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Research Article

ASSESSMENT OF POTENTIALLY TOXIC TRACE ELEMENTS IN THE SURFACE SOILS OF DONG NAI PROVINCE USING NEUTRON ACTIVATION ANALYSIS

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ABSTRACT

This study aimed to investigate the levels of contamination of potentially toxic trace elements in the soil of Dong Nai Province, Vietnam. To achieve this, the Instrument Neutron Activation Analysis (INAA) method was employed to measure the concentrations of five potentially toxic trace elements, including manganese (Mn), arsenic (As), chromium (Cr), antimony (Sb), and zinc (Zn), across 30 soil samples collected from the region. After that, the Geo Accumulation Index (Igeo) and enrichment factors (EF) were utilized to evaluate the existence of anthropogenic contaminants and the intensity of their deposition on surface soil. The results of the Igeo revealed varying levels of contamination for each of the elements investigated, with Sb showing the highest levels of contamination, followed by Zn, Cr, As, and Mn. The EF analysis also demonstrated that the mean enrichment factor value was highest for Sb, followed by As, Zn, Cr, and Mn. The results of this study provide important baseline data for future research on soil pollution in Dong Nai Province and other rapidly industrializing regions. It is hoped that the findings will serve as a valuable resource for policymakers and environmental managers seeking to mitigate the risks associated with soil contamination and promote sustainable land use practices.

Keywords: EF; Igeo; INAA; soil contamination; trace elements

I. Introduction

Soil is an important, non-renewable resource that ecosystems and humans need to survive. Soil quality, and notably the level of soil pollution, has important implications for human health (Li et al., 2017; Velea et al., 2008; Ding et al., 2018). Soil is both a source and

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a sink for pollution, and it can pass pollution on to groundwater and the food chain, and then to humans and/or animals (Khan et al., 2010). The elements present in soil have a direct impact on both the environment and human health. A sufficient intake of essential elements is necessary for health (Singh et al., 2006). Certain essential elements pose a health risk when consumed in excess. Activities such as industrial, agricultural, and transportation operations, as well as construction work, and so on, affect every component of the environment, particularly the soil. Monitoring of these environmental components is critical for recognizing and forecasting the hazardous effects of pollutants on the biosphere.

Vietnam is a country in the process of industrialization and modernization. The potentially toxic trace elements are one of the characteristics that require close attention in Vietnam's soil quality monitoring system. Numerous research on soil pollution has been conducted in Vietnam (Kien et al., 2010; Phuong et al., 2009; Nguyen et al., 2020; Thanh et al., 2022). Soil mineral and element quantification has traditionally been performed using a variety of traditional analytical techniques, including atomic absorption spectroscopy (AAS), inductively coupled plasma optical emission spectroscopy (ICP-OES), neutron activation analysis (NAA), and inductively coupled plasma mass spectrometry (ICP-MS). We employed the instrument neutron activation analysis (INAA) to evaluate the potentially toxic trace element composition of 30 surface soil samples from Dong Nai in this investigation. Due to its accuracy and reliability, neutron activation analysis (NAA) is one of the most sensitive methods for analyzing trace elements in soil (Kassem et al., 2004; Naidu et al., 2003). The primary advantage of INAA is that it is a nondestructive technique in which the sample is not subjected to any physicochemical treatment, other than drying, before analysis. This study was conducted to analyze the potentially toxic trace elements manganese (Mn), arsenic (As), chromium (Cr), antimony (Sb), and zinc (Zn) in the soil and identify the level of pollution using the geoaccumulation index (Igeo) and enrichment factor (EF). The findings can give useful data on soil pollution levels in Dong Nai, which can be used as references for land treatment, usage, and management in building and development projects.

2. Method

2.1. Study site and sampling

Dong Nai Province in southeastern Vietnam, covering an area of 5,900 square kilometers, is known for diverse agriculture and rapid urbanization. With a population exceeding 3 million, it combines industrial, residential, and agricultural land usage with more than 31 industrial zones, supporting various industries like textiles, electronics, and food processing. This study's focus on Dong Nai Province is essential due to its economic importance and potential environmental concerns, as industrial zones and agriculture may introduce trace elements into the environment through discharge activities or the use of fertilizers and pesticides.

