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Cd and Cr Metals Burden in Fruits and Vegetables of Southern European Countries: A Review

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Abstract
The Cadmium (Cd) and Chromium (Cr) burden in fruits and vegetables in Southern European countries such as Turkey, Spain, Italy, Kosovo, Croatia, Greece, and Romania are compared and analyzed in this review article to account for any significant differences. Potential sources of the metals and their health issues have been underlined. Various reasons for the extent of the metals' uptake are highlighted, including species' nature, soil pH and atmospheric conditions. In Croatia, the highest Cd burden was observed in lettuce (1.16±0.24 mg/kg), followed by carrot (0.49±0.18 mg/kg) and onion (0.28±0.08 mg/kg). The concentration of Cd in peppermint and spinach has been reported as 0.004±0.001 μg/g and 0.024±0.001 μg/g, respectively, in Turkey. Spain's strawberries, melon, and bananas have shown 0.0025 mg/kg, 0.0028 mg/kg, and 0.0005 mg/kg of Cd, respectively. The Cr level in Croatia's vegetables was less than the instrument's detection limit. In Romania, its concentration in cabbage and potato root was 17.5 ± 0.7 mg/kg and 4.7 ± 0.3 mg/kg, respectively. Similarly, in Greece, the highest levels of Cr were found in green peas (0.21 ± 0.03 μg/g), followed by onion (0.16 ± 0.07 μg/g) and cabbage (0.13 ± 0.08 μg/g). The fruits and vegetables of most regions have shown a greater burden of Cd than Cr. Several strategies could be implemented to minimize the extent to which the plants take up these metals and reduce their environmental risks.
Keywords: Cd, Cr, Fruits, Vegetables, uptake, burden

1. INTRODUCTION

Vegetables and fruits are a vital part of the human diet. They might contain heavy metals in very minute or large concentrations, which can be a health hazard in the long term. Heavy metals are metals that have high molecular weights as well as relatively high densities. Heavy metals can be poisonous and toxic even in trace quantities. Heavy metals include cadmium (Cd), lead (Pb), arsenic (As), mercury (Hg), chromium (Cr), and thallium (Th).

1.1. Sources

Determining how the pollutants are transported from one environmental segment to the other is vital. The anthropogenic sources of these two heavy metals are discussed below.

The healthy development of plants requires a continuous nutrient cycle from the soil; hence, what enters the plant via the soil or air is of significant value [1]. Two well-known metals are chromium (Cr) and cadmium (Cd), which might be present in fruits and vegetables. Rhizo-filtration (rhizo=root) is a process through which plants take nutrients from the soil and filter out metal contaminants. However, they can take up these contaminants if any of the contaminants is similar to one of the plant's required metals. The adsorption and precipitation of metals onto plant roots and absorption of metal contaminants into the plant root (these contaminants might be present in the form of a solution in the soil) are processes through which a plant can take up metal contaminants [2]. The polluted atmosphere around a plant may contain heavy metals in the air due to their volatile nature, and the plants may absorb them from the airborne deposits on the plants themselves. Many factories and industries do not treat their contaminated water before releasing it, and contaminants leach into the soil, and the water carries the rest. The contaminants carried by water are then a part of the irrigation water, and thus plants are exposed to metal contaminants like Cd and Cr. Also, some of the gaseous effluents from the industries are released into the air untreated, exposing the atmosphere to the metal contaminants that become a part of the fruits and vegetables being cultivated eventually. The presence of heavy metals in plants can also be attributed to the addition of lead compounds in petrol in order to enhance the efficiency of the engines [3]. Fertilizer use and metal-based pesticides, transportation, harvesting process, storage or sale can cause metal contamination [4]. Many of such metals reach non-contaminated areas from contaminated areas via dust. They might also leach from landfill sites and contaminate new areas of land. Rain may wash away some of the contaminated soil via erosion, and such metals can enter into non-contaminated areas when soil deposition occurs.

The uptake of metals from the soil depends on different factors such as their soluble content, soil pH, plant species, fertilizers, and soil type [6]. The atmosphere and industrial pollution can also be other factors. The more contaminated areas mean the fruits and vegetables of a region will be more contaminated. Vegetables, especially leafy vegetables, accumulate higher amounts of heavy metals because they absorb them in their leaves [7]. Only soluble and chelated metal species are more mobile and available for the plant species to uptake.

1.1.1. Cd (Cd)

Cd is a soft metal, bluish-white in colour, found in zinc ores and Cd minerals. Like zinc, it commonly exists in the +2 oxidation state. The anthropogenic source of Cd is mining wastes and industrial waste discharges, especially from metal

plating. Cd is a by-product of zinc smelting and is released into the atmosphere as vapour due to its boiling point of less than 1000 degrees. It is not easy to refine solid form a fuel like coal, so burning coal in industries leads to the emission of Cd into the atmosphere. Incineration of garbage, Ni-Cd batteries, plastics, and crockery are also considered anthropogenic sources of Cd. Due to its similarity in oxidation states with Zn, many plants uptake Cd from the irrigation water. It is absorbed similarly to tobacco leaves and then released into the atmosphere and smoke, where carbon acts as a carrier. Acid rain dissolves the Cd into it and returns it to the soil, which becomes a component of the grown fruits and vegetables.
Cd, in the form of Cd sulfide (CdS), has extensively been used in paints [5]. Phosphoric fertilizer contains high Cd quantities; hence, excessive phosphate fertilizers are found in agricultural soils [6].

1.1.2. Chromium(Cr)

Cr is a steely-grey, hard, lustrous transition metal. The two most common oxidation states of Cr are +3 and +6. Food appears to be the primary intake source for this metal, which means it is widely present in fruits and vegetables. Cr is used for electroplating and corrosion protection. In leather tanning, it binds to the leather's protein and makes it resistant to heat corrosion, water corrosion, and corrosion by bacteria. It is widely used as a wood preservative as Chromated Copper Arsenate (CCA) [5].

1.2. Health hazards

Both Cd and Cr have hazardous effects on human and animal health if present in the body in amounts greater than the tolerable intake.

1.2.1. Cd

Cd is a carcinogenic metal. It has health risks associated with damage to the DNA (DNA repair mechanisms are affected) and cell signalling [7]. It also affects the kidneys and the respiratory system if absorbed by the body in amounts greater than the sulfhydryl groups in the protein metallothionein can be complexed. Even 1 g of the Cd ion is considered lethal. As the WHO established in 2010, the tolerable monthly Cd intake was 25μg/kg body weight [8]. Other health effects associated with Cd include high blood pressure, fracture of bones, and red blood cell destruction.

1.2.2. Cr

Hexavalent Cr is more toxic than trivalent Cr due to more excellent water solubility and more significant propagation. Cr is very toxic by inhalation and dermal route and causes lung cancer, nasal irritation, nasal ulcer, and hypersensitivity reactions like contact dermatitis and asthma [9]. The chromate ion can oxidize DNA and RNA bases upon entering the cells.

2. Related Work

The occurrence of heavy metals, particularly Cd and Cr, in vegetables and fruits has been the subject of numerous studies over the past years. Several researches have been done to analyze the effects of these toxic elements that have entered the food chain via fruits and vegetables.

A comparison of the quantities of several metals has been recorded in various fruits and vegetables; peppers, eggplants, lettuces, strawberries, onions, cucumbers, and tomatoes, to name a few. Experiments have been conducted to determine the accumulation of these toxic metals and the extent of health hazards they pose. Results have been published regarding several Southern European countries, including different regions of Spain in 1990 and 2008, certain areas of Turkey in 2003-2004 and 2005-2006, Croatia, Romania, Greece, Kosovo, and Southern Italy 2013-20

3. Country-wise work on Cd and Cr

Many studies have been performed on Cd and Cr burden in various countries of Europe in the south. The results of the studies are discussed country-wise below for both metals..

