THE CAUSES OF HIGH NATURAL RADIOACTIVITY IN THE SOILS AND RADIOECOLOGICAL PROBLEMS IN AREAS WITH HIGHLY RADIOACTIVE SOILS

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There are several provinces on the globe that have a high content of natural radioactive elements in soil. Examples are the soils of Minas-Gerais State in Brazil, soils of Kerala State in India, soils of Nuie Island and soil in the South-Chinese Province Guangdong [1].

The character of soil radioactivity varies from pure uranium radioactive (U >Th, which is typical for soil of Niue 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

Table 1. The chemical composition of soil Guangdong Province by the instrumental neuron 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

Table 2. The chemical composition of soil of Auvergne region by the instrumental neuron activation analysis

Elements	Sm	Ce	Ca, %	Lu	U	Th	Cr	Yb	Au	Hf	Ba	Sr	Nd	As	Ag
Content, g/t	2.14	29.3	2.53	0.103	6.44	4.6	11.7	0.8	0.002	2.26	482	30	20.8	24.3	0.5
Elements	Br	Cs	Tb	Sc	Rb	Fe, %	Zn	Ta	Co	Na, %	Eu	La	Sb		
Content, g/t	5.8	96.51	0.33	3.2	443.8	1.19	108.9	7.01	2.19	0.48	0.65	11.3	1.28		

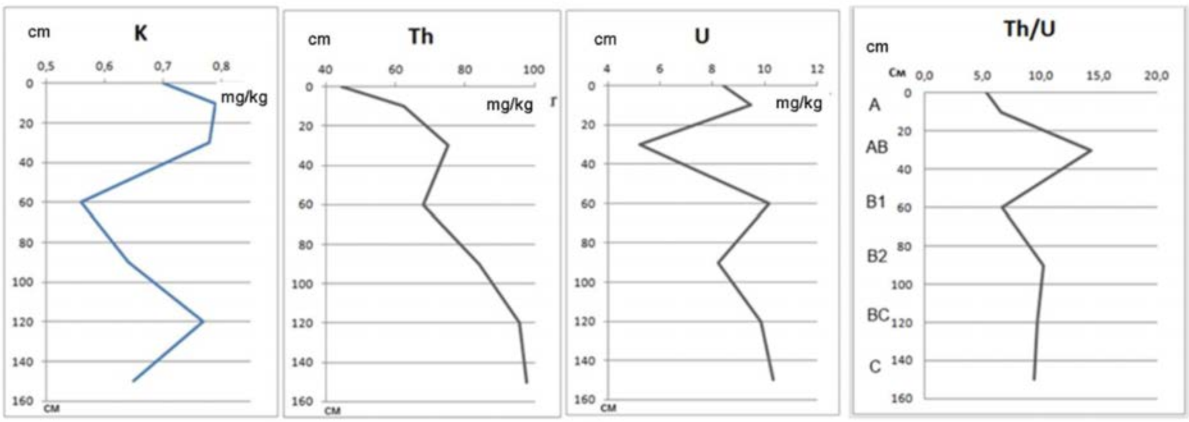


Fig. 1. *The data of measurement by method gamma-ray spectrometry*

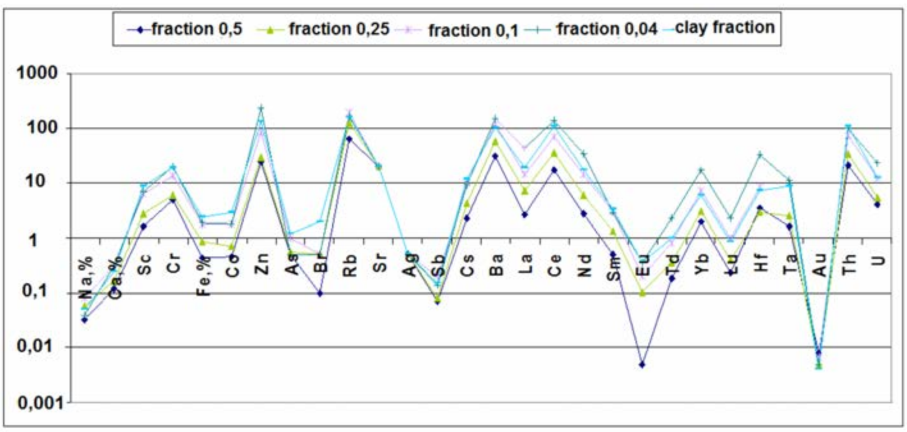


Fig. 2. *The elemental composition of soil Guangdong province by the instrumental neuron activation analysis*

