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Health Risk Assessment of Crops Cultivated out of Wastewater– A case study in Madurai

S. Sivasangari¹, T. Vel Rajan², L.Sasikumar³

Assistant Professor, Department of Civil Engineering, Sethu Institute of Technology, Tamil Nadu, India¹

Associate Professor, Department of Civil Engineering, Thiagarajar College of Engineering, Tamil Nadu, India²

Assistant Professor, Department of Civil Engineering, Kalasalingam Institute of Technology, Tamil Nadu, India³

ABSTRACT: The use of wastewater for irrigation is increasingly being considered as a technical solution to minimize soil degradation and to restore nutrient content of soil. Wastewater contains substantial amounts of beneficial nutrients and toxic heavy metals, which are creating problems for agricultural production. Heavy metals accumulation in soil and crops is of increasing concern because of the potential human health risks. The presenting study aimed to determine the human health risks associated with the food chain contamination of heavy metals routing from wastewater irrigation in the village Kallambal near chinthamani, located in Madurai district. Irrigated water, soil and vegetables were analysed for heavy metals such as Zn2+, Pb2+, Cr2+, Cd2+, Fe2+, Mn2+, Cu2+ and Daily Intake of Metals (DIM) and Health Risk Index (HRI) were also calculated. Pb2+,Cd2+, Fe2+ and Cu2+ in cultivated vegetables exceeded the permissible limits while the Transfer Factor (TF) was lower for all metals except Zn2+ and HRI was found to be maximum for *Solanum melongena* (Brinjal) and *sesbania grandiflora* (Agathikeerai) cultivated by wastewater.

KEYWORDS: Heavy metals, Wastewater irrigation, Health Risk Index (HRI), Daily intake of metals (DIM), Risk Assessment.

I. INTRODUCTION

The usage of used water for irrigation purposes has become one of the sustainable ways of reuse. It reduces the water scarcity for farmers and also it helps in crop growth with its high nutritional and organic constituents. Although it seems as a beautiful rose there are some thorns, as there are health issues for people consuming such cultivated crops, also associated with environmental risks, such as the soil fertility. Consumption of food crops contaminated with heavy metals is a major food chain route for human exposure. Heavy metals are ubiquitous in the environment, as a result of both natural and anthropogenic activities, and humans are exposed to them through various pathways. Contamination of foods by heavy metals has become an inevitable challenge these days. Health risk assessment of heavy metals on the human population is very important because such assessment would be useful to give information about any threat regarding heavy metals contamination of vegetables. This is because heavy metals are not easily biodegradable and consequently can be accumulated in human vital organs. This situation cause varying degree of illness based on acute and chronic exposures. The probability risk assessment technique has been adopted by a number of researchers to fully utilize available exposure and toxicity data. Generally heavy metals like Cadmium, Lead and Zinc are toxic at high concentrations and are urgency pollutants. Contamination of vegetables with heavy metals due to irrigation of contaminated water, addition of chemical fertilizers, industrial emissions and transportation, etc. have sever effect on human health. These food plants contain both essential and toxic metals over a wide range of concentration. It is well known that plants take up metals by absorbing them from contaminated soil as well as deposits on parts of the plants exposed to the air from polluted environments.

Madurai City Corporation is one among the ten metropolitan cities in Tamil Nadu. Having located at almost in the southern part of the state, it is the city for education and business. City covers an area of 148.99 square kilo meters. Climate prevailing is arid to semi-arid. Rainfall is occasional. Annual rainfall varies from 547.0 mm to 1264.0 mm against the normal rainfall of 891.50 mm. Groundwater table level fluctuation during 1991 to 2012 is found to vary



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between 4.0 meter and 11.10 meter below ground level. The 2011 census population is 15.64 lakh. The forecast of the wastewater generation from Madurai City Corporation indicates the quantity of 162.8 MLD at 2014 and is likely to be 338.7 MLD in the year 2044. The irrigation potential of wastewater reuse is assessed as 3000 ha with crops like groundnut, maize, millet etc... during 2014 and about 6000 ha during 2044. Scope for utilizing the existing minor irrigation tanks/ponds may be used for storage. Such tanks may also be useful in polishing the water quality as a result of natural purification. Scope for groundwater recharge through soil-aquifer treatment is also more. Mostly groundnut, maize, sorghum, sunflower, etc... are cultivated in and around Madurai due to the climatic conditions and water resources availability.

