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Inorganic Composition and Physico-Chemical Properties of the Peloid of the Salt Lake Moiyldy (Kazakhstan) As a Natural Source of Biologically Active Substances

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The purpose of the presented in this paper research is to give the physicochemical and morphological characterization of one of the most popular among Kazakhstani people peloid of Moiyldy lake. The results could lead to a more complex use of their potential as a self-healing source of biologically active components of natural origin as an environmentally safe material. A review of the available studies of natural mud in Kazakhstan has shown the absence of systematic studies of the composition and structure of peloids in the North-Eastern part of the country. For the first time, a comparative analysis of the data obtained with previous studies of peloids of other Kazakhstan regions was carried out. The results indicate that the studied samples of therapeutic mud have a medium mineralized, slightly alkaline continental origin with an insignificant content of humic carbon (3.861 %). It is characterized by an increased concentration of ammonium (253,849 mg kg^{-1}), which is the biologically active component that has anti-allergic and anti-inflammatory effects. Based on the elemental content, it is concluded that elements whose gross concentrations exceed the accepted values are bismuth (41.4 times), tantalum (10.6 times), and cesium (8.0 times). The mineralogical and elemental analysis results demonstrate diverse composition that allows biologically active elements synergistically empower their effect and expand the scope of the therapeutic application of peloid in the future.

1. Introduction

Recently, especially in the condition of quarantine measures related to the COVID-19 pandemic, interest in local mud springs (peloids) of natural origin as a therapeutic and affordable remedy has increased in Kazakhstan. Peloid is a natural product consisting of a mixture of a salty lake or mineral healing water (liquid phase) with organic and inorganic components (solid phase) obtained as a result of biological (humus) and geological effects (clay minerals), which is applied locally as a therapeutic agent in the form of application (Antonelli and Donelli, 2018). A review of the available data shows the lack of systematic scientific research on natural mud lakes in the North-Eastern region of Kazakhstan, which includes Moiyldy Lake. It contains a layer thickness of 0.6 m and a volume of about 150,000 m^3 of therapeutic natural mud, which indicates the relevance of their study to unlock the full potential of this natural and available raw material. The literature review represented numerous pieces of evidence about the indirect positive impact of peloid on the immune and inflammatory response of the body through the skin microbiome (Antonelli and Donelli, 2018), a powerful anti-inflammatory, moisturizing, rejuvenating effect (Spilioti et al., 2017), the strengthening antibacterial effect of some medicines (Arthur et al., 2020). Diverse chemical composition with a physiological activity allows scientists to assume that the first living beings did not arise in the aquatic environment but in the mud itself (Vadlja et al., 2022). The peloid of Moiyldy has been popular among the local population since ancient times for its healing properties.
The small amount of data about their composition and their informal and advertising nature demonstrates the insufficiency of research on this mud and demonstrates its relevance. For the first time, an investigation of the

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mineral composition of Moiyldy sapropel was carried out. The purposes of this study were to determine the physicochemical characteristics and textural features as well as to give a general evaluation of the composition and structure and the predicted physiological effect of native Moiyldy mud. Research in the field of mud resources of the regions of North-Eastern Kazakhstan will allow obtaining effective medicines and cosmetics based on natural physiological raw materials.

2. Methods and materials

2.1 Sampling

Samples were taken by point sampling up to 20 cm deep in one layer every 5 m in the radius of the source of Moiyldy Lake (52°23'48"N 77°04'03"E) and were further quartered according to PND F 12.1:2:2.2:2.3:3.2-03. The GPS location during sampling was determined using the Google Maps application. Sampling was in the autumn period (September 2022). The averaged samples were stored in clean polyethylene containers with a tight lid at a temperature of 4 °C in a dark place.

2.2 Methods used to determine the physicochemical parameters and the mineralogical and elemental composition of the sample.

