPA, 2022b).

$$RBC_C = \frac{1}{\frac{1}{(RBC_{oral})_C} + \frac{1}{(RBC_{dermal})_C}} \quad (8)$$

C. RBC Total, it is used as Eq. 9 (USEPA, 2022b).

$$RBC = RBC_{NC} + RBC_C \quad (9)$$

The information or assumptions used for the analysis and the default values used in the above equation are sourced from the USEPA (User's Guide – Risk Assessment - Regional Screening Levels) and USEPA (IRIS – Integrated Risk Information System – IRIS Assessments). Table 1 shows information related to the default values used to calculate RBC non-cancer and cancer effects. Table 2 shows the RfD, Kp, GIABS, and CSF values of each heavy metal.

Based on the results of a review from the USEPA (IRIS – Integrated Risk Information System – IRIS Assessments), the types of heavy metals that are carcinogenic are As, Cd, Cr, and Pb, while Cu, Hg, Mn, and Zn are not carcinogenic. Information on the types of heavy metals such as Co and Fe is not available in the USEPA (IRIS Assessments).

Table 1: Default values used to calculate RBC USEPA (2022c)

Symbol	Information	Unit	Adult	Child
Non-cancer effects				
THQ	Target hazard quotient	Unitless	1	1
BW	Body weight	kg	80	15
AT	Averaging time (365 days/y*ED)	days	9490	2190
IR	Ingestion rate	L/day	2.5	0.78
ET	Exposure time	hours/event	0.71	0.54
EF	Exposure frequency	days/y	350	350
ED	Exposure duration - resident	years	26	6
EV	Resident events	event/day	1	1
SA	Resident surface area water - adult	cm ²	19652	6365
Cancer effects				
TR	Target Risk	Unitless	1 x 10 ⁻⁶	*
ET	Resident water exposure time	hours/event	0.71	*
DFW	Resident water dermal contact factor	cm ² -event/kg	2610650	*
LT	Lifetime	years	70	*
IFW	Resident drinking water ingestion rate	L/kg	327.95	*
AT	Averaging time	days/y	365	*

Table 2: RfD, Kp, GIABS and CSF values of each heavy metal

Heavy metals	RfD	RfD Source	Volatile	Chemical type	Kp (cm/h)	GIABS	CSF oral	CSF source
As	0.0003	IRIS	No	Inorganics	0.001	1	1.5	EPA IRIS
Cd	0.0005	IRIS	No	Inorganics	0.001	0.05	15	CAL EPA ²
Co	0.0003	PPRTV	No	Inorganics	0.0004	1	*	*
Cr	0.003	IRIS	No	Inorganics	0.001	0.013	0.5	CAL EPA ²
Cu	0.04	HEAST	No	Inorganics	0.001	1	*	*
Fe	0.7	PPRTV	No	Inorganics	0.001	1	*	*
Hg	0.00016	Cal EPA ¹	Yes	Inorganics	0.001	1	*	*
Mn	0.14	IRIS	No	Inorganics	0.001	0.04	*	*
Pb	0.0014	Nag and Cummins, 2022	No	Inorganics	0.0001	1	0.0085	Cal/OEHHA
Zn	0.3	IRIS	No	Inorganics	0.0006	1	*	*

RfD = Reference dose (mg/kg-day); Kp = Dermal Permeability Constant (cm/h); GIABS = Fraction of Contaminant Absorbed in Gastrointestinal Tract (unitless); CSF = Cancer Slope Factor (mg/kg-day); CAL EPA¹ = The California Environmental Protection Agency Office of Environmental Health Hazard Assessment; CAL EPA² = California Environmental Protection Agency, U.S in Zeng *et al.*, 2015; Cal/OEHHA = California Office of Environmental Health Hazard Assessment in Parker *et al.*, 2022; PPRTV = Provisional peer reviewed toxicity values; IRIS = Integrated risk information system; HEAST = The EPA superfund program's health effects assessment summary table

Spatial distributions

Heavy metal concentrations are visualized into a spatial distribution map. Interpolation is a process where the value of an attribute in a non-sampled area is predicted using data that can be accessed from another sampling area. Inverse distance weighted (IDW) is the interpolation method used to visualize heavy metal concentrations and evaluate water quality. Interpolation considers the geographic location of the sample points. IDW is an optimal interpolation model used to assess the spatial distribution pattern of heavy metal concentrations in the study area (Saha *et al.*, 2022). This IDW interpolation technique assumes that variables that are close to each other are more similar than those that are far apart. This technique analyzes the values obtained around the predicted point to estimate the value for each unobserved point. Measured values closer to the predicted location have a greater impact on projected values than those further away (Saha *et al.*, 2022). Eq. 10 is used in this interpretation (Bhunja *et al.*, 2018).

$$Z = \frac{\sum_{i=1}^n \left(\frac{Z_i}{d_i^p} \right)}{\sum_{i=1}^n \frac{1}{d_i^p}} \quad (10)$$

where z is the estimated value at the interpolation point, z_i is the measured value at point i , n is the total

number of measured values used in the interpolation, d_i is the distance between the interpolated value and the measured value z_i , and p represents the weighting power that determines how the weight is reduced by increasing the distance.

Statistical analysis

Data are statistically analyzed using the statistical package for the social sciences (SPSS) version 26.0 with correlation analysis to investigate the possibility of a positive or negative relationship between heavy metal concentrations in groundwater. Significance is set at $p < 0.05$. Kruskal-Wallis test is used to analyze significant differences in heavy metal concentrations in areas with different topographies, namely, high topography (Pacet and Pangalengan) and low topography (Majalaya, Rancaekek, Ciparay, Baleendah, and Soreang). The test is used for data that are not normally distributed (non-parametric test) and continued with a follow-up test to determine the location of the difference in heavy metal concentrations. Differences were considered significant at $p < 0.05$.

RESULTS AND DISCUSSION

Heavy metal concentration and spatial distribution map

For sanitation and hygiene (SH)




Table S3 shows the statistical characteristics of the

Table 3: Matrix that presents the order of the average concentration of heavy metals in sanitation and hygiene water from highest to lowest

Sub-districts	As*	Cd	Co*	Cr*	Cu*	Fe	Hg	Mn	Pb	Zn*
Ciparay	2	7	5	1	2	2	4	4	6	5
Majalaya	7	2	6	6	6	1	5	6	2	6
Rancaekek	1	1	7	2	5	3	6	5	1	4
Baleendah	4	4	4	5	1	4	1	7	5	1
Soreang	5	3	2	7	7	5	3	2	3	3
Pacet	3	5	3	4	3	6	7	1	4	7
Pangelangan	6	6	1	3	4	7	2	3	7	2

*The concentration complies the water quality standard "Regulation of the Health Minister of the Republic of Indonesia No. 32 of 2017" at 100% samples.

