

NICKEL IN TUZLA TOPSOILS: ITS DISTRIBUTION AND IMPACTS

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Abstract

Nickel in Tuzla topsoils: Its distribution and impacts

The paper presents the results of geoecological - pedological research of nickel (Ni) concentrations in the soil of the city of Tuzla. The main goal of the research was to determine the extent to which the soil of the city of Tuzla is contaminated with nickel and to determine the origin of pollutants. The 240 soil samples were collected during the terrain research, covering an area of 303 km². The nickel concentration testing in soil samples was performed by mass spectrometry (ICP-MS) with a detection range of 0.1 - 10000ppm. Exceeding the maximum allowable concentrations of nickel, defined by the Ordinance on determining the allowable amounts of harmful and dangerous substances in the soil and methods of their testing was recorded in 99.16% of samples.

The range of determined nickel concentrations is 30 - 1437.7ppm, and the average value of all measured concentrations in the samples is 297.91ppm. The highest concentration of nickel was recorded in sample 64 (1437.7ppm), collected in the area of Lipnica. Out of 240 total analyzed soil samples, nickel concentrations were increased in 238 samples, in some places several tens of times larger than the allowed limit. Elevated concentrations were also recorded in sample no. 59 (1109.1ppm) in the area of Požarnica - Kovačica and in sample 189 (1005ppm) in the area of Slavinovići. At twenty locations, nickel concentrations vary from 500-1000ppm.

Keywords

Nickel (Ni), pollution, soil, Tuzla.

1. Introduction

The area of the city of Tuzla geographically belongs to the region of northeastern Bosnia, more precisely to the Spreča-Majeвица subregion. Tuzla is located in the valley of the Jala river. From the northeast, it's surrounded by medium high mountain morphostructure of Majeвица, and from the south by the Spreča valley. Tuzla's area is located between 18°55' and 18°9' E and 44°48' and 44°67' N, at an altitude from 200 m at the lowest point at the Jala riverbed up to 600 m at the eastern and northeastern border of the city. The area of Tuzla covers about 303km² and it's located on the northern slope of the Dinaric mountain system and is generally mildly tilted towards the Upper Spreča valley. There are about 111,000 inhabitants living in 66 settlements in the researched area (Stjepić Srkalović, 2015., Census, 2013.).

The geological base of the soil implies the rock surface from which the soil is created and developed under the influence of several factors. Soil material can be produced by any rock, provided that it is on the surface and thus it's subject to physical, chemical and biological influences that lead to the disintegration of its surface layer (Ćirić, 1991.). For a longer period, the area of the city and the wider surroundings of Tuzla has been marked by processes of urbanization and deruralization, industrialization and deagrarization, which has largely contributed to soil pollution, degradation and devastation (Stjepić Srkalović, et al, 2016.).

Environmental contamination with nickel, both anthropogenic and natural, is a common occurrence in the world and is recognized as a global problem. Nickel and certain nickel compounds are listed in the National Toxicology Program (NTP) as reasonably expected carcinogens. The International Agency for Research on Cancer (IARC) listed nickel compounds within group 1 (there is sufficient evidence of carcinogenicity to humans) and nickel within group 2B (agents that are likely to be carcinogenic to humans) (Lenntech.com., 2016. Periodic table - chart of all chemical elements.).

The main goal of the research was to determine the extent to which the soil of the city of Tuzla is contaminated with potentially toxic elements (PTE), and especially nickel. Igneous rocks are the primary source of Ni found in soils. The total Ni concentration in soils is directly related to the concentration in the parent material and depends on erosion. Organic matter has the ability to absorb metals and coal and oil can contain significant amounts. Power plants and garbage incinerators release nickel into the air, which binds with moisture in the air and reaches the ground through raindrops. It takes a long time for the air to be cleaned of nickel. Nickel can also end up in surface waters when part of wastewater flows. Most of the nickel compounds released into the environment will be absorbed into the soil and as a result become immobile (Lenntech.com., 2016. Periodic table - chart of all chemical elements.).

