

Conditions of Desalinization Process of Soils Flooded with Copper Mining Wastewater

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Abstract

In areas where pipelines transporting technological water from drainage of the KGHM "Polska Miedź" S.A. copper mines are installed there are occasional instances of pipe failure with the resultant flooding of the soil with saline water. Based on studies undertaken immediately after the occurrence of pipeline failures and soil flooding, as well as at a later period, an attempt was made to describe the course of natural desalinization of soil that takes place as a result of percolation of precipitation. Examination and description was made for soils from 5 locations at which, in the years 1995-2008, failures of the above type took place. The study included determination of the soil salinity and of the composition of the cations exchange capacity of the flooded soils. A decrease in soil salinity was observed, as well as a decrease in the level of the exchangeable sodium percentage, taking place under the effect of precipitation water percolation through the soil. It was stated that the rate of decrease in soil salinity is in relation to the parameter characterizing soil texture. The salt leaching process proceeds notably faster in soils with a coarse texture compared to finely textured soils, and at the same time that process is faster in the surface horizons of the soil while being delayed in the sub-surface horizons. At the same time, a decrease was observed in the scatter of salinity and ESP results upon return to the state of equilibrium.

Keywords: soil salinity, cations exchange capacity, exchangeable sodium percentage (ESP), technological water

Introduction

Soil salinity is one of the main environmental factors that limits yields in agriculture [1-3]. Under moderate climate conditions, on a large space scale, the process of salinization of soils may result from improper irrigation [4-6], while locally it may be caused by the effect of certain industrial and infrastructure installations [7-9].

Local salinization processes of this type have been observed for years in areas of installed pipelines transporting saline water from the KGHM "Polska Miedź" S.A. copper mines to post-flotation sediment reservoirs [10].

Occasional failures of the pipelines cause soil flooding and subsequent percolation of the saline water into the soil (Table 1). Higher precipitation than evaporation, combined with percolation of precipitation waters to ground waters, result in spontaneous desalinization of the flooded soils.

In the literature of the subject, apart from numerous theoretical models [11-15], one can find many attempts at empirical description of the phenomenon of soil desalinization due to water percolation. The empirical approach, in spite of its limitations, is characterized by greater simplicity and practicality [16]. One of the first attempts at such an empirical description was that by Reeve [17], who presented an equation relating the relative content of salt in soil (in relation to the initial salt content) to the amount of water

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percolating through the soil. A similar equation taking into account the level of soil salinity under the conditions of equilibrium was presented by Leffelaar and Sharma [18]. The run of the process of desalinization resulting from percolation of natural precipitation or irrigation water percolation, methods of augmentation of the process and additional effects related with the leaching of nitrogen and potassium from the soil, remain within the field of interest of numerous researchers [19-21].

The aim of the study presented here was investigation of the soil desalinization process intensity under natural conditions. Changes in the physicochemical properties of the soil and factors affecting the desalinization process were also investigated.

Materials and Methods

The study was conducted in areas flooded with saline technological water as a result of failures of transport pipelines. Five locations were selected for the study, for which analyses were made immediately after pipeline failure, and then repeated once or twice after a certain period of time. The particular locations included in the study are referred to by the names of the nearest locations (villages) (Fig. 1).

At the particular locations, samples were taken on the sampling dates at the same points (with accuracy of a few meters). At all sampling points the groundwater table was

located at depths greater than 1.5 m relative to the ground surface.

Samples for analyses were taken from a depth of 0-30 cm, which always coincided with the accumulation horizon, and from depths >30 cm, beyond the range of the accumulation horizon. For chemical analyses the samples were dried, ground, and sifted through a sieve with mesh <1 mm.

