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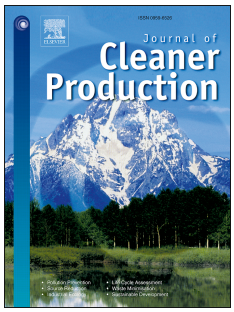
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Alexandra I. Belyanovskaya: Conceptualization, Data curation, Writing - original draft, Formal analysis.

Daria Vorobeva Writing - original draft, Formal analysis.

Bertrand Laratte: Conceptualization, Supervision, Methodology.

Natalia Guseva: Conceptualization, Supervision.

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Abstract

The chemical composition of soils reflects the degree of industrial exposure. Cu and Ni concentrations in soils of the «Severonickel» plant vicinity are higher than remote ones. In the impact area, the mean concentration of the heavy metals in the subsoils is 55 [ppm] for Ni and 33 [ppm] for Cu. Soils' chemical composition varies over different mineral horizons. The subsoil layer is the main accumulator of chemical elements, including pollutants. Erosion of the surface layer and technogenic disturbance of the soil profile can lead to subsoil spreading and a significant increase in the human health Impact Score. At the same time, the default IS calculation is focused on the 0.1 [m] depth for all zones. In the paper two factors are considered in the impact modeling modification. The human health Impact Score is calculated specifically for each genetic soil layer with the respective depth of the profile (from 0.05 [m] to 0.2 [m]) and for soils from background and impact areas. The discrepancies between default and modified Impact Scores are observed. In the Impact area, the highest IS_{hum} for Ni is 60, for Cu is 5.6 [DALY]; in the Background area, it is 11 and 3.1 [DALY] respectively. The importance of using the regionally modified values in population health impact monitoring is highlighted.

Keywords

Arctic soils, impact assessment, characteristic factors, USEtox, Kola region, copper, nickel, regional LCIA

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Introduction

The Arctic zone is characterized by extreme natural and climatic conditions, the focal nature of industrial and economic exploration of the territory, low population density, and the vulnerability of nature and public health from industrial activity. The Kola Peninsula is Russian Arctic territory where the wealth of mineral resources has become the basis for the growth of mining and metallurgical industries. At the same time, northern ecosystems are highly sensitive to the intake of technogenic origin chemical elements. Many metal ions are potentially toxic, so it is important to understand their transport and distribution in the environment and the routes through which they may be transferred to humans (Spurgeon et al., 2011). Nickel (Ni) and copper (Cu). Ni and Cu are chosen for the research because they are the main pollutants in the North-West of Russia (Barsova et al., 2019; Kyllönen et al., 2020; Moiseenko et al., 2006; Sandimirov, 2020). Exposure to heavy metals can cause a lot of health problems in humans. Long-term exposure is linked to several neurological disorders (Alquezar et al., 2020; Chin-Chan et al., 2015). Therefore, the question of assessing the impact of pollutants on individual elements of ecosystems arises, air, water, soil, including on humans and their health.

Current article is focused on the soil pollution. Soil is a biogeochemical barrier for chemical elements input to ecosystems from the polluted atmosphere. Soil is a depositing medium that accumulates chemical elements. This property becomes especially important under conditions of long-term anthropogenic load (Barsova et al., 2019). Soil is a critical repository for numerous deleterious pollutants, thereby serving as a good matrix for assessing the status quo of environmental pollution (Doyi et al., 2018). Soils degradation leads to a decrease in their sorption capacity and, consequently, to groundwater contamination with heavy metals (Shumilova and Petrov, 2016). Heavy metal pollution affects the soil's ecological function (Li et al., 2021; Yuan et al., 2021). Heavy metals emanate from a myriad of sources in the environment, including industrial wastes, and particularly in areas of the intersection of enterprises (Wuana and Okieimen, 2011; Zhang et al., 2020). Recently a lot of research has been devoted to human health impact assessment of heavy metals in mediums such

as soils, sediment, or dust. Because of their persistence and non-biodegradability, heavy metals can easily accumulate in the environment and could be transferred to plants, living organisms, and groundwaters (Alloway, 2013).

When assessing the pollution of natural environments, the gross value of chemical elements in the components of the natural environment is the most frequently determined indicator (F. Bratec et al., 2019; Perminova, 2017; Sharma et al., 2021). Nevertheless, it does not allow a comparative impact assessment between different chemical elements or components of the natural environment. For this reason, many authors now use modern methods and models of the impact and environment geoecological state assessment based on the calculation of the various indexes (Liang et al., 2015; Makarova et al., 2018; Zhang et al., 2020).

In this study, we consider assessing of the human toxicity with calculation of the life cycle impact assessment Impact Score (IS) calculation as the approach to quantify the human health impact (European Commission -- Joint Research Centre -- Institute for Environment and Sustainability, 2010). The Impact score is calculated using the total mass of the pollutant and the substance specific characterization factor. The characterization factors calculation is taken from the USEtox model.

USEtox is a scientific consensus model for comparative assessment of toxics of goods and services (Wu and Su, 2020). USEtox is model endorsed by UNEP's Life Cycle Initiative for characterizing human and ecotoxicological impacts of chemicals (Hauschild et al., 2008; Rosenbaum et al., 2008). The USEtox is specifically focused on the human health and the ecosystems impact assessment (Nordborg et al., 2017).

However, there are a lot of uncertainties connected with the LCIA models, and the USEtox in particular:

1. The inventory of metals in the model dataset needed to be developed. In the USEtox inorganics are all specified as ‘indicative’, reflecting the relatively high uncertainty associated with estimates of fate, exposure and effects for this substance group (Fantke et al., 2017). The CFs for metals in the USEtox are classified as interim (Pizzol et al., 2011). Variations in transport properties

for inorganic substances depend in complex ways on a range of media properties. However, there are not local geological information in the model. In the article we propose to reduce this limitation with the analytical results of Ni and Cu concentration in soils of Kola peninsula.

2. In the USEtox model only the concentration of the pollutant in the surface layer is considered for the Impact Score calculation (Fantke et al., 2017). However, the surface soil layer can be disturbed, or the natural genetic layer can be changed (Elsukova et al., 2019; Kashulina et al., 2014, 2010). The default IS calculation is focused on the 0.1 [m] depth for all zones. We extend the model dataset leaning on media properties as different soils profile horizons (from 0.05 [m] to 0.2 [m]).

Considering all limitations, we can propose following hypothesis, it is necessary to consider local features of the landscape: natural differentiation and technogenic transformation of the soil profile. It allows adjusting the result of the impact assessment on the living organism and adapting it for regional conditions (Belyanovskaya et al., 2022, 2019, 2020).

To investigate this hypothesis, we compare soil layers' potential toxicity for human health by calculation of the Impact score. The Impact score is calculated both in the area of influence and in the background area. It emphasizes the combined influence of natural and anthropogenic factors, which demonstrates how in small, contiguous territories there can be a sharp change in IS as a result of the variability of lithogeochemical conditions and the presence of industrial facilities. This approach allows the IS values to be taken as another factor for identifying the zone of impact of industrial enterprises from the perspective of human health impact assessment. To compare the level of impact the background area was chosen as well.

The background zone is remoted from the emission source. Studies of soil chemistry in this area revealed minimal concentrations of nickel and copper (Guseva N.V. et al., 2020; Vorobeva D. and Guseva N.V., 2021). It was difficult to locate the background zone farther because of the variable lithogeochemical background and the presence of other large industrial facilities. This approach is in accordance with studies by other authors (Opekunova et al., 2006).

Modification of the Impact score calculation allows us to characterize the degree of impact on human health (IS_{hum}), combining different methods of assessing the ecological condition of urban areas: geological approach and impact assessment, which reflect the novelty of the research. On the one hand, studies of the geological characteristics of areas exposed to industrial impact reflect the changes occurring in the natural environment due to technogenesis. On the other hand, the application of a wealth of geological data allows for the expansion of the database of impact assessment models and a more complete assessment of the quality of the natural environment. The application of an interdisciplinary approach is based on the principles of ecogeochemistry, human health, and ecosystem impact assessment.

1. Study area

1.1 Natural conditions

The Lake Imandra catchment is the central part of the Murmansk region (Figure 1-2). Murmansk region is a constituent entity of the Russian Federation with a total area of 144.900 thousand km^2, located in the north-west of Russia. Sampling points refer to three administrative units of the Murmansk region: the Apatity, Kirovsk, Mochegorsk catchment areas.

Almost the entire territory lies north of the Polar Circle and is located on the Kola Peninsula. The climate is a temperate cold climate, with average annual precipitation and an annual temperature of 553 mm and -0.8◦C, respectively. The prevailing wind direction is from north to south in summer and from south to north in winter.

The eastern part area is occupied by the alkaline Khibiny Massif, and the western part of the territory is a hilly plain with a section of industry-disturbed landscape. The geological structure here is represented by basic and ultrabasic rocks, overlapped by moraine lake-glacial and fluvioglacial

deposits. The study area as a zone of ores extraction is investigated; the mineralographic map is following (Figure 1).

Figure 1. Mineralographic map of the research area, 1:200000

Compiled on the basis of materials from GIS-Atlas «Subsoil of Russia» as of 01.09.2019.

According to the mineralographic map of the area there are natural depositions of Ni, Cu, Cr, and Pt in the area of smelter vicinity (Monchegorsk city). The impact and the background areas are placed in the boundaries of the established nonferrous metals depositions. There are non-metal minerals resources around Kirovsk city.

