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Chapter

Evaluate the Impact of Soil Contamination on Vegetables and Fruits

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Abstract

In the chapter will be presented: scientific substantiation on the models used worldwide to evaluate the contamination of soil, respectively vegetables and fruits; development of theoretical models to evaluate the impact of soil contamination by heavy metals on vegetables and fruits; testing of theoretical models in real conditions based on data obtained from laboratory; development of mathematical models to evaluate the impact on soil contamination on vegetables and fruits and thus on consumers health. The research presented in the chapter aim to develop some original models on the correlation between the level of soil contamination, respectively the remanence of heavy metal in vegetables and fruits harvested for consumption in fresh state. The statistical mathematical models elaborated by the interpolation of the experimental data are models with practical applications in both scientific research and agricultural management.

Keywords: contamination of soil, vegetables and fruits, heavy metal, mathematical models

1. Introduction

The rapid development of industry and urbanization in developing countries has led to the chaotic increase in levels of toxic heavy metals in the environment. In addition, heavy metal contamination of agricultural soils and crop plants in these countries, because of the use of industrial waste water it can have negative effects on human health. Other sources of heavy metals from agriculture include manure, fertilizers and pesticides, and contamination from the air due to excessive use of cars [1].

The most important pollution in agriculture is the accumulation of heavy metals which are very toxic to soil, water, plants and humans. Although heavy metals have an key role in nature for soil conservation, their concentration above certain limits can have toxic effects [2]. Therefore, in order to understand the phenomenon of accumulation in soil and plants, it is necessary to know the following definitions [3]:

- Bioamplification is the accumulation of toxic substances in the tissues of tolerant organisms as the trophic level increases. This increase can occur if: the substance cannot be decomposed by environmental processes; the concentration of the substance gradually increases as it moves into a food chain; the impossibility of internal degradation or elimination of the substance.

- Bioaccumulation means increasing the concentration of toxic substances in certain tissues of organisms, over a period of time, due to absorption from food and environment (air, water, soil). The longer the body's exposure to the toxic substance, the more serious the risk of intoxication, even if the level of intoxication is not very high.

- Bioconcentration is the process of accumulating a toxic substance in an aquatic organism, when its source is only water.

Bioconcentration and bioaccumulation occur within an organism, bioamplification occurs at trophic levels (food chain).

Excess accumulation of heavy metals in crops from contaminated agricultural soils it results in soil pollution and low quality of food. Soil is the key factor, it is the basis of the food chain that determines food safety. Vegetables and fruits are plants that are commonly used in food due to their content in nutrients (iron, calcium, proteins), vitamins, minerals, fiber, beneficial to health. Consumption of contaminated fruit and vegetables entail risks for health therefore many researchers have studied food safety in this regard [1, 4–8].

The key functions of metals in plants are involvement in redox reactions and an integral part of enzymes. The essential metals for plants Fe, Cu, Mo, Zn play a major role in the formation of enzymes, the transport of electrons, the sustaining metabolism. In the soil, metals are known as essential trace elements, others non-essential (Hg, Ag, Pb, Ni, etc) and as ultra-trace elements [2].

Heavy metals are present in the environment and they are pollutants in both aquatic and terrestrial ecosystems. The most hazardous heavy metals (Cr, Ni, Cu, Zn, Cd, Pb, Hg) in the environment have three characteristics: their persistence in the environment, toxicity to the soil, water, plants, and organisms, their bioaccumulation in the structure of the soil, the composition of water, the tissues and organs of plants, the body of organisms [9].

The negative effect of metals on the activity of microbes in the soil indirectly affects plant growth. Plants that grow under the stress of heavy metals consume more energy for their survival, what affects other physiological processes, such as: absorption of nutrients, photosynthesis, respiration, metabolism and reproduction, water balance. Due to metallic stress, a lot of reactive oxygen accumulates in the plant [10–12]. Plant reactions to heavy metal toxicity may include: necrosis, chlorosis, senescence and wilting, slowing growth, metabolic disorders, loss of yield, nutrient deficiency, reduced ability to fix atmospheric nitrogen, the small number of seeds and, finally, death [2].

The accumulation of soil metals in plants depends on a number of factors, such as: the structure of the plant, the life cycle of the plant, the vigor of the plant, the pH of the soil, the depth of the root system, temperature, partial pressure of oxygen, carbohydrate level, respiration rate, nutrient exchange and microbial activity [2, 13].

Generally, plants can be integrated into three categories, given their reaction against metals: exclusions, accumulators and indicators based on the mechanism of action for to survive under stress, as suggested [2, 14]. Exclusions react to entering of metals into the vegetative aerial parts making this impossible by stopping the metal in the roots. Accumulators are plants that accumulate metals in the vegetative aerial

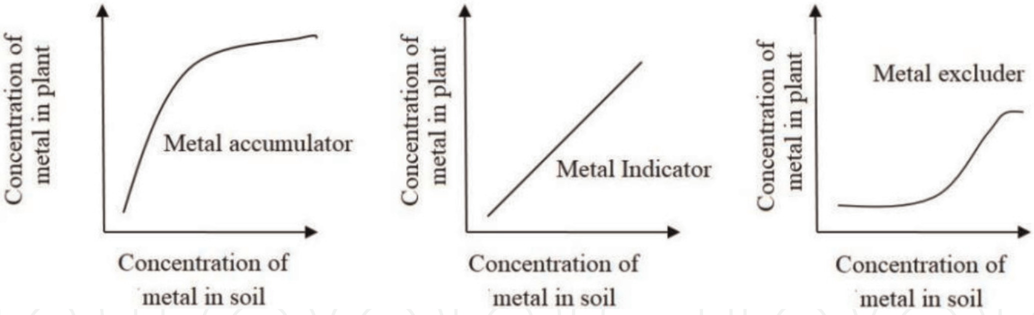


Figure 1.
The shape of curves for mathematical modeling of heavy metal concentrations in the soil-plant system [15].

parts greater than the metals in the soil. Indicators are species that continuously accumulate metals in vegetative aerial tissues tolerating metal concentrations and indicating the amount of metals in the soil. The behavior of plants in relation to the increase in the concentration of heavy metals in the soil is shown in **Figure 1** [15].

Hyperaccumulation of heavy metals is the process by which plants accumulate excess metals 0.1–1% of their dry weight, and concentrate them into roots, stems, or leaves [16]. A hyperaccumulating plant can accumulate and/or tolerate high levels amounts of metals. As an ecological adaptation, some plant species have the ability to grow in heavy-contaminated metal soils and accumulate them [2, 17]. As hyperaccumulating species, about 400 species of plants from 22 families are known. The Brassicaceae family contains a large number of these plants, which includes 87 species from 11 genera [15].

According to the paper [18] the soil factors that influence the absorption of metal by plants are: the concentration of metal in the soil, the processes and properties of the soil and the vegetable factors. These are: the refeeding of ions in the rhizosphere, the kinetic parameters that adjusts the absorption of metals from plants, the tolerance of metal by the plant.

For the assessment of phytoremediation of soils by means of plants, there are many specialized papers [12, 19–21], which uses the bioconcentration factor (BCF) and the translocation factor (TF) as parameters. Through BCF, the concentration of metals in plant tissues is determined in relation to their growth medium, while TF determines way metals are translocated in the aerial vegetative parts of plants [12, 19–21]. As plant species used to remove metals from the environment, they can be listed: peas [1, 22], lettuce [23], wood species (poplar, willow, ash) [24], oleander [25], flax and hemp [26], jute, rooster crest, field thyme [27], rapeseed and Indian mustard [28], sunflower and corn [29], cucumber [22, 30], cherry tomato [15], sweet pepper [31], cabbage and broccoli [32], spinach [33, 34].

