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Assessing the Geo-Environmental Risks of Technogenic Pollution of Agricultural Soils

Astghik Sukiasyan ^{a, *}, Arsen Simonyan ^a, Samvel Kroyan ^b, Alik Hovhannisyan ^a,
Vahram Vardanyan ^c, Alla Okolelova ^d, Armen Kirakosyan ^a

^aNational Polytechnic University of Armenia, Yerevan, Armenia

^bNational University of Architecture and Construction of Armenia, Yerevan, Armenia

^cYerevan State University, Yerevan, Armenia

^dVolgograd State Technical University, Volgograd, Russian Federation

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Abstract

Environmental problems are the result of the disturbance of the natural balance through a combination of processes in the environment and their irreversible consequences. The latter causes specific environmental problems. This is particularly true for countries with limited land, water and plant resources. Factors such as soil fertility and protection against adverse effects, inactivating and demobilising chemical pollutants that penetrate the soil, etc. must be managed to ensure a stable ecosystem. Climatic factors (ambient temperature, rainfall, wind rose, etc.) also contribute to wasteful human activities. All these factors lead to soil degradation and desertification in the territory of the Republic. There are different forms of these phenomena. The extent and irreversible consequences of heavy metal contamination of biota are well documented. However, all discussions are reduced to quantitative comparisons of heavy metal concentrations in the environment with accepted maximum permissible concentrations, without focusing on the nature of what is happening. In this article, approaches with consideration of geoecological coefficients for assessment of multi-component impact of anthropogenic pollution on arable soils of the Republic of Armenia are considered. The anthropogenic zone of the city of Hrazdan and the surrounding arable soils are considered as an example of the approach to the estimation of the degree of environmental pollution. It contributes to the differentiation of abiotic and anthropogenic factors of heavy metal pollution in environmental studies. It helps to differentiate between the abiotic and the anthropogenic sources of heavy metals in studying them. This approach makes it possible to set limits on the use of natural resources. It also contributes to the development of environmental protection measures aimed at the safety of the biota as a whole.

Keywords: heavy metals, pollution of soils, wind rose, Clark's coefficient, geoecological coefficients, monitoring.

* Corresponding author

E-mail addresses: sukiasyan.astghik@gmail.com (A. Sukiasyan)

1. Introduction

The combination of processes taking place in the environment and their irreversible consequences create environmental problems. They disrupt the ecological balance of the region (Third National Communication..., 2015). This leads to a number of environmental problems, especially in countries such as the Republic of Armenia, where the resources of land, water and plants are limited (Kroyan, 2019). Soil fertility and protection from adverse impacts, inactivating and demobilising penetrating chemical pollutants are important factors in ecosystem stability. All this is due to the complex structure and a variety of biochemical processes in the soil (Figure 1). It is known that in terms of origin, composition, structure and agronomic properties, the geochemical processes taking place under the conditions of vertical zoning of soils in Armenia have formed completely different genetic types of soils that alternate with each other from lowlands to high mountain peaks (Sukiasyan, 2018). The heterogeneous geological structure of the territory of the Republic of Armenia and the diverse nature of rock-forming excavations cause irreversible changes in the topography. As a result of these actions, there is contamination of all the components of the biota (Sukiasyan, Pirumyan, 2018).

On the other hand, the various manifestations of soil degradation (desertification, alkalisation, etc.) and vegetation cover (Mkrtchyan, Petrosyan, 2014; <https://unfccc.int>) are caused by climatic changes together with wasteful human economic activities. In this regard, geo-ecological research, assessment and monitoring of the multifactorial effects of anthropogenic soil pollution of territories is necessary (Armenia. Climate..., 1999) due to the impact of uncontrollable anthropogenic factors on the ecosystem of the region.

