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Non-typical degraded and regraded humus forms in metal-contaminated areas, or there and back again

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ABSTRACT

Chemically contaminated soils are not included in the European morpho-functional classification of humus systems and forms. However, they differ from the uncontaminated analogs in biological, chemical, and physical features. We investigated the topsoils contaminated by long-term sulfur dioxide and metal emissions from a copper smelter (southern taiga; conifer and deciduous forests; the Middle Urals, Russia). The smelter has been in operation for more than 80 years, but ten years ago, emissions almost ceased which initiated the recovery of adjacent ecosystems. We performed 1,155 humus profile descriptions on 231 sampling plots in highly contaminated, moderately contaminated, and background sites. In the contaminated sites, we found 21 non-typical humus forms that do not fit the European classification. Earthworms' extinction caused by soil toxicity resulted in a Mull-to-Mor transformation of humus profiles. Recent recolonization of poorly decomposed litter by earthworms and other soil macrodetritivores after emission cessation triggered a Mor-to-Mull shift. Because different topsoil layers react to environmental changes with unequal rates, signs of several stages of humus evolution can be imprinted in one profile. More rapid degradation of organic horizons than organic-mineral ones results in non-typical Mor forms when the thick non-zoogenic OF combines with zoogenic A horizon. Zoogenic O horizons' heterogeneity due to non-zoogenic inclusions, or inversion of the litter layers sequence, or lagging of A horizon recovery behind the O horizons, represents the discrepancies between non-typical regraded and typical humus forms. Mor-to-Mull recovery can occur with and without passing the Moder forms. In the first path, all intermediate stages at least fitted to one of the known humus systems. In the second path, intermediate humus forms are represented by non-zoogenic OF conjoining highly zoogenic OF and lacking OH horizon (we named such forms Mormull). In both paths, the recovery starts from the upper litter layer and spreads downward, i.e., the signs of the zoogenic activity change in the opposite direction than within the natural Mor profile. We consider the non-typical humus forms as non-equilibrium topsoil states that are ephemeral on the forest succession scale. We described non-typical humus forms in detail, proposed their evolutionary diagram, nomenclature, and preliminary typology, and extended the Humus Index with non-typical forms.

1. Introduction

Humus in the pedological sense (humus form, humus profile) has been investigated for nearly a century and a half. One of the most prominent achievements of recent years in this domain is the European morpho-functional classification of humus systems and forms (hereafter referred to as the Classification) (Zanella et al., 2011; Zanella et al., 2018a; Zanella et al., 2018e). The undoubted advantages of the Classification, which determine its increasing popularity, are clear diagnostic criteria, theoretical validity, internal consistency, and unification of national taxonomies and terminologies. The detailed practical guidelines (Zanella et al., 2018c; Zanella et al., 2018h), including those implemented even on mobile phones (Zanella et al., 2019), facilitate the widespread application of the Classification.

Ideally, the taxonomy should include all existing or even potentially possible humus diversity. The Classification covers most of the topsoil types within Europe and even a wider geographic area. The humus forms are most comprehensively represented for forests on automorphic soils (Zanella et al., 2018g; Zanella et al., 2018h). In the latest version of the Classification, considerable attention is paid to temporary wet soils (Zanella et al., 2018i), permanently waterlogged soils (Zanella et al., 2018c; Zanella et al., 2018d), and Para systems (Zanella et al., 2018f).

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The Classification presents man-induced humus forms in much less detail and includes only artificially constructed substrates, urban soils, and agricultural soils (Zanella et al., 2018b; Zanella et al., 2018k; Zanella et al., 2018l). Chemically contaminated soils have not been identified and are not mentioned in the Classification. This gap is frustrating since the pollution-induced biological, chemical, and physical processes result in morphological differences between contaminated soils and uncontaminated analogs. Severe industrial pollution rarely extends more than one or two tens of kilometers from a source of emissions. Although contaminated areas are negligible compared to ones with other human-affected soils, their investigation helps study soil evolution, resistance, and resilience to heavy impacts.

The main factors that can alter the topsoil morphology in metal-contaminated areas are well known. In the vicinity of non-ferrous factories, extremely high concentrations of metal(loid)s, increased soil acidity and a reduced CEC (e.g., Dudka and Adriano, 1997; Ettler, 2016) cause high soil toxicity to biota and decrease the diversity and abundance of soil fauna (e.g., Nahmani and Lavelle, 2002; Vorobeichik et al., 2019) and microflora (e.g., Nordgren et al., 1983; Mikryukov and Dulya, 2017; Mikryukov et al., 2020). In combination with high soil acidity, the detrimental effect of polymetallic pollution on earthworms is especially crucial (Bengtsson et al., 1983; Spurgeon and Hopkin, 1996; Vorobeichik, 1998; Nahmani et al., 2003; Vorobeichik et al., 2019). These soil ecosystem engineers affect soil structure, food webs, nutrient cycles, and plant litter disappearance (e.g., Lavelle et al., 1997; Lavelle et al., 2006).

Visible to the naked eye, the accumulation of a thick layer of poorly decomposed forest litter due to slow-down organic matter decomposition was noted in early studies of terrestrial ecosystem responses to industrial pollution (Strojan, 1978; Coughtrey et al., 1979; Freedman and Hutchinson, 1980). Subsequently, it was repeatedly documented in other case studies (e.g., Vorobeichik, 1995; Gillet and Ponge, 2002; Hale and Robertson, 2016). However, only a few pedological works sophisticatedly explored the changes of topsoil morphology in highly contaminated areas (Dijkstra, 1998; Gillet and Ponge, 2002; Filzek et al., 2004; Ciarkowska and Gambuś, 2005).

Previously, we have described the humus alteration induced by long-term copper smelter emissions in the southern taiga (Korkina and Vorobeichik, 2018). When approaching the smelter, the primary Mull forms are replaced by the Moder forms and then the Mor forms, similar to a humus change in a latitudinal or altitudinal gradient. In both cases, Mull-to-Mor replacement is caused by the transformation of the macroinvertebrate decomposers spectrum, namely, a successional decrease in earthworm abundance with their subsequent disappearance, first endogeic species and then epigeic ones (Vorobeichik, 1998; Vorobeichik et al., 2019).

In most cases, Moder and Mor forms in contaminated areas completely met the Classification criteria of natural Moder and Mor forms. However, sometimes there was an incomplete correspondence of contaminated humus forms to the Classification's specifications (Korkina and Vorobeichik, 2016). In particular, we found that the thick non-zoogenic OF typical for Mor forms was combined with a zoogenic A horizon atypical for this humus system. We suggested that the organic-mineral horizons react to pollution with some delay, compared to the upper-positioned organic ones, and retain the previous states' fingerprints (Korkina and Vorobeichik, 2016).

Quite recently, we found more mismatches when studying the initial stages of ecosystem recovery after an almost complete cessation of emission. In the past few years, in moderately polluted sites we have registered an increase in the abundance of earthworms and their encroachment towards the smelter (Vorobeichik et al., 2019) caused by the mitigation of excess soil acidity (Vorobeichik and Kaigorodova, 2017). Peculiarities of soil macrofauna recovery were clearly reflected in the structure of organogenic horizons. Specifically, the humus forms mismatching the Classification indicated faster recovery of organic horizons than organic-mineral ones or a delay in the recovery of some layers within the O horizons.

It turned out that such non-typical humus forms occur quite frequently, as we discuss further (see Table S1 and Table 1), so they cannot be considered just occasional curiosities. It would be more logical to include non-typical humus forms in the taxonomy than to keep "pretending" that they do not exist. The first step towards this is their complete inventory, i.e., a detailed description of different kinds of atypical topsoil morphology. The purpose of this article is to provide detailed documentation on non-typical humus forms found in contaminated areas. Also, we propose their nomenclature and typology, discuss possible mechanisms of formation, and present an evolutionary diagram.

2. Materials and methods

2.1. Study area

The study area is located on the geographical border between Europe and Asia, near the city of Revda, 50 km west of Yekaterinburg (Sverdlovsk region, Russia) (Fig. 1). The territory belongs to the southern taiga subzone in the Middle Urals' low mountains (altitudes are 100–450 m above sea level). The climate is warm-summer humid continental, Dfb according to Köppen-Geiger classification (Peel et al., 2007). The annual air temperature is + 2.0 °C; the annual precipitation is 550 mm; the warmest month is July (+17.7 °C) and the coldest month is January (–14.2 °C); the annual sum of active temperatures (>0 °C) is 2330°-days (mean values for the last 40 years, 1975–2015, according to the data of the nearest meteorological station in Revda). The snowless period is about 215 days (from April to October), the maximum height of the snow cover is about 40–60 cm.

Primary coniferous forests (*Picea abies*, *Abies sibirica*, and *Pinus sylvestris*) and secondary deciduous forests (*Betula pendula, Betula pubescens,* and *Populus tremula)* prevail. Soil formation occurs on eluvium and eluvium-diluvium of bedrock metamorphic rocks (shales, sandstones, quartzites, and silicified limestones). Soil cover is formed mainly by soddy-podzolic soils (Albic Retisols, Stagnic Retisols, and Leptic Retisols), burozems (Haplic Cambisols), and grey forest soils (Retic Phaeozems).

Middle Ural Copper Smelter is located close to the border between Urals' western and eastern slopes (N 56°50′37″ E 59°52′44″). Spruce and fir forests with nemoral flora on loam or heavy loam soils prevail on the western slope, and pine forests on sandy loam or light loam soils dominate on the eastern side.

2.2. Regional features of soil fauna

Let us briefly describe soil macrofauna features of the study area that are essential for interpreting the results of our long-term studies (Vorobeichik, 1998; Vorobeichik et al., 2012; Vorobeichik et al., 2019). Soil macrodetritivores are abundant and include the following taxa: earthworms, enchytraeids (they occupy an intermediate position between macrofauna and mesofauna), millipedes, Nematoceran larvae, Coleopteran larvae (Elateridae), and mollusks. The last three taxa are classified as phytosaprophages. Among Collembolans (35,300 ind. m^{-2}), hemiedaphic species dominate (*Folsomia manolachei*, *F. quadrioculata*, *Parisotoma notabilis*, *Isotoma viridis*, *Isotomiella minor*) (Kuznetsova, 2009).