**The concentration complies water quality standard Environmental Working Group - EWG (2021) at 100% samples.

1 2 3 4 5 6 7 = Order from highest to lowest concentration
 Average concentration of heavy metals (HMs) < Quality standard and all samples < Quality Standard.
 Average concentration of HMs < Quality standard and some samples > Quality Standard
 Average concentration of HMs > Quality standard and some samples > Quality Standard

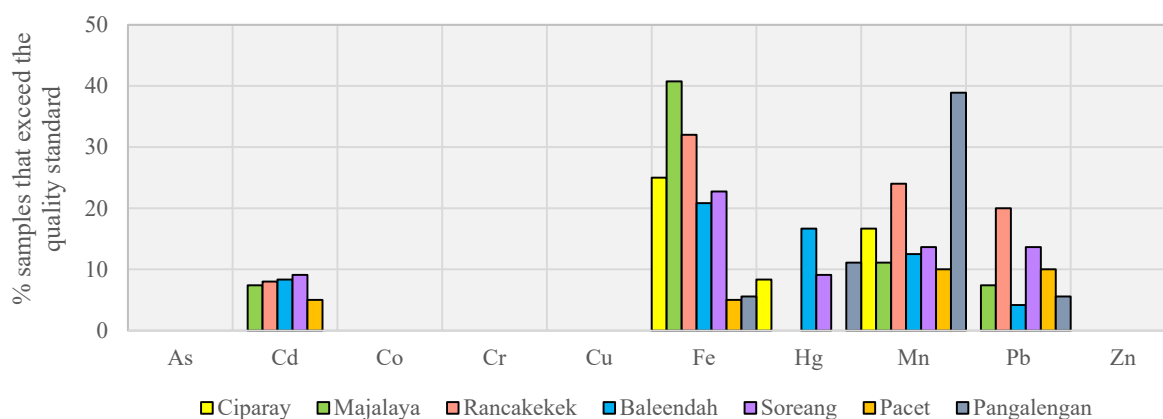


Fig. 2: Percentage of the number of samples whose concentration of heavy metals exceeds the sanitation and hygiene water quality standard in seven sub-districts

analyzed heavy metal concentrations in groundwater used for SH activities. In terms of the minimum and maximum concentration values of the 10 heavy metals, all of them were detected in groundwater samples used for SH; hence, As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Pb, and Zn are COI in this study. In detail, the order of the average concentration of heavy metals from the highest to the lowest is presented in Table 3 and Fig. 3.

Table 3 shows that in 100% of the SH water samples analyzed, the contents of As, Co, Cr, Cu, and Zn still meets the water quality standards for sanitation hygiene based on the Regulation of the Minister of Health of the Republic of Indonesia No. 32 of 2017, although the average concentration has been sorted

from the highest to the lowest. In contrast to Cd, Fe, Hg, Mn, and Pb, some of the samples do not meet the water quality standards for SH. The highest average concentration (\pm SD, in mg/L; Table S3) of Cd was found in Rancaekek (0.0015 ± 0.0025), Fe in Majalaya (1.7465 ± 3.1291), Hg in Baleendah (0.0135 ± 0.0571), Mn in Pacet (1.2254 ± 3.3744), and Pb in Rancaekek (0.0291 ± 0.0506). Fig. 2 shows the percentage of samples whose heavy metal concentrations exceed the SH water quality standard. The heavy metals of concern in groundwater for SH purposes are Cd, Fe, Hg, Mn, Pb and Zn. The level of Cd exceeds the quality standard in five sub-districts, namely, Soreang (9% of groundwater samples exceeds the quality standard) > Rancaekek (8%) = Baleendah (8%) > Majalaya

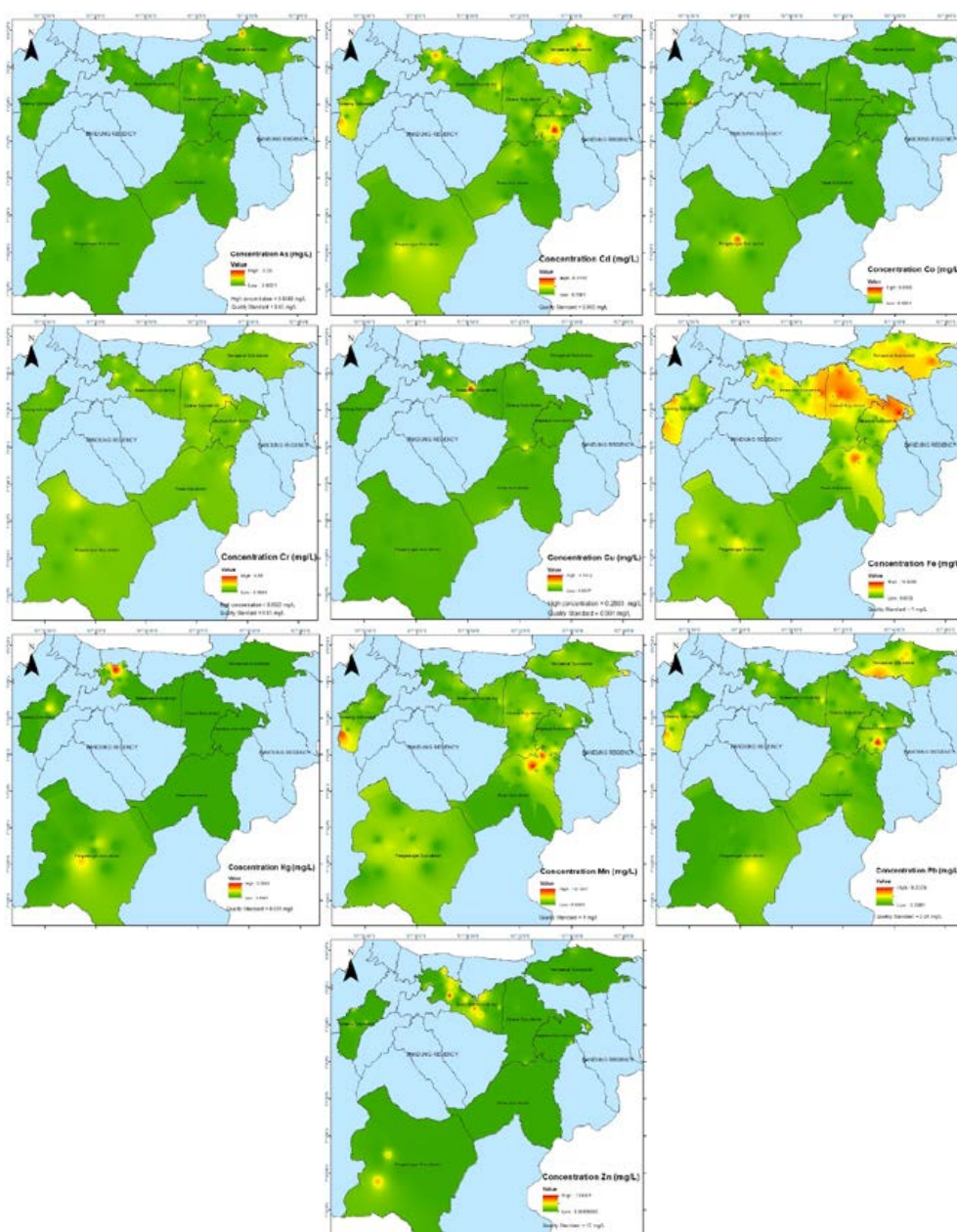


Fig. 3: Spatial distribution map of 10 heavy metals in sanitation and hygiene water

(7%) Pacet (5%); Fe in seven sub-districts, namely, Majalaya (41%) > Rancaekek (32%) > Ciparay (25%) > Soreang (23%) > Baleendah (21%) > Pangalengan (6%) and Pacet (5%); Hg in four sub-districts, namely, Baleendah (17%) > Pangalengan (11%) > Soreang (9%) > Ciparay (8%); Mn in seven sub-districts, namely,

Pangalengan (39%) > Rancaekek (24%) > Ciparay (17%) > Soreang (14%) > Baleendah (13%) > Majalaya (11%) > Pacet (10%); and Pb in six sub-districts, namely, Rancaekek (20%) > Soreang (14%) > Pacet (10%) > Majalaya (7%) > Pangalengan (6%) > Baleendah (4%). In particular, although the average concentration of

Mn is the highest in Pacet, the highest percentage of groundwater samples exceeding the quality standard was found in Pangalengan. This finding could be due to the distribution of high Mn concentrations (marked in red in Fig. 3) in Pacet.

