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FLUORIDE CONTENT AND BIOLOGICAL VALUE OF FLOWERS OF SOME CHAMOMILE (*MATRICARIA RECUTITA* L.) CULTIVARS

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Abstract

Chamomile (*Matricaria recutita* L.) is one of the most important medicinal plant in Europe. Phytotherapeutically useful are inflorescences – *Anthodium chamomillae*. Their main biologically active compound is an essential oil. Among other active agents there are polyphenols, carotenoids and ascorbic acid. The aim of the experiment was to compare fluoride content and biological value of four Polish chamomile cultivars: Promyk (2*n*), Mastar (2*n*), Złoty Łan (4*n*) and Dukat (4*n*). The experiment was carried out in 2010 in Poland. The chemical analyses included determination of the content of dry matter, total ash, total nitrogen, total protein, crude fibre, nitrates V, total and reducing sugars, saccharose, total chlorophyll, chlorophyll a and b, L-ascorbic acid, total carotenoids, total polyphenols, total flavonoids, essential oil, titratable acidity, antioxidant activity and fluoride concentration. It was proved that among the tested cultivars, the diploid cultivars of chamomile Promyk and Mastar were characterized by high biological value. They contain high amounts of total polyphenols, total flavonoids, dry matter, total ash, total nitrogen, total protein, crude fibre, total and reducing sugars and saccharose. However, the highest content of essential oil and fluoride content was determined in flower anthodia of the tetraploid cultivar Dukat.

Key words: chamomile cultivars, essential oil, polyphenols, flavonoids, antioxidant activity, fluoride.

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ZAWARTOŚĆ FLUORKÓW I WARTOŚĆ BIOLOGICZNA KOSZYCZKÓW WYBRANYCH ODMIAN RUMIANKU POSPOLITEGO (*MATRICARIA RECUTITA* L.)

Abstrakt

Rumianek pospolity (*Matricaria recutita* L.) jest jedną z najważniejszych roślin leczniczych w Europie, której głównym surowcem farmakologicznym są kwiatostany – koszyczki (*Anthodium chamomillae*). Ich głównym biologicznie czynnym składnikiem jest olejek eteryczny, a ponadto polifenole, karotenoidy i kwas askorbinowy. Celem badań było porównanie wartości biologicznej oraz zawartości fluorków w koszyczkach czterech polskich odmian rumianku: Promyk (2*n*), Mastar (2*n*), Złoty Łan (4*n*) i Dukat (4*n*). Analizy chemiczne obejmowały oznaczenie zawartości suchej masy, popiołu ogólnego, azotu ogólnego, białka ogółem, błonnika surowego, azotanów V, cukrów ogółem i redukujących, sacharozy, chlorofili ogółem, chlorofilu a i b, kwasu L-askorbinowego, karotenoidów ogółem, polifenoli ogółem, flawonoidów ogółem, olejku eterycznego. Oznaczono także kwasowość ogólną, aktywność antyoksydacyjną oraz koncentrację fluorków. Stwierdzono, że spośród badanych w doświadczeniu odmian wysoką wartością biologiczną odznaczały się diploidalne odmiany rumianku: Promyk i Mastar. Wykazano w nich dużą zawartość polifenoli ogółem, flawonoidów ogółem, a także suchej masy, popiołu ogólnego, azotu ogólnego, białka ogółem, błonnika surowego, cukrów ogółem i redukujących oraz sacharozy. Natomiast istotnie największą zawartość olejku eterycznego oraz fluorków stwierdzono w koszyczkach tetraploidalnej odmiany Dukat.

Słowa kluczowe: odmiany rumianku, olejek eteryczny, polifenole, flawonoidy, aktywność antyoksydacyjna, fluorki.

