Risk Assessment of Aluminum Residue in Drinking Water of Residents in Sandakan, Sabah

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ABSTRACT

Objective: To assess the risk of aluminum residue exposure in drinking water of residents of Batu 7, Sandakan.

Method: A cross sectional study was conducted to determine the health risk associated with the exposure to aluminum in drinking water among population. The study population comprised of male and female respondents, aged 18 and above whom use treated water as their main source of drinking water. A total of 100 respondents were involved in this study. Data was analysed using Statistical Package for Social Science (SPSS).

Results: There was significant relationship between aluminum and pH levels of drinking water samples in the study area. There was significant difference between aluminum levels in drinking water with National Standards for Drinking Water Quality. There was no health risk of aluminum exposure among respondents, with HI<1.

Conclusion: The mean of aluminum concentration was 1.03 mg/L in this study exceeded the national standard of 0.2 mg/L by 5 folds, leaving a significant difference between drinking water sample and the upper safe limit. However, the Hazard Index calculated from the findings did not exceed 1. This showed that the study area was considered safe from having risk related to nervous systems, even though the aluminum concentration exceeded the upper safe limit.

Keywords: aluminum, pH, drinking water, chronic daily intake, hazard index

1. Introduction

Water is one of our most valuable resources, which essential to sustaining life and a healthy environment. Municipal or residential use of water significantly impacts the overall water supply and its quality. Water used at home is for drinking water and cleaning; therefore, access to safe drinking water is essential to humans and other life forms even though it provides no calories or organic nutrients. Access to safe drinking water has improved over the last decades in almost every part of the world, but approximately one billion people still lack access to safe water and over 2.5 billion lack access to adequate sanitation (UN News Centre, 2008).

Aluminum is usually used as a coagulant in raw water treatment. Aluminum salts such as aluminum sulfate (alum) or polyaluminum chloride (PACl) are used extensively as coagulants in raw water treatment to enhance the removal of particulate, colloidal and dissolved substances (Rubinos et al., 2005). These coagulants are widely used as they are effective, readily available and relatively inexpensive (Frentiu et al., 2004). However, aluminum-based coagulants have come under scrutiny in recent years due to concerns about aluminum residuals in public water supplies (Kvech and Edwards, 2002) and interests concerning aluminum have considerably increased due to increased knowledge about the potential toxic effects of aluminum (Narin et al., 2004). National Standards for Drinking Water Quality by Ministry of Health Malaysia has set 0.2 mg/L as the limit to Al concentration in drinking water (MOH, 2004).

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 (WHO, 1997a). There are no
reported cases of acute aluminum poisoning of healthy individuals exposed to normal levels of aluminum, which is below 0.2 mg/L (Verissimo and Gomes, 2008). Chronic toxicity of aluminum in drinking water is associated with severe diseases of the nervous system such as Parkinson’s, amyotrophic lateral sclerosis and Alzheimer’s disease (Health Canada, 2012).

Alzheimer’s disease is a progressive mental deterioration manifested by memory loss, inability to calculate, visual spatial disturbances, confusion and disorientation (Rondeau et al., 2000). The study found that high aluminum levels in drinking water (>0.1 mg/L) were associated with an elevated risk of dementia and Alzheimer’s disease. To date, 13 epidemiological studies have been done worldwide to investigate the hypothesis of a correlation between Alzheimer’s disease and increased levels of aluminum in drinking water and nine of these studies have found a positive association (Flaten, 2001; Gupta et al., 2005).

To date, 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 level in drinking water and to evaluate the health risk among respondents from selected residential area in Malaysia.

2. Methodology

2.1 Study location

This study was conducted in Batu 7, Sandakan, Sabah. Sandakan is administered by Sandakan Municipal Council, with area of 875 square miles, with total population of 396,290 people based on population census (Sandakan Municipal Council, 2010). Batu 7 Sandakan was selected as the study location because the population density is the largest among residential areas in Sandakan and they used treated water as their main source of drinking water. The housing areas, including Kampung Tinusa, Kampung Batu Putih, Taman Gaya, Taman Khong Lok, and many more. The water source is from Kinabatangan River, Batu 5 River and groundwater (Sandakan Town Planning Department, 2014).

2.2 Study Design

A cross sectional study was conducted to estimate the health risk associated with the exposure to aluminum in drinking water among Batu 7, Sandakan population. Cross sectional study involves observation of some subset of a population of items all at the same time, in which, groups can be compared at different ages with respect of independent variables, such as IQ and memory. This study can be thought of as providing a “snapshot” of the characteristics of water quality and risk of drinking water among the population of Batu 7, Sandakan at a particular point in time.

