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HEALTH RISK ASSESSMENT OF HEAVY METALS IN SHALLOT, BANTUL REGENCY

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Abstract

Introduction: *Shallots are one of the horticultural products in great demand by the public. Excessive heavy metal content in shallot will affect people's health who consume it. This study examines the health risks of the people who consume shallot products produced in Srigading Village.* ***Methods:*** *Determination of sampling locations was carried out by purposive sampling method on shallot cultivation land that was ready to harvest, as many as 30 points, in September 2021. Soil and shallot samples were tested for the heavy metal content of Pb, Cd, Co, and Ni. The public health risk assessment was analyzed by looking at the Transfer Factor (TF) value, daily intake, health risk index, hazard index, and cancer risk.* ***Results and Discussion:*** *The concentration of heavy metals in shallots is Pb 19.14 – 30.04 mg kg-1, Cd 1.03 – 2.21 mg kg-1, Co 8.58 – 15.08 ppm, and Ni 6.00 – 10.09 mg kg-1. The average value of Transfer Factor (TF) shows metal uptake by shallots with metal levels Cd (1.07) > Pb (1.03) > Ni (0.73) > Co (0.46). The average daily dose shows that the daily consumption in children is higher than the daily dose for adults.* ***Conclusion:*** *Children are more at risk of being exposed to heavy metals compared to adults in consuming shallots based on the hazard index value. The continuous use of chemical fertilizers and chemical pesticides in shallot farming must be controlled to prevent the increase of accumulation of heavy metals in land and agricultural products.*

INTRODUCTION

Heavy metals are toxic compounds that can have negative effects on the plants, environment, animals (1), and human health (2). The presence of heavy metals naturally exists in the earth's crust but its concentration increases with the presence of anthropogenic activities, which poses a threat to food safety due to the persistent, non-degradable, and bioaccumulation properties of heavy metals (3). Exposure to heavy metals in humans can be through consumption, inhalation, and skin contact so that it can cause health risks for humans (4). Consuming agricultural products containing heavy metals is one of the pathways for heavy metals to accumulate in the human body (5). Shallots are one type of horticultural plant that is often consumed by the public because it is used as a food flavoring.

Heavy metals in land and agricultural products can come from industrial waste dumped on agricultural land (6) or in rivers that are used as a source of agricultural irrigation (5) and the application of agrochemicals in the form of fertilizers (7) and pesticides during the growing period. Soil in the industrial area of Panzhihua City, China, contains heavy metals Ni, Cr, Cu, Pb, Cd, Zn, and V which are quite high due to waste from various industries in the city (8). Vegetables grown using irrigation from industrial wastewater in Erbil City, Iraq contain heavy metals Cd, Pb, Cr, and Zn which have exceeded the critical limits determined by FAO/WHO (9). The agrochemicals in fertilizers and pesticides will accumulate in the soil and agricultural products if the application does not match the dosage and is carried out continuously over a long period of time. Soil already has a very small concentration of natural heavy metals but will increase with the source of

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fertilizers and pesticides applied to agricultural land (10). Phosphate fertilizers that are applied for a long period of time can cause the accumulation of heavy metals Pb, Zn, Cd, and others in the topsoil layer (11). The pesticide formulations used by farmers from the insecticide, herbicide, and functional groups contain an abundance of heavy metals such as Ni, Pb, As, Co, and Cr (12). The heavy metal concentrations in some of pesticide formulations have exceeded the limit when compared to the drinking water quality standards that have been determined by WHO (13) (Pb 10 ppb, As 10 ppb, Ni 20 ppb and Cr 50 ppb).

Bantul Regency has the largest shallot harvest area in D.I. Yogyakarta, which is 770 hectares or 59% of the total shallot harvest area in the DIY Province (14). The centers of shallot farming in Bantul Regency are Sanden and Kretek sub-districts. One of the villages in Sanden sub-district which has the largest shallot harvest area is Srigading sub-district with a harvested area of 77.9 ha (14). In onion cultivation, farmers in Srigading Village do not only use paddy fields but also use sand for agricultural land. Sand soil has very limited nutrient content. Therefore, one of the solutions for the farmers is to add soil enhancers, such as fertilizers. Pesticides used by farmers include 3 groups, namely herbicides, insecticides, and fungicides. The farmers used the pesticides not according to the dosage recommended in the instructions for use. The average use of production factors per hectare is 42.21 L for herbicides, 4.16 L for insecticides, and 22.83 kg for fungicides (15).

