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The Nature of High Soil Radioactivity in Chinese Province Guangdong

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Abstract

Soil is a basic component of biosphere and its important natural resource. The article deals with the analysis of soil radioactivity in Chinese province Guangdong. In the course of the analysis, it was stated that highly radioactive soil of China had been formed due to deep chemical weathering of highly radioactive potassium granites. High uranium and thorium contents in them are caused by specific conditions of weathering crust formation and subsequent pedogenesis. High dose loads for a man are formed in the development fields of such rock types.

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1. Introduction

There are several provinces on the globe that have high content of natural radioactive elements in soil. The examples are soils of Poços de Caldas province of Minas Gerais State in Brazil, that of New Island as well as soil in the South-Chinese Province Guangdong¹.

The character of soil radioactivity varies from pure radioactive ($U > Th$, which is typical for soil of New Island) to mixed uranium-thorium ($Th/U > 2.5-5$) and thorium ($Th/U > 5$, as it is in the soil of Guangdong Province). For the latter, it is suggested that its radioactivity is explained by the presence of monocyte.

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As a reason for the formation of high natural radioactive concentrations in soil, elevated concentrations of these elements in primary parent rocks and various geologic processes leading to accumulation of radioactive elements, for example, insolation processes of uranium accumulation as well as anthropogenic contamination with radioactive components in vicinity of mining factories can serve².

The purpose of the given work is to study the causes of high soil radioactivity in the South-Chinese Guangdong Province.

2. Methods

The preliminary gamma-spectrometric soil analysis (soil samples weighting 238 g) show that they are characterized by thorium radioactive nature (Th-190 Bq/kg; U (in terms of Ra) -120 Bq/kg ; K-150 Bq/ kg).

In soil, there are also some traces of technogenic isotope Cs137 that points to presence of the abundant isotope fallouts after nuclear weapon tests in the atmosphere.

The soil research by the instrumental neutron activation analysis in the Nuclear Geochemical Laboratory of the Geoecology and Geochemistry Department (made by A.F. Sudyko, an analyst) revealed that the content of Th in soil amounts 43.6 g/t, but U-9.2 g/t).

3. Results and discussion

Particular attention is drawn by the high concentration of Th in the rare earth soil (\sum TR = 134.5 g/t), particularly Ce and Nd (see Table 1).

Table 1 The chemical composition of soil Guangdong Province by the instrumental neutron activation analysis

Elements	Ca,%	Na,%	Fe,%	Th	U	Ta	La	Ce	Sm	Nd	Zn
Content, g/t	0.22	0.04	1.6	43.6	9.2	8.4	17.6	89	3.5	14.5	68.6
Elements	Eu	Tb	Lu	Yb	Cr	Au	Hf	Ba	As	Br	Cs
Content, g/t	0.2	1.2	1.1	7.4	18	0.0003	9.6	51	2.1	4.9	9.5

Low content of Ca, Fe, Na and high content of U, Th as well as rare earth elements suggest that the original substrate for soil formation was potassium granite due to the presence of silica relics and K-feldspar in soil.

For further research, silt-loam fraction of the given soil was separated. Its portion amounted 37.4 % of the total sample weight.

Sand soil fraction (of > 0.01mm in size) was subjected to the classic sieve analysis (Fig.1).

