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Original Article

Chemical Speciation, Risk Assessment, and Pollution Level of Lead Metals in Road Dust of some Industry Zones and Urban Areas in Northern Vietnam

Nguyen Thi Thu Thuy[1,*], Vuong Truong Xuan[1], Nguyen Thieu Le[2], Tu Binh Minh[2]

[1]TNU University of Science, Thai Nguyen University, Tan Thinh, Thai Nguyen, Vietnam
[2]VNU University of Science, 19 Le Thanh Tong, Hanoi, Vietnam

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Abstract: This study analyzed the chemical speciation of Pb metal in road dust samples from six provinces in northern Vietnam. Samples were extracted according to Tesser's sequential extraction procedure [22] and measured by the ICP/MS method, with the total recovery of Pb ranging from 90.9% to 107%. The analysis results showed that Pb existed mainly in the form of residual fraction F5 (38.6%) > organic fraction (29.0%), > Fe/Mn oxide fraction (19.2%) > exchange fraction, and carbonate fraction (5.00% and 8.32%, respectively). The pollution indices such as geological accumulation index (I_{geo}), individual contamination factor (ICF), and risk assessment index (RAC%) were utilized to evaluate pollution levels. The values of I_{geo} of Pb in samples ranged from 1.80 to 2.65, with a mean value of 2.24. The value of ICF varied from 0.60 to 2.87, with a mean value of 2.04. The values of RAC ranged from 4.47% to 23.2%, with a mean value of 13.3%. In general, the concentrations of Pb in most of the studied samples in the six provinces were classified as low pollution and risk levels according to Igeo, ICF, and RAC. However, in some provinces which have many industries producing electronic components, steel ore, and densely populated urban areas, the pollution level of Pb was from moderate to large, potentially affecting the environment of the studied areas.

Keywords: Chemical speciation, geological accumulation index, individual contamination factor, risk assessment index.

1. Introduction

Currently, urbanization is a global phenomenon, constantly accelerating leading to the emission of pollutants into the environment. Heavy metals are one of the most widely studied groups. Metals can be trace elements (copper, zinc,…) needed to maintain the human body's metabolism. However, they can lead to poisoning at higher concentrations. Heavy metals can be emitted from human activities and industrial products such as current and

* Corresponding author.
E-mail address: Thuyntt@tnus.edu.vn

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former mining operations, foundries, furnaces, and diffusion sources such as pipelines, by-products, combustion by-products, transportation, industry [1, 2]. Road dust contains high levels of heavy metals and many persistent organic pollutants such as PCBs, PAHs, Dioxin which lead to environmental pollution. Road dust includes both natural and manufactured particles. Particles in nature are derived mainly from minerals in the soil and gases.

In contrast, artificial particles come from human activities such as road construction (cement, asphalt, and road paint), automobiles, rubber dust, brake dust, rusted iron), industrial inputs (iron ore, plastic,...), or atmospheric deposition and natural geology [3, 4]. Many studies have also identified and evaluated the distribution and environmental implications of total concentrations of metals in urban street dust [3, 5, 6]. However, the toxicity of heavy metals is the greatest when they exist in potential mobile phases in the environment. Therefore, They can enter the human body through the food chain. While heavy metals in residual fractions are difficult to dissolve in water, so they have little impact on human health. Chemical speciation of heavy metals has been popularly carried out in many studies so far; however, they focus mainly on sediments of lakes, rivers [7, 8], and seas, or soil [9] and sludges [1, 10]. Some studies in China, Iran have assessed the risks to human health when exposed to metal in mobile phases of road dust samples, such as Tokalıog˘ lu et al., 2006; Yıldırım et al., 2016, Keshavarzi et al., 2015 [2, 11, 12].

In Vietnam, many studies assessed the pollution level of heavy metals, but most were assessed in soil, water, sediment, and plant. Authors V, D. Loi and P. T. T. Ha [8, 13, 14], analyzed and assessed the pollution level of heavy metals in Cau river sediments in Thai Nguyen province and Tri An lake, or author Pham Kim Phuong (2008), author Dieu et al., 2017, analyzed heavy metals in sediments and organisms [15, 16].

Thus, it can be seen that there are no specific statistics on the safety threshold of heavy metals in the sample of road dust, as well as the pollution level of heavy metals in the road dust in industrial zones of some provinces in northern Vietnam.

In this paper, the chemical speciation of lead in road dust was investigated using Tessier's sequential extraction procedure [17]. This procedure is widely utilized in analyzing the fractions of heavy metals in solid samples such as soil and sediment. Lead is a toxic substance capable of inhibiting the activity of many enzymes. The way lead enters the body depends on its chemical and physical form. Inorganic lead is absorbed mainly by inhalation and digestion without undergoing biological exchange. Lead combining with organic compounds enters the body mainly by contact with the skin and respiratory tract. Then it is metabolism in the liver. Symptoms of lead poisoning are fatigue, tremors, headache, nausea, seizures, the appearance of blue-black lines on gum tissue, and abdominal pain. Lead also interferes with hemoglobin synthesis and most seriously interferes with kidney function. Lead also affects the viability and development of the fetus [18].

