International Journal of Engineering & Technology, 7 (4.38) (2018) 755-759

International Journal of Engineering & Technology

Website: www.sciencepubco.com/index.php/IJET

Research paper



The Involvement of Heavy Metals in the Main Trophic Chains in Agrophytocenoses of the Sterlitamak Region of the Republic of Bashkortostan

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Abstract

The work was aimed at evaluating the involvement of heavy metals in the biogeochemical cycle in the soil-plant system in the setting of agrophytocenoses. The elemental composition of soil and plants was determined by atomic absorption. The sanitary-hygienic and ecological assessment of the condition of soils and plants in the region by heavy metals content has been provided for the first time; for individual items, the health risk of soil contamination and used agricultural products has been assessed.

For the first time, background concentrations of priority pollutants in the soil and plants have been found at different sites from contamination sources; the annual average volumes of removal of Cd, Pb, Zn, Cu, Ni, Cr, and Mn from the soil have been established; the antagonistic-synergistic interactions have been revealed at the introduction of macro- and microelements into plants.

Keywords: *Anthropogenic Load, Xenobiotics, Heavy Metals, Antagonism and Synergism Of Ions, Risk Assessment.*

1. Introduction

It has been revealed that the inadequate to normal intake of trace elements in the body, and, above all, the excess intake of xenobiotics, the abnormal relationship between chemical elements in the diet cause irreversible physiological changes in the body and even its death [1-4]. The issue of heavy metals' accumulation in plants that serve or can serve as food for animals and humans is of particular interest [5].

In the experimental conditions, the antagonistic and synergistic, as well as additive interaction of chemical elements upon their intake into plants has been found. These phenomena are insufficiently studied. However, in the natural conditions the soil always contains not one but several "heavy" elements, which influence plants and soil biota in the complex. It has been established that the plants themselves actively regulate the absorption of heavy metals and their distribution throughout the plant organs. The more toxic the element is and the higher its concentration in the soil is, the lower the regulatory capacity of plants is. Currently, the relevance of research on the chemical composition of plants is dictated by the increasing anthropogenic load on all components of the environment, resulting in the disruption of the chemical elements' circulation in the biosphere. The scale of contamination of soil, crop and livestock products with heavy metals necessitates theoretical studies on the production of agricultural products standardized by certain chemical elements. First of all, it affects such biophilic elements as N, Mg, P, Ca, Cr, Mn, Fe, Cu, Zn, and xenobiotics - Ni, Cd, Pb [6].

The Republic of Bashkortostan has incomplete information on the ratios of the elements consumed by the main agricultural plants. The Sterlitamak district of the Republic, in which agriculture is developed, along with major industrial production represented by the chemical, petrochemical and energy enterprises, is characterized by an increased technogenic load on the environment. The local soil contamination by heavy metals such as copper, zinc, cadmium, lead, nickel, chromium, manganese and iron has been found there. Therefore, research is needed to examine the ecological and physiological characteristics of different plant species, allowing to take into account their ability to accumulate heavy metals.

The purpose of the study was to explore the limits of variation, migration and accumulation of heavy metals in the soil-plant systems of agrocenoses of the Sterlitamak region of the Republic of Bashkortostan.

2. Methods

The main results have been obtained on the basis of the monitoring studies of soils and plants in the period from 2015 to 2017.

Local soil-ecological monitoring was carried out on 14 reference plots laid in the Sterlitamak district at different distances from the Sterlitamak industrial zone (Table 1).

