

# Elemental Profiling and Safety Assessment of Four Spice Vegetables: Insights into Nutritional and Toxicological Implications

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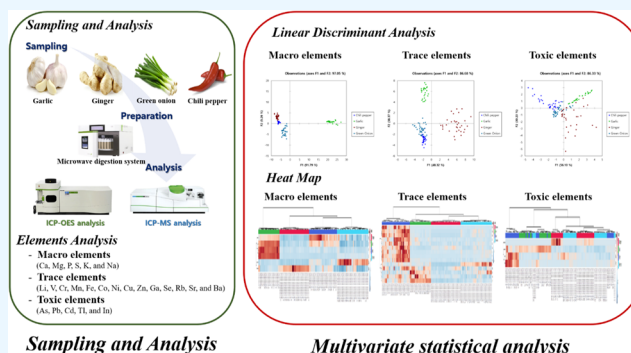


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**ABSTRACT:** Spiced vegetables are consumed globally, appreciated for their distinctive aromas and flavor profiles. Additionally, their unique elemental composition is recognized as a crucial parameter. This study aimed to evaluate the safety and toxicological risk of four commonly consumed spice vegetables: garlic (*Allium sativum*), ginger (*Zingiber officinale*), green onion (*Allium fistulosum*), and chili pepper (*Capsicum annuum*). Using ICP-OES and ICP-MS, a total of 25 elements, including macro, trace, and toxic elements, were determined. The analytical methods were validated per AOAC guidelines. Results indicated that potassium was the most abundant element across all samples, with garlic showing significantly higher concentrations of phosphorus, sulfur, and potassium. Trace element analysis revealed zinc and manganese as the most prevalent, with ginger exhibiting elevated levels of manganese, barium, and gallium. While toxic elements were detected in all samples, their concentrations remained within safe limits for human consumption. The estimated daily intake (EDI) and hazard quotient (HQ) analysis, based on Korean dietary data, confirmed minimal health risks. Heatmap analysis and linear discriminant analysis plots demonstrated the potential to distinguish between the four spice vegetables based on element profiles. This study highlights the importance of monitoring elemental compositions in spice vegetables to ensure food safety and reduce health risks from toxic elements, offering crucial insights into their nutritional value and safety.



## 1. INTRODUCTION

Garlic (*Allium sativum*), ginger (*Zingiber officinale*), green onion (*Allium fistulosum*), and chili pepper (*Capsicum annuum*) are essential spice vegetables used in cuisines worldwide.<sup>1,2</sup>

Garlic is a staple ingredient of many cuisines, including those of the Mediterranean, Asian, and Middle Eastern. It has a strong flavor and aroma and is used in various dishes,<sup>3</sup> providing numerous health benefits, such as reducing cholesterol levels and boosting the immune system.<sup>4</sup> Ginger is particularly popular in Asian cooking, where it is used as a food additive. It is known for its medicinal properties, for reducing inflammation and relieving nausea.<sup>5</sup> Green onions, also known as scallions, are a type of onions that are harvested before they fully mature. They are used to garnish the such as soups and salads and act as flavoring agents. It is popular in Chinese, Korean, and Mexican cuisines.<sup>6</sup> Chili peppers have several varieties and are used in many cuisines globally. They are known for their heat and spiciness and are used in chili, curry, and salsa dishes. They exhibit various health benefits, which include reducing inflammation and boosting metabolism.<sup>7</sup> These four spices are crucial for adding flavor and depth to dishes and imparting several health benefits. They are staples in many cuisines worldwide and will likely remain valuable for years to come.<sup>8</sup>

These four spice vegetables are not a major source of element because they are consumed in small quantities. However, they have trace amounts of specific elements. For example, garlic contains small quantities of elements such as Se, Mn, and Ca; ginger contains K, Mg, and P; and green onion contains Cu, Mn, and P.<sup>9–11</sup> Chili pepper is a good source of K, Mn, and Fe, and contains small quantities of Cu, Mn, and P as well.<sup>12</sup> These spices might not be substantial sources of nutritionally significant elements; however, they are valuable additions to a healthy diet owing to their numerous health benefits, including the anti-inflammatory and antioxidant properties.<sup>13</sup>

The content of elements in vegetables depends on the soil quality. Nutrient-rich soils facilitate plant growth. However, soils contaminated through industrial activities and waste sources may accumulate hazardous elements such as Pb, Cd, and As, thereby posing significant health risks to humans

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including lung cancer, kidney dysfunction, hypertension, and neurological issues.<sup>14,15</sup> Therefore, the safe levels of these elements in food have been globally established.<sup>16</sup> Pb exposure can damage the nervous system, whereas Cd is toxic to the skeletal and renal systems, which can cause cancer.<sup>17,18</sup> Cd is carcinogenic, and its toxicity varies based on its form and oxidation state.<sup>19</sup> Inductively coupled plasma-optical emission spectrometry (ICP-OES) and inductively coupled plasma-mass spectrometry (ICP-MS) have the advantages of selectivity, sensitivity, and multielement analysis capabilities.<sup>20</sup>

This study analyzed macro, trace, and toxic elements in four types of spice vegetables: garlic, ginger, green onion, and chili pepper using microwave digestion. In addition, the analytical methods were validated by assessing the limits of detection (LOD), limit of quantification (LOQ), accuracy, precision, and linearity. Emphasis was placed on analyzing toxic elements to assess potential health risks. Heatmap analysis and linear discriminant analysis (LDA) plots were employed to compare the quantities of all elements, highlighting the distribution and concentrations of toxic elements specifically. This comprehensive analysis aims to evaluate their potential impact on human health, ensuring food safety and mitigating risks associated with toxic element exposure.

