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TEMPORAL AND SPATIAL CHANGES IN ORGANIC AGENTS IN THE PROGRESS OF PRIMARY PEDOGENESIS DURING THIRTY YEARS

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Abstract. A special experiment was established in 1963 and initiated in 1964 to study pedogenesis under grass-herbaceous vegetation on red-brown calcareous till. The results of the first two decades have been discussed earlier. This paper deals with pedogenetic activity during the third decade of the experiment and during the total transient period of thirty years. A humus-accumulative process wavy in its intensity has continued being accompanied with the breakdown and leaching of carbonates, accumulation of nonsiliceous iron (hydro)oxides, slight progress of argillization *in situ*, and lessivage of fine silt and clay. A net accumulation of organic carbon and nitrogen was observed that was mainly guaranteed by similar amounts of humifiable phytocoenotic agents. However, also a cyclic mineralization of humus substances accumulated earlier and a loss of unstable humus supplies had taken place. Temporal periodicity of mineralization and humification tended to be characteristic of primary pedogenesis although a narrow C:N ratio indicated the perfection of humus formed. Changes in the quality indices of humus resulted in an increase in fulvicity and decrease in total solubility whereas the transformation of Ca-humates into humins and the formation of fulvic compounds with mobile sesquioxides on the account of Ca-humic—fulvic complexes were ascertained. An intensification of the connection of humic—fulvic complexes with inactive sesquioxides and clay minerals and an increase in the quantity of fulvic acids in the crystalline structure of clay minerals were characteristic of the soil formation within the third decade.

Key words: experimental modelling, primary pedogenesis, humus-accumulative process, humus quality, humic acids, fulvic acids.

INTRODUCTION

Since V. V. Dokuchayev up to the modern schools of genetic and ecological soil science pedogenesis has been interpreted as a permanent and sophisticated complex of interactions between organic and mineral substances, solar radiation, moisture and gases, living creatures and the inanimate environment in space and time. The soil profiles formed and differentiated on initial mineral strata as a result of these interactions are the principal objectives for the explanation of the origin of soil genesis and evolution in the past as well as at present, although there might have been temporal and spatial variances and changes in trends and

speeds of any interdependent processes in the formation and development of humus status in the soil as well as energetic-substantial phenomena in the whole ecosystem. Simonson (1959) suggested that the horizon differentiation is a function of additions, removals, transfers, and transformations within the soil system whereas according to Arnold (1965) material gains and losses can be interpreted as an active process in the soil development.

Any particular soil type, as well as the soil mantle of any territory, is subjected to the regularity of continuum representing a reflector of ecosystem interdependences and interactions (Targulian et al., 1979; Arnold et al., 1990). To study the modern synchronous production and pedogenetic processes in a certain ecological situation and to find out both the extent and trends of the changes taking place in ecosystem characteristics, the method of experimental modelling was introduced and is increasingly more used (Simonson, 1959; Smirnov, 1960; Arnold, 1965; Гагарина & Цыпленков, 1974; Оја, 1975). Besides special experimental models showing a rapid progress of soil formation (Мельникова & Ковеня, 1971; Оја, 1975; Reintam, 1982; Рейнтам & Погорелова, 1986) valuable pedogenetic and ecosystem information has been received as a result of the investigation and recultivation of mined territories (Таранов, 1977; Трофимов et al., 1977; Schafer et al., 1980; Daniels & Amos, 1981; McSweeney & Jansen, 1984; Ужегова & Махонина, 1984; Roberts et al., 1988a), and researches into archaeological objects (Griffith, 1981; Haidouti & Yassoglou, 1982; Reintam, 1975, 1981, 1990b, 1994; Holliday, 1985; Collings & Shapiro, 1987).

Organic matter, especially its humus compounds, has been interpreted as a motive power of pedogenesis and ecosystem functions during the whole history of genetic soil science (Zonn, 1986). Attention has always been focused on the formation of humus and its status in a soil and the progress of production phenomena in the ecosystem and organic impacts on the changes in textural, chemical, etc. characteristics of soils. Therefore, to explain the formation of humus relationships and pedogenetic activity of both annual and perennial cultivated plants on red-brown calcareous till a special experiment was established in 1963 under natural conditions.

The results of pedogenesis during the first (1964—74) and second (1974—84) decades of the experiment were preliminarily presented and discussed in Diploma (BSc) papers of Virve Olvi (1976) and Tatyana Pogorelova (1986), respectively. Then changes in the balance of substances and also in the initial constituents of red-brown till within the primary pedogenesis during the first and first two decades were published (Reintam, 1982; Рейнтам & Погорелова, 1986; Reintam & Pogorelova, 1987) and summarized in Pogorelova's PhD dissertation discussed at the Harkov Agrarian University, Ukraine, in 1989 (Погорелова, 1989). To vouch for the coherent information on the pedogenetic activities by stages the objective of this paper is to deal with the changes in the humus relationship and in some soil properties during the third decade (1984—94) of pedogenesis as well as with general results of the soil formation ascertained in the transient period of thirty years and by three decades with the help of a new sampling in 1994.

