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SOIL PHYSICS

Effect of the Size of Elementary Soil Particles on the Soil Moisture Characteristic Curve

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Abstract—Statistical analysis of water vapor sorption by light clayey brown forest soil and its elementary particles of different diameters has revealed extremely close correlations and linear relationships between the logarithm of total soil water potential (pressure) and the water contents in the separated particle-size fractions (due to the hydration of exchangeable cations in the diffuse layer near the surface of soil solid phase), as well as between the water content of particle-size fractions and the logarithm of their diameter (due to the differences in the specific surface area and mineralogy of these particles).

Keywords: soil water content, soil water potential, soil water pressure, specific soil surface area, exchangeable bases, correlation, regression

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INTRODUCTION

The soil water regime can be optimized only on the basis of sufficiently exact prognostic calculation of water fluxes in soils, which requires knowledge of the geophysical properties of different particle-size fractions of these soils, primarily the relationships of the soil water potential (or pressure) and hydraulic conductivity with the soil water content, which are frequently referred to (at the suggestion of Globus) as soil moisture characteristic curves (SMCC) [2, 8, 10]. The determination and analysis of SMCC are essential and urgent problems of soil hydrophysics, which have been elaborated over the last half of a century. Interest in these problems does not decrease so far [11–13, 21–23, 25, 26, 28, 29, 31–33, 35–43, 45, 46, 48, 49]. The aim of this work was to reveal an essential component of the SMCC: the relationship between the soil water potential (pressure) and the water contents of different particle-size fractions.

OBJECTS AND METHODS

The solution of this problem requires adequate input information on the water content of soil particle-size fractions at different levels of water pressure (potential). Such information is reported in the fundamental monograph by Rode *Theory of Soil Moisture* [9]. These are results of the precise studies of Kuron [34], one of the founders of soil physics, who is known for the high precision of his experiments. The data are presented as the content of water adsorbed by different particle-size fractions of light clayey brown soil from Wegnersau (Lower Silesia, Poland) separated by sedimentation at different relative water vapor pressures. For the determination of relationship between the SMCC and the size of elementary particles in different fractions, these data were subjected to mathematical analysis according to the following algorithm: (1) the values of water potential (J/g water) were first calculated from the data on the equilibrium relative water vapor pressure; (2) the natural logarithms of water potential modules and the pF values were then calculated; (3) the water contents of different soil particle-size fractions at different levels of equilibrium relative water vapor pressure were calculated from the data of Kuron ([34], cited from [9]).

The next essential stage of study was to find a relatively compact (elementary) analytical mathematical expression (function) for the adequate description of SMCC in the entire range of hygroscopic moisture, because such an elementary function is necessary to present the effect of the size of elementary soil particles on the SMCC in the generalized form. Different analytical expressions were earlier presented for the description of SMCC in the hygroscopic moisture range; however, each of them gives satisfactory results only within a certain part of this range [9, 22].

The search for an adequate analytical function describing the SMCC is usually performed by means of pedotransfer methods. There are several main groups of these methods [21]: (1) methods of physically based models; (2) point-regression methods; (3) functional parametric regression methods. The methods of physically based models are preferable because they derive the most representative functions (the SMCCs obtained by these methods for one soil can be used for determining the SMCCs of other analogous soils). The complexity of the structural and functional properties of soils makes the creation of physically based models difficult; however, a happy example of such model is available for the SMCC within the hygroscopic moisture range [14–20]. This model is based on the formation of a diffuse layer of exchangeably adsorbed cations in the soil solution contacting with the surface of soil solid phase; these cations are retained at this surface (usually negatively charged) by the Coulomb forces of electrostatic attraction. According to the theory developed by Gouy [30], "The distribution of ions in the solution at the solid phase surface is determined by two opposite effects. On the one hand, the thermal motion tends to uniformly distribute ions so that similar numbers of positive and negative ions are in each element of solution volume. On the other hand, because of the excess of similarly charged ions at the interface, the electrostatic forces originated from the solid phase surface act so that the elements of solution volume located near the interface contain an excess of oppositely charged ions. The solid phase surface attracts the oppositely charged ions and repulses the similarly charged ions. The equilibrium established due to these two forces (thermal motion and electrostatics) is analogous to the equilibrium of gaseous molecules in the atmosphere under the impact of gravity. The excess of oppositely charged ions occurring at the surface decreases with the distance from the interface in accordance with the barometric law" [4].