## 2. MATERIALS AND METHODS

**2.1. Instrumentation and Analysis.** The macro elements (Mg, P, S, Ca, K, and Na) were analyzed using an ICP-OES model Optima8000 (PerkinElmer, CT). Trace (Li, Be, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Sr, and Ba) and toxic elements (As, Pb, Cd, Tl, and In) were analyzed using an ICP-MS model 300D (PerkinElmer Sciex, CT). The instruments were optimized by scanning blank, standard, and samples in the programmed mass value and wavelength range. The background correction mass values and wavelengths were specifically selected for each analyte peak at the appropriate positions. Instrument drift was managed by running a multielement standard solution at intervals of once every 10 analyses. The operating parameters and conditions for the ICP-OES and ICP-MS analyses are detailed in [Supplementary Table S1](#).<sup>21</sup>

**2.2. Reagents.** Ultrapure deionized water with an 18.2 MΩ·cm resistivity was obtained using a Milli-Q Plus water purification system (Millipore, Bedford, MA). Analytical reagent-grade, concentrated 70% (v/v) HNO<sub>3</sub> (Dong Woo Fine-Chem, Iksan, Korea) was used. A multielement standard solution containing all the elements at 10 mg/L concentration (Kriat Co., Daejeon, Korea) was used. Standard calibration solutions were prepared by diluting the stock solution with 1% (v/v) HNO<sub>3</sub>, which is the same percentage as that in the samples. All glass/plastic were soaked overnight in 20% (v/v) HNO<sub>3</sub> and rinsed several times with deionized water. The plasma torch was supplied with Ar at a purity level exceeding 99.999% to ensure optimal analytical performance.

**2.3. Sample Preparation and Digestion.** A total of 140 samples of four types of spice vegetables (garlic (25), ginger (33), green onion (37), and chili pepper (35)) were collected from online and offline markets in South Korea. To homogenize the samples, more than 500 g of each was first processed. Subsequently, about 5 g of the homogenized sample was placed into a microwave-closed PTFE vessel for decomposition. For the decomposition process, 1.0 mL of 30% H<sub>2</sub>O<sub>2</sub> and 7.0 mL of concentrated 70% HNO<sub>3</sub> were added to each vessel. The microwave decomposition temper-

ature was gradually increased, starting from 80 °C for 5 min to 120 °C for 5 min, 150 °C for 5 min, 180 °C for 20 min, and subsequently, cooled to 40 °C. After cooling, the tube contents were set to 50.0 g in 50 mL polypropylene volumetric tubes.<sup>22</sup>

**2.4. Quality Assurance.** Calibration curves were constructed using five different concentrations of the standard solution and a blank for each target element. The concentrations of the elements in the samples were within the linear range of the calibration curves and above the established lower limit of linearity. The LOD and LOQ were calculated to be three and ten times that of standard deviation, respectively, obtained by determining 10 replicates of the blank divided by the slope of the analytical curve. Linearity was evaluated by constructing calibration curves for all analyte elements using a nonweighted least-squares linear regression analysis and calculating the corresponding correlation coefficients (R<sup>2</sup>). Precision was determined as the percentage coefficient of variance (CV%), which was calculated by measuring the relative standard deviation of ten replicates of one sample. Recovery experiments were conducted using two different standard concentrations, which are the same as that of the salt samples, to verify the quality control. The accuracy of the method was validated by spiking the standard samples at three different levels: low, medium, and high, with varying concentrations of the macro, trace, and toxic elements. The low level had concentrations of 5 mg/100 g for macro elements and 0.1 mg/kg for trace and toxic elements, whereas the medium and high levels had concentrations of 50 and 300 mg/100 g for macro elements and 1 and 5 mg/kg for trace and toxic elements, respectively.<sup>23</sup>

**2.5. Statistical Analyses.** The results were presented as mean ± standard deviation of triplicate measurements. One-way analysis of variance (ANOVA) and Tukey's honestly significant difference (HSD) tests were performed using SPSS software version 20 (IBM, New York) to analyze significant differences ( $p < 0.05$ ) between means. Different superscripted letters denote significant differences ( $p < 0.05$ ). LDA was performed to determine the similarities and differences between the four types of spices based on the concentrations of macro, trace, and toxic elements. A data matrix with elements (determined elements) and rows (analyzed samples) was created for the analysis using XLSTAT 2020 software (Addinsoft, Paris, France) following the method of Pilgrim, Watling & Grice.<sup>24</sup> A heatmap was generated using GraphPad Prism v7.02 (GraphPad Software).<sup>25</sup>

## 3. RESULTS AND DISCUSSION

**3.1. Validation of Analytical Method.** A total of 25 elements, including macro (Mg, P, S, Ca, K, and Na), trace (Li, Be, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Sr, and Ba), and toxic elements (As, Pb, Cd, Tl, and In), were analyzed in 140 samples from 4 types of spice vegetables. The validation of the analysis method used in the study included the measurement of linearity, LOD, LOQ, and relative standard deviation % (RSD, %) for each ICP-OES and ICP-MS method, and the results were presented in [Table 1](#).

The macro elements were analyzed using ICP-OES, wherein the linearity, RSD of the standard solutions, LOD, and LOQ of the analyzed elements ranges between 0.9989 (Mg) to 0.9996 (S), 1.4 (Ca) to 2.2% (S), 0.081 (K) to 0.106 mg/kg (Na), and 0.243 (K) to 0.318 mg/kg (Na), respectively. The trace and toxic elements were analyzed using ICP-MS, and linearity of the calibration curve, RSD, LOD, and LOQ range from

**Table 1. Analytical Figures of Merit and Calibration Parameters**

	$R^{2a}$	LOD <sup>b</sup>	LOQ <sup>c</sup>	RSD <sup>d</sup>
Macro Elements				
Ca	0.9991	0.097	0.291	1.4
Mg	0.9989	0.083	0.249	1.7
P	0.9992	0.086	0.258	2.0
S	0.9996	0.091	0.273	2.6
K	0.9993	0.081	0.243	1.4
Na	0.9989	0.106	0.318	1.8
Trace Elements				
Li	0.9999	0.269	0.807	2.1
V	0.9998	0.357	1.07	2.7
Cr	0.9999	0.258	0.774	2.4
Mn	0.9998	0.354	1.06	2.1
Fe	0.9999	0.249	0.747	3.7
Co	0.9996	0.343	1.029	2.0
Ni	0.9998	0.276	0.828	3.1
Cu	0.9996	0.377	1.13	2.3
Zn	0.9998	0.372	1.12	2.2
Ga	0.9998	0.296	0.888	2.6
Se	0.9997	0.286	0.858	3.0
Rb	0.9999	0.319	0.957	3.1
Sr	0.9993	0.370	1.11	2.3
Ba	0.9998	0.288	0.864	3.6
Toxic Trace Elements				
As	0.9993	0.226	0.678	2.5
Cd	0.9996	0.156	0.468	2.1
Pb	0.9998	0.233	0.699	3.4
Tl	0.9996	0.129	0.387	3.0
In	0.9998	0.106	0.318	2.4

<sup>a</sup> $R^2$  = Correlation coefficient. <sup>b</sup>LOD = Concentration unit of macro elements is mg/100 g, trace elements and toxic trace elements are ug/kg. <sup>c</sup>LOQ = Concentration unit of macro elements is mg/100 g, trace elements and toxic trace elements are ug/kg. <sup>d</sup>RSD = Relative standard deviation (%).

