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THE STUDY OF AIR POLLUTION OF METAL ELEMENTS IN HANOI, HUNG YEN AND DONG-NAI USING BARBULA INDICA MOSS

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Abstract. *Air pollution in Vietnam, especially the pollution caused by metal elements, is severe due to the rapid growth of industrialization. The study of air pollution caused by metal elements is a hard and expensive work. Development of the simple and cheap methods for studying metal pollution in the air is being carried out in many countries of the world. In this paper, we used Barbula Indica moss for studying heavy metal pollution in the air in Hanoi, Hung Yen and Dong-Nai provinces and the obtained results will be presented.*

Keywords: air pollution; heavy metal elements, Moss technique; neutron activation analysis.

Classification numbers: 89.60.-k; 91.65.Nd; 29.30.Hs; 82.33.Tb.

I. INTRODUCTION

In recent years, along with the rapid development of the economy, the air environment is seriously polluted by many different causes. Air pollution in Vietnam is a very worrying issue. There have been many different warnings about air pollution in Vietnam and which is high compared to other countries in the world [1, 2]. Therefore, monitoring air quality in Vietnam is an urgent need and should be carried out regularly and continuously.

Currently, the monitoring of air quality in Vietnam is conducted mainly by using automatic monitoring stations imported from abroad [3,4]. The advantage of this method is that it is possible to monitor air quality continuously. However, this method has many disadvantages, namely: i) very expensive (several dozens of billions vietnamese dong for a monitoring station and requires a few billion vietnamese dong of running cost per year); ii) need technical staff who are knowledgeable to maintain, and recalibrate analytical equipment; iii) can only be installed in locations where the power supply is available,... Due to these disadvantages, this method is often not suitable for developing countries like Vietnam. In addition, the automatic air quality monitoring stations are often used to monitor only toxic gases such as SOx, NOx, COx, etc. It is dificult to get information of metal pollution in the air by using these stations.

Another way for monitoring the air quality is to use power pumps in combination with air filters. After a certain period of air sample taking, the filters will be taken to laboratories for analyzing the concentration of metal elements accumulated on the filters. This method also has the disadvantages: it cannot be investigated on a wide area at the same time. Moreover, the time of sample taking cannot be long enough, so the analytical results are usually only considered to reflect the level of pollution in a very short period of time.

For overcoming these disadvantages of the above methods, a method for studying metal pollution in the air using biological indicators (lichen, moss, algae, ...) has been developed and widely used in European countries since the 60s of the last century [5–9]. It has been shown that the content of heavy metal elements in moss is proportional to the concentration of metal elements in the air. Mosses are mostly used due to the following reasons [10]: i) Mosses lack developed root system, therefore, they are fed only by nutrient absorption from the air; ii) Mosses lack leaves cuticula; iii) Mosses have low variability of morphology through the growing season; iv) Mosses have a high surface-to-volume ratio; v) Mosses have a high cation exchange capacity of cell walls. In addition, mosses are present everywhere, so sampling is not difficult and process of collecting moss, storage and processing for analysis is also very simple.

Many previous studies have confirmed that the method of studying metal pollution in the air by using moss is a cheap and easy one. However, so far, this method has not been implemented in Vietnam. In this paper, we will present the possibility of applying this technique to investigate the metal pollution in the air in Vietnam.

At the Institute of Physics of the Vietnam Academy of Science and Technology (VAST), a research group was formed in order to cooperate with the research team at the Joint Institute of Nuclear Research (JINR) in Dubna (Russian Federation) for using moss to study the metal pollution in the air in Vietnam. According to an agreement between two groups, the Vietnamese team will take moss samples from different provinces in Vietnam and bring them to JINR to determine the concentration of the metal elements by a method of neutron activation analysis (NAA) at the IBR-2 nuclear reactor. The first investigations have been carried out for different provinces in the north and south of the country including Thai Nguyen, Hanoi, Hue, Hoi An and Ho Chi Minh city [11, 12]. This study is a continuation of our campaign for investigation of the metal air pollution using moss as bioindicator in Hanoi, Hung Yen and Dong-Nai provinces.

II. MATERIAL AND METHODS

II.1. Moss sampling procedure

The local moss *Barbula indica* was used in this work. The moss height often ranges from 2 to 3 cm and rarely higher (up to 5 cm) but in Vietnam, it just ranges from 1 to 2 cm. As this is a short moss species, in our study, only mosses picked out from tree trunks or walls were used to minimize the influence of substrate on analytical results.

