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Health and Safety Concerns: Quantitative Studies of Leaching of Metals from Glazed Surfaces of Traditional Ceramic Potteries

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Abstract

Traditional ceramic wares have been known as a source of heavy metals poisoning. Traditional ceramic potteries may be improperly glazed, and the glaze used to make the pottery may contain over amounts of heavy metals. These over glazed ceramic wares can release deadly metal into foodstuff and constitute health hazards. In this work, Quantitative studies were done according to ASTM C 738.81 (1982) leaching standard test methods for the determination of trace amount of selected metals from glazed surfaces of traditional ceramic potteries by 4% acetic and 2% citric acid standard solutions at different temperatures. Finally, leaching potential has been done using ICP-MS analysis. The capacity of each ceramic tableware sample ranged between 250 and 350ml. The ceramic wares selected randomly from products available in the local markets at Doha (Qatar), Cairo (Egypt) and Gharyan (Libya).

Keywords

Heavy Metals, Traditional Ceramic Potteries, Acetic Acid, Citric Acid, ICP-MS

1. Introduction

Potters, since old times, have been utilizing different metal salts as parts in coating mixes to confer smooth and splendid surfaces and to improve shading to artistic items (Belgiad J. E. 2003). A coating glaze is a thinner layer of glass melded onto the surface of clay pottery. There are many health risks caused by the intake of heavy metals from the leachate of foods or liquids exposed to these glazed ceramic wares. Ingestion of even very low level of lead causes significant neurological and cognitive effects in humans (Valadez-Vega C. *et al.,* 2011). The issue with the presence of heavy metals in coated ceramic ware lies in the way that these contaminants can be go away to drinks and foods by leaching procedure, which directly related to the physical and chemical states of the food, for example, temperature and pH (González de Mejía and Craigmill 1996). Traditionally potteries tableware widely used in many Arab countries. There outer bodies prepared from local clays mixed with light burning clay minerals then fired in traditional special furnaces the firing temperature not exceeded than 1000°C (Rhodes D. 1973). After that, the fired wares covered with glaze then refired another time at 1050°C. Lead added to this type of glazed pottery to improve the chemical and color properties of the glazed surfaces to help them to avoid the harmful attack of detergents. Lead also improves the bond properties between pottery and glaze. The glazed traditional pottery not fired enough so no complete glassification of the items body occurs (OECD 1994). Moreover, when colored coating are developed, compounds of lead, cadmium, chromium, zinc, copper and other heavy metals are present. Since over glaze decoration are not subjected to high temperatures, they are more striking than those utilized as a part of high-temperature under glaze coloring methods, yet they are additionally more powerless against wear and damage (Anonymous A. 1930; Cunningham J. 1982; Colbert N. W. 1993; Stapleford G. H. 1936). Most of foods are acidic in nature therefore; weak acids such as acetic acid and citric acid are used for the leaching of heavy metals. Different studies have been demonstrated that lead and cadmium discharge relies on upon the kind of acid and that specific acids present in foodstuffs, can as powerful as 4% acetic acid (Geller R. F. and Creamer A. S. 1939; Sheets R. W. 1997; Sheets R. W. *et al.,* 1996; Somogyi A. *et al.,* 1999). The FDA has altered the extreme suitable convergences of leachable lead range from 0.5 to 3.0 μg/ml relying upon the kind of dinnerware. In addition to lead, other heavy metals are known to be noxious were distinguished in numerous sorts of glazed products and may constitute a wellbeing peril if such utensils are not utilized legitimately (Sheets, R. W. 1997; Dayan A. and Paine A. 2001; Domingo J. L. 1996; Exley C. *et al.,* 2007; Fosmire G. J. 1990; Kesteloot H. *et al.,* 1968; Krinitz B. and Hering R. 1971; Nordberg G. F. *et al.,* 2007; Pier S. M. 1975; Santamaria A. B. 2008; Sheets R. W. 1998; Sundar S. and Chakravarty J. 2010). Due to the large number of lead poisoning cases, the European Union, decided to monitor dinnerware for lead and cadmium release and set the directive 84/500/EC, which determine the specific migration limits for ceramic articles, which summarized at Table (1) (Demont M. *et al.,* 2012, WHO 1976; The Council of the European Communities, 1984; The European Parliament, 2004).

