

Original Paper

Effects of occurrence form of soil organic matter on the Atterberg limits and thermal conductivity of clays

Yue Gui¹ , Qingkun Sang² and Jie Yin³ ¹Faculty of Civil Engineering and Mechanics, Kunming University of Science and Technology, Kunming, China; ²Faculty of Tourism Management, Three Gorges Tourism Polytechnic College, Yichang, China and ³Faculty of Civil Engineering and Mechanics, Jiangsu University, Zhenjiang, China

Abstract

Because of the interfacial interactions between mineral soil particles and soil organic matter (SOM), SOM occurs in various forms in the soil, and the mineral-associated and particulate forms are fundamental. Many recent studies have concentrated on the effects of SOM content and type on the geotechnical behavior of soil. However, the influence of SOM occurrence forms is not well understood, nor is there a scientific classification standard for SOM in geotechnical engineering. The main objectives of this study were to explore the effects of SOM occurrence forms on a few physical properties of clays to develop an engineering classification standard of SOM. First, this paper reviews the interfacial interaction mechanism, factors that influence the relation between mineral soil particles and SOM, and the classification method of SOM in soil science. Three predominant clays (montmorillonite, illite, and kaolinite) were then used as the matrix, and three groups of artificial soil samples with different SOM contents (w_u , ranging from 0 to 50% by weight) were prepared by adding peat. A chemical extraction method was used to determine the amount of different forms of SOM. Moreover, the Atterberg limits w_L (w_p) and thermal conductivity λ of artificial soil samples were tested. Based on the experimental results, the relationship between the form of SOM and these physical parameters was established. The experimental results show that the w_L (w_p) vs w_u , and λ vs w_u fitted curves were not monotonic but piecewise linear and could be divided into two straight lines with different slopes; w_u corresponded to the inflection point of w_L (w_p) vs w_u , and λ vs w_u curves were closer to the threshold value $w_{u,2}$. Finally, a simple engineering classification method of SOM is proposed.

Keywords: Adsorption; Atterberg limits; Clays; Engineering classification standard of SOM; Occurrence form of SOM; Thermal conductivity

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Introduction

Soil organic matter (SOM) is the general term for various carbon-containing organic compounds in soil originating from the decomposition residues of animals, plants, and microorganisms (Kleber *et al.*, 2021). In geotechnical engineering, the SOM content in diverse soil types commonly encountered varies widely. For soils rich in SOM, such as peat, the SOM content is greater than 30% and up to 98% (Huat *et al.*, 2011). Generally, except for some light groups dominated by residual fibers, the main component of SOM is humus, which is characterized by significant hydrophilicity, extreme plasticity, and marked compressibility. SOM is an essential factor affecting physical properties such as Atterberg limits, plasticity, and shrinkage, along with mechanical properties such as the strength, deformation, and permeability of soils (Mesri and Ajlouni, 2007; O'Kelly and Zhang, 2013).

Strata containing some organic soil layers, such as dredged materials, silty soil, and peat, are prone to engineering disasters including the excessive settlement or differential settlement of foundations, pit instability, and failure of the cement mixing process

(Den Haan and Edil, 1994; Long *et al.*, 2014; Lau *et al.*, 2019; Özcan *et al.*, 2020; Long *et al.*, 2023). It is generally accepted that SOM in soil, even in small amounts, can act to the detriment of soil engineering properties (Malkawi *et al.*, 1999; Abu-Hamdeh and Reeder, 2000; Develioglu and Pulat, 2019; Hamouche and Zentar, 2020).

Many previous studies (Adejumo, 2012; Malkawi *et al.*, 1999; Hamouche and Zentar, 2020) have confirmed that the Atterberg limits increase linearly with the SOM content. However, because the liquid and plastic limits may increase at different rates with the increase in SOM content, the plasticity index (=liquid limit – plastic limit) may follow a variation law of increasing, decreasing, or constant (Malkawi *et al.*, 1999). In addition, the SOM content is the dominant factor in changes of soil bulk density (Ruehlmann and Körschens, 2009) and the specific gravity of the soil particles decreases with increasing SOM content (Malasavage *et al.*, 2012; Federico *et al.*, 2015; Li *et al.*, 2020; Yu *et al.*, 2017). Develioglu and Pulat (2019) suggested that the specific gravity of dredged soils decreases from 2.74 to 2.53 as the SOM content increases from 0 to 11%.

The presence of SOM affects the micro- and macrostructure of the soil, thus affecting shear strength and stiffness (Dhowian and Edil, 1980; Chakraborty *et al.*, 2014). Charged functional groups in the SOM can lead to cation exchange with the clay interlayer, which results in the formation of an organic coating on the clay surfaces and an alteration in the interfacial frictional regime in

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the soil mass (Bate *et al.*, 2014). Edil and Wang (2000) summarized dozens of experimental datasets and found no significant relevance between the effective internal friction angle (ϕ') and SOM content (w_w) of peaty soil. Develioglu and Pulat (2019) pointed out that the compression indexes (C_c) increase with the SOM content (w_w), ranging from 0.32 to 0.10. In practice, SOM is of various types, both natural and synthetic. The effect of different types of SOM on the engineering properties of soils has been discussed. Zeng *et al.* (2017) took humic acid as an example to illustrate that SOM may decrease the compressibility of reconstituted clays with a wide spectrum of three clay minerals. Zeng *et al.* (2016) conducted an experimental study of the physical properties of clays with organic matter both soluble and insoluble in water. For natural soil, SOM and mineral soil particles are not simply and mechanically mixed. During the long-term accumulation of SOM, the interface interactions between mineral soil particles and SOM result in different occurrence forms of SOM in the soil (Chotzen *et al.*, 2016; Lavallee *et al.*, 2020). Namely, a part of the SOM is adsorbed and covered on the surfaces of the soil particles and forms a mineral-associated SOM, whereas the rest is in a free or particulate form without the adsorption of the soil particles. Although research on the influence of SOM content and type on the engineering properties of soils has been fruitful, few studies have focused on the influence of SOM occurrence forms, and a systematic classification standard for SOM is still lacking.

The first objective of this study was to explore the effects of SOM occurrence forms on a few physical parameters of clays. Three groups, including 42 soil samples with different SOM contents, were subjected to chemical desorption tests of SOM, the Atterberg limits test, and a thermal conductivity test. Based on the testing results, the relationship between the SOM content, the Atterberg limits, and the thermal conductivity coefficient of the soils was clarified, and the relevance of the relation between the SOM occurrence form and the altered law of the above physical parameters of the soils was also revealed. The second objective of this study was to establish a classification standard for SOM in geotechnical engineering according to the influence patterns of the SOM contents and occurrence forms on the engineering properties of soils.