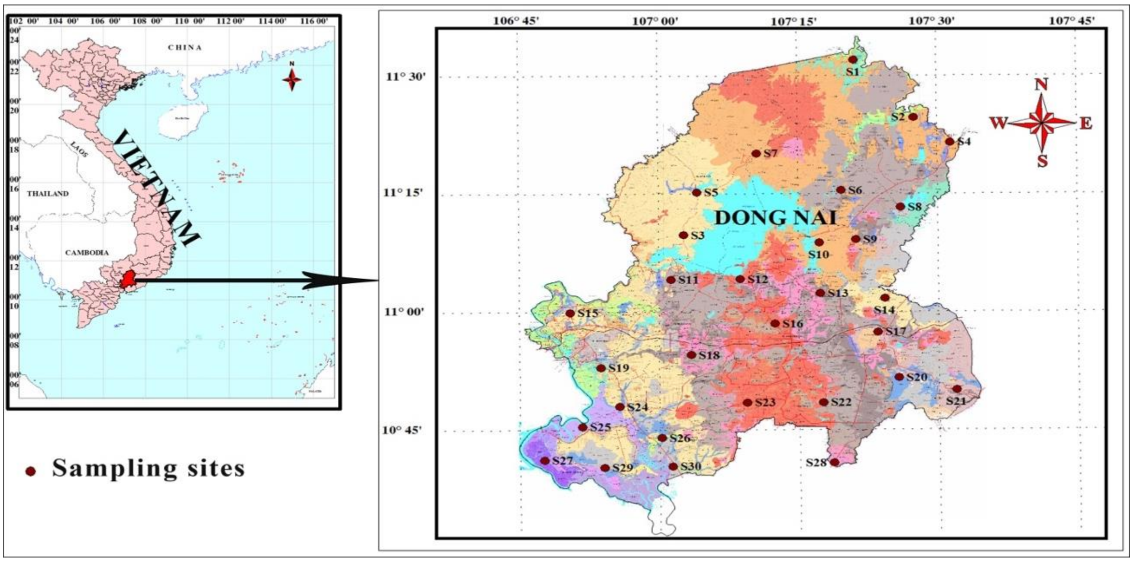


Figure 1. *The location map of the study area and sampling sites in Dong Nai*

Thirty soil samples were collected. The sample locations were recorded with a GPS (Global Positioning System). The soil core (diameter 10 cm, depth 5 cm) was taken using stainless steel pipes. After collection, samples were labeled and stored in a sealed bag. The sampling sites are shown in Figure 1. The soil samples were crushed and passed through a 2 mm sieve to remove rocks and large plant remains. After that, the soil samples were ground and sieved for a particle size fraction of approximately 0.25 mm, then dried at 100^0C for 10 h. The content of average moisture was determined to be approximately 1 to 2 %.

2.2. Instrument neutron activation analysis - INAA

A plastic box containing 30 mg (short irradiation) and 100 mg (long irradiation) of soil sample was used to determine the elemental content using k0-standardized neutron activation analysis. The packed samples were irradiated at the dry channel No. 7-1 (short irradiation) and the rotary specimen rack (long irradiation) of the Dalat research reactors. After suitable irradiation and cooling times, the samples were counted in the HPGe detector (GMX30190, FWHM approximately 2.3 keV at 1332.5 keV, and relative efficiency of 30%), respectively. The efficiency calibration of the HPGe detector was measured by counting the point sources ^{137}Cs and ^{152}Eu. The neutron spectral parameters in irradiation position (φth, α, f) were determined by using the set of monitors (^{198}Au, ^{95}Zr/^{97}Zr). The standard reference material (NIST 2711a, Montana II Soil) was determined together with the soil sample. The u-score test and relative bias of standard reference material were calculated for quality control (Ho et al., 2016). The k0-IAEA software version 9.1 was used to calculate the absolute efficiencies, neutron spectral parameters, and elemental concentrations. Table 1 shows the analytical condition for the soil sample.

