Assessment of heavy metal(loid) contents of clay bricks manufactured and utilized as structural building material in Türkiye

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Abstract

Heavy metal(loid)s (HMs) originating from natural and anthropogenic activities can have toxic effects on humans even at low concentrations. Clay bricks (CBs), generally produced by mixing clay and water, are formed by firing the air-dried mixture to make them durable and stable. During firing, the CB suffers some chemical and physical changes and turns into a new artificial material. CBs, known as masonry units, have been one of the most used building materials throughout the history of construction. CB may naturally contain HMs depending on the geochemical structure of the clay used in the production phase. In this study, major and minor oxides and HM distributions in forty-five CB samples collected from thirty-one CB factories that provide approximately one-third of the CB utilized in buildings in Türkiye were determined for the first time using an energy-dispersive X-ray fluorescence spectrometer. The average concentrations (in %, dw) of major and minor oxides in CB samples are in order of SiO_2 (49.9) > Al_2O_3 (17.8) > CaO (9.5) > MgO (8.2) > Fe_2O_3 (7.5) > SO_3 (3.6) > Na_2O (3.3) > K_2O (1.8) > TiO_2 (0.9) > P_2O_5 (0.2) > MnO (0.1). The average concentrations (in mg/kg dw) of Fe, Ti, Mn, Cr, Sr, V, Ni, Zr, Zn, Cu, Co, Pb and As in CB samples were analyzed as 52779, 5329, 736, 341, 23, 192, 190, 110, 85, 44, 39, 14 and 8, respectively. According to the enrichment factor results based on the Earth's crust average, it was revealed that Cr, Ni, and As were naturally moderately enriched.

Introduction

The term heavy metal(loid)s (HMs) is used to collectively refer to a group of metals such as titanium (Ti), vanadium (V), chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn), zirconium (Zr), mercury (Hg), lead (Pb) and metalloids such as germanium (Ge), arsenic (As), antimony (Sb) and tellurium (Te) whose densities are approximately five times greater than the density of water (Turhan et al. 2020; Miletić et al. 2023; Din et al. 2023). The source of HM pollution, which threatens the environment and human health even at very low concentrations due to its bioaccumulative nature, toxicity, and persistence in the ecosystem, can be both natural (geogenic and lithogenic) and anthropogenic (Ali et al. 2019). An enhanced content of HMs in the environment, such as rocks, soils, and waters, can take place in connection with natural processes such as volcanic eruptions, weathering of metal-bearing rocks, and acid drainage (Ali et al. 2019; Miletić et al. 2023). Global industrialization, mining, agricultural activities, and urbanization trends around the world have led to an increase in the anthropogenic share of HMs in the environment (Alloway 2013; Edelstein and Ben-Hur 2018; Ali et al. 2019; Rezapour et al. 2022; Turhan et al. 2023). HMs can be classified into two groups: essential HMs (Cu, Co, Zn, Ni, Fe, Mn, Cr III, etc.), which are required for biological physiology and function, and non-essential HMs (As, Hg, Pb, Cd, and Cr IV), whose biological functions are unknown (Tchounwou et al. 2012; Khaneghah et al. 2020; Apau et al. 2022; Ding et al. 2022; Kollander et al. 2023). Accumulation of HMs in organs causes various chronic and acute diseases affecting the entire system: immune, cardiovascular, endocrine, nervous, skeletal, etc. (Miletić et al. 2023). The toxicity of HMs depends on several factors, such as concentration, duration, dose of HM, and route of exposure (Tchounwou et al.

2012). Humans can be exposed to HMs in three ways: ingestion, inhalation, and dermal (skin) contact (Miletić et al. 2023).

The construction industry plays a vital role in the socio-economic development of any country such as sustainable economy, urbanization, and population growth (Koroneos and Dompros 2007; Arianpour and Arianpour 2022). The construction sector utilizes more building and raw materials by weight than other industrial sectors (Koroneos and Dompros 2007). Nearly 50% of all materials extracted from the Earth's crust are converted into building materials (cement, concrete, gas concrete, clay brick, pumice brick, roofing tile, ceramics marble, granite, limestone, gypsum, etc.) (Koroneos and Dompros 2007). However, construction and demolition waste constitutes a large building material waste stream and poses significant environmental impacts because these wastes can contain high levels of natural radionuclides and heavy metals. Therefore, the disposal and recycling of these waste materials presents great difficulties (Koroneos and Dompros 2007). There has been a sustainable increase in urbanization and population growth in Türkiye in the last decades. The Turkish construction sector is one of the most important industrial sectors and a crucial motivating supporter of these social developments. There is an increase in the building stock due to the increasing demands of the construction sector. In Türkiye, the number of apartments issued with building permits was 319,720 in 2019 and reached 695,804 in 2022 (TÜİK 2024). As it is known, Türkiye is a country prone to earthquakes. Türkiye's location on the border of the Eurasian and African plates makes it a seismically active region. On February 6, 2023, southeastern Türkiye was shaken by two major earthquakes that devastated eleven provinces (Mavroulis et al. 2023). Tens of thousands of buildings collapsed, including newly built apartment buildings, and many more were later demolished (Mavroulis et al. 2023). As a result of these demolitions, thousands of tons of building material wastes and debris containing toxic chemicals and HMs were generated. Unfortunately, several earthquake debris disposal sites in these provinces operated in or near agricultural areas and surface water bodies (Mavroulis et al. 2023). Also, this pollution can be carried by water and wind and reach areas far from the pollution source or remain as waste. HM concentrations in soil may be high at greater distances from the main source. This situation poses serious threats to public health and the environment. Therefore, it is important to know the HM concentrations in each building material that creates these wastes to determine the extent to which agricultural lands and water resources are polluted.

