

Article

Risk Assessment of Aluminum in Drinking Water between Two Residential Areas

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Abstract: A cross-sectional study was conducted at Sungai Lembing (SL) and Bukit Ubi (BU), Kuantan, Malaysia. The main objectives of this epidemiological study were to determine the aluminum concentration in drinking water, to compare with the government standard and to perform health risk assessment prediction among respondents from these two residential areas. A total of 100 respondents were selected from the study areas based on a few inclusive and exclusive criteria. Two duplicates of treated water samples were taken from each respondent's house using a 200 mL high-density polyethylene (HDPE) bottle and 0.4 mL (69%) pure concentrated nitric acid added as preservative. Aluminum concentrations were analyzed using Lambda 25 UV/V spectrophotometer. The result showed that the mean concentration of aluminum in drinking water from SL was 0.11 ± 0.0634 mg/L and 0.12 ± 0.0462 mg/L for BU. The mean value of Chronic Daily Intake (CDI) in SL (0.0035 ± 0.0028 mg/kg/day) was lower compared to BU (0.0037 ± 0.0021 mg/kg/day). The Hazard Index (HI) calculation showed all respondents had HI less than 1. In conclusion, there was unlikely potential for adverse health effects from aluminum intake in drinking water. However, it was necessary for some action to be taken in order to reduce aluminum levels found in drinking water in both locations.

Keywords: aluminum in drinking water; health risk assessment; chronic daily intake (CDI); hazard index (HI); environmental health; environmental hygiene; guideline

1. Introduction

Aluminum and its compounds cover about 8% of the Earth's surface [1]. It is the third most abundant element after oxygen and silicon, and this compound occurs naturally in cryolite, silicates, and bauxite rock [1]. Aluminum is a non-essential trace element with no known biological function to which humans are frequently exposed [2]. Aluminum is widespread throughout nature, air, water, plants and consequently in food [3]. The physical and chemical characteristics of aluminum make it ideal for a variety of uses in food, drugs, consumer products, and water treatment processes [2].

Aluminum salts such as aluminum sulphate (alum) or polyaluminum chloride (PACl) are used extensively as coagulants in drinking water treatment to enhance the removal of particulate, colloidal and dissolved substances [4]. Aluminum sulphate and polyaluminum chloride are the most widely used coagulants, because they are effective, readily available, and relatively inexpensive [5]. However, aluminum-based coagulants have come under scrutiny in recent years due to concerns about aluminum residuals in the public water supply [6] and interests concerning aluminum have considerably increased due to increased knowledge about the potential toxic effects of aluminum [3].

Health risks associated with exposure in drinking water containing aluminum can be distinguished into two terms: acute and chronic toxicity. For acute toxicity, there is little indication that aluminum is acutely toxic by oral exposure, despite its widespread occurrence in foods, drinking-water, and many antacid preparations [7]. There are no reported cases of acute aluminum poisoning of healthy individuals exposed to normal levels of aluminum [8].

Chronic toxicity of aluminum in drinking water is associated with severe diseases of the nervous system such as Parkinson's dementia, amyotrophic lateral sclerosis and Alzheimer's disease [2]. Alzheimer's disease is a progressive mental deterioration manifested by memory loss, inability to calculate, visual spatial disturbances, confusion and disorientation [9]. A study by Rondeau [9] found that high aluminum levels in drinking water (≥ 0.1 mg/L) were associated with an elevated risk of dementia and Alzheimer's disease [9]. To date, 13 epidemiological studies have been done worldwide to investigate the hypothesis of a correlation between Alzheimer's disease and increased concentrations of aluminum in drinking water, and nine of these studies have found a positive association [10,11].

There is limited data available to determine the risk associated with aluminum exposure in drinking water among the Malaysian population. This study was carried out to determine the aluminum concentration in drinking water and to evaluate the health risk among respondents from two selected residential areas in Malaysia.

2. Materials and Methods

The study was conducted in two residential areas of the Kuantan district in the state of Pahang Darul Makmur, Malaysia, namely Sungai Lembing (SL) and Bukit Ubi (BU). The source of SL

drinking water was from the Sungai Lembing's water treatment plant while BU drinking water was from the Bukit Ubi's water treatment plant.

The study population comprised male and female respondents aged 18 and above who use treated water as their main source of drinking water. The respondents were purposively selected using a list of names provided by the head of the village or "Ketua Kampung". The inclusive criteria used were: male or female respondents, aged 18 and above and use of treated water as their main source of drinking water. While the exclusion criteria were: respondents that use bottled water or well water as their main source of drinking water. Each house was represented by one respondent.

