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Influence of Chemical Amelioration of Solonetzic Soils on the Presence of Heavy Metals and Radionuclides in Soil Profile

Guzeeva Svetlana Anatolievna[1]

[1] Tyumen Industrial University, Lunacharskogo 2, Tyumen, 625001, Russia
e-mail: nesterka07@mail.ru

ABSTRACT
The article considers the results pertaining to the influence of solonetzic soils chemical amelioration on the presence and redistribution of heavy metals and radionuclides in meter profile of soils. Meliorative procedures were carried out 45 years ago. Phosphogypsum has been used as chemical ameliorant up to 90-s on the solonetzic soils of Tyumen region. It is a by-product of phosphates production. The objective aimed at revealing possible negative consequences connected with the introduction of the chemical elements associated with phosphogypsum. The studies showed that phosphogypsum introduction contributes to some increase in the content of active forms of heavy metals (zinc Zn, copper Cu, cadmium Cd and lead Pb) in soil. However, critical values are achieved only by active lead Pb. The evaluation of radioactive state of meadow crusted solonetzic soils after carrying out of meliorative procedures (for the same period) on the content of isotopes of thorium-232 (Th-232), calium-40 (K-40), radium-226 (Ra-226), caesium-137 (Cs-137) and strontium-90 (Sr-90) indicated that redistribution of radionuclides after chemical melioration does not cause a negative change of radiation background. Vibrations in the soil profile are connected with various initial concentration in rock formations or weathering detritus. Specific effective activity ($A_{eff} \leq 370$ Bq/kg) testifies to the soils being studied and absence of their additional introduction with introduced doses of phosphogypsum.

Keywords: phosphogypsum, amelioration, consequence, heavy metals, radionuclides.

INTRODUCTION

At present, the issue of soils contamination with heavy metals and radionuclides becomes more and more significant. The soils are natural accumulators of heavy metals and the first screen on the way of radioactive substances inflow from the atmosphere to the earth surface. Thus, they contribute to the contamination of plants, animals and other objects of the environment.

One should consider the use of ameliorants and mineral fertilizers, especially phosphoric and potassium as the main source of contamination of soils of agricultural meaning with heavy metals. In the opinion of Alexeev [2008], Berezin et al. [2008], and Vodyanitskiy et al, [2012], the reason for this is the presence of strontium, uranium, lead, vanadium and other elements in the raw material for the production of ameliorants and mineral fertilizers.

Perelman et al. [1999] notes that the more intensive element extracted from the earth's crust is, the sooner relative concentration of the biosphere takes place. According to the scientist's data, the following order of heavy metals technophility can be presented: Pb > Hg = Cd = Cu > Sn > Zn = Mo > Cr > Ni > Fe = Mn > Co. This indicates that the global biogeochemical background will increase to the fullest extent in the case of lead, mercury, cadmium and copper.

In recent times, the content of heavy metals in soils, their state and ecological evaluation has been studied by such researchers as Ilin [2012], Kazantsev et al. [2016], Merzlaya et al. [2009], Mikhalchuk [2017], Ovsyannikova et al. [2016].

Particular danger of contamination with heavy metals and radionuclides in relation to solonetzic soils is caused by the use of phosphogypsum as an ameliorant. Phosphogypsum constitutes waste product of orthophosphoric acid and mineral fertilizers (double superphosphate, ammophos and ANP fertilizer). Phosphogypsum is obtained in a form of mud with humidity up to 55% which contains small amount of impurities (3–7%) and by the content of the main component ($CaSO_4 \times 2H_2O$) it relates to gypsum raw material of 1–2 types. The chemical composition of phosphogypsum is mainly determined by the quality of used phosphate raw material.

In the Tyumen region, solonetzic soils occupy the area of about 350 ha. In 1970s and 1980s, the agrobiological and chemical methods of amelioration were actively used in the region. Phosphogypsum was introduced on the area of more than 20 thousand ha, and its doses varied from 10 to 40 t/ha [Skripin et al. 2013]. However, the monitoring of the state of gypsum solonetzic soils contamination with heavy metals and radionuclides was not carried out.

The goal of the presented studies was to carry out an ecological evaluation of the possibility of contamination of solonetzic soils with heavy metals and radionuclides during their chemical amelioration with phosphogypsum.