1.1 Geological setting and pedogeographic characteristics

1.1.1 Geological setting

The oldest structures belong to the Tuzla's Lower Miocene formations in which organogenic limestones are prevailing ("slavinovički" limestones and dolomites) with sporadic marls. Above them, the clasts were deposited with characteristic reddish coloring sandstones and conglomerates, building the "red" series. The continuation of the sedimentation cycle is made of a "layered" series, where the salt formation with accompanying dolomite, anhydrite and tufts are developed. The organogenic

limestones, clays, marly clays, sands and subsidiary conglomerates are belonging to the youngest Miocene products. The development of the lower pliocene is characterized by the deposition of several lignite (main, base and top seams). Vertical development of the Pliocene formation has the characteristics of rhythmicity: quartz sand, clays (slate and alevrite) and lignite. Quaternary formations were developed along the streams in the form of proluvial depositions (debris) and as precipitated terrace and alluvial sediments (sand and pebbles) (Figure 1) (Čičić, et al, 1988.).

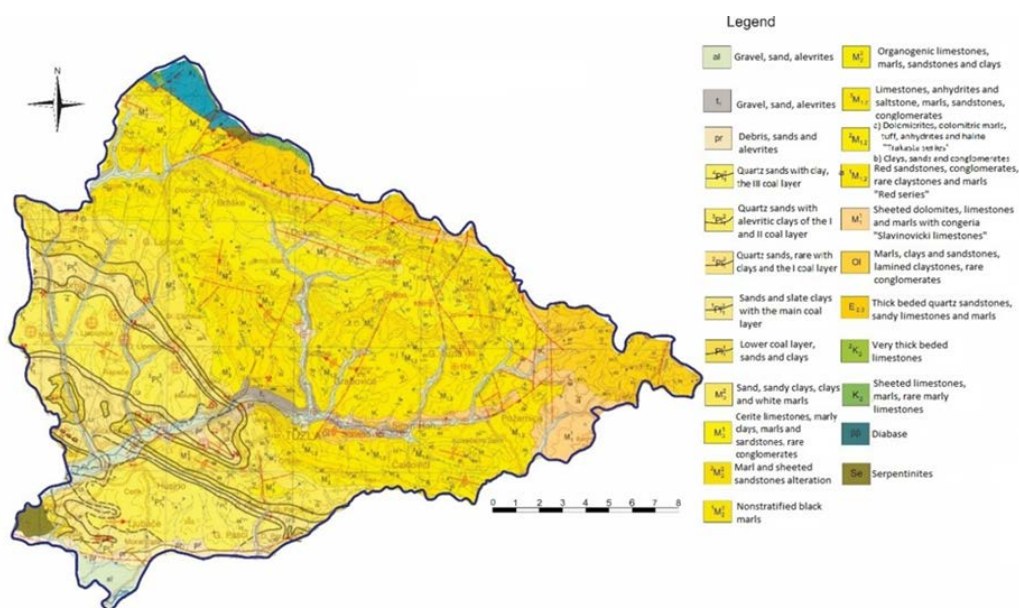


Fig.1. Geological map of Tuzla
Source: Čičić, 1988.

1.1.2 Pedogeographic characteristics

On the pedological map (R - 1: 50 000) of the Tuzla's urban area, there are 25 (automorphic and hydromorphic) soil types (Stjepić Srkalović, 2015.) (Figure 2). The most common types of soil in the researched area are yellowish-brown soils on sands and sandstones, brown degraded soil on clays and loams, brown medium deep and deep soil on limestones, grey-brown carbonate soil, grey-brown deeply soaked soils, pelosols and vertisols. It should be noted that high percentage of these soils are covered with urban infrastructure and which aren't used for agricultural purposes.