MATERIAL AND METHODS

The experiment was founded at Eerika, Tartu County, Estonia (58°22' N, 26°36' E) in the autumn of 1963. *Albi-Eutric Luvisol* profile on red-brown calcareous till was dug up to a depth of 2 m in an area of 9 m².

The pit formed was divided into four equal parts (2.25 m² each), isolated from every side with saturated felt, and filled with unchanged red-brown calcareous till dug up from a neighbouring cellar pit of the lysimeter building from a depth of 1.5—3 m. The initial bulk density (1.71 Mg·m⁻³) of the till transferred was preserved by the volume. The initial characterization of the till used was published in 1982 (Reintam, 1982). The till was practically free of organic carbon (0.06%) and nitrogen (0.02%), the contents of clay and silt-clay were 14 and 26%, respectively.

The real experiment was initiated in the spring of 1964 after the natural winter subsidence and the formation of the sown agricultural herbaceous vegetation. The experiment included four variants: (1) white clover and grasses pasture sward, (2) hop lucerne, (3) summer barley, (4) no vegetation. In the first decade the crop was not harvested and all the aboveground biomass formed was turned into natural cycling to the advantage of soil formation. Only summer barley was newly sown every spring. The fourth variant was kept free of vegetation. Because of some objective reasons it became impossible to continue the experiment according to this scheme during the second decade and all variants were spontaneously covered with grass-herbaceous vegetation with both white clover and hop lucerne ousted from the sward. The biomass was completely used as an energetic-substantial source for pedogenesis. In 1984 the standing left from the previous year was cut and weighed. The data obtained in the context together with materials published earlier were used for the quantitative evaluation of an approximate production and pedogenetic activities in the duration of twenty years (Рейнтам & Порорелова, 1986; Reintam & Pogorelova, 1987).

Since 1984 (beginning of the third decade) the dynamics of biomass formation was determined by variants. The variants were distinguished on the basis of the differences in the accumulation and/or elimination of the organic aboveground matter formed. So, at the end of the third decade the variants by their organic sources of pedogenesis were as follows:

- (1) **G—G—G+**: 1964—73 — white clover & grasses without harvesting, 1974—83 — grasses & herbs without harvesting, 1984—93 — grasses & herbs weighed and returned; **all grass—herbaceous biomass formed represented the source for pedogenesis.**
- (2) **L—G—G—**: 1964—73 — hop lucerne without harvesting, 1974—83 — spontaneous grasses & herbs without harvesting, 1984—93 — vegetation weighed and eliminated; **on the background of the former complete accumulation of organic residues, elimination prevailed during the last decade.**
- (3) **B—G—BG+**: 1964—73 — summer barley without harvesting, 1974—83 — spontaneous grasses & herbs without harvesting, 1984—87 — barley weighed, grains eliminated, straw and spontaneous hop lucerne & weeds returned, 1988—93 — spontaneous hop lucerne, grasses & herbs weighed and returned; **on the background of annual vegetation the perennial one with the intermittent accumulation and elimination of residues.**
- (4) **O—G—G+**: 1964—73 — without vegetation, 1974—83 — spontaneous herbs & grasses without harvesting, 1984—93 — vegetation weighed and returned; **on the background of a continuous absence of organic agents complete accumulation of spontaneous biomass formed to the advantage of pedogenesis.**

Three times (early May 1974, 1984, 1994) the morphological description and sampling of the profiles developed were carried out to a depth of 60 cm by the traditional and well-known way used in soil science separately for the microfabric investigations, bulk density determination (in four replications using a barrel of 50 cm³), and laboratory techniques. The solum dug up from the profile described was returned by layers and covered with a natural piece of turf from this place so as to change the situation as little as possible. The aboveground phytomass was cut three to four times in the vegetation period. The results were expressed in absolute dry weight.

Analytical techniques were carried out in the laboratories of our Institute by Raja Kährik, a research assistant. Fine earth less than 1 mm was used. The group and the fractional composition of humus and decalcinate were determined by the alternate acid-alkaline treatment applying the Tyurin—Ponomareva volumetric method (Пономарева, 1957) expressing the results obtained as the percentage of organic carbon. The total percentage of organic carbon and nitrogen were ascertained by Tyurin and Kjeldahl methods, respectively (Соколов, 1975). Nonsiliceous iron after Coffin, amorphous sesquioxides and silica after Tamm, and iron activity after Schwertmann were determined (Зонн, 1982). Supplies of all substances were calculated on the basis of the thickness of layers (horizons) described and the bulk density determined.

