

HEAVY METAL ACCUMULATION IN RICE (*ORYZA SATIVA L.*) FROM IRRIGATION WATER SOURCES OF CITARUM RIVER AND TARUM BARAT CANAL TO PUBLIC HEALTH RISK

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Abstract

Food safety are needed in Karawang District, which is the second largest rice producing area in West Java, the Citarum River water used as an irrigation source is polluted by industrial waste. This study aims to identify the heavy metal content of paddy water, soil and accumulation of rice using polluted irrigation water sources that pose a risk to public health. Total of 60 samples from water, soil, rice were measured using Atomic Absorption Spectrophotometry (AAS) and analysed for bioaccumulation factors (BAF). The questionnaire results were used to calculate farmers' health risk using deterministic and probabilistic Monte Carlo. The concentrations of water and soil in the irrigated rice fields of the Citarum River (SCT) and the irrigated rice fields of the West Tarum Canal (STBT) were still below the permitted standards. The concentrations of Pb and Cr metals in rice in SCT rice fields were above the quality standards. BAF>1 indicates Cu metal in rice in SCT and STBT rice fields can accumulate heavy metals. The results show that Hazard Index or HI>1 indicates farmers in both rice fields have a noncarcinogenic risk with a contribution level of Hazard Quotient Cu of 57.71%-63.78%. Cancer risk of Cr metal showed that exceeds the acceptable value both deterministic and probabilistic approaches, thus indicating a carcinogenic risk to farmers in both study areas. Proper monitoring of soil and irrigation is needed with prevention of consumption of contaminated rice.

Keywords: *heavy metals, bioaccumulation, rice fields, health risk analysis*

Introduction

Rice is a very important food crop for half the world's population. Rice is needed as the main food for 90% of the Indonesian population. This is because rice has a high carbohydrate content and is the main source of energy for the Indonesian population, especially in rural communities because rice is able to fulfil 35-60% of the body's calorie needs Rice is a very

important food crop for half the world's population. Rice is needed as the main food for 90% of the Indonesian population. This is because rice has a high carbohydrate content and is the main source of energy for the Indonesian population, especially in rural communities because rice can fulfil 35-60% of the body's calorie needs (Rasydy et al., 2021). Indonesia is known to be the second largest rice consuming country in the Asia with rice consumption reaching 163 kg/capita/year Indonesia is known to be the second largest rice consuming country in the world with rice consumption reaching 163 kg/capita/year (Jamal et al., 2023). However, pollution in rice fields due to high

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concentrations of heavy metals from the use of contaminated irrigation water can have an impact on the quality of the crops produced. However, pollution in rice fields due to high concentrations of heavy metals from the use of contaminated irrigation water can have an impact on the quality of the crops produced (Yu et al., 2022).

Heavy metals are potential pollutants in rice and are classified as major toxic chemicals due to their high potential risk to ecosystems and human health as they can accumulate in soil and cause contamination in the food chain. Heavy metals such as Cu, As, Cr, Cd and Pb are of major concern in soil and food contamination, especially in rice cropping systems. Their high toxicity properties can pose a great risk to human health through dietary intake of contaminated food crops via transfer of heavy metals from soil to plant tissues via roots or direct atmospheric deposition onto plant surfaces. These toxic elements accumulate in the soil, causing potential contamination in the food chain (Zulkafflee et al., 2020). Rice contaminated with heavy metals can cause health problems such as DNA damage which can reduce energy levels. At the molecular level, heavy metals can interact with thiols, amino acids to form metal complexes that can inhibit protein activity. The transfer pathway of heavy metals from industry to the environment can be through polluted water, soil and plants through daily food intake so that it can interfere with human health (Shah et al., 2023).

Several studies related to heavy metals in rice fields have been conducted, by (Aziz et al., 2023; Hasan et al., 2022; Xiao et al., 2019) characterisation of heavy metals in rice plant parts, the results show that there are heavy metal elements with significant concentrations that pose a health risk to local population groups, especially in industrial areas.

Karawang Regency was chosen as the research area because it is one of the second largest rice producing regions in West Java. In addition, the development of industrial and residential areas in this area adds to the risk of more industrial waste being discharged into the water body which is also used as a source of irrigation water for paddy fields. In 2022, there were approximately 276 companies discharging waste into the Citarum Hilir River, both from industries located in the Industrial Estate and industries outside the Industrial Estate (industrial zone). These industries are dominated by the textile, metal processing and automotive industries (DLH Kab Karawang, 2022). Based on the 2022 water quality monitoring data of DLH Karawang Regency, the Lower Citarum River detected heavy metals Cu, Ni and Zn exceeding the class 2 quality standard (DLH Karawang, 2023). Meanwhile, sediments from the West Tarum Channel were detected to contain heavy metals Cu, Pb, Ni, Cr and Hg (Moelyo et al., 2012).

