IJMS 2015 vol. 2 (1): 43-50

International Journal of Multidisciplinary Studies (IJMS)

Volume 2, Issue I, 2015

Exposure Analysis of Drinking and Dietary Contaminants in a Selected Population, Padaviya, Anuradhapura

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ABSTRACT

This study focused to analyse exposure of selected drinking and dietary contaminants and to assess the health risk for the selected population of Padaviya, Anuradhapura. Thirty families were randomly selected from which fifteen families were with the presence CKDu patients and other fifteen families were with absence of CKDu patients. Questionnaire based social survey was conducted and relevant data were collected for the risk analysis. Water, rice and soil samples were collected on family basis for the quality assessment. Nitrate-N, total hardness and fluoride varied within the range of 1.01 - 23.4 mg/L, 40.04 – 644.58 mg/L and 0.47 – 1.92mg/L respectively. All physiochemical parameters were significantly different among the wells ($P<0.05$). Water pH, conductivity and TDS in well water were below the Sri Lankan standard for portable water level (SLPWL). However, exceeded SLPWL value of NO_3-N (C10, C14, C15& N3), hardness (C12 & C13) and fluoride (C7, C15 & N3) were observed in some wells. Both iron and copper concentrations in well water were lower than the provisional maximum tolerable daily intake (PMTDI) of WHO (Fe: 2 mg/L and Cu: 2 mg/L). Dietary iron and copper concentrations in rice were higher than the PMTDI of WHO (0.5 mg/kg) except for family N7. Copper and Iron varied within the range of 1.55 – 48.4 mg/kg dw and 467.08-893.61 mg/kg dw in soil respectively. Probable exposure concentration was higher than probable non-exposure concentration in the selected population. Therefore, Relative Risk for CKDu was greater than 1 for all selected contaminants and it explains that there is a possible risk due to drinking water and eating rice for the selected contaminants. Non-cancer risk values in selected families were higher than the unity of the risk level ($1x10^{-6}$) and therefore the contaminants in drinking water and rice in Padaviya area can be considered as risk factors for prevailing chronic kidney disease.

KEYWORDS:*Exposure, contaminants, drinking, dietary, risk, recommended level*

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1. INTRODUCTION

The contamination from both natural and anthropogenic sources becomes a major issue which is responsible for serious health problems in world wide. The European Food Safety Authority recommends that women should drink about 1.6 litres of water and men should drink about 2.0 litres of water per day and one person consumes about three or two meals per day. Therefore, water and food consumption is the major exposure route for many human contaminants and degradation of the resource caused by various anthropogenic activities increases the availability of contaminants. Sri Lanka is ranked in the eighth position in the world on the list of countries that use the highest quantity of chemical fertilizer (Mudalige, 2014). The highest usage of fertilizers and agro-chemicals has been reported in North Central Province (NCP), is applied for about 128,000 ha (Weeraratna, 2013). Recent findings show that, some toxic heavy metals including fertilizers and agro chemicals are the major contaminant in Sri Lanka (Jayasumana, 2014; Bandara et al., 2008).

More than 80% of the rural drinking water supply needs are met from groundwater by means of dug wells and tube wells (Panabokke & Perera, 2005). High Fluoride levels (above 1.5mg/L) in well water in dry zone had been observed as far back as 1976. Subsequent studies have shown that 40% of wells in NCP were rich in Fluoride and a number of 456 deep tube wells in Anuradhapura district have also been found with Fluoride ranging from 0.78 to 2.68 mg/L (Lasanntha et al., 2008). The same study reported that 34% of the wells exceeded the maximum desirable level of 100 mg/L of calcium in drinking water and also 8% of the wells exceeded the maximum permissible level of 240mg/L in Anuradhapura District. Electrical conductivity in Anuradhapura district reported as 350μS/cm indicating the abundance of electrolytes in water (Dissanayake, 2005).