RESULTS AND DISCUSSION

Unlike during the previous two decades, during the third decade the total aboveground phytomass was measured. On the basis of the data obtained and materials discussed in literature (Arvisto, 1970, 1971; Гришина, 1974; Sau, 1979) the extent of possible humification was calculated following our earlier pattern (Рейнтам & Поргорелова, 1986; Reintam & Pogorelova, 1987). Differences in the absolute amounts of the aboveground organic matter amounted to one third between the variants with permanent perennial vegetation and barley background (Table 1). The spontaneous vegetation of the initial zero variant was on a par with the initial pasture sward already by the end of the second decade exceeding at present the latter by as much as 10%. These relations tend to recur in the possible extent of humification although the accumulation/elimination ratio becomes more essential there. A complete elimination of the aboveground grass—herbaceous mass on the initial background of lucerne results in an about twofold decrease in the possible pedogenetic agents compared not only with the former situation in this area, but also with the variants of phytomass restoration.

So, during the third decade the progress of soil formation could be induced (at a significance level of 20%) by plant residues, which were able to humify in the amounts of 790—1300 and 450—875 g·m⁻² in case of their complete restoration and partial elimination, respectively (Table 1). As these features are quite similar to those reported for the previous decades (Рейнтам & Поргорелова, 1986; Reintam & Pogorelova, 1987) the initial data from the literature taken for such calculations seem to be real and significant.

In spite of a general similarity a clear tendency to a decreasing aboveground phytomass can be noticed beginning from the middle of the third decade (Table 2). The periodicity of production and pedogenetic phenomena could not be excluded (Sau, 1983), but also an obsolescing of the sward and the influence of frequent draughts were possible. For example, in 1988 and 1990—93 (also in 1986) the phytomass seasonal increment

Table 1

Accumulation of organic residues into the soil in 1984—93, $g \cdot m^{-2}$

Material	G—G—G+	L—G—G—	B—G—BG+	O—G—G+
(1) Aboveground phytomass	3992	3706	3018	4449
(2) By the above- and underground phytomass ratio 1:1 (Sau, 1979) the accumulation of 30% of root residues into soil (Гришина, 1974)	1197	1112	905	1335
(3) Possible humification of root residues to the extent of 50%	588	556	452	667
(4) Possible humification of above-ground mass to the extent of 10% (Arvisto, 1970, 1971)	399	0	277	445
(5) Total possible humification	987	556	729	1112
Average annual possible humification	99	56	73	111
Average diurnal increment of aboveground phytomass	2.3	2.2	1.8	2.6
Average annual humification calculated for the period of 1964—83 (Рейнтам & Погорелова, 1986)	36	109	x	71

x, not determined.

Table 2

Accumulation of organic residues in the periods of a decade, $g \cdot m^{-2}$

Variant	Period	Aboveground phytomass		Possible extent of annual humification*
		total	annual	
G—G—G+	1984—87	2154	538.5	144
	1988—93	1838	306.3	82
L—G—G—	1984—87	1935	483.8	81
	1988—93	1771	295.2	49
B—G—BG+	1984—87	1017	254.3	51
	1988—93	2001	333.5	89
O—G—G+	1984—87	2241	560.2	149
	1988—93	2208	368.0	98

* Calculated on the basis of the criteria used in Table 1.

ceased already in late August—early September; however, in 1984—87 and 1989 it was intensive up to late October. Whereas a decrease in the aboveground phytomass could be accompanied by an increase in the root system the total amounts of organic residues potentially able to induce soil formation could remain in the limits of the former situation. At that the unification of variants with the restoration of all phytomass produced has also taken place. It tends to create prerequisites for a future thorough study of root production.

A continuous humus-accumulative process is characteristic of the top of thirty-years' soil formations except for the variant with the permanent elimination of aboveground phytomass (Table 3). This accumulation