Brick, generally known as a clay-based masonry unit, is one of the most used building materials throughout the history of construction due to their durability and high compressive strength and is mainly used in the construction of external and internal walls of buildings (Mavroulis et al. 2023; Lachheb et al. 2023). Brick is manufactured by firing a mixture of soil-based raw materials such as clay, silt, and sand at 800–1200 °C. Approximately 340 billion tons of clay are utilized worldwide every year for brick production (Aakash 2014). The Turkish clay industry has been growing since 1990 due to the high demands of the domestic building markets (Arianpour and Arianpour 2022). A total of 28 billion CBs were produced between 2018 and 2022, and approximately 5.6 billion CBs are produced annually in Türkiye (İMSAD 2022). To date, many studies on the energy consumption, usage, and carbon emissions of brick production have been published in the literature (Darain et al. 2013; Kumbhar et al. 2014; Yüksek et al. 2020; Rehman et al. 2020; Xin et al. 2023). However, there is a lack of comprehensive studies on the determination of HM concentrations in CB samples. Therefore, the investigation of HM levels contained in CBs is important and mandatory to assess the risks that workers working in the brick production line properly manage earthquake debris and determine the extent of HM pollution that abandoned brick factories can produce. The purposes of this study are to (i) determine the concentrations of major and minor oxides and HMs in CB samples collected from thirty-one CB factories using an energy dispersive X-ray fluorescence (EDXRF) spectrometry and (ii) calculate enrichment ratio (to clay average) and enrichment factor (to Earth's crustal average) of the HMs. The novelty of this study is that it is the first attempt to raise awareness about the presence of HMs accompanying CBs, one of the most used building materials, and to create a database on HM distributions of brick kilns in the Western Black Sea Region of Türkiye.

Materials and methods

Collection, preparation and analysis of sample

The CB factories where the samples within the scope of this study were collected are located in the Boyabat district of Sinop province and Tosya district of Kastamonu province in the Western Black Sea Region of Türkiye. Boyabat basin is located in the middle part of the Pontide tectonic belt, one of the tectonic units of Türkiye, and was established within the Tertiary aged depression area (Ustaömer et al. 2007). Thick subduction-accretion complexes consisting of ocean floor sediments and basic magmatic rocks in this basin developed during the closure of the Paleo-tethys and Neotethys oceans (Ustaömer et al. 2007). In the basin, in addition to the alluviums formed by the Gökırmak depression, clayey limestone (marl) observed in the raw material area, there are also conglomerate, sandstone, and mudstone units deposited in different periods (Ustaömer et al. 2007). Tosya and its immediate surroundings are located in the North Anatolian Fault Zone, one of the important tectonic areas of Türkiye. There is Devrez depression (Devrez Stream) formed due to this fault. Alluviums at the bottom of the Devrez depression extend throughout the valley. The raw material area is within the Pliocene lacustrine storage area.

Forty-five CB samples and seven clay samples were collected from thirty-one CB plants, and clay quarry, the locations of which are shown on the map in Fig. 1. Samples were brought to the sample preparation laboratory in plastic bags. Following the procedure for preparing the sample mentioned in the references (Turhan et al. 2020; Turhan et al. 2022; Altıkulaç et al. 2022; Altıkulaç and Turhan 2023). CBs were crushed and powdered to fit the calibrated powder geometry in the EDXRF spectrometer. Each CB sample was dried at 110 °C in an oven for 10 hours. Analysis of major and minor oxides, and HMs in CB and clay samples was performed using an EDXRF spectrometer (Spectro Xepos, Ametek) with a thick binary Pd/Co alloy anode X-ray tube (50 kV, 60 W), the properties of which were detailed in previous studies (Turhan et al. 2020; Turhan et al. 2022; Altıkulaç et al. 2022; Altıkulaç and Turhan 2023). NIST SRM 2709 reference material was utilized for quality assurance of the EDXRF spectrometer (Turhan et al. 2020). CB samples were placed in the automatic sampler and counted once for two hours and then analysis processes were completed. The overall uncertainty (%) of Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Sr, Zr and Pb was found to be 0.2, 2.1, 0.4, 0.3, 0.1, 9.8, 0.8, 1.2, 0.6, 3.3, 0.2, 1.2 and 2.8, respectively.