The sampling and the preparation of the moss samples were carried out in accordance with the protocols written in the monitoring manual of the UNECE ICP 2015 [13]. For each sample we recorded the following characteristics: the moment of sampling, the day, the name of the place and the GPS coordinates.

The sampling places were chosen from the areas with different pollution levels: Some were collected near the economical units which produce gaseous pollutants, others were taken near the roads which have different traffic flows and the others were also collected from the areas, far away from any sources of pollution. Thus we can have the possibility to make a comparative analysis between heavy metal air pollution in those places.

Mosses were cut out from their carpets by plastic tools and put in polyethylene zip-lock bags, both tools and bags were made from of low impurity materials. All sampling sites were located at least 200 m from main roads, 50 m from local roads with the sampling net at least of about 3 km $\times$ 3 km. However, by special characteristics of cities in Vietnam, in many places moss was covered with soil dust. Therefore, according to the protocol of "Field sampling" written in monitoring manual [13], the collected samples were thoroughly washed by distilled water and cleaned from extraneous materials (such as large soil particles, leaves, dust, etc.). Furthermore, according to this manual, the top part of the moss (living, green) was separated from the lower part (dead, brown) and only the top part was used for study. In the last stage, all moss samples were dried until their weights are unchanged.

II.2. Analytical method

The Neutron Activation Analysis (NAA) was carried out at the NAA Department of Frank Laboratory of Neutron Physics (FLNP) of the Joint Institute for Nuclear Research (JINR) in Dubna, Russia. About 0.3 g of dry weight of each sample was heat-sealed in polyethylene bag or was packed in aluminum cups, to be used for short-term irradiation or for long-term irradiation, respectively. The samples were irradiated in channels equipped with pneumatic system installed at the IBR-2 pulsed nuclear reactor of FLNP with the average power of 2 MW. Main characteristics of the irradiation channels are showed in Table 1 that have been maintained since 2000 years until now [14, 15]. To determine short-lived isotopes, each sample was irradiated for 3 min in channel 2, then was measured, after 3 to 5 min of decay, for 15 min. To determine long-lived isotopes, samples were irradiated for about 3 days in the Cd screened channel 1. After irradiation, they were repacked and measured twice, the first time after 3 days of decay for 30 min and, the second time after 20 days of decay for 1.5 h. Gamma spectra of the samples were measured by Ge (Li) detector or by HPGe detector with the resolution of 2.5 to 3 KeV or of 1.9 KeV, respectively, for the 1332 KeV line of the 60Co. The software Genie 2000 was used to store, display, and analyze the gamma spectra. The other software developed at FLNP was used to calculate concentrations of the elements in the samples. The calculation was done by the support of the standard reference materials produced by the International Atomic Energy Agency and the National Institute of Standards and Technology. The analytical errors of the concentrations of the interested elements range from 3 to 15%.

III. RESULTS AND DISCUSSION

III.1. Studied areas

For starting a campaign to study air pollution of metal elements in Vietnam using the moss bio-indicator, a number of areas in the south of Vietnam was investigated, namely in Hue, Hoi An and Ho Chi Minh city [12]. In this investigation, Hanoi, Hung Yen and Dong-Nai provinces were studied. The sampling locations in these areas are shown in Fig. 1.

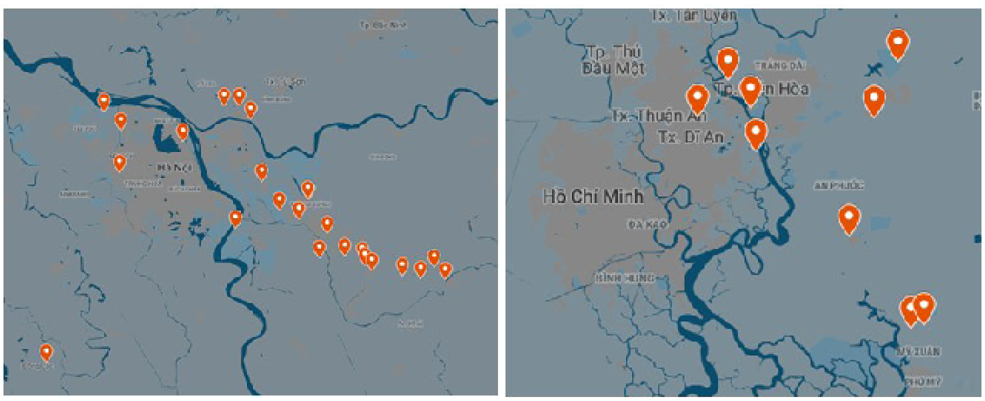

Fig. 1. The locations of sampling sites.