The way plants tolerate, absorb, transport, capture, sequester and bioaccumulate metals differs depending on several factors, such as: plant species, phenology, type of metal, soil type and quality, climate, type of source that contaminates, chemical and physical behavior of the plant, environmental factors [12].

The following will be presented different parameters for the assessment of soils and plants with heavy metals, calculated with different mathematical formulas. This parameters have been used in many papers by researchers who have studied this major environmental issue currently existing globally at the moment.

- **The contamination factor (CF)** is the ratio of the metal tracked from the soil to the background value of the heavy metal, expressed in mg/kg^{-1} dry matter, as it is presented in the papers [35, 36] and is determined with the formula (1):

$$CF = \text{Concentration of heavy metal in the sample/Background value of the heavy meta} \quad (1)$$

The contamination factor values are classified as such: $CF < 1$ (low contamination), $1 < CF < 3$ (moderate contamination), $3 < CF < 6$ (high contamination) and $CF > 6$ (very high contamination) [35, 36].

- **Geoaccumulation index (I_{geo})** can be used when desired effective environmental planning of pollution. It is used successfully to assess soil contaminated with heavy metals from natural or anthropogenic sources. It can be determined with the formula (2):

$$I_{geo} = \log_2\left[\frac{C_n}{1.5 \bullet B_n}\right] \quad (2)$$

-where, Cn shows the measured value of the metal concentration, in mg kg^{-1} dry matter; n și Bn are the geochemical background value of the corresponding metal, measured in mg kg^{-1} dry matter and 1,5 is the compensation factor in the background concentration of heavy metals. In the study [35] shale values were used as background values.

In the papers [35, 36] geoaccumulation index has been classified into the following six categories: unpolluted environment ($I_{geo} \leq 0$), unpolluted environment to moderately polluted environment ($0 < I_{geo} \leq 1$), moderately polluted environment ($1 < I_{geo} \leq 2$), moderately to heavily polluted environment ($2 < I_{geo} \leq 3$), highly polluted environment ($3 < I_{ge}o \leq 4$), strongly to extremely polluted environment ($4 < I_{geo} \leq 5$).

- **Enrichment factor (EF)** is also important to be able to assess the level of heavy metal pollution from anthropogenic sources, and it is calculated as the ratio of the concentration of the studied metal (the chosen reference metal must be in combination with very fine surface solids and occur naturally and evenly in the environment) and geochemical background, expressed also in mg $kg^{-}1$ dry matter, calculated with the formula (3):

$$EF = \text{sample metal/Background metal} \quad (3)$$

The values of enrichment factor were classified into: $EF < 1$ (soil without enrichment in Ref. metal), $1 < EF < 3$ (soil less enriched in Ref. metal), $3 < EF < 5$ (soil moderately enriched in Ref. metal), $5 < EF < 25$ (soil severely enriched in Ref. metal), $25 < EF < 50$ (soil very severely enriched in Ref. metal) si $EF > 50$ (soil extremely severely enriched in Ref. metal) [35, 37].

- **Degree of contamination (DC)** is presented in literature [35, 36] as a simple evaluation method for controlling anthropogenic pollution, as an indication of dangerous or not. A classification has been proposed for degree of contamination in three categories: $DC < 6$ (low degree of contamination), $6 < DC < 12$ (moderate degree of contamination) and $12 < DC < 24$ (considerable degree of contamination). It can be calculated as the sum of the contamination factor of each metal concerned, with the following formula (4):

$$DC = \sum nCF \quad (4)$$

- **Potential Ecological Risk** is a mathematical model that evaluates as the degree of soil pollution based on toxicity from heavy metals as well as assessment of environmental threats because of metals in response to environmental factors. The model is important because it shows which metal is more dangerous to the environment [38]. In the eqs. (5), E_{ri} is calculated for the determination of environmental risk and PERI (Eq. (6)) is calculated for determination as the sum of all the risk values presented by heavy metals in the soil.

$$E_{ri} = T_{ri} \bullet CF \tag{5}$$

where T_{ri} represents the value of the toxic or lethal response; and CF is the contamination factor.

$$PERI = \sum_{f=1}^{n} E_{ri} \tag{6}$$

The two formulas helps establish the degree of threat to soils because to heavy metals by the indication of limits and to measuring the environmental sensitivity for the metals concerned. Therefore, [36] classified the two parameters as follows: $E_{ri} < 40$ (low ecological risk), $40 < E_{ri} < 80$ (moderate ecological risk), $80 < E_{ri} < 160$ (considerable ecological risk), $160 < E_{ri} < 320$ (high ecological risk) and $E_{ri} > 320$ (very serious ecological risk). In the same way, PERI was classified as follows: PERI < 95 (low ecological risk), 95 < PERI <190 (moderate ecological risk), 190 < PERI < 380 (considerable ecological risk) and PERI >380 (very high ecological risk) .

Soil pollution with each metal was determined in the paper [1, 39] using the **pollution load index, (PLI)**, calculated as the ratio of the concentration of heavy metal in polluted soils to the concentration of heavy metal in unpolluted soils.

In the paper [40] the level of chemical pollution of the soil was determined using the **anthropogenic coefficient (Kc),** (Eq. (7)) of the concentration of a metal in a sample, calculated as the ratio of the content to the metal in a studied land (C) to the base level of the metal (CF).

$$K_c = C/CF \tag{7}$$

It is known that soil pollution due to anthropogenic activities may have different sources, therefore in the paper [40] the **total pollution index (Zc)**, (Eq. 8)) was calculated as a result of a group of heavy metals in a studied area.

$$Z_c = \sum_{i=1}^{n} K_c - (n-1) \tag{8}$$

where, K_c it is the anthropic coefficient of the concentration of a metal in a sample; n is the number of samples analyzed.

In mathematical modeling of the phenomenon of accumulation of metals in the soil-plant system, depending as a lot of factors such as was previously mentioned, it is necessary that the behavior of the plant be regularly monitored in order to study the prediction of dynamics. For this purpose, in the papers [40, 41] was determined the **biological absorption coefficient**, noted **(K_i^{bp})**, (Eq. (9)).

$$K_i^{bp} = C_i^r / C_i^p \tag{9}$$

C_i^r is the content of the i-th heavy metal in a plant, mg/kg; C_i^p is the content of the i-th heavy metal in soil, mg/kg.

In the paper [42] was determined the **normalized difference vegetation index (NDVI)**, (Eq. 10)), from mining areas where the soil is polluted with heavy metals. This index (NDVI) depend on the following factors: the topographic position index (TPI), wind speed (WP), precipitation (P), atmospheric dustfall (D), and surface temperature (W).

NDVI it is useful to be able to determine the cover of vegetation from an area, and it is calculated as the ratio of the value of the difference and the total value both near infrared bands, as well as visible infrared bands, and is calculated with the formula (10) [42]:

$$\mathrm{NDVI} = \mathrm{NIR} - \mathrm{red}/\mathrm{NIR} + \mathrm{red} \tag{10}$$

under NDVI este normalized difference vegetation index, NIR arată the value of the near-infrared light, iar roșu arată the value of the visible infrared light.

The study [42] concluded that atmospheric dust was the main factor that increased the heavy metal content in the soil, being strongly influenced by wind speed and topography of the soil.

- **Mitotic index (MI),** helps to determine the number of cells in the leaves, as a growth parameter of the plant, and is determined as a ratio between the number of cells that divide and the total number of cells in the leaves, expressed as a percentage (%), and calculated with the formula (11) [31]:

$$\mathrm{MI} = (\text{number of dividing cells}/\text{total number of cells})\ \mathrm{x}100 \tag{11}$$

In a statistical analysis [30] which describes the modeling of heavy metals in the soil-plant system, rice was used in the study and was used as algorithms: multiple linear regression (MLR), support vector machines (SVM), random forest (RF), and cubist. They have helped predict the bioaccumulation coefficient of metals in rice and to identify the potential for transfer of metals into the tissues of rice plants. The flow diagram of the study [30] carried out in China is shown in **Figure 2**.