A total of 100 respondents were selected for this study, where 50 respondents were each selected from the two study areas. A structured questionnaire which comprised two sections was used in this study. The first section contained questions regarding the respondents' background information such as age, gender, household income and education level. The second section gathered information regarding duration of residence which was used to calculate the Chronic Daily Intake (CDI) of aluminum exposure in their drinking water source. The questionnaire developed, was adapted from the Baseline, Descriptive and Time Activity Questionnaires used in the National Human Exposure Assessment Survey (NHEXAS) Arizona study [12].

Drinking water samples were collected at the respondents' kitchen tap. The tap was turned on and water was allowed to run for 3–5 minutes before it was collected. A 200 milliliter (mL) non-acidified high-density polyethylene (HDPE) bottle was used for water sample collection. Two replicates of water samples were taken from each respondent's house. The samples were then preserved using 0.4 mL 69% pure concentrated nitric acid before being analyzed at the laboratory to ensure bacterial removal from the samples and to lengthen the storage time of the samples [13]. The body weight of the respondents was measured using Tanita Digital Weight Scales. The readings were taken three times and then averaged.

The Lambda 25 UV/V spectrophotometer was used to determine the aluminum concentration in drinking water from both study areas. The Erlochome Cyanine method was used to determine the aluminum concentration in drinking water with detection limit 0–1.2 mg/L. The analytical wavelength used was 278.0 nm. The ordinate mode was single wavelength. The split UV/VIS was 1.00 nm [14].

In order to estimate health risk associated with aluminum in drinking water, chronic daily intake (CDI) was first calculated using the following equation [15]:

$$CDI (I) = (C_1 R_1 F_E D_t) / (W_B T_{AVG}) \quad (1)$$

Where CDI (I) is the chronic daily intake (mg/kg/d), C_1 is the level of aluminum concentration in drinking water (mg/L), R_1 is ingestion rate (2 L/day), F_E is exposure frequency (day/year), D_t is exposure duration (year), W_B is body weight (kg) and T_{AVG} is the average of exposure duration ($D \times 365$ days/year). To conclude the significant exposure and overall potential for non-carcinogenic health effects posed by aluminum in drinking water, the Hazard Index (HI) was calculated using the following Equation [15]:

$$Hazard\ Index\ (HI) = (CDI) / (RfD) \quad (2)$$

RfD is reference dose (Equation 2). RfD for Aluminum is 7 mg/kg/day [16]. In cases where the non-cancer HI does not exceed unity ($HI < 1$), it is assumed that no chronic risks are likely to occur at the study site [15].

Data collected from the questionnaire, aluminum concentration in drinking water, body weight of respondents, CDI and HI of both study areas were analyzed using Statistical Package for Social Science (SPSS) version 17.0 (SPSS Inc, Chicago, IL, USA). Kolmogorov-Smirnov, Shapiro Wilk and Skewness test were used to determine the normality of distribution of the variables. The data were then analyzed in two stages of analysis. The first stage was univariate analysis. Bivariate analysis was then used to test the hypothesis, which was divided into testing for statistical significant difference and testing for relationship or associations.

3. Results

3.1. Background of Respondent

Fifty respondents from both study areas were selected based on purposive sampling. Essential information including age, race, gender, and household income, and respondent weight, education level of respondents and duration of residence were collected (Tables 1 and 2).

Table 1. Age, household income, weight and duration of residence of respondents.

Variable	Sungai Lembing (SL) (n = 50)		Bukit Ubi (BU) (n = 50)	
	Range	Mean \pm SD	Range	Mean \pm SD
Age (years)	18–77	42 \pm 17	18–63	38 \pm 14
Household Income (RM)	220–3122	994.78 \pm 523.594	600–3000	1520.00 \pm 693.924
Weight (kg)	44.0–90.0	63.342 \pm 10.8277	45.0–93.0	64.118 \pm 11.0462
Duration of residence (years)	3–59	17.86 \pm 12.090	2–55	14.18 \pm 12.229

N = 100; SL = Sungai Lembing; BU = Bukit Ubi.

Table 2. Race, gender and education level of respondents.