Nowadays, chemical contaminants are a major concern for food safety because of the increased role of man-made chemicals due to our modern lifestyles. Rice is the main diet of Sri Lankans in all over the country and rice and other several popular food items have been contaminated from heavy metals such as cadmium (Bandara et al., 2008). The heavy metal contamination like cadmium was observed in rice, *Nelumbonucifera* (Lotus) rhizomes, cow's milk and in Tilapia (*Oreochromisniloticus*) (Bandara et al., 2008). The overuse and misuse of agrochemicals would be contributed to heavy metal contamination (Cd, Cr, Ni, Pb, As etc.) and for both acute and chronic renal failure (Bandara et al., 2010).

Therefore, this study was aimed to identify the major contaminants in drinking water and in selected food types in Padaviya area and to follow the risk assessment model in order to assess the exposure levels of above identified contaminants.

2. MATERIALS AND METHODS

Study Area

The study area was decided according to the data taken from Office of the Provincial Director of Health Services, North Central Province, Anuradhapura regarding prevalence of CKDu (Fig 1). Padaviya is 93 km away from Anuradhapura and it receives 1450 mm mean annual rainfall and available from October to March. A questionnaire based social survey and laboratory analyses were carried out to collect data for human health risk assessment. Thirty families were selected representing fifteen families with the presence of CKDu patients and other fifteen families with the absence of CKDu patients.

Collection of Samples

Paranagama (2013) has reported that sources of drinking water of CKDu patients are dug wells (92 %) and tube wells (8 %). Therefore, water samples were collected from the water sources such as dug wells, tube wells, etc. belongs to the selected families. On site measurement of pH, conductivity, Total Dissolved Solid (TDS), salinity, Dissolved Oxygen (DO), temperature were taken by Multi - Parameter Meter (HACH sens ION 156). The filtered water samples were stored with ice in a heat insulated box for facilitating transportation. Rice samples were collected from the selected families while soil samples were collected from their own paddy fields.

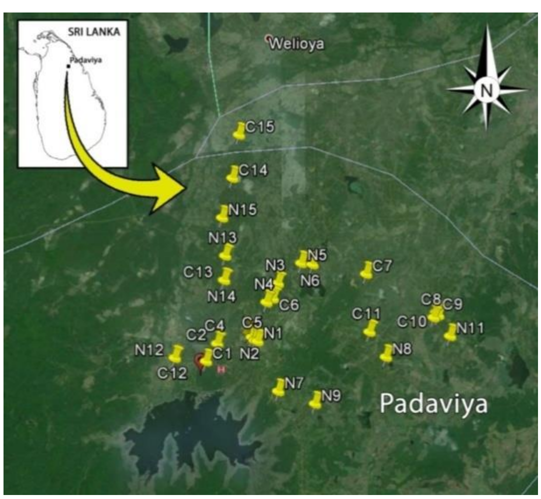


Figure 1. GPS locations of sampling in Padaviya area; Source :(Earth.google.com, 2014)

Analysis of chemical parameters

The filtered water samples were analyzed for Nitrate-N according to the Sodium Salicylate method, water soluble salts of phosphoric acid were measured by automated ascorbic acid reduction method (Rand et al., 1975, p.481-482) and organic phosphorous content of each water sample were measured by persulphate digestion method and the automated ascorbic acid reduction method (Rand et al., 1975, p. 481-482). Fluoride concentration of each filtered water sample was measured directly by HATCH-DR 4000U Spectrophotometer at a wave length of 580 nm using SPADNS solution (Rand et al., 1975, p.393-394). Also, the water samples were analysed for total hardness by EDTA titration method.