0.9993 (Sr) to 0.9999 (Li, Cr, Fe, and Rb), 2.0 (Co) to 3.7% (Fe), 0.106 (In) to 0.377  $\mu\text{g/kg}$  (Cu), and 0.318 (In) to 1.131  $\mu\text{g/kg}$  (Cu), respectively.

The recovery percentages of the standard sample spiking experiments were listed in Table 2. The recovery percentages for the macro elements in garlic range between 90.8 (S) to 98.0% (K), whereas for trace and toxic elements, the range is 90.2% (Sr) to 104% (V). For ginger, green onion, and chili pepper, the recovery percentages of the macro elements ranged from 95.8% (S) to 99.2% (K), 95.8% (Ca) to 102% (K), and 91.3% (S) to 103%, respectively. The recovery percentages of trace and toxic elements ranged from 90.2% (Sr) to 104% (V) for garlic, 92.0 (Ga) to 99.2% (As) for ginger, 91.7 (Ni) to 105% (Pb) for green onion, and 92.7 (Mn) to 103.3% (Cd) for chili pepper. All validation parameters were compliant with AOAC guidelines.

**3.2. Macro Elements.** Table 3 lists the macro elements (Ca, Mg, P, S, K, and Na) in four types of spice vegetables. K was the most abundant component in all the samples, whereas the order of abundance varied as follows: (K > P > S > Mg > Ca > Na), (K > P, Mg > Ca > Na > S), (K > Ca > P > Mg > S > Na) and (K > P > Mg > Na, Ca > S) for garlic, ginger, green onions, and chili peppers, respectively (Figure S1(A)).

K is crucial for maintaining fluid balance, muscle contractions, and nerve functions in the human body.<sup>26</sup> The

concentration of K was highest in all samples, with an average content of 537, 352, 208, and 179 mg/100 g in garlic, ginger, chili pepper, and green onion, respectively. S is essential for various bodily functions such as skin health, joint function, and detoxification. It is anti-inflammatory and enhances the body's ability to eliminate toxins.<sup>27</sup> The S quantity in garlic was exceptionally high at 61.4 mg/100 g, which is 6.8 to 18.1 times higher than in other samples. Therefore, garlic is valued for its health benefits and is widely used in various cuisines and medicinal practices. Furthermore, the phosphorus content (151 mg/100 g) in garlic is approximately five times higher than that in the other analyzed samples. Phosphorus plays a critical role in energy production, cell signaling processes, bone formation, and maintenance in the body.<sup>28</sup> However, while phosphorus is essential for various physiological functions, its bioavailability can be limited depending on its chemical form. A significant portion of phosphorus in garlic is present in the form of phytate, a storage form of phosphorus in plants that has low bioavailability in humans due to its resistance to enzymatic hydrolysis.<sup>29</sup> In this form, phytate can act as an antinutrient by chelating essential metal ions such as Ca, Fe, and Zn, thereby reducing their bioavailability and absorption in the intestine.<sup>30</sup> Ginger, green onion, and chili pepper exhibited higher concentration of Mg (34.1 mg/100 g), Ca (55.1 mg/100 g), and Na (10.7 mg/100 g), respectively. The results of this study on ginger are similar to those reported by Tabbassum et al.<sup>31</sup> Mg is crucial for muscle and nerve function, Ca is essential for strong bones and teeth, and Na helps maintain fluid balance and nerve function.<sup>32</sup>

**3.3. Trace Elements.** Table 4 shows the results for the 15 trace elements (Li, Be, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, Se, Rb, Sr, and Ba) in four types of spice vegetables. Zn and Mn are the most abundant elements in all the analyzed samples. In the case of garlic, the composition of Zn was higher than in other groups, while for ginger, the Mn composition was elevated compared to the other groups (Figure S1(B)). Zn is essential for immune function and wound healing, whereas Mn is essential for bone health and acts as an antioxidant.<sup>33</sup> Garlic has the highest Zn content at 8.18 mg/kg, which is 1.8 to 4.2 times greater than that of the other samples. The abundance of elements follows the order: Zn > Mn > Cu > Rb > Sr > Ba > Fe, Cr, Ni > Se, Ga > Co, V, Li > Be. This pattern is consistent with the findings reported by D'archivio et al.<sup>34</sup> The order of trace element concentrations in ginger was Mn > Zn > Ba > Rb, Cu, Fe > Sr, Cr, Ni > V, Ga > Li, Co, Se > Be, which is similar to that reported by Tabbassum et al.<sup>31</sup> Ginger shows remarkably higher levels of Ba and Ga at 2.31 and 0.156 mg/kg, which are 5.1 to 38.3 times and 5.2 to 52.1 times higher than those in the other samples, respectively. The quantity of Mn in ginger was exceptionally high at 31.4 mg/kg, which is 11.1 to 23.3 times higher than that of other trace elements in ginger. However, high levels of Ba exposure can cause breathing difficulties and muscle weakness, whereas Ga can lead to skin and eye irritation and long-term liver and kidney damage.<sup>35</sup> The order of abundance in green onion is Zn, Mn > Sr > Rb, Ba, Cu > Fe, Ni, Cr > Ga, V, Se, Li, Co > Be, with Zn, Mn, and Sr being the macro components. Sr can potentially improve bone density and decrease the risk of fractures.<sup>36</sup> Sr was highest in green onions at 1.57 mg/kg, 2.3 to 12.8 times higher than that in other samples. The order of abundance in chili pepper is Zn, Mn > Rb, Cu > Sr, Fe, Cr, Ni, Ba > Co > Se, Li, Ga, V > Be, with Zn and Mn being the macro components at 1.94 and 1.35 mg/kg, respectively.