The main groups among macrodetritivores are annelids, earthworms (235–285 ind. m^{-2}, cocoons excluded) and enchytraeids. Unfortunately, we do not have data on the species composition of enchytraeids and their abundance with extraction by the wet funnel method; the density of hand-sorted enchytraeids, that is individuals of body length over 2 mm, is 300–1,000 ind. m^{-2}. Epigeic earthworms feeding on plant litter and inhabiting only the O horizon are represented by the small (2–4 cm long) *Dendrobaena octaedra* and *Dendrodrilus rubidus*. Epi-endogeic species dwelling the O horizon and the upper (0–10 cm) layer of A horizon are the medium (5–10 cm) *Rhiphaeodrilus diplotetratheca* and *Lumbricus rubellus* and the very large (up to 20 cm) *Eisenia atlavinyteae*. Endogeic

Table 1
Number of mini-plots with typical and non-typical humus forms in pollution zones.

Humus Index	Typical humus form	Natural attraction or degradation				Regradation			
		Non-typical humus form	Background	Buffer	Impact	Non-typical humus form	Background	Buffer	Impact
10	Eumor	Deg Term	0	11	132	Reg Term	0	2	2
		Deg preTerm	0	9	35	Reg preTerm	–	–	–
9	Humimor	Deg Term	0	11	48	Reg Term	0	8	40
		Deg preTerm	0	8	18	Reg preTerm	0	2	23
8	Hemimor	Deg Term	0	1	0	Reg Term	0	10	13
		Deg preTerm	0	0	1	Reg preTerm	0	13	12
7	Dysmoder	Deg Term	0	5	3	Reg Term	–	–	–
		Deg preTerm	–	–	–	Reg preTerm	–	–	–
	–	–	–	–	–	Hemimormull Reg Term	0	43	39
						Hemimormull Reg preTerm	0	16	6
6	Eumoder	–	31	29	0	Reg Term	0	6	0
						Reg preTerm	0	9	0
	–	–	–	–	–	Eumormull Reg Term	0	60	7
						Eumormull Reg preTerm	0	18	3
5	Hemimoder	–	56	11	2	Hemimoder Reg Term	0	26	1
						Quasihemimoder Reg	0	8	1
	–	–	–	–	–	Oligomormull Reg Term	0	6	0
						Oligomormull Reg preTerm	0	2	0
4	Dysmull	–	257	12	4	Quasidysmull Reg	0	5	0
	–	–	–	–	–	Dysmormull Reg	0	49	15
3	Oligomull	–	6	0	0	Quasioligomull Reg	0	16	0
2	Mesomull	–	0	4	0	–	–	–	–
1	Eumull	–	0	0	0	–	–	–	–

The dash denotes the absence of a particular humus form.

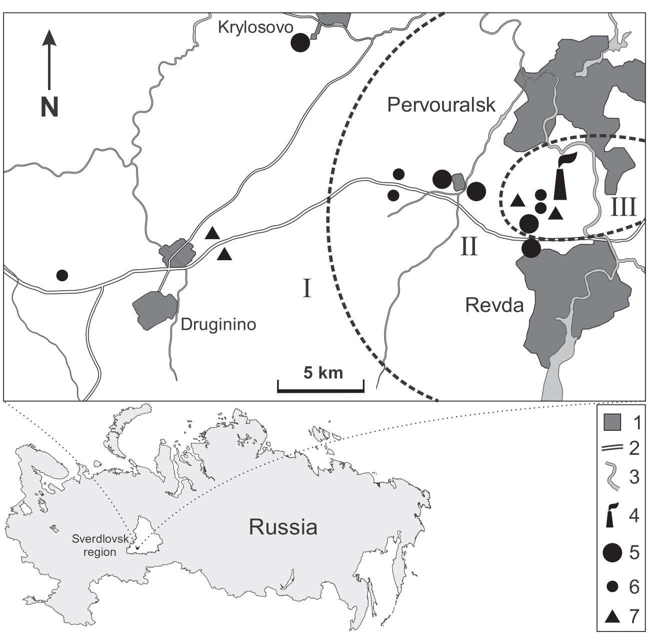

Fig. 1. Study area. Roman numbers denote pollution zones: (I) background, (II) buffer, and (III) impact. Symbols denote (1) settlements; (2) highway; (3) rivers and water bodies; (4) the Middle Ural Copper Smelter; (5) study site with circular plots in regular grid nodes; (6) study site with 3–6 plots; (7) study site with one plot.

species feeding on soil organic matter in the middle (10–20 cm) of mineral horizons are represented by the small (5–7 cm) *Aporrectodea rosea* and the large (7–12 cm) *Perelia tuberosa* and *Octolasion lacteum.* In coniferous forests, epi-endogeic species dominate (70–80% in density, mainly *R. diplotetratheca*), and in deciduous forests endogeic species are of comparable abundance. In the meadows, these endogeic species are accompanied by the large (up to 15 cm) *Aporrectodea caliginosa*, which dwell in mineral layers deeper than 20 cm. Anecic earthworms, typical for the more western European regions (e.g., *Lumbricus terrestris*), are absent; their role is played to some extent by epi-endogeic species.

Nematoceran larvae are represented by large specimens (3–4 cm long, Tipulidae and Limoniidae) and small specimens (about 0.5–1 cm, Bibionidae, Sciaridae, Chironomidae, Cecidomyiidae, and some others); their density is 100 ind. m^{-2}. The abundance of wireworms (Elateridae) is 25–60 ind. m^{-2}. According to modern concepts, most click beetle larvae are omnivores, including detritivores, although wireworms were earlier considered herbivores or carnivores, exclusively (Samoylova, 2018). The dominant species in the study area (*Athous subfuscus* and *Dalopius marginatus*) are also considered omnivores (Samoylova, 2018). The density of mollusks is 290–340 ind. m^{-2} (snails, 0.5–1 cm in diameter and slugs, 4–5 cm long).

Compared to western European or more southern regions, woodlice (Oniscoidea) and wood cockroaches (*Ectobius*) are absent or occasional. Another regional feature is that millipedes (only *Polyzonium germanicum* 1.0–1.5 cm long) have low abundance (6–23 ind. m^{-2}).

2.3. Source of pollution

The smelter has been in operation since 1940, and until recently it was one of the most significant sources of pollution in Russia. Main pollutants are sulfur, fluorine, and nitrogen gaseous compounds and toxic metal(loid)s (Cu, Pb, Zn, Cd, Fe, Hg, and As). Emissions were at a maximum in the mid-1970 s, reaching 350,000 t $year^{-1}$, then gradually decreased: 225,000 t $year^{-1}$, 148,000 t $year^{-1}$, and 63,000 t $year^{-1}$ in 1980, 1990, and 2000, respectively. After factory reconstruction in 2010, emissions decreased to about 3,000–5,000 t $year^{-1}$ (Vorobeichik and Kaigorodova, 2017).

According to 2016 data, 0.5–3 km west of the smelter, metal concentrations in the forest litter are very high: Cu – 3,484, Pb – 2,462, Cd – 17, Zn – 650 mg kg^{-1}, exceeding background values (20–30 km west) 93, 37, 7, and 3 times, respectively; while pH shifted from a background level of 5.9 to 4.9 (Korkina and Vorobeichik, 2018). Four–five km west of the smelter, metal concentrations are lower: Cu – 1,138, Pb – 903, Cd – 14, Zn – 508 mg kg^{-1}, still exceeding background values 30, 13, 6, and 2.6 times, respectively; pH is 5.1 (Korkina and Vorobeichik, 2018).

2.4. Pollution-induced ecosystem shifts

Signs of ecosystem transformation are typical for contaminated areas near copper smelters: decreased productivity of tree stands (Usoltsev et al., 2012) and reduced diversity of ground-level vegetation (Vorobeichik et al., 2014), as well as changes in soil morphology (increase in O horizon thickness, degradation of soil structure) and soil chemistry (higher acidity and lower concentrations of exchangeable Ca and Mg) (Vorobeichik, 1995; Kaigorodova and Vorobeichik, 1996; Vorobeichik and Kaigorodova, 2017; Korkina and Vorobeichik, 2018). Pollution also harms soil organisms: it slows down cellulose decomposition (Vorobeichik, 2007), inhibits the specific respiratory activity of forest litter (Smorkalov and Vorobeichik, 2016), decreases soil microbial diversity (Mikryukov and Dulya, 2017), modifies the structure of soil fungal communities (Mikryukov et al., 2020), and reduces the abundance of soil macroinvertebrates (Vorobeichik, 1998; Vorobeichik et al., 2019). Notably, earthworms, a soil decomposer key taxon, entirely disappeared in highly contaminated sites; the earthworm-free area is about 65 km^2 (Vorobeichik et al., 2019; Vorobeichik et al., 2020). The abundance of other soil detritivores, particularly enchytraeids, millipedes, mollusks (Vorobeichik et al., 2019), and collembolans (Kuznetsova, 2009), has been sharply reduced. A decrease in soil animal feeding activity was also registered with the bait-lamina test (Vorobeichik and Bergman, 2020; Vorobeichik and Bergman, 2021).