Table 3 and Fig. 2 show that Rancaekek, Majalaya, and Soreang sub-districts need attention because of their similar pattern their higher average concentrations of Pb and Cd than the other sub-districts. They also have the same pattern (Fig. 3):

1. Rancaekek has the highest average concentrations of Pb and Cd in groundwater; about 20% of groundwater samples exceeded the Pb quality standard and 8% exceeded the Cd quality standard.

2. Majalaya has the second highest average concentrations of Pb and Cd in groundwater; 7% of groundwater samples exceeded the Pb quality standard and 7% exceeded the Cd quality standard.

3. Soreang has the third highest average concentrations of Pb and Cd in groundwater; 14% of groundwater samples exceeded the Pb quality standard and 9% exceeded the Cd quality standard.

Pb and Cd were found to have a significant positive correlation (Table 7; $p < 0.05$; $r = 0.575$). Pb and Cd in the three sub-districts may have originated from anthropogenic sources. Based on Table S2, these sub-districts have high distributions of the textile industry. The release of large amounts of wastewater containing heavy metals, such as Pb and Cd, is unavoidable from the textile industry because their raw materials are mixed with fibers during the dyeing process (Velusamy et al., 2021). Another possible source of Pb and Cd in groundwater is river water. The distribution pattern of high Pb and Cd concentrations is located in the area adjacent to the river (Fig. 3 and Fig. S1). Heavy metal pollution in rivers may enter the groundwater system through infiltration and percolation. The West Java government has implemented the Citarum Harum Program as an effort to improve water quality in the Citarum River and its surroundings. This program has been regulated through Presidential Regulation Number 15 of 2018 concerning the Acceleration of Pollution Control and Damage to the Citarum River Basin. The program includes handling industrial waste, livestock waste, domestic waste, law enforcement, and others. The program succeeds in improving the water quality of the Citarum River, from moderately polluted in 2019 to slightly polluted in 2020 (JOD,

2020). However, heavy metals are non-degradable and persistent in the environment (Ali et al., 2019; Jeyakumar et al., 2023) and are difficult to clean completely in the environment; this phenomenon may be one of the reasons for the discovery of heavy metals in the present study. In addition, the presence of Hg in Baleendah, Pangalengan, Soreang, and Ciparay needs attention; in particular, Baleendah has the highest average concentration of Hg (Fig. 3), with 17% of groundwater samples exceeding the quality standard (Fig. 2); Hg in Pangalengan has the second highest average concentration, with 11% of groundwater samples exceeding the quality standard; Hg in Soreang has the third highest average concentration, with 9% of groundwater samples exceeding the quality standard; and Hg in Ciparay has the fourth highest average concentration, with 8% of groundwater samples exceeding the quality standard. The high concentrations of Hg in these sub-districts can be sourced from natural and anthropogenic sources. Naturally, Hg originates from volcanoes, geothermal springs, geological deposits, and oceans (USGS, 2019; Karbassi and Heidari, 2015). Given that Hg in the sub-district exceeds the water quality standard, it possibly originates from human activities (anthropogenic). Hg is thought to have come from illegal small-scale gold mining or from burning coal. Globally, artisanal and small-scale gold mining (ASGM) is the largest source of anthropogenic mercury emissions (37.7%), followed by stationary burning of coal (21%) (USEPA, 2022a). Based on Fig. 2 and Table 3, Fe in groundwater also shows a certain pattern from the seven sub-districts studied; it has a high average concentration from order 1 to sequence 4 in Majalaya, Ciparay, Rancaekek, and Baleendah (areas with low topography) and a low mean concentration from order 5 to 7 in Soreang, Pacet, and Pangalengan (high topography) (the concentration distribution pattern can be seen in Fig. 3). Based on the results of the statistical analysis in Table 8, Fe significantly differed among different topographical areas ($p < 0.05$). The number of groundwater samples that exceed the quality standard is higher in Majalaya (41%), Ciparay (25%), Rancaekek (32%), Baleendah (21%), and Soreang (23%) than in Pacet (5%) and Pangalengan (6%). In contrast to Fe, Mn has high average concentrations from order 1 (highest) to 3 (Table 3) in Pacet, Soreang, and Pangalengan (high topography) and low average concentrations

(orders 4 to 7) in Ciparay, Rancaekek, Majalaya, and Baleendah (low topography); however, the difference in the concentrations is not significant ($p > 0.05$; Table 8). When viewed from the percentage of samples that exceeded the groundwater quality standard for SH purposes (Fig. 2), the result has no pattern that can inform factors that influence or the source of Mn in groundwater. Fe and Mn in groundwater have a significant positive correlation (see Table 7, $p < 0.05$; $r = 0.498$), indicating that they may have originated from the same source (*natural source*) and influenced by topography, soil properties/types, and/or rock formations. The high average concentration of Fe in the present study (especially in Majalaya, Ciparay, Rancaekek, and Baleendah) is thought to be more influenced by topographic factors and soil properties/types. These sub-districts are areas with low topography (Fig. S2) and low slope (Fig. S3) and are dominated by alluvial soil types (soil derived from sediment processes or textured sediments such as clay) (Fig. S4). Meanwhile, Soreang, Pacet, and Pangalengan are located in a high topographic area; the latter two are dominated by andosol soil types (soil originating from volcanic activity), and the former is dominated by latosol soils (soil formed from weathering with high intensity and overgrown by trees). Fe in groundwater may increase with decreasing altitude, indicating that the concentration of Fe is high in areas with low topography. In areas with high topography, the residence time of Fe is relatively lower because dissolved Fe will migrate to areas with lower topography. By contrast, the concentrations of Fe and Mn are higher in areas with low topography because they have a longer residence time in groundwater (Zhai *et al.*, 2021).

The main mineral in clay is layered aluminosilicate containing Fe. Clay soil contains fine particles and small pores between particles so it is easy to become a reducing environment that is conducive to the reductive dissolution of solid Fe (Zhai *et al.*, 2021). The average concentration of Mn is higher in Pacet, Soreang, and Pangalengan than in Majalaya, Ciparay, Rancaekek, and Baleendah. This finding is thought to be influenced by rock formations found in the area. The Mn content is high in limestone or limestone formations (Force and Cox, 1916). Pacet, Soreang, and Pangalengan sub-districts are areas dominated by Plio-Pleistocene volcanic rock formations (Fig. S5). Volcanic rock products deposited during the Plio-Pleistocene period can be in the form of breccia rocks (MEMR, 2014), which are composed of limestone. Geological factors contribute to the composition of groundwater through the influence of water-rock interactions in aquifer, where Mn in rocks can be released into the aquifer at pH 4–7, namely, the decoupling of Mn (IV) in an insoluble form in rock to Mn (II) dissolved form in aquifers (Kousa *et al.*, 2021).

For drinking water or consumption water

Table S4 shows the statistical analysis of the concentrations of heavy metals in drinking water. In detail, the order of the average concentration of heavy metals from the highest to the lowest is presented in Table 4 and Fig. 5.