**Table 2. Three Different Level of Accuracy Results on Four Kinds of Spices Vegetable<sup>a,b,c</sup>**

	garlic			ginger			green onion			chili pepper		
	low level	medium level	high level	low level	medium level	high level	low level	medium level	high level	low level	medium level	high level
Macro Elements												
Ca	92.2	96.4	96.1	97.6	96.1	98.1	96.1	95.8	99.3	93.7	91.8	98.9
Mg	93.5	97.3	96.4	98.0	96.4	98.2	96.3	96.8	99.9	94.3	92.2	99.6
P	92.3	95.4	96.1	97.6	96.1	98.1	96.2	97.1	99.4	93.8	91.8	99.8
S	90.8	95.4	95.8	97.1	95.8	97.9	95.9	96.4	98.6	93.0	91.3	99.3
K	97.4	98.0	97.3	99.2	97.3	98.7	96.9	101	102	96.3	93.5	103
Na	92.5	96.6	96.2	97.6	96.2	98.1	96.2	98.2	99.5	93.9	91.9	101
Trace Elements												
Li	97.1	99.8	101	99.1	97.2	98.6	96.9	103	102	96.1	95.1	94.7
V	93.5	97.3	104	95.2	94.5	97.3	95.0	99.2	95.8	94.4	97.9	101
Cr	96.5	99.4	101	96.6	95.5	95.8	95.7	98.0	97.9	95.8	95.9	103
Mn	90.2	95.0	99.6	96.9	95.7	97.9	97.8	95.8	98.3	92.7	96.8	98.9
Fe	94.3	97.8	94.3	98.2	96.6	98.3	96.5	99.2	94.3	94.7	98.2	101.3
Co	93.6	97.4	100	98.0	95.4	98.3	96.4	97.2	104	94.4	95.9	99.9
Ni	95.0	98.4	97.2	98.5	96.8	98.4	96.6	98.4	91.7	95.1	98.4	101
Cu	93.7	97.4	95.2	95.9	95.0	97.6	95.4	95.4	96.9	94.4	98.0	101
Zn	91.1	95.6	99.8	97.2	93.9	98.0	96.0	95.9	98.8	93.2	97.1	99.0
Ga	91.7	96.0	97.9	95.4	96.0	92.0	96.1	97.3	97.1	93.5	95.3	95.8
Se	92.9	96.9	93.4	97.8	96.3	98.2	96.3	94.8	99.7	94.1	97.7	94.0
Rb	96.0	99.1	94.7	98.8	97.0	94.5	96.7	99.5	101	95.6	98.8	102
Sr	90.2	95.0	96.2	96.9	95.7	97.9	95.8	95.7	98.3	92.7	96.8	98.9
Ba	93.7	97.5	103	98.0	96.5	98.3	96.4	97.1	100	94.5	98.0	96.8
Toxic Trace Elements												
As	97.3	99.9	103	99.2	97.3	98.6	96.9	101	102	96.2	99.2	102
Cd	93.5	97.3	96.4	98.0	96.4	98.2	96.3	99.2	99.9	94.3	97.9	103
Pb	102	103	102	95.6	98.2	96.1	97.6	105	104	98.3	95.6	103
Tl	91.9	96.2	97.0	97.5	96.1	97.1	96.1	96.7	99.2	93.6	97.4	99.6
In	95.1	98.4	96.8	98.5	96.8	98.4	96.6	98.5	101	95.1	98.4	97.8

<sup>a</sup>Low level: the standard was spiked onto the sample at 5 mg/100 g for macro elements and 0.1 mg/kg for trace and toxic elements. <sup>b</sup>Medium level: the standard was spiked onto the sample at 50 mg/100 g for macro elements and 1 mg/kg for trace and toxic elements. <sup>c</sup>High level: the standard was spiked onto the sample at 300 mg/100 g for macro elements and 5 mg/kg for trace and toxic elements.

**Table 3. Concentration (mg/100 g) of Macro Elements in Four Kinds of Spices Vegetable<sup>a</sup>**

	garlic	ginger	green onion	chili pepper
Ca	14.4 <sup>bc</sup> ± 2.4 10.9–21.0	19.5 <sup>b</sup> ± 6.5 9.6–31.7	55.1 <sup>a</sup> ± 16 20.8–89.5	9.4 <sup>c</sup> ± 1.8 6.3–14.4
Mg	26.8 <sup>b</sup> ± 2.2 22.3–31.1	34.1 <sup>a</sup> ± 6.0 24.2–48.1	17.2 <sup>c</sup> ± 5.0 10.6–31.2	15.5 <sup>c</sup> ± 1.2 12.7–18.5
P	151 <sup>a</sup> ± 17 131–206	34.6 <sup>b</sup> ± 6.3 23.4–46.7	29.9 <sup>b</sup> ± 8.6 15–52.9	33.0 <sup>b</sup> ± 4.3 26.3–43.7
S	61.4 <sup>a</sup> ± 4.1 55.8–72.3	3.4 <sup>c</sup> ± 0.8 2.2–5.3	9.0 <sup>b</sup> ± 1.9 5.3–12.4	4.7 <sup>c</sup> ± 0.6 3.8–6.4
K	537 <sup>a</sup> ± 45 440–614	352 <sup>b</sup> ± 54 209–435	179 <sup>d</sup> ± 67 91.6–367	208 <sup>c</sup> ± 19 160–239
Na	5.9 <sup>b</sup> ± 1.3 4.4–9.7	6.3 <sup>b</sup> ± 1.5 3.4–9.7	4.7 <sup>c</sup> ± 2.0 2.4–9.7	10.7 <sup>a</sup> ± 1.9 7.6–16.6

<sup>a</sup>Different superscript letters a–d indicate that values within a row differ significantly ( $p < 0.05$ ). For each element, data are presented as mean value ± standard deviation value in the first row and as minimum value–maximum value in the second row.

**3.4. Toxic Elements.** Table 5 shows the results for the five toxic elements (As, Cd, Pb, In, and Tl) in four types of spice vegetables. The composition ratios of toxic elements varied among the samples. In the case of garlic, Cd had the highest proportion, while for ginger, Pb was predominant. As levels were observed at a high ratio in green onions and chili peppers.

