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The assessment of water quality and human health risk from pollution of chosen heavy metals in the Upstream Citarum River, Indonesia

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Abstract: This study explains water quality in terms of seven heavy metals in the Upstream Citarum River and analyses human health risk (non-carcinogenic risk) for adults and children. Water samples were collected from five sampling locations along the Upstream Citarum River, i.e. from Majalaya Sub-District to Dayeuhkolot Sub-District. The contents of heavy metals were analysed by the Atomic Absorption Spectrometer (AAS) variant 240 FS. The results of the analysis showed that the pollution index value, which was categorised as slightly polluted from the highest to the lowest value, was as follows: location 4 (4.220) > location 1 (3.764) > location 2 (3.219) > location 5 (2.967) > location 3 (2.800). Values of the hazard index (*HI*) for adults and children were as follows: Pb > Cr > Cd > Zn > Ni > Co > Cu. Pb and Cr have *HI* values greater than 1. This indicates that these metals can have a negative impact on public health. The *HI* in the ingestion pathway was greater than that of the dermal pathway, and the *HI* value for children was greater than that for adults. Further research is needed regarding the health risks from groundwater around the area which is used directly by the community because river water and groundwater systems are interconnected through streambeds.

Keywords: Citarum River, heavy metals, human health risk, water quality

INTRODUCTION

The Citarum River is the largest and the longest river in West Java, Indonesia. The river basin area is 6,614 km² and the river length 297 km. The Citarum River crosses 13 regencies/cities in West Java, from Mount Wayang to the mouth of Java Sea, which is in Muara Gembong, Bekasi. The Citarum River plays an important role as a source of raw water for drinking. It is also used in Java-Bali electricity generation and rice field irrigation [ROOSMINI *et al.* 2018]. The Citarum River supplies raw water for households, as well as drinking water for urban and industrial purposes in the Bandung Area. The use of surface water from the upstream Citarum River for domestic purposes is 125,500 m³·day⁻¹ and for industrial purposes 111,300 m³·day⁻¹ [ADB, World Bank 2013].

Due to its rapid industrial growth and poor waste management, the Citarum River was included in the list of the most polluted rivers in the world in 2013 [AAM *et al.* 2019]. Based on information from the Governor of West Java, in 2020, the Citarum River was still polluted with a light polluted category. The water quality improved from the moderately polluted category in 2019 to lightly polluted in 2020 due to the Citarum Harum Program. The program has been regulated through Presidential Regulation Number 15 of 2018 concerning the Acceleration of Pollution Control and Damage to the Citarum River Basin [Open Data Jabar 2020].

MARSELINA *et al.* [2021] stated that the Citarum watershed is dominated by the manufacturing industries, such as chemical, textile, leather, paper, pharmaceutical, metal, agriculture, as well as animal husbandry, food and beverage production, and others. The level of pollution in the Citarum River is very dangerous for people. SHARA [2021] said that there was a very high concentration of Cd in the River. Heavy metals are dangerous compounds because they cannot be degraded, and they can accumulate in the environment and the body of living creatures. The accumulation of heavy metals in the body has a negative impact on health [OGINAWATI et al. 2020], including non-carcinogenic and carcinogenic effects. Heavy metal pollution in the Citarum River will affect the river ecosystem and harm the health of people who use this river to meet their daily needs [SEPTIONO et al. 2015]. Heavy metals from the use of water can enter the human body in two ways, i.e. drinking contaminated water (ingestion pathway) and by dermal contact with water (dermal absorption pathway [BRIFFA et al. 2020]. Excessive levels of heavy metal cause significant damage to every organ and can present neurological defects, respiratory disorders, carcinogenicity, gastrointestinal obstruction, osteoporosis, etc. [MITRA et al. 2022].

Health risk assessment was a method commonly used to evaluate carcinogenic and non-carcinogenic health risks due to heavy metal pollution in the environment [GHADERPOORI *et al.* 2018]. Health risk assessment was a study to determine environmental conditions and characteristics that have the potential to pose a public health risk. The study was also the basis for environmental and health risk management. The evaluation of human health risks can be done by calculating the level of daily water intake and the hazard quotient of heavy metal contaminants [KAUR *et al.* 2020]. Currently, studies and publications related to heavy metal concentrations in the Upstream Citarum River area still mostly focus on heavy metal qualitative analysis without being equipped with a health risk assessment analysis. There have been health risk analyses of water in Indonesia but these have not been carried out in the Upstream Citarum River [Ashar 2007; INDIRAWATI 2018; PAHRUDDIN *et al.* 2017; TARIGAN 2015; WIBOWO 2013]. Therefore, the health risk assessment needs to be carried out in the Upstream Citarum River. Such a study was conducted to assess heavy metals concentration (Pb, Cd, Co, Cr, Cu, Ni, and Zn) and human health risk. This study was included with the spatial distribution of risk levels of heavy metals.

MATERIALS AND METHODS

SAMPLING COLLECTION

This study was carried out on April 20, 2018 at 08:00 a.m until 17:00 p.m. in sunny conditions (no rain). Water samples were collected from five sampling locations along the Upstream Citarum River, i.e. from Majalava Sub-District to Daveuhkolot Sub-District (Fig. 1). These locations were chosen for the industrial and agricultural activities around the Majalaya and Dayeuhkolot Sub-Districts that are the source of heavy metals in water. Hence, we can see the pattern of heavy metal concentrations in river water ranging from areas with small agricultural activity and a small number of industries to large agricultural areas and many industries. Water samples were collected without regard to seasonal variations. Water samples (500 cm³) were collected from the depth of approximately 50 cm below the water surface. Water samples were preserved by adding 8 drops of HNO_3 with pH < 2. Then, they were put into a cool box containing blue ice (4°C).

