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Quantification of Aluminum Contents in Cooked Foodstuffs from Three Regions in Java Using Neutron Activation Analysis

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Aluminum is widely available in nature and the third most abundant element on earth. Improper intake of aluminum can increase toxicity and correlate with Alzheimer's disease. One source of aluminum comes from food. In this study, aluminum content in foodstuffs was analyzed using neutron activation analysis. Various foodstuffs were purchased from markets in three regions in Java, namely Bangkalan (East Java), Magelang (Central Java), and Cianjur (West Java) and cooked at a temperature above 80°C until the ready-to-eat condition. The cooked samples were freeze-dried and irradiated in the G.A. Siwabessy research reactor with neutron flux of $5x10^{13}$ neutrons.$m^2.s^{-1}$. Post-irradiation samples were analyzed using gamma spectrometry. The results show that the aluminum contents in each foodstuff from one region have a strong correlation with other regions (Pearson correlation coefficient $r>0.9$, $P<0.001$), indicating that the distribution of aluminum content does not differ from one region to another. The staple food category has a relatively low aluminum content with an average value of 24 mg/kg and a maximum value of 35 mg/kg. The dish category has higher aluminum content with an average value of 51 mg/kg and a maximum value of 77 mg/kg. The vegetable category has the highest content with an average value of 156 mg/kg and a maximum value of 710 mg/kg owned by caisim. Caisim is interesting for further research because of its ability to store large amounts of several elements. In general, the intake of aluminum sourced from these foods is still below the allowed value.

1. INTRODUCTION

Aluminum occurrence in the environment is almost everywhere. It is the third most abundant element in the Earth's crust after oxygen and silicon. However, in a normal biological system, aluminum is only present in trace amounts and has an unknown role. In fact, no metal elements from groups 3, 4, or 13 and 14 that are important for biological systems are known [1,2].

Food is the main source of aluminum for humans. Aluminum is naturally present in food, and aluminum content can increase due to the cooking process or during conditioning [3–6]. The average aluminum intake for humans is 10 mg/day [3]. Estimated daily and weekly aluminum intake is an important component of risk assessment and evaluation of hazardous substances. In 2011, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a provisional tolerable weekly intake (PTWI) of 2 mg/kg body weight [7]. The intake restriction is carried out because the toxicity of aluminum increases with increasing aluminum intake in the human body. Undue intake of aluminum is associated with

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Alzheimer's disease [8,9]. Alzheimer's disease is a significant cause of dementia and begins with a progressive decline in cognitive function in the form of memory loss [10].

Aluminum content in food is different for each type of food and sampling area. It is widely determined in raw food, whereas food is commonly consumed by people in a cooked condition [11–13]. The distribution of aluminum content is important for estimating its intake by people in a particular area compared with other regions. With reference to consumption levels, daily aluminum intake can also be estimated.

Aluminum content can be determined by various spectroscopy methods such as atomic absorption spectroscopy (AAS), inductively coupled plasma atomic emission spectroscopy (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS) [14–16]. Although these techniques are quite sensitive, they require sample dissolution and can be influenced by empty reagents. Non-destructive radioanalytical methods such as X-ray fluorescence (XRF), particle-induced X-ray emission (PIXE), and neutron activation analysis (NAA) are able to determine the total aluminum concentration [17–19]. Although XRF and PIXE can determine aluminum concentrations, they are not routinely practiced at low concentration levels. NAA can easily determine the aluminum content in various types of samples, especially bulk samples. Analysis of bulk samples is feasible by NAA because of the high neutron penetration power [20]. By using NAA, many samples can be analyzed simultaneously, and the analysis process is quite short. In the present study, we used NAA to determine aluminum content in cooked foodstuffs (boiled at temperatures above 80°C). Foodstuffs are taken from three regions in Java, i.e., Bangkalan (East Java), Magelang (Central Java), and Cianjur (West Java) to find out the distribution of aluminum content in different areas.

