

**PRELIMINARY ASSESSMENT OF EXPOSURE LEVELS OF Hg AND Mn IN
SELECTED FOOD TYPES IN HAMBANTOTA DISTRICT, SRI LANKA**

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ABSTRACT

This study was conducted to assess the exposure of dietary contaminants (Hg and Mn) in selected areas of Tangalle, Beliatta, Angunukolapelassa and Balangoda (reference site). Different food types were collected from selected houses where kidney failures live and some samples were collected from the local fair during the dry period of September - October, 2013. The sample pre-preparation was mainly based on acid digestion and the concentrations of Hg and Mn were measured using atomic absorption spectroscopy with the combination of cold vapor generator for Hg analysis. Questionnaire based field survey was conducted to identify the status of the hazard and to collect relevant data from the families to apply risk assessment models. Mean concentrations of Hg in Lotus rhizomes collected from Tangalle, Angunukollapellassa, Beliatta and Balangoda areas were 1.4 ± 1.6 , 7.2 ± 0.07 , 11.8 ± 3.65 and 3.0 ± 0.88 $\mu\text{g/g}$ wet wt respectively. The mean exposures of Hg with the consumption of Lotus rhizomes for the considered populations were 0.28 ± 0.32 , 1.44 ± 0.001 , 2.36 ± 0.68 and 0.60 ± 0.16 $\mu\text{g/kg}$ body weight/day respectively. The mean exposure of Hg with the consumption of fish (*Oreochromis niloticus*, *Puntius* sp., *Labeo rohita*, *Glossogobius* sp.) were 17.5 ± 5.83 , 103.75 ± 104.54 , 30.00 ± 4.88 and 18.13 ± 76.36 $\mu\text{g/kgbw/day}$ respectively for the studied areas. Exposure of Hg via the consumption of fish exceeded the recommended reference dose of 1.60 $\mu\text{g/kg}$ body weight/day. The mean exposure of Mn via the consumption of Lotus rhizomes were 25.96 ± 35.4 , 72.60 ± 41.48 and 16.24 ± 33.89 $\mu\text{g/kgbw/day}$ respectively. Mean exposure of Mn via the consumption of *Lasia* sp in above four areas, were 11.67 ± 5.85 , 4.36 ± 4.18 , 4.34 ± 8.54 and 1.89 ± 2.15 mg/kgbw/day respectively. Accordingly, the hazard quotients for the exposure to Hg via Lotus rhizomes exceeded the unity of the model (1×10^{-6}) in all four areas and it exceeded for the consumption of rice only at Beliatta. The hazard quotients for Mn also exceeded the unity of the model at above four areas due to consumption of *Lasia* and Lotus rhizomes except at the reference site of Balangoda area. Also, high risk ratios of carcinogenic effect were observed for farmers in Angunukolapelassa as 4.00 and 5.45 for male and female respectively.

Keywords: Exposure, risk assessment, hazard quotient, Mercury, Manganese.

INTRODUCTION

Traces of heavy metals are found in every food item and evaluation of element intakes and the risk of exceeding the tolerable intakes of contaminants is important for the certification of food security. Some elements such as Hg have no beneficial biological functions and the exposures may be harmful to health. For example, organic mercury compounds are neurotoxins and are known to cause chronic renal damage; Pb can be harmful to neuropsychological development while some elements such as Mn, Cu and Zn are essential to health and may be toxic at high levels of exposure (Ysart et al., 2000). With respect to dietary exposure of Hg and its compounds has been noticed specially due to methyl mercury

residuals in fish in Germany (Wilhelm et al., 2003). Therefore, estimating dietary exposure to a substance is a considerable task and it requires information on the level of substance in a specific food and information about the types and quantities of foods consumed (Biego et al., 1998). The risks to health from some elements in food can be evaluated by comparing the values of dietary exposures with the Provisional Tolerate Weekly Intakes (PTWIs) and Provisional Maximum Tolerable Daily Intakes (PMTDIs) recommended by the Joint Expert Committee on Food Additives (JECFA) of the Food and Agriculture Organization of the United Nations (Ysart et al., 2000).

