POLLUTION

DOI: https:/doi.org/10.23649/jae.2020.3.15.2

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Received: 11.06.2020; Accepted: 25.06.2020; Published: 11.09.2020

COMPARATIVE EVALUATION OF POLLUTION BY HEAVY METALS OF PLOUGHED AND FALLOW LAND AT VARIOUS DURATION OF AGROPEDOGENESIS

Research article

Abstract

Studying the features of soil heavy metal/metalloid accumulation formed as a result of long-term agropedogenesis is of great importance for predicting safe use of soil resources in agriculture. In such regions with a long history of agricultural development as Crimea, the topsoil can keep in the soil memory the results of slow-acting processes, including any changes in the solid-phase component of the soil system. The aim of this study was to establish regional differences in the concentration of heavy metal using the chronosequences of agrogenic soil transformation method. The objects of study were the humus-accumulative horizon of 20 soils and their parent rock, for which nine of soil heavy metals / metalloids were identified for two hazard groups using X-ray Fluorescence Analysis. The natural and anthropogenic causes of differences in the accumulation of heavy metal in in soils of the Eastern Crimea (Co, Zn, Pb, V, Ni, Cu, As, Ba and Cr) and in soils of the North-Western Crimea (Co, Cr, Ba, Pb and V) have been established. This will optimize a list of priority indicators in regional systems of agro-ecological regulation and selection of territories for growing organic agricultural products.

Keywords: soil, parent rock, soil chronosequence, continually ploughed land, long-term fallow land, hazard degree.

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Получена: 11.06.2020; Доработана: 25.06.2020; Опубликована: 11.09.2020

СРАВНИТЕЛЬНАЯ ОЦЕНКА ЗАГРЯЗНЕНИЯ ТЯЖЕЛЫМИ МЕТАЛЛАМИ ПАХОТНЫХ И ЗАЛЕЖНЫХ ПОЧВ ПРИ РАЗЛИЧНОЙ ДЛИТЕЛЬНОСТИ АГРОПЕДОГЕНЕЗА

Научная статья

Аннотация

Исследование особенностей аккумуляции в почвах тяжелых металлов/металлоидов в результате длительного агропедогенеза имеет большое значение для прогноза безопасного использования почвенных ресурсов в земледелии. Почвенный покров в таких регионах с длительной предысторией аграрного освоения, как Крым, сохраняет в почвенной памяти результаты медленно действующих процессов, включая изменение твердофазного компонента почвенной системы. Цель исследования заключалась в установлении региональных различий в концентрации тяжелых металлов, используя метод хронорядов агрогенной трансформации почв. Объектами изучения стали гумусово-аккумулятивный горизонт 20 почв и их материнских пород, для которых определены девять тяжелых металлов/металлоидов по двум группам опасности, используя метод рентгеноспектрального флуоресцентного анализа. Установлены природно-антропогенные причины различий в аккумуляции тяжелых металлов/металлоидов в почвах Восточного Крыма (Co, Zn, Pb, V, Ni, Cu, As, Ba, Cr) и в почвах Северо-Западного Крыма (Co, Cr, Ba, Pb, V). Это позволит усовершенствовать перечень приоритетных показателей в региональных системах агроэкологического нормирования и выбора территорий для выращивания экологически безопасной сельскохозяйственной продукции.

Ключевые слова: почвы, материнские породы, хроноряды почв, старопахотные почвы, старозалежные почвы, оценка опасности.

1. Introduction

A justified concern about the global problem of soil resources conservation used to encourage many researchers to focus on various signs of soil degradation. Now there are already more than 40 types of degradation of cultivated soils, however, it is believed that nine to be sufficient to evaluate the degradation state during agropedogenesis [1]. This situation is also due to the fact that possible effects of soil progradation can be diagnosed for (enormously) longer periods than the current stage of agricultural activities. A soil system can reach a relative balance state with regular influencing factors if they were similar in strength.

Regions with a long (centuries-old) prehistory of arable farming can provide special information opportunities for examination of agro-pedogenesis results with due account for processes with longer characteristic periods, including changes in agro-soil solid phase.

These regions include Crimea where agriculture made economic basis in the ancient period, in particular, arable farming for grain crops and perennial plantations. Large-scale agricultural development on the lands of three regions of the Crimea in the antiquity has created a unique potential for research the most complete chronosequences of agrogenic soil transformation, including changes in soil system's solid phase that have a longer characteristic time as compared to material composition transformations in the currently developed agricultural lands [2].