Table 1. *European Directive 84/500/EEC relating to ceramic articles specific migration limits to foodstuffs.*

Category	Specifications	Pb level	Cd level
Flatware	Internal depth 25mm	0.8 mg/dm^2	0.07 mg/dm^2
Small hollowware Volume	Volume < 3L	4 mg/ L	0.3 mg/ L
Large hollowware	Volume > 3L	1.5 mg/ L	0.1 mg/ L

Intense exposure to different concentrations of heavy metals prompts sickness, anorexia, spewing, gastrointestinal irregularities and dermatitis. Heavy metals are risky in light of the fact that they tend to bioaccumulation in the ecology and human bodies (Khare H. N., et al., 2014). Young kids are more defenseless to the impacts of lead uptake since they ingest a few times the percent ingested contrasted and grown-ups and in light of the fact that their brains are more plastic and even short, exposures may impact formative procedures (Sue YJ. Mercury, 2015).

Heavy metals group such as aluminum, antinomy, arsenic, barium, beryllium, bismuth, cadmium, gallium, germanium, lead, mercury, nickel, silver, strontium, tellurium, thallium, tin, titanium, vanadium and uranium have no settled human biological uses and are considered as non-essential metals and may be classified as toxic and harmful (Chang L. W., *et al.,* 1996). In this study we examined and evaluated the sequential leaching of glazed heavy metals e.g. lead, cadmium, cobalt, zinc, iron, copper, chromium, manganese and barium from traditional glazed potteries ware collected from local Qatari markets and other samples of traditional glazed pottery derived from Egyptian and Libyan markets.

2. Materials and Methods

Traditional pottery samples collected from Souq Waqif traditional pottery shops and Omani market at Doha city. Other traditional samples derived from Al Fustat traditional ceramics area, Cairo, Egypt and another samples from Gharyan city, Libyan. Two similar pottery samples of approximately equal size and volume were collected from each glazed pottery.

Table 2. *The names of the potteries and characteristics of the chosen glazed ceramics.*

No.	Sample Code	Country	Country of Origin	Physical Characteristics
1	DOH SW_1	Qatar	Yemen	Brown colored with smooth surface
2	DOH SW_2	Qatar	Yemen	Brown colored with smooth surface
4	DOH OM_1	Qatar	Oman	Reddish Brown colored with smooth surface
5	DOH OM_2	Qatar	Oman	Brown colored with smooth surface
7	CAI FUS_1	Egypt	Egypt	Brown colored with smooth surface
8	CAI FUS_2	Egypt	Egypt	Brown colored with smooth surface
10	LIB GH_1	Libya	Libya	Blue colored with smooth surface
12	LIB GH_3	Libya	Libya	Brown colored with smooth surface

The range of volumes for each group of pottery from 250 to 300 ml in average. The names of the potteries and characteristics of the chosen glazed ceramics for the leaching studies are given in Table (2). Firstly, the tested samples washed carefully with detergent then washed with tap water followed by deionized water and dried at 65^{o}C temperature to complete dryness. Each tested item was filled with the leaching solution up to the rim until it started overflowing with test solution at dark place at the desired temperature for 24h. The heavy metals leaching method used at this study was that of the ASTM C 738.81 (ASTM 1982) and agreed by USFAD for heavy metals leaching from tableware (Wallace et al., 1985). Samples of ceramic pottery subjected to leaching by using 4% acetic acid and 2% citric acid solution to investigate the leachability of heavy metals at acidic medium at pH similar to foodstuff. Inductively Coupled Plasma Mass Spectrometry ICP-MS (Perkin Elmer NexION 300 D) analyzed all concern heavy metals.

3. Results and Discussion

Inductively Coupled Plasma Mass Spectrometry (ICP-MS) technique was the most powerful technique used for heavy metals determination. In this study, ICP-MS was used for determining the concentration of trace and ultra- trace amounts of the heavy metals that was leached out of the traditional potteries. The detection limits of the ICP-MS, Perkin Elmer NexION 300D used for multi-elements analysis are given in Table (3).