Table 3

Organic carbon and nitrogen accumulated in pedogenesis, $g \cdot m^{-2}$

Variant	Thickness of layer, cm	Organic C		N		Annual increment 1984-94	
		1984	1994	1984	1994	C	N
<i>G-G-G+</i>	0-5	590	986	53	104	39.6	5.1
	5-10	290	795	23	76	44.5	5.3
	10-20	310	404	17	60	9.4	4.3
	0-20	1190	2125	93	240	93.5	14.7
	20-40	370	184	70	29	-18.6	-4.1
	40-60	50	124	94	32	7.4	-6.2
<i>L-G-G-</i>	0-60	1610	2433	257	301	82.3	4.4
	0-5	790	701	57	66	-8.9	0.9
	5-10	520	331	43	31	-18.9	-1.2
	10-20	260	191	22	19	-6.9	-0.3
	0-20	1570	1223	122	116	-34.7	-0.6
	20-40	210	256	33	51	4.6	1.8
<i>B-G-BG+</i>	40-60	60	33	21	34	-2.7	1.3
	0-60	1840	1512	176	201	-32.8	2.5
	0-5	550	895	29	84	34.5	5.5
	5-10	260	538	16	48	27.8	3.2
	10-20	230	122	15	5	-10.8	-1.0
	0-20	1040	1555	60	137	51.5	7.7
<i>O-G-G+</i>	20-40	170	107	1	0	-6.3	-0.1
	40-60	110	67	0	0	-4.3	0.0
	0-60	1320	1729	61	137	40.9	7.6
	0-5	720	925	48	79	20.5	3.1
	5-10	660	260	32	25	-40.0	-0.7
	10-20	320	311	-16	20	-0.9	3.6
<i>O-G-G+</i>	0-20	1700	1496	64	124	-20.4	6.0
	20-40	130	194	35	35	6.4	0.0
	40-60	60	147	39	66	8.7	2.7
	0-60	1890	1837	138	225	-5.3	8.7

consists of 4-8 and 2.5-4 $g \cdot m^{-2}$ of organic carbon per one centimetre per year in the layers of 5 and 20 cm, respectively, whereas in the case of elimination a simultaneous loss of carbon accounted for 1.7-1.8 $g \cdot m^{-2}$ per centimetre of the total 20 cm per year. This means that some unstable humus compounds that had formerly accumulated belonged to breakdown processes. This became evident not only in the variant of *L-G-G-*, but also in the variant of *O-G-G+* at a depth of 5-20 cm resulting in the loss of carbon at a rate of 2.5 $g \cdot m^{-2}$ per centimetre per year there. The breakdown of underground organic residues in layers also tends to be characteristic of the transformation processes during the third decade (Table 3). It is more noticeable under the very top as well as at a depth of 20-40 cm. Probably the oxidational decomposition of the unstable carboxydes and/or carbohydrates of humus substances as well as rapid autolysis of microbial matter formed and deamination of humus molecules are due to these changes in the overall carbon-nitrogen status (Martin & Haider, 1971; Александрова, 1972). That is why the decade features are in negative balance and instead of an increment a loss of formerly accumulated humus material occurred.

However, the cease of humification and progressing destruction of the humus compounds formed could even demonstrate a dynamic cyclicality of changes in pedogenesis (Smeck et al., 1983). Loss of organic carbon and

Table 4

Periodicity of mean annual increments of organic carbon and nitrogen, $g \cdot m^{-2}$

Variant	Depth, cm	C			N		
		Decades					
		1.	2.	3.	1.	2.	3.
G-G-G+	0-5	95	-36	40	5.0	0.3	5.1
	0-20	166	-47	93	7.0	2.3	14.7
	0-60	188	7	82	7.1	18.6	4.4
L-G-G-	0-5	45	34	-9	4.1	1.6	0.9
	0-20	70	87	-35	6.3	5.9	-0.6
	0-60	77	107	-33	6.5	11.1	2.5
B-G-BG+	0-5	8	47	35	4.1	-1.2	5.5
	0-20	17	87	52	6.0	0.0	7.7
	0-60	21	111	41	6.0	0.1	7.6
O-G-G+	0-5	11	61	21	3.8	1.0	3.1
	0-20	31	139	-20	4.8	1.6	6.0
	0-60	27	162	-5	4.9	8.9	8.7

nitrogen accumulated during the first decade was ascertained after the passage of the second decade (Рейнтам & Погорелова, 1986; Reintam & Pogorelova, 1987). Similar data have been obtained by several authors (Александрова, 1972; Таранов, 1977; Ужегова & Махонина, 1984). The wavy character of mean annual increments by decades (Table 4) tends to confirm the stage periodicity of mineralization and humification described by Sau in the alternation of a 6—7-year period in a thin topsoil (Sau, 1979, 1983). The periodical consumption of unstable aromatic humus substances for the microbial activity shows an increase in depth although their narrow C:N ratio (everywhere less than 10) suggests that they are already quite perfect (Flaig, 1971) there. Probably free fulvic acids and Ca-fulvates, which are unable for permanent condensation and transformation into more stable complexes, prevail among unstable humus substances. The poor and/or negative accumulation of organic carbon could be only partly explicable with the absence of rapidly mineralizing plant residues, which would have acted as a producer of CO_2 above the soil surface and would have thus induced the formation of humus in topsoil (van Veen et al., 1991).

