

SOIL ACIDITY RELATIONS WITH SURFACE CHARGES OF SOIL CONSTITUENTS AND THEIR DEPENDENCE ON pH

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Abstract. The pH influence on the particular forms of soil acidity and the relations between soil neutralization and charge were investigated. Clay fractions of six acidic soils of different origin were examined after conversion into homoionic sodium and aluminium forms. Results indicate that soil exchange acidity should be defined not on the basis of neutral salt extraction but on the basis of surface charge available to acidic ions adsorption. The amount of pH dependent acidity neutralized with pH increase can be quantitatively described by the increase of negative surface charge and the decrease of positive surface charge.

INTRODUCTION

Soil acidity is the basic characteristic of the nature and quantity of proton donors present in the soil.

Total soil acidity may be divided onto two main parts: active acidity and potential acidity. Potential acidity is further divided onto two forms: exchangeable acidity and pH-dependent (hydrolytic) acidity [5].

Active acidity is equal to the activity of hydrogen ions in the soil solution [24]. Exchangeable acidity is defined as that part of soil acidity which can be extracted from the soil with a neutral salt solution (KCl, BaCl₂, CaCl₂). Exchangeable acidity is due mainly to the presence of exchangeable aluminium and of exchangeable hydrogen in soil exchange complex [20,25]. For a long time hydrogen ions were considered to be strongly

exchangeably adsorbed by the soil [12] but they have been proven to be adsorbed like any other monovalent cation [8]. In general, it is assumed that exchangeable hydrogen plays an important role in organic soils, but it is probable that the presence of protons in salt extracts of these soils is due to the hydrolysis of aluminium ions specifically adsorbed by organic matter [27]. Because of this, the role of exchangeable hydrogen in exchangeable acidity is often neglected and this acidity is attributed to exchangeable aluminium. The quantity of exchangeable acidity depends on the soil mineralogical composition and the organic matter present. The highest exchangeable acidity values are observed in montmorillonitic soils and the lowest in kaolinitic soils [21]. Negative surface charge balancing with Al and Fe (hydro) oxide coatings has lead to exchangeable acidity decreases [3]. Exchangeable acidity strongly increases as soil pH decreases due mainly to aluminium compound dissolution accompanied by Al adsorption [25].

The pH dependent acidity is due to the part of soil acidity that can be neutralized as pH increases until a certain value (with the exclusion of exchange acidity) [1]. As a rule, it is assumed that the pH value to which the soil is neutralized is 8.2. This value comes

from Bradfield and Allison [2] as the criterion of 100 % base saturation. During pH dependent acidity neutralization, mainly variable charge constituent reactions are involved and are connected with surface negative charge and CEC increase [23]. The consumption of the base, however, is not equal to the negative charge increase. Gillman and Sumpter [10] stated that only 50 % of base added was consumed with a CEC increase of tropical soils investigated by them. The same effect was observed for Scottish soils [18], but in this case the dose of lime applied was only 1.26 times higher than the average CEC increase. This phenomena can be related in part to the specific adsorption of calcium ions by soil organic matter [26].

The purpose of the present work was to investigate the pH influence on the particular forms of soil acidity and the relations between soil neutralization and charge.

MATERIALS AND METHODS

Clay fractions of six acidic soils of different origin were examined. Their description is presented in a previous paper [13].

From the clays organic matter, iron oxides and aluminium oxides were consecutively removed and resulting clay materials were converted into homoionic sodium and aluminium forms which were adjusted in suspensions to different pH values as described in previous publications [13-15].

After two weeks of equilibration, the solid phases were separated from the suspensions by centrifuging in double chamber vessels divided by porous membrane covered with filter paper. Titratable acidity (TA, in $\text{cmol n}^{-1}\text{kg}^{-1}$), was measured in the sediments by titration of the two combined N BaCl_2 salt extracts to pH 8.2.

Next, all clay suspensions of the lowest pH, 3, were centrifuged. One half of the equilibrium solution (supernatant) was taken out from each sample and the sediment was dispersed in the remaining one half of the supernatant. The part of supernatant taken

out and the suspension were titrated with 1 N NaOH/0.01 N NaCl solution. Supernatants were titrated automatically and suspensions periodically with adding every 24 h of titrant small portions and continuously shaking. The quantity of base to be reacted with the solid phase was calculated as a difference between the quantity of base to be reacted with the suspension and the quantity to be reacted with the supernatant at the required pH level.