Enrichment ratio and enrichment factor

The enrichment ration (ER) for HM analyzed in CB samples was calculated using the following formula:

$$ER = \frac{(C_{HM})_{CB}}{(C_{HM})_{Clay}} \qquad (1)$$

where C_{HM} is the concentration of HM analyzed in CB and clay samples. The enrichment factor (EF) is an index used to assess the level of enrichment or pollution and to infer the distribution of metals of anthropogenic origin from locations determined by individual metals in environmental samples (Parvez et al. 2023). EF is calculated using the following formula (Kowalska et al. 2018; Turhan et al. 2020):

$$EF = \frac{\left({}^{C_n}\!/\!{}_{C_{Ref}}\right)_{CB}}{\left({}^{C_n}\!/\!{}_{C_{Ref}}\right)_{Earth}} \qquad (2)$$

where C_n and C_{Ref} are the concentration of HM and reference element in the CB sample and geochemical background (Earth's crustal), respectively. In the EF calculations, aluminum (Al) was taken as a reference element. Al has no important anthropogenic input compared to the major geogenic element, and Al's ion potential is close to target elements, reducing variation in comparison (Bourennane et al. 2010; Poh and Tahi 2017). EF values close to unity represent the crustal origin of the HMs (comparable to Earth’s crust); EF < represents a possible mobilization or depletion of HMs; EF > 1 represents that the HM is of anthropogenic origin. Therefore, EF values of 1−2 are considered slightly enriched; conversely, $2 \leq EF < 5$ is moderately enriched, $5 \leq EF < 20$ is severely enriched, $20 \leq EF < 40$ is highly enriched, and $EF \geq 40$ is extremely high enriched (Turhan et al. 2020; Parvez et al. 2023).

Statistical Analysis

Statistical analysis was conducted to assess data distribution, variance homogeneity, and potential differences. The Shapiro-Wilk and Bartlett tests were used to confirm the data distribution and variance homogeneity, respectively. The results of these tests indicated that most of the data did not conform to the normal distribution and/or exhibit variance homogeneity. Thus, logarithmic transformation was applied to the dataset to address the challenges of non-normality and variance heterogeneity and to compare the data on the same scale.

After applying the logarithmic transformation, the data were re- assessed using the Shapiro-Wilk and Bartlett tests. However, some of the transformed data still exhibited deviations from normality or lacked homogeneity of variances. Therefore, the Kruskal-Wallis test was performed to investigate potential differences and subsequent pairwise Wilcoxon rank-sum tests were conducted to identify significant differences. All statistical analyses were conducted using R studio version 2023.06 (R Core Team 2022).

Results and discussion

The distribution of major and minor oxides analyzed in the CB samples is shown in Fig. 2. The concentrations (in dry weight (dw)) of SiO_2, Al_2O_3, CaO, MgO, Fe_2O_3, SO_3, Na_2O, K_2O, TiO_2, P_2O_5 and MnO varied from 46.7 to 52.9%, 14.9 to 20.3%, 7.0 to 12.8%, 6.7 to 18.0%, 5.4 to 11.1%, 0.5 to 6.7%, 2.4 to 4.0%, 1.0 to 2.1%, 0.6 to 1.7%, 0.1 to 0.3% and 0.1 to 0.2%, respectively. The average concentrations of Al_2O_3, CaO, MgO, Fe_2O_3, SO_3, Na_2O and K_2O are higher than the average concentrations of 15.9%, 9.4%, 5.4%, 1.1%, 0.1%, 2.7% and 1.1% in Earth's crust, respectively (Yaroshevsky 2006).

HM concentration (in mg/kg dw) of each CB sample and some descriptive statistical data of HMs analyzed in all CB samples are given in Table 1. The frequency distribution of the concentration of HMs is shown in Fig. 3. From Table 1, the average HM concentrations are ranked in descending order as follows: Fe > Ti > Mn > Cr > Sr > V > Ni > Zr > Zn > Cu > Co > Pb > As. Both from Fig. 3 and the results of the Shapiro-Wilk test, it can be proposed that the concentration distributions of V, Cr, Co, Ni, Zn, Sr, Zr, and Pb in all clay bricks exhibit a normal distribution ($p > 0.05$), while Ti, Mn, Fe, Cu, and As have a non-normal distribution ($p < 0.05$). According to the Bartlett test, most data did not present variance heterogeneity. Thus, a logarithmic transformation was applied to the data. The difference between HMs in the CB samples is given in Fig. 4. The result of the Kruskal-Wallis test showed that the concentration of at least one HM found in samples was different than others ($p < 2.2 \times 10^{-16}$). Wilcoxon rank-sum tests presented that the Co - Cu and V - Ni concentrations were similar in all CB samples ($p > 0.05$), while the rest of the HMs differed ($p < 0.05$).