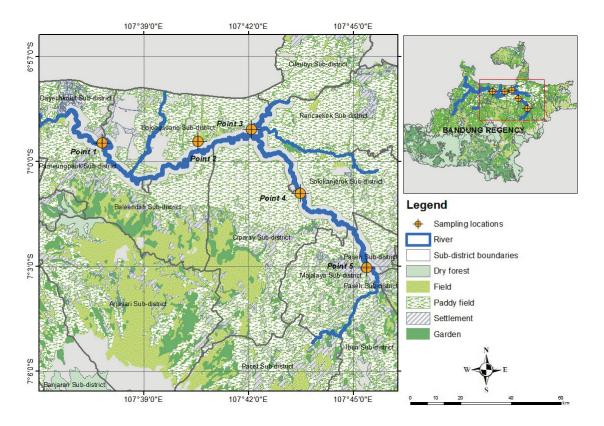


Fig. 1. Sampling locations along the Upstream Citarum River in the Bandung Regency; source: own elaboration

HEAVY METALS ANALYSIS

Water samples taken from the river were analysed for their heavy metal content, i.e. Cd, Cu, Co, Ni, Pb, Cr, and Zn. The analysis of heavy metals was carried out in the Integrated Laboratory, Agricultural Environment Research Institute, Pati, Central Java which had been accredited by SNI ISO/IEC 17025: 2008 by *National Accreditation Committee* (KAN). Water samples were analysed using the Atomic Absorption Spectrometer (AAS) variant 240 FS. Before being injected into the AAS, water samples were digested. The digestion process was carried out to test the dissolved heavy metals after water samples were filtered.

The digestion process started by inserting 100 cm³ of homogenised water samples into a 250 cm³ Erlenmeyer flask, then adding 5 cm³ of HNO₃. The samples were heated using a hot plate until almost dry (remaining volume 15–20 cm³). The samples were put into a 25 cm³ volumetric flask and aquadest added until it reached 25 cm³ size limit. The samples contained in the volumetric flask (sample + aquadest) were homogenised. Furthermore, the samples were filtered using filter paper (SNI 6989.16:2009 for Cd; 6989.6:2009 for Cu; SNI 6989.68:2009 for Co; SNI 6989.18:2009 for Ni; SNI 6989.8:2009 for Pb 6989; SNI 6989.17:2009 for Cr; SNI 6989.7:2009 for Zn). After being filtered, the samples were injected into the AAS variant 240 FS.

WATER QUALITY ASSESSMENT

Water quality assessment was carried out using the Pollution Index Method in accordance with Regulation of Environmental Minister Indonesia No. 115 of 2003. The pollution index (*PI*) method was used to determine the level of water quality pollution based on permitted quality standards with Equation (1) [FAHIMAH *et al.* 2020].

$$PI_{j} = \sqrt{\frac{\left(\frac{C_{i}}{L_{ij}}\right)_{M}^{2} + \left(\frac{C_{i}}{L_{ij}}\right)_{R}^{2}}{2}} \tag{1}$$

where: PI = pollution index for designation *j*, C_i = parameter water quality concentration *i*, L_i = water quality parameter concentration *i* listed in the designation standard of quality *j*, while M = maximum, and R = average. Index class of *PI* consisted of four classes, with a score of $0 \le PI < 1.0$ indicates good water quality (comply with quality standard); $1.0 \le PI < 5.0$ indicates slightly polluted; $5.0 \le PI < 10.0$ indicates moderately polluted, and $PI \ge 10.0$ indicates heavily polluted.

HUMAN HEALTH RISK ASSESSMENT

The US EPA human health risk assessment method [US EPA 2004] was used to quantify the level of public health risk in the Upstream Citarum River. Two pathways assessed for human health risks included the ingestion pathway and dermal absorption pathway, and two different age groups included adults and children [SHIL, SINGH 2019]. The level of non-carcinogenic human health risk (also known as hazard quotient – HQ) is quantified by dividing the value of chronic daily intake (*CDI*) with chronic reference dose (*RfD*) [AHMED *et al.* 2021; JOLODAR *et al.* 2021; SHIL, SINGH 2019]. Furthermore, the hazard index (*HI*) is the sum of *HQ* values for various heavy metals (i.e. Pb, Cd, Co, Cr, Cu, Ni,

and Zn) and various exposure pathways (i.e. ingestion pathway and dermal absorption pathway) [ZAKIR *et al.* 2020]. The values of *CDI* for both adults and children were predicted by the equation [AHMED *et al.* 2021; JOLODAR *et al.* 2021; SHIL, SINGH 2019; USEPA 2004] as follows:

$$CDI_{\text{ingestion}} = \frac{C_i \cdot EF \cdot ED \cdot IRW}{BW \cdot AT}$$
 (2)

$$CDI_{\text{dermal}} = \frac{C_i \cdot SA \cdot KC \cdot EF \cdot ET \cdot ED \cdot ABS}{BW \cdot AT}$$
(3)

where: C_i = concentration of heavy metals in water (mg·dm⁻³), IRW = ingestion rate (2.5 dm³·day⁻¹ for adults and 0.64 dm³·day⁻¹ for children) [SHIL, SINGH 2019]; BW = body weight (60 kg for adult and 15 kg for children) [US EPA 2002], EF = exposure frequency (365 days·y⁻¹) [US EPA 2020], ED = exposure duration (70 years for adults and 30 years for children [US EPA 2020], AT = average time for non-carcinogenic substance (*EF*·*ED*, in days) [US EPA 2020], SA = skin surface area for water contact (5700 cm² for adults and 2800 cm² for children) [US EPA 2020], KC = dermal permeability factor (0.001 cm·h⁻¹) [AHMED *et al.* 2021]; ET = exposure time (0.58 h·day⁻¹ for adults and 1 h·day⁻¹ for children) [SHIL, SINGH 2019], and *ABS* = fraction of dermal absorption (0.001 for all types of heavy metals except arsenic) [US EPA 2020].

Hazard quotient (*HQ*) helps to assess the health risk level (non-carcinogenic) from heavy metals in river water. In the current study, it is used to determine the non-carcinogenic health risk from heavy metals in the river water by Equation (4) [JOLODAR *et al.* 2021; KAUR *et al.* 2020; OGINAWATI *et al.* 2021]:

$$HQ = \frac{CDI_{\text{ingestion or dermal}}}{RfD} \tag{4}$$

where: RfD = reference dose (mg·kg⁻¹·day⁻¹).

If the analysed HQ < 1, a community using water or having direct contact with water will not face a potential health risk. In addition, the community is potentially at non-carcinogenic risk, if $HQ \ge 1$, with the potential level increasing with increasing value [SAHA *et al.* 2016].

In addition, the cumulative hazard index (*HI*) was measured by adding up HQ for all types of heavy metals using Equation (5) [MOHSENIBANDPI *et al.* 2018].

$$HI = \sum_{i}^{n} HQ_i \tag{5}$$

The total hazard index (HI_{total}) is estimated by summing more than one HI from several pathways, i.e. ingestion pathway and dermal contact (dermal absorption), which are calculated by Equation (6) [MOHSENIBANDPI *et al.* 2018; ODUKOYA *et al.* 2017].

$$HI_{\text{total}} = HI_{\text{ingestion}} + HI_{\text{dermal}}$$
 (6)

 $HI \ge 1$ indicates the potential for adverse non-carcinogenic effects. Therefore, environmental management actions are needed to minimise the level of risks.

