ISSN-1996-918X

Pak. J. Anal. Environ. Chem. Vol. 25, No. 1 (2024) 49 – 56

http://doi.org/10.21743/pjaec/2024.06.05

Quantitative Assessment and Source of Heavy Metals in Bricks: A Case Study of Lakki Marwat, Khyber Pakhtunkhwa, Pakistan

Muhammad Bilal[1], Javed Ali[1], Zia-ur-rehman[1], Zahid Farooq[1] and Hizbullah Khan[2*]

[1]*Department of Chemistry, Kohat University of Science and Technology, Kohat, Pakistan.*

[2]*Department of Chemistry, University of Science and Technology, Bannu, Pakistan.*

*Corresponding Author Email: dr.hizbullah@ustb.edu.pk

Received 04 August 2023, Revised 29 November 2023, Accepted 20 January 2024

Abstract

Humans' indiscriminate usage of heavy metals has a negative impact on the world's natural ecosystem. One of the main causes of contaminating the fresh and healthy atmosphere in rural Pakistan is the use of coal and soil in an improper manner by brick kilns. Different heavy metal concentrations and their impact on the surrounding areas of Pakistan's District Lakki Marwat were investigated in this study. Acid digestion methods were used to prepare samples, which were then examined using an Atomic Absorption Spectrophotometer. The concentrations of cadmium, iron, and lead in certain samples exceeded the WHO's permitted level, whereas nickel and zinc concentrations were within the limit. It was also discovered that the supply of raw materials is the key determinant in the buildup of heavy metals in the areas near brick not the age of the kiln.

***Keywords**:* Bricks kiln, Heavy metals, Coal, Atomic Absorption Spectrophotometer

Introduction

Houses, both in urban and rural areas, used to be made up of bricks for hundreds of decades, but approximately 30% of the world's population still lives in earthen structures [1-3]. The increase in the population of the world compels people to build more houses and buildings which need more brick kilns. Increases in the population have also forced researchers to search for new routes for the preparation of bricks. From the past to the present, bricks are mostly prepared from mud which contains either clay and sand or straw as binder. Presently it is also prepared from concrete, red mud, and other wastes of industries [4-6]. The muddy bricks contain 20 to 30% alumina, 2 to 5% lime, 50 to 60% silica, and less than 7% iron oxide [7]. The muddy brick is fired at 900–1000 °C to achieve its strength. The use of clay and sand in brick manufacturing depleted natural resources and harmed the environment. To supply the brick business, virgin resources are mined from riverbeds, hillsides, and fertile soil, leaving the mining area unreclaimed [8, 9]. Furthermore, typical brick manufacture necessitates a significant amount of energy, resulting in extremely low costs. As a result of widespread deforestation, wood and trees are used as a source of energy in the fire of young bricks [2]. In SAARC countries, brick kilns are considered the 3rd largest consumers of coal after power plants and steel. According to a report on bricks in Pakistan, approximately 3005 metric tons of coal were used for brick preparation in 2010 [10]. However, the improper and low quality combustion of coal in brick kilns is being practiced, causing air and soil pollution of the earth (emission factor). Different types of polluted gases: (SO_2, NO_x (nitrogen oxides), CO, CO_2, carbon, and particulates (dust) are emitted from the combustion of low quality coal, which have adverse impacts on human health and other animals and plants [11-16]. The low quality and improper use of coal also caused contamination of surrounding areas with heavy metals such as lead, nickel, chromium, iron, etc. [17-24]. Heavy metals are non-biodegradable substances that accumulate to a considerable extent in the environment. They are bio-transferred, bio-accumulated, and thus transferred into the human body [25-27]. Heavy metals are found in varying amounts throughout the biosphere. Pollution from anthropogenic activity has introduced and contributed some of these heavy metals to the biosphere in recent times. Because of their toxicity at specific concentrations, non-biodegradability, and translocation through food chains, which is responsible for their buildup in the biosphere, heavy metals in the environment are of enormous ecological relevance [8, 28-32].