The total number of collected moss samples was 35, among them 10 in Hanoi, 15 in Hung Yen and 10 in Dong-Nai. These moss samples were collected during the period from November 2016 to February 2017. The concentration of 33 metal elements for each moss sample was determined by the method of neutron activation analysis carried out using IBR-2 nuclear reactor of the Joint Institute for Nuclear Research in Dubna. The error of the concentration for most elements is less than 10%. The average values of concentration in mg.kg^{-1} (dry weight) of 33 elements in Hanoi, Hung Yen and Dong-Nai are listed in Table 1 together with the minimum and maximum values of the elemental concentration of all 33 elements.

The concentration of almost elements in the same area at different sampling points varies in a fairly wide range. This suggests that there are many pollution sources which spread these elements into the air in the investigated areas.

From Table 1, it can be seen that in Hanoi, the descending order of the mean concentration of the elements in moss samples is: Ca > K > Al > Fe > Mg > Cl > Na > Ti > Zn > Mn > Ba > Sr > V > Cr > Ce > Br > Ni > La > As > Co > Sb > Th > Sc > U > Cs > Gd > Cd > Hf > Sm > Yb > Se > Ta > Tb. In Hung Yen province, the order is: Ca > K > Al > Mg > Fe > Cl

> Na > Zn > Ti > Mn > Ba > Sr > Cr > V > Br > Ce > Ni > La > As > Co > Sb > Cs > Th > Sc > Cd > U > Gd > Hf > Sm > Se > Yb > Ta > Tb. For Dong-Nai province, the following descending order was observed: Ca > K > Al > Fe > Mg > Cl > Ti > Na > Mn > Zn > Sr > Ba > Cr > V > Ce > Ni > Br > La > Co > Sc > As > Hf > Th > Sm > Cs > Gd > Sb > U > Cd > Yb > Se > Ta > Tb. Although there is a slight difference, in general the descending order of the mean concentration in the moss samples observed in the three provinces is quite similar. It means that the sources of metal pollutions in the air in these three provinces are quite similar.

The highest concentrations belong to Ca, K, Al, Fe, Mg, Cl, Na and Ti. These elements are most abundant elements in the crust. The lowest concentrations belong to Ta and Tb. 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 buildings and transportation systems are being built in these provinces resulting in a lot of crust dust in the air.

Descriptive statistics of metal concentration data including mean, median, standard deviation, minimum, maximum, range, coefficient of variation in percent (CV=standard deviation/mean $\times$ 100%), kurtosis and skewness determined in moss samples in Hanoi, Hung Yen and Dong-Nai are presented in Table 2. The IBM SPSS software version 20 was used for statistical calculations.

From Table 2, it can be observed that all of the heavy metals under investigation in Hanoi showed moderate variation in concentration, with the coefficient of variation ranging from 17% to 68% except for Ba (117.4 %). For Hung Yen province, the coefficients of variation range from 14% to 64% except for Zn (97%) and Ta (227.1%). For Dong-Nai province, the variation in concentration is much higher than Hanoi and Hung Yen. The coefficients of variation for U, Fe, Yb, Hf, Sc, Mn, Cr, Ta, Sb, Ti, Sr, Ni and Co are higher than 75%. High values of the coefficient of variation are likely to indicate the influence of complicated origins of these elements in mosses [10]. Furthermore, for those elements whose value of skewness is in the range from -0.8 to 0.8 and the value of its kurtosis is in the range from -3.0 to 3.0 then its concentration can be considered to be normally distributed. In this case, the source of pollution of these elements can be the same. Otherwise, the source of pollution may be very complicated.

III.2. Contamination Factor of Metal Elements

Contamination factor (CF) may be useful to classify pollution situation of the investigated area [16] and for our case they are listed in Table 3. We calculated contamination factor for element i by the following formula

$$CF_i = \frac{C_i}{BG_i} \tag{1}$$

where C_i is the mean value of each element from the moss samples from the investigated area and BG_i is the average e value of the three sample sites showing the lowest concentration of the corresponding metal from the investigated area. The pollution level can be classified according to CF value as follows: CF$\leq$1: no pollution (C1), 1<CF$\leq$2: suspected pollution (C2), 2<CF$\leq$3.5: slight pollution (C3), 3.5<CF$\leq$8: moderate pollution (C4), 8<CF$\leq$27: serious pollution (C5) and 27<CF: extreme pollution. The calculated CF coefficients for Hanoi, Hung Yen and Dong-Nai areas together with their air pollution rank are listed in Table 3.