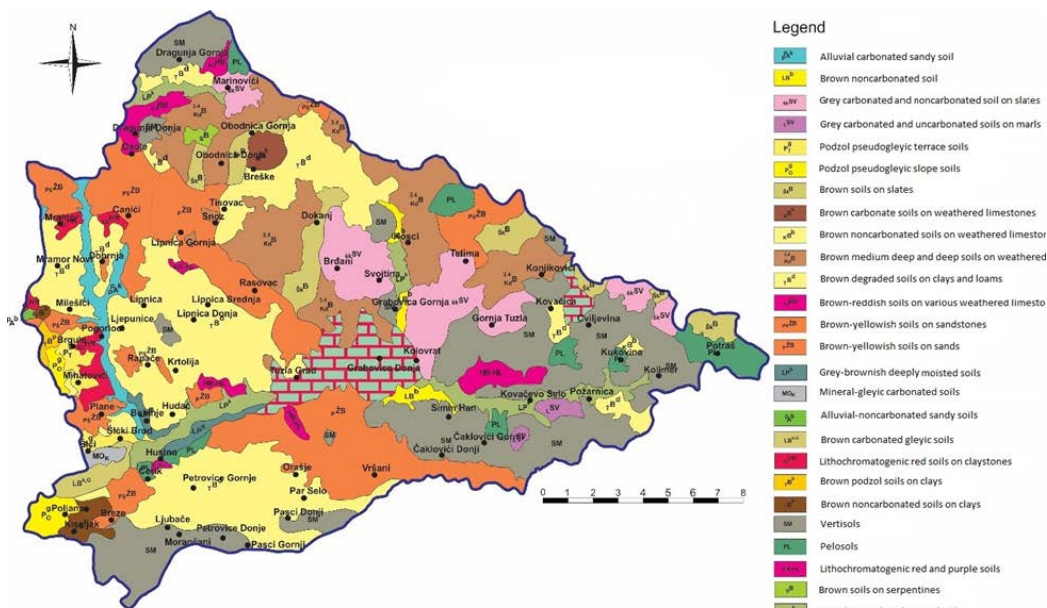


Fig.2. Pedological map of Tuzla
Source: Čičić, 1988.

2. Research methodology

During the research for the paper, various methods were used, such as analysis of the results of previous researches, defining the concept of work and the order of research, terrain researches, preparation of samples for laboratory tests and making thematic maps and tables etc.

Field work included the collection of soil samples (240 samples) from 303 km² (within the city borders of Tuzla). The sampling network in the urban part is 1km x 1km, and in the rural part 1.5km x 1.5km (Figure 3). Samples in the far eastern part of the study area were not collected because the area was mined. Samples marked with "a" after the ordinal number (246a-264a) are control samples collected near schools in the urban part of the city of Tuzla.

The samples were collected from the designated locations by a process of composite sampling. Five soil subsamples were taken and mixed together at each sampling. Samples were taken from a depth of about 30 cm and stored in PVC bags with the specified number, location and coordinates. These composite soil samples, weighing about 0.5 kg each, were dispatched to a laboratory and prepared for chemical analyses. Soil sampling was conducted according to the geochemical expert group (The Urban Geochemistry Project (URGE)) (Ottesen, et al 2008.).