Based on these considerations, this study was conducted to determine the content and accumulation of heavy metals through the transfer path from water to rice field soil until it accumulates in rice plants in rice fields that use polluted irrigation water sources so that it can pose a risk to human health.

Research Methodology

Research Location

The sampling locations were located in irrigated rice fields of Citarum River in Anggadita Village and irrigated rice fields of West Tarum Canal in Puseurjaya Village, both of these areas are industrial development designation areas shown in Figure 1. So that the rice fields are surrounded by various types of industries both inside the industrial area and outside the industrial area. The location of the sampling points was determined using a simple random sampling technique with a random tool using the

Arcgis 10.8 application and total 20 sampling points from 2 rice fields. Samples were taken in a composite manner consisting of 5 sub-samples and taken in a systematic pattern from each end and centre of the field. The sampling distance of each sub-sample is about 20-50 metres. All sampling points of the two rice fields were confirmed to receive water flow from the Lower Citarum River and the West Tarum Canal.

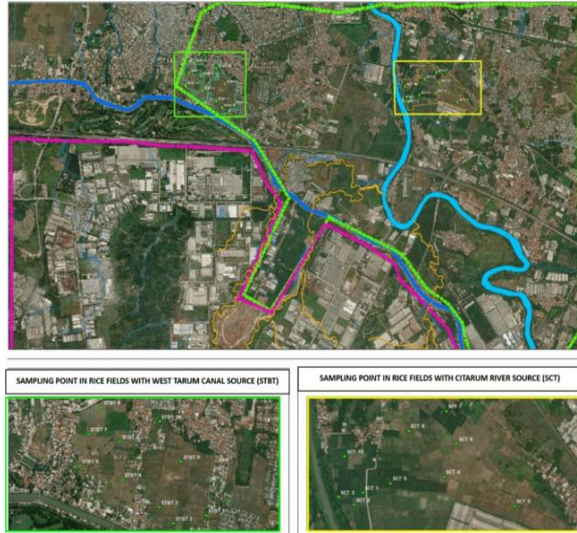


Figure 1. Research Location

Determination of Heavy Metal concentrations in Paddy Water, Soil and Rice

The selection of parameters to be examined was tailored to the types of heavy metals detected in the 2 irrigation water sources, the heavy metal characteristics of the dominant types of industries discharging wastewater into the Lower Citarum River, the risk to health and the high toxicity. The supporting parameters for this study are soil pH and temperature which were determined according to the soil pH test method SNI 6787: 2015. Water and soil sampling were conducted before the harvest period and still receiving irrigation water supply. The water sampling method refers to SNI 6989.57-2008 on surface water sampling methods and the method from Ohio EPA, 2001 was used for sampling paddy field soil. Preservation of water and paddy soil samples was carried out before the samples were analysed. All samples were

extracted using the wet deconstruction method before metal measurements were taken using the Atomic Absorption Spectrometry (AAS).

Interview and Questionnaire to Farmers

The determination of the number of respondents and the determination of respondents who will be used as subjects in this study refers to the NIOSH Occupational Exposure Sampling Strategy using a simple random sampling technique using a random number table. For a degree of confidence of 0.95 ($\alpha = 0.05$), the minimum sample size required for each population size (N) is shown in Table 1. as follows. The population in this study were all tenant farmers in 2 research locations, namely rice fields that use water sources from the Citarum River and the West Tarum Canal, totalling 20 and 25 people, so the minimum sample size for data collection of respondents in this study was 20 people.

Table 1. Determination of the minimum sample size (Leidel et al, 1977)

Population Total (N)	7-8	9-11	12-14	15-18	19-26	27-43	44-50	51-...
Sample Total (n)	6	7	8	9	10	11	12	14

Bioaccumulation Factors (BAF)

Bioaccumulation factor is the relationship between heavy metal concentrations in rice samples and heavy metal concentrations in soil samples. (Satpathy et al, 2014). The following formula is used to calculate the transfer of heavy metals from soil to plants in Eq. (1) :

$$BAF = \frac{C_{rice}}{C_{soil}} \tag{1}$$

Where:

C (rice) = concentration of heavy metals in rice,
 C (soil) = concentration of heavy metals in soil.

