Feed and Water Management May Influence the Heavy Metal Contamination in Domestic Ducks from Central Java, Indonesia



R. Susanti • Karima Widiyastuti • Ari Yuniastuti • Fidia Fibriana

Received: 13 December 2019 / Accepted: 26 March 2020 / Published online: 11 April 2020 © Springer Nature Switzerland AG 2020

Abstract Heavy metal contamination comes from the air, soil, and water, which affect the animals, plants, and humans in the food chain loop. Duck meat is one of the protein sources for humans and can be used as an indicator of heavy metal pollution. In this research, cross-sectional analytic research was carried out to analyze heavy metal contamination in duck meat. The water, feed, and duck meat (thigh and chest) samples from in intensive duck farming in Central Java, Indonesia, were taken randomly. The metal contents were measured and analyzed using Inductively Coupled Plasma (ICP-OES), followed by the manufacturer's procedure. Six tolerable heavy metals (Ag, Al. Cr. Cu. Ni, Sr) were found in water, feed, and duck meat which were still under the tolerable threshold with the highest concentration was Al and Cu, at 0.98 ± 0.025 ppm and 0.68 ± 0.117 ppm, respectively. The essential heavy metals were found in duck meat, including Fe and Zn. The dangerous heavy metal Hg contamination was found in Semarang duck meat $(5.54 \pm 0.01 \text{ ppm})$, followed by Magelang $(4.84 \pm 0.00 \text{ ppm})$ and Salatiga $(4.56 \pm$ 0.00 ppm). All water samples from five locations were contaminated with As, Hg, Cd, and Pb, which exceeded

R. Susanti (🖂) · K. Widiyastuti · A. Yuniastuti Department of Biology, Faculty of Mathematics and Natural Sciences, Universitas Negeri Semarang, Kampus Sekaran Gunungpati, Semarang 50229, Indonesia e-mail: r.susanti@mail.unnes.ac.id

F. Fibriana

the national standard. The presence of Hg in meat is positively correlated with the concentration of Hg in water (p = 0.002) and feed (p = 0.02) and influences simultaneously $(R^2 = 0.788; p < 0.05)$. The best possibility of heavy metal contamination in duck meat is the accumulation of water and feed consumption. Therefore, there is a need for special attention from the farmers on the duck feed for achieving meat safety as an effort to maintain food security.

Keywords Duck meat · Heavy metal contamination · ICP-OES

1 Introduction

In Asian countries, the number of consumers who prefer to choose meat and eggs from duck is increased rapidly due to its tasty flavor. Also, duck meat and eggs have high amino acids and fatty acids value. Public interest in duck meat has increased rapidly in the last 10 years and has widely opened up for business opportunities (Faten et al. 2014). Besides, culinary tourism, lifestyle, and the existence of social media is incredibly affecting duck meat production (Delgado et al. 2017; Petrovan and Leu 2017). Based on FAO (2017) data, Asian countries alone account for 84.2% of total duck meat produced in the world. These recent years, China holds the world record as the highest producer of duck meat, reaching almost 3,000,000 t per year. Myanmar is the highest producer of duck meat in the South East Asia region, reaching 150,000 t per year (Biswas et al. 2019). In Indonesia, duck plays a crucial

Department of Integrated Science, Faculty of Mathematics and Natural Sciences, Universitas Negeri Semarang, Kampus Sekaran Gunungpati, Semarang 50229, Indonesia

role in supplying animal-based protein. In 2017, poultry meat production reached 2.1 t, which are dominated by mallard and muscovy (Sumarmono 2019). Recorded in 2018, the domestic duck population in Central Java reaches more than 5 million (BPS 2018).

The increase in trends and lifestyles of duck meat consumption has not been balanced yet by the food safety concern, especially in developing countries. Even though the duck exhibits superiority in their immune systems, especially against disease over the other poultry animals, they are mainly live in a wet area, which is prone to heavy metal contamination (Sumarmono 2019). This phenomenon is closely related to the small-scale duck farmers who are constrained by inadequate appropriate technologies. They raise their duck in a traditional system of herding and scavenging, and sometimes it is an activity in between their crop farming activities for additional income. Therefore, the quality of duck meat has not been reached the standard that is acceptable for the global market (Biswas et al. 2019).