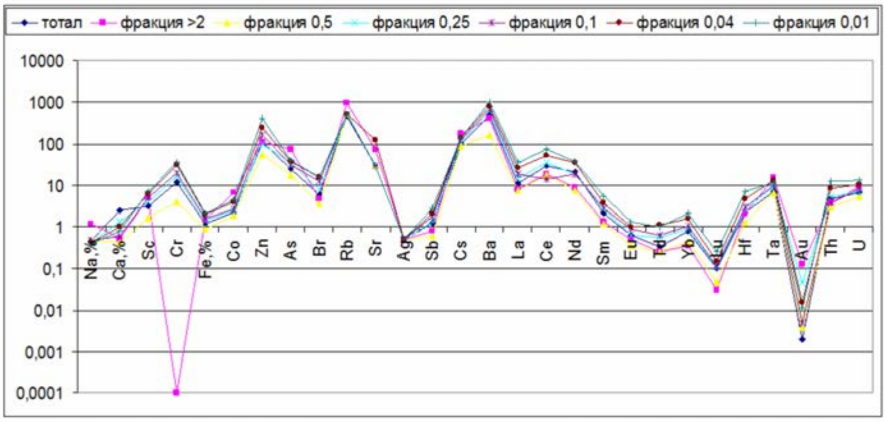


Fig. 3. *The elemental composition of soil of Auvergne region by the instrumental neuron activation analysis*

а)

Element	[wt.%]	[norm. wt.%]	[norm. at.%]	Error in %
Carbon	2,254039	2,830595	6,640686	0,327863
Oxygen	27,25863	34,23107	60,28802	2,924639
Sodium	0,94107	1,181785	1,448499	0,086243
Aluminium	2,370814	2,977241	3,109292	0,144357
Silicon	3,9757	4,992637	5,009128	0,195868
Phosphorus	11,48598	14,42396	13,12215	0,488408
Sulfur	0,016661	0,020922	0,018386	0,002248
Potassium	0,445496	0,559449	0,403197	0,039102
Calcium	0,233436	0,293146	0,206107	0,032494
Yttrium	17,23769	21,64689	6,86087	1,23777
Gadolinium	2,875756	3,611341	0,647131	0,112715
Dysprosium	5,527792	6,941736	1,20373	0,237796
Erbium	2,639268	3,314362	0,55837	0,102745
Ytterbium	2,368929	2,974873	0,484436	0,314859

б) в)

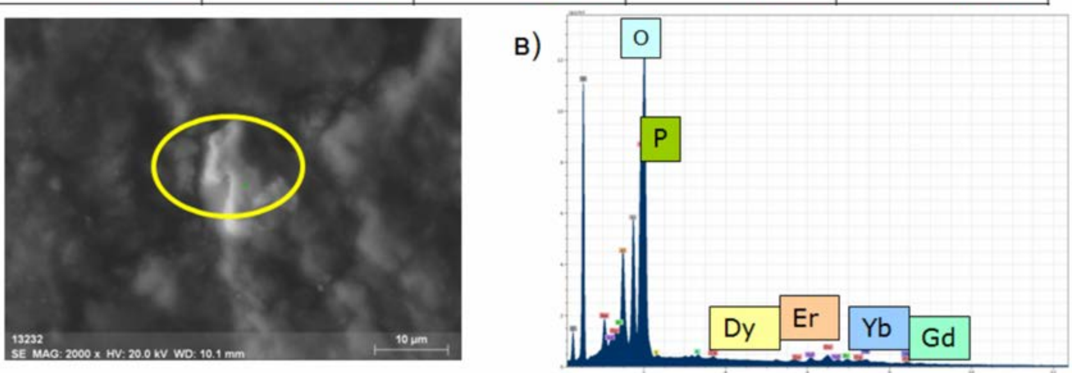


Fig. 4. *The phosphates of heavy and light rare earths in clay fraction (б), energy dispersive spectrum (в) and composition (а) in soil of Guangdong province*

latter, it is suggested that its radioactivity is explained by the presence of monocyte [3].

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 serve.

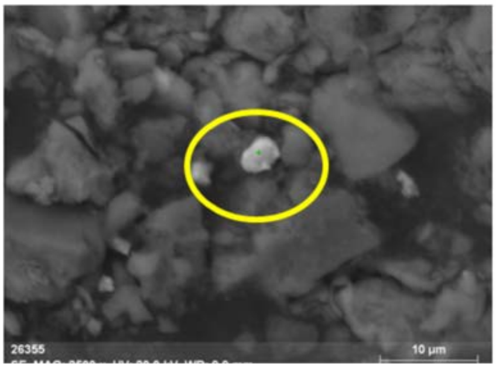
The purpose of the given work is to study the causes of high soil radioactivity in the South-Chinese Guangdong province and in the Auvergne region.

