

SELENIUM IN SOILS OF THE DANUBE DELTA NORTH-WESTERN PART

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Abstract. Soils from the dyked precincts Sireasa and Pardina of the Danube Delta were analyzed, belonging to the Calcaric Fluvisol, Calcaric Gleysol, Mollic Calcaric Gleysol, Mollic Fluvisol, Calcaro-Calcaric Kastanozem, and Calcaro-Calcaric Chernozem⁷ types. The soils are slightly alkaline, with a moderate carbonates content, low up to average humus and total nitrogen ones, and diverse, from very low to very high, of mobile phosphorus and potassium. Some of them have a salinization level up to 688 mg soluble salts per 100 g soil. The mobile and total selenium contents are high, superior to the average general content of the World's soils and to the contents of the South-Eastern Romanian Plain and Central and South Dobrogea soils. In fact, they are the highest values registered so far in Romania's soils. In general, the soils within the built-up area have higher values than those of the outside built-over one both for selenium and other chemical elements. Direct proportionality relations were established between the total selenium content and some of the agrochemical soil properties (indirect with the pH), all of them statistically ensured, and also between the total and mobile selenium contents, on one hand, and the micro elements (heavy metals) contents on the other hand. The ensuring degree of the selenium's correlations with some heavy metals increases by depth which shows the geogenic origin of the chemical elements in the Delta soils. Although the Danube Delta is a deprived area the selenium content of the analyzed soils is high without reaching, though, toxicity levels.

Introduction

Selenium is a micro element with numerous qualities in animal and human nutrition, with an anti-infections and anti-oxidant effect as a component of the glutation-peroxidase enzyme, and anti-tumor effect (Deélstra et al., 1982; Gissel-

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Nielsen et al., 1985). Its physiological and biochemical role in plant nutrition was also outlined (Läuchli, 1993; Turakainen et al., 2005), and yield increases were obtained when selenium was administered on the seed, in soil, or on plant (Lăcătușu et al., 2002).

Selenium abundance in the environment components is low. Thus, the average content oscillates between 50 and 90 $\mu\text{g}\cdot\text{kg}^{-1}$ in the lithosphere, between less than 100 and 2,000 $\mu\text{g}\cdot\text{kg}^{-1}$ in the pedosphere, from less than 50 up to 15,000 $\mu\text{g}\cdot\text{kg}^{-1}$ in the biosphere, and around 0.2 $\mu\text{g}\cdot\text{l}^{-1}$ in the hydrosphere (Kabata-Pendias and Pendias, 2001).

Selenium abundance in soil depends on a series of chemical and physical factors such as reaction, organic matter and macro and micro elements contents, the parent material nature. The content interval of the total selenium content in the upper horizon of the World's soils is 5-3,500 $\mu\text{g}\cdot\text{kg}^{-1}$, with an average value of $383 \pm 255 \mu\text{g}\cdot\text{kg}^{-1}$ (Kabata-Pendias and Pendias, 2001). The extreme values of the content interval belong to selenium deficiency, respectively toxicity areas. Selenium deficiency leads to the occurrence of some diseases in living beings such as: ovine myodystrophy, hepatic necrosis with swine, white muscle disease with horses, exudative diathesis with poultry, and the excess determines the alkaline disease occurrence with animals and people (Gissel-Nielsen et al., 1985). Selenium deficiency with people is implied in a series of Cardiovascular and Digestive Systems diseases and in many tumor diseases. Its major role for human health lies in the anti-oxidant effect of its compounds (Reilly, 2006).