Review of mineral–organic matter interface interactions and SOM classification systems

Interface interactions between mineral soil particles and SOM

The interfacial interactions between mineral soil particles and SOM are incredibly complex. In soil and clay science, research on the interfacial interactions between mineral soil particles and SOM has been fruitful (Wada, 1987; Tombácz *et al.*, 2004; Lagaly *et al.*, 2006; Sutton and Sposito, 2006; Chotzen *et al.*, 2016; Kleber *et al.*, 2021). The mineral soil particles and SOM can form mineral–SOM complexes (Fig. 1) through anion exchange, ligand exchange, hydrophobicity, entropy, cationic bridging, and hydrogen bonding (Wada *et al.*, 1987). The surface of clay particles is usually negatively charged, whereas the SOM has positively charged groups; therefore, the surface of clay particles is covered by a film of SOM under the action of the clay particle's surface electric field. The mineral-associated SOM is referred to as 'bound SOM', and SOM far away from the mineral particles and not affected by adsorption is referred to as 'free SOM'. Generally, the bound SOM is in a layered structure, which can be subdivided into two layers that directly and indirectly interact with clay minerals. The SOM directly interacting with the clay minerals is the first layer of SOM, which is adsorbed on the mineral particles' surface. In fact, the SOM indirectly interacting with the clay minerals is also affected by the adsorption energy of the mineral surface through the interaction between SOM molecules. The SOM closer to the mineral surface is subjected to a stronger adsorption force from the mineral particles. Sollins *et al.* (2006) proposed a "laminated" model to describe the onion-like layered structure formed by SOM on the surface of the mineral soil particles.

Influence factors of mineral–SOM interface interactions

Previous studies have shown that mineral–SOM interface interactions are closely related to mineral types and SOM composition (Tombácz *et al.*, 2004; Schmeide and Bernhard, 2010). Aluminum, iron, and manganese oxides exhibit the most significant adsorption on SOM, followed by clay minerals, whereas quartz and feldspar show the weakest adsorption. The adsorption of clay minerals on the SOM is mainly dependent on the specific surface area, and a

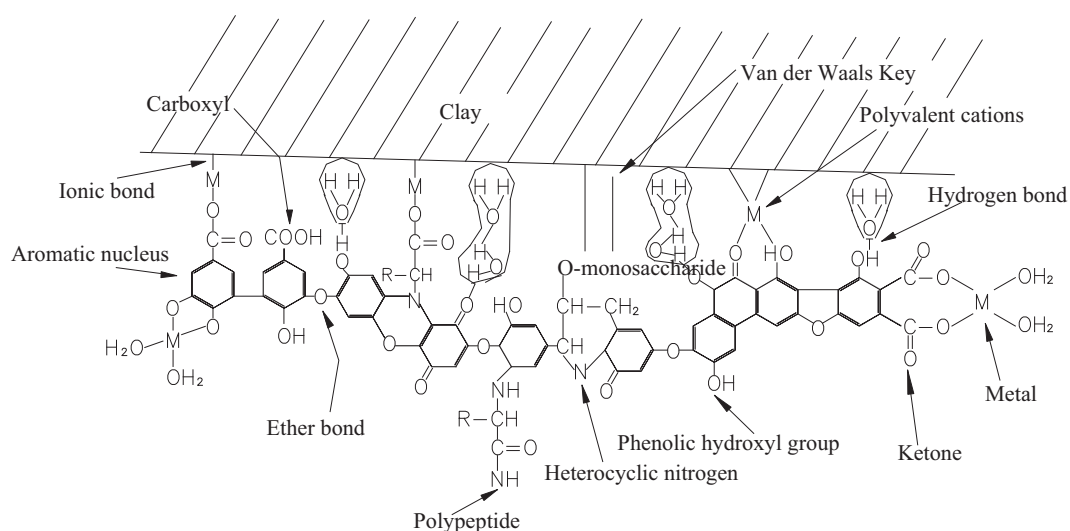


Figure 1. Schematic of mineral–SOM interface interactions.

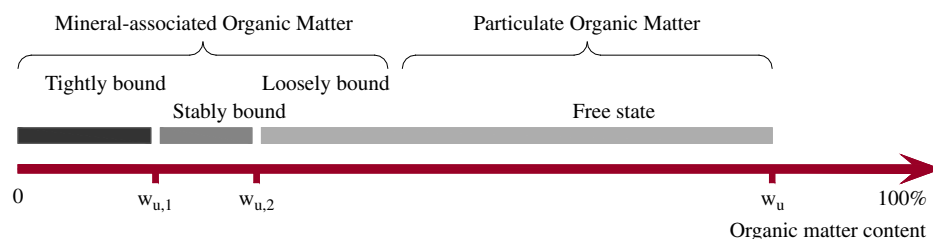


Figure 2. Morphological classification of SOM.

larger specific surface area means a stronger adsorption force. The order of the adsorption capacity of different clay minerals for SOM is montmorillonite > illite > chlorite > kaolinite.

Other factors affecting the mineral–SOM interface interactions include solution pH and ionic strength (Tombácz *et al.*, 2004). Mayes *et al.* (2012) found that the maximum adsorption capacity of SOM gradually decreased with the increase in pH because most of the SOM had ionizable acidic groups. After ionization, the negatively charged SOM and the soils with lower pH generally had more positive charges, which are conducive to the adsorption of SOM.

SOM classification systems in soil science

SOM, especially humic SOM, has a heterogeneous organic macromolecular structure and contains a variety of chemical groups with different physical properties, spatial arrangement, and reactivity (Lehmann and Kleber, 2015). Selective, preferential, and competitive adsorption under the effect of the adsorption energy of mineral soil particles leads to the differentiation of organic molecules. The physical and chemical properties of SOM closest to the mineral soil particles differ from those far away (Ndzana *et al.*, 2022). For example, the density of bound SOM is generally greater than that of free SOM.