3.1 Cd

3.1.1. Croatia

An analysis was carried out in Croatia at the city market of Varazdin for common vegetables. The results showed that a certain number of fruits and vegetables contained noticeable amounts of Cd levels, as indicated by Table (1) below [10]:

Table 1 Cd burden in vegetables in Varazdin, Croatia given as mean value ± standard deviation in mg/kg [10]

Sample	Cd
Red Potato	0.26±0.14
Onion	0.28±0.08
Carrot	0.49±0.18
Beans	0.04±0.00
Cabbage	0.2±0.07
Lettuce	1.16±0.24
White Potato	0.23±0.09

The Cd concentration was above the stipulated level for lettuce, i.e. (1.16±0.24) mg/kg. These samples consisted of the dry weight of the vegetable. After extraction of the metal, the sample was prepared for atomic absorption spectrophotometry. This technique gave the above-stated results for each of the vegetables tested. An industrial area surrounds the selected market in Croatia, and the selected vegetables are among the everyday food intake of the population. The city's mean annual temperature is 10.2°C and varies from -3.6°C to 27°C from January to July.

The study states that Cd's tolerable weekly intake (TWI) is 2.5 µg kg-1 b.w. [11]. If a comparison is made with measurements stated by EFSA, Cd's mean concentrations in the study were higher in red potato and white potato (0.06 and 0.05mg/kg compared with 0.02mg/kg), carrot had a value of 0.06mg/kg, whereas, EFSA's measurement states 0.02 mg/kg. Beans were 0.04 mg/kg compared with EFSA's 0.01 mg/kg, and lettuce levelled 0.06 mg/kg compared with EFSA's 0.02 mg/kg. The same was observed in cabbage, which levelled at least 0.02 mg/kg, compared with EFSA's 0.02 mg/kg [10]. These results show that the market's vegetables and fruits showed higher Cd levels than acceptable ones. A prolonged intake of higher levels can have a hazardous impact on health. One major reason contributing to these levels is nearby industrial regions, resulting in the heavy uptake of contaminant metals. Similarly, the harmful metal content of wild berries and fruits was determined in Croatia. The results of the Cd levels in certain berry fruits are summarized in Table (2) [12]:

Table 2 Cd burden in berry fruits in Croatia in mg/kg [14]

Sample	Cd
Lingon Berries	<0.028
Rose Hip	<0.028
Blueberries	<0.028

The results reported in Table 2 show the readings in mg/kg for the dry weight of three types of fruit berries. Cd is readily taken up by plants and roots and accumulates into food, mainly fruits and berries[13]. The detection limit of the instrument was 0.028mg/kg. The results showed that the Cd levels were less than 0.028mg/kg. Comparing the rosehip results with the rest of Southern Europe, Turkey has observed a 0.81mg/kg Cd level [4].

3.1.2. Turkey

Different research projects determined fruit and vegetable content in Kayseri, Turkey. Organically produced fruits and vegetables were tested for metallic element levels present in them, and the results for Cd for the particular project have been discussed [14]. A study in turkey states that the copper, iron, manganese, and zinc concentrations were found to be 1.6–15.5, 10.3–144, 23.0– 211, and 23.3–91.6 µg/g, respectively. The cobalt, lead, Cd, Cr, and nickel concentrations in all analyzed organic fruit samples were below the quantification limits of National Future Farmers of America (FFA).

In another study, in Turkey, black grape contained the highest levels of copper(4.52 µg/g), the highest nickel content was 9.4 µg/g in black plum, the maximum Pb content was found in apricot(12.4 µg/g), the highest Mn content was found in rosehip that valued 25.5 µg/g. The peak iron content was in apricot with a value of 64.1 µg/g. Yellow plum contained the maximum Cr content at 6.17 µg/g. Mulberry had maximum levels of cobalt, valued at 1.81 µg/g. The maximum concentration of Cd was found in apricot and rosehip, which levelled at 0.8 µg/g [4]. Comparing this with the rosehip content in Croatia, Croatia's rosehip had less than 0.028mg/kg Cd in the rosehip berry.

Amongst the vegetables, onion has the highest Cd levels; 0.97µg g^{-1} in the urban area and 0.61µg g^{-1} in the rural area, while the lowest was observed in lettuce; 0.34µg g^{-1} in the urban area and 0.24µg g^{-1} in the rural area. The varying levels amongst the vegetables depending on the plant's ability to uptake certain metals, metal concentration, and weather conditions. According to a study conducted in the Bursa province of Turkey, traffic and industrial regions were almost alike. The highest amount of Cd and lead were found in the traffic region. The Cd level was 0.81 ± 0.25 mg kg^{-1}, and the

Pb was 1.59 ± 0.45 mg kg^{-1}, respectively. The least Pb and Cd levels were found in leeks grown in rural areas. The lead levels were 0.10 ± 0.03 mg kg^{-1}, and the Cd levels were 0.05 ± 0.01 mg kg^{-1} compared with other vegetables [15].

The information is summarized [Table 3]for the Cd contamination in the vegetables from rural and urban Kayseri, Turkey[6].

Table 3 Cd burden in vegetables of Kayseri, Turkey given as mean ± standard deviation both in $\mu g g^{-1}$

Sample	Urban Area Kayseri, Turkey	Rural Area Kayseri, Turkey
Lettuce	0.34±0.01	0.24±0.01
Parsley	0.84±0.05	0.4±0.07
Peppermint	0.68±0.07	0.63±0.01
Onion	0.97±0.008	0.61±0.05
Cucumber	0.64±0.004	0.3±0.07
Okra	0.58±0.003	0.42±0.04
Green-Pepper	0.62±0.008	0.32±0.02
Pumpkin	0.6±0.001	0.41±0.01
Eggplant	0.43±0.09	0.33±0.03
Tomato	0.41±0.007	0.33±0.05
Bean	0.36±0.008	0.29±0.01

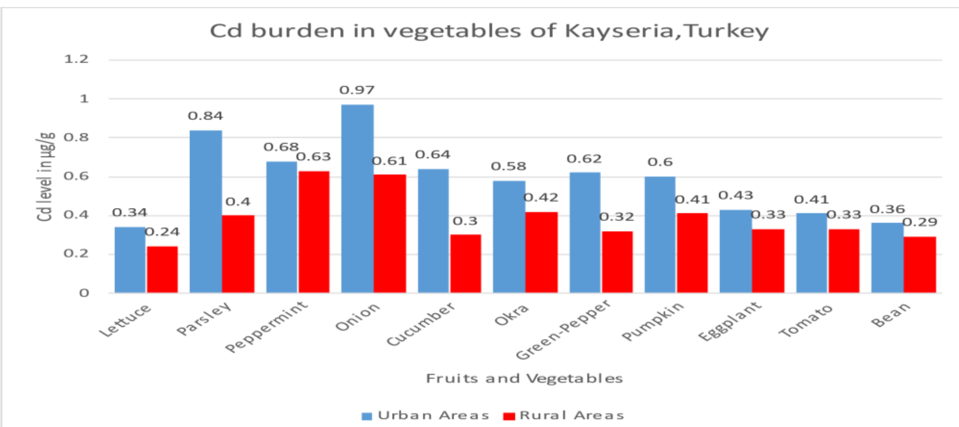

Figure 1 Cd burden in vegetables of Kayseri, Turkey

As illustrated by Figure 1, Kayseri, Turkey's urban areas exhibited higher Cd levels in all the vegetables where Cd content was determined. The primary source of Cd in the soil is phosphate fertilizers. All fruits and vegetables grown in the agriculture sector receive fertilizers. Cd is found in phosphate fertilizers; hence, soils that receive phosphate fertilizers have levels of Cd present in them, which in turn become a burden in fruits and vegetables grown in those soils. Urban areas have more pollution due to higher interaction with the anthropogenic environment. Chances of contamination are thus greater. From 2005 to 2006, research was carried out on Cd levels in a region in Turkey called Manisa [16].

