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Heavy metal contamination status in the soil-water-rice system near coal-fired power plants in Cilacap, Indonesia

Status kontaminasi logam berat dalam sistem tanah-air-padi di dekat pembangkit listrik tenaga uap batu bara di Cilacap, Indonesia

Febbyandi Isnanda Pandiangan[1*], Kholis Abdurachim Audah[2]

[1]Department of Food Technology, Faculty of Life Sciences and Technology, Swiss German University, Tangerang, Indonesia

[2]Department of Biomedical Engineering, Fakultas Life Sciences and Technology, Swiss German University, Tangerang, Indonesia

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ABSTRACT

Indonesia is intensively developing plenty of coal-fired power plants to support electricity demand growth. Some research showed the utilization of coal as an electrical energy source may produce anthropogenic contaminants (ACs) that can accumulate in plants and environmental compartments, such as water, soil, and air. This research was carried out to investigate the severity of the heavy metals contamination problem, for instance, arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) in soil, water, rice, and rice bran using ICP-MS and ICP-OES. This research demonstrated the metal contamination levels of arsenic, cadmium, mercury, and lead in the soil, water, rice, and rice bran were below the maximum limit, according to SNI, Codex, and FAO/WHO. However, only a rice sample showed the lead (Pb) level above the safe limit. Coal-fired power plant activities in Cilacap did not indicate clear evidence of soil, water, rice, and bran heavy metal contamination. Therefore, transformation to green energy (e.g., solar and geothermal) is highly recommended to minimize the potential health risks of environmental pollution due to the coal-fired power plant's by-product activities.

ABSTRAK

Indonesia saat ini sedang gencar mengembangkan banyak pembangkit listrik tenaga batu bara (PLTU) untuk mendukung pertumbuhan kebutuhan listrik. Beberapa penelitian menunjukkan pemanfaatan batubara sebagai sumber energi listrik dapat mengakibatkan pencemaran antropogenik yang dapat terakumulasi pada tanaman dan kompartemen lingkungan, seperti air, tanah, dan udara. Penelitian ini dilakukan untuk mengetahui besarnya masalah pencemaran logam berat antara lain arsenik (As), cadmium (Cd), merkuri (Hg), dan timbal (Pb) pada tanah pertanian, air, beras dan kulit ari beras menggunakan ICP-MS dan ICP-OES. Penelitian ini menunjukkan tingkat pencemaran logam arsenik, kadmium, merkuri, dan timbal dalam tanah, air, beras dan kulit ari beras berada di bawah batas maksimum menurut SNI, Codex, dan FAO/WHO. Namun, hanya satu sampel beras yang menunjukkan kadar timbal (Pb) di atas batas aman. Kegiatan PLTU di Cilacap belum menunjukkan bukti yang jelas adanya pencemaran tanah, air, beras dan kulit ari beras. Transformasi ke energi hijau, seperti energi dari tenaga surya dan panas bumi sangat dianjurkan untuk meminimalkan risiko pencemaran lingkungan akibat kegiatan produk sampingan PLTU.

**Corresponding author*
E-mail: febbyandi.pandiangan@sgu.ac.id

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INTRODUCTION

Rice is one of the essential carbohydrate sources, which acts as a staple food in some countries for more than half of the global population worldwide (Khir & Pan, 2019). Indonesia was the third largest rice producer in 2017, with more than 81 million tonnes, after China (1st) and India (2nd) (FAOSTAT, 2019). From the aspect of nutrition, rice provides around half of the recommended dietary caloric intake daily. Therefore, rice is one of the vital protein sources, particularly for the poor-income population in Asia (Muthayya et al., 2014). In Indonesia, Cilacap is readily known as a rice-producing city. In 2018, Cilacap experienced a surplus of rice production of more than 300,000 tons (Dony, 2019)

At the same time, rice also may accumulate harmful levels of toxic substances. Recently, there has been an increase in carcinogenic substances and harmful pollutants such as heavy metals contamination in paddy rice. Consequently, it can lead to human health risks via the food chain (Mao et al., 2019). The metal contaminants that are potentially contaminating rice are arsenic (As), cadmium (Cd), mercury (Hg), and lead (Pb) (Shraim, 2017). In addition, one primary source of environmental pollutants such as toxic trace metals and gasses in the environment is a result of coal-fired thermal power plants (*Pembangkit Listrik Tenaga Uap* -PLTU) activities (Rai et al., 2019).

