POLYCICLIC AROMATIC HYDROCARBONS IN MEDICINAL HERBAL TEAS

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Introduction

The presence of polycyclic aromatic hydrocarbons (PAHs) in herbal teas may result in health risk to consumers, which are generally hoping that a natural remedy may represent a healing chance. Contamination of herbal teas with PAHs can occur as a result of environmental pollution or by uptake from water or soil, as well as during processing, especially in some preservation or drying stages that allow combustion products to come into contact with plants . This research fits into current concerns directed towards establishing the PAHs' contamination level of some commercial brands of herbal teas, the focus being on the US Environmental Protection Agency's priority PAHs contaminants as established by the European Union and by the US Environmental Protection Agency : benzo[a]-anthracene, benzo[b]fluoranthene, indeno[1,2,3-cd]pyrene, benzo[k]fluoranthene, benzo[a]pyrene, dibenzo-[a,h]anthracene, naphthalene, acenaphthene, acenaphthylene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, chrysene and benzo(g,h,i)perylene. High performance liquid chromatography (HPLC) was the analytical tool used to investigate the PAHs' content from several commercial herbal teas brands from Romanian market. PAHs' content of 12 commercial herbal teas brands was investigated by high performance liquid chromatography (HPLC): basil (*Basilici Folium*), chamomile (*Chamomillae Folium*), mulberry (*Mori Folium*), plantain (*Plantaginis Folium*), dandelion (*Taraxaci Herba*), mint (*Menthae Herba*), St. John's wort (*Hyperici Herba*), black locust (*Acacia Flos*), common elder (*Sambuci Flos*), lime (*Tiliae Flos*), marigold (*Calendula Flos*) and fennel (*Foeniculi Fructus*).

Materials & methods

12 herbal tea brands were purchased from local hypermarkets. ~5 g tea samples were grounded, weighed then extracted with 20 mL hexane:acetone (1:1) for 30 minutes by ultrasonication, at room temperature ; the supernatant was decanted while the residue was re-extracted with a new volume of solvent mixture in the same conditions, the procedure being repeated twice more. The obtained extracts were combined, passed through an anhydrous Na_2SO_4 layer and evaporated to dryness in a rotary evaporator; the residue was dissolved in 2 mL acetonitrile, then the final solution was filtered through 0.47 μm PTFE membrane filters and injected in the HPLC system. Three replicates from each tea brand followed the same procedure.

PAHs were determined by a sensitive optimized HPLC method, using an Agilent 1100 system with fluorescence detection in which baseline separations for 16 PAHs (naphtalene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benz(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(a,h) anthracene, benzo(g,h,i)perylene, indeno(1,2,3-c,d)pyrene) was accomplished using an Envirosep PP column in a total run time less than 30 minutes (fig.1). Recoveries for each PAHs were established using samples spiked with a standard mixture of PAHs.

Principal component analysis (PCA) and cluster analysis were completed on autoscaled data using Matlab (The Mathworks Inc., USA).