The fact is known that large areas of the North (Finland, Sweden, Norway; Hartikainen, 2005), Central (Germany; Hartfiel and Bahners, 1988), South-Eastern European countries (Serbia; Maksimovic, 1992), and from Russia (Ermakov, 1992) are affected by the selenium deficiency. Romania also lies in a World's area with deficient selenium contents registered with animals and even with people. Thus, Salanțiu, even since 1970, highlighted the selenium deficiency in calves, lambs, sucking pigs, and young buffalos in large areas of the Transylvania Basin. More recently, Serdaru and Giurgiu (2007) analyzed 1,548 fodder samples, 1,175 cattle blood serum samples, 1,030 sheep blood serum samples, and 600 human blood serum samples collected from the Ardeal area and concluded that only 3.7% of the fodder samples, 5.0% of the cattle blood serum samples, none of the sheep blood serum samples, and only 3.3% of the human blood serum samples have normal contents, while the differences belong to the deficiency domain. Alike, Serdaru et al. (2003) analyzed 185 fodder samples from 41 Dobrogea localities and concluded that only 6.5% of them belong to the normal content domain, and the difference belongs to the deficiency domain. This situation required the introduction of selenium in the animal feed premixes. The deficiency level mostly occurs because of some low soil selenium contents.

Among the first data regarding total selenium content in Romania's soils there are those concerning the Oriental Carpathians Mountain soils and some river sediments (Ababi and Dumitrescu, 1973; Lăcătușu and Ghelase, 1992). The authors found $640 \mu\text{g}\cdot\text{kg}^{-1}$, respectively $380 \mu\text{g}\cdot\text{kg}^{-1}$ average values, the latter in hematurigenous areas. Determinations carried out in Dobrogea soils highlighted total selenium concentrations between 211 and $585 \mu\text{g}\cdot\text{kg}^{-1}$, with an average value of $314 \mu\text{g}\cdot\text{kg}^{-1}$, and mobile selenium concentrations, soluble in ammonium acetate lactate solution (AL) at $\text{pH} = 3.7$, between 0.9 and $74 \mu\text{g}\cdot\text{kg}^{-1}$, with an average value of $10 \mu\text{g}\cdot\text{kg}^{-1}$ (Lăcătușu et al., 2009, 2010 a,b). In the Central-Eastern part of Dobrogea, in the Sibioara area, where cases of ovine myodystrophy have been registered, the average total and mobile selenium contents, soluble in AL, were $140 \mu\text{g}\cdot\text{kg}^{-1}$, respectively $5 \mu\text{g}\cdot\text{kg}^{-1}$ (Lăcătușu et al., 2002). Total selenium determinations carried out in samples of the upper horizon of the soils from the South-Eastern part of the Romanian Plain, predominantly Chernozems, highlighted higher values than those of the Dobrogea soils, with 64%, on an average (Lăcătușu et al., 2010). Unlike these ones, in the Solonchaks and Solonetz of the Buzău and Călmățui Valleys Lăcătușu et al. (2011) determined total selenium contents with values around $800 \mu\text{g}\cdot\text{kg}^{-1}$, twice as much as the average of the selenium contents from many non-halomorphic soils of the World and three up to five times more than the total selenium content of the upper horizon of the South-Eastern Romanian Plain or Dobrogea soils.

Continuing the researches regarding selenium abundance in the Romania's soils the present paper highlights this chemical element's contents in some of the most recent soils of the Country, namely in the Danube Delta North-Western part.

1. Materials and methods

The researches had an expeditionary character, and soil samples were collected by the 0-20 and 20-40 cm depths, from the Danube Delta North-Western part, more precisely from the Sireasa and Pardina dyked areas (Figure 1). 60 samples were collected from outside the built-over area and 16 from within the built-up one. The latter from the localities: Tudor Vladimirescu, Ceatalchioi, Pardina and Chilia Veche.

The soil samples were analyzed in the laboratory from the general chemical characteristics (pH, humus, total nitrogen, mobile forms of phosphorus and potassium, soluble salts, carbonates, total an mobile micro elements forms) and of the total an mobile selenium contents point of view. The general chemical characteristics were determined by standardized (STAS and ISO) methods: pH – potentiometrically, with double glass and calomel electrode, in aqueous solution with the soil:water ratio 1:5; humus content by the Walkley-Black method

2. Results and discussions

2.1. The investigated soils and their general characteristics

The 38 investigated soils belong to the Calcaric Fluvisol (12), Calcaric Gleysol (9), Mollic Calcaric Gleysol (7), Mollic Fluvisol (7), Calcaro-Calcic Kastanozem (2), and Calcaro-Calcic Chernozem (1) types. They have a predominantly loamy, clayey-loamy up to sandy-loamy texture, sometimes clayey, especially the Gleysols. The statistical parameters of the main chemical characteristic of the dominant soils are presented in Table 1.