A BAF value > 1 indicates that the accumulation of heavy metals in the rice is greater than in the medium (soil). If BAF < 1, the rice absorbs

heavy metals but does not accumulate them (EPA, 2023).

Statistical test

Statistical tests used in this study include descriptive statistical tests, Pearson correlation tests, multivariate regression tests and unpaired t-tests.

Health risk analysis (Determination Approach)

Health risk analyses were conducted based on the determination of heavy metal concentrations in paddy field water, soil and rice.

Average Daily Dose (ADD) is exposure dose of heavy metals through the ingestion route is calculated using the following formula in Eq. (2).

$$ADD = \frac{C_{\text{rice}} \times IR \times EF \times ED}{AT \times BW} \quad (2)$$

Where C_{rice} is metal concentration (mg/kg); IR is ingestion rate, EF is daily exposure frequency (365 days/year); ED is exposure duration (years), based on questionnaire; BW is body weight (kg), questionnaire; Average time (AT) non-carcinogenic is ED years \times 365 days/year, AT Carcinogenic is 70 years \times 365 days/year is 25550 days (USEPA, 1989).

The calculation of risk characteristics with deterministic approach is divided into two, namely non-carcinogenic and carcinogenic. Non-carcinogenic risks are expressed by the notation HQ or Health Quotient in Equation (3). In addition, the total hazard index (HI) is calculated by summing the HQ of each element, as described by Equation (4):

$$HQ = \frac{ADD}{Rfd \text{ Ingestion}} \quad (3)$$

$$HQ = \sum HI \quad (4)$$

If the HQ value < 1 , it can be said that the risk is safe, and if the HQ value > 1 , the risk level is

said to be unsafe. To calculate the overall risk agent, the risk level is expressed by the Health Index (HI). HI is the aggregated value of the HQ value of each risk agent. If the HI value is > 1 then it is said to be unsafe.

The risk level for carcinogenic effects is expressed in the notation Cancer Risk (CR) which is the multiplication of the intake value by the cancer slope factor (CF) value for each agent. To characterise the risk for carcinogenic effects, the following calculations were performed in Eq. (5):

$$CR = ADD \times CF \quad (5)$$

The risk level is acceptable or safe if $CR \leq 10^{-6}$, and if $CR > 10^{-4}$ then the risk level is declared unsafe (Ren et al, 2021)

Health risk analysis (Probabilistic Monte Carlo)

Health risk analysis with a probabilistic approach (Monte Carlo) it is used to reduce uncertainty and variability from the deterministic approach. The parameters used for the calculation are the intake and CR values from the carcinogenic risk evaluation in humans. In this evaluation, to assess the variability of the distributions, each parameter was tested using Monte Carlo simulation. In the initial stage, probability distributions were analysed for input variables such as metal concentration, rice consumption level, body weight and exposure duration. Monte carlo simulation with 10,000 iterations was used for exposure modelling and risk assessment using @RiskPlatform software (8.6.1.0, Palisade Company, LLC).

Result and Discussion

Characteristics and Content of Heavy Metals in Water and Soil

Physico-chemical parameters can affect availability, mobility and plant uptake. The quality of paddy field water is influenced by the quality of the Citarum River and the West Tarum Canal. Table 2. shows that the average

pH and temperature of paddy field water are still in good condition because they are below the quality standards according to FAO 1999. The average concentrations of Pb, Cu and Cr in rice field water from the Citarum River source tend to be the same, which is in the range of 0.02-0.03 while the level of heavy metal concentrations in rice field water from the West Tarum Canal source shows that $Cr > Pb > Cu$. there is a significant difference ($P < 0.05$) in the concentration of heavy metal Cr in rice field water from the Citarum River and West Tarum Canal sources. This is due to the influence of the location of the sampling point and the distance of the chromium producing industry (electronic waste collection industry) in the rice field area of the West Tarum Canal source. Generally, chromium concentration levels were found to be more than 1000 ppm in mobile phone PCBs (Lakshmi et al., 2020).