In soil science, relatively well-established methods exist for classifying SOM based on the different occurrence forms. Mineral-

associated SOM can be subdivided into three types: tightly, stably, and loosely bound SOM, as shown in Fig. 2. The threshold values of SOM content correspond to $w_{u,1}$ and $w_{u,2}$. Chemical extraction methods, like that one proposed by Fu (1983), are often adapted to determine the threshold values. According to Fu's method, SOM extracted from 0.1 mol L⁻¹ NaOH solution is interpreted as the sum of loosely bound and free ones ($w_u - w_{u,2}$), and the remaining is the sum of tightly and stably bound SOM ($w_{u,2}$). The extract of a 0.1 mol L⁻¹ NaOH + 0.1 mol L⁻¹ Na₄P₂O₇ mixed solution is then interpreted as stably bound SOM ($w_{u,2} - w_{u,1}$), and the rest is tightly bound SOM ($w_{u,1}$). The above operation reflects the difficulty in overcoming the adsorption energy of mineral soil particles by chemical energy. Significant amounts of SOM would be in the free state when the soil is rich in SOM. However, there are no clear limits on or identification of free and loosely bound SOM.

Materials and methods

Clays and SOM

In this study, artificial soil was used to eliminate the adverse effects of the compositional complexity and diversity of natural soils. Three predominant clays, including commercially purchased montmorillonitic clay (Fuchen Chemical Reagent Co., Ltd, Tianjin, China), illitic clay, and kaolinitic clay (Sinopharm Chemical Reagent Co., Ltd, Shanghai, China), were used as the matrices in this project (Fig. 3). Table 1 details the properties and mineral compositions of the clays. The specific gravity of the samples was measured according to ASTM D854 (ASTM, 2014a). The liquid limit and the plastic limit test were conducted following ASTM D4318 (ASTM, 2010).

Amorphous peat was used as a natural source of SOM to change the SOM content of artificial soil. The peat was collected from Kunming City, Yunnan Province, China, on the north shore of Dianchi Lake, at the soil surface layer (10–40 cm). Before mixing it with the matrix clays, a series of preparation procedures was developed as follows: (1) manually remove the residual resin and leaves in the peat with small tweezers; (2) place the peat in a beaker with enough distilled water, and then thoroughly stir it to a slurry followed by the removal of the light SOM floating on the surface of the suspension by an ear-washing ball after resting for ~5 min; (3) pour the slurry into a 1000 mL cylinder, add distilled water to the volume of 800 mL, and then stir repeatedly with a stirring bar until the slurry is uniform; (4) after standing for 5 min, take the suspension from the middle section of the cylinder with a 75 mL pipette, then discard the remaining 200 mL of relatively heavy slurry because it contains more mineral soil particles; (5) dewater the suspension slurry by centrifuging before drying it at room temperature; and (6) grind the dried peat and pass it through a 0.5 mm sieve. After the above treatments, the peat powder's SOM content



Figure 3. Clays and peat.

Table 1. Physical and mineral properties and composition of clays

Items	Properties of testing clays	Montmorillonitic clay	Illitic clay	Kaolinitic clay
Physical properties	Specific gravity, G_s	2.59	2.65	2.55
	Liquid limit, w_L (%)	187.2	88.3	57.9
	Plastic limit, w_p (%)	68.3	42.4	33.7
	Specific surface area, S_c ($m^2 g^{-1}$)	292	34	19.5
Particle-size composition	Silt ($\sim 0.06\text{--}0.002$ mm) (%)	64.5	71.2	69.6
	Clay (<0.002 mm) (%)	32.2	22.6	20.4
Mineral composition percentage	Montmorillonite (%)	62.1	4.3	—
	Illite (%)	4.5	60.3	—
	Kaolinite (%)	—	—	83.9
	Quartz (%)	21.7	26.6	13.1
	Mica (%)	5.2	4.7	—
	Others (%)	6.5	4.1	3.0

(determined by the loss on ignition method) was $>96\%$, and the ash content was $<3\%$.

Soil sample preparation

The clays were dried and then mixed with peat in the required proportions to make the following SOM contents: $w_{cl} = 2.5, 5, 7.5, 10, 12.5, 15, 20, 25, 30, 35, 40, 45,$ and 50% . After the clay and peat were mixed, distilled water was added and the mixed soil samples were stirred thoroughly to a soft, liquid plastic state. The prepared soil samples were then put into sealed plastic bags and placed in a standard maintenance room (at a temperature of $20\pm 2^\circ\text{C}$ and humidity of $>98\%$) for 90 days to allow for a thorough mixture reaction.

Test methods for determining SOM occurrence form in soil samples

A chemical extraction method was used to determine the amounts of SOM in different forms. In the present study, the method proposed by Fu (1983) was adopted, and its flowchart is shown in Fig. 4. The corresponding specific procedure is as follows.

Step 1: Desorption of free and loosely bound SOM

The soil sample was dried at 60°C , then ground and passed through 0.5 mm sieves. A sample of 2.5 g of sieved soil was placed into a 50 mL centrifuge tube (six duplicate samples for each group), which was followed by adding 30 mL of NaOH solution into the centrifuge tube (0.1 mol L^{-1}); the samples were then stirred evenly with a glass rod, and after being kept in an incubator (30°C) for 12 h, they were centrifuged at 3000 rpm for 15 min. If the suspension was turbid, a small amount of saturated Na_2SO_4 solution was added and mixed, then allowed to stand for 10 min before being centrifuged again. After centrifugation, the supernatant was poured out, and the NaOH solution was added to the remaining soil samples. The above treatment was repeated until the extraction solution was nearly colorless. After then the distilled water was added into the extraction solution and mixed with a glass rod, and the solution was washed till its pH was close to 7.0 by centrifugation. After extraction, the soil sample was removed from the centrifuge tube in which the free and loosely bound SOM was fully desorbed.

Step 2: Desorption of stably bound SOM

After Step 1, 2.5 g of soil was placed in a 50 mL centrifuge tube (two duplicate samples). Next, 30 mL of the mixed solution of 0.1 mol L^{-1} NaOH and 0.1 mol L^{-1} $\text{Na}_4\text{P}_2\text{O}_7$ (with a pH value of ~ 13) was added, and the mixture was stirred evenly with a glass rod. It was then placed in a 30°C incubator for >12 h before being centrifuged at 3000 rpm for 15 min. If the suspension was turbid, a little saturated Na_2SO_4 solution was mixed in and then left to stand for 10 min before being centrifuged once more. The supernatant was poured away after centrifugation, and 0.1 mol L^{-1} NaOH and 0.1 mol L^{-1} $\text{Na}_4\text{P}_2\text{O}_7$ were added to the remaining soil samples. The aforementioned repeated treatments were applied until the extraction solution was nearly colorless (Fig. 4), at which point some distilled water was added to the extraction solution and stirred well with a glass rod. Next, centrifugation was used to wash the extraction solution until it was pH neutral. Finally, the soil sample was taken out of the centrifuge tube, where the stably bound SOM was fully desorbed.