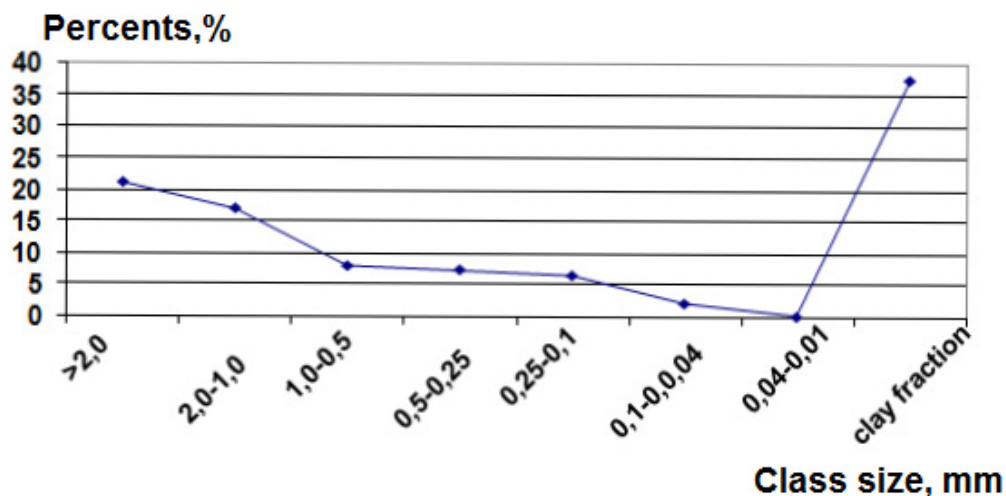


Fig.1 Granulometric composition of the Guangdong Province soil

The fraction of > 2 mm in size is accounted for 21%, and it is presented in the form of quartz intergrowth and feldspar. It was excluded from further chemical and mineralogical study.

Out of separated sand fractions of 1.0-0.5 mm; 0.5-0.25 mm; 0.25-0.1 mm; 0.1-0.04 in size, light and heavy fractions were separated using bromoform. The light fraction comprised all minerals with the specific gravity less than 2.9: quartz, feldspar, colorless mica etc. But the heavy fraction included mostly ore (accessory) minerals. The portion of heavy minerals as a part of sand fraction amounted 0.2%, i.e. 0.05 % of the total soil volume.

The heavy fraction was separated into magnetic, weakly magnetic and nonmagnetic fractions by means of superpowerful Sm-Nd (samarium – neodymium) magnet.

Further mineralogical analysis of those fractions was made with the use of optical (binocular microscope) and electron-microscopic (electron microscope Hitachi S-3400N) research methods.

Isolated grain soil fractions were tested for the presence of radioactive, rare-earth and a number of other elements by the instrumental neutron activation method (Fig.2). The data analysis showed that maximum accumulation of radioactive elements was found in fraction – 0.04 and clay fraction. In addition, maximum accumulation of U was revealed in the fine sand fraction (22.7 g/t), but Th – in the clay fraction (110.4 g/t). Thorium-uranium relationship in them varies from 4.3 to 9, respectively. In fine sand (<0.04) and clay (<0.01) fractions, there was maximum accumulation of rare-earth, Ta and a number of other elements.

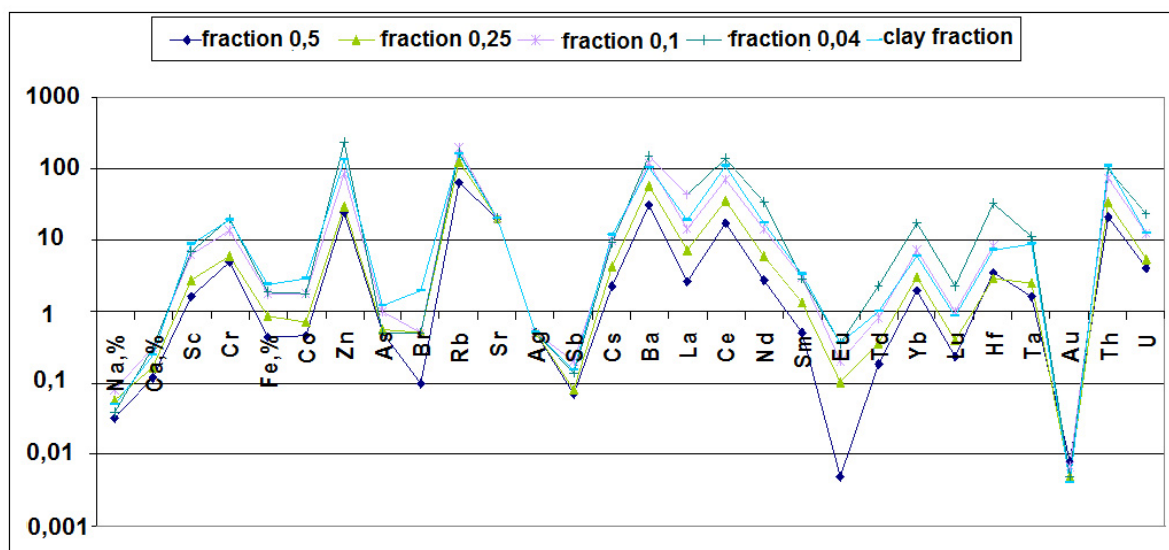


Fig.2. The elemental composition of soil Guangdong Province by the instrumental neutron activation analysis

Therefore, silt-loam soil fraction was subjected to further more detailed study. Its chemical analysis showed that 81.45 % was caused by the presence of Si and Al oxides with the ratio $\text{SiO}_2 : \text{Al}_2\text{O}_3 = 1.5$, whereas the content of Fe oxides amounted 3.63 %. The content of alkaline elements was low and equaled 1.34 %, besides, 1.3% was accounted for by K_2O (see Table 2).