Table 3. shows the characteristics of paddy field soil, soil pH tends to be acidic in both paddy fields because it is in the range of 4.4-6. This can increase the mobility and concentration of heavy metals from the soil to rice plants. temperature in $STBT > SCT$ rice fields because sampling was done during the day. The average concentration of metals in irrigated soil from both rice fields showed that the concentration was still below the quality standard. This study is in line with the soil Pb concentration in agricultural land of the Upper Citarum River, which is still below the critical limit of the presence of allowable heavy metal concentrations (Handayani et al., 2022). There is a significant difference ($P < 0.05$) in the concentrations of heavy metals Pb and Cu in the irrigated paddy fields of the Citarum River and the West Tarum Canal. Based on the Pearson correlation coefficient matrix, There is a significant difference ($P < 0.05$) in the concentration of heavy metals Pb and Cu in the irrigated rice field soil of Citarum River and West Tarum Canal. Based on the Pearson

correlation coefficient matrix, there is a significant positive strong correlation between pairs of elements of soil samples in STBT rice fields, namely in Cu-Pb (0.627) and Cr-Pb (-0.748) metals. so this indicates that there are contaminants from the same/ homogeneous source.

Table 2. Average characteristics and concentrations of heavy metals in paddy field water in 2 rice field areas

Parameters	Rice Field Water Source		P-value	Irrigation Water Quality Standard ^a
	Citarum River (SCT)	West Tarum Channel (STBT)		
pH	7.53±0.14	7.51±0.32		5-9
Temperature (°C)	26.33±0.84	29.55±2.83		35
Lead (Pb : mg/L)	0.028±0.02	0.04±0.01	0.241	2
Copper (Cu : mg/L)	0.033±0.01	0.03±0.01	0.935	0.2
Chromium (Cr : mg/L)	0.031±0.03	0.09±0.06	0.003	0.1

Source: a. FAO, 1999, ± Deviation standards, P value: Significance level for differences in SCT and STBT ($P > 0.05$ no Significance level for differences place

Table 3. Average characteristics and concentrations of heavy metals in paddy soil in 2 rice fields

Parameters	Rice Field Water Source		P-value	Soil Standard ^{a,b}
	Citarum River (SCT)	West Tarum Channel (STBT)		
pH	5.34±0.50	5.27±0.39		
Temperature (°C)	27.02±1.05	28.95±2.19		
Lead (Pb : mg/kg)	1.08±0.49	5.09±6.14	0.030	80 ^a
Copper (Cu : mg/kg)	6.49±0.65	5.85±5.22	0.039	140 ^b
Chromium (Cr : mg/kg)	4.72±5.80	6.93±8.61	1.000	90 ^a

Source: a: GB 15618-1995 ; b: European standards 2002; ± Deviation standards , P value: Significance level for differences in SCT and STBT ($P > 0,05$ no Significance level for differences place.

Heavy Metal Accumulation in Rice

Table 4 shows that the highest average concentration in rice from rice fields irrigated by the Citarum River is $Cu > Cr > Pb$. The order of magnitude of metal concentrations in rice follows the order of metal concentrations in soil from rice fields irrigated by the Citarum River. The highest Cu concentration and the lowest Cr and Pb concentrations suggest the accumulation of metals in rice may result from metal pollution from soil (Ahmed et al., 2019). These results were reinforced by multiple regression tests that showed the influence of Cu concentration in rice was 98.1% influenced by Cu content of water and soil, while the highest order of metal concentration in rice from irrigated rice fields of West Tarum Canal was $Cu > Pb > Cr$.

Table 4. Average concentrations of lead, copper and chromium in rice in the two study areas

Parameters	Rice		P-value	Rice Standard _{a,b}
	Citarum River (SCT)	West Tarum Canal (STBT)		
Lead (Pb:mg/kg)	0.54±0.23	0.80±0.31	0.05	0.2 ^a
Copper (Cu:mg/kg)	7.54±1.29	4.93±0.70	0.00	20 ^b
Chromium (Cr:mg/kg)	2.21±1.80	0.40±0.45	0.01	1 ^b

Source: a, WHO/FAO 2022; b, WHO/FAO 2002; ± Deviation standards, P value: Significance level for differences in SCT and STBT ($P > 0.05$ no Significance level for differences place

This study is also in line with previous research in 3 industrial areas in Bangladesh where Cu concentrations are still below critical limit of WHO/FAO in respectively area, i.e. 38.12 ± 11.21 mg/kg (Savar), 25.34 ± 8.56 mg/kg (Gazipur), dan 19.74 ± 5.87 mg/kg (Ashulia) (Hasan et al., 2022). The concentration of Pb metal exceeded the WHO/FAO quality standard (2022) in both SCT and STBT fields. The average Cr metal was only detected to exceed the quality standard in SCT while the average Cu concentration was still far below the quality standard. There were significant differences

($P < 0.05$) in the concentrations of heavy metals Pb, Cu and Cr in rice from irrigated rice fields of the Citarum River and West Tarum Canal.