The Worldwide Open Proficiency Tests for Nuclear and Related Analytical Techniques Laboratories (PTNATIAEA/19, lab code: 202) performed a quality control process for the measurements by analyzing international comparison clay samples and sediment samples with the same procedure.

Table 1. *Analytical conditions for soil sample in the Dalat research reactor (short, medium, and long half-live isotopes)*

Sample mass	Irradiation time	Irradiation position (φth, α, *f*)	Decay time	Counting time	Measured radionuclides
~30 mg	1 m	Chanel No. 7-1 ($4×10^{12}$ n.cm-2.s-1, -0.0676, 9.86)	10 m	180 s	^{28}Al.
			1 h 30 m	600 s	^{56}Mn.
~100 mg	10 h	Rotary rack ($3.5×10^{12}$ n.cm-2.s-1, 0.071, 39.5)	4 d	1,200 s	^{76}As.
			15 d	10,800 s	^{51}Cr, ^{124}Sb, ^{65}Zn.

2.3. The assessment of soil contamination

The Geo Accumulation Index (Igeo)

The Igeo of the soil samples was examined to assess the level of soil pollution. The Geo accumulation Index was first created by Müller for metal concentrations in the 2-micron fraction and was subsequently developed for global standard shale values (Müller et al., 1969; Müller et al., 1979). Igeo was evaluated based on the values suggested by Equation 1 indicates the Igeo account:

$$Igeo_i = Log_2 \frac{C_i}{1.5\ B_i} \quad \text{(Equation 1)}$$

Where Ci is the measured concentration of the element in soil, Bi is the geochemical background value, and the constant 1.5 enables us to study natural changes in the concentration of a specific substance in the environment and to discover extremely small anthropogenic influences. From Class 0 (Igeo=0, Uncontaminated) to Class 6 (Igeo>5, very contaminated), Müller has established seven distinct levels of pollution according to the Geoaccumulation Index (Table 2). At least one hundred-fold increase in abundance above baseline is reflected in Class 6, the highest possible classification.

Table 2. *Classification of Geoaccumulation Index*

Class	Value	Soil quality
0	Igeo≤0	Uncontaminated
1	0< Igeo ≤1	Uncontaminated to moderately contaminated
2	1< Igeo ≤2	Moderately contaminated
3	2< Igeo ≤3	Moderately to heavily contaminated
4	3< Igeo ≤4	Heavily contaminated
5	4< Igeo <5	Heavily to extremely contaminated
6	Igeo≥5	Extremely contaminated

Enrichment factor (EF)

The enrichment factor (EF) can be utilized to differentiate between the metals originating from human activities and those from natural procedures, and to assess the degree of anthropogenic influence (Li et al., 2017; Nguyen et al., 2020; Buat-Menard et al., 1979;

Ali et al., 2013), and equation 2 is used to calculate it as follows:

$$EF = \frac{(Ci/Cref)soil}{(Ci/Cref)background} \quad \text{(Equation 2)}$$

Where (Ci/Cref) soil is the ratio between the concentrations of the element of interest and the reference element in the soil sample and (Ci/Cref) background is the ratio between the concentrations of the element of interest and the reference element in the background. The reference (or conservative) elements are those whose concentrations in the earth's crust are so high that anthropogenic effects do not significantly alter them, or which are inactive in biogeochemical cycles. The most often used reference elements are Fe, Al, Ca, Ti, Sc, or Mn (Kowalska et al., 2018). Al is selected as the reference element in the present study. The soil quality classification is in accordance with the EF as indicated in Table 3.

Table 3. *Classification of Enrichment factor*

Class	Value	Soil quality
0	EF<2	Deficiency to minimal enrichment
1	2≤ EF <5	Moderate enrichment
2	5≤ EF <20	Significant enrichment
3	20≤ EF <40	Very high enrichment
4	EF ≥ 40	Extremely high enrichment

Background values or reference data, such as the concentrations of reference elements, are essential for calculating indices like Igeo and EF. Baseline data helps determine the amount of each element present in soil samples in relation to environmental indices such as the geo-accumulation index (Igeo) and enrichment factor (EF). In this study, Rudnick and Gao's upper continental crust values (Rudnick et al., 2003) were used as the background values.