SPATIAL DISTRIBUTION PATTERNS AND DATA ANALYSIS

Spatial distribution patterns of heavy metals hazard index were described using ArcGIS 10.2 developed by ESRI. Recently, ordinary kriging and inverse distance weight (IDW) have been commonly used interpolation methods to describe the spatial distribution of heavy metals [MENG *et al.* 2019]. Interpolation kriging was drawn for the non-carcinogenic *HI* distribution pattern for adults and children. Kriging interpolation has been widely used to determine the accurate spatial distribution of heavy metals and the level of health risks in humans. The kriging method is an accurate method for describing heavy metal contamination [WU *et al.* 2021]. The spatial distribution pattern of heavy metals *HI* was obtained by integrating five sampling points in the GIS kriging geostatistical procedure. For data analysis, the raw data were processed using Microsoft Excel to get the resulting pollution index and risk level.

RESULTS AND DISCUSSION

WATER QUALITY IN TERMS OF HEAVY METALS

Data regarding the concentration of seven types of heavy metals in river water and their comparison with quality standards are shown in Figure 2. Pb, Cd, and Zn exceeded the quality standards at all sampling points, while Co and Ni did not exceed the quality standards at all sampling points. The highest concentrations of Cr and Cu exceeding the quality standard are at point 4.

The data are analysed to determine the pollution index. The pollution index for Pb, Cd, Co, Cr, Cu, Ni, and Zn in five sampling points is shown in Table 1. As presented in the table, the mean pollution index for seven heavy metals in five sampling points was 3.394, which indicated that the Upstream Citarum River around the Majalaya and Dayeuhkolot Sub-District was slightly polluted. The pollution index value from the highest,

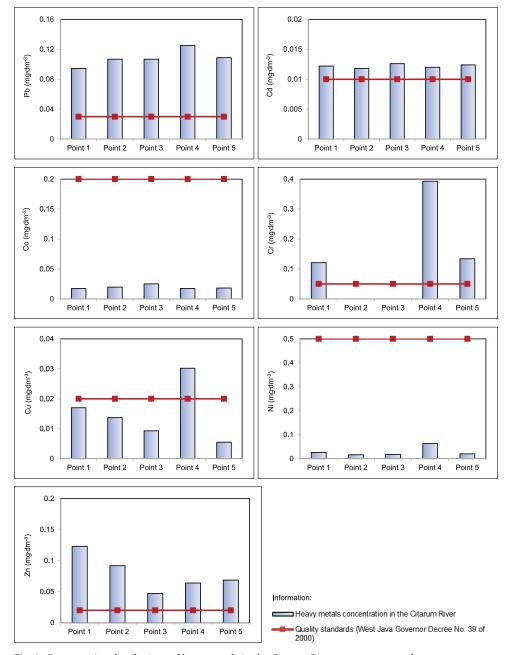


Fig. 2. Concentration distributions of heavy metals in the Citarum River; source: own study

Table 1.	Water	quality	assessment
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Sampling point	Pollution index of chosen seven heavy metals	Category
1	3.764	slightly polluted
2	3.219	slightly polluted
3	2.800	slightly polluted
4	4.220	slightly polluted
5	2.967	slightly polluted
Pollution index mean	3.394	slightly polluted

Source: own study.

which indicates moderate pollution, to the lowest value, which indicates compliance with the quality standard, was sorted as follows: location 4 (4.220) > location 1 (3.764) > location 2 (3.219) > location 5 (2.967) > location 3 (2.800).

The variation in the level of pollution index, which indicates the occurrence of pollution, was caused by various factors, one of which was the management factor in the vicinity of the study site. Furthermore, the industries in the vicinity of the study location are suspected of contributing to the high heavy metal pressure in the Upstream Citarum River. The highest concentration of heavy metals was found in the college tanning industry and the textile industry [DAs *et al.* 2011]. The industry uses more heavy metal chemicals during the production process and this may affect human health [TADESSE, GUYA 2017].

Therefore, some heavy metal concentrations that significantly exceeded the quality standard in the Upstream Citarum River need attention before accumulating in the sediment. They are very difficult to degrade and have a negative impact on aquatic animals and human health through the food chain or by direct contact with environmental media polluted by heavy metals.

HUMAN HEALTH RISK ASSESSMENT

Exposure factors and reference values used to predict intake levels and levels of health risk from heavy metal exposure are listed in Table 2. The reference dose (RfD) of chosen heavy metals for ingestion and dermal pathways are shown in Table 3.

The calculation of the $CDI_{ingestion}$ and CDI_{dermal} for noncarcinogenic risk using the reference value, i.e. ingestion rate, exposure frequency, exposure duration, body weight, average

Table 2. Exposure factors and reference values for health risk assessment

Factor's name	Symbol	Adult	Children	Unit	Reference	
Ingestion rate	IRW	2.50	0.64	$dm^3 \cdot d^{-1}$	SHIL and SINGH [2019]	
Exposure frequency	EF	365	365	$d \cdot y^{-1}$	US EPA [2020]	
Exposure duration	ED	70	30	у	US EPA [2020]	
Body weight	BW	60	15	kg	US EPA [2002]	
Average time	AT	25,550	10,950	h	US EPA [2020]	
Skin surface area for water contact	SA	5,700	2,800	cm ²	US EPA [2020]	
Dermal permeability factor	KC	0.001	0.001	$cm \cdot h^{-1}$	Анмед <i>et al.</i> [2021]	
Exposure time	ET	0.58	1	$h \cdot d^{-1}$	SHIL and SINGH [2019]	
Fraction of dermal absorption ¹⁾	ABS	0.001	0.001	-	US EPA [2020]	

¹⁾ For all types of heavy metals except arsenic (As). Source: own study.

