

Monitoring of heavy metals and trace elements contamination in the soil and vegetables and air pollution in the Republic of Macedonia

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Abstract: The concentration of toxic heavy metals Lead (Pb) and Cadmium (Cd), trace element Zinc (Zn) and metalloid Arsenic (As) in some vegetable and soil samples have been evaluated in four cities named as; Skopje, Veles, Ohrid and Kocani, of Republic of Macedonia. In general, the excess of concentration of such toxic metals, trace element and metalloid in vegetables and soil are dangerous and can cause serious problems to all organisms and heavy metal bio accumulation in the food chain especially can be riskful to the human health. A total of 30 samples of few vegetables and 30 samples of soils have been collected from the surface layer (0–20 cm) from the respective areas in the Republic of Macedonia. In our present work, we determined the concentrations of above metals / elements by inductively coupled plasma – optical emission spectroscopy (ICP-OES) method and found that these concentrations are in relation to the prescribed standards criteria of WHO-World health organization and FAO-Food and Agriculture Organization.

Keywords: Toxic heavy metals, health risks, concentration in (mg/kg), World Health Organization (WHO) and Food and Agriculture Organization (FAO).

Introduction

Naturally, all vegetables and soils contain varying concentrations of heavy metals, trace elements and metalloid depending on the type of parent material from which the soil was formed and from where these vegetables were cropped [1]. All these heavy metals, trace element and metalloid may be added to meadow soils through the use of mineral and organic fertilizers, direct defecation and urination by animals, pesticides, soil amendment practices (e.g. liming and gypsum applying) [2, 3], waste disposal, waste incineration, urban effluent, traffic emissions, atmospheric deposition (e.g. radionuclides, vehicle and industrial productions) [4-6] and long-term application of wastewater in agricultural land [7-10].

The accumulation of toxic heavy metals, trace element and metalloid in the agricultural soil is a growing apprehension due to the food safety matters and prospective health risks as well as its harmful effects on the ecosystem of soil [10, 11]. These heavy metals, trace elements and metalloids have strange characteristics and few of them are such as; (i) these metals, trace elements and metalloids can be essential or useful to the crops i.e. vegetables until particular levels but can be toxic when exceeding precise thresholds, (ii) they do not decay with time, (iii) they are always present at a background level of non-anthropogenic basis, their input in soils being related to weathering of parent rocks and pedogenesis and (iv) they frequently occur as cautions which strongly interact with the soil matrix, consequently, heavy metals in soils can become itinerant as a result of varying environmental conditions. Therefore, based on the above consequences, this study reflects to a “Chemical timing bomb (CTB)” [10-13]. On behalf of the literature survey [1-16], the present work has been carried out on the percentage concentration and accumulation of toxic heavy metals in vegetables and soil of some regions in the Republic of Macedonia which are settled in the Balkan Peninsula, and the results are presented and found satisfactory with a standard curriculum of WHO [17] and FAO [18] organizations. Hence, one can assume that, it is worth meaning to investigate the level of toxic heavy metals in vegetable and soil.

A. Macro and Micro Nutrients

The Macro nutrients / elements are required by the human body in good quantities for the appropriate metabolism mechanism and functioning of body organs. These are such as; Calcium (Ca), Magnesium (Mg), Sodium (Na), Potassium (K) and Iron (Fe). On the other hand, the micro nutrients / elements are required by the human body in trace quantities and are essential for maintaining various body functions and metabolic functions. Metals such as Silicon (Si),

Nickel (Ni), Boron (B), and Vanadium (V) are essentially for human and animal's body (but toxic at higher levels). These have some biological mechanisms in the crops as well as the animals, but essentiality for humans and their requirements are under research [19]. The toxicity details of a few of them which cause human disorders expressed such as [20] and also we have studied their concentration in vegetables (crops) and soil in the Republic of Macedonia in the present work.