According to Bandara et al. (2010) the prevalence of end-stage renal failure is reaching epidemic levels in the North Central Province (NCP) of Sri Lanka and based on the local news it is now spreading in the southern province. Several reasons such as type II diabetes, hypertension overuse and misuse of agrochemicals have been suggested as the causes for both acute and chronic renal failure (Bandara et al., 2010). Even though, there are some evidences to support an environmental aetiology to CRF and geographical variation of occurrence of the incidence in Sri Lanka (Wanigasuriya et al., 2007) the aetiology of chronic renal failure in the North Central Province still remains unknown.

Agriculture is the main form of economic activity and over 55% of the total population depends on agriculture in its various forms in Hambantota district (Hdccc.lk, 2014). Mainly Hambantota district produce 5.8% of inland and aquaculture fish production from the total production (The Ceylon Chamber of Commerce, 2014). Therefore, the objective of the study is to determine the concentration of essential (Mn) and contaminant trace metals (Hg) in selected food types collected from the local market and from the houses in the studied areas. Also, it is focused to estimate the exposure levels of the metals in food types and hazard quotient values to compare with the recommended values for male, female and children of the selected populations.

MATERIALS AND METHODS

Dietary samples of rice, Lotus rhizomes, *Lasia* rhizomes and fish muscle (*Oreochromis* sp, *Labeo* sp, *Glossogobius* sp, *Cyprinus carpio* and *Puntius* sp, were the common fish species in the studied areas) were collected from houses, and local fairs at Tangalle, Beliatta, Angunukolapalessa and Balangoda area (out of the Hambantota district). A questionnaire based family survey was conducted for 30 families at each area to collect details (age, sex, occupation, body weight, food sources, consumption rates of the selected food types, health conditions and life style..etc) for risk assessment and to select the householders who are suffering from kidney disease and cancer.

Sample preparation and Analysis

Pre washed samples of Lotus rhizomes, *Lasia* rhizomes, rice and fish muscle were dried at 105°C until become the weights constant. The dried samples were grinded by using a quartz mortar and pestle and sieved via 200µm mesh and three replicates of 0.5g of sieved samples were digested with 5ml of C, HNO₃ and 1ml of H₂O₂ in closed pyrex beakers at 55°C during 30 minutes. The digested samples were filtered via 45 µm GF/C filter papers and the filtrates were diluted up to 100ml with deionized water. The same procedure was followed to prepare the blank solution for the analysis. Hg and Mn concentrations were determined using AAS (Varian 220) and the accuracy of the method for each metal was determined according to the analytical methods in APHA (1999).

The working series of Mn as 1 mg/L, 3 mg/L, 5 mg/L and 7 mg/L was prepared using analytical grade MnSO_4 and C_2H_2 was used as the oxidant for the flame of AAS. The working series of Hg as 0.0005 mg/L, 0.001 mg/L, 0.002 mg/L and 0.004 mg/L was prepared using 1000 ppm standard solution of Hg (sigma chemicals). The cold vapor generator was used to evaporate the samples and 10% SnCl_2 was used as the reductant for the analysis of Hg. The limit of detection for the two metals was determined according to the standard deviation of the background variation for a signal at three times (Thermo Elemental, 2001, p 18). The sensitivity of the instrument was adjusted according to the manual of AAS and the percentage error of the obtained values was determined using known concentrations of each metal.

Determination of Exposure and Hazard Quotient

Exposure

Dietary Exposure of contaminants was calculated separately using the following equation and those values were used for Hazard Quotient calculations (Joseph and Thomas, 1987).

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

CDI: Chronic Daily Intake by food consumption (mg/kg-day)

CF: chemical concentration in food (mg/g)

CR: consumption rate (g/day)

EF: exposure frequency (days/year)

ED: exposure duration (years)

BW: body weight (kg)

AT: averaging time (period over which the exposure is averaged—days)

Hazard Quotient

The Reference Dose of Mn is 140 $\mu\text{g}/\text{kg}/\text{day}$ (Epa.gov, 2014) and the reference dose for Hg is 0.23 $\mu\text{g}/\text{kg}/\text{day}$ (Washam, 2010). The HQ was calculated using the equation given below for each food type and if the HQ exceeds 1×10^{-6} (unity) there will be a potential non-carcinogenic effect (EPA criteria, 2010).

$$\text{Hazard Quotient (HQ)} = \text{Reference Dose} \times \text{Exposure}$$

Statistical analysis

Descriptive data analysis (mean, standard deviation, standard error, maximum and minimum values) was carried out using Windows excel 2007 version. One way ANOVA was carried out to compare the mean heavy metal concentrations among the sampling areas further with the Post hoc comparison of means using Turkey's multiple range tests at the significance level of 0.05. All the statistical analysis was carried out using SPSS software programme (SPSS v 16.0).