Apart from agricultural land and settlements around the study location, there were also industries that could be a source of heavy metal pollution in the Upstream Citarum River. In inhabited areas, heavy metals can be released from shampoos used by local communities. This is supported by ISLAM et al. [2019], who stated that Fe, Co, Ni, Cu, Zn, Cd, Cr, Al, Mg, Pb, Mn, Hg, As, and Ag are found in shampoos, with high concentrations of heavy metals in synthetic shampoos compared to herbal shampoos. Heavy metals were also contained in household detergents, soaps, and environmental cleaning and health products because they were discharged directly into the environment without processing [IWEGBUE et al. 2019]. In agricultural areas, heavy metals can originate from fertilizers and pesticides used by farmers [GIMENO-GARCÍA et al. 1996]. There was a significant association of heavy metal concentration in the soil and fertilizer application in agriculture [ATAFAR et al. 2009].

Table 3. Reference doses (*RfD*) of chosen heavy metals for major pathways

Heavy metal	<i>RfD</i> _{ingestion}	<i>RfD</i> _{dermal}
Pb ¹⁾	0.0014	0.000524
Cd ¹⁾	0.001	0.000025
Co ²⁾	0.02	0.0576
Cr ¹⁾	0.003	0.003
Cu ¹⁾	0.04	0.012
Ni ¹⁾	0.02	0.0054
Zn ¹⁾	0.02	0.06

Source: own elaboration based on the literature: ¹⁾ Adimalla [2019], ²⁾ LANIYAN and ADEWUMI [2019].

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time, skin surface are for water contact, dermal permeability factor, exposure time and the fraction of dermal absorption presented in Table 2. The *HQ* value can be found by dividing the *HQ* value by the $RfD_{\text{ingestion}}$ or RfD_{dermal} shown in Table 3.

CDI values, *HQ* values, and *HI* values from the ingestion pathway and dermal absorption pathway in children and adults were used to estimate the level of non-carcinogenic risk. The results of the *CDI* and *HI* for adults and children on the ingestion pathway are presented in Table 4 and the dermal absorption pathway are presented in Table 5.

According to Tables 4 and 5, the mean HQ values for adults and children were sorted as follows: Pb > Cr > Cd > Zn > Ni > Co > Cu. The highest value of $HQ_{\text{ingestion}}$ was detected in Pb, which contributed 3.2262 for adults and 3.306 for children. The highest values of HQ_{dermal} were also detected in Pb, which contributed 0.0043 for adults and 0.0145 for children. The HQ value of the

Table 4. Chronic daily intake (CDI) and hazard index (HI) for adults and children ingestion pathway

T di	$CDI_{\text{ingestion}} (\text{mg·kg}^{-1} \cdot \text{d}^{-1})$							111)		
Location	Pb	Cd	Со	Cr	Cu	Ni	Zn	HI ¹⁾		
	Adult									
1	0.0039	0.0005	0.0007	0.0050	0.0007	0.0011	0.0051	5.37		
2	0.0045	0.0005	0.0008	0.0000	0.0006	0.0007	0.0038	3.96		
3	0.0045	0.0005	0.0010	0.0000	0.0004	0.0007	0.0020	3.91		
4	0.0052	0.0005	0.0007	0.0164	0.0013	0.0026	0.0027	10.01		
5	0.0045	0.0005	0.0008	0.0056	0.0002	0.0008	0.0029	5.84		
Mean HQ (CDI/RfD)	3.2262	0.5083	0.0408	1.8025	0.0158	0.0598	0.1646	-		
			(Children						
1	0.0040	0.0005	0.0007	0.0052	0.0007	0.0011	0.0053	5.49		
2	0.0046	0.0005	0.0008	0.0000	0.0006	0.0007	0.0039	4.05		
3	0.0046	0.0005	0.0011	0.0000	0.0004	0.0008	0.0020	4.00		
4	0.0053	0.0005	0.0007	0.0168	0.0013	0.0027	0.0027	10.25		
5	0.0046	0.0005	0.0008	0.0057	0.0002	0.0009	0.0029	5.98		
Mean HQ (CDI/RfD)	3.306	0.5205	0.0417	1.8458	0.0161	0.0612	0.1685	-		

¹⁾ $HI = HQ_{Pb} + HQ_{Cd} + HQ_{Co} + HQ_{Cr} + HQ_{Cu} + HQ_{Ni} + HQ_{Zn}$. Explanation: HQ = hazard quotient. Source: own study.

Table 5. Chronic daily intake (CDI) and hazard index (HI) of adult and children for dermal absorption pathway

	$CDI_{dermal} (mg \cdot kg^{-1} \cdot d^{-1})$								
Location	Pb	Cd	Со	Cr	Cu	Ni	Zn	$HI^{1)}$	
	Adult								
1	0.000005	0.000001	0.000001	0.000007	0.000001	0.000001	0.000007	0.007	
2	0.000006	0.000001	0.000001	0.000000	0.000001	0.000001	0.000005	0.005	
3	0.000006	0.000001	0.000001	0.000000	0.000001	0.000001	0.000003	0.005	
4	0.000007	0.000001	0.000001	0.000022	0.000002	0.000003	0.000004	0.023	
5	0.000006	0.000001	0.000001	0.000007	0.000000	0.000001	0.000004	0.008	
Mean HQ (CDI/Rfd)	0.0043	0.0007	0.0001	0.0024	0.0000	0.0001	0.0002	_	
			(Children					
1	0.000018	0.000002	0.000003	0.000023	0.000003	0.000005	0.000023	0.024	
2	0.000020	0.000002	0.000004	0.000000	0.000003	0.000003	0.000017	0.018	
3	0.000020	0.000002	0.000005	0.000000	0.000002	0.000003	0.000009	0.018	
4	0.000023	0.000002	0.000003	0.000073	0.000006	0.000012	0.000012	0.045	
5	0.000020	0.000002	0.000003	0.000025	0.000001	0.000004	0.000013	0.026	
Mean HQ (CDI/Rfd)	0.0145	0.0023	0.0002	0.0081	0.0001	0.0003	0.0007	-	

¹⁾ *HI* as in Tab. 4.

Explanation as in Tab. 4. Source: own study.

ingestion pathway was higher than that of the dermal absorption pathway, and the HQ value of the ingestion pathway was greater than 1 which indicates a health risk unacceptable to the dominant community from the ingestion pathway than from the dermal absorption pathway. The contribution of HQ from the ingestion pathway to total HI was the highest in the exposure pathway and accounted for more than 99% of the total risk. Similar results were illustrated in the Bangladesh Gomti River where the HQ value in the ingestion pathway was higher than the dermal absorption pathway, but the HQ value in the river was still categorised as safe (HQ < 1) even though there were differences in values between the two-exposure pathway [AHMED et al. 2021; CHONOKHUU et al. 2019]. The risk of ingestion pathway is higher than that of the dermal absorption of heavy metals because water is drunk directly so that in a short time heavy metals contained in water can enter the human body, whereas if heavy metals enter the body through skin absorption, it takes time for heavy metals to enter the body. In addition, in the ingestion pathway, heavy metals that enter the body can be in the form of dissolved and particulate heavy metals, while in the absorption pathway, heavy metals that enter the body may only be dissolved heavy metals.

