SOME RECENTLY STUDIED RESULTS ON ENVIRONMENTAL RADIOACTIVITY AND HEAVY ELEMENTS CONTENT IN RARE EARTH MINE AREA AT DONG-PAO, LAI-CHAU

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Abstract

The radioactivity of nuclides and content of heavy elements in soil, water and air are commonly important criteria in the environmental assessment of the mining areas. The study within the years of 2016-2018 on radioactivity and heavy elements content in the water and air samples at Lai Chau Rare Earth project (belonged to Dong Pao rare earth mine area) is to support the environmental survey and assessment program running by environment agency to manage the individually environmental mining area. The total area of rare earth mine is about 11 km² with approximately potential 11.7 million tons of ore concentrate, where geological distribution of ore is un-identical. The implemented project area is located at Ban-Hon and Ban-Giang communes, Tam-Duong District, Lai-Chau Province, which covers 7 ore bodies. The research samples were measured onsite or taken, processed and analyzed according to the Vietnam standards. The results of radioactivity of some nuclides, the concentration of toxic gases such as CO, SO₂, H₂S in air samples and those of heavy elements in soil and water samples at this area were reported according to the Vietnam standards on environment. The results of study contributed to the environmental survey and assessment in this area.

Keywords: Radioactive nuclides, heavy elements, environment, rare earth mine, Dong-Pao

Introduction

The content of heavy metals and environmental radiation is always naturally different from one region to the others. However, humans can still live normally in high radiated background places (100 times more natural radiation than the global average), where some mines existed underground for very long time. Currently, the pollution of air, soil, water and /or radioactive background have been studied commonly and seriously evaluated in many different regions in the world.

Yangjiang in China, Kerala in India, Guarapari in Brazil and Ramsar in northern Iran are among the world's well-known areas with high levels of natural radiation [1 - 4]. Rare earth deposits have been found in the northern Vietnam and are concentrated in the northwestern metallogeny zone [5]. Rare earths present the potential economic opportunities as well as the health and the environmental risks. The results of natural radioactive environment survey in the northwestern Vietnam showed that the gamma radioactive dose varying from 0.2 to 3.0 μ Sv.h⁻¹ [6, 7]. In all rare earth deposits areas the gamma dose level exceeds the safety limit of 0.6 μ Sv.h⁻¹[6]. The results of studies on U, Th and Rn have been conducted in this mountainous area, showing that the gamma radiation levels range from 0.2 to 3.0 μ Sv.h⁻¹; the radiation of Rn and Th are 10 times higher than the IAEA standard [5-9].

Air quality management issues relating to mining are mainly concentrated around the impacts of the particles. These include the dust deposition, and the health impacts associated with PM10 and PM2.5 and the mineralogy and chemical composition of the particles. The sources of dust on mines result mainly from blasting, handling, processing or transporting of

materials, or it can be come from the disturbed areas or the waste disposal facilities, including waste rock and tailings, by wind erosion [10-12].

Water is the earth's most valuable compound that is fundamental for human as well as all other living forms and seems to be inexhaustible. Unfortunately, the outlook for the world's fresh water supply is not very hopeful. Moreover, shortage of fresh water throughout the world can be directly attributed to human misuse in the form of pollution. Water is used for number of purposes like drinking, bathing and washing etc. so it must be free from toxic materials for healthy human and aquatic life. Among surface waters, springs' water is usually considered as safe for drinking. However, springs' water or surface water at the rare earth mining area should be somewhat at risk due to the dissolution of natural radioactive nuclides beside of other components such as pH, odor, turbidity, hardness, TDS, EC, alkalinity, DO, Cl⁻, NO₂⁻, NO₃⁻, SO₄²⁻, heavy metals and microbiological index (total Coliform, Pseudomonas aeruginosa, Enterococcus and Staphylococcus auerus) [13-15].