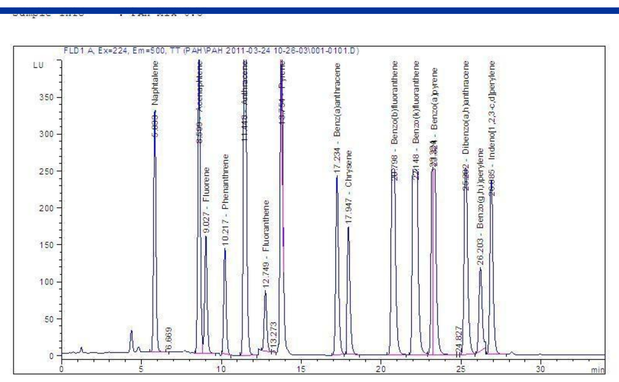


Figure 1. HPLC chromatogram of PAHs – reference mixture

Results

- The maximum overall PAH's contamination in the studied herbal teas was recorded for dandelion (131.15 ppb), mulberry (27.78 ppb) and St.John's wort (18.28 ppb), all of these exposing high surface area of leaves to environmental contamination with PAHs (table 1).
- From the 15 quantified PAHs, only three were present in all samples (acenaphthene, fluorene and benzo(b)fluoranthene), while anthracene and benzo(g,h,i)perylene were quantified in only two samples.
- The maximum levels of individual PAHs were recorded in dandelion: 30.79 ppb naphthalene, 28.91 ppb pyrene, 23.79 ppb phenanthrene and 10.28 ppb benzo(b)fluoranthene. Dandelion herbal tea exhibit also the largest PAH's range (14 PAHs).
- The lowest PAHs concentrations were recorded in basil and fennel (1,96 and 2.20 ppb), these being the least contaminated also in terms of PAH's range (only 7 from the 15 studied PAHs).
- Carcinogenic PAHs ranged from 0.45 ppb (lime) to 36.54 ppb (dandelion), with St.John's worth revealing the maximum concentration of benzo(g,h,i) perylene from all samples.
- Principal component analysis of available experimental data reveals a close correlation between the recorded concentrations of naphtalene, anthracene, pyrene, benzo(a)pyrene, benzo(b)fluoranthene and benzo(k)fluoranthene, while acenaphtene and dibenzo(a,h)anthracene are anti-correlated; bez(a)anthracene's contribution to the variability of data is small. Based on this, a PCA model was developed for 14 variables and 2 principal components, explaining 76.33% variance (figures 3 and 4), in which "mulberry" is an outlier, "dandelion" is outside the model border, while the other samples joins together in one cluster..

Table 1. Mean values of recorded PAHs' concentrations in the studied herbal teas [ppb] – N.D = not detected

	Basil	Fennel	Lime	Platain	Black locust	Chamomile	Mint	Marygold	Elder	St. John's wort	Mulberry	Dandelion
Naphtalene	N.D.	N.D.	N.D.	N.D.	0.04	N.D.	1.40	N.D.	7.48	N.D.	N.D.	**30.79**
Acenaphtene	0.43	0.81	0.52	0.44	0.63	2.29	**5.25**	2.51	0.07	4.67	**8.02**	0.91
Fluorene	0.06	**0.86**	0.21	0.46	0.54	0.26	0.48	0.46	0.56	0.47	2.71	2.14
Phenanthrene	N.D.	N.D.	N.D.	N.D.	N.D.	2.63	N.D.	2.21	**3.43**	N.D.	7.25	23.79
Anthracene	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	0.21	N.D.	N.D.	4.14
Fluoranthene	N.D.	N.D.	N.D.	N.D.	0.55	N.D.	N.D.	N.D.	2.49	2.52	7.33	3.94
Pyrene	N.D.	N.D.	**1.30**	**0.85**	N.D.	N.D.	0.27	N.D.	1.29	N.D.	0.13	28.91
Benz(a)anthracene	0.13	0.32	0.11	0.13	0.21	0.22	0.28	**4.98**	N.D.	0.10	0.25	N.D.
Chrysene	**0.92**	0.07	0.08	N.D.	**2.40**	**3.34**	2.04	0.59	0.04	0.22	1.51	1.16
Benzo(b)fluoranthene	0.24	0.06	0.16	0.39	0.13	0.19	0.23	0.51	0.15	0.15	0.30	10.28
Benzo(k)fluoranthene	0.05	N.D.	0.07	0.30	0.05	0.04	0.04	0.01	0.14	0.08	0.16	6.11
Benzo(a)pyrene	N.D.	0.03	0.03	0.19	0.06	0.04	0.06	0.14	0.10	0.14	0.11	8.89
Dibenzo(a,h) anthracene	0.13	0.05	N.D.	0.68	0.11	N.D.	N.D.	0.09	0.10	0.14	N.D.	0.15
Benzo(g,h,i)perylene	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.	**6.73**	N.D.	2.10
Indeno(1,2,3-c,d)pyrene	N.D.	N.D.	N.D.	N.D.	0.15	N.D.	N.D.	N.D.	N.D.	3.07	N.D.	7.85
Total	1.96	2.20	2.48	3.43	4.87	9.01	10.06	11.50	16.07	18.28	27.78	**131.15**
Total carcinogenic	1.47	0.53	0.45	1.69	3.12	3.83	2.66	6.33	0.54	10.62	2.34	**36.54**