The geological and climatic features of the region determine the soil characteristics. Four types of soils have developed on the Kola Peninsula: tundra, podzol, boggy, to a small extent soddy and derived from them. Most typical of the Kola Peninsula is podzol soil type. The mechanical composition is dominated by sandy and sandy loam soils, largely heaped (Elkshina and Kupriyanova, 1970).

1.2 Industrial influence

Landscapes of the Kola Peninsula have been already investigated due to the high level of anthropogenic tension (Kashulina, 2017; Revich, 2020). This impact leads to massive air, soil, and water pollution (Bazova, 2013; Dauval'ter and Kashulin, 2015).

The presented results allow us to evaluate the impact of the copper-nickel plant on the chemical composition of the components of the natural environment through the polluted atmosphere. Emissions in 2016 were Ni 245 t/yr Cu 462 t/yr (Barkan and Lyanguzova, 2018). In the zone of impact of copper-nickel factory emissions, atmospheric precipitation contains significant amounts of

pollutants: Ni 262 µg/l, Cu 540 µg/l (Evtyugina and Asming V., 2013). In surface waters (Ni 46 µg/l, Cu 17 µg/l) concentrations of these elements are significantly higher than in groundwater (Ni 4.9 µg/l, Cu 0.56 µg/l) (Evtyugina et al., 2016). When atmospheric precipitation infiltrates through the soil, most of the heavy metals are bound in the soil horizons, where they accumulate (Ni 54.99 ppm, Cu 33.26 ppm) (Table 7, Annex). The soil is thus a natural buffer that protects groundwater from aerotechnogenic nickel and copper intrusion. Therefore, the stage of infiltration of atmospheric precipitation through the soil plays a significant role in the chemical composition of groundwater, and soils themselves.

There are extremely high concentrations of Ni and Cu in the organogenic soil horizons and the medium peat layers (Kashulina, 2017). According to the investigation of (Koptsik et al., 2015) soils of technogenic wastelands formed under the influence of long-term emissions of Severonickel Combine are acidic, depleted in nutrients, polluted with heavy metals, and badly eroded Natural illuvial humic podzols have lost their upper organogenic and podzolic horizons as a result of erosion and removal of protective vegetation cover. Atmospheric air pollution with SO_4, disruption of vegetation cover and changes in the morphological composition of soils increases their ability to accumulate heavy metals (Kashulina et al., 2014, 2010). Accumulation of Ni and Cu in soils correlates with winter fallouts (Kashulina, 2017).

The concentration and the composition of the pollutants in soils correlate with the emission from local industry (Barkan and Lyanguzova, 2018). Smelters are one of the largest sources of environmental pollution (Opekunova, 2016, 2004; Opekunova et al., 2006). The study is restricted to one of the major industrial enterprises of the Murmansk region. The copper-nickel smelting plant «Severonickel» (The Kola Mining and Metallurgical Company) built in the 1930s is located in Monchegorsk at an altitude of about 123 m. Currently, copper-nickel matte is processed at this site.

The reconstruction of the production in Monchegorsk resulted in a 56.6% reduction in nickel emissions from 2013 (Revich, 2020). Emissions from The Kola Mining and Metallurgical Company («Severonickel» in Monchegorsk and «Pechenganickel» Nickel-Zapolayrniy) decreased from 1627.1

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tons in 1998 to 348.9 in 2017 for Ni, and from 1062.2 to 486.9 tons for Cu over the same period. A certain indicator of atmospheric air pollution by metals is their content in the surface soil layer (Ettler, 2015a; Evdokimova et al., 2011; Evseev and Krasovskaya, 2017). The content of copper and nickel in Monchegorsk soil exceeds MAC up to 5 times (Opekunova et al., 2006).

Mixed environmental conditions (natural anomalies and anthropogenic impact) generate the need to assess the impact of the environment on public health.

2. Materials and methods

2.1 Methods of soil sampling

This research paper is focused on Ni and Cu distribution and the impact assessment in soils of the Kola Peninsula, taking into account their concentration in impact and background areas (Figure 2). Impact areas are normally more polluted than remote areas (Ettler, 2015). The background zone was taken 30-50 km from the emission source while observing a decrease in the concentration of the main pollutants (Ni and Cu).

According to the prevailing wind direction, 13 soil sections (0-60 cm) were sampled by layer during the expedition in July 2016 (total number of samples 28). The sampling locations are shown in Figure 2 and Table 2 in the Annex.

Figure 2. Sampling locations (A.) and study area (B.), industry-disturbed landscape (C.) and main wind direction of Monchegorsk city (D.)

To assess the accumulation of heavy metals in different types of soils, the structure of their vertical profile (from the surface of the soil down to the parent rock) is of particular importance. As a rule, the soil profile consists of several layers, called genetic horizons, as they were formed as a result of

soil-forming processes caused by the genesis of the soil (National Atlas of Soils of the Russian Federation [Nacional'nyj atlas pochv Rossijskoj Federacii], 2011).

All the samples we studied were taken at points belonging to the podzol soil type formed in cold areas with a good leaching regime. On the Kola Peninsula, podzols with a low thickness of 15-50 cm are typical. Podzols with higher thicknesses (layer of more than 50 cm) are not widespread (Elkshina and Kupriyanova, 1970).

Podzols are characterized by a differentiated profile (Figure 3) consisting of litter 3–8 cm thick; whitish, lightened due to the removal of coloring iron compounds, and humus of the podzolic or eluvial horizon (topsoil) with a thickness of 2 to 20-30 cm; illuvial horizon (subsoil) of brown or ocher tones, formed as a result of the illuvial accumulation of aluminum-ferruginous-humus complex compounds, gradually turning into the parent rock (substratum) (National Atlas of Soils of the Russian Federation [Nacional'nyj atlas pochv Rossijskoj Federacii], 2011).

Figure 3. Genetic horizon of podzol

Each soil layer plays a particular role in the accumulation processes of pollutants. In the current research, we turn attention in particular to the subsoil. The smaller number of topsoil samples is due to the eroded upper horizons at some sites, and the smaller number of substratum samples is due to the highly cluttered area and the inability to deepen the soil section (Table 1). Thus, the majority of samples are taken from the depth of 0.06-0.2 m – the subsoil. This depth corresponds to the illuvial horizon according to the Russian classification of soils (National Atlas of Soils of the Russian Federation [Nacional'nyj atlas pochv Rossijskoj Federacii], 2011). The subsoil is the main mineral horizon that accumulates inorganic components, coming from the overlying layers (Adams and Moore, 1983).

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Table 1. Soil horizons collected for the investigation

N	Soil layer	Depth [m]	Quantity of samples
1	Topsoil	0.06-0.08	6
2	Subsoil	0.06-0.2	16
3	Substratum	0.2-0.3	5

Sampling was carried out taking into account the local features of the structure of the soil profile: by in accordance with the genetic horizons, but not evenly spaced in depth. Some samples taken at the same depth belong to different genetic horizons (Annex, Table 3).

Soils intended for the determination of heavy metals are sampled with tools that do not contain metals. Soil samples are packed in containers ensuring the preservation of small soil particles (bags made of synthetic film, dense fabric, or water-resistant paper).

2.2. Sample preparation and analysis

Chemical analysis and sample preparation was conducted in the Research Laboratory for Hydrogeochemistry (Tomsk Polytechnic University, Tomsk, Russia).

Sample preparation. Soil samples were naturally air-dried in the room prevented chemical pollution. Debris was removed, and they were then sieved through a sieve (mesh size 1 mm) for later use.

To determine the total metal concentration, a soil sample of 2-3 g was weighed, placed into a PP digestion vessel, and then concentrated HNO_3 of high purity at a ratio of 1:10. Sample decomposition is carried out in a microwave oven with a blank sample at 110-115°C in 5-7 steps for 2 minutes. Then the solution is diluted with 20% HNO_3.

Then chemical elements were measured by an inductively coupled plasma mass spectrometer (ICP-MS method) NexION 300D (PerkinElmer, USA).

Two heavy metals (Ni and Cu) were chosen for the research. These chemicals have similar toxic properties, and they are the main pollutants of the area (Motuzova et al., 2004).

Nickel and its compounds s are toxic and carcinogenic (Rabinovitch and Rizhova, 2016). The most common types of health impairment caused by Ni are respiratory, skin and cardiovascular diseases (Chashschin et al., 1994). The increased nickel content in soils leads to endemic diseases: ugly forms appear in plants and in animals; eye diseases are associated with the accumulation of nickel in the cornea (McIlveen and Negusanti, 1994). The clark of nickel in World soils by Vinogradov is 40 mg/kg (Dobrovol'skij V.V., 2003). More recent clark estimates coincide with this value (Butovsky, 2005). In U.S. soils, Ni clark is much lower than world clark - 17 mg/kg (Dobrovol'skij V.V., 2003). Nickel enters the soil as a result of fuel combustion and industrial emissions. In soils Ni accumulates strongly in agrodern-podzolic gley soil (Vodyanitsky, 2008).

Copper is the third most used metal in the world ("VCI, Copper history/Future, Van Commodities Inc.," 2011). This metal has dual nature: essential at an optimum level, while toxic at high levels (Ameh and Sayes, 2019). Cu expresses organophilic properties (McBride, 1989). Cu clark in U.S. soils coincides with world clark and is equal to 20 mg/kg (Dobrovol'skij V.V., 2003). The main source of copper in the soil is copper-nickel smelters (Ilyin, 1991).

3. Data processing technique

3.1. Descriptive statistics

Descriptive data analysis, including mean, standard deviation (SD), minimum and maximum concentrations, variation coefficient, etc., was carried out. Together with SD, variation coefficient (VC), which is SD/mean, was used to reflect the degree of discrete distribution of different metal element concentrations, and to indicate indirectly the activeness of the selected element in the examined environment. Histograms were also utilized to show the distributions of the metals (Annex, Figure 1).