(i) Lead (Pb):- Metal element Lead (Pb) is a toxic heavy element which enters into the body (human / animals) through the air, water and food and cannot be removed by washing fruits and vegetables [20, 21]. It is a serious cumulative body poison, which can affect many organ systems in the body. Its exposure to high levels can hang out damage the kidney, brain and ultimately cause of death and long-term exposure result in decreased performance in some tests that evaluate the activities of the nervous system; weakness in fingers, wrists, or ankles; small increases in blood pressure; and anemia. Others are abdominal pain, anemia, arthritis, attention deficit, back problems, blindness, cancer, constipation, convulsions, depression, diabetes, migraine headaches, thyroid imbalances and tooth decay [20, 21].

(ii) Cadmium (Cd):- This is also a toxic heavy element which has been obtained generally from vegetables (crops) / soils and influence on the cardiovascular system of humans. In some studies on rats and rabbits, cadmium exposure was shown to increase blood pressure, or to cause cardiac lesions [20]. However, studies of exposed human have found positive, negative and no association between cadmium exposure and hypertension. This suggests that if the metal (Cd) does affect blood pressure, the magnitude of the effect is insignificant as compare with to other determinants of hypertension. Death rates for cardiovascular disease do not appear to be elevated in populations exposed to heavy metal (Cd) by inhalation or in the diet. Overall, the weight of evidence suggests that cardiovascular effects are not a sensitive end point indicator for metal (Cd) toxicity [20, 21]. Evidence that Cadmium (Cd) exposure may affect kidney vitamin D metabolism with subsequent disturbances in Calcium (K) balance and bone density, suggests that decreased bone density, particularly in elderly women, may be a significant adverse effect of kidney (Cd) accumulation [22 and references therein].

(iii) Arsenic (As):- Arsenic (As) is the main constituent of more than 200 mineral species, of which about 60% are arsenate, 20% sulfide and sulfosalts and the remaining 20% include arsenides, arsenites, oxides and elemental arsenic. The most common of the arsenic minerals is arsenopyrite (FeAsS), and arsenic is found associated with many types of mineral deposits, especially those including sulfide mineralization (B. J. Alloway, 1995) [22 - 23]. Volcanic action is the most important natural source of arsenic, followed by low-temperature volatilization [22]. Inorganic arsenic is found in groundwater used as drinking-water in several parts of the world: in Taiwan (Chen et al., 1994) [24] and more recently in most districts of Bangladesh (Dhar et al., 1997) [25] and (Biswas et al., 1998) [26]. Mining, smelting of non-ferrous metals and burning of fossil fuels are the major industrial processes that contribute to anthropogenic arsenic contamination of air, water and soil. Historically, use of arsenic-containing pesticides has left large tracts of agricultural land contaminated. The use of arsenic in the timber preservation treatment has also led to environmental pollution.

(iv) Zinc (Zn):- It is a trace element and is an essential nutrient for humans that is necessary for the function of a large number of metalloenzymes, including alcohol dehydrogenase, alkaline phosphatase, carbonic anhydrase, leucine aminopeptidase, and superoxide dismutase. Zn deficiency has been associated with dermatitis, anorexia, growth retardation, poor wound healing, hypogonadism with impaired reproductive capacity, impaired immune function, and depressed mental function; an increased incidence of congenital malformations in infants has also been associated with Zn deficiency in the mothers. Zn deficiency may also have an impact on the carcinogenesis of other chemicals, although the direction of the influence seems to vary with the carcinogenic agent [22 and references therein].

The main emphasis of the present experimental work is to determine the percentage concentration values of toxic heavy metals, metalloid and the trace element. The most important factor is to controll the spatial distribution of toxic heavy metals, metalloid and the trace element and to identify the possible origins (sources) of these metals in the soil samples collected from four different cities in the Republic of Macedonia. In conclusion of this, one can say that these worthy results led to accomplish scientific way for the contamination control and further monitoring of the toxic heavy metal accumulation in the soil. Similar work has been carried out by Afshin Qishlaqi and Farid Moore [10 and references therein], and we found a good agreement between our results and the results obtained by others [10-16].

