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Studying airborne trace elements in coastal areas of South Central Vietnam using moss and INAA techniques

Điều tra nguyên tố vết trong không khí bằng kỹ thuật rêu và phân tích kích hoạt neutron ở một số khu vực thuộc duyên hải miền Trung Việt Nam

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Abstract

Thirteen moss samples were collected in special geological and economical cities and townships in the South Central region in Vietnam (Quang Nam and Khanh Hoa provinces). Twenty-four elements were determined in the moss samples by Neutron Activation Analysis at the reactor IBR-2 in the Joint Institute for Nuclear Research in Dubna, Russia. The analysis results of moss samples are partly reflected the air quality in the research locations. Our study is aimed to investigate air pollution and identify its sources in order to find a suitable process for improving the environment in the studied areas in Vietnam.

Keywords: air pollution; Vietnam; moss technique; neutron activation analysis.

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Tóm tắt

Mười ba mẫu rêu đã được thu thập tại các thành phố, huyện có nét đặc trưng về vị trí địa lý, hoạt động kinh tế thuộc vùng Nam Trung Bộ Việt Nam (thuộc tỉnh Quảng Nam và Khánh Hòa). Hai mươi bốn nguyên tố đã được xác định trong các mẫu rêu bằng kỹ thuật phân tích kích hoạt neutron tại lò phản ứng IBR-2 thuộc Viện Liên hiệp Nghiên cứu hạt nhân Dubna, Liên bang Nga. Kết quả phân tích hàm lượng các nguyên tố trong các mẫu rêu đã phần nào phản ánh chất lượng không khí tại các địa điểm nghiên cứu. Nghiên cứu của chúng tôi nhằm mục đích điều tra và xác định nguồn gốc gây ô nhiễm không khí để tìm ra quy trình phù hợp nhằm cải thiện môi trường tại các khu vực nghiên cứu ở Việt Nam.

Từ khóa: ô nhiễm không khí; Việt Nam; kỹ thuật rêu; phân tích kích hoạt neutron.

1. Introduction

In several decades, the increase in environmental pollution is associated with industrial development and population growth. One main form of pollution is air pollution, which has caused different diseases in the community. Therefore, assessing air pollution, in the form of particles and gases in the air, is one of the most important and urgent tasks in Vietnam today.

Moss technique has been widely used for monitoring atmospheric deposition in Europe for a long time. However, the moss technique is a fairly new application in Vietnam, and the moss species used are different from the ones used in Europe. Moss lacks a true root system and obtains nutrients directly from the atmosphere. Moss has good ability for bioaccumulation of heavy metals and the moss leaves are highly permeable to ions of trace elements and mineral salts. Therefore, moss is used effectively to study air deposition of metals as well as other trace elements [1, 2]. The recommended moss species in Europe are *Pleurozium schreberi*, *Hylocomium splendens*, and *Scleropodium purum* [3]; however, these species are not found or are rare in Vietnam. The local epiphytic moss *Barbula*, which is widely distributed across Vietnam, tropical Asia [4] and other tropical and sub-tropical regions [5], has been used in our study. We have compared the accumulation ability of chemical elements in mosses, and *Barbula s.p* is chosen as appropriate indication to bio-monitoring air pollution in Vietnam [6, 7].

Previously, there have been several programs using different methods to study atmospheric environment in Vietnam [8, 9, 10], including the moss technique of the authors [11, 12, 13]. The moss technique is a low cost method for airborne trace elements investigation.

Our group is using the moss technique to investigate air pollution in featured coastal areas of South Central Vietnam, such as Khanh Hoa and Quang Nam provinces as described in this work.