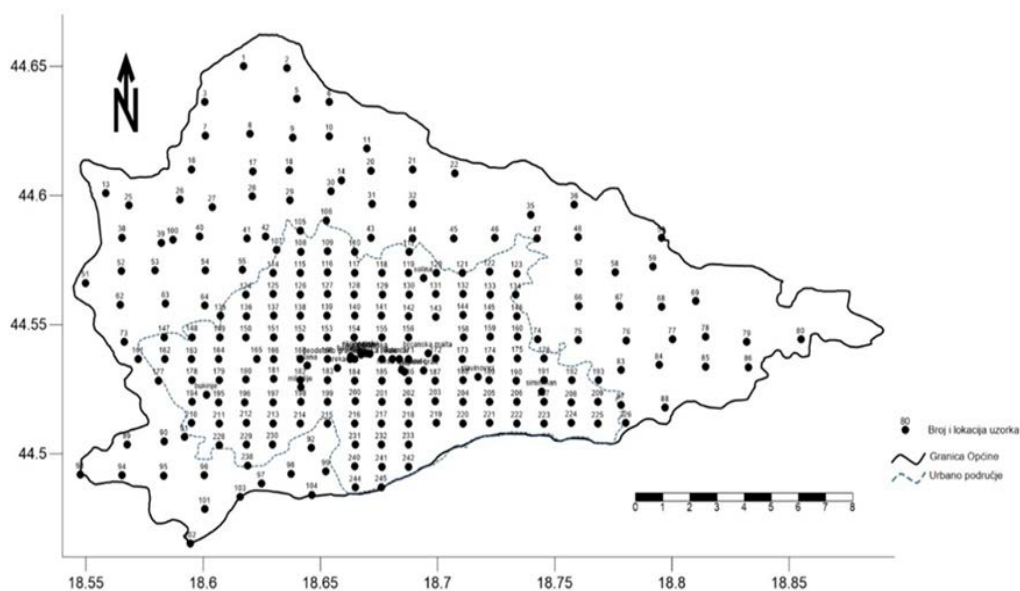


Fig. 3. Sample locations

The preparation of soil samples for laboratory analysis (sowing, drying, grinding, weighing) was carried out on the Faculty of mining, geology and civil engineering at the University of Tuzla. The dried and sieved soil samples were ground in an agate mortar to the fineness of a powder. After that, 12 - 15g were extracted from each sample.

Determination of PTE concentrations and a number of other trace elements (59 elements in total) was performed by the ICP-MS method (Inductively Coupled Plasma - Mass Spectrometry, code MA250) at Bureau Veritas Commodities Canada Ltd. laboratory in Vancouver - Canada (ISO 9002 accreditation). International standards STD OREAS25A-4A and STD OREAS45E were used for calibration. This method can determine a large number of elements with a very low detection limit, because the sensitivity is very high. The detection limit of this method for Ni is 0.1ppm - 10000ppm. Graphical processing of the results was performed in the Golden software Surfer 12 software package.

3. Results and discussion

Nickel is a trace element to a minor element in the iron group with siderophilic, chalcophilic and lithophilic properties. According to its representation in the Earth's crust, it is in 23rd place. The mean nickel content in igneous rocks is 75ppm, in shales about 68ppm, in sandstones 2ppm and carbonates 10ppm. The mean value of nickel in the most common soil types is about 40ppm (Table 1). Soils on ultrabasic rocks may contain concentrations of 100ppm (Halamić, Miko, 2009).

Table 1: Maximum allowable Ni concentrations in soils.

Element	Sandy soil (ppm)	Powder-loamy soil (ppm)	Clayey soil (ppm)
Ni	30	40	50

Source: Ordinance on determining the permitted quantities of harmful and dangerous substances in the soil and methods of their testing. Official Gazette of the Federation of Bosnia and Herzegovina, No. 72/09, Sarajevo.

