Exposure and Health (2019) 11:167–180
https://doi.org/10.1007/s12403-019-00297-w

S.I. : DRINKING WATER QUALITY AND PUBLIC HEALTH

Non-carcinogenic Health Risk Assessment of Aluminium Ingestion Via Drinking Water in Malaysia

Minhaz Farid Ahmed[1] · Mazlin Bin Mokhtar[1] · Lubna Alam[1] · Che Abd Rahim Mohamed[2] · Goh Choo Ta[1]

Received: 23 June 2018 / Revised: 27 December 2018 / Accepted: 5 January 2019 / Published online: 19 January 2019

Abstract

Exposure to aluminium (Al) is inevitable in the daily life because of its abundance in the environmental media through natural processes. Meanwhile, several studies have reported a positive association between Alzheimer's disease and a higher level of Al ingestion through drinking water. The present study is the first of its kind in Malaysia which predicts the human health risk of Al ingestion via drinking water at the Langat River Basin, Malaysia. Water samples were collected during 2015–2016 from the four stages of drinking water supply chain at the basin to determine the concentrations of Al by the Inductively Coupled Plasma Mass Spectrometry (ICP-MS). The determined mean concentration of Al in river 2.50E−01 ± 1.89E−01 mg/L crossed the Malaysian drinking water quality standard of 0.2 mg/L. The higher concentration of Al in the Langat River might be due to natural weathering of Al-bearing minerals. The mean Al concentrations in the treated water by the treatment plants, household's tap and after filtration water, respectively, were found to be within the Malaysian drinking water quality standard. This study suggests that there is no potential human health risk of Al ingestion through drinking water (HQ = 3.81E−03 ± 1.82E−03) at 95% confidence level in the basin because the hazard quotient (HQ) value is less than 1. However, the authorities need to be careful of excessive ingestion of Al via drinking water because the water treatment plants in the basin follow the conventional method to treat raw water. The turbidity in the tropical Langat River changes very frequently; thus, the doses of $Al_2(SO_4)_3$ for water disinfection are very crucial. Therefore, reverse osmosis technology can be introduced in the treatment plants because the United States Environmental Protection Agency has recommended that it can remove all types of metal > 90% from treated water.

Keywords Malaysia · Aluminium · Drinking water · Chronic daily intake · Alzheimer's disease

Introduction

Aluminium (Al) in the air, water and soil are mainly due to the natural weathering processes of Al-bearing minerals; however, in the surface water, the presence of Al is usually lower than 0.1 mg/L. Although the Al concentration in water is low, the acid mine drainage as well as acid rain can significantly increase the Al concentration in the surface water body (ATSDR 2008; WHO 1997). Moreover, the natural weathering of Al-enriched mineral is another vital source of Al^{3+} in the surface water and groundwater. Accordingly, the acidic environment also increases the mobility of Al content in the acidic water (i.e. pH < 4) through the dissolution of Al-bearing minerals such as gibbsite as well as the natural weathering of clay and rock minerals (WHO 1997). Hydrous aluminium in clay mineral (Aris et al. 2014) and erosion of ferralsols (i.e. oxisols and ultisols) enriched with Al (Lim et al. 2013a, b) in the Langat River Basin attributes to the higher dissolved concentration of Al in the Langat River. Moreover, the excess use of $Al_2(SO_4)_3$ (aluminium sulphate) in the conventional flocculation and coagulation method of tropical river water disinfection might have also contributed to the higher level of Al concentration in the supply water at the household level in the Langat River Basin Malaysia (Hajira and Shaharuddin 2015). Therefore, the Malaysian National Drinking Water Quality Standard (NDWQS) along with the World Health Organization (WHO) has fixed the

✉ Mazlin Bin Mokhtar
mbmlestari@hotmail.com; mazlin@ukm.edu.my

[1] Institute for Environment and Development (LESTARI), Universiti Kebangsaan Malaysia (UKM), 43600 Bangi, Selangor Darul Ehsan, Malaysia

[2] School of Environmental and Natural Resource Sciences, Universiti Kebangsaan Malaysia (UKM), 43600 Bangi, Selangor Darul Ehsan, Malaysia

maximum limit of Al at 0.2 mg/L in the drinking water (MOH 2010; WHO 2017). United States Environmental Protection Agency (USEPA) has reported that Al ingestion via drinking water does not possess carcinogenic human health risk at the secondary maximum contaminant level, i.e. 0.05–0.2 mg/L (USEPA 2017). However, USEPA has listed Al in the secondary water contaminant category in order to provide some guideline to reduce higher Al concentration from the supplied water at household level. The higher Al concentration in the supplied drinking water might stop people using water from their public water system. The non-carcinogenic human health risk can be estimated by hazard quotient (HQ) of Al ingestion via drinking water. If the calculated HQ of Al ingestion via drinking water is higher than 1, then it indicates significant non-carcinogenic human health risk. Meanwhile, a few studies have reported the non-carcinogenic health risk such as Dementia or Alzheimer's disease associated with the higher level of Al ingestion via drinking water (Lina and Shaharuddin 2017; Li et al. 2017, 2018). Similarly, kidney dysfunction, bone or brain diseases are also thought to be associated with the long-term Al ingestion via drinking water (ATSDR 2008).

Al halogenides, Al hydride and lower aluminium alkyls react strongly in water due to its physiochemical and geological characteristics (WHO 1997). Al is abundant in the earth crust, which accounts for higher than 8%, it is highly present in the soil-derived dust, especially in the aluminium silicate clay (Lim et al. 2012, 2013a; WHO 1997). Other anthropogenic activities such as mining, agriculture, coal combustion are also the examples of aluminium-enriched dust release to the environment. Therefore, the increased mobilization of Al in the water is mainly due to the acidic environment consequences from the anthropogenic activities. Usually, Al is found in monomeric form in acidic water that increases its mobility and bioavailability. However, dissolved Al concentration can be found in both the monomeric and polymeric forms along with the organic and inorganic ligands. Hence, the Al speciation is determined not only by the pH level in water but also with the dissolved organic carbon (DOC), fluoride, sulphate, phosphate and suspended particulates (WHO 1997).

Therefore, Al ingestion through drinking water and food is apparent, and WHO (1997) has already reported that humans absorb about 3%, approximately 0.4 mg/day total daily uptake of Al via drinking water. However, human health problems such as nausea, vomiting, diarrhoea, mouth ulcers, skin ulcers, skin rashes, arthritic pain and other related diseases were reported in England where around 20,000 populations were accidentally exposed to a higher level of Al through the supply of water. Similarly, two out of twenty epidemiological studies globally confirmed the significant association between Al ingestion via drinking water and Alzheimer's disease (WHO 1997; Martyn et al. 1997). However, there is no significant study in Malaysia that has determined the human health risk of Al ingestion via drinking water. Therefore, this study predicts the human health risk of Al ingestion through drinking water at the Langat River Basin and suggests better management of drinking water.

