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A Study of the Organic and Nonorganic Food Ingredients with Instrumental Neutron Activation Analysis

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Abstract: Organic food is welcomed by the general public because people think organic food is more environment-friendly and can introduce a healthy lifestyle. This popular notion is under scrutiny recently. Compared with conventional food, does the organic food we obtained from local farms and/or supermarket chains are actually chemically healthier? In this research, organic fruit and vegetables with USDA certification from local farmers and popular supermarket chains, along with their conventional counterparts, were collected and studied by a radioanalytical method—instrumental neutron activation analysis (INAA). Samples were irradiated by thermal and epithermal neutrons from the PULSTART nuclear reactor. After that, regular gamma-ray spectroscopy was applied to obtain the qualitative and quantitative information of target isotopes. Our preliminary study indicated that there is not much difference in the trace elements content between organic food and its conventional counterpart. Some heavy metals, which are commonly regarded as the source of harmful components, are detected in both categories. In terms of methodology, INAA is proved to be a sensitive radioanalytical tool to tell the elemental information on atomic or nuclear levels. However, as a nuclear technique, it lacks the capability to probe the properties of compounds on the molecular level, which may be the real difference between organic and nonorganic food.

Keywords: Organic Food, Instrumental Neutron Activation Analysis (INAA), Element Analysis

1. Introduction

In recent decades, organic food has gained lots of attention from the general public [1]. The difference between organic and nonorganic food is that nonorganic food is usually produced by conventional agriculture, which depends heavily on chemical intervention, such as fertilizers, herbicides, and pesticides, to provide plant nutrition, control weeds, and eliminate insects. On the contrary, organic agriculture follows natural principles such as biodiversity and composting to generate healthy organic food, which reduces pesticide residues and nutrient pollution, improves soil tilth and productivity, and lowers energy use and the carbon-emission level in the atmosphere [2].

Besides the assets of sustaining the whole ecosystem, organic agriculture also brings some "unexpected" benefits to the public: previous research indicates that people associate happiness and pleasure with organic food consumption. Consumers have linked organic food with healthiness and increased wellbeing. Eating organic food satisfies the consumers' need for wellbeing and a healthy lifestyle [3-5]. Triggered by these reasons, the production and consumption of organic food have proliferated over the past two decades [6, 7]. For instance, USDA-certified organic crop acreage more than doubled between 1997 and 2005, and organic operations and acreage have expanded to every state and commodity sector [2]. At the same time, organic sales have decupled since 1997 in the United States. Organic food sales increased from \$3.6 billion in 1997 to \$52.5 billion in 2018 (see Figure 1). Sales of organic foods increased annually between 5-20 percent during this period. Market penetration has also grown steadily; organic food products accounted for almost 6 percent of total food sales in 2019 in the United

States [8].