Table 2: Concentrations of nickel (Ni) in the soil of the city of Tuzla

Sample	Ni (ppm)	Sample	Ni (ppm)	Sample	Ni (ppm)	Sample	Ni (ppm)	Sample	Ni (ppm)	Sample	Ni (ppm)
1	338,9	48	492,8	97	227,4	140	587,8	182	398,7	223	288,3
2	239,5	50	325,7	98	170,9	141	311,5	183	426,5	224	51,1
3	342,8	51	194,3	99	150,7	142	197,6	184	146,8	225	159,2
5	271	52	431,4	100	71,6	143	265,2	185	342,9	226	247
6	398	53	569,3	101	213,2	144	241,2	186	351,2	228	183,3
7	196,8	54	262,6	102	367,1	145	275,9	187	839,2	229	132,3
8	168,6	55	184,8	103	205,8	146	195,9	188	287,8	230	313
9	195,3	57	217,1	104	152,1	147	724,9	189	1005	231	229,6
10	199,5	58	250,2	106	535,5	148	395,6	190	403,7	232	71,1
11	133,3	59	1109,1	107	196,8	149	351	191	305,3	233	40,8
13	132,6	62	345,7	105	599,7	150	183,7	192	197,2	238	136,8
14	133,2	63	159,8	108	174,8	151	168,9	193	315,1	240	71,5
15	125	64	1437,7	109	570,9	152	82,1	194	312,6	241	128,4
16	429,6	66	207,8	110	231,4	153	297,2	195	421,2	242	300,8
17	324,2	67	188,6	112	261	154	266,1	196	145,1	244	229,8
18	245,9	68	191,8	114	218,5	155	451,6	197	190	245	372,8
19	312,5	69	193	115	574,5	156	241,3	198	300,6	246a	240,6
19a	262,8	71	194,2	116	763,5	158	312,1	199	344,1	247a	184,9
20	208	72	70,3	117	225	159	224,5	200	551,3	248a	249,8
21	221,6	73	299,2	118	241,9	160	201,8	201	180,5	249a	998,8
22	179,2	74	264,6	119	199,2	161	169,5	202	434	250a	368,7
25	74,3	75	197,6	120	510,9	162	191,9	203	400,4	251a	253,2
26	264,2	76	120,3	121	469,8	163	699,1	204	272,7	252a	269,5
27	795,8	77	368,9	122	233,9	164	505,1	205	200,9	253a	414
28	467,6	78	103,3	123	199,5	165	88,3	206	277,8	254a	269,6
29	300,9	79	229	124	58	166	238,5	207	208,4	255a	30
30	214,5	80	139	125	113,5	167	426	208	368,4	256a	326,8
31	194,2	83	323,1	126	607,3	168	124,8	209	267,8	257a	327,8
32	251,6	84	775,7	127	452,5	169	123,8	210	239,4	258a	297,7
35	305,5	85	632,7	128	244,6	170	311	211	560,8	259a	357,7
36	216,6	86	190,1	129	274	171	402,8	212	117,8	260a	298,7
38	166,8	87	291	130	249,4	172	453,7	213	164,1	261a	208
39	192,8	88	214,6	131	193,8	173	440,8	214	834,6	262a	274,4
40	130,6	89	391	132	345,4	174	247	215	222,5	263a	220
41	206,4	90	430,7	133	220,8	175	809,8	216	108,5	264a	208,4
42	258,1	91	200,6	134	205,2	176	833,4	217	434,1		
43	312,6	92	133,8	135	250,2	177	77,1	218	268,8	Med.	248,2
44	258,7	93	385,3	136	158,9	178	315,6	219	371,5	Avg.	297,91
45	206	94	264,9	137	75,6	179	571,6	220	402,1		
46	194,4	95	61,6	138	153,8	180	184,4	221	280,9		
47	242,7	96	133,5	139	214,1	181	235,8	222	345,6		

Nickel is not a biogenic element, but it is essential for some organisms. It is highly toxic to plants in concentrations above 50ppm, with the exception of endemic species on serpentinite soils that accumulate nickel. High concentrations of nickel in the soil hinder the growth of plants. It is moderately toxic to mammals. Nickel is known to be an allergen. Phosphate fertilizers increase the availability of nickel, while limestone and potassium fertilizers reduce its availability. Nickel deficiency causes retardation during animal growth. About 70 plant species that accumulate nickel (eg walnuts, cocoa, kale, etc.) are known, while other plants do not accumulate it (eg cereals, potatoes, carrots, etc.) (Halamić, Miko, 2009).