In the discussion on the behavior of soil particles in water [9], Rode wrote, "Many researchers, beginning from Gouy, consider a particle of soil colloid as a nucleus surrounded with a diffuse layer of ions. In soil colloids, these are cations in most cases, because the particle usually has a negative charge. The cations forming the diffuse layer constitute a part of exchangeable cations (dissociated ions) .... They tend to spread out but are retained by the electrostatic attraction of the oppositely charged complex colloidal ion, which forms the nucleus of the ionic atmosphere. The cations dissociated by the colloidal particle generate the osmotic force. Due to the hydration of these ions, the soil acquires the capacity to uptake and retain water."

Shein analyzed this problem in the *Course of Soil Physics* and came to an analogous conclusion: "The mineral particles having exchangeable cations on their surface will repulse from one another...; a zone of increased ion concentration appears between separate particles. This should increase the osmotic pressure, which will 'pump' free moisture between the particles. These forces tend to gather water in the soil" [22].

The force of water retention by the soil hydrating these cations at different contents of soil water depends on the change of cation concentration in water with the distance from the electrically charged surface of soil solid phase. As early as 100 years ago, Gouy [24, 30] supposed that the relationship between the concentration of cations and their distance from the solid phase surface has an exponential character; i.e., the logarithm of cation concentration ($\log C_c$) in the diffuse layer is related to the distance from the electrically charged surface (L) by an inverse linear relationship

$$\log C_c = a - bL, \tag{1}$$

where a and b are constant values for a given object.

The validity of this supposition was to be strictly theoretically proved. This became possible due to fundamental discoveries in theoretical physics. The first of them was made more than 150 years ago (in 1859) by Maxwell [7], who theoretically derived the law of the statistical distribution of molecules among energy levels. This is one of the key scientific accomplishments behind statistical physics (an essential branch of the modern theoretical physics). Ten years later (in 1869), another great scientist Boltzmann [7] extends the scope of this law, which made it applicable for describing the distribution of particles by their potential energy in the external force field. For the gravity field of the Earth, the so-called barometric law was derived from this distribution law; the barometric law describes the changes in the atmospheric pressure at the elevation to several kilometers above sea level (in the first approximation, i.e., without consideration for the effect of the nonstationary state of the atmosphere). According to the barometric law, the logarithm of the atmospheric pressure linearly decreases with the elevation over the Earth surface [5, 7].

The laws describing two types of interactions—electrostatic (Coulomb law) and gravitational (gravity law) ones—are completely analogous (the attraction forces of masses and electric charges are inversely proportional to the squared distance between them) [5]; therefore, the Boltzmann barometric law can describe not only the decrease of atmospheric pressure with the elevation over the Earth surface, but also the structure of the diffuse layer of cations: the logarithm of their concentration linearly decreases with the distance from the negatively charged surface of the soil solid phase. The barometric law is frequently written in the form

$$\ln p_x = \ln p_o - (mgx/kT), \tag{2}$$

where p_o and p_x denote the atmospheric pressure at the sea level and height x, respectively; m is the molecular mass of the gas; g is the gravity acceleration; k is the Boltzmann constant; and T is the absolute temperature [5, 7].

According to the Clapeyron–Mendeleev law, the gas pressure (p) is proportional to its concentration (c) in the low pressure range; therefore, Eq. (2) can be put into the form

$$\ln c_x = \ln c_o - (mgx/kT) = \ln c_o - (mg/kT)\,x. \tag{3}$$

Then, because of the exact physical analogy of the Newton law of gravitation and the Coulomb law of electrostatic interaction, the following expression will be valid for the cations attracted by the negatively charged surface of the soil solid phase:

$$\ln C_{cx} = \ln C_{co} - (qk_1/kT)\,x, \quad (4)$$

where C_{co} and C_{cx} are the concentrations of cations on the solid phase surface and at distance x from this surface; q is the cation charge; and k_1 is the coefficient in the Coulomb law [5].

