DISTRIBUTION OF ^{137}CS AND ^{239,240}PU PHYSICOCHEMICAL FORMS IN WATERLOGGED AND NON-BOGGY SOILS

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Sequential extraction is a widely used technique for determining the solid speciation of a trace metal or radionuclide, which explains under what chemical or physical conditions the metal will be remobilised. The technique utilises a succession of chemical extractants, chosen to selectively dissolve the various geochemical phases likely to be affected by changes in environmental conditions within the sediment or the surrounding water. Metals and radionuclides associated with a given phase will be released upon dissolution of the host phase (Lucey at al. 2004). Sequential extraction technique, in which the sediment or soil is separated into defined, sequentially-dissolved phases via a series of increasingly aggressive chemical extractions, can be used to determine their geochemical associations indirectly. This can provide useful information on the potential migration of the radionuclides in soils (Kimber et al 2015).

Two samples of non-boggy and waterlogged soils were taken in an untouched place near the lake Bedugnis, which is located in Varėna district. Physicochemical forms of tested radionuclides were studied in soil layers with maximum concentrations of the radionuclides since their content may be considered as the main source of further vertical migration of radionuclides. In both cases, maximum concentrations of the radionuclides were obtained at the same depth (6-8 layers).

The goal of our study was to evaluate the vertical distribution of ^{239,240}Pu and ^{137}Cs in the waterlogged and non-boggy soil samples as well as to determine their physicochemical forms in soil layers with the maximum concentrations of the radionuclides for the aim to define possible differences in the radionuclide migration.

The geochemical forms of Pu and ^{137}Cs as well as their migration opportunities in waterlogged and non-boggy soils significantly differ. Thus, in waterlogged soil where anaerobic conditions are usually formed, the most part of radiocesium (~99 %) was bound in the residue due to the presence of clay materials and its migration opportunities were restricted. In contrary, the greatest migration opportunities of radiocesium were found in non-boggy soil under aerobic conditions. Thus, in the non-boggy soil core (6-8 layers), its exchangeable fractions varied in the range of 10.1 – 12.2 %. A radiocesium part in the residue amounted only ~61 - 71 %.

Table 1. Activity concentration of radionuclides and properties of 6 - 8 layers of the **non-boggy** soil core.

Depth	6-th layer	7-th layer	8-th layer
pH (H$_2$O)	3.91	4.21	4.43
Organic matter quantity, %	35.5	16.3	6.9
^{239,240}Pu, Bq/kg	1.47	4.85	1.98
^{137}Cs, Bq/kg	66.6	66.0	28.0

Table 2. Activity concentration of radionuclides and properties of 6 - 8 layers of the **waterlogged** soil core.

Depth	6-th layer	7-th layer	8-th layer
pH (H$_2$O)	4.28	4.19	3.94
Organic matter quantity, %	86.8	69.1	13.9
^{239,240}Pu, Bq/kg	0.89	0.84	0.64
^{137}Cs, Bq/kg	18.6	18.0	18.7