RESULTS AND DISCUSSION

Since exchange acidity is defined as the sum of acidic ions extracted from the soil with a neutral salt solution, it has to be equal to the value of titratable acidity (TA). Following this definition, the TA value is equal to the amount of acidic exchangeable ions in the CEC.

The measured values of titratable acidity for the sodium and aluminium clays investigated are presented in Table 1.

On the basis of the data presented in previous papers [14-16], when one calculates the part of surface negative charge that can be available for acidic ions at given pH conditions (the value of surface negative charge minus the quantity of exchangeable sodium, both measured at the same pH), it is seen that titratable acidity is greater than the charge available for acidic ions in entire pH range. This is particularly pronounced for samples containing organic matter (natural clays) and can possibly be attributed to chemically bonded surface proton release due to increasing salt concentration (suppressing of diffuse double layer). It has also been seen that titratable acidity occurs even at very high pH values which can be caused by the release of protons from sesquioxides surfaces and edges of clay minerals as well as by organic matter.

It can be stated then that the definition of exchangeable acidity should not be based on the neutral salt extraction but rather on surface charge and exchangeable basic cation measurements. It ought to be defined as

Table 1. Titratable acidities ($\text{cmol n}^{-1}\text{kg}^{-1}$), of the clays investigated extracted at different pH levels

pH	Sodium forms											
	1A*	1B	1C	1D	2A	2B	2C	2D	3A	3B	3C	3D
3.0	10.8	3.3	4.6	3.9	9.7	3.8	4.1	4.5	14.7	6.2	6.5	7.8
3.5	8.7	3.1	4.4	3.8	8.0	3.0	3.7	3.9	10.0	4.0	4.7	5.2
4.0	8.8	2.9	3.8	3.5	6.4	2.7	3.3	3.6	9.3	3.4	3.7	4.1
4.5	8.2	2.3	3.1	2.5	5.4	2.0	3.2	2.5	8.9	3.4	3.9	3.6
5.0	7.6	1.8	2.0	2.2	5.4	2.0	2.4	2.4	8.3	2.7	3.2	2.5
6.0	7.9	2.3	2.4	1.9	5.5	2.7	2.5	2.4	7.8	1.9	2.0	2.1
7.0	7.3	2.7	2.2	1.9	5.8	3.3	2.1	2.1	8.6	2.6	2.3	2.0
8.0	7.3	3.2	2.0	1.7	6.2	3.4	2.4	2.1	9.4	2.8	2.1	1.8
8.5	7.8	3.7	2.1	1.7	6.6	3.4	2.7	2.1	9.7	3.5	2.6	1.9
pH	4A	4B	4C	4D	5A	5B	5C	5D	6A	6B	6C	6D
3.0	11.8	4.1	4.1	4.3	10.2	5.9	6.8	7.6	5.3	3.1	3.3	3.5
3.5	9.9	3.0	3.1	3.4	7.9	4.6	5.4	4.9	4.4	2.4	2.8	2.9
4.0	8.2	2.8	2.5	2.8	6.7	3.4	4.1	4.2	4.7	2.7	2.4	2.4
4.5	8.3	2.3	2.1	2.1	5.3	3.5	3.4	3.4	3.8	2.2	2.2	2.1
5.0	8.6	2.0	1.9	2.0	4.9	3.0	2.8	2.8	3.6	1.9	1.9	1.7
6.0	8.3	2.3	2.1	2.1	4.6	2.8	2.5	2.9	3.0	1.9	2.0	1.3
7.0	8.5	2.2	2.2	2.0	4.8	3.1	2.4	2.3	3.3	2.1	1.5	1.8
8.0	8.3	2.0	2.1	1.8	4.7	3.1	2.8	2.6	3.6	2.1	1.9	1.6
8.5	8.2	2.1	2.4	1.7	4.9	3.1	2.9	2.6	3.9	2.4	2.1	1.5
pH	Aluminium forms											
	1A	1B	1C	1D	2A	2B	2C	2D	3A	3B	3C	3D
3.0	29.4	25.3	25.6	26.2	26.3	20.1	22.1	24.3	55.7	46.8	49.6	53.3
3.5	31.7	25.1	26.4	26.8	27.1	21.0	22.3	24.8	58.5	48.0	51.1	56.2
4.0	33.8	27.1	28.1	28.5	27.9	20.9	22.5	25.2	63.1	49.4	53.7	59.5
4.5	19.2	11.3	12.2	12.3	16.3	8.5	9.6	10.3	30.6	19.8	20.9	23.0
5.0	15.6	6.1	6.8	7.8	12.7	5.7	5.4	6.6	20.8	10.1	10.5	11.1
6.0	8.1	2.3	2.4	1.9	6.7	2.2	2.2	1.9	10.9	3.5	3.4	3.5
7.0	7.3	2.8	2.2	2.0	6.4	2.5	2.1	2.0	10.6	3.2	3.3	3.3
8.0	7.6	3.3	3.1	2.8	6.2	3.3	2.4	2.3	10.7	3.6	3.2	3.1
8.5	7.9	3.4	3.1	2.8	6.1	3.6	2.9	2.8	11.4	3.9	3.1	3.1
pH	4A	4B	4C	4D	5A	5B	5C	5D	6A	6B	6C	6D
3.0	28.8	23.1	24.2	25.1	40.6	40.7	44.8	47.1	20.1	17.4	19.1	22.8
3.5	30.9	23.4	23.7	25.4	42.9	42.0	45.7	47.7	20.4	17.9	19.2	23.4
4.0	34.2	23.8	24.5	25.8	45.7	43.4	46.1	49.6	20.7	18.1	19.9	24.1
4.5	20.9	10.3	10.9	11.1	20.3	16.8	17.4	21.1	10.3	8.2	9.5	10.1
5.0	13.8	7.4	8.4	8.5	12.9	10.7	11.6	12.1	8.0	4.9	5.1	5.9
6.0	9.3	3.3	3.4	3.4	7.1	3.3	3.4	3.5	6.1	3.3	3.3	3.4
7.0	9.5	3.2	3.2	3.2	6.6	3.1	3.3	3.3	5.4	3.1	3.2	2.8
8.0	9.3	3.9	3.1	3.1	6.2	3.5	3.1	3.1	5.2	3.5	3.2	2.9
8.5	9.2	4.1	3.1	3.1	6.2	3.8	3.1	3.1	5.2	3.7	3.1	3.1