Thus, the current study was conducted to investigate the soil pollution load, to understand the appropriateness of wastewater-irrigated soils for vegetable cultivation, and to assess the metal uptake by the food crops and the potential health risks associated with human consumption.

II. RELATED WORK

A) STUDYAREA

Madurai City Corporation is one among the ten metropolitan cities in Tamil Nadu. Having located at almost in the southern part of the state, it is the city for education and business. City covers an area of 148.99 square kilo meters. Climate prevailing is arid to semi-arid. The forecast of the wastewater generation from Madurai City Corporation indicates the quantity of 162.8 MLD at 2014 and is likely to be 338.7 MLD in the year 2044. The irrigation potential of wastewater reuse is assessed as 3000 ha with crops like groundnut, maize, millet etc... during 2014 and about 6000 ha during 2044. Agriculture land at kallambal and kallanthiri villages located in Madurai city was selected as the study area. Two main zones were selected on the basis of Municipal wastewater irrigation and Fresh water irrigation. Surface water from nearby tanks was used for irrigation at the fresh water irrigated zone; it was located in kallanthiri village near Alagarkovil (North of city), whereas wastewater from urban drains (from Kaalavasal, periyar bus stand, palanganathametc) was used for irrigation at the wastewater irrigated zone; it was located in kallambal village near Chinthamani (south of city).



Figure 1: Madurai District Map showing Study Areas

The underlined places in Fig 1 were the study areas showing the Fresh water and Wastewater irrigation zones, (i.e.) Kallambal village near main town Thiruparankundram from where the wastewater has been collected and Alagarkovil located in Madurai North from where the freshwater was collected.



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B) IDENTIFICATION OF CROPS

The study areas were visited and the most viable crops cultivated there had been identified, out of which three crops were selected which were commonly found in both the sites. The description of vegetables examined were shown in Table1, those crops were selected based on the frequent consumption by the people. The crops identified were *Solanum melongena, Cyamopsistetragonoloba* and *sesbania grandiflora*.

PLANT SPECIES	FAMILY	ENGLISH NAME	PART USED
Solanum melongena	Solanaccae	Brinjal	Fruit
Cyamopsistetragonoloba	Fabaceae	Guar (or) Cluster bean	Seed pots
sesbania grandiflora	Amaranthaccae	August tree (or) Agathi	Leaves

C) SAMPLING

Water samples that were used for irrigation practices were collected from each site i.e from freshwater and wastewater irrigation sites, in pre cleaned high-density polyethylene bottles. Soil from agricultural land was collected by digging, using a plastic scooper. Non soil particles e.g. stones, wooden pieces, rocks, gravels, organic debris were removed from soil. Soil was oven dried and this dried soil was sieved through a 2 mm sieve and stored in the labeled polythene sampling bags. A diversity of vegetables were grown in the study area; Solanum melongena, Cyamopsistetragonoloba., sesbania grandiflora were collected from each site of sampling zone and stored in polythene bags and kept in laboratory. Hence, totally 5 types of samples were collected from each irrigation sites. Edible parts of vegetables were then oven dried and ground into powdered form for making the plant digests.

D) DIGESTION OF SAMPLES

1 g of soil and vegetable samples was digested by 25 ml Tri acid mixture i.e. HNO_3 , H_2SO_4 , $HClO_4$ at 9:2:1 ratio until the transparent solution appeared. Water samples were filtered and it was made to 100 ml of extract by adding distilled water and stored for further analysis. The tri acid mixture extraction was used for the heavy metal analysis only. Separate extraction for phosphorous and potassium was made, which was then used for determining total phosphorous and potassium present in samples.