2. Materials and methods

2.1. Sampling areas

Thirteen samples of *Barbula s.p* moss were used to study air pollution in featured areas in the South Central regions of Vietnam. Seven samples were collected in Hoi An city (A1) and Dien Ban town (A2) in Quang Nam province, and six samples were collected in the coastal Nha Trang city (B1) and Khanh Vinh district (B2) in Khanh Hoa province. The sampling was carried out in the end of the rainy season in each region. Each moss sample was collected from 5 to 10 locations in an area of 1kmx1km. The sampling distance was about 3 kilometers in the inner cities, and about 6-10 kilometers in their vicinities (Dien Ban and Khanh Vinh). NhaTrang is a tourist city; besides, fishing and shipping have contributed significantly to the local economy. There are different deposits with medium reserve of minerals and construction materials. Granite and sand are the two most exploited minerals. Hoi An is a small ancient tourist city with a lot of the traditional craftwork villages, such as bronze casting, pottery, carpentry and leather works; the urbanization level is low. Table 1 displayed the information of moss samples.

Table 1. Information of the areas for collecting *Barbula s.p* moss samples

	Area	Symbol	Position	Collection place	Latitude	Longitude
1	Quang Nam Province	A	A1	Hoi An city	15°53′01″N 15°54′20″N	108°17′32″E 108°19′12″E
			A2	Dien Ban Town	15°52′50″N 15°53′19″N	108°18′51″E 108°22′05″E
2	Khanh Hoa Province	B	B1	Nha Trang city	12°18'05″N 12°24'20″ N	109°17'30″E 109°19'44″ E
			B2	Khanh Vinh district	12°18'85″N 12°25'91″N	108°81'51″E 108°91'69″3E

2.2. Moss sampling and preparation

Barbula sp. moss forms clusters or mats of short plants about 0.5–2 cm tall, with unbranched sterm; and the moss leaves occur all around each stem. In this form, its upper part can be easily detached from its base and the substrate influence could be minimized. Therefore, *Barbula sp.* is the most suitable type of moss used for the air deposition survey in the entire country of Vietnam.

Moss was cut out from its carpets using plastic tools and put in polyethylene zip-lock bags. Both the tools and bags were made from low impurity materials. The samples were cleaned to remove extraneous materials (large soil particles, leaves, etc) and then thoroughly washed with distilled water and only the green top part of moss was used for study. Finally, the samples were dried to constant mass before being prepared for analysis. The moss sampling and preparations were executed in accordance with the protocols [3].

2.3. Analytical method

The Instrumental Neutron Activation Analysis (INAA) used to determine elemental concentrations of the samples was executed in the sector of Neutron Activation Analysis and Applied Research of Frank Laboratory of Neutron Physics (FLNP) – Joint Institute for Nuclear Research (JINR) in Dubna, Russia. Sample irradiation was carried out in channels equipped with the pneumatic system installed at the IBR-2 pulsed nuclear reactor of FLNP with the average power of 2 MW. The irradiation channels are used on beamlines B1 and B2. The thermal neutron flux density in beamline B2 is about $2x10^{12}$ $cm^{-2}s^{-1}$. The second irradiation beamline B1 has a cadmium casing, so irradiation is mainly implemented using resonant and fast neutrons. The flux density of resonant neutrons in beamline B1 is about $1.5x10^{11}$ $cm^{-2}s^{-1}$. As a rule, beamline B2 is used for short irradiation (SLI), and B1 for long irradiation (LLI).

The samples were irradiated two times in different conditions to determine short- and long- lived isotopes. About 0.3 g of dry-weight material of each sample was heat-sealed in a polyethylene bag, or packed in aluminum cup, used respectively for short-term irradiation or for long-term irradiation. For the short-term irradiation, each sample was irradiated for 3 minutes and measured for 15 minutes after 3 to 5 minutes of decay. For the long-term irradiation, the samples were irradiated for about 3 days. After irradiation, they were repacked and measured twice: the first time after 4 days of decay they were measured for 30 minutes. The second time, after 20 days of decay, they were measured for 1.5 hours [14, 15, 16].

The Ge (Li) or HPGe detectors with high resolution were used to measure the gamma spectra of the studied samples. The software Genie 2000 was used to store, display, and analyze the gamma spectra. Other softwares developed at FLNP were employed to determine elemental concentrations of the samples. Calculation of the element concentrations was done with the support of the standard reference materials produced by International Atomic Energy Agency and National Institute of Standards and Technology [16].