2. Methodology

2.1. Sample Collection and Preparation Road Dust

Six samples in the study were taken at the road in front of industrial plants in the areas of Thai Nguyen (TN - Diem Thuy and Yen Binh industrial park), BacNinh (BN - Yen Phong 1, 2C), Bac Giang (BG-Van Trung industrial park), Quang Ninh (BN - coal mining Ha Long); Hai Phong (HP - road area in front of Hai Phong Vsip; Hanoi (HN - Road axis Thanh Xuan, Nguyen Trai). Each dust sample consists of multiple samples collected on-road sections, then mixed into a bulk sample of approximately 300g of dust. Samples were collected by using polyethylene brushes at a distance of 0.5 to 1 m from the edge of the pavement, 5 to 10 m each. All dust samples were stored in sealed polyethylene bags, labeled, and then transported to the laboratory.

2.2. Sequential Extraction Procedure (SEP) in Road Dust

In this study, Tessier sequential extraction procedure was used to analyze the chemical speciation of lead in road dust. This process consists of four stages and five fractions: exchangeable fraction (F1); carbonate-associated fractions (F2); reducible fraction or fraction associated with Fe and Mn oxides (F3); oxidizable fraction (F4, fraction bound to organic matter and a residual fraction (F5). The total concentration of Pb (in 1g road dust) was analyzed as a residue fraction in Tessier sequential extraction procedure [17]. The blank was processed according to the actual sample analysis procedure, using 1ml of distilled water instead of 1g of the dry sugar dust sample. Samples were repeated three times. Total concentrations and chemical speciation of the Pb in road dust were determined by the ICP/MS method. Recovery of Pb through the sequential extraction procedure was performed by the sum concentration of the five fractions (F1, F2, F3, F4, and F5) compared with the pseudo-total concentrations of Pb in road dust.

2.3. Contamination Assessment Methods

Some environmental indexes such as Igeo, ICF, and RAC were applied to assess the pollution level of Pb in road dust for the ecological environment [3, 6, 19-21].

I_{geo} (Geoaccumulation index) is an index to evaluate the level of pollution in road dust, with the following formula:

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n} \quad (1)$$

Where: Cn is the content of Pb in the sample; Bn: background value of Pb in soil [3, 6] 1,5: is the constant used to neutralize variations due to lithogenic actions. In this study, we use the background value of Pb in soil from China National Environmental Monitoring Center [22].

Individual contaminant factor (ICF) was used the following equations [20] :

$$ICF = \frac{F_1 + F_2 + F_3 + F_4}{F_5} \quad (2)$$

The influence of the IFC index is classified into the following levels: low (< 1); medium (1-3); high (3-6); very high (> 6) [21].

RAC (Risk Assessment Code) is an indicator to assess the ability of metals in road dust to release into the environment, and lead to the risk of accumulation of heavy metals in the human body [20]. In road dust, in exchangeable fraction (F1), carbonate-associated fractions (F2) metal bond is relatively weak, so it is easy to release into the environment and enter the human body through many contact routes, thus causing toxicity for the environment and humans [23]. The RAC index is calculated as follows:

$$RAC\ \% = \%F1 + \%F2 \quad (3)$$

Assessing the level of risk index RAC%: low (< 10%); medium (10 - 30%); high (30 - 50%); very high (> 50%).

3. Results and Discussion

3.1. Chemical Partitioning of Pb in Road Dust

Chemical partitioning and concentrations of the five fractions for Pb obtained by the SEP were shown in Tables 1 and Figure 1.

Recovery of Pb in the sum of the five fractions was from 90.9 to 107%. The results show that the difference with the pseudo-total concentrations of Pb was not more than 10%. Thus, the analytical method shows accurate results. From the analysis results, it can be seen that Pb was presented mainly in residues fractions (F5).The concentrations (%) of Pb in five fractions were in the following order: residues fractions (F5, 38.6%) > fraction bound to organic matter (F4, 29.0%) > fraction associated with Fe and Mn oxides (F3, 19.2%) > carbonate-associated fractions (F2, 8.32%) > exchangeable fraction (F1, 5.00%). The Pb percentage in F4 was equivalent to the results obtained in the studies of Tokalıogˇ lu and et al., 2006 (29.7%) [2]; Yıldırım and et al., 2016 (28.0%) [4]. Lead was present high in F4 because lead can form strong complexes with organic compounds found in road dust.

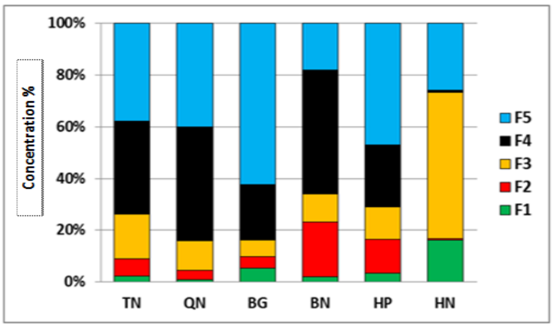

Figure 1. The chemical fractionation patterns of *Pb in the road dust.*

The mobility concentration of Pb based on the sum of the proportions of exchangeable, carbonate-associated, Fe and Mn oxides; and oxidizable fractions for six road dust samples was ranged from 11.3 to 32.4 mg/kg. The highest concentration of Pb in TN (with 60% in mobility phases) and the lowest in HP. The concentration of Pb in this study was lower than that in Li et al., 2013 [20]. It can be seen in industrial parks with many factories producing electronics and building materials such as TN, BN, and urban areas such as Hanoi are significant sources of emissions of Pb in the mobility phase. This result can lead to health risks to the population of the study sample area.