Fig.1. Distribution of ^{137}Cs activity concentrations (fractions, %) in a) sixth, b) seventh and c) eighth layers of the waterlogged and non-boggy soil cores in the physicochemical forms of soil components.
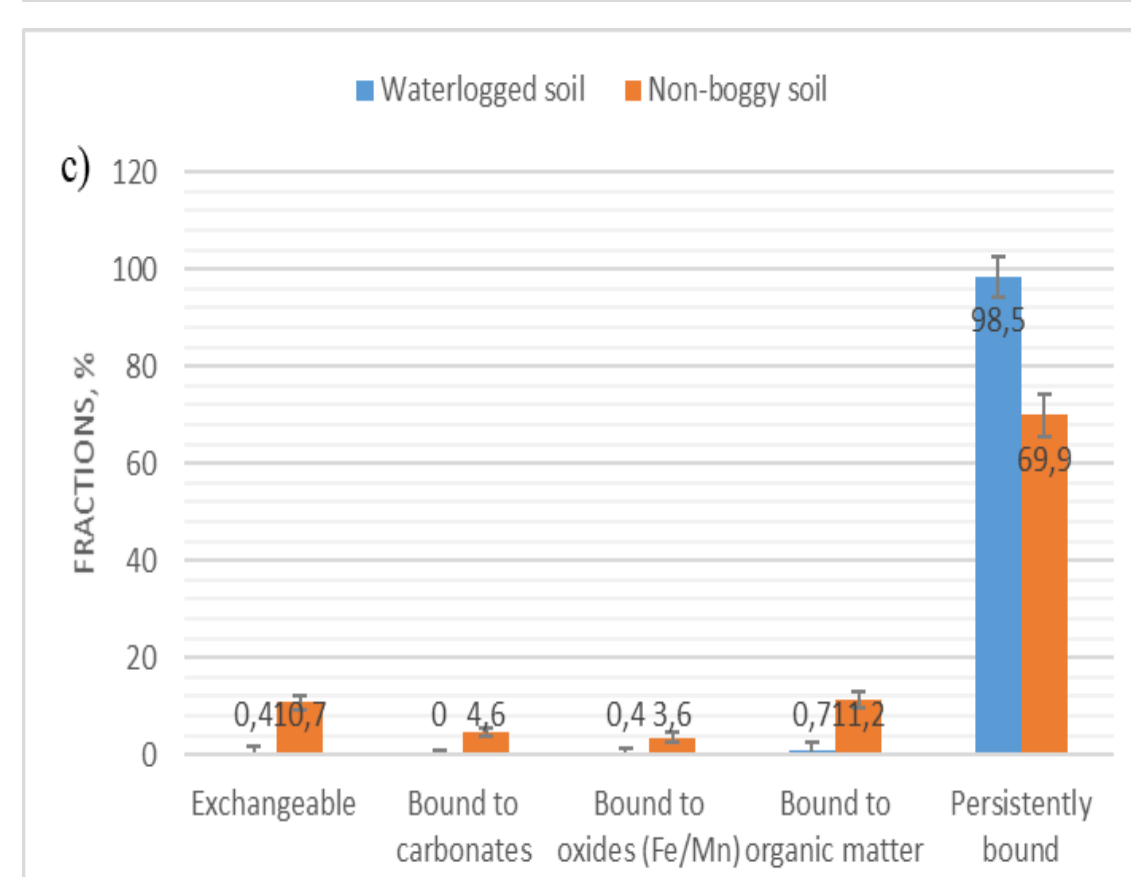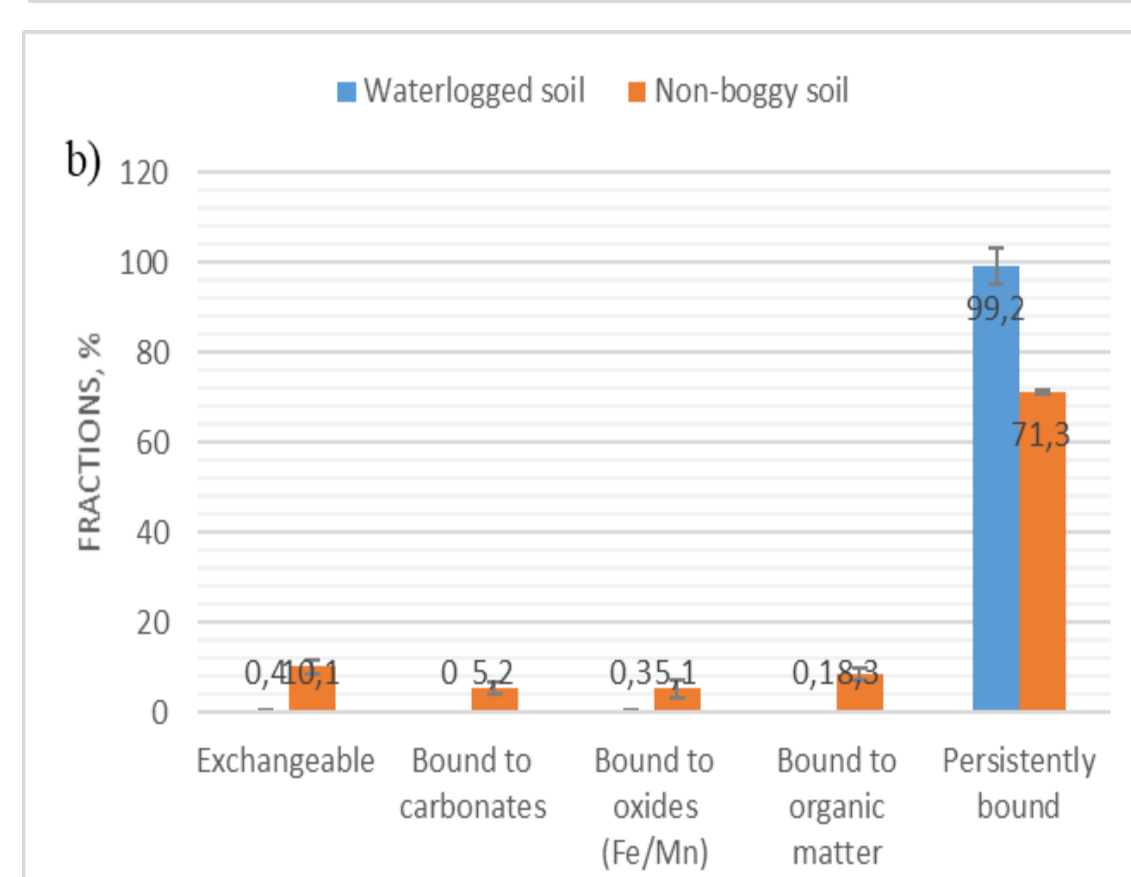