Table 3. *Perkin Elmer NexION 300a (ICP-MS) Minimum detection limit for multi-element analysis for metals of interest in the leaching studies (ppb, µg/l).*

Metals	Pb	Cd	Zn	Fe	Cu	Cr	Mn	Ba
(*µg/l*)	0.0004	0.01	0.10	0.10	0.009	0.01	0.03	0.002

The results for heavy metals concentrations leached from the traditional potteries were given in Tables (4-7) and Figures (1-4). The results ranged from 18.140 to 1760.205 µg/l, 34.135 to 1825.118 µg/l, 17.336 to 1613.136 µg/l and 61.175 to 1984.247 µg/l for lead leaching by using 4% acetic acid at 35°C, 45°C, 65°C and for lead leaching by using 2% citric acid at 45°C respectively. The results shows that lead was released from all the traditional wares samples but significantly, high level of lead was leached from Libyan and Egyptian samples.

Table 4. *Metal released into 4% acetic acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 35^{0}C ND means "not detected".*

Metals (*µg/l*)	**DOH SW_1**	**DOH SW_2**	**DOH OM_1**	**DOH OM_2**	**EGY FUS_1**	**EGY FUS_2**	**LIB GH $_1$**	**LIB GH $_3$**
Lead (Pb)	63.009	18.140	147.494	40.473	1282.102	446.234	1760.205	431.155
Cadmium (Cd)	ND	ND	20.476	ND	ND	46.676	301.360	33.308
Zinc (Zn)	740.781	403.940	843.038	481.563	425.439	976.059	698.433	426.328
Iron (Fe)	465.732	376.312	263.122	174.080	775.756	860.442	255.016	280.305
Cupper (Cu)	187.054	58.153	ND	50.254	79.243	ND	109.318	47.086
Chromium (Cr)	ND	ND	ND	ND	ND	ND	120.253	136.147
Manganese (Mn)	193.038	244.139	228.129	132.153	672.446	836.047	318.139	619.637
Barium (Ba)	29.391	16.817	64.086	115.132	214.049	173.267	428.105	726.114

Cadmium was below the detectable limits for some samples of the traditional wares but detected for others. For Doha, traditional samples cadmium could be detected in one sample with the concentration 20.476µg/l at 35°C with 24h contact period by using 2% acetic acid but at the same conditions, it leached by concentrations 46.676, 301.360 and 33.308 µg/l for traditional samples FUS_2, GH_1 and GH_3, respectively.

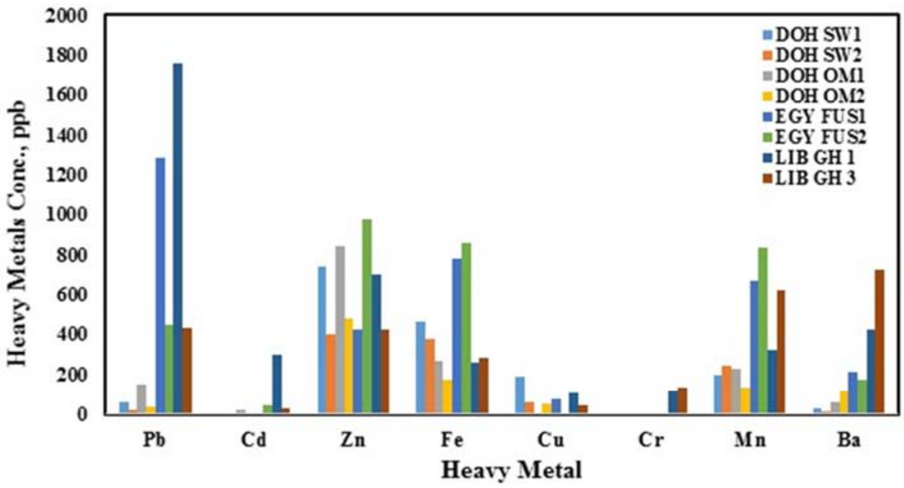

Figure 1. *Metal released into 4% acetic acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 35°C.*

At 45°C cadmium could not be detected in four samples, SW_1, SW_2, OM_2 and FUS_1 while it is detected in the other four samples OM_1 (19.082 µg/l), FUS_2 (38.572 µg/l), GH_1 (180.019 µg/l) and GH_3 (274.553 µg/l). Moreover, at 65°C cadmium was not detected in three samples but detected in the other five samples ranged from 12.266 µg/l to 137.119 µg/l and the maximum concentration found in Libyan samples. By using 2%, citric acid cadmium was leached to the acidic solution of five samples ranged from 18.006 to 365.067 µg/l but not leached from samples SW_1, SW_2 and OM_2.

