PERGAMON

Quaternary International 76/77 (2001) 113–121

Clay mineral composition and palaeoclimatic interpretation of the Pleistocene deposits of Ukraine

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Abstract

The Pleistocene deposits on the plains regions of Ukraine are subaerial and subaqueous formations of diverse types. Loesses and fossil soils represent 16 climatic stages (8 warm and 8 cold) of the Pleistocene and environmental changes are reflected in their clay mineral composition. Clay fractions (< 0.001 mm) of all 16 stages from 34 key sites and boreholes in different regions were analysed mineralogically. In the loesses and palaeosols the main clay minerals are of the smectite group (montmorillonite, nontronite, beidellite), hydromicas, mixed layer hydromica-montmorillonite minerals, kaolinite and halloysite. Chlorite, goethite, calcite, gypsum, and quartz are also present. There are differences in the amounts of the main minerals in palaeosols and loess horizons of different ages. Regional differences in the clay mineral composition of two Middle Pleistocene stages — the Zavadovka and Dnieper — were studied. Changes in mineral composition at 12 key Pleistocene sites in different regions indicate temporal (rhythmic) and spatial (regional) changes that reflect the palaeoclimatic conditions. © 2001 Elsevier Science Ltd and INQUA. All rights reserved.

1. Introduction

Pleistocene sediments cover almost the entire plains area of Ukraine. They are subaerial and subaqueous deposits of various types, but loesses and soils predominate. The thickness of this cover ranges from a few metres to about 60 m. During the Pleistocene the climate oscillated between warm and cold conditions, which resulted in the alternation of soil forming and loess forming stages. According to Veklich et al. (1984a, 1993), eight warm and eight cold Pleistocene stages can be recognized in Ukraine (Table 1).

Warm stages (interglacials and interstadials) are represented by fossil soils. As the environmental conditions and lengths of these stages were variable, different types of soils were formed (Sirenko, 1974; Sirenko and Turlo, 1986). Conditions of sedimentation in the cold stages were also different, so the different loess horizons vary in thickness, colour, granulometry, chemical and mineral composition, etc. Evidence for environmental conditions is often preserved in the clay fraction of sediments and soils, as this is the most active part of their mineral material. This paper presents the results of studies of the clay mineralogy (< 0.001 mm) of the Pleistocene deposits (from the Ilyichevsk to the Holocene, Table 1). These were derived from 34 stratified key sections and boreholes in different parts of the Ukrainian plains region (Middle and Porozhistoye Pridniprovye, Pobuzhye, Donbass, Lower Pridnistrovye, left bank of the Lower Danube, Kerch Peninsula and Prichernomorye: Perederij, 1974–1976, 1981, 1984, 1988a, b, 1989, 1995a, b, 1996, 1998a, b, c, 1999a, b; Veklich and Perederij, 1977; Veklich et al., 1984a, b, 1991, 1993, 1997; Matviishina et al., 1990). As all these detailed studies were published in Russian, this paper presents the first summary to be published in English.

2. Methods

Samples of 25–50 g were taken from all genetic horizons of the palaeosols. In loesses and glacial deposits they were taken from the lower, middle and upper parts of the section. The fraction < 0.001 mm was separated using the methods of Gorbunov (1963) and Gorbunov and Gradusov (1966) until the suspension was completely clear and the whole silt fraction was removed.

The analytical methods used included X-ray diffraction, differential thermal analysis, total chemical analysis, and transmission electron microscopy. X-ray diffraction was undertaken on oriented aggregates which were untreated, saturated with glycerine or ethylene glycol, heated to 550°C or treated with warm HCl (to distinguish the 14 Å reflections of smectite and chlorite) using "Dron-I" and "Dron-05" equipment with Cu radiation and a

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PII: S1040-6182(00)00095-1

Table 1
Palaeogeographical stages and detailed stratigraphy of the late (upper) Cenozoic in the loess regions of Ukraine (Veklich et al., 1984a, 1993)

Stratigraphical scale		Palaeogeographical stages Stratigraphical horizons			Age of lower bound (ka)	Duration (ka)	Alpine scheme
Period system	Main subdivision	Title	Soils	Index			
Q	Holocene	Holocene	■	hl		10 (13, 5)	
	Late (Upper)	Prichernomorje		pc	21	11 (72, 5)	W_3
	Pleistocene QIII	Dofinovka	■	df	50	29	$W_{2\text{-}3}$
		Bug		bg	75	25	W_2
		Vitachev	■	vt	90	15	$W_{1\text{-}2}$
		Udaj		ud	100	10	W_1
		Priluki	■	pl	130	30	R_3
	Middle	Tyasmin		ts	170	40	R_2
	Pleistocene QII	Kajdaki	■	kd	240	70	$R_{1\text{-}2}$
		Dnieper		dn	290	50	R_1
		Zavadovka	■	zv	420	130	M-R
		Tiligul		tl	480	60	M_3
	Early (Lower)	Lubny	■	lb	640	160	$M_{2\text{-}3}$
	Pleistocene QI	Sula		sl	730	90	M_2
		Martonosha	■	mr	920	190	$M_{1\text{-}2}$
		Priazov		pr	1000	80	M_1
N	Late (Upper)	Shirokino	■	sh	1290	290	G-M
	Neogene N_2^3	Ilichevsk		il	1400	110	G_2
		Kryzhanovka	■	kr	1610	210	$G_{1\text{-}2}$
		Berezan		br	2200	590	G_1
		Beregov	■	bv	2430	230	