3. Results and discussion

The concentrations of twenty-four elements were determined in the *Barbula sp.* moss samples by INAA with the relative error from 3 to 15%, include: Na, Mg, Al, Cl, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Zn, As, Se, Br, Sr, Sb, Ba, La, Th, U. The concentrations of elements in the moss samples in Quang Nam, Khanh Hoa were displayed in Tables 2 and 3, respectively.

Table 2. The elements concentration in *Barbula sp.* moss collected in Quang Nam province

Element	Hoi An city				Dien Ban Town		
	A1_1	A1_2	A1_3	A1_4	A2_1	A2_2	A2_3
Na	536 ± 16	1510 ± 44	2420 ± 71	1150 ± 34	833 ± 25	1440 ± 42	1270 ± 38
Mg	2068 ± 65	1984 ± 59	2730 ± 82	1627 ± 51	1089 ± 37	1075 ± 34	1362 ± 44
Al	1700 ± 48	6150 ± 171	7790 ± 217	3210 ± 89	1900 ± 54	7370 ± 206	2090 ± 59
Cl	3270 ± 282	2100 ± 182	2320 ± 201	4000 ± 344	1270 ± 110	1450 ± 126	1800 ± 156
K	18500 ± 1535	15800 ± 1589	14800 ± 1656	25100 ± 2083	8880 ± 856	12000 ± 1228	13200 ± 1473
Ca	9770 ± 777	16400 ± 1266	30300 ± 2314	10700 ± 839	9160 ± 729	9330 ± 746	6720 ± 544
Sc	0.439 ± 0.024	1.39 ± 0.074	1.9 ± 0.101	0.688 ± 0.038	0.792 ± 0.043	1.44 ± 0.076	0.516 ± 0.029
Ti	205 ± 24.6	305 ± 32.4	465 ± 49.0	186 ± 22.1	92.4 ± 13.3	368 ± 38.5	123 ± 15.9
V	4.12 ± 0.346	9.76 ± 0.775	44.70 ± 3.473	5.11 ± 0.419	3.39 ± 0.288	11.80 ± 0.937	3.97 ± 0.33
Cr	4.77 ± 0.456	15.60 ± 1.991	15.40 ± 1.689	4.66 ± 0.696	6.73 ± 1.112	13.10 ± 1.691	3.80 ± 0.629
Mn	98.2 ± 3.32	87.5 ± 2.96	112 ± 3.82	45.7 ± 1.57	29 ± 1.01	89.7 ± 3.03	34.9 ± 1.22
Fe	4920 ± 226	5140 ± 241	5270 ± 249	2190 ± 109	1920 ± 95	4810 ± 224	1580 ± 77
Co	0.73 ± 0.044	3.81 ± 0.22	2.44 ± 0.143	0.837 ± 0.051	0.876 ± 0.053	1.8 ± 0.106	1.01 ± 0.06
Ni	2.25 ± 0.37	6.15 ± 0.69	7.65 ± 0.89	2.85 ± 0.29	2.67 ± 0.42	4.82 ± 0.62	2.47 ± 0.25
Zn	357 ± 7.07	254 ± 5.56	249 ± 5.53	471 ± 10.03	144 ± 3.25	456 ± 9.76	183 ± 4.06
As	1.29 ± 0.065	4.96 ± 0.248	11.6 ± 0.58	2.97 ± 0.149	2.01 ± 0.101	4.63 ± 0.232	2.76 ± 0.138
Se	0.637 ± 0.044	0.958 ± 0.186	0.786 ± 0.159	0.602 ± 0.121	0.673 ± 0.136	0.696 ± 0.138	0.817 ± 0.158
Br	6.43 ± 0.176	19.5 ± 0.53	50.1 ± 1.363	7.69 ± 0.21	9.04 ± 0.247	7.37 ± 0.201	6.95 ± 0.19
Sr	27.5 ± 2.01	53.6 ± 3.75	100 ± 6.77	29 ± 2.22	24.3 ± 1.85	40.9 ± 2.99	27.2 ± 2.50
Sb	0.532 ± 0.051	1.57 ± 0.152	1.2 ± 0.115	0.819 ± 0.08	0.886 ± 0.086	1.59 ± 0.154	0.817 ± 0.080
Ba	24.4 ± 1.34	96.7 ± 6.11	127.0 ± 7.99	53.3 ± 3.38	34.2 ± 2.21	82.7 ± 5.19	95.8 ± 6.00
La	0.909 ± 0.038	4.69 ± 0.185	7.9 ± 0.311	2.24 ± 0.09	1.59 ± 0.065	5.38 ± 0.211	1.37 ± 0.057
Th	0.388 ± 0.021	1.9 ± 0.098	3.45 ± 0.177	0.861 ± 0.045	0.555 ± 0.029	2.07 ± 0.107	0.56 ± 0.029
U	0.142 ± 0.006	0.538 ± 0.036	2.21 ± 0.148	0.233 ± 0.016	0.171 ± 0.012	0.63 ± 0.042	0.187 ± 0.013

