Mercury Concentration in Vegetables of Pakistan Irrigated by Different Water Sources

[1]QAMAR SHAHEEN, [2]ZAID MAHMOOD*, [2]MUHAMMAD IMRAN,
[3]SYEDA ROBINA GILLANI AND [4]ILYAS AHMED FARIDI
[1]*Food Testing Laboratory, Health Department, Lahore, Pakistan.*
[2]*Institute of Chemistry, University of the Punjab, Lahore, Pakistan.*
[3]*Chemistry Department, University of Engineering & Technology, Lahore, Pakistan.*
[4] *Institute of Public Health, Health Department, Lahore, Pakistan.*

(Received on 1st March 2010, accepted in revised form 19th October 2010)

Summary: Mercury levels were determined in twenty samples of each vegetable *i.e.*, Spinach *(Spinacia oleracea)*, Lettuce *(Lactuca sativa)*, Carrot *(Daucus cariota)*, Capsicum *(Capsicum fistulosus)*, Sweet pea *(Lathyrus Odoratus)*, Potato *(Solanum tuberosum)* and Cabbage *(Brassica oleracea)*, with a special reference of source of water of irrigation, *i.e.*, tube well water, canal water and municipal sewage water. All the samples of vegetables were collected during the year 2006, 2007 and 2008 from the five districts of Pakistan viz Lahore, Kasur, Multan, Bahawalpur and R.Y.Khan. Statistical analysis such as Test of significance and multiple comparison were applied on the data obtained. The results showed that the concentration of Mercury in vegetables irrigated by canal water, sewage water and tube well water was in the range of 3.1-88.9 ppb and 9.0-130.6 ppb. It can be concluded from this study that the uptake of mercury by vegetables collected from above five districts of Pakistan was in the following order. Leafy vegetables > Root vegetables > seedy vegetables.

Introduction

The introduction of heavy metals in food chain was established in the middle of 19th century and water of irrigation was considered as one of the major contributors of entry of mercury in the food chain [1]. Vegetables constitute essential diet components by contributing protein, vitamins, iron, calcium and other nutrients, which are usually in short, supply [2]. They also act as buffering agents for acidic substances produced during the digestion process. Metal accumulation in vegetables may pose a direct threat to human health [3, 4]. Vegetables take up metals by absorbing them from contaminated soils, as well as from deposits on different parts of the vegetables exposed to the air from polluted environments [5].

Mercury exists in water in a number of inorganic and organic forms however most of the mercury in the atmosphere is elemental and inorganic mercury while in water, soil, plants, and animals it is primarily methyl mercury [6]. The global cycling of mercury begins with the evaporation of mercury vapor from land and sea surfaces. Volcanoes can be an important natural source [7]. Methyl mercury biomagnifies up the food chain as it is passed from a lower food chain level to a subsequently higher food chain level through consumption of prey organisms or predators. Fish and fish products are the major, if not the only, source of methylmercury in humans [8]. In human body, about 95% of methyl mercury is absorbed in the gastrointestinal tract, although the exact site of absorption is not known. It is distributed to all tissues in a process completed in about 30 hr. About 5% is found in the blood compartment and about 10% in brain [9]. It is reported in the literature that methyl mercury is converted to inorganic mercury in the brain [10]. Several co-workers studied the characteristics of mercury accumulation in vegetable and determined the total mercury concentrations in various samples of agricultural crops and foods [11, 12].

Food safety and environmental protection is a burning issue of the present era all over the world. At international level a lot of research work is being carried out under different headings like heavy metal contamination, uptake of heavy metal in food items, basket survey, heavy metal contamination- a threat to life etc. In Pakistan it is yet to be investigated that what is the extent of heavy metal pollution in food items in different areas of the country. We have a little information about this issue and this part of our research work is supposed to be very useful to act as base-line information to identify the pattern of food problems in Pakistan.

Results and Discussion

Range of Concentration of Mercury in Vegetables, Irrigated with Canal Water in Five Districts of Punjab, Pakistan

Table-1: Range of concentration of mercury (ppb) in vegetables irrigated with canal water in five districts of punjab, pakistan 2006-2008.