Thus, according to the Maxwell–Boltzmann–Helmholtz–Gouy theory, the concentration of cations continuously and steadily (according to the exponential law) decreases with the distance from the charged solid phase surface.

The nanoscopic thickness of the soil water layer, in which the major part of the diffuse layer of cations occurs, makes the experimental measurement of their concentration at different distances from the solid phase surface impossible by direct methods; however, this distribution can also be indirectly assessed from other cation properties. Among them are the ability of cations to be hydrated (i.e., to fix water). The hydration energies of different ions reach high values (kJ/mol): K^+, 314; Na^+, 398; H^+, 1060; Mg^{2+}, 1910; Fe^{3+}, 4355; Al^{3+}, 4640; Cl^-, 376; OH^-, 460; SO_4^{2-}, 1060 [6]. Therefore, cations (especially di- and trivalent ones), which are attracted by the negatively charged surface of the soil solid phase, strongly retain water molecules. This energy significantly exceeds that of the molecular interaction of water with the soil solid phase due to hydrogen bonds (60 kJ/mol) and the van der Waals forces (<8 kJ/mol) [6].

Each ion binds a certain number of water molecules; therefore, the concentration of cations (C_c) in the given point of the solution is directly proportional to the degree of water binding, i.e., the osmotic pressure of soil solution (P) in accordance with the van't Hoff law [6]:

$$P = RTC_c, \quad (5)$$

where R is the universal gas constant.

Consequently, the higher the concentration of cations (C_c) in any layer of soil solution, the higher the osmotic pressure and, hence, the lower the total soil moisture pressure (potential) in this layer. Substituting Eq. (5) to Eq. (4), we obtain

$$\ln|P_x| = \ln|P_o| - (qk_1/kT)x = A - Fx, \quad (6)$$

where $P_o = P$ on the solid phase surface, $A = \ln|P_o|$ and $F = qk_1/kT$.

Thus, it follows from the Maxwell–Boltzmann–Helmholtz–Gouy theory that the osmotic pressure of soil solution (P) also (as well as the concentration of cations) should decrease with the distance from the charged soil surface according to the exponential law. However, the diffuse layer thickness does not exceed several tens of nanometers (i.e., several tens of water molecule layers); therefore, the direct experimental measurements of the osmotic pressure of soil solution at so short distances from the solid phase surface are not yet feasible. At the same time, the values functionally related to it are measurable. One of these values is the equilibrium relative water vapor pressure (p/p_o) [6]:

$$P_x = (R\,T/V)\ln(p/p_o)_x, \quad (7)$$

where V is the water mole volume.

Consequently, if a state close to the thermodynamic equilibrium between the air's water vapor and the surface layer of soil water in contact with the air is ensured, the P_x value for the surface water layer at distance x from the soil solid phase surface can be determined from the p/p_o value using Eq. (7). The determination procedure is known [1, 2, 7, 8, 21].

The direct experimental measurement of the thickness of the water layer covering the soil solid phase surface in the wet soil is also impossible; however, under the supposition that the thickness of this water layer little varies over the surface, the mean layer thickness (x, L) can be approximately calculated as the quotient of the volumetric soil water content (W, L^3 water/M soil) by the soil specific surface area (S, L^2/M soil):

$$x = W/S. \quad (8)$$

This supposition is apparently valid for clayey and loamy soils, because, at low contents of soil water, the major part of soil moisture forms a thin layer on the surface of microscopic platy crystals of clay minerals (including montmorillonite-group minerals) [1].

Substituting (8) to (6), we obtain

$$\ln|P_W| = A - FW/S = A - BW, \quad (9)$$

where $A = \ln|P_o|$ and $B = F/S$.

This relationship was experimentally revealed for sediments in 1948 [44] and for soils in 1966 [19]. In [19], an attempt was also made to theoretically derive this relationship (with some simplifying assumptions). Later on, other authors also proposed its use [2]. Data confirming this relationship only for a relatively narrow P range (from –5 to –200 atm) were reported earlier [19]. Next, the lower limit of this range was extended to –2600 atm [17, 20, 27].

