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CONTENT OF HEAVY METALS, MACRO- AND MICROELEMENTS IN APPLE FRUITS

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Abstract

The research evaluated the content of heavy metals, micro, and macronutrients in fruits of eight apple varieties - Red Del. Superchief, Fujion pbr, Enterprise, Pinova, Orion, Red Del. Mestar, Gemini and Crimson crisp, grown in orchard near Brestnik village, situated at 3.5 km distance from the potential source of pollution (the Holding KCM 2000) near Plovdiv, Bulgaria. Significant varietal differences in the composition of the apple fruit were observed. The highest content of Pb, Cd, Zn, Cu, Fe, Mn, P, Mg and K were found in the variety Red Del. Superchief. Variety Pinova contained Ca and Hg, the variety Orion - Cr and Ni, and the variety Gemini - B, Na and As. However, the content of Cd, Zn, As, Cr and Ni in the fruit of all tested varieties was below the maximum permissible level. The Pb content in the apples of the varieties Enterprise, Pinova and Red Del. Mestar was low and did not exceed the maximum permissible level for heavy metals in food, thus these apple varieties can be included in the daily human diet without any restrictions.

Keywords: apples, cultivars, nutrients, toxic elements, fruits

INTRODUCTION

Apple (*Malus domestica*) in one of the most consumed fruits worldwide due to its organoleptic characteristics (aroma and taste) and high nutritional value. Apples contain sugars, dietary fiber, pectin, vitamins (C and A), organic acids and various biologically active compounds (Wu et al., 2007). Chemical composition and nutritional value of apples depend primarily on the variety, soil and climatic conditions, growing technology and postharvest storage (Cadar et al., 2014). The chemical composition of apples affects their quality and human health.

Heavy metals are among the most hazardous substances present in the environment because their presence in soils results in the contamination of plants and, consequently, food. Consumption of contaminated food (fruits and vegetables) is the primary cause of toxic metals transfer into the human body (Rai et al., 2019). Cadmium (Cd), lead (Pb), and mercury (Hg) are among the most toxic heavy metals causing severe health problems in humans (Tchounwou et al., 2012).

In recent years, the number of studies related to research on the safety of food and food products contaminated with pesticides, heavy metals or toxins has increased significantly (Arnold & Gramza-Michalowska, 2023). Plants grown in contaminated areas have a higher concentration of heavy metals than those grown in an uncontaminated environment (Imeri et al., 2019). The uptake and accumulation of heavy metals in plants depends on a number of factors such as soil pH, concentration of heavy metals in the soil, aerosol contamination, agrometeorological conditions, distance from the source of contamination, exposure time (Kabata-Pendias, 2010). However, in the literature the information about the influence of cultivar on the production of orchard species in areas with industrial pollution is scarce.

The objective of this study was to determine the content of heavy metals, macroand microelements in fruits of eight apple varieties (*Malus domestica*) cultivated on contaminated with heavy metals soil.

MATERIALS AND METHODS

The study was carried out in a nursery orchard of the Experimental field of the Agricultural University, Plovdiv, located in the village of Brestnik, 3.5 km away from the source of heavy metal contamination (the Holding KCM 2000, Plovdiv). The soil was characterized by a low alkaline reaction (pH 7.6), an average organic carbon content (2.24%) and an average content of nitrogen - 0.22% N, phosphorus - 387.3 $mg.kg^{-1}$, and potassium - 6780 $mg.kg^{-1}$. Lead content in soil reached 83.1 $mg.kg^{-1}$, Cd - 4.0 $mg.kg^{-1}$, Zn - 215.3 $mg.kg^{-1}$, Cr -107.2 mg/kg^{-1}, Ni -112.9 $mg.kg^{-1}$, Hg - 0.5 $mg.kg^{-1}$, Cu - 71.3 $mg.kg^{-1}$, Fe - 29581.9 $mg.kg^{-1}$, Mn - 884.2 $mg.kg^{-1}$, Ca - 18065 $mg.kg^{-1}$, Mg - 10040.1 $mg.kg^{-1}$. Among the analyzed heavy metals, only the content of Cd (4.0 $mg.kg^{-1}$) exceeded the maximum permissible level of 3.0 $mg.kg^{-1}$(Angelova et al., 2020).