When the coal gets combusted as the result of coal-fired power plant activity to generate electricity, trace elements bound in it are spread through the atmosphere in the form of bottom ash, fly ash, and flue gas, where Cd and Pb are highly detected in the bottom ash, As is highly detected in fly ash and Hg is highly detected in the flue gas. Hg is highly detected in the flue gas because it is more volatile than others. The compositions of chemicals contained in bottom ash, fly ash, and flue gas from coal combustion are 12-18% of Cd, 70-80% of Hg, and traces of As, Fe, Pb, and Cr (Reddy et al., 2005) which are known as anthropogenic contaminants (ACs). Hazardous chemicals, pollution, and particulate matter as the result of coal combustion to generate electricity will subsequently disperse over large areas and are most likely to contaminate the environment (Munawer, 2018). Those hazardous chemicals might contaminate the plants and paddy plants grown near the coal-fired power plants.

The water used to wash the impurities coals may contaminate nearby water sources such as groundwater, rivers, and lakes, which will affect the life of aquatic flora and fauna (Finkelman et al., 2021). The emission of fly ash residues and pollutants, as the result of coal combustion, will contaminate the soil. Therefore, it harms agricultural activities besides spreading through the atmosphere (Guttikunda & Jawahar, 2014). Rice has the ability to bioaccumulate metal contaminants. Rice is planted under flooded plants with extensive irrigation water, an agricultural practice that may lead to the accumulation of As and other harmful metals in rice crops when irrigating water or the soil is already polluted (Mao et al., 2019).

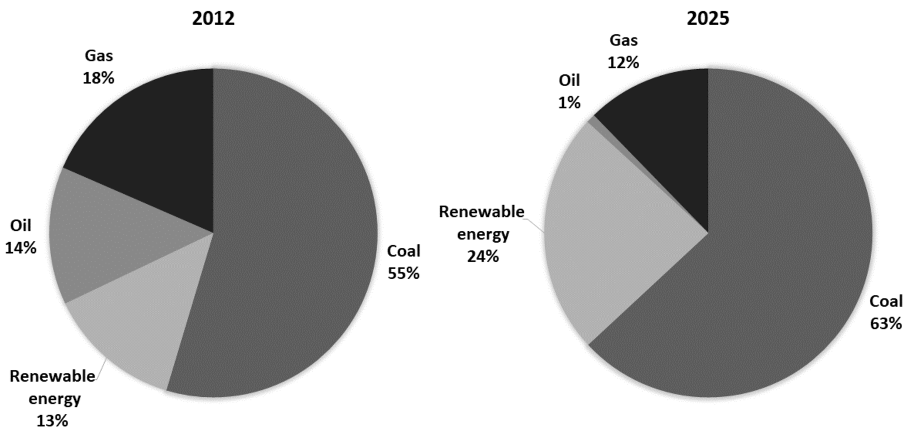

Figure 1. Indonesia's energy source composition for electricity generation

Heavy metal poisonings may happen through water, air, and food exposure. These heavy metals bioaccumulate in the body and have various harmful effects on different human tissues and organs. Heavy metals are able to disrupt cellular activities such as cell growth, differentiation, proliferation, apoptosis, and damage-repair mechanisms. Other metal toxicity effects include the generation of reactive oxygen species (ROS), weakened antioxidant defense, enzyme inactivation, oxidative stress, and even genome instability that eventually leads to a carcinogen (Balali-Mood et al., 2021). Inorganic arsenic is a chronic, non-threshold carcinogen (Signes-Pastor et al., 2016).

