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THE HEAVY METAL CONTENT IN SELECTED KIND OF SPICES

Ľuboš Harangozo[1], Marek Šnirc[1], Július Árvay[1], Daniel Bajčan[1], Judita Bystrická[1], Pavol Trebichalský[1], Ján Kovarovič[1], Ivona Jančo

Address(es): Ing. Ľuboš Harangozo, PhD.,
[1]Slovak University of Agriculture in Nitra, Faculty of Biotechnology and Food Sciences, Department of Chemistry, Tr. A. Hlinku 2, 949 76 Nitra, phone number:+4213764134376

*Corresponding author: harangozolubos@gmail.com

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ABSTRACT

Hazardous (heavy) metals are widely spreaded in our environment, which is endangered by these hazardous metals that infiltrate water, air, soil, plants and subsequently in the food chain. This has a major impact on human health and animal health, because these metals can cause serious health problems. For this reason, it is very important to control the occurrence of heavy metals in the environment, water, air and soil, and then eliminate them to the permissible limit values.
Spices have been used in the past as natural flavors, used to highlight or create flavor. Nowadays, spices are commonly found in different trade chains and different brands. Consumer information is a very important decision-making factor when purchasing, but there are not always, on the packaging, all the information on harmful substances. For this reason, it is important to clarify the content of the risk metals, as it can prevent health problems and difficulties, and consumers can choose a healthy food (spice). Regarding this fact, the aim of this study was to closely determine the amount of hazardous (heavy) metals in selected kinds of spices (basil, black pepper, marjoram, sweet red paprika) that can cause serious health issues, if the limited levels of these heavy metals described in the Codex Alimentarius of Slovak republic are overstepped. The analyzed spices were purchased from three local markets. The amount of lead (5.59 $mg.kg^{-1}$) and cadmium (1.38 $mg.kg^{-1}$) were exceeding the highest acceptable limits stated in the Codex Alimentarius SR (Cd 0,50 $mg.kg^{-1}$, Pb 5 $mg.kg^{-1}$) in all three samples of basil. The content of cuprum and zinc was under the highest acceptable limits stated in the Codex Alimentarius of the Slovak Republic. According to these findings, we would recommend more inspections focused on the level of heavy metals in spices.