Table 1: Topographic survey of the sites under study

Site	Geographical coordinates		Nearest inhabited locality
	Latitude	Longitude	
Background	53.8605	55.4072	Pervomayskoye village
1.	53.7614	55.9512	Roshchinskiy village
2.	53.8448	55.9122	Bolshoe Aksakovo village
3.	53.7454	56.0404	Noviy Krasnoyar village
4.	53.8326	56.1636	Bolshoy Kuganak village
5.	53.7239	56.1146	Belskoye village
6.	53.7225	56.1618	Zabelskoye village
7.	53.5153	55.9269	Naumovka village
8.	53.4449	55.9174	Kantyukovka village

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9	53.4033	55.7548	Maksyutovo village
10.	53.4662	55.6067	Tyuryushlya village
11.	53.6626	55.5466	Buguruslanovka village
12.	53.5973	55.4640	Maksimovka village
13.	53.7806	55.7246	Sadovka village
14.	53.8833	55.5553	Severnoye section village

The soil cover in the study area is represented by leached heavy loamy chernozem, characterized by subacid and neutral reaction of the medium.

The soil contamination degree in the region was assessed by comparing them with background values, which were the sites with similar soils with the lowest content of the elements under study. The soils' availability and the levels of their contamination with heavy metals were assessed in accordance with the scale developed based on years of research [7].

The soil was sampled as per GOST 17.4.4.02 – 84. In the selected samples, humus, pH, the content of the Ca and Mg exchange forms, the content of P2O5 and K2O, the content of gross and mobile forms of heavy metals such as Cu, Zn, Cd, Pb, Ni, Cr, Mn and Fe, were determined.

The content of trace elements in soil and plant samples was determined using a C–115–M1 atomic absorption spectrophotometer.

For the sites under study, the total soil pollution indicators (Zc) was:

$$Zc = \sum_{i=1}^{n} Ki - (n-1), \quad (1)$$

where n was the number of elements to be determined; and Ki was the concentration ratio – the ratio of the content of this element in the soil to its background content. For an integrated assessment of the toxic load on the soil, the pollution gradient indicator was used

$S_n = 1/n * \sum C_i / C_f$, RU, (2)

where Ci was the concentration of only those elements, the content of which in the soil exceeded the background one; C_f was the concentration of chemical elements in the background area; and n was the number of chemical elements included in the analysis.

3. Results

The specifics of the location and specialization of industrial zones in the city of Sterlitamak, the uneven traffic load, the preferential direction of the winds have determined the nature of the pollutants' distribution in the region.

The industrial enterprises of Sterlitamak are concentrated in its northern and northeastern parts. These territories are more susceptible to contamination by heavy metals, in particular, Cu, Zn, Cd, Pb and Ni due to the prevailing southern circulation of atmospheric air.

Analysis of the total amounts of pollutants from all sources suggests a significant role of the Sterlitamak district in the environmental pollution of the Republic of Bashkortostan.

In the city of Sterlitamak, the density of pollutant emissions per 1 ha is the highest one among the cities of the Republic, and averages 7.1 tons per year, 110 times exceeding the average density in the region (Figure 1). At the same time, according to the flow of pollutants per 1 resident, Sterlitamak ranks 5th in the Republic.

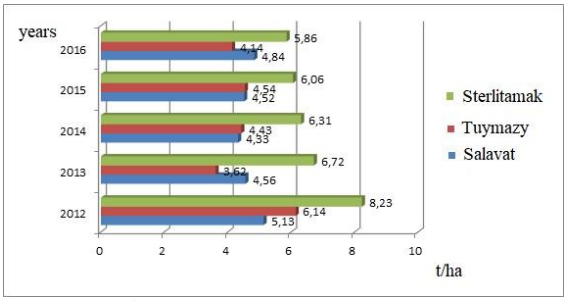


Fig. 1: Dynamics of the pollutants' emission into the atmospheric air in the cities of the Republic of Bashkortostan with the highest amount of emissions, t/ha

Soil pollution with heavy metals takes place at a local level and covers mainly the areas located near industrial enterprises, thereby causing an unsatisfactory environmental situation.

For the ecological and sanitary-hygienic assessment of soil pollution, background and approximate permissible concentrations (APC) of the studied elements were used. The highest total pollution coefficient (9.32) was recorded in the South-Western part of the Sterlitamak district. In area 9, background values for cadmium were exceeded by 4.1 times; for lead – by 3.8 times; for zinc and nickel –by 2.1 and 2.0 times, respectively, and for copper – by 1.4 times. During the air masses' transfer, this area receives from the adjacent territories an additional amount of pollutants resulting from the activities of the industrial enterprises located in the nearby Salavat and Ishimbay. At the same time, the gross content of heavy metals in all soil samples is within the APC limits (Table 2, Figure 2).