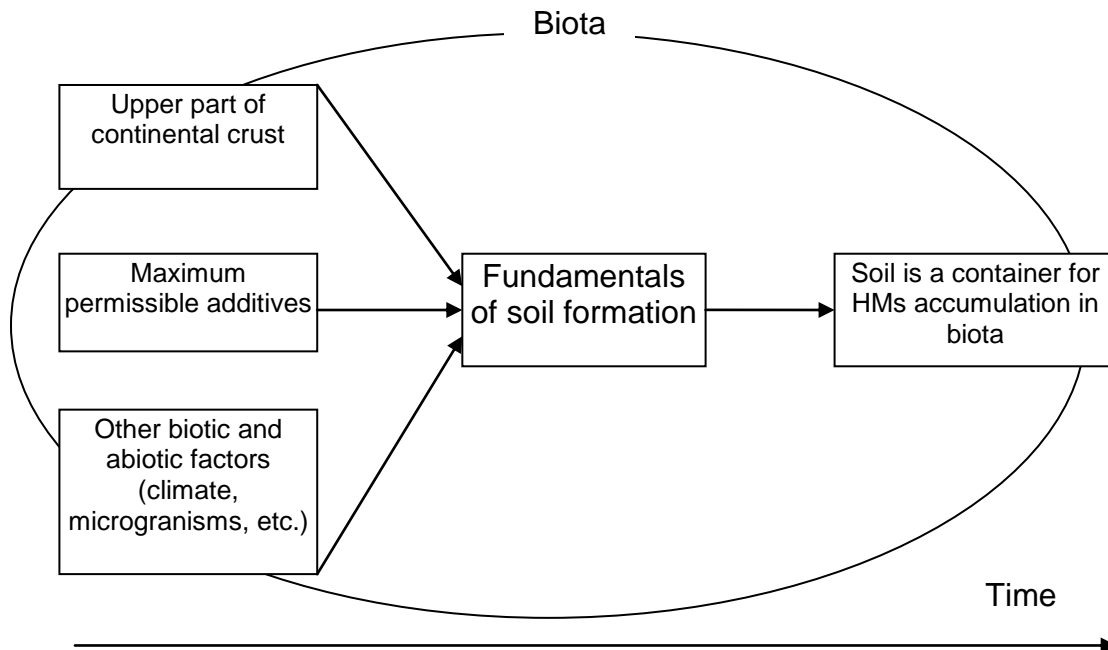


Fig. 1. Multi-component soil formation

One of the factors contributing to land degradation in Armenia is land contamination by mining industry, thermal power plants, chemical fertilizers for agriculture, transportation, etc. (Tepanosyan et al., 2018; Hunanyan, 2012; Danielyan, Khoetsyan, 2009). A combination of different monitoring approaches is necessary for the assessment of pollution of environmental components: biological condition of vegetation, soil water regime, pH response, nutrient inputs, organic carbon and other agrochemical indicators (Darzi-Naftchali, Ritzema, 2018; Shokri et al., 2022; Tchounwou et al., 2012; Ali et al., 2021).

In this article, the approaches to assess the multi-component impact of anthropogenic pollution on the arable soils of the Republic of Armenia using geo-ecological coefficients including Clark's coefficient are discussed. As an example of the approach to the estimation of the degree of environmental pollution including the direction of the wind rose, the anthropogenic zone of the city of Hrazdan, and the surrounding arable soils are considered.

2. Materials and method

The subject of the study was soil sampling from experimental sites near at the following locations: Odzun – 41°03'06"N, 44°36'55"E, Shnogh – 41°08'52"N, 44°50'16"E, Teghut – 41°07'05"N, 44°50'45"E), Hushakert – 40°04'52"N, 43°55'35"E, Hrazdan – 40°30'30"N, 44°50'45"E), Hushakert – 40°04'52"N, 43°55'35"E, Hrazdan – 40°30'0"N, 44°46'0"E, Gavar – 40°21'32"N 45°07'36"E, Martuni – 40°08'24"N 45°18'23"E. Soil samples under dry weather conditions were taken by the envelope method from the depth of growth of the root system of the plant under study. Non-metallic instruments were used for point sampling. At least five incremental samples taken from the same sampling site were mixed to form a pooled soil sample. Sample analysis was performed using a Thermo Scientific™Niton™XRF Portable Analyzer (USA) by directing X-rays at the sample for a total of up to 210 seconds. Changes in the concentration of some of the HMs have been determined in soil samples taken from different regions of the country. The chemical composition of the geological structure of the rocks in our experimental sites is complex. Therefore, it is reasonable to consider Clark's coefficient (K_k) of chemical elements as a reference (Vodiansky, 2012; Müller, 1981). Then, based on the results, some geochemical coefficients for assessing the degree of soil contamination were calculated (Sukiasyan, 2018). All experiments had up to 5 technical replicates. The data were statistically processed. The Student's t-criterion was used to process the results. The observed differences are statistically significant. The calculated values of the criterion were greater than the critical value at a significance level of $p < 0.05$.