Vegetables	Lahore			Kasur			Multan			Bahawalpur			R Y khan		
	06	07	08	06	07	08	06	07	08	06	07	08	06	07	08
Spinach *(Spinacia oleracea)*	18.6-40.2	13.4-45.01	15.4-61.3	13.9-30.4	18.2-62	13.2-74.6	36.2-88.9	13.9-43.6	15.6-42.9	13.1-35.4	14.6-58.9	12.6-59.2	12.1-34.6	14.0-58.5	12.2-56.3
Lettuce *(Lactuca sativa)*	14.3-.34.2	13.5-42.9	14.5-64.1	12.1-30.2	19.1-60	12.1-64	11.3-24.9	13.1-40.4	11.9-54.4	14.9-38.5	12.5-51.2	13.6-51.3	12.2-38.9	11.6-50.3	14.6-43.9
Carrot *(Daucus cariota)*	11.5-28.6	9.9-31.2	10.2-51.3	9.8-21.6	12.1-48.2	9.3-49.8	8.6-19.3	9.8-22.3	10.2-43.1	9.8-29.9	10.1-43	10-48.2	8.6-31	9.2-41.5	10.1-45
Capsicum *(Capsicum fistulosus)*	8.8-18.2	7.8-21	7.4-34.9	6.9-15.4	9.8-32	6.0-31.6	7.3-14.1	7.2-23.1	6.8-29.5	6.9-17.5	7.1-28.8	6.5-31.8	5.9-20.2	6.3-30.1	6.8-32.3
Sweet pea *(Lathyrus odoratus)*	4.8-10.3	3.9-12.8	4.9-18.6	3.8-8.6	4.8-16.1	3.8-18.9	3.8-6.9	3.8-9.9	3.5-14.8	3.6-9.8	4.1-14	3.9-15.2	3.1-9.8	4-13.2	3.9-14.1
Potato *(Solanum tuberosum)*	8.0-18.2	6.1-21.3	6.9-30.2	7-15.1	8.9-30	5.9-30.1	5.9-11.3	6.2-24.9	6.1-24.3	6.9-17.8	6.1-23.8	6.1-30.4	5.1-18.3	5.8-29.2	5.8-27.3
Cabbage *(Brassica oleracea)*	17.3-35.5	12.2-42.9	13.8-60	12.9-28.8	7.6-58.3	11.2-35.5	10.6-23.3	12.1-40	13.2-50.6	13.2-35.2	13-58.6	12.1-58.3	11-37.2	11.2-55.6	12.9-59.6

Table-1 shows the range of concentration of mercury in seven vegetables, *i.e.*, Spinach *(Spinacia oleracea)*, Lettuce *(Lactuca sativa)*, Carrot *(Daucus cariota)*, Capsicum *(Capsicum fistulosus)*, Sweet Pea *(Lathyrus odoratus)*, Potato *(Solanum tuberosum)* and Cabbage *(Brassica oleracea)*, irrigated with canal water in the five districts of Punjab, during the year 2006-2008. Range of concentration of mercury in spinach is 12.1-88.9 ppb in all the five districts, in lettuce it is 11.3-64.1 ppb while in carrot it is 12.1-49.8 ppb. Similarly in capsicum, sweet pea, in potato, cabbage, it is in the range 6.0-34.9 ppb, 3.1-18.9 ppb, 5.1-30.4 ppb, 7.6-59.6 ppb respectively in all the five districts of Punjab.

Range of Concentration of Mercury in Vegetables, Irrigated with Sewage Water in Five Districts of Punjab, Pakistan

Table-2 shows the range of concentration of mercury in seven vegetables, *i.e.*, Spinach *(Spinacia oleracea)*, Lettuce *(Lactuca sativa)*, Carrot *(Daucus cariota)*, Capsicum *(Capsicum fistulosus)*, Sweet Pea *(Lathyrus odoratus)*, Potato *(Solanum tuberosum)* and Cabbage *(Brassica oleracea)*, irrigated with canal water in the five districts of Punjab, during the year 2006-2008. Range of concentration of mercury in various vegetables was found to be 18.2-130.6 ppb for spinach and 18.6-101.6 ppb, 16.9-98.6 ppb, 13.4-98.4 ppb, 9.0-51.3 ppb, 18.9-98.6 ppb, 21.6-110.3 ppb for lettuce, carrot, capsicum, sweet pea, potato and cabbage respectively among all the five districts of Punjab.

