Hop and Medicinal Plants, Year XXIV, No. 1-2, 2016
ISSN 2360 – 0179 print, ISSN 2360 – 0187 electronic

CHEMICAL RISK DUE TO HEAVY METAL CONTAMINATION OF MEDICINAL PLANTS

MUNTEAN Edward[1], MICHALSKI Rajmund[2], MUNTEAN Nicoleta[3], DUDA Marcel[4]

[1]University of Agricultural Sciences and Veterinary Medicine Cluj Napoca, Faculty of Food Sciences and Technology, Department of Food Sciences, Romania, edimuntean@yahoo.com

[2] Institute of Environmental Engineering, Polish Academy of Sciences, Sklodowska-Curie 34, Zabrze, Poland, rajmund.michalski@ipis.zabrze.pl

[3]Regional Public Health Center Cluj-Napoca, Romania, str. Louis Pasteur nr. 6, 400046, Cluj–Napoca, Romania

[4]University of Agricultural Sciences and Veterinary Medicine Cluj Napoca, Faculty of Agriculture, Department of Phitotechny, Romania

Abstract. The paper highlights some features related with the quality of medicinal plants, the focus being directed towards the heavy metal content of some commercially available products between 2013-2015: basil, chamomile, mulberry, plantain, dandelion, mint, St. John's wort, marigold, fennel and dill. Four heavy metals were investigated (cadmium, lead, zinc and copper) using atomic absorption spectrophotometry, measurements being performed using a Shimadzu AA-6300 double beam atomic absorption spectrophotometer with graphite furnace, after microwave-assisted digestion. The concentration ranges were as follows: for lead from 0.07 μg/kg (chamomile - in 2013) to 45.01 μg/kg (dill - in 2013), for cadmium from 0.88 μg/kg (plantain - in 2013) to 25.63 μg/kg, for zinc from 1.45 mg/kg (fennel – in 2013) to 22.38 mg/kg (marigold – in 2015) and for copper from 2.34 mg/kg (dill – in 2013) to 13.49 mg/kg (marigold – in 2015). The contaminants' contents of herbal teas are variable due to the factors like differences between the plants species, geographical area and exposure to different pollution sources or conditions during drying process.

Keywords: heavy metals, contamination, medicinal plants, phytopharmacy

Introduction

The benefits of consumption herbal teas or other remedies obtained from medicinal plants are multiple: they induce relaxation, contribute to heart disease therapy, relieves digestive problems, detoxifies the body, strengthens the immune system, provides antioxidants to the body, helps prevent colds, stimulates internal organ function, have antiparasitic and

antimicrobial effects, hepato-protective effect, antitumor effects etc. (Gupta et al., 2013; Kazemi et al., 2010; Li et al., 2013; Noor et al., 2013).

Besides biologically active constituents, medicinal plants contain usually variable amounts of hazardous substances, such as heavy metals, pesticides, polyhydroxy aromatic compounds, etc. (El Nahhal, 2004; Muntean et al., 2013; Princewill-Ogbonna and Ogbonna, 2011; Sarma et al., 2012; Street, 2012; Subramanian et al., 2009). Pollution is the major cause of this situation, which can lead to unwanted side-effects of using such remedies, many cases of liver, kidney or other organ damage being recorded (Ebrahim et al., 2012; Rodriguez-Fragoso et al., 2008). Unfortunately, in most countries there is no rigorous regulation and control for medicinal plants and the unreliable quality of herbal products can raise serious problems for certain patients. In most cases, medicinal plants addresses chronic conditions, so that the therapeutic effects are obtained after lengthy treatments which may take several months or even longer, so that a toxic load can represent a risk, especially due to the accumulation potential of some contaminants, the situation being compounded by the fact that in many cases herbal remedies are used by a sensitive segment of population, such as pregnant women, children or old patients with chronic diseases (Alica et al., 2015; Ernst, 2004; Kosalek et al., 2009; Muntean et al., 2014; Nasri and Shirzad, 2013; Nasri and Rafieian-Kopaei, 2014).