3. Results and discussion

Environmental quality is effectively monitored by bioindication methods based on the determination of the ability of plants to absorb chemical elements from the growth soil (Parmar et al., 2016; Holt, Miller, 2010).

Among the dominant environmental pollutants are heavy metals (HMs), which are considered to be essential elements that are necessary as macro- and micro-nutrients for the biological activity of organisms (Pierzynski et al., 2000). Therefore, research on the mechanisms of HMs transport in the food chain (soil-plant-human or soil-plant-animal-human) deserves special attention. They are highly dependent on the metal's chemical form, stimulating processes such as mineral deposition and dissolution, ion exchange, absorption and removal, biological immobilisation and mobilisation, as well as plant uptake and accumulation (Wuana, Okieimen, 2011).

However, many HMs are dangerous even at low concentrations. They can accumulate in the body and contribute to adverse effects (Yan et al., 2020). In addition, unlike organic pollutants, most HMs are not susceptible to microbiological or chemical degradation and can accumulate in the soil for long periods of time (Lasat, 2002). It is important to conduct comprehensive studies that will allow the study of the multi-component effects of anthropogenic pollution of natural soil zones in the Republic of Armenia. This will contribute to the ecological assessment of the quality and safety level of the environment, as well as to the development of monitoring methods. The Republic of Armenia is a mountainous country, with 77 per cent of its territory located at an average altitude of 1,850 metres above sea level. It is characterized by a complex combination of different relief structures, which leads not only to limited land and water use, but also to adverse engineering and geological conditions (high seismicity, abundance of geodynamic processes, etc.) of natural resources in general (Sagatelyan, 2004). The geographical position of the country and its complex mountainous terrain cause the diversity of natural conditions throughout the country. There are 14 types, 27 subtypes and many families, varieties and types of soils in Armenia, and the total number of soils is 228 (Resources ..., 2013). The most common types are mountain-meadow, mountain-brown forest, mountain-brown forest (the latter two types are grouped together with the forest humus-limestone types in the Acrisol reference soil group); mountain-chnozem, mountain-chestnut (chestnut soils) and mountain-brown semi-desert (calcisols). The scattered variety of soils corresponds to the diversity of bioclimatic and lithological-geomorphological conditions of soil formation. The modern evolution of soils in the region is mainly determined by anthropogenic factors (Sagatelyan, 2004; Hunanyan, 2010), although they still exist in some places.

Agriculture is one of the developing sectors of the Armenian economy. The main source of water for irrigation of arable land is surface water and a small amount of groundwater (mainly in the Ararat Valley). Due to the presence of industrial, agricultural, cultural, municipal and other wastewater in these areas, the composition of water in rivers is subject to spatial and temporal

changes. In addition, the use of polluted river water for irrigation leads to lower crop yields and deterioration of crop quality (Simonyan et al., 2018). In regions with a developed mining industry, HMs in irrigation water is in excess of permissible standards (Tutunjyan, Pirumyan, 2011). Potassium, iron, zinc and copper contents in river water fluctuate within the same limits in zones II and III (Figure 2).