Table 4 Cd Levels in vegetables in Manisa, Turkey(2005-2006) given as mean ± standard deviation in µg/g

Sample	Cd
Lettuce	0.005±0.001
Nettle	0.001±0.001
Peppermint	0.004±0.001
Spinach	0.024±0.001
Dill	0.001±0.001
Chard	0.007±0.001
Pulsane	0.013±0.001
Rocket	0.024±0.001

Cd levels of vegetable samples ranged between 0 and 0.060 mg/kg. The highest content was found in rocket and spinach, with a value of 0.024 µg/g. Dill and nettle contained the lowest range in Manisa. The results of Table 3 and Table 4 show that Cd levels have varied from place to place and from time to time. According to a study, Cd contents in food items from a market in Greece were between 0.0002mg/kg and 0.527mg/kg [17]. According to another study, Dill's leaves contained between 0.023mg and 0.028 mg Cd [18].

Studies carried out in Corlu, Tukey showed results in which Cd values in onions, lettuce, spinach, beans, and cereal exceeded the limit codex allimentarus.[19]

3.1.3. Greece

Greece is a country in Southern Europe, and studies have been conducted in this region regarding the Cd levels[Table 5] in fruits and vegetables [20].

Table 5 Cd levels in "certified organic foodstuff" in the Greek market (mean ± standard deviation in ng/g)

Sample	Cd
Potato	8.6±2.7
Parsley	5.9±2.3
spinach	37±10.5
Celery	5.3±1.8
Amarantus	21.7±6.7
Blitum	7.3±2.4
Lettuce	2.1±0.3
Dry Onion	2.9±1
Fresh small	1.8±0.6
onion	5.1±2.1
Leek	<1.0
Fresh Garlic	3.3±1.4
Cucumber	6.5±6.9
Beet	5.9±5
Cabbage	4.5±0.6
Cauliflower	12.1±9.7
Broccoli	4.9±2.1
Green Pepper	5.0±3.5
Carrot	2.4±1.5
Aubergine	1.5±8.7
Fresh green	3.2±1.0
beans	<1.0
Artichoke	<1.0
Tomato	<1.0
Apple	<1.0
Kiwi	<1.0
Orange	<1.0
Mandarin	
Apricot	
Peach	

The results show that significant uptake of Cd was present in the vegetables, and the fruits' levels were below 1ng/g of the wet weight of the fruits. The highest value was in potato at 8.6 ng/g, and the lowest Cd-contaminated vegetable was cucumber, in which the amount was less than 1.0 ng/g. Because of controlled conditions, the results show that organically produced foodstuffs have lower Cd levels than conventionally grown foods. Anthropogenic Cd sources have contaminated most human diets with toxic metals, especially vegetables, which tend to up-taking metals more than the fruits found in the same areas. Greek farmers use many phosphate fertilizers, which are a source of Cd. Also, traditional farming practices like manure may deposit metals like Cd into the soil, becoming a part of the fruits and vegetables.

3.1.4. Kosovo

Kosovo is a partially recognized state in southeastern Europe. The following study was conducted in the Mitrovica region, Kosovo[Table 6]. The city of Mitrovica lies in the north of Kosovo [23]. The study concentrates on heavy metal content in different parts of apple species like fruit, leaf, shoot, and apple types per rootstock reported in Table [11].

Table 6 Cd concentration (mg /kg) in parts of apple species depending on rootstock type from the Mitrovica region.

Rootstock	Tissues	Cd (mean ± standard deviation)
	Shoot	0.67±0.11
M106	Leaf	0.51±0.08
	Fruit	0.37±0.17
	Shoot	0.52±0.15
M26	Leaf	0.48±0.07
	Fruit	0.22±0.11
	Shoot	0.36±0.07
M9	Leaf	0.31±0.06
	Fruit	0.24±0.07

Table 7 Cd concentration (mg/ kg) in parts of apple species depending on rootstock type from the reference site.

Rootstock	Tissues	Cd (mean ± standard deviation)
	Shoot	0.23±0.17
M106	Leaf	0.16±0.15
	Fruit	0.009±0.001
	Shoot	0.17±0.24
M26	Leaf	0.19±0.16
	Fruit	0.08±0.05
	Shoot	0.13±0.09
M9	Leaf	0.06±0.04
	Fruit	0.003±0.001

Among all of the apple species' parts, the minimum Cd uptake is by the fruit in every type of rootstock. It can be observed from Table 6 and Table 7 that the levels of Cd in the Mitrovica region in all apple rootstocks are higher than those in the reference site. The fact that the area is an industrial region explains the higher Cd levels observed when tested for Cd contamination.

3.1.5. Spain

A study has been carried out in Valencia, a city in Spain. Results reported in Table [8] show the Cd level in the fruits and vegetables in the region with Cd heavy metal [21].

Table 8 Cd burden in fruits and vegetables in Valencia in mg/kg

Sample	Cd
Lettuce	0.0098
Green Beans	0.0051
Onions	0.0040
Garlic	0.0088
Peppers	0.0058
Cucumber	0.0065
Carrots	0.0046
Tomato	0.0055
Melon	0.0028
Banana	0.0005
Strawberry	0.0025
Olives	0.0038
Pumpkin	0.0046

It was inferred from Table [8], that lettuce in Valencia contains the highest mean Cd level of 0.0098mg/kg among other fruit and vegetables. As observed in other countries, fruits have much lower Cd levels than vegetables, containing Cd concentrations from 0.078mg/kg to 0.353mg/kg. Comparing these levels with that of Greek shows that Greek fruits, including oranges, have levels less than 1.0ng/kg. It has been observed that lettuce and onions in the countries of southern Europe discussed above have high Cd levels compared with other vegetables. The plant metal uptake mechanism plays a vital role in this process. Soil conditions from place to place vary a lot. Lettuce, however, is one of the vegetables named in Cd uptake in almost all countries.

3.1.6. Romania

Research in Romania showed that certain soils and vegetables were tested for the presence of seven different heavy metals, including Cd [Table 9]. Samples were collected from two contaminated mining sites, Ruschita (R) and Moldova Noua (M), and an exact reference site.

Table 9 Cd levels in the soil of sampling areas, given as mean value ± standard deviation in mg/kg

Sampling area	Cd
R site	2.00 ± 0.23
M site	0.40 ± 0.02
Reference site	0.17 ± 0.01

Results of the Cd levels found in the soil of the three sampling regions, R, M, and the reference area are shown in Table 9. From Figure 2, it can be observed that contaminated areas R (2.00 ± 0.23 mg/kg) and M (0.40 ± 0.02 mg/kg) contained significantly higher Cd concentrations than that observed in the reference area (0.17 ± 0.01 mg/kg).