2.3 Study Population and Sampling Method

The study population comprised male and female respondents aged 18 and above whom use treated water as their main source of drinking water. The sampling method in this study was purposive sampling method. Respondents were selected based on the inclusion and exclusion criteria. The inclusion criteria were those who use treated water as their main source of drinking water, and have been living in Batu 7, Sandakan for at least 10 years. While the exclusion criteria were respondents that use bottled water, well water or personal water filtration systems as their main source of drinking water.

2.4 Water Sampling

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 millilitre (mL) non-acidified high-density polyethylene (HDPE) bottles were used for water samples collection. Two replicates of water samples were taken from each sampling point. Then, samples were preserved by 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 (MOH, 2004).

2.5 Study Instrumentation and Data Collection

2.5.1 Questionnaire

A questionnaire was modified from the Baseline, Descriptive and Time Activity Questionnaires used in National Human Exposure Assessment Survey (NHEXAS) Arizona study (Kavcar et al, 2009). The questionnaire was translated back to back to the national language. A pre-test of questionnaire was conducted to ensure that all the questions were easily understood by respondents.
2.5.2 Thermo Scientific - SOLAAR S Series AA Spectrometer

Aluminum residue in water samples was measured using Thermo Scientific - SOLAAR S Series AA spectrometer. This equipment performed efficient and accurate trace elemental analysis. The detection range was from 0.1 to 1.5 mg/L. It came with dedicated flame, furnace or combined flame and furnace options. It was fast, easy-to-use and fully automated AAS analyzers (Thermo Scientific, 2015).

2.6 Health Risk Assessment

The chronic daily intake (CDI) was calculated using the following equation (USEPA, 1989):

\[
 \text{CDI (I)} = \left( C_i \cdot R_i \cdot F_E \cdot D_t \right) / (W_b \cdot T_{AVG})
\]

where CDI (I) is the chronic daily intake (mg/kg/d), \( C_i \) is the level of aluminum level in drinking water (mg/L), \( R_i \) 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 (USEPA, 1989):

\[
 \text{Hazard Index (HI)} = \frac{\text{CDI}}{\text{RfD}}
\]

where the RfD is reference dose. RfD for Aluminum is 7 mg/kg/day (FAO and WHO, 1989). 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 (USEPA, 1989).

3. Results

3.1 Aluminum and pH level in Drinking Water

Table 1 shows the aluminum concentration and pH level of water samples. Based on the Malaysian Drinking-water Standard, aluminum concentration in drinking water samples must not be more than 0.2 mg/L and the normal pH level was between 6.5 to 9. The mean ± SD of Al residue was 1.026 mg/L ± 0.17 while pH ranged from 6.53 to 7.96. The mean level of Al residue in this study exceeded the standard while pH value was within the standard.

<table>
<thead>
<tr>
<th>Variables</th>
<th>Mean ± SD</th>
<th>Minimum</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al concentration (mg/L)</td>
<td>1.03 ± 0.17</td>
<td>0.63</td>
<td>1.39</td>
</tr>
<tr>
<td>pH level</td>
<td>7.61 ± 0.27</td>
<td>6.53</td>
<td>7.96</td>
</tr>
</tbody>
</table>

N = 100

3.2 Correlation of Aluminum Concentration with pH Value of Water Samples

Spearman’s correlation indicated negative association between pH level and Al concentration (\( r = -0.377, p < 0.01 \)) (Table 2).

### Table 2. Correlation between aluminum concentration with pH value of water samples

<table>
<thead>
<tr>
<th>Variable</th>
<th>Unstandardized Coefficients</th>
<th>Standardized Coefficients</th>
<th>T</th>
<th>p</th>
</tr>
</thead>
<tbody>
<tr>
<td>(Constant)</td>
<td>2.361</td>
<td>0.466</td>
<td>5.070</td>
<td>&lt;0.001</td>
</tr>
<tr>
<td>pH_level</td>
<td>-1.176</td>
<td>-0.278</td>
<td>-2.870</td>
<td>0.005</td>
</tr>
</tbody>
</table>

*a dependent variable: aluminum concentration

3.3 Comparison of Aluminum Concentration between Housing Areas of Batu 7, Sandakan

There was no significant difference of Al concentration between housing areas in this study with the p value of 0.629 (> 0.05). The mean ± SD of Al concentration for Kg Batu Putih, Kg Tinusa, Taman Sri Rimba and Taman Khong Lok were 0.9±0.2 mg/L, 1.1±0.2 mg/L, 1.0±0.1 mg/L, and 1.1±0.1 mg/L, respectively. The ANOVA test showed that there is at least one pair of housing areas that has differences (p = 0.002, <0.05). Based on Tukey’s post hoc test, the mean Al concentration for Kg Tinusa and Taman Khong Lok are slightly higher than Kg Batu Putih and Taman Sri Rimba. There was significant difference between Taman Khong Lok and Kg Batu Putih (p = 0.021, less than 0.05) and Kg Tinusa and Kg Batu Putih (p = 0.002, less than 0.05).