Statistical Evaluation of Mercury Concentration in Spinach with Respect to Water of Irrigation

Table-3 shows the average concentration level of Mercury in spinach irrigated with Canal water and sewage water was 32.6 ± 0.97 and 44.6 ±1.18 respectively. The highest concentration level was seen in spinach irrigated with sewage water *i.e.* 130.6 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 141 (48 from canal water and 93 from sewage water).The average concentration level of Mercury in spinach was statistically different with respect to water of irrigation (*p-value = 0.000*).

Table-2: Range of concentration of mercury (ppb) in vegetables irrigated with municipal sewage in five districts of punjab, pakistan 2006-2008.

Vegetables	Lahore			Kasur			Multan			Bahawalpur			R Y khan		
	06	07	08	06	07	08	06	07	08	06	07	08	06	07	08
Spinach *(Spinacia oleracea)*	28.6-37.2	38.1-56.9	35.4-100.2	28.3-38.5	30.2-58.6	35.5-89.6	45.1-130.6	18.2-28.6	27.3-58.3	20.1-32.2	25.8-54.3	19.6-80.1	20-31.6	28.9-56.2	31.2-89.5
Lettuce *(Lactuca sativa)*	26.3-35.2	38.1-54.6	40-101.6	25.9-35.3	28.6-56.1	31.3-98.5	19-30.2	22.3-50.1	29.1-91.8	18.6-28.6	23.3-51.1	26.8-98.4	18.6-29.3	21.1-48.9	19.9-86.4
Carrot *(Daucus cariota)*	21.1-31.6	39-61.2	35.4-98.6	24.3-36.1	30.4-67.6	31.5-84.2	16.9-28.2	21.9-58.6	21.8-80	19.6-24.5	25.5-50.8	21.6-84.1	20-37.2	27.2-59.8	21.8-84.2
Capsicum *(Capsicum fistulosus)*	21-32.9	33.2-46.5	29.6-98.4	20.9-31	25.6-51.9	31.2-90	13.4-21.9	21.8-46.3	28.4-81.9	19.6-28.2	21.5-50	19.8-69.1	18.3-30	25.6-50.7	29.2-78.7
Sweet pea *(Lathyrus odoratus)*	13.6-19.5	21.6-29.8	18.4-51.3	14.6-21.5	15.2-31.9	19.3-49.2	9-15.6	14.2-28.3	16.3-43	9.1-14.2	15.6-27.3	14.6-50	10.5-16.2	14.6-29.2	19.1-39.8
Potato *(Solanum tuberosum)*	28.6-39.2	39.6-55.3	35.7-98.6	28.8-38.2	30.1-56.5	38.9-98.2	19.2-31.1	29.6-54.3	31.4-83	18.9-27.8	27.6-51.9	28.4-83.1	19.2-29.2	27.2-58.1	19-76.3
Cabbage *(Brassica oleracea)*	31.6-57.5	41.2-60.3	41.9-110.3	35.5-46	31.8-70.2	41.6-105.2	25-38.2	31.6-60.2	37.4-98.2	22.2-34.1	33.2-61.9	35.4-93.3	21.6-38.2	28.1-63.2	25.3-88.9

Table-3: Statistical evaluation of Mercury in Spinach & Lettuce with respect to Water of irrigation

	SPINACH			LETTUCE		
Source	**CW***	**SW****	**Total**	**CW**	**SW**	**Total**
Mean	**32.6**	**44.6**	**38.6**	**29.5**	**41.9**	**35.7**
Std. Dev	**±16.7**	**±20.5**	**±19.7**	**±12.831**	**±20.5**	**±18.2**
Std. Error	**±0.97**	**±1.18**	**±0.80**	**±0.74**	**±1.19**	**±0.74**
>PAL[1]	**48**	**93**	**141**	**24**	**70**	**94**
N²	**300**	**300**	**600**	**300**	**300**	**600**
F=62.01 , p-value=0.000				***F=78.6 , p-value=0.000***		

* CW= Canal water, ** SW= Sewage water
1. > PAL. Greater than permissible ambient level
2. N. Total No of Samples

Statistical Evaluation of Mercury Concentration in Lettuce with Respect to Water of Irrigation