Step 3: Determination of SOM content with different occurrence forms

After treating the soil samples following Steps 1 and 2, the samples were placed in the blast drying box at 60°C before they were ground and screened. The SOM content thresholds for the soil samples could then be determined using the loss on ignition method per ASTM D2974 (ASTM, 2014b).

Atterberg limit and thermal conductivity tests

One challenge of this project was to identify the relevant parameters from the range of indicators characterizing the soil engineering properties that are sensitive to the changes in the SOM occurrence form. Based on the pre-study, Atterberg limits and thermal conductivity were identified as the relevant parameters, because they can readily demonstrate the way SOM occurrence forms affect the soil's physical properties. It is well known that the Atterberg limits are the fundamental index of soil properties and that thermal conductivity can reflect the macrostructural characteristics of soils. Therefore, assessing the effects of the SOM content and SOM

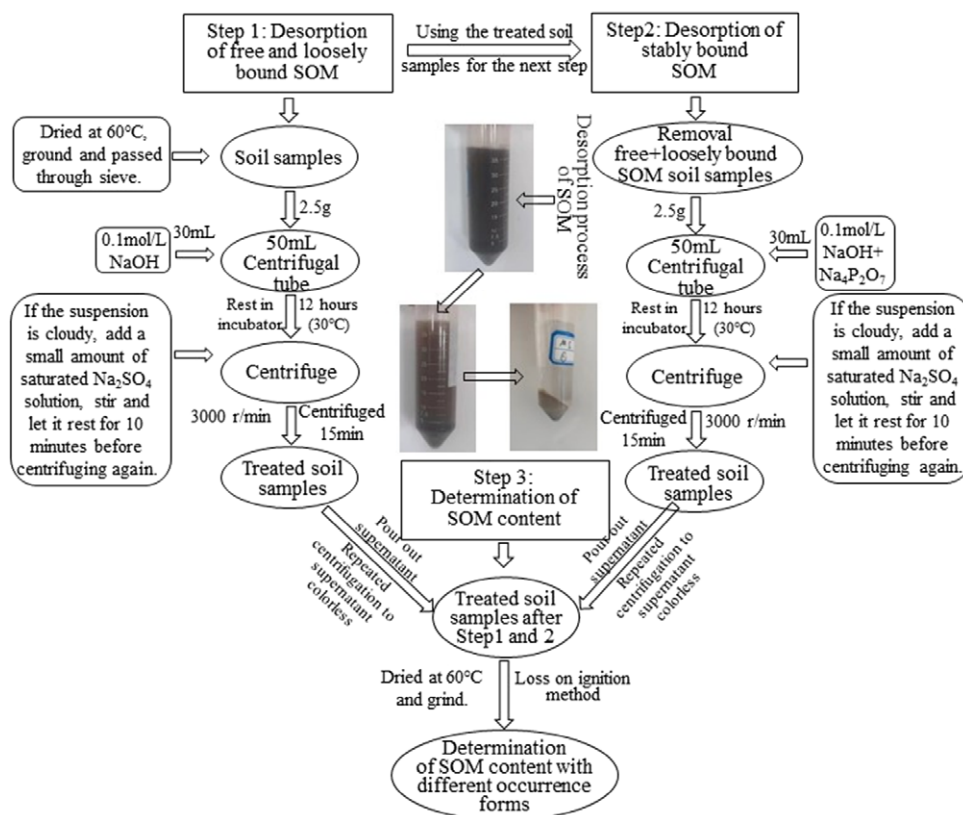


Figure 4. Flow diagram of desorption of SOM.

occurrence forms on these relevant physical parameters allows mechanical behavior to be anticipated.

The Atterberg limit tests were conducted in accordance with ASTM D4318 (ASTM, 2010). Through thermal conductivity tests, the thermal conductivity coefficient (λ) of the soil samples was measured with a KD2 Pro thermal analyzer (Decagon) that consists of a controller and a dual probe. The probe (1.3 mm in diameter and 30 mm in length with a dual-probe spacing of 6 mm) can complete the test quickly and stably. All tested soil samples were saturated and had a constant dry density ($\rho_d = 1.3 \text{ g cm}^{-3}$) and were statically pressed in layers in a mold (39.1 mm inner diameter and 40 mm height); the specimens were sealed and placed in a standard curing room for 24 h before testing. A thin layer of heat-conductive silicone grease was applied to the surface of the probe to ensure good contact between the probe and the specimen, to reduce the gap between the probe and the holes (drilled by an electric hand drill to facilitate the probe insertion), and to eliminate test error. In addition, each sample was measured several times on different parts, and the average values were taken as the final experimental data.

Results and Discussion

Adsorption characteristics of clays on SOM

The relations between the amounts of different SOM occurrence forms and the SOM content of three clays are shown in Fig. 5. As the SOM content increased, the adsorption amount of tightly and stably bound SOM increased gradually and tended to stabilize. This variation law indicates that under interfacial interactions, the adsorption of different forms of SOM was gradually saturated. The corresponding saturated adsorption capacity of three clays

can be obtained intuitively from Fig. 5. For example, the saturated adsorption capacity of montmorillonitic clay was 11.5% for tightly bound SOM and 6.1% for stably bound SOM.

The saturated adsorption capacity of tightly bound SOM corresponds to the threshold $w_{u,1}$, and the total saturated adsorption capacity of tightly and stably bound SOM corresponds to $w_{u,2}$. When the SOM content in the soil is $< w_{u,1}$, most of the SOM is tightly bound. SOM mainly exists in a tightly and stably bound form when the SOM content does not exceed $w_{u,2}$. When the SOM content is $> w_{u,2}$, loosely bound and free SOM occurs in the soil. As shown in Fig. 5, the threshold values $w_{u,1}$ and $w_{u,2}$, were 11.5% and 17.6% for montmorillonitic clay, 6.8% and 10.4% for illitic clay, and 5.0% and 7.5% for kaolinitic clay, respectively. This demonstrates that the thresholds ($w_{u,1}$ and $w_{u,2}$) are related to the types of clay minerals.

Effect of SOM content and occurrence form on Atterberg limit of clays

Figure 6 shows the relations between the Atterberg limits (liquid limit, w_L , and plastic limit, w_p) and the SOM content, w_u . The w_L and w_p of all three clays showed an increasing trend as w_u increased. As the SOM content increased, w_L (w_p) increased slowly at low levels of SOM content, w_u , whereas it increased quickly when w_u exceeded a particular value. Therefore, the variation pattern of Atterberg limits with SOM content can be roughly divided into Stages I and II. Accordingly, the variation of the Atterberg limit with the SOM content is not monotonous but a segmented linear relationship.