The apple trees were planted in 2020 in a 4x2 m scheme or at a density of 125 trees per acre with a retaining structure and drip irrigation. New, for the country, were varieties Gemini, Red Del. Superchief, Orion, Enterprise, Pinova, Crimson crips, Fujion prb, which are resistant to economically important diseases. Trees from the variety Red Del. Mestar were used as a control. The varietal rootstock in all cultivars was MM106.

Fruits from the studied cultivars were taken for analysis at different time. For early cultivars such as Gemini, Red Del. Superchief, Red Del. Mestar fruits were collected in mid-August and early September. For mid-early cultivars such as Orion, Enterprise and Pinova the fruits were collected in mid and late September. For late cultivars – Fujion prb and Crimson crips – fruits were collected in mid-October. The fruits were analyzed in their fresh condition for the content of macroelements, microelements, and heavy metals using the method of microwave mineralization. The quantitative measures were carried out by ICP method (Jobin Yvon Emission - JY 38 S, France). Digestion and analytical efficiency of ICP was validated using a standard reference material of apple leaves (SRM 1515, National Institute of Standards and Technology, NIST). SPSS was used for data statistical processing.

RESULTS AND DISCUSSION

The content of macroelements, microelements and heavy metals in the fruits of the eight apple cultivars is presented in Fig. 1. Additional information such as minimum, maximum and average values are presented in Table 1. The results showed that in the apple fruits the macroelements K, P, Mg, Na, Ca predominated, and were followed by B, Fe, Zn, Mn and Cu content. The apple fruits also contained traces of the toxic metals such as Pb, Cd, Hg, As, Ni and Cr. The content of toxic metals in most foods was regulated by Commission Regulation (EC) No 1881/2006, which was repealed by Commission Regulation (EC) No 2023/915. According to the Regulation, the concentration of Pb and Cd in fruit is limited to 0.10 and 0.02 $mg.kg^{-1}$ fresh weight, respectively (European Commission, 2023).

The average content of Pb in apple fruit varies from 0.057 $mg.kg^{-1}$ in the Pinova variety to 0.210 $mg.kg^{-1}$ in the Red Del. Superchief variety (average content 0.116 $mg.kg^{-1}$). The Pb levels in the fruit of the apple cultivars Red Del. Superchief, Fujion pbr, Orion, Gemini and Crimson crisp exceeded the maximum permissible level (0.1 $mg.kg^{-1}$) (European Commission, 2023). The content of Pb in apples of the varieties Enterprise, Pinova and Red Del. Mestar was low and did not exceed the maximum permissible level for heavy metals in food, thus these apple varieties can be included in the daily diet without any restrictions.

A higher content of heavy metals was usually determined in apples from orchards located near industrial plants. (Bednarek et al., 2007). High levels of Pb were also found in apples in Romania (0.413-1.149 $mg.kg^{-1}$) (Prundeanu et al., 2020). Zhen (2008) reported that apple fruits planted near highways were contaminated with Pb and Cd – 0.082 and 0.010 mg/kg, respectively).

The average Cd content in the apple fruits reached up to 0.002 $mg.kg^{-1}$. In the variety Gemini, Cd was below the detection limit of the method. The Cd content in the apple fruit of all tested varieties was below the maximum permissible level (0.02 $mg.kg^{-1}$) (European Commission, 2023).

According to Kabata-Pendias (2010), the Cd content in apples was relatively low and ranged from 0.003 to 0.03 $mg.kg^{-1}$. Analysis of Cd content in apples from Poland (0.002-0.0057 $mg.kg^{-1}$) (Campeanu et al., 2009; Bednarek et al., 2007) and Portugal (0.0143 $mg.kg^{-1}$) (Waheed et al., 2004) was also low. Values of Cd content ranging from 0.025 to 0.127 $mg.kg^{-1}$ was detected in apples from Romania (Prundeanu et al., 2020) and up to 0.16 $mg.kg^{-1}$in Turkey (Hamurcu et al., 2010).