Tab. 1 – Statistical parameters of the main chemical characteristics of the soils from the Danube Delta North-Western part

Statistical parameter	pH _{H₂O}	CaCO ₃	Humus	Total N	P _{AL}	K _{AL}
		%			mg·kg ⁻¹	
Calcaric (mollic) gleyic Fluvisol, n = 42						
Xmin	7,3	0,6	0,5	0,025	3	60
Xmax	8,5	11,3	6,3	0,584	23	656
X	7,9	7,2	2,9	0,144	14	200
σ	0,2	2,5	1,7	0,119	10	118
cv (%)	35	35	58	83	76	59
Xg	7,8	7,0	2,8	0,138	12	189
Calcaric (mollic) fluvic Gleysol, n = 30						
Xmin	7,4	1,0	1,7	0,097	5	64
Xmax	8,1	10,5	7,9	0,525	235	620
X	7,8	6,3	4	0,23	39	224
σ	0,2	2,3	1,8	0,117	53	137
cv (%)	3	36	44	51	136	61
Xg	7,7	6,1	3,7	0,224	36	220

The soils have alkaline reaction, no exceptions, and belong to the slightly alkaline domain, with pH (measured in aqueous solution) values ranging from 7.3 to 8.5. The calcium carbonate (CaCO₃) content is medium, almost no exception, with values ranging between 4.2 and 11.3%. The exception is represented by eight values ranging from 1.0 to 1.9%. The humus, assessed depending on the texture, has a large values domain, significant for very low, low, and medium contents out of which low and medium contents are equally dominant. The total nitrogen contents vary alike, in the low and medium values zone. As regards the mobile phosphorus and potassium forms supply, soluble in the ammonium acetate lactate solution at pH 3.7, they belong to large values intervals, between 3 and 7 mg·kg⁻¹

in the outside built-over areas and between 9 and 235 mg·kg⁻¹ in the soils within the built-up area for phosphorus and between 6 and 656 mg·kg⁻¹ in the outside built-over areas and between 92 and 620 mg·kg⁻¹ in the soils within the built-up area for potassium. Practically, these values cover the whole supply domains both for phosphorus and potassium.

Some of the analyzed soils (6) contain soluble salts beyond the 100 mg per 100 g soil limit, considered to be the threshold between non-salinized soils and the saline ones. The latter's salinization level reaches 688 mg/100 g soil. The dominant salts are: calcium sulphate (CaSO₄), with values up to 66.1%, magnesium sulphate (MgSO₄), with values up to 15.8%, and sodium sulphate (Na₂SO₄), with values up to 24.7%. Calcium bicarbonate (Ca(HCO₃)₂), with values up to 23.7%, magnesium bicarbonate (Mg(HCO₃)₂), with values up to 9.6%, and different proportions of sodium, potassium, calcium, and magnesium chloride, reaching maximum values of 19.6; 50.8; 13.5; respectively 6.1% occur secondarily. The mentioned maximum values belong to different samples.

Therefore the Danube Delta analyzed soils, mainly located in the Sireasa and Pardina dyked areas, are made up of Fluvisols and Gleysols, both calcareous, with a slightly alkaline reaction, with a medium carbonates content, with low and medium humus and total nitrogen contents, and diverse levels of mobile phosphorus and potassium supply, from very low to very high. Some of the soils are salinized, reaching up to 688 mg/100 g soil. The soluble salts consist mainly of sulphates, mostly calcium, and bicarbonates and chlorides follow in a decreasing order.