RESULTS & DISCUSSION

The Table 1. gives the mean values of Mn and Hg concentrations in different food types collected from the selected areas. Hg was not detected in *Lasia* collected from Tangalle, Angunuklappassa and Beliatta areas and in rice collected from Angunukolappassa and Balangoda areas. The highest concentration of Hg (33 $\mu\text{g}/\text{g}$ wet weight) was detected in fish

samples collected from Angunukolaplassa area and the value exceeded the value of 0.5 $\mu\text{g/g}$ wet (Choi, 2011). Also, comparatively lower values of Hg has been recorded in locally produced fish Tianjin, China ranging from 0.001 to 0.007 $\mu\text{g/g}$ wet wt and it has been recorded in vegetables ranging from 0.0003 to 0.007 $\mu\text{g/g}$ wet wt (Wang et al., 2005). Therefore, it is important to consider that the Hg concentration in food types collected from all four areas exceeded the above given values except Hg in *Lasia* rhizomes and the rice samples taken from Angunukollapellassa and Balangoda areas. The concentration of total Hg in rice has been reported as 0.1 $\mu\text{g/g}$ in China by Jackson and Punshon (2015) and the detected Hg concentration in rice at present study were nearly 10 times higher than the reported value. Also, it is interesting to say that the relatively lower concentrations of Hg were detected in Lotus and *Lasia* rhizomes and in rice than in fish from the study areas.

The Mn concentrations in *Lasia* from all four areas also exceeded the maximum permitted level of 140 $\mu\text{g/day}$ (U.S. EPA, 2012). Mn is one of the most plentiful metals in the earth's crust and it is important to control and regulation of glucose homeostasis (Biego et al., 1998). The quantity of Mn supplied by foods has been determined as 2.5 mg/day by Biego et al. (1998) and the estimated quantity of Mn for analysed food types was within the range of 150-338 mg/day in the present study. The estimated value of Mn was comparable to the recommended values of 2.5–5 mg/day for an adult (US National Academy of Sciences, 1980) and the recommended range of 2–9 mg/day (World Health Organization, 1994).

Table 1: The mean concentrations (\pm SD) of Mercury and Manganese at each area
ND: Not Detected

Heavy Metal	Food type	Sampling area			
		Tangalle	Angunukolapalssa	Beliatta	Balangoda
Mercury ($\mu\text{g/g}$ wet wt)	Lotus (n = 6)	3.0 \pm 2.8	7.2 \pm 5.2	11.8 \pm 12.6	1.4 \pm 1.2
	<i>Lasia</i> (n = 6)	ND	ND	0.0006 \pm 0.0007	ND
	Fish (n = 6)	5.6 \pm 3.8	33 \pm 33	9.6 \pm 10.5	5.8 \pm 6.0
	Rice (n = 6)	1.0 \pm 1.0	ND	1.48 \pm 1.3	ND
Manganese ($\mu\text{g/g}$ wet wt)	Lotus (n = 6)	130 \pm 132	363 \pm 275	81 \pm 84	ND
	<i>Lasia</i> (n = 6)	14592 \pm 10469	10895 \pm 9723	5428 \pm 5276	2369 \pm 2522
	Fish (n = 6)	ND	ND	854 \pm 922	ND

The daily intake of rice as the staple food was contaminated daily food item in those areas respectively for male, female and children according to the questionnaire based survey. The rice consumption of adult and children was reported as 491.5 g/person/day and 289.6 g/person/day respectively in the study of Wang et al. (2005) and in the same study revealed that the fish consumption rates as 57.5 g/person/day and 92.6 g/person/day respectively. The hazard quotient (HQ) values are given in the Table 2 for male, female and child in the selected population. The total HQ values were $>1 \times 10^{-6}$ only in Beliatta area due to Hg exposure and the significant contribution was observed due to the high consumption rate of rice. According to the HQ values in Table 2 children have already the adverse health effects. However, HQ value is a highly conservative and relative index and the value > 1 may not show the reality of the adverse health effects (Wang et al., 2005).