The variables $\ln|P_W|$ and W are related by a very close correlation (the coefficient of correlation between them is –0.99 for a significance level below 0.05) [3]. Such close correlation between the soil properties is very rare; it indicates the adequacy of the physically based model derived on the basis of fundamental physical laws discovered by Maxwell, Boltzmann, Helmholtz, and Gouy [7, 30, 47].

RESULTS AND DISCUSSION

The water content in the separated particle-size fractions of soil strongly depended on the size of elementary soil particles in the fraction (Table 1). At the relative water vapor pressure (p/p_o) equal to 0.942 (which corresponds to the maximum soil hygroscopicity determined by the Mitscherlich method and the water potential of –8.1 J/g water), the water content of

Table 1. Values of relative water vapor pressure (p/p_0), total soil water potential (P, J/g water), natural logarithm of P module ($\ln|P|$), pF, and the corresponding water contents (% of dry soil weight) for light clayey brown forest soil and its particle-size fractions. The soil sample contains 12.19% particles of <2 μm, 12.65% particles of 2–5 μm, 13.04% particles of 6–20 μm, and 62.12% particles of >20 μm

p/p_o	$-P$, J/g	$\ln\|P\|$	pF	Soil water content, %	Water content (%) of separate particle-size fraction, μm			
					<2	2–6	6–20	>20
0.942	8.1	2.08	4.91	7.61	20.60	13.52	9.80	2.05
0.868	19.3	2.96	5.27	6.24	16.82	11.32	8.50	1.63
0.748	39.3	3.67	5.60	5.03	13.60	9.33	6.98	1.34
0.582	73.3	4.29	5.87	3.96	10.57	7.40	5.59	1.02
0.383	130	4.92	6.11	2.91	8.00	5.50	4.16	0.82
0.177	234	5.45	6.37	1.97	5.45	3.85	2.83	0.54
0.069	363	5.90	6.56	1.21	3.48	2.48	1.80	0.35
0.034	459	6.12	6.66	0.88	2.82	1.78	1.25	0.25

the particle-size fraction <2 μm reached 20.6%, while the water content of the fraction >20 μm was only 2.05%. The water contents in the intermediate size fractions were expectedly intermediary. The maximum hygroscopic water content in the whole soil was 7.63%.

At the lowest relative water vapor pressure equal to 0.034 (corresponding to the water potential of −459 J/g water), the water content in the particle-size fraction <2 μm reached 2.82%, while the water content of the fraction >20 μm was only 0.25%. The water content of the whole soil was 0.88%.

Each particle-size fraction of soil contains elementary particles of different sizes, and one averaging parameter that characterizes the diameter of particles in this fraction should be selected for assessing the effect of the size (diameter) of soil particles on their hydrophysical properties. The water content in the soil under some relative water vapor pressure is proportional to the specific surface area of its particles, which in turn is approximately proportional to their diameter [21]; therefore, the middle point of the range for the diameters of the particles in the fraction may be taken as such an averaging parameter. The middle point corresponds to 4 μ for the fraction of 2–6 μm and 13 μm for the fraction of 6–20 μm. However, it is more difficult to select the averaging diameter for the fractions of <2 μm and >20 μm because only one limit is specified for their particle diameter range. The absent limits were defined using the Kuron rule for the separation of fractions [34]. According to this rule, the maximum diameter of particles in the fractions of 2–6 and 6–20 μm is higher than their minimum diameter by 3 to 3.3 times. Then, the lower limit of the fraction <2 μm may be taken at 2/3.3 = 0.6 μm, and the upper limit of the fraction >20 μm may be taken at 20 × 3 = 60 μm. Thus, the middle points of the diameter ranges for these fractions are 1.3 and 40 μm, respectively. In this case, the average diameter of particles in the coarsest fraction is higher than that in the finest fraction by 31 times. At the same time, the water content in the finest fraction is lower than that in the coarsest fraction by only 10 times at a relative water vapor pressure of 0.942 and by 11 times at a relative water vapor pressure of 0.034. Consequently, when the diameter of the particles decreases, the water content in their fraction increases three times less than their diameter.