The present report focus on the radioactivity and heavy elements content in the water and air samples at Lai Chau Rare Earth project (belonged to Dong Pao rare earth mine area). The work was done within the years of 2016-2018 to support the environmental survey and assessment program running by environment agency to manage the individually environmental mining area over the 4 mining bodies (F3, F7, F9 and F10). The total area of rare earth mine is about 11 km² with approximately potential 11.7 million tons of ore concentrate, where geological distribution of ore is un-identical. The implemented project area is located at Ban-Hon and Ban-Giang communes, Tam-Duong District, Lai-Chau Province, which covers 7 ore bodies.

Materials and methods

The study areas

The Dong Pao rare earth mining area (coordinates: $103^{\circ}33$ 'E, $22^{\circ}18$ 'N) is belong to Tam Duong commune, Tam Duong District, Lai Chau province (Fig.1). Dong Pao rare earth mining area has rugged mountainous terrain with heights of 500–2,000 m above-sea-level (ASL). The center of the mine in the south has height of 1,138 m ASL. Mountains have slopes of 40° – 50° , some-times with steep cliff. The topography is in the form of weathering denudation. In the limestone areas like Dong Pao there is also karst topography with caves, karst sinkholes [6,9]



Fig.1 Topographic map of 4 ore area F3, F7, F9, F10 delegation to survey

Symbol of surface water: NM

Air symbol: KK

Radon symbol: R

Chemicals, apparatus and equipment

The DURRIDGE RAD7: is a truly versatile radon and thoron detector.

Optimal 7 – MRU: is a micro-flow infrared sensor; is able to measure continuously simultaneously concentration of CO, CO₂, SO₂, NO, NO_x, H₂S, O₂, temperature, pressure, especially for lower range measurement.

Extech EN300 Environmental Meter: Measures Humidity, Temperature, Air Velocity, Light and Sound.

The CEL712 Microdust Pro: provides real-time measurement and display of airborne dust, fumes and aerosols.

Laboratory equipment such as ICP-MS, AAS, Low background Alpha/Beta counting, Gamma Spectrometry with HGe detector and others: air hot oven, hot plate, balance, etc.,

Water sampling tools, sample container, preserving and other chemical needs.

Chemicals used in the laboratory were at analytical grade.

Measuring technique

The measurements were conducted in the winter-spring time (December to April). The annual average radon concentrations were estimated by averaging measured concentrations in this time period. This could be a reasonable estimation as the area has tropical climate, and, as it was reported in [17], tropical climate areas could have no seasonal changes in radon concentration. The total duration of each study period is one week.

The RAD7 is a sophisticated measuring instrument. During exposure time, the detectors were hung on a stable rod, at the height of 1.5–2 m from the land floor. The machine is setup with 20 cycles and 30 minutes/cycle time. At the end of the exposure time the detector pairs were recovered and transferred to the laboratory for processing.

Monitoring parameters such as noise, temperature, dust, SO_2 , CO_x , NO_x ... are measured by handheld devices at Institute for Technology of Radioactive and Rare Elements. Surface water samples are taken into PE bottles according to Vietnamese standards. Samples are stored according to standards and transported to the laboratory. The criteria of analysis at the area and at the laboratory are evaluated based on Vietnam standards on surface water QCVN 08: 2008/BTNMT.

Result and discussion

1. Radon activity concentrations in air

The measurement was carried out over the 4 rare earth mining bodies. The exposure at each point location was marked as R1, R2...which corresponds the geographic coordinates (X and Y) showed in the below.

	v	V	Average	Rn	Rn	Rn	Sample	
TT	A	I	Dose rate	min	max	TB	code	Location
	coordinate	coordinate	(µSv/h)	(Bq/m^3)	(Bq/m^3)	(Bq/m^3)	Rn	
1	351200	2465976	0,262	35.7	82	59	R1	Road to F7
2	351346	2466313	0,613	71.3	153	112	R2	F7
3	351239	2466440	0,864	215	358	287	R3	F7