Table 4 shows that 100% of the consumption water samples analyzed for Cr, Cu, and Zn content still meet the drinking water quality standards in accordance with the Regulation of the Minister of Health of the Republic of Indonesia No. 492 of 2010, although the average concentrations are sorted from the highest

Table 4: Matrix that presents the order of the average concentration of heavy metals in drinking water from highest to lowest

Sub-districts	As	Cd	Co	Cr	Cu*	Fe	Hg	Mn	Pb	Zn*
Ciparay	1	3	4	1	3	1	5	3	4	5
Majalaya	7	5	6	5	5	7	7	6	5	6
Rancaekek	5	1	2	2	4	3	1	4	1	4
Baleendah	4	7	7	7	6	6	3	7	7	1
Soreang	3	6	1	6	7	5	2	5	3	2
Pacet	2	2	3	4	2	2	6	1	2	7
Pangalengan	6	4	5	3	1	4	4	2	6	3

* The concentration complies the water quality standard Regulation of the Health Minister of Indonesia No. 492/2010 at 100% samples

The quality standard for heavy metal concentrations refers to Regulation of the Health Minister of Indonesia No. 492/2010, except for Co which refers to the Agency for Toxic Substances and Disease Registry – ATSDR (2004).

- 1 2 3 4 5 6 7 = Order form highest to lowest concentration
- Average concentration of heavy metals (HMs) < Quality standard and all samples < Quality standard
- Average concentration of HMs < Quality standard and some samples > Quality standard
- Average concentration of HMs > Quality standard and some samples > Quality standard

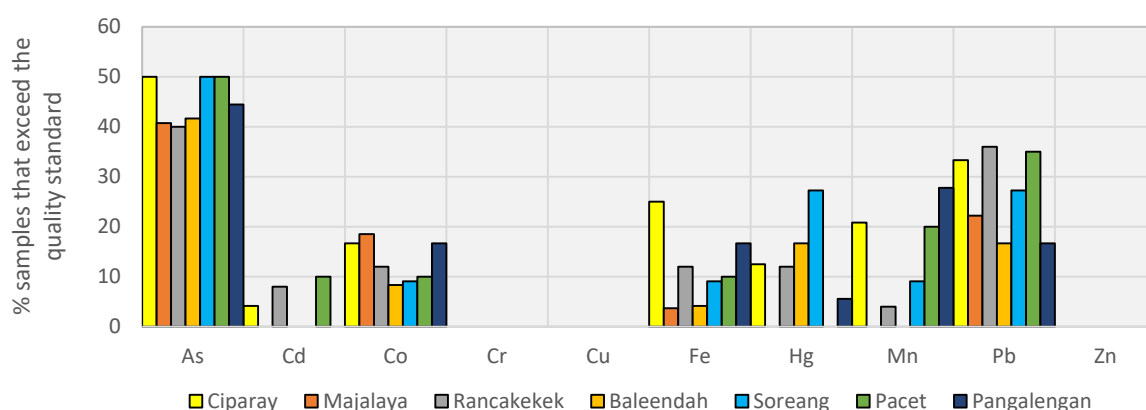


Fig. 4: Percentage of the number of samples whose heavy metal concentrations exceed the consumption water quality standard in seven sub-districts

Table 5: Percentage of total consumption water sample that exceed the quality standard for groundwater and from refill water/spring/ rainwater

Heavy Metals	Type of water consumption	Ciparay	Majalaya*	Rancaekek**	Baleendah	Soreang	Pacet	Pangalengan
As	Well water	41.67	18.18	10.00	30.00	27.27	100.00	50.00
	Refill water, etc	58.33	81.82	90.00	70.00	72.73	0.00	50.00
Cd	Well water	0.00	0.00	0.00	0.00	0.00	100.00	0.00
	Refill water, etc	100.00	0.00	100.00	0.00	0.00	0.00	0.00
Co	Well water	75.00	0.00	0.00	50.00	100.00	100.00	100.00
	Refill water, etc	25.00	100.00	100.00	50.00	0.00	0.00	0.00
Fe	Well water	83.33	100.00	33.33	100.00	50.00	100.00	100.00
	Refill water, etc	16.67	0.00	66.67	0.00	50.00	0.00	0.00
Hg	Well water	66.67	0.00	0.00	25.00	16.67	0.00	100.00
	Refill water, etc	33.33	0.00	100.00	75.00	83.33	0.00	0.00
Mn	Well water	60.00	0.00	100.00	0.00	100.00	100.00	100.00
	Refill water, etc	40.00	0.00	0.00	0.00	0.00	0.00	0.00
Pb	Well water	50.00	0.00	11.11	25.00	33.33	100.00	33.33
	Refill water, etc	50.00	100.00	88.89	75.00	66.67	0.00	66.67

* In Majalaya, 1 sample of drinking water from springs did not detect various types of heavy metals

** In Rancaekek, 1 sample of drinking water from rainwater detected As and Pb with concentrations exceeding the quality standard.

to the lowest. The levels of As, Cd, Co, Fe, Hg, Mn, and Pb do not meet the drinking water quality standards in few samples, similar to previous reports (Abeer et al., 2020; Hossain et al., 2023; Rahman et al., 2023). As and Co do not exceed the quality standard for SH purposes (Table 3, Fig. 2) but exceed that of the quality standard for drinking water; this finding is due to the fact that the quality standard for drinking water is stricter than for SH purposes. The highest average concentrations (\pm SD, in mg/L; Table S4) of As were found in Ciparay (0.0129 ± 0.0108), Cd in Rancaekek (0.0017 ± 0.0047), Co in Soreang (0.0018 ± 0.0041), Fe in Ciparay (0.5528 ± 1.5324), Hg in Rancaekek (0.0232 ± 0.1016), Mn in Pacet (1.2254 ± 3.3744), and Pb in Rancaekek (0.0171 ± 0.0316) (details

regarding the spread of high concentrations in Fig. 5). The percentage of samples whose heavy metal concentrations exceed the quality standards for consumption water is presented in Fig. 4.

Among the seven heavy metals that were found to be above the quality standards, As and Pb were detected in the largest number of samples and exceeded the quality standards for drinking water, with average values of 45% and 27%, respectively (Fig. 4). Other types of heavy metals are sorted as follows: Co (13%) > Mn (11.7%) > Fe (11.52%) > Hg (10.57%) > Cd (3.17%). Based on Table 5, As in drinking water is more dominant, with concentrations exceeding the quality standards in water other than groundwater (refill water or rainwater) (90% in Rancaekek >

81.82% in Majalaya > 72.723 in Soreang > 70% in Baleendah > 58.33% in Ciparay > 50% in Pangalengan), compared with groundwater (100% in Pacet > 50% in Pangalengan > 41.67% in Ciparay > 30% in Baleendah > 27.27% in Soreang > 18.18 % in Majalaya > 10% in Rancaekek). Pb with concentrations that exceed the quality standard is more dominant in drinking water from water sources other than groundwater (Table 5). In refill water, the heavy metals detected may be related to the raw water source used at the Refill Drinking Water Depot (DAMIU); the technology used cannot remove heavy metals, and its maintenance process is not optimal. According to Hasanawi and Salami (2022), ultrafiltration (UF) is the processing technology generally used by DAMIU in Bandung Regency as reported by 83% of the respondents, and ozonation is used by 17% of the respondents. The UF process can effectively remove microorganisms and other pathogenic bacteria (Pei and Duo, 2022) and colloidal particles (Nguyen, 2014) but cannot remove heavy metals in certain speciation forms, such as dissolved charged ions (Bernat et al., 2007). Hence, UF can remove heavy metals in particulate form (adsorbed on particulates) but cannot remove those in dissolved forms because UF does not have a charged membrane on the surface and its pore size is too large (Nguyen, 2014). In drinking water sourced from groundwater, the sources of heavy metals have been described previously, except for As and Co. The source of arsenic in groundwater is thought to be of natural origin. As shown in Table 7, As and Fe are positively and significantly correlated in the Spearman test, but the correlation is very weak ($p < 0.05$; $r = 0.162$). As can bind to Fe(III)-oxyhydroxides or Fe oxides in soils. The reductive dissolution of Fe(III)-oxyhydroxides or iron oxides is the main geochemical mechanism of the release of soil As into groundwater (Maity et al., 2011). Arsenopyrite (FeAsS) is the most abundant mineral that contains As and is generally found in the environment and in various rock-forming minerals, such as sulfides, oxides, phosphates, carbonates, and silicates (Smedley and Kinniburgh, 2002). Given the weak correlation between As and Fe, other sources of As in groundwater may exist, such as from textile industry (Singha et al., 2021), use of pesticides in agriculture (Hooda, 2010; DHSS, 2013; Kayode et al., 2020), use of fertilizers in agriculture (Jayasumana et al., 2015), and coal-fired power plants (DHSS, 2013). Furthermore, the source of Co in groundwater is