It can be seen from Table 3 that Hanoi is suspected pollution by most elements (1<C2$\leq$2) except for Cd, Ba and U. By these elements, Hanoi is moderately polluted (2<CF$\leq$3.5). In

Table 1. The concentration of elements in the collected moss samples in $mg.kg^{-1}$ (dry weight).

	El.	Hanoi			Hung Yen			Dong-Nai		
		Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
1	Na	946.9	601	1380	912.9	717	1830	927	522	2100
2	Mg	4876	3630	6640	3989	3340	6380	4095	2160	7620
3	Al	7882	4950	13900	5871	4530	14700	8552	1800	20400
4	Cl	1212	485	2300	2748	2830	3620	1683	462	2580
5	K	8764	6450	10700	9931	8600	12200	11930	8980	14200
6	Ca	27310	13500	62600	21026	15800	27600	14585	8470	33600
7	Sc	1.44	0.83	2.46	0.95	0.708	2.25	1.90	0.352	5.75
8	Ti	587	370	967	356.70	298	749	1378.48	91.3	3930
9	V	21.15	11.8	39.1	14.70	13.4	24.1	19.50	3.22	45.4
10	Cr	19.12	10.5	32.3	21.28	10.9	46.3	30.00	7.03	103
11	Mn	152.4	100	225	127.8	200	226	132.5	47.6	412
12	Fe	5431	2850	8310	3536	4050	6610	6641	1290	19400
13	Ni	7.75	4.95	15.5	6.40	5.22	11.8	8.60	1.98	32.3
14	Co	2.22	1.50	3.58	1.49	1.11	3.42	3.11	0.76	13.70
15	Zn	443	158	1230	370.1	298	1340	97.93	57.8	156
16	As	4.09	2.59	7.83	2.49	1.56	3.85	1.58	0.79	2.59
17	Se	0.32	0.18	0.52	0.33	0.273	0.68	0.21	0.136	0.30
18	Br	8.64	4.29	16.10	8.42	5.56	15.10	7.08	3.50	16.8
19	Sr	66.92	28.3	131.0	36.27	34.3	47.2	53.30	21.9	204
20	Cd	0.86	0.34	1.39	0.71	0.7	1.66	0.50	0.20	0.78
21	Sb	1.68	0.78	3.28	1.18	0.72	2.22	0.63	0.20	2.29
22	Ba	141.08	52.5	601	55.65	61.1	111	40.57	19.8	55.5
23	Cs	1.11	0.61	1.82	1.17	0.71	2.04	0.67	0.33	1.09
24	La	4.76	3.06	8.42	3.56	2.79	8.41	4.19	1.55	9.77
25	Ce	9.33	6.07	15.8	7.03	5.14	17.3	9.52	3.14	22
26	Sm	0.67	0.36	1.22	0.51	0.4	1.21	0.75	0.25	1.56
27	Gd	0.87	0.45	1.67	0.62	0.53	1.32	0.67	0.24	1.31
28	Tb	0.10	0.06	0.16	0.07	0.05	0.16	0.09	0.03	0.16
29	Yb	0.35	0.55	0.35	0.29	0.26	0.61	0.32	0.11	0.57
30	Hf	0.84	0.44	1.23	0.52	0.38	1.35	1.34	0.46	3.6
31	Ta	0.14	0.09	0.23	0.27	0.12	2.46	0.19	0.03	0.50
32	Th	1.63	1.00	2.74	1.12	0.83	2.72	1.09	0.36	2.21
33	U	1.16	0.44	2.02	0.67	0.88	1.17	0.59	0.18	1.72

Hung Yen area, there is no pollution by Th and U (C1). Air pollution by 17 elements including Na, Mg, Cl, K, Ca, V, Mn, Fe, As, Se, Br, Sr, Ba, Cs, Gd, Yb and Hf is suspected (C2). By other elements including Al, Sc, Ti, Cr, Ni, Co, Cd, Sb, La, Ce, Sm and Tb, air pollution is slight (C3). Especially, air pollution by Zn and Ta is moderate (C4). In case of Dong-Nai province, it can be summarized as follows. Air pollution by 11 elements namely Na, Mg, K, Ca, Zn, As,

Table 2. Descriptive statistics of metal concentration data.