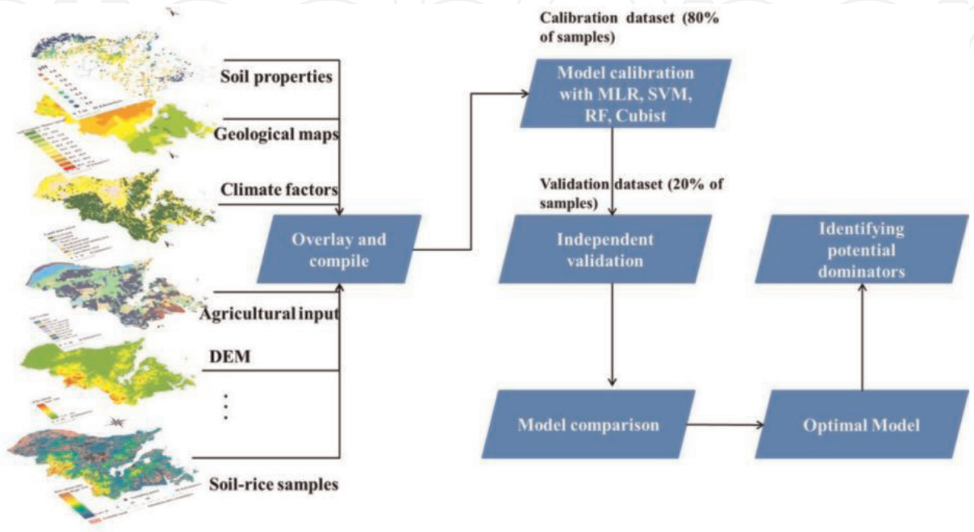


Figure 2.
Flowchart of the modeling of heavy metals in soil–rice system [30].

In order to establish the relationship between soil and the environment, in science and engineering, many forms of models have been used, such as they are: the Philip model, the Richard equation, the Hortonn model [43].

Modeling the distribution of heavy metals and their bioaccumulation in plants has been a major concern for many researchers because the concentration and distribution of heavy metals because the concentration and distribution of heavy metals concern both soil and crop quality. Therefore, the use of a precise model is essential for estimating the real values of heavy metals in the soil and their distribution for effective management in agriculture.

Three models (linear, logarithmic, polynomial, and quadratic models) have often been developed to test their adequacy to experimental data correlated to heavy metals [43, 44]. Validation of these models by direct analysis and comparison of the experimental data with the precise ones, indicates the percentage deviation (DV), determined with the relation (Eq. (12)):

$$D_v = \left[\frac{D_p + D_e}{D_e}\right] x100 \tag{12}$$

where, D_v is deviation, D_p is predicted values and D_e is experimented values [43].

Regression models has been widely used in the prediction of soil properties, especially the degree of pollution, because of their ease and wide use. These models show a global model of the problem studied and through a single regression equation can represent the process [43, 45].

Statistical models for predicting metal concentrations in plants have as main independent variables metal concentrations in the soil. Based on the available analytical data, the concentrations are total or only the bioavailable portion.

2. Experimentation of impact of soil contamination on vegetables and fruits

2.1 General principles of experimentation

It has been shown that when environmental conditions allow heavy metals to infiltrate the soil and, implicitly, groundwater, whole food chain is degraded and there is a direct risk of pollution. The dissemination of metals in food chains place as follows: contaminated soil with toxic metals - plants that absorb metals from the soil in the root, stem, leaves and fruit- humans and animals who eating plants contaminated with dangerous toxic metals and drinking water that can circulate through groundwater and surface water that has metals drains in them.

The danger of contamination of soil and plants with heavy metals depends primarily on the species of the plant and the properties of the soil, and second, the amount and concentration of the metal in the soil. But on the other hand the existence of chemical elements that can change the effect of metals and other substances, can reduce or amplify the absorption or adsorption processes in the soil. The adverse effect depends on the mobility and solubility of heavy metals in the soil. As control measures, there must be optimal conditions for passing metals from the soil solution into stable forms.

In plants, the concentration of metals varies depending on the species of the plant, its period of development, the vegetative organs (roots, stems, leaves, shoots, etc. branches, buds, flowers, fruits). There are plant species that can preferentially accumulate heavy metals according to their vegetative organs, as well as varying amounts. Therefore, in polluted areas, especially mining areas, it is not recommended, to ingest especially green vegetables, because toxic metals quickly get into the leaves due to foliar absorption.

Therefore, for areas at risk of soil pollution and implicitly of plants, the population must selectively consume the plants that grow on those contaminated soils. The selection of plants for consumption is based on the following criteria:

- the frequency with which a given species of plant is eaten;
- taxonomy, the family to which the genus or species of the plant belongs;
- the surface on which it grows and in what quantity a certain species is found to that place;
- the period of growth and vegetation of the plant;
- the vegetative part of the plant that can be eaten (only the fruit, only the leaves);
- the resistance of the plant to diseases and pests;
- the ability to spread the plant.

In this study, the following vegetables and fruits were used in a controlled environmental experimentation:

- vegetables - carrot (*Daucus carota*) grown as root, parsley (Petroselinum spp.) grown as leafy vegetables, cucumbers (*Cucumis sativus*), then increase the fresh lettuce over time;
- fruits - strawberries (Fragaria spp.); raspberry (*Rubus idaeus*).

These vegetables and fruits have been chosen because they are frequently eaten, and grown by the inhabitants of suburban areas of Romania, being essential and rich in micro-macronutrients, proteins, antioxidants and vitamins beneficial to the human body.

The selected vegetables also have a high capacity of accumulating metals without the phytotoxicity of the plants being observable by consumers. The vegetables studied had a relatively short life cycle (about 60 days) and developed well in pots, in the greenhouse, where a controlled environment was created.

2.2 Actual development of experiments

The heavy metals studied were copper, lead and zinc introduced into potted soils in four different concentrations for the solution of each metal, namely: 1.5%, 3.0%, 4.5% si 6.0%. The solutions were prepared individually, using distilled water, as a solvent and copper sulfate, lead acetate and zinc sulphate, as reagents. 250 ml solution of each metal (Cu, Pb, Zn) with the concentrations for each metal was placed, at 1 kg of soil.

Soil contamination with metal solutions and their concentrations was carried out by homogenizing them evenly in the soil prior to planting in pots, no additional until harvest.

In pots, carrot seeds, parsley, cucumber and salad seedlings, raspberry bushes and strawberry stems were planted, which were 1 year old at the time of planting.

At the same time, the reference samples were also made, which consisted of planting seeds, seedlings, bushes and stems in fertile, uncontaminated soil.

The basic properties of the soil used in the experiments were: PH 6.2, moisture 15.4%, particle elements <18 mm 85–95%, total phosphorus 0.8%, total potassium 1%, total nitrogen 2%.

The analysis of the metal content for samples from soils and plants was made on ash resulting from the samples. The methods and techniques used were consistent with the recommendations developed by [46, 47] using an atomic absorption spectrophotometer [48]. The method used was the spectrophotometric method (atomic absorption in flame) [48, 49].

2.3 Experimental results

Table 1 presents the content of metals in soil uncontaminated and in soli contamination with Cu, Pb and Zn separated concentrations.

The experimental data obtained for vegetables are presented in the **Table 2** and for fruits in the **Table 3**. Based on them, the mathematical modeling was done.

Also, aspects with the plants during the experimental research are shown in the **Figure 3**.

Metal	Concentrations of metals in soil, [%]				
	0	1.5	3.0	4.5	6.0
	Content initial of metals in soil (C_{is},), [mg/kg]				
Cu	17.6	58.9	267.2	525.1	680.8
Pb	6.75	48.7	84.7	117.7	285.2
Zn	39.8	202.7	534.8	921.7	1052.3

Table 1.
The content of metals in soil function of four concentrations.