As shown in Fig. 6, the slope (i.e. slope = Δw_L (Δw_p)/ Δw_u) of the fitted line represents the increment of w_L (w_p) for a 1% increase of

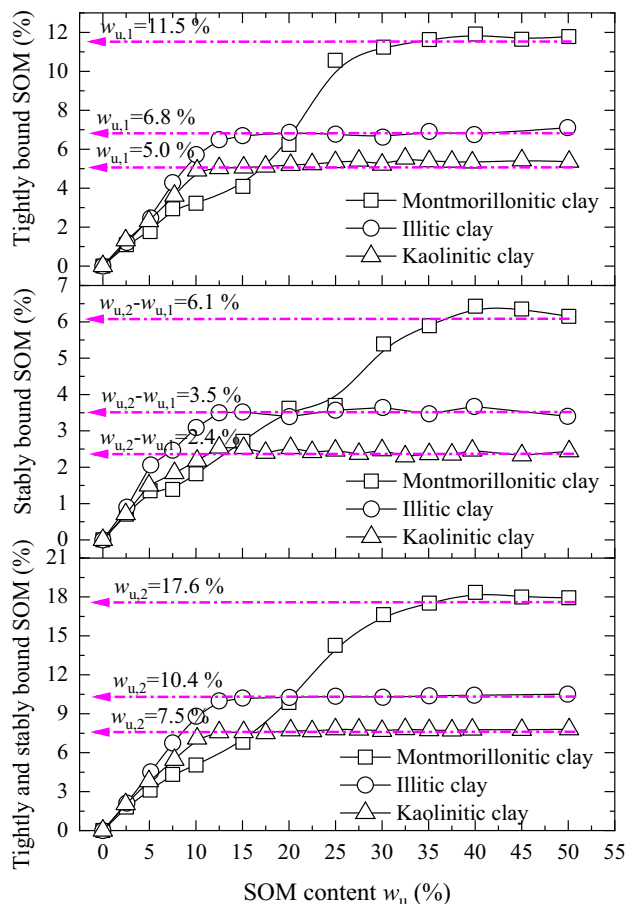


Figure 5. Variation of amounts of different SOM occurrence forms with the SOM content.

w_u in the soil. In Stage I, the slopes of w_L vs the w_u fitted lines were 1.62, 1.41, and 1.76 for montmorillonitic, illitic, and kaolinitic clay, respectively, and the slopes of w_p vs the w_u fitted lines were 0.78, 0.54, and 1.11 for montmorillonitic, illitic, and kaolinitic clay, respectively. In Stage II, the slopes of w_p vs the w_u fitted lines were 2.76, 2.44, and 2.83, for montmorillonitic, illitic, and kaolinitic clays, respectively, and the slopes of w_p vs the w_u fitted lines were 4.0, 3.67, and 3.08, respectively. The w_u at the inflection points of the two stages is related to the clay types, and its value was about 20% for montmorillonitic clay, 11% for illitic clay, and 10% for kaolinitic clay.

Previous studies (Husein *et al.*, 1999; Adejumo, 2012; Develioglu and Pulat, 2019; Hamouche and Zentar, 2020) had concluded a positive linear correlation between the Atterberg limit (w_L and w_p) and the SOM content. As shown in Table 2, the slopes of the linear relationship fitted from their experimental data are of the same order of magnitude as in this study. Notably, they studied soils with low SOM content (mostly below 15%), which should explain the absence of a fitted relationship with a bi-linear line similar to the present paper.

It is generally recognized that the presence of SOM leads to an increase in the Atterberg limit of soils, which is the water-binding capacity of soils (Booth and Dahl, 1986; Rashid and Brown, 1975; Develioglu and Pulat, 2019; Hamouche and Zentar, 2020). SOM particles are highly colloidal, and their surface area and water adsorptive capacity were far in excess of those exhibited by clay minerals (specifically, kaolinite and illite; Husein *et al.*, 1999). In

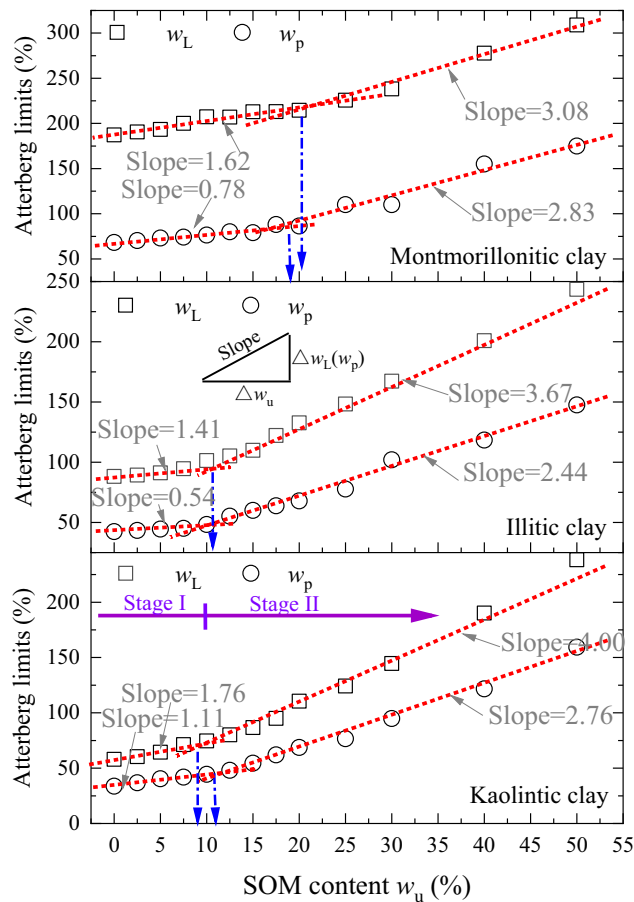


Figure 6. Variation of Atterberg limits with SOM content.

Stage II, the slopes of the fitted straight lines were greater than those in Stage I, indicating that the SOM in Stage II had a much greater adsorption capacity.