Table 1. Concentrations of Pb by Tessier's sequential extraction method

Samples	Concentrations of Pb in five fractions								
	F1	F2	F3	F4	F5	Mobility phase (F1+ F2 + F3 + F4)	Total concentration of five fractions	Pseudo-total	Recovery (%)
TN	1.19	3.46	8.98	18.8	19.7	32.4	52.1	48.8	107
QN	0.05	0.20	0.64	2.43	2.23	3.31	5.55	6.10	90.9
BG	0.80	0.64	0.98	3.18	9.26	5.60	14.9	15.7	94.6
BN	0.41	4.55	2.31	10.3	3.86	17.5	21.4	23.5	91.0
HP	0.06	0.24	0.23	0.44	0.87	0.98	1.84	1.76	105
HN	1.70	0.05	5.95	0.09	2.72	7.80	10.5	10.5	99.1
Mean	0.70	1.52	3.18	5.86	6.45	11.3	-	-	-
SD	0.66	1.96	3.52	7.31	7.13	10.8	-	-	-
CV %	93.9	128	110	124	110	95.8	-	-	-

3.2. Environmental Implications of Pb in Road Dust

i) Geoaccumulation Index: I_{geo}

I_{geo} values for Pb were ranged from 1.80 to 2.65 (Figure 2). The highest value was found in the province TN with many industrial zones related to the production of iron and steel, electronic components, and urban areas with a large population and many means of transport (HN). The average values were ranged from 1 to 2 for the QN, BN, HP provinces, so the pollution level is medium. Elsewhere, I_{geo} values were the pollution level from medium to high (Igeo > 2). Thus, the risk of Pb metal pollution can be seen in areas with many industries and urban areas with high population density.

ii) Individual contaminant factor (ICF)

To test the relative retention time of Pb in road dust, we calculated the individual pollution factor (ICF). A higher metal contamination factor indicates a lower retention time and a higher risk to the local environment. The average IFC value of Pb was 2.04, ranging from

0.60 to 2.87 (Figure 2). Most of the values in the provinces were from 1 to 3, indicating low or moderate influence. However, the ICF value of Pb in BN is pretty high at 4.54, that their retention time and mobility in road dust is high, a possible risk to the local environment [19].

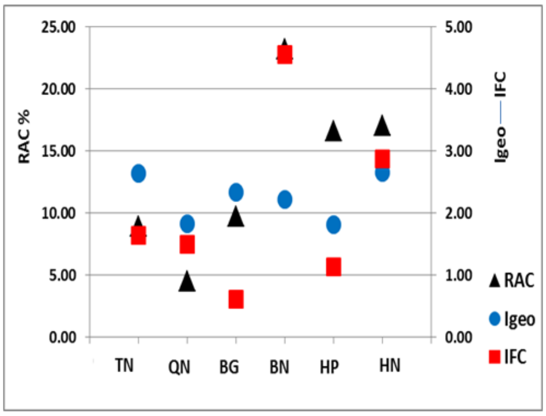

Figure 2. Igeomate, IFC, and RAC index of Pb in road dust.

iii) Risk Assessment Code - RAC

The average RAC value of Pb was 13.3%, ranging from 4.47 to 23.2%. As shown in Figure 2, the RAC value in provinces decreased in the order: BN > HN > HP > BG > TN > QN. This result was consistent with the fact in the sampling area. QN province was in the coal mining area, the metal presented more in organic matter and residual fraction, so the RAC value was the lowest among the six provinces. According to the risk assessment criteria, the RAC values were low to medium, and the influence level was insignificant.

4. Conclusion

In this study, chemical speciations of Pb in road dust from 6 northern provinces of Vietnam (TN, QN, BN, BG, HP, HN) were determined. The analysis results showed that Pb was present in the most residual fractions (F5, 38.6%), and the lowest in exchangeable (F1, 5.00%), carbonate-associated fractions (F2, 8.32%). On the other hand, the percentage of lead in the total mobile phases is relatively high (with 60% in TN). We have also assessed pollution levels as well as risks to the environment and ecosystems through Igeo (1.80 - 2.65), ICF (0.60 to 2.87), and RAC indexes (4.47 to 23.2%). In general, the pollution levels of Pb in six provinces were low or medium. However, in some areas with industrial production related to electronic components or iron and steel (BN, TN) or in densely populated urban areas (HN), the pollution level levels were high. Therefore, it could be concluded that the risk of lead metal pollution in road dust of some industrial parks and large urban areas.

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