The accumulation of Ni and Cu has a similar nature in the research areas, thus for the rank correlation analysis, the whole sampling was used. To calculate the correlation between Ni, Cu, and other heavy metals, the whole sampling variety was taken.

3.2 The Impact assessment and Characteristic factors calculation with the USEtox model

3.2.1 General framework

The basics of the USEtox model developed on the Microsoft Excel platform (Figure 1, Annex). According to the USEtox model, Impact score (IS, Formula 2) - the LCIA impact score for human toxicity is expressed at midpoint level as cancer or non-cancer disease cases and endpoint level as several of disability-adjusted life years [DALY] (Fantke et al., 2017). The LCIA impact score for potential impacts of the chosen elements (Cu, Ni) is calculated using a weighted summation of the released pollutants – Ni and Cu in soils (the mass – M, [kg], Formula 2) and substance specific characterization factors (CF) obtained with the USEtox model (Formula 1).

$$IS = CF \times M_{x,i}$$

Formula 1. The Impact Score calculation (Fantke et al., 2017)

Note: CF – characterization factor, M - the total mass of the element

Characterization factors for the potential human health damage [DALY/kg] of Ni (II) and Cu (II) for the geo-zone «Central Asia» is taken from the USEtox model (Fantke et al., 2017) (Annex, Table 1). The total mass of the element in soils (M) (Formula 2) is calculated according to the formula developed by T. Bratec (F. Bratec et al., 2019; T. Bratec et al., 2019; Perminovaet al., 2017). The total mass of the pollutant is derived from the concentration of the chemical element in soils ($C_{x,i}$) in each studied area. This data is obtained analytically with the ICP-MS– geological data.

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$$M = \frac{C_{x,i} \times V_s \times p_x}{10^6} [kg]$$

Formula 2. M - total mass of the element calculation (F. Bratec et al., 2019; T. Bratec et al., 2019; Perminova et al., 2017)

Note: $C_{x,i}$ – metals concentration in soils; V_s - soils volume; 4) p_s - bulk density of soils [kg_i/m_i^3].

3.2.2 Impact score calculation modifications

In the current paper, we present the approach to modify the Impact score calculation. The modification leads to reduce the lack of geological data limitation of the USEtox method. In the USEtox only the topsoils (0.1 [m]) are considered in the CF calculation. In the research paper, we modify the total soils volume calculation with different heights of the soil layer (Formula 3).

$$V_i = h_i \times S_i$$

Formula 3. The volume of soils calculation

Note: h_i – the height of soils, [m]; S_i – the square footage of the research area, [m^2]

Physical parameters of the soils are taken into account. For each studied area, the total volume of soils is considered (Formula 3, V_s [m^3]). The volume of soils here represents square footage (S_i) of the studied region and the height of the soil's horizon (h_i). The bulk density of soils (p_i), which is the table value taken from the USEtox documentation [kg_{soil}/m^3_{soil}], and V_s is the volume of soils of each considered region [m^3].

The areas under research (background and impact areas) belong to 3 administrative units of the Murmansk region: Kirovsk, Apatity, Monchegorsk catchment areas. Different sizes of the regions are taken into account with the square footage parameter (S_i) (Annex, Table 2).

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4. Results and discussions

4.1 Statistical analysis results

Analysis of the chemical composition of the mineral horizons of the Kola Peninsula soils shows that the subsoil horizon accumulates the majority of the estimated pollutants (heavy metals, radioactive, and REEs) (Annex, Table 4) in all sampling points. A large number of these pollutants have high concentration coefficients (CC <2;3;5). Maximum CC is also noted in subsoils. The main components of emissions to the environment (Ni, Cu) (Opekunova et al., 2006) are characterized by a high concentration in all the considered mineral horizons of soils, both in the control and in the zone of influence. On the leeward side 5 km from the mill, there is a decrease in Ni and Cu concentrations (Figure 4-5), these elements accumulate the most.

Figure 4. The Ni concentrations in the Background (B) and in the Impact (I) areas, in different distance from the source of the pollution

Note: Clark - S.R. Taylor, S.M. McLennan, 1985 – average concentration of Ni and Cu in the upper continental crust (Taylor and McLennan, 1985)

N – number of sampling point, Distance [km] – distance from the source of pollution

Figure 5. The Cu concentrations in the Background (B) and in the Impact (I) areas, in different distance from the source of the pollution

Note: Clark - S.R. Taylor, S.M. McLennan, 1985 – average concentration of Ni and Cu in the upper continental crust (Taylor and McLennan, 1985)

N – number of sampling point, Distance [km] – distance from the source of pollution

Ni and Cu in this case or represent the most stable components of emissions or they are accumulated due to natural factors.

Minimal concentrations of Ni and Cu (CC<1) are noted in soils of the background area, while other heavy metals, radioactive and rare-earth elements in these samples have CC>1. The wide spectrum of element accumulation in soils of the Kola Peninsula in points distant directly from the production site is explained by its mixed geoecological situation. Data analysis demonstrates the difficult ecological and geochemical situation in the study area. Opekunova et al. (Opekunova et al., 2006) also revealed the content of a group of heavy metals in soils in the vicinity of Monchegorsk. Since this work is devoted to the accumulation of heavy metals, further attention in the work is focused on this group of chemical elements.

The wide range of accumulation of heavy metals in the soils of the Kola Peninsula is also characterized by the presence of significant correlations between them. Heavy metals in general and Ni and Cu as the main industrial pollutants of the research area have close chemical properties and correlate in the soil (Figure 6). There is the strongest positive correlation between Cu and Ni (Spearman rank correlation coefficient $p_{0.05}$=0.9), (Annex, Figure 2). Heavy metals (Cr, Cd, Co) have significant positive correlations with Cu and Ni. These elements according to factor loading analysis are under the influence of one factor (Annex, Figure 3). Due to the absence of the other deposition, but Cu and Ni in the area, the main factor affecting the whole group of the heavy metals is probably industrial.

In the following diagram (Figure 6), we observe the graphical expression of the Ni-Cu correlation. The strong positive correlation of these metals can be explained by the same source of accumulation as well as their chemical mechanisms. There is probably a mix of the industrial specificities of the area supports and the natural sources.

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Figure 6. Cu - Ni correlation diagram, polynomial trend line ($n_{samples}$=17), R^2 - reliability of approximation

The statistical interpretation of the obtained results demonstrates the distribution of Ni and Cu in the sampling variability. Ni and Cu concentrations in soils follow the lognormal distribution in the studied soils (Figures 7-8). The lognormal distribution and the high variety coefficient reflect the heterogeneity of the metal accumulation in soils. They are usually formed when several dependent causes are present. Thus, it can be caused by the massive intake of chemicals from industrial contamination and the natural deposition of the elements in the area. Due to the lognormal distribution of the data the nonparametric statistics were applied to the sampling.

Figure 7. The Cu and Ni distribution in soils of the Impact zone [ppm]

Figure 8. The Cu and Ni distribution in soils of the Background zone [ppm]

According to the Mann-Whitney U test (Annex, Table 5); the averages of the compared sampling varieties (Ni and Cu in soil background and impact zones) have a significant difference (Table 2). The values of Ni and Cu in two investigated areas were compared, therefore.

Descriptive statistics of Ni and Cu concentrations in soils are presented for impact and background areas in Table 2. The average concentration of Ni varies from 24.24 – 88.17 [ppm] in the Impact area, and from 3.34 to 12.62 [ppm] in the background; Cu concentrations are in the gap from 14.17 to 60.66 [ppm] in the Impact and 6.76 -24.60 [ppm] in the background. For further comparison, the arithmetical means were taken to avoid the underestimation of the results and to show the local anomalies of the metal accumulation.

Table 2. Descriptive statistics

	Impact area		Background area	
	Ni	Cu	Ni	Cu
Mean	55.0	33.3	9.3	11.4
SE	7.8	4.7	1.2	2.3
Min	24.2	14.2	3.3	6.8
Max	88.2	60.7	12.6	24.6
Median	50.4	30.1	9.7	9.7
VC	45	44	33	53
*S.R. Taylor, S.M. McLennan, 1985	20	25	20	25

*Note: mean – arithmetical mean [ppm], SE - Standard error, SD – standard deviation, CV – variation coefficient [%], * S.R. Taylor, S.M. McLennan, 1985 – average concentration of Ni and Cu in the upper continental crust* (Taylor and McLennan, 1985)

The leading role of the anthropogenic nature of the heavy metal accumulations in the soils of the impact area is supported in the Table 2. First, the mean concentrations of Ni - Cu by soil profile for impact areas significantly over their concentration in soils of the background area. Ni arithmetic mean concentration in soil impact areas is nearly 6 times higher than the background value. Whereas Cu is approximately 3 times. The composition of the industrial pollution of the ecosystem from the «Severonickel» vicinity includes Cu_2S, $CuFeS_2$, $(Ni, Fe)_9S_8$, metallic Ni, and Cu (Barkan V.Sh., 2008). Thus, pollutant composition is reflected in the high concentration of heavy metals in soils. The mean values of Ni-Cu in the plant vicinity increase the continental crust as well (Taylor and McLennan, 1985), at the same time values of the conditionally clean area is lower than literature data. The anthropogenic origin of the pollution is supported by the fact that one of the largest industrial enterprises in the Murmansk region is located in the study area (Sandimirov, 2020).

The background soils exceeding the maximum permitted concentration of the heavy metals in soils are probably polluted too. Unlike the impact area, where the depositions of Ni and Cu are reported (Figure 1) soils of the conditionally clean area have no natural occurrence of these metals. At the same time wind destination is the main factor in the pollutants spread.