METHODS

Field test was conducted in 1972 in the state farm Vagijskiy of Omutinskiy region. Phosphogypsum was introduced in half dose and in full dose (43 t/ha). The area of land was 400 m^2, test replication is threefold. Selection of soil samples took place in 2017 layer by layer up to the depth of 1 m.

The extraction of the active forms of heavy metals was carried out using acetate-ammoniacal buffer solution with pH=4.8. A chemical analysis on determination of quantitative content of active forms of heavy metals was carried out on the spectrophotometer AAS-3m according to the ‘’Methodical instructions on the determination of heavy metals in soils and products of plant growing’’, approved by the Ministry of Agriculture of RF, CINAO 1992, and GOST P to GOST 26932–96 and GOST 26933–96.

Radiometry of the soil samples was carried out on the spectrometric complex ‘’Progress’’. Determination of caesium-137 was carried out with the method of scintillation gamma spectrometry and strontium-90 by means of scintillation beta- spectrometry method.

RESULTS

The data obtained from the analysis on the content of heavy metals in soil sample is given in the Table 1. The studies showed that in the case of introducing large doses of phosphogypsum in meliorated solonetzic soils, a tendency of increasing heavy metals content it noted, especially in 0–20 cm layer. With an increase of phosphogypsum dose this phenomenon was more intensive. Thus, heavy metals content in the layer 0–20 cm increases when introducing phosphogypsum in doses 21 and 43 t/ha: zinc (Zn) by 1.1–1.2 times; copper (Cu) by 1.4–1.8 times; cadmium (Cd) by 1.4–1.7 times and lead (Pb) by 2.1–2.6 times. An analogous regularity was demonstrated on the whole 1-meter depth of soil profile. It is important to note that exceedings by Zn, Cu and Cin respect to MPC did not take place.

The accumulation of lead during the phosphogypsum introduction causes the largest danger. At the same time, its content in the initial parent rock increases MPC (6 mg/kg) and reaches 9.4–11.8 mg/kg. Introduction of phosphogypsum led to the increase of lead by the soil profile. In 0–20 cm layer, its size reached the MPC level and made up half-dose 5.05–7.09 mg/kg, by full dose 6.34–8.43 mg/kg.

The obtained results testify that the introduction of this industrial waste for solonetzic soils amelioration is mostly connected with the accumulation of lead in the soil. At the same time, the possibility of its accumulation in the ground waters is not excluded.

According to Bykova et al. [2013] and Dobrovolskiy et al. [2012], soil and vegetation cover of the biosphere occupies significant place in the radioecological studies. Radionuclide exchange between atmosphere and hydrosphere is carried out through it as through global nonliving membrane. At the same time, soils become the main source of accumulation of radionuclides in the surface natural environment.

Evaluation of radioactive state of meadow solonetzic soils by the content of thorium-232 (Th-232), potassium-40 (K-40) and radium-226 (Ra-226) showed that these soils do not cause danger in respect to the selection of place of living and agricultural works. Thus, the maximum specific effective activity of natural radionuclides of the soil reached 118.7 Bq/kg. According to GOST data (30108–94), A_{eff} should not exceed 370 Bq/kg.

Table 1. Influence of phosphogypsum on heavy metals content in meadow crusted solonetzic soils (mg/kg) for 1972–2017

Depth, cm	Zn	Cu	Cd	Pb
Control (without phosphogypsum)				
0–10	1,67	0,34	0,15	3,99
10–20	1,48	0,48	0,67	1,73
20–40	2,30	0,87	0,79	0,06
40–60	4,13	1,50	1,20	6,83
60–80	4,71	1,74	1,06	9,37
80–100	4,16	1,45	1,18	11,79
Phosphogypsum 21t/ha				
0–10	1,78	0,59	0,47	7,09
10–20	1,53	0,58	0,66	5,05
20–40	2,78	0,97	0,89	1,53
40–60	2,44	1,25	1,35	11,86
60–80	4,32	1,16	1,62	12,06
80–100	5,95	1,43	1,35	13,36
Phosphogypsum 43 t/ha				
0–10	2,05	0,77	0,63	8,43
10–20	1,59	0,73	0,78	6,34
20–40	5,89	1,13	0,90	4,05
40–60	5,21	1,68	1,59	14,51
60–80	5,52	1,72	1,31	17,90
80–100	5,87	1,44	1,35	17,90
MPC	**23**	**3**	**1–5**	**6**

Thus, distribution of natural nuclides in meadow crusted solonetzic soils does not cause a negative change of radiation background. The vibrations in the soil profile are connected with their different initial concentration in rocks, weathering detritus and also with the quality of introduced ameliorant.