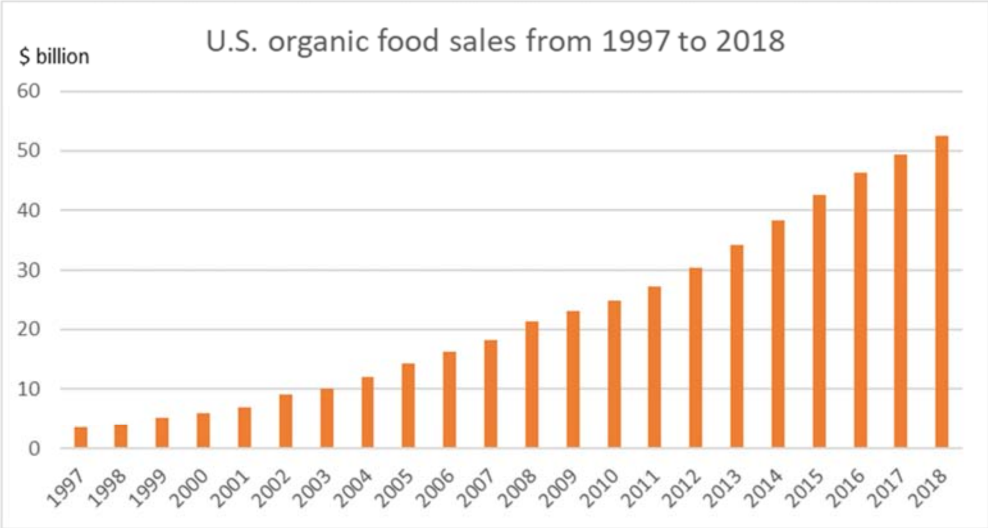

Figure 1. *U.S. organic food sales from 1997 to 2018 [9].*

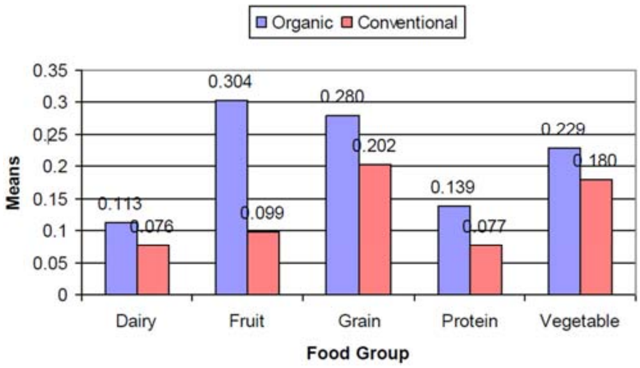

Figure 2. *Mean costs in US dollars per ounce by organic status and grocery venue [10].*

However, organic products cost more than their conventional counterparts due to more expensive farming practices, tighter government regulations, and lower crop yields. Figure 2 compares the mean costs per ounce by organic status and grocery venue with its conventional counterparts. One can see that prices for organic products are generally higher [10]. On average, organic foods are priced at 69% higher than conventional foods, with little variation between stores and time periods [11]. Since consumers pay more for organic food, the reasonable questions to follow are: is the quality of organic food worth the price difference? Does the price difference deserve the quality of organic food? Is organic food really healthier based on strictly chemical analysis? Is there a sensitive analytical method to distinguish organic and conventional food quickly? To answer these questions, instrumental neutron activation analysis (INAA), one of the most sensitive radioanalytical methods for multiple element analysis [12], is introduced in this project to study the chemical elements in organic and nonorganic food ingredients. Some pioneering and similar research was conducted on organic and nonorganic oranges and coffee with INAA and data mining techniques [13, 14].

2. Experimental

2.1. Sample Collection

The USDA-certified organic food ingredients and corresponding nonorganic counterparts are collected in local farms and some local chained grocery stores, such as Costco, Trader Joe's, and Whole Foods. The focus is on vegetables and fruits. After collection, all the samples were firstly cut into smaller pieces. For instance, apple, yam, cucumber, and radish were cut into blocks of size around 1 cm x 1 cm x 1 cm. Celery stems were cut about 5 cm long, and the crown size of broccoli and cauliflower were around 3-4 cm, followed by 20 hours of heat treatment at 60oC in the oven (Thermo Isotemp, Thermo Fisher, CA, USA). The relatively small size of grapes and celery leaves were kept in whole before heat treatment at 60 oC for 20 hours. Some samples (e.g., F16L, F24L, F26L) were residential and collected from family-grown gardens where no pesticides or other chemicals were used. Dried foods were commercially packaged products purchased from the local grocery store. These dried foods were not further treated and used as-is. One commercially available soil sample (F13L, purchased from local Lowes') and one local soil sample (F14L, from the local family-grown gardens) were collected in zip-lock bags and were treated in the oven at 60 oC for 20 hours. Some samples are listed in Table 1.