Heavy metals in vegetables will impact on the health of people who consume them (16). Heavy metals that are accumulated in human's body will have a detrimental impact on health in the form of non-carcinogenic and carcinogenic risks (17). Exposure to high concentrations of Cd metal will cause health problems such as bone complications. It also can cause cancer due to the highly toxic nature of Cd metal (18). Heavy metal Pb with high concentrations in the body can interfere a person's health because it can cause arterial pressure and various diseases (19). Infants and children are generally vulnerable to Pb metal (20).

There are plenty of health risks to humans caused by consuming agricultural products containing heavy metals. Therefore, it is very important to frequently conduct a health risk assessment of agricultural products consumed by the community, one of which is shallots. This study aims to provide information about the public health risks related with the consumption of shallots produced on agricultural fields in the Srigading Village.

METHODS

This research was conducted in the agricultural land of Srigading Village, Sanden sub-District, Bantul Regency. The sampling location was chosen by conducting a purposive sampling method on shallot cultivation land that was ready for harvest. Soil samples were taken from 30 plots of land. Soil sampling was carried out at a composite soil depth of 0-25 cm with each sample consisting of 5-10 individual samples (subsamples) set diagonally on each plot of land (21). Plant samples (shallots) were taken at the same plot of land as the soil sampling location. The soil samples obtained from the field were prepared by drying the soil to obtain a constant moisture content, then pulverized using a mortar pestle to make a powder (diameter 0.5 mm). The preparation stage for the shallot samples was carried out by cleaning the shallot obtained from the field by draining them. Then the onion samples were peeled, cut into small pieces, and mashed using a blender. Then, the soil and shallot samples that had been prepared were analyzed for the total heavy metal concentration Pb, Co, Ni, and Cd. The method of heavy metal analysis in soil and shallot samples using Atomic Absorption Spectrophotometer (AAS). The method was applied at Indonesian Soil Research Institute with modifications to the volume of the sample analyzed, the volume of the application of concentrated nitric acid solution, and the stages of destruction (22-23). Observation of heavy metal parameters in the sample was carried out at the Integrated Laboratory of Indonesian Agricultural Environment Research Institute (Balingtan), Ministry of Agriculture. The results of the analysis of heavy metals in soil and shallots were compared with the quality standards set by Alloway (24) and BPOM RI (25).

The data on public health risk assessment were analyzed by examining Transfer Factor (TF), Average Daily Dose (ADD), Health Risk Index (HRI), and Hazard Index (HI), and Cancer Risk (CR).

Transfer Factor (TF)

The value of Transfer Factor (TF) is the ratio between the concentration of heavy metals in soil and shallots. The TF value indicates the ability to transfer heavy metals from the soil to the shallots (26).

$$TF = \frac{C\ plant}{C\ soil}$$

The C_{plant} dan C_{soil} represents concentrations of heavy metals in vegetable and soil extracts based on dry weight. Plants are divided into three categories based on the transfer factor value (TF) obtained, namely plants as metal accumulators with a transfer factor value (TF) > 1, plants as metal excluders with a transfer factor value (TF) < 1, and plants as metal indicators with a value of transfer factor (TF) is close to one. Plants that are consumed by humans, such as shallots, are certainly not expected to have a TF value >1.

Average Daily Dose (ADD)

Average Daily Doses (ADD) is determined by the following formula:

$$ADD = \frac{Ci \ x \ IR \ x \ EF \ x \ ED}{BW \ x \ AT}$$

Ci is the heavy metals concentration in vegetables, IR is the level of consumption in children and adults, EF is the frequency of exposure, ED is the duration or length of the exposure, AT is average time to non-carcinogenic exposure, and BW is the body weight of adults and children. ADD estimates were used to assess health risks in terms of their non-carcinogenic and carcinogenic effects (27).

Health Risk Index (HRI)

$$HRI = \frac{ADD}{RfD}$$

The Health Risk Index (HRI) of people who consume vegetables contaminated with heavy metals is estimated by the ratio between the reference oral dose (RfD) and the daily intake of metals for each metal. HRI value <1 indicates that people who are exposed to heavy metals are at a safe health risk.

The reference oral doses (RfD) were 0.001 and 0.004 mg/kg/day for Cd and Pb, respectively (28). Ni 0.02 mg/kg/day (29), Co 0.043 mg/kg/day (30).