(Figure S1(C)). Exposure to As, Cd, Pb, In, and Tl results in various detrimental health effects, including skin lesions, cardiovascular diseases, and certain types of cancer.<sup>37</sup> Cd exposure can lead to kidney damage, lung disease, and osteoporosis,<sup>38</sup> whereas Pb exposure is associated with developmental delays in children, behavioral problems, and neurological damage.<sup>39</sup> Exposure to In and Tl can cause lung disease, cholesterol granulomas, gastrointestinal symptoms, hair loss, and neurological damage.<sup>40,41</sup> Pb was exceptionally high in ginger at 0.072 mg/kg, which is 7.3 to 42.2 times higher than that in other samples owing to the characteristics of ginger. Cd and Pb are detected in all the samples, with the order for concentration detection as Cd > As > Pb > In, Tl for garlic and Pb > As > Cd > In, Tl for ginger. A root vegetable that easily absorbs Pb components from the soil into its roots.<sup>42</sup> Voica et al. reported Pb, As, and Cd concentrations ranging from 0.192–3.010, 0.030–0.048, and 0.011–0.070 ug/g, respectively.<sup>43</sup> In comparison, our study shows lower levels of these elements overall. The order of detection is As > Pb > Cd > In, Tl, in green onion, and As > Cd > Pb > Tl > In in chili pepper. Pb was the highest in ginger among all samples, and should be below 0.3 mg/kg as set by Codex, EU, Australia, and China. All the samples evaluated in this study meet this standard and are confirmed to be harmless to humans. The estimated daily intake (EDI, mg/kg/day) of toxic elements was calculated using food intake data from all age groups based on the Korean Health Statistics 2021: Korea National Health and

Table 4. Concentration (mg/kg) of Trace Elements in Four Kinds of Spices Vegetable<sup>a</sup>

	garlic	ginger	green onion	chili pepper
Li	0.001 <sup>b</sup> ± 0.001 0.001–0.004	0.078 <sup>a</sup> ± 0.053 0.012–0.170	0.013 <sup>b</sup> ± 0.009 0.001–0.032	0.005 <sup>b</sup> ± 0.003 0.001–0.010
V	0.003 <sup>b</sup> ± 0.002 0.001–0.005	0.174 <sup>a</sup> ± 0.091 0.031–0.332	0.019 <sup>b</sup> ± 0.016 0.002–0.057	0.001 <sup>b</sup> ± 0.001 0.001–0.003
Cr	0.147 <sup>b</sup> ± 0.079 0.070–0.332	0.453 <sup>a</sup> ± 0.26 0.079–0.944	0.080 <sup>b</sup> ± 0.025 0.042–0.137	0.084 <sup>b</sup> ± 0.023 0.039–0.128
Mn	2.83 <sup>b</sup> ± 0.26 2.39–3.31	31.4 <sup>a</sup> ± 19.0 7.40–68.9	2.51 <sup>b</sup> ± 1.30 1.06–5.40	1.35 <sup>b</sup> ± 0.32 0.724–2.07
Fe	0.198 <sup>b</sup> ± 0.051 0.142–0.313	0.800 <sup>a</sup> ± 0.41 0.110–1.56	0.121 <sup>b</sup> ± 0.055 0.033–0.221	0.096 <sup>b</sup> ± 0.022 0.048–0.138
Co	0.005 <sup>c</sup> ± 0.002 0.002–0.008	0.049 <sup>a</sup> ± 0.027 0.012–0.118	0.010 <sup>bc</sup> ± 0.007 0.003–0.030	0.017 <sup>b</sup> ± 0.008 0.007–0.036
Ni	0.129 <sup>b</sup> ± 0.057 0.051–0.268	0.376 <sup>a</sup> ± 0.18 0.137–0.790	0.113 <sup>b</sup> ± 0.037 0.065–0.219	0.081 <sup>b</sup> ± 0.026 0.035–0.151
Cu	1.47 <sup>a</sup> ± 0.66 0.856–3.19	0.994 <sup>b</sup> ± 0.44 0.424–1.96	0.419 <sup>c</sup> ± 0.20 0.157–0.922	0.569 <sup>c</sup> ± 0.081 0.409–0.711
Zn	8.18 <sup>a</sup> ± 0.97 6.76–9.54	4.61 <sup>b</sup> ± 1.80 2.51–9.66	2.97 <sup>c</sup> ± 0.85 1.88–5.44	1.94 <sup>d</sup> ± 0.31 1.37–2.61
Ga	0.019 <sup>bc</sup> ± 0.007 0.005–0.031	0.156 <sup>a</sup> ± 0.082 0.046–0.351	0.030 <sup>b</sup> ± 0.025 0.003–0.078	0.003 <sup>c</sup> ± 0.001 0.001–0.005
Se	0.027 <sup>a</sup> ± 0.014 0.013–0.059	0.009 <sup>c</sup> ± 0.005 0.001–0.025	0.015 <sup>b</sup> ± 0.011 0.003–0.048	0.005 <sup>c</sup> ± 0.002 0.001–0.009
Rb	1.04 <sup>a</sup> ± 0.38 0.418–1.76	1.10 <sup>a</sup> ± 0.60 0.237–2.24	0.601 <sup>b</sup> ± 0.23 0.139–0.944	0.655 <sup>b</sup> ± 0.26 0.374–1.28
Sr	0.633 <sup>b</sup> ± 0.21 0.331–1.21	0.685 <sup>b</sup> ± 0.26 0.280–1.33	1.57 <sup>a</sup> ± 0.57 0.927–3.40	0.122 <sup>c</sup> ± 0.040 0.040–0.244
Ba	0.393 <sup>b</sup> ± 0.16 0.133–0.641	2.31 <sup>a</sup> ± 0.90 0.900–3.83	0.456 <sup>b</sup> ± 0.25 0.132–0.946	0.060 <sup>c</sup> ± 0.016 0.034–0.098

<sup>a</sup>Different superscript letters a–d indicate that values within a row differ significantly ( $p < 0.05$ ). For each element, data are presented as mean value ± standard deviation value in the first row and as minimum value–maximum value in the second row.