Materials and Methods Used

A. Area Descriptions and Samples

The present study area is located in the Republic of Macedonia is a country situated in the central part of the Balkan Peninsula, positioned between 40°50' and 42°20' North geographic latitude and between 20°27'30" and 23°05' East geographic longitude. On the west, it borders Albania (191 km), on southern Greece (262 km), on east Bulgaria (165

km) and Serbia on the north (231 km). The country has a population of 2.045 million inhabitants with 0.44 million people living in the capital of Skopje. Diverse climate types and sub-types are combination of main ones - Mediterranean, continental and Alpine climate. It is a mountainous country with many lowlands. The average altitude of its territory is 850 meters. According to the country's spatial plan, 1.9% of the territory is covered by water (lakes), 19.1% are plains and valleys, while its largest part consists of 79% of hills and mountains. Plains and valleys in Macedonia occupy a total area of 4,900 km². The total territory of the Macedonia's highlands is 354.26 km² or 1.38% of its territory. Midland areas cover 7,528.22 km² or 29.28% of the country. Lowland areas below 1,000 meters are spread throughout the country. The total area of lowlands is 17,830.52 km², or 69.34%. Forests occupy around 30% (947.653 ha) of the territory of the Republic of Macedonia. The total agricultural arable land covered 546.000 ha in 2005, out of which arable land and gardens account for 82 %. Pastures occupy 682.000 ha.

The agricultural sector is deemed as one of the most important sectors for the Macedonia's economy. And also, here the soil is highly operative. Hence, the pollution of soil, water, and air became inevitable as a result of the anthropogenic influences. For present experimental work, all the samples of vegetables and soil were collected from the four cities named as; Skopje, Veles, Ohrid and Kocani in the Republic of Macedonia during the time period of November to December 2012. A map of study regions / sites has been depicted in figure 1, where the red spots indicates the four cities; Skopje, Veles, Ohrid and Kocani in the Republic of Macedonia.



Figure. 1: Map for the study regions / sites in the Republic of Macedonia.

We have collected 30 vegetable samples, each sample is safe to eat and in quantitatively 1.0 kg, (i.e. 5 samples of Carrot, Cabbage, Cauliflower, Leaks, 4 samples of Parsley, 3 samples of onion, 2 samples of Beets and 1 sample of Parsnips) from the above said four cities. At the same time, 30 samples of soil were collected where these vegetables were cropped. The vegetable samples were collected along the same gradients. Soil samples were taken in the immediate vicinity of the roots of the crop samples from 0 to 20.0 cm depth. We collected our samples for the analyses in such order that; the suitable part for eating of each vegetable is taken into account. All the soil or vegetable samples were quartered separately to provide sub-samples. The samples (vegetables) details along with the regions / sites are given in Table 1, and much more details in about the present samples are given in our recent article submitted to the journal, entitled "Indian Journal of Horticulture", Jan. 2014" with "ID number – DE/IJOR/SUB/IHJ/20011409343" by author Biljana B. P. et. al, [27].

B. Method Used and Analysis

Before doing experimental analysis, it necessary to gave a pre-treatment analysis to vegetable and soil samples. The soil samples were air-dried in the laboratory and sieved < 3 mm. A part of the soils were ground in a porcelain mortar < 100 mesh. The suitable part for eating of the all the vegetables were washed twice under a strong jet of water then again washed with de-ionized water and later on dried on the filter paper. After this, the vegetable samples have been ground in a blender. Finally, both the samples of vegetables and soil were dried in a special type of drier at temperature 85 °C until 10 to 12 hours in our laboratory, and later on all these samples were kept in the twofold transparent polyethylene bags at room temperature very carefully.

The present experimental work has been performed such as it is depicted in Fig. 2 by a flow chart. Much more descriptions in about experimental procedure, one can be seen in our recent contribution submitted by authors Biljana B. P. et. al, in a journal “Indian Journal of Horticulture”, Jan. 2014” with “ID number – DE/IJOR/SUB/IHJ/20011409343” [27].

