Assessment of Soil Contamination by Heavy Metals in the Area Affected by Petrol Stations

Svitlana Domuschy[1][a], Valentina Trigub[1][b] and Elgudja Kulidjanov[2]

[1]*Department of Geography of Ukraine, Soil Science and Land Cadastre, Odesa I. I. Mechnikov National University, 2, Champagne Lane, Odesa, Ukraine*

[2]*Odesa branch of the State institution "Soils Protection Institute of Ukraine", 19, Laboratorna Street, Mizykevycha village, Ovidiopol district, Odesa region, Ukraine*

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Abstract: The results of experimental researches on determination of the content of mobile forms of heavy metals in the soils of the city of Odessa (Ukraine) within the limits of influence of petrol stations are presented. The negative role of road transport emissions on modern climate change is outlined. The assessment of the ecological condition of the city soil cover by the concentration coefficients and the total indicator of heavy metal pollution was carried out. It was found that the soils within the limits of influence of all petrol stations have a much higher level of accumulation of the studied HM compared to the background content. Installed those soils in terms of concentration coefficients have unsatisfactory ecological status in terms of zinc (100%), copper (73%) and lead (50%) content of the study area. It was found that according to the indicator of total pollution, 55% of the studied area belongs to the dangerous category of soil pollution. The priority pollutants that have the highest levels of pollution intensity are zinc and copper, which can significantly affect the increase in the overall morbidity of children and adults in the city. A correlation was established between the intensity of soil pollution and the total pollution rate (r = 0.99), which confirms the negative impact of vehicle emissions on the soil cover of the city.

1 INTRODUCTION

In modern cities there is a rapid increase in the number of petrol stations (PS). These environmentally hazardous sites are often located in residential areas of cities, causing high levels of local pollution, which is closely related to transportation, storage, filling of tanks and spilling of petroleum products. Peculiarities of petrol station environmental pollution are high concentration of pollutants in the surface layer of the atmosphere with subsequent deposition on the soil and plant surface, as well as the lack of scattering of pollutants by wind currents. Pollution from stationary sources located on the territory of the petrol station is supplemented by emissions from vehicles. At the petrol station site, the speed of cars is significantly reduced, and during the operation of the internal combustion engine at low speeds, the emission of toxic substances into the atmosphere increases. It is known that exhaust gases contain more than 200 different toxic compounds, including heavy metals (HM) - Pb, Mn, Cd and others (Ibragimova, 2006; Ekzan, 2005). Heavy metals, as a result of participation in various migration cycles, pollute all vital areas: the atmosphere, hydrosphere, pedosphere. Their high content causes morphological and physiological abnormalities and adversely affects the main functions of living organisms (bioproductivity, generative capacity, etc.) (Ibragimova, 2006).

With a high degree of contamination of soils with heavy metals, there is a change in some chemical and physical properties of the soil. As long as heavy metals are strongly bound to the constituent parts of the soil and difficult to access, their negative impact on the soil and the environment will be insignificant.

Thus, the study of the content of soluble forms of heavy metals in the soil of cities is an urgent problem

[a] https://orcid.org/0000-0003-3375-2682
[b] https://orcid.org/0000-0002-4436-2017

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in the process of ecotoxicological assessment of environmental pollution by petrol station emissions.

2 ANALYSES OF RECENT RESEARCH AND PUBLICATIONS

Problems of environmental pollution by heavy metals and petroleum products near petrol stations and highways have been previously covered in scientific publications of Radomska, 2010; Sheikina and Mysliuk, 2008; Franchuk and Radomska, 2009; Isabel M. Morales Terrés et al., 2010; Fernández-Villarrenaga et al., 2005. Scientists paid special attention to determining the concentration of heavy metals (Pb, Mn, Cd, Cu, Zn) in the soil cover along the highways of industrial and urban areas (Hryshko et al., 2012; Bilyk et al., 2009). The question of studying the range of possible toxic consequences of Petrol stations for workers and civilians dedicated works Radomska, 2015; Brugnone et al., 1997; Karakitsios, 2007, Lynge et al., 1997; Antropchenko et al., 2016.