In background areas, *Oxalis acetosella*, *Dryopteris* spp., *Calamagrostis arundinacea*, *Aegopodium podagraria*, *Ajuga reptans*, *Circaea alpina*, *Cerastium pauciflorum,* and typical boreal small herbs dominated ground-level vegetation (Vorobeichik et al., 2014). In moderately contaminated sites (the buffer zone), only approximately 70% of the forest floor was covered with herbaceous plants with dominant *Cirsium heterophyllum*, *Athyrium filix-femina*, *C. arundinacea*, *Linnaea borealis*, and *Dryopteris carthusiana* (Mikryukov and Dulya, 2017). In highly contaminated sites (the impact zone), the whole area was bare, and relictual ground-level vegetation consisted of only *Agrostis capillaris*, *Deschampsia caespitosa, Brachypodium pinnatum*, *Equisetum sylvaticum*, *Lathyrus vernus*, *Sanguisorba officinalis*, *Vaccinium myrtillus*, and *V. vitis-idaea* (Vorobeichik et al., 2014).

Despite the fact that the emissions have almost ceased since 2010, there has been no reduction in soil metal concentrations in highly contaminated sites (Vorobeichik and Kaigorodova, 2017) or revegetation (Vorobeichik et al., 2014). However, in moderately polluted sites, due to the mitigation of excess soil acidity (Vorobeichik and Kaigorodova, 2017), soil macroinvertebrates began to recover within the last few years (Vorobeichik et al., 2019; Vorobeichik et al., 2020).

2.5. Study sites and sampling design

We located study sites west of the smelter in background (30–16 km from the smelter), buffer (7–4 km), and impact (3–1 km) zones (Fig. 1). These zones reflect successive stages of pollution-driven forest degradation, and we distinguished them by topsoil metal concentrations and vegetation state. We placed four study sites in each zone, 12 sites in total. The distance between the sites was 10–20 km in the background zone and 0.5–3 km in impact and buffer zones. Within a study site, we placed 25 × 25 m sampling plots, or circular plots with a radius of about 10 m. We used two sampling designs within each site: a random design with 150–500 m distance between sampling plots or a regular one using a grid with 100 m steps. A total of 231 sampling plots were established: 70, 80, and 81 in background, buffer, and impact zones, respectively.

Study sites were located on gentle slopes of ridges in forests with a different stand composition (spruce-fir, pine, birch, and mixed forests). Loam and heavy loam soddy-podzolic soils (Albic Retisols) and podzolized burozems (Leptic Retisols) prevailed. Near the smelter, soddy-podzolic soils are transformed into soddy-podzolic gleyish soils (Stagnic Retisols (Toxic)).

2.6. Humus form assay

On each sampling plot, we used five circular mini-plots with a radius of about 1.5 m, the distance between the centers of the circles being 8–10 m. We placed the mini-plots at random, excluding nearby trunk areas with a radius of 1–2 m around large trees (more than 30 cm in diameter), windblown complexes, and any visible pedoturbations, such as wild boar diggings, mole tunnels, eroded soil, or soil displaced or compacted by man.

In the center of a mini-plot, we dug a pit to 20–25 cm depth and performed the topsoil profile's morphological survey. In different directions from the small pit at a distance of 0.5–1 m, we performed two additional topsoil descriptions down to the upper boundary of the A horizon (from 2 to 5 cm in the background zone to 10–15 cm in buffer and impact zones). Thus, on each mini-plot, the description of the humus form was based on three observations which represented one humipedon. In total, we performed humus assays on 1,155 mini-plots in 2019.

We identified horizons, morphologically described, and diagnosed humus form following the Classification (Zanella et al., 2018g; Zanella et al., 2018h; Zanella et al., 2018j). The thickness of organic (OL, OF, OH) and organic-mineral (A (g)) horizons was measured with an accuracy of 3–5 mm. For organic horizons, we estimated the density, the ratio of indistinguishable organic matter to distinguishable plant residues, their botanical composition, color, fragmentation degree, and arrangement relative to each other. We also registered organic matter (humic component) color and structure (granular, powdery, or microfibrous; presence, size, and shape of animal droppings). For organic-mineral horizons, we analyzed color, structure, organic matter features, texture, redox characteristics, and other morphological features. We also examined the shape and thickness of the transition area between lower organic and upper organic-mineral horizons. If the diagnosis of humus form was unfeasible in the field, morphology was studied at mesoscale under a reflected light microscope (x10) in a laboratory.

For the non-zoogenic A horizon, we considered more types of structure than the Classification suggests. In particular, clody, or blocky angular with an irregular shape and size of aggregates, or weak blocky subangular, or powdery structure was regarded as non-zoogenic, that is, formed due to the degradation of the pre-existing blocky subangular or granular structure.

3. Results

3.1. Diversity of humus forms

Of the high diversity of observed humus forms, nine forms morphologically fully corresponding to the Terrestrial humus systems (from Mesomull to Eumor) were referred to as **typical**. In addition, we have described 21 topsoil variants, for which no correspondence can be found in the Classification (Table S1, Fig. 2, and Fig. 3). Such forms we referred to as **non-typical**. Of them, 14 forms, at least fitted to one known humus system, were referred to as **with-analog**. The remaining seven non-typical forms corresponding to none of the known humus systems were termed **without-analog**.

Of the typical forms, all Mull forms (Mesomull, Oligomull, and Dysmull) and two Moder forms close to the Mull system (Hemimoder and Eumoder) were referred to as **natural**, since they occurred in the background area. The remaining typical form of the Moder system (Dysmoder), all typical forms of the Mor system (Hemimor, Humimor, and Eumor), and all non-typical forms we considered more or less induced by pollution were referred to as **pollution-induced**.

The proportion of typical and non-typical forms differed between pollution zones (Table 1). In the background area, only typical forms were revealed. In the buffer zone, non-typical forms predominated (79% of all topsoil profiles), while in the impact zone, typical and non-typical forms were equally represented (47% and 53%, respectively).

3.2. Nomenclature of humus forms in contaminated areas

We have not changed the names for natural forms. The names for pollution-induced forms are formed as follows: base name + mandatory

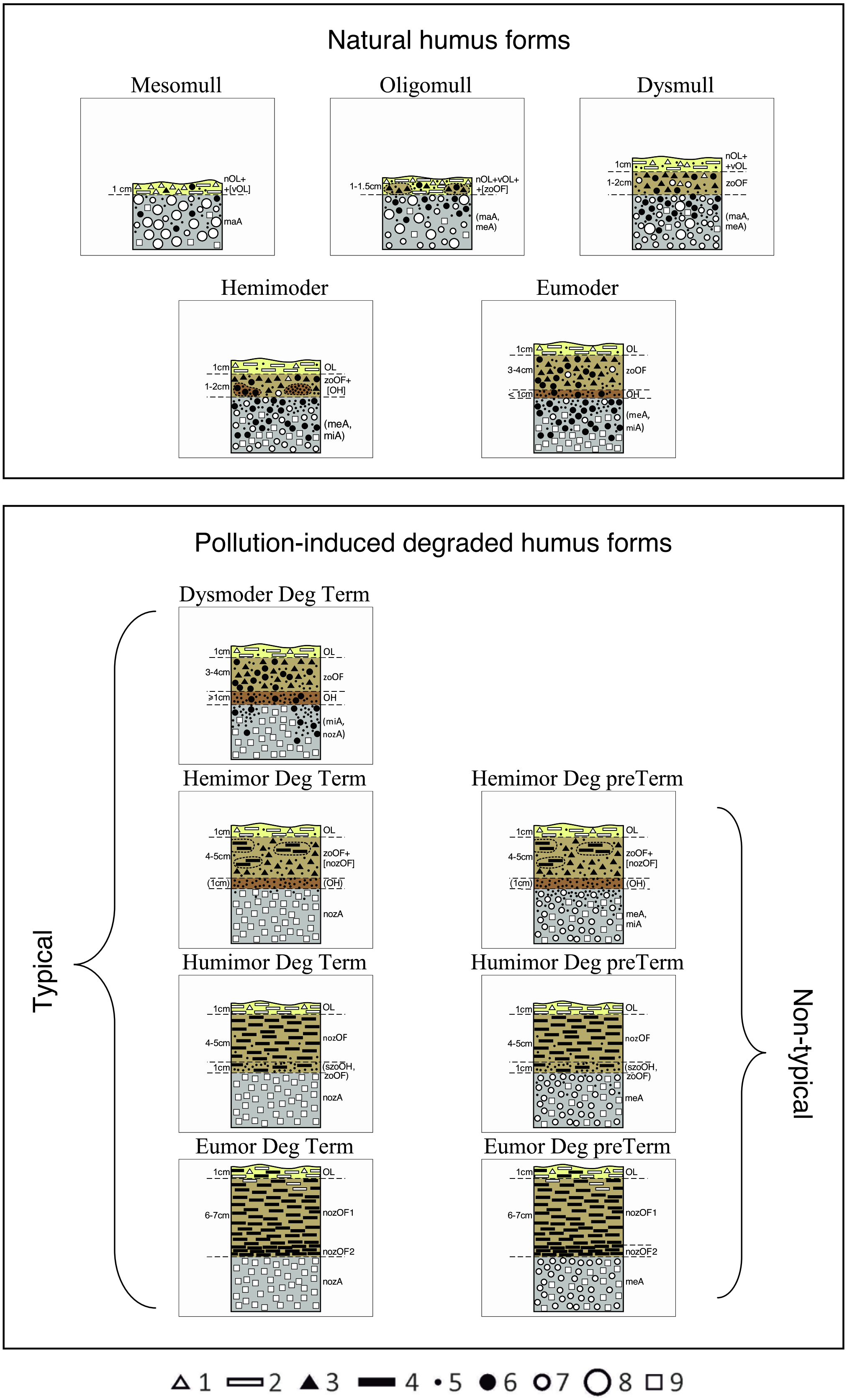

Fig. 2. Schemes of natural and degraded humus profiles. Degraded profiles are arranged in decreasing order of zoogenic activity. Here and in Fig. 3, the size of organic horizons is proportional to their real thickness, and symbols denote (1) intact litter; (2) slightly altered litter; (3) fragmented litter; (4) mycogenically transformed litter; (5) microdroppings; earthworms' (6) organic mesodroppings, (7) organic-mineral mesodroppings, and (8) organic-mineral macrodroppings; (9) mineral particles and soluble organic matter. Yellow, light brown, dark brown, and grey denote OL, OF, OH, and A horizons.