NI filter. The differential thermal analytical work used "Q-1500" equipment, total chemical analysis was undertaken using standard methods, and the transmission electron microscopy used magnifications of 17,500 and 20,000.

Clay minerals were identified using diagnostic criteria published by Gorbunov (1963, 1974), Gorbunov and Gradusov (1966), Ginzburg (1946, 1953), Brindley (1965), MacEwan et al. (1965), Weaver (1962), Jackson (1965). The smectites were identified by a 14 Å reflection increasing to 17.6 Å after saturation with glycerine or ethylene glycol, and by endothermal reactions at 120–180° and 600–700°C. Hydromicas were identified by basal reflections at 10.5 and 3.34 Å in the diffraction diagrams, by the K_2O content and the results of differential thermal analysis.

Kaolinite was identified by the 7.2 and 3.58 Å reflections in untreated, glycerine-saturated samples heated to 550°C, an endothermal reaction at 560–570°C and an exothermal reaction at 925–1000°C. The mixed layer hydromica–montmorillonite minerals were identified by a series of reflections at 14–30 Å and 10–14 Å, increasing to 18–20 Å after saturation with glycerine, by the appearance of 3.21–3.23 Å reflection after heating to 550°C and by a double endothermal peak at 130–150°C. Chlorite was identified by 14, 7 and 4.7 Å reflections in untreated samples and their absence after treatment with warm HCl. Goethite was identified by 4.18, 2.69, and 2.45 Å reflections in untreated aggregates and by an endothermal reaction at 300–360°C.

Amounts of the main clay minerals (smectites, hydromicas, kaolinites) were estimated using the methods of Brindley (1965) and Gorbunov (1974), by measuring the areas beneath the X-ray reflection peaks multiplied by various factors. The composition of mixed layer minerals was determined according to the relative areas beneath the reflections of the component layer types.

3. Results

The results for key sites are given in Figs. 1–4. These figures have no vertical scale because the thickness of the deposits of different stages is very variable. The horizontal scale of clay mineral abundance (predominant, many, present) is also approximate. The sections were selected to represent the main regions and all 16 stages of the Pleistocene in Ukraine. The deposits of two stages (Zavadovka and Dnieper) are discussed in detail in order to demonstrate changes of environmental conditions in the Middle Pleistocene.

The palaeosols contain 30–50% clay and the intervening loess layers only 10–15%. In the Lower Pleistocene (Shirokino, Martonosha, Lubny) and Middle Pleistocene (Zavadovka) soils the clay content is 40–56%, suggesting greater intensity of mineral weathering or clay

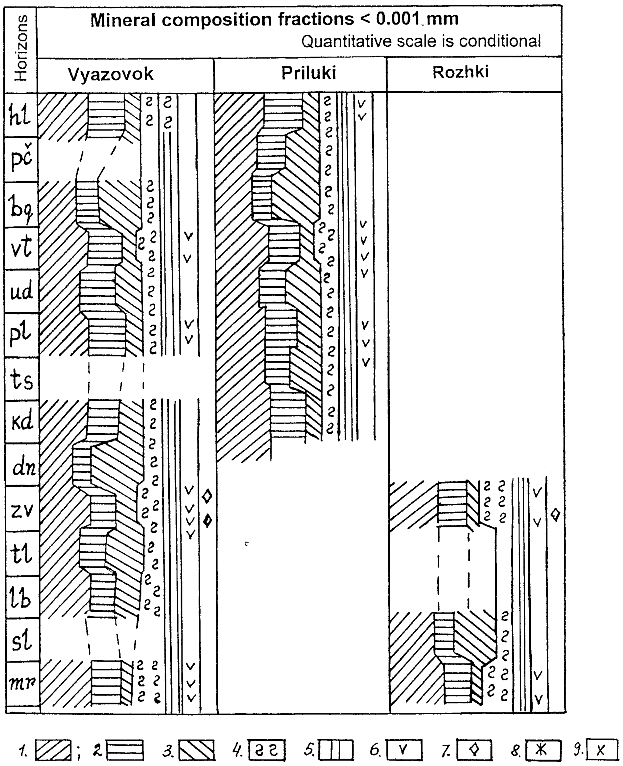

Fig. 1. Clay mineral composition (< 0.001 mm) of the Pleistocene deposits at the key sites of Middle Pridnieprovye, Ukraine. For horizon names see Table 1. Legend: 1 — smectite; 2 — mixed layer minerals; 3 — hydromica; 4 — kaolinite; 5 — quartz; 6 — goethite; 7 — calcite; 8 — gypsum; 9 — chlorite.