Vegetable	Concentration of heavy metals, [%]	The contents of metals in vegetables (C_p), [mg/kg]		
		Cu	Pb	Zn
Carrot roots	0	7.6	5.4	20.2
	1.5	9.3	8.3	70.3
	3.0	8.5	9.0	73.5
	4.5	10.8	35.3	119.4
	6.0	10.5	44.0	110.5
Parsley leaves	0	4.5	6.1	32.4
	1.5	10.1	7.5	176.9
	3.0	8.1	14.9	185.0
	4.5	10.4	24.8	245.8
	6.0	10.7	45.2	349.9

Vegetable	Concentration of heavy metals, [%]	The contents of metals in vegetables (C_p), [mg/kg]		
		Cu	Pb	Zn
Cucumbers	0	5.2	2.7	34.1
	1.5	6.0	12.8	45.0
	3.0	8.5	12.0	65.3
	4.5	8.3	9.5	84.5
	6.0	10.0	11.8	106.3

Table 2.
The experimental data from vegetables.

Fruit	Concentration of heavy metals, [%]	The contents of metals in fruits (C_f), [mg/kg]		
		Cu	Pb	Zn
Strawberry	0	4.1	2.7	9.9
	1.5	17.5	—	—
	3.0	15.5	4.0	25.8
	4.5	—	7.1	—
	6.0	—	3.2	24.8
Raspberry	0	15.7	6.2	47.9
	1.5	13.8	—	—
	3.0	10.9	—	—
	4.5	19.8	5.3	35.4
	6.0	—	3.0	—

Table 3.
The experimental data from fruits.

3. Development of mathematical models

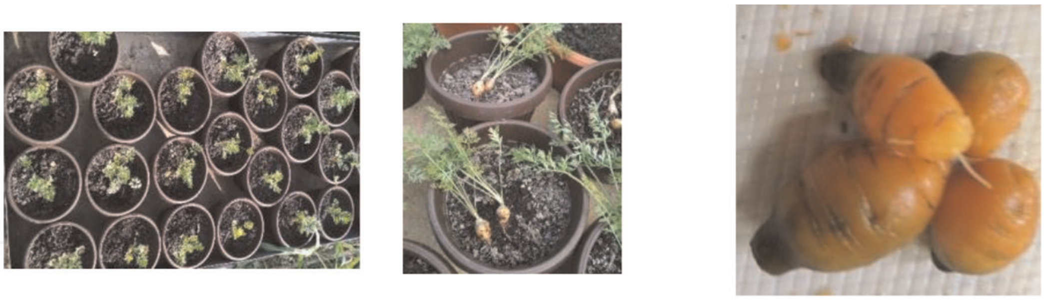
3.1 Study of the transfer coefficient of plants studied

The transfer coefficient reflects the heavy metal uptake ability from the soil by the plant as a function of the heavy metal concentration in the soil.

The definition formula is (Eq. (13)):

$$C_t = \frac{C_{fp}}{C_{is}} \tag{13}$$

For vegetables and fruits, the variation of the transfer coefficient (C_t) depending on the initial concentration of heavy metal in the plants (C_{fp}) and on the initial concentration of heavy metal in the soil (C_{is},) is represented in the following tables (**Table 4** for vegetables and **Table 5** for fruits).



Carrots

Parsley

Cucumbers

Strawberry

Raspberry

Figure 3.
Aspects with the plants during the experimental research.

3.2 Statistical models regarding the phenomenon of accumulation of heavy metals in plants studied

Using the numerical data from the experiments, the statistical mathematical models were obtained by interpolation.

- Interpolation formulas for the amount of heavy metal accumulated at the end of vegetation period

The general form of the interpolation polynomials (degrees one - four) is (Eq. (14)):

$$C_p(C_{is}) = c_0 + c_1 C_{is} + c_2 C_{is}^2 + c_3 C_{is}^3 + c_4 C_{is}^4 \tag{14}$$

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Vegetable	**Concentration of heavy metals, [%]**	**The transfer coefficient (C_t)**		
		Cu	**Pb**	**Zn**
Carrot roots	0	0.433	0.800	0.508
	1.5	0.157	0.169	0.347
	3.0	0.032	0.106	0.137
	4.5	0.020	0.299	0.130
	6.0	0.015	0.154	0.105
Parsley leaves	0	0.257	0.904	0.814
	1.5	0.172	0.155	0.873
	3.0	0.030	0.176	0.346
	4.5	0.020	0.211	0.267
	6.0	0.016	0.158	0.333
Cucumbers	0	0.297	0.404	0.857
	1.5	0.102	0.263	0.222
	3.0	0.032	0.142	0.122
	4.5	0.016	0.081	0.092
	6.0	0.015	0.041	0.101

Table 4.
The variation of the transfer coefficient from vegetables.

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Fruit	**Concentration of heavy metals, [%]**	**The contents of metals, [mg/kg]**		
		Cu	**Pb**	**Zn**
Strawberry	0	0.233	0.400	0.249
	1.5	0.297	—	—
	3.0	0.058	0.047	0.048
	4.5	—	0.060	—
	6.0	—	0.011	0.024
Raspberry	0	0.892	0.919	1.204
	1.5	0.234	—	—
	3.0	0.041	—	—
	4.5	0.038	0.045	0.038
	6.0	—	0.011	—

Table 5.
The variation of the transfer coefficient from fruits.

3.2.1 Transfer coefficient interpolation

The transfer coefficient was determined according to (Eq. (13)). Similar to (Eq. (14)) and the transfer coefficient (Eq. 15))was determined:

$$C_t(C_{is}) = c_0 + c_1 C_{is} + c_2 C_{is}^2 + c_3 C_{is}^3 + c_4 C_{is}^4 \tag{15}$$

Table 6 shows the coefficients for the interpolation of the transfer coefficient Ct.

The experimental data for the transfer coefficient indicated a potential monotonic, asymptotic decrease, the mathematical modeling was done according to formula (16):

$$C_t(C_{is}) = \frac{c_0}{C_{is}} + c_1 \tag{16}$$

The coefficients of the polynomials (Eq. (15)), shown in **Table 7**, and those of the hyperbola (Eq. (16)), shown in **Table 6**.

Vegetable	Metal	Hyperbola coefficients	
		c0	c1
Carrot	Cu	7.55	$9.20\cdot10^{-3}$
	Pb	4.44	0.13
	Zn	15.41	0.14
Parsley leaves	Cu	4.34	0.02
	Pb	5.25	0.11
	Zn	18.87	0.39
Cucumbers	Cu	5.07	0.01
	Pb	2.10	0.10
	Zn	31.55	0.06

Table 6.
The hyperbolic interpolation equations corresponding to the transfer coefficient.