Hazard Index (HI)

USEPA has developed a Hazard Index (HI) calculation as the sum of the Health Risk Index (HRI) values obtained for all metals in the observed soil (31). The potential health risks to the community caused by consuming more than one heavy metal can be estimated through the Hazard Index (HI). The hazard index value can be used to determine the toxicity of vegetables consumed by the community. HI is calculated as the sum of HIR.

$$HI = HRIPb + HRICd + HRNi$$

The Hazard Index (HI) value > 1 indicates a possible non-carcinogenic effect, while a possible non-carcinogenic risk is indicated by HI value < 1 (32).

Cancer Risk (CR)

The cancer risk is calculated by multiplying the average daily dose by the Slope Factors (SF). ADD is the average daily intake of metals per day in units of mg/kg/day, which is an indication of the average daily dose of heavy metal exposure for life. SF is a cancer slope factor indicating the risk of a lifetime mean dose of 1 mg/kg/day or depending on the type of heavy metal. Acceptable cancer risk of $1x10^{-6}$ to $1x10^{-4}$. The Slope Factor (SF) value for Pb was 0.0038 mg/kg/day and the SF value for Cd was 0.0085 mg/kg/day.

$$Cancer\ Risk\ (CR) = ADD \times SF$$

RESULTS

Heavy Metals in Shallot Fields

The concentration of heavy metal Pb ranged from 18.80-30.95 mg kg^{-1}**.** Co, Ni, and Cd metal concentration in the soil ranged from 1.58-2.21 mg kg^{-1}, 3.04-6.63 mg kg^{-1}, and 3.33-7.49 mg kg^{-1}. The graph of heavy metal concentrations of Pb, Cd, Co, and Ni in the soil can be seen in Figure 1 and statistical data on heavy metal concentrations in shallot fields can be seen in Table 1.

Table 1. Statistical Data on Heavy Metal Concentrations in Shallot Farmland (n=30)

Statistics	Pb	Cd	Co	Ni
	mg kg^{-1}			
Mean	27.94	1.96	5.71	6.39
Median	28.18	1.98	5.86	6.74
Standard Deviation	2.23	0.14	0.78	1.01
Minimum	18.80	1.58	3.04	3.33
Maximum	30.95	2.21	6.63	7.49
Count	30	30	30	30
CV (%)	7.99	7.12	13.61	15.81
Quality Standard (Alloway, 1995)	100	3	50	100

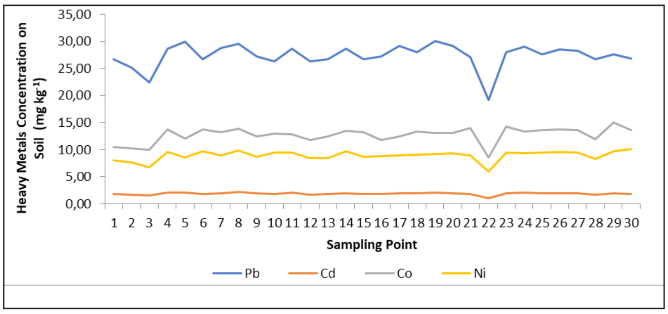

Figure 1. The Heavy Metal Concentration at Each Point of Soil Sampling in The Shallot Field

Heavy Metals in Shallots

The concentration of heavy metals in shallots is Pb 19.14 – 30.04 mg kg^{-1}, Cd 1.03 – 2.21 mg kg^{-1}, Co 8.58 – 15.08 ppm, and Ni 6.00 – 10.09 mg kg^{-1}. The graph of heavy metal concentrations of Cd, Pb, Ni, and

Co in shallots can be seen in Figure 2 and statistical data on heavy metal concentrations in shallots can be seen in Table 2.

Table 2. Statistical Data on Heavy Metal Concentrations in Shallot (n=30)

Statistics	Pb	Cd	Co	Ni
	mg kg^{-1}			
Mean	27.38	1.87	12.73	8.92
Median	27.59	1.91	13.14	9.10
Standard Deviation	2.20	0.21	1.41	0.90
Minimum	19.14	1.03	8.58	6.00
Maximum	30.04	2.21	15.08	10.09
CV (%)	8.03	11.45	11.08	10.09
Quality standard Alloway, 1995	50	5	15	5
BPOM RI, 2018	0.20	0.05	-	-