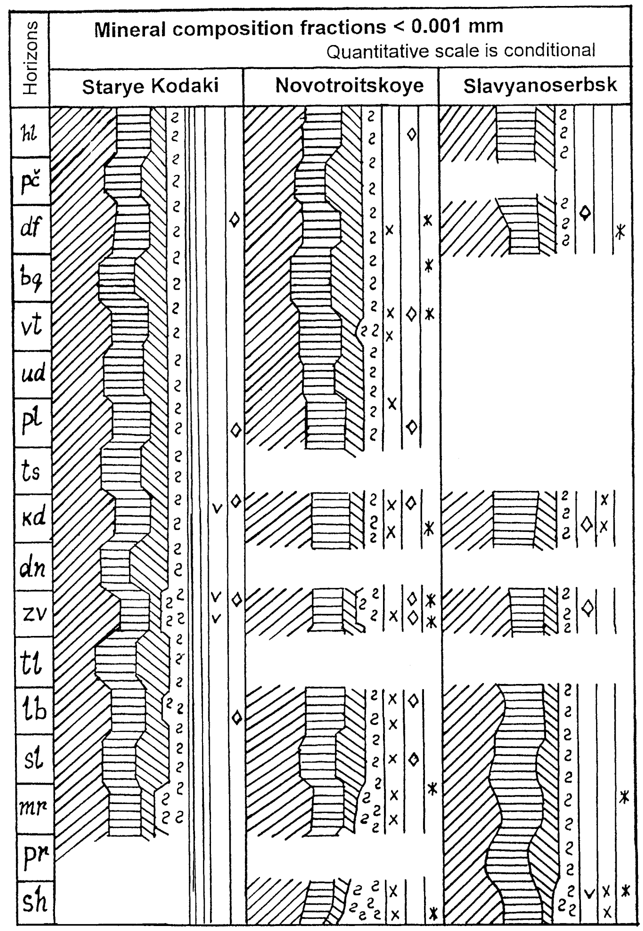

Fig. 2. Clay mineral composition (< 0.001 mm) of the Pleistocene deposits at the key sites in Porozhistoye, Pridnieprovye and Donbass. For horizon names see Table 1. For legend see Fig. 1.

illuviation during these stages. The clay contains minerals of the smectite group (montmorillonite, nontronite, beidellite), hydromicas, mixed layer hydromica–montmorillonite, kaolinite and halloysite in various proportions. Chlorite, goethite, calcite, gypsum and quartz are also present in small amounts. The overall similarity of clay mineral assemblages throughout the Pleistocene results from derivation of deposits from those of earlier stages and the weak transformation of minerals by weathering during formation of palaeosols. However, there are small quantitative and qualitative differences in minerals of the smectite group and the hydromicas between palaeosols and loess layers of different ages, and these suggest palaeoenviromental changes during the Pleistocene. The assemblages of the palaeosols and unaltered loess and till layers are considered separately.

4. The clay fraction of the palaeosols

The main minerals in the clay of the palaeosols are smectites. In the Middle and Porozhistoye Pridnieprovye, Pobuzhye and Donbass regions, they are montmorillonite, but in the south (Prichernomorye and Lower Pridniestrovye regions) in the Shirokino and Martonosha palaeosols (Lower Pleistocene) and sometimes in the Zavadovka soils (Middle Pleistocene), they are nontronite and beidellite. Smectites also dominate the Kajdaki (Middle Pleistocene), Priluki and Vitachev (Upper Pleistocene) soils.

The Lower Pleistocene (Shirokino, Martonosha) and the Middle Pleistocene (Zavadovka) palaeosols contain slightly more kaolinite than those of the Upper Pleistocene. Given the greater thickness of these soils, and their cinnamonic or reddish–cinnamonic colours, greater clay contents (40–55%), enrichment in Al_2O_3 and Fe_2O_3 (with $SiO_2/Al_2O_3 = 3.4$–4.2, $SiO_2/Fe_2O_3 = 13.8$–14.7, $SiO_2/R_2O_3 = 2.7$–3.2) more intense mineral weathering under a warm, variably humid climate is indicated. Kaolinite also occurs in the brown forest and burozem-like soils of the Vitachev stage of the upper Pleistocene. Mixed layer minerals occur in all the palaeosols.

