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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.

Journal Prevention

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3 Authors

- 4 ^{1, 2} Alexandra Belyanovskaya alexandra.belyanovskaya@outlook.com
- 5 ¹ Daria Vorobeva vorobeyda18@gmail.com
- 6 ¹ Natalia Guseva gusevanv@tpu.ru
- 7 ³ Bertrand Laratte bertrand.laratte@ensam.eu
- 8
- ⁹ ¹ Division for Geology of School of Earth Sciences & Engineering, Tomsk Polytechnic University,
- 10 30, Lenin Avenue, Tomsk, 634050, Russia
- ¹¹ ²Laboratory for Sedimentology and Paleobiosphere Evolution University of Tyumen, Russia
- ³ Arts et Métiers Institute of Technology, University of Bordeaux, CNRS, Bordeaux INP, INRAE,
- 13 I2M Bordeaux, F-33400 Talence, France

15 Abstract

The chemical composition of soils reflects the degree of industrial exposure. Cu and Ni concentrations 16 in soils of the «Severonickel» plant vicinity are higher than remote ones. In the impact area, the mean 17 concentration of the heavy metals in the subsoils is 55 [ppm] for Ni and 33 [ppm] for Cu. Soils' 18 19 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 20 disturbance of the soil profile can lead to subsoil spreading and a significant increase in the human 21 health Impact Score. At the same time, the default IS calculation is focused on the 0.1 [m] depth for 22 all zones. In the paper two factors are considered in the impact modeling modification. The human 23 health Impact Score is calculated specifically for each genetic soil layer with the respective depth of 24 25 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 26 highest IS_{hum} for Ni is 60, for Cu is 5.6 [DALY]; in the Background area, it is 11 and 3.1 [DALY] 27 respectively. The importance of using the regionally modified values in population health impact 28 29 monitoring is highlighted.

30

31 Keywords

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

34

36 Introduction

37

The Arctic zone is characterized by extreme natural and climatic conditions, the focal nature of 38 industrial and economic exploration of the territory, low population density, and the vulnerability of 39 nature and public health from industrial activity. The Kola Peninsula is Russian Arctic territory where 40 the wealth of mineral resources has become the basis for the growth of mining and metallurgical 41 industries. At the same time, northern ecosystems are highly sensitive to the intake of technogenic 42 origin chemical elements. Many metal ions are potentially toxic, so it is important to understand their 43 transport and distribution in the environment and the routes through which they may be transferred 44 45 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 46 al., 2020; Moiseenko et al., 2006; Sandimirov, 2020). Exposure to heavy metals can cause a lot of 47 48 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 49 50 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 51 input to ecosystems from the polluted atmosphere. Soil is a depositing medium that accumulates 52 chemical elements. This property becomes especially important under conditions of long-term 53 anthropogenic load (Barsova et al., 2019). Soil is a critical repository for numerous deleterious 54 pollutants, thereby serving as a good matrix for assessing the status quo of environmental pollution 55 (Doyi et al., 2018). Soils degradation leads to a decrease in their sorption capacity and, consequently, 56 to groundwater contamination with heavy metals (Shumilova and Petrov, 2016). Heavy metal 57 pollution affects the soil's ecological function (Li et al., 2021; Yuan et al., 2021). Heavy metals 58 emanate from a myriad of sources in the environment, including industrial wastes, and particularly in 59 areas of the intersection of enterprises (Wuana and Okieimen, 2011; Zhang et al., 2020). Recently a 60 lot of research has been devoted to human health impact assessment of heavy metals in mediums such 61

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 inparticular:

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.

91 2. In the USEtox model only the concentration of the pollutant in the surface layer is considered
92 for the Impact Score calculation (Fantke et al., 2017). However, the surface soil layer can be
93 disturbed, or the natural genetic layer can be changed (Elsukova et al., 2019; Kashulina et al., 2014,
94 2010). The default IS calculation is focused on the 0.1 [m] depth for all zones. We extend the model
95 dataset leaning on media properties as different soils profile horizons (from 0.05 [m] to 0.2 [m]).