Table-3 shows the average concentration level of Mercury in lettuce irrigated with Canal water and sewage water was 29.5 ± 0.74 and 41.9 ±1.19 respectively. The highest concentration level was seen in lettuce irrigated with sewage water *i.e.* 101.6 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 94 (24 from canal water and 70 from sewage water). The average concentration level of Mercury in lettuce was statistically different with respect to water of irrigation (*p-value = 0.000*)

Statistical Evaluation of Mercury Concentration in Carrot with Respect to Water of Irrigation

Table-4 shows the average concentration level of Mercury in carrot irrigated with Canal water and sewage water was 22.3 ± 0.54 and 41.1±1.05 respectively. The highest concentration level was seen in carrot irrigated with sewage water *i.e.* 98.6 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 71 (1 from canal water and 70 from sewage water). The average concentration level of Mercury in carrot was statistically different with respect to water of irrigation (*p-value = 0.000*)

Table-4: Statistical evaluation of Mercury in Carrot & Potato with respect to Water of irrigation

	CARROT			PATATO		
Source	**CW***	**SW****	**Total**	**CW**	**SW**	**Total**
Mean	**22.3**	**41.1**	**31.7**	**14.7**	**43.1**	**28.9**
Std. Dev	**±9.41**	**±18.1**	**±17.2**	**±5.98**	**±19.8**	**±20.4**
Std. Error	**±0.54**	**±1.05**	**±0.70**	**±0.35**	**±1.14**	**±0.83**
>PAL[1]	**1**	**70**	**71**	**0**	**77**	**77**
N²	**300**	**300**	**600**	**300**	**300**	**600**
F=255.2 , p-value=0.000				***F=567.2 , p-value=0.000***		

* CW= Canal water, ** SW= Sewage water
1. > PAL. Greater than permissible ambient level
2. N. Total No of Samples

Statistical Evaluation of Mercury Concentration in Potato with Respect to Water of Irrigation

Table-4 shows the average concentration level of Mercury in potato irrigated with Canal water and sewage water was 14.7 ± 0.35 and 43.05±1.14 respectively. The highest concentration level was seen in potato irrigated with sewage water *i.e.* 98.6 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 77 (0 from canal water and 77 from sewage water). The average concentration level of Mercury in potato was statistically different with respect to water of irrigation (*p-value = 0.000*)

Statistical Evaluation of Mercury Concentration in Capsicum with Respect to Water of Irrigation

Table-5 shows the average concentration level of Mercury in capsicum irrigated with Canal water and sewage water was 16.3 ± 0.40 and 38.6±1.04 respectively. The highest concentration level was seen in capsicum irrigated with sewage water *i.e.* 98.4 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 54 (0 from canal water and 54 from sewage water). The average concentration level of Mercury in capsicum was statistically different with respect to water of irrigation (*p-value = 0.000*)

Table-5: Statistical evaluation of Mercury in Capsicum & sweet pea with respect to Water of irrigation

	CAPSICUM			SWEETPEA		
Source	**CW***	**SW****	**Total**	**CW**	**SW**	**Total**
Mean	**16.3**	**38.6**	**27.5**	**8.56**	**22.9**	**15.8**
Std. Dev	**±6.94**	**±18.0**	**±17.6**	**±3.72**	**±10.6**	**±10.8**
Std. Error	**±0.40**	**±1.04**	**±0.72**	**±0.22**	**±0.61**	**±0.44**
>PAL[1]	**0**	**54**	**54**	**0**	**5**	**5**
N²	**300**	**300**	**600**	**300**	**300**	**600**
F=401.2 , p-value=0.000				***F=493.2 , p-value=0.000***		

* CW= Canal water, ** SW= Sewage water
1. > PAL. Greater than permissible ambient level
2. N. Total No of Samples

Statistical Evaluation of Mercury Concentration in Sweet Pea with Respect to Water of Irrigation

Table-5 shows the average concentration level of Mercury in sweet pea irrigated with Canal water and sewage water was 8.56 ± 0.22 and 22.9 ±0.61 respectively. The highest concentration level was seen in sweet pea irrigated with sewage water *i.e.* 51.3 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 5 (0 from canal water and 5 from sewage water).