The content analysis of mobile and gross forms of heavy metals at the above sites showed the ambiguity of their distribution in the soil (Table 3).

Based on the concentration of gross forms of heavy metals in the soil, at site 1, the largest exceedances of background values for Cu (by 1.6 times) and Cr (by 1.9 times) were noted, at site 9 – for Ni (by 2 times), at site 11 – for Pb, Cd and Zn (by 3.8; 4.1 and 2 times, respectively). The third site was characterized by the highest total pollution ratio in gross forms. At the same time, there were no exceedances of the maximum permissible concentration limits (MPC) for any of the elements.

The content of mobile forms of heavy metals indicates a significant excess of their concentrations compared to background values: Cd – by 3.9-7.6 times; Pb – by 2.3-2.8 times; Zn – by 3.5-4.5 times; Cu – by 9.6-16.6 times; Ni – by 1.1-1.9 times; and Cr – by 1.1-1.8 times.

The largest share of mobile forms to the gross ones was noted for cadmium and manganese - up to 38 and 27%, respectively.

Based on the pollution gradient, 7 areas characterized by different toxic load, namely 1.01 (14); 1.26 (13); 1.50 (5); 1.88 (3); 2.15 (8); 2.25 (1); 2.29 (Table 3), were identified.

The analysis of the spring wheat samples grown in these sites showed a very strong relationship (r = 0.98) between the gradient of soil pollution with toxicants and their content in the grain.

Table 2: Excess background concentrations of heavy metals and APC values in the soil of the studied sites

No.	Cu	Zn	Cd	Pb	Ni	Total pollution ratio
1	1.55 0.23	1.75 0.33	3.86 0.14	2.15 0.10	1.93 0.10	7.24
2	0.86 0.13	1.44 0.27	3.71 0.13	2.01 0.10	1.91 0.10	5.95
3	1.45 0.21	1.63 0.31	2.14 0.08	2.63 0.13	1.55 0.13	5.40
4	1.05 0.15	1.78 0.34	3.29 0.12	3.31 0.16	2.60 0.16	8.04
5	1.18 0.17	1.08 0.21	2.86 0.10	2.39 0.12	0.62 0.12	4.14
6	1.04 0.15	1.85 0.35	2.43 0.09	2.05 0.10	2.01 0.10	5.38
7	1.23	1.74	3.86	2.13	1.93	6.90

	0.18	0.33	0.14	0.10	0.10	
8	1.26 0.18	2.30 0.44	3.43 0.12	1.86 0.09	1.89 0.09	6.73
9	1.37 0.20	2.06 0.39	4.14 0.15	3.75 0.18	1.99 0.18	9.32
10	1.33 0.19	2.04 0.39	2.71 0.10	1.78 0.09	1.66 0.09	5.53
11	1.24 0.18	1.99 0.38	3.86 0.14	2.32 0.11	2.03 0.11	7.44
12	0.74 0.11	1.41 0.27	2.86 0.10	2.11 0.10	0.92 0.10	4.04
13	0.87 0.13	1.08 0.21	2.00 0.07	1.78 0.09	1.42 0.09	3.14
14	0.86 0.13	1.07 0.20	1.71 0.06	1.98 0.10	1.30 0.10	2.92

*in the numerator - the excess of background values; in the denominator - the excess of the APC

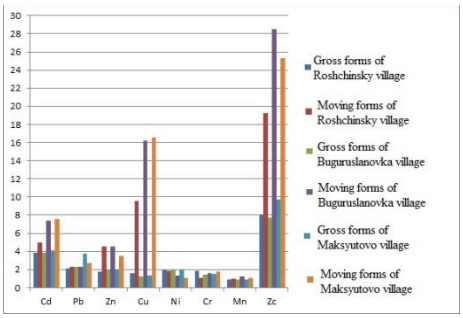


Fig. 2: The ratios of total soil pollution of sites

Table 3: The average content of heavy metals in the soil of the sites remote from pollution sources, mg/kg