Indonesia has many PLTU with various power capacities. Cilacap is the potential city where several PLTU are projected to be built. PLTU Karangkandri is one of the coal-power plants with the most prominent electricity capacity in Indonesia, operating since 2006. Indonesia is targeting to provide an electric energy capacity of 35,000 MW, and of March 2018, the project's construction phase reached 48%, equivalent to 16,994 MW (Suprateka, 2018). Figure 1 shows Indonesia's electricity power generation, in which coal as a source composition is the first choice of fuel and accounts for up to 55% in 2012 and would only get more massive in the upcoming years, particularly on the projection 2025 (Rahardjo, 2014). Coal is one of the inexpensive energy sources with an abundant amount in Indonesia. An Indonesia economic energy price comparison study by Liun et al. (2014) reported that conventional energy sources, for instance, fossil fuels (coal), were still superior to being used for electricity generation with cheaper economic value compared to other energy sources. The electricity per unit of coal was $3.3p per kWh, which is more affordable than solar, wind, and biofuel, which accounted for $37.73p per kWh, $12p per kWh, and $22.2p per kWh, respectively. On the other hand, based on mathematical simulation modeling, the total economic value (TEV) as a result of a coal-fired power plant activity was Rp 18 billion per year, consisting of the burden of air emission costs and the potential losses on the agricultural side (Jenned & Dewi, 2017). In addition, coal-fired thermal power plants may cause high costs to human health and the environment (Munawer, 2018). However, the Indonesian government is planning to stop the current operation of the coal-fired power plants, starting in 2025, and replace them with more renewable energy power plants (Umah, 2021).

Several recent studies have further confirmed some effects of coal-fired power plant activity on heavy metal content in rice. In 2016, research was published about rice safety assessment around a thermal power plant in Shaoguan City, Guangdong Province, China, showing that the average Cd and Pb contents in rice grain had exceeded the maximum permissible limits according to regulatory food standards in China (Wang et al., 2016). Furthermore, a potential impact of Hg emission from the coal-fired power plant affects the accumulation of Hg in rice in the area of Hunan Province, China (Xu et al., 2017).

In Indonesia, there are currently limited published studies that specifically examine the effect of coal-fired power plant activity on heavy metal contamination in rice and soil-water compartments. However, several published studies were about the risk assessment of heavy metals in Indonesian rice and paddy plants affected by environmental pollution activities such as mining (Rauf et al., 2020) and agricultural activities in industrial areas (Astuti et al., 2021). A study carried out by Silalahi & Putra (2018) showed that some rice samples in Medan, Sumatera Utara were detected to have high As levels exceeding the World Health Organization (WHO) maximum limit in white and brown rice. Gold mining activities also had the potential to pollute the environment and contaminate rice with Hg levels exceeding the safe limit (Istikasari et al., 2017). Sukarjo et al. (2018) found that the concentration of Pb in irrigated rice exceeded the national and world (WHO) standards for food safety, which correlated with the emergence of cancer. In addition, using fertilizers, such as inorganic fertilizers, also contributes to soil pollution (Rauf et al., 2020) and, ultimately, Hg, Pb, and Cd translocation in rice and rice crops (Sukarjo et al., 2018).

With little scientific works of literature concerning the rice contamination from the coal-fired power plant in Indonesia, therefore, the study to investigate heavy metal (As, Cd, Hg, Pb) concentrations in rice, water, rice bran, and cultivated soils in a rural area near the coal-fired power plants in Cilacap is needed. Through this research, the community would understand the severity of heavy metal contamination in soil and water and heavy metal bioaccumulation in rice and rice bran. Moreover, the analysis and discussion presented in this study would convey valuable information for future research exploring the risk assessment and bioaccumulation of heavy metals in organisms.

MATERIALS AND METHODS

This research investigated four metals, arsenic, lead, mercury, and cadmium, to find the relationship between PLTU activities and environmental pollution. As was a metal that attracts most researchers because of its ability to bioaccumulate in all paddy plants, especially in grain, known as one of the staple foods in the world. Hg, Pb, and Cd also had potential adverse health effects if they were accumulated in the body for a long time or consumed in high dosage. Metal and metalloid concentrations were found to vary widely between soil, bran, water, and rice. Because the As species distribution is still under research and not fully understood, only total As concentrations were measured in this study.