INTRODUCTION

Chamomile (*Matricaria recutita* L., syn. *Matricaria chamomilla* L. or *Chamomilla recutita* (L.) Rauschert, family *Asteraceae*) is one of the most important medicinal plant native to southern and eastern Europe, and adjoining Asian countries (SINGH et al. 2011). It is cultivated in Poland on 650--750 ha (SEIDLER-ŁOŻYKOWSKA 2007)

The medicinal part of the plant is the flower (*Matricariae flos*) and the medicinal substance is an essential oil (*Aetheroleum chamomillae*) (FP VIII, 2008). Over 120 constituents have been identified in chamomile (MCKAY, BLUMBERG 2006). Among its active agents we can find volatile and non-volatile components (GOSZTOLA et al. 2006, SALAMON 2009). The flowers contain 0.24-1.9% essential oil, of which the main components are a-bisabolol, bisabolol oxides, (E)-β-farnesene, chamazulene, and en-yn-dicycloethers (ORAV et al. 2001, RAAL et al. 2003, SALAMON 2009). Its blue colour is due to chamazulene, a secondary product of hydrodistillation and it is a result of proazulene (matricin) transformation (SVÁBNÉ 2000).

Among non-volatile components of chamomile flowers the most important are flavonoids, coumarins and pectine-like mucilages (GOSZTOLA et al. 2006). Flavonoids exhibit strong antioxidant and free radical scavenging activity, which also plays an important role in their anticarcinogenic effect

(Švehlíková, Repčák 2000). The most abundant chamomile flavonoid is apigenin, localized only in the white ligulate florets of the anthodium (Carle et al. 1993). Besides flavonoids and other phenolic compounds, there are other plant antioxidants, for example liposoluble carotenoids and tocopherols and water soluble vitamin C (Ivanova et al. 2005, Chrpová et al. 2010). Also, chlorophylls exhibit significant antioxidant activity. Antioxidant activity of main carotenoids and chlorophylls present in herbs decrease in the order: chlorophyll a > lutein > pheophytin a > chlorophyll b > β-carotene > pheophytin b (Suzuki, Shioi 2003).

Over the last twenty years, much information has accumulated on the role of fluoride in cellular respiratory processes and associated free radical reactions (Rzeuski et al. 1998). Fluoride is also known to be an inhibitor/activator of numerous enzymes. Although the relationship in human fluorosis between free radical generation, lipid peoxidation, and antioxidant systems has been investigated extensively, various studies have produced conflicting results (Chlubek 2003)

Chamomile is used as a component of herbal teas, single ingredient herbal tea or tisane as well as a valuable ingredient of many galenic preparations (tinctures, extracts) (Böttcher et al. 2001, Mckay, Blumberg 2006). Both the flowers and the essential oil have been reported to possess anti-inflammatory, antiseptic, antispasmodic, sedative and antiulcer properties (Mericli 1990, Sashidhara et al. 2006, Salamon 2009). They are used in the pharmaceutical, beverage and cosmetics industries (Reda et al. 2010).

The aim of the experiment was to compare fluoride content and biological value of four cultivars of chamomile flowers.

MATERIAL AND METHODS

The experiment was carried out in 2010 at the Horticultural Experiment Station in Dołuje, which belongs to the Department of Horticulture of West Pomeranian University of Technology in Szczecin. The laboratory part of the experiment was conducted in the Department of Biochemistry and in the Laboratory of Storage and Processing (Department of Horticulture) of West Pomeranian University of Technology in Szczecin. The research material consisted of fresh flowers of chamomile (*Matricaria recutita* L.) cultivars. Two diploid cultivars: Promyk and Mastar, and two tetraploid cultivars: Złoty Łan and Dukat were tested.

The experiment was established in randomized blocks with four replications. The experimental plot area was 1.44 m^2 (1.2 m × 1.2 m). The field was prepared with agronomic techniques proper for chamomile cultivation (Mordalski 2010). Mineral fertilization was quantified according to the results of chemical analysis of the soil. The seeds of chamomile were sown on

6th April, at a distance of 40 cm between rows. During the growing season, the crop management included mainly irrigation, weeding and soil cultivation.