These metals may negatively have effects on either ground water quality, soil ecology, and agricultural production, so it ultimately have a harmful impact on the health of animals and plants through the food chain. These effects are influenced by the biological availability of hazardous metals, which is influenced by metal ion speciation in the soil. As a result, determining the quantities of free metal ions in soil becomes extremely critical. The concentration of free metal ions in soils is determined not only by the total metal content but also by the metal species present [25, 33]. Therefore, the aim of this study was to analyze how trace metal deposition takes place in the surrounding areas of the brick kilns of Lakki Marwat. It was also checked whether the age of kiln establishment has an effect on the concentration of heavy metals or not.

Materials and Methods

Chemicals and Reagents

Nitric acid (HNO_3) and hydrogen peroxide (H_2O_2) of analytical grade were purchased from Sigma Aldrich Chemie GmbH, Germany.

Sample Collection

Samples were taken from different brick kilns of the district Lakki Marwat, Khyber Pakhtunkhwa, Pakistan in June 2020. Those areas were selected which were away from the main road (National Highway, N-55) or other industrial areas. From each kiln, five samples (coal, sand, soil, brick, and plant) were collected. The details of the sample collection are given below in Table 1.

For the baking of bricks, coal is used as fuel, which is brought from the different mines of Pakistan. Unused coal samples were selected from different kilns in solid form. Sand and soil samples were taken from different places around 500 meters away from each kiln. For taking soil samples, first of all, the soil surface was dug about one foot deep. The soil was taken from the upper surface till down in order to get uniform soil from up till down from each kiln. Random samples of brick were collected from each kiln. Sand samples were collected from nearby sand mountains. Sand is usually brought from these mountains for the preparation of raw brick. Similarly, two plants samples (*Acacia Arabica* and *Zizyphus maurtiana Lam*) were collected from two kilns i.e. B_3 and B_8. These two species of plants were selected from those regions which were grown near kilns. All these samples were stored in polyethylene bags and were brought to the labs for further analysis. The samples of soil, sand, and bricks were crushed and sieved between 200 µm to 400 µm particle size.

Table 1. **Sample collection points.**

Year of Establishment	1990	1991	1996	1999	2000	2001	2005	2009	2009	2010
Kiln Place	Naurang	Naurang	Tarikhel	Kalan	Gandi Chowk	Muslim Bagh	Tari Khel	Behram Khel	Adamzai	Sarai Gambila
Kiln Name	Barkat	Latif	Salim	Asmat	Dawood	Hashim	Rafi	Bashir	Ismail	Mutabar
Sample Code	B_1	B_2	B_3	B_4	B_5	B_6	B_7	B_8	B_9	B_{10}

Sample Treatment

The sample was prepared by a wet acid digestion method. 2 g of each sample (coal, soil, sand, brick, and plant) was taken into five separate china dishes [34]. 5 mL of HNO_3 was added in each sample, and, then the sample was heated at 80 ^oC till the samples became semi-solid. Then 5 mL of HNO_3 was poured into the sample, followed by 5 mL of Hydrogen peroxide (H_2O_2) and heated until semi-solid samples were obtained again. All the samples were taken out from the heating plate and were kept at room temperature for some time. When the temperature of the samples dropped to room temperature, 5 mL of HNO_3 was added and the samples were left at room temperature for 24 hours. The material was filtered with filter paper after twenty-four hours. To get all of the metal nitrates down in the flask, distilled water was run through the samples numerous times through filter paper.

Sample Analysis

Determination of nickel, cadmium, zinc, iron, and lead in each sample, was carried out using Atomic Absorption Spectrometer (PerkinElmer PinAAcleTM 900T) equipped with the intuitive winlab32TM for A.A. software running under Microsoft window. Before analysis of the sample, calibration of the Atomic Absorption Spectrometer was done using standard solutions of each metal (Nickle, Cadmium, Zinc, Iron and Lead) purchased from PerkinElmer.