Sampling

The samples were collected nearby PLTU Karangkandri (latitude -7.682091, longitude 109.095120) and PLTU Adipala (latitude -7.681936, longitude 109.136238) in Cilacap. The distance between the city center of Cilacap and both PLTU are only about 10 kilometers. The chosen sampling method was non-probability sampling due to the difficulties of applying random sampling because of the severe dry season at the end of 2019. For soil samples, the topsoil (horizon A) was collected from a cultivated paddy field by a small hoe and put into a zipper polyethylene bag, labelled, and stored in a cooler box. Paddy IR 64 as the rice and bran samples were collected at several rice milling facilities around the sampling sites and put into a zipper polyethylene bag, labeled, and stored in a cooler box. Water sampling was taken from the irrigation channel around the paddy field, put into the plastic bottle, labeled, and stored in a cooler box. This research investigated three rice samples (two polished white rice and one polished red rice), two irrigation water samples, four soil samples, and three rice bran samples, as shown in Figure 2. All samples were brought to Saraswanti Indo Genetech laboratory, Bogor, Jawa Barat, for heavy metals testing

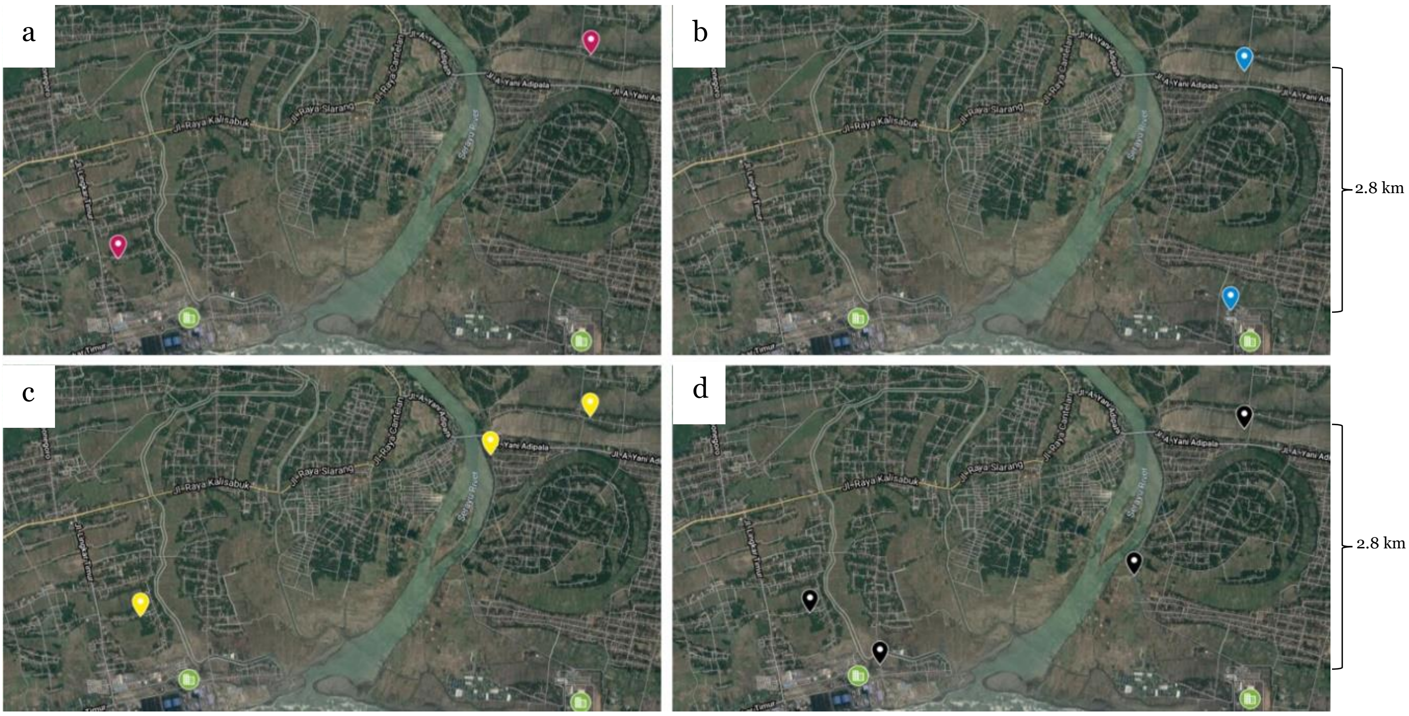

Figure 2. The sampling locations. (a) rice, (b) water, (c) bran, (d) soil, and the green circles indicating the coal-fired power plants (*Pembangkit Listrik Tenaga Uap* (PLTU) locations.

Sample preparation and metal analysis

In this study, ICP-MS and ICP-OES were chosen as an accurate analytical methods to reveal the presence of harmful heavy metals such as, As, Cd, Hg, and Pb (Dhanalakshmi & Gawdaman, 2013; Wilschefski & Baxter, 2019) in the samples. For the determination of heavy metals in rice, water, and bran, ICP-MS was used following the method 18-13-14/MU/SMM-SIG as explained by Carey et al. (2015) with some modifications. A 0.5–1.5 g solid sample or liquid sample was added to the vessel. A 10 ml concentrated nitric acid (HNO_3) was added and kept for 15 