In the current study, the average Hg content of the fruit was low - 1.720 $\mu g.kg^{-1}$. Similar results were obtained for apples in Poland (Bednarek et al., 2007).

The average Ni content in apple fruits reached up to 0.042 $mg.kg^{-1}$. Higher values were found in the cultivar Orion (0.188 $mg.kg^{-1}$), the values were significantly lower than the maximum permissible for fruit (0.5 $mg.kg^{-1}$) (Regulation №31 of 29 July 2004). According to Kabata-Pendias (2010), the Ni content in apples is low (0.003-0.08 $mg.kg^{-1}$). Higher values were found in Poland (0.182 $mg.kg^{-1}$) (Campeanu et al., 2009), Romania (0.054-0.257 $mg.kg^{-1}$) (Prundeanu et al., 2020), Serbia (0.444 $mg.kg^{-1}$) (Mitic et al., 2007) and Pakistan (1.0-8.9 $mg.kg^{-1}$) (Zahoor et al., 2003).

The average Cr content in apple fruit reaches 0.032 $mg.kg^{-1}$. Higher values were found in the variety Orion (0.057 $mg.kg^{-1}$), with values significantly lower than the maximum permissible for fruit (0.1 $mg.kg^{-1}$). The Cr content of apple fruit is known to be low (0.013 $mg.kg^{-1}$) (Kabata-Pendias, 2010). In Romania, values in the range of 0.05-0.101 $mg.kg^{-1}$ (Prundeanu et al., 2020), 0.24 $mg.kg^{-1}$ in Turkey (Hamurcu et al. 2010), and 1.48-6.43 $mg.kg^{-1}$ in Pakistan (Zahoor et al., 2003) have been reported.

The As content of apple fruit ranges from 0.021 to 0.057 $mg.kg^{-1}$ (average 0.040 $mg.kg^{-1}$) and is lower than the tolerance level for fruit (0.5 $mg.kg^{-1}$) (Regulation №31 of 29 July 2004).

The trace element content of apple fruit decreases in the following order: B> Fe> Zn> Mn> Cu (Table 1).

The average content of Zn in fruits reached 0.673 $mg.kg^{-1}$, values significantly lower than the permissible level for fruits (10 $mg.kg^{-1}$) (Commission Regulation (EC) No 2023/915). The order of Zn accumulation was Red Del. Superchief > Orion > Crimson crisp > Fujion pbr > Enterprise > Red Delicious > Pinova. Studies have shown that Zn content in apples can vary considerably - 0.271 $mg.kg^{-1}$ in Portugal (Waheed et al., 2004), 0.181-0.453 $mg.kg^{-1}$ in Poland (Campeanu et al., 2009), 0.4 $mg.kg^{-1}$ in USDA National Nutrient Database (USDA, 2009), 0.909-4.458 $mg.kg^{-1}$ in Romania (Prundeanu et al. 2020) and 1.185 $mg.kg^{-1}$ in Hungary (Hegedus et al., 2008).

The Cu content of the fruits ranged from 0.338 $mg.kg^{-1}$ in the variety Orion to 0.972 $mg.kg^{-1}$ in the variety Red Del. The values estimated in Superchief variety (average 0.548 $mg.kg^{-1}$) were significantly lower than the permissible level for fruit (5.0 $mg.kg^{-1}$) (Regulation №31 of 29 July 2004).

Similar values were found in apples from Poland - 0.414 $mg.kg^{-1}$ (Krejpcio et al., 2005), Romania - 0.409 $mg.kg^{-1}$ (Prundeanu et al., 2020), Serbia - 0.869 $mg.kg^{-1}$ (Mitic et al., 2013) and Hungary -1.1 $mg.kg^{-1}$ (Hegedus et al., 2008).