Keywords: risk mineral compounds, culinary herbs, limit, Codex Alimentarius

INTRODUCTION

The spices are obtained from spice plants. They are used as a whole spice plants or just a part of the root plant like underground or aboveground. They are used to improve sensory properties, but also have many health benefits to human organism. Species have been used in the past to overburden the odor of foods (**Habán *et al.*, 2001**). Culinary herbs have always been used for their organoleptic properties, because they have a distinctive aroma and a pleasant and delicious flavor. Aromatic herbs (basal, coriander, mint, oregano, rosemary, etc.) are widely used in the food industry, distilleries, candy processing (as aroma), and even as preservatives (**Cozzolino *et al.*, 2016**).
Spices are also used in the pharmaceutical industry. Consumers demand natural products and therefore synthetic substances are replaced by natural ones. One of the alternatives of natural active ingredients are herbs, spices and their extracts as a substitute. Their effects were tested primarily on humans, but also on laboratory animals (**Zotte *et al.*, 2016**).
Numerous studies have documented antioxidant, anti-inflammatory and immunomodulatory effects of species that may be associated with the prevention and treatment of some diseases (**Zheng *et al.*, 2016**).
In folk medicine, they were used for a number of biologically active substances and beneficial effects on health, for example, some antioxidants from species, such as curcumin from curcumin, eugenol from cloves and capsaicin from paprika (**Srinivasan, 2014**).
It has been experimentally proven the management of cellular oxidative stress based on antioxidant properties and their ability to block the production of reactive oxygen species and interference with signaling pathways (**Rubio *et al.*, 2013**).
Black pepper (*Pipper nigrum L.*, family *Piperaceae*) is one of the most widely used spices in the world, known for its acrid and penetrating component piperine. Piperine is in principle a bioactive compound, which has been reported to have anti-carcinogenic, antiasthmatic, antimicrobial, anti-inflammatory and anti-viral effects (**Meghwal and Goswami, 2013**).
Pharmacological studies have also showed anti-cancer and antioxidant effects (**Selvendiran *et al.*, 2006**). Interestingly, piperine also plays an important role in increasing the bioavailability of many drugs (**Chithra *et al.*, 2014**) by increasing intestinal absorption, suppressing drug metabolism in the human body, in pulmonary and liver tissues through inhibition of CYP3A4 and P-glycoprotein (**Makhov *et al.*, 2012**). Piperine as a pungent alkaloid has been identified as the most effective adjuvant to increase the efficacy of tumor necrosis induced apoptosis ligand (**Sattarinezhad *et al.*, 2015**).
Capsicum annum L. is an important agricultural crop, not only for its economic importance, but also for its role in the food, and its unique flavor and aroma. Sweet paprika is studied extensively for its high content of bioactive substances with antioxidant activity, such as carotenoids, vitamin C and A, phenolic substances (**Garcia-Mier *et al.*, 2015**) and micro and macro elements. Sweet paprika is a rich source of antioxidants in food (**Saini and Keum, 2016**).
The experimental proven potential of red paprika is in reducing oxidative stress (**Cervantes-Pay *et al.*, 2014**), inflammation (**Spiller *et al.*, 2008**), fat intake and body weight (**Kawabata *et al.*, 2006**).
The majoran (*Origanum majorana L.*) belongs to the family *Lamiaceae*. Old Egyptians, Greeks and Romans also used it. It has come to attention recently, mainly for its useful physiological function and antimicrobial activity (**Roby *et al.*, 2013**). It is used fresh or dried in food, beverages, perfumes and cosmetics, for its pleasant and highly desirable aroma (**Jiang *et al.*, 2011**). Essential oils of spices have been used as preservatives in studies, due to their antimicrobial activity against pathogens transmitted by food (**Marques *et al.*, 2015**).
Ocimum basilicum L. is a rich source of a certain class of secondary metabolites, known as phenolic acids. Basil is rich in rosmarinic, caftaric and cichoric acids and has antioxidant effects that have a number of beneficial effects on human health.
Purple varieties of basil also contain anthocyanins, a group of red-blue plant dyes, belonging to one of the largest classes of secondary metabolites, called flavonoids. Anthocyanins have anti-inflammatory, anti-carcinogenic and neuroprotective effects that are beneficial to humans (**McCance *et al.*, 2016**).

Heavy metal contamination is very dangerous from a health point of view, due their carcinogenicity and toxicity (**Baraka *et al*., 2007**). These metals enter to the human body through oral administration, inhalation, skin and hair contact (**Eastman *et al*., 2013; Censi *et al*.,2011**). In the body they cause acute and chronic toxicity, damage of the blood component, lung, kidney, liver, and central nervous and immune system, and sometimes they lead to letalite (**Zeng *et al*., 2016; Vallverdu-Coll *et al.*, 2015**).

Lead (Pb) is a systemic toxic substance that causes serious damage to virtually of all organs, but mainly affects the central nervous system, especially brain development (**Duruibe *et al.*, 2007**). Lead is a poison that also causes a reduction in hemoglobin formation and kidney dysfunction (**Nadeem *et al.*, 2006**). Pb (II) has been identified as one of the most toxic heavy metals because its effects are detrimental to the bloodstream, kidney and human reproductive system (**Yu *et al*., 2011; Huang *et al*., 2010**).

Cadmium accumulates in the human body in the kidneys, liver, breasts, prostate, but also in the brain and lungs (**Notarachille *et al*., 2014**). Cadmium is dangerous to humans for its toxicity; it has undesirable effects on kidney function and also increased exposure to cadmium results in an increased risk of cancer (**Satarug *et al*., 2010**).

Exposure to cadmium has also shown significant effects on human and animal health, including chemical pulmonary inflammation, bone deformity and renal impairment (**Bertin and Averbeck, 2006**). Excessive exposure to Cd^{2+} may lead to a disease that is affecting the cardiovascular system and GIT (**Shah and Nahakpam, 2012**).

MATERIAL AND METHODS

The spices – black pepper (*Piper nigrum L.)*, red sweet paprika (*Capsicum annuum L.*), basil (*Ocimum basilicum L.*), marjoram (*Origanum majorana L.*) were bought in the three local markets (A, B, C) (totally 20 samples). The obtained results were compared with the Codex Alimentarius of the Slovak Republic. All spices were grown out of the Slovak Republic.