3. Result and discussion

In the present study, 30 soil samples from Dong Nai were collected and analyzed by irradiating the samples in the Dalat research reactor. By detecting gamma rays emitted from irradiated samples, five elements As, Cr, Mn, Sb, and Zn in soil samples were quantified. Elemental concentrations of NIST 2711a and Montana II Soil (International exchange soil for comparison) have been determined and used for checking the precision and accuracy of the method. Table 4 shows a summary of elemental concentrations in the soil sample. The following parameters are shown: (min-minimal value, max-maximal value, Mean-mean value, SD-standard deviation, and Ref- the reference element).

Manganese (Mn) concentrations in the samples ranged from 13.82 to 5,642 mg/kg, indicating significant variations in Mn concentrations, with a mean of 1,038 mg/kg. The mean concentration is comparable to the average upper continental crust concentration of 1,000 mg/kg (Rudnick et al., 2003). Comparatively, a previous study (Nguyen et al., 2020) reported that the concentration of manganese in agricultural soil in Nam Dinh Province ranged from 10,000 to 170,000 mg/kg, which was higher than the levels observed in the current study area. The lowest value of arsenic (As) is 0.84 mg/kg, and the highest value is 37.52 mg/kg. The mean value is 9.4 mg/kg. The mean value of As is higher than the average of the upper continental crust, which is 4.5 mg/kg (Rudnick et al., 2003). Many studies have looked at how much As is in the different soils used for farming in Viet Nam. The topsoil used for farming in Nam Dinh, Vietnam, had the highest amount of As, at 22.46 mg/kg (Nguyen et al., 2020). Samples S5 and S6 are higher than that number. The average concentration of Cr in the upper continental crust is 92 mg/kg (23). This study measured Cr concentrations ranging from 17.56 to 843.6 mg/kg, with a mean of 197.78 mg/kg. The Cr content of 50 % of all samples exceeds the upper continental crust average (92 mg/kg). The mean concentration is greater than that of agricultural soils in Nam Dinh (37.82 mg/kg) (Rudnick et al., 2003). The average Sb concentration was 1.36 mg/kg, ranging from 0.17 to 13.24 mg/kg. Compared to the study conducted by Rudnick and Gao in 2003 (Rudnick et al., 2003), 73.3% of all samples have a higher Sb concentration in the upper continental crust than 0.4 mg/kg. According to Li et al. (2017), the As concentration in Baoji, China ranged from 7.8 to 179.07 mg/kg, which was substantially higher than the concentration in the study area. The range of Zn concentration in the study area was 8.6 to 328 mg/kg, with an average of 139.82 mg/kg. The sample with the lowest value was S29, and the sample with the greatest value was S28. The average Zn concentration in the upper continental crust was estimated to be 67 mg/kg (Rudnick et al., 2003), with 83.3% of the samples examined having higher concentrations. The reported average Zn concentrations at various locations in Vietnam ranged from 27.17 to 195.49 mg/kg (Kien et al., 2010; Phương et al., 2009; Nguyen et al., 2020; Thanh et al., 2022). Generally, the distribution of five elements in Dong Nai soils was in the following order: Mn>Cr>Zn>As>Sb. The mean concentration of Mn (1038 mg/kg), Cr (197.8 mg/kg), Zn (139.8 mg/kg), As (9.4 mg/kg), and Sb (1.36 mg/kg) was higher than the average upper continental crust value given in Rudnick and Gao 2003 (Rudnick et al., 2003).