The concentrations of Fe in the CB samples varied from 37510 (CB27) to 77420 (CB43) mg/kg with an average value of 52779 mg/kg, which is lower than the Earth's crust average of 46500 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Fe are 1.1 and 1.0, respectively. The average ER value shows that Fe in CB is 10% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Fe. The concentrations of Ti in the CB samples varied from 3639 (CB19) to 10220 (CB43) mg/kg with an average value of 5329 mg/kg. The average Ti concentration is higher than the Earth's crust average of 4500 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Ti are 0.9 and 1.0, respectively. The average EF value indicates deficiency to minimal enrichment of Ti. The concentrations of Mn in the CB samples varied from 498 (CB27) to 1438 (CB42) mg/kg with an average value of 736 mg/kg. The average Mn concentration is lower than the Earth's crust average of 100 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Mn are 0.9 and 0.6, respectively. The average EF value indicates deficiency to minimal enrichment of Mn. The concentrations of Cr in the CB samples varied from 244 (CB3) to 440 (CB42) mg/kg with an average value of 341 mg/kg. The average Cr concentration is 4 times higher than the Earth's crust average of 83 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Cr are 0.8 and 3.5, respectively. The average EF value indicates moderate enrichment of Cr. The concentrations of Sr in the CB samples varied from 149 (CB24) to 324 (CB12) mg/kg with an average value of 233 mg/kg, which is lower than the Earth's crust average of 340 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Sr are 1.2 and 0.6, respectively. The average ER value shows that Sr in CB is 20% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Sr. The concentrations of V in the CB samples varied from 141 (CB3) to 237 (CB18) mg/kg with an average value of 192 mg/kg, which is 2 times higher than the Earth's crust average of 90 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for V are 1.2 and 1.8, respectively. The average ER value shows that V in CB is 20% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of V. The concentrations of Ni in the CB samples varied from 129 (CB27) to 270 (CB42) mg/kg with an average value of 190 mg/kg, which is approximately 3 times higher than the Earth's crust average of 58 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Ni are 1.1 and 2.8, respectively. The average ER value shows that Ni in CB is 10% more enriched than in clay, while the average EF value indicates moderate enrichment of Ni. The concentrations of Zr in the CB samples varied from 75 (CB35) to 166 (CB43) mg/kg with an average value of 110 mg/kg, which is lower than the Earth's crust average of 170 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Zr are 1.1 and 0.6, respectively. The average ER value shows that Zr in CB is 10% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Zr. The concentrations of Zn in the CB samples varied from 59 (CB27) to 118 (CB43) mg/kg with an average value of 85 mg/kg, which is slightly higher than the Earth's crust average of 83 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Zn are 1.2 and 0.9, respectively. The average ER value shows that Zn in CB is 20% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Zn. The concentrations of Cu in the CB samples varied from 27 (CB5) to 82 (CB42) mg/kg with an average value of 44 mg/kg, which is slightly lower than the Earth's crust average of 47 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Cu are 1.2 and 0.8, respectively. The average ER value shows that Cu in CB is 20% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Cu. The concentrations of Co in the CB samples varied from 21 (CB5) to 59 (CB42) mg/kg with an average value of 39 mg/kg, which is approximately 2 times higher than the Earth's crust average of 18 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Co are 1.1 and 1.9, respectively. The average ER value shows that Co in CB is 10% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Co. The concentrations of Pb in the CB samples varied from 8 (CB41) to 20 (CB18) mg/kg with an average value of 14 mg/kg, which is slightly lower than the Earth's crust average of 16 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for Pb are 1.2 and 0.8, respectively. The average ER value shows that Pb in CB is 20% more enriched than in clay, while the average EF value indicates deficiency to minimal enrichment of Pb. The concentrations of As in the CB samples varied from 4 (CB45) to 15 (CB19) mg/kg with an average value of 8 mg/kg, which is approximately 5 times higher than the Earth's crust average of 1.7 mg/kg (Yaroshevsky 2006). The average values of ER and EF calculated for As are 1.4 and 3.8, respectively. The average ER value shows that As in CB is 40% more enriched than in clay, while the average EF value indicates moderate enrichment of As.

In addition, the correlation coefficients were calculated for HM concentrations in CB samples to investigate potential relationships. The results are presented in the correlogram given in Fig. 5. The correlogram shows some significant correlations between HM concentrations in CB samples. The correlation test reveals significant correlations between most HM concentrations in CB samples, with one remarkable exception of As concentration in CB samples. This could mean that As contamination may have different sources or causes of contamination compared to others, leading to weaker correlations.