Determination of heavy metals (by AAS)

By analytical weight were weighted 1 g samples of dried spices. Mineralization of samples was in a mixture of distilled water with concentrated nitric acid in a ratio 1:1. The weighed samples were put into Teflon vessels with 5 cm^3 of distilled water with 5 cm^3 of concentrated nitric acid. Closed vessels with the samples were mineralized (Tab. 1) by microwave digestion unit MARS X-press (USA).

Table 1 Experimental conditions for element determination by FAAS.

Stage	Power (W)	Power (%)	Initialization Time (min)	Temp. (°C)	Duration time (min)
Initialization	800	90	15	160	0
Mineralization	800	90	0	160	20
Cooling	–	–	–	–	20

After mineralization were analyzed samples filtered through quantitative filter paper MUNKTELL (Germany) grade 390.84 $g.m^{-2}$ (green) to volumetric flasks (50 cm^3).

Flasks were refilled with distilled water to the mark and after that was the determination of heavy metals by VARIAN AA 240FS (Australia). The detection limits and sensitivity are displayed in table 2.

Table 2 Detection limit and sensitivity of the investigated elements.

Element	Detection limit ($mg.l^{-1}$)	Sensitivity ($mg.l^{-1}$)
Cd	0.001	0.01
Pb	0.02	0.1
Cu	0.002	0.03
Zn	0.006	0.008
Co	0.005	0.05
Cr	0.003	0.04
Ni	0.008	0.06
Mn	0.003	0.02
Fe	0.005	0.04

Analysis determination has not a deviation more than 3%, the gas flow: air: 13.5 $l.min^{-1}$, acetylene 2.0 $l.min^{-1}$.

Statistical analysis

For testing the normal division data were used the Shapiro-Wilk test and the Kolmogorov-Smirnov test. As a result, there were chosen non-parametric test, there was used a Kruskal-Wallis test with a significance level of p-0.05 to testing the statistical hypotheses. To describe the data set was used the arithmetic mean and the standard deviation. All calculations were performed using MS Excel 2016 and XLSTAT (**Addinsoft, 2014**).

RESULTS AND DISCUSSION

The following graphs show the content of the risk metals in selected types of spices, obtained from markets A, B and C, and then their comparison with the Codex Alimentarius (CA) of the Slovak Republic. Results are shown in figures 1-7, which indicated the average values of each element and limit value according to Codex Alimentarius.

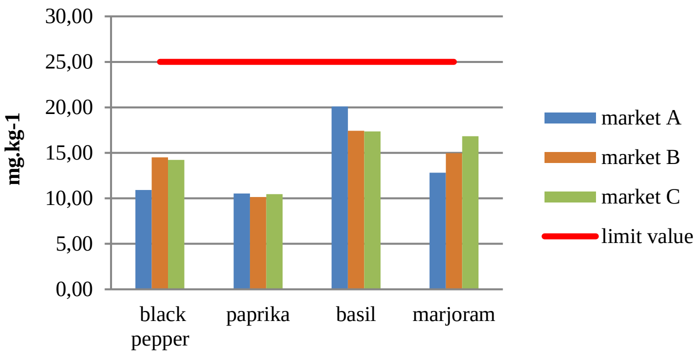

Figure 1 The copper content in selected spices from three markets in $mg.kg^{-1}$, compared to limit in CA of the Slovak Republic.

The highest copper content was found in basil from market A (20.11 $mg.kg^{-1}$), in samples from market B and C was copper content approximately in the same level (17.40 $mg.kg^{-1}$). The copper content in sweet red paprika was approximately 10 $mg.kg^{-1}$ in all tested samples. The copper content in black pepper samples ranged from 10.93 $mg.kg^{-1}$ (A) to 12.44 $mg.kg^{-1}$ (B). In marjoram, the copper content ranged from 12.83 $mg.kg^{-1}$ (A) to 16.81 $mg.kg^{-1}$) (B). There were observed statistically significant differences in copper content between marjoram producers. Also, there were observed statistically significant differences in copper content between black pepper samples from tested producers (Table 3). All the tested samples were bellow the limit value according to Codex Alimentarius. **Bielicka-Gieldon and Rylko (2013)** reported a copper content in black pepper of 10.6 ± 0.03 $mg.kg^{-1}$, in sweet paprika12.6 ± 0.05 $mg.kg^{-1}$, which is in accordance whit our results. **Valtcho, Lyle, and Baoshan (2006)** reported a copper content in basil 22.5 $mg.kg^{-1}$. Significant differences of the copper content were also found in marjoram and black pepper in market A, as opposed to markets B and C. Increasing copper content in selected market spices is in the following order: sweet paprika < black pepper < marjoram < basil.