Results and Discussion

Cadmium

Cadmium is a highly poisonous element and is responsible for several diseases. A small amount of cadmium has a negative effect on the arteries of the human kidney. It interacts with enzymes and causes Itai-itai [1], a painful condition. The acceptable maximum amount of Cadmium is 0.02 mg/kg, according to the WHO [35]. Table 2 shows that in both plants: *Zizyphus maurtiana Lam* and *Acacia Arabica*, the concentration of cadmium is higher than the permissible limits recommended by WHO. The table also indicates that in almost all samples of different kilns, cadmium was detected. The coal sample had the highest levels of cadmium of all the samples. This gives a clue that the main cause of cadmium, around different kilns, is the coal that is brought to kilns from different mines in Pakistan. Cadmium concentrations in soil, brick, and sand samples obtained from various brick kilns in Lakki Marwat were also found to be greater than WHO's maximum allowed values [35]. From the results, it was found that Ismail (B9) and Salim (B3) kilns gave the highest concentration. If you see the years of establishment, it indicates that it is not the age of the kiln but the source of raw materials that is the main factor for the accumulation of heavy metals in the surroundings of the kiln.

Table 2. **Cadmium concentration (mg/L) in samples taken from different kilns of Lakki Marwat.**

Sample	Year	Coal	Soil	Sand	Brick	Plant name	
B_1	1990	0.03± 0.00	0.02± 0.00	0.01± 0.00	0.02± 0.00	**Zizyphus maurtiana**	
B_2	1991	0.02± 0.00	0.03± 0.00	0.02± 0.01	0.03± 0.01	***Stem***	***Leaf***
B_3	1996	0.16± 0.01	0.08± 0.00	0.05± 0.01	0.05± 0.00	0.16 ±0.0	0.05 ±0.0
B_4	1999	0.02± 0.00	0.02± 0.00	0.03± 0.00	0.01± 0.00		
B_5	2000	0.03± 0.01	0.03± 0.00	0.02± 0.00	0.02± 0.01		
B_6	2001	0.01± 0.00	0.01± 0.00	0.02± 0.00	0.02± 0.00		
B_7	2005	0.03± 0.00	0.05± 0.00	0.04± 0.00	0.03± 0.01	***Acacia Arabica***	
B_8	2009	0.02± 0.00	0.03± 0.00	0.04± 0.00	0.03± 0.00	Stem	Leaf
B_9	2009	0.16± 0.01	0.08± 0.00	0.05± 0.00	0.04± 0.00	0.53 ±0.0	0.17 ±0.0
B_{10}	2010	0.03± 0.00	0.08± 0.00	0.04± 0.00	0.06± 0.00		

Nickel

Human activities may be to blame for the greater concentration of plants. Nickel Itch is the most prevalent illness induced by nickle. A small amount of Ni is required by the body since it is predominantly found in the pancreas, where it plays a crucial role in insulin production, whereas higher concentrations have distinct side effects [36]. According to WHO guidelines, the maximum acceptable limit for Nickel is10 mg/kg in plants [37]. The concentration of nickel in Acacia Arabica was below the acceptable level, as shown in Table 3. Nickel concentrations in soil, bricks, and sand were found to be below the allowed level [37]. In coal, the concentration of nickel was also recorded below the normal range. Among different kilns, Salim (B3), Bashir (B8), and Ismail (B9) kilns showed the highest amount, which were located in the same area. These results indicated that not only coal is solely responsible for heavy metal accumulation in the region, but also the geological presence of salt of particular metals may contribute to it [38].

Table 3. **Nickel concentration (mg/L) in samples taken from different kilns of Lakki Marwat.**