Conclusion

In this study, the HMs (Fe, Ti, Mn, Cr, Sr, V, Ni, Zr, Zn, Cu, Co, Pb, and As) contents of the CB samples manufactured and utilized as a building material in Türkiye were studied for the first time in detail. As a result of the study, it was observed that (i) all HMs analyzed in CB samples, except Ti, Mn, and Cr, were naturally enriched according to the clay raw material used in CB production and (ii) Cr, As and Ni analyzed in CB samples were found to be at moderate levels compared to the Earth's crust average.

The HM concentration values in CBs obtained in this study will constitute a guiding information and database for the future, both for earthquake waste management and for regulations to be prepared regarding the production, transportation, and use of these materials. To eliminate situations that may threaten the health of workers, it must be mandatory to take necessary precautions, such as preventing workers from breathing brick dust.

Declarations

Availability of data and material Not applicable.

Ethical Approval

Not applicable.

Consent to Participate

Not applicable.

Consent to Publish

Not applicable.

Authors Contributions

Turhan, was leading the research project on assessment of heavy metal of clay brick samples. Turhan, Duran, Bakır, Hançerlioğulları, Kurnaz and Ünal collected the samples and made them ready for analysis. Turhan and Altuner performed the statistical analyses. Turhan evaluated the data and wrote the manuscript. All authors read and finalized the manuscript.

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Not applicable.

Competing Interests

The authors declare no competing interests.

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Table

Table 1 is available in the Supplementary Files section.