The preliminary gamma-spectrometric soil analysis shows that they are characterized by thorium radioactive

а)

Element	AN	series	Net	[wt.%]	[norm. wt.%]	[norm. at.%]	Error in wt.% (1 Sigma)
Carbon	6	K-series	12815	0	0	0	0
Oxygen	8	K-series	33720	38,092	53,265	77,957	4,664
Aluminium	13	K-series	18358	4,524	6,326	5,490	0,247
Silicon	14	K-series	35828	7,474	10,451	8,713	0,348
Scandium	21	K-series	372	0,199	0,278	0,145	0,038
Iron	26	K-series	3840	3,563	4,982	2,089	0,139
Copper	29	K-series	259	0,413	0,578	0,213	0,055
Zirconium	40	L-series	28157	13,255	18,535	4,758	0,539
Hafnium	72	L-series	771	1,755	2,454	0,322	0,109
Thorium	90	M-series	1497	1,465	2,049	0,207	0,089
Uranium	92	M-series	768	0,775	1,083	0,107	0,065

б)



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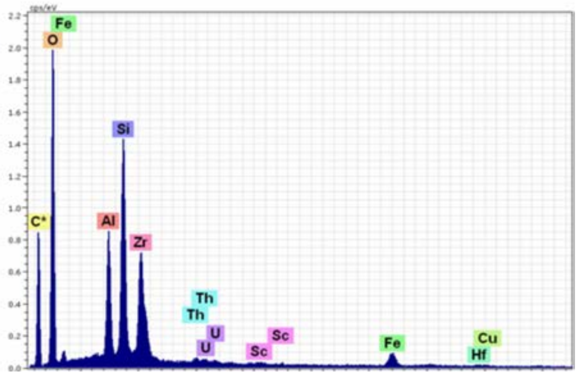

Fig. 5. *The mineral of monacyte in clay fraction (б), energy dispersive spectrum (в) and composition (а) in soil of Guangdong province*

nature (Th-190 Bq/kg; U (in terms of Ra)-120 Bq/kg; K-150 Bq/ kg) in soil of Guandong province.

In soil, there are also some traces of technogenic isotope Cs-137 that points to presence of the abundant isotope fallouts after nuclear weapon tests in the atmosphere.

The Guandong province soil research by the instrumental neuron 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). Particular attention is drawn by the high concentration of Th in the rare earth soil (Σ TR = 134.5 g/t), particularly Ce and Nd (see Table 1). Low contents 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.

As for Auvergne region soil, the operational soil research by the instrumental neuron activation analysis has revealed that the content of U in soil amounts 6,44 g/t, but Th – 4,6 g/t) (Table 2). Low content of Ca, Fe, Na and high content of U, Th as well as rare earth elements suggests that the original substrate for soil formation was two-mica granites.

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 >0.01 mm in size) was subjected to the classic sieve analysis.

Granulometric analysis shows that the C horizon is characterized by increased content of fine fraction (0.04 mm). While increasing horizons (from C to A), the percentage (%) of the fine dust fraction decreases.

According to data of measurement by of method gamma-ray spectrometry (Fig. 1) potassium is an active migrant. Intense leaching of K occurs in the B1 horizon. The distribution of thorium in the soil profile differentiated weaker than the other elements.

However, the existing variations in the distribution of Th allow us to trace the trend of its accumulation in the horizons B2, BC, C closer to the soil-forming rocks. The redistribution of uranium in the soil profile indicates a high mobility of the element in the illuvial-eluvial process. Significant removal of U is observed in the AB horizon and an intense accumulation of U from soil solutions is found in the humus horizon A.

The data analysis of instrumental neuron activation showed that maximum accumulation of radioactive elements was found in fraction – 0.04 and clay fraction of soil from Guangdong province. 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).