* 1-Mollic Gleysol; 2-and 3-Eutric Cambisols; 4-Stagnogleyic Phaeozem; 5-Orthic Luvisol; 6-Podzoluvisol. A-natural clays; B-after organic matter removal; C-after Fe removal; D-after Al removal.

the part of the actual negative charge available for acidic cations, as it is done by Gillman [9] in his method of exchangeable acidity measurement in tropical soils.

Using this last definition, the exchangeable acidity for the samples investigated was

calculated as the difference between the actual surface charge and the exchangeable sodium ions. Data from papers [14,15, and 16] were used and the results are presented in Fig. 1.

Exchangeable acidity of sodium clays is caused mainly by exchangeable hydrogen ion

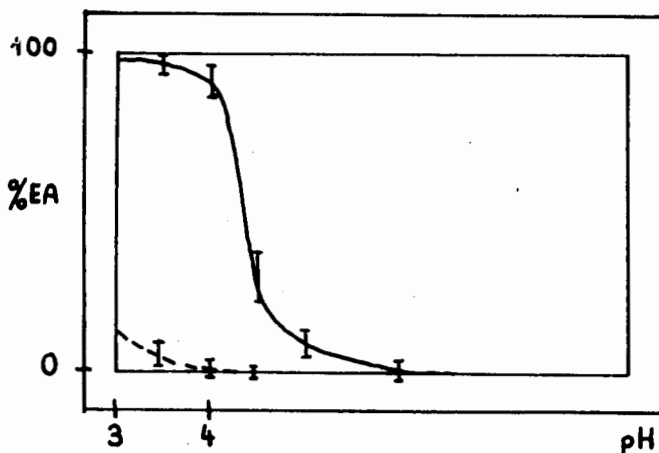


Fig. 1. Exchangeable acidity (EA) as a percentage of the charge available for acidic ions in the actual CEC. Plots are for samples in aluminium forms (solid line) and for sodium forms (dashed line). I - indicates the range of experimental points.

adsorption. Its zero value above pH 4-4.5 is in agreement with many authors [4,6,11]. Note that, even at low pH values the exchange acidity of sodium clays is low.