Study Area

UNESCO HELP (Hydrology for Environment, Life and Policy) Langat river basin is situated at the west part in peninsular Malaysia and it covers about 2350 km^2 along its course of 200 km (UNESCO 2010). Langat River is unique in characteristic since it drains through three different constituency and the basin shares Selangor State (78.14%), Negeri Sembilan State (19.64%) and the Federal Territories of Kuala Lumpur (0.33%) and Putrajaya (1.90%) (DOE 2003). The major tributaries of Langat River are the Semenyih, Beranang and Labu river (Fig. 1); however, there are about 40 smaller tributaries of Langat River (LUAS 2013) and the basin receives an average annual rainfall of 1500 to 2900 mm (Lim et al. 2012). The total populations at Langat River Basin were 1,184,917 in 2000 with a growth rate of 7.64% (Elfithri 2016a, b). The populations in the basin increased to 4,065,023 during 2013 (DOS Census 2013). Langat River is one of the prime drinking water sources for the basin and it provides water to half of the population in the Selangor State of Malaysia (Ahmed et al. 2016a, b).

Lembaga Urus Air Selangor (LUAS 2013) has already identified the location of point sources of pollution in the Langat River Basin (Fig. 1). There are several industrial zones within the basin along with the Nilai industrial zone in the Negeri Sembilan State. Hence, the discharge of the unregulated effluent in the environment is a real threat of the water pollution due to the lack of implementation of Environmental Quality Act, 1974. Juahir et al. (2011) have also reported that sewage discharges (10.80%) and industrial discharges (9.09%) are the potential chemical pollution in the Langat river. The undisturbed upstream water sampling stations were covered with the thick tropical forest. The shifting cultivation is also observed in the Sungai Pangsoon, Sungai Lolo, Sungai Serai to Sungai Langat areas. This area is considered as an unpolluted upstream area of Langat River (Yap 2013). The upstream area of Langat River Basin is also a place of native people of Hulu Langat (i.e. villages of indigenous people) who are highly dependent on the river for washing and bathing purposes. The common characteristics of water sampling stations 1–3 (i.e. Pangsoon, Lolo and Serai) were the presence of cobbles and pebbles and sometimes sand and gravel, while the water sampling point 4 (i.e. Langat) was moderately polluted by the land clearance, agriculture activities as well as slightly industrial effluent discharges.

Fig. 1 Location of point sources of pollution at the Langat River Basin Malaysia Modified from LUAS (2013)

Cheras Mile 11, Bukit Tampoi, Salak Tinggi and Sungai Labu water treatment plants (WTPs) belong to the polluted downstream area of the Langat Basin. The water sampling point 5 (i.e. Cheras Mile 11) receives a massive amount of point and non-point sources of pollution because of its location in the built-up area (i.e. residential and industrial) along with the cultivation, rubber and palm oil plantation. On the other hand, the Bukit Tampoi (i.e. water sampling point 6) and Salak Tinggi WTP (i.e. water sampling point 7) areas accumulate all the significant residential, industrial, agricultural, mining wastes from Kajang, Bangi, Dengkil and Semenyih areas of Selangor State, while Sungai Labu WTP (i.e. water sampling point 8) area was also experiencing higher pollution from Nilai Industrial Zone of the Negeri Sembilan State as well as the palm oil and residential waste from Sepang, Selangor State (Ahmed et al. 2016a). In these three water sampling locations (i.e. Bukit, Salak and Labu), the salinity is higher compared to other points in the Langat River because of its proximity to the Strait of Malacca. Furthermore, the colour of the water from the Langat WTP area to Salak Tinggi WTP area is light brownish to deep brownish mainly due to the pollution from sewage and oil palm plantation (Yap 2013). Accordingly, the Dengkil WTP area have many manufacturing and metal finishing industries along with the vast oil palm plantation area, while the surface water of Salak Tinggi area is highly polluted of the inorganic wastes from the paper-based industries. The Bangi and Semenyih areas of the Langat River are polluted from the ex-mining as well as the sand and gravel extraction activities (Ahmed et al. 2016a). Kim (2012) reported that the discharges of Al-enriched wastewater in the Langat River by the water treatment plants are one of the major sources of higher Al concentration in the river.

Methods

Physico-chemical Analysis

A total of 46 water samples were collected from the drinking water supply chain at the Langat River Basin, Malaysia. Three replicates of each water sample were collected during 2015–2016 from eight stations in the Langat River precisely from where the eight water treatment plants (WTPs) in the basin collect water for drinking water treatment purposes (Fig. 2). Samples were also collected from the outlet of those eight WTPs. Supply water samples were collected from the kitchen's tap of fifteen households at the Langat Basin. Similarly, household filtration water samples were collected from the same fifteen households of the basin. These fifteen households at the Langat Basin were selected based on commonly use of five types of household water filtration system i.e. carbon water filter, reverse osmosis water filter, alkaline water filter, ultraviolet water filter and distilled water filter. In situ physiochemical water quality parameters such as dissolved oxygen (DO), conductivity (SPC), salinity and pH were also recorded at the river sampling points using the calibrated Professional Plus Water Quality Instrument (6050000, YSI Incorporated, USA). Chelex 100 resin column ion exchange method was applied to analyse dissolved Al in the water samples (Daud and Mohamed 2013; Paulson et al. 1991) by the Inductive Coupled Plasma-Mass Spectrometry (ICP-MS).

Health Risk Assessment

To assess the human health risk of metal (i.e. Al) ingestion through drinking water, the model of chronic daily intake (CDI) of chemicals (Eq. 1) and non-carcinogenic hazard quotient (HQ) (Eq. 2) based on chemical ingestion through drinking water were established by the United States Environmental Protection Agency (USEPA 1989, 1991), where Cdw refers to Al (1.21E−01 ± 5.75E−02 mg/L) concentration in drinking water, IR refers to water ingestion rate (1.996 L/Day) from questionnaire survey, EF refers to exposure frequency (365 days/year), ED refers to exposure duration (74 years), BW refers to Body Weight (63.193 kg) from questionnaire survey, AT refers to average time (27,010 days) and RfD refers to oral reference dose (1.00 mg/kg-day) (USEPA 1991; Ab Razak et al. 2016). This model has already been widely used by scholars and institutes (Adimalla and Li 2018; Li et al. 2016; Wu and Sun 2016; Wu et al. 2017; Zhang et al. 2018). The risk can be calculated as follows:

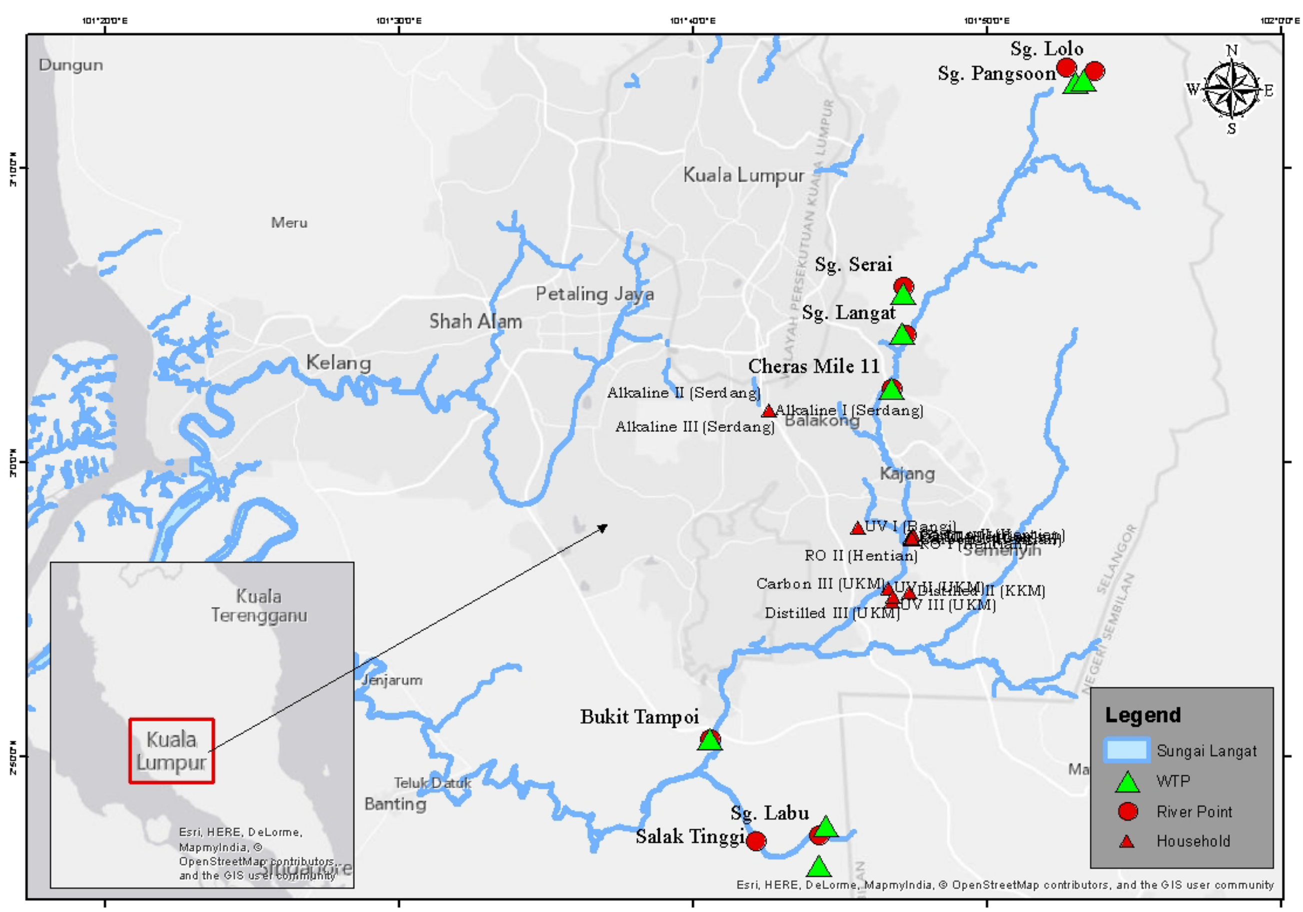

Fig. 2 Water sampling locations at the Langat River Basin, Malaysia

$$\mathrm{CDI}\left(\frac{\mathrm{mg}}{\mathrm{kg}}\Big/\mathrm{day}\right) = \frac{\mathrm{Cdw}\left(\frac{\mathrm{mg}}{\mathrm{L}}\right)\times \mathrm{IR}\left(\frac{\mathrm{L}}{\mathrm{day}}\right)\times \mathrm{EF}\left(\frac{\mathrm{day}}{\mathrm{year}}\right)\times \mathrm{ED\ (years)}}{\mathrm{BW(kg)}\times \mathrm{AT(days)}} \tag{1}$$

$$\mathrm{HQ} = \frac{\mathrm{CDI}\left(\frac{\mathrm{mg}}{\mathrm{kg}}\Big/\mathrm{day}\right)}{\mathrm{RfD}\left(\frac{\mathrm{mg}}{\mathrm{kg}}\Big/\mathrm{day}\right)}. \tag{2}$$

Household Questionnaire Survey

According to the latest population census by the Department of Statistics Malaysia, the total number of households are 1,494,865 (DOS 2013) at the Langat River Basin, Malaysia. Therefore, 402 household questionnaire survey was conducted randomly at the basin using Eq. 3 (Yamane 1967; Alam 2014) to get the average daily drinking water intake by the population at the basin as well as their body weight to calculate the CDI of Al (Eq. 1).

$$n = \frac{N}{1 + N(e)^2}, \tag{3}$$

where n is sample size; N is population size; e is level of precision, i.e. 0.05 at 95% confidence level.

Results and Discussions

Aluminium Concentration in the Langat River

The higher dissolved concentration of Al in the upstream water sources of Langat River is mostly from the natural weathering of iron and silica bedrock in Titiwangsa Granite Hill Range along the tropical climatic conditions. The soil properties of Hulu Langat in the Langat River Basin—comprised of 10% sand, 60% silt and 30% clay—enriched with the aluminosilicate minerals within 2 m from the surface soil (Fasihnikoutalab et al. 2016). Therefore, the highest mean concentration of dissolved Al was recorded in the upstream Serai point 556.9 ± 95.54 μg/L (Table 1) in the range of 457.87 – 648.53 μg/L followed by Lolo point 529.96 ± 70.45 μg/L (range: 459.53 – 600.4 μg/L) and Pangsoon point 231.17 ± 128.59 μg/L (range: 131.61 – 376.35 μg/L) in the upstream that are undisturbed forest areas with abundance of sand, gravel and pebbles in the streams following to the main channel of Langat River. There is a total of 43 earth material, 21 granite quarries, and 86 sand and gravel extraction sites in the Langat River Basin (Ahmed et al. 2016a; Juahir 2009). Therefore, these mineral extraction sites also contribute to the higher concentration of Al in the Langat River.

Similarly, the highest dissolved oxygen (DO) 9.44 mg/L at Pangsoon followed by the higher DO at Lolo 7.84 mg/L and Serai 8.72 mg/L points (Fig. 3) and relatively lower conductivity in the upstream stations than the downstream stations indicate the higher biological activities by the phytoplankton (i.e. diatoms). The higher biological activities attribute to release much oxygen in the clean water to dissolute Al from the aluminosilicate river bedrock (Lim et al. 2013a, b). The hydrous aluminium in the clay minerals (i.e. magnesium-aluminosilicate) of the basin (Aris et al. 2014; Fasihnikoutalab et al. 2016) and erosion of ferralsols (i.e. oxisols and ultisols) enriched with Al (Lim et al. 2013a) attribute to the higher dissolved concentration of Al in the Langat River apart from the man-made activities.