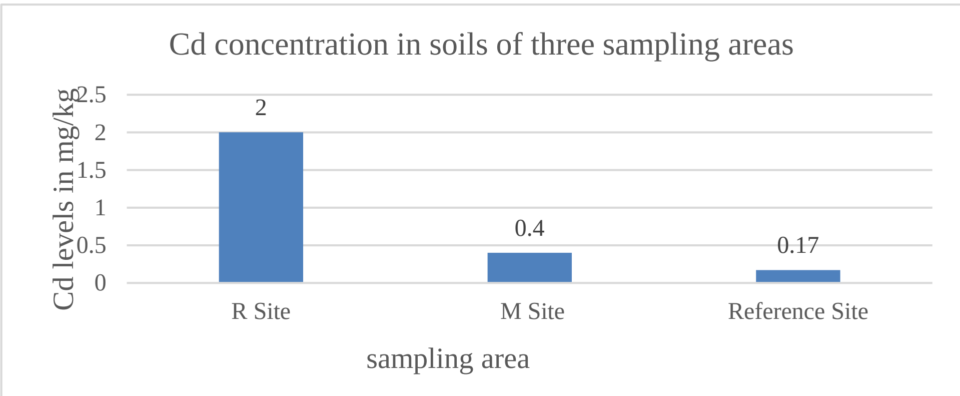

Figure 2 Cd levels in soil of sampling areas

The Cd levels detected in the selected vegetables from each of the three sampling sites are reported in Table (10). It is evident from the data that the vegetables from contaminated sites contained greater concentrations of Cd than those grown in the clear reference area. Parsley root, for example, contained 0.2 ± 0.12 mg/kg of Cd in the mining site R followed by 0.04 ± 0.02 mg/kg in M. The least amount of Cd was detected in the reference area (0.01 ± 0.00 mg/kg). Similarly, the carrot root of R and M contained 0.08 ± 0.02 mg/kg and 0.03 ± 0.00 mg/kg of Cd, respectively, while that of the reference area contained 0.01 ± 0.00 mg/kg. In the case of onion, the Cd burden found in M and the reference area was the same, with a value of 0.01 ± 0.00 mg/kg.

Table 10: Cd levels in vegetables of sampling areas, given as mean value ± standard deviation in mg/kg

Sample	Cd level in R site	Cd level in M site	Cd level in the Reference site
Parsley root	0.20 ± 0.12	0.04 ± 0.02	0.01 ± 0.00
Carrot root	0.08 ± 0.02	0.03 ± 0.00	0.01 ± 0.00
Onion	0.06 ± 0.01	0.01 ± 0.00	0.01 ± 0.00
Carrot leaf	0.01 ± 0.00	0.04 ± 0.00	0.01 ± 0.00
Parsley leaf	0.09 ± 0.01	0.05 ± 0.00	0.03 ± 0.00
Cabbage	0.12 ± 0.02	0.06 ± 0.03	0.01 ± 0.00
Lettuce	0.09 ± 0.01	0.09 ± 0.01	0.02 ± 0.00

As shown above, the samples of carrot leaf from site M were observed to contain higher amounts of Cd (0.04 ± 0.00 mg/kg) than those present in site R and the reference area (0.01 ± 0.00 mg/kg). Additionally, greater levels were detected in the cabbage collected from site R than in others. The maximum allowable limit (MAL) of Cd in parsley root, carrot root, and onion is 0.1 mg/kg, while that in carrot leaf, parsley leaf, cabbage, and lettuce is 0.2 mg//kg. It must be noted that the MAL for Cd was not exceeded in any of the tested vegetables [22]. Figure 3 shows the graphical representation of the data collected. Most vegetables grown in the contaminated site R contain a significant Cd burden than those selected from the other sites. Mining activities are one of the primary anthropogenic sources of heavy metal contamination and explain Cd's higher levels.

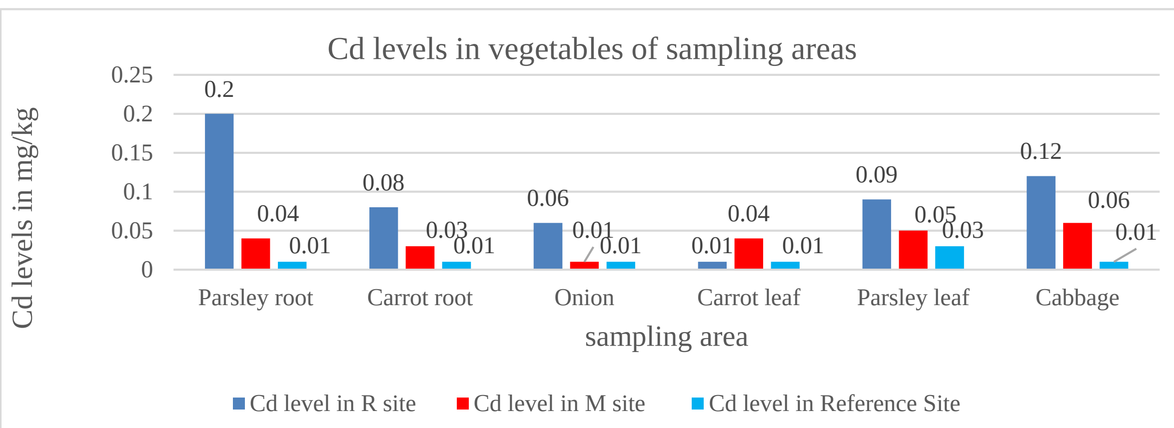

Figure 3 Cd levels in vegetables of sampling areas

3.1.7. Italy

Data for metals in fruits and vegetables were collected for an industrialized city in Italy by Sardinia's name. The findings in the study are discussed [23]. From Table 11, it can be observed that fresh leafy vegetables contained the highest concentration of Cd, with a mean value of 0.353 mg/kg, followed by roots and onions (0.195 mg/kg) and fresh fruiting vegetables (0.182 mg/kg). In contrast, the least Cd levels were detected in fresh citrus fruits.

Table 11 Mean Metal Levels in Foods from Italy(mg/kg) wet weight

Food Categories	Element	Mean concentration
Pulses Fresh and processed	As	0.126
	Cd	0.0426
	Pb	0.102
	Zn	13.240
Leafy Vegetables, Fresh	As	0.065
	Cd	0.353
	Pb	0.374
	Zn	12.783
Tomatoes, fresh	As	0.041
	Cd	0.101
	Pb	0.301
	Zn	6.867
Fruiting vegetables, fresh	As	0.045
	Cd	0.182
	Pb	0.139
	Zn	7.357
Roots and onions, Fresh	As	0.120
	Cd	0.195
	Pb	3.313
	Zn	4.933
Other Vegetables, fresh	As	0.061
	Cd	0.078
	Pb	0.074
	Zn	4.933
Citrus fruit, fresh		
	As	0.119
	Cd	0.009
Other fruit, fresh	Pb	0.152
	Zn	5.829
	As	0.055
	Cd	0.031
	Pb	0.379
	Zn	9.757

According to risk assessment in this study, the mean exposure in the European countries via food was of the value 2.3 µg/kg by weight per week, where the lower bound was 1.9 µg/kg by weight per week, and the upper bound was 3.0 µg/kg by weight per week. 3.0 µg/kg by weight per week was the mean value for the high contact, whose minimum value was 2.5 µg/kg b.w. Per week and the maximum value was 3.9 µg/kg b.w. Per week. Vegetarians majorly consume fruits and vegetables, and their intake is about 5.4 µg/kg by weight per week [23].

3.2 Chromium(Cr)

3.2.1. Croatia

Samples of seven types of vegetables were collected in 2016 from Varazdin, one of Croatia's most largely populated cities. Anomic absorption spectrometry (AAS) analysis was done to quantify the levels of several heavy metals in white potatoes, red potatoes, onions, carrots, beans, lettuce, and cabbage [Table 12]. The results of the Cr content discovered in each specie is listed below [10].

Table 12 Cr burden in some vegetables of Varazdin, Croatia, dl=detection limit of the instrument

Sample	Cr
White potato	<dl
Red potato	<dl
Onion	<dl
Carrot	<dl
Beans	<dl
Lettuce	<dl
Cabbage	<dl
Total	<dl

Additionally, three types of berries, namely rosehip, lingonberries, and blueberries, have also been studied in 2018 for their Cr burden, and the results are summarised below [12].

Table 13 Cr level in some berries of Croatia in mg/kg d.w.