2. METHODOLOGY

Sample collection

Various foodstuffs (21 types) were purchased from the local market in three areas in Java, i.e., Bangkalan (East Java), Magelang (Central Java), and Cianjur (West Java). These areas are 3 out of 100 regions with high stunting cases in Java. The part of foodstuffs used as a sample is the part that is generally eaten by people. The foodstuffs were divided into three categories of dishes (D), staples (S), and vegetables (V).

Sample preparation and irradiation

Foodstuffs were cleaned and boiled using water without purification at temperatures above 80°C until the ready-to-eat condition. The equipment used in the boiling process is in the same and clean condition for each sample. To remove the water content, cooked samples were dried through the freeze-drying process at -90°C and a pressure of 0.025 Pa. Dried samples were mashed with mortar to the size of 100 mesh. Samples were weighed and put into low-density polyethylene (LDPE). The samples were irradiated with standard reference materials (SRM) in polyethylene capsules. The irradiation process was carried out in the rabbit system at the G.A. Siwabessy multi-purpose reactor with thermal neutron flux 5×10^{13} neutron.cm^{-2}.s^{-1}. Details of the irradiation parameters are presented in Table 1.

Data acquisition and analysis

Irradiated samples were acquired using gamma spectrometry with a high purity germanium detector (HPGe) and multi-channel analyzer (MCA). The HPGe detector has an efficiency of 30% and a resolution of 1.63 keV for the peak of 1332.5 keV Co-60. All samples were counted with a counting time of 120 s. Data were analyzed using the comparative method with SRM.

Internal quality control of the measurement

Internal quality control ensure that the method used for characterizing materials with biological matrices in this study is valid. For the validation method, Tomato leaves from NIST were used. Statistical evaluation of the test results uses three statistical parameters: U-test score, Z-score, and relative standard deviation (RSD,%). The evaluation uses a U-test score to determine whether the SRM test results obtained differ significantly from the value of the certificate. U-test scores are calculated using equation (1).

$$U_{score} = \frac{|x_m - x_r|}{\sqrt{\mu_m^2 + \mu_r^2}} \qquad (1)$$

where x_m, μ_m, x_r, and μ_r are the values of measurement results, measurement uncertainties, certified standard, and standard uncertainties in sequence. Z-score shows the measurement of the proximity of the measurement value with the reference value. The z-score value is calculated using equation (2).

$$Z_{score} = \frac{x_m - x_r}{\mu_r} \qquad (2)$$

where the performance results are considered satisfactory if $Z_{score} \leq 2$, it is questionable if $2 < Z_{score} < 3$, and unsatisfactory if $Z_{score} \geq 3$. RSD is a measure of the type of error called random error, a type of error that cannot be controlled very well [21,22].

Table 1. Analysis parameters of the elements of interest

Isotope target	Isotope product	Half time (s)	E (keV)	Abd (%)	T_{irr}	T_{decay}
Al-27	Al-28	134	1778.99	100	60 s	5 – 10 m

3. RESULTS AND DISCUSSION

The results of the method validation showed that our measurement performance was excellent. The RSD value is quite low, less than 10%. The Z-score is -0.33 (<2), indicating that the measurement results are acceptable. The U-test score is 1.44 (<1.64), indicating that the measurement results do not differ significantly from the assigned value [23–25].

The results of the analysis of aluminum content in cooked foodstuffs are presented in Table 3. Data from each region are tested using Pearson correlation. The results show that the regions have a strong correlation each other (Pearson correlation coefficient $r > 0.9$, $P < 0.001$) and aluminum contents of each region are co-vary (see Table 4). The strong correlation indicates that the distribution of aluminum content does not differ from one region to another in the foodstuff, and each foodstuff has a different deposit of aluminum content.

Table 2. Comparison of the concentration value of the measurement results and the value of the certificate on the SRM

Element	Certified value	Measured value	RSD (%)	Z-score	U-test score	Ratio (measured/ certified)	SRM type
Al	598 ± 12	558 ± 25	6.7	-0.33	1.44	0.93	Tomato leaves

Table 3. Aluminum concentration in cooked foodstuff samples.