Against the background of an evident humus accumulateness of the soils formed during the period of thirty years a highly significant correlation between the pedogenetic agencies of phytomass origin and real result of humus-accumulative pedogenesis is found under the conditions of permanent vegetation and complete return of the produced organic matter into the soil (Table 5). The substitution of perennial plants (grasses) for an annual plant (summer barley) has led to the intensification of humus-accumulative phenomena, with the possible and actual sources of organic substances considered being nearly in the same order. In the conditions of spontaneous herbage the progress of the humus-accumulative process is highly significant in the topsoil, but the predominance of breakdown reactions is not of less significance during the third decade. Losses of humus substances that were earlier formed are characteristic of the third decade in the case of the removal of the aboveground production.

The humus-accumulative process results in the formation and progress of a humus horizon in the soil. Gagarina and Tsyplenkov (Гаргарина & Цыпленков, 1974) described the formation of a leached from carbonates thin humus-accumulative microaggregated soil profile on a pure loess under an oak stand of forest steppe during ten years. The formation of

Table 5

Possible and real humification by layers, $g \cdot m^{-2}$

Variant	Aboveground mass for humification	Organic C in top layer of 5 cm	Total possible phytomass for humification	Organic C in top layer of 20 cm	Organic C in layer of 60 cm
<i>G-G-G+</i>	399	396	987	935	823
<i>B-G-BG+</i>	277	345	729	515	409
<i>O-G-G+</i>	445	205	1112	-204	-53
<i>L-G-G-</i>	—	-89	556	-347	-328

a humus horizon of 2.5–5 cm on till under various grass swards was ascertained already within six to seven years (Sau, 1983). In Virginia a distinct surface humus horizon had even formed within three years (Roberts et al., 1988a).

In the conditions of our experiment the passage of the first decade showed the presence of a dwarf humus horizon of 3.5–4 cm only under the clover–grasses sward and lucerne, while under barley it was practically absent (Reintam, 1982). Within the following decade the humus-accumulative process developed quite intensively and a humus horizon was found under all variants (Reintam & Pogorelova, 1986; Reintam & Pogorelova, 1987). Yet nowhere its thickness exceeded 3.5–5 cm.

In spite of the cyclic intensity of humus accumulation induced by the above- and underground organic matter of plant origin the process was especially developed in the third decade. The humus horizon is a diagnostic one everywhere now, it reaches down to 8 and 7 cm in the variants of *G-G-G+* and *B-G-BG+*, respectively, and exceeds 5 cm in the other cases. The mean annual increment amounts to 1.7–3 mm in depth whereas the rate of more than 2.5 mm tends to be characteristic of the last decade. A transitional *ABm*-horizon has formed below that of humus (*A*). It contains already 0.4–0.5% of organic carbon and 0.05–0.06% of nitrogen, extends to a depth of 10–15 cm, in *G-G-G+* and *O-G-G+* even to 20 cm. Some mezo- and macromorphological signs of argillization *in situ* as well as of slight lessivage can also be observed. But these were still too shallow to meet actual argillic, cambic, and/or luvic criteria (Schafer et al., 1980).

Ten years ago (within the twenty-year period of soil formation) 37–43% of pedogenetic organic carbon and 50–75% of nitrogen were in the top layer of 5 cm. After the passage of thirty years these features formed 40–52 and 33–35(61)%, respectively. In the layer of 20 cm 81–90% of carbon has already accumulated and only 2–8% occurs deeper than 40 cm. Thus the intensification of humus accumulation has quantitatively developed in the topsoil and spatially penetrated deeper into the soil. The formation of a new macro- and microstructure and the decrease in bulk density in the topsoil enabled a slight upward growth of the humus soil profile, too (Reintam, 1990a). Such a progress of humus-accumulative pedogenesis is more obvious under permanent herbaceous vegetation (Tables 3 and 4), where the storage of organic carbon in the topsoil is rather small (40%), but its distribution within 20 cm is more noticeable and homogeneous than in all other cases. In spite of the negative annual increment an analogy can be drawn in *L-G-G-* where the development of soil processes was ensured by the participation of root residues within the whole period investigated.