Effect of SOM content and occurrence form on thermal conductivity of clays

Figure 7 shows the relation between thermal conductivity λ and SOM content, w_u . It can be seen that λ decreases as w_u increases, and the change process of λ with w_u can be divided into two stages, steep reduction (Stage I) and gentle reduction (Stage II). The slope (i.e. slope = $\Delta\lambda/\Delta w_u$) of the fitted straight line reflects the decrement in λ due to a 1% increment in w_u . It can be illustrated that the absolute value of the slope of the fitted straight line in Stage I (0.032–0.006) was greater than that in Stage II (0.014–0.001), indicating that the decreasing rate of λ with increasing w_u in Stage I was much larger than that in Stage II. Additionally, the value of w_u corresponding to the intersection point of the two stages was also dependent on the types of clay minerals. The value of w_u corresponding to the intersection point of the two stages was $\sim 17.5\%$ for montmorillonitic clay, $\sim 10.0\%$ for illitic clay, and $\sim 8.0\%$ for kaolinitic clay.

There are relatively few reports in the literature on the fitted relationship between the SOM content and the thermal conductivity of soils (Table 3). Ekwue *et al.* (2006) tested the thermal conductivity, λ , of three types of soils (i.e. sandy loam, clay loam, and clay) with different SOM contents (w_u mostly $< 15\%$), and a similar linear relationship between λ and w_u was obtained. With

Table 2. Predictive equations of w_L (w_p) from SOM content w_u

Type of soils	SOM content w_u /%	Regression predictive equation	R^2	Samples
Inorganic material + industrial compost	~0–15%	$w_L=1.89 \times \{OM(\%)\} + 25.13$	0.99	Hamouche and Zentar (2020)
		$w_p=1.80 \times \{OM(\%)\} + 16.70$	0.95	
Dredged marine sediments		$w_L=3.08 \times \{OM(\%)\} + 15.16$	0.96	
		$w_p=3.00 \times \{OM(\%)\} + 4.32$	0.96	
dark spongy/organic clay soils	~0–8.6%	$w_L=1.02 \times \{OM(\%)\} + 42.0$	0.98	Adejumo (2012)
		$w_p=0.52 \times \{OM(\%)\} + 25.1$	0.97	
Dredged material	~0–11	$w_L=0.61 \times \{OM(\%)\} + 31.59$	0.79	Develioglu and Pulat (2019)
		$w_p=0.41 \times \{OM(\%)\} + 26.26$	0.98	
Illitic soils + peat	~0–30%	$w_L=0.68 \times \{OM(\%)\} + 81.40$	0.70	Malkawi <i>et al.</i> (1999)
		$w_p=1.10 \times \{OM(\%)\} + 44.50$	0.95	
Montmorillonitic clay + peat	~0–20%	$w_L=1.62 \times \{OM(\%)\} + 156.8$	0.97	This study
		$w_p=0.78 \times \{OM(\%)\} + 62.73$	0.95	
	~20%–50%	$w_L=3.08 \times \{OM(\%)\} + 150.8$	0.96	
		$w_p=2.83 \times \{OM(\%)\} + 22.23$	0.95	
Illitic clay + peat	~0–11%	$w_L=1.41 \times \{OM(\%)\} + 74.8$	0.97	
		$w_p=0.78 \times \{OM(\%)\} + 62.73$	0.95	
	~11–50%	$w_L=3.67 \times \{OM(\%)\} + 49.8$	0.96	
		$w_p=2.44 \times \{OM(\%)\} + 26.3$	0.95	
Kaolinitic clay + peat	~0–10%	$w_L=1.76 \times \{OM(\%)\} + 50.42$	0.97	
		$w_p=1.11 \times \{OM(\%)\} + 27.47$	0.95	
	~10–50%	$w_L=4.00 \times \{OM(\%)\} + 26.8$	0.96	
		$w_p=2.76 \times \{OM(\%)\} + 18.83$	0.95	

clay loam as the matrix, although peat moss was used to increase the SOM content, Abu-Hamdeh *et al.* (2000) observed that increasing the soil organic matter (SOM) content, denoted as w_u , resulted in a decrease in thermal conductivity (λ). Their findings also revealed an inflection point on the λ versus w_u curve at approximately $w_u = 20\%$. This inflection point is consistent with the experimental results obtained in this study. However, they did not analyze this phenomenon in detail, so the result may be due to the small amount of experimental data.

Analysis of experimental data and related mechanisms

The main test results and related analytical data mentioned above are shown in Table 4. The following main conclusions were drawn: (1) For the three types of clays, the slope ratios (S_2/S_1) of the segmented fitted straight lines of the w_L vs w_u curves ranged from 1.90 to 2.60; the slope ratios of the w_p vs w_u curves were in the range of 2.49 to 4.52 for the slope ratios of the w_p – w_u relationship curves; and the slope ratios (S_1/S_2) of the λ vs w_u curves varied from 2.29 to 6.00. (2) The w_u corresponding to the inflection point on the w_L (w_p) versus w_u curve was similar to that on the λ – w_u curve. (3) The threshold values of $w_{u,2}$ were very close to the w_u corresponding to the inflection point. The w_u corresponding to the inflection point of the λ vs w_u curve was closer to $w_{u,2}$ than that of the w_L (w_p) vs w_u curve.

Summarizing the above experimental data, it can be readily deduced that the macroscopic physical properties of clays are not only related to the SOM content but are also significantly influenced by the SOM occurrence form. Based on the adsorption characteristics of SOM on clays and referring to the microstructure of SOM-rich soils such as peaty soils, a model of the transformation of SOM occurrence forms in clays (Fig. 8) was established as a means to explore the mechanism of the variation patterns of the above-mentioned experimental data.

- (i) Atterberg limits reflect the ability of the soil to adsorb water. SOM, especially humus, is an amorphous polymer organic compound with a huge specific surface area and hydrophilic groups. On a mass fraction basis, the specific water adsorption capacity of SOM was 10 to 40 times that of soil minerals (Wang *et al.*, 2011), and the maximum water absorption could exceed 500% of its weight. Therefore, as the SOM content increases, the Atterberg limits of the soil also increase. However, because of the adsorption of mineral soil particles, the capacity of bound SOM to adsorb water is not as strong as that of loosely bound and free SOM. This is the reason that the w_L (w_p) vs w_u curve was phased around $w_u = w_{u,2}$. According to the experimental results (S_2/S_1), the adsorption capacity of loosely bound and free SOM on water was roughly 1.90 to 4.52 times that of tightly and stably bound SOM.