difficult to discuss because the results of the Spearman correlation were not significant compared with those of the other heavy metals ($p > 0.05$ in Table 7). The results of the Kruskal Wallis analysis did not show any significant differences based on topography ($p > 0.05$ in Table 8), but try to explain based on the dominant land use. Table 3 shows the average concentration of Co in seven sub-districts, where the highest average concentration was found in Pangalengan, Soreang, and Pacet (dominant in areas with a high percentage of agriculture; Table S1) and the lowest concentration was detected in Rancaekek, Ciparay, Majalaya, and Baleendah (predominant in areas with a low percentage of agriculture; Table S1). Thus, Co is thought to originate from agricultural activities, such as from the use of fertilizers. Co is also an essential micronutrient for plant growth (Hu et al., 2021). One of the fertilizers sold freely in Indonesia contains 0.27 ppm Co. However, the type of fertilizer used in the study area remains unknown. Rainwater is another source of drinking water for one of the respondents in Rancaekek; the sample has As concentration that exceeded the quality standard. The increased concentration of arsenic (As) is due to mineral dust particles entering the system, when rainwater interacts with the roof catchment (Quaghebeur et al., 2019) and may be the reason for the high Pb in rainwater.

Chemical of potential concerns For sanitation and hygiene (SH)

Groundwater is used by respondents for sanitation and hygiene. Heavy metals in water have the potential to enter the respondent's body through the dermal pathway because water is only used for SH and not used for drinking, so the RBC calculated refers to only the RBC of the dermal pathway and does not consider the RBC of the oral pathway. Based on information from IRIS USEPA, metals that have carcinogenic risks are As, Cd, Cr, and Pb, while Co, Cu, Fe, Hg, Mn, and Zn are not carcinogenic; hence, RBC is calculated only on non-cancer effects and does not consider cancer effects. Table S5 shows the matrix for determining COPCs based on the comparison of the concentration values of heavy metals from SH water with RBC or SL in adults. Table S5 shows that the maximum concentration of As is smaller than that of RBC As (0.0448 mg/L < 1.80298 mg/L), and the same is true for other metals such as Cd, Co, Cr, Cu, Fe, Hg, Mn, Pb, and

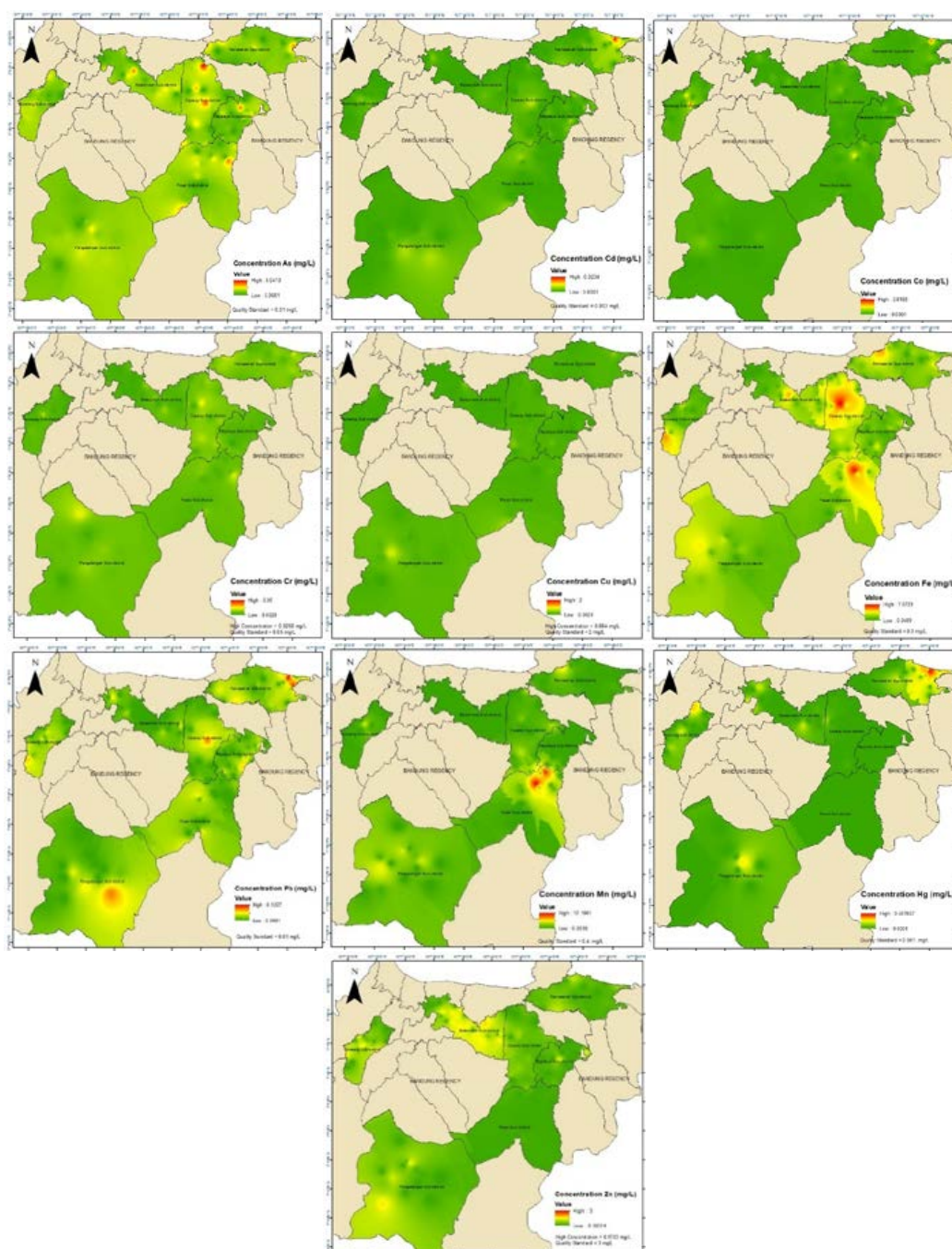


Fig. 5: Spatial distribution map of 10 heavy metals in drinking water

Zn, with values of 0.0157 mg/L < 59.7928 mg/L; 0.0171 mg/L < 4.48447 mg/L; 0.0323 mg/L < 1379.84 mg/L; 0.1414 mg/L < 239.172 mg/L; 14.9385 mg/L < 4185.54 mg/L; 0.2803 mg/L < 0.95669 mg/L; 12,2943 mg/L <

20927.52 mg/L; 0.2588 mg/L < 99.92687 mg/L; and 1.0918 mg/L < 2989.65 mg/L, respectively. All heavy metal concentrations were concluded to be lower than the RBC values, so they were not retained as COPCs.