The flower harvest was done at full-bloom stage, on 5th July. The chemical analyses of raw plant material included determination of the content of dry matter (drying at 105°C to constant weight), total ash (incineration of samples in 500°C), total protein (using factor 6.25 for amount of nitrogen determined by the method of Kjeldahl), crude fibre (KLEPACKA 1996), nitrates V (by the colorimetric method, ZALEWSKI 1971), total and reducing sugars, and saccharose (by the method of Luff-Schoorl), total chlorophyll, chlorophyll a and b (LICHTENTHALER, WELLBURN 1983), vitamin C as L-ascorbic acid (by the method of Tillmans), titratable acidity (KREŁOWSKA-KUŁAS 1993), total carotenoids (LICHTENTHALER, WELLBURN 1983), total polyphenols – in a spectrophotometer, using gallic acid as the reference and Folin-Ciocalteu reagent (SINGLETON, ROSSI 1965). Scavening effect of chamomile flowers on the DPPH-radical was determined according to the method of YEN and CHEN (1995). Raw homogenised material was diluted 400 times in 100% methanol. DPPH percent inhibition was calculated according to ROSSI et al. (2003). Some of the raw plant material was dried in a through-flow laboratory dryer set at 35°C. The concentration of flavonoid compounds (using the spectrophotometric method – FP VI, 2002) and essential oil content (with use of distillation of the sample together with water in Deryngs apparatus, FP VI, 2002) were determined in the dried and pulverized material.

Fluoride content was assayed according to the method of SZYMCZAK and GRAJETA (1982). Fluoride concentration was determined in the presence of TISAB III buffer using the potentiometric method with an Orion Research ion-selective electrode.

The results were statistically evaluated by the analysis of variance. Significance of the differences was tested by Tukey's test at p=0.05.

RESULTS AND DISCUSSION

Measurements of the content of chemical compounds of chamomile flowers showed significant variation between the tested cultivars.

The highest content of dry matter was determined for cultivar Promyk (23.80%), lower for Mastar (21.53%) and the least for Dukat and Złoty Łan (lower on average by 4.28% in comparison with Promyk and by 2.01% comparing to Mastar) – Table 1.

According to BÖTTCHER et al. (2001), the dry matter content of chamomile flowers varies between 18.5 and 23.5% according to the sowing date.

In the present experiment, the content of crude fibre, total ash, total nitrogen and total protein were also determined (Table 1). The highest con-

Table 1

Content of dry matter, crude fibre, total ash, total nitrogen and total protein of chamomile flower anthodia according to the cultivar

Cultivar	Dry matter (%)	Crude fibre (% f.w.)	Total ash (% f.w.)	Total nitrogen (% f.w.)	Total protein (% f.w.)
Złoty Łan	19.40*c*	1.93*b*	1.81*bc*	0.58*c*	3.60*c*
Promyk	23.80*a*	1.87*b*	2.23*a*	0.71*a*	4.44*a*
Dukat	19.64*c*	1.84*b*	1.76*c*	0.67*b*	4.16*b*
Mastar	21.53*b*	2.29*a*	2.00*b*	0.60*c*	3.72*c*
Mean	21.09	1.98	1.95	0.64	3.98

tent of crude fibre was noted for cv. Mastar, and lower for Złoty Łan, Promyk and Dukat (on average by 0.41% in comparison with Mastar).

The cultivar Promyk was characterized by the highest content of total ash, total nitrogen and total protein. Less total nitrogen and total protein occurred in cv. Dukat and the lowest amounts of these nutrients were in cv. Mastar and Złoty Łan. The cultivars Mastar and Złoty Łan were characterized by a lower total ash content than Promyk. However, there were no significant differences between Złoty Łan and Dukat, which had the lowest total ash content.

The highest content of total sugars, reducing sugars and saccharose was determined for cv. Mastar (Table 2). The cultivar Dukat was characterized by a significantly lower content of total sugars and reducing sugars than cv. Mastar (0.39 and 0.12% less, respectively). The lowest content of total sugars was found in cv. Złoty Łan, and reducing sugars – in the cultivars Promyk and Złoty Łan. In comparison with cv. Mastar, the cultivars Promyk, Dukat and Złoty Łan were characterized by a significantly lower content of saccharose.