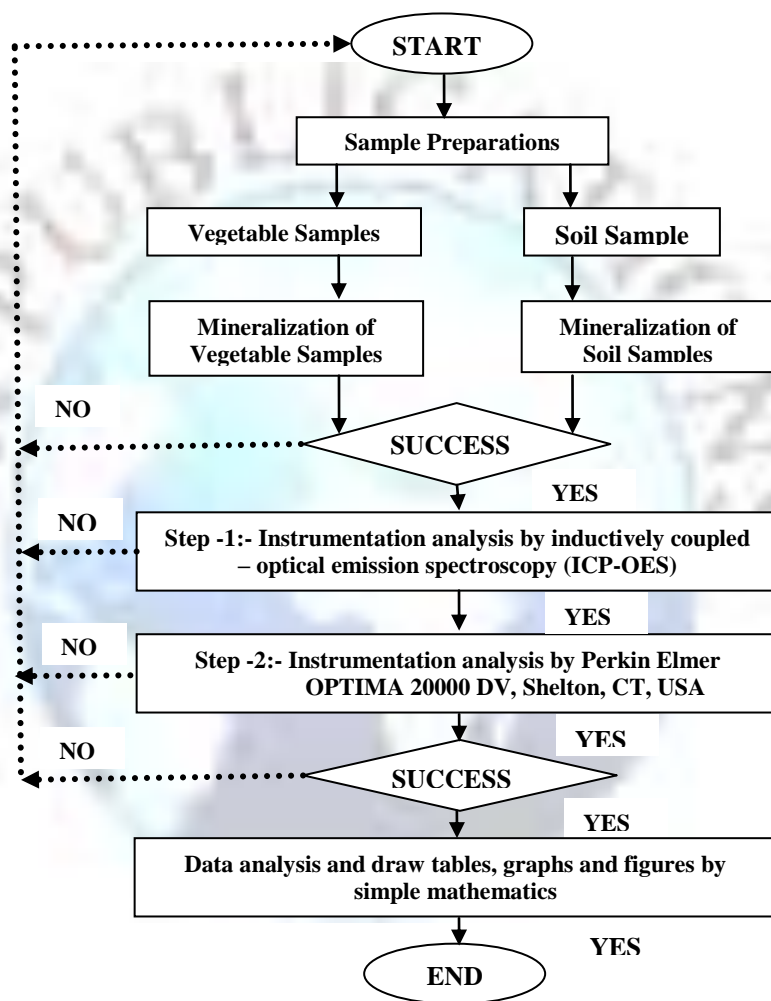


Figure. 2: A flow chart for the performance of the present experimental work

Results and Discussions

On behalf of our study and statistical data analysis, first we have determined the concentration of heavy metals, metalloids and trace elements in the soil from where the vegetable samples have been cropped. The measured concentration values along with the standard values for such elements in the soil have been depicted in Table 1 for all the regions in R. of Macedonia. All measured values of the concentrations of heavy metals, metalloids and trace elements in the soil samples, i.e. where the vegetable sample was cropped, and also were in well-defined standard / criteria prescribed by the Regulations of the Republic of Macedonia (S.I list naRM118/2008G) [28] also from the World Health Organization (WHO) [17] and Food and Agriculture Organization (FAO) [18].

Table 1: For the concentration values of heavy metals, metalloid and the trace elements in the soil, where the vegetable samples have been cropped in all regions.