2.2. Neutron Activation

The PULSTAR reactor in the Burlington Laboratory at North Carolina State University was the source for our neutron irradiation (see Figure 3). The PULSTAR reactor is a 1 MW pool-type research reactor filled with 4% enriched, pin-type uranium dioxide pellets fuel under zircaloy cladding [15]. For the short-lived isotope study, neutron irradiation was performed by a pneumatic terminus system (PTS), which connects the tube nearby the reactor core and a hot cell facility adjacent to the reactor. PTS was coupled with a fast rabbit transfer system with a tube size of 3.18 cm in diameter. The thermal neutron flux of the irradiation point at PTS is around $1.0\times10^{13}/(cm^2\cdot s)$. For each activation of short-lived isotopes, the irradiation time is about 20 seconds, predefined by a controlled timer. For the activation of medium-lived and long-lived isotopes, neutron irradiation was conducted in the four dry sample pool standpipes, 8.89 cm in diameter at the reactor core's backside. At the standpipes, the average thermal neutron flux is around $7.7\times10^{12}/(cm^2\cdot s)$ and the average epithermal neutron flux is around $1.8\times10^{11}/(cm^2\cdot s)$. The neutron activation for medium-lived and long-lived radionuclides lasted continuously for about 10 hours. SRM 1648a (urban particulate matter) from the National Institute of Standards and Technology in US was applied as the comparator [16]. NIST reference materials were chosen because they have many well-known elements and widely used in radioanalytical practices. In addition, several Cu/Ti/Mn, La/Mo, Se/Ag solutions were made from diluting SPEX CertiPrep standard solutions with 2% HNO3. They were irradiated at the same time in the same position matrix with samples as both flux monitors and QA/QC controls. After neutron activation, all samples were moved out of the standpipes and cooled down in the reactor pool for one week before pulling out for gamma-ray spectra collection with two HPGe detectors in the adjacent gamma-ray counting room.

Table 1. *List of Organic and Nonorganic Some Food/Vegetable Samples.*

Label	Sample	Location	Remarks
F1L	Organic Purple Yam	Swansea, SC	Organic fresh vegetable
F2L	Red Delicious Apple	Costco, Columbia, SC	Organic processed dry apple
F3L	Freeze-dried banana	Trader Joe's, Thailand	Processed dry fruit
F4L	Freeze-dried Fuji Apple	Trader Joe, USA	Processed dry fruit
F5L	Dry okra	Trader Joe's, Thailand	Processed dry veg
F6L	Freeze-dried red grape	Trader Joe, USA	Processed dry fruit
F7L	Freeze-dried strawberries	Trader Joe, USA	Processed dry fruit
F8L	Freeze-dried blueberry	Trader Joe, USA	Processed dry fruit
F9L	Roasted plantain Chips	Trader Joe Peru	Processed dry food
F10L	Purple Yam	Swansea, SC	Organic fresh vegetable
F11L	Organic celery	Trader joe CA	Organic fresh vegetable
F12L	Celery	Trader joe CA	Organic fresh vegetable
F13L	Organic soil	Lowe’s, NC MEZCLA OMRI	Organic soil
F14L	Soil local residential	Columbia, SC	Soil local residential
F15L	Grape 4499	CHILI, Whole Foods	Fresh fruit
F16L	Organic Radish	Local	Organic grown
F17L	Organic cauliflower	Trader Joe's, USA	Organic fresh vegetable
F18L	Organic broccoli	Trader Joe's, USA	Organic fresh vegetable
F19L	Cucumber	Costco	Fresh vegetable
F20L	Organic Cucumber	Trader Joe's, CA	Organic fresh vegetable
F21L	Cucumber	Trader Joe's, CA	Fresh vegetable
F22L	Persian Cucumber	Trader Joe's, CA	Fresh vegetable
F23L	Organic Persian Cucumber	Trader Joe's, CA	Organic fresh vegetable
F24L	Organic Celery stem	Local	Organic vegetable
F25L	Organic Celery leaves	Local	Organic vegetable
F26L	Organic celery leaves yellow dry	Local	Organic vegetable
F27L	Organic Beans	Walmart	Organic Beans
F28L	Organic Pearl	Walmart	Organic Pearl

