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# Health Risks from Heavy Metal for Medical Toothpastes Derived from Herbal in Iraqi Pharmacies

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## ABSTRACT

Although toothpastes are considered as topical cosmetics that are not normally ingested, it is evident that they may contribute to the introduction of heavy metals through buccal and gastrointestinal absorption. The purpose of this study was to determine the heavy metals such as zinc, iron and lead in herbal toothpastes used in Iraq. Heavy metal analysis was conducted by using atomic absorption spectroscopy. Also, some carcinogens and non-carcinogens parameters (human health risk) due to zinc, iron and lead concentrations were calculated, such as chronic daily intake (CDI), total non-cancer risk (HI) and total cumulative cancer risk (TCCR). Results showed that the average value of zinc, iron and lead for all toothpastes were 44.55±37.72, 205.81±63.45 and 6.42±1.15 mg/kg, respectively. In the case of human health risk, the average values of HI and TTCR were 0.009±0.001 and 2.25±0.76 x$10^{-6}$, respectively. Zinc and lead for most samples in the present study were within the limits for WHO standards. While, iron concentrations in all toothpaste's samples fell outside the limits for WHO standards. HI and TTCR analysis revealed that toothpastes were safe from heavy metal related to health risk.

**Key words:** Heavy metals, toothpastes, atomic absorption spectroscopy, Iraqi pharmacies

## INTRODUCTION

Heavy metals are regarded as micropollutants since their excess presence has an adverse effect both on health and the environment. However, their occurrence in nature is not harmful to our environment as they are present only in very small amounts (Abojassim and Rua, 2020). They have gained significant importance due to their persistence, high toxicity and bioaccumulation properties. Heavy metals have been proved to have a negative impact on human health and their chronic toxicity to humans is associated with many chronic diseases like mental disorders, hypertension, pneumonia, gastrointestinal disorders, vascular disease, coronary heart disease, myocardial infarction, central nervous functions, kidney disorder and cancer (Mishra *et al.,* 2019). They can be exposed through direct and indirect sources like drinking water, food, air, soil (Rua and Abojassim, 2022; Oladoye *et al.,* 2022) and body care products (Swierczek *et al.*, 2019; Usman *et al.,* 2021; Mohammadzadeh *et al.*, 2022). Besides the reports on the evaluation of the possible contact and adverse health effects of the heavy metals from traditionally regulated sources, the exposures to metal toxins from toothpaste and mouthwashes have also gained importance in the recent years (Vella and Attard, 2019; Agbo, 2021). However, the investigation on toothpaste or oral care products remained comparatively ignored and needs more study especially in developing countries like Iraq. As far as we know, there are still no evaluation reports on the toothpaste and mouthwashes available in the markets of Iraq. Therefore, the aim of this study was to assess heavy metals (Zn, Fe and Pb) in different commercial kinds of toothpaste derived from of Herbal (plants) available in the local pharmacies in Iraq and the related potential health risk. As well as, it investigated the potential toxic metal hazards of toothpastes by comparing levels of Zn, Fe and Pb with permissible limits given by World Health Organization (WHO) and United States Environmental Protection Agency (USEPA). The daily intake of metals approach (carcinogens and non-carcinogen), total non-cancer risk and total cumulative cancer risk

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were adapted in the risk assessment of these toothpastes.

## METHODOLOGY

Ten toothpastes were selected from different Iraqi pharmacies in March 2022. These included herbal toothpastes. Although the samples were taken from pharmacy setting, these products were available at supermarkets, beauty shops and other outlets in various other countries. The samples were properly checked for their sample code, name of the manufacturer, type of herbal, origin, and manufacturing date (Table 1).

Ten grams of raw samples were oven-dried at 105°C until a constant weight was obtained on cooling in a desiccator. Toothpaste samples (1 g each) were digested according to the procedures (Das and Ting, 2017; Rua and Abojassim, 2022). Heavy metals in toothpaste were estimated with an atomic absorption spectrophotometer (AA-7000, Shimadzu, Japan) using flame-AAS method for Zn, Fe and Pb at wavelengths 213.9, 248.3 and 283.3 nm, respectively.