Name of regions / site	Heavy metal (Pb) in mg/kg	Heavy metal (Cd) in mg/kg	Metalloid (As) in mg/kg	Trace element (Zn) in mg/kg
Skopje (the cement Usje surroundings).	40.00*	0.60*	15.00*	110.00*
	39.53**	1.69**	14.32**	133.30**
Skopje (Jurumleri)	40.00*	0.60*	15.00*	110.00*
	22.64**	0.32**	10.67**	99.31**
City Ohrid	40.00*	0.60*	15.00*	110.00*
	28.42**	0.04**	13.47**	106.60**
City Kocani	40.00*	0.60*	15.00*	110.00*
	20.66**	1.22**	10.54**	86.30**
Veles (Basino village)	40.00*	0.60*	15.00*	110.00*
	42.62**	1.22**	20.34**	166.30**

* Means:- the standard value prescribed by the Regulations of the Republic of Macedonia (S.I list naRM118/2008G) [28] also from the World Health Organization (WHO) [17] and the Food and Agriculture Organization (FAO) [18].
 And ** means:- the measured by experimentally in our Laboratory in R. of Macedonia.

Further the concentration values of all such elements / metals in vegetable samples have been explained by Figures 3 - 7. Therefore, the Figure 3 has been shown for the concentration values in the vegetable samples in the region of Skopje (the cement Usje surroundings). From this figure, one can see that the maximum concentration of the toxic heavy metals in our present work for Pb is 1.261 mg/kg in Leek and Cd is 1.027 mg/kg in Carrot, the metalloid As is 0.057 mg/kg in Cabbage and the trace element Zn is 0.251mg/kg in Carrot. These results have shown here are in a good agreement with the results find by other workers in the same field [9-16]. We are also reporting the minimum concentration of such elements first time and these are in such orders; Pb is 0.031 mg/kg in Parsley; Cd is 0.053 mg/kg in Parsley, As is 0.012 mg/kg in Parsley and Zn is 0.026 mg/kg in Cauliflower. It is also clear from the figure 2 that the concentration of metalloid (As) is very low as compared to the concentrations of other heavy metals Pb and Cd and the trace element Zn is on an average in the region of Skopje (the cement Usje surroundings).

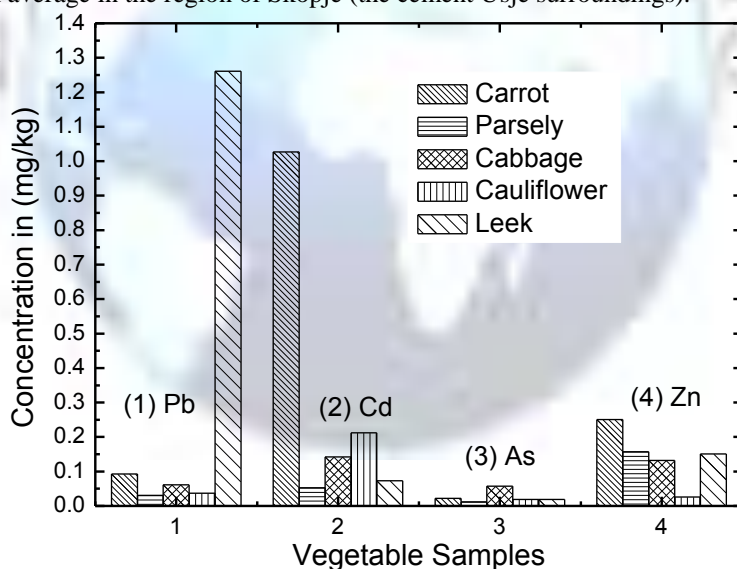


Figure. 3: For the concentration values of heavy metals (Pb & Cd), metalloid (As) and trace element (Zn) in the vegetable samples in the region of Skopje (the cement Usje surroundings).

Similarly, the Figure 4 has been shown for the concentration values of heavy metals (Pb and Cd), metalloid (As) and trace element (Zn) in the vegetable samples in the region of Skopje (Jurumleri). From this figure, one can find that the maximum concentration of the toxic heavy metals; Pb is 0.078 mg/kg in Onion and Cd is 0.029 mg/kg in Carrot, the metalloid As is 0.021 mg/kg in Parsley and the trace element Zn is 0.133 mg/kg in Beets. These results have shown here are in a good agreement with the results find by other workers in the same field [9-16]. The minimum concentrations of such elements are in such orders; Pb is 0.023 mg/kg in Cauliflower; Cd is 0.013 mg/kg in Cauliflower, (As) is 0.012 mg/kg in Onion and Leek and Zn is 0.009 mg/kg in Carrot. It is also clear from the Figure 4 that the concentration of metalloid (As) is very low as compared to the concentrations of other heavy metals Pb and Cd but the trace element Zn is much higher in the region of Skopje (Jurumleri).