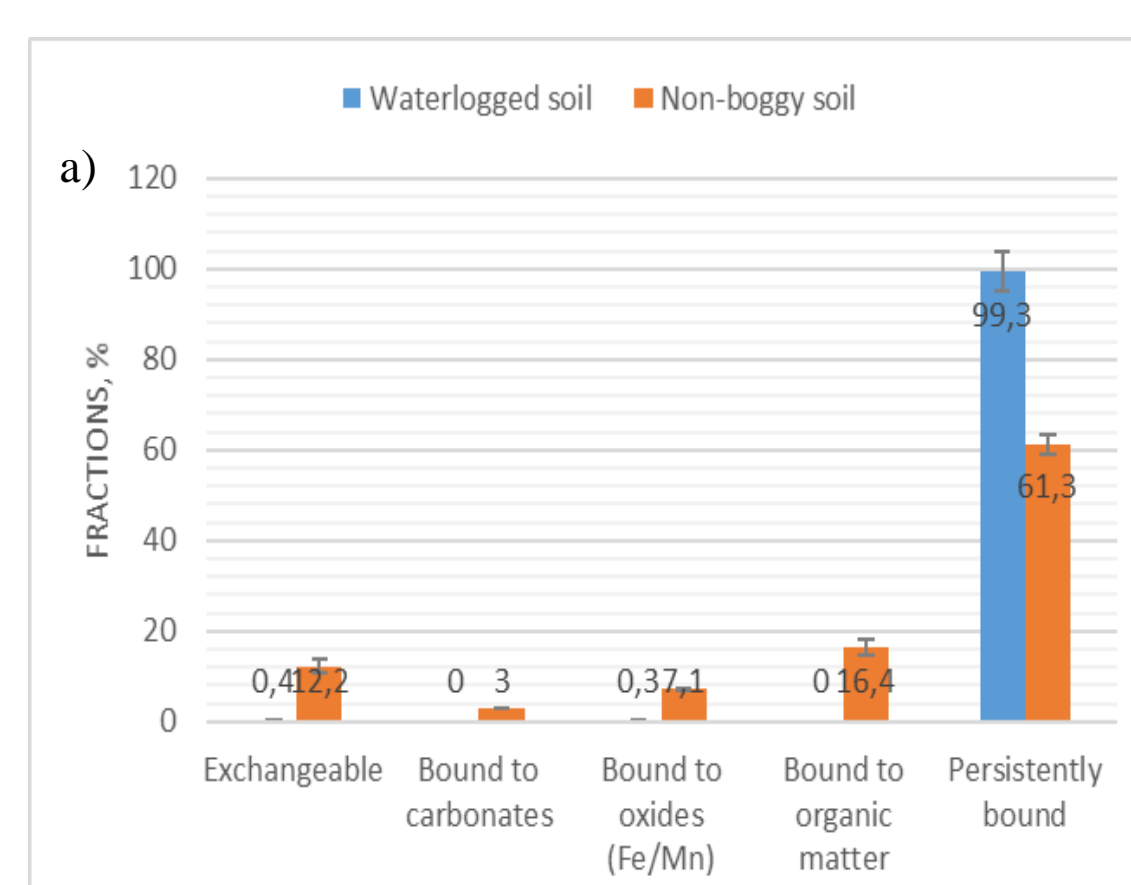
The data on the ^{137}Cs activity concentrations (fractions,%) in the physicochemical forms (extracts) of the studied sixth – eighth layers (6-8 cm) of the non-boggy and waterlogged soil cores are shown in Fig.1 .