It is known that pollution of the earth's surface by transport emissions near and on the territory of petrol stations is accumulating gradually. The degree of pollution depends on the number of vehicles passing through the adjacent route, road quality, storage conditions (transportation) of fuel at the petrol station and persists for a long time, even after road closure, road closure, highway, or complete road and asphalt removal) (Franchuk & Radomska, 2009).

Various chemical elements, especially heavy metals that accumulate in the soil, are absorbed by plants and through them through the food chain pass into the body of animals and humans. Some of them dissolve and are carried away by groundwater, polluting rivers and other bodies of water and already with drinking water can enter the human body (Bilyk et al., 2009, Chaika et al, 2018).

The aim of the article is to study the ecological condition of urban soils in relation to their contamination with soluble forms of heavy metals within the limits of influence of petrol stations in the city of Odessa (Ukraine).

3 MATERIALS AND METHODS

Contamination of the environment with chemical elements, and especially heavy metals, is usually determined in relation to the background content of these elements and (or) the maximum allowable concentration (MAC) in it.

The level of soil and plant pollution depends on the type of plants, forms of chemical compounds in the soil, the presence of elements that counteract the effects of heavy metals and substances that form complex compounds with them, adsorption and desorption, the number of available forms of these metals in soil and soil-climatic conditions. As noted, the negative impact of heavy metals significantly depends on their mobility (solubility) (Bilyk et al., 2009).

The object of the study was urban soils within the limits of influence of petrol stations. Petrol stations in different parts of the city of Odessa were selected to study the contamination of urban soils with heavy metals, where soil samples were taken at a distance of 10-15 m from fuel storage tanks. All petrol stations in the city are located next to highways, which are also an additional source of pollution.

The control area was located at a considerable distance from the sources of industrial and motor transport pollution (the territory of the city's botanical garden, № 11) (Figure 1).

Figure 1: Map-scheme of soil sampling within the limits of influence of petrol stations in Odessa.

The location and geographical coordinates of the selection points are given in Table 1.

Sampling was carried out mainly by the envelope method from a depth of 0-15 cm. Preparation of samples was carried out according to standard methods. In the testing center of the Odessa branch of the State Institution "Soil Protection Institute of Ukraine" were conducted studies to determine the content of mobile forms of heavy metals Mn, Co, Cd, Pb, Cu, Zn (according to DSTU 4770.1: 2007, DSTU 4770.5: 2007, DSTU 4770.3: 2007, DSTU 4770.9: 2007, DSTU 4770.6: 2007, DSTU 4770.2: 2007 ammonium acetate buffer with pH 4.8 on the atomic absorption spectrophotometer AAS 115) (National

Standard of Ukraine…, 2005). To compare our research on the assessment of soil contamination of the city of Odessa with heavy metals, we used the average data of zonal soils of the territory - the chernozems of the south (Kulidjanov et al., 2014).

Statistical processing of the obtained results was performed using the MS Excel package. The degree of contamination of the soil of the urban environment with heavy metals was carried out according to the indicator of the concentration factor of chemical elements (Ks) and the total pollution index.

Table 1: Soil sampling points.

№	The name of the study area	Location	Geographical coordinates
1	Petrol station № 1 «Socar»	the Lustdorf road Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
2	Petrol station № 2 «WOG»	Balkivska Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
3	Petrol station № 3 «Catral»	Balkivska Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
4	Petrol station № 4 «OKKO»	Hrushevsky Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
5	Petrol station № 5 «Alcor-Oil»	Southern road	$46^0$26'35'' n. l. $30^0$43'37'' east l.
6	Petrol station № 6 «BRSM Oil»	Black Sea Cossacks Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
7	Petrol station № 7 «OKKO»	Krasnova Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
8	Petrol station A№ 8 «Motto»	Avenue of the Heavenly Hundred	$46^0$26'35'' n. l. $30^0$43'37'' east l.
9	Petrol station № 9 «WOG»	Lustdorf road Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.
10	Petrol station № 10 «X-Oil»	Gagarin Avenue	$46^0$26'35'' n. l. $30^0$43'37'' east l.
11	Control (the territory of botanical garden)	French Boulevard Street	$46^0$26'35'' n. l. $30^0$43'37'' east l.