Table 5. Concentration(mg/kg) of Toxic Elements in Four Kinds of Spices Vegetable<sup>a</sup>

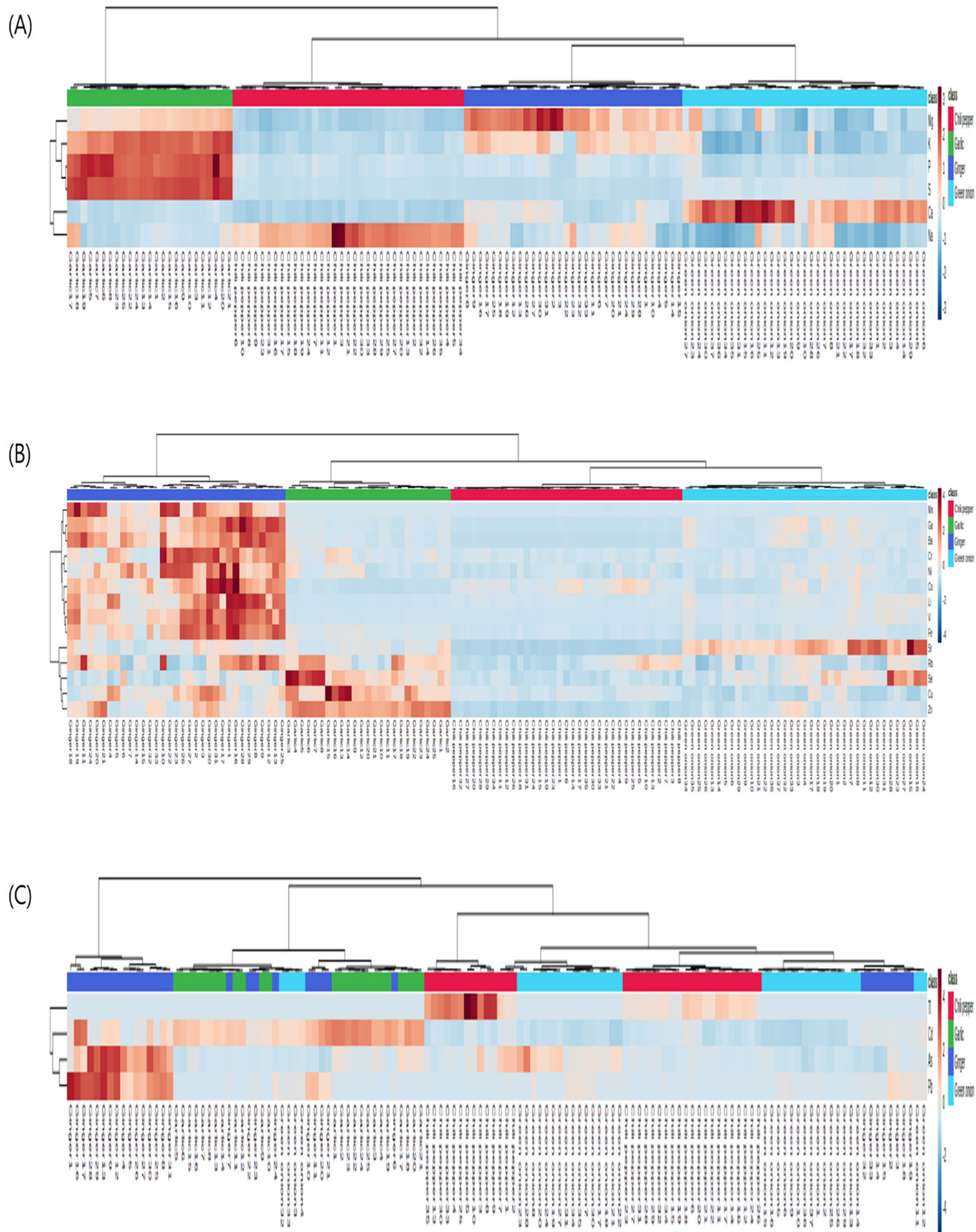
	garlic	ginger	green onion	chili pepper
As	0.012 <sup>b</sup> ± 0.004 0.006–0.022	0.023 <sup>a</sup> ± 0.02 0.003–0.057	0.013 <sup>b</sup> ± 0.008 0.003–0.042	0.012 <sup>b</sup> ± 0.006 0.007–0.028
Cd	0.029 <sup>b</sup> ± 0.007 0.017–0.039	0.019 <sup>b</sup> ± 0.01 0.008–0.044	0.006 <sup>c</sup> ± 0.005 0.001–0.021	0.007 <sup>c</sup> ± 0.003 0.003–0.014
Pb	0.002 <sup>b</sup> ± 0.001 0.001–0.004	0.072 <sup>a</sup> ± 0.06 0.010–0.182	0.010 <sup>b</sup> ± 0.008 0.001–0.027	0.003 <sup>b</sup> ± 0.002 0.001–0.009
Tl	ND <sup>b</sup>	ND <sup>b</sup>	ND <sup>b</sup>	0.001 <sup>a</sup> ± 0.001 0–0.003
In	ND <sup>b</sup>	ND <sup>b</sup>	ND <sup>b</sup>	ND <sup>b</sup>