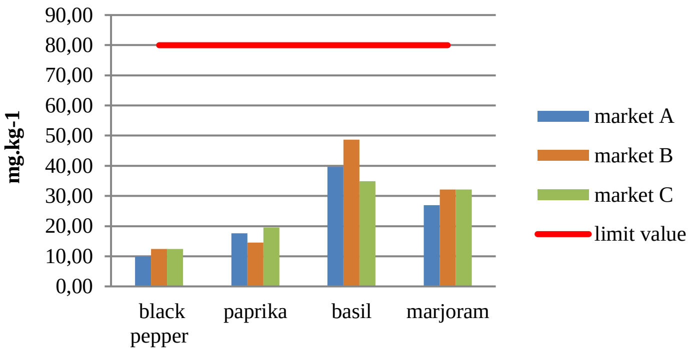

Figure 2 The zinc content in selected spices from three market in $mg.kg^{-1}$, compared to limit in CA of the Slovak Republic.

The lowest zinc content was detected in black pepper, approximately 12.4 $mg.kg^{-1}$for all the three markets. The highest content of zinc was observed in basil and ranged from 34.86 $mg.kg^{-1}$ to 48.65 $mg.kg^{-1}$. In the case of sweet red paprika, the content of zinc ranged from 14.58 $mg.kg^{-1}$ to 19.51 $mg.kg^{-1}$. The lowest content of zinc in marjoram was observed in the samples from market not work A (26.9 $mg.kg^{-1}$), the other samples had approximately the same values around 32.1 $mg.kg^{-1}$. All the tested values were bellow the limit value according to Codex Alimentarius. There were observed statistically significant differences between the producers in zinc content in basil and sweet red pepper (Table 3). **Bielicka-Gieldon and Rylko (2013)** reported the zinc content in black pepper 15.9 ± 0.1 $mg.kg^{-1}$, which is comparable to our achieved results. The highest zinc concentration was measured in basil, with the highest content in market network B (48.65 $mg.kg^{-1}$). The increasing content of zinc in selected market network spices is in the following order: black pepper < sweet red paprika < marjoram < basil.

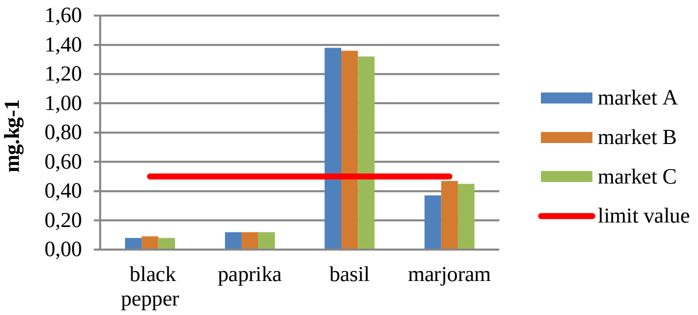

Figure 3 The cadmium content in selected spices from three markets in mg.kg^{-1}, compared to limit in CA of the Slovak Republic.

The highest amount of cadmium was measured in basal from market A (1.38 mg.kg^{-1}), which was eight times more than was measured in black pepper and sweet red paprika of all three markets. The content of cadmium in basil in all three markets exceeded the limit of Codex Alimentarius of Slovak Republic. The cadmium content in marjoram ranged from 0.37 mg.kg^{-1} (A) to 0.47 mg.kg^{-1} (B). There were observed no statistically significant differences between the producers in cadmium content. The cadmium content in sweet red pepper was approximately 1.12 mg.kg^{-1} in all three markets. **Bojňanská, Frančáková and Pavlisová (2002)** analyzed a cadmium content of 0.44624 mg.kg^{-1} in maize and compared to our measured values, it coincides with all three markets. **Valtcho, Lyle, and Baoshan (2006)** reported a cadmium content in basil 0.7 mg.kg^{-1}. Seven Hungarian spice paprika samples (7/22) were contaminated with cadmium, the average concentration was 0,086 ± 0,002 mg.kg^{-1} In case of the Serbian (2/5) and Peruvian samples (2/2), these values were 0,170 ± 0,001 and 0,254 ± 0,001 mg.kg^{-1}, respectively. Cadmium contamination was not detectable in the Spanish, Chinese and Bulgarian samples **(Molnar *et al.*, 2018).** The increasing content of cadmium in selected species of market network is in the following order: black pepper < sweet red paprika < pepper < marjoram < basil.