The concentration coefficient of the studied heavy metals, which is characterized by the ratio of the content of the chemical element in the soil to its background value, was calculated by the formula (1) (Madzhd et al., 2016):

$$Kc = Ci / Co, \quad (1)$$

where: Ci - the content of the chemical element in a particular object, mg/kg; Co - background content of the chemical element in the soil, mg/kg.

The total pollution rate was determined by the formula (2) (Madzhd et al., 2016):

$$Zc = \sum_{i=1}^{n} Kc - (n-1), \quad (2)$$

where n is the number of total elements; Kc - concentration factor.

The level of ecological condition of the soil cover of the study area was determined in accordance with the indicators of dangerous of soil contamination according to the total indicator Zc, proposed by Yu. V. Saet (1990). There are 4 categories of soil pollution: admissible if Zc <16 - is characterized by the lowest level of morbidity in children and a minimum of functional disorders in the adult population; moderately dangerous, if Zc = 16-32 - is characterized by an increase in the overall incidence; dangerous if Zc = 32-128 - is characterized by an increase in the overall level of morbidity, the number of frequently ill children, children with chronic diseases, dysfunction of the cardiovascular system; very dangerous if Zc> 128 - is characterized by increased morbidity of children, impaired reproductive function in women (increased cases of toxicosis during pregnancy, premature birth, stillbirth, infant malnutrition) (Madzhd et al., 2016)

The risk of soil contamination with heavy metals in relation to public health was determined by the intensity of pollution. The intensity of soil contamination (Pj) was calculated by the formula (3):

$$Pj = \sum(Kc \times Mi), \quad (3)$$

where: Kc is the coefficient of concentration of the trace element; Mi - the value of the hazard index (toxicity) of a chemical element according to the hazard class (4.1 and more - the first class; 2.6-4.0 - the second class; 0.5-2.5 - the third class; up to 0.5 - fourth grade) (Chaika et al, 2018).

The level of environmental hazard was determined according to a scale developed taking into account the impact of pollution intensity indicators on the health of the population (Table 2).

Table 2: Characteristics of environmental hazards of soil pollution (Chaika et al, 2018).

Category of soil pollution intensity	Admissible	Permissible	Dangerous	Very dangerous
Intensity of soil pollution, P_j	15 and under	16-30	31-50	51 and more
Changes in population health indicators	The lowest level of morbidity	Increasing the overall morbidity of the population	Increase in the general morbidity, chronic diseases, disturbance of a functional condition of cardiovascular system	Increase in the general morbidity of children, reproductive dysfunction in women

4 RESULTS OF THE STUDY AND THEIR DISCUSSION

It is known that environmental pollution by emissions from road transport leads to both short-term and long-term negative consequences. During the operation of road transport engines, a wide range of gases and solids are released, the impact of which leads to the intensification of global warming, acid rain, pollution of all components of the environment.

Pollution by road transport has a negative impact in several areas:

- global warming;
- pollution of air, water and soil;
- impact on the health of the population (Serdyukova & Barabanshchykov, 2018).

Car emissions contain various greenhouse gases, such as carbon monoxide and nitrogen oxide, which are able to block the sun's rays reflected from the Earth's surface, changing air temperature, which is one of the main factors in global warming (When will the Arctic…, 2016).

In turn, the harmful effects of global warming on the environment are manifested in such negative consequences as desertification, increased melting of snow and ice, rising sea levels, the emergence of severe storms and other extreme natural phenomena (When will the Arctic…, 2016).

It is well known that the air temperature is always higher within the city. One of the reasons for this phenomenon is the presence of heavy traffic.

Emissions from road transport, especially old cars, are toxic to living organisms and can cause a variety of diseases (such as lung cancer); negatively affect the growth and development of plants. However, the greatest danger from environmental pollution from road transport emissions is the reduction of the ozone layer. After all, it is the presence of the ozone layer that prevents harmful ultraviolet (UV) rays from entering the atmosphere, which can cause many diseases, including skin cancer and others. (Jenny, 2018).