Table 5. *Metal released into 4% acetic acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 45°C ND means "not detected".*

Metals (*µg/l*)	**DOH SW_1**	**DOH SW_2**	**DOH OM_1**	**DOH OM_2**	**EGY FUS_1**	**EGY FUS_2**	**LIB GH $_1$**	**LIB GH $_3$**
Lead (Pb)	84.023	30.002	156.120	49.042	1471.044	577.033	1825.118	523.243
Cadmium (Cd)	ND	ND	19.082	ND	ND	38.572	180.019	274.553
Zinc (Zn)	980.337	460.239	906.142	513.436	186.113	1050.26	752.340	650.341

Metals (*µg/l*)	DOH SW_1	DOH SW_2	DOH OM_1	DOH OM_2	EGY FUS_1	EGY FUS_2	LIB GH $_1$	LIB GH $_3$
Iron (Fe)	324.536	472.221	274.412	198.136	850.229	890.037	345.202	322.152
Cupper (Cu)	317.537	294.407	218.271	373.075	422.218	352.203	294.237	362.057
Chromium (Cr)	ND	ND	46.341	57.343	36.524	28.423	228.458	476.392
Manganese (Mn)	208.141	292.137	328.347	229.244	795.366	914.427	473.016	928.502
Barium (Ba)	364.058	429.218	201.553	332.009	532.147	642.357	618.133	937.352

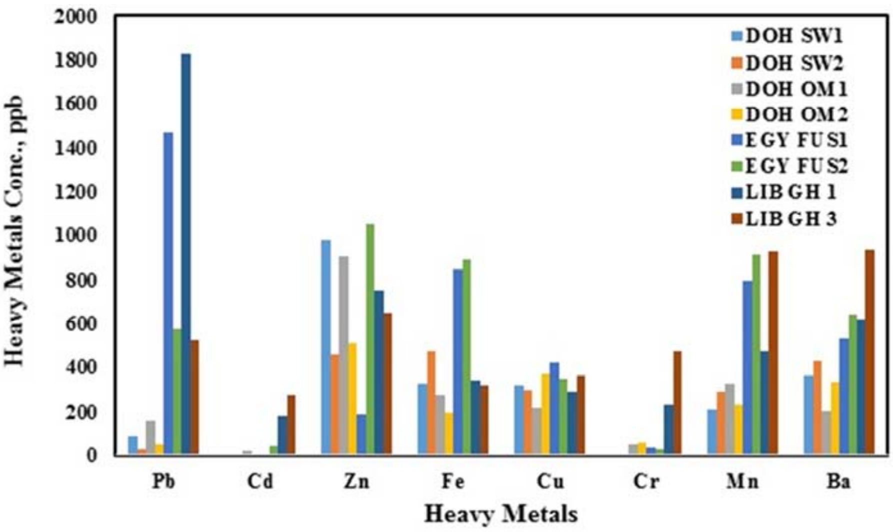

Figure 2. *Metal released into 4% acetic acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 45°C.*

The leached zinc from the traditional glazed pottery samples wares varied depending on the temperature and the leachate acidic solution, zinc leached from all samples under investigations. The results indicated that 2% citric acid solution more powerful than 4% acetic acid solution in the leaching process of zinc.

Table 6. *Metal released into 4% acetic acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at* 65^0C *ND means "not detected".*

Metals (*µg/L*)	DOH SW_1	DOH SW_2	DOH OM_1	DOH OM_2	EGY FUS_1	EGY FUS_2	LIB GH $_1$	LIB GH $_3$
Lead (Pb)	34.228	25.424	94.332	52.493	1051.115	421.247	1613.136	268.238
Cadmium (Cd)	ND	ND	12.266	ND	30.377	33.415	137.119	104.218
Zinc (Zn)	634.125	289.450	592.147	324.358	158.131	574.080	403.115	296.532
Iron (Fe)	394.194	297.257	195.344	106.446	539.436	693.125	291.195	314.785
Cupper (Cu)	224.366	210.210	147.794	269.444	253.228	281.336	207.448	179.603
Chromium (Cr)	65.643	22.476	160.283	156.261	77.088	105.299	254.193	582.859
Manganese (Mn)	158.147	254.346	302.659	192.389	554.875	647.609	374.885	788.268
Barium (Ba)	204.831	225.852	132.875	136.974	142.049	367.668	588.536	574.028