In the soil material sampled the following analyses of chemical and physicochemical properties were made:

- electrical conductivity of soil was measured by conductivity meter (Mettler Toledo Seven Multi 2009) with the soil-water ratio of 1:5, according to ISO 11265 standard (1994); conversion to $\text{mg}\cdot\text{kg}^{-1}$ of soluble salts was made based on the calibration curve plotted for potassium chloride;
- pH was measured in a 1 M solution of sodium chloride by glass electrode pH-meter;
- hydrolytic acidity was determined with the use of calcium acetate, followed by titration with sodium hydroxide in the presence of a few drops of phenolphthalein (Lab – mate burette – High Tech Lab);
- organic carbon content was determined using the wet oxidation method with external heating, followed by titration with ferrous ammonium sulfate;
- exchangeable cations were measured with an extract of 2 N ammonium acetate by flame photometer for Ca, K, and Na, and with atomic absorption spectrophotometer Philips 9100X for Mg; for samples where salt content

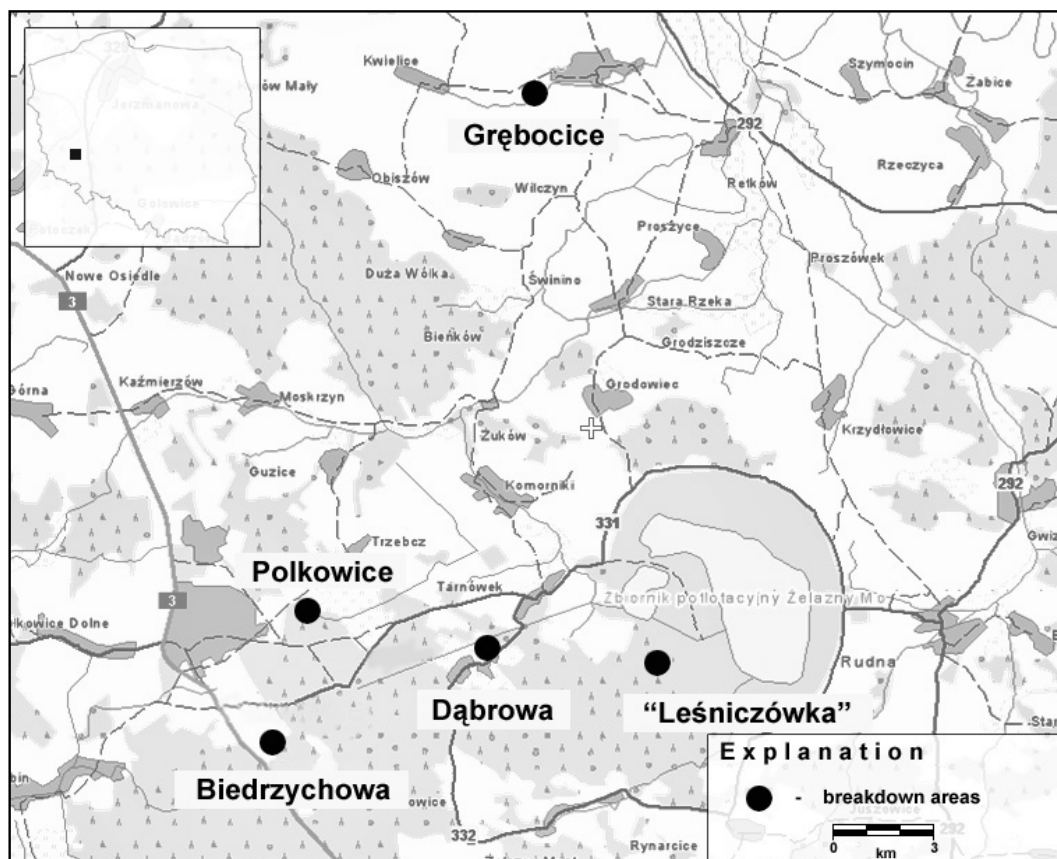


Fig. 1. Location of examined objects.

exceeded 2,000 mg·kg⁻¹ the measurements were followed by leaching of the soluble forms with 40% ethanol;

- soil texture – by sieving and sedimentation method with the use of hydrometer acc. to standard ISO/FDIS 11277 (2005);
- contents of Zn, Cu, Pb, Cr, Ni, and Cd in the technological water were measured with the use of atomic adsorption spectrometry.

Results and Discussion

According to the soil classifications of the Polish Soil Science Society, all examined soils belong to one soil division, autogenic soils. They represent three different soil subtypes. In the “Grębocice” area there are dystric cambisols. The haplic podzols and haplic arenosols are found in the remaining locations.