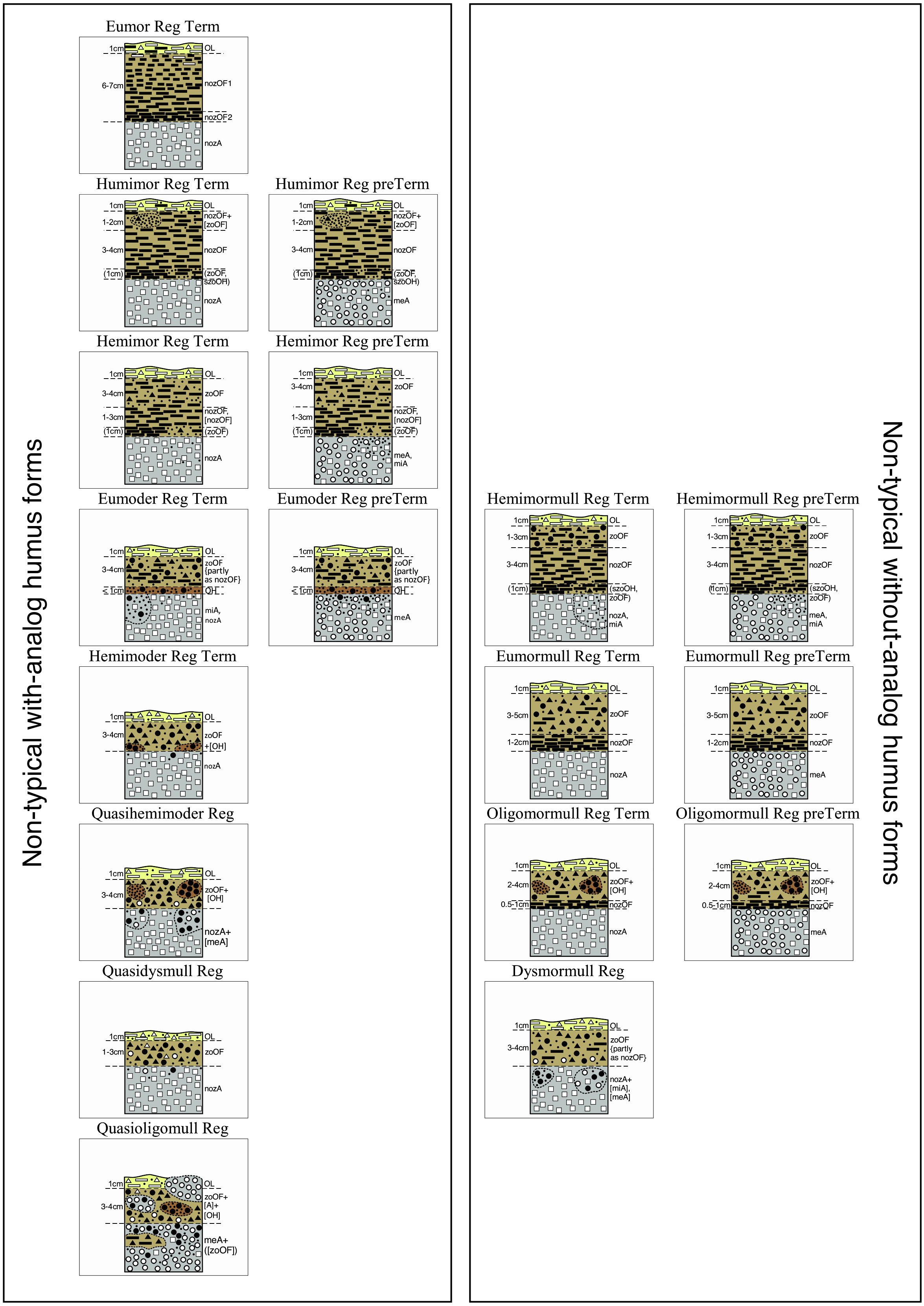

Fig. 3. Schemes of terminal (left) and preterminal (right) regraded humus profiles. Profiles are arranged in increasing order of zoogenic activity.

qualifier + facultative qualifier. The base name of with-analog forms was the name (unchanged or with the prefix Quasi) of a Terrestrial humus form which the diagnostic criteria they met. Importantly, we prioritized the similarity of organic rather than organic-mineral horizons. Base names for without-analog forms ("Mormull") originated from the combination of Mor and Mull humus systems; also, we added several standard prefixes (Hemi, Eu, Oligo, Dys), following the general principles of forming names in the Classification.

As we discuss further (see *3.3* and *3.4*), two opposite processes unfold in the study area: the degradation (decrease in zoogenic activity) and regradation (recovery of zoogenic activity in previously degraded soils). Accordingly, with the mandatory qualifiers "Deg" (from "degraded") and "Reg" (from "regraded"), we indicated the profiles shaped by the first and second processes, respectively.

We also paid attention to the degree of degradation finality and considered this process terminated if organic and organic-mineral horizons reached the same degree of degradation, reflected by a facultative qualifier "Term" (from "terminated"). Incomplete degradation, that is, the delay in organic-mineral horizon degradation compared to organic horizons, was specified with "preTerm" (from "pre-terminated"). We denoted the "starting point" of the recovery in regraded forms, since it can start after either a completed or incomplete degradation. At advanced stages of recovery, it is impossible to unambiguously diagnose the cause of organic-mineral horizons zoogenicity, which could be inherited from a form with an incomplete degradation or formed *de novo*. In this case, we added no facultative qualifier to the name.

3.3. *The typology of humus forms in contaminated areas*

We propose a two-level typology of humus forms in the contaminated areas (Table 2). At the higher hierarchical level, we used the criterion of presence/absence of discrepancy with the Classification (i.e., presence/absence of signs that distinguish non-typical forms from typical ones). At the lower hierarchical level, we identified the "localization" of the discrepancy, defining two groups of forms: 1) with the discrepancy between the O and A horizons, and 2) with the discrepancy within the O horizons.

3.3.1. *Discrepancy between O and A horizons*

In this group, there are two types: 1) degradation of organic-mineral horizons delayed compared to organic ones, and 2) recovery of organic-mineral horizons delayed compared to organic ones.

In the first type, the structure of organic horizons corresponds to Mor forms, but the A horizon retains a blocky subangular or granular structure, which is atypical for this system. Although there are no signs of currently active macroinvertebrates (no living earthworms or their burrows) and organic coprolites are destroyed, organic-mineral aggregates are similar in size and shape to the aggregates from the meA horizon of the background zone. We associated this type with the Hemimor Deg preTerm, Humimor Deg preTerm, and Eumor Deg preTerm. Most of these forms were found in the impact zone and less in the buffer zone, accounting for 13.3% and 4.3% of all humus profiles, respectively (Table 1).

We associated the second type with the Hemimoder Reg Term and Quasidysmull Reg forms: organic horizons are similar to typical Hemimoder and Dysmull forms, respectively, but the A horizon remains non-zoogenic with massive, powdery, clody, or weak blocky subangular structure. Both the Hemimoder Reg Term and Quasidysmull Reg were observed in 6.5% and 1.3% plots of the buffer zone, respectively, while the Hemimoder Reg Term was encountered only once in the impact zone. The same discrepancy is possible (but not always manifested) in the Quasihemimoder Reg and Dysmormull Reg: the A horizon's zoogenic structure begins to form only in some parts of the topsoil profile.

Table 2
Typology of the humus forms in contaminated areas.

<table>
<tr><td colspan="2" rowspan="3"></td><td colspan="3">The discrepancy within O horizons</td></tr>
<tr><td rowspan="2">Discrepancy is absent</td><td colspan="2">Discrepancy is present</td></tr>
<tr><td>Inversion of layers or horizons</td><td>Heterogeneity within horizons</td></tr>
<tr><td rowspan="3">The discrepancy between O and A horizons</td><td>Discrepancy is absent</td><td>**Eumull** – nOL/maA
Mesomull – nOL+[vOL]/maA
Oligomull – nOL+vOL/[zoOF]/(meA, maA)
Dysmull – nOL/vOL/zoOF/(meA, maA)
Hemimoder – OL/zoOF/[OH]/(meA, miA)
Eumoder – OL/zoOF/OH{<10 mm}/(miA, meA)
Dysmoder Deg Term – OL/zoOF/OH{≥10 mm}/(miA, nozA)
Hemimor Deg Term – OL/[nozOF]/zoOF/(OH)/nozA
Humimor Deg Term – OL/nozOF/(szoOH, zoOF)/nozA
Eumor Deg Term – OL/nozOF1/nozOF2/nozA</td><td>Eumor Reg Term –
OL/nozOF1{many mycelia}/nozOF2/nozA
Humimor Reg Term –
OL/nozOF+[zoOF{micro}]/nozOF/(szoOH, zoOF){may be absent}/nozA
Hemimor Reg Term –
OL/zoOF{micro}/(nozOF, [nozOF])/(zoOF){may be absent}/nozA</td><td>Eumoder Reg Term –
OL/zoOF{partly as nozOF}/OH{<10 mm}/(miA, nozA)</td></tr>
<tr><td>Discrepancy is present – Delay in A horizon degradation</td><td>*Dysmoder Deg preTerm* –
OL/zoOF/OH{≥10 mm}/(meA, miA)
Hemimor Deg preTerm –
OL/[nozOF]/zoOF/(OH)/(meA, miA)
Humimor Deg preTerm –
OL/nozOF/(zoOF, szoOH)/(meA, miA)
Eumor Deg preTerm –
OL/nozOF1/nozOF2/(meA, miA)</td><td>*Eumor Reg preTerm* –
OL/nozOF1{many mycelium}/nozOF2/(meA, miA)
Humimor Reg preTerm –
OL/nozOF+[zoOF{micro}]/nozOF/(szoOH, zoOF){may be absent}/(meA)
Hemimor Reg preTerm –
OL/zoOF{micro}/(nozOF, [nozOF])/(zoOF)){may be absent}/(meA, miA)
Hemimormull Reg preTerm –
OL/zoOF/nozOF/(zoOF, szoOH){may be absent}/(meA)
Eumormull Reg preTerm – OL/zoOF/nozOF/meA
Oligomormull Reg preTerm – OL/zoOF+[OH{pockets}]/nozOF/meA</td><td>Eumoder Reg preTerm –
OL/zoOF{partly as nozOF}/OH{<10 mm}/(meA)</td></tr>
<tr><td>Discrepancy is present – Delay in A horizon regradation</td><td></td><td>Hemimormull Reg Term –
OL/zoOF/nozOF/(zoOF, szoOH){may be absent}/(nozA, miA)
Eumormull Reg Term –
OL/zoOF/nozOF/nozA
Oligomormull Reg Term –
OL/zoOF+[OH{pockets}]/nozOF/nozA</td><td>Hemimoder Reg Term – OL/zoOF/[OH]/nozA
Quasihemimoder Reg –
OL/zoOF+[OH{pockets}]/nozA+[meA]
Quasidysmull Reg – nOL/vOL/zoOF/nozA
Quasioligomull Reg –
OL/([A])/zoOF+([A]+[OH])/meA+([zoOF])
Dysmormull Reg –
OL/zoOF{partly as nozOF}/nozA+([meA], [miA])</td></tr>
</table>