Related to food safety concerns, one of the health problems that threaten the community is heavy metal contamination as a result of industrial activities. Heavy metals, for example, mercury (Hg), lead (Pb), cadmium (Cd), lead (Sn), and arsenic (As) in waters are persistent, can be bioaccumulated, and toxic to the environment (Islam et al. 2015; Budijono et al. 2017). Today, heavy metal contamination has not only been found in the environment but has also entered livestock meat products as a result of the interaction in the food chain. Most livestock such as cows, goats, chickens, broilers, pigeons, and ducks have been contaminated with a high amount of trace elements and or heavy metals. As a top consumer in the ecosystem, heavy metals accumulation can have adverse effects on human health (Saha et al. 2016: Kalisińska 2019).

In laboratory testing, Cd has been proved to cause significant problems in poultry and waterfowl raising. It can cause decreased food intake, decreased growth rate and muscle mass, thin duck eggshells, reduced egg production, kidney toxicity, altered avoidance behavior, disrupted calcium metabolism, and metallothionein induction. The female ducks can accumulate heavy metals and release them in their eggs (Wayland et al. 2005). Also, various types of organs in Egyptian ducks exhibit high arsenic (As) contamination (Elsharawy 2015). The bioaccumulation of mercury (Hg), in the form of organic or methylated in the ecosystem, is linked to high toxicity risk and adverse effects on wildlife and human. It is including their behavioral, immunological, and reproductive systems (Ackerman et al. 2016). Hg toxicity causes sufferers to experience tremors and decreased hearing function, vision, and even memory. It is also a global concern since methylmercury (MeHg) biomagnifies through food chains and food webs (Driscoll et al. 2013; Lavoie et al. 2014). Duck lives in the aquatic and terrestrial ecosystem, which are susceptible to heavy metals contamination from water, feed, and soil in their environment. In the traditional farming system, duck is prone to heavy metals contamination, biomagnification, and bioaccumulation. This issue is related to a particular form of a farmhouse with no outflow. Aendo et al. (2020) found that the high concentration of Pb and Cd in duck meat raised in large-scale duck farms may incur the health risks of people who consume the duck in long-term of the period in Thailand.

The typical duck farming system in Indonesia or other developing countries is divided into two systems, i.e., free-grazing duck farming and backyard or house duck farming. The backyard duck raising system in a close system has a house and pond inside the farmhouse. Therefore, the risk of heavy metals bioaccumulation is high in this close farmhouse with no outflow. The bioaccumulation has the potential to negatively affect the duck in traditional breeding and farming systems since mostly they have a close system with no outflow (Sando et al. 2007). The duck receives the water for drinks and wallows from nearby rivers or ponds which are flow to the cage by self-made water pipe systems. The daily feed of ducks is often mixed with vegetable wastes or fish and shrimp products waste. Therefore, there are many potential sources of heavy metals contamination in duck-based products. There are no comprehensive data on the heavy metals contamination in duck meat in Indonesia. Based on this issue, it is crucial to explore heavy metals elements in the duck meat, feed, and water of duck farming to find the most potent source of heavy metals contamination in local duck meat, especially in the Central Java region. The results of this research could be used as a reference for the farmers as well as the stakeholders to give more concern on duck farming system for better food safety in the future, not only in Central Java, Indonesia but also in all traditional duck farming systems in the developing countries in the world.

2 Materials and Methods

2.1 Sample Collection

This study was an exploratory observational, involving duck populations from five intensive farms in Central Java, Indonesia. A total of 25 duck samples were collected from five different locations, namely Semarang and Pati in the coastal lowlands; Salatiga and Magelang, as a representation of inland lowlands and Temanggung to represent mountain highlands as presented in Table 1. Livestock locations were varied to represent the topographical conditions in Central Java, which were considered as one of the most abundant duck farmings.

Duck samples were local varieties, 18-months-old female ducks, and sampling method was conducted randomly. The collected water, feed, and duck meat samples were analyzed by performing Inductively Coupled Plasma (ICP-OES) Optima 8300 machine following the manufacturer procedure (Naschan et al. 2017). The results of the analysis were then compared with the concentration of the predetermined threshold of some heavy metal groups, which are non-essential and harmful to the body (Table 2).