However, different results were presented by SHIL and SINGH [2019], namely the value of HI_{adult} was higher than that of $HI_{children}$, while this study showed that the HI for children was higher than that for adults. Hazard index (HI) measured for different age groups is shown in Figure 3. Suppose the value of HI ($HI_{ingestion} + HI_{dermal}$) is compared in two different age groups, namely adults and children. In that case, as presented in Figure 3, the HI value for children is greater than that for adults. Higher HI scores in children can make them more vulnerable [ANYANWU, NWACHUKWU 2020]. Children tend to be at higher risk than adults, as their relatively lower body weight implies that the impact of water contaminated with heavy metals can be relatively higher.

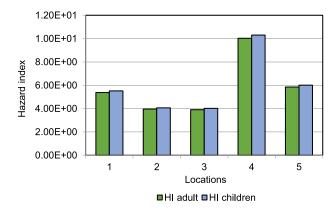


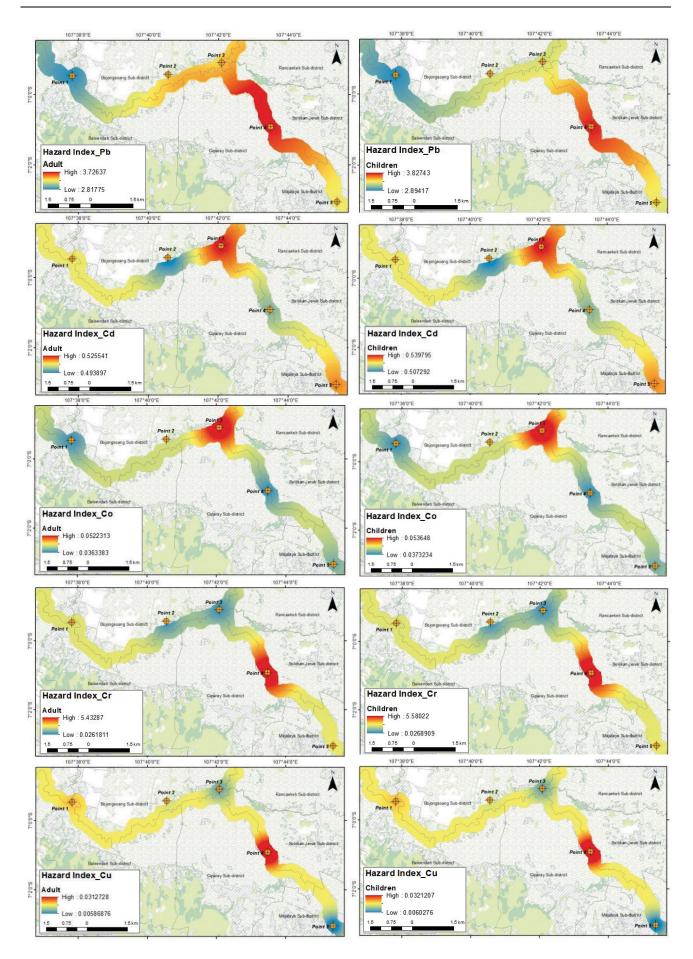
Fig. 3. Measured hazard index (HI) for different age grouped people; source: own study

For more details, the spatial distribution patterns of heavy metals *HI* regarding Pb, Cd, Co, Cr, Cu, Ni, and Zn for adults and children are shown in Figure 4. The measured *HI* of Pb ranged from 2.818 to 3.726 for adults and from 2.894 to 3.827 for children. The maximum *HI* of Pb was observed in sampling point 4 (around Ciparay and Solokanjeruk Sub-District). The measured *HI* of Cd ranged from 0.494 to 0.526 for adults and from 0.507 to 0.540 for children. The maximum *HI* of Cd was observed in sampling point 3 (around Bojongsoang and Rancaekek Sub-District). The measured *HI* of Co ranged from 0.494 to 0.526 for adults and from 0.507 to 0.540 for children.

0.036 to 0.052 for adults and from 0.037 to 0.054 for children. The maximum HI of Co was observed in sampling point 3 (around Bojongsoang and Rancaekek Sub-Districts). The measured HI of Cr ranged from 0.026 to 5.433 for adults and from 0.027 to 5.580 for children. The maximum HI of Cr was observed in sampling point 4 (around Ciparay and Solokanjeruk Sub-District). The measured HI of Cu ranged from 0.006 to 0.031 for adults and from 0.006 to 0.032 for children. The maximum HI of Cu was observed in sampling point 4 (around Ciparay and Solokanjeruk Sub-District). The measured HI of Zn ranged from 0.109 to 0.251 for adults and from 0.112 to 0.258 for children. The maximum HI of Zn was observed in sampling point 1 (around Bojongsoang and Baleendah Sub-District). These results indicate that HI values of Pb and Cr are more than 1 (HI > 1), which means that the presence of heavy metals Pb and Cr, which were categorised as slightly polluted, can pose a risk to the health of people living in the vicinity of the study area, especially the area that has the highest level of risk was at sampling point 4.

The high concentration of Pb and Cr around Ciparay and Solokan Jeruk (sampling point 4) may be due to the influence of the textile industry waste, where the Upper Citarum area is an area that has a high density of companies, mainly the textile industry. Industries that do not comply with wastewater quality standards can contribute to high levels of Pb and Cr pollutants at sampling point 4. Heavy metals, such as Pb and Cr, are found in textile industry effluents with concentrations exceeding the quality standard [TOLUIZTE et al. 2020]. In addition, based on data from the Bandung Regency Statistics Center (2019), the number of textile industries in the Ciparay Sub-district is 83 and in Solokan Jeruk is 22, while in the regions above, namely Baleendah and Rancaekek Sub-districts, the number of textile industries is higher, 83 and 76 respectively. The high number of industries in the previous area can also affect the high concentration level at sampling point 4 because heavy metals are cumulative and heavy metals can flow with the river from upstream to downstream and undergo the process of deposition in other areas. With a weak flow rate and large particle sizes, the deposition process will take place quickly and with a close distance from the source of contamination. However, when the flow velocity is high, the deposition process will take longer and further away from the source [FAHIMAH et al. 2020]. Furthermore, apart from industry, heavy metals can also be sourced from agricultural practices [COSTA, LIA 2022] as well as mixed sources of irrigation and transportation activities [WANG et al. 2021].