Natural humus forms are in bold; hypothetical forms are in italics. Humus forms are arranged within a cell as follows: for degraded forms, zoogenic activity decreases top-down; for regraded forms, zoogenic activity increases top-down. In the formulas for non-typical forms, main differences from typical ones are underlined. Slash separates horizons. The sign "+" denotes a combination of several sub-horizons in one horizon. In parentheses, options for horizon structure are separated by commas; if a single horizon is indicated in parentheses, this means that it is optional. A discontinuous (fragmentary) horizon is in brackets. Explanations (e.g., horizon thickness or peculiarities) are in braces.

3.3.2. Discrepancies within O horizons

This group presents a greater variety of forms, classified into two types: 1) forms with an inverted sequence of layers, and 2) forms with non-zoogenic fingerprints in zoogenic O horizons.

3.3.2.1. The inversion of layers. Usually, in Mor forms, layers with signs of mycogenic and zoogenic activity are located in the lower part of the litter. In "inverted" humus profiles, layers are arranged in another way.

In the Eumor Reg Term (twice accounted in impact and buffer zones), organic horizons are non-zoogenic. However, the degree of mycogenic decomposition of plant litter is higher in the upper rather than in the lower part of the nozOF. Specifically, many mycelia are visible to the naked eye, plant remains are more fragmented and softer, and the content of fine fibers (as dust) is higher in the upper layer than in the lower one.

Similar inversions are observed when a zoogenic OF horizon is formed above a non-zoogenic OF rather than below it. Such cases can be divided into two subtypes. The first subtype presents profiles with signs of zoogenicity associated with the activity of microarthropods and small macroinvertebrates (enchytraeids, millipedes, Nematoceran larvae, some other taxa, and microdroppings of<1 mm) except earthworms. The second subtype possesses signs of earthworm activity in profiles such as live earthworms and mesodroppings of 1–3 mm.

We ascribed the Humimor Reg Term (9.9% and 2% in impact and buffer zones, respectively), Hemimor Reg Term form (3.2% and 2.5%), Humimor Reg preTerm (5.7% and 0.5%), and Hemimor Reg preTerm (about 3% in each zone) to the first subtype (Table 1). In the Humimor Reg Term, microdroppings are present as clusters or evenly distributed between beds of needles and leaves in the upper part of the nozOF, while the thick nozOF is preserved below. Therefore, the sequence of horizons zoOF/nozOF is reversed compared to typical Humimor. In the Hemimor Reg Term, the layer of non-zoogenically transformed residues is not as thick or discontinuous. In the Humimor Reg preTerm and Hemimor Reg preTerm, regradation occurs in soils where the degradation was not finalized, and signs of recovery (reversion zoOF/nozOF) combine with incomplete degradation of the A horizon.

We described the second inversion subtype as Mormull forms. They have no analogs among natural forms since the nozOF is combined with a highly zoogenic OF, but the OH horizon is absent. In the Hemimormull Reg Term, the zoOF is thin (1–3 cm, and zoOF $\leq$ nozOF in thickness), and retains beds of leaves and needles, but organic micro- and mesodroppings are present. In the Eumormull Reg Term, the zoOF is thicker; in its upper part, plant residues are highly fragmented, loose, and well mixed with organic coprolites, while layered residues of large leaves are preserved only fragmentally. In the Oligomormull Reg Term, coprolites accumulate in such quantities that they form clusters resembling fragments of OH in the zoOF. In all three forms, nozOF is preserved under zoOF, and its thickness decreases along the series Hemimormull—Eumormull—Oligomormull. The nozA, or thin 0.5–1 cm szoOH, or zoOF, formed by microarthropods and small macroinvertebrates except earthworms, can be located under the non-zoogenic OF. The Hemimormull Reg Term and Eumormull Reg Term are the most common forms in the buffer zone (10.8% and 15.0%, respectively), while the Oligomormull is rare (1.5%). In the impact zone, the frequency of the Hemimormull Reg Term is higher than the Eumormull Reg Term (9.6% and 1.7%, respectively), while the Oligomormull is absent (Table 1).

3.3.2.2. Heterogeneity within O horizons. In contrast to typical Eumoder, the formed *de novo* zoOF of the Eumoder Reg Term and Eumoder Reg preTerm bears nozOF imprints. Namely, needles and leaves lying in well-manifested layers, with micro- and mesodroppings between them, are accompanied by fragments of non-zoogenically transformed plant residues. The A horizon retains its non-zoogenic structure (massive, clody, or powdery), or converts to miA (Eumoder Reg Term), or inherits the meA structure from incompletely degraded forms (Eumoder Reg preTerm). These forms were found only in the buffer zone (3.8%).

In the Quasihemimoder Reg (2% in the buffer zone and met once in the impact zone), discontinuous OH fragments are present as clusters of organic coprolites in the whole zoOF, except for its lower part. The A horizon retains a non-zoogenic structure, or fragments of blocky subangular structure appear.

In the Quasioligomull Reg (found only in the buffer zone, 4%), zoOF is heterogeneous and thicker than in typical Dysmull. The volume of clusters of organic-mineral coprolites on its surface and within a layer exceeds the number of plant residues, and zoOF can be intermittent. Preserved OF fragments (layered leaves), buried 5–7 cm under a layer of organic-mineral coprolites, constituting *de novo* meA, signifies that this profile is replacing a formerly degraded humus form.

The Dysmormull Reg (3.7% and 12.3% in the impact and buffer zones, respectively) inherits certain features from the Mor forms, especially weakly fragmented layers are preserved in some loci in OF, and the A horizon is non-zoogenic.

3.4. Evolution diagram of humus forms

Gradual transitions between forms, which coincide well with gradients of increasing/decreasing pollution, allowed us to interpret the diversity of humus forms from the perspective of their genesis and draw up an evolutionary diagram (Fig. 4). All changes occur in two opposite directions: 1) degradation of topsoil structure under the pollution impact due to macrofauna suppression leads to Mull-to-Mor transformation, while 2) Mor-to-Mull transformation results from the recovery of topsoil structure after cessation of pollutant inputs and decrease in soil toxicity, which initiated the macrofauna revival. Based on the logic of transitions between humus forms, two hypothetical forms are shown that were not found, but probably exist.

3.4.1. Humus form degradation

In the background zone, Mull humus forms (mainly Dysmull) dominate (73.4%). Moder humus forms are less common, with Hemimoder (closest in morphology and zoogenic activity to Dysmull) prevailing among them (16.0%), and Eumoder (8.9%) found more in wet sites (Table 1 and Fig. 2). In the buffer zone, Mull humus forms are less frequent (15.8% excluding regraded forms), and Moder humus forms prevail (45%). Mor humus forms (40%) occur when approaching the boundary between buffer and impact zones (5–4 km from the smelter). In the impact zone, Mor humus forms (mainly Eumor) dominate (96% excluding regraded forms), while Mull and Moder humus forms are rare (4% in total). Note that when calculating the prevalence of humus forms, we did not separate Eumor and Hydro Eumor because these humus forms are similar to each other in the Terrestrial humus system and Hydro Intergrades.

Thus, the Mull humus forms are replaced by the degraded Moder and Mor humus forms along the pollution gradient. In most cases, degraded humus forms do not differ from the typical variants described in the Classification. During Mull-to-Mor transformation, topsoil morphology changes as follows. The OH horizon joins the OL and zoOF horizons, then a discrete layer of litter without signs of zoogenic transformation (nozOF) appears (in the Hemimor) and later becomes continuous. The thickness of zoogenic zoOF and OH decreases and then disappears by forming an extreme form, as Eumor.

Changes also occur in the organic-mineral horizon: the primary zoogenic structure (granular and subangular blocky) of the A horizon degrades and transforms into a non-zoogenic structure (powdery, clody, or massive). If the degradation of the A horizon for some reason lags behind that of organic horizons, then non-typical Mor humus forms with incomplete A horizon degradation are formed.