Contrary to the common opinion that medicinal plants are safe because they are natural, a significative number of papers highlighted the severe consequences from side effects from certain herbal products which have been reported from the use of some herbal products either due to the toxic effects of the herbs, or due to the effects of contaminants; by using unreliable medicinal plants one can be exposed to different risks, such as poisoning due to their improper use (dozens of plants contain substances with toxic effects on the liver, stomach, intestine, nervous system and blood) or as a result their contamination with chemicals originating from industrial emissions, agriculture, energy production and transport (Chan, 2003; Caldas et Machado, 2004; Jabeen et al., 2010; Muntean et al., 2010).

Despite heavy metals are natural components of the Earth's crust, they being common components of various matrices, the concentration of several heavy metals has increased in last years as a result of anthropogenic activities, many of them posing toxicological risk on human health at higher concentrations (Elkekes et al., 2010; Rodriguez et al., 2008). This paper focuses on medicinal plants' contamination with heavy metals, which can occur in all stages of their development as a result of inputs from the environment (soil, water, or air - rainfall, atmospheric dust, plant protective agents and fertilizers) and also later, during the manufacturing processes (Karimi et al., 2008; Nagajyoti et al., 2010). This study is a continuation of a

former one (Muntean et al., 2013), extending the previous research with two more years: 2014 and 2015.

Materials and methods

Herbal teas samples were acquired from the local supermarkets in Cluj-Napoca, targeting the same brands during the whole study period (2013-2015): basil (*Basilici folium*), chamomile (*Chamomillae folium*), mulberry (*Mori folium*), plantain (*Plantaginis folium*), dandelion (*Taraxaci herba*), mint (*Menthae herba*), St. John's wort (*Hyperici herba*), marigold (*Calendula flos*), fennel (*Foeniculi fructus)* and dill (*Anethi fructus*).

The nitric acid solution used (65%) was of ultra pure grade (Merck, Germany), while hydrogen peroxide and standards solutions for atomic absorption spectroscopy containing 1000 mg/ L metal in HNO_3 were from Fluka. All solutions were prepared using ultrapure water with a specific resistance of 18.2 MΩ / cm, obtained from Direct Q 3UV Smart (Millipore).

Around 0.5 g homogenized samples were accurately weighed and transferred in Teflon reaction vessels, then 5 mL of HNO_3 65% and 3 mL of H_2O_2 were added; wet digestion was accomplished with a Berghoff Microwave Digestion System MWS-3+ (190^0C for 2 hours).

Measurements were performed using an AA-6300 – Shimadzu double beam atomic absorption spectrophotometer (Shimadzu Corporation, Japan) with both flame and graphite furnace atomization, equipped with deuterium lamp for background correction and hallow-cathode lamps for each of the studied elements. Air–acetylene flame was used for determination of all the elements. Calibration curves were prepared using five concentrations, the linear correlation coefficients obtained ranging between 0.985 - 0.996. The operation conditions were those recommended for each metal in the instrument's method. All measurements were carried out in triplicates. Instrument control, data acquisition and spectrophotometric data analysis were accomplished using WizAArd software (Shimadzu Corporation, Japan); experimental data were finally processed in Excel 2003 (Microsoft) and mean values were reported.

Results and discussions

The content of lead, cadmium, zinc and copper determined in selected commercial herbal teas were summarized in figures 1-4. Figure 1 reveals different contamination patterns with lead, dill and basil being the most contaminated plant matrices, while chamomile and fennel were found

to contain only small amounts. Lead concentrations ranged from 0.07 µg/kg (chamomile) to 45.01 µg/kg (dill), both values being from 2013.