Statistical Evaluation of Mercury Concentration in Cabbage with Respect to Water of Irrigation

Table-6 shows the average concentration level of Mercury in cabbage irrigated with Canal water and sewage water was 27.1 ± 0.70 and 49.2 ±1.14 respectively. The highest concentration level was seen in cabbage irrigated with sewage water *i.e.* 110.3 ppb. Total samples exceeding the PAL for Mercury in food, *i.e.*, 50 ppb were 135 (22 from canal water and 113 from sewage water).

Table-6: Statistical evaluation of Mercury in Cabbage with respect to Water of irrigation

CABBAGE			
Source	**Canal water**	**Sewage water**	**Total**
Mean	**27.1**	**49.2**	**38.2**
Std. Dev	**±12.1**	**±19.7**	**±19.7**
Std. Error	**±0.70**	**±1.14**	**±0.81**
>PAL[1]	**22**	**113**	**135**
N[2]	**300**	**300**	**600**
F=274.8 , p-value=0.000			

1. > PAL. Greater than permissible ambient level
2. > N. Total No of Samples

Discussion

Industrial development has facilitated our lives in many ways. It has made easy to travel, introduces a very high rate of agricultural development, induced new methods of food preparation and thus enabled us for easy living. At the same time there are many problems connected with this development and mainly environmental pollution is the key problem to be resolved in this regard. There are numerous types of environmental pollution as various toxicants are added into the environment due to industrial development [13]. Heavy metals supply a major fraction of this problem and among them mercury is considered as one of the major air pollutants threatening the life of humans on this planet [14]. The heavy metals affect not only the fruits and vegetables but also the human beings which are consumers. National and international regulations on food quality have decreased the maximum permissible levels of heavy metals in human edible food therefore it became necessary to control the concentrations of these metals in food [15]. The present work includes the estimation of mercury in different vegetables based on the water of irrigation as a major source of heavy metal contamination in the vegetables due to environmental pollution. The results discussed above give an overall picture of mercury contamination in vegetables estimated in five different districts of Punjab. The selected districts are Lahore, Kasur, Multan, Bahawalpur and Rahim Yar Khan. The present work was conducted to estimate the concentration of mercury in different vegetables depending upon the source of irrigation. The results show that the range of concentration of mercury in the vegetables grown in five different districts of Punjab is in between 3.1-130.6 ppb irrigated from canal and sewage water. Among them the concentration of mercury did not exist in the vegetables irrigated by tube well water and the results show the maximum concentration of mercury contents in leafy vegetables irrigated by sewage water. Nearly one fourth of all of the vegetables samples (irrigated by sewage water) showed the concentration of mercury more than that of PAL (Permissible ambient level). The results thus show a very genuine demand of further research on this topic. The research may be conducted on the biological aspects of identification, evaluation and control of effects of mercury on human health coming through the vegetables due to pollution of water of irrigation.

Experimental

Sample Collection

Twenty Samples of each vegetable, *i.e.*, Spinach *(Spinacia oleracea)*, Lettuce *(Lactuca sativa)*, Carrot *(Daucus cariota)*, Capsicum *(Capsicum fistulosus)*, Sweet Pea *(Lathyrus odoratus)*, Potato *(Solanum tuberosum)*, Cabbage *(Brassica oleracea)*, were collected from five districts of Pakistan like Lahore, Kasur, Multan, Bahawalpur and R.Y.Khan during the year 2006, 2007 and 2008.

Sample Preparation

All Samples of vegetables were washed three times with distilled and HCl (0.05 M), followed by washing with de-ionized water to ensure dislodging and removal of dust particles. Samples were then dried in a fan-forced oven at 60 ±5 °C for 48 hours and finally grinded using a stainless steel grinder. The grinded samples were passed through a 0.2-mm sieve and stored in plastic vials for further analysis of mercury [16].

Reagents and Glassware

Hydrochloric acid, HNO_3, Hydrogen Peroxide, $KMnO_4$ s , $K_2S_2O_8$, $SnCl_2$ solution, NaCl-Hydroxylamine sulfate solution were purchased from Merck/BDH. All glassware was cleaned with a sodium dichromate cleaning solution followed by detergent, deionized water, 1 M nitric acid and finally with doubly deionized water. The glassware was retained for mercury analysis and kept in 1*M* nitric acid when not in use.