Vegetable	Metal	Coefficients of the interpolation polynomials					Error*,%
		c0	c1	c2	c3	c4	
Carrot	Cu	0.277	$-4.713\cdot10^{-4}$	0	0	0	175.587
		0.362	$-1.67\cdot10^{-3}$	$1.762\cdot10^{-6}$	0	0	122.469
		0.462	$-3.508\cdot10^{-3}$	$8.829\cdot10^{-6}$	$-6.731\cdot10^{-9}$	0	93.579
		0.598	-0.01	$5.342\cdot10^{-5}$	$-1.031\cdot10^{-7}$	$6.608\cdot10^{-11}$	$5.909\cdot10^{-13}$
	Pb	0.458	$-1.402\cdot10^{-3}$	0	0	0	158.923
		0.712	$-7.62\cdot10^{-3}$	$2.01\cdot10^{-5}$	0	0	113.81
		0.976	-0.028	$2.556\cdot10\text{–}^{4}$	$-5.917\cdot10^{-7}$	0	1.025
		0.972	-0.027	$2.419\cdot10^{-4}$	$-4.861\cdot10^{-7}$	$-2.259\cdot10^{-10}$	$3.623\cdot10^{-13}$
	Zn	0.446	$-3.642\cdot10^{-4}$	0	0	0	60.035
		0.545	$-1.094\cdot10^{-3}$	$6.61\cdot10^{-7}$	0	0	16.433

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Vegetable	Metal	Coefficients of the interpolation polynomials					Error*,%
		c0	c1	c2	c3	c4	
		0.571	$-1.472\cdot10^{-3}$	$1.596\cdot10^{-6}$	$-5.799\cdot10^{-10}$	0	10.878
		0.546	$-9.403\cdot10^{-4}$	$-7.101\cdot10^{-7}$	$2.759\cdot10^{-9}$	$-1.534\cdot10^{-12}$	$1.934\cdot10^{-13}$
Parsley leaves	Cu	0.2	$-3.272\cdot10^{-4}$	0	0	0	112.486
		0.252	$-1.06\cdot10^{-3}$	$1.077\cdot10^{-6}$	0	0	47.272
		0.279	$-1.833\cdot10^{-3}$	$4.051\cdot10^{-6}$	$-2.832\cdot10^{-9}$	0	16.952
		0.303	$-2.759\cdot10^{-3}$	$1.014\cdot10^{-5}$	$-1.598\cdot10^{-8}$	$9.02\cdot10^{-12}$	$4.973\cdot10^{-13}$
	Pb	0.5	$-1.654\cdot10^{-3}$	0	0	0	171.259
		0.824	$-9.559\cdot10^{-3}$	$2.555\cdot10^{-5}$	0	0	106.208
		1.07	-0.028	$2.461\cdot10^{-4}$	$-5.541\cdot10^{-7}$	0	31.212
		1.184	-0.046	$6.82\cdot10^{-4}$	$-3.932\cdot10^{-6}$	$7.223\cdot10^{-9}$	$2.63\cdot10^{-12}$
	Zn	0.853	$-5.935\cdot10^{-4}$	0	0	0	49.634
		0.964	$-1.404\cdot10^{-3}$	$7.339\cdot10^{-7}$	0	0	39.635
		0.824	$6.817\cdot10^{-4}$	$-4.426\cdot10^{-6}$	$3.201\cdot10^{-9}$	0	23.812
		0.71	$3.178\cdot10^{-3}$	$-1.526\cdot10^{-5}$	$1.889\cdot10^{-8}$	$-7.205\cdot10^{-12}$	$7.119\cdot10^{-13}$
Cucumbers	Cu	0.19	$-3.136\cdot10^{-4}$	0	0	0	169.64
		0.245	$-1.094\cdot10^{-3}$	$1.148\cdot10^{-6}$	0	0	123.307
		0.286	$-2.288\cdot10^{-3}$	$5.738\cdot10^{-6}$	$-4.372\cdot10^{-19}$	0	99.395
		0.415	$-7.355\cdot10^{-3}$	$3.904\cdot10^{-5}$	$-7.634\cdot10^{-8}$	$4.936\cdot10^{-11}$	$4.653\cdot10^{-12}$
	Pb	0.31	$-1.142\cdot10^{-3}$	0	0	0	89.319
		0.435	$-4.183\cdot10^{-3}$	$9.829\cdot10^{-6}$	0	0	6.995
		0.433	$-4.033\cdot10^{-3}$	$8.062\cdot10^{-6}$	$4.44\cdot10^{-9}$	0	6.853
		0.418	$-1.798\cdot10^{-3}$	$-4.75\cdot10^{-5}$	$4.349\cdot10^{-7}$	$-9.206\cdot10^{-10}$	$7.649\cdot10^{-13}$
	Zn	0.587	$-5.594\cdot10^{-4}$	0	0	0	155.184
		0.831	$-2.355\cdot10^{-3}$	$1.626\cdot10^{-6}$	0	0	91.914
		1.013	$-5.077\cdot10^{-3}$	$8.362\cdot1^{0-6}$	$-4.179\cdot10^{-9}$	0	48.423
		1.136	$-7.765\cdot10^{-3}$	$2.002\cdot10^{-5}$	$-2.107\cdot10^{-8}$	$7.757\cdot10^{-12}$	$1.062\cdot10^{-12}$

Table 7.
The coefficients of the interpolation polynomials.

3.2.2 Study of the variation of the final concentration in fruit depending on the initial concentration of heavy metal injected into the soil

Based on **Table 3** which shows the variations in the content of heavy metals: Cu, Pb, Zn in strawberry and raspberries fruits grown in soil injected with heavy metal solutions separated by different concentrations, results the formula (Eq. (17)) and values from the interpolation equations presented in **Table 8**.

The general form of the interpolation polynomial (grades one - two) is:

$$C_f(C_{is}) = c_0 + c_1 C_{is} + c_2 C_{is}^2 \tag{17}$$

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Fruit	Metal	Coefficients of the interpolation polynomials			
		c0	c1	c2	c3
Strawberries	Cu	4.14	14.02	−3.41	—
	Pb	2.71	−4.97	2.76	−0.32
	Zn	9.94	18.37	−6.05	0.56
Raspberry	Cu	15.7	1.91	−3.06	0.63
	Pb	4.14	14.02	−3.41	—
	Zn	47.9	−21.28	4.11	—

Table 8.
The interpolation equations corresponding to the fruits.

4. Experimental research on the accumulation of heavy metals over time in fresh lettuce

Experimental research focused on growing and monitoring a lettuce crop over a 68-day period, the optimal vegetation period is 45–50 days [50]. The lettuce culture was carried out this time again in a controlled environment, in pots and in the greenhouse. The soil was contaminated with zinc sulfate solution in three concentrations: 1.5%, 3.0% and 4.5%, respectively, prepared solutions similar to those in the previous experiment.

The soil was homogenized with each solution before planting the lettuce seedlings in pots, as shown in the **Figure 4**. One kg of soil was homogenized with 250 ml solution, under the same experimental conditions, without further addition. 11 samples were placed for each concentration of the soil in zinc, harvesting being made at distances of about seven days each.

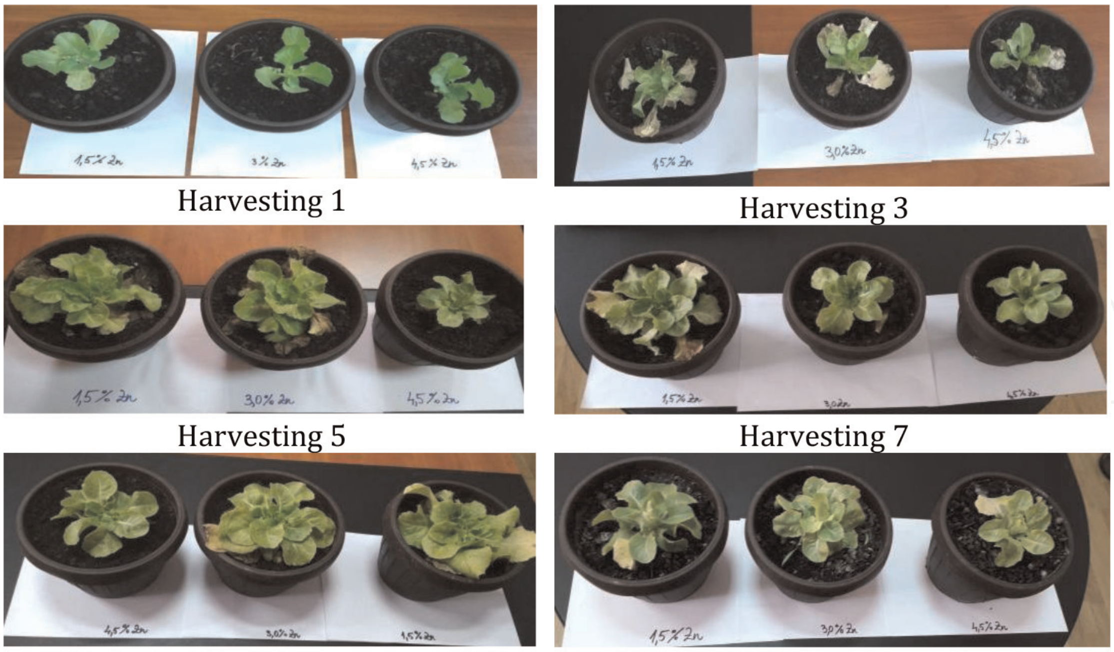
The crop has also developed in the greenhouse, the environmental conditions being favorable for the development of plants and having slight variations.