Table 1. Geographic coordinates of measurement and sample collection at studied location

	v	V	Average	Rn	Rn	Rn	Sample	
TT	A	I	Dose rate	min	max	TB	code	Location
	coordinate	coordinate	(µSv/h)	(Bq/m^3)	(Bq/m^3)	(Bq/m^3)	Rn	
4	351120	2466592	1,052	251	524	388	R4	F3
5	351175	2466407	0,638	372	412	392	R5	F3
6	351106	2466407	0,682	210	326	268	R6	F3
7	351706	2468032	0,586	143	215	179	R7	F9
8	351450	2468219	0,762	150	185	168	R8	F9
9	351483	2468618	0,542	75	215	145	R9	F9
10	350712	2469390	0,542	175	248	212	R10	F10
11	351548	2468441	0,672	215	258	237	R11	F10
12	350480	2468861	0,611	114	254	184	R12	F10



Fig.2 Rn average activity at the measured positions

The results of the measurements of radon concentrations in the rare earth field of Lai Chau province are given in Fig.2. Position R4, R5 showed the highest radon activity concentration. The mean radon concentration at the measurement points are ranging from 60 to 400 Bq/m⁻³. Comparing to worldwide geometric mean value of 37 Bq/m⁻³ reported in UNSCEAR 2000 about limits in housing [16], the radon level in the Dong Pao is 2 to 10 times higher than the world average.

Some analytical results of representative air and surface water samples at the research area are given in the following Tables 2. In general, the environmental quality of air and water in this area is still good because the analyzed data are under the limit of Vietnam Standards due to the fact that the project has not started to exploit the rare earth ore.

No	Analyte	Unit	KK1	KK2	KK29	KK30	KK45
1	Temperature	⁰ C	11.7	12.3	10.8	11.3	11.2
2	Noise Intensity	dBA	42.1	42	42.3	42.3	41.9.41
3	TSP	mg/m ³	0.058	0.092	0.058	0.062	0.083
4	SO_2	mg/m ³	0.182	0.149	0.116	0.149	0.155
5	CO	mg/m ³	0.213	0.162	0.129	0.162	0.197
6	NO ₂	mg/m ³	0.070	0.070	0.070	0.092	0.099
7	H_2S	mg/m ³	0.01	0.03	0.02	0.01	0.02
8	NH ₃	mg/m ³	0.02	0.01	0.01	0.02	0.01
9	PM _{2.5} Particle	$\mu g/m^3$	27.62	43.81	31.00	29.52	46.00
10	PM ₁₀ Particle	$\mu g/m^3$	12.73	20.19	14.29	13.61	21.20
11	Radioactive Dose	μSv/h	0.528	0.262	0.582	0.521	0.743
12	Sb	$\mu g/m^3$	0.02	0.03	0.03	0.03	0.03

Table 2. Some analytical results on air samples at Rare earth Lai-Chau project area

N <u>o</u>	Analyte	Unit	KK1	KK2	KK29	KK30	KK45
13	As	$\mu g/m^3$	0.001	0.001	0.001	0.001	0.001
14	Cd	$\mu g/m^3$	0.01	0.02	0.02	0.02	0.01
15	Pb	$\mu g/m^3$	0.02	0.01	0.01	0.01	0.01
16	Cu	$\mu g/m^3$	0.21	0.27	0.21	0.27	0.19
17	Zn	$\mu g/m^3$	0.24	0.27	0.12	0.27	0.26
18	HCl	$\mu g/m^3$	0.28	0.39	0.26	0.19	0.22
	Measured pos	sition	F7	Road to F7	F3	F9	F10
	X Axis		351244	351200	351064	351848	350013
	Y Axis	2466357	2465976	2466667	2467901	2469152	

However, the measured dose rate is in Table 1. It could be seen that all inhabitants in the investigated areas may have been exposed to high annual effective doses. Comparing to the worldwide average val-ues of 1.25 mSv.year⁻¹, Dong Pao area may be exposed to geometric mean dose of 20 - 65 mSv.year-1, nearly 20 - 60 times higher than the world average.