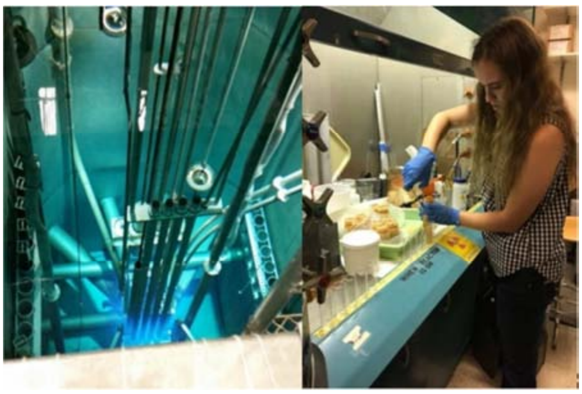

Figure 3. *Neutron irradiation at the PULSTAR reactor (left: reactor core and irradiation tubes at the PULSTAR reactor; right: a student worked by the hot cell to unpack the samples after irradiation of short-lived isotopes).*

2.3. Spectra Collection

Two HPGe detectors from Mirion Technologies with efficiencies of 42% and 38% were used to measure short and medium-lived isotopes. Another 25% efficiency HPGe spectrometer from Ortec linked with an Automatic Sample Exchange System (ASES) was designated for the measurement of long-lived isotopes. Figure 4 shows the counting electronics associated with the HPGe detectors. Standard nuclear electronics, such as pre-amplifier, high voltage bias, analog to digital converter (ADC), multichannel analyzers (MCA), and Windows 7 based Personal Computers, were applied to process the signals from HPGe spectrometers. An Eu-152 point-source was applied to check the detector efficiency calibration of all HPGe spectrometers regularly. Eu-152 was selected because it has 11 characteristic energy lines that cover the whole energy range of our interest. The computer program used in the gamma-ray spectra collection was standard Genie 2K from Mirion Technologies. The measurement includes six possible positions (S, A, B, C, D, F), which correspond to certain predefined distances (0 to 5 inches) above the detector surface. In the measurement, the position was carefully chosen to ensure the dead time of the counting system was less than 10%.

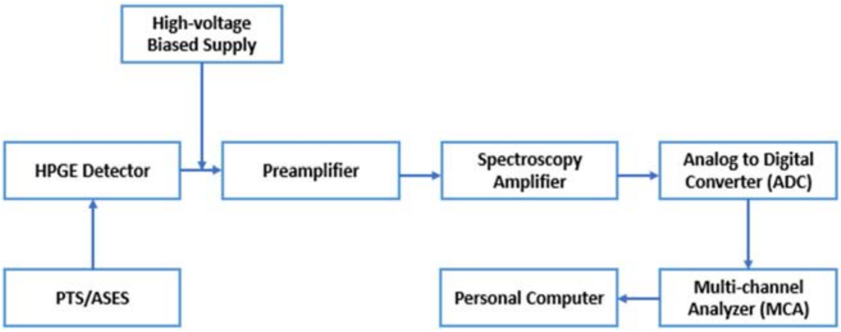

Figure 4. *The Block diagram of the counting electronics associated with the High Purity Germanium (HPGe) Detectors.*

3. Data Analysis and Discussions

3.1. INAA Validation

Three spectra, which correspond to short-lived, medium-lived, and long-lived isotopes, are collected for each sample to reduce the background and achieve higher sensitivity. Figure 5 indicates a typical gamma spectrum for long-lived isotopes collected by the Genie 2K program, overlapped with the arrow to identified peaks and corresponding nuclides. We followed Tables 10, 11, and 12 in the MURR INAA database [17] for isotope identifications. The calculated concentrations correspond to short-lived isotopes (e.g., Ba, Ti, Ge, I, Br, Mn, etc.), medium-lived isotopes (e.g., Na, K, Ca, W, Ce, Au, etc.), and long-lived isotopes (e.g., Sc, Cr, Fe, Co, Ni, Zn, Ag, etc.). Their amount in the sample ranged from normal, rare, to trace.