According to Ginzburg (1946, 1953), Petrov (1967), Zhus (1966), Gorbunov (1969, 1974) and Weaver (1962), the presence of mixed layer minerals suggests weathering changes in the clay minerals. Interstratified hydromica–montmorillonite is the most common. It is

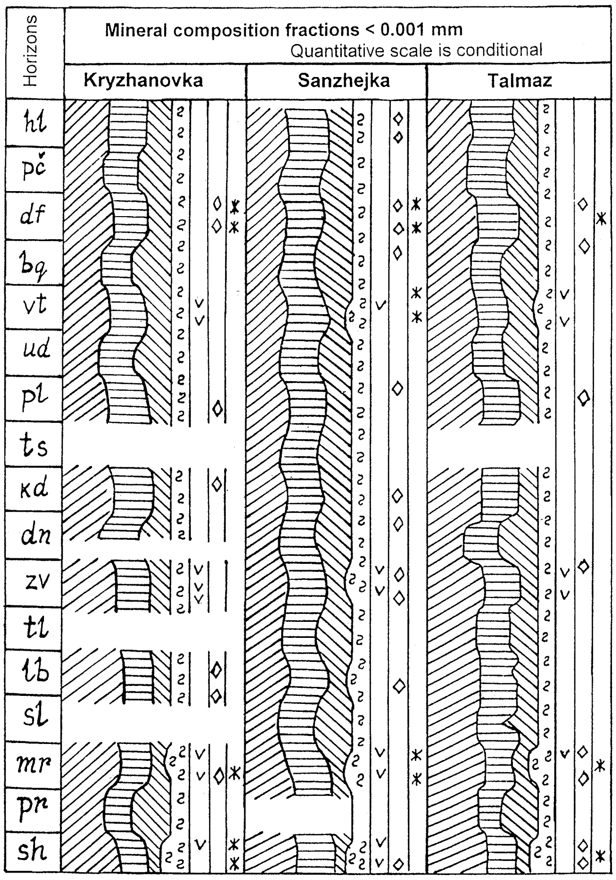

Fig. 3. Clay mineral composition (< 0.001 mm) of the Pleistocene deposits at the key sites in Prichernomorye and Lower Pridniestrovye. For horizon names see Table 1. For legend see Fig. 1.

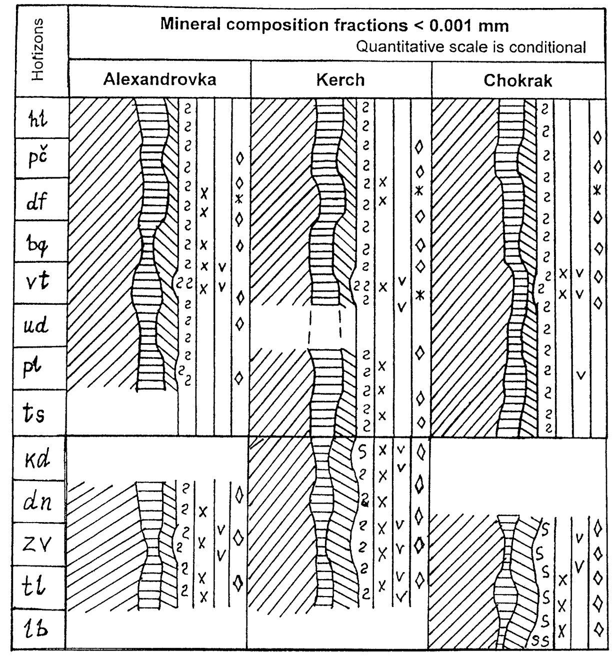

Fig. 4. Clay mineral composition (< 0.001 mm) of the Pleistocene deposits at key sites of the Kerch Peninsula. For horizon names see Table 1. For legend see Fig. 1.

identified by reflections at 10–14 Å and 14–28 Å and results in a decrease in the intensity of, and the area beneath the 10 Å peak of the soil samples compared with the loess samples, and in a decrease in K_20. This suggests that hydromicas inherited from the loess parent materials became montmorillonitized by the soil forming processes resulting from palaeoenvironmental changes (aqueous and thermal regimes) in subsequent stages. According to Rentgarten and Konstantinova (1965), increases in temperature and humidity favoured microbial activity and the production of organic acids, which would have removed interlayered potassium from the hydromica to produce montmorillonite-like layers. The greater abundance of the mixed layer minerals in the palaeosols, especially those with hydromorphic features, therefore suggests wetter and warmer conditions during palaeosol formation compared with those of loess deposition. The unusual forms of the kaolinite (oval, with almost no hexahedral forms) and montmorillonite particles in profiles of the Kerch Peninsula suggest other changes in clay mineral composition in the hydromorphic palaeosols.

The differences in mineral composition of clay fractions within the Dofinovka, Vitachev, Priluki (upper Pleistocene), Kajdaki, Zavadovka (Middle Pleistocene) and Lubny (Lower Pleistocene) palaeosols suggest climatic changes during soil development, with conditions during the optimum phase of soil formation that were warmer and wetter than in the initial and final phases.