Sampling of the vegetal samples was done in time up to 68 days after planting (**Figure 5**) and each time the lettuce was harvested, the soil sample was taken from the pot, after it was homogenized.

The determination of zinc from the contaminated soil and from the whole fresh lettuce plant (root and leaves) was performed by the spectrophotometric method (atomic absorption in the flame) [27, 28].

The variation in the zinc content of lettuce is shown graphically in **Figure 6**. It is generally observed that the concentration of zinc in lettuce leaves increases for each of the concentrations of contaminated soil.

Figure 4.
Planting the lettuce seedling.



Harvesting 1

Harvesting 3

Harvesting 5

Harvesting 7

Harvesting 9

Harvesting 10

Figure 5.
Pots with plants from the three types of crops harvested at several.

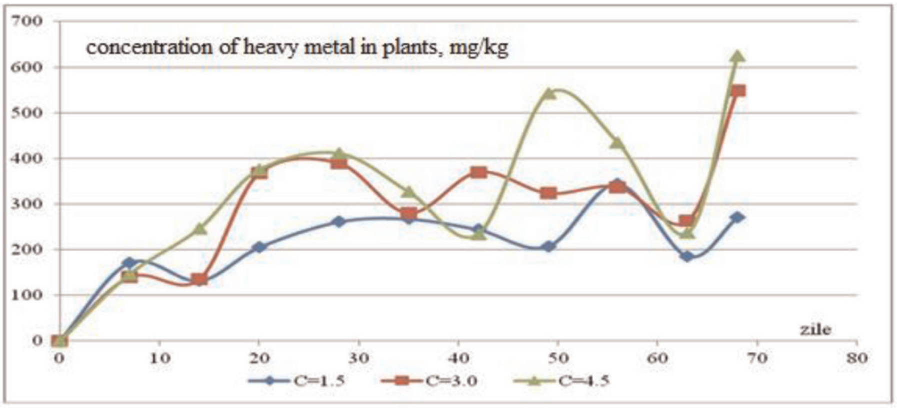


Figure 6.
Variation in time of the concentration of heavy metal (Zn) in lettuce.

At certain time periods, experimental data show a decrease in zinc in plants. If the phenomenon is real, and there was no error in the measurement, it can be concluded that the removal of metal from the plant was natural, in this regard, new experiments are indicated to be able to determine and which factors influenced this process.

If we count the zinc concentrations obtained in plants, we notice that: for soil contaminated with 4.5%, 7 of the 11 crops have the highest concentration of zinc; for soil contaminated with 3.0%, 3 of the 11 crops have the highest concentration of zinc; for soil contaminated with 1.5%, 1 of the 11 crops have the highest concentration of zinc. This observation is also supported by the average lettuce harvest, which recorded 228.9 mg/kg, for the crop developed in soil contaminated with 1.5% Zn, 315.3 mg/kg, for the crop developed in soil contaminated with 3.0% Zn si 357.9 mg/kg, for the crop developed in soil contaminated with 4.5% Zn.

The conclusion is that after the concentration of the soil in zinc the order of accumulation of metal in the lettuce is: 4.5% > 3.0% > 1.5%.

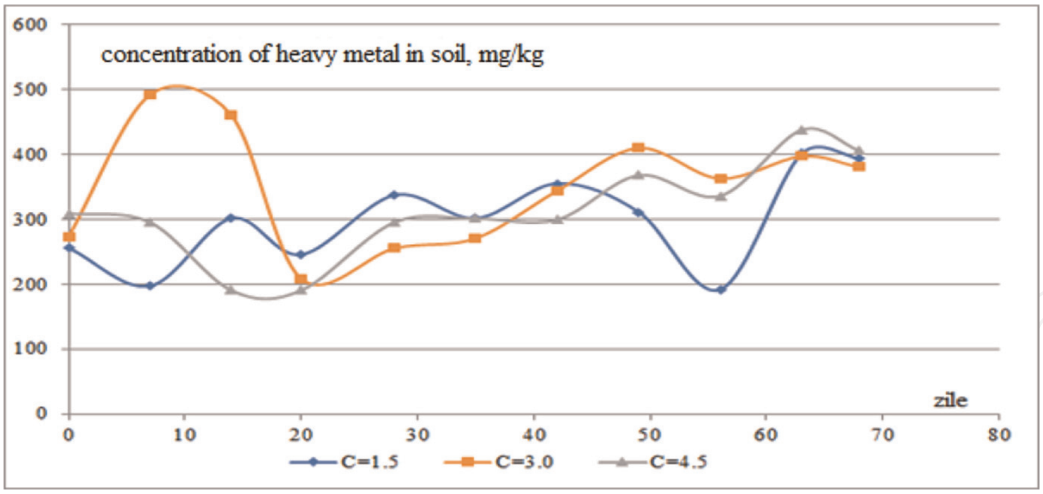


Figure 7.
Variation in time of the concentration of heavy metal (Zn) in soil.

The variation in the zinc content of lettuce is shown graphically in **Figure 7**. It is observed that, over time, the concentration of zinc oscillates, even slightly increasing. Again, it is questioned whether the phenomenon is real or whether measurement errors have occurred. If in the lettuce it can be believed that the metal could not be absorbed by the plant and that the mass has increased, in the case of soil, this conclusion is hard to say, although some soil chemicals can pass into plants as well.

5. Statistical modeling of experimental data on the accumulation of heavy metals over time in fresh lettuce

By interpolating the experimental data, functions were obtained that indicate the variation over time of the zinc concentrations from the soil into the plants. **Figure 8** showed linear interpolation, the results being similar to the increase in the concentration of metals in the soil-plant system. It has been observed that in soils with high concentrations of zinc there has been a large and rapid accumulation.

The interpolated functions obtained can help calculate other important parameters that influence the growth and development of plants on soils contaminated with zinc.

In **Figure 9** graphically represents the variations of Zn concentrations in lettuce grown on the soils of the three infestation categories, in the form interpolated by second-degree polynomials and in the form of experimental data. The same order is

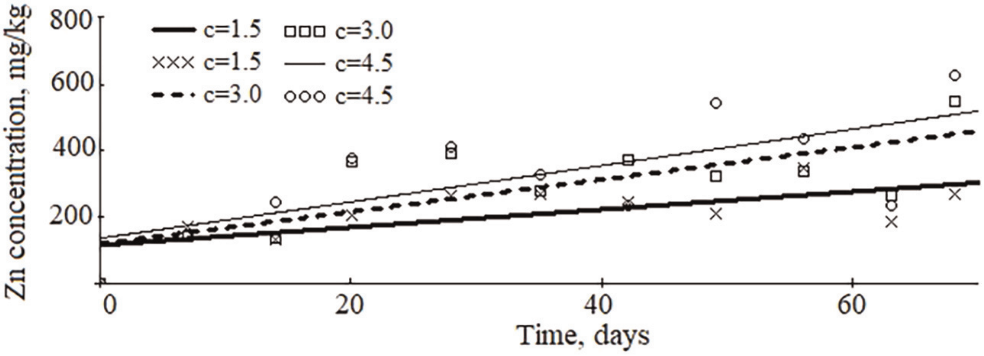


Figure 8.
Linear interpolation of the increase of Zn concentration in fresh lettuce.