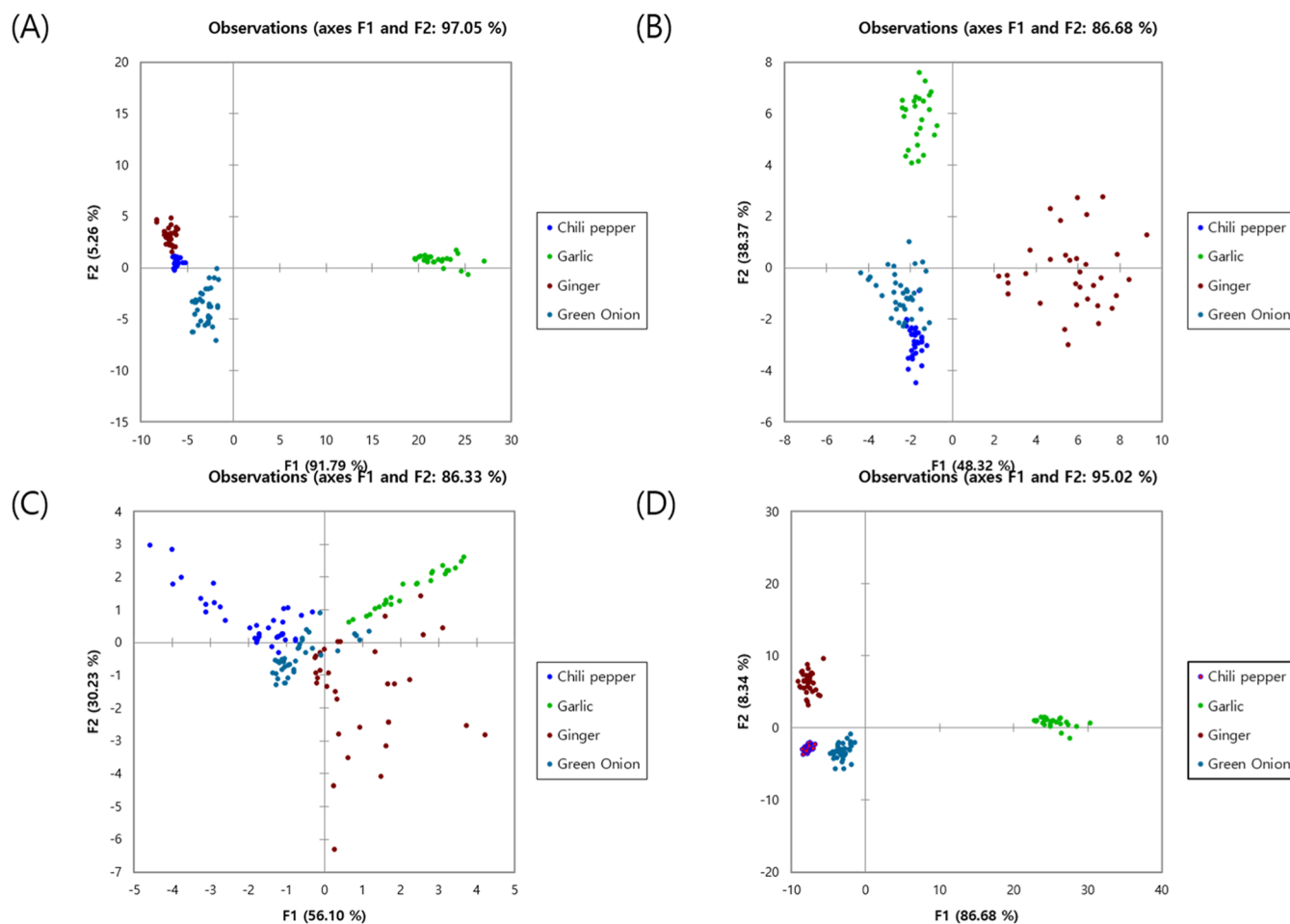
<sup>a</sup>Different superscript letters a–d indicate that values within a row differ significantly ( $p < 0.05$ ). For each element, data are presented as mean value ± standard deviation value in the first row and as minimum value–maximum value in the second row. <sup>b</sup>ND: Not detected.

Table 6. Risk Assessment of Toxic Elements: Estimated Daily Intake (EDI) and Hazard Quotient (HQ) in Four Spice Vegetables

		As		Cd		Pb		Tl		In	
	IR <sup>a</sup> (g/kg bw/day)	EDI <sup>b</sup> (mg/kg/day)	HQ <sup>c</sup>	EDI (mg/kg/day)	HQ	EDI (mg/kg/day)	HQ	EDI (mg/kg/day)	HQ	EDI (mg/kg/day)	HQ
garlic	0.50	6.03 × 10 <sup>−6</sup>	0.0201	1.46 × 10 <sup>−5</sup>	0.0292	1.00 × 10 <sup>−6</sup>	0.0003	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>
ginger	0.08	1.90 × 10 <sup>−6</sup>	0.0064	1.57 × 10 <sup>−6</sup>	0.0031	5.96 × 10 <sup>−6</sup>	0.0000	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>
green onion	1.03	1.3 × 10 <sup>−5</sup>	0.0446	6.17 × 10 <sup>−6</sup>	0.0123	1.03 × 10 <sup>−5</sup>	0.0000	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>	NA <sup>d</sup>
chili pepper	0.04	4.9 × 10 <sup>−7</sup>	0.0017	2.90 × 10 <sup>−7</sup>	0.0006	1.24 × 10 <sup>−7</sup>	0.0000	4.14 × 10 <sup>−8</sup>	0.0041	NA <sup>d</sup>	NA <sup>d</sup>

<sup>a</sup>IR(bw): body weight-adjusted intake rate (g/kg bw/day), up to the p99. <sup>b</sup>EDI (mg/kg/day)=C(avg) × IRbw, C(Avg)=Average elemental concentration in food (mg/kg). <sup>c</sup>HQ = EDI/RfD, RfD = Reference Dose (mg/kg/day). <sup>d</sup>NA: not available.





**Figure 2.** LDA plots of four kinds of spice vegetables for (A) macro elements, (B) trace elements, (C) toxic elements, and (D) total elements.

Nutrition Examination Survey (KNHANES VIII-3).<sup>44</sup> The hazard quotient (HQ) of toxic elements was evaluated using the reference dose (RfD, mg/kg/day).<sup>45</sup> Values suggested by the U.S. Food and Drug Administration (FDA) and the World Health Organization (WHO).<sup>46–48</sup> As a result, the HQ values for all four toxic elements were below 1, indicating a low likelihood of harmful effects from dietary exposure (Table 6). Other toxic elements, such as As and Cd, did not pose a threat to human health.

**3.5. Differentiation of Elements in Four Types of Spice Vegetables.** To confirm differences between the samples, the concentration of each sample was visualized as a heatmap, as given in Figure 1, based on the elements such as macro, trace, and toxic elements. The LDA plot results identify the differences between the sample groups, as presented in Figure 2.

The heatmap analysis in Figure 1(A) reveals that ginger and green onions are in a similar group, whereas garlic was the most independent in its position. This result corroborates with the LDA plot results, as shown in Figure 2(A).

The LDA plots based on the macro elements reveal that garlic was located independently in the positive direction of the X-axis, distinct from ginger, chili pepper, and green onion (Figure 2(A)). This is likely influenced by the elevated levels of K, P, and S. As the concentrations of K, P, and S increase, they independently contribute to higher values on the X-axis, and due to the high values of K, P, and S in garlic, it positions distinctly in this regard. Ginger and chili pepper are closely

distributed, whereas green onion is independent owing to their relatively high Ca content (Figure S2(A)).

The heatmap analysis, as shown in Figure 1(B), reveals similar results for chili pepper and green onion, whereas ginger was the most independent. The LDA plots based on trace elements in Figure 2(B) show that ginger was independently located in the positive direction of the X-axis, with relatively high quantities of Mn, Ba, and Ga compared to those in other samples. Garlic is positioned above 4 on the Y-axis value. This is attributed to the higher content of Zn, Se, and Cu, resulting in higher values on the Y-axis. Due to the elevated levels of these elements in garlic, it independently positions itself in the positive range on the Y-axis. Green onion and chili pepper were located in the negative region of the X- and Y-axes, possibly owing to the lower content of trace elements compared to that in other samples (Figure S2(B)).