Figures

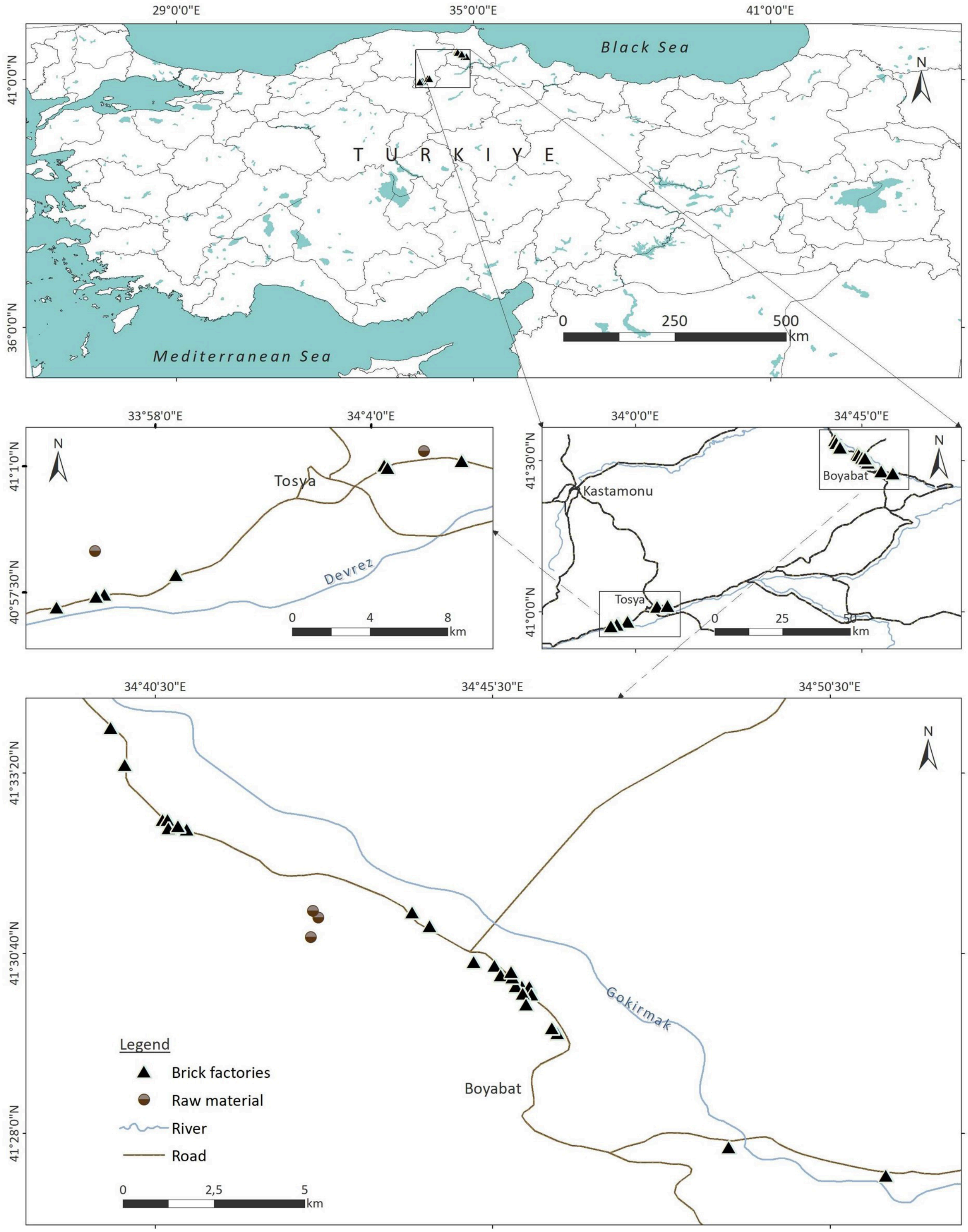

Figure 1

Geological map of locations of CB factories and clay quarry

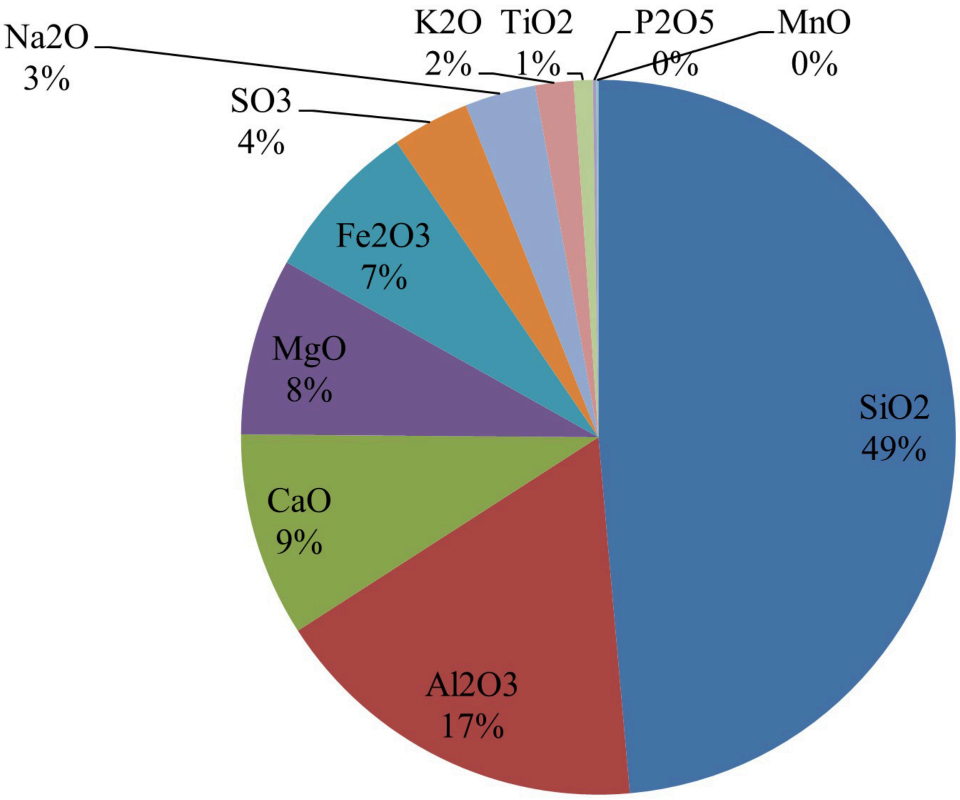

Figure 2

Major and mineral oxides of CB soil samples in percentage