Skin contact (dermal), in contrast to oral consumption, does not pose a potential risk (Luo *et al.*, 2022). As such, determination of COPCs will be continued by calculating RBC from oral intake. In addition, the potential risk of children is higher than that of adults (Gao *et al.*, 2019; Xiao *et al.*, 2019), so determination of COPCs will be continued by calculating RBC in children. Table S6 shows the matrix for determining COPCs based on the comparison of the concentrations of heavy metals from SH water with RBC or SL in children. Similar to the conclusion of COPCs in adults, Table S6 shows that the concentrations of As, Cd, Co, Cr, Cu, Fe, Hg, Mn, Pb, and Zn in SH water are smaller than the RBC in children. Thus, these metals are not retained as COPCs. The results of determining COPCs cannot be seen only by the comparative analysis of the use of SH water for adults and children. Without considering the risk of oral intake, the heavy metals inferred as COPCs have not been seen because oral intake is the main pathway for potential health risks. Table S6 shows that the order of RBC from highest to lowest: Mn (15929.126 mg/L) > Fe (3185.8253 mg/L) > Zn (2275.5895 mg/L) > Cr (1050.2721 mg/L) > Cu (182.0472 mg/L) > Pb (63.7165 mg/L) > Cd (45.5118 mg/L) > Co (3.4134 mg/L) > As (1.3654 mg/L) > Hg (0.7282 mg/L). The smaller the RBC value is, the more likely it is to pose a risk to human health, so it needs to be maintained as COPCs; however, it will depend on the concentration of heavy metals detected in the field.

For drinking water or consumption water

The RBC of drinking water was calculated for direct contact (oral pathway) with drinking water to determine COPCs posing a risk to human health. These RBCs were calculated in adults (Table 5) and children (Table 6). In drinking water, the RBC on skin contact (dermal) was not considered and only referred to RBC from the oral pathway and RBC cancer effects only.

Table 5 confirms that heavy metals such as As, Cd, Co, Hg, Mn, and Pb can be maintained as COPCs because their maximum concentrations are greater than RBC, with values of 0.0416 mg/L > 0.0101 mg/L; 0.0234 mg/L > 0.0167 mg/L; 0.0171 mg/L > 0.0100 mg/L; 0.5090 mg/L > 0.0053 mg/L; 12.2943 mg/L > 4.6720 mg/L; and 0.1229 mg/L > 0.0559 mg/L, respectively. From the 160 samples analyzed, the concentration of As greater than the COPC value is 45.57%, Cd is 0.63%, Co is 1.27%, Hg is 7.59%, Mn is 1.27%, and Pb is 5.06%. Therefore, heavy metals such as As, Cd, Co, Hg, Mn, and Pb have the potential to pose a risk to human health; moreover, some sample points need attention because 45.57% have the potential to pose health risks from As (less than 10%) and from low percentage (Cd, Co, Hg, Mn and Pb). Arsenic concentrations in drinking water to levels that pose a health risk were found in Pakistan (Abeer *et al.*, 2020), India (Ravindra and Mor, 2019),

Table 5: Matrix for determining COPCs from the comparison of heavy metal concentrations for consumption water with RBC or SL for adults; n = 160

Const.	Max. Conc.	Cancer Effects			Non Cancer Effects			RBC = RBC _c + RBC _{nc} (Screening Level)	Retain as COPC		
		RBC oral	RBC dermal	Calc. Goals	RBC oral	RBC dermal	Calc. Goals		Based on Max. Conc.	Based on 160 samples	
		(RBC _o) _c	(RBC _d) _c	RBC _c	(RBC _o) _{nc}	(RBC _d) _{nc}	RBC NC			Yes (%)	No (%)
		mg/L	mg/L	(mg/L)	mg/L	(mg/L)					
		TCR = 1 x 10 ⁻⁶			THQ = 1						
As	0.0416	0.0001	**	0.0001	0.0100	**	0.0100	0.0101	Yes	45.57	54.43
Cd	0.0234	0.0000	**	0.0000	0.0167	**	0.0167	0.0167	Yes	0.63	99.37
Co	0.0171	*	**	-	0.0100	**	0.0100	0.0100	Yes	1.27	98.73
Cr	0.0268	0.0002	**	0.0002	0.1001	**	0.1001	0.1003	No	0.00	100.00
Cu	0.0840	*	**	-	1.3349	**	1.3349	1.3349	No	0.00	100.00
Fe	7.6741	*	**	-	23.3600	**	23.3600	23.3600	No	0.00	100.00
Hg	0.5090	*	**	-	0.0053	**	0.0053	0.0053	Yes	7.59	92.41
Mn	12.2943	*	**	-	4.6720	**	4.6720	4.6720	Yes	1.27	98.73
Pb	0.1229	0.0092	**	0.0092	0.0467	**	0.0467	0.0559	Yes	5.06	94.94
Zn	0.9703	*	**	-	10.0114	**	10.0114	10.0114	No	0.00	100.00

*RBC_{oral} is not calculated because water is used only for SH

**RBC for cancer effects is not calculated because heavy metals is non-carcinogenic

Table 6: Matrix for determining COPCs from the comparison of heavy metal concentrations for consumption water with RBC or SL for children; n = 160

Const.	Max. Conc.	Cancer Effects			Non Cancer Effects			RBC = RBC _c + RBC _{nc} (Screening Level)	Retain as COPC		
		RBC oral	RBC dermal	Calc. Goals	RBC oral	RBC dermal	Calc. Goals		Based on Max. Conc.	Based on 160 samples	
		(RBC _o) _c	(RBC _d) _c	RBC _c	(RBC _o) _{nc}	(RBC _d) _{nc}	RBC NC			Yes (%)	No (%)
		mg/L	mg/L	(mg/L)	mg/L	(mg/L)					
		TCR = 1 x 10 ⁻⁶			THQ = 1						
As	0.0416	*	*	-	0.0060	**	0.0060	0.0060	Yes	63.29	36.71
Cd	0.0234	*	*	-	0.0100	**	0.0100	0.0100	Yes	0.63	99.37
Co	0.0171	*	*	-	0.0060	**	0.0060	0.0060	Yes	2.53	97.47
Cr	0.0268	*	*	-	0.0602	**	0.0602	0.0602	No	0.00	100.00
Cu	0.0840	*	*	-	0.8022	**	0.8022	0.8022	No	0.00	100.00
Fe	7.6741	*	*	-	14.0385	**	14.0385	14.0385	No	0.00	100.00
Hg	0.5090	*	*	-	0.0032	**	0.0032	0.0032	Yes	8.86	91.14
Mn	12.2943	*	*	-	2.8077	**	2.8077	2.8077	Yes	1.27	98.73
Pb	0.1229	*	*	-	0.0281	**	0.0281	0.0281	Yes	9.49	90.51
Zn	0.9703	*	*	-	6.0165	**	6.0165	6.0165	No	0.00	100.00

*) For Children, RBC_{cancer effects} are not calculated**) RBC_{oral} is not calculated because water is used only for SH