Several batches of NIST 1648a were irradiated at different positions. Some were labeled as NIST 1648a, and others were just labeled "unknown" materials. The table below indicated the calculated values of the main elements in the "unknown" materials compared with certified values in NIST 1648a. The discrepancy in the table is calculated by.

$$\text{Discrepancy\%} = (\text{measure value} - \text{certified value})/(\text{certified value}) \times 100\%$$

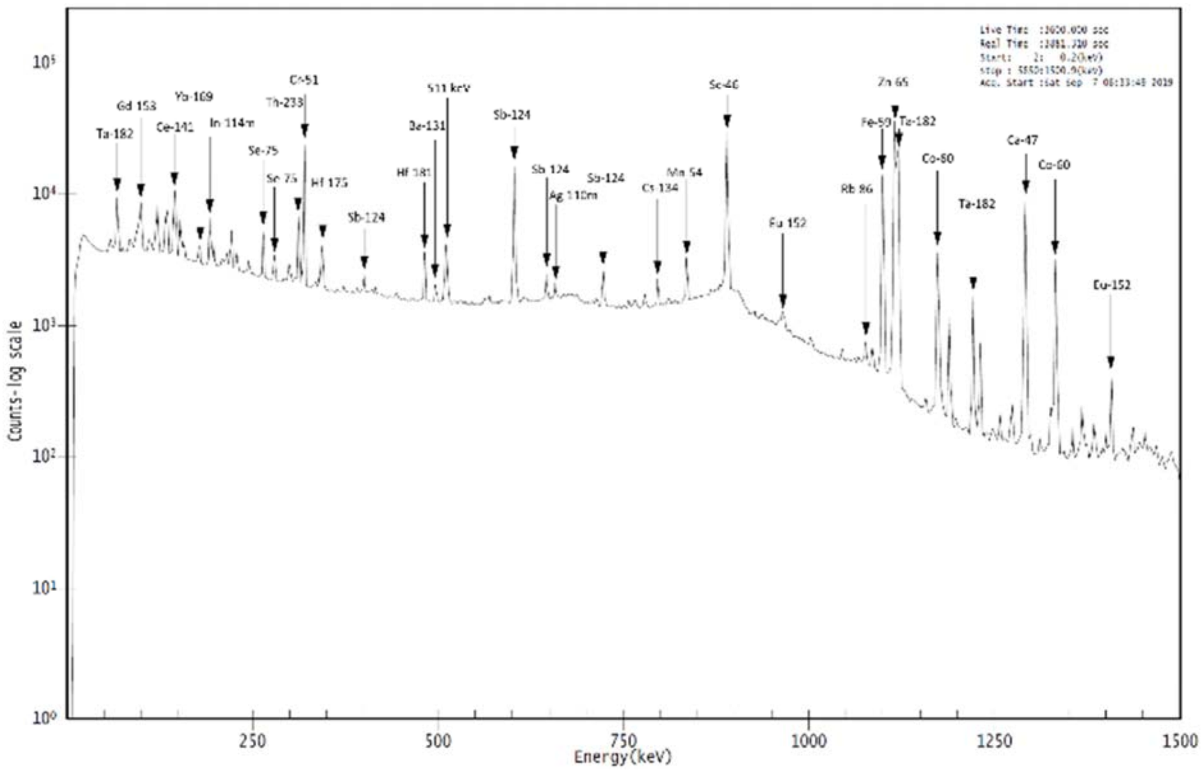

Figure 5. *A gamma spectrum for long-lived isotopes (NIST 1648a) with peaks and corresponding nuclides.*

Table 2. *Elements in NIST1648a and Their Corresponding Concentrations.*

Elements	Calculated value (mg/kg)	Certified values (mg/kg)	Discrepancy (%)
Na	4280±150	4240±60	0.96%
Mg	8030±1260	8130±120	-1.20%
Al	35000±1400	34300±1300	2.10%
Cl	4550±200	4543±47	0.26%
K	10250±4790	10560±490	-2.92%
Ti	360±60	402±13	-10.84%
V	127±12	127±11	-0.05%
Cr	340±55	402±13	-15.53%
Mn	810±47	790±44	2.09%
Fe	34100±4500	39200±2100	-13.12%
Br	535±30	502±10	6.69%