Analysis of heavy metals

Filtered water sample were acidified with10% Conc. Nitric acid (Analar grade). Collected rice samples were thoroughly washed several times with deionized water and the sample was dried at 105°C in an oven until obtained a constant weight. Dried samples were ground to fine powder using mortar and pestle and fine powder was sieved using 250 µm sieve. Then, 5 mL of concentrated HNO_3 (65%) and 2.5 mL of H_2O_2 (30%) were added to each 0.5g of sieved rice samples and the solution was heated on Kjeldhal Heating Digester under fume hood at 80°C, for 2 - 3h, till the clear transparent solutions were obtained. The final solutions were filtered through Whatman No. 41 filter paper (Jalbani et al., 2014). All soil samples were air dried on polypropylene sheets at room temperature for several days until they were deprived of moisture. Then the soil samples were well ground using porcelain mortar and pestle and were sieved through a 250 µm mesh sized sieve. The pre-digestion step was done at room temperature for 24 h with 10 mL of a (3:1) mixture of 12M HCl and 17M HNO_3. The suspension was digested on GK 06 Kjeldhal Heating digester under fume hood at 130°C for 15 min. The obtained suspension was cooled at room temperature and filtered through Whatman No. 41 filter paper (Peña-Icart et al., 2011).

A series of standard metal solutions were prepared for Cu using the stock solutions of 1,000 mg/L (BDH chemicals). Concentrations of Cu were measured using Atomic Absorbance Spectrophotometer (SpectrAA 220 AAS). Iron concentration of rice, soil and water samples were measured according to the Thiocyanate colorimetric method (University of Canterbury, 2011).

Human Health Risk Assessment

The data, which had taken from questionnaire based social survey, were prepared to be used for the risk assessment.

Exposure Analysis of water and dietary intake

$$I = \frac{C \times IGR \times EF \times ED \times CF}{BW \times AT \times 365}$$

Where,

I = Intake of chemical (mg/kg/day)

C = Average chemical concentration in food (mg/kg)

IGR = Ingestion rate (mg/day)

EF = Exposure frequency (days/year)

ED = Exposure duration (years)

CF = Conversion factor (If units in above parameters don't match)

BW = Average Body weight (kg)

AT = Average exposure time (years)

$$CDI = \frac{C \times CR \times EF \times ED \times CF}{BW \times AT}$$

Where,

CDI = Chronic daily intake of water (mg/kg/day)

C = Chemical concentration in water (mg/L)

CR = Consumption rate (L/day)

EF = Exposure frequency (days/year)

ED = Exposure duration (years)

CF = Conversion factor (If units in above parameters don't match)

BW = Average Body weight (kg)

AT = Average exposure time (days)

Source : (enHEALTH, 2012)

Determination of Non_Cancer Risk

$$Risk = RFD \times Exposure$$

Where,

RFD = Reference Dose

Determination of Relative Risk (RR)

$$RR = \frac{PEC}{PNEC}$$

Where,

PEC = Probable Exposure Concentration

$PNEC$ = Probable Non Exposure Concentration

Statistical analysis

Oneway ANOVA was carried out to determine the difference of the water quality parameters among the selected wells. The multiple comparison of the water quality parameters was performed using Tukey HSD test (SPSS, Version 16.0).

3. RESULTS AND DISCUSSION

The physical parameters of pH, conductivity and Total dissolved solid (TDS), did not exceed Sri Lankan standard for portable water level/SLPWL (SLS 614, 1983) and there was a significant difference among the above mentioned parameters in 30 wells ($P < 0.05$). Water pH and electrical conductivity

(EC) varied within the range of 6.86 – 7.95 and 16.25 - 755µs/cm respectively (Table 1).

Table 1. Physical parameters of well water

	Value	Significantly different wells
pH		
Mean± SD	7.37± 0.31	C1, C6, C7 & N10
Range	6.86 – 7.95	
Electrical Conductivity (µs/cm)		
Mean± SD	477.23 ± 211.78	C1, C8, C9, C10, C13, N2 & N10
Range	16.25 – 755.0	
TDS (mg/L)		
Mean± SD	236.85 ± 106.4	C1, C4, C8,C9, N2, N5, N6,N8, N9 & N10
Range	6.3 – 378	
Temperature (°C)		
Mean± SD	29.5 ± 2.16	-
Range	27.4 - 36.1	

C : CKD patients present in the family (Case)
N : No one present with CKD in the family (Non_Case)

Jayawardana et al. (2010) has reported that changing of pH values from weak acidic to weak basic (4.0 to 8.2 with the average of 7.2) with very low EC values (2.40 mS/m) indicated that dissolved ionic species are very low in the water. They have also explained that, in dry zone, fluoride values increase in the condition with slightly alkaline pH (7.5-8.2) and relatively low EC (1.0-2.5 mS/m) (Jayawardana et al., 2010). Mechenich & Andrews (2004) have reported, that the much greater hardness may indicate the presence of contaminants which may occur naturally or be influenced by human activity. In the current study, TDS values of most of the wells were much greater than two times of the hardness (Ex. Well no. C3).