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

To investigate this hypothesis, we compare soil layers' potential toxicity for human health by 100 calculation of the Impact score. The Impact score is calculated both in the area of influence and in 101 the background area. It emphasizes the combined influence of natural and anthropogenic factors, 102 103 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 104 approach allows the IS values to be taken as another factor for identifying the zone of impact of 105 106 industrial enterprises from the perspective of human health impact assessment. To compare the level of impact the background area was chosen as well. 107

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 113 health (IS_{hum}), combining different methods of assessing the ecological condition of urban areas: 114 geological approach and impact assessment, which reflect the novelty of the research. On the one 115 hand, studies of the geological characteristics of areas exposed to industrial impact reflect the changes 116 117 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 118 a more complete assessment of the quality of the natural environment. The application of an 119 interdisciplinary approach is based on the principles of ecogeochemistry, human health, and 120 ecosystem impact assessment. 121

122

123 1. Study area

124

125 **1.1 Natural conditions**

126

127 The Lake Imandra catchment is the central part of the Murmansk region (Figure 1-2). Murmansk 128 region is a constituent entity of the Russian Federation with a total area of 144.900 thousand km², 129 located in the north-west of Russia. Sampling points refer to three administrative units of the 130 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 isfollowing (Figure 1).

140

141

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

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

144

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, 153 1970).

154

155 **1.2 Industrial influence**

156

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).

160 The presented results allow us to evaluate the impact of the copper-nickel plant on the chemical 161 composition of the components of the natural environment through the polluted atmosphere. 162 Emissions in 2016 were Ni 245 t/yr Cu 462 t/yr (Barkan and Lyanguzova, 2018). In the zone of 163 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, 164 Cu 17 µg/l) concentrations of these elements are significantly higher than in groundwater (Ni 4.9 165 μ g/l, Cu 0.56 μ g/l) (Evtyugina et al., 2016). When atmospheric precipitation infiltrates through the 166 soil, most of the heavy metals are bound in the soil horizons, where they accumulate (Ni 54.99 ppm, 167 168 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 169 precipitation through the soil plays a significant role in the chemical composition of groundwater, 170 and soils themselves. 171

There are extremely high concentrations of Ni and Cu in the organogenic soil horizons and the 172 medium peat layers (Kashulina, 2017). According to the investigation of (Koptsik et al., 2015) soils 173 of technogenic wastelands formed under the influence of long-term emissions of Severonickel 174 Combine are acidic, depleted in nutrients, polluted with heavy metals, and badly eroded Natural 175 illuvial humic podzols have lost their upper organogenic and podzolic horizons as a result of erosion 176 and removal of protective vegetation cover. Atmospheric air pollution with SO₄, disruption of 177 vegetation cover and changes in the morphological composition of soils increases their ability to 178 179 accumulate heavy metals (Kashulina et al., 2014, 2010). Accumulation of Ni and Cu in soils correlates with winter fallouts (Kashulina, 2017). 180

The concentration and the composition of the pollutants in soils correlate with the emission from local 181 182 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 183 major industrial enterprises of the Murmansk region. The copper-nickel smelting plant 184 «Severonickel» (The Kola Mining and Metallurgical Company) built in the 1930s is located in 185 Monchegorsk at an altitude of about 123 m. Currently, copper-nickel matte is processed at this site. 186 187 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 188 («Severonickel» in Monchegorsk and «Pechenganickel» Nickel-Zapolayrniy) decreased from 1627.1 189

190	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					
191	certain indicator of atmospheric air pollution by metals is their content in the surface soil layer (Ettler,					
192	2015a; Evdokimova et al., 2011; Evseev and Krasovskaya, 2017). The content of copper and nickel					
193	in Monchegorsk soil exceeds MAC up to 5 times (Opekunova et al., 2006).					
194	Mixed environmental conditions (natural anomalies and anthropogenic impact) generate the need to					
195	assess the impact of the environment on public health.					
196						
197	2. Materials and methods					
198 199	2.1 Methods of soil sampling					
200	This research paper is focused on Ni and Cu distribution and the impact assessment in soils of the					
201	Kola Peninsula, taking into account their concentration in impact and background areas (Figure 2).					
202	Impact areas are normally more polluted than remote areas (Ettler, 2015). The background zone was					
203	taken 30-50 km from the emission source while observing a decrease in the concentration of the main					
204	pollutants (Ni and Cu).					
205	According to the prevailing wind direction, 13 soil sections (0-60 cm) were sampled by layer during					
206	the expedition in July 2016 (total number of samples 28). The sampling locations are shown in Figure					
207	2 and Table 2 in the Annex.					
208						
209						
210	Figure 2. Sampling locations (A.) and study area (B.), industry-disturbed landscape (C.) and main					
211	wind direction of Monchegorsk city (D.)					
212						
213	To assess the accumulation of heavy metals in different types of soils, the structure of their vertical					
214	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 216 Federation [Nacional'nyj atlas pochy Rossijskoj Federacii], 2011). 217 All the samples we studied were taken at points belonging to the podzol soil type formed in cold areas 218 with a good leaching regime. On the Kola Peninsula, podzols with a low thickness of 15-50 cm are 219 220 typical. Podzols with higher thicknesses (layer of more than 50 cm) are not widespread (Elkshina and Kupriyanova, 1970). 221 Podzols are characterized by a differentiated profile (Figure 3) consisting of litter 3–8 cm thick; 222 whitish, lightened due to the removal of coloring iron compounds, and humus of the podzolic or 223 eluvial horizon (topsoil) with a thickness of 2 to 20-30 cm; illuvial horizon (subsoil) of brown or 224 ocher tones, formed as a result of the illuvial accumulation of aluminum-ferruginous-humus complex 225 226 compounds, gradually turning into the parent rock (substratum) (National Atlas of Soils of the Russian Federation [Nacional'nyj atlas pochy Rossijskoj Federacii], 2011). 227