The heatmap analysis results in Figure 1(C) do not show a clear differentiation between the sample groups because compared to macro and trace elements, toxic elements are remarkably influenced by the translocation of environmental components into the plant. The LDA plot results based on toxic elements in Figure 2(C) show that garlic and ginger were located in the positive region of the x-axis, where garlic and ginger were influenced by the concentrations of Cd and Pb, respectively. Chili pepper and green onion were located in the negative region of the X-axis, with chili pepper having a higher concentration of Tl than that in green onion (Figure S2(C)).



The LDA plot results (Figure 2(D)) for all the elements exhibit that garlic was independently located in the positive region of the X-axis, possibly owing to the high concentrations of macro elements such as P, S, Se, Zn, and K. Ginger was located in the positive region of the Y-axis and was influenced by the trace elements Fe, Ba, and Mn and the toxic element Pb (Figure S2(D)). The chili pepper and green onion are positioned independently and can be distinguished from each other. However, overall, it has been confirmed that they share the most similar elemental composition characteristics.

Comparing the two groups of red peppers and green onions, there was a significant difference in the concentrations of elements such as Ca, Na, and Tl. Green onions had higher concentrations of Ca and Tl, while red peppers were found to have a higher Na content.

As a result, it is situated in the negative X-axis region of the plot. The results of the elemental composition effectively demonstrate that four types of spice vegetables could be independently distinguished and positioned according to the element quantities.

#### 4. DISCUSSION

Elemental composition analysis has been extensively performed on various foods and their raw materials to evaluate their nutritional value, classification, and potential health risks associated with exposure.<sup>12,49,50</sup> Garlic, ginger, green onion, and chili pepper are essential ingredients in Korean cuisine, including kimchi, and are frequently used in dried powder form to enhance flavor. Considering the significance of elemental composition in nutritional evaluation and food safety, a comprehensive investigation into the elemental profiles of these spice vegetables offers valuable insights.

The results of this study confirmed that K was the most predominant element across all samples, with garlic exhibiting particularly high concentrations of P, S, and K. These findings align with previous studies that demonstrated nutritionally significant elements variations among garlic varieties, enabling differentiation between cultivars and production origins based on their elemental compositions.<sup>50</sup> Trace element analysis revealed Zn and Mn as the most predominant trace elements across all samples, whereas ginger exhibited significantly elevated levels of Mn, Ba, and Ga. The concentrations of Mn and Zn in chili powder (*Capsicum annuum*, Nightshade) were significantly higher than those reported for chili peppers in previous studies (*Capsicum annuum* L.), with Ca and Mg levels also varying depending on the variety.<sup>51</sup>

Mineral balance plays a critical role in dietary health. The optimal Ca/Mg ratio in food is generally considered to be between 1:1 and 2:1,<sup>52</sup> whereas the recommended Ca:P ratio for bone health ranges from 1:1 to 1.5:1.<sup>53</sup> In this study, green onion exhibited a Ca/Mg ratio of 3.2:1, indicating a relatively higher calcium content, while garlic, ginger, and chili powder had higher Mg concentrations compared to Ca. These findings highlight the necessity of maintaining a balanced mineral intake to prevent nutritional deficiencies or imbalances.

To evaluate potential health risks, the EDI and HQ values were calculated. The results indicated that all HQ values remained below 1, suggesting that the consumption of these spice vegetables poses no significant health risk. The toxic element analysis (As, Cd, Pb, Tl, and In) confirmed their presence in all samples, with ginger showing the highest Pb concentration. However, all detected levels were within the safety limits established by the WHO, the FAO, and national

food safety regulatory authorities.<sup>45,46</sup> Furthermore, the EDI and HQ values for kimchi, which includes all four spices, indicated that consumption at the 95th percentile (P95) remained within safe limits.<sup>54</sup>

Multivariate statistical analyses, including heatmap clustering and LDA, demonstrated the potential to differentiate the four spice vegetables based on their elemental profiles. Variations in elemental composition are mainly influenced by environmental factors, such as soil composition, fertilization, and water quality.<sup>55</sup> Additionally, intraspecies varietal differences may also explain discrepancies observed between the findings of this study and prior research.

#### 5. CONCLUSIONS

This study systematically analyzed the elemental composition and safety of four commonly used spice vegetables: garlic, ginger, green onion, and chili pepper. A total of 25 elements, including macro elements (Mg, P, S, Ca, K, Na), trace elements (Li, Be, V, Cr, Mn, Fe, Co, Ni, Cu, Se, Ga, Rb, Sr, Ba), and toxic elements (As, Pb, Cd, Tl, In), were quantified using ICP-OES and ICP-MS. The results confirmed that garlic contained the highest concentrations of P (151.0 mg/100 g), S (61.4 mg/100 g), and K (537.0 mg/100 g), while ginger exhibited elevated Mg (34.1 mg/100 g), green onion had the highest Ca content (55.1 mg/100 g), and chili pepper was rich in Na (10.7 mg/100 g).

The analysis of toxic elements indicated that their concentrations in all samples were within internationally recognized safety limits, posing no significant health risks. The calculated EDI and HQ values further confirmed that the consumption of these spice vegetables, even at high intake levels (up to the 99th percentile), remains safe. Statistical analyses, including LDA and heatmap clustering, effectively differentiated the four spices based on their elemental compositions, with garlic showing distinctively high K, P, and S concentrations, while ginger, chili powder, and green onion were characterized by variations in trace and toxic elements.

These findings provide valuable insights into the nutritional and toxicological aspects of spice vegetables, emphasizing the importance of regular monitoring to ensure food safety. By addressing elemental composition and potential health risks, this study contributes to the development of improved food safety regulations and promotes safer consumption practices.

#### ■ ASSOCIATED CONTENT

##### Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.5c00578>.


ICP-OES and ICP-MS operating conditions and measurement parameters (Table S1); percentage of each element on four kinds of spices vegetable for (A) macro elements, (B) trace elements, and (C) toxic elements (Figure S1); influence of each element on the LDA plot for (A) macro elements, (B) trace elements, (C) toxic elements, and (D) total elements (Figure S2) (PDF)

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
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## Notes

The authors declare no competing financial interest.

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