Variable	SL (n = 50)	BU (n = 50)
	Frequency (%)	Frequency (%)
Race		
Malay	50 (100)	50 (100)
Gender		
Male	23 (46)	30 (60)
Female	27 (54)	20 (40)
Education level		
No education	2 (4)	0 (0)
Primary school	12 (24)	5 (10)
Lower Secondary Certificate (PMR)	10 (20)	9 (18)
Higher Secondary Certificate (SPM)	18 (36)	28 (56)
Sijil Tinggi Pelajaran Malaysia (STPM)	3 (6)	0 (0)
Higher education	5 (10)	8 (16)

N = 100; SL = Sungai Lembing; BU = Bukit Ubi.

3.2. Aluminum Concentration in Drinking Water

The results showed there was no significant difference in aluminum concentration in drinking water between the two study areas (Table 3). Table 4 shows the number of drinking water samples that violated the Malaysian Drinking Water Standard. Based on the result, samples from SL showed a higher frequency of violation compared to BU (Table 4).

Table 3. Differences of Aluminum concentration in both study locations.

Variable	SL (n = 50)		BU (n = 50)		t	p
	Range	Mean ± SD	Range	Mean ± SD		
Aluminum concentration (mg/L)	0.02–0.28	0.11 ± 0.0634	0.05–0.26	0.12 ± 0.0462	−0.861	0.391

N = 100; SL = Sungai Lembing; BU = Bukit Ubi.

Table 4. Aluminum violation in two study locations against Ministry of Health Guideline.

Variable	SL (n = 50)	BU (n = 50)
	Frequency (%)	Frequency (%)
Aluminum concentration (mg/L)	6 (12)	4 (8)

N = 100; SL = Sungai Lembing; BU = Bukit Ubi.

3.3. Difference of Aluminum Concentration in Drinking Water Compared with Malaysian Drinking Water Standard

The difference in aluminum concentration in drinking water from SL and BU compared with the Malaysian Drinking Water Standard was analyzed with a reference value of 0.2 mg/L. The results in Table 5 show a low significant difference in aluminum concentration in drinking water for SL and BU compared with the Malaysian Drinking Water Standard.

Table 5. Difference of aluminum concentration in drinking water from SL and BU with the Malaysian Drinking Water Standard.

Variable	SL (n = 50)			BU (n = 50)		
	Mean ± SD	t ^	p	Mean ± SD	t ^	p
aluminum concentration (mg/L)	0.11 ± 0.0634	−9.582	0.001 *	0.12 ± 0.0462	−11.689	0.001 *

N = 100; SL = Sungai Lembing; BU = Bukit Ubi; ^ Test Value = 0.2 (Compare with Malaysia Guideline from Ministry of Health, Malaysia); * Significant at p value ≤ 0.05.

3.4. Respondents' Chronic Daily Intake

Respondents' CDIs for both study areas were calculated using the CDI equation (1). Table 6 shows the CDI of respondents from both study areas. The difference in respondents' CDIs were analyzed using a non parametric test (Mann Whitney U test) due to non-normal data distribution. Based on the result, there was no significant difference in CDI between the two study areas (Table 6).

Table 6. Chronic Daily Intake (CDI) of respondents.

Variable	SL (n = 50)		BU (n = 50)	
	Range	Mean ± SD	Range	Mean ± SD
Chronic Daily Intake (CDI) (mg/kg/day)	0.0008–0.0112	0.0035 ± 0.00224	0.0012–0.010	0.0040 ± 0.00027
	Median (IQR)	Median (IQR)	z	p
	0.0035 (0.0028)	0.0037 (0.0021)	−1.041	0.298

N = 100; SL = Sungai Lembing; BU = Bukit Ubi.

3.5. Hazard Index (HI)

The HI was used to determine the health risk of aluminum by dividing CDI by RfD. The RfD of aluminum is 7 mg/kg/day [16]. Based on previous literature [16], there are two classes of hazard index; greater or less than 1. Table 7 shows the HI of respondents and its categories from both study areas respectively.

Table 7. HI of respondents and its categories.

Variable	SL (n = 50)		BU (n = 50)	
	Range	Mean ± SD	Range	Mean ± SD
Hazard Index (HI)	0.000115–0.001714	0.000532 ± 0.00032	0.000174–0.001491	0.000576 ± 0.00027
	Frequency (%)		Frequency (%)	
HI < 1	50 (100)		50 (100)	
HI > 1	0 (0)		0 (0)	

N = 100; SL = Sungai Lembing; BU = Bukit Ubi; HI = Hazard Index.

4. Discussion

4.1. Respondent Background

In this study 100 respondents were selected as the study sample, consisting of 50 respondents from SL and 50 respondents from BU. The gender selection between male and female was similar in both study areas. The mean household income of respondents in BU was greater than those in SL. The reason why is because BU is located in an urban area that offers opportunity for higher salaries and incomes compared to SL, which is located in a rural area with limited employment opportunities.