In terms of texture, the locations studied were characterized by certain differences. Soils found in the “Biedrzychowa” location contained from 3 to 11% of fractions < 0.02 mm and from 0 to 4% of fractions < 0.002 mm. The soils, however, were characterized by a highly varied content of fractions > 1.0 mm, at even up to 34% (Fig. 2).

Soils from the “Dąbrowa” location were medium-textured. At the surface horizons of the soils the content of fractions < 0.02 mm varied from 14 to 24%, and the content of fractions < 0.002 mm varied from 2 to 5%. At depths of 30-95 cm there were soils with coarser texture. Those contained from 4 to 15% of fractions < 0.02 mm, including from 1 to 4% of fractions < 0.002 mm. At greater depth, > 95 cm, there were soils containing ca. 30% of fractions < 0.02 mm, including 15 % of fractions < 0.002 mm. All of the soils under study had a fraction > 1 mm content of up to 10% (Fig. 2).

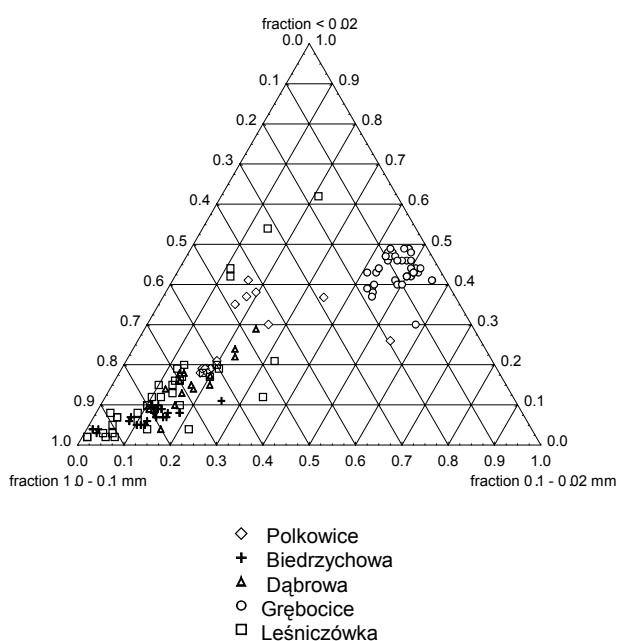


Fig. 2. Texture of examined soils.

Table 1. Average concentrations of selected substances in the wastewater transported by the pipelines from copper mine.

Item	Parameter	Unit	“Biedrzychowa”	“Dąbrowa”
2	Soluble substances	mg·kg ⁻¹	3,050	15,300
3	Chlorides		528	6,500
4	Sulphates		1,310	2,600
6	Calcium		-	980
7	Magnesium		-	230
8	Sodium		-	3,800
9	Potassium		-	110
12	Chromium		0.0083	-
13	Copper		0.221	0.16
14	Zinc		0.148	0.123
15	Lead		0.02	0.158
16	Nickel		0.045	0.27
17	Cadmium		0.0009	-

The texture of the soils from the “Polkowice” location was also profile-varied. The surface horizons of those soils containing 18-19% of fractions < 0.02 mm and from 4 to 6% of fractions < 0.002 mm. At deeper levels there were soils containing from 28 to 41% of fractions < 0.02 mm, and from 19 to 25% of fractions < 0.002 mm. The soils had only a slight content (up to 3.5%) of fractions > 1 mm.

The texture of soils from the “Leśniczówka” location were also differentiated. In the surface horizons of those soils the content of fractions < 0.02 mm was from 5 to 19%, and fractions < 0.002 mm from 0 to 5%. The content of fractions > 1 mm fell within the range from 0 to 20%. At deeper levels soils showed considerable differences of texture (Fig. 2).

Soils from the “Grębocice” location were the most finely textured. The texture of these soils was also the most uniform. All samples studied (with one exception) contained from 37 to 49% of particles < 0.02 mm and from 7 to 22% of fraction < 0.002 mm. Those soils had a low content of fractions > 1.0 mm, up to 3%.

Accumulation Horizons (Epipedons)

Acidity measured in the 1 M sodium chloride solution for epipedons, during the first cycle of measurement (within a short time following pipeline failure) varied from pH 3.7 to pH 7.9, with median 6.8. During the last cycle of measurement the pH varied from 4.4 to 7.6 with median 6.6. The values of median for individual locations hadn't shown systematic changes.