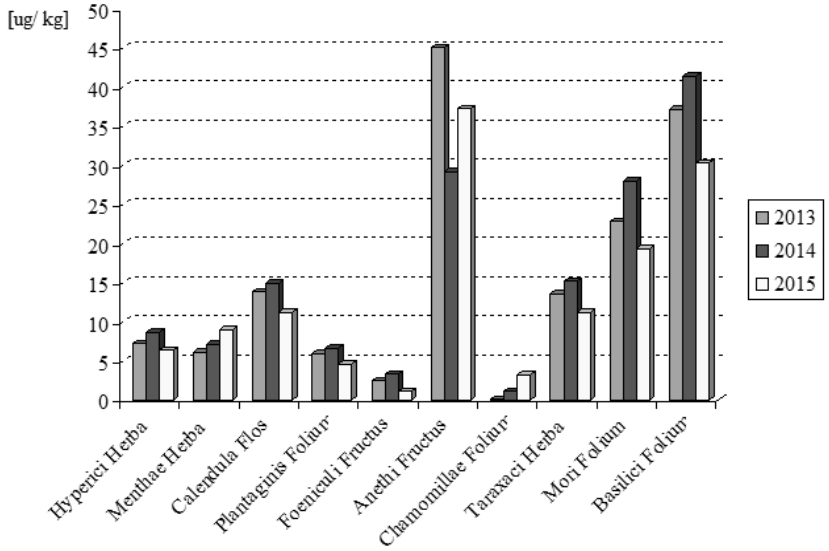

Fig. 1. Average lead concentration in selected medicinal plants

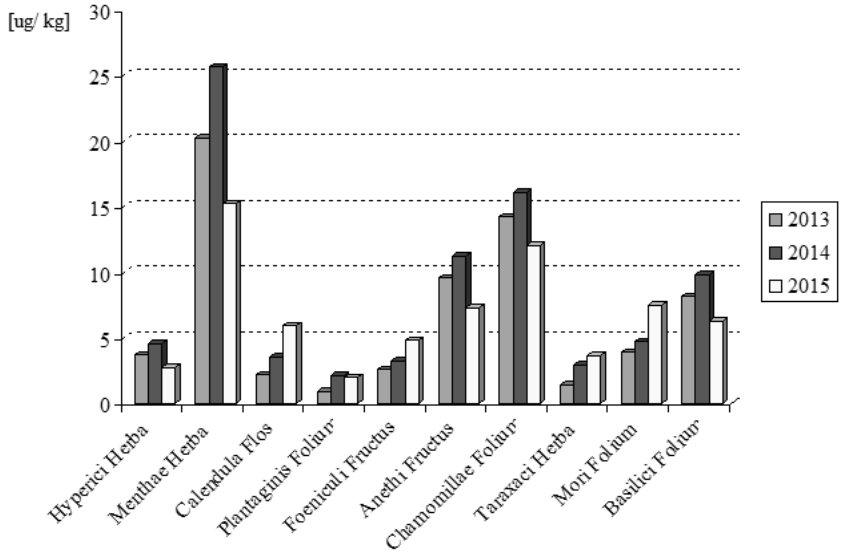

Fig. 2. Average cadmium concentration in selected medicinal plants

From figure 2 one can distinguish the maximum content of cadmium from mint, this being followed by chamomile, dill and basil; the concentration range for cadmium was from 0.88 µg/kg (plantain - in 2013) to 25.63 µg/kg (mint – in 2014).

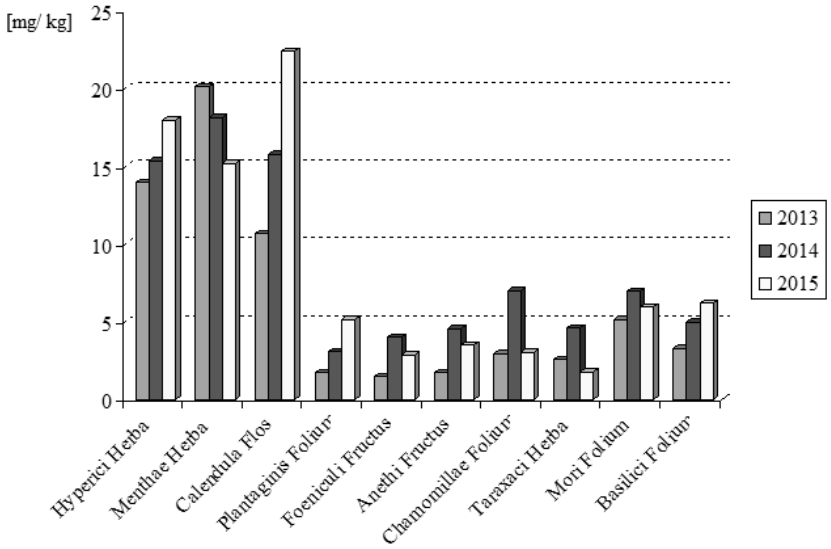

Fig. 3. Average zinc concentration in selected medicinal plants

The status of zinc content is different, the major concentrations of this element being recorded in St. John's wort, mint and marigold (fig.3); the zinc concentrations ranged from 1.45 mg/kg (fennel – in 2013) to 22.38 mg/kg (marigold – in 2015).