Volumetric mineralogy was determined by X-ray diffraction (XRD) using an X-ray diffractometer of modular design X'Pert High Score manufactured by PANalitical (Netherlands). The analyses were in the range of diffraction angles 2θ from -12° to +140° with a minimum scanning step of 0.001°. The data of diffractograms were decrypted using the Crystallography Open Database (COD) and Inorganic Crystal Structure Database (ICSD) card files. The granulometric composition of the mud according to the pipette method was determined (GOST 12536-2014, 2019). To study the surface microrelief and particle size distribution (PSD), a low-vacuum analytical scanning electron microscope (SEM) manufactured by JEOL Ltd. (Japan) with the INCA EnergyPenta FET X3 energy dispersive microanalysis system of OXFORD InstrumentsAnalyticalLimited (UK) microscope was used. ImageJ software has been used to calculate particle size distribution. The construction of size distribution curve was with help of the OriginPro 2018 program. The quadrupole mass spectrometer with inductively coupled plasma (ICP-MS) Agilent 7500cx manufactured by Agilent Technologies (USA) was used to determine the gross concentrations of elements. Measurement of the pH value of the peloid was provided by using a two-channel S47 SevenMulti pH/conductivity meter (Russia) equipped with an InLabExpertPro pH electrode with a measurement accuracy of ±0.001 (GOST 26423-85, 2011). The maximum hygroscopic moisture of the peloid was identified by saturation of the sample with vaporous moisture, followed by the determination of soil moisture (GOST 28268-89, 2006). Concentrations of mobile forms of nitrate nitrogen were determined using the extraction of nitrates with a 1M solution of potassium sulfate with a ratio of 1:2.5 of the mass of the mud sample and the volume of the solution and subsequent determination of nitrates in the extract using the method of potentiometry (GOST 26951-86, 1986). Phosphorus and potassium ions were determined according to the modified Machigin photometric method at a red light filter with a wavelength of 710 nm to determine phosphorus and a wavelength of 770 nm to determine potassium (GOST 26205-91, 1992). The extraction of mobile sulfur from the sample was conducted with a solution of potassium chloride, followed by precipitation of sulfates with barium chloride and subsequent turbidimetric determination of them in the form of barium sulfate, in which optical density of the suspension was measured. Soluble starch as a suspension stabilizer is used (GOST 26490-85, 1985). Sodium, potassium, ammonium, calcium, and magnesium cations were determined by capillary electrophoresis "KAPEL-104T" on the software "Elforan" according to PNDF 16.1:2:2.2:2.3.74-2012; chloride ions, sulfate ions in aqueous extract according to PNDF 16.1:2:2.3:2.2.69-10. The content of total organic carbon, carbon of humic acids, and carbon of fulvic acids in the aqueous extracts by the pyrophosphate express method was determined.

3. Results and discussion

The mud sample is a jelly-like homogeneous mass of dark gray color without large inclusions of organic residues, having an oily sheen and the smell of hydrogen sulfide. According to the obtained physicochemical indicators (Table 1), this peloid is characterized as slightly alkaline, highly mineralized, with a low content of humic carbon, according to the classification given by Bokuchava (2009).

The analysis of the granulometric and mineral composition of the peloid shows its close relationship with the physicochemical properties and the aggregate state. The highest concentration of the silt fraction (<0.001 mm) and the absolute content of non-clay minerals (Table 2) in the sample indicate that the quartz contained has mostly a fine-grained structure.

Table 1: Physico-chemical parameters of Moiyldy mud

Parameter	Value	Parameter	Value
pH	8.973±0.088	Granulometric composition, %	
Maximum hygroscopic moisture, %	27.820±0.380	1-0.25 mm	6.533
Mass fraction of the dense residue, %	10.000±0.053	0.25-0.05 mm	2.317
Group composition of humus, %		0.05-0.01 mm	11.395
C total	0.0390	0.01-0.005 mm	3.861
C (humic acids)	0.0012	0.005-0.001 mm	5.745
C (fulvic acids)	0.0378	<0.001 mm	70.150

Table 2: Phase mineralogical composition of the Moiyldy mud

Card number in the COD and ICSD database	Name of the compound	Chemical formula	Semi-quantitative analysis data, %
98-009-0145	Quartz low	SiO_2	45.3
98-001-8166	Calcite	$CaCO_3$	23.5
98-018-1148	Halite	NaCl	29.3
98-009-6074	Hematite	Fe_2O_3	1.9

Table 3: Concentrations of water-soluble and mobile ions in Moiyldy peloid

Water aqueous forms of composition, mg kg^{-1}		Mobile forms of composition, mg kg^{-1}	
Cl^-	29,966±756	P – P_2O_5	33.11±0,31
SO_4^{2-}	125,500±6024	K – K_2O	15,556±147
Ca^{2+}	800±16	S – SO_4^{2-}	125.25±4,70
Mg^{2+}	1,980±24	N-NO_3^-	>109
Na^+	5,275±121		
K^+	213.33±8,19		
NH_4^+	253,849±19 750		