Table 1. Content of heavy metals, macroelement and microelements (mg.kg^{-1}) in apple fruit

	Min x ± sd	Max x±sd	Average x±sd
Pb	0.057±0.01	0.210±0.14	0.119±0.11
Cd	nd	0.004±0.001	0.002±0.001
Zn	0.569±0.052	0.944±0.103	0.673±0.094
Cu	0.338±0.050	0.972±0.151	0.548±0.106
Fe	1.604±0.152	4.954±0.286	2.669±0.206
Mn	0.353±0.082	0.694±0.156	0.516±0.105
P	124.9±6.8	226.5±10.1	162.4±8.4
Ca	36.2±1.2	64.0±2.1	47.6±1,6
Mg	57.2±1.4	110.3±2.8	72.3±1.6
K	780.9±8.7	1156.3±10.4	1017.5±10.1
Na	39.9±1.0	88.1±2.4	57.1±1.4
B	1.624±0.182	4.923±0.309	2.749±0.245
Cr	nd	0.057±0.014	0.012±0.015
Ni	0.002±0.001	0.188±0.028	0.042±0.013
As	0.021±0.010	0.057±0.015	0.040±0.012
Hg*	0.910±0.102	2.514±0.196	1.720±0.144

Legend: *- µg.kg^{-1}, nd- not detectable,
x- average value(mg.kg^{-1}) from 5 repetitions;
sd - mean standard deviation

The average Fe content reaches up to 2.669 mg.kg^{-1} and Mn up to 0.516 mg.kg^{-1}. The distribution of elements in the studied apple varieties was specific for each element and follows the order: Red Del. Superchief> Pinova> Orion> Crimson crisp> Enterprise> Red Delicious> Gemini> Fujion, for Mn: Red Del. Superchief> Pinova> Red Delicious> Crimson crisp> Orion> Gemini> Fujion> Enterprise. Studies have shown that the Fe content of apples varies widely - 0.4 mg.kg^{-1} in Canada (Rupasinghe & Clegg, 2007), 0.94 mg.kg^{-1} in Spain (Gorinstein et al., 2001), 1.2 mg.kg^{-1} in the USDA National Nutrient Database (USDA, 2009), 1.3 mg.kg^{-1} in Hungary (Hegedus et al., 2008), 2.926 mg.kg^{-1} in Romania (Prundeanu et al., 2020) and 40.89 mg.kg^{-1} in Serbia (Mitic et al., 2013).