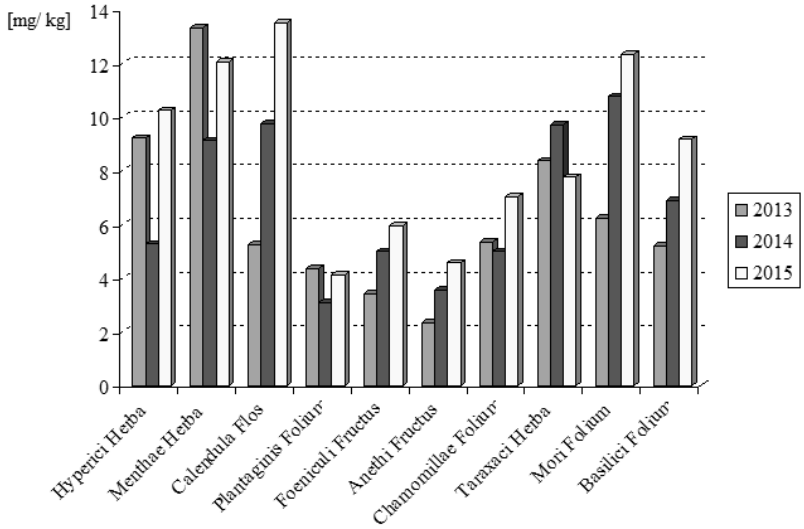

Fig. 4. Average copper concentration in selected medicinal plants

Smaller concentrations were recorded for copper, ranging from 2.34 mg/kg (dill – in 2013) to 13.49 mg/kg (marigold – in 2015), the highest concentrations being recorded in mint and marigold (fig.4).

Conclusions

The beneficial role of herbal teas and of other herbal remedies is undeniable, but the heavy metal content of medicinal plants is affected by environmental pollution, as was shown by the analysis of the selected medicinal plants; lead, cadmium, zinc and copper were recorded in a wide range of concentrations. The maximum content of the most hazardous heavy metals (sum of lead and cadmium) were recorded in dill (139.39 μg/kg overall, during the three years of study) and basil (131.01 μg/kg). The heavy metals contents of herbal teas are variable due to the factors like differences between the plants species, geographical area and exposure to different pollution sources, as well as processing.

Good manufacturing practice shall provide a proper control of the raw material in order to assure the quality of the herbal teas, with a special emphasis on a strict survey of the harvest areas and on processing in order to maintain the heavy metals' content at the lowest possible value. Since the medicinal plants and their extracts are used in traditional medicine, there is a possible hazard of heavy metal poisoning, if they come from the polluted areas and therefore these raw materials should be collected from nonpolluted regions and they should be analyzed for heavy metal content in order to avoid their cumulative toxicities in long-term use.

Due to their hazard, the content of heavy metals has to be one of the main criteria for the use of medicinal plants as raw materials in the production of traditional remedies; hence it is essential to have a proper quality control of such raw materials used to ensure safety and efficacy of herbal products.

References

1. Alica B., Martina F., Marek B., Ľubomír L., Tomáš T., Lucia Z. (2015). Selected parameters of quality and safety of herbal tea. Potravinárstvo, 9(1):183-189.
2. Caldas E.D., Machado L.L. (2004) Cadmium, mercury and lead in medicinal herbs in Brazil. Food and Chemical Toxicology, 42(4):599-603.