E) ANALYSIS FOR MACRO-NUTRIENTS

Total nitrogen (N) was determined using the Kjeldahl procedure. By using the extraction made for phosphorous analysis, presence of total phosphorous was determined using spectrophotometer. By using the extraction made for potassium analysis, the presence of total potassium in the samples was determined using Flame photometer.

F) HEAVY METAL ANALYSIS

Concentrations of heavy metals were determined by using Atomic Adsorption Spectrophotometer. Extraction made using tri acid mixture was used for the heavy metal analysis.

G) DATA ANALYSIS

i. Transfer factor: Soil to plant metal transfer was computed as transfer factor (TF), which was calculated by using the equation. TF = Cplant / Csoil

Where, Cplant is the concentration of heavy metals in plants and Csoil is the concentration of heavy metals in soil.

ii. Daily intake of metals (DIM): Daily intake of vegetables in adult was calculated by the following equation

DIM = Cmetal x Cfactor x Dfood intake / Baverage weight

Where, C_{metal} - concentration of heavy metal, $D_{food intake}$ – Daily food intake and $B_{average}$ weight - Average body weight, C_{factor} - Conversion factor (0.085), Daily intake of vegetables – Approximately considering 4 members were



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there in a family consuming $\frac{1}{2}$ kg of Brinjal and 300g of Agathi for a week. And 50 g of guar for 3 days in a week. An average body weight of 60 kg was considered. These assumptions were made regarding the local cultural practices that exposed to these vegetables. iii. Health risk index (HRI): To assess the human health risk of heavy metals, it is necessary to calculate the level of human exposure to that metal by tracing the route of exposure of pollutant to human body. There subsist many exposure routes for heavy metals that depend upon a contaminated media of soil and vegetables on the recipients. In the present study, metal concentrations in the collected vegetables were used to calculate the health risk index (HRI). The value of HRI depends upon the daily intake of metals (DIM) and oral reference dose (R_{fd}). R_{fd} is an estimated per day exposure of metal to the human body that has no hazardous effect during life time (US-EPA, 2006).The health risk index for Zn2+, Pb2+, Cr2+, Cd2+, Fe2+, Mn2+ and Cu2+ by consumption of contaminated vegetables was calculated by the following equation, HRI = DIM /

 R_{fd}

Where, DIM represents the daily intake of metals, R_{fd} represents reference oral dose. R_{fd} value for Zn2+, Pb2+, Cr2+, Cd2+, Fe2+, Mn2+ and Cu2+ is 0.3, 0.004, 1.5, 0.001, 0.02, 0.033 and 0.04 (mg/kg bw/day) respectively (US-EPA,2006).

III. EXPERIMENTAL RESULTS

A. CHARACTERISTICS OF FRESH WATER AND WASTEWATER

The physico-chemical characteristics of the wastewater and freshwater which were used for the cultivation of these vegetables were analyzed and tabulated as below in Table 2. The chemical parameters measured were temperature, pH, electrical conductivity (EC), chemical oxygen demand (COD), biochemical oxygen demand (BOD), total dissolved solids (TDS), total solids (TS) and total suspended solids (TSS).

Parameters	Fresh water	Wastewater
	samples	samples
pH	7.56	6.33
Electrical conductivity (µs/cm ⁻¹)	246.5	1.403
Total solids (mg/L)	122	717
Total dissolved solids (mg/L)	122	694.3
Chemical oxygen demand (mg/L)	Nil	124
Total suspended solids (mg/L)	Nil	22.7
Biological oxygen demand (mg/L)	Nil	10.5