4.1. Clay mineral composition of soils of the Zavadovka stage

The Zavadovka stage (Middle Pleistocene) in Ukraine is represented by a widespread palaeosol that is thickest (2.0–3.8 m) in the regions of Porozhistoye Pridnieprovye, the Pridnieper highlands and the Pridonetsk plain (Sirenko, 1974). A characteristic feature of this palaeosol is the clear zonality indicating variation of environmental conditions in time and space. Five zones are recognized (Veklich, 1965; Veklich et al., 1973; Sirenko and Turlo, 1986). In the Middle Pridnieprovye region, there are brown forest cinnamon-like soils, and in the Porozhistoye Pridnieprovye and on the Pridonesk plain cinnamonic and meadow–cinnamonic soils are found. In the south there are reddish–cinnamonic soils, and red–brown soil in the Prichernomorye. The different soil types have different clay mineral assemblages (Fig. 5).

The brown forest and cinnamonic-like soils in the Middle Pridnieprovye and left bank plains regions (Fig. 1: key sites Rozhki and Vyazovok) contain large amounts of silt (35–42%) and iron and magnesium oxides, and the clay fractions mainly contain smectite group minerals with moderate amounts of mixed layer minerals and kaolinite. This clay mineral composition indicates rather

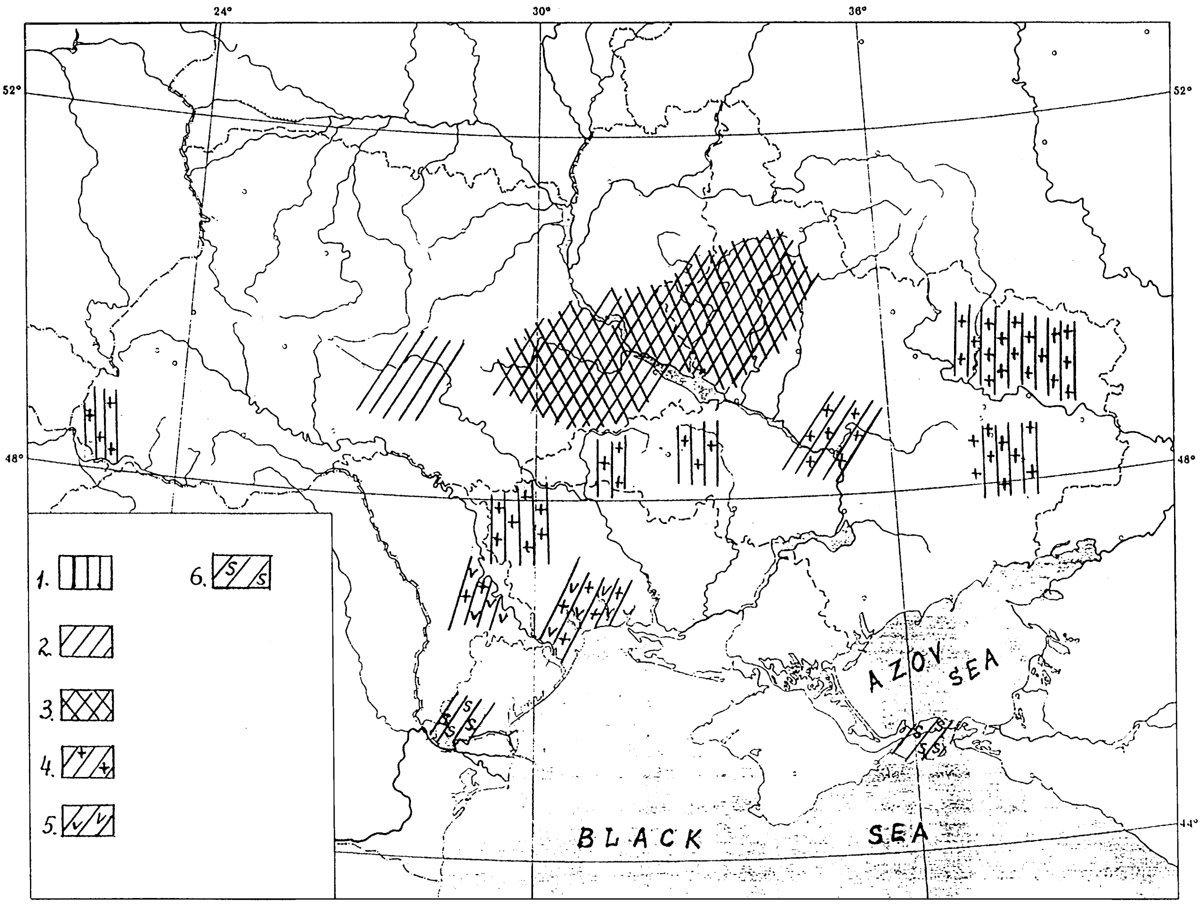

Fig. 5. Regional variation in clay mineralogy (< 0.001 mm) of the Zavadovka stage fossil soils. 1 — smectite; 2 — smectite and mixed layer minerals; 3 — smectite, mixed layer minerals and kaolinite; 4 — smectite, mixed layer minerals and calcite; 5 — smectite, mixed layer minerals and gypsum; 6 — smectite and kaolinite.

warm humid conditions. Minerals of the smectite group also dominate the cinnamonic soils of the Porozhistoye Pridnieprovye (Fig. 2: key site Staryye Kodaki). In the Pridonetsk plain (key site Slavyanoserbsk), the quantities of silt (30–35%) and kaolinite are somewhat less, suggesting warmer but drier conditions.