Sample	Year	Coal	Soil	Sand	Brick	Plant name	
B_1	1990	N.D± 0.00	0.29± 0.01	0.19± 0.01	0.19± 0.00	**Zizyphus maurtiana**	
B_2	1991	0.03± 0.01	0.07± 0.01	0.16± 0.00	0.07± 0.00	***Stem***	***Leaf***
B_3	1996	0.60± 0.01	0.08± 0.01	0.07± 0.01	0.07± 0.01	N.D	N.D
B_4	1999	0.07± 0.01	0.05± 0.00	0.07± 0.01	0.07± 0.01		
B_5	2000	0.03± 0.01	0.04± 0.00	0.06± 0.00	0.05± 0.01		
B_6	2001	0.08± 0.00	0.41± 0.01	0.28± 0.00	0.28± 0.00	***Acacia Arabica***	
B_7	2005	0.01± 0.01	0.37± 0.00	0.06± 0.01	0.01± 0.01	Stem	Leaf
B_8	2009	0.07± 0.00	0.60± 0.01	0.07± 0.01	0.05± 0.00	0.02± 0.00	0.04± 0.00
B_9	2009	0.15± 0.00	0.29± 0.01	0.18± 0.00	0.21± 0.01		
B_{10}	2010	0.06± 0.01	0.07± 0.01	0.07± 0.01	0.06± 0.00		

Zinc

Zinc is a micronutrient that is important in a variety of physiological and metabolic processes in many organisms [36]. Higher zinc amounts, on the other hand, can be toxic to the body. In plants, the WHO recommends the maximum amount of 50 mg/kg [37]. There was no zinc concentration found in *Zizyphus maurtiana Lam*, although nickel concentrations were found to be below the permitted level in Acacia Arabica. Table 4 shows that in brick, sand, soil, and coal samples the amount of zinc recorded is below the permissible range. Among these, the sand sample of the B1 sample gives the highest value, which is 3.39 mg/L.

Table 4. **Zinc concentration (mg/L) in samples taken from different kilns of Lakki Marwat.**

Sample	Year	Coal	Soil	Sand	Brick	Plant name	
B_1	1990	01± 0.00	0.41± 0.00	3.39± 0.00.	N.D	**Zizyphus maurtiana**	
B_2	1991	N.D	N.D	0.03± 0.00	N.D	***Stem***	***Leaf***
B_3	1996	0.01± 0.00	N.D	N.D	0.01± 0.0	N.D	N.D
B_4	1999	N.D	N.D	0.02± 0.00	0.01± 0.0		
B_5	2000	N.D	N.D	N.D	N.D		
B_6	2001	N.D	N.D	N.D	N.D	***Acacia Arabica***	
B_7	2005	N.D	0.04± 0.00	N.D	N.D	Stem	Leaf
B_8	2009	N.D	N.D	N.D	N.D	0.11± 0.01	0.15± 0.01
B_9	2009	N.D	N.D	N.D	N.D		
B_{10}	2010	N.D	N.D	0.05± 0.00	0.03± 0.0		

Iron

Iron is an essential element for both humans and animals. It's a necessary part of hemoglobin [36]. The WHO recommends a level of iron in plants of 20 mg/kg [37]. Table 5 shows that the concentrations of iron in both plants were within the allowed level. However, in bricks, coal, sand, and soil, concentrations of iron were recorded above the permissible range as shown in Table 5. Among different kilns, the soil of B7 and B10 showed the highest concentration of iron. The red colour of soil also clues to the high concentration of iron.

Table 5. **Iron concentration (mg/L) in samples taken from different kilns of Lakki Marwat.**

Sample	Year	Coal	Soil	Sand	Brick	Plant name	
B_1	1990	11.46± 0.16	12.67± 0.26	11.54± 0.09	10.68± 0.43	**Zizyphus maurtiana**	
B_2	1991	11.03± 0.69	12.88± 0.03	11.23± 0.38	11.01± 0.17	***Stem***	***Leaf***
B_3	1996	10.38± 0.28	9.89± 0.11	12.94± 0.28	9.31± 0.18	N.D	0.20± 0.99
B_4	1999	1.10± 1.34	12.48± 0.23	12.57± 0.26	6.03± 0.82		
B_5	2000	12.64± 0.13	12.39± 0.19	11.38± 0.32	9.87± 0.23		
B_6	2001	11.97± 0.10	12.18± 0.21	12.38± 0.07	11.92± 0.33	***Acacia Arabica***	
B_7	2005	2.90± 0.76	13.45± 0.58	11.68± 0.15	11.30± 0.03	Stem	Leaf
B_8	2009	12.22± 0.74	12.24± 0.01	12.00± 0.18	9.47± 0.15	1.78± 0.63	4.15± 0.19
B_9	2009	8.84± 0.12	13.08± 0.91	12.06± 0.15	12.57± 0.46		
B_{10}	2010	8.77± 0.03	13.47± 0.10	12.66± 0.11	13.19± 0.13		