Metal	1	9	11	APC MPC	Background content
Cu	29.74±1.49 0.27±0.02	23.78±1.19 0.27±0.02	26.32±1.32 0.21±0.01	1323	19.20±0.96 0.06±0.003
Zn	73.36±4.40 4.03±0.24	83.25±4.99 6.80±0.47	86.39±5.18 6.47±0.39	22023	41.86±2.51 0.42±0.03
Cd	0.27±0.01 0.09±0.004	0.27±0.01 0.09±0.004	0.29±0.01 0.11±0.004	2 -	0.07±0.003 0.04±0.002
Pb	13.55±0.81 0.80±0.05	14.59±0.88 1.18±0.07	23.56±1.41 1.21±0.07	130 -	6.29±0.38 0.16±0.01
Ni	69.19±4.84 1.33±0.09	72.84±5.10 0.97±0.07	71.47±5.00 0.74±0.05	804	35.86±2.51 0.71±0.05
Cr	151.50±4.55 0.56±0.02	114.18±3.43 0.71±0.02	123.23±3.70 0.60±0.02	- 6	79.95±2.40 0.58±0.17
Mn	531.16±5.31 78.76±2.36	474.67±4.74 114.03±3.42	475.14±4.75 126.79±3.80	- -	544.50±5.44 70.53±2.12
Fe	31796±3334 8.05±0.63	26831±285 1.88±0.23	26177±218 2.78±0.23	- -	24339±232 1.93±0.24

*1. In the numerator - the gross form of heavy metals, in the denominator - the mobile form of heavy metals; 2 "-" – the standards are absent.

With an increase in the pollution gradient by 2.3 times, the total content of the studied elements in the grain increased by 2.1 times (Table 4).

The content of the studied elements in the grain varied greatly: for Ni – by 1.8 times; for Zn – by 1.9 times; for Mn and Fe – by 3.4 times; for Cu – by 4.1 times; for Cd – by 11.5 times; and for Pb – by 12.0 times.

The removal of heavy metals by the spring wheat grain also varied greatly: for Cu – by 3.3 times; for Zn – by 1.6 times; for Cd – by 8.5 times; for Pb – by 10.9 times; for Ni – by 1.5 times; for Mn and Fe – by 3.1 times (Table 5).

Table 4: Changes in the chemical composition of spring wheat grain depending on the toxic load on the soil

Site	Level of toxic load	Cu	Zn	Cd	Pb	Ni
14	1.01	1.32 0.1	21.2 0.4	0.02 0.2	0.08 0.2	0.09 0.11
13	1.26	1.56 0.2	29.9 0.6	0.03 0.3	0.03 0.06	0.10 0.12
5	1.50	1.79 0.2	31.1 0.6	0.05 0.5	0.13 0.3	0.08 0.11
3	1.88	5.03 0.5	38.2 0.8	0.04 0.4	0.21 0.3	0.11 0.11
8	2.15	4.01 0.4	41.3 0.8	0.05 0.5	0.03 0.06	0.12 0.14
1	2.25	5.43 0.5	41.2 0.8	0.20 2.3	0.36 0.7	0.12 0.13
11	2.29	4.69 0.4	42.5 0.8	0.23 2.3	0.25 0.2	0.14 0.16

*the numerator indicates the content, mg/kg, the denominator - exceedance of the maximum permissible concentration

Table 5: Removal of heavy metals with the spring wheat grain, g/ha

Experiment Option	Cu	Zn	Cd	Pb	Ni	Mn	Fe
N	2.805	45.05	0.043	0.170	0.191	29.33	195.5
M	3.036	58.19	0.058	0.058	0.195	91.29	195.6
E	3.405	59.13	0.095	0.247	0.152	74.18	359.5
C	9.236	64.98	0.068	0.221	0.187	71.34	134.6
H	7.034	72.44	0.088	0.053	0.210	64.42	131.6
A	8.063	66.04	0.369	0.577	0.192	70.59	408.1
K	5.860	62.73	0.365	0.191	0.222	52.45	253.9

The ability to absorb, accumulate and use chemical elements in plants is genetically determined. However, within the same genotypic form, the nature of their absorption may change under the influence of external conditions, leading to the need to study the antagonism and synergism of ions as they enter the plants [8-9].