Water use has been identified as the main pathway associated with heavy metal exposure to humans [NAG, CUMMINS 2022] and has a negative impact on health. Pb can affect the nervous system, kidneys, and blood circulation, especially in children, infants, and fetuses [Guo et al. 2018]. Pb can also affect brain and intellectual development in children, induce apoptosis in organ tissues [MANI et al. 2019] and in some cases, irreversible neurological damage [NAG, CUMMINS 2022]. Moreover, Cr(VI) damages cells in several ways, such as increased oxidative stress, DNA adduct formation, and chromosomal breakdown. Cr(VI) also causes toxicity in various ways, namely it reduces the activity or efficiency of the immune system, competing with cofactor fixation sites for enzyme activity, suppressing important enzymes, such as oxidative phosphorylation, and causing changes in cell architecture, especially in the lipoprotein region of the membrane [SHARMA et al. 2022].



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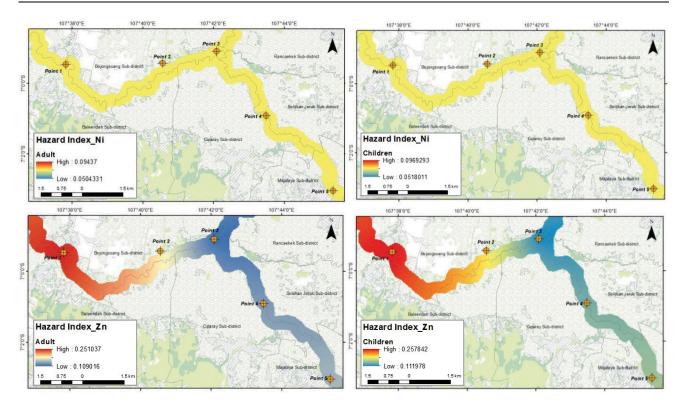


Fig. 4. Spatial distribution patterns of heavy metal hazard index (HI) for adults and children; source: own study

CONCLUSIONS

This research was conducted to determine the water quality in terms of heavy metals and hazard index (HI) in Upstream Citarum River around the Majalaya and Dayeuhkolot Sub-Districts. The pollution index value, which was categorised as slightly polluted from the highest to the lowest value, was sorted as follows: location 4 (4.220) > location 1 (3.764) > location 2 (3.219) > location 5 (2.967) > location 3 (2.800). The HI values for adults and children were sorted as follows: Pb > Cr > Cd > Zn > Ni > Co > Cu. Pb and Cr metals have *HI* values greater than 1, which indicates that these metals have the potential to have a negative effect on public health. The value of HI in the ingestion pathway is larger than that of the dermal pathway, and the HI value for children is larger than that for adults. The findings of this study can provide insight to stakeholders and the public about water quality and the level of risk to health when water is used for daily life. Moreover, it can support the government in making regulatory policies and priority programs for the prevention of metal contamination in water systems. Further research is needed regarding the health risks from groundwater around the area which is used directly by the community because river water and groundwater systems are interconnected through streambeds.

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REFERENCES

- AAM M.R., MUSNANSYAH A., WITARSYAH D. 2019. Penerapan manajemen interoperabilitas open data Citarum menggunakan opendatasoft [Application of Citarum open data interoperability management using opendatasoft]. eProceedings of Engineering. Vol. 6(2) p. 8051–8065.
- ADB, The World Bank 2013. Downstream impacts of water pollution in the Upper Citarum River, West Java, Indonesia: Economic assessment of interventions to improve water quality. Water and sanitation program: Technical paper. Asian Development Bank and The International Bank for Reconstruction and Development / The World Bank pp. 32.
- ADIMALLA N. 2019. Heavy metals contamination in urban surface soils of Medak Province, India, and its risk assessment and spatial distribution. Environmental Geochemistry and Health. Vol. 42 p. 59–75. DOI 10.1007/s10653-019-00270-1.
- AHMED A.S.S., HOSSAIN M.B., BABU S.M.O.F., RAHMAN M.M., SARKER M. S.I. 2021. Human health risk assessment of heavy metals in water from the subtropical river, Gomti, Bangladesh. Environmental Nanotechnology, Monitoring & Management. Vol. 15, 100416. DOI 10.1016/j.enmm.2020.100416.