Specifically, the moss sample collected at location A1_3 (near Cua Dai beach) has the highest value for most elemental contents including: Br, Ca, Al, Mg, Ni, Cr, Sr , Ti, Mn, Fe, V, Sc, Cs, Ba, La, Ce, Sm, Gd, Tb, Ta, Th and U. In particular, the high conectration element of As (11.6 mg/kg) is different from the remaining locations. The maximum concetration of Co, Se, Cr elements were found in the moss sample A1_2 and the second largest included elements Cd and Sb; A1_2 location is at the intersection of Ba Trieu - Hai Ba Trung - Tran Hung Dao streets, where a large number of tourist vehicles circulate. The minimum concentration of almost all of the elements was found at A1_1, the location is far from the center of Hoi An ancient town (low population density). Moss samples A2_1, A2_2 and A2_3 were collected at locations in Dien Ban town, where there are sparsely populated areas, mainly growing crops by households. The results of most concentrated factors are also reflected at a low level in these samples, except that the concetrations of heavy metal elements Si, Sb (max), and Zn, Cr, Ni were also high in moss samples collected at location A2_2.

Table 3. The element concentration in *Barbula sp.* moss collected in Khanh Hoa province

Ele-ment	Nha Trang city				Khanh Vinh district	
	B1_1	B1_2	B1_3	B1_4	B2_1	B2_2
Na	697 ± 35	1000 ± 50	748 ± 37	945 ± 47	2240 ± 112	428 ± 21
Mg	1573 ± 94	1366 ± 82	2772 ± 139	1577 ± 95	1774 ± 88=9	1472 ± 74
Al	6690 ± 268	12000 ± 480	6450 ± 258	2800 ± 112	34600 ± 1384	18900 ± 756
Cl	162 ± 14.6	206 ± 18.5	458 ± 36.6	2900 ± 232.1	1090 ± 87.2	457 ± 36.6
K	7370 ± 663	8820 ± 793	6940 ± 625	13100 ± 1179	10200 ± 918	6390 ± 575
Ca	13400 ± 1474	8550 ± 940	30600 ± 3366	29300 ± 3223	8500 ± 935	12100 ± 1331
Sc	0.76 ± 0.023	1.18 ± 0.035	1.10 ± 0.033	0.30 ± 0.009	6.70 ± 0.201	2.30 ± 0.069
Ti	231 ± 23.1	286 ± 28.6	475 ± 38.1	153 ± 16.8	1750 ± 122.5	724 ± 57.9
V	4.4 ± 0.26	3.7 ± 0.26	11 ± 0.55	6.3 ± 0.32	39 ± 1.95	14.3 ± 0.86
Cr	3.46 ± 0.35	3.7 ± 0.41	16.4 ± 0.98	2.77 ± 0.30	22.5 ± 1.80	8.3 ± 0.91
Mn	96 ± 3.8	124 ± 4.9	180 ± 7.2	113 ± 4.5	187 ± 7.5	525 ± 21.0
Fe	2130 ± 106.5	2400 ± 120	5530 ± 276.5	1040 ± 52	17600 ± 880	7460 ± 373