Organic carbon content during the first cycle of measurement varies from 0.39% to 3.57%, with the average

Table 2. Information concerning the pipeline failures in the area of investigations.

Name of location	Time of pipeline failure	Dates of sampling			amount of spilled water [m ³]	Flooded area [m ²]	Type of land use
		I	II	III			
Dąbrowa	06.1995	06.1995	06.1997	06.1998	unknown	60,000	arable lands
Precipitation between sampling dates [mm]		1093.7		723.1			
Grębocice	12.2003	09.2004	05.2006	04.2008	unknown	65,000	arable lands
Precipitation between sampling dates [mm]		770.4		1,042.4			
Biedrzychowa	11.2008	12.2008	04.2009	-	800	20,000	house gardens, lawns
Precipitation between sampling dates [mm]		117.3					
Leśniczówka	10.2003	12.2003	02.2004	-	unknown	3,000	young stand, forest
Precipitations between sampling dates [mm]		145.4					
Polkowice	09.1998	10.1998	04.1999	-	24	6,000	allotment gardens
Precipitations between sampling dates [mm]		211.5					

value 1.49%. During the last cycle of measurement this parameter was in the range 0.45% to 3.57%, with average value 1.62%.

Within a short time following pipeline failure we observed notable differences of soil samples, both in terms of their salinity and of the exchangeable sodium percentage (ESP). With the passage of time, a decrease was observed in

soil salinity and, to a lesser extent, in the exchangeable sodium percentage (ESP). The difference in soil salinity in the locations immediately after pipeline failure and after the passage of a specific period of time was statistically significant at the level of $\alpha < 0.001$ for the locations "Biedrzychowa" and "Grębocice", and at the level of $\alpha < 0.05$ for the locations "Polkowice", "Dąbrowa" and "Leśniczówka." At the same