Sample	Cr
Rose Hip	<0.018
Lingonberries	<0.018
Blueberries	<0.018

The tolerable daily intake for Cr^{3+} has been reported as 300 g/kg b.w., while no exact amount has been mentioned for Cr^{6+}[10]. Tables 12 and 13 show that the Cr burden exceeded the instrument's detection limit in all specified vegetables and berries. Hence, these selected species may not significantly add to the population of Croatia by the Cr burden.

Figure 4 shows that berry fruits take up less Cr than Cd uptake. For Cd, the levels found were less than 0.028 mg/kg, while for Cr, the values were less than 0.018 mg/kg in all three types of berry fruit.

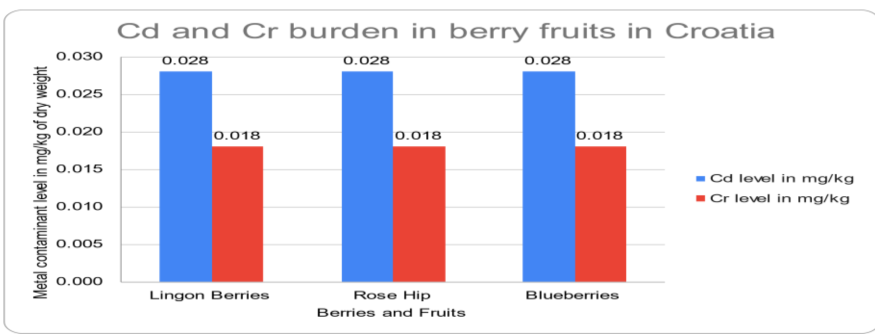

Figure 4 Cd and Cr burden in berry fruit in Croatia

3.2.2. Spain

Various fruits and vegetables in the Valencia, Spain region were assessed to determine the toxicity level of the heavy metals present in them. The samples were collected in 2010-2011, and the results for the analysis of Cr are shown in Table 14 [21]. From the values listed, it can be observed that olives and pickles contain the most generous amount of Cr with a mean weight of 0.3809 mg/kg, followed by spinaches and swiss chard, having about 0.0677 mg/kg. In contrast, peppers showed the least contamination by the metal, and their average amount was approximately 0.0119 mg/kg. The samples of all the fruits and vegetables were observed to contain levels higher than the LOQ (Limit of Quantitation), except in garlic, in which 20% of the samples had levels less than the LOQ.

Table 14 Levels of Cr in mg/kg fresh mass

Sample	>LOQ (%)	Mean
Oranges	100	0.0470
Strawberries	100	0.0129
Spinaches and swiss chard	100	0.0677
Lettuces	100	0.0285
Green beans	100	0.0180
Onions	100	0.0121
Garlic	80	0.0310
Peppers	100	0.0119
Aubergine, courgette, and cucumber	100	0.0178
Carrots and pumpkins	100	0.0136
Tomatoes	100	0.0189
Olives and pickles	100	0.3809
Apples and pears	100	0.0392
Sherry and plum	100	0.0325
Melon and watermelon	100	0.0219
Bananas	100	0.0549
Peach and apricot	100	0.0265
Grapes	100	0.0367
Cauliflower, cabbage, and broccoli	100	0.0434

3.2.3. Kosovo

In September-November 2017, a study was carried out in the Mitrovica region of Kosovo to investigate Cr metal concentrations in different apple types [24].

Table 15 Cr concentration (mg/ kg) in parts of apple species depending on rootstocks type from the Mitrovica region.

Rootstock	Tissues	Cr (mean ± standard deviation)
	Shoot	2.39±0.19
M106	Leaf	5.78±0.56
	Fruit	6.88±1.63
	Shoot	2.47±0.33
M26	Leaf	4.61±0.44
	Fruit	6.31±1.25
	Shoot	2.12±0.28
M9	Leaf	4.62±0.47
	Fruit	5.36±0.91

As evident from the data provided in Table 15, all the apple species' fruits contained more significant amounts of Cr than the other tissues. For example, the Cr burden in the fruit of M106 was 6.88±1.63 mg/kg, while its leaf and shoot contained 5.78±0.56 mg/kg and 2.39±0.19 mg/kg, respectively. The highest level was found in the fruit of the rootstock of M106 (6.88±1.63 mg/kg), followed by M26 (6.31±1.25 mg/kg). On the other hand, M9 reported the least contamination with a value of 5.36±0.91 mg/kg.

Compared with other Southern European countries such as Spain and Croatia, Kosovo has reported higher Cr levels. The observed significant burden could be due to the sampling regions' industrial location in Kosovo, leading to higher contamination.

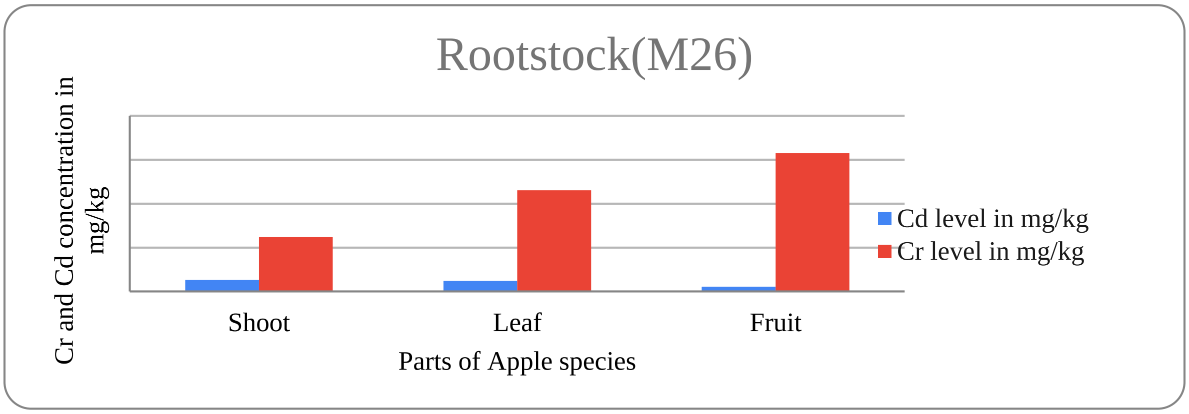

Figure 5 Cd and Cr level in rootstock (M26)

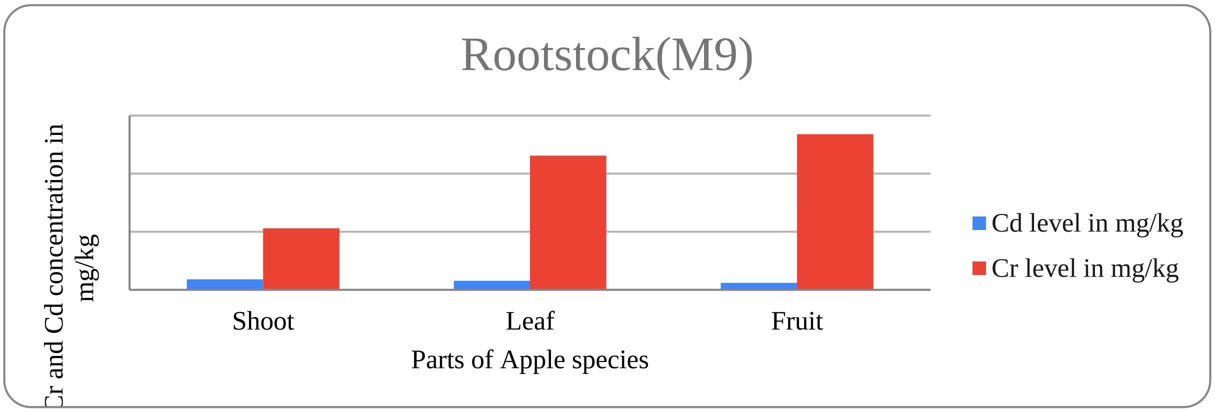

Figure 7 Cr and Cd level in rootstock (M9)