Many researchers said that water hardness, high level of fluoride and contamination of heavy metals can effect on human health (Bandara et al., 2010; Bandara et al., 2008; Dissanayake, 2005). In the present study total hardness and fluoride varied within the range as 40.04 – 644.58 mg/L and 0.47 – 1.92 mg/L respectively. The concentration of nitrate-N varied from 1.01 to 23.4 mg/L among the 30 wells. Paranagama (2013) has observed nitrate-N varied from 0.3×10^{-6} to 5.82×10^{-6} mg/L in Padaviya. All chemical parameters were significantly different among the wells ($P<0.05$). Some parameters as NO_3-N (C10, C14, C15& N3), hardness (C12 & C13) and fluoride (C7, C15 & N3) concentrations exceeded the SLPWL (SLS 614, 1983) (Table 2). Both iron and copper concentrations in well water were lower than the provisional maximum tolerable daily intake (PMTDI) of WHO (Fe: 2 mg/L and Cu: 2 mg/L). Dietary iron concentrations in rice were higher than the PMTDI of WHO (0.8 mg/kg) and copper concentrations in rice were higher than the PMTDI of WHO (0.5 mg/kg) except in the sample taken from family N7. Copper and Iron varied within range of 1.55 – 48.4 mg/kgdw and 467.08 - 893.61 mg/kgdw respectively in the soil.

It was noted that the wells located closer to the paddy fields have exceeded the nitrate concentration than SLPWL (SLS 614, 1983). Gunatilake and Iwao (2010) also have reported that, during the period of fertilizer application to paddy fields the drinking water wells located nearby paddy fields are more vulnerable to nitrate contamination.

Organic phosphorous in well water varied from 0.1 to 0.39 mg/L. Paranagama (2013) has reported that phosphate varied range of 61.1×10^{-6} - 80.25×10^{-6} mg/L in well water. Generally, organic matter decomposition is lower in well water than in surface water and microbial decomposition is the main way of entering orthophosphate into the well water Young et al (2010). Therefore, low concentration of orthophosphate was observed in the well water is said that phosphate concentrations in wells found in different soil formations show distinctive variations.

The fluoride concentration in groundwater of dry zone, Sri Lanka lies in a range of 0.1 to 4.7 mg/L (Johnson et al., 2012). Fluoride may leach into groundwater due to weathering of fluoride-rich minerals in the basement rocks and climate and geological formation of dry zone are the most preferred factors for elevated fluoride level in water (Jayawardana et al., 2010).Furthermore, above study mentioned that the dry zone aquifers are well known for higher fluoride concentrations and fluoride-related endemic diseases and chronic renal failures. As a major component in acidic soils, iron hydroxides serve as an important sink for fluoride in soil resulting into the enhancement of fluoride concentration in water under acidic conditions (Chandrajith et al., 2012). In dry zone, soil consists with reddish-brown earth which predominates by iron rich biotite gneisses of the Khondalite series (Kalpage et al., 1963). In the current study, iron content in soil varied from 467.08-893.61 mg/kg dw. The soil Iron content of each sampling site was poorly correlated with fluoride content of relevant water samples. Jayawardana et al. (2010) has shown in oxidized environment in the field of Fe_2O_3 and it should be the result for poor relationship of iron with fluoride.