Table 4. *Summary of elemental concentrations (mg/kg) in the soil sample*

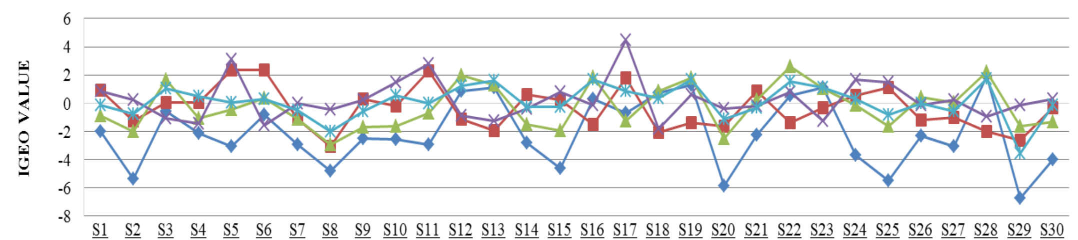
Element	Min	Max	Mean	SD	Reference*
Mn	13.82	5,642	1,038	1,433	1,000
As	0.84	37.52	9.40	10.59	4.8
Cr	17.56	843.60	197.78	215.22	92
Sb	0.17	13.24	1.36	2.50	0.4
Zn	8.60	328.00	139.82	93.99	67
Al**	6,010	137,900	59,681	41,211	154,000

** Reference value (Rudnick R.L. and Gao .S 2003).*

*** Enrichment factor (EF): Al is selected as the reference element in the present study.*

Assessment based on the Geo Accumulation Index

The results of the Igeo analysis indicate varying levels of contamination at different sites across the study area. Igeo classified all soils as uncontaminated to moderately to heavily contaminated with Mn, As, Cr, Sb, and Zn (Figure 2). The mean Igeo values were in the following order: Sb>Zn>Cr>As> Mn. For manganese (Mn), sites S12, S16, S18, and S22 have an uncontaminated to moderately contaminated soil quality (Class 1), while sites S13, S19, S23, and S28 have a moderately contaminated soil quality (Class 2). The remaining sites show no evidence of manganese pollution. For arsenic (As), sites S1, S3, S4, S9, S14, S15, S21, and S24 are classified as moderately contaminated (Class 1), while sites S17 and S25 are moderately contaminated (Class 2), and sites S5, S6, and S11 are moderately to heavily contaminated (Class 3). Chromium (Cr) contamination levels vary, with sites S3, S12, S13, S16, S19, S2, S23, and S28 having moderate to heavy contamination (Classes 2 and 3), sites S6, S18, S21, S26, and 27, having moderate contamination (Class 1), and the remaining sites showing no contamination. Antimony (Sb) contamination ranges from moderately contaminated (Class 1 and 2) to highly contaminated (Class 5), with sites S5, S11, and S17 showing the highest level of contamination. Finally, for zinc (Zn), sites S3, S12, S13, S16, S19, S22, S23, and S28 have a moderate to high level of contamination (Class 2), while sites S4, S5, S6, S10, S17, S18, S24, and S26 are uncontaminated to moderately contaminated (Class 1), and the remaining sites show no evidence of Zn pollution.



	S1	S2	S3	S4	S5	S6	S7	S8	S9	S10	S11	S12	S13	S14	S15	S16	S17	S18	S19	S20	S21	S22	S23	S24	S25	S26	S27	S28	S29	S30
Mn	-2,0	-5,3	-0,5	-2,1	-3,0	-0,8	-2,9	-4,8	-2,5	-2,5	-2,9	0,88	1,09	-2,8	-4,6	0,29	-0,6	0,75	1,30	-5,9	-2,2	0,54	1,11	-3,6	-5,5	-2,3	-3,0	1,91	-6,7	-4,0
As	0,93	-1,2	0,06	0,07	2,36	2,38	-1,1	-3,0	0,27	-0,2	2,27	-1,1	-1,9	0,61	0,26	-1,5	1,80	-2,0	-1,3	-1,6	0,85	-1,4	-0,3	0,54	1,13	-1,2	-1,0	-2,0	-2,6	-0,3
Cr	-0,8	-2,0	1,66	-1,0	-0,4	0,33	-1,1	-2,9	-1,6	-1,6	-0,7	1,99	1,27	-1,5	-1,9	1,83	-1,2	0,86	1,82	-2,4	0,24	2,61	1,03	-0,1	-1,6	0,42	0,03	2,20	-1,6	-1,3
Sb	0,87	0,22	-1,0	-1,4	3,10	-1,6	-0,0	-0,4	0,22	1,49	2,77	-0,9	-1,2	-0,4	0,77	-0,1	4,46	-1,8	0,63	-0,4	-0,2	0,80	-1,2	1,65	1,46	-0,1	0,25	-0,9	-0,1	0,28
Zn	-0,1	-0,7	1,06	0,49	0,02	0,31	-0,5	-2,0	-0,5	0,53	-0,0	1,25	1,59	-0,2	-0,2	1,65	0,85	0,34	1,64	-1,1	-0,3	1,54	1,12	0,27	-0,8	0,01	-0,5	1,71	-3,5	-0,1