2.2 Sample Preparation

2.2.1 Water Sample Preparation

Water samples were collected from duck drinking water. A clean and sterile bottle was used to collect 1500 mL of water from each farmhouse. The water samples were then brought to the laboratory for further analysis. The pH of 100 mL water sample was adjusted to 2.0 by adding 65% HNO₃, and then the distilled water was added to the sample until the volume reached 200 mL.

2.2.2 Feed Sample Preparation

Feed samples were collected by taking 1500 g from each location to the laboratory. It was then oven-dried at 70 °C for more than 48 h. After the drying process, the samples were then ground to get fine feed powder by a blender machine. Subsequently, 6 g feed powder was added with 2 mL 65% HNO₃ until the solution was immersed. The feed solution was then stirred until to get a homogeneous solution. Then, the feed solution was dried using a furnace with a temperature of 500 °C for 2 h. The feed ash was then dissolved in

50 mL distilled water and was further used for metal detection.

2.2.3 Meat Sample Preparation

Duck samples were randomly picked from each farm and were then slaughtered. The duck meat was collected from the thighs and thorax part. It was cut into 2 cm \times 2 cm \times 3 cm size and put into sterile Petri plates to measure the weight. The meats were dried in an oven at 70 °C for more than 72 h. After drying, 6 g of meat sample was added with 2 mL of 65% HNO₃ and was mixed homogenously. The meat samples were dried using a furnace at 500 °C for 2 h. The meat ash was then dissolved with distilled water up to 50 mL before heavy metal analysis.

2.2.4 Data Analysis

The data collected were analyzed using SPSS 22.0. The tests included multivariate and correlation tests; the data were then presented with Ms. Excel.

3 Results

Based on the results of the heavy metal analysis, the metal content of each sample was varied, as shown in Figs. 1, 2, and 3. The results of detected heavy metals in this study were classified into three major groups, i.e., (1) tolerable heavy metals, (2) essential metals, and (3) dangerous heavy metals. Tolerable heavy metals are the mineral traces found in duck's body, and it could be toxic if it is found in high concentration. In this research, this group consists of six heavy metals (Ag, Al, Cr, Cu, Ni, Sr) which their concentrations are under the maximum threshold that can be received by the duck. Essential metals are metals that are needed by the body to carry out the physiological activities of the body. Essential metals in the body include Co that is needed by the body as a constituent of vitamin B12, Fe as the core of hemoglobin and is involved in binding oxygen, Mn, Se, and Zn act as specific enzyme cofactors, especially for enzymes involved in metabolism and immunity (Reis et al. 2010). Hazardous heavy metals are toxic heavy metals that have been determined nationally and globally. The standards have determined the maximum limit for the existence of heavy metals in force in each country.

Sampling location	Farmhouse building characteristics	Distance to human settlement (m)	Source of feed	Source of water
Semarang	Bamboo building with a dirt floor	± 500	Fish and shrimp product waste	Well
Pati	Bamboo building with a dirt floor	± 1000	Concentrate feed, bran, fish product waste	Well
Salatiga	Bamboo building with a raw earth floor	± 3000	Concentrate feed, bran	Well
Magelang	Bamboo building with a raw earth floor	±5000	Concentrate feed, bran, cabbage waste	Fish ponds
Temanggung	Bamboo building with a raw earth floor	±5000	Concentrate feed and bran	Ponds

Table 1 Characteristics of the farmhouse in five different sampling locations

The tolerable heavy metals are heavy metals that allow contamination in low concentration, as can be seen in Fig. 1.

Concentrations of hazardous heavy metals such as Al and Cu were most commonly found in feed, with concentrations reaching 0.98 ± 0.025 ppm and 0.68 ± 0.117 ppm, respectively. However, Cu metal was not only found in duck feed but also found in water, which reached 0.97 ± 0.019 ppm. Metal concentrations exceeding the threshold were also found in duck meat, which reached 0.86 ± 0.256 for Cu, and 0.51 ± 0.512 for Al. The high metal content in meat is probably due to the high rate of biotransformation and bioaccumulation of metals in the duck body.

The essential metals (trace minerals) such as Co, Fe, Mn, Se, and Zn were found in feed, water, and meat samples. For certain types of metals, iron (Fe) and Zinc (Zn) were detected more abundantly in the meat than in feed and water, as shown in Fig. 2. Those metals were found in meat in quite high concentrations. However, the concentration of those two metals was safe. Both metals are needed to carry out the body's metabolic functions. The concentration of iron in the meat or muscle reached 2.63 ± 0.467 ppm, while Zn reached 1.41 ± 0.897 ppm. It is in line with the physiological role of the two minerals. Therefore, the presence of these metals plays a role in metabolism as an electron supplier and receiver. It would be very natural to find high metal concentrations in meat.