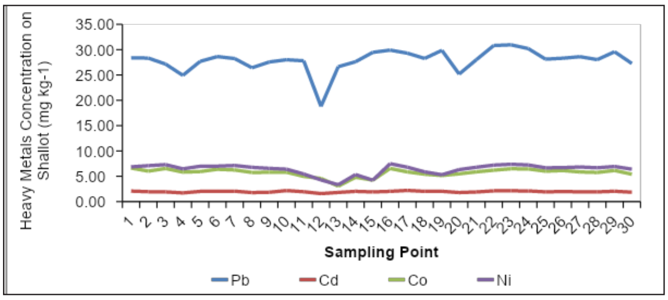

Figure 2. The Heavy Metal Concentration at Each Sampling Point of Shallots

Transfer Factor (TF)

The average value of TF on heavy metals Pb and Cd was > 1, each TF value was 1.029 and 1.073. The average value of TF on heavy metals Co and Ni was < 1, each TF value was 0.456 and 0.727. Value of the heavy metal transfer factor in each sample of shallot can be seen in Figure 3, and the average value of heavy metal transfer factor in shallots can be seen in Figure 4.

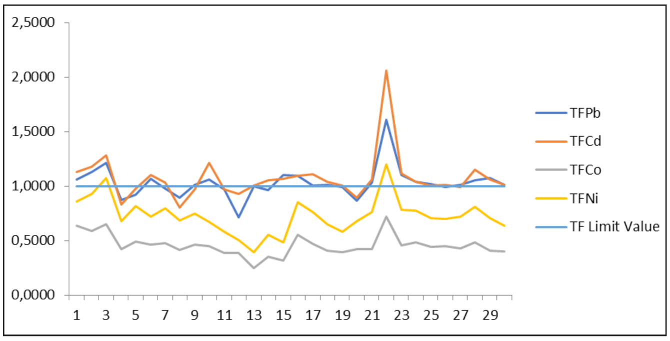

Figure 3. Heavy Metal Value Transfer Factor in Shallots

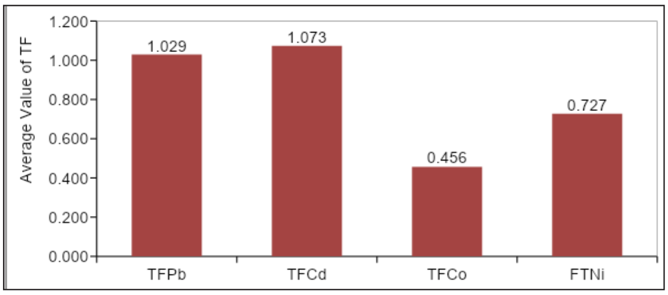

Figure 4. The Average Value of Heavy Metal Transfer Factor in Shallots

Average Daily Dose (ADD)

The highest daily dose of metal in adults and children was Pb with Pb concentrations of 0.38 mg/kg/day and 1.76 mg/kg/day. The lowest dose of metal was Cd with a concentration of 0.03 mg/kg/day for adults and 0.12 mg/kg/day for children. The average value of daily doses in children is higher than adults because children's weight is less than adults so that the divisor variable is smaller. The average daily dose for children and adults in consuming shallots can be seen in Table 3.

Table 3. The Average Daily Dose of Shallots in the Community

Variable	Pb	Cd	Co	Ni
	mg/kg/day			
Adult	0.38	0.03	0.08	0.09
Children	1.76	0.12	0.36	0.40

Hazard Index (HI) and Health Risk Index (HRI)

The value of the health risk index is in Table 4. It shows a value that is far from the expected value, which is <1. The highest HRI value is shown in Pb which reaches a value of 107.51 in adults and a value of 501.70 in children. The lowest value for metal Co is 1.79 in adults and 8.35 in children. The hazard index value shows a value of 166.52 in adults and a value of 777.10 in children.

Table 4. The Health Risk Index (Non-Carcinogenic) and Danger Index of Shallot

Variable	Pb	Cd	Co	Ni	HI
Adult	107.51	52.92	1.79	4.31	166.52
Children	501.70	246.95	8.35	20.09	777.10

Cancer Risk (CR)

The cancer risk value of Pb 1.43 x 10^{-3} in adults and 6.67x10-3 in children, metal Cd 2.2 x 10^{-4} for adults and 1.05x10^{-3}. The value of cancer risk (CR) can be seen in Figure 5.