Table 6

Supplies of CaCO_3 in fine earth, $\text{kg} \cdot \text{m}^{-2}$

Variant	Depth, cm	Year			
		1964	1974	1984	1994
G—G—G+	0—5	5.8	5.3	4.5	4.9
	0—20	23.3	18.0	22.6	15.4
	0—60	69.8	44.6	37.1	29.0
L—G—G—	0—5	5.8	5.0	4.1	4.4
	0—20	23.3	24.4	22.9	22.4
	0—60	69.8	61.8	52.4	60.6
B—G—BG+	0—5	5.8	5.5	6.8	6.9
	0—20	23.3	25.3	29.1	22.9
	0—60	69.8	67.3	80.6	43.9
O—G—G+	0—5	5.8	5.6	4.5	6.1
	0—20	23.3	27.6	25.1	28.4
	0—60	69.8	70.4	81.6	83.1

The substitution of perennial vegetation for barley resulted in a relative enrichment of the thin topsoil not only with organic carbon, but also with nitrogen. At that entire pedogenic nitrogen has accumulated in the top solum of 20 cm. In other cases the distribution of nitrogen (9—25 and 11—29% in the layers of 20—40 and 40—60 cm, respectively) is much more homogeneous than that of carbon. This demonstrates a rather great importance of microbial albumins to primary humus-accumulative pedogenesis.

Weathering of carbonates is characteristic of pedogenesis on calcareous parent strata from the early stages of the process (Гагарина & Цыпленков, 1974; Reintam, 1982; Haidouti & Yassoglou, 1982). It leads, on the one hand, to their leaching and, on the other, to the breakdown of calcareous skeleton and enrichment of fine earth with carbonates mobilized from pebble and/or gravel (Table 6). The last phenomenon seems to be more widely spread within the last decade in the conditions of rapid intensification of biological activity than earlier. Simultaneously an obvious leaching is typical of the permanent cover of perennials and intensive humus-accumulative processes occur there.

Changes in the carbonate regime (amongst these the bilateral movement of calcareous weathering—pedogenetic products) are evidently proportional to the accumulation of organic residues, their transformation, and humification. Predominant fulvicity of humus accumulated is of importance to the organic—mineral interactions.

Compared with the former status (Reintam, 1982; Рейнтам & Погорелова, 1986; Reintam & Pogorelova, 1987) the group and fractional composition of humus has been quite unstable (Tables 7 and 8). A decrease (by 2—3 times) in the quantity of humic acids, called by Grishina and Orlov (1977) humification degree, and an accompanying increase in fulvicity could be interpreted as a favoured polycondensation of humic acids and transformation of products into humins in the previous arid season. However, inhibition of the transformation of primary fulvic products into humic ones could not be excluded either in the conditions of a series of mild winters followed with a severe one (Александрова, 1980). Fulvic acids that remained uncondensated on the background of almost stable solubility of humus are able to proceed to the crystalline structure of clay minerals. These fulvic compounds, extractable by 0.5 M sulphuric acid, showed within the last decades an increase everywhere except for the thin top of G—G—G+.

Composition of 30-year-old humus in percentage per total organic carbon

Characteristics	Variant & Depth, cm											
	G-G-G+			L-G-G-			B-G-BG+			O-G-G+		
	0-8	10-15	15-20	0-5	5-10	10-20	0-7	7-11	11-20	0-5	5-10	10-20
Total org. C, %	1.46	0.48	0.11	1.09	0.48	0.18	1.22	0.43	0.10	1.22	0.37	0.23
Nitrogen, %	0.17	0.06	0.05	0.12	0.06	0.04	0.13	0.05	0.02	0.12	0.05	0.03
C:N	8.6	8.0	2.2	9.1	8.0	4.5	9.4	8.6	5.0	10.2	7.4	7.7
Humic acids (H.a.)	7.5	0.0	0.0	4.4	0.0	0.0	4.6	0.7	0.0	3.4	0.3	0.0
1	2.9	0.0	0.0	0.2	0.4	4.4	0.0	0.0	3.0	0.2	0.5	0.0
2	3.4	4.6	5.5	5.1	6.0	2.2	3.9	5.8	7.0	4.6	5.4	3.5
Σ	13.8	4.6	5.5	9.7	6.4	6.6	8.5	6.5	10.0	8.2	6.2	3.5
Fulvic acids (F.a.)	3.4	4.2	0.0	4.6	5.6	6.1	1.8	5.8	8.0	4.6	11.1	0.0
1	10.6	8.8	2.7	9.5	9.6	7.8	11.9	9.8	6.0	12.0	3.8	8.3
2	6.8	1.7	22.7	2.6	0.2	0.6	0.6	1.6	19.0	0.1	18.1	1.3
3	8.0	11.3	20.0	12.8	5.6	17.8	8.1	7.2	21.0	12.0	13.5	12.2
Σ	28.8	27.0	45.4	29.5	21.0	32.3	22.4	24.4	54.0	28.7	46.5	21.8
Extracted by 0.5 M H ₂ SO ₄	13.4	22.7	32.7	16.3	21.4	13.9	13.7	22.8	14.0	17.1	19.7	17.0
Total extractable	56.0	54.3	83.6	55.5	48.8	52.7	44.6	53.7	78.0	54.0	72.4	42.3
Nonsoluble residue (humins)	44.0	45.7	16.4	44.5	51.2	47.3	55.4	46.3	22.0	46.0	27.6	57.7
H.a.: F.a.	0.5	0.2	0.1	0.8	0.3	0.2	0.2	0.3	0.2	0.3	0.3	0.1
1st fr.: 2nd fr.*	1.9	5.2	0.1	5.0	16.0	1.6	27.5	6.6	0.3	51.3	0.2	6.4

* 1st fr. (H.a.+F.a.): 2nd fr. (H.a.+F.a.).