Accumulation of dangerous heavy metals, mercury (Hg), was found in duck meat exceeding the safe threshold, i.e., 4.42 ± 0.845 ppm, followed by water, which was 2.23 ± 0.264 ppm and feed which was 3.31 ± 0.173 ppm (Fig. 3).

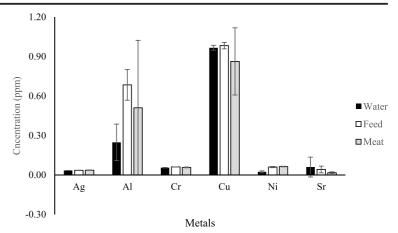
The high accumulation of heavy metals in muscle tissue or meat is probably caused by a lack of a mechanism in the duck body to transform and excrete several types of metal; instead, it accumulates in various tissues and organs. However, some essential metals that accumulate in the duck tissues because of duck need them as the activity regulator—for example, Fe to carry oxygen or Zn as a cofactor of the enzyme. Therefore, the high input of heavy metals, which is not excreted, is accumulated in the tissues and possibly has an impact on the reduced nutritional value for meat consumption.

Broadly, the three primary heavy metal contaminants in duck meat in all sample locations were beyond the threshold. Significantly (p < 0.05), the highest Hg

Metals	Food and agric	culture organization (p	pm)	Indonesia Nat	tional Standard (ppr	1)
	Water	Feed	Meat	Water	Feed	Meat
Hg	0.001	0.1	0.1	0.001	0.05	0.03
Pb	0.01	0.3	0.1	0.01	0.3	0.3
Cd	0.003	0.5	0.5	0.003	0.1	0.3
As	0.01	0.5	0.5	0.05	0.5	0.5

Table 2 The heavy metal threshold in a water environment, duck's feed, and duck meat, according to FAO (2017) and SNI (2009)

Fig. 1 Tolerable heavy metals concentration in water, feed, and duck meat



content was found in Semarang, while the lowest was in Temanggung. For contamination of Al, the highest was found in Magelang and the lowest in Salatiga. However, different results are shown by Cu concentration, that there was no significant difference between heavy metal concentrations in various cities Fig 4.

4 Discussion

Each sample location has various characteristics; for instance, duck farms in Semarang are located in coastal areas bordering the electric power industry and shipyards. Besides, the condition of the northern waters of the island of Java has experienced heavy pollution by heavy metals from upstream. The coast of Semarang is the biggest producer of animal feed made industrial fish waste from the Java sea. As for information and interestingly, the duck feed was produced from crustaceans, mollusk, and small fish from the contaminated ocean

Fig. 2 Essential metals concentration in water, feed, and duck meat

that possibly being one of the primary sources of heavy metals contamination. Crustaceans such as shrimp are one of the main ingredients in making the fish and duck feed to increase their productivity. It would be possible to allow contamination in the water of northern Java to accumulate heavy metals in crustaceans such as shrimps, which then enter the food chain and food webs. The calculation of this contamination could be performed by the method of Aendo et al. (2020). Furthermore, the distribution of this feed is wide to other areas. In this research, the duck samples were collected from the intensive farming system with intensive treatments of a close system. Several duck farmings are located next to community settlements with polluted air from factories. The other heavy metal sources are contaminated water near the duck farmings that are usually in the form of small rivers or small ponds. However, the source of duck drinking water pollution was not identified in this study. However, the best possibility of contamination for each region may differ based on the water

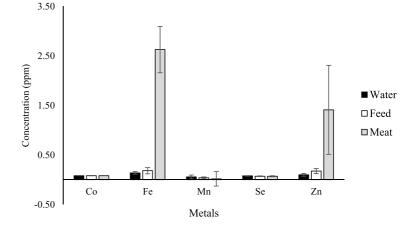
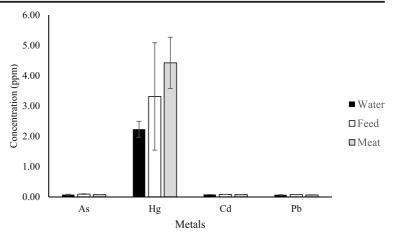