3.4.2. Humus form regradation

The Mor-to-Mull transformation is associated with the conversion of

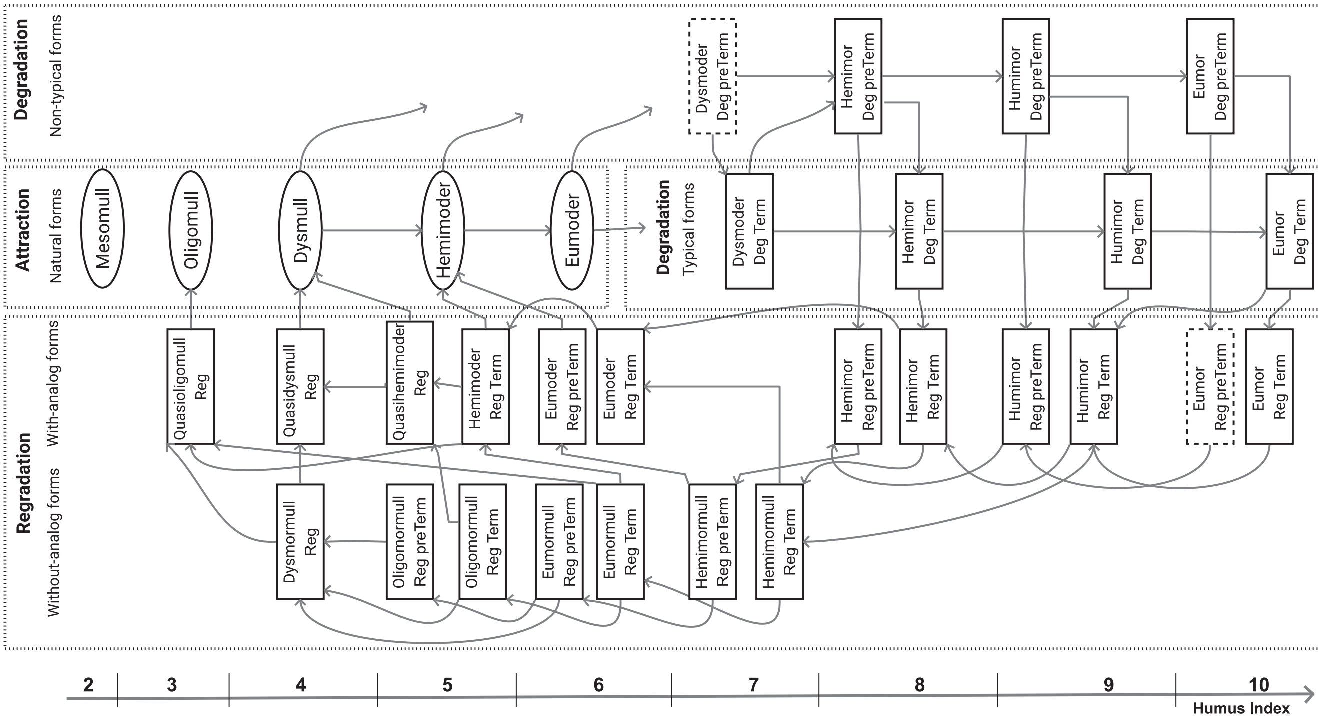

Fig. 4. Evolution diagram of humus forms in contaminated areas. For humus form names and descriptions, consult Table S1, Table 2, Fig. 2, and Fig. 3. Natural humus forms (as attractors) are shown in ellipses, pollution-induced forms are in rectangles, hypothetical forms that have not been found are denoted with a dashed line. Arrows designate putative transitions between humus forms. Zoogenic activity decreases with Humus Index increase.

non-zoogenic O horizons into zoogenic ones and recovery of A horizon structure. Non-zoogenically transformed litter fragments or intact plant residues lie as well-manifested layers. This sign is preserved in regraded profiles, which indicates the formation of zoogenic horizons in place of pre-existing non-zoogenic ones, not caused by the processing of fresh litter.

Based on the morphology of non-typical humus forms (see *3.3*), we assume that the Mor-to-Mull transformation can follow two different paths (Fig. 3 and Fig. 4). The first path is through Moder intermediate stages, and for all these stages there are analogs among the typical humus forms. The second path is without passing Moder intermediate stages through humus forms for which there are no analogs among typical ones (Mormull). In both paths, the conversion of nozOF to zoOF starts from the upper litter layer and spreads downward in the profile. A preliminary stage is possible, not associated with the zoogenic transformation of plant residues but rather with the activation of microbial decomposition in the upper layers (Eumor Reg Term).

In the first path, in the early stages of recovery, an increase in the activity of microarthropods and small macroinvertebrates except earthworms (enchytraeids, Nematoceran larvae, millipedes) in the upper part of the nozOF, in Eumor Deg Term or Humimor Deg Term, leads to the formation of Humimor Reg Term. Then, more active colonization of nozOF by these invertebrates produces Hemimor Reg Term. Similar processes occur during the recovery of those degraded forms in which degradation of the A horizon has not been completed (Eumor Deg preTerm transforms into Humimor Reg preTerm and then into Hemimor Reg preTerm). In the series Eumor Reg—Humimor Reg—Hemimor Reg (for both terminal and preterminal humus forms), the intensity and thickness of nozOF decreases, while the manifestation of zoogenic features increases.

Further transformation of Hemimor Reg Term and Hemimor Reg preTerm may be associated with a deeper processing of plant residues in the former non-zoogenic OF by small macroinvertebrates and colonization of the upper litter layers by epigeic earthworms. As a result, nozOF disappears, and an OH horizon appears in Eumoder Reg Term. Under favorable conditions for the soil animals (including both the improvement of local conditions and the presence of faunal refuges in the vicinity), signs of non-zoogenic OF disappear gradually, but the zoOF horizon remains thicker than in typical Moder humus forms (4–5 cm, up to 7–9 cm). The non-zoogenic structure is preserved in the A horizon, or fragments of granular structure (meA) appear. We observed two kinds of these humus forms: Hemimoder Reg Term with a delay in forming the zoogenic structure of the A horizon and the Quasihemimoder Reg with discontinuous OH fragments within zoOF. These humus forms can further evolve to the Moder humus system (i.e., into a typical Hemimoder) or the Mull humus system.

The activization of macroinvertebrates, except endogeic earthworms, leads to a further movement towards the Mull humus system: the thickness of OF decreases, and its structure becomes similar to the OF of a typical Dysmull, however, the recovery of the zoogenic structure of the A horizon is still lagging behind organic horizons (Quasidysmull Reg). The recolonization of endogeic earthworms noticeably changes the topsoil morphology: organic-mineral coprolites generate a zoogenic structure within the A horizon (meA). These changes are seen in Quasioligomull Reg and during the transition from Quasidysmull Reg to a typical Dysmull.

In the second path of regradation, the upper layers of non-zoogenic horizons are colonized by earthworms, without or with very weak initial processing of plant residues by other invertebrates. The result is the formation of humus forms for which there are no analogs among natural ones (Mormull). They arrange the Mor-to-Mull transformation series without going through intermediate Moder stages (Hemimormull—Eumormull—Oligomormull—Dysmormull). The thickness of nozOF decreases until it completely disappears in the Quasidysmull, in which only residual non-zoogenic fingerprints remain in OF. In addition to such Mor-to-Mull transformations, each form can pass into other non-typical with-analog humus forms.

3.5. Incorporation of non-typical forms in the Humus Index

Ponge et al. (2002) proposed the Humus Index (HI) to convert qualitative information on topsoil morphology into a quantitative form. The HI is the ordinal number of a humus form in increasing order of macrodetritivore participation to organic matter destruction. HI was initially proposed for Terrestrial humus systems but was expanded later to include Hydro and Para humus forms (Korkina and Vorobeichik, 2018). For this, Hydro and Para humus forms were ranked in a similar series, and then were correlated with the series for Terrestrial humus systems, so that humus forms close in zoogenic activity, but belonging to different series, had the same HI.

We performed this procedure for non-typical humus forms (Table 1 and Fig. 4) using the same number of points as for the HI of Terrestrial humus systems. The HI increases from 1 (Eumull) to 10 (Eumor); the higher the HI value, the lower the plant litter decomposition rate, and the lesser macrodetritivores participate in plant litter destruction.

We constructed a series of homologies based on the following rules. Incompletely degraded humus forms and regraded humus forms with a delay in forming the zoogenic structure of the A horizon, were assigned the HI of those typical forms with which they were similar in O horizon structure. For regraded humus forms associated with microarthropod and small macroinvertebrate activity, the HIs of Mor humus forms, except Eumor, were assigned (i.e., 9 and 8). For regraded humus forms associated with earthworm activity, the HIs of Moder humus forms were assigned (i.e., 7–5). For regraded humus forms close to typical Dysmull and Oligomull, we assigned the HIs of Mull humus forms (i.e., 4 and 3).

4. Discussion

4.1. Non-typical humus form as non-equilibrium soil state

The essence of the described non-typical humus forms is that signs of different stages of topsoil evolution are presented in the same profile. Like the mythical Chimeras, non-typical humus forms assemble mechanically “unit” parts of different profiles. The main reason for their formation is the unequal response rate of different layers to environmental changes: one layer reacts slower than another, which at some point in time, preceding a “finale”, looks like a response delay. In other words, non-typical humus forms exhibit continuous soil transformation triggered by abrupt environmental changes.

It is fruitful to use an attractor concept that has come into soil science from theoretical physics, to understand pedogenic processes taking place in contaminated areas. An attractor is the domain of the equilibrium state of the system. A particular humus system is an ecosystem strategy (Ponge, 2003) and an attractor determined by local climate, geology, orography, and vegetation (Zanella et al., 2018a; Zanella et al., 2018j). Notably, a humus system is “an attractive hole” in the “ecological landscape” from which it is “climbing” under the impact, and to which it is “falling” after termination of the impact, “like a golf ball in a green” (Zanella et al., 2018j, p. 97).

The Mull system should be considered an attractor for the studied area, since it dominates in undisturbed forests; here, relatively good water and heat supply determine the high feeding activity of earthworms and the rapid utilization of plant litter. Before starting the smelter operation, the same highly zoogenically active humus forms occurred in its vicinity. After long-term pollution, these forms can be found only at a considerable distance from the smelter, that is, in the background area. In other words, all humus forms in the contaminated sites are deviations from the Mull humus system: such Mor humus forms originated from Mull and will become Mull when the ecosystem recovery initiated by the pollution cessation will be complete. As we discuss further (see *4.3*), besides soil toxicity, the absence of sources of immigration for earthworms and other macroinvertebrates can limit ecosystem recovery.