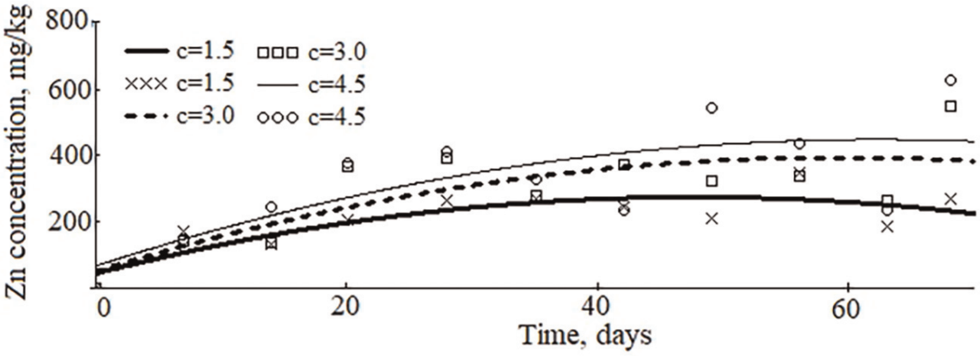


Figure 9.
Quadratic interpolation of the increase in heavy metal concentration in fresh lettuce.

easily visible between the heavy metal concentrations in the plants of the three categories of soil infestation, as in the case of linear interpolation.

From **Figure 9** it is observed that in plants the arrangement of curves of variation of heavy metal concentrations and their increasing monotony is permanent both for the third degree polynomial and for the separation of curves, without any common points between the three curves being observed.

The interpolation curves by third degree polynomials, for increasing the concentration of heavy metals in plants, are represented in **Figure 10** and for fourth degree polynomials, are represented in **Figure 11**.

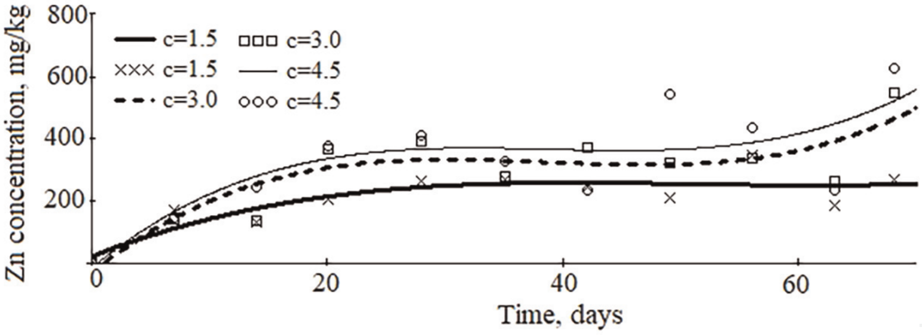


Figure 10.
Cubic interpolation of the increase in the concentration of heavy metal (Zn) in fresh lettuce.

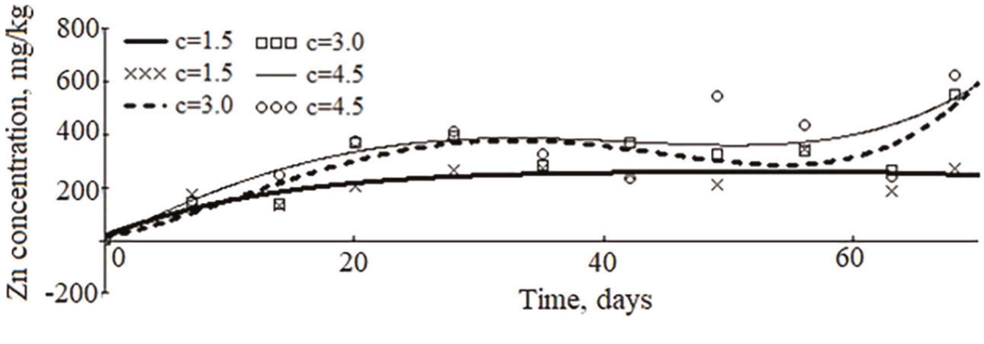


Figure 11.
Polynomial interpolation of the fourth degree for the increase of heavy metal (Zn) concentrations in fresh lettuce crops.

It is observed from **Figures 10** and **11** that, generally, the order and monotony of interpolation curves of the variation in the concentration of heavy metals in plants is observed by polynomial curves, with a few exceptions, the curve corresponding to the soil contaminated by 3% Zn slightly reaches the curve corresponding to the soil contaminated by 4.5% and it is very almost, toward the end of the study period, to the curve corresponding to plants grown in soil contaminated with 1.5% Zn.

6. Conclusions

6.1 Conclusions on the development of vegetables in contaminated soil

- In most cases of vegetables grown in soil contaminated with different concentrations of heavy metal have been observed a tendency to increase the amount of heavy metal accumulated in the plant as the amount of heavy metal in the soil increases.

- On vegetables, in general, the conclusion regarding the increasing monotony representing the variation of the heavy metal concentration in the plant remains valid. It is observed that the variation of the concentration of heavy metal in plants appear more often in the case of contaminations with copper, and less with lead and zinc.

- In the case of the transfer coefficient to vegetables, the trend is decreasing as the amount of heavy metal in the soil increases. The general conclusion is that for the vegetables studied (carrot, parsley and cucumber) bioaccumulation with, Pb and Zn is less as the amount of metals in the soil is higher. Therefore, the more contaminated the soil, the more difficult it is to phytoremediation with these plant species.

6.2 Conclusions on the development of fruits in contaminated soil

- Fruits have a longer vegetation period than vegetables, and experiments to accumulate metals in them are more complicated because, the general distribution of metals requires complex study, possibly over several seasons and on all vegetative plant organs (root, stem, branches, leaves, flowers, fruits and seeds). This way of working requires a long time and a large number of analyzes for a single plant. The analysis in this study focused on the concentration of Cu, Pb and Zn in fruits, ripe, therefore the conclusions for them are mediating.

- For experiments on strawberries with a single heavy metal (Cu, Pb, Zn), it was observed that the variation in the final content in the fruit has a maximum. Therefore, while the concentration in the soil increases, the absorption capacity of metal from the soil into strawberries decreases. It can be concluded that strawberries are developing mechanisms of protection.

- For experiments on raspberries with a single heavy metal (Cu, Pb, Zn), it was observed that the values of the transfer coefficient decrease as the concentration of metals in the soil increases. The order of absorption of metals in raspberry fruits being Zn > Cu > Pb.

- Also, in the case of strawberries and raspberries, the best absorption was observed in the low concentration of 1.5%.

6.3 Conclusions on mathematical modeling of vegetables and fruits

Mathematical modeling was performed based on the interpolation of experimental data obtained in this study.

- Polynomial equations are statistical models with practical applications in both research and agriculture. The formulas obtained can help with various calculations such as:
    - the number of plants corresponding to the implementation of the phytoremediation process of soils contaminated with heavy metals;
    - the compliance of plant production within the limits provided for human consumption;
    - the optimization of phytoremediation and crops with regard to the marketing and consumption.
- In this case, the first and second degree interpolation polynomials were the most used because they showed small variations between the experimental data. The curves obtained for third and fourth degree polynomials showed greater variations between the experimental data and their use may be at risk of showing errors, although it has been found that the fourth degree polynomial passes very close to experimental points when it is restricted to the set of experimental points only.
- Overall, the interpolation curves obtained in this study indicate the increase in the final concentration of Cu, Pb and Zn in the roots of carrot, parsley leaves, cucumbers and strawberry and raspberry fruits and the decrease in the transfer coefficient of the three metals studied at the end of the vegetation periods.
- For the transfer coefficient to vegetables, the experimental data presented in **Table 2** showed a hyperbolic decrease in this coefficient. The hyperbolic interpolation equation (Eq. (16)), proved effective, with few exceptions.