The higher Al concentration in the upstream of Langat River might also have influenced with the lower pH through flocculation and conservative behaviour than the mid to downstream of the river. The decreasing trend of Al (i.e. $R^2 = 0.47$; Fig. 3) specially in the mid to downstream (i.e. Langat to Labu point) might be because of adsorption of $Al(OH)^{4-}$ onto the suspended particulate matter as well as the bacterial reduction to dissolute Al in slightly alkaline water (i.e. pH ranges 7.61 – 8.72) than the upstream of the river. Similarly, the significant, increasing trend of salinity $R^2 = 0.73$ (Fig. 3) along with the sharp decreasing trend of dissolved oxygen (DO) $R^2 = 0.91$ (Fig. 3) under the

Table 1 Al (μg/L) and physiochemical parameters in several locations of Langat River

Location	Al (μg/L)	DO (mg/L)	SPC (μS/cm)	SAL (psu)	pH
Pangsoon	231.17 ± 128.59	9.44 ± 0.05	35.53 ± 0.51	0.02 ± 0.01	7.92 ± 0.11
Lolo	529.96 ± 70.45	7.84 ± 0.30	50.40 ± 1.37	0.02 ± 0.00	7.75 ± 0.01
Serai	556.90 ± 95.54	8.72 ± 0.20	16.67 ± 0.06	0.01 ± 0.00	7.76 ± 0.02
Langat	214.48 ± 2.92	7.21 ± 0.15	60.07 ± 0.06	0.03 ± 0.00	8.56 ± 0.07
Cheras	144.18 ± 36.68	6.52 ± 0.03	138.77 ± 0.64	0.06 ± 0.00	8.45 ± 0.10
Bukit	104.09 ± 66.91	5.57 ± 0.19	179.37 ± 0.15	0.08 ± 0.00	7.97 ± 0.21
Salak	143.68 ± 46.45	4.60 ± 0.28	122.30 ± 3.46	0.05 ± 0.01	8.17 ± 0.13
Labu	77.65 ± 41.42	2.12 ± 0.03	155.13 ± 0.15	0.07 ± 0.00	7.61 ± 0.04
Average	250.26 ± 61.12	6.50 ± 0.15	94.78 ± 0.80	0.04 ± 0.00	8.02 ± 0.09

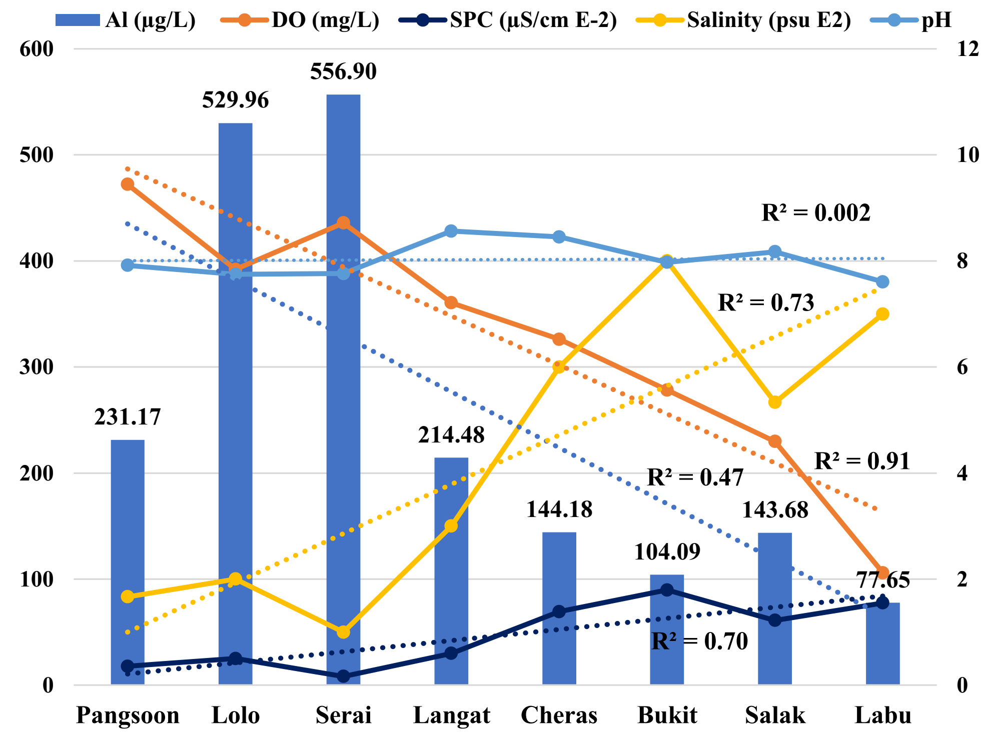

Fig. 3 Al (µg/L) concentration from upstream to downstream in the Langat River, Malaysia (2015)

slightly alkaline condition indicates adsorption of Al that is also influenced by the bacterial reduction from upstream to downstream. Therefore, the lowest concentration of Al was observed at the downstream Labu point 77.65 ± 41.42 µg/L in the range of 38.09–120.70 µg/L, where slightly alkaline water pH 7.61 ± 0.04 (Fig. 3) and little biological activities (i.e. lowest DO = 2.12 ± 0.03 mg/L) as well as higher electrical conductivity 155.13 ± 0.15 µS/cm (Fig. 3) influenced the sorption of authigenic aluminosilicate formation (Mackin and Aller 1984). The conservative behaviour of Al in the slightly alkaline water along with the comparatively higher salinity (0.07 psu) at the Labu point than the other points of Langat River indicate the sorption and deposition process of aluminosilicate formation (Wang et al. 2015). Therefore, the decrease in Al concentration from mid to downstream areas might be because of adsorption of Al as well as the formation of authigenic aluminosilicate with the increase of salinity towards downstream.

Aluminium Concentration in Drinking Water Supply Chain

Mean aluminium (Al) concentrations in the treated water at the outlets of water treatment plants (WTPs), tap and filtration water at the household level in the Langat Basin were within the drinking water quality standard of the Ministry of Health (MOH) Malaysia 0.2 mg/L, WHO 0.2 mg/L and USEPA 0.087 mg/L; however, the maximum Al concentration in the household's tap 2.25E−1 mg/L and filtration 2.71E−1 mg/L water crossed the drinking water quality standard of Al concentration (Table 2). The higher Al concentration in the tap water might be due to the corrosion of piping structure causing the partial solubilization of Al in the water distribution system (Farizwana et al. 2010; Mora et al. 2009). Similarly, the higher Al concentration in household filtration water might be due to poor management of filtration system. The mean Al concentration in Langat River