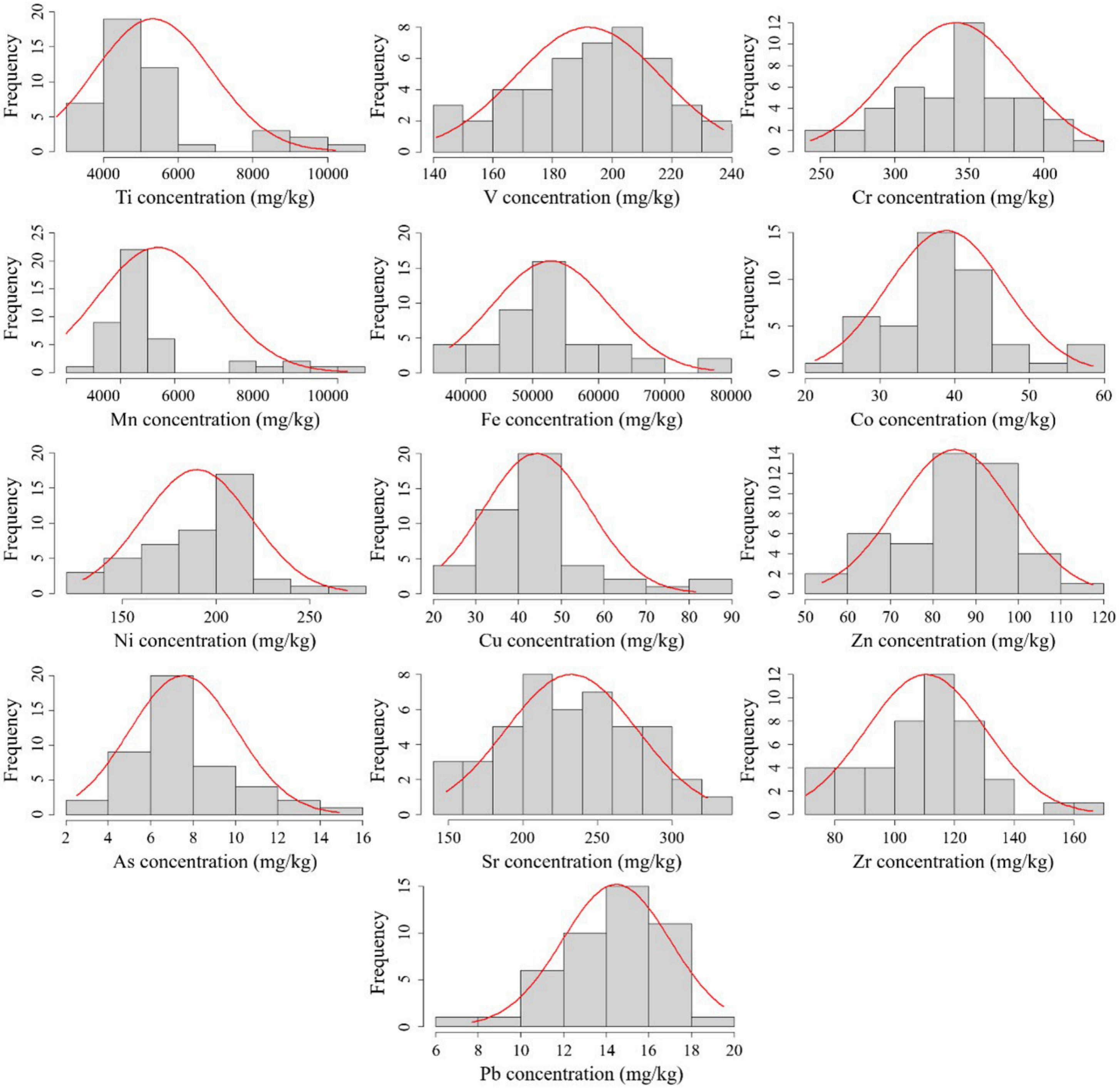

Figure 3

Frequency distributions of HM concentrations in CB samples

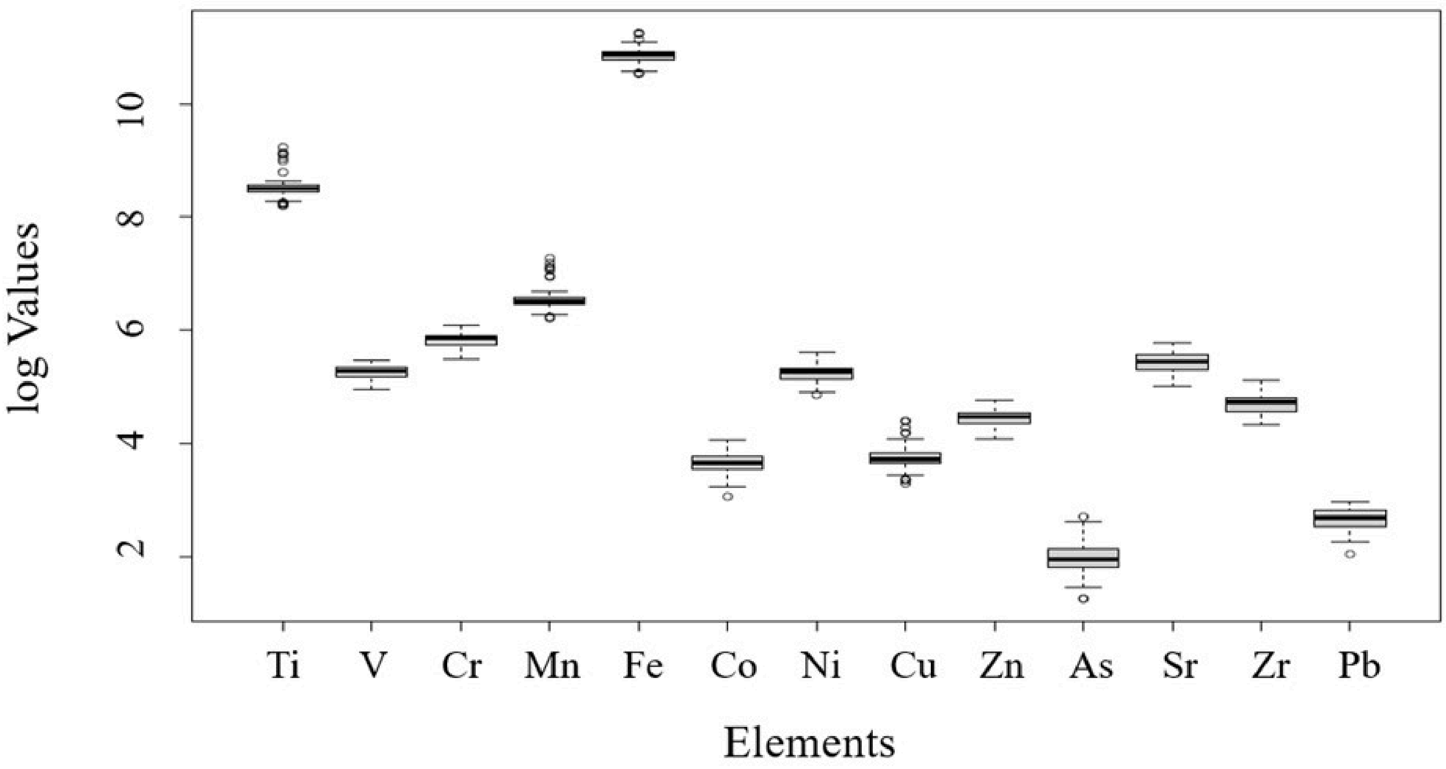

Figure 4

Boxplot for HMs in CB samples