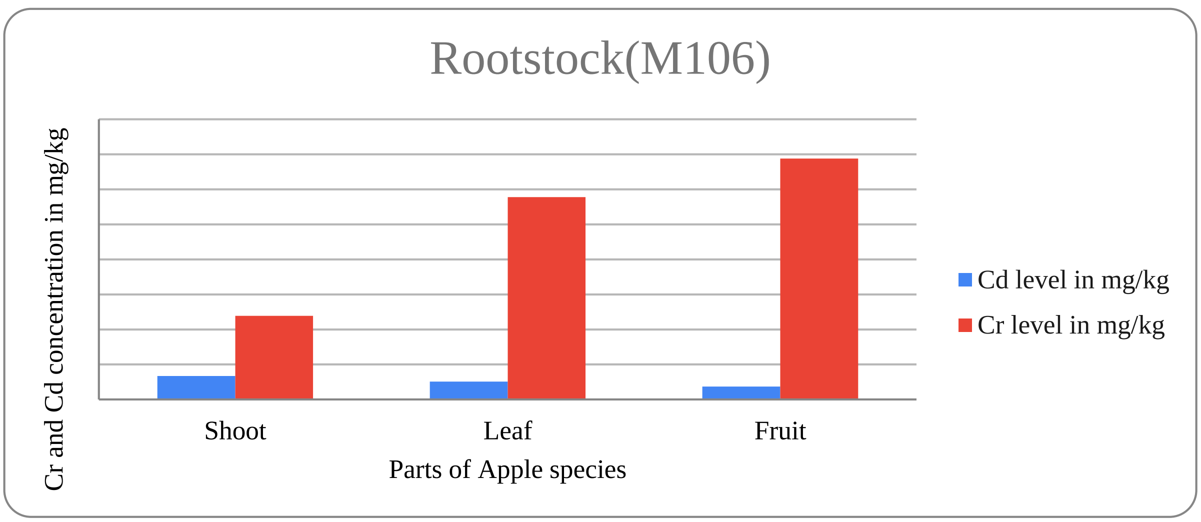

Figure 7 Cr and Cd level in rootstock (M106)

Figures 5, 6, and 7 compare apple species with different rootstocks in the Mitrovica region. The apple species are more prone to Cr uptake than Cd in Spain. The fruit from M106 contains 6.880 mg/kg Cr, while Cd is in a minimal quantity relative to Cr at 0.370 mg/kg.

3.2.4. Romania

Samples from three different crop roots, cabbage, potato, and celeriac root from an agricultural area in Ploiesti, were tested for Cr's presence [25]. The results showed a significant difference in the accumulation of Cr by all three crop roots.

Table 16 Cr levels are given as mean value ± S.D in mg/kg

Crop type	Cr
Celeriac root	7.2 ± 0.4
Potato root	4.7 ± 0.3
Cabbage root	17.5 ± 0.7

Cabbage root reportedly had the most significant uptake of Cr, which was observed to be around 17.5 ± 0.7 mg/kg, while potato root showed the least level of Cr burden (4.7 ± 0.3 mg/kg). If the crop root analysis results are compared with the same plants' soil content, it can be noted that the soil levels were undoubtedly higher than the roots for all the tested species. The celeriac root contained 7.2 ± 0.4 mg/kg of Cr, while its soil had 64.1 ± 3.0 mg/kg.

3.2.5. Greece

Various vegetables and fruits were examined for their Cr content in different areas of Greece, and the analysis results are shown below in Tables 17 and 18 [26].

Regarding the vegetables, Cr concentration ranged between 0.021 µg/g to 0.28 µg/g (Table 17). The maximum concentration was found in canned green peas (0.21 ± 0.03 µg/g), followed by onions (0.16 ± 0.07 µg/g), canned tomato (0.13 ± 0.04 µg/g) and cabbage (0.13 ± 0.08 µg/g). Mushrooms, by comparison, were observed to contain the smallest traces of Cr (0.04 ± 0.01 µg/g). It can also be stated that all canned vegetables, except spinach, contained higher metal accumulations than fresh ones. Tomato contained 0.09 ± 0.03 µg/g of Cr, while 0.13 ± 0.04 µg/g was observed in canned tomatoes.

Table 17 Cr levels in some vegetables given as mean value ± standard deviation in µg/g

Vegetables	Cr
Beans, green	0.09 ± 0.04
Beans, green, canned	0.10 ± 0.05
Cabbage	0.13 ± 0.08
Carrots	0.06 ± 0.05
Carrots, canned	0.07 ± 0.03
Celery	0.07 ± 0.03
Cucumber	0.07 ± 0.02
Eggplant	0.09 ± 0.06
Lettuce	0.05 ± 0.02
Mushrrom	0.04 ± 0.01
Mushroom, canned	0.07 ± 0.02
Onion	0.16 ± 0.07
Pea, green	0.18 ± 0.10
Pea, green, canned	0.21 ± 0.03
Potato	0.04 ± 0.02
Red beets	0.06 ± 0.01
Spinach	0.08 ± 0.03
Spinach, canned	0.08 ± 0.03
Tomato	0.09 ± 0.03
Tomato, canned	0.13 ± 0.04

From Table 18, it can be stated that the fruit samples did not contain very high amounts of Cr. The maximum level of Cr was found in apples (0.08 ± 0.02 µg/g), while apricot and melon both had inadequate amounts (0.02 ± 0.01 µg/g). Unlike vegetables, no significant difference was found in the levels of canned and uncanned fruits.

Table 18 Cr levels in some fruits given as mean value ± standard deviation in µg/g

Fruits	Cr
Apple	0.08 ± 0.02
Apricot	0.03 ± 0.01
Apricot, canned	0.02 ± 0.01
Grape	0.03 ± 0.02
Lemon	0.04 ± 0.01
Melon	0.02 ± 0.01
Orange	0.03 ± 0.01
Peach	0.05 ± 0.01
Peach, canned	0.05 ± 0.01
Pear	0.05 ± 0.01
Pear, canned	0.06 ± 0.04

3.2.6. Turkey

Some selected fruits and vegetables were chosen to be tested for their heavy metal contents in Kayseri and Tokat, Turkey, in 2005. The observations are summarized in Table 19 [27].

Table 19 Cr contents of canned foods marketed from Turkey given as mean value ± standard deviation in µg/g

Sample	Cr
Pea	0.19 ± 0.02
Mixed vegetable	0.33 ± 0.03
Tomato	0.52 ± 0.04
Stuffed grape leaves	0.32 ± 0.03
Pickle	0.50 ± 0.05
Bean	0.37 ± 0.04

The Cr levels varied between 0.19 ± 0.02 µg/g and 0.52 ± 0.04 µg/g. Peas contained the least amount of Cr (0.19 ± 0.02 µg/g), while tomato was analyzed to show the highest level (0.52 ± 0.04 µg/g) of the heavy metal.

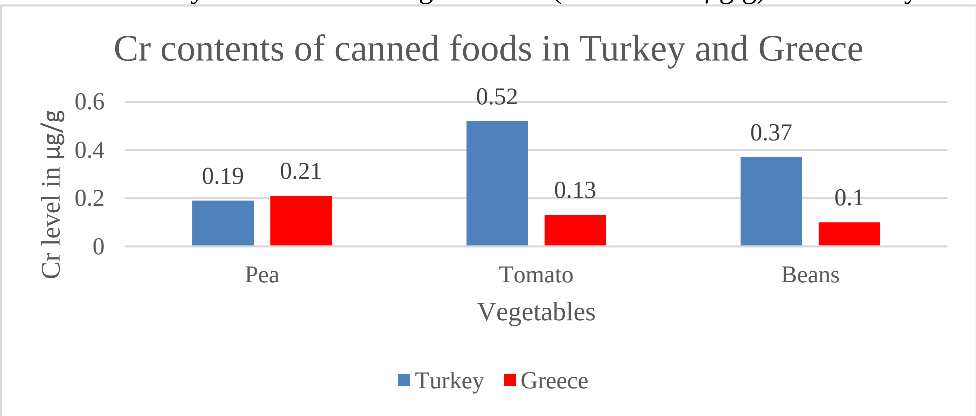

Figure 8 Cr levels in Turkey and Greece

When compared with the data collected from other southern European countries, the Cr content in tomatoes is significantly higher in Turkey (0.52 ± 0.04 µg/g) than in Greece (0.13 ± 0.04 µg/g). Similarly, the canned beans analyzed in Turkey contained 0.37 ± 0.04 µg/g, while 0.10 ± 0.05 µg/g was reported in Greece. However, no significant difference is found in peas (Figure 8) [26].