Table 2: Physico-chemical Characteristics of Fresh water and Wastewater

B. MACRO-NUTRIENTS CONCENTRATION IN SAMPLES

Table 3:	Macro nutrients	present in	the sampl	es

Samples	Total nitrogen %	Total phosphorous %	Total potassium %
Fresh Water	0.0005	0.05	0.138
Waste Water	0.003	0	2.677
Fresh Watered soil	0.014	0.488	8.18
Waste Watered soil	0.0145	1.330	9.05
Fresh Water S.m (Solanum melongena)	0.350	0.967	1.442
Waste Water S.m (solanum melongena)	0.701	0.492	1.759
Fresh Water C.t (Cyamopsistetragonoloba)	2.242	1.590	1.374
Waste Water C.t (Cyamopsistetragonoloba)	3.923	1.580	1.127
Fresh Water S.g(sesbania grandiflora)	4.623	1.027	1.958
Waste Water S.g(sesbania grandiflora)	6.024	1.130	2.389

Total nitrogen (N) was determined using the Kjeldahl procedure. By using the extraction made for phosphorous analysis, presence of total phosphorous was determined using spectrophotometer. By using the extraction made for



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potassium analysis, the presence of total potassium in the samples was determined using Flame photometer and the results were tabulated in Table 3.



Figure 2: Concentration of Macro- Nutrients

By using these values a graphical representation was made in order to interpret the observed values which were shown in Fig 2. From the figure we can observe the increased values of potassium in fresh watered and waste watered soils. And there were increased Nitrogen content in *Sesbania grandiflora* samples.

C. CONCENTRATION OF HEAVY METALS IN WATER AND SOIL SAMPLES

Concentrations of heavy metals were determined by using Atomic Adsorption Spectrophotometer. Extraction made using tri acid mixture was used for the heavy metal analysis. Heavy metal concentration in irrigated water at fresh water zone and wastewater zone was analysed and results were shown in the table. While comparing the heavy metals concentration of both fresh water and wastewater cultivated crops, majority heavy metals did not show any significant differences, although the concentration of Pb2+ and Cu2+ showed significant differences which can be seen in Table 4.

SAMPLES	UNIT	Zn	Pb	Cr	Cd	Fe	Mn	Cu
Fresh Water	mg/L	bdl	0.026	bdl	0.0137	0.2	bdl	bdl
Waste Water	mg/L	bdl	0.342	bdl	0.0079	0.057	bdl	bdl
Fresh Watered Soil	mg/kg	94.6	bdl	bdl	4.3	3600	345.9	140.8
Waste Watered soil	mg/kg	0.45	157.8	bdl	12.9	2828.6	44	124.3

Table 4: Heavy Metal concentration in soil and water

*bdl – below detectable level

D. HEAVY METAL CONCENTRATION IN VEGETABLE SAMPLES

Heavy metal concentrations were analysed for the vegetable samples cultivated in both the irrigation sites and the results were shown in the table. In food crops grown in wastewater, first in Solanum melongena (Brinjal) the extent of heavy metal enrichment was seen for Pb2+, Fe2+ and Cu2+.



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Table 5: Heavy metal concentration in vegetable samples

SAMPLES	Heavy metals concentrations in Vegetable Samples (mg/kg)						
	Zn	Pb	Cr	Cd	Fe	Mn	Cu
Fresh Water Solanum	15.7	263.15	bdl	143.8	171.4	15.7	201.6
melongena							
Waste Water Solanum	51.35	157.8	bdl	100.7	542.8	9.4	bdl
melongena							
Fresh Water	39.1	bdl	bdl	26.6	257.1	15.7	69
Cyamopsistetragonoloba							
Waste Water	64.8	26.3	71.4	34.5	285.7	6.3	82.8
Cyamopsistetragonoloba							
Fresh Water sesbania	16.7	263.15	bdl	12.9	285.7	4.4	174
grandiflora							
Waste Water sesbania	5.4	552.6	bdl	2.87	400	31.4	91.1
grandiflora							

*bdl – below detectable level

Table 5 shows the increased Cr and Cd contents especially in the crops grown in wastewater comparing to that of crops grown in fresh water. Results of *sesbania grandiflora* grown in wastewater indicates more risk in consuming Pb content.