The highest concentrations of iron, zinc and copper are found in zone I, which is largely due to the geochemical conditions of the area. The content of these metals decreases by 1.4 to 2.5 times (for zinc by 3.6 to 8.7 times) towards the south-east of the republic (zones II and III). On the contrary, from the north-western part of the country (zone I) to the central part (zone II) and the south-eastern part (zone III), the potassium content in Armenian river waters increases by 1.6 to 1.8 times. Selenium in river water increases uniformly and slowly, by a factor of 2 from zone I to zone II, and remains at low levels. In contrast to selenium, chromium in river waters increases significantly in Zone III (by a factor of 3), which is due to the development of mining and metallurgical industry in the Syunik region (Margaryan et al., 2014). The transport of HMs in soils is highly dependent on their chemical form. It stimulates the proliferation of processes such as mineral deposition and dissolution, ion exchange, adsorption and desorption, complex formation, biological immobilisation and mobilisation, uptake and accumulation by plants (Levy et al., 1992).

Soil is the ultimate site of deposition and accumulation of HMs. In contrast to organic pollutants, the majority of HMs are not subject to microbial or chemical degradation (Kirpichtchikova et al., 2006). Their total concentration in the soil persists for a long time (Adriano, 2001).



Fig. 2. Geographic zones of the studied rivers and their corresponding regions in Armenia: I zone is Northwestdirection with regions: Shirak, Lori, and Tavush; II zone is Middle with regions: Aragatsotn, Kotayk, and Gegharkunik; III zone is Southeast with regions Armavir, Ararat, Vayots Dzor, and Syunik

At the same time, the presence of HMs in soil prevents the biodegradation of organic pollutants (Maslin, Maier, 2000). For an ecosystem, the contamination of soil by HMs poses potential risks through direct contact with contaminated soil via the food chain and through the use of contaminated groundwater (Mc Laughlin et al., 2000). The most important sources for the accumulation of HMs in the soil cover are summarised in Table 1. Among them, exploitation of metal mines, waste disposal in unprotected landfills and combustion of coal and petrochemical residues are more mobile and therefore more bioavailable than pedogenic or lithogenic (Korchagina, 2014).

Table 1. Anthropogenic sources of heavy metal accumulation in soils

Source of pollution	Production type	Concentration coefficient (K_k)*	
		from 2 to 10	more than 10
Non-ferrous metallurgy	Production of non-ferrous metals directly from ores and concentrates	Pb, Zn, Cu, Ag	Sn, Bi, As, Cd, Sb, Hg, Se
	Secondary processing of non-ferrous metals	Pb, Zn, Sn, Cu	Hg
	Production of hard and refractory non-ferrous metals	W	Mo
	Titanium production	Ag, Zn, Pb, B, Cu	Ti, Mn, Mo, Sn, V
Iron and steel industry	Production of nonferrous metals	Co, Mo, Bi, W, Zn	Pb, Cd, Cr, Zn
	Iron ore Production	Pb, Ag, As	Zn, W, Co, W
Metal processing industry	Manufacture of superphosphate	Sr, Zn, F	Cu, Cr, As and rare-earth chemical elements
	Plastics production	-	Y, Ag
	Cement production	-	Hg, Sr, Zn
	Production of concrete products	-	-
Printing industry	Font foundries, printing houses	-	Pb, Zn, Sn
Solid domestic waste of large cities used as fertilizer	-	Pb, Cd, Sn, Cu, Ag, Sb, Zn	Hg
Sewage sludge	-	Pb, Cd, W, Ni, Sn, Cr, Cu, Zn	Hg, Ag
Wastewater for irrigation	-	Pb, Zn	Cu

* Note: K_k is the ratio of element content in the studied object to its background content.