Most active Moder humus forms (Hemimoder and Eumoder) can also be considered attractors for the studied area, at least in some habitats that are less favorable for earthworms, where enchytraeids dominate instead. Interactions between earthworms and enchytraeids are complex and remain poorly understood; often, enchytraeids are suppressed in the presence of earthworms (e.g., Räty and Huhta, 2003). The inverse hyperbolic dependence between earthworms and enchytraeids abundance gives reason to believe Mull and Moder systems as topsoil alternative stable states (Ponge, 2003). “The biological forces that drive to the production of the Mull and Moder humus forms are antagonistic. However, that does not mean that they are exclusive to each other. Rather, a spectrum exists, and this is mostly the rule” (Bernier, 2018, p. 310). Considering this rule, most likely, within the Mull-to-Mor series, the boundary between natural and pollution-induced humus is region-specific *ceteris paribus*. In the studied area, Dysmoder is the first pollution-induced form (Fig. 4); we suppose that in more northern regions, Dysmoder may be the last natural form, and in more southern areas, Eumoder may be the first pollution-induced form.

An attractor is an equilibrium system state conforming to external conditions. The described non-typical humus forms are a non-equilibrium system state, since the topsoil is out of balance. A non-equilibrium state is unstable since the system “tends to leave” it until it reaches a new equilibrium with external conditions. With such instability in mind, we can conclude that non-typical humus forms in the Mor-to-Mull transformation are “ephemeral” at the forest successional scale, and during further recovery, they will quickly turn into typical Mull forms. We predict that the lifespan of such non-typical humus forms is only within the first tens of years.

The lifespan of degraded humus forms is much longer and, most likely, comparable to the duration of the smelter’s operation (in the study area, at least 80 years). The soil can retain metals very firmly, even for many decades if not centuries after pollution cessation (Tyler, 1978). Therefore, degraded humus forms can be considered soil alternative stable states (Beisner et al., 2003).

Most surface soils are polygenetic due to temporal variations of environments, so the polygenetic profiles exhibiting the combination of several specific pedogenic processes are widespread (Targulian and Krasilnikov, 2007). In contrast to such profiles, non-typical humus forms indicate just one process—the decrease or increase of zoogenic activity—proceeding in different layers at unequal rates.

4.2. Putative mechanisms of the formation of non-typical humus forms

The studied area is fascinating, as now we are dealing with the results of two oppositely directed processes: soil degradation, i.e., moving far from the attractor, and soil regradation, i.e., going back to the attractor. In its pure form, the first process could be studied before emission reductions, that is, until the mid-2000 s. We can still reconstruct it using space-for-time substitution by describing humus form changes when approaching the smelter. The second process began after the almost complete cessation of emissions in 2010 and became especially noticeable since 2018. We analyzed humus regradation by repeated observations forasmuch as we re-examined previously investigated sites in 2015–2016 (Korkina and Vorobeichik, 2016, 2018). Accordingly, we observe a complex mosaic of different stages of these two processes in space at the present moment.

We built two series of humus forms: degraded and regraded (Fig. 4). Transitions between the humus forms within each series and transitions between series are speculative and show the putative evolutionary paths of humus forms. These transitions were reconstructed based on ideas about the delay in response to environmental changes of some horizons or layers compared to others.

4.2.1. Degraded non-typical humus forms

With soil degradation, the delay in the response of organic-mineral horizons, i.e., the retention of residual signs of primarily zoogenic structure in the A horizon (preterminal Eumor, Humimor, and

Hemimor), is associated with the resistance of soil aggregates to the destructive effects of sulfur acid and polymetallic dust from the smelter emissions. We do not know why this resistance occurs—only speculations about this are possible.

On the one hand, locally increased iron, aluminum, and calcium concentrations, due to the parent rock heterogeneity or organic matter or clay accumulation in micro-depressions, can account for aggregate resistance. These soil constituents are among the main factors of soil aggregate stability (Amézketa, 1999).

On the other hand, it cannot be ruled out that localities with preserved signs of the zoogenic structure are the last “outposts” of earthworms in a highly contaminated area, in which they could exist much longer than in the rest of the area. Accordingly, the destruction of aggregates could begin later and was not completed until performing the humus assay. In turn, the delay of earthworm extinction may be associated with locally favorable microhabitat conditions, such as increased humidity or high deciduous litterfall or less soil toxicity due to the soil buffering capacity to acids or less polymetallic dust deposition. We found high spatial heterogeneity of metal concentrations, soil toxicity (Vorobeichik and Pozolotina, 2003), earthworm abundance (Vorobeichik et al., 2020), and detritivore feeding activity (Vorobeichik and Bergman, 2020) in the smelter vicinity. Such areas, still poorly affected by pollution, could act as sources for the immigration of earthworms and other macroinvertebrates and thus might increase the rate of topsoil regradation if present not too far from the smelter (also see *4.3*).

4.2.2. Regraded non-typical humus forms

With soil regradation, the delay in recovering the organic-mineral horizons is due to the need to form a zoogenic structure *de novo*. However, macroinvertebrate activity is concentrated only in forest litter and has not yet reached the organic-mineral horizon (see below). The characteristic times of the delay in forming the zoogenic structure are much shorter than the erasure of the zoogenicity signs in the A horizon during degradation: it is most likely rated several years, not in decades.

The delay in recovering some layers within O horizons is associated with the macroinvertebrate vertical stratification. Epigeic earthworms dominate the background area; endogeic species are abundant only in deciduous forests (see *2.2*). Endogeic species are more sensitive to pollution than epigeic ones, since they are the first to disappear along the pollution gradient (Vorobeichik, 1998). When approaching the smelter, earthworms redistributed from the upper part of the A horizon into the forest litter; at the limit of their range (i.e., 4–5 km from the smelter), earthworms dwell in litter exclusively (Vorobeichik, 1998). Pollution induces a similar change in the vertical stratification of other soil macroinvertebrates: in the smelter vicinity, most remaining taxa dwell only in forest litter (Vorobeichik et al., 2007). In non-typical Mor humus forms, soil macrodetritivores also “strive upward,” but only within the litter. In contrast to typical Mor humus forms, the most zoogenically active litter layer is the upper one (see *3.3.1*), most likely because the lower layer is still toxic.

We assume that possible reasons for the lower toxicity of the upper layers can be (i) slight contamination and acidity of the newly fallen plant litter, (ii) metal leaching from the upper layers and illuviation into lower ones. The main route of metal entry into leaves is aerial, i.e., associated with adhesion of polymetallic dust, while root intake is much less important (Kozlov et al., 2000). Therefore, with the cessation of emissions, metal concentrations in the leaves and, consequently, in plant litter, should decline sharply. This fact was mentioned for birch leaves (Belskaya, 2018) and herbaceous plants (Nesterkov, 2019). A decrease in the acidity in the upper litter layers can be associated with a lower input of sulfur acid due to the almost complete cessation of SO_2 emissions and organic acids due to the growth of deciduous trees, and not conifers, during reforestation (Vorobeichik and Kaigorodova, 2017).

Metal leaching from the upper litter layers and illuviation into lower ones are likely under the udic soil moisture regime and naturally weak acidic rains in the study area. The realness of this mechanism is indirectly confirmed by comparing the metal concentrations in O and A horizons before and after emission reduction. After pollution cessation, metal concentrations in the litter decreased, while in the A horizon they increased compared to the previous period (Vorobeichik and Kaigorodova, 2017). However, our hypothesis about the lower toxicity of the upper litter layers requires direct verification, and we are going to do it in the future.

The specificity of non-typical Mor humus forms is that macroinvertebrates colonize the litter with an uncommon vertical gradient of favorability. In the natural Mor humus forms, the deeper into the litter, the more favorable the environment for mesofauna (Zanella et al., 2011; Zanella et al., 2018h). Such direction can be associated with a more stable microclimate and higher food availability due to microflora thriving (e.g., Didden and De Fluiter, 1998). In the contaminated Mor humus forms, the gradient of environmental favorability is oppositely directed, i.e., the deeper into the litter, the less favorable environment due to toxicity.

It would be interesting to compare the earthworm colonization of contaminated and uncontaminated areas regarding humus changes. Unfortunately, most of the study of consequences of European earthworm species invasion into North America (e.g., Bohlen et al., 2004; Hale et al., 2005; Eisenhauer et al., 2007) did not include humus form descriptions *per se*. Lejoly et al. (2021) compared humus forms in earthworm-invaded and earthworm-free zones, but they analyzed changes that occurred many years after the invasion, that is, a stable state of soils. In laboratory experiments, earthworms usually colonize the mechanically homogenized humus (e.g., Haimi and Huhta, 1990); hence there are no humus form assays. The earthworm colonization of uncontaminated litter would likely occur throughout its entire thickness, which would not lead to the formation of non-typical Mor humus forms.

4.3. Drivers of topsoil degradation and regradation

It is safe to say that the main driver of soil degradation is long-term polymetallic contamination combined with acidification, which led to high soil toxicity and subsequent macrofauna extinction. Based on the sequence of taxa disappearance along the pollution gradient (Vorobeichik, 1998; Kuznetsova, 2009; Vorobeichik et al., 2012; Vorobeichik et al., 2019), we can conclude that endogeic earthworms are most sensitive to soil contamination, followed by epigeic earthworms, then enchytraeids, and collembolans. Accordingly, in moderately contaminated sites, enchytraeids and collembolans “compensate” the decreased activity of earthworms, leading to the formation of Moder humus forms. Enchytraeids can persist better in severe soil conditions than earthworms (Pelosi et al., 2020) because they are much more tolerant than earthworms to soil acidity (Didden, 1993), salinity (Owojori et al., 2009), and metal contamination (Bart et al., 2017; Amossé et al., 2018). In highly contaminated sites, all decomposer taxa are inhibited, and thus, Mor humus forms prevail.