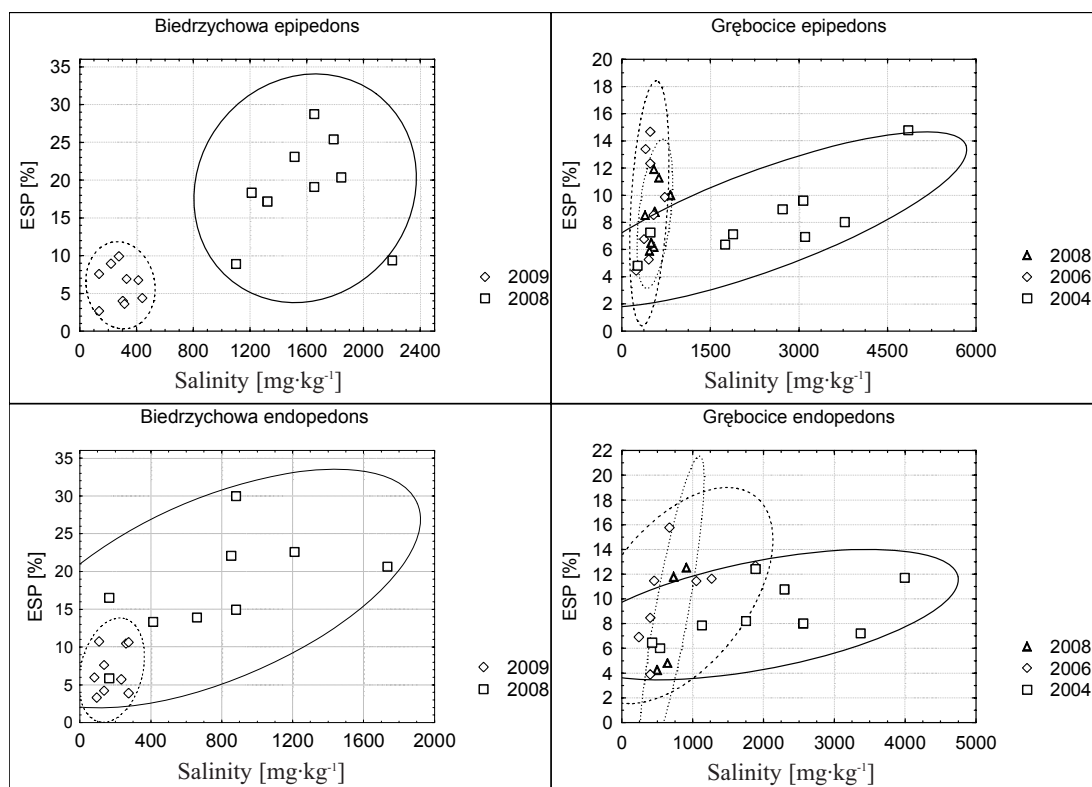


Fig. 3. Salinity and exchangeable sodium percentage for epi - and endopedons from the "Biedrzychowa" and "Grębocice" locations.

Table 3. Average values and standard deviations of salinity, cation percentage, and base saturation in the accumulation horizons (epipedons) of soils of the particular locations in the successive cycles of analyses.

Location	Parameter	Salinity [mg·kg ⁻¹]		ESP [%]		ECaP [%]		EMgP [%]		EPP [%]		V [%]	
"Biedrzychowa"	sampling date	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009
	average	1,588	286	18.92	6.07	52.94	57.62	18.48	13.56	1.47	1.86	91.8	79.1
	std. deviation	344	107	6.64	2.53	17.47	18.96	8.67	4.79	0.58	0.60	8.16	15.4
	No. of samples	9	9	9	9	9	9	9	9	9	9	9	9
"Dąbrowa"	sampling date	6.1995	6.1997	6.1998	6.1997	6.1995	6.1997	6.1995	6.1997	6.1995	6.1997	6.1995	6.1997
	average	448	161	6.33	3.76	50.85	54.96	11.14	12.91	4.29	4.71	72.6	76.3
	std. deviation	250	68	3.86	0.90	14.26	15.94	2.79	4.89	2.05	1.89	16.5	15.4
	No. of samples	11	10	11	10	11	10	11	10	11	10	11	10
"Lesniczówka"	sampling date	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004
	average	1,017	188	13.55	2.52	62.95	62.84	15.96	7.48	1.55	1.46	93.8	89.2
	std. deviation	651	66	7.69	1.03	12.53	31.57	3.14	3.69	0.77	0.60	8.7	8.8
	No. of samples	7	4	7	4	7	4	7	4	7	4	7	4
"Gręboice"	sampling date	9.2004	4.2008	9.2004	4.2008	9.2004	4.2008	9.2004	4.2008	9.2004	4.2008	9.2004	4.2008
	average	3,028	497	8.86	10.11	82.68	78.95	6.34	8.12	1.53	2.12	99.4	99.3
	std. deviation	1075	115	2.86	3.50	4.99	3.85	2.50	1.41	1.23	1.05	0.55	0.40
	No. of samples	7	7	7	7	7	7	7	7	7	7	7	7
"Polkowice"	sampling date	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999
	average	651	423	6.82	5.78	68.76	77.03	14.64	10.43	5.31	3.54	95.5	96.8
	std. deviation	146	53	2.55	0.61	5.42	1.70	1.28	0.63	1.89	1.80	1.86	1.13
	No. of samples	6	5	6	5	6	5	6	5	6	5	6	5

ECaP – exchangeable calcium percentage, EMgP – exchangeable magnesium percentage, EPP – exchangeable potassium percentage, V – base saturation

time, the decrease in the exchangeable sodium percentage was statistically significant at $\alpha < 0.001$ for the "Biedrzychowa" location, at $\alpha < 0.005$ for the "Dąbrowa" location (when comparing 1995 and 1998), and at $\alpha < 0.05$ for the "Leśniczówka" location. For the "Polkowice" and "Grębocice" locations the changes in ESP were not statistically significant, even at $\alpha < 0.05$. It should be noted that the soils of locations for which the change in the ESP parameter was not statistically significant have notably finer texture than the soils from other locations. At the same time, one can observe a distinct tendency for point clustering on the salinity – ESP graph with the passage of time from the moment of pipeline failure (Fig. 3). This is illustrated by the change in standard deviation of both parameters immediately after the failure and in the course of subsequent analyses (Table 2). The reduction of salinity standard deviation was statistically significant for all location except "Polkowice," and the reduction of ESP standard deviation was statistically significant for all location except "Grębocice."