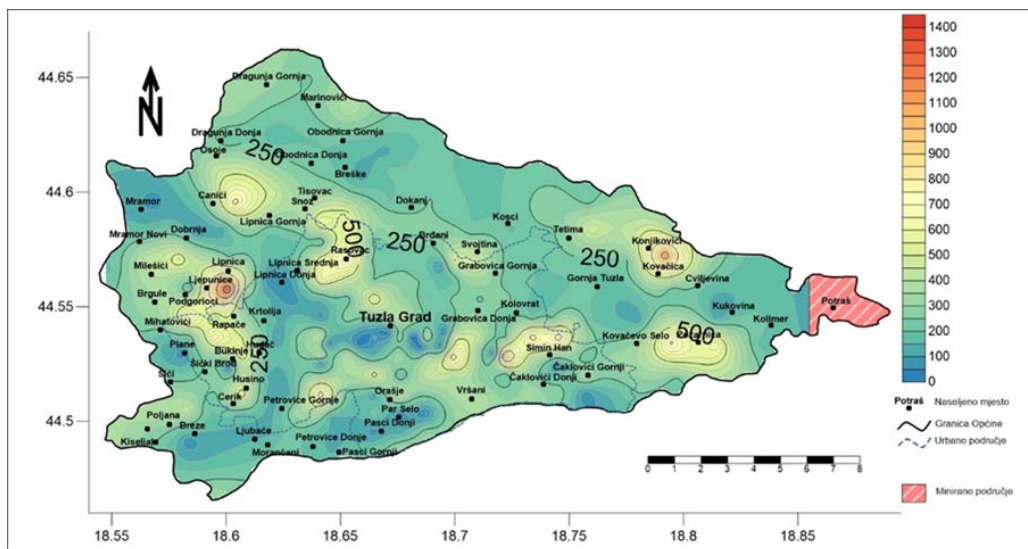


Fig. 4: Concentrations of nickel (Ni) in the soil of the city of Tuzla In ferrous metallurgy, nickel is a steel refiner with a high degree of recycling. Its world production is 8×10^5 t/year. Environmental pollution with this metal occurs through industrial dust, waste and wastewater (Halamić, Miko, 2009).

Nickel is mostly used in the preparation of alloys. Nickel alloys are characterized by strength, ductility and resistance to corrosion and heat. About 65% of nickel, which is consumed in the Western World is used to make stainless steel, whose composition can vary, but usually the iron with 18% chromium and 8% nickel. 12% of all nickel produced goes into super alloys. The remaining 23% of consumption is divided between alloy steels, rechargeable batteries, catalysts and other chemicals, coins, casting products and formwork (Lenntech.com., 2016. Periodic table - chart of all chemical elements.).

According to the Ordinance on determining the permitted amounts of harmful and dangerous substances in the soil and methods of their testing, the maximum permissible nickel concentrations in soils vary from 30ppm for sandy soils, 40ppm for dusty-loam soils and 50ppm for heavy clay soils (Table 1). The average nickel concentration for the study area is 297.91ppm, and the median is 248.2ppm, which indicates a huge increase in nickel concentrations above the allowable values in the soil of the study area (Table 2).

Nickel concentrations in the soil of the city of Tuzla (Table 2) range from 30ppm (minimum value determined in sample 255a) in the yard of the Secondary School of Economics and Trade, to a maximum of 1437.7ppm in sample number 64 in the area of Lipnica. Out of 240 total analyzed soil samples, nickel concentrations were increased to 238, in some places several tens of times the allowed limit. The highest concentrations were recorded in samples 64 (1437.7ppm) in Lipnica, 59 (1109.1ppm) in the area of Požarnica - Kovačica and in sample 189 (1005ppm) in the area of Slavinovići. At twenty locations, nickel concentrations vary from 500-1000ppm. Igneous rocks are the primary source of Ni found in soils. The total Ni concentration in soils is directly related to the concentration in the parent material and depends on erosion. Ni losses from the soil occur due to dissolution, leaching and run-off water during erosion processes. The increase in Ni in the soil occurs naturally through the accumulation of eroded material or as a consequence of the application of fertilizers on agricultural land (Chauhan et al, 2008). The investigated area is dominated by Lower Miocene organogenic carbonates, Miocene organogenic limestones, clays, marly clays, sands and subordinate conglomerates, and Quaternary formations developed along the stream bed in the form of proluvial sediments (crumbs) and as precipitated terrace and alluvial sediment. (Stjepić Srkalović et al, 2018). No igneous rocks have been determined in the investigated area, and therefore their erosion is not possible. Since this is a hilly area, intensive agriculture that would require the application of certain fertilizers is not present. Agriculture is represented on smaller plots, most often in the form of greenhouse production, which is not enough to contaminate the soil in the entire area of the city of Tuzla with this element. Organic matter has the ability to absorb metals, and coal or oil can contain significant amounts of nickel. Power plants and garbage incinerators release nickel into the air, which binds with moisture in the air and reaches the ground through raindrops. It takes a long time to clean the nickel contaminated air. Nickel can also end up in surface water when it is part of wastewater flows. Most of the nickel compounds released into the environment will be absorbed into the soil and as a result become immobile. However, in acidic soils, nickel will become more mobile and will often leach into groundwater (Lenntech.com., 2016. Periodic table - chart of all chemical elements.). Coal has been exploited in the area of the city of Tuzla for decades, which resulted in the construction of the Tuzla Thermal Power Plant in 1959. Also the impact of other thermal power plants in relative proximity, such as TPP Ugljevik, Stanari and Obrenovac shouldn't be neglected. All these pollutants contributed to environmental pollution in the study area.