In the meadow-cinnamonic soils of these regions, the silt contents (up to 53%) and the amounts of iron oxides and kaolinite are considerably greater, suggesting wetter conditions with more intense hydromorphism and mineral weathering.

The dark cinnamonic palaeosols of the southern part of Donbass (Fig. 2: key site Novotroitskoye) and of Middle Pobuzhye and the right bank of the Dnieper are thinner and contain less clay, but are calcareous, suggesting formation in steppe conditions. The Zavadovka palaeosols of the Trans-Carpathian regions (Adamenko and Grodetskaya, 1987) and S.W. Ukraine (Sokolovsky, 1958; Samodurov, 1957) are similar in clay mineral composition. In the southern Ukraine (Prichernomorye, Pridniestrovye), however, they are reddish–cinnamonic and red–brown in colour, and are thinner and contain less clay, as well as being more saline. Smectite and mixed layer hydromica–montmorillonite minerals prevail in their clay fractions, and calcite and gypsum also occur (Fig. 3: key sites Kryzhanovka, Sanzhejka, and Talmaz). This clay mineral composition suggests drier conditions during soil formation.

The greyish–cinnamonic soils of the Kerch Peninsula (Fig. 4: Kerch, Alexandrovka, the Chokrak Lake) and the brownish soils on the left bank of the Lower Danube are notable for considerable hydromorphism, a dominance of smectite and some increase in the quantity of kaolinite.

The variation in clay mineral composition of the Zavadovka soils indicates warm and humid conditions in northern regions and warm, arid conditions in southern and southeastern regions.

5. The clay fractions of loess and till horizons

The clay fractions of the loess horizons are characterized by large amounts of hydromicas and fewer smectites and mixed layer minerals than in the palaeosols. In the thicker loess horizons {Tiligul (Lower Pleistocene), Dnieper (Middle Pleistocene) and Bug (Late Pleistocene)}, there are geographical differences in the quantities of these minerals. In the north they contain more hydromicas but fewer smectite minerals than in the south (Donbass, Lower Pridniestrovye, Prichernomorye, and the Kerch Peninsula). In the thinner loess horizons, such as Sula (Lower Pleistocene), Udaj (Middle Pleistocene) and Prichernomorye (Late Pleistocene), the minerals are more weathered. This is indicated by the presence of amorphous materials recognised by diffuse low-angle reflections and their appearance in transmission electron micrographs. The extent of weathering in these horizons increases to the south: their hydromica content decreases and the smectite and mixed layer minerals increase. These trends suggest latitudinal differences in climatic conditions during deposition of these loesses. The largest quantities of hydromica occur in the various deposits of the Middle Pleistocene Dnieper horizon (moraine, lacustrine, glaciofluvial, loess) of the glaciated region (Middle and Porozhistoye Pridnieprovye).

5.1. Mineral composition of the Dnieper stage

The maximum extent of glacial ice cover in Ukraine occurred during the Middle Pleistocene Dnieper stage. The ice occupied considerable areas in the north (Polesje), in Middle Pridnieprovye. Along the Dnieper Valley, the ice sheet moved south to the Porozhistoye Pridnieprovye (Fig. 6). The deposits of the Dnieper stage are of three main facies: glacial (two till layers), subaqueous (alluvial, lacustrine, glaciofluvial) and subaerial (loesses and loessic loams of pale yellow or brown colour: Dorofeyev, 1961). The deposits are quite thick (5–12 m) in the Pridnieprovye region and are subdivided by the presence of 1–3 embryonic palaeosols. To the west and south, loess thickness on watersheds decreases to 2–4 m in Prichernomorye and 0.3–0.7 m in the Donbass and Kerch Peninsula. Loess is completely absent locally (Dorofeyev, 1961; Veklich, 1968).

The clay fractions of the various facies of the Dnieper stage in Ukraine are composed mainly of hydromicas, smectites and mixed layer minerals (Figs. 1–4). In the glaciated area, they show vertical differences that may be summarised as follows.