3.2.7. Italy

Between 2014 and 2016, particular fruit and vegetables were inspected for some trace elements, including Cr. The analysis was done with the help of inductively coupled plasma mass spectrometry (ICP-MS). Table 20 illustrates the findings of the study conducted [28].

Table 20 Average Cr content (fresh weight) in some fruits and vegetables, given as mean value ± standard deviation in mg/kg

Sample	Cr
Apricot	0.008 ± 0.003
Peach	0.016 ± 0.042
Prune	0.005 ± 0.003
Olive	0.034 ± 0.033
Grape	0.010 ± 0.002
Tomato	0.007 ± 0.007
Pepper	0.015 ± 0.017
Pea	0.013 ± 0.012
Lettuce	0.085 ± 0.056
Cabbage	0.007 ± 0.004

The highest levels were reported in lettuce (0.085 ± 0.056 mg/kg) followed by olives (0.034 ± 0.033 mg/kg) whereas tomatoes (0.007 ± 0.007 mg/kg) and cabbage (0.007 ± 0.004 mg/kg) contained the least amount of Cr.

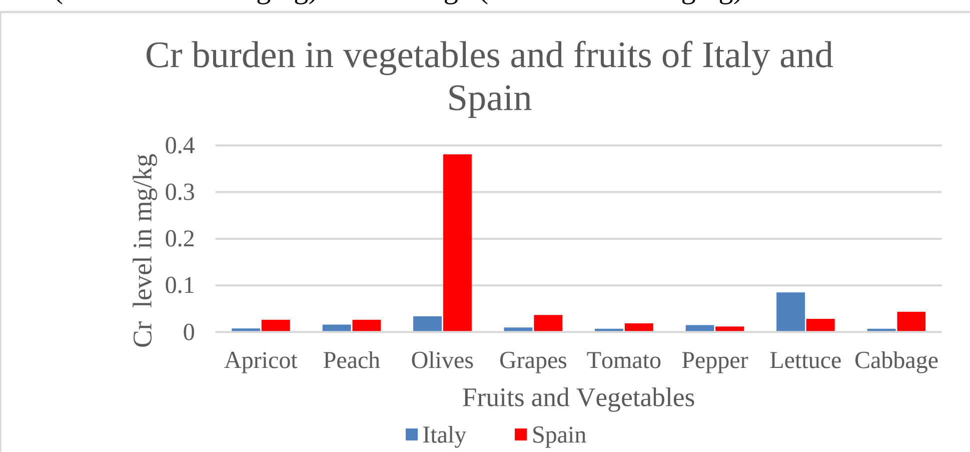

Figure 9 Cr levels in Italy and Spain

As illustrated in Figure 9, most of the fruits and vegetables tested in Spain showed a higher Cr presence than those analyzed in Italy, with a few exceptions, such as lettuce and peppers. Cr in tomatoes was 0.0189 mg/kg in Spain, whereas the amount in Italy was 0.007 ± 0.007 mg/kg. Additionally, grapes (0.0367 mg/kg), peaches (0.0265 mg/kg), and olives (0.3809 mg/kg) also showed greater content of Cr in Spain. [21]

In Italy, olives have substantial quantities (0.034 ± 0.033 mg/kg) of Cr found in them compared to other fruits and vegetables. Spain contained concentrated Cr levels that the fruits and vegetables in Italy.

3.3 Cd and Cr with respect to the location

In general, different countries in southern Europe have different metal concentrations. These levels vary from country to country. Industrial areas generally have greater Cd and Cr levels than non-industrialized areas; this may result from the factory effluent leaching into the lithosphere by rain or the atmosphere. Rural and urban areas also have differences in the values of metal uptake. Generally, urban areas contain industries; thus, a greater metal concentration is present in the environment, which goes into the soil or water the plants and fruits take up. The greater the concentration in the surrounding soil and environment, the greater is the level of these metals in fruits and vegetables. Soil collected from Sicily, Italy, showed higher Pb, Cr, Zn, and Hg levels than other European cities. The primary cause in this city was associated with traffic pollution that spread all over the city. The Cd levels were a natural enrichment by weathering and pedogenic processes.[29]. This comparison with other European rural areas indicates that the urban soils have more significant metal levels. Thus, there is greater uptake in fruits and vegetables.

3.4 Health risks of Cd and Cr

According to research carried out in the past, continuous/consistent exposure to Cd (Cd) in edibles and water results in the deposition of Cd in kidneys. This phenomenon can cause dysfunction in the kidneys. (ATSDR, 1993). Nriagu and Pacyna, in 1998, concluded in their research in 1988 that nickel (Ni) proves lethal in case it crosses the allowed concentration in food [30]. Not only this, but Cd can cause many disorders, as discussed in an earlier study; an extensive Cd contact may result in lung disorders like alveolitis, emphysema, and bronchiolitis. Renal effects (kidney-related) may also be caused by Cd's breathing in volatile forms [31]. Lead and Cd are included in the most bounteous heavy metals and are toxic. A continuous intake of these heavy elements in the food is associated with several disorders, especially cardiovascular problems, nervous kidney issues, bone disorders and dysfunctionalities. [32].

3.4.1. Valencia, Spain

In Valencia, Spain, a study showed the metal uptake in infants and children; "The per capita estimated weekly intake was 82% and 55% for infants, while children (3–10) were 77% and 50%. The PTWI considered is 7 μg/kg b.w. Per week, as reported by the Regulation (EC) 1881/2006 setting maximum levels for certain contaminants in foodstuffs". [21]. The infants and children were more at risk compared to adults. Children have growing bodies. Their food intake is used in the development and growth of the body more when compared to adults. When heavy metals enter their body, and during the growth process can become a part of their lipid and protein tissue, which can be very dangerous as it can cause many dysfunctionalities and abnormalities. Cr is especially dangerous in this regard as it can oxidize the DNA and RNA on entering the bodies, which can also cause genetic disorders.

3.4.2. Turkey

Gastrointestinal malignancies are observed all over the world. The metal levels were studied for different fruits, vegetables, and soil in a cancerous region in Eastern Turkey. The standard levels and study results are shown in the figures below [33].

Table 21 Heavy metals in fruits and vegetables (standards, ppm)

Element	Vegetables and fruits
Pb	6-9
Cd	<0.5
Cu	2-20
Ni	1-10
Co	0.02-0.5
Mn	10-20
Zn	5-100
Cr	0.1-1

As shown in Table 21, the Cd levels were significantly higher in all the samples' fruits and vegetables. The mean Cd level in fruits and vegetables should be less than 0.5 ppm, but the cancerous region had a mean value of 25 ppm with a standard deviation of 28 ppm. According to the study, the mean zinc levels were 44 ppm, while the allowed levels in fruits and vegetables can be 5 ppm-100 ppm. Similarly, other metals like copper and cobalt were found to have higher levels than the standard levels present in fruits and vegetables. The study concluded that such high levels of the metals eventually resulted in gastrointestinal cancers when long-term exposure to these metals occurred. Hence, the conclusion was drawn that metals such as Cd, cobalt, and lead contributed to gastrointestinal cancer in the region.