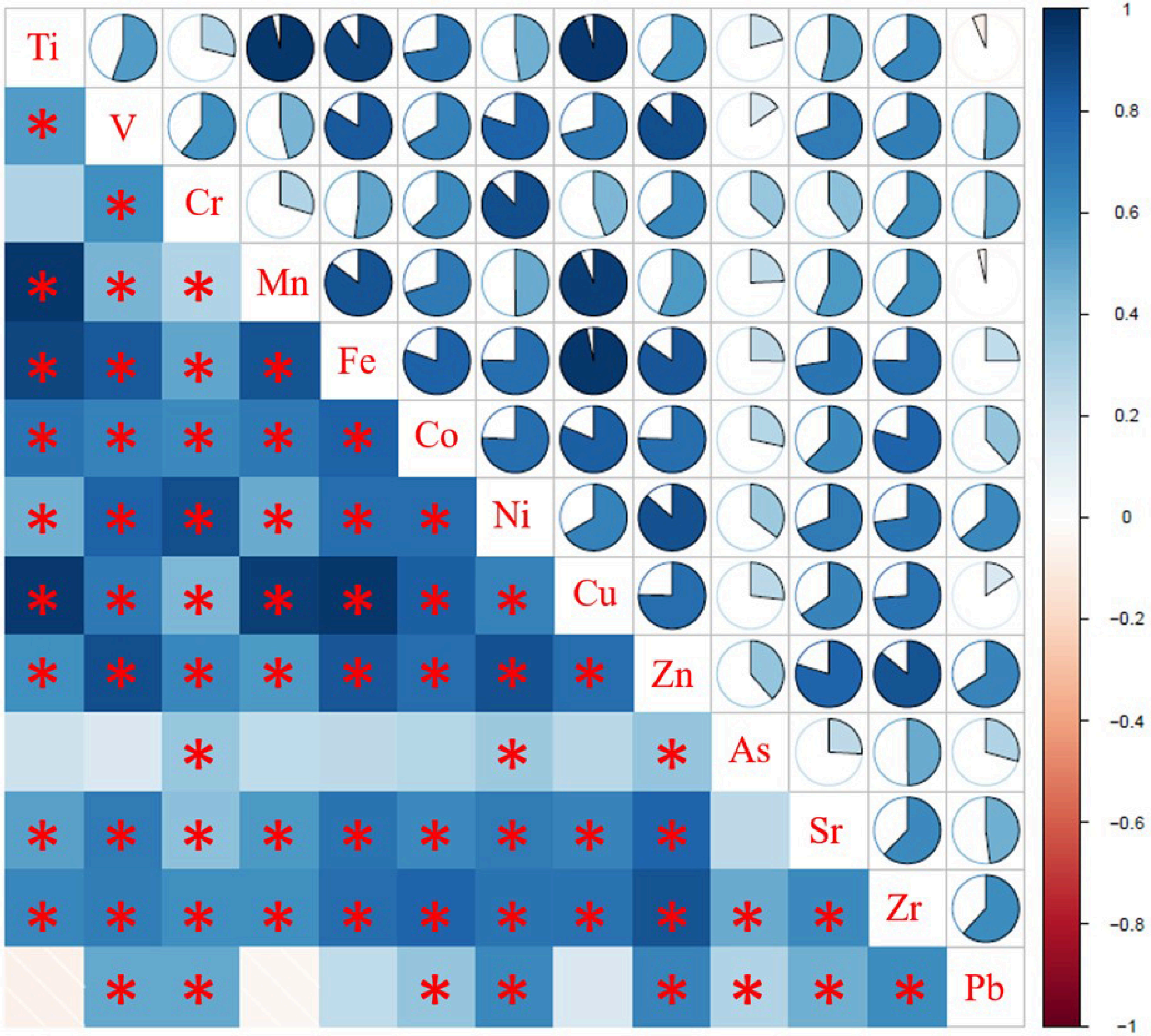

Figure 5

Correlogram plot on HMs in CB samples. * denotes a statistically significant correlation ($p < 0.05$)

Supplementary Files

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- TABLE.docx