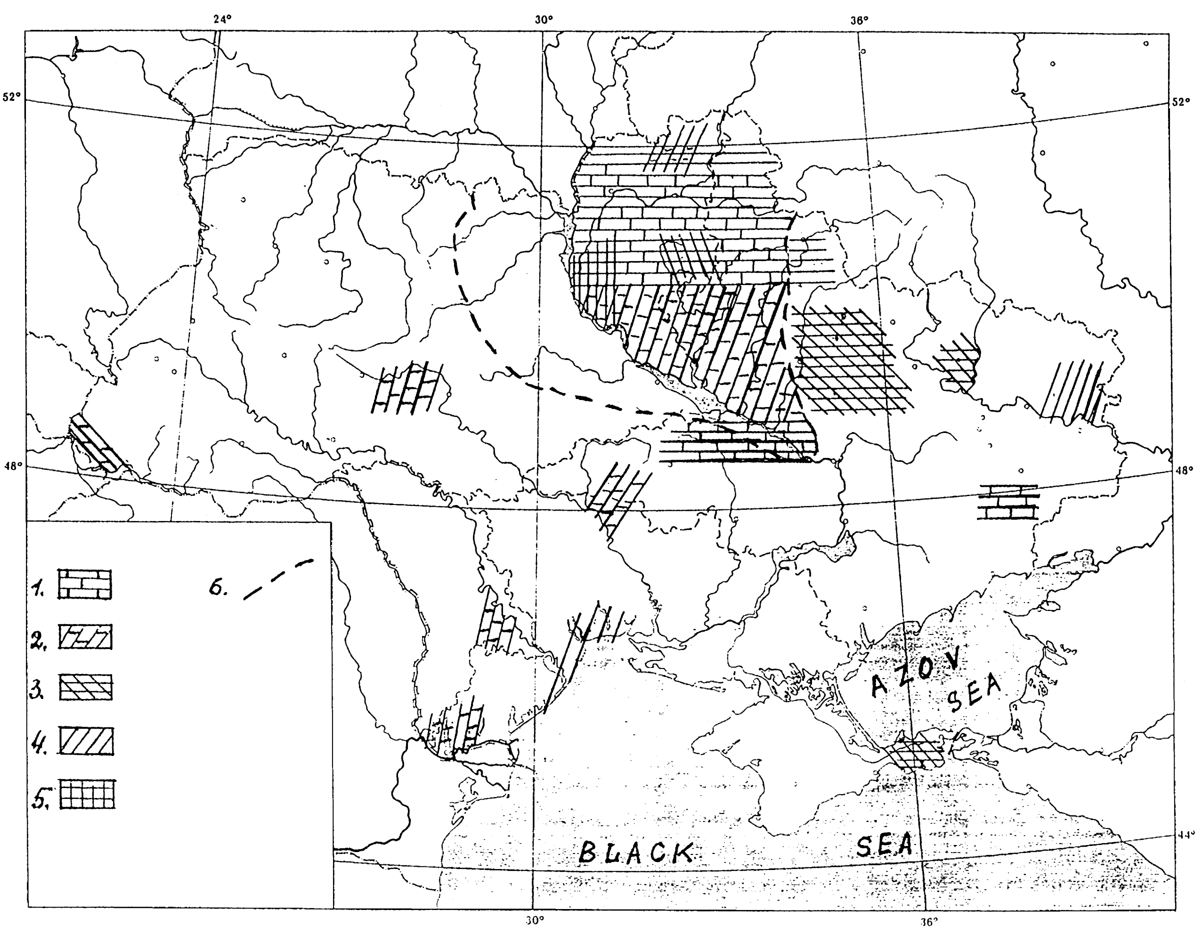

Fig. 6. Regional variation in clay mineralogy (< 0.001 mm) of the Dnieper stage deposits. 1 — hydromica, smectite; 2 — smectite, hydromica; 3 — hydromica, smectite, kaolinite; 4 — smectite, mixed layer minerals; 5 — hydromica, kaolinite; 6 — Dnieper glacial boundary.

1. The loess deposits beneath the moraines contain interstratified hydromica–montmorillonite with a sharp increase in the montmorillonite component in the lower part, possibly because smectite was inherited from the underlying soils of the Zavadovka stage, and there was no transformation of the inherited minerals in the periglacial conditions. As the loess thickens, its smectite content gradually decreases and hydromica increases.
2. In the glacial deposits, hydromica is the main clay mineral. Small quantities of smectite, kaolinite, chlorite and mixed layer minerals can be explained by derivation from the various source materials over which the glacier advanced. During glacial transportation, even over large distances, there were no chemical changes in the clay, although some physical changes can be observed. Thus, in the glacial and glaciofluvial deposits, the kaolinite is usually represented by fragments, hexagonal crystals being rare. Geographical differences in mineral composition of the glacial deposits result mainly from changes in underlying materials. The maximum content of hydromicas occurs in the northern and western parts of the glacial region, whereas in southern and southeastern regions, especially in the distal part of the ice tongue, smectite increases, especially in the lower part of the glacial deposits (Dorofeyev, 1961; Melnik, 1962).
3. The strata above the glacial sediments are 0.3–2.0 m thick and consist of loesses and loessic loams, with some lacustrine and glaciofluvial deposits. The mineral composition of the clay in these deposits is qualitatively similar to that in the underlying glacial sediments, but there are quantitative differences in the proportions of mica and smectite components in the mixed-layer minerals. In the Lower Pridniprovye region, the deposits of the Dnieper stage of the nonglacial area consist of thick loess strata characterized by interstratified hydromica–montmorillonite with small amounts of kaolinite and chlorite. In the Middle Pobuzhye region, smectites and hydromicas are dominant in the Dnieper loesses. However, on the left bank of the Siversky Donets River, smectites and mixed layer minerals are dominant and hydromicas are less abundant. A similar clay mineral assemblage occurs in the North Prichernomorye region. In the Trans-Carpathians, Lower Pridnistrovye and on the left bank of the Lower Danube, smectites and hydromicas predominate.