2.2. Total and mobile selenium contents

The statistical parameters of the total selenium content in the analyzed soils highlight a value interval between 0.307 and 1.776 mg·kg⁻¹, with medium values of 0.600 mg·kg⁻¹ for the arithmetic mean (\bar{x}) and 0.576 mg·kg⁻¹ for the geometric mean (x_g), median (Me), and module (Mo). Separately by the two geometric horizons (0-20 and 20-40 cm) one can notice that the first one contains more selenium than the underlying one (Table 2).

If these values are compared to the average total selenium contents in the World's soils (from Kabata-Pendias and Pendias, 2001), of $0,383 \pm 0,255$ mg·kg⁻¹, one can notice that the Danube Delta analyzed soils contain 1.6 times more selenium in the 0-40 cm layer and 1.7 times more in the 0-20 cm layer.

As compared to the Romania soils from the South-Eastern Romanian Plain and Central and Southern Dobrogea (Lăcătușu et al., 2010), the Danube Delta analyzed soils contain, on an average, 2.5, respectively 4.2 times more total selenium. The phenomenon can be easily understood if the fact is taken into account that the Danube Delta soils are formed by the Danube alluvia which

consist, in their turn, of diverse natural materials transported by the river in its course and of anthropic materials discharged in its waters by the riverside countries' inhabitants.

Tab. 2 – Statistical parameters of the total selenium content ($\text{mg}\cdot\text{kg}^{-1}$)

Statistical parameter	Depth, cm		
	0-40	0-20	20-40
n	78	39	39
x_{\min}	0,307	0,377	0,307
x_{\max}	1,776	1,776	0,980
\bar{x}	0,600	0,648	0,552
σ	0,201	0,239	0,141
x_g	0,576	0,619	0,536
cv (%)	34	37	26
Me	0,576	0,584	0,552
Mo	0,575	0,538	0,552

Analyzing and comparing the statistical parameters of the mobile selenium content (Table 3) with the average mobile selenium content in the South-Eastern Romanian Plain and Central and Southern Dobrogea (Lăcătușu et al., 2010), the same conclusion is reached: the analyzed Delta soils contain more mobile selenium, 1.7 times, than those of the South-Eastern Romanian Plain and 6 times as compared to the Central and Southern Dobrogea ones. The phenomenon's explanation is the one mentioned above.

2.3 Selenium correlations in the analyzed Delta soils

Proportionality relations were established between the total selenium content of the Delta analyzed soils, on the 0-40 cm depth, and the main soil agrochemical characteristics (pH, humus and total nitrogen content, mobile phosphorus and potassium, soluble in the ammonium acetate lactate solution at $\text{pH} = 7$, supply level), entirely statistically ensured (figures 2-6).

The total selenium content – reaction (pH) correlation is reverse, and the correlation coefficient has a negative value ($r = -0,465^{**}$). The real pH domain in which the correlation is significant is 7.2-8.5. Therefore, in the alkaline reaction domain the total selenium content decreases as the pH value increases, at least for the mentioned reaction interval.

The other total selenium correlations, with humus, total nitrogen, and mobile phosphorus and potassium forms, are direct and have high, distinctly significant values both for the correlation coefficient (r) and ratio (η). The correlations with

humus and total nitrogen stand out as being tighter as compared to those with mobile phosphorus and potassium. In the latter case most of the values are distributed in smaller content intervals, otherwise normal for such soils.