The establishment of relationships between the risk of soil heavy metal (HM) accumulation and human activities is one of the urgent tasks of agroecology [3], [4], [5]. It is assumed [6] that all chernozems are characterised by microelement redundancy, at that it is noted accumulation of Pb, Cu, Co and Zn in 0-22 centimetres layer in the century dynamics of chernozems, while during 50 years mobile forms of HM such as Pb, Cu, Co and Zn increased in comparison to the background.

The aim of this work was to establish special aspects of changes in concentration of soil heavy metal/metalloids under the influence of different arable farming periods, using a method of chronosequences of agrogenic soil transformation.

2. Methods

2.1. Study area and soil sampling

The objects of study are the humus-accumulative horison of 20 soils and loesslike loams in two regions of the Crimea: ancient farming zone, distant chora of Tauric Chersonesos and Eastern Crimea, chora of Bosporan towns. The representative members of agrogenic soil transformation series were formed for each of the two of the regions. Justification of the objects of research in the soil chronosequence of agrogenic transformation is given earlier [7], as well as the accepted encoding: virgin land (VL); modern-day ploughed land (MPL); continually ploughed land (CPL); post-antique long-term fallow land (PLFL). The Tarkhankut is part of the Crimean steppe plain, where the main soils are Calcic and Petrocalcic Chernozems [7]. It is reasonable to involve geochemical background when studying the local soil standard within the botanical reserve “A section of the virgin steppe on the Tarkhankut Peninsula” (100 ha) west of the village of Krasnoselskoe (Chernomorskiy region). Virgin soil (according to the morphological structure of the profile) on Kerch Peninsula is a medium-loamy southern carbonate Chernozem (1.6 km south of the village of Geroyevskoye). The continually ploughed land is defined by the locations of the agricultural areas near Bosporan small towns and agricultural settlements with a final activity period from 4^{th} c. BC to 5^{th} c. AD. The post-antique long-term fallow land was studied near ancient settlements with the end of life in 1^{st} c. BC – 2^{nd} c. AD. Soil sample preparation included cleaning from inclusions and grinding it into powder.

2.2. Analytical and evaluation methods

Standard procedures have been applied for chemical analyses of soils: assessment of the CO_2 in carbonates by acidometry; the organic carbon (Corg) after Tyurin; the available P_2O_5 by Machigin’s method; particle size distribution by method pipettes treated with sodium pyrophosphate. X-ray Fluorescence Analysis (XRF) method was used to determine the contents of chemical elements. The concentrations of macro- and trace elements in soils (S) and parent rock (P) were determined in powdered samples. Accepting Liu’s proposition [8] concerning the eluviation coefficient (Ke), the modified variant of the formula: Ke = (Si / (Mn + Ca + K + Mg + Na)).

The list of soil heavy metals of highly hazardous and moderately hazardous classes, which is fore sawn by the general toxicological GOST 17.4.1.02-8, with addition of modern ecotoxicological data of Dutch ecologists according to degree of hazard of soil heavy metals / metalloids (HMM) [9], [10] allowed us to determine a summary series from nine HMMs with allocation of two groups according to the degree of danger: (V, Ni, Cu, Cr, As, Ba)> (Pb, Zn, Co).

The values of maximum permissible concentration (MPC_i) for the main HMM were determined based on the review [11], in addition to Ba, for which the limiting value is conditionally taken from the maximum concentration range in soils (1000 ppm).

Hazard degree (HD) of soil contamination HMM was evaluated by the formula:

$$HD = \frac{1}{n}\sum_{i=1}^{n}\frac{S_i}{MPC_i} \quad (1)$$

S_i is the content of HMM and their maximum permissible concentration (MPC_i).

3. Results

A certain idea of soil buffering and the conditions for the interaction of HMM with the active phase of the soil system is provided by data on the contents of Corg, $CaCO_3$, MnO, total Fe, and particles size <0.01 mm. The granulometric composition of the soils of the North-western Crimea is significantly different. Horizon A of virgin land (Petrocalcic Chernozems) has a loam composition with high carbonate content, and agrosoil (Calcic Chernozems) has a heavier composition: loam clay and silty clay loam with a significant $CaCO_3$ content in modern-day ploughed land. Agricultural soils contain more Fe and Mn than virgin land and fallow land. Because of bioaccumulation, the humus-accumulative horizon of virgin soil and long-term fallow land has a 22-29% higher content of potential pollutants compared to parent rock (Table 1, ratio S/P).