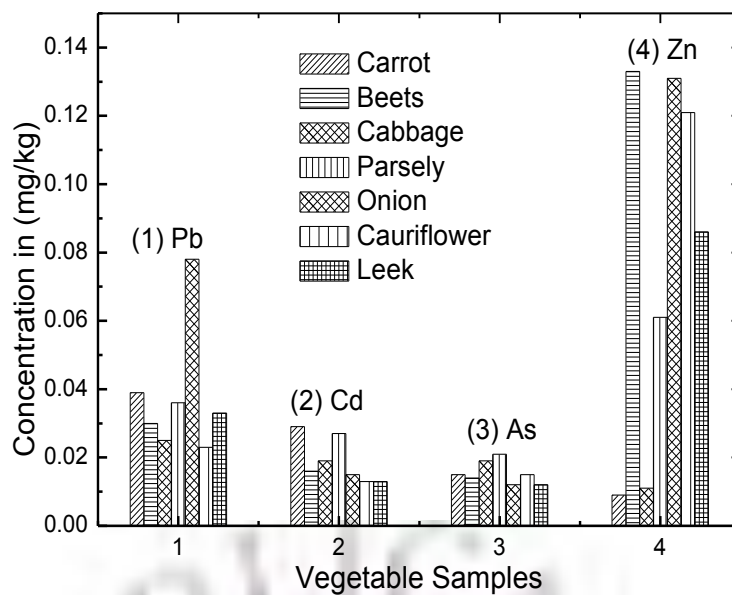


Figure. 4: For the concentration values of heavy metals (Pb & Cd), metalloid (As) and trace element (Zn) in the vegetable samples in the region of Skopje (Jurumleri).

Figure 5 has been show for the concentration behaviour for heavy metals, metalloid and the trace element in the vegetable samples in the city Ohrid. From this figure, we have drawn the conclusion that the maximum concentration of the toxic heavy metals; Pb is 0.093 mg/kg in Carrot and Cd is 0.091 mg/kg in Leek, the metalloid (As) is 0.066 mg/kg in Beets and the trace element Zn is 0.095 mg/kg in Cauliflower. It is also clear from the Figure 5 that the concentration of heavy metal (Cd) is very low except the vegetable sample parsley as compared to the concentrations of other heavy metals Pb, metalloid (As) but the trace element Zn is much higher in the region

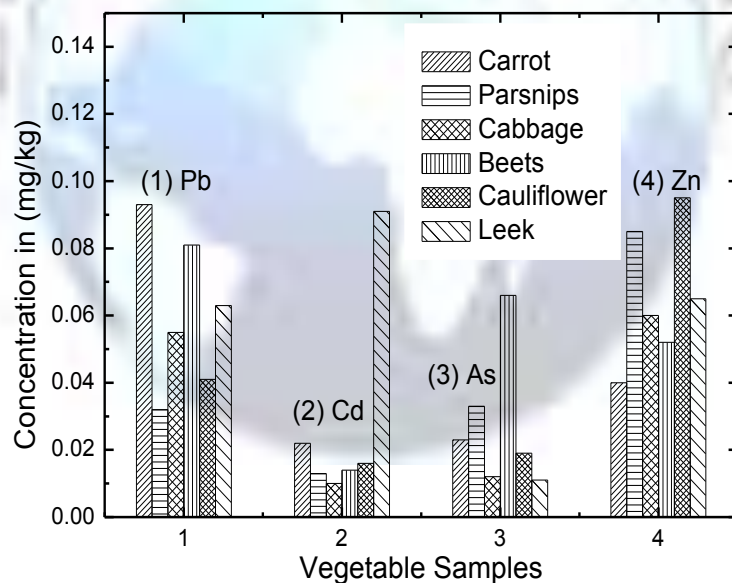


Figure. 5: For the concentration values of heavy metals (Pb & Cd), metalloid (As) and trace element (Zn) in the vegetable samples in the city Ohrid.