Figure 3: Heavy metal concentration in samples

From the graphical representation we can observe that in *Cyamopsistetragonoloba* (Kothavarangai) and *sesbania grandiflora* (AgathiKeerai) high enrichment of Pb2+ and Cu2+. From Fig 3 the heavy metal concentration in vegetables grown at wastewater zone the trend appeared as Pb2+ > Fe2+ > Cu2+ > Cd2+ > Mn2+ > Zn2+ > Cr2+ while in vegetables grown at surface water zone the trend appeared as Pb2+ > Fe2+ > Cd2+ > Cd2+ > Zn2+ > Mn2+ > Cr2+ while in vegetables grown at surface water zone the trend appeared as Pb2+ > Fe2+ > Cd2+ > Cd2+ > Mn2+ > Zn2+ > Mn2+ > Cr2+.

E. TRANFER FACTOR, DIM AND HRI VALUES FOR VEGETABLE SAMPLES

Soil to plant metal transfer was computed as transfer factor. The metal transfer factor in vegetables from the study area were investigated and tabulated as below. Values of DIM calculated for people exposed to these vegetables were presented in the table. These data revealed that the values of daily intake of metal were high for vegetables grown at waste water zone comparing to the vegetables grown at fresh water zone. By considering an average of 4 members in a family an assumption was made that, consumption of all three crops in the range of; ½ kg of Brinjal and 300 g of Agathi greens for a week and 50 g of Guar for 3days in a week. Later the consumption of 1 person per day is calculated.



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An average body weight of 60 kg per person is being assumed for DIM calculation. The health risk index for heavy metals by consumption of vegetables grown in waste water and freshwater zonesfor people was calculated and values were tabulated as below.

SAMPLES		Zn	Pb	Cr	Cd	Fe	Mn	Cu
Fresh Water Solanum	TF	0.166	0	0	3.33	0.0476	0.045	1.432
melongena	DIM	0.0004	0.007	0	0.004	0.005	0.0004	0.006
	HRI	0.0015	1.864	0	4.074	0.2	0.013	0.143
Waste Water Solanum	TF	11.4	1	0	7.81	0.192	0.21	0
melongena	DIM	0.0015	0.004	0	0.003	0.0154	0.0003	0
	HRI	0.0049	1.12	0	1.92	0.77	0.008	0
Fresh Water	TF	0.413	0	0	6.186	0.071	0.045	1.432
Cyamopsistetragonoloba	DIM	0.0002	0	0	0.0002	0.002	0.0001	0.0005
	HRI	0.0009	0	0	0.188	0.2	0.013	0.143
Waste Water	TF	14.4	0.167	0	2.67	0.1	0.143	0.666
Cyamopsistetragonoloba	DIM	0.0005	0.0002	0.0005	0.0002	0.0002	0	0.0006
	HRI	0.0015	0.047	0.0003	0.244	0.1	0.0014	0.015
Fresh Water sesbania	TF	0.177	0	0	3	0.079	0.127	1.236
grandiflora	DIM	0.0002	0.004	0	0.0002	0.004	0.0006	0.002
	HRI	0.0008	0.93	0	0.183	0.2	0.019	0.062
Waste Water sesbania	TF	12	3.5	0	0.22	0.141	0.714	0.733
grandiflora	DIM	0	0.0078	0	0	0.006	0.0004	0.0013
	HRI	0.0003	1.957	0	0.041	0.283	0.0135	0.032