In the thin loesses of the Dnieper stage, mixed layer hydromica–montmorillonite minerals are probably inherited from the underlying Zavadovka soils, as well as resulting from soil forming processes of the subsequent Kajdaki stage. However, in the thick loess strata of the Lower Pridniestrovye, Pobuzhye and in the deposits overlying the till in the glaciated area, these minerals may have resulted from cryogenic degradation of hydromicas into mixed layer minerals or because of contemporaneous terrigeneous input with slow loess deposition. The Dnieper deposits of the Kerch Peninsula were strongly influenced by groundwater and soil forming processes in the Kajdaki stage: their clay fractions consist of smectite, hydromica and kaolinite.

The regional variation in the clay mineral composition of the different facies of the Dnieper stage is summarised in Fig. 6.

6. Conclusions

The clay minerals of sediments and palaeosols in the Pleistocene deposits of different regions of Ukraine indicate repeated rhythmic changes of climate. The clay assemblages indicate a clear alternation of deposits of cold periods (loesses, loessic loams, moraines) and palaeosols of warm stages.

Dominance of hydromica in the thick Pleistocene loesses of the Dnieper and Tiligul stages is associated with periglacial and glacial conditions. In deposits of other cold stages this mineral is also fairly abundant, but in the palaeosols it is much less common. Warming and increases of humidity in periods of soil formation led to changes in mineral composition, with formation of smectites, mixed layer hydromica–montmorillonite and kaolinite in various proportions.

Within the palaeosols there was a gradual decrease in clay content with time from 56% in the Lower Pleistocene to 20–25% in the Late Pleistocene, and a decrease in kaolinite content, suggesting a gradual decrease in the intensity of weathering. In deposits of the cold stages, the amounts of hydromicas gradually increased from the Early to the Late Pleistocene.

Some climatic zonality can be inferred from the lateral variations in clay composition in certain stages of the Pleistocene. This is seen in the loess horizons of the Bug, Dnieper and Tiligul stages in which the hydromica content gradually decreases and the smectite content increases towards the south. In the palaeosols of almost all warm stages, especially in the Martonosha, Zavadovka, Kajdaki, Priluki and Vitachev palaeosols, there are regional differences in the clay mineral composition. In the Martonosha and Zavadovka soils in southern regions (Prichernomorye, Lower Pridniestrovye), the smectite group minerals are often represented by nontronite or beidellite and, in other regions by montmorillonite. The amounts of calcite and gypsum amounts increase there compared with other regions.

The Kajdaki soils in Prichernomorye show considerable amounts of calcite, and in eastern regions (Donbass) the calcite and chlorite content increases. As for their mineral composition, the Priluki soils are similar to the Kajdaki soils, but in northern regions (Middle

Pridnieprovye) considerable amounts of goethite occur in them. Large quantities of kaolinite and goethite are peculiar to the Vitachev soils of the Middle Pridnieprovye. In the eastern (Donbass) and southern (Prichernomorye) areas the kaolinite and goethite content decreases, while the amount of calcite and gypsum increases.

Within profiles of the Lubny, Zavadovka, Kajdaki, Priluki and Vitachev palaeosols can also be seen indications of different environmental conditions varying from cold (loess formation) to warm periods in which formation of weakly developed soils of the initial phase occurred with a slightly changed mineral material inherited from the underlying loesses. Increase in temperatures and humidity towards the climatic optimum favoured the formation of full-profiled, well developed soils, resulting in enhanced mineral weathering, change of structure, and transformation of the less stable minerals into the mixed layer types under climatic and biotic impacts. In the final phase, with a change towards cool and more arid conditions, the soil forming processes weakened in their intensity and mineral species responded accordingly. This is seen mostly in the palaeosols of the Middle and Porozhistoye Pridnieprovye, Donbass, Pobuzhye and Lower Pridniestrovye regions. Thus, clay mineral investigation provides clear evidence for temporal and spatial differences in environmental conditions during the Pleistocene in the territory of Ukraine.

Acknowledgements

The author is pleased to express her gratitude to Professor John Catt for his considerable help in producing the English text.

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