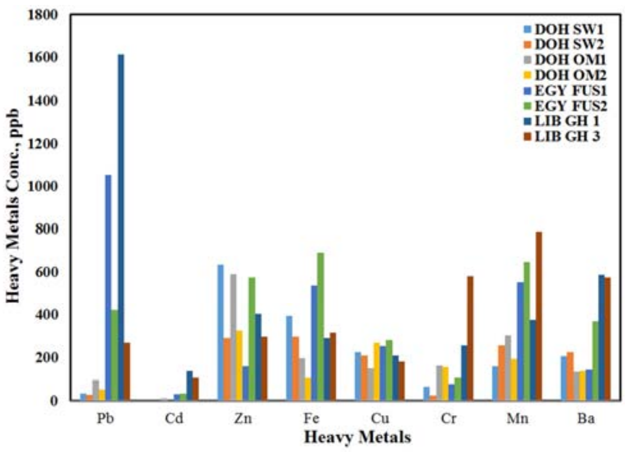

Figure 3. *Metal released into 4% acetic acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 65°C.*

Zinc leached range from 258.522 µg/l to 976.059 µg/l, 186.113µg/l to 980.337µg/l and 158.131 µg/l to 634.125µg/l at the temperatures 35°C, 45°C and 65°C respectively. The amounts of zinc leached by using 2% citric acid ranged from 438.388µg/l to 1320.179µg/l at temperature 45°C.

The leached Iron from the examined traditional glazed potteries changed according to the temperature change and the used acid. The results indicated that, iron leached from all samples with different concentration values ranged from 160.144 to 860.442 µg/l, 198.136 to 920.226 µg/l and 106.446 to 814.339 µg/l at leaching temperatures 35°C, 45°C and 65°C, respectively by using acetic acid but equal to 394.789 to 1076.30 µg/l when using citric acid. The leachability of citric acid more than the leachability of the acetic acid at the desired temperature.

The leaching of copper from the traditional glazed ceramic wares by using 4% acetic acid at 35°C differs from sample to another the concentration of copper ranged from 47.086 µg/l to 187.054 µg/l and not detected at samples such as SW_1, OM_2 and FUS_2. However, at temperature 45°C the concentration of leached copper increased taking the range from 187.344µg/l to 422.218 µg/l, but at 65°C the concentration range take the sequence from 137.285 µg/l to 347.164 µg/l lower than that at 45°C.

Table 7. *Metal released into 2% citric acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 45^{0}C ND means "not detected".*

Metals (*µg/l*)	DOH SW_1	DOH SW_2	DOH OM_1	DOH OM_2	EGY FUS_1	EGY FUS_2	LIB GH $_1$	LIB GH $_3$
Lead (Pb)	92.068	64.242	263.386	84.238	1680.21	734.217	1984.247	1021.167
Cadmium (Cd)	ND	ND	34.193	ND	35.196	42.368	365.067	348.225
Zinc (Zn)	1040.690	831.364	1130.262	702.632	438.388	1320.179	866.764	927.075
Iron (Fe)	454.006	758.025	621.117	394.789	1038.12	1069.037	573.022	723.022
Cupper (Cu)	478.115	612.124	562.146	576.132	603.158	534.128	489.963	758.833
Chromium (Cr)	104.388	89.045	174.495	324.416	103.066	214.063	286.868	397.913
Manganese (Mn)	253.159	388.144	463.482	329.847	897.158	942.154	578.774	1003.004
Barium (Ba)	428.204	463.857	247.173	361.852	628.625	679.183	786.164	1018.046

The data remarked to that the Egyptian and Libyan samples leached higher heavy metals than the traditional Qatari samples. The leaching concentration ranged from 410.112 µg/l to 758.833µg/l, also these values considered as higher as against USEPA permissible limits (1975).