3. Chan K. (2003), Some aspects of toxic contaminants in herbal medicines. Chemosphere, 52:1361 - 1371.
4. Ebrahim A.M., Eltayeb M.H., Khalid H., Mohamed H., Abdalla W., Grill P., Michalke B. (2012). Study on selected trace elements and heavy metals in some popular medicinal plants from Sudan. Journal of Natural Medicines, 66(4):671-679.
5. El Nahhal Y. (2004). Contamination and safety status of plant and food in Arab countries. Journal of Applied Sciences, 4(3):411-417.
6. Elekes C.C., Dumitriu I., Busuioc G., Iliescu N.S. (2010) The appreciation of mineral element accumulation level in some herbaceous plants species by ICP-AES method. Environment Science and Pollution Research, 17:1230–1236.
7. Ernst E.(2004), Risks of herbal medicinal products. Pharmacoepidemiology and drug safety, 13(11):767-771.
8. Gupta A., Khamkar P.R., Chaphalkar S.R. (2014). Review on medicinal plants to target and inhibit the epidermal growth factor receptor signaling in cancer and tissue repair therapy. International Journal of Advances in Pharmacy, Biology and Chemistry, 3(1), 210-213.
9. Jabeen S., Tahir Shah M., Khan S., Qasim Hayat M. (2010), Determination of major and trace elements in ten important folk therapeutic plants of Haripur basin, Pakistan. Journal of Medicinal Plants Research, 4:559 – 566.
10. Karimi G., Hasanzadeh M.K., Nili A., Khashayarmanesh Z., Samiei Z., Nazari F., Teimuri M., (2008), Concentrations and health risk of heavy metals in tea samples marketed in Iran, Pharmacology, 3:164–174.
11. Kazemi S., Asgary S., Moshtaghian J., Rafieian M., Adelnia A., Shamsi F. (2010). Liver-protective effects of hydroalcoholic extract of allium hir-tifolium boiss. In rats with alloxan-induced diabetes mellitus. ARYA Atherosclerosis, 6(1):11-5.
12. Kosalec I., Cvek J., Tomić S. (2009). Contaminants of medicinal herbs and herbal products. Archives of Industrial Hygiene and Toxicology, 60(4): 485-501.
13. Li S., Li S.K., Gan R.Y., Song F.L., Kuang L., Li H. B. (2013). Antioxidant capacities and total phenolic contents of infusions from 223 medicinal plants. Industrial Crops and Products, 51, 289-298.
14. Muntean N., Muntean E., Creta C., Duda M. (2013). Heavy metals in some commercial herbal teas. ProEnvironment/ ProMediu, 6:591-594.
15. Muntean E., Muntean N., Mihăiescu T. (2010). Cadmium and lead soil pollution in Copsa Mica area in relation with the food chain. Research Journal of Agricultural Science, 42(3):731-734.

16. Nagajyoti P.C., Lee K.D., Sreekanth T.V.M. (2010), Heavy metals, occurrence and toxicity for plants: a review. Environmental Chemistry Letters, 8(3): 199-216.
17. Nasri H., Rafieian-Kopaei M. (2014). Medicinal plants and antioxidants: why they are not always beneficial? Iranian Journal of Public Health, 43(2):255-257.
18. Nasri H., Shirzad H. (2013). Toxicity and safety of medicinal plants. Journal of HerbMed Pharmacology, 2(2):21- 22.
19. Noor A., Bansal V.S., Vijayalakshmi M. A. (2013). Current update on anti-diabetic biomolecules from key traditional Indian medicinal plants. Current Science, 104(6), 721-727.
20. Princewill-Ogbonna I.L., Ogbonna P.C. (2011). Heavy metal content in soil and medicinal plants in high traffic urban area. Pakistan Journal of Nutrition, 10:618-624.
21. Rodriguez-Fragoso L., Reyes-Esparza J., Burchiel S.W., Herrera-Ruiz D., Torres E. (2008). Risks and benefits of commonly used herbal medicines in Mexico. Toxicology and Applied Pharmacology, 227(1):125-135.
22. Sarma H., Deka S., Deka H., Saikia R.R. (2012). Accumulation of heavy metals in selected medicinal plants. In Reviews of environmental contamination and toxicology, Springer New York: 63-86.
23. Street R.A. (2012). Heavy metals in medicinal plant products—An African perspective. South African Journal of Botany, 82:67-74.
24. Subramanian R., Gayathri S., Rathnavel C., Raj V. (2012). Analysis of mineral and heavy metals in some medicinal plants collected from local market. Asian Pacific Journal of Tropical Biomedicine, 2(1):S74-S78.