228

229

230 Figure 3. Genetic horizon of podzol

231

Each soil layer plays a particular role in the accumulation processes of pollutants. In the current 232 research, we turn attention in particular to the subsoil. The smaller number of topsoil samples is due 233 234 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 235 samples are taken from the depth of 0.06-0.2 m – the subsoil. This depth corresponds to the illuvial 236 horizon according to the Russian classification of soils (National Atlas of Soils of the Russian 237 Federation [Nacional'nyj atlas pochy Rossijskoj Federacii], 2011). The subsoil is the main mineral 238 239 horizon that accumulates inorganic components, coming from the overlying layers (Adams and Moore, 1983). 240

242	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

243

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).

247 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 ofsynthetic film, dense fabric, or water-resistant paper).

250

251 **2.2. Sample preparation and analysis**

252

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

255 Sample preparation. Soil samples were naturally air-dried in the room prevented chemical pollution.

256 Debris was removed, and they were then sieved through a sieve (mesh size 1 mm) for later use.

257 To determine the total metal concentration, a soil sample of 2-3 g was weighed, placed into a PP

digestion vessel, and then concentrated HNO₃ 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% HNO₃.

261 Then chemical elements were measured by an inductively coupled plasma mass spectrometer (ICP-

262 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 265 common types of health impairment caused by Ni are respiratory, skin and cardiovascular diseases 266 267 (Chashschin et al., 1994). The increased nickel content in soils leads to endemic diseases: ugly forms appear in plants and in animals; eve diseases are associated with the accumulation of nickel in the 268 cornea (McIlveen and Negusanti, 1994). The clark of nickel in World soils by Vinogradov is 40 269 270 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). 271 Nickel enters the soil as a result of fuel combustion and industrial emissions. In soils Ni accumulates 272 strongly in agrodern-podzolic glev soil (Vodvanitsky, 2008). 273

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).

279

280 **3. Data processing technique**

281 **3.1. Descriptive statistics**

282

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.
- 292

293 **3.2** The Impact assessment and Characteristic factors calculation with the USEtox model

- 294 **3.2.1 General framework**
- 295 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).

302

- $IS = CF \times M_{x,i}$
- Formula 1. The Impact Score calculation (Fantke et al., 2017)

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

306

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.

314
$$M = \frac{C_{x,i} \times V_s \times p_x}{10^6} [kg]$$

- 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 $[kg_i/m_i^3]$.

318

319 **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).

324

325 $V_i = h_i \times S_i$

326 Formula 3. The volume of soils calculation

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

328

Physical parameters of the soils are taken into account. For each studied area, the total volume of soils is considered (Formula 3, V_s [m³]). 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}/m³_{soil}], and V_s is the volume of soils of each considered region [m³].

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).