China (Jiang *et al.*, 2021), and Iran (Maleki and Jari, 2021). The results in children (Table 6) are the same as that in adults (Table 5), that is, the types of heavy metals that are retained as COPCs are As, Cd, Co, Hg, Mn, and Pb, but the percentage of sample points that have potential health risks children are bigger than adults, with values of 63.29% for As, 0.63% for Cd, 2.53% for Co, 8.86% for Hg, 1.27% for Mn, and 9.49% for Pb. Children have underdeveloped immune systems and unique activity patterns that make them more susceptible to heavy metal exposure (Wang *et al.*, 2019). Compared with adult body size, children absorb 40% to 90% more heavy metals ingested (ATSDR, 1990). Low-income children may be at particular risk due to poor diet lacking nutrients that can otherwise help inhibit heavy metal absorption (Bradman *et al.*, 2001). Exposure to the risk of heavy metals through consumption water intake can threaten the welfare of the community; the groups most vulnerable to this risk are children so they require special treatment (Ismael *et al.*, 2022). This study reveals that six of 10 types of heavy metals are chemicals of potential concerns (COPCs) and are sorted based on potential risks to health: As > Hg > Pb > Co > Mn > Cd. Ingestion is the main pathway for potential risk, and children are more likely to be at risk than adults. Heavy metals detected in drinking water can accumulate in the human body and can cause

side effects on human health. The bioaccumulation of heavy metals Hg, Pb, Cd, and As has a variety of toxic effects on tissues and organs of the body. They can damage tissues and even organs, reduce organ function, and cause neurological disorders, cancer, and even death (Malik and Sandhu, 2023; Pandey and Kumari, 2023). Hg, Pb, Cd, and As interfere with cellular activities including growth, proliferation (cell cycle repetition), differentiation, damage repair processes, and apoptosis. The mechanism of action for heavy metals is similar, namely, by inducing toxicities including reactive oxygen species (ROS) generation, weakening of antioxidant defenses, enzyme inactivation, and oxidative stress. Heavy metals can induce toxicity in biological systems by binding to sulfhydryl groups and forming ROS. Several toxic metals including Cd and As can cause genomic instability. Defects in deoxyribonucleic acid (DNA) repair after the induction of oxidative stress and DNA damage by Cd and As have been considered to be the cause of their carcinogenicity (Balali-Mood *et al.*, 2021). Carcinogenicity of As, Cd, and Pb may occur due to ROS generation in cells by selectively activating transcription factors, indicating that cell death may be associated with exposure to carcinogenic metals. The mechanism of carcinogenesis induced by As, Cd, and Pb is as follows.

- As can bind to DNA-binding proteins and interfere

with the DNA repair process, thereby increasing the risk of carcinogenesis (Engwa *et al.*, 2019).

- Generation of ROS by Pb is the key in changing the structure and sequence of chromosomes (Silbergeld *et al.*, 2000; Ohiagu *et al.*, 2022)
- Cd has been implicated in promoting apoptosis, oxidative stress, DNA methylation, and DNA damage (Engwa *et al.*, 2019).

Long-term arsenic exposure can cause skin lesions (pigmentation, keratosis, and skin carcinoma cancer) (Banerjee *et al.*, 2023). The effects of Hg poisoning on the human body are not limited to redness of the hands and feet; kidney failure; cardiovascular, liver, brain, and hormonal problems; and intestinal ulceration (Jyothi and Farook, 2020). Hg poisoning causes psychiatric disorders due to central nervous system (CNS) dysfunction and affects listening and speaking disorders. Intention tremor (often shaking) is a disorder that occurs in speech and mouth disorders (Kark *et al.*, 1971). Attention-deficit hyperactivity (ADHD) and ASD (autism spectrum disorder) are some of the disorders associated with mental retardation; people who are exposed to Hg also manifest abnormal behavior (Jyothi and Farook, 2020). Co can be absorbed through the gastrointestinal tract and accumulate in the liver, kidneys, pancreas, heart, skeleton, and skeletal muscles. Chronic increases in serum Co may result in adverse long-term biological effects such as immune modulation and oxidative DNA damage. Higher Co concentrations are associated with neurologic, cardiac, hematological, and endocrine toxicity. Higher Co levels in tissues compete with calcium uptake and affect other signaling processes involving hypoxic

responses, oxidative stress, and energy metabolism (Uddin and Rumman, 2020).

Statistical results

Based on the results of normality testing with Shapiro Wilk, the concentration of 10 types of heavy metals in groundwater has an abnormal distribution ($p < 0.05$), so analysis was carried out using Spearman's correlation (non-parametric). The Spearman correlation coefficient between heavy metals in groundwater is presented in Table 7.

In Table 7, a significant positive correlation exists between heavy metals. As is significantly positively correlated with Fe ($p < 0.05$), but the correlation is very weak ($r = 0.162$). Significant and weak positive correlations also occur in Co and Mn, Cr and Fe, Cr and Hg, Cr and Mn, Cr and Pb, and Cu and Pb. Cd has a significant and quite strong positive correlation with Pb ($p < 0.05$; $r = 0.575$). A strong and significant positive correlation also occurs between Cr and Cu ($p < 0.05$; $r = 0.567$) and Fe and Mn ($p < 0.05$; $r = 0.498$). The strong correlations among heavy metals indicate that they may have originated from the same source (Zhao *et al.*, 2021).

Kruskal–Wallis analysis was used to test differences in heavy metal concentrations in groundwater in the seven sub-districts (Table 8). Cr, Cu, Fe, and Zn are significantly different among several sub-districts ($p < 0.05$). Fe and Zn are significantly different in areas with high and low topography. High concentrations of Fe and Zn are found in areas at low topography and vice versa. Low concentrations of Fe and Zn are found in areas of high topography. In addition to topographic differences, the two divisions of the region have

Table 7: Spearman correlation coefficient between heavy metals in groundwater

	As	Cd	Co	Cr	Cu	Fe	Hg	Mn	Pb	Zn
As	1.000	-0.034	-0.097	0.080	0.035	0.162*	-0.140	0.082	0.105	0.079
Cd		1.000	0.071	0.063	0.106	0.060	0.071	0.094	0.575*	0.070
Co			1.000	0.006	0.040	0.092	0.024	0.266*	-0.022	0.022
Cr				1.000	0.567*	0.327*	0.208*	0.177*	0.238*	0.123
Cu					1.000	0.072	0.113	0.086	0.210*	0.060
Fe						1.000	-0.025	0.498*	0.125	0.101
Hg							1.000	0.043	0.083	0.009
Mn								1.000	0.139	0.024
Pb									1.000	0.090
Zn										1.000

*. Correlation is significant at the 0.05 level (2-tailed).

Table 8: Results of the Kruskal – Wallis analysis to determine significant differences in heavy metal concentrations in groundwater in regions with different topography

Heavy Metals	Kruskal-Wallis H	df	Asymp. Sig.
As	0.009	1	0.923
Cd	1.254	1	0.263
Co	1.465	1	0.226
Cr	0.016	1	0.898
Cu	2.980	1	0.084
Fe*	18.133	1	0.000
Hg	0.057	1	0.811
Mn	2.237	1	0.135
Pb	3.773	1	0.052
Zn*	3.939	1	0.047

*indicates significant value (≤ 0.05)

different soil types (Fig. S4). In low topography, the dominant type of soil is clay. According to Zhai *et al.* (2021), Fe is found to be high in clay soils and has a lower residence time in high topography because it can migrate to areas with low topography; meanwhile, Zn is suspected of migrating even though the concentrations in all samples do not exceed the quality standard (Table 3). The concentrations of Cr and Cu still meet the quality standards for drinking water and for sanitation and hygiene (Table 3). The concentrations of Cr and Cu significantly differ among several sub-districts ($p < 0.05$) and have a strong positive correlation, indicating that they originate from natural sources, such as the main material in soil and/or geological activity. Hence, the significant difference between Cr and Cu is thought to be influenced by the characteristics of each sub-district.