El.	Hanoi				Hung Yen				Dong-Nai			
	SD	Kurt	Skew	CV (%)	SD	Kurt	Skew	CV (%)	SD	Kurt	Skew	CV (%)
Na	294.5	-1.64	0.41	31.1	372.8	1.28	1.35	40.84	539.2	2.03	1.64	58.16
Mg	1135	-1.19	0.55	37.33	1198	0.06	0.99	30.03	1865	-0.12	0.98	45.53
Al	2942	0.73	1.26	37.33	3524	2.09	1.54	60.03	6238	-0.27	0.92	73.47
Cl	520	1.15	0.8	42.88	529.9	0.19	-0.23	19.28	767.7	-1.45	-0.41	45.62
K	1543	-1.68	-0.04	17.61	1436	-1.09	0.05	14.47	1746	-0.84	-0.56	14.63
Ca	15108	2.58	1.63	55.32	3015	0.42	0.29	14.34	7603	6.09	2.34	52.13
Sc	0.52	0.01	0.87	36.11	0.59	1.07	1.34	62.18	1.7	2.94	1.58	89.32
Ti	217.7	-0.94	0.83	37.08	176.0	0.72	0.92	49.33	1445	0.89	0.94	104.8
V	9.27	-0.08	0.84	43.84	4.64	0.28	0.72	31.59	14.22	-0.55	0.47	72.91
Cr	6.98	-0.55	0.72	36.51	12.21	-0.05	1.02	57.37	29.61	5.53	2.24	98.68
Mn	45.84	-1.46	0.38	30.08	47.83	-0.32	0.61	37.43	119.5	3.75	1.89	90.18
Fe	1176	-1.05	0.14	32.71	1562	-0.32	0.83	44.18	5757	2.44	1.47	86.68
Ni	3.26	3.03	1.69	42.06	2.95	-0.72	0.44	46.13	9.53	5.83	2.34	110.8
Co	0.7	-0.16	0.87	31.69	0.77	1.68	1.18	51.67	4.16	6.79	2.55	133.9
Zn	304.0	5.72	2.21	68.63	361.7	3.01	1.85	97.73	34.32	-0.77	0.58	35.05
As	1.56	3.28	1.68	38.23	0.88	-1.23	0.47	35.59	0.5	1.82	0.61	31.35
Se	0.12	-0.92	0.54	37.65	0.12	4.43	1.82	36.04	0.06	-1.63	0.38	30.45
Br	3.2	3.1	1.28	36.97	2.87	0.63	1	34.06	4	5.03	2.09	56.54
Sr	36.06	-0.45	0.85	53.89	6.58	-0.98	0.17	18.15	57.7	7.96	2.78	108.3
Cd	0.38	-1.78	0.04	44.98	0.42	-0.03	0.85	58.3	0.18	-0.46	0.1	36.48
Sb	0.7	2.47	1.27	41.54	0.53	-0.07	0.7	45.42	0.64	7.34	2.63	101.7
Ba	165.6	8.69	2.9	117.4	23.44	0.91	0.96	42.12	12.5	-1.1	-0.48	30.81
Cs	0.36	0.49	0.81	32.53	0.44	0.08	0.88	37.79	0.25	0.73	0.57	37.26
La	1.57	2.82	1.58	32.92	2.11	1.37	1.44	59.35	2.7	1.03	1.18	64.33
Ce	2.83	2.49	1.35	30.36	4.29	1.65	1.53	60.98	6.34	0.29	0.99	66.59
Sm	0.25	1.83	1.01	37.36	0.3	1.24	1.36	57.91	0.46	-0.82	0.47	60.48
Gd	0.32	5.12	1.84	36.63	0.3	0.76	1.16	48.81	0.39	-1.31	0.5	58.45
Tb	0.03	1.51	1.15	31.05	0.04	1.18	1.36	43.96	0.05	-2.22	-0.05	57.87
Yb	0.11	0.19	0.39	30.39	0.13	1.66	1.3	43.96	0.16	0.51	1.37	86.98
Hf	0.27	-1.63	0.03	32.31	0.31	2.89	1.8	58.68	1.16	0.51	1.37	86.98
Ta	0.05	-0.6	0.78	32.95	0.61	14.6	3.8	227.1	0.19	-1.46	0.85	100.2
Th	0.55	0.82	1.07	33.53	0.71	1.36	1.52	63.75	0.58	0.46	0.55	53.08
U	0.55	-1.21	0.26	47.25	0.26	-0.87	0.47	39.23	0.48	3.63	1.88	81.82