In intensive agriculture, large amounts of fertiliser are regularly applied to the soil to ensure the necessary crop growth. Approximately 10 % of the chemicals registered for use as pesticides, fungicides, insecticides and herbicides are based on compounds containing Cu, Hg, Mn, Pb or Zn (McMullen et al., 2009). They contain N, P and K as impurities as well as trace amounts of HMs (e.g. Cd and Pb), which significantly increase their content in the soil after long-term application (Song et al., 2013). In turn, application of animal manure, composts and compost mixes also accumulates trace elements As, Cd, Cr, Cu, Pb, Hg, Ni, Se, Mo, Zn, Tl, Sb, etc. in soil (Silva et al., 2012). One of the principles of sustainable waste management is the analysis of the potential impact of waste on the environment and the assessment of its potential impact. It is known that Pb, Ni, Cd, Cr, Cu and Zn are most commonly found in particulate matter of biological origin. When they enter the soil, they lead to leaching and contamination of groundwater (Li et al., 2014). Contaminants are therefore classified according to their hazardous environmental effects (Salt et al., 1998). However, regardless of the source of HMs (biotic or anthropogenic), any changes in environmental concentrations have global consequences. By their very nature, HMs contaminate all components of the biosphere (Król, 2011).

Fugitive emissions are often distributed over much smaller areas and close to the soil surface. The type and concentration of HMs thus released is dependent on the specific conditions that cause their deposition on the soil surface (Crommentuijn et al., 1997). Further development of the rock weathering scenario is accompanied by the penetration of simple and complex HMs ions into clay minerals. This is associated with subsequent binding to soil organic matter (Dobrovolsky, 1983). Changes in the concentration of HMs in air, surface water and groundwater result from this distribution.

The upper (sedimentary and granitic-metallic) layer of the continental crust is the main reservoir for the accumulation of HMs. Differences in the prevalence of geochemical anomalies of the studied chemical elements were revealed according to the calculated results of K_k in the surface soil horizon of the experimental sites (Table 2).

Table 2. The values of Clark`s coefficient of soil samples from different soil-climate regions of the Republic of Armenia

Chemical element	Hushakert	Odzun	Shnokh	Teghut	Hrazdan	Gavar	Martuni	Condition control
Ba	0.18	0.42	1.21	0.49	0.85	0.89	1.36	0.98
Sr	0.69	0.58	0.55	0.49	1.66	2.03	2.84	1.29
Rb	1.20	20.8	2.29	2.61	4.42	3.43	8.58	4.08
As	3.14	1.96	8.38	1.70	8.02	4.83	7.19	3.02
Pb	0.80	1.04	4.34	0.68	8.47	5.52	5.66	2.14
Zn	1.32	0.87	3.39	0.72	1.30	1.18	2.28	2.35
Cu	1.64	2.66	16.14	1.24	2.09	2.12	3.68	2.23
Ni	1.11	0.88	1.27	1.19	1.31	1.65	2.85	1.67
Co	14.24	12.75	14.58	7.08	4.94	8.62	7.61	9.48
Fe	1.08	0.54	0.66	0.52	0.72	0.95	1.55	0.90
Mn	1.30	0.55	0.87	0.59	1.09	1.07	1.92	1.16
Cr	4.02	1.07	0.76	0.78	1.13	1.28	2.94	2.07
V	1.93	1.01	0.96	0.91	1.05	1.11	1.93	1.12
Ti	0.78	0.72	0.78	0.81	0.95	1.12	2.05	1.21
Ca	1.23	1.20	0.66	0.93	0.51	0.61	1.87	1.15
K	0.29	0.51	0.55	0.62	0.71	0.59	1.23	0.61
S	trace	0.41	0.53	0.312	1.36	0.98	1.42	0.32

*Note: Marked chemical elements have no Clark's value to calculate the coefficient (K_k), and if $K_k > 10$, the content of concentrations of chemical elements in soil samples is high; if $1.6 < K_k < 10$, then the content of concentrations of chemical elements in soil samples is average; if $1.0 < K_k < 1.5$, then the concentration of the chemical element in soil samples is poor (Grigoryev, 2009).