4. Results and discussions

339 4.1 Statistical analysis results

341	Analysis of the chemical composition of the mineral horizons of the Kola Peninsula soils shows that
342	the subsoil horizon accumulates the majority of the estimated pollutants (heavy metals, radioactive,
343	and REEs) (Annex, Table 4) in all sampling points. A large number of these pollutants have high
344	concentration coefficients (CC <2;3;5). Maximum CC is also noted in subsoils. The main components
345	of emissions to the environment (Ni, Cu) (Opekunova et al., 2006) are characterized by a high
346	concentration in all the considered mineral horizons of soils, both in the control and in the zone of
347	influence. On the leeward side 5 km from the mill, there is a decrease in Ni and Cu concentrations
348	(Figure 4-5), these elements accumulate the most.
349	
350	
351	Figure 4. The Ni concentrations in the Background (B) and in the Impact (I) areas, in different
352	distance from the source of the pollution
353	Note: Clark - S.R. Taylor, S.M. McLennan, 1985 – average concentration of Ni and Cu in the upper
354	continental crust (Taylor and McLennan, 1985)
355	N-number of sampling point, Distance $[km]$ – distance from the source of pollution
356	
357	
358	Figure 5. The Cu concentrations in the Background (B) and in the Impact (I) areas, in different

- 360 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 accumulateddue to natural factors.

Minimal concentrations of Ni and Cu (CC<1) are noted in soils of the background area, while other 366 heavy metals, radioactive and rare-earth elements in these samples have CC>1. The wide spectrum 367 368 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 369 ecological and geochemical situation in the study area. Opekunova et al. (Opekunova et al., 2006) 370 also revealed the content of a group of heavy metals in soils in the vicinity of Monchegorsk. Since 371 this work is devoted to the accumulation of heavy metals, further attention in the work is focused on 372 this group of chemical elements. 373

The wide range of accumulation of heavy metals in the soils of the Kola Peninsula is also 374 characterized by the presence of significant correlations between them. Heavy metals in general and 375 Ni and Cu as the main industrial pollutants of the research area have close chemical properties and 376 correlate in the soil (Figure 6). There is the strongest positive correlation between Cu and Ni 377 (Spearman rank correlation coefficient p_{0.05}=0.9), (Annex, Figure 2). Heavy metals (Cr, Cd, Co) have 378 379 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, 380 but Cu and Ni in the area, the main factor affecting the whole group of the heavy metals is probably 381 382 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.

387

389 Figure 6. Cu - Ni correlation diagram, polynomial trend line ($n_{samples}=17$), R² - reliability of 390 approximation

391

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.

399

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]

400

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.

410

411 Table 2. Descriptive statistics

<sup>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.</sup>

	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)

415

The leading role of the anthropogenic nature of the heavy metal accumulations in the soils of the 416 impact area is supported in the Table 2. First, the mean concentrations of Ni - Cu by soil profile for 417 impact areas significantly over their concentration in soils of the background area. Ni arithmetic mean 418 419 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 420 «Severonickel» vicinity includes Cu₂S, CuFeS₂, (Ni, Fe)₉S₈, metallic Ni, and Cu (Barkan V.Sh., 421 2008). Thus, pollutant composition is reflected in the high concentration of heavy metals in soils. The 422 mean values of Ni-Cu in the plant vicinity increase the continental crust as well (Taylor and 423 424 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 425 enterprises in the Murmansk region is located in the study area (Sandimirov, 2020). 426

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.

444 As we can see from the wind direction vector, the climate of the research area determines the equal 445 influence of wind transfer. At the same time, zones of landscape degradation reflect the main wind 446 destination (Figure 2) and the pollution flow too. Thus, the sampling point 5 km (point N 12, Annex, 447 Table 3) away from the source of the contamination is placed in the Impact area but from the leeward. 448 Consequently, we observe the reduction of Ni and Cu concentrations at this point.

449 Massive intake of the pollutants is usually characterized by the heterogeneity of their accumulation, 450 with the consequent augmentation of the concentration coefficient. Here, we see that the 451 concentration of Ni and Cu calculated for soils from the background vary less than for the impact

452 area. This can be explained by the multiple inputs (pollution and natural deposition) of those453 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.

465

466 4.2 Human health impact assessment

467

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).

472 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

underestimation of the negative influence on the population health. Local data propose an approach
to counteract over- and underestimation of metal toxicity in the default USEtox assessment (Hedberg
et al., 2019). Despite the fact that USEtox model succeeds in mimicking the results of the spatially
differentiated model (Kounina et al., 2014), soil parameters as thickness of the topsoil layer, or linking
soils with subsoil (Karim et al., 2019) should be considered.