Table 2 Al (mg/L) in drinking water supply chain at Langat Basin Malaysia

Sample	Min.	Max.	Mean	Std.	Skewness	Kurtosis	MOH[a]	WHO[b]	USEPA[c]
River	3.81E−2	6.49E−1	2.50E−1	1.89E−1	0.74	0.66	0.2	0.2	0.087
WTP	5.16E−2	1.98E−1	1.31E−1	4.86E−2	0.05	−1.38			
Tap	7.19E−2	2.25E−1	1.48E−1	4.66E−2	−0.26	−1.50			
Filter	2.11E−2	2.71E−1	1.21E−1	5.75E−2	0.96	−0.48			

[a]Drinking water quality standard proposed by Ministry of Health Malaysia (MOH 2010)
[b]Guidelines for drinking water quality proposed by World Health Organization (WHO 2017)
[c]Toxic reference value proposed by the United States Environmental Protection Agency (USEPA 1999)

2.50E−01 ± 1.89E−01 mg/L crossed the stipulated drinking water quality standard of MOH, WHO and USPEA mainly due to the natural weathering of hydrous aluminium in the clay mineral (Aris et al. 2014) as well as discharge of aluminium-enriched sludge by the WTPs in the environment at the Langat Basin (Kim 2012). The skewness (< 1) and kurtosis (< 1) analysis of Al concentration data of the river, treated water, tap and filtration water indicate normal distribution of the data (Table 2).

Figure 4 describes the conventional water treatment process at the Langat Basin since all the WTPs within the basin follow the conventional method in treating drinking water.

Raw Water

The changes in physical–chemical characteristics of the raw water are subject to seasonal variation. Therefore, it is very important to monitor the quality of raw water continuously to optimize the treatment process as well as to anticipate the changes. The quantity and the visual quality of the raw water should also be monitored regularly. These parameters are important to determine the quality of the chemical to be dosed for the treatment and this is the only means to detect any significant changes in raw water quality.

Intake Area

The raw water channelled through course screen and grit chamber before flowing into the pumping area. It is then pumped into the aeration chamber through the raw water pipe.

Chemical Treatment

Rapid dispersion of coagulant is accomplished by the hydraulic mixing at the inlet wire. Gentle agitation of the raw water that has been dosed with aluminium sulphate flows through mixing channel. The mixing channel contains wooden baffles that provide gentle agitation to promote the growth of the floc particles. The purpose of the sedimentation tank is to reduce the amount of suspended particles in the water by retaining the water within the tank in order to deposit the settleable floc at the base of the tank. As the water flows into the sedimentation tanks, the water then enters the lower portion of the LOVO type of sedimentation tanks where the sludge can be settled. The settle water then flows into the settle water channel.

Filter

The purpose of filtration is to strain the suspended particle and remove colloidal clay, colouring matter and bacteria that may still present in the settle water.

Liquid Chlorine

Liquid chlorine is delivered as a liquid form under 68 kg steel cylinder pressure and no preparation is required to dose it directly into the pipeline through using gas chlorinator system. Chlorine should be dosed to achieve a free residual of between 1.5 and 2.5 mg/L. Fluoride in the form of sodium silico fluoride is dosed in the range of 0.4–0.6 mg/L to prevent dental problems.

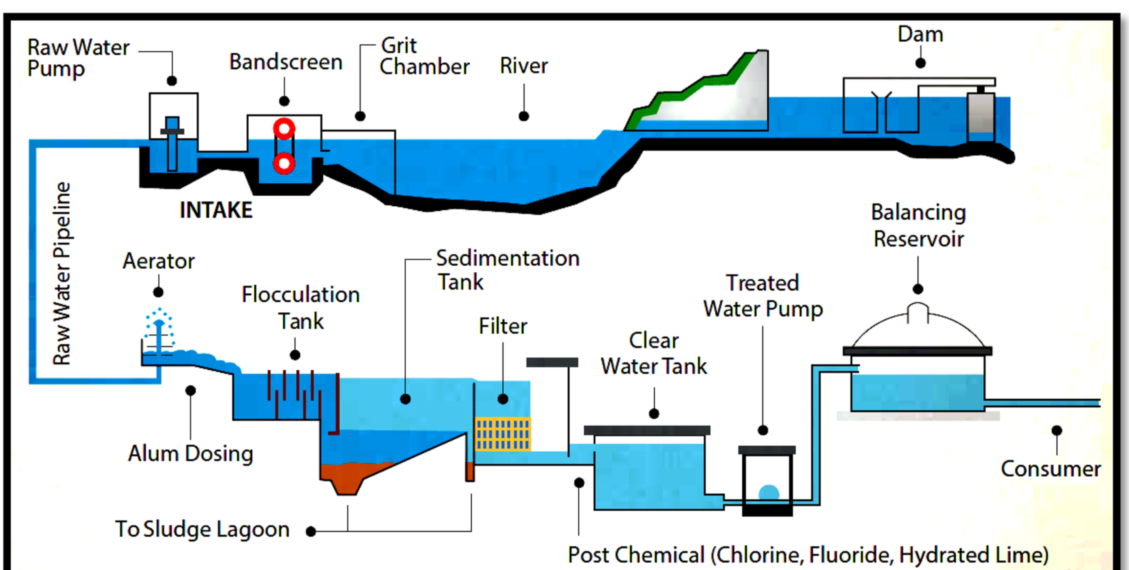

Fig. 4 Typical Conventional Treatment Plants' Process Flow Chart (PASSB). Performance Report for 2015

pH Correction in Treated Water

Hydrated lime is dosed to reduce the aggressive characteristic of the water especially the acidity. The lime slurry is dosed by gravity inside the clear water tank.

Balancing Reservoir

Treated water is finally stored in a reservoir before supplying to the consumer.

The mean dissolved Al concentrations in treated water (131.43 ± 48.56 µg/L), tap water (148.31 ± 46.93 µg/L) and household filtration water (120.68 ± 57.5 µg/L) (Table 3) were within the Malaysian drinking water quality standard of 200 µg/L. The carbon water filtration system at the household level in the Langat Basin was found to be very poor in removing Al concentration from the supply water. For instance, higher concentrations of dissolved Al were determined in the Carbon I, II and III filtration water than the supply water. The prime reason for the higher concentration of dissolved Al in the filtration water is the accumulation of Al in the cartridge along with lack of awareness of consumers to clean the filtration system regularly. Moreover, carbon filter (i.e. particulate filtration) can remove particle size about 1 µm, whereas Al ions may be < 0.0001 µm (Aquathin 2017). Theoretically reverse osmosis (RO) filtration system has the highest efficiency to remove all types of metal ions including Al ions (i.e. about 0.0001 µm). However, the higher concentration of Al in the filtration water by RO III is higher than the concentration of supply water which might be due to the expiring of the cartridge.