Some indices of the state of humus within 30 years

Year	Variant & Depth, cm											
	G-G-G+			L-G-G-			B-G-BG+			O-G-G+		
	0-5	5-10	10-20	0-5	5-10	10-30	0-5	5-10	10-20	0-5	5-10	10-20
	Degree of humification (Grishina & Orlov, 1977)											
1974	11	11	27	0	0	x	4	x	x	8	x	x
1984	16	16	9	18	14	13	14	14	13	13	10	17
1994	14	5	6	10	6	7	9	7	10	6	4	4
	Role of active humic acids (1st fr.), % of total H.a.											
1974	46	43	21	42	0	x	75	x	x	17	x	x
1984	39	50	28	40	30	41	33	39	33	53	23	23
1994	54	0	0	45	0	0	54	11	0	42	0	0
	Role of stable humic acids (3rd fr.), % of total H.a.											
1974	6	0	0	0	0	x	0	x	x	0	x	x
1984	45	46	72	34	35	59	67	61	67	47	77	77
1994	25	100	100	53	94	33	46	89	70	56	100	100
	Solubility of humus, % of total C											
1974	74	88	94	70	68	x	71	x	x	68	x	x
1984	56	66	51	57	44	74	58	59	96	56	76	76
1994	56	54	84	56	49	53	45	54	78	54	42	42
	Extracted by 0.5 M H ₂ SO ₄ , % of total C											
1974	18	23	17	22	16	x	27	x	x	22	x	x
1984	13	16	13	11	8	11	18	17	21	14	16	16
1994	13	23	33	16	21	14	14	23	14	17	17	17
	1st fraction of humic and fulvic acids : 2nd fraction of humic and fulvic acids											
1974	0.6	0.3	0.2	0.7	0.2	x	0.8	x	x	0.3	x	x
1984	1.2	0.5	0.6	1.9	0.9	0.6	0.5	0.7	0.3	1.8	0.5	0.5
1994	1.9	5.2	0.1	5.0	16.0	1.6	27.5	6.6	0.3	51.3	6.4	6.4
	Humic acids : Fulvic acids											
1974	0.3	0.2	0.6	0.1	0.0	x	0.1	x	x	0.2	x	x
1984	0.6	0.5	0.3	0.6	0.7	0.3	0.5	0.5	0.2	0.5	0.4	0.4
1994	0.5	0.2	0.1	0.8	0.3	0.2	0.2	0.3	0.2	0.3	0.1	0.1

x, not determined.

Table 9

Supplies and dynamics of pedogenetic nonsiliceous Fe oxide in the solum of
50 cm, $g \cdot m^{-2}$

Variant	Supply		Mean annual increment	
	1974	1994	1964—74	1974—94
G—G—G+	1830	5354	183	176
L—G—G—	930	4236	93	165
B—G—BG+	480	5559	48	254
O—G—G+	180	3365	18	159

The total quantity of humic—fulvic complexes connected with stable sesquioxides and clay minerals (the third fraction of humus acids) was rather stable during two decades (Table 7; Рейнтам & Погорелова, 1986; Reintam & Pogorelova, 1987). Their percentage (11—20% of total organic carbon) forms as a result of the interfractional transformation of fulvic compounds into humic ones. The relative increase in the share of the latter (Table 8) is probably connected with the transformation of calcic humates into humins. An increase in the role of humic—fulvic complexes of the third fraction within the third decade at a depth of 5—10 cm at the level of significance tends to be accompanied by an increase in mobility and fulvicity of humus as well as by a negative balance of humification products (Tables 3 and 4).

An inhibited transformation of fulvates into humates is probably explicable by relative stability of active R_2O_3 -fulvates in the conditions of base saturation and neutral reaction, which results in their accumulation *in situ* and in the transformation of Ca-humates into humins. That is why the humic acids connected both with mobile (active) sesquioxides and alkaline earth metals are often absent deeper than 5—10 cm, but with a simultaneous increase in fulvicity and decrease in solubility of humus the ratio between the first and second fractions (all humus acids connected with active sesquioxides and alkaline earths, respectively) has changed towards the benefit of the former. Such qualitative changes could take place step by step within the whole last decade characterized in general by warm summers and mild winters, but also due to the climatic contrasts between the seasons of the very last year.