Finally, Figure 6 and 7 have been shown for the concentration of such heavy metals / metalloid and the trace element in the agriculture regions / where the vegetable sample have been cropped, the city Kocani and Veles (Basino village) respectively. From figure 6, we can conclude that the concentration values for heavy metal Pb is much more higher for Onion and Leek as compare to all vegetable samples as well as all elements; heavy metal, metalloid and trace element. And also heavy metal Cd is much more less as compared to (As) and trace element (Zn).

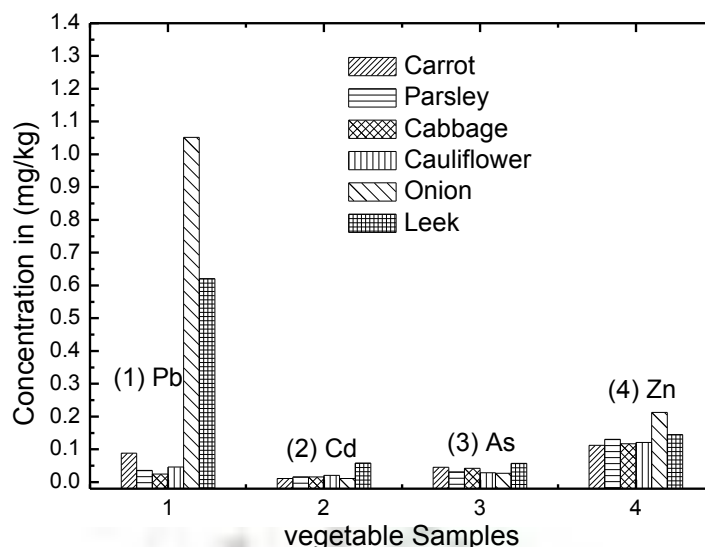


Figure. 6: For the concentration values of heavy metals (Pb & Cd), metalloid (As) and trace element (Zn) in the vegetable samples in the city Kocani.

Same from Figure 7, we can say that that the concentration values for heavy metal Pb is much higher for Onion and Cabbage as compare to all vegetable samples as well as all elements; heavy metal, metalloid and trace element.

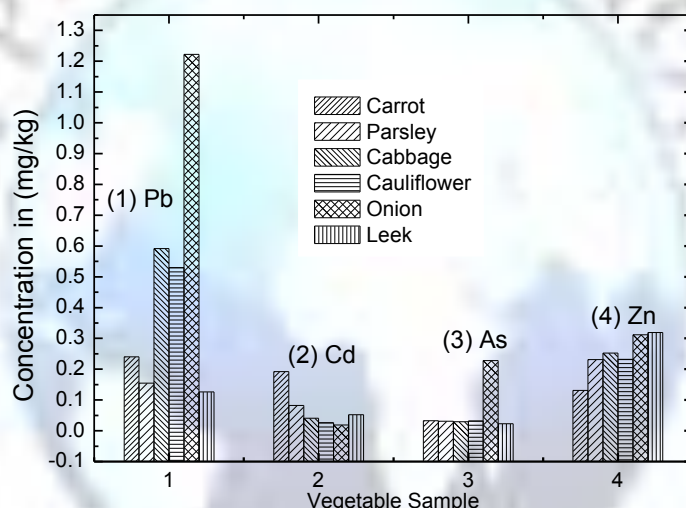


Figure. 7: For the concentration values of heavy metals (Pb & Cd), metalloid (As) and trace element (Zn) in the vegetable samples in the region of Veles (Basino village).