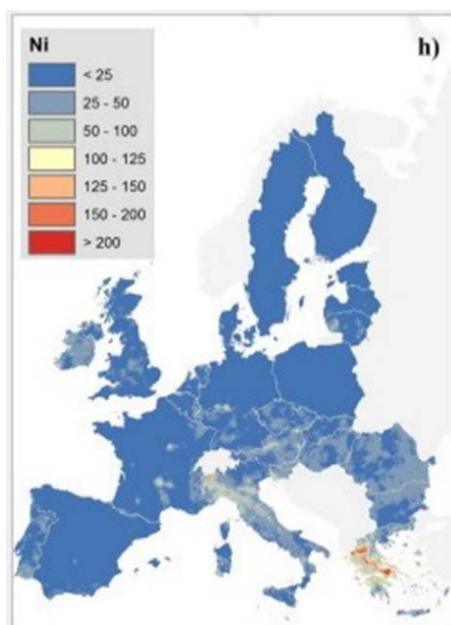


Fig. 5: Ni concentrations in European soils

Source: Toth, et al., 2016.

The last glaciation affected the geographical redistribution of nickel in the surface layer of the soil. Northern of 55° latitude the Ni concentration is generally low, although in the Baltic States it has been found that about 5% of the samples have a Ni concentration above 100ppm. This share is comparable to the share across the EU where over 95% of the area contains <100ppm nickel. It should be mentioned that, according to the MEF (2007.)¹, this is a guideline for environmental risk assessment. The areas with the highest concentrations of this element (Map 5) are located in the Piedmont region in northwestern Italy and northern Greece, including the northern part of the Peloponnese (Toth, et al, 2016).

Nickel intake in the body can increase after ingesting large amounts of vegetables from contaminated soil, especially since plants accumulate it. Smokers ingest higher amounts of nickel through the respiratory system. Nickel exposure can be through air, water, food or cigarettes. The intake of this trace element in the body can also take place through the skin, i.e. in contact with the skin and pollutants. Nickel in small quantities in the body does not pose a danger to human health.

Intake of excessive amounts of nickel has the following consequences: higher chances of developing lung, nose, larynx and prostate cancer, lung embolism, respiratory failure, congenital defects, asthma and chronic bronchitis, allergic reactions such as skin rashes, heart disorders, etc. (Lenntech.com., 2016. Periodic table - chart of all chemical elements.).

Nickel and certain nickel compounds are listed in the National Toxicology Program (NTP) as reasonably expected carcinogens. The International Agency for Research on Cancer (IARC) listed compounds of nickel within group 1 (there is sufficient evidence

¹ MEF (2007) - Ministry of the Environment, Finland, Government Decree on the Assessment of Soil Contamination and Remediation Needs, 214/2007 (March 1, 2007).

of carcinogenicity to humans) and nickel within group 2B (agents that are likely to be carcinogenic to humans). OSHA² does not regulate nickel as a carcinogen. According to the ACGIH³, nickel has been confirmed as category A1, i.e as a human carcinogen. There is not much available data on the effects of nickel on plant and animal organisms. We know that high concentrations of nickel on sandy soils can significantly harm plants, and high concentrations of nickel in surface waters can reduce algae growth rates. Slow growth has also been observed in microorganisms due to the presence of nickel, but they usually develop resistance to nickel after some time (Lenntech.com., 2016. Periodic table - chart of all chemical elements.).