Table 2 Chemical analysis of Guangdong Province soil (%)

Elements	Na_2O	MgO	Al_2O_3	SiO_2	P_2O_5	K_2O	CaO	TiO_2	MnO	Fe_2O_3	BaO	V_2O_5	Cr_2O_3	NiO	LiO
Content, %	0.04	0.16	31.7	49.7	0.02	1.3	0.18	0.38	0.02	3.63	0.01	0.008	0.006	0.005	12.9

The content of organic hydrogen in soil was low, only 0.14 %. 24.6 % of its total amount was accounted for as fulvic acid hydrogen.

The research of mineral composition of silt-loam fraction by the X-ray structure analysis show that kaolinite dominates in it ($\text{Al}_2[\text{OH}_4]\text{Si}_2\text{O}_5$), there are a great deal of hydrargillite ($\text{Al}[\text{OH}]_3$), chlorite and quartz as well as insignificant admixtures of mica, K-feldspar, illite-chloritic aggregates, and hematite.

The given rock can be classified as a ferrallitic soil group of humid tropical and subtropical regions in terms of the set of chemical indicators. The region is characterized by high degree of soil-forming material weathering.

In view of the results of the optical and electron-microscopic analysis in heavy fraction, it was stated that clay particles crooked with fine mixture of hydrargillite, and adhesions of hematite predominate in it. In these aggregates, the unclear mineral phase is visible. In terms of its composition, it represents neodymium and iron oxides (Fe-47.9%, Nd-13.6%, O-35.3%). In some cases, the grains of thorium-containing zircon (Th up to 2.2%), cassiterite are noticeable.

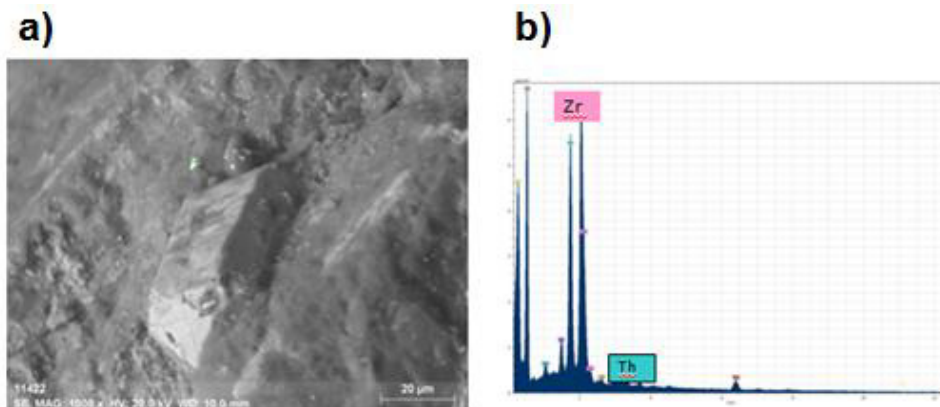


Fig.3 The grains of thorium-containing zircon in heavy fraction
 a) general view
 b) energy dispersive spectrum

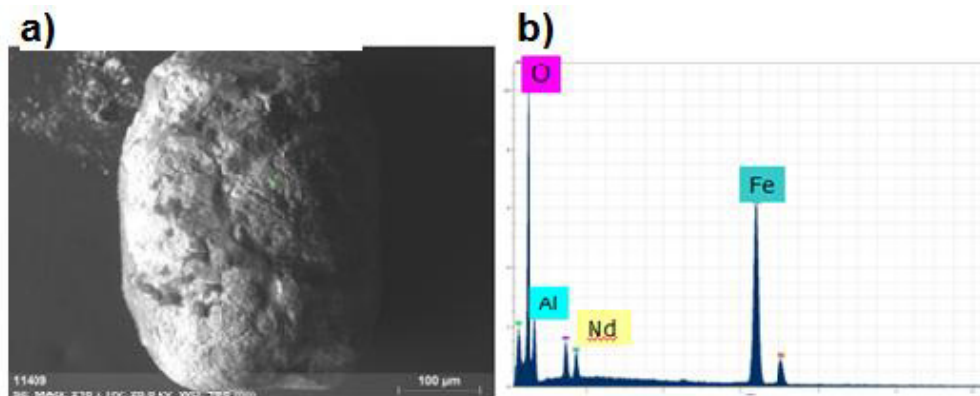


Fig.4 The neodymium iron oxides in heavy fraction
 a) general view
 b) energy dispersive spectrum