Fig. 3 Dangerous heavy metals concentration in water, feed, and duck meat

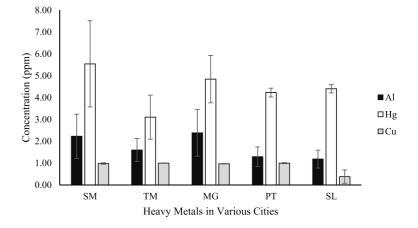


obtained for duck farming systems. In Pati and Semarang, the duck farming locations are crossed by several rivers, where along with the flow from upstream to downstream, there are several factories that produce waste (Tjahjono et al. 2018). In Temanggung, Magelang, and Salatiga, the duck farming sites are dominated by rice fields that use a lot of pesticides, fertilizers, and chemicals to increase crop productivity. Some chemicals used have a heavy metal content which then pollutes livestock water sources (Dewi et al. 2016). In addition, the equation of the five regencies and cities is that each sampling location is close to a highway and crowded with motor vehicles.

All types of metals could be eliminated from the body. However, the results of this research indicate that the rate of bioaccumulation is faster than the elimination of heavy metals, or the elimination rate was least than 50% as in Ag, Co, Hg, and Ni. Some types of heavy metals were also accumulated to more than 100% of the total inputs, such as Fe and Zn (Table 3). The

mechanism of absorption and bioaccumulation is possibly influenced by internal factors of duck physiology, and by interactions among heavy metals. The results of correlation analysis between concentrations of heavy metals indicate that other metals influence the concentration of certain metals. Interactions that arise either positively, which means the metal is equally increasing or a negative interaction, which means an increased concentration will affect or decrease other metals. Fe concentration in meat has a positive correlation with Ni and Zn content and has a negative correlation with As, Cd, and Sr, and vice versa (Table 4). Hg concentration was significantly positively correlated with Al, Se, and Zn and negatively correlated with Cd and Sr (Table 4). However, there is a limitation in this research in which the mapping of the source of contamination has not been done. However, there is a possibility that metal contamination in the highlands and rice fields originated from the use of excess fertilizer (Pradika et al. 2019). Accumulation of heavy metals in crustaceans depends on

Fig. 4 Three dominant heavy metals found in duck meat in five different locations (SM: Semarang; TM: Temanggung; MG: Magelang; PT: Pati; SL: Salatiga)



environmental water conditions, both physical and chemical (salinity, pH, and temperature). Komari and Utami (2013) added that the nature of crustaceans foraging at the bottom of the water body resulted in crustaceans to be contaminated with heavy metals. The presence of residues in meat indicates the presence of heavy metal contamination from the feed, drinking water, and soil. Polluted feed, drinking water, and soil that enters the duck body could be deposited in duck organs. Hg, Pb, and Cd are dangerous substances, where both Pb and Cd can contaminate duck through the air. Cd and its oxide compounds are most commonly found in the air and accumulate in animal tissue or are excreted in milk or eggs at a certain level that exceeds the limit of contaminants in food.

Heavy metals are pollutants that harm human health (as a final consumer) for an extended period accumulation, because their continued effects (delayed effect) the nervous system, cause mutations in genes, inhibit cell metabolism and increase cancer growth probability (Matović et al. 2015). With the increased consumption of duck meat, various ways of maintaining patterns, and environmental conditions that have been contaminated, it is estimated that heavy metals can enter and accumulate into the body of the duck. According to Darmono in Asnawi et al. (2015) reported that some metals are essential for living things, including Ca and Mg metals, which are useful for the formation of scales and cuticles in fish and shrimp. Fe, Cu, Zn, and Mn are useful in the formation of hemocyanin in the blood and as an enzymatic system in aquatic animals. Moreover, several heavy metals microelements have no biological function and cause toxicity to live organisms, including Pb, Hg, As, and Cd. Based on the significance value which shows that there is a positive correlation between meat with water and feed on the Cd, the correlation is 0.00. Cd retention and mobility depend on pH, temperature, ionic strength, cation exchange capacity, surface area, and concentration of the ligand complex (Galunin et al. 2014).