3.2. Result and Discussion

Figure 6 indicates the primary elements and their concentrations in yam, okra, and grape. One can notice that each fruit or vegetable has a certain kind of curve or signature of its elemental concentrations. Each plant has a unique element distribution curve, or in other words, the shape of the curve corresponds to the specific vegetable or fruit. The elemental signature is relatively close for each plant, whether the sample originated from organic or nonorganic crops. In the plot, one can also see that okra is quite nutritious. Many nutrition elements have a higher concentration than those of other plants, including Na, K, Mn, etc. In the yam sample, some elements, such as Fe, have higher concentrations than found in other plants. However, yam also has some trace elements that cannot be detectable in other plants, such as La, Ce, Sm, Hf, W, etc. Some pessimistic people may say that yam is very "toxic" because many trace metals are heavy metals. However, as seen in recent research [18-20], if the concentrations of these heavy metals are in the PPM or PPB levels, these trace amounts of heavy metals will cause no harm to the human body. The reason why the yam has more heavy metals is unknown. Probably, because the yam is buried in the soil, some heavy metals from the soil have conveniently penetrated from the skin of the yam.

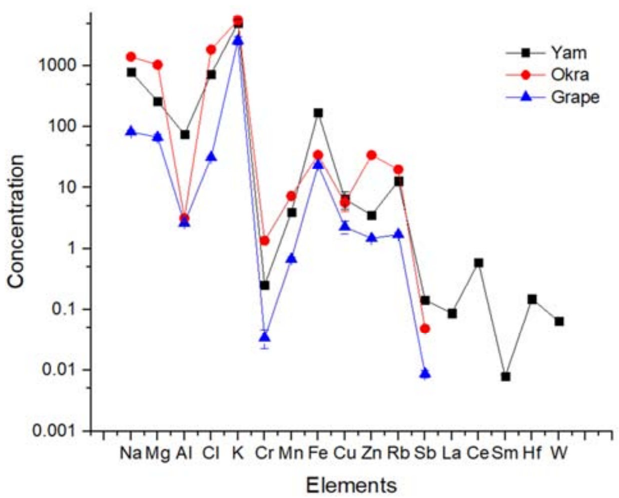

Figure 6. *Major Element Concentrations of Yam, Okra, and Grape.*

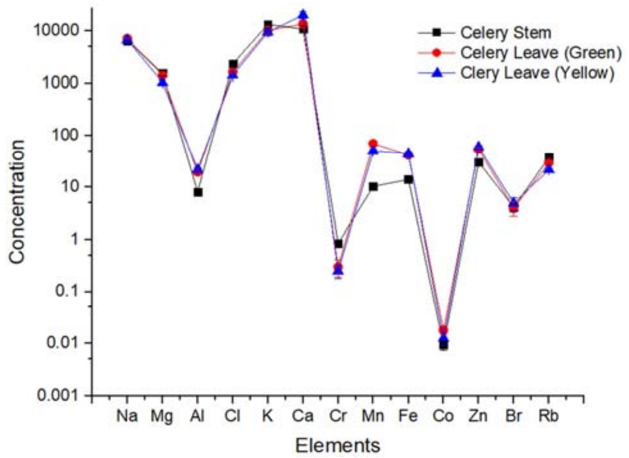

Figure 7. *Element Concentrations in Celery in different parts (Stem and Leaf).*

The elemental concentrations are different in different parts of the plants. For example, in Figure 7, one can notice that celery stems and leaves have slightly different Mn and Fe concentrations. Element Fe and Mn have high concentrations in the stem but not too much in leaves. The similar elemental pattern of yellow and green leaves indicates that the color difference is not caused by differences in elemental concentrations. Whether the plant is green or yellow, the major elements are quite similar.