During the operation of the car with internal combustion engines, the sources of emissions of harmful substances are: exhaust gases, crankcase gases, evaporation from the power supply systems, uncontrolled spillage of consumables. Exhaust gases from transport contain a large amount of lead, which together with salts of other metals enters the trophic chain "soil - groundwater - plants - animals - humans".

Thus, the increased content of lead in the human body leads to anemia, renal failure, mental retardation, an increase in the number of nervous diseases; high content of zinc - to negative changes in the composition of the blood, reduces the body's resistance to infections, promotes the development of cancer cells, delays growth and sexual development; excess copper content leads to cancer, central nervous system disorders, decreased blood vessel plasticity (Voloshyn, and Mezentseva, 2007).

In the aerogenic type, heavy metal compounds accumulate in the upper humus horizons, forming complex complexes with organic matter and have a significant period of removal from the soil, in particular, zinc - from 70 to 510, cadmium - 13-110, copper - 310-1500, lead - 770-5900 (Orlov, et al., 2001). Therefore, determining the content of heavy metals in soils is of great importance for assessing their danger within large cities, including the city of Odessa.

The current situation in Ukraine is characterized by a reduction in environmental pollution by industrial emissions, but the level of air pollution and urban soil pollution remains high, due to a significant increase in road transport and, accordingly, Petrol stations.

The data shown in Table 3 show that soils within the influence of all petrol stations have a much higher level of accumulation of the studied heavy metals compared to the background content, and lead - with the content of MAC. The content of heavy metals is respectively: for manganese - 1.3-1.7; zinc - 7-51; cobalt - 0.6-4; copper - 4-24; cadmium - 0-4; lead - 1.5-14 background content levels.

According to the calculations, it is established that the priority pollutants that can cause a negative impact on the components of the city's environment is lead. Lead is a heavy metal of the first class of danger and it is considered one of the most toxic chemical elements, even in small quantities. Compared with the MAC, urban soils at all study sites (except the control area) are significantly higher in the content of mobile lead (from 1 to 4 MAC).

Table 3: The content of heavy metals in the studied soils.

Place of selection	Content of chemical elements in soils, mg / kg					
	Mn	Zn	Co	Cu	Cd	Pb
PS № 1	55,8	3,2	0,3	1,3	0,4	1,5
PS № 2	48,1	22,0	1,6	2,6	0,6	19,5
PS № 3	47,3	8,2	0,6	0,8	0,3	5,6
PS № 4	52,3	19,3	0,8	5,4	0,5	25,7
PS № 5	47,3	22,3	0,9	5,3	0,2	13,5
PS № 6	42,4	3,6	0,2	2,0	0,3	2,6
PS № 7	51,6	5,7	0,5	1,1	0,2	3,5
PS № 8	50,2	20,7	1,2	4,0	0,5	22,6
PS № 9	51,6	12,8	0,6	3,3	0,1	7,5
PS № 10	45,2	12,8	0,9	2,3	0,4	11,1
Average content at the petrol station	49,2	13,1	0,8	2,8	0,4	11,3
Control	39,2	3,2	0,3	1,3	0,0	1,5
Background content	33,9	0,4	0,4	0,2	0,2	1,9
MAC	-	23,0	5,0	3,0	0,7	6,0

Copper is a chemical element that belongs to the heavy metals of the II class of danger. In about 40% of the studied areas, the maximum concentration limit of mobile forms of Cu was exceeded. The maximum excess is about 2 MAC (Petrol station № 4 (5.4 mg/kg).

Manganese - belongs to the III class of danger. The presence of manganese more than normal adversely affects the human body, which is expressed in the destruction of the central nervous system. Manganese values in all studied soil samples exceed the background content by 1.5-2 times. The highest content of manganese was observed at petrol station № 1 (55.8 mg/kg).

Zinc is a heavy metal of the first class of danger. In conditions of high humidity is characterized by high migration into the soil. For the distribution of mobile forms of zinc, the trend of maximum content in soil samples taken near petrol station № 2 (22.0 mg/kg) and petrol station № 5 (22.3 mg/kg), which is close to the MAC.