The experimental data were analyzed with SPSS software. Carcinogens and non-carcinogens parameters due to zinc, iron and lead concentrations such as chronic daily intake (CDI), hazard quotient (HQ), hazard index (HI), cancer risk (CR) and total cumulative cancer risk (TCCR) were calculated using following formulae (Orisakwe *et al.*, 2016; Fakhri *et al.*, 2018; Sharafi *et al.*, 2019; Nduka *et al.* 2020; Paul *et al.*, 2020).

$$CDI(\frac{mg}{kg}.d^{-1}) = \frac{Cs \times IR \times EF \times ED \times CF}{BW \times AT} \quad ...(1)$$

$$HQ = \frac{CDI_{nca}}{RFD_0} \quad ...(2)$$

$$HI = \sum_{1}^{K} \frac{CDI_k}{RFD_k} \quad ...(3)$$

$$CR = CDI_{ca} \times SF \quad ...(4)$$

$$TCCR = \sum_{1}^{k} CDI_k \times SF_k \quad ...(5)$$

Where, CS depicted exposure point concentration: mg/kg, IR was ingestion rate of toothpaste : 0.264 g/person/day (Paul *et al.*, 2020), EF was exposure frequency: 350 d/y, ED was exposure duration: 30 y (Orisakwe *et al.*, 2016 ), BW was body weight: 70 kg (Nduka *et al.* 2020), AT was averaging time for carcinogens 365 × 70 d for carcinogens, while for non-carcinogens 365 x EDd, CF was units conversion factor ($10^{-6}$ kg/mg) (Nduka *et al.*, 2020), RFDo was chronic reference dose of the toxicant (mg/kg/d); Zn = 0.3, Fe = 0.7 and Pb = 0.004, in unit mg/kg/d) (Fakhri *et al.*, 2018) and SF was slope factor (SF = 1/(6 × ED) mg/kg/d) (Nduka *et al.,* 2020).

## RESULTS AND DISCUSSION

Ten most medical toothpaste (derived from herbal or medical plants) samples widely used in Iraq and other countries from pharmacies were analyzed. The range value of zinc, iron and lead concentrations in unit mg/kg were 1.59-402.34, 36.75-654.24 and 1.00-12.05, respectively (Table 2). While, the averages with standard error (S. E.) concentrations of zinc, iron and lead were 44.55±37.72, 205.81±63.45 and 6.42±1.15 mg/kg, respectively. The maximum concentrations of zinc, iron and lead were found in samples $T_5$ (Crest, Chamomile plant, made in Germany), $T_8$ (Dabur, blackseed plant, made in UAE) and $T_4$ (Dabur, Olive plant, made in UAE), respectively, while the minimums concentrations were found in samples $T_6$ (Colgate, Seaweed plant, made in China), $T_{10}$ (Dabur, Aloe vera plant, made in UAE), respectively. The box plot of concentration spread out for zinc, iron and lead

**Table 1.** Toothpaste samples under study

| S. No. | Sample code | Name of manufacturer | Type of herbal plant | Origin | Manufacturing data |
|---|---|---|---|---|---|
| 1. | $T_1$ | Himalaya mint fresh | Fennel and clove | UAE | 2021 |
| 2. | $T_2$ | Dabur | Neem | UAE | 2020 |
| 3. | $T_3$ | Dabur | Basil | UAE | 2021 |
| 4. | $T_4$ | Dabur | Olive | UAE | 2021 |
| 5. | $T_5$ | Crest | Chamomile | Germany | 2021 |
| 6. | $T_6$ | Colgate | Seaweed | China | 2020 |
| 7. | $T_7$ | Colgate | Fleur delotedeasia | China | 2020 |
| 8. | $T_8$ | Dabur | Blackseed | UAE | 2021 |
| 9. | $T_9$ | Dabur | Salt and lemon | UAE | 2021 |
| 10. | $T_{10}$ | Dabur | Aloe vera | UAE | 2021 |