3.4 Health Risk Assessment

The mean ± SD of water daily intake rate (DI) in this study was 1.64 ± 0.47 L/day and ranged between 0.4 to 3 L/day (Table 3). The mean ± SD of chronic daily intake (CDI) was 0.030 ± 0.014 and ranged between 0.005 and 0.081. The Hazard Index (HI)
associated to the health risk of aluminum exposure among respondents was less than 1 with a mean ± SD of $4.28 \times 10^{-3} \pm 1.9 \times 10^{-3}$. The range of HI was between $0.07 \times 10^{-2}$ to $1.15 \times 10^{-2}$.

**Table 3.** The daily intake rate of water (DI), chronic daily water intake (CDI) and hazard index (HI) of respondents in this study

<table>
<thead>
<tr>
<th>Variable</th>
<th>Mean ± SD</th>
<th>Minimum</th>
<th>Maximum</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume of water (L/day)</td>
<td>$1.64 \pm 0.47$</td>
<td>0.40</td>
<td>3.00</td>
</tr>
<tr>
<td>CDI</td>
<td>0.030 ± 0.014</td>
<td>0.005</td>
<td>0.081</td>
</tr>
<tr>
<td>HI</td>
<td>$0.42 \times 10^{-3} \pm 0.19 \times 10^{-2}$</td>
<td>$0.07 \times 10^{-2}$</td>
<td>0.012</td>
</tr>
</tbody>
</table>

N = 100

4. Discussion

4.1 Aluminum Level in Drinking Water

The use of aluminum sulphate as a coagulant in the water treatment process has major and substantial public health benefits. If there is an additional of certain material during the aeration process for acidic raw water, this process may influence aluminum levels in treated water. With the use of aluminum in water treatment, it is impossible not to have some low level of aluminum in treated water (Diaconu et al., 2009).

The concentration of a chemical in water may be reduced before the water reaches consumers – physical, chemical, and biological processes may reduce the concentration of particular chemicals between their sources and consumers (WHO, 2007b). The design and process operation at water treatment plant also influences the Al levels in treated water that is delivered to consumers. The addition of aluminum sulphate in raw water as a coagulant that is not removed during the treatment will remain as residual Al in the treated water (Srinivasan et al., 1999). This could cause the Al level in finished water to be higher than allowable levels as found in this study, where the lowest aluminum level detected was 0.63 mg/L. Based on Health Canada (2008), high contamination in raw water or inadequate pH control during treatment could also contribute to higher aluminum levels in drinking water.

In this study, there is possible excessive alum dosage used during the water treatment process, which has caused high aluminum residues in drinking water. The level of Al in this study was five times higher than reported in Qaiyum et al. (2011) and Aminah (2012). The mean aluminum concentration in two villages in Batu Pahat by Qaiyum et al. (2011) were 0.200 mg/L and 0.22 mg/L, and the mean aluminum concentration of villages in Kuala Terengganu by Aminah (2012) was 0.206 mg/L. The Al level in a study conducted by Rubinos et al. (2005) in Northwest Spain was varied from 0.008 to 0.650 mg/L. The varying differences of aluminum level in various places could be affected by different water sources being used.

4.2 Relationship between Aluminum and pH Value

Based on a study conducted by Srinivasan et al. (1999), there are a few important factors that determine the solubility of aluminum residue in finished water, which are pH, temperature and turbidity. From the study, aluminum was found soluble at extremely acidic (pH<6) and alkaline (pH>8.5) condition, but is insoluble at near natural pH values 7.0 to 7.5. In water treatment process, addition of aluminum sulphate will caused low pH in water, which is then suitable to coagulate raw water. Lime is added next to increase back the pH to neutral before being distributed as low pH will cause pipe erosion. However, in neutral pH, although the aluminum in water is insoluble, it remains as residue.