Table 22 Cd concentration in fruits and vegetables in the sampled cancer region (ppm)

Centre no.	Cd
1	30
2	35
3	29
4	35
5	32
6	26
7	13
8	13
9	18
10	17
11	25
12	23
13	12
14	22
15	35
16	24
17	30
18	29
19	24
Mean	25
S. D	28

Figure 10 shows the graphical representation of Cd levels reported in different centers of a cancerous region in Turkey. The maximum level was 35ppm, while the minimum level was 12ppm.

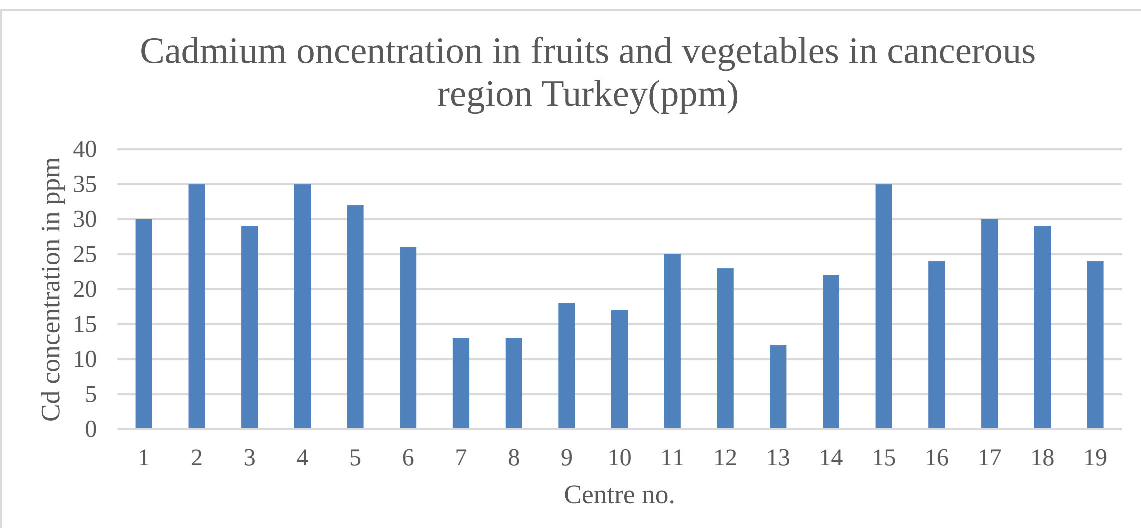

Figure 10 Cadmium concentration in fruits and vegetables in a cancerous region in Turkey

3.3 Safe cultivation levels in Serbia

In the Vojvodina Province of Serbia, a study was conducted on the health risks associated with the metal contained in fruits and vegetables[34]. Potatoes, onions, and carrots are the widely eaten foods in Vojvodina, Serbia, hence were collected for the study. The maximum levels established by the European Commission Regulation (EC 1881/2006) for Cd are 0.1mg/kg, and those for lead are 0.1mg/kg. Cd was considered safe for cultivation because the Cd level was less than the European Commission Regulation's ordained maximum limit and implemented by Serbian Regulation 2019.

A study was carried out in Vojvodina, Serbia. The daily intakes for adults and children were calculated, and the basis was the mean detected concentration found in the fruits and the vegetables. The study was based on regular items of fruits and vegetables. The study showed that the metal intake was minimal compared to the safe limit of the metal intake. This meant that the population was not at risk of high metal contaminants; the children and adults were not much at risk.

The target hazard quotient was calculated for each heavy metal under study, and the ratio of the intake of the metal level and the respective metal level allowed. The target hazard quotient for a single heavy element was calculated to be less than 1, so it was concluded that consuming vegetables with a single heavy element does not cause many health disorders. However, the study also concluded that it is more hazardous for children to have heavy elements than adults. The study also revealed that Cd was the element with more mobility than others; hence vegetables in the region were more prone to Cd uptake. Cultivating vegetables less prone to Cd uptake in the area was allowed.

4. Recommendations

1. It has been observed that washing vegetables and fruits with 10% acetic acid were more effective in removing heavy metals than tap water, 5% acetic acid, 5% NaCl, and 10% NaCl. Since 10% of acetic acid is acidic; thus, it has a lower pH value, so it is more effective in cleaning heavy metals. However, washing vegetables with 8% ginger solution was most effective in removing heavy metals due to the pH changes.
2. Three processes, washing, shelling, and soaking in vinegar, reduce heavy metals' rates from 63 to 81 (%) for Cd and 46 to 56 (%) for Cr.
3. Before eating, washing vegetables and fruits is a common, easy, and effective way to reduce metal ions from them. Studies reveal that metals from anthropogenic sources are usually in water-soluble form; they stick on vegetables' outer surfaces. Thus washing them with clean/ tap water removes metal ions as they dissolve in water, which takes them away, leaving behind clean vegetables that are safer to eat.[35]
4. It is advised to people living in urban areas where industries are more than farms that they should not eat leafy green vegetables but instead consume others because the risk of heavy metals in these vegetables is highest.
5. It is also advised that farmlands should be located away from traffic routes and industries or vice versa. Selling vegetables on highways or traffic routes should also be banned.[36]
6. It is recommended that vegetables be grown near sewerage water; the household kitchen garden vegetables should be preferred as the kitchen water irrigates them.
7. Washing with chemical solutions of hydrogen peroxide, citric acid, and biological extracts of lemon, radish, and sodium carbonate decreased heavy metal amounts in vegetables and fruits.[37]
8. In cities near industries, fruits are significantly contaminated with heavy metals like Cr (Cr) and Cd (Cd). It is advised to cultivate fruits away from the mining areas because there is a greater concentration of heavy metals.
9. We need to control soil contamination to reduce the concentration of heavy metals in fruits and vegetables. For that purpose, using less fertilizers and pesticides is advised because excessive use leads to soil contamination.
10. To reduce Cr contamination of food, avoiding metal cans for packaging and processing food items is recommended because it can lead to a higher Cr concentration in food items stored in these cans.
11. It has been observed that the concentration of Cd and other heavy metals in fruits and vegetables can be controlled by introducing organic agricultural practices. These practices are to be done under the controlled use of Cd phosphorus fertilizers in cultivation.
12. Another reason for soil contamination with heavy metals is the gaseous effluent released from the industries. When industries release their gaseous wastes untreated in the atmosphere, it comes down on earth as acid rain, which increases Cd uptake in food by contaminating the soil. Many factories and industries do not treat their contaminated water before releasing it, and contaminants leach into the soil, exposing plants to metal contaminants like Cd and Cr. It is recommended that the government keep strict vigilance on these industries to avoid food contamination with heavy metals hazardous to human health.

5. Conclusion

The review article focused on the Cd and Cr levels in fruits and vegetables of different countries in southern Europe. It indicated the metal uptake was dangerous, whether high or low. There have been not one but several studies that have been carried out on the heavy metal uptake in fruits, vegetables, and crops. Different metals have a different rate of uptake by plants, whether it is fruits or vegetables, depending on certain factors, as discussed earlier. Greater uptake of Cr than Cd has been observed in fruits than the vegetables in southern European countries. Generally, metals have been observed to be more abundant in leafy vegetables than fruits. These metals are very deadly to human health once they have crossed the allowed levels in the body. The Cd's ability to cross the blood-brain barrier in the body can have a very harmful impact on health. These metals have different detrimental effects on living organisms in different oxidation states. They can react with compounds in the atmosphere under certain conditions to form more lethal forms. Thus, the use of these metals needs to be minimized as much as possible in different compounds. The anthropogenic source eventually enters the biosphere, risking the lives of humans and animals.

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