**Table 2.** Results of Zn, Fe and Pb in samples under study

| Sample code | Concentrations ppm) mg/kg) | | |
|---|---|---|---|
| | Zn | Fe | Pb |
| $T_1$ | 11.30 | 72.50 | 9.04 |
| $T_2$ | 5.63 | 189.80 | 11.05 |
| $T_3$ | 3.18 | 147.94 | 4.02 |
| $T_4$ | 2.72 | 151.68 | 12.05 |
| $T_5$ | 402.34 | 528.31 | 5.02 |
| $T_6$ | 1.59 | 63.67 | 1.00 |
| $T_7$ | 6.10 | 57.82 | 5.02 |
| $T_8$ | 5.90 | 654.24 | 2.01 |
| $T_9$ | 3.05 | 155.43 | 10.04 |
| $T_{10}$ | 3.78 | 36.75 | 5.02 |
| Average±S. E. | 44.55±37.72 | 205.81±63.45 | 6.42±1.15 |

in mg/kg is given in Fig. 1. The medians of the Zn and Fe were closer to the bottom of the box, and the distribution was skewed. The median was near the center of the Pb, this meant that the distribution of the data set was normally distributed and symmetrical. The percentage of the results of Zn concentrations found in toothpaste samples (Fig. 2) ranged from 0 to 2%, which were lower than standard values (10%). However, all samples were below the standard value according to WHO which equalled 50 mg/kg (WHO, 2005), except $T_5$ sample. The percentage of the results of Fe concentrations found in toothpaste samples (Fig. 3) ranged from 2 to 32%, which was higher than standard values (1%). However, all samples had higher standard values according to WHO which equalled 20 mg/kg (WHO, 2005). While, for Pb concentrations, the percentage results (Fig. 4) ranged from 1 to 16%, which with standard values (13%). However, three samples ($T_2$, $T_4$ and $T_9$) had higher values according to WHO which equalled 10 mg/kg (WHO, 2005).

The Pearson correlation indicated positive correlation among the Zn and Fe present in

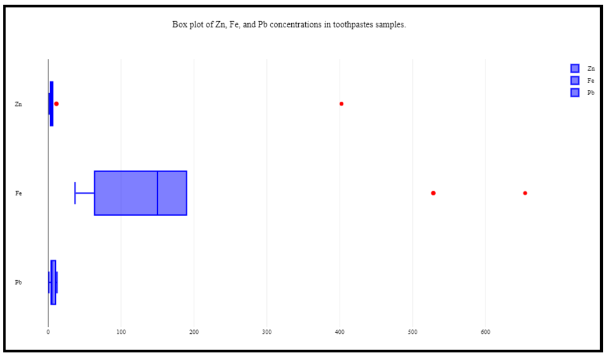


Fig. 1. The box plot of Zn, Fe and Pb concentrations in toothpastes samples.

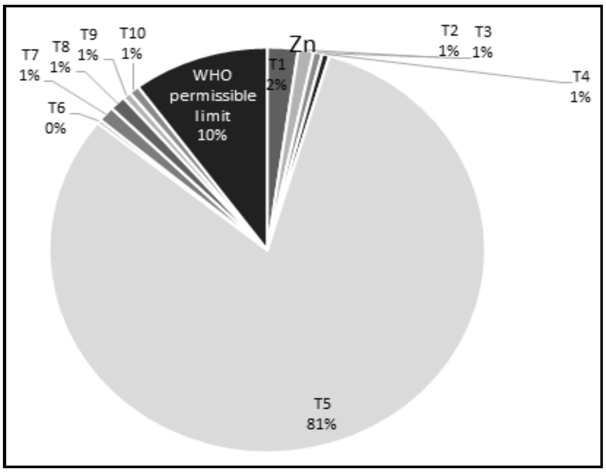


Fig. 2. The percentage of concentrations of Zn of the investigated 10 toothpaste samples in comparison with the WHO permissible limit.