Table 2

Content of total and reducing sugars, saccharose, nitrates V and titratable acidity of chamomile flower anthodia according to the cultivar

Cultivar	Sugars (% f.w.)			Titratable acidity (% citric acid f.w.)	Nitrates V (mg $NaNO_3$ kg^{-1} f.w.)
	total	reducing	sacchrose		
Złoty Łan	1.15*d*	0.94*c*	0.20*bc*	0.035*a*	364.54*a*
Promyk	1.33*c*	1.01*c*	0.31*b*	0.040*a*	210.16*a*
Dukat	1.46*b*	1.21*b*	0.25*b*	0.030*a*	359.44*a*
Mastar	1.85*a*	1.33*a*	0.50*a*	0.035*a*	475.55*a*
Mean	1.45	1.12	0.31	0.035	352.42

The yield of essential oil of *Matricaria recutita* depends on the plant genotype as well as the environmental conditions under which the plants are grown (LUTOMSKI, CZABAJSKA 1993, RAAL et al. 2003, SASHIDHARA et al. 2006, SALAMON 2007). GRGESINA et al. (1995) compared the essential oil content in different parts of chamomile plants (a diploid cultivar). The highest concentration of this compound was determined for yellow florets (0.49%), lower for flower heads (0.43%) and petals (0.28%) and the lowest for stems and leaves (0.08%). In the opinion of BÖTTCHER et al. (2001), the essential oil content of chamomile ranges from 0.56 up to 0.86 cm^3 per 100 g dry drug subject to the sowing date.

In the research by GOSZTOLA et al. (2006), the essential oil content differed significantly between examined chamomile populations, ranging from 0.2% (a wild type) up to 0.93% (a tetraploid cultivar). In our work, the highest content of essential oil (2.69 cm^3 100 g^{-1} d.m.) was determined for the tetraploid cultivar Dukat (Table 3). Less of this compound was determined in cv. Złoty Łan (4*n*) and Promyk (2*n*). However, there were no significant differences found between the cultivars Promyk and Mastar (2*n*), which contained the least essential oil.

Table 3

Content of essential oil, total polyphenols, total flavonoids and antioxidant activity of chamomile flower anthodia according to the cultivar

Cultivar	Essential oil (cm^3 100 g^{-1} d.m.)	Total polyphenols (mg 100 g^{-1} f.w.)	Total flavonoids (% d.m.)	Antioxidant activity (% DPPH)
Złoty Łan	1.36*b*	316.64*b*	0.50*a*	8.14*a*
Promyk	0.93b*c*	420.35*a*	0.48*a*	7.66*a*
Dukat	2.69*a*	308.78*b*	0.37*b*	6.97*a*
Mastar	0.78*c*	354.10*ab*	0.49*a*	8.62*a*
Mean	1.44	349.97	0.46	7.85

Significantly more polyphenols were determined in the cultivars Promyk and Mastar (Table 3). However, there were no significant differences found between the cultivars Mastar, Złoty Łan and Dukat.

The content of total flavonoids determined in the experiment varied from 0.37 to 0.50% d.m. The cultivars Złoty Łan, Mastar and Promyk were characterized by a significantly higher content of flavonoids. The lowest content of flavonoids was determined in cv. Dukat. GRGESINA et al. (1995) found on average 0.93% of flavonoids in chamomile flowers.