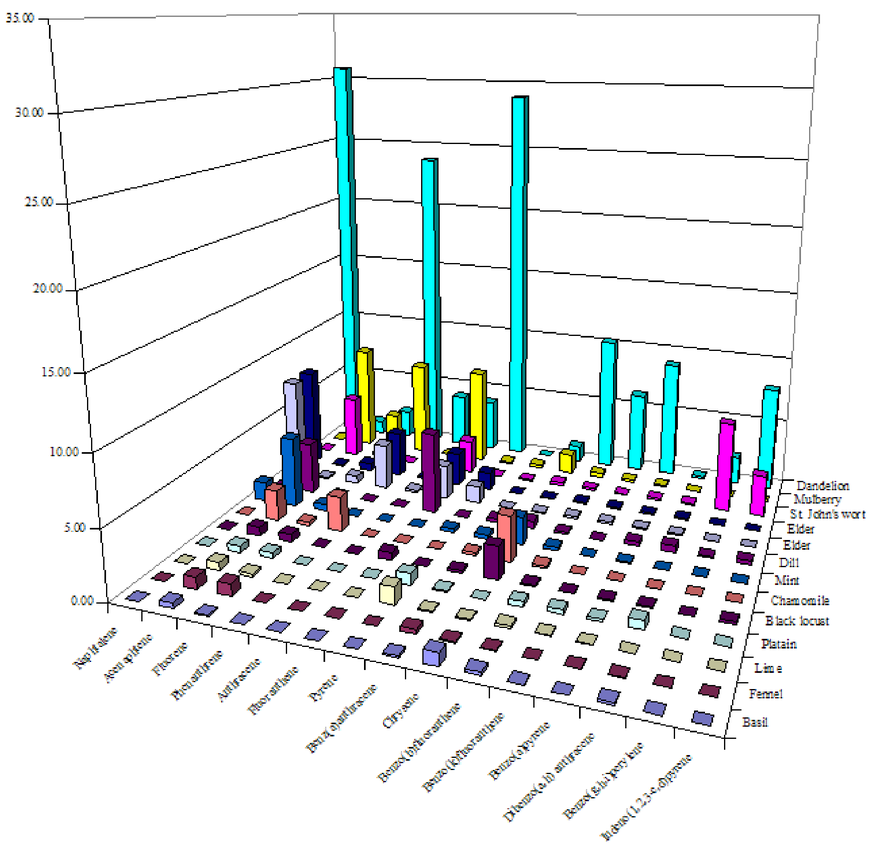


Figure 2. Loadings plot for PAHs

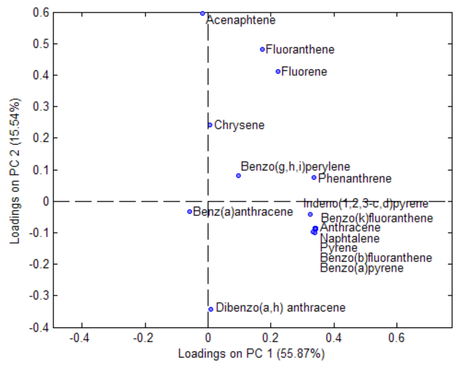


Figure 3. PCA scores

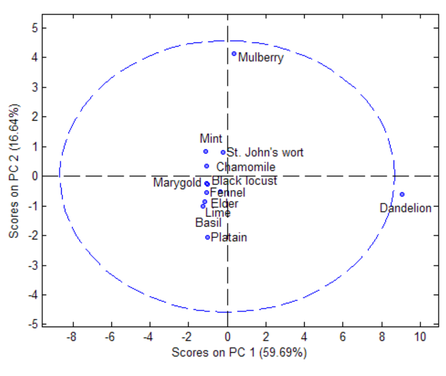
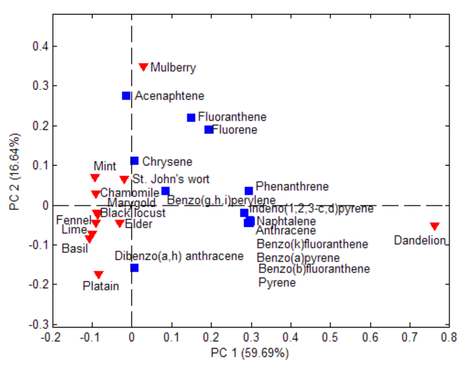


Figure 4. PCA biplot



Conclusions

- 15 PAHs were determined in 12 herbal teas' brands, each product showing a particular contamination pattern; PAHs' contamination is more important for ***herba*** (dandelion, mulberry, St.John's wort)> ***flos*** (common elder, black locust, lime) > ***semen*** (fennel) –with some exceptions.
- The content of carcinogenic PAHs' are higher than 50% from the total PAHs' content only for basil, black locust, marigold and St. John's wort.
- The obtained data are comparable with the published ones [1, 2], but showing much smaller PAHs' concentrations.

References

1. M.Ciecierska, M.Obiedziński (2009), The GC-MS analysis of polycyclic aromatic hydrocarbons content in selected fruit and herbal teas, Herba Polonica, 55(2), 18-24.

2. D.Lin, Y.Tu, L. Zhu (2005), Concentrations and health risk of polycyclic aromatic hydrocarbons in tea. Food and Chemical Toxicology 43, 41-48.