Lead

The hazardous properties of lead are well-known. To various degrees, lead affects all organs and functions of the human body. It builds up in the bones, kidneys, aorta, spleen, and liver as we become older. About 120 mg of lead is found in the human body, largely in the skeleton and in lower amounts in hair and blood [17]. The WHO recommends a lead limit of 2 mg/kg in plants [25, 39]. Table 6 illustrates that in both plants, concentrations of lead are recorded below the permissible range. The table shows that in almost all samples, lead is detected. However, different samples (bricks, coal, sand, and soil) have different concentrations of lead. In samples, B3, B8, B9, and B10, the lead concentration crossed the permissible limit.

Table 6. **Lead concentration (mg/L) in samples taken from different kilns of Lakki Marwat.**

Sample	Year	Coal	Soil	Sand	Brick	Plant Name	
B_1	1990	0.34± 0.28	0.97± 0.98	1.10± 0.48	0.83± 0.82	***Zizyphus maurtiana***	
B_2	1991	1.13± 0.31	1.26± 0.56	0.78± 0.29	1.15± 0.33	***Stem***	***Leaf***
B_3	1996	1.57± 0.16	1.45± 0.10	0.97± 0.25	2.01± 0.27	0.55± 0.02	0.51± 0.08
B_4	1999	0.86± 0.27	1.11± 0.11	1.67± 0.14	0.65± 0.39		
B_5	2000	1.34± 0.63	0.58± 0.25	0.56± 0.54	1.33± 0.36		
B_6	2001	0.50± 0.62	N.D	N.D	0.44± 0.08	***Acacia Arabica***	
B_7	2005	1.13± 0.54	1.12± 0.30	1.27± 0.06	1.10± 0.19	Stem	Leaf

Heavy metals found in raw materials are integrated into baked bricks, according to the findings. In kiln baking, coal is utilized as a fuel. It was thought that coal was one of the sources of heavy metals and that coal transferred the majority of heavy metals in bricks. However, current findings suggested that heavy metal concentrations in some coal samples are lower than heavy metal concentrations in sand and soil [34]. This indicates that wholly coal is not a cause of heavy metals contamination in bricks. Heavy metals may be incorporated into bricks from coal, soil, and sand. The kilns used the soil and sand which are present in the passage of water which comes from the mountains of District Lakki Marwat. It is believed that these mountains may be the main source of heavy metals. This soil is then used in brick formation and causes heavy metal contamination of bricks. When maximum concentrations of different heavy metals in bricks were compared, it was found that the order of different heavy metals was Iron > Zinc > Lead > Nickel > Cadmium. From the year of the establishment of different kilns, it was concluded that the age of the kiln is not the main factor for the accumulation of heavy metals in bricks.

Conclusion

In this work, different heavy metals were evaluated in soil, sand, bricks, and plants in the surrounding areas of the brick kilns of Lakki Marwat, Khyber Pakhtunkhwa, Pakistan. From overall results, it was found that in some samples, concentrations of cadmium, iron, and lead crossed the permissible limit of WHO whilst the nickel and zinc concentrations were below the permissible limit. It is also concluded that kilns that are near hilly areas (Betanni hills) crossed the permissible limit of WHO. So, it can be clued that wholly coal and the age of kilns are not the only cause of heavy metals contamination in bricks but sources of raw materials (soil, sand, and other components) used for the preparation of raw bricks also significantly contribute to heavy metals accumulation in the bricks kilns.

Acknowledgements

The author, Hizbullah Khan is grateful to the Higher Education Commission of Pakistan (HEC) for providing financial support, the project number is 20-10669.

Conflict of Interest

The authors declare no conflict of interest.

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