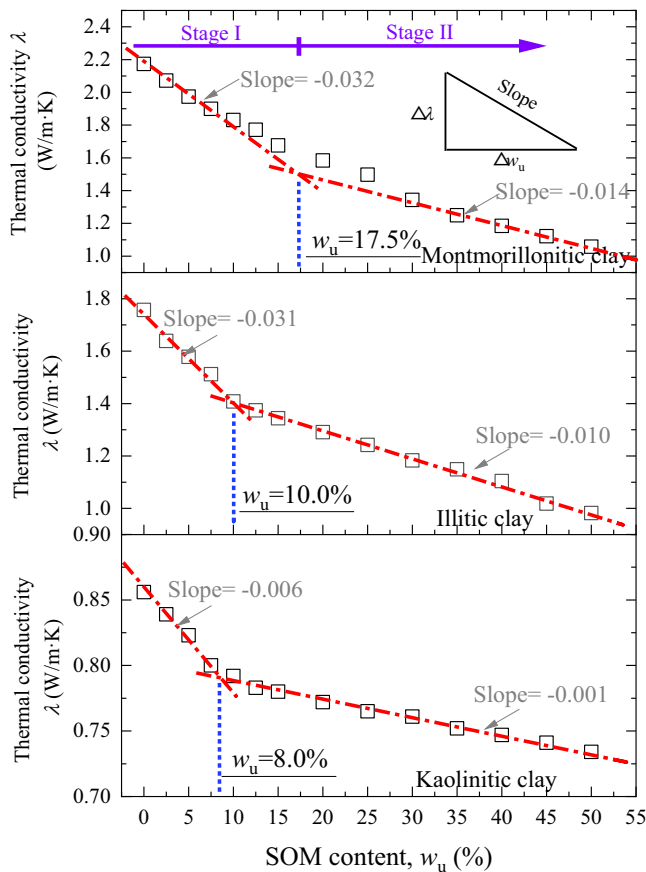


Figure 7. Changes in thermal conductivity with SOM content.

- (ii) The thermal conductivity, λ , of soils is determined by the thermal conductivities of the solid, liquid, and gas single-phase substances in the soil and the proportional relationship of each phase component (Abu-Hamdeh *et al.*, 2000; Xu *et al.*, 2019; Keskin *et al.*, 2023). As far as single-phase substances are concerned, the λ of mineral particles is several times that of water and nearly 100 times that of air, and the λ of SOM is about 0.44 times that of water, i.e. $\lambda_{\text{mineral}} > \lambda_{\text{water}} > \lambda_{\text{OM}} > \lambda_{\text{air}}$ (Cheshire *et al.*, 2000; Zhao *et al.*, 2019). The

thermal conductivity λ of soils decreases because the proportion of mineral soil particles in the soil decreases correspondingly as the SOM content increases (Fig. 7). Mineral soil particles conduct heat through point and surface contacts. When the SOM content w_u is in the range of 0 to $w_{u,2}$, the tightly and stably bound SOM covers the surface of mineral soil particles (Fig. 8) like an insulation layer. The SOM, especially in the loosely bound and free states, has a spongy porous structure and a high light texture; thus, when $w_u > w_{u,2}$, the mineral soil particles are wrapped into clumps by the SOM (Fig. 8). Therefore, when the SOM content w_u increases from 0 to exceeding $w_{u,2}$, the solid phase thermal conductivity of the soil changes from mineral soil particles to SOM. This is the reason for the different slopes of the fitted straight lines in Stage I and Stage II (Fig. 7).

Engineering classification of SOM based on its occurrence form

Understanding the effects of SOM occurrence forms on the physical properties of soils is the first step to proposing a scientific engineering classification standard of SOM. In this section, the engineering classification method of SOM is discussed.

When the classification of the SOM is overly subdivided, as in soil science, its testing process is complicated and is not utilized to analyze engineering problems. We suggest that the SOM in the soil should be divided into only two categories: bound and free SOM (Fig. 9). The limited value (w_j) between the two types of SOM is equivalent to the threshold value $w_{u,2}$. Specifically, the bound SOM includes the tightly and stably bound states of SOM whereas the free SOM includes the loosely bound and free states of SOM.

With the establishment of the SOM engineering classification system, the analysis methods for soils containing SOM must also be adjusted accordingly. Generally, soils containing SOM consist of a mineral phase, an organic phase, a liquid phase, and an air phase.

When the SOM content of the soil is low, the SOM is predominantly bound SOM. Although the presence of bound SOM degrades the engineering properties of mineral soil particles, the properties of soil containing bound SOM are still similar to mineral-based ones. Therefore, bound SOM does not need to be divided into independent phases and can be seen as part of the mineral soil particles. It is also reasonable to consider the three-

Table 3. Predictive equations of thermal conductivity from the SOM content

Type of soils	OMC w_u /%	Regression predictive equation	R^2	Samples
Sandy loam	~1.7–12.2%	$\lambda = -0.059 \times \{OM(\%)\} + 2.04$	0.97	Ekwue <i>et al.</i> (2006)
Clay loam	~4.7–14.9%	$\lambda = -0.049 \times \{OM(\%)\} + 1.78$	0.98	
Clay	~2.7%–13.1%	$\lambda = -0.019 \times \{OM(\%)\} + 0.99$	0.98	
Clay loam + peat moss	~5–30%	$\lambda = -0.006 \times \{OM(\%)\} + 0.35$	0.96	Abu-Hamdeh <i>et al.</i> (2000)
Montmorillonite + peat	~0–11%	$\lambda = -0.032 \times \{OM(\%)\} + 2.15$	0.99	This study
	~11–50%	$\lambda = -0.014 \times \{OM(\%)\} + 1.74$	0.99	
Illite + peat	~0–10%	$\lambda = -0.031 \times \{OM(\%)\} + 1.74$	0.98	
	~10–50%	$\lambda = -0.010 \times \{OM(\%)\} + 1.49$	0.99	
Kaolinite + peat	~0–10%	$\lambda = -0.006 \times \{OM(\%)\} + 0.86$	0.96	
	~10–50%	$\lambda = -0.001 \times \{OM(\%)\} + 0.79$	0.98	

Table 4. Statistics and analysis of test results

Soil type	Types of curves	Slope of the fitted straight lines		Slope ratio		SOM content at intersection point w_u /%		Threshold values	
		Stage 1 (S_1)	Stage 2 (S_2)	S_2/S_1	S_1/S_2	$w_{u,1}$ /%	$w_{u,2}$ /%		
Montmorillonitic clay	$w_L \sim w_u$	1.62	3.08	1.90	—	20.0	—	11.5	17.6
	$w_p \sim w_u$	0.78	2.83	3.63	—	18.0	—		
	$\lambda \sim w_u$	-0.032	-0.014	—	2.29	—	17.5		
Illitic clay	$w_L \sim w_u$	1.41	3.67	2.60	—	11.2	—	6.8	10.4
	$w_p \sim w_u$	0.54	2.44	4.52	—	11.0	—		
	$\lambda \sim w_u$	-0.031	-0.010	—	3.10	—	10.0		
Kaolinitic clay	$w_L \sim w_u$	1.76	4.00	2.27	—	9.0	—	5.0	7.5
	$w_p \sim w_u$	1.11	2.76	2.49	—	11.0	—		
	$\lambda \sim w_u$	-0.006	-0.001	—	6.00	—	8.0		