4. Conclusion

Nickel concentrations in the soil of the city of Tuzla (Table 2) range from 30ppm (minimum value determined in sample 255a), to a maximum of 1437.7ppm in sample number 64. Out of 240 total analyzed soil samples, nickel concentrations were exceeded in 99.16% (n=238) of samples, in some places several tens of times the allowed limit. The highest concentrations were recorded in samples 64 (1437.7ppm) in Lipnica, 59 (1109.1ppm) in the area of Požarnica - Kovačica and in sample 189 (1005ppm) in the area of Slavinovići. At twenty locations, nickel concentrations vary from 500-1000ppm.

The average concentration of nickel for the study area is 297.91ppm, and the median 248.2ppm, which indicates a huge increase in nickel concentrations above the allowable values in the soil of the study area.

The total Ni concentration in soils is directly related to the concentration in the parent material and depends on erosion. The increase of Ni in the soil occurs naturally through the accumulation of eroded material or as a consequence of the application of fertilizers on agricultural land.

Considering the geological structure and geomorphological characteristics of the investigated area, it can be concluded that high concentrations of nickel in soils are not a consequence of natural processes, as well as intensive agricultural production. Power plants and garbage incinerators release nickel into the air, which binds with moisture in the air and reaches the ground through raindrops. It takes a long time for the air to be cleaned of nickel. Nickel can also end up in surface waters when part of wastewater flows. Most of the nickel compounds released into the environment will be absorbed by the soil and as a result become immobile. The researched area has been an industrial center for many years, and the thermal power plant in Tuzla, as well as those in the relative vicinity of this area, have greatly contributed to environmental pollution.

Given that it is a carcinogenic element, even more detailed research should be conducted on the concentrations of nickel in soil, water, plants and air, with the aim of reducing carcinogenic diseases in Tuzla. Pedoecological research is of crucial importance given that about 95% of the food needed by humans is produced directly or indirectly on and in the soil, and all pollutants in the soil eventually ends up in the human body. It should be noted that there are other ways to intake harmful substances from the soil, such as: by cultivating agricultural land, playing in parks or school playgrounds, using water from inadequate wells, etc.

² OSHA - Occupational Safety and Health Administration

³ ACGIH – American Conference of Governmental Industrial Hygienists

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Summary

The main goal of the research was to determine the extent to which the soil of the city of Tuzla is contaminated with nickel and to determine the origin of pollutants. The 240 soil samples were collected during the terrain research, covering an area of 303 km². Laboratory analysis was performed at Bureau Veritas Commodities Canada Ltd. laboratory in Vancouver - Canada, by the method of ICP-MS (Inductively Coupled Plasma - Mass Spectrometry), code MA 250. The nickel concentration testing in soil samples was performed by mass spectrometry (ICP-MS) with a detection range of 0.1 - 10000ppm. Exceeding the maximum allowable concentrations of nickel, defined by the Ordinance on determining the allowable amounts of harmful and dangerous substances in the soil and methods of their testing was recorded in 99.16% of samples.

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Power plants and garbage incinerators release nickel into the air, which binds with moisture in the air and reaches the ground through raindrops.

Most of the nickel compounds released into the environment will be absorbed by the soil and as a result become immobile. The researched area has been an industrial center for many years, and the thermal power plant in Tuzla, as well as those in the relative vicinity of this area, have greatly contributed to environmental pollution. Given that it is a carcinogenic element, even more detailed research should be conducted on the concentrations of nickel in soil, water, plants and air, with the aim of reducing carcinogenic diseases in Tuzla.