Digestion Method

0.5 g and 1 g of grinded samples were weighed and placed in 50 ml poly-propylene centrifuge tubes and then dried without charring at 55 ºC in a circulating oven. The resulting extract from circulating oven was mixed with $KMnO_4$. The digestion was carried out with conc. HCl and HNO_3. To this mixture 10 ml of 5 % $K_2S_2O_8$ solution was added. The mixture was heated to boiling for 2 hrs in a water bath. Excess of $KMnO_4$ was removed by adding NaCl-hydroxylamine sulfate solution.

Analysis

A Perkin-Elmer model Aanalyst 800 atomic absorption spectrophotometer equipped with a MHS-15 Mercury/Hydride system was used for Cold Vapour atomic absorption spectrophotometry. A Perkin-Elmer electrodeless discharge lamp was used as a light source. The acid digested solution was treated with stannous chloride ($SnCl_2$) to reduce mercury into its elemental form, which was volatilized to vapors. Under aeration the vapors of mercury were carried by air into the absorption cell. The absorbance was measured at the wavelength of 253.7 nm.

Conclusion

The present work was conducted to estimate the concentration of mercury in different vegetables depending upon the source of irrigation. Nearly one fourth of all of the vegetables samples (irrigated by sewage water) showed the concentration of mercury more than that of PAL. It presents a serious status of vegetable contamination and requires further investigations for the formulation of remedial measures in this regard. The toxicity of mercury suggests that immediate steps should be taken at policy level of food safety and water sanitation. Conversion of sewage water into grey water should be mandatory and covered by the legislation.

Acknowledgement

The authors are thankful to the University of the Punjab for funding this project. The provision of analysis facility and related support by *Food Testing Laboratory*, *Health Department*, *Lahore*, *Pakistan and* Institute of Public Health, Health Department, Lahore, Pakistan is highly acknowledged

References

1. L. Jarup, *British Medical Bulletin*, **68**,167 (2003).
2. H. C. Thompson, W. C. Kelly,Vegetable Crops, 5th Ed. McGraw Hill Publishing Company Ltd., New Delhi.Tata McGraw- Hill Publishing company Ltd. pp. 513-515 (1990).
3. M. K. Türkdogan, F. Kilicel, K. Kara, I. Tuncer and I. Uygan, *Toxicol*. *Pharmacol*., **13**, 175 (2003).
3. M. Damek-Poprawa and K. Sawicka-Kapusta, *Toxicology*, **186** (1-2), 1 (2003).
4. G. Zurera-Cosano, R. Moreno-Rojas, J. Salmeron-Egea and R. P. Lora, *Journal of the Science of Food Agriculture*, **49**, 307 (1989).
5. J. C. Clifton, *Pediatric Clinics of North America*, **54**, 237 (2007).
6. W. F. Fitzgerald and T. W. Clarkson, *Environmental Health Perspectives*, **96**, 159 (1991).
7. J. K. Virtanen, T. H. Rissanen, S. Voutilainen and T. P. Tuomainen, *The Journal of Nutritional Biochemistry*, **18**, 75 (2007).
8. E. Cernichiari, R. Brewer, G. J. Myers, D. O. Marsh, L. W. Lapham, C. Cox, C. F. Shamlaye, M. Berlin, P. W. Davidson and T. W. Clarkson, *NeuroToxicology*, **16**, 613 (1995).
9. L. Magos, A. W. Brown, S. Sparrow, E. Bailey, R. E. Snowden and W. R. Skipp, *Archives of Toxicology*, **57**, 260 (1985).
10. D. Liu and C. Qing, *The journal of Applied Ecology*, **13**, 315 (2002).
11. L. Perring, M. I. AlonsoI, D. Andrey, B. Bourqui and P. Z. Fresenius, *Journal of Analytical Chemistry*, **370**, 76 (2001).
12. R. Jedrzejczak, *Food Additives and Contaminant*, **19**, 996 (2002).
13. K. C. Jones, C. J Sy mon and A. E. Johnston, *Science of the Total Environment*, **67**, 75 (1987).
14. J. C. Moreira, *Science of the Total Environment*, **188**, 61 (1996).
15. A. Husain, Z. Baroon, M. Al-Khalafawi, T. Al-Ati and W. Sawaya, *Environmental International*, **21**, 803 (1995).
16. S. M. I. Huq, J. C. Joardar, S. Parvin, R. Correll and R. Naidu, *Journal of Health Population Nutrition*, **24**, 305 (2006).