Bioaccumulation Factor

The bioaccumulation factor (BAF) for heavy metal uptake by rice in the Citarum River source rice field is shown in Figure 2. The accumulation and transfer of heavy metals in rice may pose potential health risks. The trend of BAF values for heavy metals in the West Tarum Canal source rice fields is the same as in the Citarum River source rice fields, which is in the order of $Cu > Pb > Cr$. The average BAF value found for Cu metal in SCT rice fields was 1.17 ± 0.22 . Sampling point SCT 6 showed a higher BAF value compared to other sampling points. This indicates that on average, rice in SCT rice fields can accumulate and absorb heavy metal Cu from paddy soil. The BAF of Cu metal was also observed in the study of Aziz et al, 2019 which stated that the BAF value was > 1 in 3 different land use areas with the highest level of IP (industry) $>$ RP (rural) $>$ TP (transportation). Whereas Pb and Cr metals showed relatively low BAF values low < 1 means that the bioavailability of heavy metals in the soil is low so that rice is not able to absorb these metals. This result is similar to the results of previous studies which state that rice is a non-accumulator of heavy metal Pb (Ashraf et al., 2015).

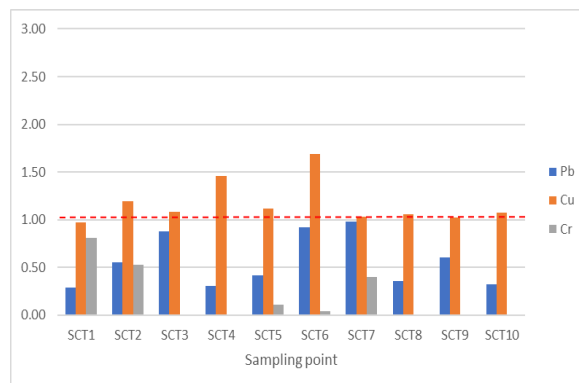


Figure 2. BAF rice field SCT

Figure 3 shows the BAF in the STBT rice fields. Sampling point STBT 4 showed a higher BAF value on Cu metal compared to other sampling points. The average BAF value found in Cu metal in STBT rice fields is 1.43 ± 0.91 . Meanwhile, sampling point STBT 8 showed the highest BAF value of Pb (1.08) and Cr (2.00) metals. This indicates that the rice plants from these sampling points can accumulate and absorb heavy metals from the rice field soil.

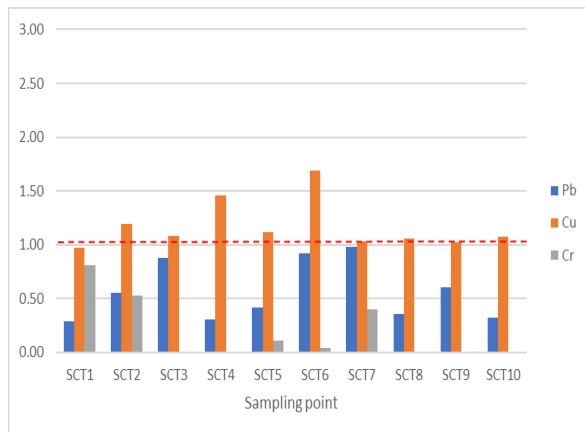


Figure 3. BAF rice field STBT

Health Risk Analysis (HRA)

The result of the non-carcinogenic risk characterisation of heavy metals is the aggregation of HQ values of each element expressed as HI values. The results of HQ and HI calculations are summarised in Table 5. The results of the evaluation of non-carcinogenic hazards in farmers from the Citarum River rice fields show that the HQ of Pb and Cr < 1 while Cu > 1, which means that the accumulation of Cu metal in the body of farmers in Anggadita Village can pose a non-carcinogenic risk. Excessive exposure to Cu in the body can cause liver damage and gastrointestinal disorders (NIH, 2022). The contribution rate of HQCu to HI is 57.71%, HQPb 41.84% and HQCr 0.44%. Therefore, Cu and Pb are the main contributing factors that pose non-carcinogenic risks.