The intensive pollution of the area with heavy metals reported by many (Evseev and Krasovskaya, 2017; Kashulina, 2017; Kyllönen et al., 2020; Liisa et al., 1997) and significant level of the pollution leads us to propose the anthropogenic nature of their accumulation and distribution.

We assume that the industrial intake of Cu and Ni corresponds to the general trend of their concentration. In this case, the spatial distribution from the source of pollution should be investigated. Therefore, the variation of the heavy metals accumulation in different sampling points is observed. As we can see the distance from the source of pollution is a critical parameter of any environmental monitoring. Data represented in the table was also compared according to the sampling point (background: clean area). The graphical interpretation of the obtained results demonstrates the main trends of the distribution of heavy metals in the vicinity of the industrial plants (Figure 4-5).

We observe that Ni and Cu follow the same distribution trend. We see that the majority of the soils taken in the zones far from the source of the industrial pollution contain lower concentrations of both metals.

As we can see from the wind direction vector, the climate of the research area determines the equal influence of wind transfer. At the same time, zones of landscape degradation reflect the main wind destination (Figure 2) and the pollution flow too. Thus, the sampling point 5 km (point N 12, Annex, Table 3) away from the source of the contamination is placed in the Impact area but from the leeward. Consequently, we observe the reduction of Ni and Cu concentrations at this point.

Massive intake of the pollutants is usually characterized by the heterogeneity of their accumulation, with the consequent augmentation of the concentration coefficient. Here, we see that the concentration of Ni and Cu calculated for soils from the background vary less than for the impact

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area. This can be explained by the multiple inputs (pollution and natural deposition) of those chemicals in the anthropogenic loaded area.

Accumulation of heavy metals is also changed by the genetic characteristics of the soil's horizons. Investigated in the current paper «B» horizon of soils or «subsoil» are normally covered with the litter and top-soils and less contaminated (Elsukova et al., 2019). However, leaning on the obtained results subsoil horizons of the Imandra lake vicinity are also polluted. Pollutants migrate from the surface layer to the deeper horizons. Similar processes are noted in zones of industrial vicinity by other authors (Kim et al., 2020).

The accumulation capability of the deeper soil layers highlights the necessity of their investigation as a possible source of pollution. The results of our studies show that soils of the Kola Peninsula accumulate a significant value of Cu and Ni, in the impact and background areas, which poses a potential threat to human health. In this regard, it is necessary to assess the toxicity of individual elements for the human body.

4.2 Human health impact assessment

Site-specific characteristics of the researched areas (as maternal rock composition) and massive environmental pollution of the area with Ni and Cu determine negative conditions for the population health in the area.

To simplify the reading of the resulting charts, we repeat the sampling map below (Figure 10).

We observe 2 trends in the obtained results:

1) The trend of Ni and Cu gross values (concentrations) distribution correlates with the IS values. The first mineral horizon (topsoils) shows the lowest element's concentrations (Annex, Table 3). It is logical to assume that IS of the surface layer will be the lowest one. We conclude, that scoping the "active" layer only (0.1 [m]) (de Souza et al., 2017), when heavy metals vary greatly depending on the local properties of the soil considered (Hellweg et al., 2005; Stoessel et al., 2018), can lead to an

478 underestimation of the negative influence on the population health. Local data propose an approach
479 to counteract over- and underestimation of metal toxicity in the default USEtox assessment (Hedberg
480 et al., 2019). Despite the fact that USEtox model succeeds in mimicking the results of the spatially
481 differentiated model (Kounina et al., 2014), soil parameters as thickness of the topsoil layer, or linking
482 soils with subsoil (Karim et al., 2019) should be considered.
483 2) When heavy metals concentrations depend on the soil profile properties, consequent variation
484 between default and modified IS values is observed. That supports the hypothesis that consideration
485 of specific features of the soil profile-such as erosion of the upper horizons significantly changes the
486 results of the human health impact assessment. Obtained results show that the distribution of elements
487 in soil profiles is characterized by an increase in Cu, Ni content with depth. Thus, dusting of the
488 topsoil horizons can be considered as specific geographic conditions of the studied area. Models
489 should be adapted and parameterized for different local conditions (Gentil et al., 2020).
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Figure 9. Ni (A.), and Cu (B.) Impact score calculation modified (m) and default (d) calculated for different soil layers in the Background and Impact area

Note: h, [m] – height of the soil horizon, N – number of sampling point, Distance [m] – distance from the source of pollution

Figure 10. Sampling locations (A.) and study area (B.)

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Specific geographic conditions of the studied area do not allow the calculation of the standard height of the soil horizon to be done. Due to the geographical conditions in the vicinity of Lake Imandra low power podzols with poorly developed humus layer and litter are formed. These soils due to their depth of ~0.1 [m] may correspond to the B horizon, which accumulates the main mass of chemical elements. Also, the topsoils can be disturbed by industrial activity and intensive environmental pollution, that leads to subsoils scalping (Wu et al., 2020; Yu et al., 2014). Soils from the deeper horizons can be spread with the wind flow and consequently affect the local population. Environmental fate of the pollutants as their distribution through the soil horizon should be connected with the human toxicity assessment (Núñez and Finkbeiner, 2020). It is important to admit the geological and ecological characteristics of the region in the impact assessment process. For example, for further impact assessment research such site-specific characterization as landscape conditions or their technogenic disruption.

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5. Conclusion

Studies of heavy metal pollution in the Kola Peninsula are relevant, in terms of assessing the state of the environment and in terms of the impact on public health. Pollution of the surroundings of Severonickel Plant by heavy metals is undeniable and is emphasized both by this work and by other authors (Bazova, 2013; Evseev and Krasovskaya, 2017; Kashulina, 2018; Kyllönen et al., 2020; Liisa et al., 1997).

The results of our work demonstrate a wide range of accumulation of chemical elements in the subsoil horizon, both in the area adjacent to the plant and in the conditional background area. Factor and the correlation analysis between Ni and Cu and a similar trend of their accumulation, allow us to propose that they have similar source of origin. The results emphasize the role of subsoils in the accumulation of the main number of pollutants in the studied soils. To ensure a more comprehensive and accurate assessment of metal contamination results, two assessment methods of impact score, and characteristic factors calculation with the USEtox model were applied.

As it was noted earlier, the research area is characterized by man-made disturbance of the soil profile and erosion of the upper horizons. While the results of the work highlight the features and height of soil horizons in the profile as a critical parameter to assess the impact on the environment. For such areas it is necessary to make an adapted calculation of the human health Impact Score, taking into account the characteristics of the local landscape.

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Contaminated Arctic soils human health Impact assessment: A regional Life Cycle Impact Assessment approach

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Annex

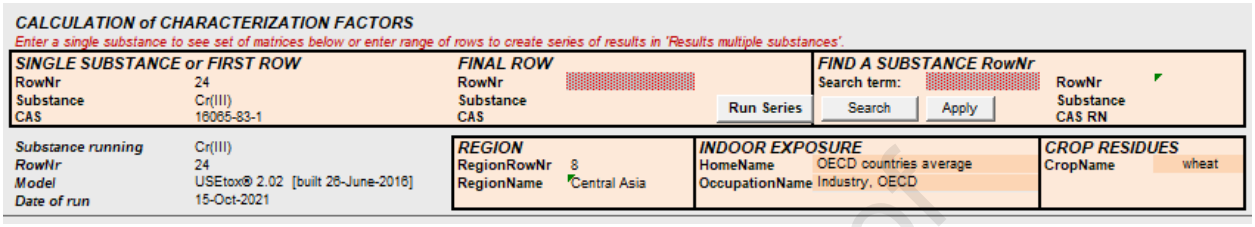


Figure 1. The USEtox model interface

Table 1. Characterization factors for the potential human health damages, CF [DALY/kg]

CF hum	**Ni (II)**	**Cu (II)**
Nat.soilC	9.0E-07	1.2E-07

Table 2. Square of sampling areas

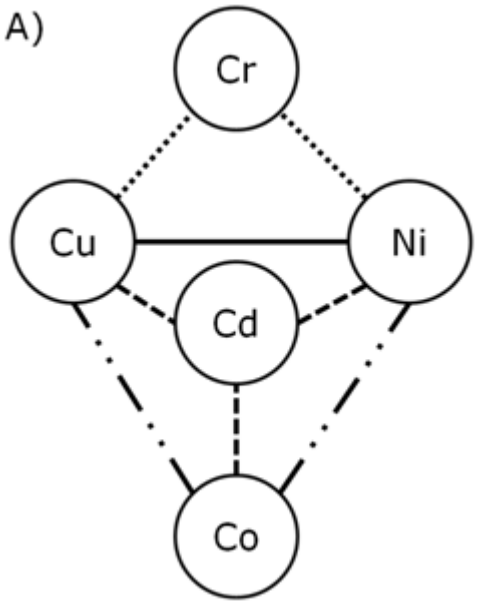
N	**Sampling area**	**S [km^2]**	**Sampling points**
1	Apatity catchment area	2500	1, 13
2	Monchegorsk catchment area	3400	5, 6, 7, 12, 8, 9, 10, 2
3	Kirovsk catchment area	3600	4, 3

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15 Table 3. Ni and Cu gross values and calculation results of IS