Figure 2. *Igeo values calculated for the sites*

Assessment based on enrichment factor

The results of EF across 30 sampling sites revealed varying levels of enrichment for five elements, namely Mn, As, Cr, Sb, and Zn. The enrichment factors were classified into four categories, ranging from no enrichment to very high enrichment (Figure 3). The mean EF values followed the following order: Sb>As>Zn>Cr>Mn. For Mn, twenty sites (S1 to S8, S10, S11, S14, S15, S17, S20, S24 to S27, S29, and S30) showed no enrichment (class 0), while sites S9, S12, S16, and S21 to S23 exhibited moderate enrichment (class 1), and sites S13, S18, S19, and S28 were classified as having significant enrichment (class 2). For As, the highest enrichment was observed in site 11 (class 3). Twelve sites, including S5, S6, S9,

S10, S14, S15, S17, S20, S21, S25, S29, and S30, showed significant enrichment (class 2), seven sites S1, S2, S4, S7, S8, S18, S24 had moderate enrichment (class 1), and ten sites S3, S12, S13, S16, S19, S22, S23, S26, S27, S28 showed low enrichment (class 0). In the case of Cr, ten sites S3, S12, S13, S16, S18, S19, S21, S22, S26, S29) showed significant enrichment (class 2), thirteen sites S4 to S9, S11, S14, S15, S20, S23, S27, S30 had moderate enrichment (class 1), six sites S1, S2, S10, S17, S24 and S25 showed no enrichment (class 0), and one site S28 exhibited very high enrichment (class 3). For Sb, site S17 showed extremely high enrichment (class 4), fifteen sites S2, S5, S7 to S11, S14, S15, S20, S21, S24, S25, S29, S30 showed moderate to very high enrichment (classes 2 and 3), seven sites S1, S18, S19, S22, S26, S27, S28 had moderate enrichment (class 1), and seven sites S3, S4, S6, S12, S13, S16, S23 showed no enrichment (class 0). Finally, for Zn, all 30 sites exhibited moderate to significant enrichment, with thirteen sites S1, S2, S3, S5, S6, S7, S12, S23 to S27, and S29 showing moderate enrichment (class 1) and the remaining seventeen sites having significant enrichment (class 2).