Determination of point zero charge was performed on the tested soils, which showed that:

• soil particles of the non-boggy soil sample have a positive surface charge which means that cation sorption due to electrical forces does not occur;

• and waterlogged soil sample have negative surface charge which indicates the presence of a negative charge on the surface of soil particles, promoting the sorption of cations.

Data on ^{239,240}Pu activity concentrations (fractions, %) in physicochemical forms (extracts) in the studied sixth – eighth layers (6-8 cm) of the non-boggy and waterlogged soil cores are shown in Fig.2.

Fig.2. Distribution of ^{239,240}**Pu** activity concentrations (fractions, %) in a) sixth, b) seventh and c) eighth layers of the waterlogged and non-boggy soil cores in the physicochemical forms of soil components.
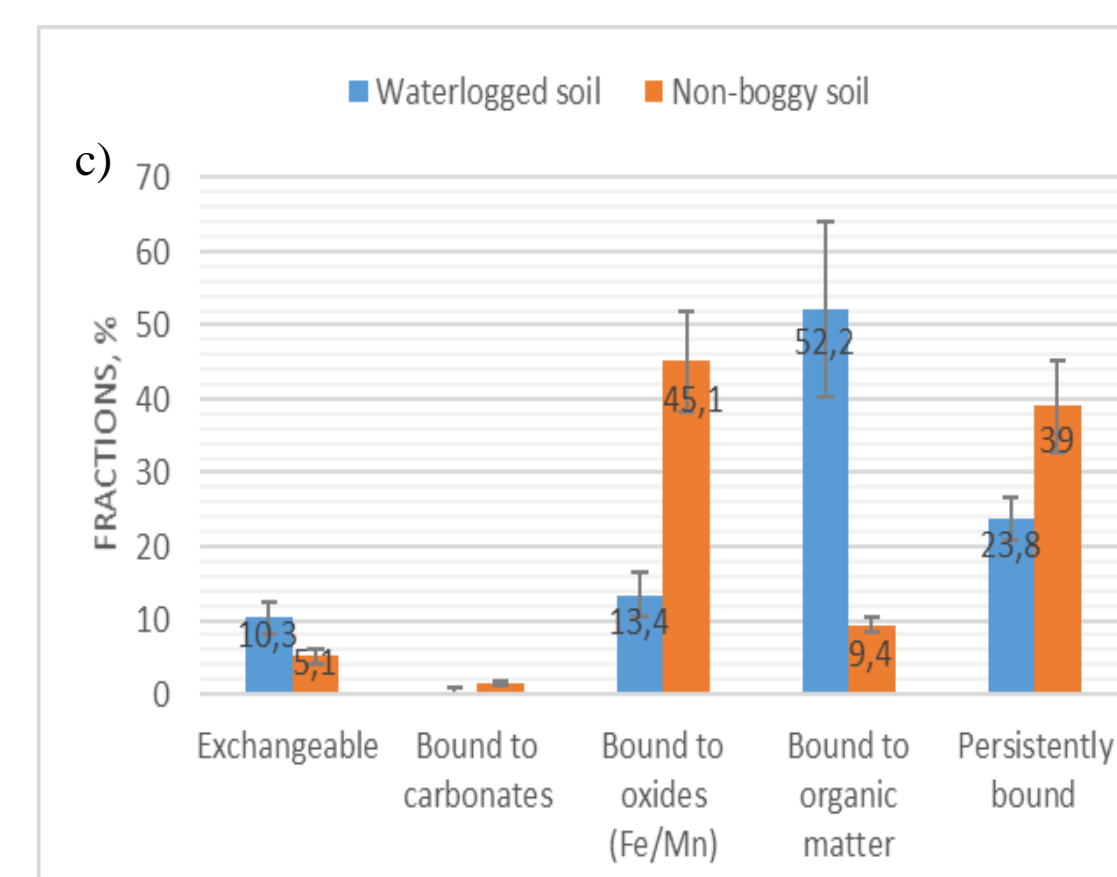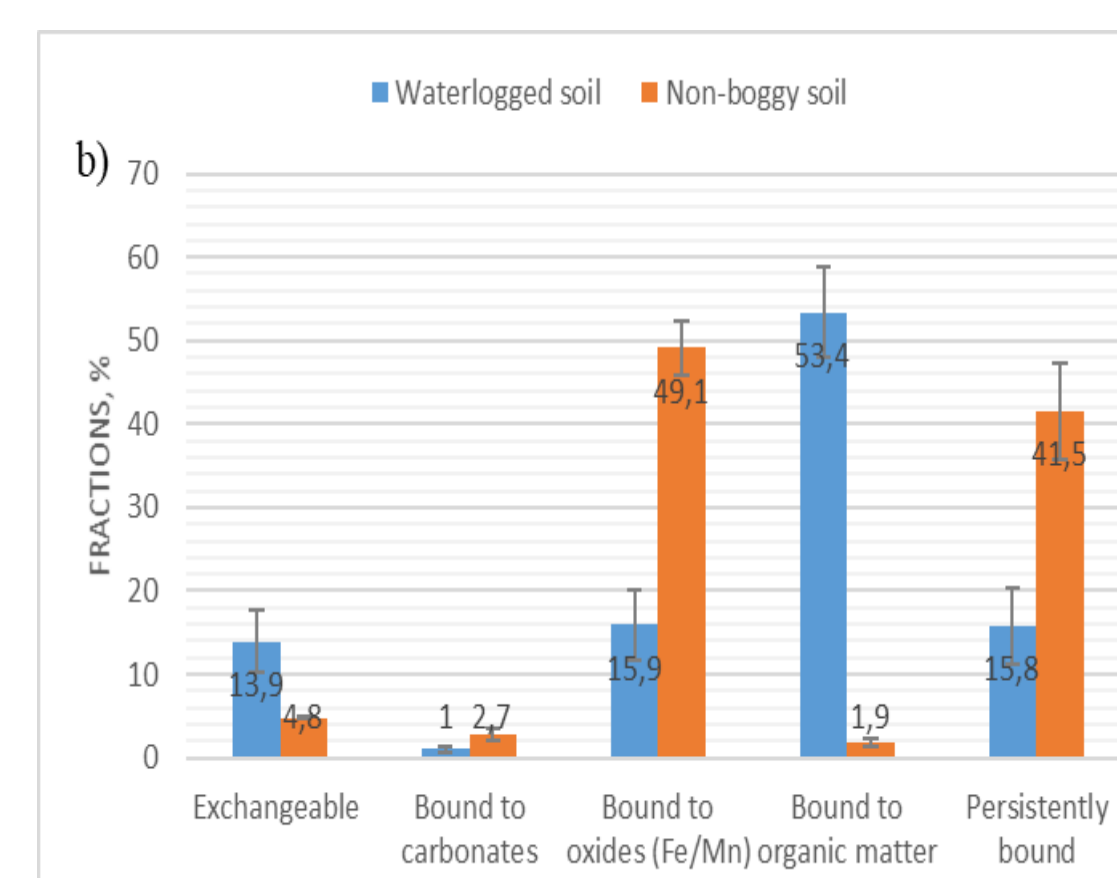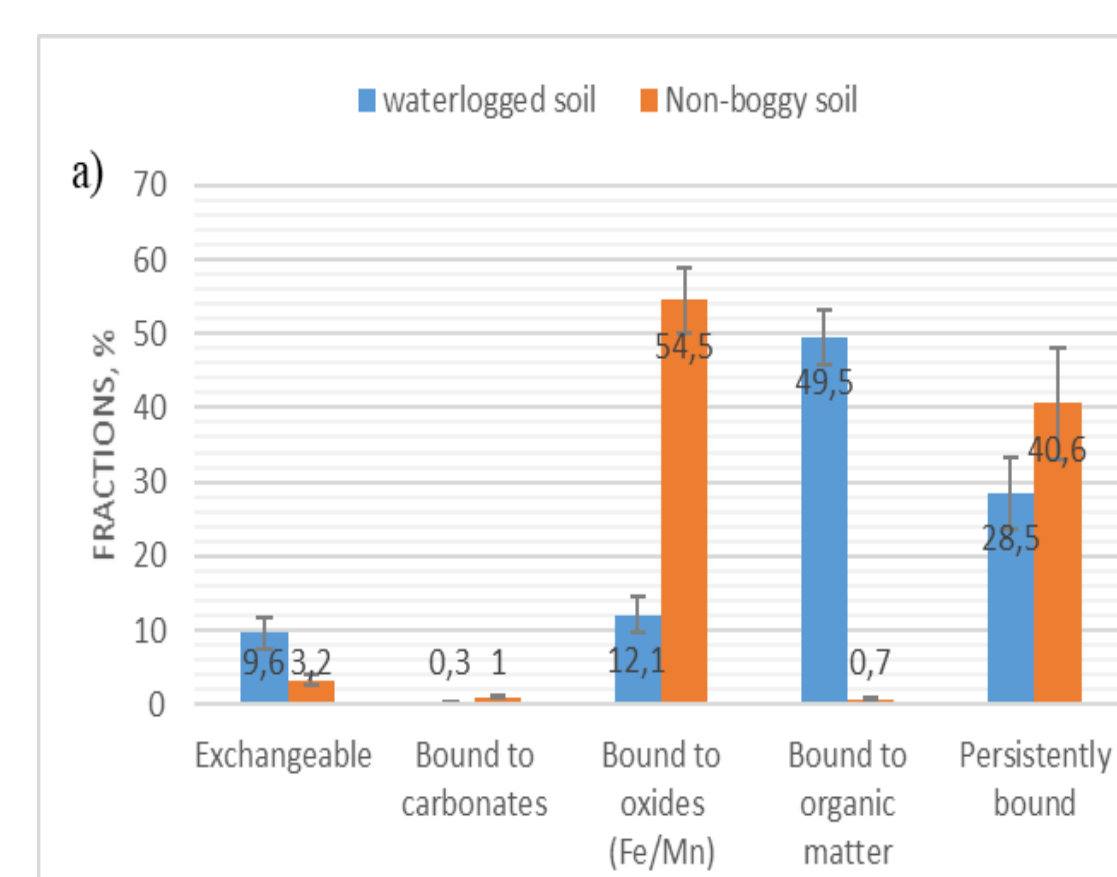
• In the studied layers of non-boggy soil, most of the ^{239,240}Pu was in the oxide fraction (~45 - 54 %), lower in the residual (~23 - 40 %) fraction and then in the exchangeable one (~3 %).

• In contrary, in the same layers of the waterlogged soil core, the most part of the ^{239,240}Pu was concentrated in the organic matter fraction (49-53 %), less in the residual (16 - 28 %), oxide (12 - 16 %) fractions and in the exchangeable (10 - 14 %) fraction.

• The latter indicates a large migration capacity of plutonium under anoxia conditions and that absorption of ^{239,240}Pu by clay materials is not a dominant factor.

• This means that in the studied soils in the area of Lake Bedugnis the radionuclide mobilizing factors act as opposite for radiocesium and plutonium

Most of the stable elements were accumulated in the residual, organic matter and the oxide fractions. The distribution of Al in waterlogged soil was very similar to that of ^{239,240}Pu (organic fraction> oxide and residual fractions).

The results of this study indicate that the physicochemical forms of radionuclides in the soil are an indicator of their mobile capabilities and this method can be applied to the assessment of radionuclide migration potential of the contaminated area in different soils.

References:
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