Can INAA determine the difference between organic and nonorganic food ingredients? The answer probably is a no, at least from our experimental results. In our experiments, the results demonstrated that NAA is not an viable method in telling the difference between organic and nonorganic crops. For instance, one can notice in Figure 8, concentrations of different elements in organic and nonorganic cucumbers are quite similar. Or in other words, the signature curve of cucumber is unique, whether the samples are from organic or nonorganic sources. Besides, the different sub-species of cucumber, ordinary cumber and Persian cucumber, have quite similar curves even though they are different in appearance. Therefore, we suppose the primary difference between organic and nonorganic crops is not from their elemental concentrations. If one relies solely on elemental analysis to identify the organic food and vegetables from nonorganic ones, it may difficult to distinguish them. The difference between organic and inorganic food ingredients may not be at the atomic level of some rare earth metals. Instead, it is due to the molecular level of organic combination among the carbon (C), hydrogen (H), and oxygen (O), such as phenol, aldehyde, organic acid, etc. Since the limited sampling size and limited food categories of this research, the claim about the limitation of INAA in determining the difference between organic and non-organic food ingredients cannot be extrapolated into a more general context and need to be interpreted with caution. Similar work conducted with other food ingredients with a more extensive sampling size has shown positive results with elemental discrimination of INAA and ICP-OES techniques [13, 14, 19]. Assisted with data mining and the process of Knowledge Discovery in Databases (KDD), INAA has demonstrated to be a promising tool for using elemental concentrations to discriminate between organic and conventional green coffees [13].

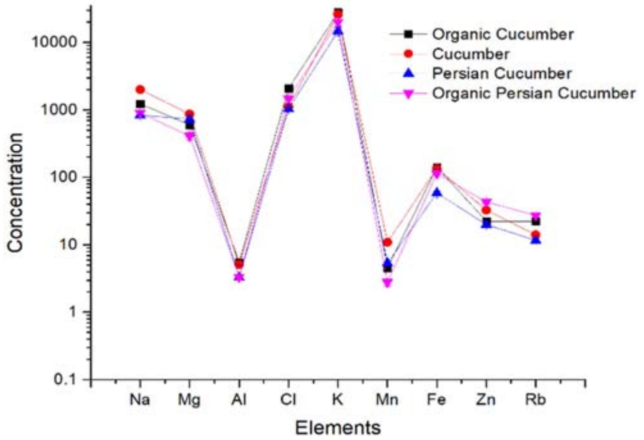

Figure 8. *The difference among organic cucumber, Persian cucumber, and their nonorganic counterparts.*

Does organic food have more 'nutritious' elements than its conventional counterpart? It is probably not based on simple element analysis, but the direct answer to this question is unclear since food nutrition is a very complex issue. If we only look at the plants' elements, we can conclude that organically and conventionally produced food are similar in their nutrient content. However, nutrition is not a concept defined by elements in the food but also based on its freshness and possibility of contamination. For instance, organic produce is not as widely available as other produce. That means, depending on where you live, it may be shipped from farther away from that nonorganic produce. In some cases, it may sit longer on the shelf before it is sold. During the lag time between harvest and consumption, certain nutrients can degrade within produce even if they are of the same elemental composition. Another factor to consider is pesticides. Usually, organic produce carries significantly fewer pesticide residues than does conventional produce. If the residues on products, whether organic or nonorganic, do not exceed government safety thresholds, we can safely say that the amount of pesticides found on fruits and vegetables poses a minimal health risk.

4. Conlusion

We can draw a preliminary conclusion from the study as below:

1. INAA is a versatile and sensitive radioanalytical method in element analysis. It can determine concentrations of many elements in food and vegetables simultaneously with high sensitivity and precision.
2. Each food or vegetable may have a particular curve of elemental distribution, and this curve can be used to characterize the food or vegetable, and it was impacted by different biological species, different food processing methods, or even by a different part of the plant.
3. However, the implementation of INAA on telling the distinction of organic and nonorganic food and vegetable did not reach the expectation. Organic and nonorganic plants did not differ too much in elemental concentration. It is more likely that the distinction is not on the atomic or nuclear level but the molecule level. Some heavy metals are more likely to show up in the root buried by the soil.

For future work on this project, more samples need to be collected, including fruit and vegetables with organic and nonorganic counterparts, soil, pesticides, and fertilizers. Since the distinction between organic and conventional food is likely from the molecule level of organic compounds, it is necessary to introduce organic analysis instrumentation, such as gas chromatography (GC), Fourier Transform Infrared (FT-IR) Spectrometer, NMR spectrometers, etc.

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