Food categories	**Samples**	**Code**	**Bangkalan**	**Cianjur**	**Magelang**
Dishes	Chicken	D1	31 ± 2	25 ± 2	36 ± 2
	Beef	D2	28 ± 1	45 ± 3	46 ± 10
	Mackerel tuna	D3	45 ± 2	26 ± 3	34 ± 9
	Shrimp	D4	64 ± 7	123 ± 14	45 ± 7
	Oyster mushroom	D5	74 ± 7	82 ± 12	60 ± 12
	Tofu	D6	44 ± 3	46 ± 4	55 ± 4
Staples	Rice	S1	14.8 ± 0.6	43 ± 8	10.8 ± 0.6
	Brown rice	S2	45 ± 2	30 ± 4	24 ± 2
	Glutinous rice	S3	11.2 ± 0.7	12.1 ± 0.7	20 ± 3
	Black glutinous rice	S4	25 ± 1	22 ± 1	14 ± 2
	Potato	S5	26 ± 2	20 ± 3	20 ± 1
	Peanuts	S6	45 ± 3	36 ± 3	23 ± 1
	Soybeans	S7	25 ± 2	22 ± 1	20 ± 4
Vegetables	Long beans	V1	47 ± 3	37 ± 3	38 ± 6
	Bean sprouts	V2	42 ± 2	28 ± 2	19 ± 2
	Leek	V3	127 ± 5	146 ± 6	206 ± 10
	Papaya leaf	V4	159 ± 7	142 ± 10	86 ± 30
	Caisim	V5	996 ± 40	777 ± 89	358 ± 20
	Bitter ground	V6	83 ± 3	72 ± 3	44 ± 2
	Eggplant	V7	60 ± 6	48 ± 3	38 ± 2
	Young jackfruit	V8	69 ± 16	65 ± 11	60 ± 2

Table 4. Pearson correlation test results.

		Bangkalan	Cianjur	Magelang
Bangkalan	Pearson Correlation	1	.993**	.909**
	Sig. (2-tailed)		.000	.000
	N	21	21	21
Cianjur	Pearson Correlation	.993**	1	.927**
	Sig. (2-tailed)	.000		.000
	N	21	21	21
Magelang	Pearson Correlation	.909**	.927**	1
	Sig. (2-tailed)	.000	.000	
	N	21	21	21

**. Correlation is significant at the 0.01 level (2-tailed).

The range of aluminum content in each cooked foodstuff is presented using a boxplot (can be seen in Figure 1). The result shows that the staple category has a relatively low aluminum content (14 – 35) mg/kg, with an average value of 24 mg/kg and a maximum value of 35 mg/kg. The dish category has a higher aluminum content than the staple category (31 – 77) mg/kg, with an average value of 51 mg/kg and a maximum value of 77 mg/kg. The vegetable category has a relatively high aluminum content (30 – 710) mg/kg, with an average value of 156 mg/kg and a maximum value of 710 mg/kg. The categories based on aluminum content from the lowest are staples, dishes, and vegetables, respectively.

Vegetables tend to have high aluminum content because of their life in water and their ability to store large amounts of the element [26]. Caisim has the highest aluminum content (average and maximum value of 710 and 996 mg/kg, respectively). Caisim, in its raw condition, is also reported to have a high potassium content [27]. Caisim is interesting for further research, considering its ability to store a large amount of several elements.

Aluminum intake can be estimated by multiplying the aluminum content in each food item by its consumption level. Table 5 shows that aluminum intake from each food item is relatively low. Rice has the biggest contribution in aluminum intake (about 5 mg/kg) due to the high level of consumption. In general, the daily aluminum intake sourced these foods is still below the permitted value.