2. Surface water analysis

20 samples of surface water were preserved and returned to the laboratory to perform various standard analyzes. The analysis results of some representative samples are shown in Table 3. The results of the whole sample are reported in the analytical results.

Analyte	Unit	NM-01	NM-05	NM-06	NM-10	NM-11
pН		7.27	6.04	6.26	7.38	6.10
BOD5	mg/l	3	12	18	2	5
COD	mg/l	0.64	19.68	0.64	0.48	8
Total suspended solid	mg/l	42	232	244	85	213
Amonia (NH ₄ ⁺)	mg/l	0.14	1.24	13.92	0.11	0.22
Nitrate (NO_3)	mg/l	0.05	1.10	5.90	< 0.05	< 0.05
Nitrite (NO_2^-)	mg/l	0.052	1.10	5.895	< 0.05	< 0.05
Phosphate (PO_4^{3-})	mg/l	< 0.08	< 0.08	< 0.08	< 0.08	< 0.08
Sodium (Na)	mg/l	0.64	0.50	0.43	1.60	0.50
Cloride (Cl ⁻)	mg/l	0.26	6.39	8.67	0.24	0.46
Iron (Fe)	mg/l	0.22	0.52	0.59	0.09	0.08
Manganese (Mn)	mg/l	0.002	5.56	1.58	0.0009	0.01
Aluminum (Al)	mg/l	0.004	0.88	0.61	0.0025	0.41
Zinc (Zn)	mg/l	0.009	0.004	0.002	0.0005	0.0003
Lead (Pb)	mg/l	0.0001	0.009	0.042	0.0003	0.003
Cadmium (Cd)	mg/l	0.0003	0.0001	0.0001	0.00003	0.00002
Nikel (Ni)	mg/l	0.0001	0.0009	0.0002	0.00001	0.00001
Copper (Cu)	mg/l	0.0007	0.003	0.004	0.0002	0.0005
Chromium IV	mg/l	0.002	0.001	0.001	0.001	0.0003
Mercury (Hg)	mg/l	0.0001	0.01	0.01	0.00001	0.0001
Asenic (As)	mg/l	0.0007	0.002	0.006	0.0011	0.0006
Cyanide (CN ⁻)	mg/l	< 0.001	< 0.001	KPH	< 0.001	< 0.001
Floride (F ⁻)	mg/l	0.96	0.48	0.53	1.16	2.49
Sulfate (SO ₄ ²⁻)	mg/l	3.42	13.46	41.98	47.63	2.28
Bromide (Br)	mg/l	< 0.06	< 0.06	0.19	< 0.06	< 0.06
Coliform	MPN/100ml	0	39	49	22	23
E- Coli	MPN/100ml	121	572	680	182	150

 Table 3. Analytical results of representative surface water samples

Analyte	Unit	NM-01	NM-05	NM-06	NM-10	NM-11
Total Phenolic	mg/l	0.001	< 0.001	< 0.001	0.001	0.001
Total α radioactivity	Bq/l	0.3	0.34	0.19	0.36	0.28
Total β radioactivity	Bq/l	0.39	1.07	1.85	0.53	0.5

The results of analyzing some representative surface water samples recorded in Table 3 show that, due to the impact of hilly and mountainous terrain, the dry and cold season and the operation of the plant are still very limited, so the quality of surface water in the points Sampling is generally quite clean, except for some points (water in small pits and pools) with higher ammonium content and biochemical parameters than the threshold of QCVN 08: 2008 / BTNMT due to people's habit. Local livestock and poultry grazing, and domestic wastewater of people and animals flow into (sample NM05, NM06).