As follows from Table 3, for all the locations under study the standard deviation for salinity decreased clearly with the passage of time. The standard deviation of the ESP parameters decreased for the locations "Biedrzychowa," "Dąbrowa," "Leśniczówka," and "Polkowice," and remained at an unchanged level for "Grębocice."

The cations exchange capacity did not display any systematic changes in the course of the successive analyses on the locations, nor did the levels of exchangeable calcium, magnesium and potassium percentage, or the base saturation (V) of the soils (Table 2).

Subsurface Horizons (Endopedons)

A less clear-cut image of the desalinization process is observed in the case of the deeper soil horizons (endopedons).

For endopedons during the first cycle of measurement, pH varied from pH 3.9 to pH 7.8 with median 6.4. During the last cycle of measurement the pH varied from 4.1 to 7.7 with the same median value 6.4. The values of median for individual locations also hadn't shown the systematic changes.

In those soils horizon salinity decreased with the passage of time in all locations, but the difference between salinity immediately after pipeline failure and that observed in the course of later studies was statistically significant only in the case of "Biedrzychowa" ($\alpha < 0.005$) and "Grębocice" ($\alpha < 0.05$). For the other locations the differences were not statistically significant, though at all of them a decrease in salinity was observed. Standard deviation of salinity decreased in statistically significant meaning ($\alpha < 0.005$) for 3 locations and remained at an unchanged level in the location "Leśniczówka" and "Polkowice" (Table 4).

The exchangeable sodium percentage varied in the subsurface horizons of the soils in an irregular manner. Standard deviation for the ESP parameter decreased in statistically significant meaning ($\alpha < 0.05$) only for "Biedrzychowa" and "Leśniczówka."

As in the case of the epipedons, the total cations exchange capacity did not display any systematic changes in the course of the successive analyses on the locations, nor did the levels of exchangeable calcium, magnesium and potassium percentage, or the base saturation of the soils.

Generally speaking, both for the surface and the subsurface horizons of the soils one can note trends toward regular changes in both parameters describing salinity in coarsely textured soils, i.e. a decrease in the value of salinity and ESP and a decrease in the spread of values of both parameters as expressed by standard deviation. In the soils with fine texture the changes are of a less regular character and are time delayed.

Decreases in the value of standard deviation with simultaneous decrease in salinity means that for soils with higher initial salinity the salt leaching process proceeds at a faster rate.

Likewise, it can be noted that in soils with coarse and medium texture the process of sodium displacement proceeds faster for soils with a higher initial ESP value.

Decrease in Salinity with Relation to Precipitation and to Initial Salinity

As has been mentioned above, decrease in soil salinity in the surface horizons, over time, is greater the higher the initial salinity. At the same time, the effectiveness of salt leaching from soil decreases with time [22]. It can be assumed that soil salinity decreases due to the leaching of easily soluble compounds from the group of chlorides and sulphates by precipitation water percolating through the soil. Formally, the above observation can be formulated in the method given below, similar to the equation proposed by Leffelaar and Sharma [18]:

$$\frac{\Delta S}{\Delta H} = -\beta S \quad (1)$$

...where: S is soil salinity ($\text{mg}\cdot\text{kg}^{-1}$), ΔS is the decrease of soil salinity ($\text{mg}\cdot\text{kg}^{-1}$), ΔH is the amount of water that percolates through the soil to the groundwater or to the deeper soil horizons (expressed in cm of water column), while β is a coefficient of proportionality dependent on the properties of the soil.

Therefore, for a specific soil:

$$\frac{1}{S} \frac{\Delta S}{\Delta H} = -\beta \quad (2)$$

...is a constant value that can be interpreted as the relative amount of salt leached from a given soil horizon ($\text{mg}\cdot\text{kg}^{-1}$) due to percolation of 1 cm^3 of water through a 1 cm^2 section of the soil. Parameter β jointly characterizes those soil properties that affect the rate of desalinization.

The values of parameter β were calculated for the epipedons and endopedons in the particular locations using as the value of H atmospheric precipitation in the region of the pipeline failure determined on the basis of data from Polkowice meteorological station 1995-2009.