The distribution of chemical elements in the environment is influenced by many factors: the distance from the source of pollution and its power; the quantity and composition of gas and dust emissions reaching the soil; the transformation of the element and the state of its compounds;

the properties and composition of the soil, the dynamics of soil processes, etc. With the most modern technological production schemes, a significant amount of highly toxic compounds can be released into the environment, polluting the atmospheric air, the water and the soil of the regions in equal measure. The danger of elevated levels of HMs in an ecosystem is that most chemical elements are highly biologically active (Singh et al., 2011). HMs are the second most dangerous substances (after pesticides), but far ahead of carbon dioxide and sulphur dioxide, which may become more dangerous than nuclear power plant waste and solid waste in the future (Santa-Cruz J et al., 2021; Aschmann, 2019). As a result of their dispersion through air currents, they reach the land cover and the water surface, while as a result of migration they accumulate and enter the trophic chain in various components of the biota (Mishra et al., 2019; Etim et al., 2021; Dovletyarova et al., 2022; Sukiasyan et al., 2020). Consider the situation of arable land contamination using the example of the industrial zone surrounding the city of Hrazdan, which is one of the most important industrial and energy centres in the Republic of Armenia. The Hrazdan Thermal Power Station (HTPS) and the Hrazdan Cement Plant (HCP) are located here. Other important enterprises are also located here. In this case, it is mainly the quality, grade and type of natural material that determines the toxicity of the flue and exhaust gases emitted into the air. In addition, being close to a gas turbine power station only exacerbates the uncontrolled pollution that can be caused by heat engines and plants burning hydrocarbon fuels. This is the reason why pollution in Hrazdan constantly exceeds the maximum permissible concentrations by almost 8 times (Resources..., 2013). The degree of danger of pollutant emissions from thermal power plants may be due to the low quality of the raw materials used, which implies the presence of a large number of accompanying impurities. Airborne pollutants enter the soil where they accumulate as a result of migration and enter various biota (Kumar et al., 2008; Sukiasyan, Kirakosyan, 2020). Such a unique natural reservoir can "reflect" not only in situ pollution processes, but also those that occurred long ago (Levshakov, 2011).

A qualitative analysis of the soils in terms of the degree of contamination is crucial to solving the problem of the ecological state of the region (Blaser et al., 2000). In our studies, the soil cover is mainly represented by chestnut soils and chernozems. These soils are characterised by the loss of the upper fertile layer during industrial pollution (Kroyan et al., 2021). Special consideration was given to the peculiarities of the wind direction in the area. In cement production, the pollutants that spread in the atmosphere and are gradually deposited on the soil contain different HMs (Arfala et al., 2018). It has been shown that the largest percentage decrease in HMs content with distance from the source has been recorded for Pb, slightly less for Zn and Cu, averaging 52 % (Rühling, Tyler, 2001).

In a series of experiments, the influence of anthropogenic pollution in the vicinity of industrial enterprises in Armenia was determined in samples of arable soils taken at a distance of 0.5, 1.5, 2.5, 5, 10, 15 and 25 km from the source of pollution. In particular, the peculiarities of the direction of the wind rose in the area were taken into account. According to the results, the highest values of the content of the studied HMs in the anthropogenic HCP zone were recorded in the range of 0.5-2.5 km as the following comparative series $Zn > Pb > Cu$. The average concentration of Zn and Pb decreased by 78% and Cu by 53% at a distance of 25 km from the plant. The order of comparison for the pollutants under investigation was $Zn > Cu > Pb > Co > Mo$ (Sukiasyan et al., 2021). Based on the values of concentrations of some chemical elements, coefficients were calculated (the risk of soil contamination (K_o) and the total pollution index (Z_c)). These coefficients made it possible to assess the degree of anthropogenic load and the level of soil pollution (Sukiasyan, 2018; Kasimov, Vlasov, 2015; Müller, 1981). Thus, in the vicinity of the HCP, a low level of pollution was indicated by the values of Z_c within 5 km. Changes in the coefficient were about 90 % at a further distance of 25 km from the source of contamination. The highest risk of soil contamination in the vicinity of the HCP was lead ($K_o = 2.9$). This risk was maintained up to a distance of 5 km from the source, after which there was a sharp decrease. A similar situation was observed for zinc ($K_o = 2.3$). Pollution by copper compounds was consistently high (Sukiasyan et al., 2021). These experiments were continued to determine the effect of the wind rose at a distance of up to 1 km using the example of the Hrazdan industrial zone. Concentrations of As and Pb decreased on average by 27 % with increasing distance from the industrial zone in the city of Hrazdan (Table 3). In quantitative terms, the concentration of lead was three times higher than that of arsenic along the sampling line.