For aluminium clays the exchange acidity is mainly due to exchangeable aluminium. Proton adsorption may be neglected. Exchange acidity vanishes at pH values of about 6. This is in agreement with many authors who have described the role of soil exchange aluminium with changes of pH [7,21,22].

From Fig. 1 it is seen that for the systems investigated the presence of particular soil components (organic matter, iron and aluminium oxides) has no influence on the percentage of exchange acidity in the CEC. The only important factor is the ionic composition of the surface what is strongly connected to the ionic composition of the soil equilibrium solution. So the exchange is governed by two factors: (i) by the composition of the soil solution (including pH) from a qualitative point of view and (ii) by the soil solid phase composition (influencing the magnitude of CEC) from the quantitative point of view.

The pH dependent acidity was evaluated using the results of titrations of clay suspensions and their equilibrium solutions

(supernatants). The results, presented in Table 2, are differences between the quantity of base consumed with the reaction of the suspensions and with the reaction of the supernatants from pH 3 to a given pH. These differences give the quantity of base reacted with the solid phase only. If this difference is assumed to be equal to the potential acidity of the solid phase, neutralized with pH increase to a given value (ΔPA), then ΔPA will be equal to the exchange acidity neutralized with a pH increase to a given value (ΔEA), plus the part of the pH dependent acidity neutralized to this pH (ΔHA). Using the values of exchange acidity from a previous paper [16] and assuming that for pH 3 the neutralized pH dependent acidity is equal to zero, one can easily calculate ΔHA values for any investigated pH value:

$$\Delta HA = \Delta PA - \Delta EA \quad (1)$$

The calculations show that for sodium clays, the quantity of potential acidity neutralized with the reaction of the base is practically equal to the sum of exchange acidity neutralized plus the increase of surface negative charges minus the quantity of positive charge decrease (the values of the above quantities were calculated on the

Table 2. Quantities of base used to neutralize the solid phase of clay suspensions from pH 3 to different pH values

pH	Sodium forms											
	1A	1B	1C	1D	2A	2B	2C	2D	3A	3B	3C	3D
3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
3.5	3.5	3.9	4.1	4.2	4.7	3.2	2.4	2.3	10.8	7.6	5.7	6.4
4.0	10.5	7.8	7.8	7.2	8.8	6.0	4.1	3.5	16.5	12.0	10.1	11.0
4.5	18.6	11.9	12.0	10.2	13.3	8.8	5.5	4.1	23.5	16.2	12.9	14.0
5.0	23.3	14.0	13.8	12.7	17.9	11.3	6.9	4.8	31.2	21.4	17.5	17.3
6.0	31.2	16.8	15.6	13.6	25.6	14.9	8.7	6.5	43.2	28.2	22.9	18.3
7.0	44.1	19.2	16.5	14.9	32.4	17.7	10.0	7.6	53.9	34.5	26.4	18.9
8.0	57.2	21.5	18.0	15.5	38.4	19.5	11.1	8.5	63.6	37.8	28.8	19.8
8.5	63.4	22.3	18.5	15.7	41.7	22.0	11.9	8.8	69.0	39.2	29.9	20.4
pH	4A	4B	4C	4D	5A	5B	5C	5D	6A	6B	6C	6D
3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
3.5	5.5	3.5	3.4	2.9	7.5	6.4	5.0	4.8	5.1	3.7	2.6	2.1
4.0	11.5	5.8	6.2	4.9	12.9	10.3	7.6	6.9	8.3	6.1	4.9	4.5
4.5	17.6	8.9	8.4	6.7	17.1	12.2	9.2	10.1	12.2	8.3	7.1	6.0
5.0	21.3	11.1	9.8	8.5	22.3	16.3	11.3	11.6	15.1	10.2	8.2	6.8
6.0	28.4	12.7	11.1	9.7	29.8	19.9	13.9	12.2	21.4	14.2	11.3	8.3
7.0	41.6	15.2	12.0	10.2	35.1	23.0	15.7	12.8	25.2	15.9	13.4	8.8
8.0	54.9	16.3	13.4	11.0	41.4	23.8	17.8	13.9	27.5	19.3	15.0	9.1
8.5	63.8	17.2	13.7	11.4	44.6	25.2	18.6	14.5	29.6	20.0	15.6	9.8
pH	Aluminium forms											
	1A	1B	1C	1D	2A	2B	2C	2D	3A	3B	3C	3D
3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
3.5	4.1	2.9	3.2	2.9	3.6	2.3	1.5	1.1	15.8	5.4	3.9	3.4
4.0	9.2	6.2	6.1	5.5	7.2	4.7	2.6	1.6	12.9	8.9	6.7	6.6
4.5	32.8	26.1	26.4	25.5	26.0	20.6	18.4	18.5	52.8	42.3	41.3	50.8
5.0	42.4	33.2	32.4	32.6	34.6	26.7	24.1	24.1	69.9	56.5	54.9	65.0
6.0	52.8	38.4	36.5	36.2	44.8	32.1	28.0	27.8	86.6	67.4	64.8	71.3
7.0	65.7	40.5	37.9	37.4	51.5	34.9	29.2	28.9	97.4	73.3	68.5	72.1
8.0	78.9	42.7	39.5	37.9	57.5	37.3	30.3	29.7	106.0	76.2	70.8	73.3
8.5	85.0	43.6	39.9	39.6	61.9	39.4	31.4	30.3	112.0	78.0	72.0	74.2
pH	4A	4B	4C	4D	5A	5B	5C	5D	6A	6B	6C	6D
3.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
3.5	4.5	2.4	2.2	1.9	5.5	4.5	3.2	2.7	4.3	2.9	1.8	1.1
4.0	9.8	4.6	4.6	3.4	10.3	7.3	4.7	3.6	7.2	5.0	3.8	3.0
4.5	31.3	21.0	21.4	20.8	39.8	35.7	35.5	39.2	22.8	16.9	16.8	18.3
5.0	39.8	27.7	26.9	26.7	52.7	46.8	45.5	49.7	28.4	21.4	21.1	23.2
6.0	48.9	31.3	30.4	29.7	64.0	54.2	52.4	54.8	36.3	26.8	25.7	26.5
7.0	62.6	33.6	31.3	30.2	68.6	57.8	54.1	55.8	39.8	29.0	27.9	26.9
8.0	75.7	34.8	32.5	31.5	75.1	58.5	56.3	56.6	42.8	32.1	29.6	27.4
8.5	84.5	35.4	33.1	32.0	78.2	59.8	57.3	57.4	44.7	32.7	30.3	28.0