Peloid is also enriched with halite and calcite. According to Pozo et al. (2013), high concentrations of halite can cause an increase in the content of the silty fraction and affect the textural features of the peloid. This mineralogical composition presumably will not be able to retain a large amount of moisture and will have low heat capacity (Baricz et al., 2021) and plasticity (Oumar et al., 2022). The presence of calcite in the sample may cause a positive physiological effect since carbonates stimulate subcutaneous blood circulation and epidermal renewal (Cara et al., 2000). According to Table 3, ammonium, sodium, magnesium, sulfate, and chloride ions are present mostly in mobile and bioavailable forms. It is known that sodium and potassium ions increase skin permeability, promote moisture retention, activate the ion transport system through the cell membrane, and take part in the elimination of toxins and cell stimulation (Potpara et al., 2017). Magnesium ions play an important role in the migration and proliferation of endothelial cells and are necessary for cellular metabolism (Pygmalion et al., 2010). When comparing with the results of studies of peloids from other regions of Kazakhstan, it can be concluded that the sample is similar to the natural mud of Zhalanashkol Lake in Southern Kazakhstan (Tokpanov, 2016), Lake Bolshoy Sor in Western Kazakhstan (Khalelova et al., 2020), but differs in a high content of ammonium ions.Based on chemical analysis, the natural mud of Moiyldy Lake can be attributed to the ammonium-sodium–sulfate–chloride type of continental origin and described in the form of Eq(1):

$$M100\frac{SO_4\,1307.29\mathrm{Cl}\ 84.41}{\mathrm{NH_4}\,1{,}410\mathrm{Na}22.93[\mathrm{Mg}8.25]}, \%\ mmol; \mathrm{pH}8{,}97 \qquad (1)$$

Tokpanov et al. (2019) described the mineral bischofite ($MgCl_2$), which has anti-inflammatory, regenerating, and analgesic effects (Kuskov and Lysikova, 2004). Ammonium salts have a keratolytic effect of an anti-allergic and anti-inflammatory nature, reducing peeling and facilitating the course of skin diseases such as psoriasis and dermatitis (eczema) (Medasani, 2004). The particle size distribution according to the results obtained by SEM (Figure 1a-b) is consistent with the fact that the average particle size corresponds to a fine-grained silty fraction, and the mud of the Moiyldy can be related to the sandy-silt type (Figure 1c) by Shepard (1954). These results may indicate the coastal nature of the mineralogical composition with a low percentage of the clay fraction. The content of the silty fraction up to 70.15 % indicates a long maturation process of the studied sample and brings them closer to the composition of thermal peloids from Turkey (Karakaya et al., 2010). According to the literature, particle size distribution plays an important role in the mobility of metal ions. Accordingly, assuming the low content of humic substances and particle sizes of less than 2 µm, the peloid of Lake Moiyldy is likely to have a relatively low ability to retain metal ions, including toxic ones (Quintela et al., 2012). Assessing the sensation during skin applications, the peloid can have a soft tactile effect with a large specific surface area without physical irritation (Bergamaschi et al., 2020). Such peloids are preferred in the use of peloidotherapy (Gomes et al., 2013). Analysis of the results from the three illuminated areas (Figure 1 a) shows that the oxygen and sodium have the largest proportion by weight, and the oxide and chloride form exceeds the sulfide form in quantity. Thus, in Selected Area 1, gallite and sodium sulfates may predominate to a greater extent; in Selected Area 2 – gallite, to a lesser extent calcium aluminosilicates; in Selected Area 3 - sodium sulfates, calcium carbonates, calcium aluminosilicates, and also the content of iron sulfide and oxide is possible.

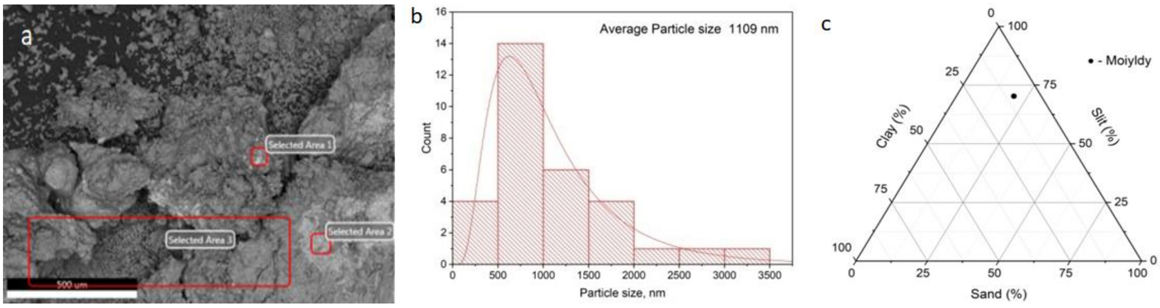

Figure 1: Moiyldy mud sample morphology: SEM images of scale of 500 µm (a); PSD (b-c)

The total concentrations of elements (Table 4) contained in the studied samples are compared with the levels in the continental crust (CC) and carbonate deep-sea sediments (CDSS) (Mielke, 1979). The sample has a relatively high content of bismuth in comparison with CC by 41.4 times (there is no information on CDSS). Also, increased amounts were found for sodium by 2.0 and 2.4 times, lithium by 1.9 and 7.5 times, cadmium by 4.4 and 7.3 times, silver by 2.7 and 2.3 times, antimony by 2.4 and 3.1 times, and arsenic by 1.6 and 2.9 times, in comparison with CC and CDSS.