CONCLUSION

This study analyzed water quality through comparison of quality standards in regulations, determined chemicals of potential concerns, and statistically analyzed 10 heavy metals in groundwater used for sanitation and hygiene as well as water sources other than groundwater for consumption. The areas with the highest average concentrations of heavy metals in groundwater for hygiene and sanitation purposes were as follows: in Rancaekek (0.0015 ± 0.0025 mg/L) for Cd, Majalaya (1.7465 ± 3.1291 mg/L) for Fe, Baleendah (0.0135 ± 0.0571 mg/L) for Hg, Pacet (1.2254 ± 3.3744 mg/L) for Mn, and Rancaekek (0.0291 ± 0.0506 mg/L) for Pb. The concentrations of Fe in groundwater for SH at 23%, Mn at 18%, Pb at 9%, Hg at 6%, Cd at 6% of the study area were higher than the maximum allowable

limits. Moreover, 46%, 27%, 13%, 11%, 11%, 11%, and 3% of the consumption water samples contained As, Pb, Co, Fe, Hg, Mn, and Cd, and the values were higher than the maximum limit. In drinking water, samples that exceeded the maximum limits of As, Cd, Hg and Pb were dominant sourced from refill water, while Fe, Mn and Co were sourced from groundwater. Thus, As, Cd, Co, Fe, Hg, Mn, and Pb exceeded the quality standards for drinking water, while only As and Co did not exceed the quality standards for water hygiene and sanitation. Arsenic and cobalt quality standards are more stringent for drinking water compared with water sanitation and hygiene. Lead–cadmium and iron–manganese in groundwater showed a positive Spearman correlation ($p < 0.05$) and may have originated from the same source. About 100% of the drinking water samples had copper and zinc concentrations that did not exceed the quality standard. Iron and zinc in groundwater differed significantly based on differences in topography and soil types ($p < 0.05$). This study reveals that six out of 10 heavy metals are chemicals of potential concern and are sorted based on potential risks to health, that is, arsenic > mercury > lead > cobalt > manganese > cadmium. Ingestion is the main pathway for potential risk, and children are more likely to be at risk than adults. Therefore, evaluating the quality of community drinking water in Bandung Regency should be conducted in collaboration with stakeholders and decision makers, who must immediately take sustainable actions to protect public health; possible actions include determining areas that have a high level of vulnerability of groundwater to heavy metal pollution and are unfit for use as a source of drinking water, considering alternative sources of drinking

water, and/or the application of engineering to reduce health risks. Evaluation of water sources, technology, and maintenance processes and checking of water quality before and after technology application from Refill Drinking Water Depots should be conducted on a regular basis to ensure that raw and processing water meets the quality set out in regulations. These processes will accelerate the cessation of use in the event of water quality deterioration and trigger the necessary actions to prevent further deterioration and remedy. People need to be careful in choosing drinking water sources that are free from heavy metal pollution. Communities should be provided with information on how to determine the quality of water suitable for drinking.

AUTHORS CONTRIBUTION

N. Fahimah, the first author and corresponding author, has contributed with conceptualization, data curation, formal analysis, methodology, software, visualization, roles/writing - original draft, writing – review, and editing. I.R.S. Salami, has contributed with supervision, writing – review, editing, and funding acquisition. K. Oginawati has contributed with supervision, writing – review, and editing. S.J.Yapfrine and Y.N. Thaher have contributed with data curation. A. Supriatin has contributed with project administration and data curation.

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest regarding the publication of this work. In addition, the ethical issues including plagiarism, informed consent, misconduct, data fabrication and, or falsification, double publication and, or submission, and redundancy have been completely witnessed by the authors. The author's statement there are no human or animal respondents in this study.

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APPENDIX. SUPPLYMENTARY DATA

The following is the Supplementary data to this article can be found from the below link:

https://docs.google.com/document/d/1320cCjD-9myvdB0h36i4EgKGHCW4aR50M/edit?usp=share_link&oid=110524797063764488087&rtpof=true&s-d=true

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ABBREVIATIONS

%	Percent
ADHD	Attention deficit hyperactivity
$(RBC_d)_c$	RBC from dermal absorption route for cancer effects
$(RBC_d)_{NC}$	RBC from dermal absorption route for non cancer effects
$(RBC_o)_c$	RBC from oral route for cancer effects
$(RBC_o)_{NC}$	RBC from oral route for non cancer effects
As	Arsenic
ASD	Autism spectrum disorder
ASGM	Artisanal and small-scale gold mining

<i>AT</i>	Averaging Time	<i>HMs</i>	Heavy metals
<i>AT</i>	Averaging time	<i>ICP OES</i>	Inductively coupled plasma optical emission spectrometric
<i>BW</i>	Body weight	<i>IDW</i>	Inverse distance weighted
<i>C</i>	Cancer effect	<i>IFW</i>	Resident drinking water ingestion rate
<i>CAL EPA</i>	California Environmental Protection Agency	<i>IR</i>	Ingestion rate
<i>Cal/ OEHHA</i>	California Office of Environmental Health Hazard Assessment	<i>IRIS</i>	Integrated risk information system
<i>Cd</i>	Cadmium	<i>Kp</i>	Dermal permeability constant
<i>cm/h</i>	Centimeters per hour	<i>L/day</i>	Liters per day
<i>cm²</i>	Square centimeter	<i>L/kg</i>	Liters per kilogram
<i>CNS</i>	Central nervous system	<i>LT</i>	Lifetime
<i>Co</i>	Cobalt	<i>mg/L</i>	Milligram per liter
<i>COI</i>	Chemical of interest	<i>mg/L</i>	Milligrams per liter
<i>COPCs</i>	Chemical of potential concerns	<i>Mn</i>	Manganese
<i>Cr</i>	Chromium	<i>NC</i>	Non cancer effect
<i>CSF</i>	Cancer slope factor	<i>NCDs</i>	Noncommunicable diseases
<i>Cu</i>	Copper	<i>NH4+</i>	Ammonium
<i>DAMIU</i>	Refill drinking water depot	<i>p</i>	The weighting power that determines how the weight is reduced by increasing distance.
<i>Days/y</i>	Days per year	<i>Pb</i>	Lead
<i>DFW</i>	Resident water dermal contact factor	<i>PPRTV</i>	Provisional peer reviewed toxicity values
<i>df</i>	Degree of freedom	<i>RBC</i>	Risk based concentration
<i>d_i</i>	The distance between the interpolated value and the measured value zi	<i>Rfd</i>	Reference of dose
<i>DNA</i>	Deoxyribonucleic acid	<i>ROS</i>	Reactive oxygen species
<i>ED</i>	Exposure Duration - resident	<i>SA</i>	Resident surface area water - adult
<i>EF</i>	Exposure frequency	<i>SD</i>	The standard deviation
<i>ET</i>	Exposure time	<i>SH</i>	Sanitation and hygiene
<i>ET</i>	Resident water exposure time	<i>SL</i>	Screening level
<i>EV</i>	Resident events	<i>SNI</i>	Indonesian national standard
<i>Fe</i>	Iron	<i>THQ</i>	Target hazard quotient
<i>GIABS</i>	Fraction of contaminant absorbed in gastrointestinal tract	<i>TR</i>	Target risk
<i>GPS</i>	Global positioning system	<i>WWTPs</i>	Waste water treatment plants
<i>HEAST</i>	The EPA superfund program's health effects assessment summary table	<i>z</i>	The estimated value at the interpolation point
<i>Hg</i>	Mercury	<i>z_i</i>	The measured value at point
		<i>Zn</i>	Zinc

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