However, the survey area of the working group is a relatively rich area of rare earths which contains a significant amount of Th and radioactive nuclei (ore bodies F3, F7, F9, F10), so the total discharge activity value Alpha and Beta radioactivity have a sample of conventional water environment (samples MN01, MN05, MN10, MN11 in table 2). This can be explained by the "bandits" exploitation in the F3 ore bodies is very rampant, cannot be prevented, because the rain washes the ore on the surface to stream branches, puddles and water holes, to the season dryness remains, creating water areas with high risk of contamination because rare earth minerals contain a certain amount of radioactive nuclei (Th, U, Ra). In general, the collected and analyzed surface water samples were able to see the radioactivity of some representative surface water collected in early days of the year 2018.

	U-238	Th-232	Ra-226
LOD	0.18	0.36	0.12
NM - 03	0.42	0.42	0.34
NM - 13	0.30	0.36	0.26
NM - 18	0.38	0.36	0.30
NM - 23	0.40	0.38	0.34
NM - 32	0.21	0.36	0.14
NM - 35	0.19	0.36	0.20

Table 4. Radioactivity of representative surface water samples (Bq/L)

(Water sample was pre-concentrated at least 5 times; Measurement on Gamma spectrometry HGe Detector-TCVN 7175:2011, ISO 10703:2007)

Conclusion

The study was done within the period of years 2016-2018 on 4 rare earth mining bodies. The quality of air and surface water at the survey area (four mining fields as F3, F7, F9 and F10), where the Lai Chau rare earth processing plant would be located seemed rather good. All criteria were in the limit of Vietnam standards on air (QCVN 05:2013/BTNMT, 06:2009/BTNMT, 26:2010/BTNMT) and surface water (QCVN 08: 2008).

The further survey is necessary for the monitoring the air quality and the assessment about the impact of industrial activity on the environment at that area should be continually done in order to protect the environment from the high risk on pollution, which would be caused by the human activity.

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MỘT SỐ KẾT QUẢ NGHIÊN CỨU HIỆN TRƯỜNG PHÔNG PHÓNG XẠ VÀ HÀM LƯỢNG NGUYÊN TỐ KIM LOẠI NẶNG TẠI KHU VỰC MỎ ĐẤT HIẾM ĐÔNG PAO-LAI CHÂU

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Tóm tắt

Hoạt độ các nhân phóng xạ và hàm lượng một số nguyên tố kim loại nặng trong đất, nước và không khí là những tiêu chí quan trắc quan trọng trong đánh giá tác động môi trường của các khu vực khai khoáng nói chung. Nghiên cứu hiện trường trong giai đoạn 2016-2018 về nồng độ hoạt độ các nhân phóng xạ và hàm lượng các nguyên tố kim loại nặng trong mẫu nước và không khí ở khu vực dự án đất hiếm Lai Châu (thuộc mỏ đất hiếm Đông Pao) nhằm đóng góp vào công tác giám sát, đánh giá các điều kiện môi trường trong quá trình quản lý môi trường từng khu vực thuộc mỏ. Toàn bộ khu vực mỏ chứa đất hiếm rộng khoảng hơn 11 km² với trữ lượng khoảng 11,7 triệu tấn tinh quặng, phân bố địa chất không đồng nhất. Khu vực triển khai dự án nằm tại 2 xã Bản Hơn và Bản Giang, huyện Tam Đường, tỉnh Lai Châu, gồm có 07 thân quặng. Các mẫu nghiên cứu được đo tại hiện trường, được thu gom, xử lý và phân tích theo các quy chuẩn, tiêu chuẩn hiện hành. Kết quả xác định nồng độ hoạt độ một số kim loại nặng trong mẫu đất, nước tại khu vực này cũng được báo cáo, căn cứ các quy chuẩn Việt Nam về môi trường. Kết quả nghiên cứu đã đóng góp vào công tác đánh giá giám sát hiện trạng môi trường tại khu vực này.

Một số kết quả đo hoạt độ Rn trung bình tại khu vực khảo sát được biểu diễn trong hình 1.