Antagonistic and synergistic interactions can be associated with the ability of one element to inhibit or stimulate the absorption of other elements [10].

These reactions can occur both inside the cells and on the surface of the membranes, and in the environment surrounding the roots of plants. The interaction processes are controlled by many factors, and their mechanisms are still poorly understood [7, 11, 12].

To study the mutual influence of chemical elements, including macroelements, when entering the plants, data on their content, obtained from pollution sources at different sites, were mathematically processed using the correlation analysis [13].

The correlation analysis revealed a strong inverse relationship between the content of macro- and microelements in the spring wheat grain for such pairs as Cu-N, Mn-P, Zn-N, Ni-K, Zn-K, Cu-K (Figure 3).

	N	P	K	Ca	Mg	Cu	Zn	Cd	Pb	Ni	Mn	Fe
N	1											
P	0,77	1										
K	0,68	0,25	1									
Ca	0,23	0,37	0,21	1								
Mg	0,33	0,16	0,08	0,01	1							
Cu	-0,82	-0,36	-0,92	-0,16	-0,12	1						
Zn	-0,86	-0,66	-0,89	-0,33	-0,11	0,86	1					
Cd	-0,59	-0,27	-0,59	0,26	-0,39	0,48	0,59	1				
Pb	-0,55	-0,18	-0,51	0,47	-0,45	0,48	0,48	0,96	1			
Ni	-0,47	-0,12	-0,88	-0,09	0,05	0,66	0,76	0,73	0,60	1		
Mn	-0,68	-0,85	-0,27	-0,22	0,31	0,42	0,60	0,19	0,11	0,18	1	
Fe	-0,59	-0,50	-0,08	0,27	-0,64	0,18	0,28	0,73	0,77	0,11	0,29	1

Fig. 3: Correlation matrix of the relationship between macro- and microelements in the spring wheat grain.

From the entire data array, strong feedback was obtained for six pairs of elements: Cu-N, Mn-P, Zn-N, Ni-K, Zn-K, and Cu–K. A strong direct relationship (0.73>r>0.96) was typical for such pairs of elements as Ni-Cd, Fe-Cd, Ni-Zn, Fe-Pb, P-N, Zn-Cu, as well as Pb and Cd.

As a result of bioaccumulation, the concentration of heavy metals in a given agricultural product can reach values that represent a real danger to the human body [6, 14].

Thus, the assessment of the trace element composition of vegetable products in the first of the test sites showed that even in the conditions of acceptable content of heavy metals in the soil in all types of vegetables there was an excess of Pb in potatoes, cabbage and onions; the content of cadmium in the onion was at the MPC level (Table 6).

In this regard, the potential risk associated with human consumption of vegetables grown under man-made loads was estimated.

Table 6: The content of heavy metals in vegetable products, mg/kg

Plants type	Cd	Pb	Cr
Potato	0.02 ± 0.001	0.06 ± 0.008	0.35 ± 0.041
Carrot	0.01 ± 0.001	0.08 ± 0.005	0.19 ± 0.08
Beet	0.02 ± 0.001	0.09 ± 0.003	0.18 ± 0.003
Radish	0.02 ± 0.002	0.15 ± 0.01	0.11 ± 0.007
Cabbage	0.01 ± 0.001	0.14 ± 0.09	0.22 ± 0.01
Onion	0.03 ± 0.002	0.86 ± 0.06	0.29 ± 0.031
MPC	0.03	0.5	0.2

In accordance with the R 2.1.10.1920-04 guidelines, a risk was assessed for the health of the population in the area under study when consuming vegetables contaminated with cadmium, lead and chromium, selected in the first site [11, 12, 15, 16].