minutes. Microwave digested in an instrument with programs: ramp 150°C for 10 minutes and held 150°C for 15 minutes until the digestion step was complete, i.e., a clear solution was reached. The destruction results were cooled and put in a measuring flask of 50 ml. The vessel was rinsed with double distilled water quantitatively and set with the destruction results in a 50 ml volumetric flask. A 0.4 ml standard internal mixture of germanium (Ge), indium (In), bismuth (Bi), and rhodium (Rh) 10 mg/l was added and diluted with double distilled water to the mark, homogenized. The solution was filtered with a 0.20 μm RC/GHP filter. The intensity of the sample solution in the ICP-MS system was measured with As analyte with internal Ge standard, Hg and Pb analytes with internal Bi standard, and Cd analyte with In internal standard. This instrument's limit of detection (LOD) was 0.00035 μg/g, 0.0005 μg/g, and 0.0005 μg/g for Pb, Hg, and Cd, respectively.

ICP-OES was used following the method 18-13-1/MU/SMM-SIG as explained by Astuti et al. (2021) with some modifications for the determination of heavy metals in soil. The samples were digested as follows: a 1 g sample was poured into a 100 ml round-bottom flask. Then, 10 ml of concentrated nitric acid (HNO_3) was added to the sample and heated using microwave digestion with programs: ramp 150°C for 10 minutes and hold 150°C for 15 minutes until the digestion step was complete, i.e., a clear solution was reached. After that, the solution was transferred into a 50 ml volumetric flask. The internal standard 0.50 ml yttrium was added and filled with distilled water. Then, the solution was filtered using filter paper. The solution was prepared for ICP-OES analysis of heavy metals. The LOD of this instrument was 0.004 μg/g for Hg. All samples were tested at Saraswanti Indo Genetech laboratory (SIG), Bogor, Jawa Barat. Heavy metal contents in samples were reported in (μg/g) wet weight. For the Certified Reference Materials (CRMs), this research used the internal standards that were developed by the SIG laboratory.