Cobalt is a chemical element that belongs to the II class of danger. Analyzing the distribution of mobile forms of cobalt, it was found that 100% of the studied areas do not exceed the MAC, but are much higher than the background content.

Cadmium - belongs to the I class of danger. Its compounds are extremely toxic, even in small concentrations. The average cadmium content (0.35 mg / kg) in all studied soil samples taken near the petrol station does not exceed the MAC, but is much higher than the background content.

As a quantitative indicator of the activity of radial migration, we used the concentration factor (Kc), which characterizes the degree of accumulation of substances in the system component relative to the selected standard. For the standard we used the background content of heavy metals. Background values of heavy metal concentrations were used to calculate Kc (Kulidjanov et al., 2014). The value of the concentration coefficient (Kc) indicates the activity of the processes of leaching (Kc <1) and accumulation (Kc > 1) of substances in the genetic horizons (or a separate horizon) of the soil.

The ecological condition of the soil by the concentration factor is defined as unsatisfactory if the excess of the concentration factor is ≥ 5.0 times; satisfactory - 3.0-5.0; normal 1.0 - 2.9; optimal ≤ 1.0 (Madzhd et al., 2016).

According to the calculation of the coefficient of concentration of chemical elements (Table 4), it was found that the content of manganese, cobalt and cadmium does not pose an environmental hazard to soils, as Ks does not exceed 5 times. Unsatisfactory ecological condition of the soil according to the coefficients of zinc concentration (Ks ≥ 5), are characterized all studied areas, including the control area. 73% of the study area is characterized by high coefficients of copper concentration (Ks ≥ 5). According to the coefficients of lead concentration, 50% of the study area has a satisfactory and 50% - unsatisfactory ecological status of soils (Petrol stations № 2, 4, 5, 8, 10).

According to the approximate assessment scale of soil contamination hazard according to the total pollution index (Zc), 55% of the study area belongs to the hazardous category of soil contamination. The most critical situation is within the influence of petrol station № 4 (Zc = 73.9), petrol station № 5 (Zc = 72.2), petrol station № 8 (Zc = 70.6), which indicates the high ability of urban soils to absorb and retain heavy metals (Table 4). It can be assumed that in these areas we can see an increase in the overall incidence of urban population, an increase in the number of children who are often ill, children with chronic diseases, an increase in the proportion of people with cardiovascular disorders.

Table 4: The value of the coefficients of concentration of chemical elements (Kc) and the total pollution index (Zc) in the studied soils.

Place of selection	Indicators, Kc						Zc
	Mn	Zn	Co	Cu	Cd	Pb	
PS № 1	1,7	7,4	0,9	6,0	2,7	0,8	4,3
PS № 2	1,4	50,1	4,2	12,0	3,9	10,5	67,1
PS № 3	1,4	18,6	1,7	3,6	1,7	3,0	14,8
PS № 4	1,5	43,8	2,2	24,4	3,2	13,8	73,9
PS № 5	1,4	50,6	2,3	24,0	1,6	7,3	72,2
PS № 6	1,3	8,2	0,6	9,2	2,0	1,4	7,7
PS № 7	1,5	13,0	1,3	4,8	1,0	1,9	8,4
PS № 8	1,5	47,0	3,2	18,2	3,6	12,2	70,6
PS № 9	1,5	29,0	1,6	15,0	0,9	4,0	37,1
PS № 10	1,3	29,2	2,5	10,6	2,5	5,9	37,0
Ave-rage values on the petrol station	1,5	29,7	2,1	12,8	2,3	6,1	39,3
Control	1,2	7,4	0,9	6,0	0,3	0,8	1,4

According to the calculations (Table 5), it is established that the priority pollutants that may adversely affect human health are zinc (Pj = 121.6) and copper (Pj = 51.0). Accordingly, according to the scale of environmental dangerous assessment, soil pollution belongs to the fourth - very dangerous category of pollution intensity, which may increase the overall morbidity of children and adults in the city.