Table 3. The value of Clark's coefficient (K_k), the coefficient of soil contamination hazard level (K_o) and the total contamination index (Z_c) near the Hrazdan industrial zone

Distance from the source of pollution (Hrazdan industrial zone), km	As		Pb		Zn		Cu		Ni		Co		Z_c
	K_k	K_o	K_k	K_o	K_k	K_o	K_k	K_o	K_k	K_o	K_k	K_o	
1.0	7.27	4.07	7.44	3.95	1.39	1.16	1.97	1.69	1.43	10.66	1.79	3.36	16.29
0.8	8.11	4.54	9.92	5.27	1.07	0.89	1.54	1.32	1.45	10.81	0	-	17.09
0.6	9.02	5.05	9.73	5.17	1.19	0.98	1.86	1.59	1.15	8.61	0	-	17.95
0.4	9.48	5.31	9.76	5.19	1.23	1.02	1.70	1.45	1.40	10.46	0	-	18.58
0.2 (near industrial zone)	9.88	5.53	10.34	5.49	1.01	0.84	1.60	1.37	1.14	8.49	0	-	18.96

*Note: The value of $K_o > 1$ pollution level was high danger; ** the summary pollution level was classified as low with $Z_c < 16$ contamination is considered as non-dangerous; with $16 < Z_c < 32$ contamination is moderately dangerous; with $32 < Z_c < 128$ contamination is dangerous; with $Z_c > 128$ contamination is extremely dangerous (Müller, 1981).

Soil samples show an average 25.5 % increase in zinc and nickel and a 19% increase in copper concentrations away from the source of pollution. Cobalt is also of particular interest. Its traces were found in soil samples close to the source of pollution. This is why the value of Clark's coefficient is zero. Already along the sampling line, at a distance of one kilometre, its concentration in soil samples was 84.0 mg/kg, which made it possible to calculate K_k . Then, the calculation of some coefficients K_o and Z_c , taking into account Clark's coefficient for each of the HMs, allows to determine the possibility of its migration, taking into account the wind rose (see Table 3).

In a situation where the coefficient is $K_o > 1$, especially in the vicinity of the source of contamination, it can take on exorbitant values. According to the established criteria, the risk of contamination increases in direct proportion to the content of HMs in the studied samples, taking into account their maximum allowable concentration (Sukiasyan, 2018). A moderately hazardous level of contamination was found in the soil samples collected in the study areas at a distance of 0.2 km from the source of contamination. The value of the total pollution index (Z_c) decreased by 16 % with increasing distance from the enterprises. As we move away from the sources up to 1 km, there is a significant increase in the concentrations of other elements (Ni, Co, Cu and Zn). The processes of migration of HMs in anthropogenically polluted areas may be due to the dominance of the southwest direction of the wind rose in this region, and a comparative line is presented as a sequence: As < Ni < Co < Cu < Zn < Pb.

4. Conclusion

Information on the extent and irreversible consequences of HMs contamination of biota is abundant. However, without going into the nature of the anomaly, all discussions about the created situation are ultimately reduced to a comparison of changes in the concentration of HMs in the environment with the accepted maximum permissible concentrations. The proposed analysis of the current situation on the example of the assessment of the technogenic zone of the city of Hrazdan brings to the fore in geo-ecological studies the concept of differentiation between abiotic and anthropogenic pollution of HMs and possible ways of their migration on the basis of the developed approach to the assessment of pollution. The choice of this approach makes it possible to apply clear limits to the use of natural resources and, to some extent, contributes to the development of conservation measures aimed at preserving the biota as a whole.

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