Manulang et al. (2014) added that heavy metals that are dissolved in water bodies naturally form free ions, inorganic, and organic pair ions. Arsenic and cadmium are included in the B3 waste category based on their toxicity. According to Government Regulation No.18 (1999) concerning Management of Hazardous and Toxic Waste Material article 1, it states that the impact caused by As and Cd can damage the environment and human health; therefore, it is categorized as B3 waste. The latest regulations governing B3 are PP No. 74 of 2001 concerning the management of hazardous and toxic materials. The quality standard of heavy metal As in drinking water for human consumption is 0.01 ppm.

Lead (Pb) is an environmental contaminant that occurs naturally and mostly comes from anthropogenic activities such as motor vehicle emissions. Pb is a metal that occurs in organic and inorganic forms that dominate the environment. Food is one of the primary sources of Pb exposure, followed by air (especially lead dust from gasoline) and drinking water. Vegetable food can be contaminated with Pb through absorption from the surrounding air and soil (Ali et al. 2013). In humans, the consumption of Pb arises from contaminated plants or animal products. Another source of Pb consumption is through the use of tin-based vessels or lead-based glaze. After being in the bloodstream, Pb is distributed among blood, soft tissue, and mineralized tissue (Ming-Ho 2005). Children are susceptible to Pb because of its critical effects on the nervous system, which can interfere with growth (Castro-González and Méndez-Armenta 2008). Today the release of Pb into the atmosphere increases sharply due to the burning of oil and gas. Pb can cause neurotoxic, nephrotoxic, and anemia effects in animals and humans. Pb is widely used in the non-food industry, and most causes poisoning effects on living things.

Mercury (Hg) or mercury is a metal that already exists naturally, is the only metal that is at room temperature in the form of liquid. The pure metal is silvery/ grayish-white, odorless liquid, and glossy. Humans use mercury oxide (HgO) and mercury sulfide (HgS) as dyes and cosmetics (whitening creams). Also, it is used in fluorescent lighting, batteries, thermometers, industrial paints, gold smelting, insect repellent, and others.

Food contamination can occur at any point in the food chain. Therefore, food safety must be a shared responsibility to avoid various diseases. Some heavy metals are an essential element that is needed by every living thing, but at certain levels, it can be toxic (Juniawan et al. 2016). Long-term consumption of foods containing heavy metals can cause health problems through the process of bioaccumulation. Bioaccumulation means an increase in the concentration of chemicals in biological organisms over time (Martin and Griswold 2009). Bioaccumulation is a tiered transfer (tropic transfer) of

Metals	Concentration (ppm)			
	Input	Meat	Balance	%
Ag	0.067 ± 0.00	0.034 ± 0.00	-0.033 ± 0.00	-48.50 ± 0.21
Al	0.931 ± 0.26	1.734 ± 0.54	0.803 ± 0.77	86.27 ± 5.89
As	0.165 ± 0.26	0.074 ± 0.00	-0.091 ± 0.02	-54.98 ± 6.26
Cd	0.151 ± 0.00	0.074 ± 0.00	-0.077 ± 0.00	-50.93 ± 0.15
Co‡	0.164 ± 0.00	0.082 ± 0.00	-0.082 ± 0.00	-49.97 ± 0.58
Cr	0.115 ± 0.00	0.056 ± 0.00	-0.058 ± 0.00	-50.96 ± 2.20
Cu	1.947 ± 0.02	0.862 ± 0.27	-1.086 ± 0.27	-55.75 ± 13.87
Fe‡	0.318 ± 0.07	2.626 ± 0.49	2.308 ± 0.43	724.68 ± 95.08
Hg	5.545 ± 2.00	4.422 ± 0.89	-1.123 ± 1.08	-34.85 ± 20.21
Mn‡	0.095 ± 0.04	0.018 ± 0.01	-0.077 ± 0.04	79.42 ± 20.73
Ni	0.082 ± 0.01	0.061 ± 0.00	-0.022 ± 0.01	-24.72 ± 13.82
Pb	0.141 ± 0.00	0.062 ± 0.01	-0.079 ± 0.01	-55.65 ± 7.37
Se‡	0.147 ± 0.01	0.071 ± 0.01	-0.076 ± 0.02	-51.41 ± 11.43
Sr	0.099 ± 0.06	0.016 ± 0.00	-0.084 ± 0.06	-79.64 ± 13.51
Zn‡	0.272 ± 0.08	1.409 ± 0.95	1.137 ± 0.92	434.57 ± 299.97