Table 4. Average values and standard deviations of salinity cation percentages and base saturations in the subsurface horizons (endopedons) of soils of the particular locations in the successive cycles of analyses.

Location	Parameter	Salinity [mg·kg ⁻¹]		ESP [%]		ECaP [%]		EMgP [%]		EPP [%]		V [%]						
"Biedrzychowa"	sampling date	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009	12.2008	3.2009					
	average	774	179	17.73	6.93	47.36	49.43	20.54	14.59	1.73	2.19	87.36	73.16					
	std. deviation	504	81	6.94	3.03	12.41	20.00	4.75	4.67	0.92	0.71	6.39	15.52					
	No. of samples	9	9	9	9	9	9	9	9	9	9	9	9					
"Dąbrowa"	sampling date	6.1995	6.1997	6.1998	6.1997	6.1995	6.1997	6.1995	6.1997	6.1998	6.1997	6.1995	6.1998					
	average	513	160	79	3.31	5.05	2.77	42.58	49.99	48.39	4.45	8.81	8.51	4.45	5.76	68.00	65.97	
	std. deviation	668	178	76	1.32	1.14	1.51	11.65	14.41	10.83	1.46	2.44	1.56	1.46	2.13	13.70	15.05	12.41
	No. of samples	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7	7
"Lesniczówka"	sampling date	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004	12.2003	2.2004					
	average	565	470	14.89	6.97	39.99	60.10	14.92	9.88	2.06	1.76	71.86	78.71					
	std. deviation	380	499	9.76	4.80	10.77	7.67	5.47	6.36	0.97	0.45	17.28	10.53					
	No. of samples	7	12	7	12	7	12	7	12	7	12	7	12					
"Grębocice"	sampling date	9.2004	5.2006	4.2008	9.2004	5.2006	4.2008	9.2004	5.2006	4.2008	9.2004	5.2006	4.2008					
	average	1998	800	705	8.73	10.28	9.03	74.32	76.13	63.28	2.60	3.02	1.27	98.14	98.46	99.63		
	std. deviation	1209	564	149	2.31	3.70	4.12	5.49	8.81	5.36	1.18	1.41	0.39	1.58	0.99	0.13		
	No. of samples	9	8	5	9	8	5	9	8	5	9	8	5	9	8	5	9	
"Polkowice"	sampling date	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999	10.1998	4.1999					
	average	756	349	8.06	7.53	56.40	69.65	21.65	14.52	4.61	4.59	90.71	96.30					
	std. deviation	286	93	3.17	1.39	4.64	2.98	1.25	2.20	3.54	0.88	3.76	1.36					
	No. of samples	3	3	3	3	3	3	3	3	3	3	3	3					

ECaP – exchangeable sodium percentage, EMgP – exchangeable magnesium percentage, EPP – exchangeable potassium percentage, V – base saturation

For the surface horizons of soils of the studied locations, the values of the parameter β calculated from equation [2] fell within the range -0.0012 cm^{-1} (a slight increase in salinity in the Grębocice location in the period 2006-08) to 0.0699 cm^{-1} (rapid decrease in salinity in the location “Biedrzychowa” in the period 2008-09). The values of parameter β for the particular soil horizons and periods of analyses are given in Fig. 4. As was expected, the values of the parameter calculated for a given soil in different periods vary only slightly. In the case of the subsurface horizons (endopedons), the rate of desalinization varied from