Co	0.69 ± 0.02	0.85 ± 0.03	1.8 ± 0.05	0.49 ± 0.02	6.1 ± 0.18	7.2 ± 0.22
Ni	2.06 ± 0.29	4.85 ± 0.44	8.5 ± 0.51	2.27 ± 0.25	10.5 ± 0.74	5.9 ± 0.47
Zn	84 ± 4.2	229 ± 11.5	301 ± 15.1	39 ± 1.9	200 ± 10.0	70 ± 3.5
As	1.64 ± 0.08	1.80 ± 0.09	2.10 ± 0.11	3.50 ± 0.18	12.80 ± 0.51	14.80 ± 0.74
Se	0.215 ± 0.026	0.120 ± 0.014	0.203 ± 0.024	0.124 ± 0.015	0.176 ± 0.021	0.110 ± 0.045
Br	9.8 ± 0.3	6.8 ± 0.2	15.0 ± 0.5	9.2 ± 0.3	3.3 ± 0.1	24.0± 0.7
Sr	200.0 ± 16.0	64.0 ± 5.1	76.0 ± 6.1	241.0 ± 19.3	74 .0± 5.9	43.0 ± 3.4
Sb	0.23 ± 0.01	0.49 ± 0.03	0.78 ± 0.05	0.22 ± 0.01	1.37 ± 0.08	0.40 ± 0.02
Ba	31.4 ± 1.9	42.0 ± 2.5	60.0 ± 3.6	45.0 ± 2.7	131.0 ± 6.6	107.0 ± 5.4
La	3.80 ± 0.11	4.90 ± 0.15	4.25 ± 0.13	1.37 ± 0.06	16.70 ± 0.50	13.80 ± 0.41
Th	3.01 ± 0.12	8.60 ± 0.26	1.73 ± 0.07	0.47 ± 0.02	11.00 ± 0.33	7.40 ± 0.22
U	1.43 ± 0.06	2.10 ± 0.08	0.97 ± 0.04	0.42 ± 0.02	3.30 ± 0.10	3.10 ± 0.12

These mosses were collected in 2 areas of Khanh Hoa province. The samples at Nha Trang City (area 1) includes B1_1, B1_2, B1_3, B1_4; and at Khanh Vinh district (area 2) includes B2_1 (near National Highway 27C) and B2_2.

The analysis results shown that the range concentration of many toxic, heavy metal and group rare-earth elements in area 2 is higher than area 1. There were arsenic (12.8 – 14.8 mg/kg), (1.8 – 3.5 mg/kg); vanadium:(14.3 – 39 mg/kg), (3.7 – 11 mg/kg); chromium (8.3 –22.5 mg/kg), (2.77 – 16.4 mg/kg); nickel (5.9 – 10.5 mg/kg), (2.06 – 8.5 mg/kg); cobalt (6.1 – 7.2 mg/kg), (0.49 – 1.8 mg/kg); lathanum (13.80 – 16.70 mg/kg), (1.37 – 4.90 mg/kg), thorium (7.4 – 11.0 mg/kg), (0.47 – 8.60 mg/kg), uranium (3.10 – 3.30 mg/kg), (0.42 – 2.10 mg/kg), respectively.

In area 1, almost the concentrations of elements such as Na, Al, Si, Sc, Ti, V, Cr, Fe, Ni, Rb, Zr, Sb, Cs, Ba, La, Ce, Nd, Sm, Eu, Gd, Th and U were determined minimum in moss samples collected at the locations B1_1, B1_2 and B1_4. The moss sample was collected at location B1_3 near the coast and the city center (Loc Tho ward) ; the concentration of Mg, Ca, Zn and Sn elements was determining maximum and the second highest concentration was the bromine (Br) element. The air quality in Nha Trang city is good [17].