The greatest changes in the qualitative composition of humus are determined by the formation of R_2O_3 -fulvic complexes on the account of the transformation of Ca-fulvic—humic ones (Tables 7 and 8). Formerly it was established that the humification of organic residues begins with the formation of free fulvic acids (Reintam et al., 1982). Such a procedure seemed to be also typical of perennial herbage material although the relative amount of free fulvic acids showed a clear tendency to decrease by decades ensuring at that sufficient weathering-pedogenetic activity and the transformation into more complicated humus substances discussed above. The accumulation of nonsiliceous iron oxides characteristic of pedogenetic weathering of ferri- and alumosilicates and argillization is noteworthy (Table 9) in the conditions of diminishing calcareousness (Table 6), but even in case of neutral reaction (pH value 7.0—7.5) and high base saturation (over 98%).

While the differences between pedogenetic agents (organic residues in the variants) and results (nonsiliceous products of silicates transformation) were quite big within the first decade of interactions a marked

unification took place during the third one both in the extent of prerequisites for soil formation (Tables 1 and 2) and in its results (Table 9). An astonishing homogeneous progress of iron mobilization was characteristic of the permanent impact of perennial herbage. The previous weak process under barley and without vegetation had highly intensified by the end of the third decade. These changes, which are connected with an increase in extractable iron and total nitrogen, reflect rapid pedogenesis in unchanged parent materials in a humid environment (Roberts et al., 1988a).

According to Roberts et al. (1988a) dissolution and leaching of carbonates, oxidation, organic matter incorporation and decomposition, shrink and swell, freeze and thaw processes rapidly transform the surface properties of soils. That is why the specific surface area had increased 1.3—1.7 times in the thin topsoil and 1.1—1.4 times in the rest of the solum in a comparatively short period of time within the third decade of pedogenesis. The higher the intensity of carbon and/or nonsiliceous iron accumulation, the greater was the increase in the specific surface area.

Iron activity (after Schwertmann) exceeded 40% only in the thin 5-cm topsoil of two variants (*B—G—BG+* and *O—G—G+*). In all other cases it was within the limits of 20—30%. As the pH value was always higher than 6.5 and 7.0 in salt and water solutions, respectively, such an iron relationship tends to demonstrate a rapid recrystallization of amorphous pedogenetic products, characteristic of brunification (Зонн, 1982). The content of amorphous iron hydroxides was quite homogeneous (0.25—0.35%) within the last two decades. This shows their continued formation and recrystallization, which can probably give rise to seasonal and layered changes in the balance of the humus-accumulative process.

The mobilization of amorphous Al-oxide is wavy by decades being induced by intensive transformation of aluminosilicates due to the influence of fulvic humus. Under the protective activities of the latter as well as of amorphous silica the translocation of clay is possible and lessivage can develop (Reintam, 1967). Some slight signs of translocative differentiation of solum have already been ascertained, but their interpretation and discussion should be presented in another paper.

CONCLUSIONS

Humus-accumulative pedogenesis continued during the 30 years of the experiment. The mean annual increase in the depth of the humus horizon formed was more than 2.5 mm within the last decade. The net accumulation of organic carbon in the profile of 60 cm was ensured by approximately equal amounts of humifiable phytocoenotic agents in the conditions of permanent vegetation and complete return of the produced organic matter into the soil as well as by the substitution of perennial plants for an annual one. In other conditions the mineralization of formerly accumulated unstable humus was ascertained. Temporal periodicity of mineralization and humification is characteristic of primary pedogenesis because of the changes in unstable humic—fulvic relationships although the narrow C:N ratio tends to demonstrate a high abundance of nitrogen and perfection of humus formed.

As compared with the previous decades the solubility of humus has decreased, but fulvicity has increased. An inhibited transformation of fulvates into humates is explicable by the relative stability of active fulvates of sesquioxides in the presence of high base saturation, calcareousness, and probable transformation of Ca-humates into humins. The main qualitative changes in the humus composition are determined by the

formation of R_2O_3 -fulvic complexes on the account of the transformation of Ca-fulvic—humic ones. This results in an increase in the relative amounts of humic—fulvic complexes connected with clay minerals and fulvic acids bound in their crystalline structure.

The continued accumulation of nonsiliceous crystalline iron oxides is characteristic of pedogenesis within the third decade being expressed by a rapid recrystallization of amorphous compounds formed. Leaching of carbonates, oxidation processes, accumulation and decomposition of organic residues and unstable humus substances, their enrichment with nitrogen, and formation and transformation of nonsiliceous ferric compounds represent continued primary pedogenetic phenomena on calcareous red-brown till. A progress of argillization *in situ* and clay translocation (lessivage) can also be observed.

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