Table 1 – The key of physico-chemical indicators for soil chronosequence (North-western Crimea)

Index	Member of soil chronosequence			
	VL	MPL	CPL	PLFL
Corg, %	2.73	1.43	1.39	2.00
<0.01 mm, %	23.15	53.78	42.68	35.19
CaCO3, %	26.0	8.97	23.27	16.97
Fe, %	2.81	3.28	2.88	2.66
MnO, %	0.09	0.11	0.12	0.09
V, ppm	80.61	84.33	99.81	67.55
Ni, ppm	48.48	50.44	48.11	45.33
Cu, ppm	42.93	50.40	39.13	38.39
Cr, ppm	82.41	97.98	85.43	82.39
As, ppm	8.21	8.27	8.58	8.20
Ba, ppm	434.98	428.56	518.64	426.87
Pb, ppm	17.11	23.18	18.04	16.98
Zn, ppm	84.70	75.61	80.64	73.76
Co, ppm	12.63	18.00	15.01	10.95
S/P	1.29	1.07	1.10	1.22
Ke	1.38	1.53	2.25	2.51
HD	0.68	0.70	0.57	0.53

The eluviation coefficient (Ke) can safely diagnose the duration of easily mobile soil components leaching in relation to quartz: the Ke value tends to increase from the modern arable land to soils with development prehistory and post-antique long-term fallow land. The comparison of average excess concentrations of HMM in soil chronosequence members compared to virgin land (Table 1) showed that the maximum excess was found for modern-day ploughed land (1.12 times), close to continually ploughed land (1.07), and fallow land (0.93) did not accumulate. Agricultural loads on horizon A against the background of decreased Corg content can lead to the loss of potential pollutants as well. Long-term agricultural land development reduces the risk of soil contamination by nine HMMs, in particular, the content of Co> Pb> Cu> Cr is reduced compared to modern-day ploughed land. The granulometric composition of the soils of the Eastern Crimea in the composition of the agrogenic transformation series is significantly different. Unlike horizon A virgin land on the coastal terrace with a loam composition at high carbonate content, agricultural soils (Calcic Chernozems) have a heavier composition (silty clay loam and sandy clay), which is typical for the Kerch Peninsula and contain less $CaCO_3$. In addition, agricultural soils have a slightly higher Fe and Mn content than virgin land (Table 2). The Eastern Crimea bioclimatic conditions did not contribute to the accumulation of potential pollutants in the humus-accumulative horizons of all soil chronosequence members compared to parent rock (Table 2). As compared both to the background (NW Crimea soil), the high eluviation coefficient values for arable soils and fallow land show that plowing has an extremely strong influence on the geochemical transformation of horizon A. The average excess concentration of HMM in soil chronosequence members compared to virgin land (Table 2) is maximum in continually ploughed land (1.80 times) and is close to modern-day ploughed land (1.53) and fallow land (1.46). An overall hazard degree assessment of soil contamination by nine HMMs has shown that even under active leaching of easily mobile components of ploughed land and post-antique long-term fallow land they are characterized by 1.4 times stronger accumulation of pollutants as compared to wild land, while continuously ploughed land has the largest accumulation of HMMs, which cumulatively does not exceed the established limits of MPC, mainly due to Co, Ba, and Pb. The relic evidences of previous agricultural loads have been preserved in horizon A of the post-antique long-term fallow land as compared to virgin land by a higher concentration of HMMs, which can be represented as an ordered descending series: Co>Pb>(Zn, V)>As>Ni>Ba>Cu.

Table 2 – The main indicators of physico-chemical properties for members of soil chronosequence (Eastern Crimea)

Index	Member of soil chronosequence			
	VL	MPL	CPL	PLFL
Corg, %	1.69	2.11	1.98	2.79
<0.01 mm, %	31.89	55.52	74.51	60.19
CaCO3, %	21.25	6.00	6.15	8.47
Fe, %	2.47	3.53	4.84	3.71
MnO, %	0.08	0.11	0.15	0.09
V, ppm	62.38	96.24	120.73	102.09
Ni, ppm	35.95	53.47	70.17	51.01
Cu, ppm	38.78	51.86	69.21	45.72
Cr, ppm	101.59	110.82	124.14	114.51
As, ppm	10.53	11.93	13.42	15.19
Ba, ppm	439.43	532.18	536.38	554.34
Pb, ppm	13.81	26.35	26.95	22.93
Zn, ppm	43.18	66.11	96.77	71.01
Co, ppm	8.29	20.90	21.44	14.96
S/P	0.97	1.04	0.99	1.06
Ke	2.83	5.69	4.22	4.55
HD	0.60	0.84	1.01	0.82