RESULTS AND DISCUSSION

In Indonesia, the maximum limit of heavy metal contamination in certain foods, including rice, is regulated under Peraturan Menteri Pertanian Republik Indonesia No 53/Permentan/kr.040/12/2018 on the Safety and Quality of Fresh Food of Plant Origin. Globally, the European Food Safety Authority (EFSA) set the maximum heavy metal contaminants in rice for European countries. At the same time, Codex also sets the limit to protect people on a broader scale. However, Codex and EFSA did not regulate the Hg safety level in rice due to the low-risk Hg bioaccumulation level towards crops compared to seafood. The comparison of maximum heavy metal contaminants in polished rice is shown in Table 1. As contamination in paddy rice is an inevitable problem since its natural ability to absorb it through the roots subsequently distributes and accumulates As into all parts of the paddy plant, including the rice grain. Nevertheless, the level of As significantly depends on the variety of rice (Rahman et al., 2014), color, and grain size (Sun et al., 2008). The maximum limit of As in rice is regulated by EFSA and Food and Agriculture Organization (FAO) at 0.2 μg/g (EC, 2015).

Table 1. Maximum heavy metal contaminants in polished rice

	As (μg/g)	Cd (μg/g)	Hg (μg/g)	Pb (μg/g)
Indonesia	0.5[1]	0.4[1]	0.05[1]	0.3[1]
Codex	0.2[4]	0.4[4]	-	0.2[4]
EFSA	0.2[3]	0.2[1]	-	0.2[2]

[1]SNI (Indonesian National Standard) 7387:2009

[2]Commission Regulation (EC) No 1881/2006 of 19 December 2006

[3]Commission Regulation (EU) 2015/1006 of 25 June 2015

[4]General Standard for Contaminants and Toxins in Food and Feed 1995 (Codex Alimentarius, 1995)

The result of heavy metal content, e.g., As, Cd, Hg, and Pb, in soil, water, rice, and bran, is presented in Table 2. This research found that the total As in rice was below the maximum limit established by EFSA, FAO, and the Indonesian National Standard (SNI). SNI set the permissible metal level for polished rice as 0.4 µg/g, 0.05 µg/g, and 0.4 µg/g for Cd, Hg, and Pb, respectively (BSN, 2009), while Kementan set 0.4 µg/g and 0.2 µg/g for Cd and Pb (Kementan, 2018). Referring to SNI, Kementan, European Union (EU) standard, and Codex, it could be concluded that the two rice samples are safe to consume regardless of white or red rice. However, there was one rice sample above the Pb safe limit. The levels of Hg in the rice samples were below LOD, while research suggested the Hg emissions from coal-fired power plants could increase Hg levels in rice (Xu et al., 2017). However, the risk assessment of rice consumption was considered to be studied to know the potency of adverse health effects in a long-term accumulation. It was clearly understood that bran had a higher level of As than grain because of the defense mechanism in the paddy crops (Meharg & Zhao, 2012). For the Cd and As contamination levels, these results agreed with a similar observation on these parameters in a global rice grain study with Indonesian rice median was 0.21 µg/g and <0.20 µg/g, respectively for Cd and As (Carey et al., 2020; Shi et al., 2020).

Table 2. Level of As, Cd, Hg, and Pb in soil, water, rice bran, and rice near the coal-fired power plants (PLTU) in Cilacap.

Sample	As (µg/g)	Cd (µg/g)	Hg (µg/g)	Pb (µg/g)
Soil 1	3.33	0.29	ND	2.01
Soil 2	4.09	0.44	ND	12.37
Soil 3	3.83	0.38	ND	10.01
Soil 4	1.78	0.22	ND	8.47
Water 1	0.0059 (µg/l)	ND	ND	ND
Water 2	0.0029 (µg/l)	ND	ND	0.0032 (µg/l)
Bran 1	0.42	0.03	ND	0.31
Bran 2	0.12	0.03	ND	ND
Bran 3	0.19	0.09	ND	0.39
Rice 1	ND	0.13	ND	0.76
Rice 2	ND	0.09	ND	ND
Rice 3 (Red)	ND	0.17	ND	ND