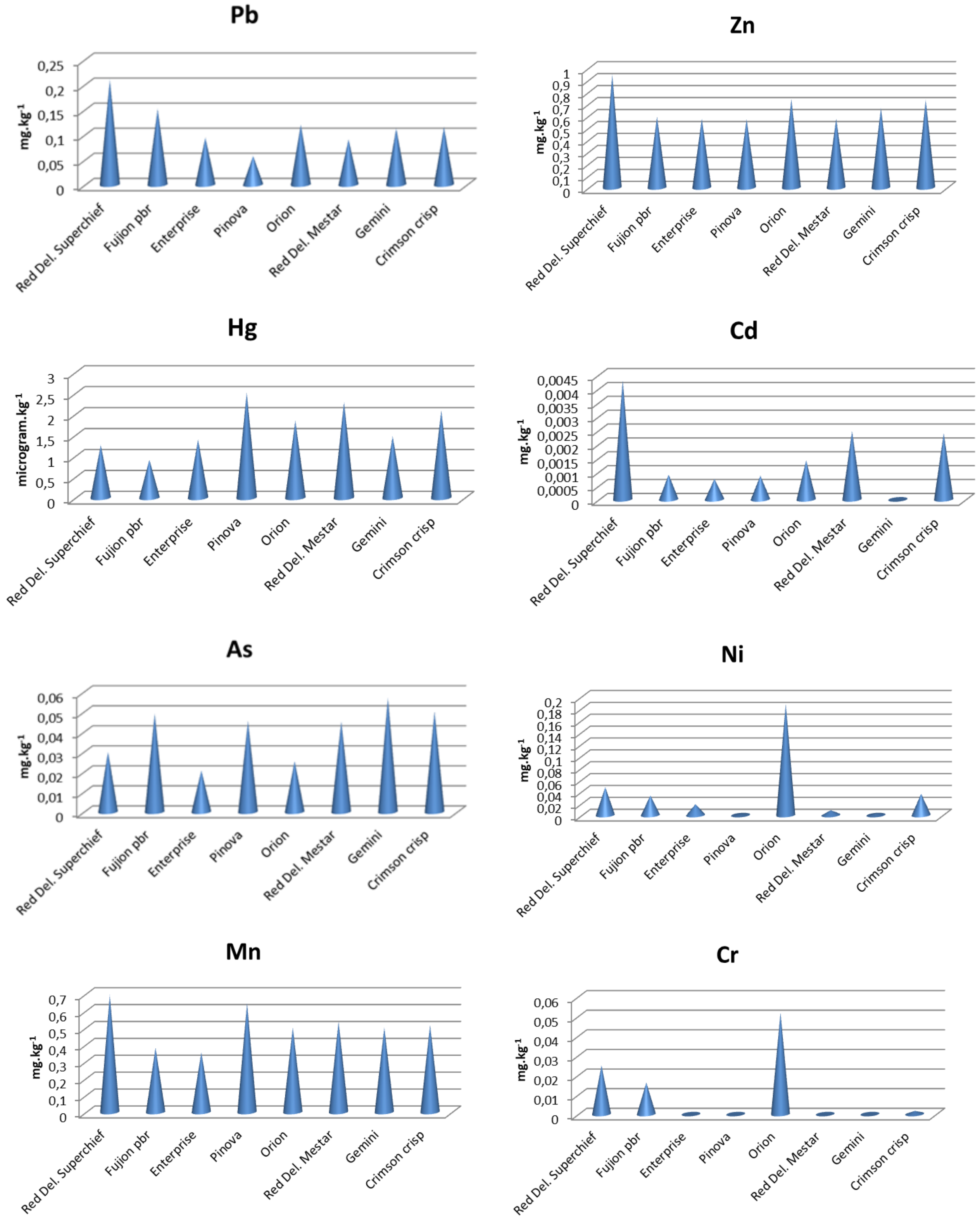
Mn content ranges from 0.228 to 0.895 mg.kg^{-1} in Romania (Prundeanu et al., 2020), 0.3 mg.kg^{-1} in Spain (Gorinstein et al., 2001), 0.307 mg.kg^{-1} in Poland (Bednarek et al., 2007), 0.4 mg.kg^{-1} in USDA National Nutrient Database (USDA, 2009), 0.527 mg.kg^{-1} in Serbia (Mitic et al., 2013), 0.9 mg.kg^{-1} in Hungary (Hegedus et al., 2008)

Studies have shown that apple fruits contain minerals such as K, Mg, Ca, Na and P and trace elements such as Zn, Mn, Cu and Fe (Jemaneh & Chandravanshi, 2021). However, the mineral content varies with cultivars, production technology and growing region (Sachini et al., 2020). Most of the mineral content in apples is low, including Fe, Mg, Mn, Zn and Cu.

The content of macroelements in apple fruits decreased in the following order: K> P> Ca> Mg (Table 1) and significant varietal differences in apple fruit composition were observed (Fig. 1). The highest contents of Pb, Cd, Zn, Cu, Fe, Mn, P, Mg and K were found in Red Del. Superchief, Ca and Hg in Pinova, Cr and Ni in Orion, and B, Na and As in Gemini.