Comparative data analysis (Table 1, 2) enables to see differences in chronosequences of agrogenic soil transformation for the two studied Crimean regions. In the North-Western Crimea, the horizon A of the virgin soil is the most enriched with Co, Cu, Pb, Ni compared to parent rock, while the same indicator (S/P) was noted to be exceeded for a smaller number of HMMs (As and Cu) in the Eastern Crimea. This is due to differences in the bioclimatic potential of these two regions. A comparison of the eluviation coefficient values shows that the process of mobile oxides leaching is twice as intensive in the Kerch Peninsula as in the Western Crimea. Climatic differences between the two regions (North-West and East Crimea) are demonstrated by a comparison of climatic parameters at the Chernomorskoe and Kerch weather stations. The territory of the Kerch Peninsula has more humid climate (the annual precipitation is more than 96 mm) with a lower volatility (35 mm) than in the North-Western Crimea. In addition, the concerned regions with selected soils differed in farming duration, which was longer on the lands of the Bosporos than on the far chora of the Chersonesos.

The newly developed soils (MPL and CPL) may differ from the post-antique long-term fallow lands (PLFL) with a higher level of HMM contamination due to the use of phosphoric fertilizers, which are the main source of contamination of arable soils (up to 50%) [12]. The ploughing horizon of old-arable and newly developed soils is provided with mobile phosphates highly (0.5-0.8 mg kg^{-1}) in the North-Western Crimea and moderately (0.20 mg kg^{-1}) in the Eastern Crimea. Pollutants that enter the soil interact with its active phase (organic matter, clay minerals, etc.), which changes their activity and hazard respectively [9]. The soils studied by us differ in the content of carbon, carbonates and physical clay (particle size < 0.01 mm). The most significant abiotic control factors for trace elements behaviour [13] include the presence of Fe/Mn-enriched particles, which is typical for silt. As compared to the virgin analogue, any cultivated soils have a significant advantage in particles proportion of <0.01 mm, with a slightly higher Fe and Mn, lower or closer Corg content.

The study allows us to formulate the following main conclusions:

1. A comparison of hazard degree values (taking into account the maximum permissible concentration levels) in four soil chronosequence members has shown that the Calcic and Petrocalcic Chernozem (NW Crimea) are more polluted by HMMs than the wild land if it is modern-day ploughed land. However, both modern-day ploughed land and continuously ploughed land have excess content of such HMMs as Co, V, Cr, and Pb, compared to the background.

2. Chernozems southern calcic (Eastern Crimea) are more polluted by HMMs as compared to the background content of both modern-day ploughed land and post-antique long-term fallow land. Longer-term arable farming than in NW Crimea has resulted in excess content of nine HMMs as compared to the wild land, which a ranked series reflects: Co > (Zn, Pb) > V > Ni > Cu > As > Ba > Cr.

3. If the Eastern Crimea virgin soil is only enriched in two elements (As, Cr) as compared to its analogue in NW Crimea, the past and present agro-soils on the Kerch Peninsula differ in a more significant accumulation of all HMMs (by an average of 29%), especially in the As, Pb, Cu, Cr, Co, V content. The reasons that determine these features are the longer agricultural pressure in the ancient period of arable farming on the European Bosporus.

4. When using the geometric mean estimated contents of HMMs in soils of agrogenic transformation series relative to the background content, it is found that accumulation of the most dangerous HMMs (V, Ni, Cu, Cr, As, Ba) in the Eastern Crimea soils is 2.8 times higher than the estimated value for the soils of NW Crimea; however, the differences in the accumulation of less dangerous HMMs (Pb, Zn, Co) are non-significant in both regions.

5. The established trends in HMM accumulation as a result of long-term agropedogenesis make it necessary to maintain priority control over the content of Co>(Cr, Ba, Pb, V) in the soils of NW Crimea and over the content of Co>(Zn, Pb)>V>Ni>Cu>As>Ba>Cr in Eastern Crimea soils in the regional systems of agro-ecological regulation and selection of territories for growing organic agricultural products.

Funding

This work was funded by the Russian Science Foundation, project no. 20-67-46017.

Conflict of Interest

None declared.

Конфликт интересов

Не указан.

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