SD: Standard deviation; CV: Coefficient of variation; Kurt: kurtosis; Skew: skewness

Se, Br, Cd, Ba and Cs is suspected (C2). Air pollution for 16 elements including Al, Cl, Cr, Mn, Ni, Sr, Sb, La, Ce, Sm, Gd, Tb, Yb, Hf, Th and U is slight (C3). Especially by 6 elements including Sc, Ti, V, Fe, Co and Ta, the air pollution is moderate (C4). In general according to the values of contamination factors listed in Table 3, it could be concluded that the air pollution in Hung Yen and Dong-Nai is more serious than in Hanoi. It might be understood because Dong-Nai and Hung Yen belong to the provinces with the fastest industrial growth rate in the country. There are many big industrial zones located in these provinces and therefore, air pollution is a result of this rapid industrial growth.

Table 3. Contamination factor.

	Element	Hanoi		Hung Yen		Dong-Nai	
		CF	Rank	CF	Rank	CF	Rank
1	Na	1.46	C2	1.62	C2	1.67	C2
2	Mg	1.32	C2	1.42	C2	1.59	C2
3	Al	1.46	C2	2.16	C3	2.62	C3
4	Cl	1.69	C2	1.39	C2	2.16	C3
5	K	1.24	C2	1.24	C2	1.21	C2
6	Ca	1.72	C2	1.23	C2	1.49	C2
7	Sc	1.55	C2	2.3	C3	3.88	C4
8	Ti	1.48	C2	2.17	C3	5.77	C4
9	V	1.79	C2	1.59	C2	3.86	C4
10	Cr	1.49	C2	2.31	C3	2.95	C3
11	Mn	1.46	C2	1.81	C2	2.67	C3
12	Fe	1.58	C2	1.86	C2	3.95	C4
13	Ni	1.5	C2	2.23	C3	3.09	C3
14	Co	1.45	C2	2.07	C3	3.8	C4
15	Zn	1.98	C2	4.69	C4	1.48	C2
16	As	1.46	C2	1.66	C2	1.44	C2
17	Se	1.63	C2	1.51	C2	1.43	C2
18	Br	1.57	C2	1.61	C2	1.65	C2
19	Sr	1.99	C2	1.31	C2	2.14	C3
20	Cd	2.03	C3	2.34	C3	1.51	C2
21	Sb	1.63	C2	2.06	C3	2.43	C3
22	Ba	2.47	C3	1.89	C2	1.58	C2
23	Cs	1.46	C2	1.73	C2	1.49	C2
24	La	1.37	C2	2.12	C3	2.24	C3
25	Ce	1.38	C2	2.11	C3	2.44	C3
26	Sm	1.64	C2	2.02	C3	2.54	C3
27	Gd	1.41	C2	1.87	C2	2.35	C3
28	Tb	1.43	C2	2.03	C3	2.61	C3
29	Yb	1.49	C2	1.89	C2	2.09	C3
30	Hf	1.5	C2	1.79	C2	2.69	C3
31	Ta	1.49	C2	6.1	C4	3.64	C4
32	Th	1.43	C2	0.65	C1	2.12	C3
33	U	2.14	C3	0.9	C1	2.33	C3

IV. CONCLUSION

This study is the continuation of our previous work to characterize the atmospheric pollution of heavy metal elements using moss technique in three provinces in the north and south of Vietnam, namely Hanoi, Hung Yen and Dong Nai. *Barbulea Indica* moss (35 samples) has been used as bio-indicator of air pollution of metal elements. The instrumental neutron activation analysis has been used using IBR-2 nuclear reactor at the Joint Institute for Nuclear Research (Dubna, Russia). For each moss sample, the concentrations of 33 elements were determined with the error being less than 10%. By the obtained results, the contamination factor for all elements has been determined and the pollution situation based on the values of this factor for these elements in three investigated provinces has been categorised. In order to find the pollution sources, the multivariate statistical analysis should be applied and this analysis will be carried out latter. Finally, we would like to emphasize that using moss technique is a low cost way for mapping air pollution in Vietnam. We will continue to apply this technique for further investigation of air pollution in other provinces in the country.

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