The high content of bismuth can be explained by the fact that in sulfide minerals, the content of bismuth varies by several orders of magnitude and tends to concentrate more together with galena (Mielke, 1979). Suárez et al. (2015) noted that elements Cr and Pb exhibit limited mobility because they are in less leachable fractions and, despite toxicity, do not negatively affect the quality of natural mud used in peloid therapy. Van Veenhuizen et al. (2021) studied the biosorption of lead by anaerobic bacteria, which also play a key role in the formation of peloids. The reduced copper content may point to a negligible clay character of the sample. The presence of increased concentrations of these elements can be explained by the location of the lake in one of the major industrial centers of Kazakhstan. However, the accumulation of heavy metals in bottom sediments is not always associated with anthropogenic pollution, as it is also affected by the local geochemical background.

Table 4: Total concentrations (C) of elements in Moiyldy peloid

Element	Li	Be	Na	Mg	Al	P	K	Sc	Ti	V	Cr	Mn	Fe
C,	37.44	0.1819	47500	19570	36440	768.5	11373	11.87	1540	45.24	39.5	480.1	16550
mg kg^{-1}	±3.76	±0.019	±5300	±1900	±3900	±78	±1400	±1.12	±188	±5.1	±4.4	±53	±1900
Element	Cu	Cd	Dy	Ge	As	Sb	Ag	Y	Zn	Nb	Mo	Sr	Zr
C,	18.8	0.6579	2.099	0.8994	2.894	0.471	0.2054	9.658	25.7	7.287	1.992	781.4	71.28
mg kg^{-1}	±2.2	±0.10	±0.32	±0.1	±0.32	±0.09	±0.03	±1.6	±3.3	±1.3	±0.32	±85	±8.5
Element	Nd	Tl	Pr	La	Sm	Eu	Gd	Tb	Ce	Ho	Th	Hf	Ta
C,	21.69	0.1242	4.126	13.909	2.514	0.298	1.107	0.1642	37.2	0.267	1.63	1.746	0.950
mg kg^{-1}	±3.6	±0.018	±0.52	±2.0	±0.2	±0.45	±0.15	±0.025	±5.1	±0.04	±0.21	±0.28	±0.15
Element	Pb	Bi	Ga	Er	Ba	Co	Ni	Rb	Cs	W	U		
C,	14.14	0.3521	9.659	0.7202	252.7	7.414	18.02	51.37	3.207	0.842	2.39		
mg kg^{-1}	±2.1	±0.05	±1.1	±0.1	±39	±0.81	±2.1	±6.5	±0.42	±0.11	±0.32		

4. Conclusions

Physico-chemical and mineralogical analysis of the Moiyldy natural mud showed that this peloid has a homogeneous, finely dispersed colloidal mass of dark gray color with a highly developed specific surface area. Mineralogical analysis has shown that the peloid mainly consists of fine-grained quartz (45.3 %) and belongs to the sandy-silt type, which indicates its coastal continental origin. The predominant content of the silt fraction (70.15 %) makes this peloid similar to the thermal peloids of Turkey and proves its sufficiently long maturation. The peloid, due to its texture, has a soft tactile effect on the skin and a potential reactivity. The analysis of the gross content of elements in the mud showed increased concentrations of magnesium, sodium, potassium, lithium, phosphorus, titanium, iron, and aluminum among the main elements and microelements; arsenic, chromium, cadmium, lead, scandium, neodymium, uranium among rare earth elements and heavy metals, in comparison with their clarke values in CDSS. Most of them are biogenic and may have a complex positive effect on the skin and joints. Extremely high concentration of bismuth (41.4 times) could be explained by its tendency to concentrate with sulfide minerals in immobile form. Most of these metals belong to chalcophiles. The study of surface morphology shows that the oxide and chloride forms of minerals are more present. The study of the quantity of the main cations and anions of natural mud shows that it can be attributed to the ammonium-sodium–sulfate–chloride type and stands out by a significant content of ammonium cations having a keratolytic effect in the fight against skin diseases. The results show its rich mineral composition that might explain the symbiotic therapeutic effect. The research promotes new developments to expand the range of its use and obtain products based on it.

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