Point	Horizon	Distance [m]	h [m]	Ni		Cu		Point	Horizon	Distance [m]	h [m]	Ni		Cu	
				C in soils [ppm]	IS [Daly/kg]	C in soils [ppm]	IS [Daly/kg]					C in soils [ppm]	IS [Daly/kg]	C in soils [ppm]	IS [Daly/kg]
Impact area								Background area							
5	Subsoils	2000	0.2	88.2	60	60.7	5.5	11	Topsoils	34500	0.1	1.2	0.3	1.2	0.04
6	Topsoils	2000	0.1	53.9	17	33.0	1.4		Subsoils		0.1	9.7	7	7.8	0.7
	Subsoils		0.2	87.8	28	32.1	1.6		Subsoils		0.3	9.3	4	9.0	0.5
7	Substratum	2000	0.4	61.9	57	33.1	4		Substratum		0.4	4.6	4	5.9	0.6
12	Topsoils	5000	0.2	32.5	12	20.0	1	1	Subsoils	33000	0.2	12.6	2	10.4	0.2
	Subsoils		0.3	36.0	26	27.3	2.6	13	Topsoils	37500	0.1	1.0	0.2	0.4	0.01
	Subsoils+ Substratum		0.5	29.5	28	25.9	3.3		Subsoils		0.2	7.6	3	9.7	0.5
8	Topsoils	7000	0.1	7.7	2	24.9	1.1		Subsoils		0.3	10.9	4	11.3	0.6
	Subsoils		0.2	61.2	45	57.8	5.6		Substratum		0.5	10.6	7	14.7	1.3
9	Subsoils	7500	0.1	75.8	28	32.4	1.6	4	Subsoils	44500	0.3	3.3	1	6.8	0.2
	Subsoils		0.1	70.7	29	22.1	0.8	3	Subsoils	47500	0.2	11.3	11	24.6	3.1
	Substratum		0.3	45.8	54	30.3	4.8								
10	Topsoils	14700	0.1	12.3	3	9.8	0.4								
	Subsoils		0.1	24.2	8	14.2	0.6								
	Subsoils		0.2	37.1	12	29.9	1.3								
	Substratum		0.4	30.8	34	31.6	4.6								
2	Subsoils	27500	0.2	39.5	27	30.3	2.7								

16



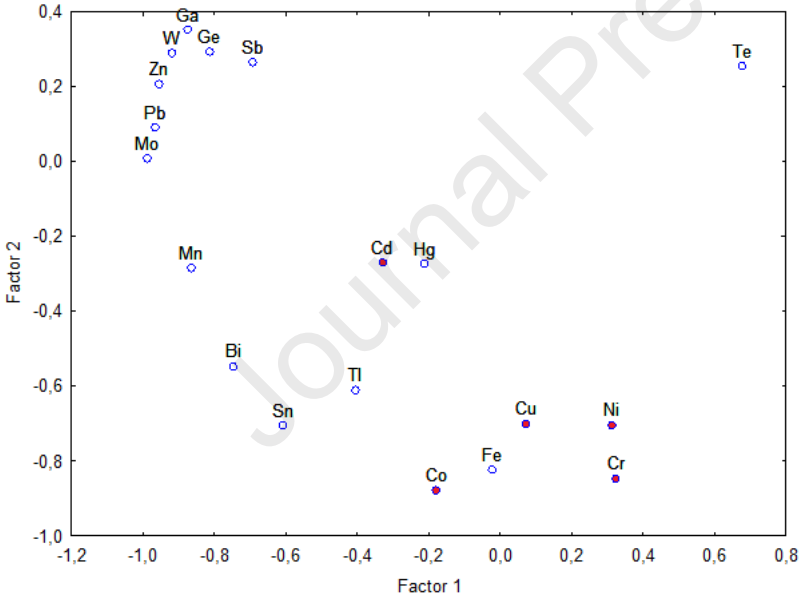
B)

Lines' type	The correlation (1-pearson r, p=0.05)
———	0.9
– · · — · · —	0.8
.....	0.7
– – – – – –	0.5

17

18 Figure 2. Correlations cluster between heavy metals in Arctic soils, N= 17.

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21 Figure 3. The factor loading (unrotated) analysis

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23 Table 4. The concentration coefficient (normalized to the mean of the sampling variety) of chemical

24 elements in soils of the Impact area (a), and the Background area (b)

N	Concentration coefficient			
	A2	B		C
		B1	B2	
The Impact area				
2		$Mo_{5,6}$-$Mn_{5,1}$-$Tl_{3,9}$-$Cs_{3,4}$-$Co_{3,2}$=Sn-Li_{3}-$Ti_{2,7}$-$V_{2,5}$=Bi-$Fe_{2,4}$-$Pb_{2,1}$-Rb_{2}-$Cr_{1,9}$-$As_{1,7}$=W=U-$Zn_{1,6}$=In-**$Cu_{1,4}$**=Sb-**$Ni_{1,3}$**-$Sc_{1,2}$-$Hg_{1,1}$=Th-Cd_{1}		
5		**$Ni_{2,8}$=Cu**-$Cd_{1,8}$-$Co_{1,7}$-$Cr_{1,6}$=Bi-$Re_{1,5}$-$V_{1,4}$=Ir=Hg=Th-$Sc_{1,3}$-$Al_{1,2}$=Fe=In=Sn-$Li_{1,1}$		
6	**$Ni_{1,7}$-$Cu_{1,5}$**=Cd-$Bi_{1,3}$-$Pb_{1,2}$-Sb_{1}	$Sc_{3,4}$-$Al_{3,3}$-$Hg_{3,1}$-**$Ni_{2,8}$**-$Cr_{2,7}$-$In_{2,6}$-$Ir_{2,5}$-$Sb_{2,4}$-$Re_{2,3}$-$V_{2,2}$-As_{2}-$Fe_{1,9}$-$Sn_{1,8}$-$Ti_{1,7}$=Os-**$Cu_{1,5}$**=Ga=Au-$Pt_{1,4}$-$Bi_{1,3}$-$Co_{1,2}$-$Mo_{1,1}$=Cd		
7				$Cr_{2,1}$=Co-**Ni_{2}**-$Sc_{1,7}$-$Tl_{1,6}$-**$Cu_{1,5}$**=Rb-$V_{1,3}$=Mn=Ba-$Fe_{1,2}$-$In_{1,1}$-Bi_{1}
8	$Au_{1,3}$-$Sb_{1,2}$=Re-**$Cu_{1,1}$**=Pb-Os_{1}	$Cd_{3,4}$-$Cr_{2,6}$=**Cu**-$Sb_{2,1}$-Hg_{2}=**Ni**-$In_{1,8}$-$Fe_{1,6}$-$Al_{1,4}$=V=Co=Ga=Ir=Bi-$Sn_{1,3}$-$Sc_{1,2}$=Os-$As_{1,1}$=Au-Pb_{1}		
9		**$Ni_{2,4}$**-$Cd_{2,3}$-Sc_{2}=Hg-$Al_{1,9}$-$Co_{1,8}$-$Cr_{1,6}$=Fe=Re=Ir-$Cu_{1,5}$=In-$V_{1,4}$-$Ti_{1,2}$-$Li_{1,1}$=Mn=Ga=Sn-Zn_{1}	**$Ni_{2,3}$**-$Co_{1,7}$-$Sc_{1,6}$-$Cd_{1,5}$-$Al_{1,4}$-$Cr_{1,2}$=Fe-$V_{1,1}$-Li_{1}=**Cu**=In=Re=Ba	$Rb_{1,7}$=Tl-$Co_{1,6}$-**$Ni_{1,5}$-$Cu_{1,4}$**-$Ba_{1,3}$-$Sc_{1,2}$=Ti-$Li_{1,1}$=V=Cr=Mn=Fe=Hg-Al_{1}
10	Pb_{2}-$Bi_{1,2}$-$Cd_{1,1}$	$In_{2,7}$-$Au_{2,6}$-$Ti_{2,5}$-$Fe_{2,3}$-$V_{2,2}$-$Cr_{2,1}$-Hg_{2}-$Sc_{1,8}$=Sn-$Cs_{1,4}$-$Al_{1,3}$=Tl-$Co_{1,2}$=Ga=Rb-$Li_{1,1}$-Cd=Lu-Mn_{1}	Re_{2}-$Tl_{1,5}$-$Li_{1,4}$=Co=**Cu**-$Sc_{1,3}$-**$Ni_{1,2}$**-$Rb_{1,1}$=In-Al_{1}=Ba=Cs	$Tl_{1,6}$-$Co_{1,4}$=**Cu**-$Sc_{1,3}$=Lu=Re=Hg-$Rb_{1,2}$=Y=Ce=Yb-$Li_{1,1}$=Ba=Er=Tm
12	$Ti_{1,2}$=V-$Sn_{1,1}$-**Ni_{1}**	$V_{1,6}$=Cr-$Ti_{1,5}$-$Sn_{1,4}$-$Fe_{1,3}$-$Li_{1,2}$=**Cu**-$Co_{1,1}$=**Ni**=Cs=Tl=Bi-Rb_{1}	$Tl_{1,9}$-$Rb_{1,8}$-$Cr_{1,2}$=**Cu**-$Li_{1,1}$=V=Ag=Cs-Co_{1}	
The Background area				
11		$Li_{1,4}$-$Fe_{1,3}$-$Ti_{1,2}$=V-$Sn_{1,1}$-In_{1}=Bi	$Li_{1,1}$	
1		Na_{15}-$Sr_{5.6}$-$K_{4.1}$-B_{4}-$Pd_{3.8}$-Be_{3}-$Hf_{2.8}$-$As_{2.6}$=Ba-		