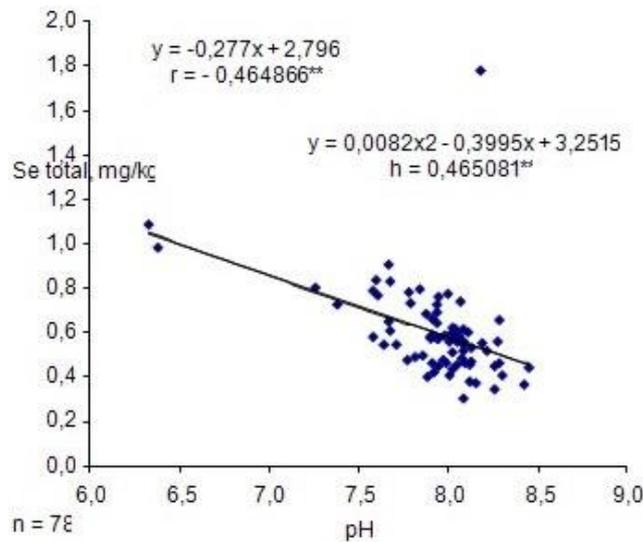


Fig. 2 – Correlation between the total selenium content and soil reaction (pH) in the analyzed soils, on the 0-40 cm depth

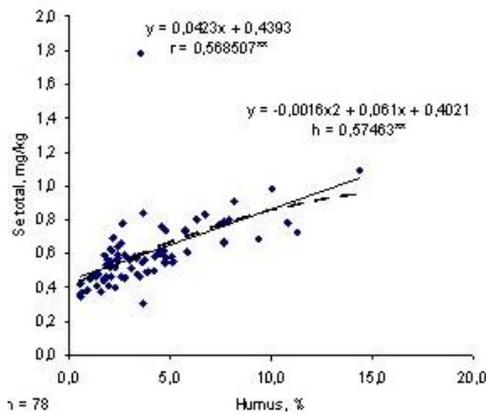


Fig. 3 – Correlation between the total selenium and the humus contents in the analyzed soils, on the 0-40 cm depth

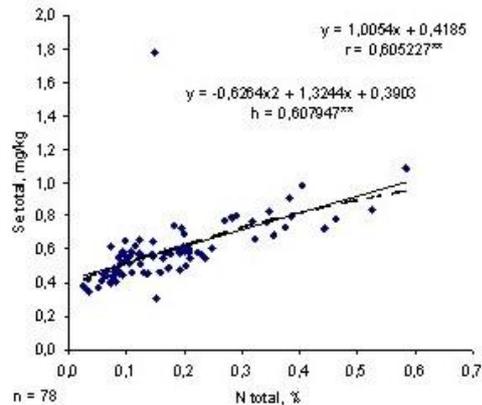


Fig. 4 – Correlation between the total selenium and the total nitrogen contents in the analyzed soils, on the 0-40 cm depth

Significant correlation ratios and coefficients were also computed between the total and mobile selenium contents, on one hand, and the micro elements (heavy metals) ones, on the other hand (Tables 4 and 5). High values are noticed, of over 0.500, both for the correlation ratios (η) and coefficients (r), most of them distinctly significant, except for the correlation ratios and coefficients of the manganese and cadmium (Table 4) which are insignificant in the case of the total selenium correlations.

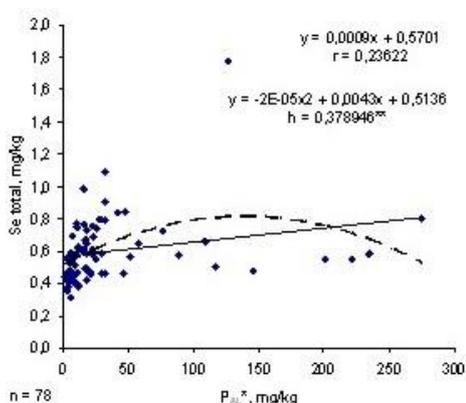


Fig. 5 – Correlation between the total selenium content and the mobile phosphorus one, soluble in ammonium acetate lactate at pH = 7 in the analyzed soils, on the 0-40 cm depth