- ANYANWU E.D., NWACHUKWU E.D. 2020. Heavy metal content and health risk assessment of a South-eastern Nigeria River. Applied Water Science. Vol. 10, 210. DOI 10.1007/s13201-020-01296-y.
- ASHAR T. 2007. Analisis risiko asupan oral pajanan mangan dalam air terhadap kesehatan masyarakat [Risk analysis of oral intake of manganese exposure in water on public health]. Kesmas National Public Health Journal. Vol. 2(3). DOI 10.21109/kesmas.v2i3.265.
- ATAFAR Z., MESDAGHINIA A., NOURI J., HOMAEE M., YUNESIAN M., AHMADIMOGHADDAM M., MAHVI A. 2009. Effect of fertilizer application on soil heavy metal concentration. Environmental monitoring and assessment. Vol. 160 p. 83–89. DOI 10.1007/ s10661-008-0659-x.
- BRIFFA J., SINAGRA E., BLUNDELL R. 2020. Heavy metal pollution in the environment and their toxicological effects on humans. Heliyon. Vol. 6(9). DOI 10.1016/j.heliyon.2020.e04691.
- CHONOKHUU S., BATBOLD C., CHULUUNPUREV B., BATTSENGSEL E., DORJSUREN B., BYAMBAA B. 2019. Contamination and health risk assessment of heavy metals in the soil of major cities in Mongolia. International Journal of Environmental Research and Public Health. Vol. 16(14), 2552. DOI 10.3390/ijerph16142552.
- COSTA C., LIA F. 2022. Temporal variations of heavy metal sources in agricultural soils in Malta. Applied Sciences. Vol. 12(6), 3120. DOI 10.3390/app12063120.
- DAS M., AHMED MD.K., ISLAM S., ISLAM MD M., AKTER M.S. 2011. Heavy metals in industrial effluents (tannery and textile) and adjacent rivers of Dhaka City, Bangladesh. Terrestrial and Aquatic Environmental Toxicology. Vol. 5 p. 8–13.
- FAHIMAH N., OGINAWATI K., SUHARYANTO 2020. Fate and spatial distribution of Pb, Cd, Cu, and Zn in the water column and in the surface sediment of Indonesian estuary (Citarum River Estuary). E3S Web of Conferences. Vol. 148(1), 07007. DOI 10.1051/e3sconf/202014807007.
- GHADERPOORI M., MIRZAEI N., SHARAFI K., REZAEI S., BAGHERI A., AHMADPOUR M., FARHANG M. 2018. Comparison of bottled waters current brands in term of important chemical parameters (nitrate, fluoride, chloride, sulfate) effecting health. International Journal of Pharmaceutical Research. Vol. 10(3) p. 328–333. DOI 10.31838/ijpr/2018.10.03.018
- GIMENO-GARCÍA E., ANDREU V., BOLUDA R. 1996. Heavy metals incidence in the application of inorganic fertilizers and pesticides to rice farming soils. Environmental Pollution. Vol. 92(1) p. 19–25. DOI 10.1016/0269-7491(95)00090-9.
- GUO G., LEI, M., WANG Y., SONG B., YANG J. 2018. Accumulation of As, Cd, and Pb in sixteen wheat cultivars grown in contaminated soils and associated health risk assessment. International Journal Environmental Research and Public Health. Vol. 15(11), 2601. DOI 10.3390/ijerph15112601.
- INDIRAWATI S.M. 2018. Model pengelolaan risiko dampak pencemaran logam berat (Pb dan Cd) berbasis studi ARKL pada masyarakat terpapar di kawasan pesisir belawan [The model for risk management for the impact of heavy metal pollution (Pb and Cd) based on the ARKL study on exposed communities in the Belawan Coastal Area]. Kota Medan. Universitas Sumatera Utara.
- ISLAM F., MORSHED A.J.M., RAHMAN MD, PARUL A., ISLAM MD, MAHMUD M., SAYEED A., MEHERUNNESA M., LEE Y.H. 2015. Determination of heavy metals and trace elements in worldwide branded shampoo available in local market of Bangladesh by Atomic Absorption Spectrometry. Asian Journal of Chemistry. Vol. 27 p. 3756–3762. DOI 10.14233/ajchem.2015.18967.
- IWEGBUE C.M.A., EMAKUNU O.S., LARI B., EGOBUEZE F.E., TESI G.O., NWAJEI G.E., MARTINCIGH B.S. 2019. Risk of human exposure to metals in some household hygienic products in Nigeria.

Toxicology Reports. Vol. 6 p. 914–923. DOI 10.1016/j.toxrep. 2019.08.014.

- JOLODAR N.R., KARIMI S., BOUTEH E., BALIST J., PROSSER R. 2021. Human health and ecological risk assessment of pesticides from rice production in the Babol Roud River in Northern Iran. Science of the Total Environment. Vol. 772, 144729. DOI 10.1016/j. scitotenv.2020.144729.
- KAUR L., RISHI M.S., DAN SIDDIQUI A.U. 2020. Deterministic and probabilistic health risk assessment techniques to evaluate noncarcinogenic human health risk (Nhhr) due to fluoride and nitrate in groundwater of Panipat, Haryana. India. Environmental Pollution. Vol. 259, 113711. DOI 10.1016/j.envpol. 2019.113711.
- LANIYAN T.A., ADEWUMI A.J. 2019. Health risk assessment of heavy metal pollution in groundwater around and exposed dumpsite in Southwestern Nigeria. Journal of Health & Pollution. Vol. 9(24), 191210. DOI 10.5696/2156-9614-9.24.191210.
- MANI M.S., KABEKKODU S.P., JOSHI M.B., DSOUZA H.S. 2019. Ecogenetics of lead toxicity and its influence on risk assessment. Human and Experimental Toxicology. Vol. 38 p. 1031–1059. DOI 10.1177/ 0960327119851253.
- MARSELINA M., SABAR A., FAHIMAH N. 2021. Spatial and temporal assessment of surface water quality using water quality index The Saguling Reservoir, Indonesia. Journal of Water and Land Development. No. 49 p. 111–120. DOI 10.24425/jwld.2021. 137103
- MENG Y., CAVE M., ZHANG C. 2019. Comparison of methods for addressing the point-to-area data transformation to make data suitable for environmental, health and socio-economic studies. The Science of the Total Environment. Vol. 689 p. 797–807. DOI 10.1016/j.scitotenv.2019.06.452.
- MITRA S., CHAKRABORTY A.J., TAREQ A.M., EMRAN T.B., NAINU F., KHUSRO A., ..., SIMALGANDARA J. 2022. Impact of heavy metals on the environment and human health: Novel therapeutic insights to counter the toxicity. Journal of King Saud University – Science. Vol. 34(3), 101865. DOI 10.1016/j.jksus.2022.101865.
- MOHSENIBANDPI A., ESLAMI A., GHADERPOORI M., SHAHSAVANI A., JEIHOONI A.K., GHADERPOURY A., ALINEJAD A. 2018. Health risk assessment of heavy metals on PM_{2.5} in Tehran air, Iran. Data in Brief. Vol. 17 p. 347–355. DOI 10.1016/j.dib.2018.01.018.
- NAG R., CUMMINS E. 2022. Human health risk assessment of lead (Pb) through the environmental-food pathway. Science of the Total Environment. Vol. 810, 151168. DOI 10.1016/j.scitotenv. 2021.151168.
- ODUKOYA A.M., OLOBANIYI S.B., OLUSEYI T.O., ADEYEYE U.A. 2017. Health risk associated with some toxic elements in surface water of Ilesha gold mine sites, southwest Nigeria. Environmental Nanotechnology Monitoring and Management. Vol. 8 p. 290– 296. DOI 10.1016/j.enmm.2017.10.005.
- OGINAWATI K., SIDHI R., SUSETYO S. H. 2020. Lead exposure in trader communities in industrial area of the battery recycling plant: Tangerang, Indonesia. Journal of Ecological Engineering. Vol. 21 (4) p. 264–270. DOI 10.12911/22998993/118297.
- OGINAWATI K., SUSETYO S. H., ROSALYN F.A., KURNIAWAN S.B., ABDULLAH S.R.S. 2021. Risk analysis of inhaled hexavalent chromium (Cr⁶⁺) exposure on blacksmiths from industrial area. Environmental Science and Pollution Research. Vol. 28(11) p. 14000–14008. DOI 10.1007/s11356-020-11590-6.
- Open Data Jabar 2020. Sungai Citarum [online]. [Access 19.05.2022]. Available at: https://opendata.jabarprov.go.id/id/infografik/sungai-citarum
- PAHRUDDIN M. 2017. Risiko pajanan logam berat pada air sungai [Exposure risk of heavy metals on river water]. Jurnal Kesehatan

Lingkungan. Vol. 14. No. 2 p. 525–532. DOI 10.31964/jkl. v14i2.47.