For the size fractions of 2–6 and 6–20 μm, the decrease in the average diameter of their particles by 3.25 times results in an increase in the water content of the fraction by 1.4 times at both levels of relative water vapor pressure. Hence, in these fractions too, the decrease in the diameter of particles results in a significantly lower increase of water content.

Thus, the water content in the selected fractions of soil elementary particles increases less rapidly than their diameter decreases and, hence, less rapidly than their specific surface area increases [21]. As will be shown below, this can be due to the difference in the mineralogy of these fractions.

The relationships between $\ln|P_W|$ and W (Table 2, figure) are described by linear functions (9) (coefficients of correlation between these variables are −0.99 for the significance level below 0.05) [3].

The value of parameter A ($\ln|P_o|$) in this relationship depended little on the size of particles (Table 2). Its value varied in a narrow range from 6.62 to 6.75. The physical sense of this parameter is the value of $\ln|P|$ at the zero water content. Its value indicates the specific adsorption energy for the first water vapor molecules on the solid phase surface of over-dried soil (dried at 105°C), if Eq. (9) would remain valid until the water content of the soil was zero. However, this assumption is unprovable, because the zero relative air humidity is technically unattainable (water vapor, although very

Table 2. Parameters A and B of the relationships $\ln(|P_W|) = A - BW$ for the whole light clayey brown forest soil and its particle-size fractions (μm), where W is the water content of soil (g water/g soil), P_W is the total potential of soil water (J/g water) at the given W value, r is the coefficient of correlation between the values of $\ln(|P_W|)$ and W (for the significance level below 0.05), and d is the average diameter of elementary soil particles in the fraction (μm)

Object	A	B	r	d, mm	log(d)	(1/B) × 100
Soil	6.62	59.2	–0.99			1.69
>20 μm	6.68	225.0	–0.99	40	1.60	0.45
6–20 μm	6.75	45.6	–0.99	13	1.11	2.22
2–6 μm	6.74	33.8	–0.99	4	0.60	2.94
<2 μm	6.68	22.3	–0.99	1.3	0.11	4.55

rarefied, is present even in the open space). The value of P_o varies from –760 to –850 J/g water, the average value being –795 J/g water, which is equivalent to –7950 atm or 190 cal/g water. This value corresponds to the relative water vapor pressure equal to 0.0028.

This specific soil hydration energy exceeds the energy necessary for ice melting (79.7 cal/g water at 0°C) by 2.4 times. Hence, it significantly exceeds the energy of water attraction by ice crystals; therefore, ice crystals cannot subtract this water from the soil at 0°C. In addition, when the total potential of soil water decreases by 1 J/g water, its freezing temperature decreases by 0.83°C, and the first portions of water adsorbed by the dry soil cannot form ice crystals within the Earth's temperature range.

For the verification of these data about the parameter A value, it should be determined by an independent method. Data reported in Table 4 of Rode's monograph [9] were also used for this purpose. Rode indicated that the radius of dehydrated calcium ions is 0.106 nm, and the radius of hydrated ions is 0.96 nm. Correspondingly, their volumes are 0.0054 and 3.7 nm^3; the volume of water bound by one ion is 3.7 nm^3. At the same time, Hendricks and Jefferson concluded that the density of bound water is 0.88; as a result, the volume of one water molecule is 0.0325 nm^3 [8]. Consequently, one hydrated calcium ion binds 3.7/0.0325 = 114 water molecules, and one gram-molecule of ions binds 114 gram-molecules (i.e., 2050 g) of water. The hydration energy of calcium ions is –1570000 J/mol [6]; therefore, the total potential of bound water is 1570000/2050 = –768 J/g water, and the total water pressure is –7680 atm.