Total hardness ranged between 40.04 – 644.58 mg/L and the water hardness in most of selected wells was lower than the recommended level of 600 mg/L in Sri Lanka except two wells (C12 & C13). The average groundwater hardness was observed in Padaviya by Fonseka et al. (2012) as 466 ± 34 mg/L. However, it is important to consider total hardness as an important public health issue (Emmanuel et al., 2013). Similarly, Jayasumana et al. (2014) has reported a correlation between the places with high ground water hardness and the geographical distribution of the CKDu in Sri Lanka andthese areas contain Ca, Mg, Fe and Sr ions in water also. Iron content of drinking water varied from 0.0011 to 0.0084 mg/L in the selected wells and those values did not exceed the recommended level of 0.3 mg/L. Paranagama (2013) revealed somewhat lower values than in the present study ranged as 0.00014 - 0.00046 mg/L in Padaviya area. Jayawardana et al.(2010) has reported low values of iron in water samples because iron is generally stable in acidic pH.

Table 2. Chemical parameters of well water

	Value	**Significantly different wells**
Nitrate-N (mg/L)		
Mean± SD	3.51± 5.32	C2, C10, C13, C14, C15 & N3
Range	1.01 - 23.4	
Total Hardness (mg/L)		
Mean± SD	161.48 ± 152.05	Most wells except C2, C4 , C5, C6, C7, C11, N1, N2, N4, N5 & N7
Range	40.04 – 644.58	
Fluoride (mg/L)		
Mean± SD	0.73 ± 0.37	C7, C15, N3, N8, N9 & N15
Range	0.47 – 1.92	
Organic phosphorous		
Mean± SD	0.08 ± 0.11	C1, C3, N5 & N10
Range	0.1 – 0.39	

C : CKD patients present in the family (Case)
N : No one present with CKD in the family (Non_Case)

Copper and iron in rice collected from selected families were observed as Fe; 2.29 - 35.21 mg/kg and Cu; 0.3 - 33 mg/kg). In rice samples, Cu and iron concentrations were higher than the provisional maximum tolerable daily intake (PMTDI) of WHO (Fe; 0.5 mg/kg and Cu; 0.8 mg/kg) except only from family (N7).

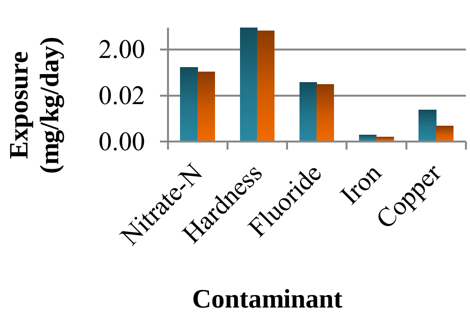
Copper and iron content of soil varied from 1.55 to 48.4 mg/kg (dw) and from 467.08 to

893.61 mg/kg (dw) respectively. The iron rich soil in dry zone has an ability to retain arsenic ions and can be accumulated those (Jayasumana et al., 2014). Recently, arsenic plays main role of chronic renal failure in dry zone of Sri Lanka (Paranagama, 2013). It implies that arsenic is not present naturally in soils nevertheless has been introduced from the surface, most probably due to anthropogenic activities. Today, most of the researchers have been investigated that agrochemicals and fertilizers are the major source of arsenic (Berg et al., 2001).

Jayasumana et al. (2014) has reported that a chemical which contain in "round up" is a major causative agent for chronic kidney disease and the chemical is Glycophosate. Hard water in dry zone with high concentration of calcium, magnisium and strontium are combined with glycophosate and easily forms complexes. Ferric ions also play a significant role in the process of adsorption of glyphosate in soil. As a result of current study, high value of total hardness and iron content in soil are correlated with the case group of CKDu.

Collected information from the social survey was important for exposure analysis of each drinking and dietary contaminants. Total exposure in case group (PEC) of all contaminants was higher than total contaminants in non_case group (PNEC) (Figure 2). It implies intake amount of relevant contaminant per day in the case group was high.