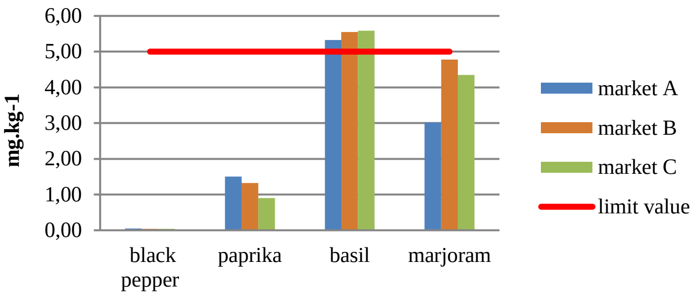

Figure 4 The lead content in selected spices from three markets in mg.kg^{-1}, compared to limit in CA of the Slovak Republic.

The lowest content of lead in market A, B, C was recorded in black pepper (0.05, 0.04, 0.04 mg.kg^{-1}). The highest lead content was measured in basil, which even exceeded the highest permissible levels for all three markets, compared to the Codex Alimentorius of the Slovak Republic. In comparison to the lead content in basil, in at all three brands its content is three times higher than in sweet red paprika. Increasing lead content in selected market network spices is in the following order: black pepper < sweet red paprika < marjoram < basil. The lead content in sweet red pepper ranged from 0.90 mg.kg^{-1} (C) to 1.50 mg.kg^{-1} (A). In case of marjoram, the content of lead ranged from 3.02 mg.kg^{-1} (A) to 4.78 mg.kg^{-1} (C). **Valtcho, Lyle, and Baoshan (2006)** reported a lead content in basil Leaves from 26.8 to 33.6mg.kg^{-1}. Lead was detected in 1 out of 22 Hungarian spice paprika samples, at a level of 0,334 ± 0,0013 mg.kg^{-1}, below the legal limit **(Molnar *et al.*, 2018).** There were observed statistically significant differences between the producers in lead content in marjoram, basil and sweet red pepper (Table 3).

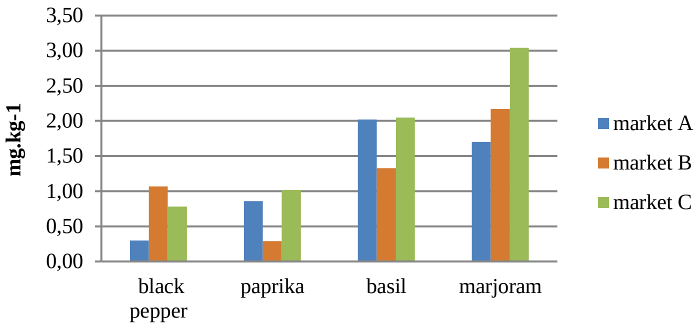

Figure 5 The chromium content in selected spices from three markets in mg.kg^{-1}, compared to limit in CA of the Slovak Republic.

Compared to these four spice types, the lowest chromium content was recorded in the black pepper and sweet red paprika. The chromium content in the marjoram from market C (3.04 mg.kg^{-1}) is more than three times higher as the average amount of chromium in black pepper and sweet red paprika of all three market networks. In the case of basil, the chrome content from market network A (2.02 mg.kg^{-1}) and market network C (3.05 mg.kg^{-1}) is approximately the same. The rising content of chromium in selected species from market A is in the following order: black pepper < sweet red paprika < basil < marjoram. There were observed statistically significant differences between the producers in chromium content in marjoram, black pepper and sweet red paprika (Table 3).