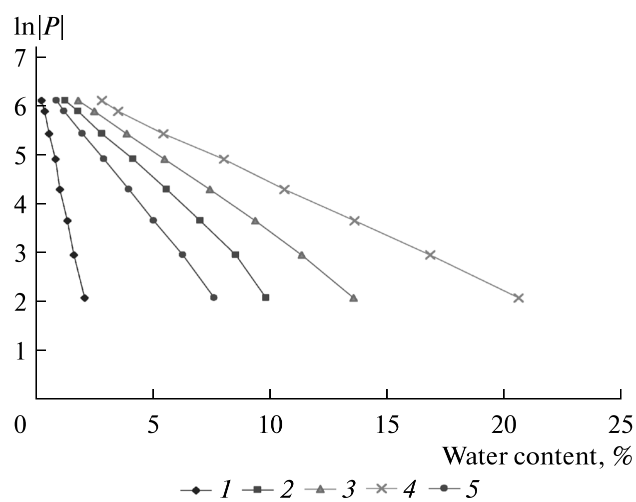
Thus, the values for the total pressure of the first moisture portions adsorbed by the dry soils (A) obtained by independent methods differ by only (–7950) – (–7680) = –270 atm, or 3.4% of their average value. This points to the high accuracy of the hygroscopic method.

The value of parameter B, on the contrary, significantly depended on the size of the elementary soil particles (Table 2). The parameter value regularly increased with their size. When the average effective diameter of elementary soil particles increased by 31 times (from 1.3 to 40 μm), the value of parameter B increased by 10 times. Therefore, for assessing the effect of the size of elementary soil particles on the SMCC, a regular (and relatively simple) relationship between these parameters should be found. Such relationship was revealed between the values of $1/B$ and the logarithms of the average diameters ($\log d$, μm) of elementary soil particles in different particle-size fractions. In the range 40 μm > d > 1.3 μm, it was linear:

$$1/B = 0.048 - 0.026\log d. \quad (10)$$

The coefficient of correlation for this relationship is –0.99 for the significance level below 0.05 [3].

The nonlinear relationship between the values of parameter B and the average diameter of particles in different size fractions can be due to the differences in the mineralogy of different-sized particles. The particles in the finest fractions mainly consist of clay minerals of the montmorillonite group and hydromicas (illite) [1, 8]. The platy shape of their crystals increases their specific surface area, and hydrated exchangeable cations occur in the diffuse layer not only on the surface of the crystals, but also in their interlayer spaces.



Natural logarithms of total soil water pressure modules (ln|P|, where P is expressed in J/g water) as functions of water contents for the light clayey brown forest soil and its particle-size fractions: (*1*) >20 μm; (*2*) 2–6 μm; (*3*) whole soil; (*4*) <2 μm; (*5*) 6–20 μm.

Therefore, the total effective specific surface area of montmorillonite reaches 500 m^2/g. The particles of the coarser size fractions are rounded in shape; they mainly consist of quartz and feldspars, which have no interlayer spaces. Therefore, only the exchangeable cations occurring in the diffuse layer on the external surface of their crystals are hydrated, and the effective specific surface area does not exceed 100–200 m^2/g.

Substituting the B value from Eq. (10) to Eq. (9), we obtain

$$W = (0.048 - 0.026\log d)\,(A - \ln|P_W|)\,. \qquad (11)$$

From Eq. (11), the water content of separate particle-size fractions can be determined at any level of total water potential (pressure) within the hygroscopic range. This information is essential for predicting the water regime of soils [13].

CONCLUSIONS

(1) The coefficient of correlation and parameters of the linear regression relationship between the water contents of separated particle-size fractions and the logarithms of soil water potential (pressure) module (or pF values) were determined in the hygroscopic moisture range of light clayey brown soil. The close correlation between these soil properties is due to the fact that water is bound by exchangeable cations forming the diffuse layer at the electrically charged surface of the soil solid phase.

(2) The coefficient of correlation and parameters of the linear regression relationship between the water contents of the separated size fractions of elementary soil particles and the logarithms of their average diameters were determined. The close correlation between these soil properties can be due to the differences, not only in the specific surface area of different-sized fractions, but also their mineralogy (the clay particles are formed by crystals of clay minerals with the large effective specific surface area, and the coarser particles consist of quartz and feldspars, whose effective specific surface area is significantly smaller).

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