Table 5. The level of consumption of some foodstuffs and estimated daily aluminum intake [28,29]

Foodstuffs	Consumption rate per day (kg)	Estimated daily aluminum intake[a] (mg)
Beef	0.0013	0.05
Chicken	0.0173	0.54
Fish and shrimp	0.0463	2.60
Tofu	0.0226	1.10
Rice	0.2210	5.06
Sticky rice	0.0007	0.01
Potato	0.0063	0.14
Peanut	0.0007	0.02
Soybean	0.0001	0.00
Long beans	0.0064	0.26
Bean	0.0026	0.08
Leek	0.0004	0.06
Eggplant	0.0073	0.36

[a] Multiplication results with the measured values

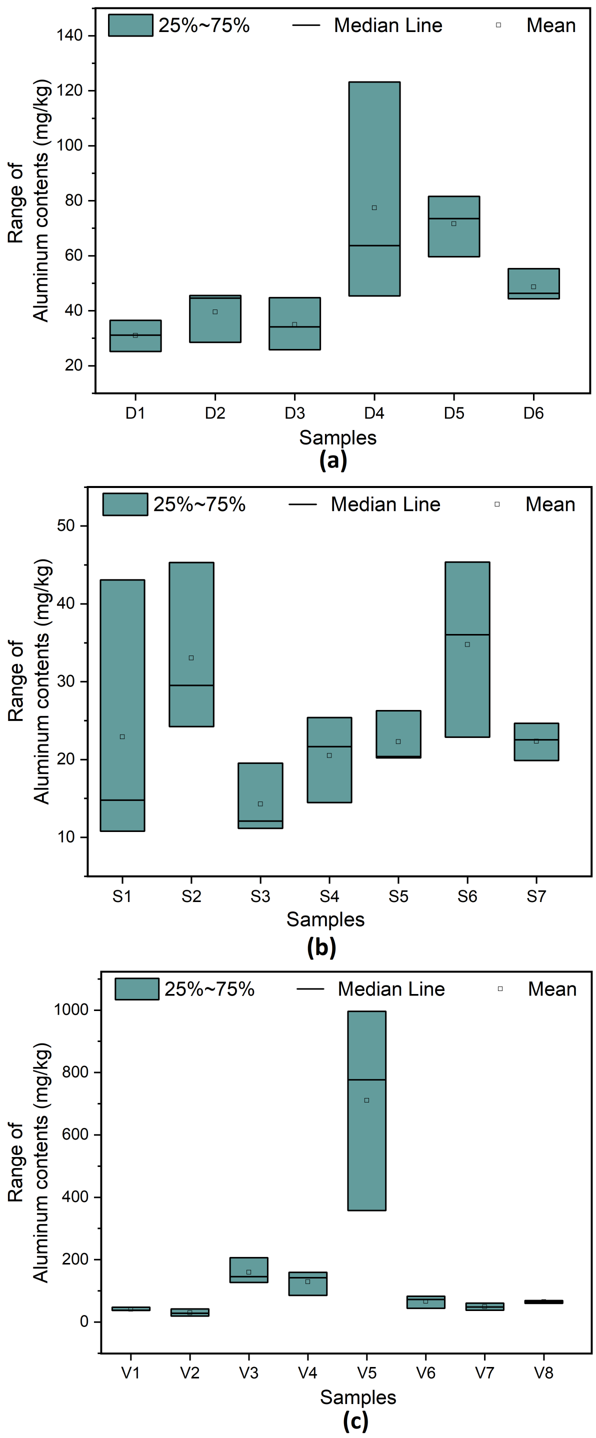

Fig. 1. Boxplot of aluminum contents in cooked foodstuffs (a) dishes, (b) staples, (c) vegetables

4. CONCLUSION

The results show that the aluminum contents in each foodstuff from one region have a strong correlation with other regions (Pearson correlation coefficient $r > 0.9$, $P < 0.001$). The staple food category has a relatively low aluminum content with an average value of 24 mg/kg and a maximum value of 35 mg/kg. The dish category has more content with an average value of 51 mg/kg and a maximum value of 77 mg/kg. The vegetable category has the highest content with an average value of 156 mg/kg and a maximum value of 710 mg/kg owned by caisim. Caisim is interesting for further research because of its ability to store large amounts of several elements. In general, the intake of aluminum sourced from these foods is still below the allowed value.

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