а)

Element	AN	series	Net	[wt.%]	[norm. wt.%]	[norm. at.%]	Error in wt.% (1 Sigma)
Carbon	6	K-series	12815	0	0	0	0
Oxygen	8	K-series	33720	38,092	53,265	77,957	4,664
Aluminium	13	K-series	18358	4,524	6,326	5,490	0,247
Silicon	14	K-series	35828	7,474	10,451	8,713	0,348
Scandium	21	K-series	372	0,199	0,278	0,145	0,038
Iron	26	K-series	3840	3,563	4,982	2,089	0,139
Copper	29	K-series	259	0,413	0,578	0,213	0,055
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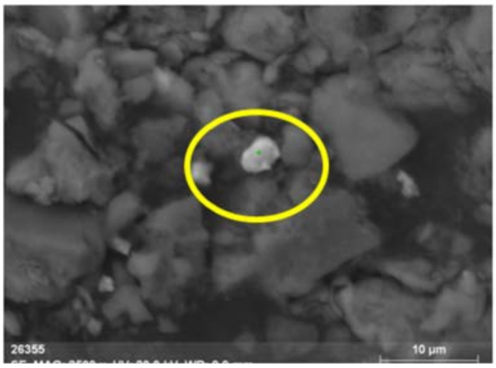
Hafnium	72	L-series	771	1,755	2,454	0,322	0,109
Thorium	90	M-series	1497	1,465	2,049	0,207	0,089
Uranium	92	M-series	768	0,775	1,083	0,107	0,065

б)



в)

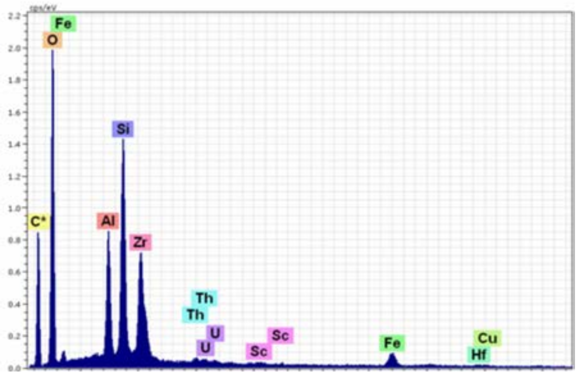


Fig. 6. *The mineral zircon with impurities of thorium and uranium in clay fraction (б), energy dispersive spectrum (в) and composition (а) in soil of Guangdong province*

The data same analysis showed that maximum accumulation of radioactive elements was found in fraction – 0.04 and clay fraction of Auvergne region soil (Fig. 3). In addition, maximum accumulation of U was revealed in the fine sand fraction, but Th – in the clay fraction. Thorium-uranium relationship in them is 0.7.

An important evaluation indicator of soil condition is thorium-uranium ratio. High (>5) thorium-uranium ratio in the soils is characteristic for areas with thorium-bearing geological formations. This is the area found in soils of Guangdong province (the ratio of thorium to uranium in the fractions varies from 4.3 to 9). The given soil 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.

Reduced thorium-uranium ratio has observed in uranium-bearing soils areas. The soils of the region of Auvergne have low thorium-uranium ratio in the range 1–2.4. The region is located within the French Massif Central. There are uranium ore deposits (Limousin, Foretz, Morvan). The deposits are confined to the two-mica granite massifs (San Silvestre et al.) with a high content of uranium, which are located along the fault Argens. The soils of this region are autochthonous.

In view of the results of the optical and electron-microscopic analysis (electron microscope Hitachi S-3400N) of heavy fraction (soil of Guangdong province), it was stated that clay particles crooked with fine mixture of hydrargillite with adhesions of hematite predominating 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%) and cassiterite were noticeable. In the clay fraction phosphates with heavy and light rare earths (Fig. 4), monacyte (Fig. 5), zircon (Fig. 6), xenotime (Fig. 7), 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), 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.

Further mineralogical analysis of fine fractions of soil from Auvergne region was made with the electron-microscopic (electron microscope Hitachi S-3400N) research methods. Barium phosphate, mineral galena were detected, the most common mineral formations were titanium oxides and iron oxides. Uranium-bearing and thorium-bearing minerals were not detected.

According to research by N. Wang Also, there is high

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Element	AN	series	Net	[wt.%]	[norm. wt.%]	[norm. at.%]	Error in wt.% (1 Sigma)
Carbon	6	K-series	98930	0	0	0	0
Oxygen	8	K-series	417508	51,451	58,143	75,971	5,489
Aluminium	13	K-series	317013	11,186	12,641	9,794	0,560
Silicon	14	K-series	322399	9,710	10,973	8,168	0,439
Phosphorus	15	K-series	103487	3,909	4,418	2,982	0,177
Potassium	19	K-series	2820	0,121	0,137	0,073	0,030
Iron	26	K-series	15615	1,430	1,616	0,605	0,065
Copper	29	K-series	2267	0,326	0,369	0,121	0,037
Yttrium	39	L-series	104303	6,832	7,721	1,815	0,296
Dysprosium	66	L-series	8434	1,155	1,306	0,168	0,059
Erbium	68	L-series	4742	0,729	0,824	0,103	0,048
Ytterbium	70	L-series	5358	0,949	1,072	0,130	0,054
Thorium	90	M-series	7494	0,691	0,780	0,070	0,049