Table 6: Transfer factor, DIM and HRI values for vegetable samples

IV. DISCUSSIONS

In the present study, two various irrigation sites were identified in which one uses fresh water for irrigation and the other uses wastewater for irrigation purposes. The samples including water, soil and crops cultivated in respective sites were collected and analysed using the knowledge of literature review. The water, soil and vegetable samples were analysed for the major nutrients (NPK) and from the study, significant increase in total P and total K were estimated; especially in Agathi greens. When comparing the total NPK values of freshwater with that of wastewater; significant decrease in macro nutrients was notable in fresh water. There were increased nutrients values for soil samples collected from the wastewater irrigation site while comparing with that of fresh water zone. In the present study, while comparing the heavy metals concentration of both fresh water and wastewater cultivated crops, majority heavy metals did not show any significant differences, although the concentration of Pb2+ and Cu2+ showed significant differences. The present study revealed that wastewater irrigated soil was enriched in Cd2+ and Pb2+. In food crops grown in wastewater, first in *Solanum melongena* (Brinjal) the extent of heavy metal enrichment was seen for Pb2+, Fe2+ and Cu2+. In *Cyamopsistetragonoloba* (Kothavarangai) and in *sesbania grandiflora* (AgathiKeerai) high enrichment was seen for Pb2+ and Cu2+ > Cu2+ > Cu2+ > Cn2+ > Cr2+ while in vegetables grown at surface water zone the trend appeared as Pb2+ > Fe2+ > Cd2+ > Cu2+ > Zn2+ > Cr2+.

The TF values were high in vegetables for Zn2+ and Cd2+ grown in wastewater zone. TF for Cr2+ was lowest for all the vegetables compared to other metals. Trend of metal TF from soil to vegetables grown at wastewater zone was in the order of Zn2+ > pb2+ > Cd2+ > Cu2+ > Mn2+ > Fe2+ > Cr2+. And the trend of metal TF from soil to vegetables grown at fresh water zone was in the order of Cd2+ > Cu2+ > Zn2+ > Mn2+ > Fe2+ > pb2+ > Cr2+. From the results arrived for DIM values it is clear that Chromium and Copper possesses low values and some other metals such as Lead, Cadmium and Zinc carries high DIM values and hence their HRI index would be increased due to high daily intake of those metals. In waste water zone the maximum HRI were found for Agathi greens and in Brinjal; especially for pb2+



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and Cd2+. The maximum HRI was found for *Solanum melongena* (Brinjal) grown at both zones. In fresh water zone only *Solanum melongena* shows high HRI values for pb2+ and Cd2+.

It is suggested that an attention is required for the implementation of proper means to monitor and regulate the municipal effluents. Risk assessment of intake of metal- contaminated vegetables to human health was characterized by HRI. If the value of HRI is less than 1 then the exposed population is said to be safe and if it is equal or greater than 1 then the population will experience health risk. This risk assessment method has been by researchers and proved to be valid and true. DIM and HRI of the study area suggest that *Cyamopsistetragonoloba* grown at wastewater zone was almost safe for consumption but *sesbania grandiflora* and *Solanum melongena* pose severe health risk with regard to the pb2+ and Cd2+.

V. CONCLUSION

The crops cultivated out of wastewater and freshwater in the villages kalambal and kallanthiri were collected and analysed for the heavy metals concentrations and macro nutrients availability. While comparing the heavy metals concentration of both fresh water and wastewater cultivated crops, majority heavy metals did not show any significant differences, although the concentration of Pb2+ and Cu2+ showed significant differences. The present study revealed that wastewater irrigated soil was enriched in Cd2+ and Pb2+. In food crops grown in wastewater, first in*Solanum melongena* (Brinjal) the extent of heavy metal enrichment was seen for Pb2+, Fe2+ and Cu2+. In *Cyamopsistetragonoloba* (Kothavarangai) and in *sesbania grandiflora* (AgathiKeerai) high enrichment was seen for Pb2+ and Cu2+. These data revealed that the values of daily intake of metal were high for vegetables grown at waste water zone comparing to the vegetables grown at fresh water zone. DIM and HRI of the study area suggest that *Cyamopsistetragonoloba* grown at wastewater zone was almost safe for consumption but *sesbania grandiflora* and *Solanum melongena* pose severe health risk with regard to the pb2+ and Cd2+.

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