Potassium is the dominant element among the macroelements in the fruit (average K content - up to 1017.5 mg.kg^{-1}), The order of K distribution was Red Del. Superchief > Fujion pbr > Red Delicious > Enterprise > Crimson crisp > Gemini > Pinova > Orion. Sachini et al. (2020) reported that K was predominant in apples from southern Brazil, followed by P and Mg, while Ca was in much smaller amount. The obtained in the current study results were similar since the content of P reached up to 162.4 mg.kg^{-1}, Mg - up to 72.3 mg.kg^{-1} and Ca - up to 47.6 mg.kg^{-1}.

The distribution of nutrients in the studied apple varieties was specific for each element and can be presented in the following order: for P: Red Del. Superchief > Red Del. Mestar > Enterprise> Fujion pbr>Crimson crisp> Pinova> Orion > Gemini, for Ca: Pinova> Red Del. Superchief > Fujion > Crimson crisp> Enterprise> Gemini > Red Del. Mestar > Orion; for Mg: Red Del. Superchief> Red Del. Mestar> Enterprise> Pinova>Crimson crisp> Fujion pbr> Orion; and for K: Red Del. Superchief> Fujion pbr> Red Del. Mestar> Enterprise> Crimson crisp> Gemini> Pinova> Orion.


Pb
mg.kg-1
0,25
0,2
0,15
0,1
0,05
0
Zn
mg.kg-1
1
0,9
0,8
0,7
0,6
0,5
0,4
0,3
0,2
0,1
0
Hg
microgram.kg-1
3
2,5
2
1,5
1
0,5
0
Cd
mg.kg-1
0,0045
0,004
0,0035
0,003
0,0025
0,002
0,0015
0,001
0,0005
0
As
mg.kg-1
0,06
0,05
0,04
0,03
0,02
0,01
0
Ni
mg.kg-1
0,2
0,18
0,16
0,14
0,12
0,1
0,08
0,06
0,04
0,02
0
Mn
mg.kg-1
0,7
0,6
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0,4
0,3
0,2
0,1
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Cr
mg.kg-1
0,06
0,05
0,04
0,03
0,02
0,01
0
Red Del. Superchief
Fujion pbr
Enterprise
Pinova
Orion
Red Del. Mestar
Gemini
Crimson crisp