Health Risk Assessment via Aluminium Ingestion

Aluminium (Al) concentration (Cdw) in the drinking water supply chain i.e. river water (2.50E−01 ± 1.89E−01 mg/L; $t = 6.48$; $p = 1.0E{-}06$), treated water by WTP (1.31E−01 ± 4.86E−02 mg/L; $t = 13.26$; $p = 2.95E{-}12$), household's tap water (1.48E−01 ± 4.66E−02 mg/L; $t = 21.37$; $p = 7.30E{-}25$) and household's filtration water (1.21E−01 ± 5.75E−02 mg/L; $t = 14.8$; $p = 6.53E{-}18$) in 2015 at Langat Basin (Table 4) were significantly (95% level) within the maximum limit 0.2 mg/L of drinking water quality standard of Ministry of Health Malaysia (MOH 2010) and World Health Organization (WHO 2017), respectively. The chronic daily intake (CDI) of Al through river (7.90E−03 ± 5.98E−03 mg/kg-day), treated (4.15E−03 ± 1.53E−03 mg/kg-day), tap (4.68E−03 ± 1.47E−03 mg/kg-day) and household filtration (3.81E−03 ± 1.82E−03 mg/kg-day) water in 2015 (Table 4) were also within the maximum tolerable daily intake of Al via drinking water for adult at 0.1 mg/kg-day recommended by the United States Department of Agriculture (ATSDR 2017). Therefore, Al ingestion through drinking water at the Langat Basin has no potential health hazard, because the HQ for river water (7.90E−03 ± 5.98E−03; $t = 6.48$; $p = 1.0E{-}06$), treated water (4.15E−03 ± 1.53E−03; $t = 13.26$; $p = 2.95E{-}12$), tap water (4.68E−03 ± 1.47E−03; $t = 21.37$; $p = 7.30E{-}25$) and household filtration water (3.81E−03 ± 1.82E−03; $t = 14.08$; $p = 6.53E{-}18$) in 2015 (Fig. 5 and Table 4) were significantly within the safe limit i.e. HQ < 1 at 95% confidence interval.

Unfortunately, the Ministry of Health Malaysia (MOH) does not monitor the river water quality for Al concentration

Table 3 Al (µg/L) concentration in treated, tap and household filtration water at Langat River Basin, Malaysia

WTP	Al (µg/L)	Tap water	Al (µg/L)	Household filtration water	Al (µg/L)
Pangsoon	184.15 ± 13.59	Serdang I	109.05 ± 48.35	Alkaline I (Serdang I)	105.45 ± 15.7
Lolo	175.5 ± 3.79	Serdang II	103.95 ± 48.72	Alkaline II (Serdang II)	31.13 ± 10.03
Serai	74.93 ± 23.34	Serdang III	110.82 ± 0.21	Alkaline III (Serdang III)	163.33 ± 8.92
Langat	103.87 ± 26.92	Hentian Kajang I	109.93 ± 10.36	RO I (Hentian Kajang I)	58.57 ± 2.01
Cheras	169.55 ± 15.61	Hentian Kajang II	166.35 ± 10.63	RO II (Hentian Kajang II)	137.22 ± 32.1
Bukit	161.71 ± 31.06	Hentian Kajang III	169.50 ± 7.31	RO III (Hentian Kajang III)	174.71 ± 11.9
Salak	114.67 ± 34.03	Hentian Kajang IV	138.05 ± 62.57	Carbon I (Hentian Kajang IV)	138.72 ± 36.3
Labu	67.02 ± 8.78	Hentian Kajang V	106.85 ± 15.63	Carbon II (Hentian Kajang V)	262.54 ± 8.03
Average	131.43 ± 48.56	Hentian Kajang VI	192.10 ± 22.08	Carbon III (UKM II)	128.13 ± 32.2
		Hentian Kajang VII	188.46 ± 4.28	Distilled I (UKM III)	74.41 ± 22.95
		UKM I	194.19 ± 12.20	Distilled II (Hentian Kajang VI)	121.62 ± 2.73
		UKM II	109.40 ± 23.53	Distilled III (UKM I)	88.11 ± 8.34
		UKM III	108.30 ± 19.38	UV I (Bangi I)	146.21 ± 34.7
		UKM IV	195.36 ± 3.09	UV II (UKM IV)	87.54 ± 40.80
		Bangi I	222.28 ± 21.43	UV III (Hentian Kajang VII)	92.51 ± 10.60
		Average	148.31 ± 46.93	Average	120.68 ± 57.5

Table 4 T test of Al concentration, HQ and LCR for household filter water in 2015

Water sample	Parameter	Mean	Std. deviation	*t* value	*p* value	95% confidence interval of the difference	
						Lower	Upper
River	Cdw (mg/L)	2.50E−01	1.89E−01	6.48	1.00E−06*	1.70E−01	3.30E−01
	CDI (mg/kg-day)	7.90E−03	5.98E−03	6.48	1.00E−06*	5.38E−03	1.04E−02
	HQ	7.90E−03	5.98E−03	6.48	1.00E−06*	5.38E−03	1.04E−02
WTP	Cdw (mg/L)	1.31E−01	4.86E−02	13.26	2.95E−12*	1.11E−01	1.52E−01
	CDI (mg/kg-day)	4.15E−03	1.53E−03	13.26	2.95E−12*	3.50E−03	4.80E−03
	HQ	4.15E−03	1.53E−03	13.26	2.95E−12*	3.50E−03	4.80E−03
Tap	Cdw (mg/L)	1.48E−01	4.66E−02	21.37	7.30E−25*	1.34E−01	1.62E−01
	CDI (mg/kg-day)	4.68E−03	1.47E−03	21.37	7.30E−25*	4.24E−03	5.13E−03
	HQ	4.68E−03	1.47E−03	21.37	7.30E−25*	4.24E−03	5.13E−03
Filter	Cdw (mg/L)	1.21E−01	5.75E−02	14.08	6.53E−18*	1.03E−01	1.38E−01
	CDI (mg/kg-day)	3.81E−03	1.82E−03	14.08	6.53E−18*	3.27E−03	4.36E−03
	HQ	3.81E−03	1.82E−03	14.08	6.53E−18*	3.27E−03	4.36E−03