The average intensity of lead contamination of the studied soils (Pj = 24.9) corresponds to the second category (permissible); manganese (Pj = 3.6), cobalt (Pj = 8.2) and cadmium (Pj = 9.5) corresponds to the first zone (admissible category of soil contamination intensity), for which the minimum level of morbidity is possible.

The control area for the intensity of manganese, cobalt, cadmium and lead pollution belongs to the admissible category of pollution; zinc and copper are permissible and dangerous, respectively.

Table 5: Intensity of soil pollution Pj.

Soil sampling points	Indicators Pj					
	Mn	Zn	Co	Cu	Cd	Pb
PS № 1	4,1	30,2	3,5	23,8	11,0	3,4
PS № 2	3,6	205,4	17,0	47,8	16,1	43,1
PS № 3	3,5	76,1	6,7	14,2	6,9	12,2
PS № 4	3,9	179,7	8,8	97,4	13,1	56,6
PS № 5	3,5	207,4	9,3	96,0	6,6	29,7
PS № 6	3,1	33,7	2,5	36,7	8,2	5,6
PS № 7	3,8	53,1	5,2	19,1	4,1	7,8
PS № 8	3,7	192,5	13,0	72,7	14,8	49,9
PS № 9	3,8	118,8	6,4	60,0	3,8	16,6
PS № 10	3,4	119,6	9,8	42,4	10,1	24,4
Pj average on the petrol station	3,6	121,6	8,2	51,0	9,5	24,9
Control	2,9	30,2	3,5	23,8	1,1	3,4

Heavy metal content is interrelated with individual components of urban ecosystems. The values of the correlation coefficients showed the existence of a mathematically proven relationship between the intensity of soil pollution and the total rate of pollution. It should be noted that a close correlation is observed (r = 0.99) between these indicators (Figure 2).

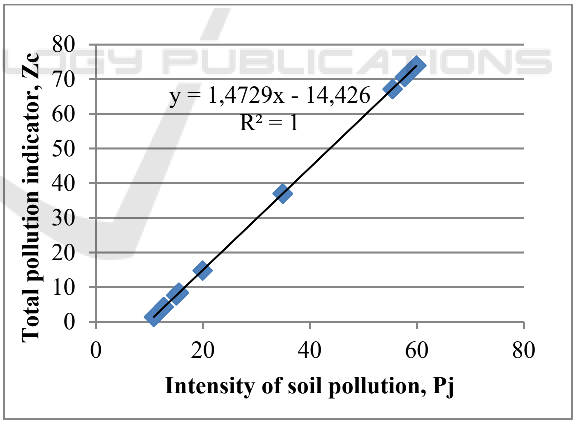

Figure 2. Correlation between the intensity of pollution and the total rate of soil pollution.

Studies show that there is a high probability of man-made impacts of road transport emissions on the soils of the study area, a high degree of their contamination with heavy metals, which can have a negative impact on the health of urban populations. Additional research is needed to confirm and study in more detail the impact of road transport emissions on the morbidity of the population.

5 CONCLUSIONS

As a result of research to determine the content of heavy metals in the soil within the influence of petrol stations, the following was found:

1. Heavy metals have a high ability to accumulate in the soil. Analysis of the content of heavy metals in soils is a representative indicator of the ecological state of the territory. It was found that the content of heavy metals within the influence of petrol stations significantly exceeds their background content. The main sources of pollution are emissions from motor transport.

2. For spatial interpretation, correct and visual presentation of data on soil contamination by various chemical compounds, it is advisable to use dimensionless indicators, in particular the concentration factor. It is established that soils have unsatisfactory ecological condition according to concentration coefficients: according to zinc content - 100%; copper - 73%; lead - 50% of the study area.

3. It was found that 55% of the study area belongs to the dangerous category of soil pollution (in the total pollution indicator).

4. The priority pollutants that have the highest levels of pollution intensity are zinc and copper, which can significantly affect the increase in the overall morbidity of children and adults in the city.

5. A correlation was established between the intensity of soil pollution and the total pollution indicator (r = 0.99), which confirms the negative impact of motor transport on the soil cover of the city.

6. Emissions from road transport have a significant negative impact on modern climate change, leading to warming, "damage" to the ozone layer, the fall of acid rain.

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