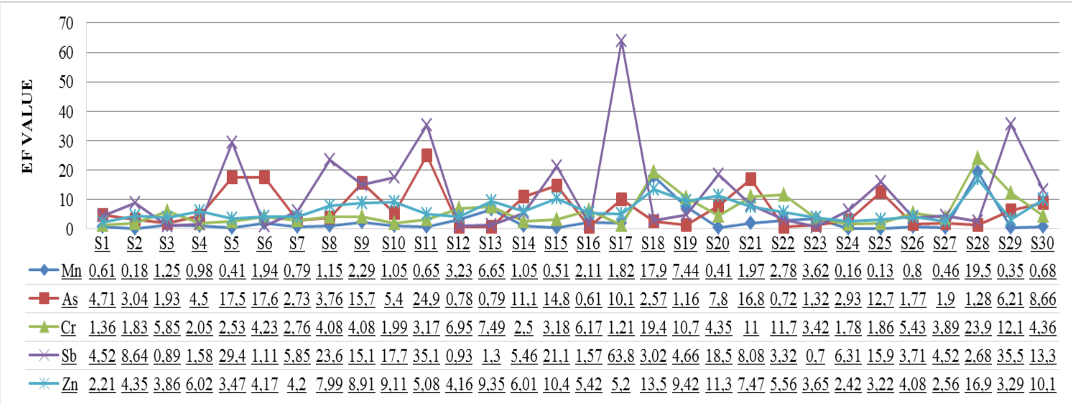


***Figure 3.** EF values calculated for the sites*

4. Conclusions

In this study, the levels of 5 potentially toxic trace elements in 30 soil samples from Dong Nai were analyzed by Instrument neutron activation analysis. The result showed that based on the results of the Igeo, it can be concluded that the study area has varying levels of contamination of the five potentially toxic trace elements, namely manganese (Mn), arsenic (As), chromium (Cr), antimony (Sb), and zinc (Zn). The contamination levels range from uncontaminated to moderately to heavily contaminated for different sites, with Sb showing the highest contamination levels, followed by Zn, Cr, As, and Mn. Based on the results of EF, the results showed varying levels of enrichment for each element, with Sb exhibiting the highest mean enrichment factor value, followed by As, Zn, Cr, and Mn.

The elements may be in the soil because of natural processes, like rocks breaking down over time, or because people do things that cause pollution. It is crucial to distinguish between natural and man-made elements and know that natural background values vary based on location and study size. We need to monitor the concentration of elements in the soil throughout time to find out how much of each element comes from nature and how much comes from people. This will help determine human-caused pollution and soil quality. This study covered only five potentially toxic trace elements and temporarily compared the earth's crust element values. More trace elements other than those considered in this study should be analyzed to provide comprehensive data on the element profile of soil.

Overall, this study provides important data on soil pollution levels in Dong Nai province, which can be used as a reference for land treatment, usage, and management in building and development projects. The results of this study can also help inform policymakers and stakeholders about the current state of soil quality in the region and guide future efforts to mitigate soil pollution.

❖ ***Conflict of Interest:*** *Authors have no conflict of interest to declare.*

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ĐÁNH GIÁ MỘT SỐ NGUYÊN TỐ VI LƯỢNG TIỀM NĂNG ĐỘC HẠI NGUY HIỂM TRONG CÁC MẪU ĐẤT BỀ MẶT CỦA TỈNH ĐỒNG NAI BẰNG PHƯƠNG PHÁP PHÂN TÍCH KÍCH HOẠT NEUTRON

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TÓM TẮT

Mục đích của nghiên cứu này là điều tra mức độ ô nhiễm các nguyên tố vi lượng có khả năng gây độc trong đất của tỉnh Đồng Nai, Việt Nam. Để đạt được điều này, phương pháp Phân tích kích hoạt Neutron (INAA) đã được sử dụng để đo nồng độ của năm nguyên tố vi lượng có khả năng gây độc hại, bao gồm mangan (Mn), asen (As), crom (Cr), antimon (Sb) và kẽm (Zn), trên 30 mẫu đất được thu thập từ khu vực. Sau đó, chỉ số tích tụ địa chất (Igeo) và hệ số làm giàu (EF) được sử dụng để đánh giá sự tồn tại của các chất gây ô nhiễm do con người gây ra và cường độ lắng đọng của chúng trên bề mặt đất. Kết quả của Igeo cho thấy các mức độ ô nhiễm khác nhau đối với từng nguyên tố được điều tra, với Sb cho thấy mức độ ô nhiễm cao nhất, tiếp theo là Zn, Cr, As và Mn. Phân tích EF cũng chứng minh rằng giá trị hệ số làm giàu trung bình là cao nhất đối với Sb, tiếp theo là As, Zn, Cr và Mn. Kết quả của nghiên cứu này cung cấp dữ liệu cơ bản quan trọng cho nghiên cứu trong tương lai về ô nhiễm đất ở tỉnh Đồng Nai và các khu vực công nghiệp hóa nhanh chóng khác. Hi vọng rằng những phát hiện này sẽ đóng vai trò là nguồn tài nguyên quý giá cho các nhà hoạch định chính sách và quản lí môi trường đang tìm cách giảm thiểu rủi ro liên quan đến ô nhiễm đất và thúc đẩy các hoạt động sử dụng đất bền vững.

***Từ khóa*: EF; Igeo; INAA; ô nhiễm đất; nguyên tố vi lượng**