The majority of respondents were educated to the level of Higher Secondary Certificate (SPM) for both study areas. The mean weight of respondents from SL was lower than BU but the difference in mean weight between the study areas was only 0.776 Kg. This study found that the duration of residence in SL differs by about three years compared to BU.

4.2. Aluminum Concentration in Drinking Water

Aluminum concentration in drinking water samples from SL ranged from 0.02 to 0.28 mg/L with a mean of 0.11 mg/L and standard deviation of ±0.0634 mg/L while samples from BU ranged from 0.05 to 0.26 mg/L with a mean 0.12 of mg/L and standard deviation of ±0.0462 mg/L. Aluminum concentration in drinking water for both areas were lower compared to the typical concentration of

aluminum in Australia which varies from 0.01 to 0.9 mg/L [17] and from another study done in Aomori Prefecture, Japan that ranged between 0.01 to 2.11 mg/L [18]. Another study done in Selangor, Malaysia found that aluminum concentration in drinking water ranges from 0.0063 mg/L to 0.03 mg/L, which is lower than the finding in this study [19]. In 1999, Canadian researchers reported that the levels of aluminum in Toronto's drinking water ranged from 0.019 to 0.29 [20], which is higher than in this study. Another study in Texas, USA showed that concentrations of aluminum up to 0.53 mg/L were observed in treated water [21].

Based on a previous study, the mean value of both areas were higher compared to the mean values from Kuala Pilah, Malaysia (0.03 mg/L) [22]. The mean aluminum concentration in drinking water from public water supplies in Galicia, Northwest Spain was 0.12 mg/L, which was similar to the mean from BU [4]. In addition, the mean aluminum concentration in this study was lower than the mean value reported in a study from the Moroccan city of Marrakech (0.21 mg/L) [23]. Aluminum concentration in drinking water measured in the Netherlands was found to be 0.6 mg/L, which was higher than both (SL and BU) study areas [24]. There were 6 (12%) violations of aluminum concentration in drinking water which exceeded the Malaysia Drinking Water Standard (0.2 mg/L) in SL while in BU there were 4 (8%) violations. The number of violations was lower than a violation of 15.9% in 60 samples in the Moldova territory, Romania [25]. Based on an annual report entitled "Drinking Water 2008" of The Drinking Water Inspectorate of England, a total of 6,165 samples was tested for aluminum in the central region of England. Severn Trent Water and Dŵr Cymru Welsh Water achieved 100% compliance with the aluminum standard. Compliance with the aluminum standard in BU was 92% compared to a 100% compliance in the central region of England [26].

The presence of aluminum in water distribution systems can be due to aluminum in the source water, aluminum leached from distribution system materials and aluminum introduced to the water from aluminum containing coagulants [27]. Other factors that may affect aluminum concentration in drinking water are temperature, pH and turbidity of the water [28].

4.3. Difference of Aluminum Concentration in Drinking Water between the Two Study Areas

The results of this study found that there were no significant differences in aluminum concentration in drinking water between the two study areas. This shows that the quality of water received in terms of aluminum concentration was almost the same even though the source of water supply was different in each area. The finding was similar to a study by Azmir [19] in Selangor (Malaysia), who found that there was no difference in aluminum concentration in drinking water between the two areas [19].

The results of this study finding were different from Japan where the mean aluminum concentration in drinking differed between two different water treatment plants [18]. Other European studies showed aluminum concentration in drinking water to be different between sampling locations in Spain [29]. A researcher in Malaysia also reported a difference in aluminum concentration in drinking water between two locations in Johor (Malaysia) [30].

4.4. Difference of Aluminum Concentration in Drinking Water from the Two Study Areas with Malaysia Drinking Water Standard

Statistical analysis showed significant difference in aluminum concentration in drinking water from SL and BU with Malaysia Drinking Water Standard ($p < 0.05$, $t = -9.582$) and ($p < 0.05$, $t = -11.689$) respectively. This shows the quality of water received in terms of aluminum concentration in both study areas met the Malaysia Drinking Water Standard (0.2 mg/L). The lower aluminum concentration in drinking water shows that a portion of the alum added to the raw water during water treatment was low; therefore the residual aluminum in the treated water becomes lower. The most apparent source of aluminum in drinking water comes from the corrosion of aluminum utensils, tanks or pipes or from incorrect dosing of aluminum sulphate as coagulant at the treatment works. Ideally, water going into supply should contain less than 0.2 mg/L [31]. Previous studies conducted in Malaysia by Azmir [19] in Selangor and Nora [30] in Johor found significant differences in aluminum concentration in drinking water between their study areas compared with the Malaysian Drinking Water Standard, which was similar to this study [19,30].