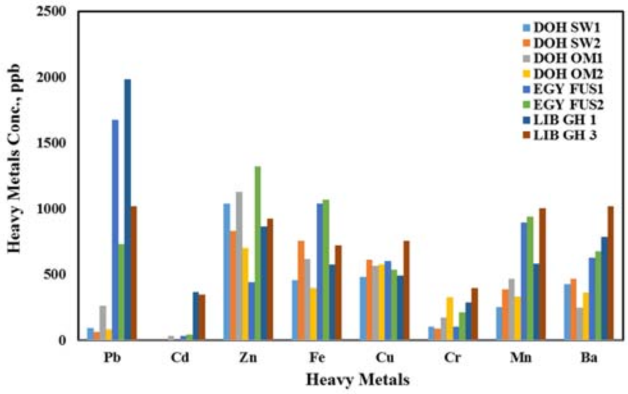

Figure 4. *Metal released into 2% citric acid leachate solution in ppb (µg/l) from traditional glazed pottery samples after a contact period of 24 hours at 65°C.*

The concentration of chromium leached from the traditional ceramic wares by using 4% acetic acid at 35°C was remarked for three samples only OM_3, GH_1 and GH_3 there values were 104.113, 120.253 and 136.147 µg/l respectively but not detected for the other nine samples. For traditional samples leached chromium by using acetic acid at temperature 45°C and 65°C was found to be ranged from 28.423 µg/l to 476.392 µg/l and 22.476 µg/l to 582.859 µg/l respectively. We can remark that the leached amount of chromium increased by increasing the temperature and the samples of Doha (SW_1, SW_2 and SW_3) not leached any chromium at 45°C but chromium started leached at 65°C but by small amounts. Moreover, the Libyan samples (GH_1, GH_2 and GH_3) still leached higher heavy metals than the other samples. By using 2% citric acid, we remarked that more chromium leached at the worked temperature 65°C and ranged from 89.045 to 485.538 µg/l and citric acid is the most powerful leaching agent for the heavy metals under the working conditions. The concentration of manganese leached from the traditional wares by using acetic acid was remarked in all examined samples but by different ranges according to the temperature change. The concentration of manganese at 35°C, 45°C and 65°C ranged from 132.153 to 836.047 µg/l, 208.141 to 928.502 µg/l and 158.147 to 788.268 µg/l respectively when using 4% acetic acid but when using 2% citric acid at 65°C the leached manganese ranged from 253.159 to 1003.004 µg/l. Barium also leached by detectable concentrations for all samples and temperatures in the range from 29.391 to 726.114 µg/l at 35°C, from 175.081 to 937.352 µg/l at 45°C and from 132.875 to 588.536 µg/l at 65°C by using 4% acetic acid. Barium was leached by using 2% citric acid at 650C the concentration ranged from 241.258 to 1018.046 µg/l.

The results obtained for the traditional glazed pottery samples indicated that by using acetic acid or citric acid the samples released high amounts of heavy metals higher than that released by the same leached acid in the case of modern ceramic samples (Mohamed et al., 1995). Lead leached from approximately of all traditional samples by high concentration at different temperatures and leaching agents for 24 leaching time. The lead leached from the internal surface of wares more over the leaching limit of lead as per Directive 84/500/EC. Libyan and Egyptian samples leached the highest amounts of lead in the ICP-MS results than Qatari Doha samples. Libyan samples had a highly decoration design on the internal surface that was exposed to the leaching solution test and lead could be potentially released from those colored glazes.

Especially lead from all heavy metals is poisonous in all forms (Gosselin et al., 1984) and in small concentrations (Nriagu, 1988; Ferguson, 1990). Lead cases physiological and neurological effects in children cases. Lead as a toxic element interfere with the enzymes function, signal systems and membranes, perhaps by combining with certain proteins active sites. Recent studies (Bergdhal et al., 1997) remarked that lead binding to a certain red blood cell proteins, like g-aminolevulic acid dehydratase. This binding leads to inhibition of the enzymatic activity. Very low blood-lead concentrations in children can case, accumulation effects which lead to neurological damage (Harvard Medical School, 1992).

4. Conclusion

The potential human health risk from the tested traditional glazed pottery samples in this study is from the wrong glazed decoration and internal covered layers. The leachable lead content of these wares is high enough to constitute a human health hazard. Results of this study remarked that heavy metals hazards are smaller when compared with the newly-purchased glazed ceramic wares than from dinnerware manufactured before 1970 (Sheets, 1997, 1998a).

Therefore, it is recommended that the relevant regulatory agencies in the countries should come forward to enact necessary regulations to control the use of these compounds in the industry and issue guidelines affecting proper glazing and firing procedures for these articles in the furnaces to minimize the possibility of toxic heavy metal leaching into the food chain.

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