According to the data of Sorbat et al. [2016], the most dangerous artificial radionuclides being uranium decay products are cesium-137 (Cs-137) and strontium (Sr-90). Cesium-137 resembles potassium in terms of its chemical properties. It is easily dissolved in the water and can accumulate in soft tissues and spleen. It is easily excreted from the organism. As far as the chemical properties are concerned, strontium-90 is close to calcium. In a living organism, it takes active part in the building and renovation of bone tissues. Therefore, strontium-90 accumulates in the bones and excretes from the organism very slowly, becoming a source on internal radiation of stem cells of red bone marrow with high-energy β-particles for many years.

It is known that strotium-90 and cesium-137 are absorbed by the soils according to the type of exchange-ionic absorption. However, the absorbed cesium-137 is fixed rather more firmly than strontium-90. More recent studies revealed that part of the absorbed cesium-137 is fixed in kidneys without exchange.

The studies of meadow crusted solonetzic soils showed that out of the two presented artificial radionuclides, the main saturation took place due to cesium-137 and strontium-90 (Table 2) to a lesser degree.

The content of cesium-137 in solonetz of virgin lands in soil profile varied from 8.5 to 15.3 under MPC 185 Bq/kg. The concentration of strontium-90 did not exceed 1.9 under MPC 55.5 Bq/kg. The introduction of phosphogypsum in full and in half doses did not lead to a notable increase of content of the presented radionuclides in meter layer.

Ratio strontium-90/cesium-137 (Sr/Cs) in meliorated solonetzic soils of the south of Tyumen region layer 0–20 cm that made up in average 0.1–0.2 that in general corresponds to Ural region. An increased ratio of strontium-90/cesium-137 (Sr/Cs) in meter layer testifies to the prevalence of radioactive cesium in the soil.

Table 2. Influence of phosphogypsum on the content of cesium and strotnium-90 in the profile of meadow crusted solonetzic soil, Bq/kg for1972–2017

Depth, cm	Cs	Cs/Sr	Sr	Sr/Cs
Control (without gypsuming)				
0–10	15,0	13,4	1,1	0,1
10–20	8,5	4,5	1,9	0,2
20–40	15,3	12,5	1,2	0,1
40–60	12,1	12,3	1,0	0,1
60–80	13,9	7,5	1,8	0,1
80–100	15,3	12,5	1,2	0,1
Phosphogypsum, 21t/ha				
0–10	11,6	8,7	1,3	0,1
10–20	11,7	9,5	1,2	0,1
20–40	13,8	9,8	1,4	0,1
40–60	11,0	11,0	1,0	0,1
60–80	7,6	5,0	1,5	0,2
80–100	10,7	7,9	1,4	0,1
Phosphogypsum, 43t/ha				
0–10	10,5	5,1	2,1	0,2
10–20	9,3	8,6	1,1	0,1
20–40	14,2	7,6	1,9	0,1
40–60	11,0	10,3	1,1	0,1
60–80	7,9	5,0	3,3	0,4
80–100	15,0	6,9	2,2	0,2
MPC	**185**	**-**	**55,5**	**-**

CONCLUSION

1. Introduction of phosphogypsum as meliorant to the meadow crusted solonetzic soil contributes to the insignificant increase of heavy metals content in it.
2. The content of active form of zinc, copper, cadmium on the meadow solonetzic soil is lower than maximal permissible concentrations; however, it exceeds the background indices.
3. The greatest danger in meliorated solonetzic soils is caused by the accumulation of lead.
4. The content of radionuclides in soil profile did not exceed MPC which testifies to the favorable radiation state of solonetzic soils.
5. After carrying out meliorated events, these soils can be used in tillage without limitation during cultivation of the main cultures.

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