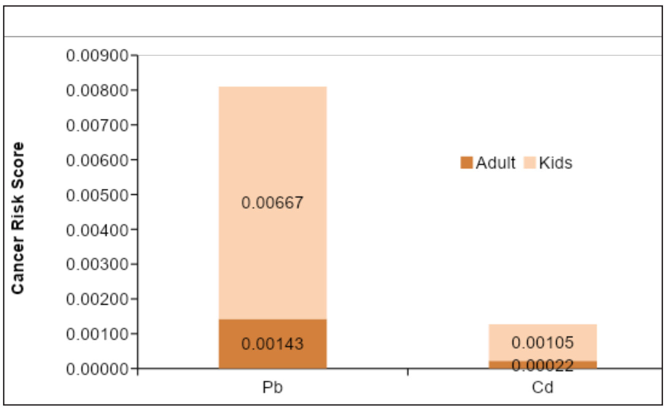

Figure 5. The Value of Cancer Risk in Shallots

DISCUSSION

Heavy Metals in Shallot Fields

Heavy metals Ni, Cd, Co, and Pb were detected at all soil sampling points in the shallot farmland of Srigading Village. The average concentration of heavy metals in the soil from the highest to the lowest concentration was Pb (27.94 mg kg^{-1})>Ni (6.39 mg kg^{-1})>Co (5.71 mg kg^{-1})>Cd (1.96 mg kg^{-1}).

The concentration of heavy metal Pb ranged from 18.80-30.95 mg kg^{-1}. This indicates that the concentration of Pb in the shallot farmland in Srigading Village is below the quality standard set by Alloway, which is 100 mg kg^{-1} (20). The concentration of Pb in agricultural land in Srigading, apart from being sourced from agrochemicals, is also probably due to Pb released from motorized vehicles because the soil sampling location is < 500 m from the road.

Pb metal originates from vehicle exhaust gas, which is due to the incomplete combustion of gasoline engines resulting in toxic exhaust gasses. Gasoline-fueled vehicles can contribute 79-97% Pb concentrations in ambient air (33) because the combustion process of a gasoline-fueled vehicle engine is able to produce exhaust gas with a lead content of 0.09 grams of lead per 1 km (34). The concentration of Pb in the air will be influenced by the density of vehicles crossing the road. Therefore, the denser the vehicles, the higher the concentration of Pb in the air (35).

The concentrations of Ni, Co, and Cd in the soil ranged from 3.33-7.49 mg kg^{-1}, 3.04-6.63 mg kg^{-1}, and 1.58-2.21 mg kg^{-1}. The concentrations of Ni, Co, and Cd were all below the quality standard set by Alloway (24). The average Cd metal is 1.96 mg kg^{-1}, it shows a value that is not far from the Cd metal quality standard, which is 3 mg kg^{-1}. Furthermore, the sources of additional Cd metal concentrations on agricultural land in Srigading Village need to be controlled. The application of fertilizers and pesticides contributed greatly to the addition of Cd in the soil, such as phosphate fertilizers which are the largest Cd metal input in the soil (36).

Heavy Metals in Shallots

Shallots grown in Srigading Village are Thai varieties of shallots with a planting age of 50 days. Concentrations of Pb, Cd, Co, and Ni were detected in all shallot samples taken from the Srigading Village agricultural land. The average heavy metals concentration in shallot from the highest to the lowest concentration was Pb (27.38 mg kg^{-1})>Co (12.73 mg kg^{-1})>Ni (8.92 mg kg^{-1})>Cd (1.87 mg kg^{-1}).

The concentration of heavy metals Pb and Cd in shallots was still below the quality standard set by Alloway. The quality standard for Pb metal is 50 mg kg^{-1} and the Pb metal concentration in shallots is 19.14-30.04 mg kg^{-1}. Meanwhile, the standard for Cd metal is 5 mg kg^{-1} and the concentration of Cd in shallots is 1.03-2.21 mg kg^{-1}. The concentration of Co and Ni is beyond the quality standard set by Alloway. The quality standard for Co metal is 15 mg kg^{-1} and the concentration of Co in shallots ranges from 8.58-15.08 mg kg^{-1}. Meanwhile, the quality standard for Ni metal is 5 mg kg^{-1} and the concentration of Ni metal is between 6.00-10.09 mg kg^{-1}.

The Pb metal concentration ranged from 19.14 - 30.04 mg kg^{-1}, the value of Pb metal was very high when compared to the quality standard of 0.20 mg kg^{-1}. Cd heavy metal concentration in shallots ranges from 1.03 to 2.21 mg kg^{-1}. This indicates that the concentration of Cd metal in shallots is beyond the quality standard determined by BPOM RI (25) which is 0.05 mg kg^{-1}. The quality standards of Co and Ni heavy metals in shallots have not been determined by BPOM RI. Therefore, it is difficult to determine the status of the concentrations of Co and Ni metals in shallots. Pb and Cd heavy metal concentration in shallots that are far above the established quality standards will have an impact on food safety.