The European Union (EU) has set the highest limit for As in agricultural soils as 20 µg/g (Kumarathilaka et al., 2018). The As level in samples was below that maximum limit; therefore, the As contamination was shallow. The fertilization activities in the paddy rice were also interesting to be observed. Alternative sources of As in paddy soils were cattle manure, the fertilizer that contains phosphate (PO_4^{3-}), and arsenical-based pesticides on cotton-rice rotations (Meharg & Zhao, 2012). Soil Environmental Quality Standards (SEQSs) are a notable tool for applying soil protection policies. The US soil environment quality standard for heavy metals sets the maximum level for Cd, As, Hg, and Pb in soils as 37 µg/g, 22 µg/g, 23 µg/g, and 400 µg/g, respectively (Chen et al., 2018). The metal levels in soil presented in this research were low compared to the US soil standard. The rice field strongly influenced the variation in metal content in rice, as the concentration of heavy metals in rainfed rice was relatively higher than in irrigated rice fields. It was possible because the intensity of using fertilizers and pesticides was higher in the upstream areas. At the same time, leaching by rainwater was less than the more intensive leaching of irrigation water in irrigated rice fields (Sukarjo et al., 2019).

WHO regulated the permissible metal level in irrigating water as 10 µg/l for As, 3 µg/l for Cd, and 10 µg/l for Pb, while Kementerian Kesehatan Republik Indonesia-Ministry of Health in No.492/MENKES/PER/IV/2010 had set the maximum metal contamination limit in drinking water as 0.001 mg/l for Hg, 0.010 mg/l for As, 0.003 mg/l for Cd, and 0.01 mg/l for Pb (Kemenkes, 2010). Therefore, if referring to the standard, the water samples presented in this research were safe for consumption. However, those elements were considered the most toxic metals even at low concentrations, which may lead to bioaccumulation (chronic manifestations) of the diversity of toxic effects on various body tissues and organs

(Balali-Mood et al., 2021). Furthermore, the water samples here were suitable for irrigating crops due to the low metal contaminants. The World Health Organisation (WHO) recommends that total As in drinking water should be under 10 μg/l, while this research found the As level in water was significantly below the limit (WHO, 2018).

The low levels of heavy metals detected in rice, water, bran, and soil samples were possibly due to the use of environmentally friendly technology in Cilacap's power plant facility that probably significantly reduced emissions and pollutants. Several technologies that were proven to reduce emissions and pollutants significantly were Super Critical (SC) and Ultra Super Critical (USC) technology combined with FGD (Flue Gas Desulfurization) and SCR (Selective Catalytic Reduction) (Wang & Zhang, 2019). It was in line with research conducted in 2015, which found that the concentration of minor and trace elements in feed coal was below the world maximum threshold, and particulate matter 2.5 ($PM_{2.5}$) produced from the Cilacap PLTU was also below the national ambient standard and WHO guidelines (Lestiani et al., 2015). One of the critical factors in determining the substantial build-up of As, Cd, Cu, Pb, and Zn in agricultural areas was the predominant wind direction of the power plant, which determines the spread area of emissions and pollutants from coal-fired power plant activity (Wang et al., 2016). In the future, cleaner energy sources will be mandatory, mainly for producing electricity. Among modern renewable energies, wind, geothermal, and solar energy might be the most practical due to their relative maturity, market penetration, abundance, and the capacity to provide base-load (geothermal) or distribution (wind and solar) electricity (Li et al., 2020).

CONCLUSIONS

Coal has a crucial role in electricity generation in the world. Coal is holding a large portion as a source of energy for electricity generation In Indonesia. Currently, Indonesia is intensively developing plenty of coal-fired power plants (PLTU) to support electricity demand growth. Research and safety measurements must be carried out to ensure environmental protection and food safety. This research suggested that most rice, bran, soil, and water samples near coal-fired power plants in Cilacap were below the maximum limit of As, Cd, Hg, and Pb contamination. However, those elements were considered the most toxic metals even at low concentrations, which may lead to bioaccumulation in plants and humans. Transformation to green energy (e.g., solar and geothermal energy) is highly encouraged to minimize the risk of environmental pollution due to the PLTU by-product activities. Nonetheless, there is still a possibility of heavy metal bioaccumulation in plants and humans; thus, further study, such as risk assessment, is needed.

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