In the clay fraction phosphates, heavy and light rare earths, monazite, thorite and rare earth cerium phase with thorium were identified. The most common minerals are iron and titanium oxides, copper and zinc compounds (such as brass), zircon, and barite. Besides, silver gray trace minerals in the form of sulfide silver (it is not possible to state accurately), micromineral formation of bismuth and sulfur dioxide, zirconium (baddeleyite), copper-nickel compound are likely to be present.

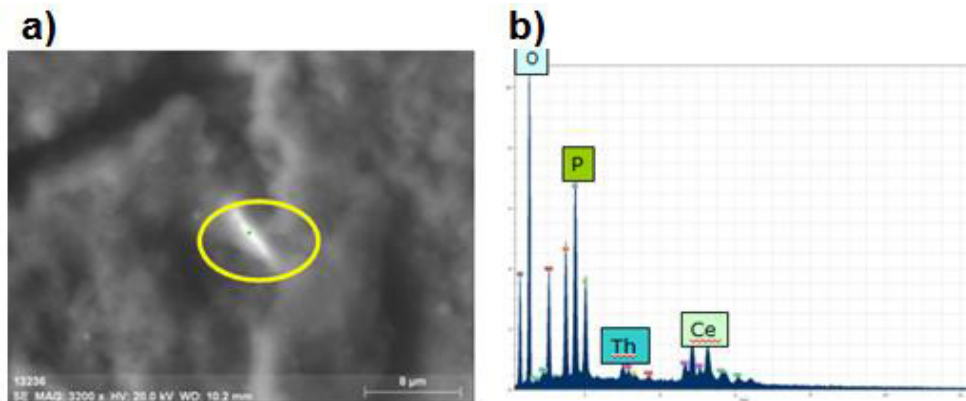


Fig.5 The mineral of monazite in clay fraction

- a) general view
b) energy dispersive spectrum

The leaching of uranium was conducted using various solvents (ammonium carbonate, nitric acid, water) to determine the location of the forms.

A series of experiments with the addition of hydrogen peroxide to ammonium carbonate at various stages of leaching was conducted to identify the nature of the uranium compounds. The samples were examined with the analyzer Fluorat 02-Panorama.

As a result of six boilings with water, not more than 0.24 g/ton of uranium was isolated.

Leaching with addition of nitric acid (HNO₃) was more effective (0.91 g/t). The ammonium carbonate (5% solution of (NH₄)₂CO₃) needs a solution, especially hexavalent uranium. To isolate the tetravalent uranium requires its pre-oxidation, which was carried out by adding a small amount of hydrogen peroxide (H₂O₂) performed at different stages of leaching.

There is low extraction of uranium (0.9 g/t), which was conducted with pure solution of ammonium carbonate only.

On leaching after three boilings with ammonia solution a solution of hydrogen peroxide was added that increased the percentage of leaching significantly. When adding hydrogen peroxide at the beginning of boiling, 1.12 g/ton of uranium are isolated as a result of 1 boiling. Leaching in this way (1.5 g/t) turns out to be the most effective.

4. Conclusion

The experiments in uranium leaching from clay fraction showed that uranium is a part of highly soluble compounds and a part of close connection with the clay component in the form of isomorphic impurity in accessory minerals.

As a current hypothesis, it can be suggested that in our case we are dealing with the sorption concentration mechanism of U, Th, rare-earth elements in kaolinite-gibbsite soil aggregate. In this case one cannot exclude the fact that proper rare-earth nanominerals are formed on this catalytic barrier that can explain the presence of iron - neodymium phase in them. The high natural radioactivity of soils in Chinese Guangdong Province is associated with a high content of radioactive elements in the predominantly fine clay fraction.

The elevated concentration of radioactive and rare-earth elements in the studied soil is likely to be characterized as “ionic” ore type occurring in the territory of China.

In the course of the work it was stated that highly radioactive soil of China had been formed due to deep chemical weathering of highly radioactive potassium granites. High uranium and thorium contents in them are caused by specific conditions of weathering crust formation and subsequent pedogenesis. According to Hiroshige Morishima et al.³ high loads for a human are formed in the field development of such rocks.

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