Statistical mathematical models determined with equations (Eqs.(14), (15) and Eq. (17)) whose coefficients are given in the **Tables 7–9**, for each case, they can be used for interpolation in the calculation of concentrations of heavy metals in the plant or calculation of transfer coefficients, only for the experimental range used in this study, for the initial concentration of each heavy metal in the soil.

- Mathematical modeling for fruits is poorly documented because at certain concentrations, plants have not developed. It is recommended for the two species of studied berries (strawberries and raspberries), interpolation curves of maximum second degree, because the experimental distributions are not linear. Interpolation was also attempted using third degree polynomial curves, which passed through experimental points but had a total zero error.

Vegetable	**Metal**	**The coefficients of the interpolation polynomials**					**Error*,%**
		c0	**c1**	**c2**	**c3**	**c4**	
Carrot	Cu	8.128	$3.867\cdot10^{-3}$	0	0	0	15.069
		8.119	$3.985\cdot10^{-3}$	$-1.745\cdot10^{-7}$	0	0	15.068
		8.406	$-4.221\cdot10^{-3}$	$3.138\cdot10^{-5}$	$-3.005\cdot10^{-8}$		14.225
		6.55	0.069	$4.497\cdot10^{-4}$	$1.01\cdot10^{-6}$	$-7.13\cdot10^{-10}$	$3.418\cdot10^{-13}$
	Pb	4.182	0.149	0	0	0	79.321
		−0.143	0.255	$-3.42\cdot10^{-4}$	0	0	74.097
		9.338	−0.465	$8.131\cdot10^{-3}$	$-2.129\cdot10^{-5}$	0	41.472
		−0.224	1.015	−0.029	$2.638\cdot10^{-4}$	$-6.097\cdot10^{-7}$	$2.269\cdot10^{-12}$
	Zn	33.157	0.083	0	0	0	37.801
		23.598	0.153	$-6.353\cdot10^{-5}$	0	0	33.614
		16.381	0.261	$-3.304\cdot10^{-4}$	$1.655\cdot10^{-7}$	0	31.78
		−6.277	0.759	$2.493\cdot10^{-3}$	$3.297\cdot10^{-6}$	$-1.438\cdot10^{-9}$	$1.107\cdot10^{-12}$
Parsley leaves	Cu	6.938	$5.925\cdot10^{-3}$	0	0	0	44.074
		6.603	0.011	$-6.986\cdot10^{-6}$	0	0	43.437
		6.084	0.026	$-6.411\cdot10^{-5}$	$5.441\cdot10^{-8}$	0	42.371
		0.882	0.231	$1.412\cdot10^{-3}$	$2.968\cdot10^{-6}$	$1.998\cdot10^{-9}$	$1.142\cdot10^{-12}$
	Pb	3.636	0.148	0	0	0	27.501
		2.875	0.166	$-6.016\cdot10^{-5}$	0	0	27.014
		6.903	−0.14	$3.54\cdot10^{-3}$	$-9.047\cdot10^{-6}$	0	0.7
		6.747	−0.116	$2.94\cdot10^{-3}$	$-4.396\cdot10^{-6}$	$-9.944\cdot10^{-9}$	$2.548\cdot10^{-13}$
	Zn	65.286	0.241	0	0	0	46.616
		60.405	0.277	$-3.245\cdot10^{-5}$	0	0	46.484
		−15.377	1.409	$-2.834\cdot10^{-3}$	$1.738\cdot10^{-6}$	0	8.212
		−30.093	1.733	$4.238\cdot10^{-3}$	$3.772\cdot10^{-6}$	$-9.342\cdot10^{-10}$	$2.966\cdot10^{-13}$
Cucumbers	Cu	5.647	$6.269\cdot10^{-3}$	0	0	0	19.454
		5.284	0.011	$-7.583\cdot10^{-6}$	0	0	17.047
		4.498	0.034	$-9.401\cdot10^{-5}$	$8.232\cdot10^{-8}$	0	3.285
		4.847	0.02	$3.612\cdot10^{-6}$	$-1.131\cdot10^{-7}$	$1.34\cdot10^{-10}$	$1.053\cdot10^{-13}$
	Pb	7.789	0.018	0	0	0	74.402
		4.457	0.1	$-2.635\cdot10^{-4}$	0	0	58.818
		0.164	0.426	$-4.1\cdot10^{-3}$	$9.641\cdot10^{-6}$	0	9.272
		−0.86	0.584	$8.039\cdot10^{-3}$	$4.016\cdot10^{-5}$	$-6.526\cdot10^{-8}$	$5.97\cdot10^{-13}$
	Zn	30.884	0.066	0	0	0	14.042
		33.759	0.045	$1.911\cdot10^{-5}$	0	0	12.642
		28.007	0.13	$-1.935\cdot10^{-4}$	$1.319\cdot10^{-7}$	0	7.389
		32.489	0.032	$2.342\cdot10^{-4}$	$-4.875\cdot10^{-7}$	$2.845\cdot10^{-10}$	$4.609\cdot10^{-13}$

Table 9.
The interpolation equations corresponding to the vegetables.

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- It was observed that the variation of the heavy metal content in berries (fruit) does not recommend linear interpolation (linear regression), as these distributions have potential extreme points (minimum or maximum).

6.4 Conclusions on the development over time of fresh lettuce on soils with different concentrations of zinc

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- The overall conclusion is accumulate zinc in the lettuce regardless of Zn concentration in the soil.
- At certain intervals, the concentration of zinc decreases in the lettuce.
- This phenomenon show that the removal of zinc from the lettuce can be done naturally, while the factors that influence this process remain to be studied.
- The accumulation of Zn in lettuce increases as the concentration of Zn in the soil increases (4.5% > 3.0% > 1.5%).
- At the higher concentration of zinc (4.5%) for contaminated soil, the accumulation of metal in lettuce is higher, in the chosen range of these experiments.

6.5 Conclusions on mathematical modeling for the development of fresh lettuce over time on soils with different concentrations of zinc

The mathematical modeling presented in this chapter was done through a mediation of experimental data.

Increased polynomic interpolation resulted in a good approximation of experimental data, but there were not enough reasons from the point of view of the phenomenon studied to get over the interpolation of the third degree polynomial. The functions determinated are particular and useful only in the experimental field chosen in this study.

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Therefore, the statistical model can be used to validate the theoretical model, which is general both in terms of working conditions (soil type, metal type, metal concentrations, atmospheric conditions) and the plant species used (root species, leaf species, fruit species).

The results obtained from experiments and interpolation suggest indications for future research: New experiments on several plant vegetation cycles, possibly with modification of working parameters that can influence the bioaccumulation process of heavy metals in plants.

6.6 General conclusions

At the optimum vegetation period (final harvest, in the stage of ripening and human consumption), the plants (carrots, parsley, cucumbers, strawberries, Raspberries) have continuously accumulated (monotone growth) heavy metals (Cu, Pb, Zn) from contaminated soil.

At the end of the growing season, the transfer coefficient decreases as the concentration of heavy metals (Cu, Pb, Zn) in the soil increases.

The data presented in this chapter can be both the foundation of dynamic mathematical models that can simulate the life of a plant and the starting point for the development of future research, that can emphasize self-defense mechanisms developed by plants, adaptations to new environmental conditions, possibilities for restoring the qualities of some plants over time.

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Conflict of interest

The authors declare no conflict of interest.

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