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N	Concentration coefficient			
	A2	B		C
		B1	B2	
		$Zr_{2.5}$-$Ca_{2.4}$-$La_{2.3}$=Nd-$Ce_{2.2}$=Pr-$Th_{2.1}$=Rb-Eu_{2}-$Sm_{1.9}$=U-$Ho_{1.8}$=Gd=Tb=Dy-$Y_{1.7}$=Rh-$Pt_{1.6}$=Yb=Er-$Au_{1.5}$=Tm-$Zn_{1.4}$=Lu=Mn-$Ta_{1.3}$=Ga=Ge=Nb-$Cs_{1.2}$-$Si_{1.2}$-$P_{1.1}$-…-**$Cu_{0.5}$-$Ni_{0.4}$**		
13	$Sn_{1,2}$	$Fe_{2,3}$-$In_{2,1}$-$V_{1,9}$-$Li_{1,7}$=Al=Sc=Ti-$Pb_{1,5}$-$Cr_{1,3}$=Th-$Ga_{1,2}$=Hg-Bi_{1}	$Li_{1,9}$-$Sc_{1,4}$=In-$Fe_{1,3}$-$Al_{1,2}$-Ti_{1}=Cr	$Li_{1,2}$-$Th_{1,1}$
4		$Sr_{8.4}$-$Sb_{8.3}$-$I_{6.5}$-$Be_{6.1}$-$W_{5.7}$-$Ta_{5.2}$-Br_{5}-$Pd_{4.9}$-$P_{4.7}$-$Nb_{4.6}$-$Mo_{4.5}$-$Cs_{4.3}$-$Ga_{3.7}$-$Rh_{3.5}$-$Ca_{3.4}$-$Zr_{3.2}$-$La_{3.1}$=Hf-Rb_{3}=Zn=Ru-$Na_{2.8}$=Ag=Hg-$Al_{2.7}$=SO4-$Cl_{2.6}$-$Ce_{2.3}$=Pr-$Pt_{2.2}$-$As_{2.1}$=Nd=U=Cd-$B_{1.9}$-$Pb_{1.8}$=Ge-$Eu_{1.7}$=Gd=K=Tb-$Ba_{1.6}$=Sm=Bi=Y-$Er_{1.5}$=Dy=Au=Ho=Tm-$Mn_{1.4}$=Lu=Yb-$Sn_{1.1}$-Ti_{1}-…-**$Cu_{0.3}$-$Ni_{0.1}$**		
3		Zr_{21}-Hf_{20}=Nb-Ta_{17}=Eu-Pr_{16}=La=Pd=Sm=Nd-U_{15}=Gd=Tb=Dy=Ho=Ce=Er=Y=Tm-Yb_{14}=W-Ag_{13}=Pt=Ge-Sr_{12}=Lu-Be_{11}-$Th_{9.9}$-$Os_{9.4}$-$Mo_{9.2}$-$As_{8.3}$-Ba_{8}-$B_{6.1}Ga_{5.3}$-$Mn_{4.9}$-$Rh_{4.8}$-$Ca_{4.7}$-$Sb_{4.4}$-$Pb_{4.2}$=Zn-$P_{4.1}$=Cs-$Rb_{3.5}$-Al_{3}-$Na_{2.7}$-$K_{2.5}$-Cd_{2}=Ru-$Sn_{1.9}$=Bi=Li=Cl=Se-$Ir_{1.8}$=Ti-$Si_{1.6}$-$Tl_{1.5}$=Au-$I_{1.4}$-$In_{1.2}$=Hg=Mg-$Co_{1.1}$=**Cu**=Fe=Sc-$SO4_{1}$-…-**$Ni_{0.4}$**		

Table 5. Mann-Whitney U Test

	Rank Sum I	Rank Sum B	U	Z	p-value	Z	2*1sided
Cu	123	30	2.00	3.17	0.002	3.17	0.0004
Ni	125	28	0.00	3.37	0.001	3.37	0.0001
The critical value, $p_{0.05}$ = 14							

Note: I – impact area (N=10); B – Background area (N=7)

Table 6. The sampling points with the distance from the pollution source

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N	N of sampling point	The distance [km]	Height of the soil horizon [m]
The Impact area			
1	5	2.00	0.15
2	6	2.00	0.08
3	12	5.00	0.16
4	12	5.00	0.21
5	8	7.00	0.16
6	9	7.50	0.08
7	9	7.50	0.06
8	10	14.70	0.07
9	10	14.70	0.07
10	2	27.50	0.15
The Background area			
11	11	34.5	0.5
12	11	34.5	0.5
13	1	33	0.5
14	13	37.5	0.4
15	13	37.5	0.5
16	4	44.5	0.4
17	3	47.5	1.6

Table 7. Ni and Cu emissions in the studied area

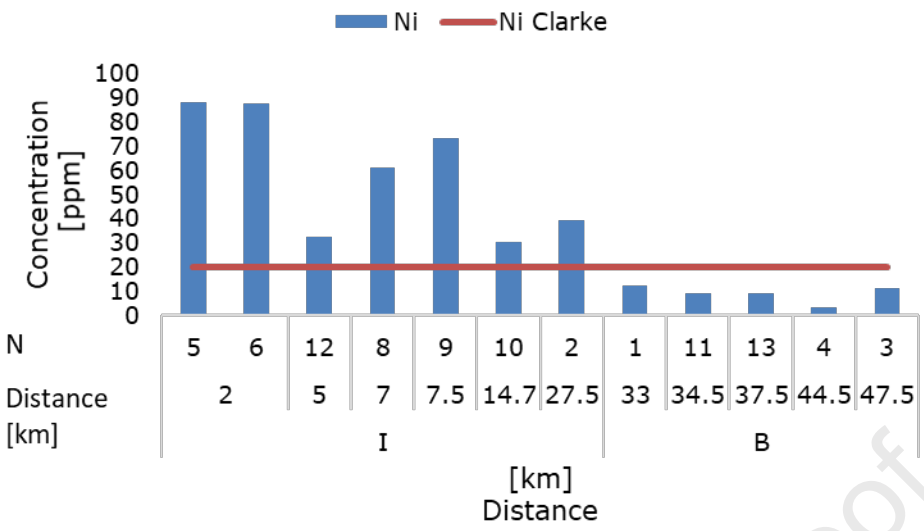
Area	Impact area		Background		References
Element	Ni	Cu	Ni	Cu	
Emission, ton/year	245	462			(Barkan and Lyanguzova, 2018)

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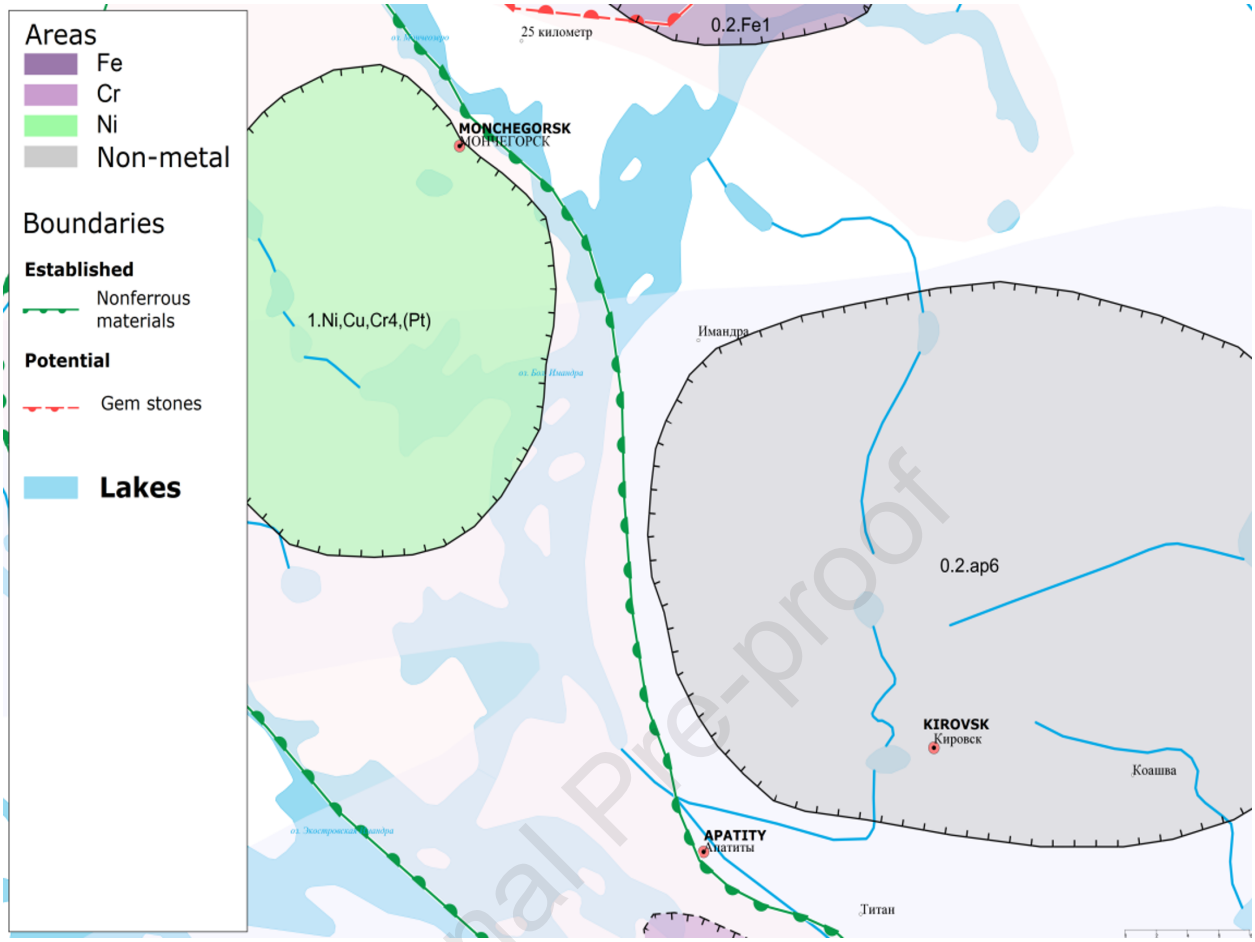
Precipitation, µg/l	262	540	14.3	19.4	(Evtyugina and Asming V., 2013)
Surface water, µg/l	46.0	17.0	0.1	0.3	(Evtyugina et al., 2016)
Groundwater, µg/l	4.9	0.6	0.02	0.1	
Soil, ppm	55.0	33.3	9.3	11.4	Own results