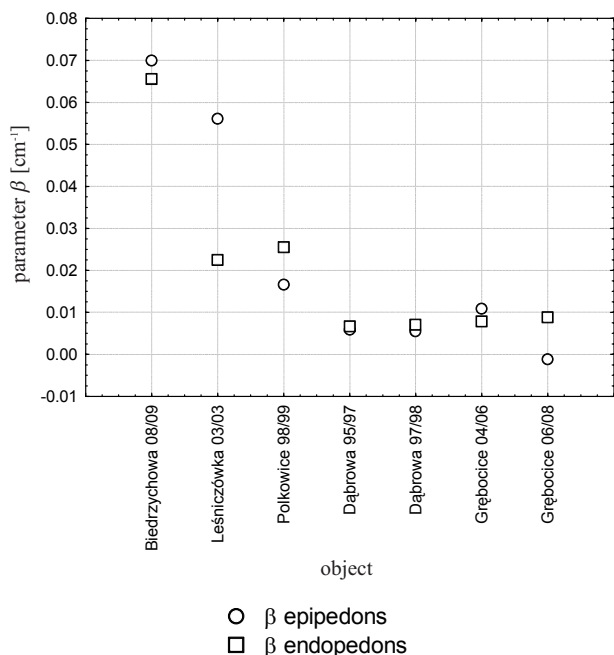


Fig. 4. The values of β parameter for epi- and endopedons from examined locations.

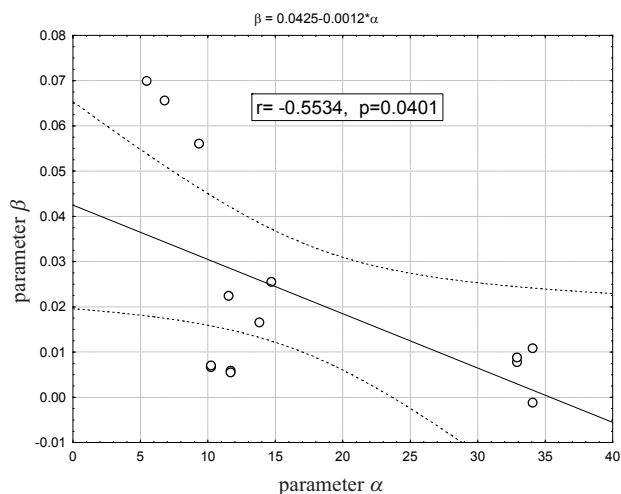


Fig. 5. The relation between parameter α – characterizing the texture of soil and parameter β – characterizing the rate of soil desalinization.

0.0067 cm^{-1} in the location “Dąbrowa” in the period 1995-97 to 0.0655 cm^{-1} in the location “Biedrzychowa” in the period 2008-09. For coarse-textured soils, the rate of desalinization of the accumulation horizons was greater than of the subsurface horizons, while for finely textured soils the rate of desalinization of both horizon groups was similar (Fig. 4). Certain differences in the rate of desalinization of the surface and subsurface soil horizons may result from the fact that a part of the salts leached from the accumulation horizons may be deposited in the subsurface horizons.

An attempt also was made at quantitative estimation of the effect of soil texture on the rate of the natural process of desalinization of soils as expressed by the value of parameter β . It was found that parameter β significantly correlates with the parameter describing soil texture $\alpha = f_0^{0.002} + f_{0.002}^{0.05}/3$, where $f_0^{0.002}$ is the content of clay fraction while $f_{0.002}^{0.05}$ is the content of fraction with particle diameters from 0.002 mm to 0.05 mm (Fig. 5).

A significant role in the processes of desalinization of soil also is played by the distribution of precipitations in time and by such other characteristics of soil as the aggregate structure, compaction, or organic matter content [4].

Conclusions

1. With the passage of time from soil flooding with saline water from a cooper mine, as a result of natural processes, there takes place a decrease in the salinity of the soil surface horizons. It is statistically significant on light soils already after 3 months between analyses. Decrease in salinity and exchangeable sodium percentage with the passage of time is also observed in the case of subsurface horizons. However, those tendencies are not as distinct as for the surface horizons.
2. The observed decrease in the exchangeable sodium percentage of the surface horizons was statistically significant only in the case of soils developed from formations with coarse texture.
3. Both for the surface and the subsurface horizons one can observe tendencies toward regular changes in the values of both parameters describing salinity in light soils, i.e. a decrease in the value of salinity and ESP, and a decrease in the scatter of values of both parameters as expressed by their standard deviations. In soils with fine texture those changes are less regular in character and are delayed in time.
4. It was found that for soils with higher initial salinity the process of salt leaching from the surface horizons proceeds faster than for soils with lower initial salinity.
5. The rate of soil desalinization can be characterized by means of the parameter described by equation [2] that gives a synthetic presentation of soil susceptibility to the leaching of easily soluble salts. The value of the parameter depends statistically significantly on the particle size distribution (texture) of the soil.

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