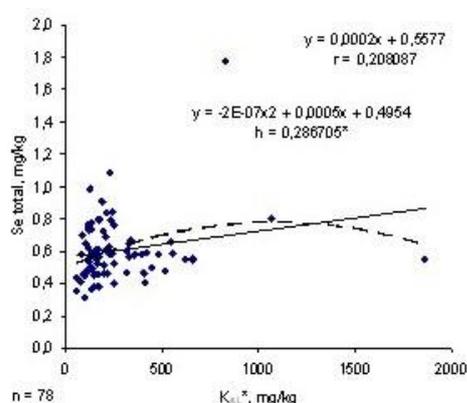


Fig. 6 – Correlation between the total selenium content and the mobile potassium one, soluble in ammonium acetate lactate at pH = 7 in the analyzed soils, on the 0-40 cm depth

Tab. 4 – Correlation ratios (r) and coefficients (η) of the total micro elements (heavy metals) and selenium contents in some soils of the Danube Delta

	Zn	Cu	Fe	Mn	Pb	Ni	Co	Cr	Cd
η	0,566**	0,745**	0,721**	0,279	0,389*	0,693**	0,467**	0,681**	0,296
r	0,525**	0,739**	0,721**	0,100	0,371*	0,719**	0,466**	0,681**	0,160

The correlations of the mobile selenium with these chemical elements (Table 5) have much lower values of the correlation ratios and coefficients and are insignificant for some chemical elements (copper, iron, manganese, chromium on the 0-20 cm depth and zinc, copper, iron, manganese, nickel on the 20-40 cm depth). Obvious and distinctly significant are the mobile selenium correlations with the heavy metals (in the true meaning of the word) lead and cadmium both on 0-20 and 20-40 cm depth. It is clearly

Tab. 5 – Correlation ratios (r) and coefficients (η) of the total micro elements (heavy metals) and mobile selenium contents in some soils of the Danube Delta

0-20 cm									
	Zn	Cu	Fe	Mn	Pb	Ni	Co	Cr	Cd
η	0,334*	0,259	0,120	0,164	0,395**	0,317*	0,285*	0,406	0,557**
r	0,308*	0,169	0,114	0,163	0,314**	0,204	0,276*	0,401	0,463**
20-40 cm									
	Zn	Cu	Fe	Mn	Pb	Ni	Co	Cr	Cd
η	0,311	0,322	0,317	0,228	0,472*	0,208	0,553**	0,417*	0,603**
r	0,306	0,321	0,258	0,199	0,456**	0,191	0,551**	0,384*	0,533**

noticed that the correlation intensity is stronger at the 20-40 cm depth for the statistically ensured correlations. This mainly certifies the geogenic origin of the chemical elements, including selenium, in the analyzed soils.

Conclusions

The analyzed Delta soils of the Sireasa and Pardina dyked areas belong to the following types: Calcaric Fluvisol, Calcaric Gleyosol, Mollic Calcaric Gleyosol, Mollic Fluvisol, Calcaro-Calcic Kastanozem, and Calcaro-Calcic Chernozem. They are slightly alkaline, have a moderate carbonate content, low up to medium humus and total nitrogen contents, and diverse, from very low to very high, of mobile phosphorus and potassium.

Some of the soils have a salinization level up to 688 mg soluble salts per 100 g soil. The salts are predominantly calcium, magnesium, and sodium sulphates.

The analyzed Delta soils have high mobile and total selenium contents, superior to the general average content of the World's soils and to the South-Eastern Romanian Plain and Central and South Dobrogea soils contents.

The higher selenium values were registered out of the Romania's soils analyzed so far.

Generally, in the soils within the built-up area higher values were registered than in the outside built-over areas ones both for selenium and other chemical elements.

Between the total selenium content and some of the soils agrochemical features direct proportionality relations (reverse with the pH) were established, entirely statistically ensured.

Between the total and mobile selenium contents, on one hand, and micro elements (heavy metals) contents, on the other hand, direct proportionality relations were established, mostly statistically ensured.

The increase of the insurance degree of the selenium correlations with some micro elements (heavy metals) with the soil profile depth certifies the geogenic origin of the chemical elements in the Delta soils.

Although the Danube Delta is a deprived area the selenium content of the analyzed soils is high without reaching, though, toxicity levels.

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