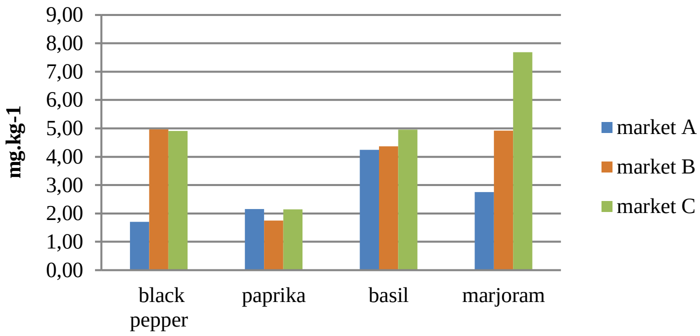

Figure 6 The nikel content in selected spices from three markets in mg.kg^{-1}, compared to limit in CA of the Slovak Republic.

The lowest average values of nickel were recorded in sweet red paprika of all markets (1.75- 2.16 mg.kg^{-1}). The highest nickel content was measured in marjoram in network market C (7.69 mg.kg^{-1}). The nickel content in black pepper ranged from 1.70 mg.kg^{-1} (A) to 4.97 mg.kg^{-1} (B). There were observed statistically significant differences between the producers in nickel content in marjoram, and basil (Table 3). **Bielicka-Gieldon and Rylko (2013)** reported a nickel content in sweet red paprika of 1.29 ± 0.14 mg.kg^{-1} and the nickel values that we measured in the sweet pepper were slightly higher. The increasing content of nickel in selected species of network market A is in the following order: black pepper < sweet red paprika < marjoram < basil. The increasing content of nickel in selected spices of market B is in the following order: sweet red paprika < basil < marjoram < black pepper. The increasing content of nickel in the selected species of market C is in the following order: sweet red paprika < black pepper < basil < marjoram.

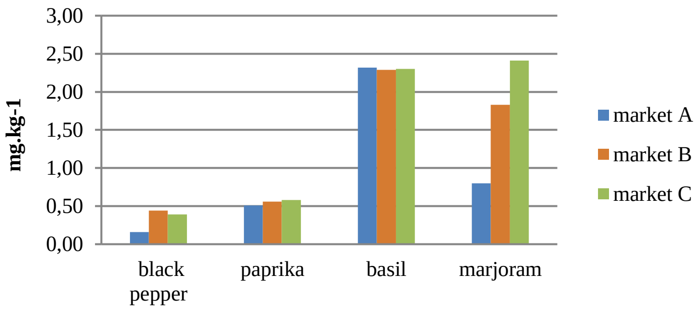

Figure 7 The cobalt content in selected spices from three markets in mg.kg^{-1}, compared to limit in CA of the Slovak Republic.

Cobalt content in black pepper and sweet red paprika was found in small quantities. The highest cobalt content, at almost the same level (approximately

2.3 mg.kg^{-1}) in all markets was recorded in basil. Cobalt content in marjoram from market A (0.80 mg.kg^{-1}) was three times lower than in market C (2.41 mg.kg^{-1}). Increasing cobalt content in selected species of market A was recorded in the following order: black pepper < sweet red paprika < marjoram < basil. Increasing cobalt content in selected species of market B was in the following order: black pepper < sweet red paprika < marjoram < basil. Increasing cobalt content in selected species market network C was in the following order: black pepper < sweet red paprika < basil < marjoram. As can be seen in the table 3, there were observed statistically significant differences between the producers in cobalt content in marjoram, sweet red red paprika and black pepper.

Statistic results

Table 3 Summary statistical results of Kruskal-Wallis Test (P-values).

Element	MARJORAM	BASIL	SWEET RED PEPPER	BLACK PEPPER
Zn	0.0665	**0.0273**	**0.0273**	0.0650
Cu	**0.0273**	0.0628	0.0509	**0.0321**
Co	**0.0273**	0.5164	**0.0312**	**0.0265**
Ni	**0.0273**	**0.0273**	0.0552	0.0608
Cr	**0.0273**	0.0549	**0.0273**	**0.0273**
Cd	0.0509	0.0769	0.8601	0.2082
Pb	**0.0273**	**0.0447**	**0.0273**	0.6074

CONCLUSION

In the analysis were found significant differences in the contents of the hazardous metals, but just some of them exceeded the limit values that are given in the CA of the Slovak Republic. Exceeded limit values were found for lead and cadmium in all market networks, and in marjoram from market network C was the nickel content at the level of the highest limit according to CA of the Slovak Republic. On the basis of some exceeded maximum limits in comparison with heavy metals in spices with CA of the Slovak Republic, it would be advisable to increase the control interval for heavy metals by focusing mainly on their content in basil.

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