In the case of oral exposure to chemical carcinogens coming from food, the SF value is set for oral (SF_o) intake (Table 7).

Table 7: Indicators of a carcinogenic hazard of heavy metals

Pollutant	Classification of carcinogenicity		Reference Dose, RfD, mg/kg	Carcinogenic potential SF_o
	IARC	EPA		
Cadmium	1	B1	0.0005	0.38
Lead	2A	B2	0.0035	0.047
Chromium	3	A	0.05	0.42

To assess the exposure, the average daily doses (Figure 4), as well as the magnitude of the individual carcinogenic risk when eating vegetables grown amidst anthropogenic stress (Table 7) were calculated.

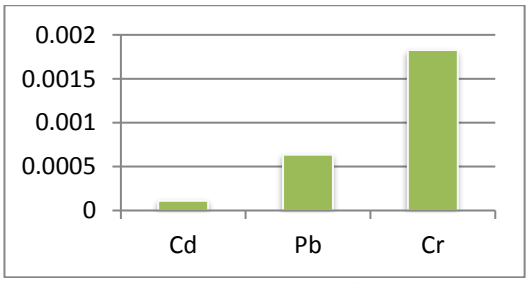


Fig. 4: The average daily dose for life, mg/(kg×day)

The magnitude of the individual carcinogenic risk in the use of the vegetables grown under the anthropogenic stress was calculated (Table 8).

The individual carcinogenic risk in case of oral administration with food products of such heavy metals as cadmium and lead corresponds to the maximum permissible risk.

Table 8: Individual risk to the population due to contamination of food with heavy metals

Metals	Cd	Pb	Cr
Individual risk	4.18E-05	2.99E-05	7.67E-04
Range	2	2	3

The assessment of individual carcinogenic risk to the population due to the intake of chromium from vegetables indicates an unacceptable level of danger for the population as a whole (risk level is more than $1 \cdot 10^{-4}$, but less than $1 \cdot 10^{-3}$) and requires the development and implementation of planned health measures based on the results of an in-depth assessment of various aspects of the existing problems, as well as the determination of their priority over other hygienic, environmental, social and economic problems in the area being assessed [12].

The accumulation of heavy metals in vegetables is currently an urgent problem of agricultural production, especially if the agricultural land is under the man-made load. The accumulation of heavy metals in plants often does not lead to any change in their presentation, which makes it particularly important to have high-quality sanitary and hygienic control of vegetables coming to consumers [17, 18].

Measures aimed at reducing the risk of consuming vegetables contaminated with heavy metals should be implemented both at the cultivation of agricultural products and at the stage of their use by consumers.

4. Conclusion

The analysis of migration and accumulation of heavy metals in the soil- plant system has allowed establishing the following patterns:

The sanitary-hygienic and environmental assessment of the arable soil of the agrocenoses in the Sterlitamak region of the Republic of Bashkortostan indicates the absence of the exceeded MPC of heavy metals and the acceptable levels of soil contamination, but there is a tendency for accumulation of elements such as Cu, Zn, Cd and Ni in the upper soil horizons.

The majority of the studied agricultural plants have accumulated a significant amount of heavy metals in the productive part without signs of the metabolic process disruption and lower yields.

The correlation analysis has allowed to establish a strong feedback between the content of macro- and microelements in the spring wheat grain for such pairs as Cu-N, Mn-P, Zn-N, Ni-K, Zn-K, Cu-K, and a strong direct connection was characteristic for Ni-Cd, Fe-Cd, Ni-Zn, Fe-Pb, P-N, Zn-Cu, and Pb-Cd.

The assessment of the risk of soil and plants' contamination with heavy metals highlighted the need for continuous monitoring of Cr, Cd and Pb contents in them.

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