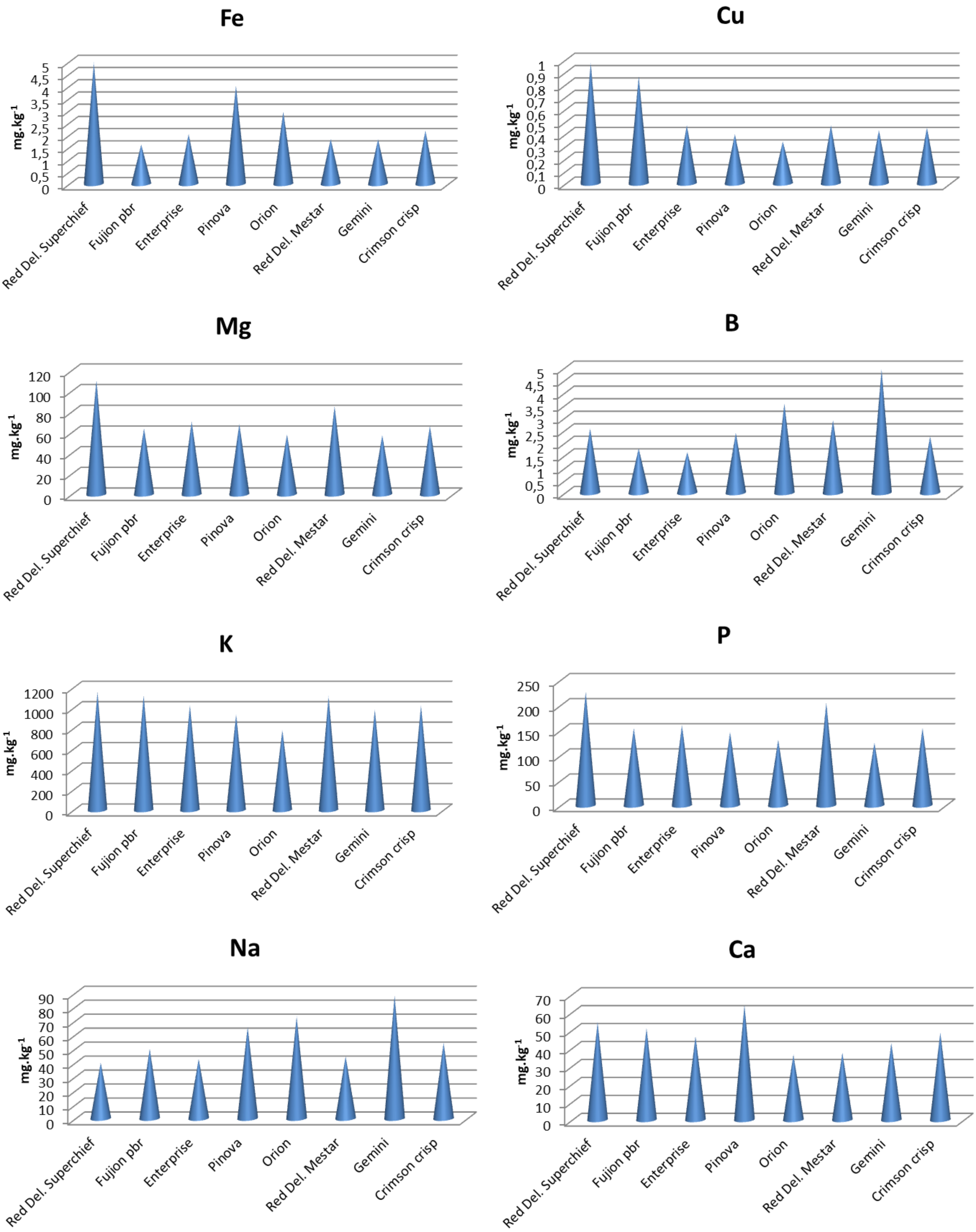

Fig. 1. Mean concentrations of heavy metals, macro- and microelements (mg.kg^{-1}) in apple fruit

The obtained values for trace elements (Cu, Zn, Mn, and Fe) and macronutrients (Na, K, Mg, and P) were higher than those reported in the USDA National Nutrient Database (2009). The differences in the results obtained are probably due to differences in the apple cultivars, environmental and geographical factors (soil, climate, etc.).

Jemaneh & Chandravanshi (2021) found different mineral content in red and green apple varieties. Green apple fruits have higher Ca and Al content than the red apple fruits, while Ni content in the green apple fruits was lower than in the red apple fruits. The authors also reported that the analyzed red and green apples did not contain toxic metals such as Cd and Pb, which does not comply with the results in the current study.

The mineral content not only affects the health of the consumers but also the quality of the apple fruit. Ca and Fe are macronutrients that affect the quality of fruit in its fresh state (Campeanu et al., 2009). Calcium plays a critical role in the ripening and aging processes (Marcelle, 1995). Apple fruits with low Ca content are susceptible to pathological diseases and have low storage potential (Nour et al., 2010).

CONCLUSION

There were significant varietal differences in the content of heavy metals, macroelements and microelements in the analyzed apple fruits. The highest contents of Pb, Cd, Zn, Cu, Fe, Mn, P, Mg and K were found in Red Del. Superchief, Ca and Hg in Pinova, Cr and Ni in Orion, and B, Na and As in Gemini. The content of Cd, Zn, As, Cr, and Ni in the fruit of all tested varieties was below the maximum permissible level. However, the Pb content of the fruit of the varieties Red Del. Superchief, Fujion pbr, Orion, Gemini and Crimson crisp exceeded the maximum permissible level (0.1 $mg.kg^{-1}$). The Pb content of Enterprise, Pinova and Red Del. Mestar apples was low and did not exceed the maximum permissible level for heavy metals in food, thus these apple varieties can be included in the daily human diet without any restrictions.

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