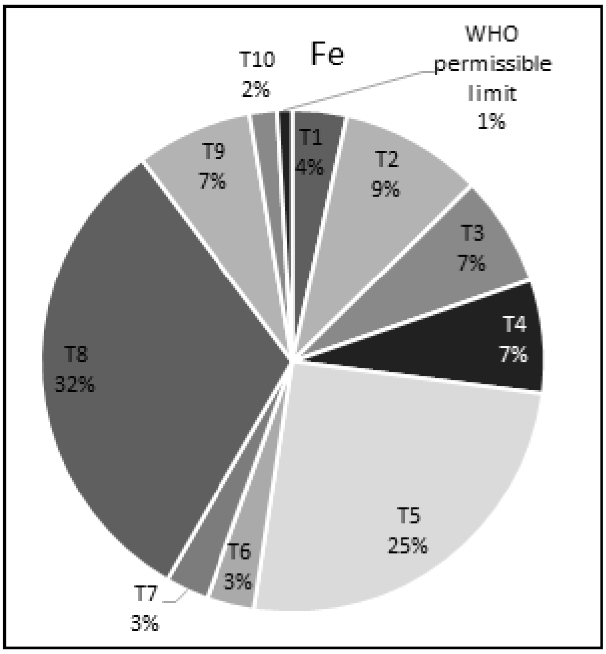


Fig. 3. The percentage of concentrations of Fe of the investigated 10 toothpaste samples in comparison with the WHO permissible limit.

the toothpaste samples, with r value as 0.5371, while the Pearson correlation indicated negative correlation for Zn with Pb and Fe with Pb which equalled -0.124 and -0.2709, respectively. Statistically, it was found that, the P-value among the metals present in the toothpaste samples was not significant at $P < 0.05$. The average values of three heavy metals in the present study (Fig. 5) were in order as: Fe > Zn > Pb.

From the results of heavy metals (Table 2), all the kinds of toothpaste contained different levels of Zn, Fe and Pb. Heavy metals were

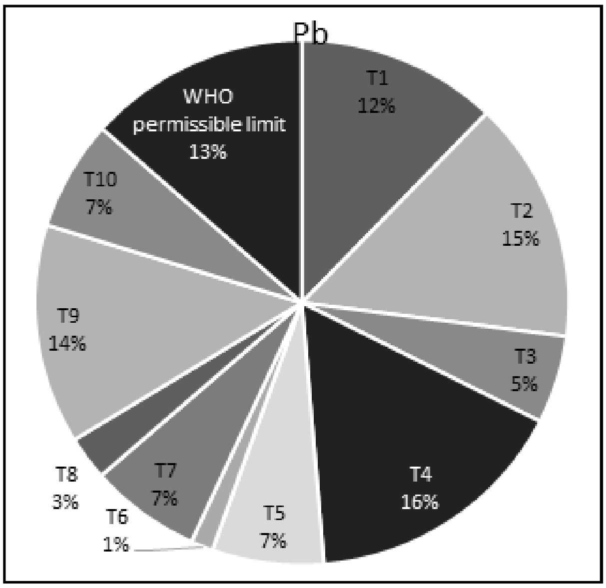


Fig. 4. The percentage of concentrations of Pb of the investigated 10 toothpaste samples in comparison with the WHO permissible limit.

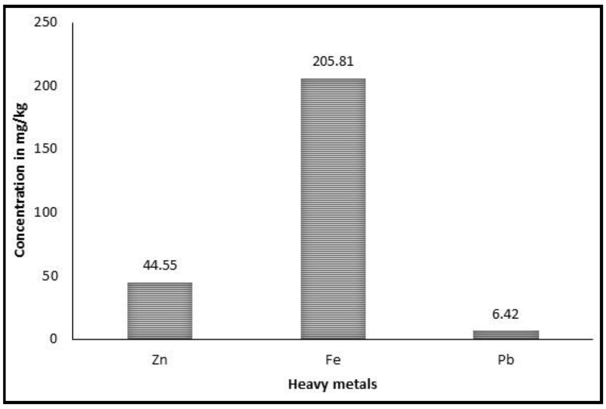


Fig. 5. The comparison of the average values of concentrations for Zn, Fe and Pb the investigated 10 toothpaste samples.

considered as mere contaminants for toothpaste as their roles were not clearly defined in the toothpaste formulation. However, their presence in the toothpaste may be accounted for abrasives, materials used from the plant sources (Paul *et al.*, 2020), accidental cross-contamination during processing, and the deliberate introduction of metals as therapeutic ingredients for more efficacy (Paul *et al.*, 2020). Consequently, these metals may become harmful when they are ingested above the tolerance level, and daily use may have a significant adverse health effect.