Based on the National Standard for Drinking Water Quality, the suitable pH for drinking water is 6.5 to 9.0. The range of pH in this study was 6.53 to 7.93, which are all within the acceptable limit. The results show a negative correlation between aluminum and pH level in drinking water samples. When the pH level of water reduces, the aluminum concentration will be increased. This trend was supported by Srinivasan et al. (1999).

4.3 Comparison of Aluminum Concentration and pH Level with Malaysian Drinking-water Standard

Based on the National Standard for Drinking Water Quality, the maximum level for aluminum in drinking water must not exceed 0.2 mg/L. All of the water samples in this study exceeded the standard by 3 to 7 folds, which was far higher from the standards and previous studies. This could be because of high aluminum dosage during coagulation process in water treatment, which caused high aluminum residue in drinking water. More aluminum sulphate will be added into raw water if the water source has high turbidity and microorganisms levels, and is highly contaminated with organic matters. Usually, the aluminum level in water treatment plant is higher than in raw water as alum will be added during the coagulation process. A study conducted by Diaconu et al. (2009) in Romania found that there was a variation of the aluminum level in water sample (raw
water, coagulation, filtration and drinking water) collected from water treatment plants. The highest aluminum level was at the coagulation process and getting less in drinking water.

4.4 Exposure Assessment

All of the respondents in this study had hazard index lower than 1 which an indication of negligible health risk. It is possible that the 0.2 mg/L limit value set by the Malaysian guideline and the US EPA are actually far safer than the actual safe limit value of aluminum level, as the study for safe limit value was not conducted adequately on humans, but rats (WHO, 1997). However, even though the HI shows an acceptable level of risk but exposure to Al in several studies was associated to several diseases. For example, a study by Rondeau et al. (2000) has found that aluminum level of more than 0.1 mg/L was associated with an elevated risk of dementia and Alzheimer’s disease. Other than that, there are several variables that need to be considered, which can affect the HI value such as the respondents CDI including aluminum concentration in water, average daily intake of water, and body weight. These variables were used to calculate the respondent’s CDI, which have the possibility to affect the HI as different respondents have different value of variables. When comparing CDI of aluminum intake with Reference Dose (RfD) which is 7 mg/kg/day, CDI for aluminum intake were far lower than RfD.

By comparing the CDI of this study with previous studies, there were apparent differences spotted. In this study, the CDI mean value was 0.030 mg/kg/day, while the previous study conducted in two villages in Kuala Terengganu by Aminah (2012) was 0.0057 mg/kg/day. Other study conducted by Qaiyum, Shaharuddin and Syazwan (2011) in Batu Pahat, the CDI mean value was 0.00619 mg/kg/day which almost the same with the CDI in Kuala Terengganu. This is clearly because the aluminum concentration in this study was far higher than the previous studies. The higher the aluminum concentration, the higher the CDI.

For the HI, both studies had determined that 100% of respondents had HI of lower than 1. However, there were slight differences between the studies. The HI in Batu Pahat were 0.00088 (Mukim Parit Lubok) and 0.00101 (Parit Raja), meanwhile in Kuala Terengganu was 0.0008. Another study conducted by Dzulfakar (2011) in Kuantan, both HI for Sungai Lembing and Bukit Ubi were 0.00053 and 0.00058, respectively. In this study, the HI was 0.0043, which was slightly higher than the HI of previous studies. Meanwhile, the reference dose (7 mg/kg/day) to calculate the HI was not adequately studied if there were differences between water and food ingestion. This possibly influence the HI level.

5. Conclusion

This study found that the mean aluminum concentration was 1.026 mg/L, which has exceeded the national standard of 0.2 mg/L by 5 folds, therefore left a significant difference between drinking water sample and the upper safe limit. However, the Hazard Index calculated from the findings did not exceed 1. This showed that the study area was considered safe from having risk related to diseases including Alzheimer’s and Parkinson’s diseases.

ACKNOWLEDGEMENT

We would like to thank the residents of Batu 7 Sandakan for giving their fullest cooperation in making this study a success. Also, gratitude and thanks to laboratory staff at the Department of Environmental and Occupational Health, Faculty of Medicine and Health Sciences, and Faculty of Sciences, Universiti Putra Malaysia.

CONFLICT OF INTEREST

The authors confirm that this article has not been published, part of or whole, in any other journals.

ETHICAL ISSUES

This study has received ethical approval from the Ethics Committee for Research involving Human Subjects (JKEUPM), Faculty of Medicine and Health Sciences, Universiti Putra Malaysia (Approval: UPM/TNCP1/RMC/1.4.18.1 (JKEUPM)/F2).

REFERENCES