Table 2: Total hazard quotient for mercury exposure

Site	Food item	Male	Female	Child
Tangalle	Lotus rhizome	-	-	-
	<i>Lasia</i> rhizome	-	-	-
	Rice	0.88×10^{-6}	0.87×10^{-6}	2.35×10^{-6}
	Fish	0.0016×10^{-6}	0.0016×10^{-6}	0.0082×10^{-6}
	Total	0.8816×10^{-6}	0.8716×10^{-6}	0.3614×10^{-6}
Angunukolapelassa	Lotus rhizome	0.0008×10^{-6}	0.0007×10^{-6}	0.0868×10^{-6}
	<i>Lasia</i> rhizome	-	-	-
	Rice	-	-	-
	Fish	0.0067×10^{-6}	0.0074×10^{-6}	0.0446×10^{-6}
	Total	0.0075×10^{-6}	0.0081×10^{-6}	0.1314×10^{-6}
Beliatta	Lotus rhizome	0.0023×10^{-6}	0.0005×10^{-6}	0.0067×10^{-6}
	<i>Lasia</i> rhizome	0.0006×10^{-6}	0.0007×10^{-6}	0.0033×10^{-6}
	Rice	11.22×10^{-6}	15.05×10^{-6}	83.76×10^{-6}
	Fish	0.0027×10^{-6}	0.0034×10^{-6}	0.015×10^{-6}
	Total	11.225×10^{-6}	15.054×10^{-6}	83.785×10^{-6}
Balangoda	Lotus rhizome	0.0004×10^{-6}	0.0005×10^{-6}	0.0067×10^{-6}
	<i>Lasia</i> rhizome	-	-	-
	Rice	-	-	-
	Fish	0.0001×10^{-6}	0.0001×10^{-6}	0.0004×10^{-6}
	Total	0.0005×10^{-6}	0.0006×10^{-6}	0.0071×10^{-6}

Table 3: Total hazard quotient for manganese exposure

Site	Food item	Male	Female	Child
Tangalle	Lotus rhizome	19.06×10^{-6}	18.76×10^{-6}	1561.74×10^{-6}
	<i>Lasia</i> sp	1588×10^{-6}	1563×10^{-6}	24862×10^{-6}
	Rice	-	-	-
	Fish	-	-	-
	Total	1607.06×10^{-6}	1581.76×10^{-6}	26423.74×10^{-6}
Angunukolapelassa	Lotus rhizome	52.74×10^{-6}	57.9×10^{-6}	190.91×10^{-6}
	<i>Lasia</i> sp	1115.4×10^{-6}	1280×10^{-6}	24312×10^{-6}
	Rice	-	-	-
	Fish	-	-	-
	Total	1168.14×10^{-6}	1337.90×10^{-6}	24502.91×10^{-6}
Beliatta	Lotus rhizome	1123×10^{-6}	1427×10^{-6}	58.79×10^{-6}
	<i>Lasia</i> sp	555.73×10^{-6}	705.97×10^{-6}	10120×10^{-6}
	Rice	-	-	-
	Fish	4.60×10^{-6}	4.96×10^{-6}	100.48×10^{-6}
	Total	1683.33×10^{-6}	2137.93×10^{-6}	10279.27×10^{-6}
Balangoda	Lotus rhizome	233.86×10^{-6}	275×10^{-6}	5573×10^{-6}
	<i>Lasia</i> sp	-	-	-
	Rice	-	-	-
	Fish	-	-	-
	Total	233.86×10^{-6}	275×10^{-6}	5573×10^{-6}

CONCLUSION

A potential health risk was observed due to consumption of rice contaminated with Hg only at Beliatta area. Although, Hg concentration was relatively higher in fish muscle the HQ value was lower than the reference risk level because of low consumption of fish at the studied areas. Manganese was also observed as a risk factor with the consumption of Lotus rhizomes at all the sites exceeding the reference level of the risk value (1×10^{-6}). Manganese was detected only in fish samples collected from Beliatta area and for the better conclusion more number of samples should be analysed.

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