The main driver of soil recovery is the recolonization of contaminated areas by earthworms. Accordingly, most of the regraded humus forms are associated with the feeding activity of these decomposers. Direct observations confirm the recolonization of the contaminated area. Specifically, a comparison of two consecutive surveys of soil macroinvertebrates revealed that in 2015, earthworms were encountered 2 km from the smelter, whereas earlier (in 1990–2014) they did not occur closer than 4 km (Vorobeichik et al., 2019). The European mole (feeding almost exclusively on earthworms) also moved closer to the smelter during this period (Vorobeichik and Nesterkova, 2015). Earthworms are relatively immobile: the colonization rate by them of a previously unsettled territory is usually 4–6 m year^{-1}, reaching 14–28 m year^{-1} (Eijsackers, 2010, 2011). However, in highly polluted areas, earthworms can survive for a long time in some refuges. We have revealed that decaying tree trunks (Vorobeichik et al., 2020) and riparian zones of small forest rivers (we prepare these results for publication) can be such microhabitats; their existence can explain a relatively rapid earthworm dispersal in the contaminated areas.

Besides earthworms, a driver of the formation of some non-typical humus forms (Humimor Reg and Hemimor Reg) may be an increase in the abundance of other, smaller in size, macrodetritivores. These can be enchytraeids, larvae of some Nematocera and Coleoptera families, millipedes, and mollusks (see *2.2*). The abundance of enchytraeids, millipedes, and mollusks, until recently was sharply reduced near the smelter, but they did not disappear as earthworms (Vorobeichik et al., 2019). Larvae of Elateridae and Nematocera (except Tipulidae and Limoniidae) are the only taxa among macrodetritivores the abundance of which has not been reduced near the smelter (Vorobeichik et al., 2019). Microdroppings in the non-zoogenic OF, often visible as clusters, are most likely produced by bibionid or sciarid larvae, which are usually highly aggregated (Benefer et al., 2010). By analogy with earthworms, we can expect a rapid colonization of contaminated areas by other taxa of macrodetritivores, including large Nematoceran larvae (Tipulidae and Limoniidae), since the flight mobility of imagoes facilitates the colonization.

4.4. Proposals for inclusion of non-typical humus forms in the Classification

Zanella et al. (2018j) noted that “even for trained people, to be puzzled with some humus forms for which the use of customary keys is a complete failure. Are they novel to science? Do they result from a disturbance that affected well-known humus forms? Or do we need to consider them as intermediate or transitional forms which cannot be included in any extant classification? Each of these opportunities must be examined” (p. 101). We have examined these opportunities.

First of all, we had made great efforts to ensure no pedoturbations when performing the humus assays (see *2.6*). Moreover, most non-typical humus forms occurred not singularly, and many of them were frequent, although sampling plots were at a considerable distance from each other; therefore, it is unlikely that they are “a fortuitous assemblage of organic and/or mineral components” (Ibid). Thus, we believe that non-typical humus forms are lawful stages in soil evolution, so we propose incorporating them in the Classification. The detailed description of non-typical forms and their preliminary taxonomy are the first steps towards their inclusion into the Classification.

To the best of our knowledge, no one has previously described such non-typical humus forms in contaminated areas. In all severe impacts investigations, topsoil evolution was described as a shift in typical humus forms presented in the Classification, for example, during the overgrowth of felling areas (Bernier and Ponge, 1994; Trap et al., 2011; Salmon, 2018) or after the invasion of alien trees (Gentili et al., 2019; Ferré and Comolli, 2020). Rapid changes in organogenic horizons were observed during the invasion of European earthworms into North American forests (e.g., Bohlen et al., 2004; Hale et al., 2005; Eisenhauer et al., 2007), but studies did not concern humus forms.

It would be erroneous to apply the term “Intergrades” to non-typical humus forms, although the morphology of Intergrades also combines traits from different humus systems (for example, between Terrestrial and Histic humus systems). In the Classification, Intergrades are understood as intermediate forms for long-existing, i.e., equilibrium, hence stable soil states, but formed at intermediate values of forming factors. Unlike Intergrades, non-typical forms are present only in a non-equilibrium soil state and are therefore unstable.

It would also be incorrect to consider pollution-induced humus forms as stages in Amphi—Tangel but not in the Mull—Moder—Mor series. Although morphological features of some pollution-induced degraded humus forms are close to Amphi humus forms, soil-forming processes differ between Amphi and Moder humus forms. Amphi humus forms are formed under periodically unfavorable conditions (e.g., droughts), which cause vertical migration of earthworms down in soil profiles, but endogeic or anecic earthworms remain the principal decomposers in organic-mineral horizons (Zanella et al., 2018h). Contrary to this, environmental conditions are permanently adverse in highly contaminated areas, and earthworms entirely disappear but do not migrate to deep mineral horizons. Moreover, Amphi humus forms developed generally on calcareous rocks, but almost all soils are formed on siliceous rocks in the study area.

Colombini et al. (2020) described a new humus form in detail, formed on Technosols (a mixture of by-product coke waste, building residues, and soil from abandoned industrial sites). The authors suggested naming this form “Techno-moder” and including it in the Classification. The authors emphasized their work’s novelty, noting the absence of analogs for Techno-moder in natural humus forms and the weak progress in studying humus in technogenically transformed soils. The soils studied by us cannot be classified as Technosols since they do not contain anthropogenic artifacts. In the World Reference Base, Technosols are considered either sealed soils with a layer of dense artificial material in the upper 5 cm or containing artifacts of more than 20% by volume (including those displaced by mining industry rocks and production wastes) or soils with an artificial geomembrane (IUSS Working Group WRB, 2014). Thus, we are dealing with initially natural forest soils, transformed by toxicants, not with Technosols. However, the lacuna in the Classification concerning technogenically transformed soils fully applies to our object of study.

5. Conclusions

In metal-contaminated areas, topsoil evolution is considerably different from natural ones. The increase in soil toxicity causes the extinction of earthworms and other macroinvertebrates, resulting in a Mull-to-Mor transformation. Pollution cessation induces the recolonization of poorly decomposed litter by earthworms, and subsequently, a Mor-to-Mull shift. Such “a voyage there and back again” was imprinted in topsoil morphology: since different topsoil layers react to environmental changes at unequal rates, signs of several stages of humus evolution can be present in the same profile. Besides, the gradient of environmental favorability within contaminated organic horizons in Mor humus forms is in opposite direction in uncontaminated ones. Such peculiarity also leads to the formation of bizarre profiles. We described in detail many non-typical humus forms which are absent in uncontaminated areas.

Methodologically, we based our study on a soil genesis analysis, wherein we followed the traditions of Russian soil science, originating in Vasily Dokuchaev’s ideas. Based on a polygenetic model of pedogenesis, soil properties cannot be explained only by current factors and processes: information about the history of environmental change is necessary to understand soil profile formation (Targulian and Krasilnikov, 2007; Targulian, 2019). The described non-typical humus forms are polygenetic profiles within organogenic horizons. The novelty of our work is determined by the time scale at which we analyzed soil processes. In contrast to paleoenvironmental reconstructions, operating at a centennial and millennial scale, we investigated topsoils dynamics at a decadal and annual scale. The mineral matrix can firmly preserve the imprints of previous stages of soil evolution, while the transformations of organic horizons proceeding over a much shorter time lapse quickly erase signs of the past environment. Such a “short memory” requires a real-time assay to reconstructing topsoil dynamics.

For soil biologists, “the issue is not only a better explanation of the diversity and complexity of underground life, but also the ability to refine the prediction of the humus system’s behavior in the course of land management or natural ecosystem dynamics such as land-use change and climate warming” (Bernier, 2018, p.311). For forestry, soil change studies at a scale of many decades or centuries are essential because such scale corresponds to the lifetime of forest-forming trees. Investigations of humus form shifts were performed at such a scale (e.g., Bernier and Ponge, 1994; Chertov and Nadporozhskaya, 2018). However, the characteristic times of soil resistance and resilience to severe influences, e.g., metal pollution, acid rains, oil spills, or recreation activities, are shorter (years or decades). Therefore, it is vital to analyze humus changes at such a time scale when performing, e.g., environmental impact assessment or an ecosystem restoration success evaluation. The expected shifts in ecosystems due to global climate change can also be rapid, so it is essential to investigate "quick" soil evolution for this reason. From the applied point of view, it is helpful that we have incorporated non-typical forms into the Humus Index, which can be used as a monitoring tool.

Zanella et al. (2018j) discussed the problem of possible intermediate humus forms; they noted that "although the proposed classification allows a wide range of humus forms to be classified…, it remains still possible that some forms were not taken into account, being unknown or rarely encountered" (p. 101). The authors assumed that such forms could be found "in high mountain humus forms belonging to the group Mor–Moder–Tangel–Amphi where much more investigations remain to be done, on various geologic and aspect conditions." Questions about other cases promising the discovery of unknown humus forms were as follows: "What happens … when the parent rock is blocky? What happens when the only visible humus is in crevices as in karstic environments? Can we identify humus forms in suspended soils of tropical rain forests? And what about city environments such as roofs covered with plants, pavements, etc. What about agricultural soils which are disturbed … by tillage, pesticides, and fertilizers?" (Zanella et al., 2018j, p.101). We were not dealing with high mountain or agricultural soils or exotic objects as karstic crevices, suspended soils, or green roofs. However, we revealed many unknown humus forms in boreal forests, that indicate the high specificity of "poisoned pedogenesis" (Targulian and Krasilnikov, 2007) in metal-contaminated areas. The discovery of these non-typical humus profiles confirms the fruitfulness of "the envision of humus forms as dynamic forms needing time to be elaborated but also prone to evolve under changing environmental conditions or disturbances" (Zanella et al., 2018j, p.101).

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Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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