(a)



(b)

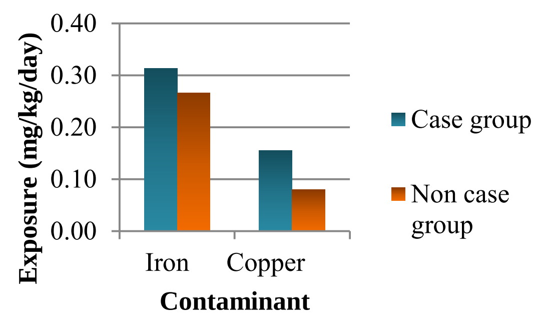


Figure 2. Total Exposure of selected contaminants in (a) water (b) rice

The relative risk of each drinking and dietary contaminants were greater than 1 (one) according to PEC and PNEC of selected families (Table 3). It explains that, there is a relative risk from each contaminant in the selected population.

Table 3. Relative Risk (RR) of selected contaminants

Contaminants	**RR of drinking contaminants**	**RR of dietary contaminants**
NO_3^--N	1.58	-
Hardness (Ca^{+2} & Mg^{+2})	1.40	-
Fluoride	1.20	-
Iron	1.26	1.18
Copper	5.06	1.95

(b)Non_cancer risk of each contaminant in selected families was higher than the unity of the criteria ($1x10^{-6}$) (Table 4). It reveals, in a population of one million people, additional person or persons would be expected to develop risk from considered contaminants in the study. When comparing the risk values separately for groups considering highest and lowest concentrations male group was the most vulnerable group for drinking (Table 4) and dietary contamination than other two groups (Table 5).

Table 4. Non-cancer risk for selected drinking contaminants

Contaminants	Code	Drinking Risk		
		Male	Female	Children
NO_3^--N				
HC	C14	1.1724	0.8245	0.0000
LC	C7	0.0844	0.0824	0.0000
Hardness				
HC	C12	76.7066	85.6754	53.4081
LC	C3	3.1797	3.4396	7.2565
Fluoride				
HC	C7	0.0055	0.0053	0.0000
LC	N8	0.0007	0.0009	0.0000
Iron				
HC	C7	3.5×10^{-6}	3.4×10^{-6}	0.0000
LC	C2	1.1×10^{-6}	1.1×10^{-6}	0.0000
Copper				
HC	C7	1.9×10^{-4}	1.8×10^{-4}	0.0000
LC	N12	0.0000	0.0000	3.6×10^{-5}

HC: Highest concentration among 30 families
LC: Lowest concentration among 30 families

Table 5. Non-cancer risk for selected dietary contaminants

Contaminants	Code	Dietary Risk		
		Male	Female	Childre n
Iron				
HC	C6	1.6×10^{-3}	1.5×10^{-3}	1.2×10^{-3}
LC	N6	5.2×10^{-4}	4.1×10^{-4}	9.3×10^{-4}
Copper				
HC	C9	3.6×10^{-4}	5.1×10^{-4}	8.8×10^{-4}
LC	N9	6.7×10^{-5}	6.5×10^{-5}	0.0000

HC: Highest concentration among 30 families
LC: Lowest concentration among 30 families

Summary of social survey

Padaviya is a rural village in Sri Lanka and the lives of people are slowly flowing by fulfilling their requirements from the village. Main occupations are paddy cultivation, chena cultivation and cultivation of seasonal crops. About 90% of people use ground water from dug wells, tube wells, springs, etc. Recently, some filtering water facilities have been settled everywhere by the government and private sector. However, the villagers have to pay for their own drinking water. The majority of people are suffering from CKDu and it is becoming an uncertainty health problem. Most of the farmers do not imitate safety methods in applying agrochemicals and also some habits such as; chewing beetle and taking alcohol were observed.

CONCLUSION

Since all physiochemical parameters were significantly different among the wells it should be paid individual attention for the wells on their quality of the water. However, pH, conductivity and TDS in well water were below the Sri Lankan standard for portable water level (SLPWL). The exceeded values than the SLPWL was observed for NO_3-N, hardness and fluoride in some wells. Both iron and copper concentrations in well water were lower than the provisional maximum tolerable daily intake (PMTDI) of WHO.

Total exposure in case group (PEC) of all contaminants was higher than total