The results of the human health for non-cancer risk parameters to the Zn, Fe and Pb concentrations in herbal toothpastes that used in Iraq are shown in Table 3. The results showed that the average values with stander error for chronic daily intake non-carcinogens ($CDI_{nca}$) for Zn, Fe and Pb concentrations were (0.161±0.13) $\times 10^{-3}$, (0.744±0.22) $\times 10^{-3}$ and (0.023±0.01) $\times 10^{-3}$ (mg/kg/d), respectively. Also, it was found the average values of hazard quotient (HQ) $\times$ $10^{-3}$ for Zn, Fe and Pb concentrations were 0.38±0.32, 2.48±0.76 and 5.81±1.04, respectively. Thus, the results of hazard index (HI) in the present study ranged from 0.002 to 0.013, with an average value of 0.009±0.001. The results of the human health for cancer risk parameters to the Zn, Fe and Pb concentrations in herbal toothpastes that used in Iraq are shown in Table 4. The results showed that the average values with standaerd error for chronic daily intake carcinogens ($CDI_{ca}$) for Zn, Fe and Pb concentrations were (0.069±0.05) $\times$ $10^{-3}$, (0.319±0.09) $\times$ $10^{-3}$ and (0.010±0.001) $\times$ $10^{-3}$ (mg/kg/d), respectively. Also, it was found that the average values of cancer risk (CR) $\times$ $10^{-3}$ for Zn, Fe and Pb concentrations were 0.38±0.32, 1.77±0.54, and 0.055±0.01, respectively. The results of total cumulative cancer risk (TCCR) $\times$ $10^{-6}$ in the

**Table 3.** Results of $CDI_{nca}$, HQ and Hi for non-cancer risk

| S. No. | Sample code | CDInca in mg/kg/d × $10^{-3}$ | | | HQ × $10^{-3}$ | | | HI |
|---|---|---|---|---|---|---|---|---|
| | | Zn | Fe | Pb | Zn | Fe | Pb | |
| 1. | $T_1$ | 0.041 | 0.262 | 0.033 | 0.097 | 0.87 | 8.17 | 0.009 |
| 2. | $T_2$ | 0.020 | 0.686 | 0.040 | 0.049 | 2.29 | 9.99 | 0.012 |
| 3. | $T_3$ | 0.012 | 0.535 | 0.015 | 0.027 | 1.78 | 3.63 | 0.005 |
| 4. | $T_4$ | 0.010 | 0.549 | 0.044 | 0.023 | 1.83 | 10.89 | 0.013 |
| 5. | $T_5$ | 1.455 | 1.911 | 0.018 | 3.464 | 6.37 | 4.54 | 0.013 |
| 6. | $T_6$ | 0.006 | 0.230 | 0.004 | 0.014 | 0.77 | 0.91 | 0.002 |
| 7. | $T_7$ | 0.022 | 0.209 | 0.018 | 0.052 | 0.70 | 4.54 | 0.005 |
| 8. | $T_8$ | 0.021 | 2.366 | 0.007 | 0.051 | 7.89 | 1.82 | 0.010 |
| 9. | $T_9$ | 0.011 | 0.562 | 0.036 | 0.026 | 1.87 | 9.08 | 0.011 |
| 10. | $T_{10}$ | 0.014 | 0.133 | 0.018 | 0.02 | 0.44 | 4.54 | 0.005 |
| Average±S. E. | | 0.161±0.13 | 0.744±0.22 | 0.023±0.01 | 0.38±0.32 | 2.48±0.76 | 5.81±1.04 | 0.009±0.001 |