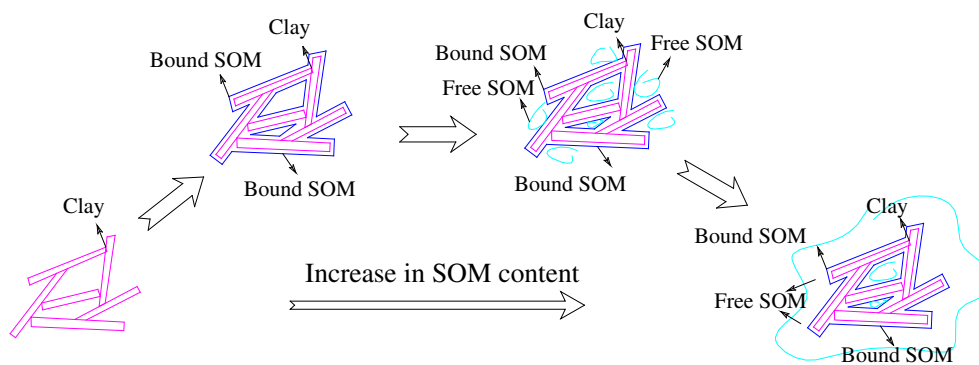
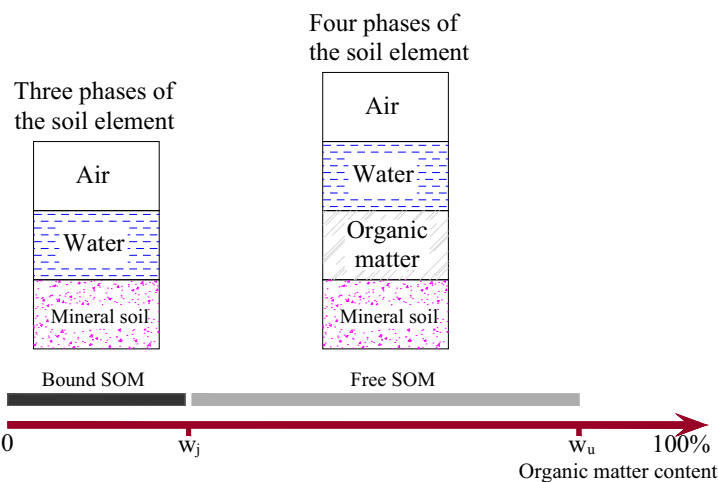


Figure 8. Sketch of the transformation of SOM occurrence form in clays.



1) when the SOM content of the soil is low, the SOM is predominantly bound SOM. Although the presence of bound SOM degrades the engineering properties of mineral soil particles, the properties of soil containing bound SOM are still similar to mineral-based ones. Therefore, bound SOM doesn't need to be divided into independent phases and can be seen as part of the mineral soil particles.

2) when the SOM content is high, the SOM is mainly in the free state except for some that is bound; confusing SOM with mineral soil is inappropriate because of SOM's natural properties, especially the loosely bound or free states, which are closer to colloidal material. Therefore, it is more reasonable to establish a four-phase system, i.e., mineral soil, organic matter, water, and air, to analyze organic-rich soils such as peaty soil or peat.

Figure 9. Engineering classification of SOM.

phase system suitable for containing bound SOM soil, a common practice nowadays.

However, when the SOM content is high, the SOM is mainly in the free state except for some that is bound; confusing SOM with mineral soil is inappropriate because of SOM's natural properties, especially the loosely bound or free states, which are closer to colloidal material. For example, the shape and volume of SOM would change under a load, which is incompatible with the assumption of Terzaghi's one-dimensional consolidation theory that the solid phase cannot be compressed. Therefore, it is more reasonable to establish a four-phase system (Fig. 9), i.e. mineral soil, organic matter, water, and air, to analyze organic-rich soils such as peaty soil or peat. Landva and Pheeney (1980) analyzed peat's unique multi-phase component characteristics, and Kogure *et al.* (1993) proposed a conceptual model of the multi-phase system for peat soils. However, it is worth noting that the progress from a three-phase to a four-phase system, the complexity of the components, and the multi-phase interface interactions coupling can also make the research significantly more difficult.

To eliminate as much as possible the incredible complexity and variability in the composition of soils that naturally contain SOM, artificial soils were used in this paper. However, two drawbacks need to be noted: (1) the accumulation process of SOM in natural soils occurs over an extremely long period of geological history, and it is not easy or not even possible to fully simulate the process manually; and (2) natural sedimentary soils have a certain degree of primary anisotropy due to the directional arrangement of particles during deposition, and the adsorption of SOM by mineral soil particles is not as uniform as that of artificial soils. Overall, a comprehensive and realistic artificial simulation of the adsorption of SOM by mineral soil particles is impossible. However, the work in this paper is still relevant to understanding the influence of SOM occurrence forms on the engineering properties of soil.

Conclusions

This paper presents an experimental investigation into the effect of SOM occurrence forms on the Atterberg limit and thermal conductivity, and a simple engineering classification method for SOM was proposed based on the experimental results and mechanism analysis. The main conclusions can be summarized as follows:

- (1) The Atterberg limits (w_L and w_p) did not increase monotonically with increasing SOM content w_u of the soils, but had a piecewise-linear relationship, and the fitted curves could be divided into two straight lines with different slopes.
- (2) With the increase in SOM content, w_u , the decrease in thermal conductivity, λ , follows two linearly decreasing phases: steeply decreasing and gently decreasing. The mechanism is that when w_u exceeds the threshold $w_{u,2}$, the SOM occurrence form changes from the bound state to the free state, and the main subject of the soil solid phase heat transfer path changes from mineral soil to organic matter.
- (3) The macroscopic physical properties of clays are not only related to the SOM content but are also significantly influenced by the SOM occurrence form.
- (4) It has been suggested that SOM can be divided into bound and free states, and the limit value of SOM content is approximately equal to the threshold value $w_{u,2}$. It is more reasonable to establish a four-phase system of mineral soil, organic matter, water, and air to analyze organic-rich soils such as peaty soil and peat.

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Competing interests. The authors declare that they have no competing interests.

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