Several laboratories have examined the antioxidant potential of chamomile. In the study of REKKA et al. (1996), chamazulene poorly interacted with 1.1-diphenyl-2-picrylhydrazyl (DPPH). BUŘIČOVA and RÉBLOVÁ (2008) determined the antioxidant activity of water and ethanol extracts of 17 medicinal plants using the DPPH radical. Chamomile flowers had low antioxidant activity comparing to the other species, lower than for example sweet balm, thyme, oregano and mint herb, elder and nettle flowers. Contrary to this, MORAES-DE-SOUZA et al. (2008), YOO et al. (2008), AL-ISMAIL and ABURJAI (2004), classified chamomile flowers as a plant with high or medium antioxidant activity. In our study, the antioxidant activity of chamomile flowers reached on average 7.85% DPPH (moderate antioxidant activity) and there were no significant differences between the tested cultivars. The antioxidant activity of herbs and other plants mostly depends on the content of flavonoids, polyphenols, L-ascorbic acid and carotenoids.

Moreover, the chamomile cultivars compared in the experiment did not differ significantly in the content of L-ascorbic acid, total carotenoids, total chlorophylls, chlorophyll a, chlorophyll b and titratable acidity, whose average values for the tested cultivars were, respectively: 39.72 mg 100 g^{-1} f.w., 143.53 mg kg^{-1} f.w., 210.36 mg kg^{-1} f.w., 139.69 mg kg^{-1} f.w., 50.33 mg kg^{-1} f.w. and 0.035% citric acid f.w. (Tables 2, 4).

Table 4

Content of dry matter, crude fibre, total ash, total nitrogen and total protein of chamomile flower anthodia according to the cultivar

Cultivar	L-ascorbic acid (mg 100 g^{-1} f.w.)	Total carotenoids (mg kg^{-1} f.w.)	Chlorophyll (mg kg^{-1} f.w.)		
			total	a	b
Złoty Łan	37.92*a*	134.76*a*	212.00*a*	145.78*a*	48.50*a*
Promyk	38.88*a*	162.25*a*	223.26*a*	153.79*a*	47.84*a*
Dukat	41.76*a*	138.73*s*	192.19*a*	120.84*a*	48.99*a*
Mastar	40.32*a*	138.37*a*	213.98*a*	138.35*a*	55.97*a*
Mean	39.72	143.53	210.36	139.69	50.33

Beside medicinal components, herbs also contain nitrates and nitrites, which are well extractable and easily pass to water solutions (ÖZCAN, AKBULUT 2007, GRZESZCZUK, JADCZAK 2008).

According to FIGURA and PLUTA (2006), the content of nitrates (V) in chamomile flowers is on average 598.06 mg $NaNO_3$ kg^{-1} d.m. However, ÖZCAN and AKBULUT (2007) did not detect any nitrates (III, V) in chamomile flowers. The content of nitrates V determined in our experiment did not differ significantly between the cultivars and reached on average 352.42 mg $NaNO_3$ kg^{-1} f.w. (Table 2).

The highest fluoride content was determined in the cultivar Dukat, where it equalled 5.39 mg F kg^{-1} d.m. In comparison with cv. Dukat, the cultivars Promyk, Mastar and Złoty Łan were characterized by significantly less fluoride, ranging from 2.29 to 2.84 mg F kg^{-1} d.m. (Table 5).

Table 5

Fluoride content in chamomile flower anthodia according to the cultivar

Cultivar	Fluoride content (mg F kg^{-1} d.m.)
Złoty Łan	2.29*c*
Promyk	2.26*c*
Dukat	5.39*a*
Mastar	2.84*b*
Mean	3.19

There are few studies on the fluoride content in herbal plants, but Ekmeli-Alturfan et. al. (2009) showed that the fluoride content in infusions from chamomile and another herbal plants was 0.03 mg F dm^{-3}. Similar findings are reported by Chan and Koh (1996)

CONCLUSIONS

1. Among chamomile cultivars compared in the experiment, cv. Promyk and Mastar were characterized by higher biological value. Flower anthodia of cv. Promyk contained significantly more dry matter, total ash, total nitrogen and total protein, while those of cv. Mastar were richer in crude fibre, total and reducing sugars, and saccharose. Moreover, both cultivars were characterized by a high content of total polyphenols and total flavonoids.

2. The highest content of the essential oil and fluoride was determined in flower-heads of the cultivar Dukat.

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