However, the analysis results of samples in area 2 were interesting. The maximal concentrations of elements Al (34600 mg/kg), Fe (17600 mg/kg), V (39 mg/kg), Ti (1750 mg/kg) were found in the sample collected at the site B2_1. The main source of these elements can be considered to be from the exploitation of the heavy-mineral ilmenite and the deposits for brick clay in the area. The Al_2O_3 is one of the main components of clay bricks, and $FeTiO_3$ is one of the main components of ilmenite [18, 19]. In addtion, maxim concentration group rare-earth elements were also found at B2_1. There were namely La (16.7 mg/kg), Th (11.0 mg/kg) and U (3.3 mg/kg) and B2_2 such as La (13.8 mg/kg), Th (7.4 mg/kg) and U (3.1 mg/kg). Source of these elements is related to bauxit and Rhyolite [20, 21, 22]. The analysis results reflect the current situation in Vietnam, which is mining deposits with little reserves that are scattered over the country have not been strictly controlled. This is an important source of air pollution around where the activity takes place.

The range (min and max) and median concentrations of some elements in the moss samples in our work and Ha Noi city [11] were displayed in Table 4.

Table 4. Minimum, maximum and median concentrations of elements in Quang Nam, Khanh Hoa and Ha Noi city

Element	Our work						Nguyen et al (2010)		
	Quang Nam (7)			Khanh Hoa (N=6)			Ha Noi (N=12)		
	Min (mg/kg)	Max (mg/kg)	Median (mg/kg)	Min (mg/kg)	Max (mg/kg)	Median (mg/kg)	Min (mg/kg)	Max (mg/kg)	Median (mg/kg)
Mg	1075	2730	1627	1366	2772	1575	929	9162	3838
Al	1700	7790	3210	2800	34600	9345	**754**	**24281**	**9911**
Cl	***1270***	***4000***	***2100***	162	2900	458	195	4003	1368
K	***8880***	***25100***	***14800***	6390	13100	8095	1809	36463	11881
Ca	6720	30300	9770	***8500***	***30600***	***12750***	9773	38295	17877
Ti	92	465	205	***153***	***1750***	***381***	195	1309	676

Cr	3.80	15.60	6.73	2.77	22.50	6.00	**4.23**	**51.03**	**27.98**
Mn	29	112	88	96	525	152	95.42	370	137
Fe	1580	5270	4810	1040	17600	3965	**1622**	**11057**	**6127**
Ni	2.25	7.65	2.85	2.06	10.5	5.38	0.12	8.79	4.40
Zn	144	471	254	39	301	142	**115**	**1576**	**285**
As	1.29	11.6	2.97	1.64	14.8	2.8	**0 .96**	**79.25**	**7.89**
Br	6.43	50.11	7.69	3.30	24.00	9.50	**4.45**	**207.38**	**9.91**
Sr	24.3	100	29	43	241	75	**53.36**	**610.06**	**147.09**
Ba	24.4	127	82.7	31.4	131	52.5	**187.60**	**4930.06**	**1267.95**

Table 4 shows that the range (min, max) and median concentrations of elements Cl and K in Quang Nam are higher than in Khanh Hoa. These elements were related to typical fertilizers [23]. It has reflected that the featured tourism of Quang Nam was herbs, spices and vegetables villages. However, the range and median concentrations of featured beach elements which are Ca, and Ti in Khanh Hoa were higher than in Quang Nam.

The range and median concentrations of elements Al, Ti, Cr, Zn, As, Br, Sr, Ba in Hanoi are higher than in our work. These pollutants originate from human activities in the high population and urbanization areas [24, 25]. These elements are most abundant elements in the crust. This reflects that the density of dust in the air is very high and air pollution in these provinces is seriously caused by floating dust. Vietnamese people feel this because a lot of building and transportation systerms are being built in these provinces resulting in a lot of crust dust in air [13].

4. Conclusion

Hoi An and Nha Trang cities are renowned tourist destinations, which play important roles in the Vietnamese economy. The analysis results of moss samples partly reflected the air quality in the research locations. The sources of air pollution were detected by moss techniques in the study areas. Our study is aimed to investigate air pollution and identify its sources in order to find a suitable process for improving the environment in the studied areas in Viet Nam.

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