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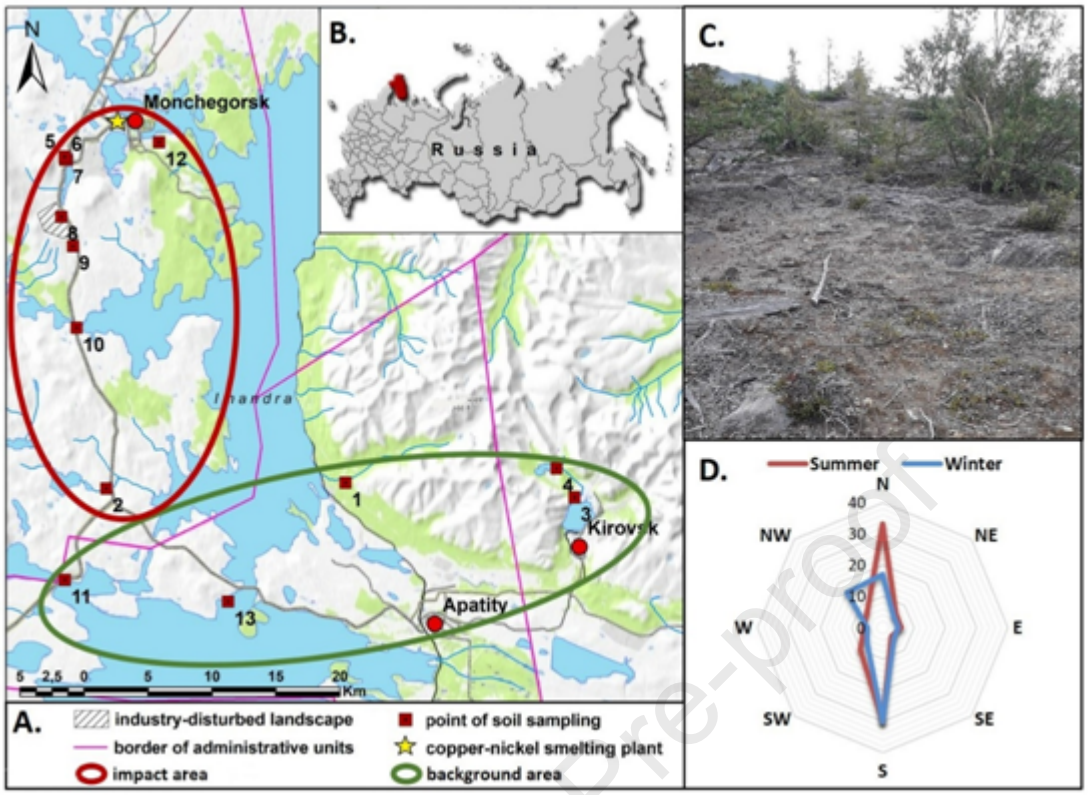
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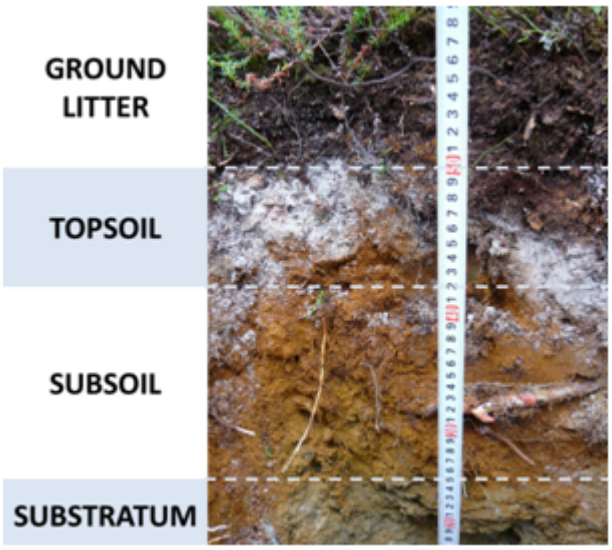
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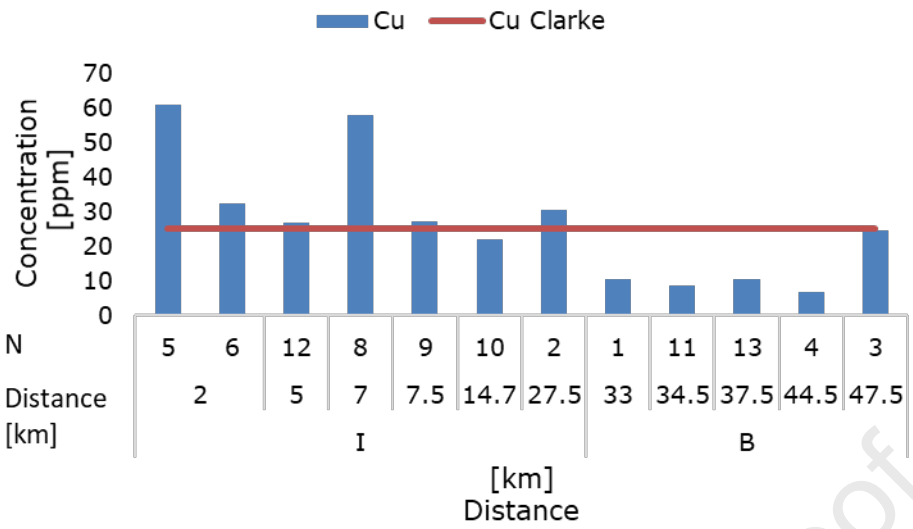
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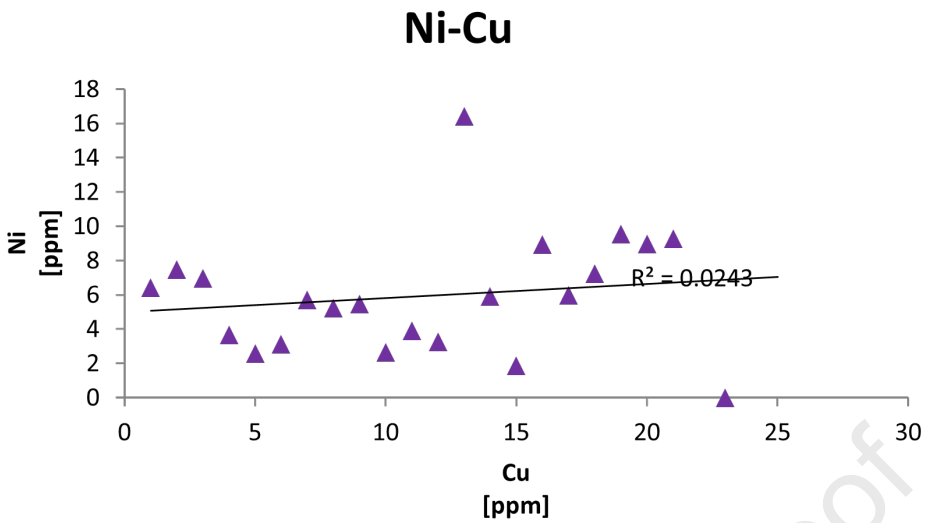
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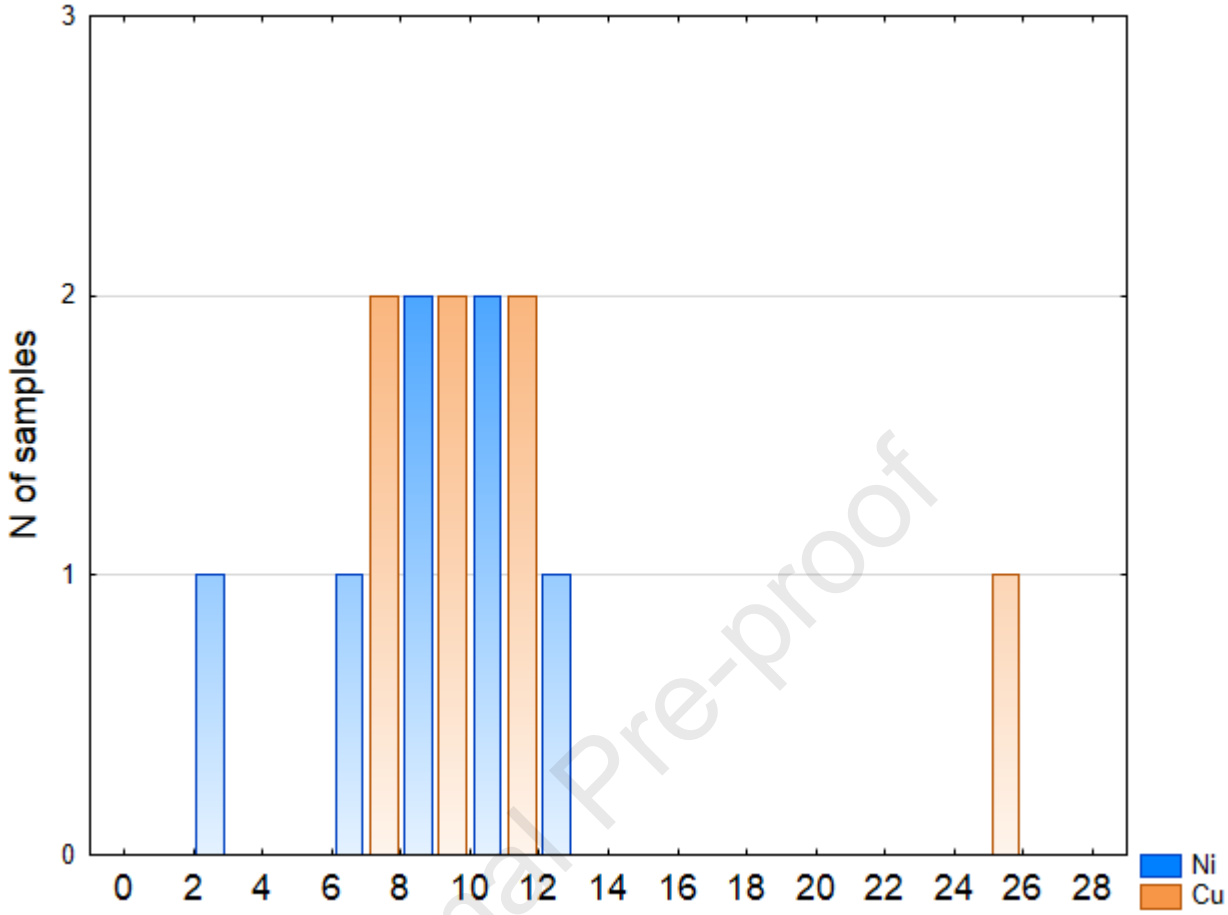
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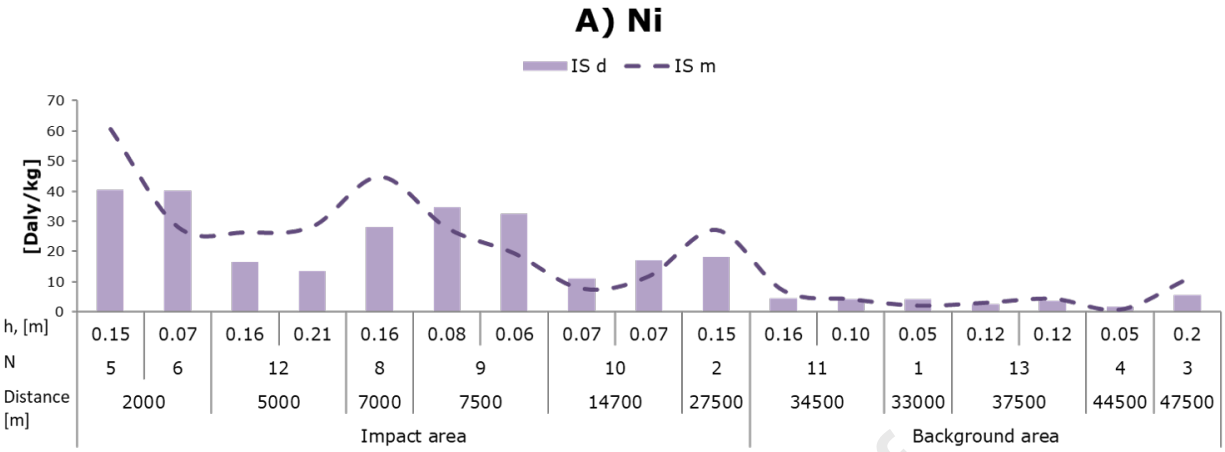
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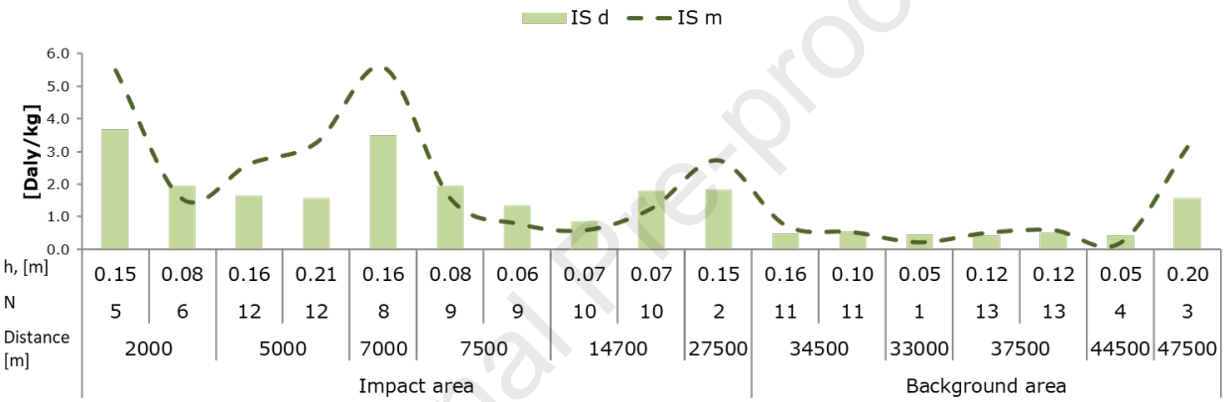