Aluminum concentrations higher than the drinking water standard of 0.3 mg/L are commonly present in the drinking water supplied to the Venezuelan population [32]. Based on an annual report entitled "Drinking Water 2008" of The Drinking Water Inspectorate England, the majority of samples analyzed complied with the standard. Non compliance was due to a localized disturbance of mains deposits and associated with elevated levels of iron in samples taken from the local network [23].

4.5. Respondents Chronic Daily Intake

There are several variables that can affect the respondents CDI including aluminum concentration in water (C1), duration of exposure (Dt), and body weight (WB). These variables were used to calculate the respondent's CDI.

This study found that CDI of respondents from SL ranged from 0.0008 to 0.0112 mg/kg/day while CDI of respondents from BU ranged from 0.0012 to 0.0104 mg/kg/day. The mean of CDI among respondents from both study areas was lower than the Aluminum Reference Dose (Rfd) of 7 mg/kg/day [16]. The mean CDI from both study areas was also lower than in a previous daily intake study, the PAQUID Cohort study in France, which reported a CDI of 0.025 mg/kg/day [33]. The mean CDI found by a Malaysian researcher in two study sites in the state of Selangor, namely Sungai Michu and Sungai Buah was 0.0516 mg/kg/day and 0.0391 mg/kg/day respectively [19], while another researcher found the CDI from two study locations in the state of Johor to be 1.41 mg/kg/day and 0.351 mg/kg/day [30]. The mean CDI from both studies were higher compared to mean CDI from both study areas in this study.

This study found no significant difference between the CDI of the two study areas. This shows that respondents' CDIs were almost similar since there was no significant difference in aluminum concentration in drinking water between both study areas. This study was different from a previous study by Nora [30] who found a significant difference in CDI between areas studied [30].

4.6. Hazard Index (HI)

This study found that the HI for SL respondents ranged from 0.000115 to 0.001714 while the HI for BU respondents ranged from 0.000174 to 0.001491. The mean value of HI for SL was 0.000532 which was lower compared to the HI from BU of 0.000576. The respondents' HI mean was less than 1 due to low CDI values. If the HI is more than 1 it indicates a risk of aluminum exposure to the respondents and if less than 1, it means respondents from both locations were not vulnerable to a risk of aluminum exposure.

The risk of aluminum exposure was shown in several studies which found a relationship between aluminum concentration in drinking water and the cognitive function in Alzheimer's disease [34]. A previous study from Novodvinsk, Northwest Russia found the HI for adults to be 0.009 [35]. This value was higher than the HI in both, SL and BU. A study by Azmir [19] found that the HI for Sungai Michu residents, Malaysia ranged from 0.0023 to 0.0145 with a mean of 0.004 while which in Kampung Buah residents, Malaysia the HI ranged from 0.0031 to 0.0136 with a mean of 0.0056 [19]. The range and mean value of HI from a previous study was higher than this study [19]. The overall results of HI for this study indicates that the health risk of aluminum in drinking water was relatively safe even some of the water samples taken exceeded the level recommended by the Malaysia Ministry of Health guideline (0.2 mg/L) by 10%. A similar result was observed by other researchers in the state of Johor and other districts [36-38] where many studies areas had lower HI values.

5. Conclusions

The main objective of this study, to determine the concentration of aluminum in drinking water and its health risk to respondents, has been achieved. This study found that mean aluminum concentration of both study areas was lower than the Malaysian Drinking Water Standard. Comparison between both study areas with Malaysia Drinking Water Standard showed no significant difference which means that the respondents from both study areas were supplied with treated water that met the standard. Only 10 (10%) samples exceeded the level of aluminum concentration. To conclude, 90 (90%) of respondents received water supply within the permitted limit set by the authorities.

Health risk assessment prediction by calculating CDI and HI of respondents in this study, found that the CDI of both locations was lower than the Aluminum Reference Dose (Rfd) of 7 mg/kg/day. In addition, the HI calculation showed results of less than "1" which indicates no risk exposure to aluminum in drinking water. Respondents from both studies are considered to be safe from the risk of aluminum in drinking water which is related to diseases including Alzheimer Disease.

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