- ROOSMINI D., SEPTIONO M.A., PUTRI N.E., SHABRINA H.M., SALAMI I.R.S., ARIESYADY H.D. 2018. River water pollution conditions in upper part of Brantas River and Bengawan Solo River. In: IOP Conference Series: Earth and Environmental Science. Vol. 106 (1), 012059. DOI 10.1088/1755-1315/106/1/012059.
- SAHA N., RAHMAN M.S., JOLLY Y.N., RAHMAN A., SATTAR M.A., HAI M.A. 2016. Spatial distribution and contamination assessment of six heavy metals in soils and their transfer into mature tobacco plants in Kushtia District, Bangladesh. Environmental Science and Pollution Research. Vol. 23(4) p. 3414–3426. DOI 10.1007/ s11356-015-5575-3.
- SEPTIONO M. A., ROOSMINI D. 2015. Heavy metal distribution in water, sediment, and fish at upper Citarum River and its potential exposure pathway to human. In: Proceedings of the 5th Environmental Technology and Management Conference, Bandung, Indonesia p. 23–24.
- SHARA S., MOERSIDIK S.S., SOESILO T.E.B. 2021. Potential health risks of heavy metals pollution in the downstream of Citarum River. In: IOP Conference Series: Earth and Environmental Science. Vol. 623(1), 012061. DOI 10.1088/1755-1315/623/1/012061.
- SHARMA P., SINGH S.P., PARAKH S.K., TONG Y.W. 2022. Health hazards of hexavalent chromium (Cr(VI)) and its microbial reduction. Bioengineered. Vol. 13(3) p. 4923–4938. DOI 10.1080/ 21655979.2022.2037273.
- SHIL S., SINGH U.K. 2019. Health risk assessment and spatial variations of dissolved heavy metals and metalloids in a tropical basin system. Ecological Indicators. Vol. 106, 105455. DOI 10.1016/j. ecolind.2019.105455.
- SNI 6989.7:2009. Air dan air limbah Bagian 7: Cara uji seng (Zn) secara Spektrofotometri Serapan Atom (SSA) – nyala [Water and waste water – Part 7: Test method for zinc (Zn) by Atomic Absorption Spectrophotometry (AAS) – flame]. Jakarta. Badan Standardisasi Nasional.
- SNI 6989.8:2009. Air dan air limbah Bagian 8: Cara uji timbal (Pb) secara Spektrofotometri Serapan Atom (SSA) – nyala [Water and waste water – Part 8: Test method for lead (Pb) by Atomic Absorption Spectrophotometry (AAS) – flame]. Jakarta. Badan Standardisasi Nasional.
- SNI 6989.16:2009. Air dan air limbah Bagian 16: Cara uji kadmium (Cd) secara Spektrofotometri Serapan Atom (SSA) – nyala [Water and waste water – Part 16: Test method for cadmium (Cd) by Atomic Absorption Spectrophotometry (AAS) – flame]. Jakarta. Badan Standardisasi Nasional.

- SNI 6989.17:2009. Air dan air limbah Bagian 17: Cara uji krom total (Cr-T) secara Spektrofotometri Serapan Atom (SSA) – nyala [Water and waste water – Part 17: Test method for chrome total (Cr-T) by Atomic Absorption Spectrophotometry (AAS) – flame]. Jakarta. Badan Standardisasi Nasional.
- SNI 6989.18:2009. Air dan air limbah Bagian 18: Cara uji nikel (Ni) secara Spektrofotometri Serapan Atom (SSA) – nyala [Water and waste water – Part 18: Test method for nickel (Ni) by Atomic Absorption Spectrophotometry (AAS) – flame]. Jakarta. Badan Standardisasi Nasional.
- SNI 6989.68-2009. Air dan air limbah Bagian 68: Cara uji kobal (Co) secara Spektrofotometri Serapan Atom (SSA) – nyala. [Water and waste water – Part 68: Test method for cobalt (Co) by Atomic Absorption Spectrophotometry (AAS) – flame]. Jakarta. Badan Standardisasi Nasional.
- TADESSE G.L., GUYA T.K. 2017. Impacts of Tennery effluent on environment and human health. Journal of Environmental and Earth Science. Vol. 7(3) p. 88–97.
- TALOUIZTE H., MERZOUKI M., BENLEMLIH M., AMRAOUI M.B. 2020. Chemical characterization of specific micropollutants from textile industry effluents in Fez City, Morocco. Journal of Chemistry. Vol. 2020, 3268241. DOI 10.1155/2020/3268241.
- TARIGAN L.B. 2015. Analisis risiko kesehatan lingkungan mangan pada air bersih di masyarakat desa supul Kecamatan Kuatnana Kabupaten Timor Tengah Selatan Tahun 2015 [Environmental health risk analysis of manganese in clean water in the community of Supul village, Kuatnana District, South Central Timor Regency in 2015]. Kupang. Jurusan Kesehatan Lingkungan, Politeknik Kesehatan Kementerian Kesehatan RI pp. 60.
- USEPA 2004. Risk assessment guidance for superfund. Vol. I. Human health evaluation manual. P. E. Supplemental guidance for dermal risk assessment. Washington, DC. Office of Superfund Remediation and Technology Innovation, U.S. Environmental Protection Agency pp. 156.
- WIBOWO S.B. 2013. Dampak kualitas perairan hubungannya terhadap risiko kesehatan di perairan Donan Cilacap-Jawa Tengah [The impact of water quality on its relationship to health risks in the Donan waters of Cilacap-Central Java]. Bogor. Sekolah Pascasarjana, Institut Pertanian pp. 97.
- ZAKIR H.M., SHARMIN S., AKTER A., RAHMAN M.S. 2020. Assessment of health risk of heavy metals and water quality indices for irrigation and drinking suitability of waters: A case study of Jamalpur Sadar area, Bangladesh. Environmental Advances. Vol. 2, 100005. DOI 10.1016/j.envadv.2020.100005.