Transfer Factor (TF)

In food crops and horticulture, the expected TF value is < 1 or close to 0. Therefore, these plants have no potential as heavy metal accumulator agents and are safe for public consumption. The value of the metal transfer factor in shallots indicates the ability of shallots to accumulate heavy metals from the soil in tubers. Transfer factor values are more often used in phytoremediation plants with expected values >1. This value indicates that plants can be used as heavy metal accumulator agents.

The average value of the heavy metal transfer factor in shallots, from the highest to the lowest, was Cd>Pb>Ni>Co. The average value of TF on Pb and Cd has exceeded 1. It shows that shallots in Srigading Village can accumulate quite high Pb and Cd heavy metals. Cd metal generally has the highest TF value of other heavy metals in soil because Cd metal in soil can be absorbed by various types of vegetables, including shallots (37).

The average value of TF on Co and Ni metals does not exceed 1. It indicates that shallot plants do not accumulate Co and Ni metals from the soil.

Accumulation of heavy metals in plants consumed by the public is not expected because it will have an impact on the health of the people who consume them.

The high concentration of heavy metals in shallots in the Srigading Village can also be caused by the low content of organic matter in agricultural soil. As a result, the fertility level of the agricultural soil is also low. The low content of organic matter in the soil will affect the absorption of heavy metals by plants. In this state, heavy metals will be released from the soil bonds and form ions that move freely in the soil solution. If other metals are not able to inhibit its existence, then there will be absorption by plant roots (38).

Plant roots are the first part of plants that can absorb heavy metals from the soil. It has large atomic weights and diameters cause heavy metals to have relatively small mobility. During the absorption, heavy metals will accumulate in the closest place to the roots, namely tubers.

Average Daily Dose (ADD)

The average daily dose value of heavy metals in shallots consumed by children is higher than adult value. This indicates that by consuming shallots, children will be more at risk of severe exposure than adults (39). Children have a high daily intake and differ greatly in value from adults because of the weight factor which is much lighter than adults (40).

Hazard Index (HI) and Health Risk Index (HRI)

The value of the health risk index shown in Table 4. It shows a value that is far from the expected value, which is <1. The highest HRI value is shown in Pb which reaches a value of 107.51 in adults and a value of 501.70 in children. The lowest value for metal Co is 1.79 in adults and 8.35 in children. This shows that children are more potentially exposed to the dangers of heavy metals compared to adults. The public health risks caused by consuming shallots will vary depending on age, health conditions, and place of residence (41). The hazard index value shows a value of 166.52 in adults and a value of 777.10 in children. A hazard index value of more than 1 indicates a future risk such as cancer.

Cancer Risk (CR)

The value of cancer risk (CR) shown in Figure 5. It shows that the risk in children is greater than in adults. Shallots grown on agricultural land in Srigading Village have a cancer risk if consumed by the community This is indicated by the cancer risk value of Pb 1.43×10^{-3} in adults and 6.67×10^{-3} in children, while metal Cd 2.2×10^{-4} for adults and 1.05×10^{-3} for children. The risk value is acceptable if the value is below 10^{-6} (42). It shows that the CR value of heavy metals Pb and Cd is not well accepted for children and adults. The CR value Pb metal in adults shows that there is a possibility of 1 adult at risk of cancer out of 1,000 adults who consume shallots, and the CR value Pb metal in children indicates that there are seven children at risk of cancer out of 1000 children who consume shallots. The CR value of Cd metal for adults shows that there are two people who are at risk of developing cancer out of 10,000 people who consume shallots, and there is one child who may be at risk of developing cancer from 1,000 children who consume shallots.

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CONCLUSION

The results obtained in this study regarding the concentration of Cd and Pb metals in shallots are beyond the quality standards set by BPOM. However, the Pb and Cd metals in shallots are still below the quality standard set by Alloway. The hazard index indicates that children are more at risk of being exposed to heavy metals compared to adults in consuming shallots. The continuous use of chemical pesticides and chemical fertilizers in shallot farming must be controlled so as not to further increase the accumulation of heavy metals in agricultural products and land. The use of botanical pesticides can be a shortcut for farmers to reduce the accumulation of heavy metals on land and agricultural products.

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