б)

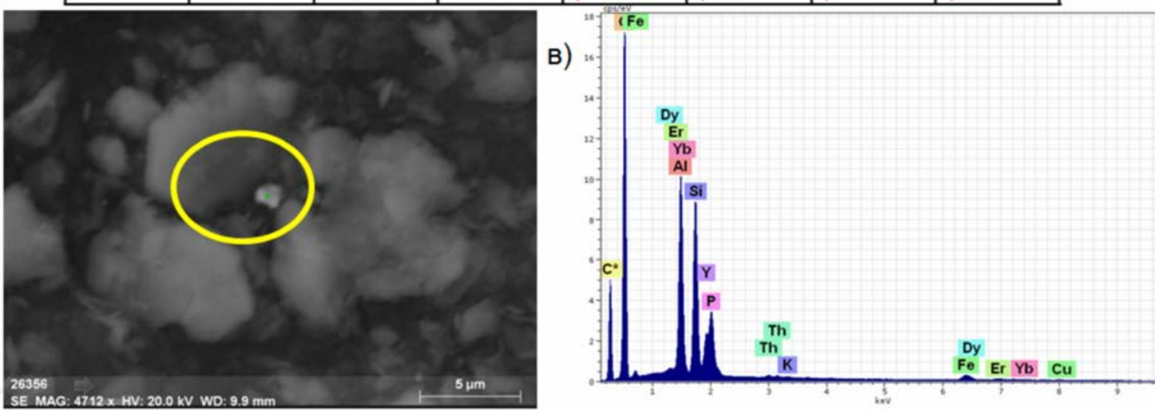

Fig. 7. *The mineral xenotime with impurities of thorium and heavy rare earths in clay fraction (б), energy dispersive spectrum (в) and composition (а) in soil of Guangdong province*

radon hazard in Guangdong province. The distribution of soil-gas ^{220}Rn obviously has geological features. Most sites with high ^{220}Rn concentrations in soil-gas are mainly related to the outcrops belonging to the Jurassic and Cretaceous granite. The average value is 100.41 kBq/m^3 (excludingthe abnormal point), and the maximum reaches 1199 kBq/m^3 [4]. Zhuhai is one of the highest radioactivity background areas in China. The measured highest terrain air-absorbed dose rate at the height of 1m above ground is 266.54 nGy·h^{-1} in Zhuhai, and the average value is 66.28±11.78 nGy·h^{-1} in sediment, 145.90±32.19 nGy·h^{-1} in weathered granite and 145.16±26.33 nGy·h^{-1} in Zhuhai, respectively. It is the reason for the fact that broken fresh granite detritus and weathered granite sands are used as road construction materials and building ground foundation materials [5].

Scientists Gal Frederick, Gadalia Alain noted high concentrations of radon in soil air near city Vichy (Lake Pavin) in Auvergne region. Study in the field of radioecology shows that on the territory of the French Massif Central activity of radionuclides has increased (>150 Bq/m^3) in the houses of the Auvergne region [2].

Conclusion

In the course of the work it was stated that highly radioactive soil of Guangdong province has 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. The concentration of radionuclides in soils depends on their content in the underlying rocks.

The soils of the region of Auvergne have high content of uranium and low thorium-uranium ratio in the range 1–2.4. Uranium ore deposits (Limousin, Foretz, Morvan) are located in Auvergne region. The deposits are confined to the two-mica granite massifs (San Silvestre et al.) With a high content of uranium, which are located along the fault Argens. The soils of this region are autochthonous. As a current hypothesis, it can be suggested that in our case the sorption concentration mechanism of U, Th, and rare-earth elements has revealed in kaolinite-gibbsite soil aggregate. The high natural radioactivity of soils in China's Guangdong Province is associated with the high content of radioactive elements in the predominantly fine clay fraction.

According to electron microscopic analysis of the soils of Guangdong province, mineral - concentrators of rare-earth and radioactive elements are monazite, phosphates of heavy rare earths, xenotime, and zircon.

The elevated concentrations of natural radionuclides in the soils lead to high concentrations of radon, which, in turn causes ecological problems. Scientists have proven negative effects on human health associated with high average doses of radiation. In the areas with increased content of radionuclides chromosomal and genetic changes, leukemia, lung cancer, sarcoma, nasopharyngeal carcinoma etc. can be observed.

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