*Significant at 95 % confidence interval

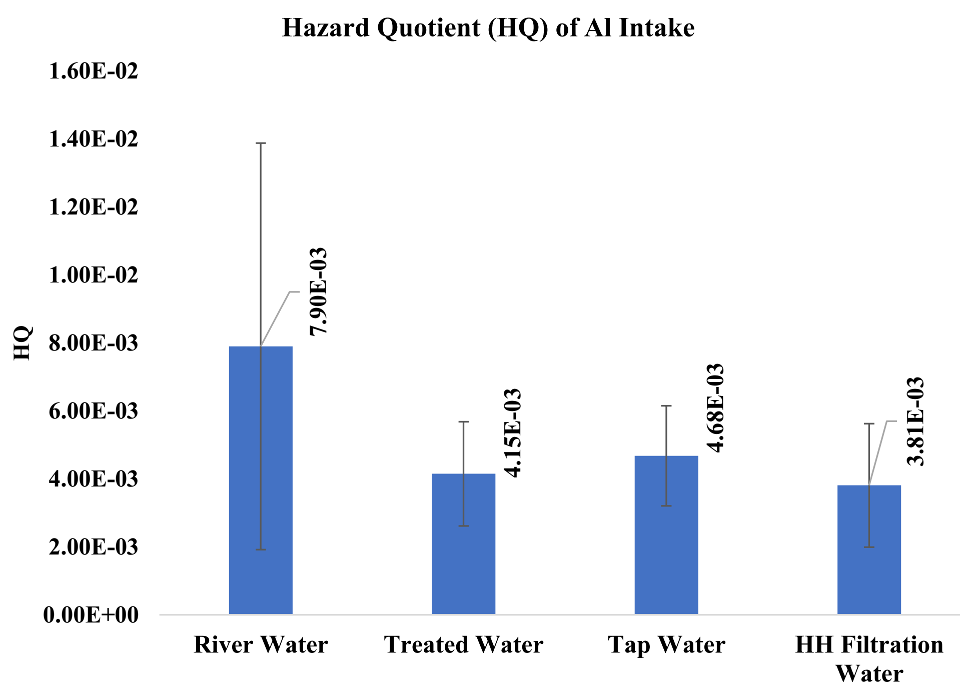

Fig. 5 HQ of Al Intake via Drinking Water at Langat River Basin, Malaysia

as a requirement of drinking water quality standard, although MOH has fixed the maximum Al concentration at 0.2 mg/L in the treated water (MOH 2010). Al concentration in Langat River during 2001 was 0.38 mg/L (Yusuf 2001); however, in 2015, the Al concentration in Langat was determined to be 2.50E−1 ± 1.89E−1 mg/L, showing a decrease of 35.87% within these 15 years. Hence, it can be assumed that in the next 5 years i.e. 2020, the Al concentration in Langat will be decreased to a total of 47.43% compared to the concentration in 2001. Thus, the predicted Al concentration in 2020 at Langat River will be 0.18 mg/L, which is within the Malaysian national drinking water quality standard at 0.2 m/L. Although Al concentration in Langat River is decreasing with time, however, the turbidity in the river is changing very frequently mainly because of the flash floods in the tropical climate, and the pollution from agricultural and industrial zones (Ibrahim et al. 2015; Al-Badaii et al. 2013). Therefore, the doses of $Al_2(SO_4)_3$ is very crucial for the treated water disinfection by the water treatment plants (WTPs) in the Langat Basin. The conventional method of WTPs in the Langat Basin is unable to fully remove the Al concentration in the treated water, however, the reverse osmosis membrane technology is capable of removing metal ions > 90% in treated water (USEPA 2005). The sharp decrease of dissolved Al concentration in Langat River from upstream to downstream might be because of adsorption of Al as well as the formation of authigenic aluminosilicate with the increase of salinity towards downstream. Contrary, Kim (2012) reported that the treatment plants of the Langat Basin are the major sources of Al-enriched wastewater discharges in the environment. Therefore, the Selangor Water Management Authority (LUAS) has started to monitor the point sources of pollution as well as to reduce the river pollution through forming 'Pollution Control Taskforce' which consists of several agencies (LUAS 2017). These activities might have also contributed to the sharp decrease of dissolved Al concentration towards downstream in the Langat River. Similarly, the Al concentration in drinking water (i.e. from household water filtration) at Langat River Basin in 2015 was determined $1.21E-1 \pm 5.75E-2$ mg/L. Therefore, the predicted Al concentration in drinking water at Langat Basin will be 0.11 mg/L in 2020 due to a decrease of 11.96% compared to 2015. Therefore, the calculated HQ both for the river 5.69E−03 and household filtration water 3.47E−03 in 2020 at the Langat Basin is within the safe limit since the HQ < 1.

Table 5 Demographic characteristics of Langat River Basin, Malaysia

	N	%
Age group		
18–25 years old	176	43.8
26–35 years old	144	35.8
36–45 years old	53	13.2
46–55 years old	22	5.5
> 55 years old	7	1.7
Total	402	100.0
Gender		
Male	160	39.8
Female	242	60.2
Total	402	100.0
Education		
SPM or below	76	18.9
Diploma	79	19.7
Degree	177	44.0
Graduation	57	14.2
Other	13	3.2
Total	402	100.0
Monthly household income		
< 2000 MYR	155	38.6
2001–4000 MYR	140	34.8
4001–6000 MYR	62	15.4
6001–10,000 MYR	26	6.5
> 10,000 MYR	19	4.7
Total	402	100.0
Use of household filtration system		
Yes	259	64.4
No	143	35.6
Total	402	100.0

Total ($N = 402$)

Association between Alzheimer's disease and Aluminium ingestion

Association between Aluminium (Al) in drinking water and human's Alzheimer's disease (AD) has been reported through several epidemiological studies in Norway (Flaten 1990), England (Martyn et al. 1997), France (Rondeau et al. 2000), North America (Flaten 2001), Canada (Meshitsuka et al. 2002), Japan (Kawahara and Kato-Negishi 2011) and Malaysia (Dzulfakar et al. 2011; Ab Razak et al. 2016; Hajira and Shaharuddin 2015). Some of the studies have also determined a higher amount of Al in the brain, neurofibrillary tangles, and senile plaques of humans who are suffering from dementia and other neurological diseases (ADFM 2017). It is believed that the long-term Al ingestion via drinking water by human beings even at a lower concentration could damage the central nervous system and skeletal as well as the haematopoietic systems (ATSDR 2008). Therefore, it is strongly assumed that Al ingestion via drinking water has a positive association with the human's Alzheimer's disease.

Among the surveyed population in the Langat River Basin, more than 98% populations were within 18 to 55 years (Table 5) and the number of female respondents were 60.2%. It is also interesting to note that 44% respondents have completed degree and the awareness to use household filtration system is 64.4%, although maximum 38.6% household's monthly income is less than MYR 2000. Moreover, among the surveyed populations, 132 (32.8%) respondents reported that at least one member of their household is suffering from critical disease (Table 6). It was also found from the survey that 1% population in the Langat River Basin is suffering from Alzheimer's Disease and in Malaysia the mortality rate of Alzheimer's Disease is 10.8 per 100,000 populations (GRAPHIQ 2017).

Although 67.2% population in the basin is not suffering from any major diseases (Table 6), however, the remaining 32.8% population's diseases were categorized based on their use of household drinking water filtration system. It was found that the use of filtration system at household has no influence over the Alzheimer's Disease in the basin. Since 75% populations were suffering from Alzheimer's disease although they are using filtration system (Table 7), it indicates poor efficiency of filtration system to remove Al from drinking water.