**Table 4.** Results of $CDI_{ca}$, CR and TTCR for cancer risk

| S. No. | Sample code | $CDI_{ca}$ in mg/kg/d × $10^{-3}$ | | | CR × $10^{-6}$ | | | TTCR × $10^{-6}$ |
|---|---|---|---|---|---|---|---|---|
| | | Zn | Fe | Pb | Zn | Fe | Pb | |
| 1. | $T_1$ | 0.018 | 0.112 | 0.014 | 0.10 | 0.62 | 0.08 | 0.799 |
| 2. | $T_2$ | 0.009 | 0.294 | 0.017 | 0.05 | 1.63 | 0.10 | 1.778 |
| 3. | $T_3$ | 0.005 | 0.229 | 0.006 | 0.03 | 1.27 | 0.03 | 1.336 |
| 4. | $T_4$ | 0.004 | 0.235 | 0.019 | 0.02 | 1.31 | 0.10 | 1.433 |
| 5. | $T_5$ | 0.624 | 0.819 | 0.008 | 3.46 | 4.55 | 0.04 | 8.057 |
| 6. | $T_6$ | 0.002 | 0.099 | 0.002 | 0.01 | 0.55 | 0.01 | 0.571 |
| 7. | $T_7$ | 0.009 | 0.090 | 0.008 | 0.05 | 0.50 | 0.04 | 0.594 |
| 8. | $T_8$ | 0.009 | 1.014 | 0.003 | 0.05 | 5.63 | 0.02 | 5.701 |
| 9. | $T_9$ | 0.005 | 0.241 | 0.016 | 0.03 | 1.34 | 0.09 | 1.451 |
| 10. | $T_{10}$ | 0.006 | 0.057 | 0.008 | 0.03 | 0.32 | 0.04 | 0.392 |
| Average± S.E. | | 0.069±0.05 | 0.319±0.09 | 0.010±0.001 | 0.38±0.32 | 1.77±0.54 | 0.055±0.01 | 2.21±0.76 |

present study ranged from 0.571 to 8.057, with an average value of 2.21±0.76. The global of standard values for chronic daily intake non-carcinogens ($CDI_{nca}$) and carcinogens ($CDI_{ca}$) due to Zn, Fe and Pb for USEPA (USEPA, 2002) were equal 0.3, 0.7, and 0.004 mg/kg/d, respectively (Fig. 6). Therefore, the values of $CDI_{nca}$ and $CDI_{nca}$ for all heavy metals as well as all samples of the present study were lower than the global limit. Also, it was found the values of hazard quotient (HQ) in all 10 toothpaste samples in the present study were lower than the global limit (1) according to USEPA (USEPA, 2002). The range of the global value of standard values for cancer risk (CR), and total cumulative cancer risk (TCCR) of carcinogens due to Zn, Fe and Pb for USEPA (USEPA, 2002) was equal $10^{-4}$ to $10^{-6}$. Therefore, it was found the values of CR and TCCR in all 10 toothpaste samples in the present study were within the global range limit. Hence, the value of the results of human health risk due to zinc, iron and lead elements was little; therefore, the risk of non-cancer and cancer was negligible.

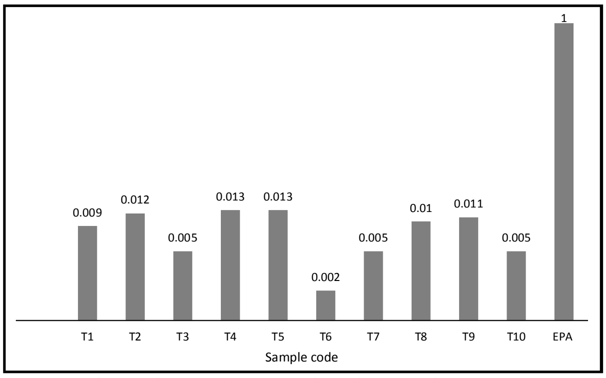


Fig. 6. The values of Hazard Index of the investigated 10 toothpaste samples in comparison with the EPA permissible limit.

## CONCLUSION

The results of Zn and Pb concentrations in most samples were lower than global limit according to WHO. Contrarily, the results of Zn concentrations in all samples were higher according to WHO. Carcinogens and non-carcinogens parameters due to zinc, iron and lead concentrations in all samples of the present study were within the acceptance of the permissible limit according to USEPA. Statistically, low and non-significant correlation coefficients were found between three heavy metals. As a result, 10 medical toothpaste (derived from herbal or medical plants) samples widely used in Iraq and other countries pharmacies did not constitute a health hazard.

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