Table 3 Heavy metal accumulation from the environment to duck meat

Note: Negative mark shows total residue that possibly eliminated from duck's body. Positive mark indicates existed deposit or possibly contaminated from the environment. Percentage (%) is calculated from metals concentration in duck's meat divided by metals input. ‡ mark representing essential metals for metabolism

contaminants between species. It occurs in lipophilic conditions (chemicals that accumulate in fat) and move to other tropic levels. The heavy metals in poultry will increase over time because it is bioaccumulative. The heavy metal accumulates more quickly than its elimination and excretion by metabolism (Ming-Ho 2005; Martin and Griswold 2009); therefore, duck meat can be used as an indicator of heavy metal pollution in humans.

5 Conclusion

Based on the results of the study, three categories of heavy metals contamination were found, i.e., (1) tolerable heavy metals, (2) essential metals, and (3) dangerous heavy metals. Six tolerable heavy metals (Ag, Al, Cr, Cu, Ni, Sr) were found in water, feed, and duck meat which were still under tolerable threshold with the highest concentration was Al and Cu. The essential heavy metals were found in duck meat including Fe and Zn. The highest Hg contamination was found in Semarang duck meat (5.54 ± 0.01 ppm), followed by Magelang (4.84 ± 0.00 ppm) and Salatiga (4.56 ± 0.00 ppm). All water samples from five locations were contaminated with As, Hg, Cd, and Pb which exceeded the national standard. The presence of Hg in meat is positively correlated with the concentration of Hg in water (p = 0.002) and feed (p = 0.02) and influences simultaneously ($R^2 = 0.788$; p < 0.05). The best possibility of the heavy metal contamination in duck meat is the accumulation from water and feed consumption. Therefore, there is a need for special attention from the farmers on the duck feed for achieving meat safety as an effort to maintain food security.

Acknowledgments The authors would like to thank the Directorate General of Research and Development, Strengthening the Ministry of Research, Technology, and Higher Education for the research funding support through National Competitive Fundamental Research Grant Fiscal Year 2019 No. 192/SP2H/LT/ DRPM/2019, 11 March 2019.

Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

Metal	Pears	Pearson's correlation	ation												
	Ag	Al	As	Cd	Co‡	Cr	Cu	Fe‡	Hg	Mn‡	Ni	Pb	Se‡	Sr	Zn‡
Ag		- 0.339	-0.438	-0.205	- 0.232	-0.635^{*}	- 0.655**	0.283^{**}	0.065^{*}	0.107^{**}	0.194^{**}	0.104^{**}	0.128^{**}	0.012**	-0.073^{**}
Al			0.017	0.660^*	0.000	0.698^*	0.534^{**}	-0.512^{**}	0.567^{*}	-0.758^{**}	1	0.431^{**}		-0.283^{**}	0.367^{**}
As				0.397	-0.027	0.477^{*}	0.581^{**}	-0.674^{**}	-0.577^{*}	0.138^{**}				0.530^{**}	1
Cd					0.074	0.336^{*}	0.212^{**}	-0.843**	0.260^{*}	-0.464^{**}				0.126^{**}	
Co‡						0.000^{*}	0.000^{**}	0.000^{**}	0.000^{*}	-0.001^{**}		0.004^{**}	0.000^{**}	-0.003^{**}	0.000^{**}
Cr							0.976^{**}	-0.388^{**}	0.164^*	-0.287^{**}	-0.398^{**}	-0.092^{**}	0.069^{**}	-0.216^{**}	0.287^{**}
Cu								-0.345^{**}	-0.017^{*}	-0.153^{**}		-0.189^{**}	-0.101^{**}	-0.116^{**}	0.212^{**}
Fe‡									0.236^{*}	0.540^{**}		-0.444^{**}	0.390^{**}	-0.570^{**}	0.560^{**}
Hg										-0.180^{**}	-0.448^{**}		0.889^{**}	-0.870^{**}	0.567^{**}
Mn‡												-0.911^{**}		-0.204^{**}	-0.260^{**}
ïZ												-0.029^{**}		0.143^{**}	0.317^{**}
Pb														0.443^{**}	0.091^{**}
Se‡														-0.773^{**}	0.816^{**}
Sr															-0.683^{**}
Zn‡															

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