Table 6 Types of diseases that people are suffering at the Langat River Basin, Malaysia

Type of disease	Number of households[a] (%)
Leukaemia	4 (1.0)
Liver cancer	2 (0.5)
Alzheimer's disease	4 (1.0)
Diabetes	72 (17.9)
Kidney failure	4 (1.0)
Other[b]	46 (11.4)
Total	132 (32.8)
No major diseases	270 (67.2)
Total	402 (100)

Other diseases refers to fever, skin diseases, etc. and in some cases lost memory of elder members in the family which they do not think as a disease

[a]At least one member in the household is suffering from diseases

In Malaysia, the river provides almost 98% of the raw water requirement for drinking purposes (Leong et al. 2007; Santhi et al. 2012) and Langat River in Selangor also provides drinking water to almost half of the population (Ahmed et al. 2016a, b). Therefore, the treatment of the raw water by the water treatment plants (WTPs) in the Langat River Basin is very important to supply safe water at the household level to achieve the SDG target of 6.1. However, all the WTPs in the basin are based on the conventional method which uses $Al_2(SO_4)_3$ (aluminium sulphate) for water disinfection. The doses of $Al_2(SO_4)_3$ depend on the water turbidity, colour, TDS, etc. Hence, Langat River being situated in the tropical climate experiences uncertain flash floods due to heavy rainfall. Thus, the pollution of the river is obvious and the treatment method of the WTPs needs to be upgraded as Farizwana et al. (2010) and Qaiyum et al. (2011) reported very higher Al concentration 9.90E−1 ± 1.52E0 mg/L and 2.10E−1 ± 4.15E−2 mg/L, respectively, at the same geographical location in the water supply of Johor, Malaysia. The conventional water treatment at the plants of Langat River Basin is unable to remove fully the Al concentration. The plant authorities are adding aluminium sulphate in treating raw water for disinfection because of frequent changes of turbidity in the raw water in the tropical climatic condition. Therefore, reverse osmosis (RO) membrane technology—Best Available Technology (BAT), and Small System Compliance Technology (SSCT) declared by USEPA—can be useful because of its capacity to remove inorganic ions > 90% from treated water (USEPA 2005). Theoretically, RO can remove particle size of about 0.0001 µm and most of the metal ions ranging from 0.0001–0.001 µm in the water including Al 0.000143 µm (Aquathin 2017). Several studies have already found an association between Alzheimer's disease and Al ingestion through drinking water for a long time, so the upgradation of water treatment method from conventional to reverse osmosis would be effective to supply safe drinking water at the household level in the basin.

Conclusion and Recommendations

Aluminium (Al) concentration (2.50E−01 ± 1.89E−01 mg/L) in the Langat River is naturally higher due to the weathering of aluminium and ferralsols clay minerals at the basin, and it has crossed the Malaysian standard of drinking water quality (0.2 mg/L). However, the treated water by the treatment

Table 7 Association between drinking water and potential diseases at the Langat River Basin, Malaysia

Use of household filtration system	Types of diseases						Total
	Leukaemia	Liver cancer	Alzheimer's	Diabetes	Kidney failure	Other	
Yes							
Count	3	1	3	47	2	27	83
% within use of filter	3.6	1.2	3.6	56.6	2.4	32.5	100.0
% within types of diseases	75.0	50.0	75.0	65.3	50.0	58.7	62.9
% of total	2.3	0.8	2.3	35.6	1.5	20.5	62.9
No							
Count	1	1	1	25	2	19	49
% within use of filter	2.0	2.0	2.0	51.0	4.1	38.8	100.0
% within types of diseases	25.0	50.0	25.0	34.7	50.0	41.3	37.1
% of total	0.8	0.8	0.8	18.9	1.5	14.4	37.1
Total							
Count	4	2	4	72	4	46	132
% within use of filter	3.0	1.5	3.0	54.5	3.0	34.8	100.0
% within types of diseases	100.0	100.0	100.0	100.0	100.0	100.0	100.0
% of total	3.0	1.5	3.0	54.5	3.0	34.8	100.0

plants, and households' tap and filtration water were within the Malaysian drinking water quality standard except for a few samples. Therefore, Al ingestion through the drinking water in the basin possesses no potential human health risk since the calculated hazard quotient (HQ) value is less than 1 at 95% confidence level. Meanwhile, globally several studies have reported positive association between a higher level of Al ingestion and Alzheimer's disease in humans. Although 1% population have reported with Alzheimer's diseases at the Langat River Basin in association with Al intake via drinking water, however, further epidemiological study is needed to investigate the causal relationship between Alzheimer's disease and Al ingestion via drinking water to achieve the SDG 3 and 6. Since all the treatment plants in Langat Basin follow the conventional method that is unable to remove Al concentration fully from the treated water, thus reverse osmosis technology can be installed in the plants to get safe drinking water because of its capacity to remove metals > 90% from treated water. Besides, the roles and responsibilities of various actors in managing drinking water at the Langat Basin can be better coordinated via a more effective and proactive leadership role by the local authorities, because they got the mandate of drinking water management through the Local Government Act, 1976 in Malaysia.

Acknowledgements The study is supported by the Grants: KRA-2017-022; TRGS/1/2015/UKM/02/5/2 and GUP-2015-029 of the Universiti Kebangsaan Malaysia. The authors are also grateful to the Laboratory of Chemical Oceanography, Faculty of Science & Technology, Universiti Kebangsaan Malaysia as well as to the water treatment plant authorities i.e. Puncak Niaga (M) Sdn. Bhd. and Konsortium Air Selangor Sdn. Bhd. to allow to collect water samples from the outlets of their plants. The authors are also thankful to the Institute of Climate Change, UKM to prepare the study area map.

Appendix

See Table 8.

Table 8 Household questionnaire for demographic information

Age	○ 18 to 25	○ 26 to 35	○ 36 to 45	○ 46 to 55	○ > 55
Gender	○ Male		○ Female		
Religion	○ Islam	○ Christian	○ Buddha	○ Hindu	○ Other
Education	○ SPM or below	○ Diploma	○ Degree	○ Graduation	○ Other
Family Income (Monthly)	Below RM2000	RM2001 to RM4000	RM4001 toRM6000	RM6001 to RM10000	Above RM10000
Occupation					
Place of Residence	○ Urban	○ Semi-urban	○ Rural	Residence Duration: Year/Month…………	
Weight of the respondent (KG or Pound)		○	Daily Intake of drinking water (No. of glass or Liter)		○
○ Do you use water directly from water supply pipe or water filters at households for cooking? (1) Water pipe, (2) Filter housing, (3) Other…………………………….					
On an average what is your household water bill or the bill of last month: RM……………					
Do you use any drinking water filtration system at your household: (1) Yes (0) No					
If you have filter at house, what is the type from below:					
○ Carbon Filter	○ Reverse Osmosis	○ Distillation	○ UV Light	○ Alkaline	○ Other:
If you have filter at house, what is the reason from below:					
○ Available	○ Cheaper	○ Effective	○ Convenient	○ Other:	
Have you or any member of your family Suffered from any of the following diseases:					
○ Leukemia	○ Liver cancer	○ Alzheimer's Disease	○ Diabetes Mellitus	○ Kidney Disease	○ Other:
Email (Optional)			Cell Phone (Optional)		

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