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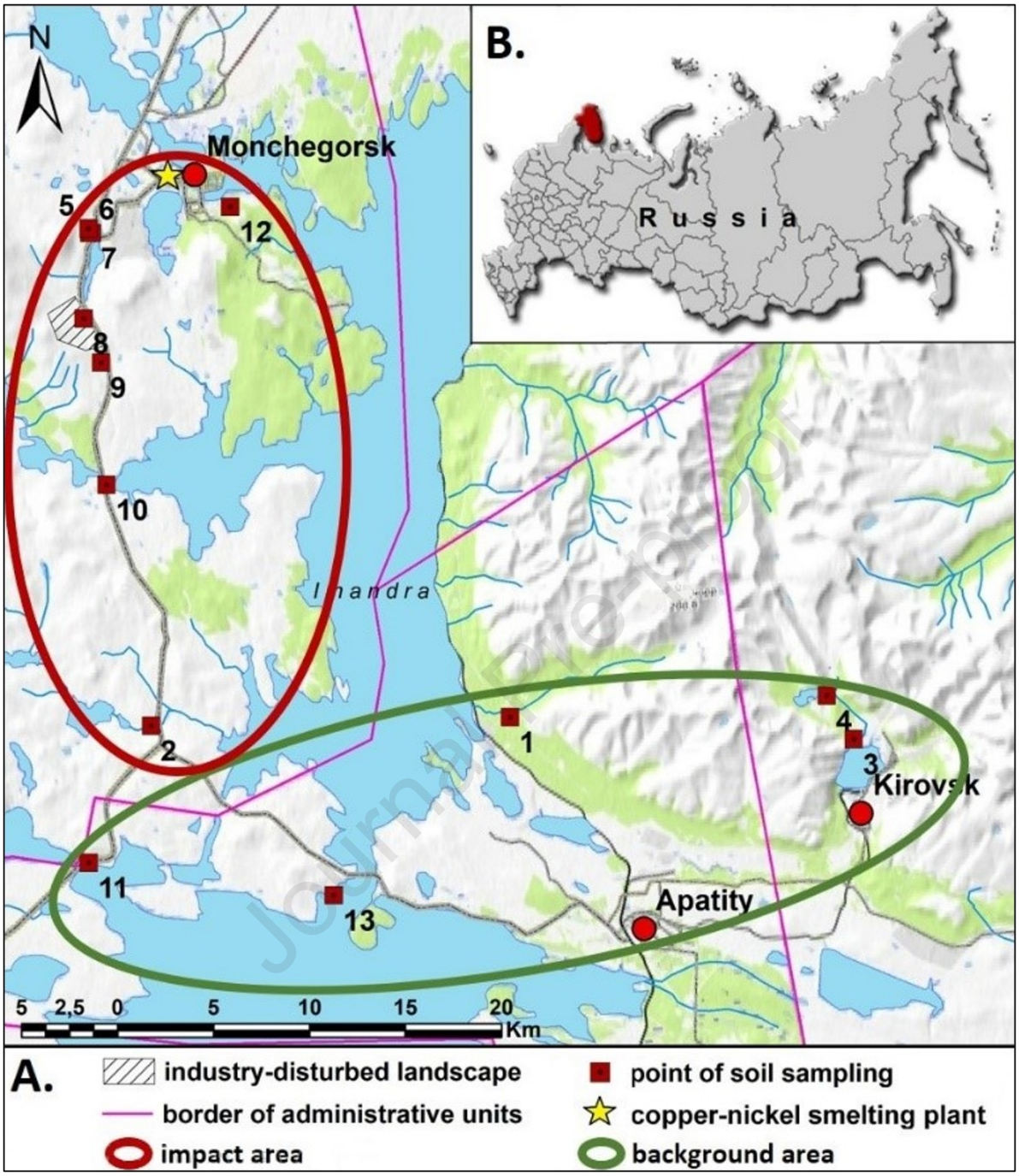


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Highlights

- Soils of «Severonickel» plant vicinity accumulate Ni and Cu in the impact and the background areas;
- The genetic soil profile of the Severonickel» plant vicinity is characterized with eroded upper horizons;
- The subsoil horizon accumulates the majority of the estimated pollutants;
- Human health Impact Scores for arctic soils increase in the subsoil horizon;
- The depth of the soil's horizons profile is a critical parameter for the human health impact score calculation.

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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