Samples abbreviations are the same as in Table 3.

basis of data presented in previous papers [13-16]. Therefore, it can be concluded that the pH dependent acidity of the samples investigated is equal to the increase of CEC minus the decrease of anion exchange capacity (AEC).

The analogical calculations for aluminium clays lead to the same conclusions but with less accuracy. It was assumed that for aluminium forms the values of positive surface charge are the same as for sodium forms. So, the quantities of pH dependent

acidities are the same for sodium and aluminium forms and the differences in the quantities of the base needed for neutralization either form to the same pH are the results of differences in their exchange acidity values (initial exchange acidities of aluminium forms exceed the respective values for sodium forms by about 10 times).

It is important to note that a significant role in base consumption and in pH dependent acidity is played by the neutralization of positive surface charge, even in these temperate climatic zone soils which were investigated. This can provide an additional explanation for the observed phenomena that base (lime) consumption exceeds CEC increase in the soils of our climatic region [17].

CONCLUSIONS

1. Soil exchange acidity should be defined not on the basis of neutral salt extraction since the neutral salt extraction leads to the overestimation of this value when compared to the surface charge available to acidic ion adsorption.

2. The amount of pH dependent acidity neutralized with pH increase can be quantitatively described by the increase of negative surface charge and the decrease of positive surface charge.

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ZALEŻNOŚĆ POSZCZEGÓLNYCH FORM KWASOWOŚCI GLEBOWEJ OD pH

Badano wpływ pH na poszczególne formy kwasowości glebowej oraz związek neutralizacji gleby z jej ładunkiem powierzchniowym. Do badań użyto sodowych i glinowych form frakcji ilastych wydzielonych z kilku gleb kwaśnych. Stwierdzono, iż do opisu kwasowości wymiennej bardziej nadaje się definicja oparta na wielkości ładunku powierzchniowego dostępnego dla jonów kwaśnych, aniżeli oparta na rugowaniu tychże jonów za pomocą soli obojętnej. Ilość kwasowości zależnej od pH neutralizowana podczas wzrostu pH badanych próbek dawała się ilościowo opisać poprzez zmiany ładunku powierzchniowego.