The results of the evaluation of non-carcinogenic hazards in rice from rice fields of the West Tarum Canal showed that there were

no metals with HQ > 1 for Pb, Cu and Cr metals. However, HI in farmers who consume rice from the West Tarum Canal of 1.201 exceeds the value of 1, which means that farmers in puseurjaya village who consume rice have a non-carcinogenic risk. The contribution rate of HQPb to HI was 63.78%, HQCu 36.13% and HQCr 0.08%. Therefore, Pb and Cu are the main contributing factors to the non-carcinogenic risk of the farmers in the region.

Table 5. Non carcinogenic risks for farmers in Citarum River rice fields and West Tarum Canal

Respondent	HQ	Pb	Cu	Cr	HI
Farmers in Rice Fields irrigated by the Citarum River	Min	0.371	0.472	0.004	
	Max	1.188	1.509	0.012	
	Mean	0.757	1.044	0.008	1.809
Farmers in Rice Fields irrigated by West Tarum Canal	Min	0.652	0.264	0.001	
	Max	1.566	0.888	0.002	
	Mean	0.766	0.434	0.001	1.201

The carcinogenic risk was analysed using the Cancer Risk (CR) to determine whether or not exposure to heavy metal contaminants causes carcinogenic effects for humans. The CR values shown in Table 6 are calculated dividing the intake by the SF of each contaminant Pb (0.0085 mg/kg/day) and Cr (0.5 mg/kg/day). The results of the CR and TCR calculations are summarised in Table 7. The results of the carcinogenic hazard evaluation in rice from the rice fields of the West Tarum Canal and Citarum River showed that the average Cr metal from both farmers had a carcinogenic risk. Previous studies have reported the carcinogenic risk mediated by Cr exposure through rice has the greatest carcinogenic risk which is similar to this study (Lu et al., 2021). While Pb metal showed lower results than acceptable in both farmers, this is in line with research from Zulkaflee (2019) with the LCR value of lead (Pb) exposure in adults ranging from 2.0×10^{-6} to 6.0×10^{-6} .

Table 6. Carcinogenic risks for farmers in rice fields of the Citarum River and Tarum Barat Canal

Respondent	CR	Pb	Cr
Farmers in Rice Fields irrigated by the Citarum River	Min	5.9.E-06	1.39.E-03
	Max	2.27.E-05	5.36.E-03
	Mean	1.33.E-05	3.14.E-03
Farmers in Rice Fields irrigated by West Tarum Canal	Min	8.0.E-07	2.49.E-05
	Max	2.40.E-05	7.13.E-04
	Mean	7.5.E-06	2.22.E-04

The potential carcinogenic risk due to lifetime exposure to Pb and Cr through consumption of rice grown in irrigated rice fields of the citarum river and west tarum canal is shown in Table 7.

Table 7. Carcinogenic risk for farmers in rice fields of Citarum River and West Tarum Canal with probabilistic approach

Heavy metals	Cancer Risk	
	Farmers in Rice Fields irrigated by the Citarum River	Farmers in Rice Fields irrigated by West Tarum Canal
	Pb	0.20.E-05
Cr	3.03.E-02	5.85.E-03

The results of Cr values among farmers in SCT and STBT are greater than the acceptable risk of 1.0×10^{-4} , which implies that the probability of long-term exposure may pose a carcinogenic risk to the local community. This study is in line with research from Zhao (2023) in China, the CR value for rice chromium metal is greater than the acceptable risk of 1.0×10^{-4} . While the CR value for Pb metal in both farmer origins, SCT (0.20×10^{-5}) and STBT (0.42×10^{-5}), still shows a value below the acceptable risk. These results show that there are differences in the value of carcinogenic risk of Cr metal among farmers in SCT and STBT, but the carcinogenic risk of Pb metal tends to be the same in both probabilistic and deterministic approaches.

Conclusions

The average concentrations of rice field water and soil in Citarum River irrigated rice fields

(SCT) and West Tarum Canal irrigated rice fields (STBT) are still below the quality standards of FAO 1999, GB 15618-1995 and European standards 2002. Meanwhile, the concentrations of Pb and Cr (SCT) in rice were above the WHO/FAO quality standards. The value of the bioaccumulation factor (BAF) shows that Cu metal in SCT and STBT rice fields has a $BAF > 1$ value, meaning that rice from both areas can accumulate and absorb heavy metal Cu from paddy soil. Based on the results of the health risk analysis, an HI value > 1 indicates that non-carcinogenic risks have the potential to occur in farmers in both rice fields with a HQCu contribution level of 57.71%-63.78%. The CR value for Cr metal shows a value above the acceptable risk in both deterministic and probabilistic approaches, thus indicating the existence of a carcinogenic risk to farmers in both study areas.

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