Finally, the studied average values of heavy elements, metalloied and trace elements have been found on the order of (Pb - 0.478 mg/kg) in Basino village > (Cd - 0.301 mg/kg) in Usje > (As - 0.063 mg/kg) in Basino village > (Zn – 0.246 mg/kg) in Basino village. The similar studies with the same method have been performed by K. Ehsanul et. al., in 2012 [29], L. Chutima et al., in 2012 [30] and by O. Etem et. al., in 2012 [31] and achieved the same results. The concentration of the accumulation level of the studied elements in both the samples Soil and vegetables in this experimental research was: Pb > Zn > Cd > As.

Air Pollution

Air pollution in the Republic of Macedonia originates mostly from energy sector, industrial sector and transport due to the use of low quality natural resources such as coal (brown coal), iron and other metal ores (chromium, lead, zinc). Energy and transport sectors, apart from their greatest impacts in terms of air pollution, are also sectors where significant technical measures and structural changes should be implemented. However, the share emissions from waste burning at open landfills, as well as emissions from agriculture resulting from the application of fertilizers and enteric fermentation with domestic animals in air pollution is also significant. Air pollution from the mentioned sources is done at local level, however the movement of pollutants into the atmosphere causes problems of national, regional and global levels. Therefore, solving the problem of transboundary air pollution requires international cooperation.

The Institute of Public Health of the Republic of Macedonia – Skopje / Ohrid follows sediments of 4 m.m. in the zone of Housing, Industrial Zone, and the periphery administrative and business area of the city of Skopje. Therefore, through the analysis of heavy metals such as; Iron (Fe), Manganese (Mn), Copper (Cu), Zinc (Zn), Lead (Pb), Cadmium (Cd) with a special technique named Atomic Absorption Spectrometer (AAS) method is a unique way to tolerate it. In this regard, we used the apparatus PERKIN ELMER HGA 700 graphite and AAS PERKIN ELMER-3110, with flame detector. In order to this, the source data from Republic of Macedonia (Air pollution Laws, Official Gazette, number. 20/74) [32] is an evaluation of air hygienic quality, which has been realized in the “Breathing area”. And the results of air pollution during the years 2009 - 2012, in the Republic of Macedonia in terms of average annual concentration of sediments is lowest in the city of Kocani and its values / per years are in the order of 19.47 mg/m² in the year 2012, (22.95 mg/m² in 2011, 58.21 mg/m² in 2010 and 33.24 mg/m² in 2009), while the city with higher air pollution in sediments, according to the annual average concentrations, but under MAC (maximum allowable concentration) is Skopje and Ohrid town 168.53 mg/m² in 2012 (152.26 mg/m² in 2011, 137.63 mg/m² in 2010 and 130.61 mg/m² in 2009). The hygienic quality of the air in the Republic of Macedonia during the 2009 - 2012 is depicted in Table 2. The Source of these data/results is “Air pollution Laws, Official Gazette, number. 20/74” [33].

Table 2: Monitoring values of the hygienic quality of the air in the Republic of Macedonia in 2012 (pollutant specified – sediments).

Institut of Health Care	Number of samples for measurment	Average of annual concentrations (mg/m ²)	Min - maximum (mg/m ²)	Number of samples above the * MAC
Skopje	333	174.0	29.5 - 659.9	44
Veles	84	100.4	10.7 - 628.7	6
Bitola	46	51.1	24.0 - 225.9	0
Kochani	48	22.95	1.14 - 44.18	0

* MAC - maximum allowable concentration (300 mg/m²/mes).

Conclusions and Final Remark

The toxicity of the metals in vegetables and soils depends upon the comparative intensity of exposure of crops to the contaminated soils as well as the deposition of toxic elements in the polluted air by sedimentation. Different metals show the toxicity at different concentrations and can be potentially toxic at sufficiently high concentrations. However, certain metals exhibit toxic effects, even at relatively low concentrations.

The results show that toxic heavy metal, metalloid and trace element accumulation was a severe phenomenon occurring in vegetables, soils and crop food stuffs consumed in all around four cities of the Republic of Macedonia.

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