When heavy metals concentrations depend on the soil profile properties, consequent variation between default and modified IS values is observed. That supports the hypothesis that consideration of specific features of the soil profile-such as erosion of the upper horizons significantly changes the results of the human health impact assessment. Obtained results show that the distribution of elements in soil profiles is characterized by an increase in Cu, Ni content with depth. Thus, dusting of the topsoil horizons can be considered as specific geographic conditions of the studied area. Models should be adapted and parameterized for different local conditions (Gentil et al., 2020).

Figure 9. Ni (A.), and Cu (B.) Impact score calculation modified (m) and Figure 10. Sampling locations (A.) and study area (B.)

default (d) calculated for different soil layers in the Background and

Impact area

Journal Pre-proof Note: h, [m] - height of the soil horizon, N - number of sampling point,

Distance [m] – distance from the source of pollution

Specific geographic conditions of the studied area do not allow the calculation of the standard height 492 of the soil horizon to be done. Due to the geographical conditions in the vicinity of Lake Imandra low 493 power podzols with poorly developed humus layer and litter are formed. These soils due to their depth 494 of ~0.1 [m] may correspond to the B horizon, which accumulates the main mass of chemical elements. 495 496 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 497 spread with the wind flow and consequently affect the local population. Environmental fate of the 498 pollutants as their distribution through the soil horizon should be connected with the human toxicity 499 assessment (Núñez and Finkbeiner, 2020). It is important to admit the geological and ecological 500 characteristics of the region in the impact assessment process. For example, for further impact 501 502 assessment research such site-specific characterization as landscape conditions or their technogenic disruption. 503

505 5. Conclusion

506

507 Studies of heavy metal pollution in the Kola Peninsula are relevant, in terms of assessing the state of 508 the environment and in terms of the impact on public health. Pollution of the surroundings of 509 Severonickel Plant by heavy metals is undeniable and is emphasized both by this work and by other 510 authors (Bazova, 2013; Evseev and Krasovskaya, 2017; Kashulina, 2018; Kyllönen et al., 2020; Liisa 511 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.

524

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526

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- Contaminated Arctic soils human health Impact assessment: A regional Life Cycle Impact Assessment approach
 A. Belyanovskaya, D. Vorobeva, N. Guseva, B. Laratte
- 5
- 6 Annex

CALCULATION of CHARACTERIZATION FACTORS Enter a single substance to see set of matrices below or enter range of rows to create series of results in 'Results multiple substances'.								
SINGLE SUBSTANCE RowNr Substance CAS	or FIRST ROW 24 Cr(III) 16065-83-1	FINAL ROW RowNr Substance CAS		Run Series	FIND A SUBS Search term: Search	Apply	RowNr Substance CAS RN	
Substance running RowNr Model Date of run	Cr(III) 24 USEtox® 2.02 [built 28-June-2016] 15-Oct-2021	REGION RegionRowNr 8 RegionName [®] Cen	IND Hom Itral Asia Occu	OOOR EXPOS neName C upationName I	SURE DECD countries ndustry, OECD	average	CROP RESID CropName	WES wheat

8 Figure 1. The USEtox model interface

9

7

- 10
- 11 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

- 13
- 14 Table 2. Square of sampling areas

Ν	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

Poin	Horizon	Distanc	h	Ni		Cu		Poin	Horizon	Distanc	h	Ni		Cu	
t		e [m]	[m	C in	IS	C in	IS	t		e [m]	[m	C in	IS	C in	IS
]	soils	[Daly/kg	soils	[Daly/kg]	soils	[Daly/kg	soils	[Daly/kg
				[ppm]	[ppm]					[ppm]	[ppm]
]]]]	
Impac	et 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	Substratu	2000	0.4	61.0	57	33.1	4		Substratu		0.4	16	4	50	0.6
	m			01.9		55.1			m			4.0		5.9	
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+		0.5		28		3.3		Subsoils		0.2		3		0.5
	Substratu			29.5		25.9						7.6		9.7	
	m						~~~								
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		Substratu m		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
	Substratu m		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								
	Substratu m		0.4	30.8	34	31.6	4.6								
2	Subsoils	27500	0.2	39.5	27	30.3	2.7	1							

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

B)



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





21 Figure 3. The factor loading (unrotated) analysis

- Table 4. The concentration coefficient (normalized to the mean of the sampling variety) of chemical
- elements in soils of the Impact area (a), and the Background area (b)

		Conce		
Ν		В	C	
	A2	B1		
		ipact area	<u> </u>	
2		$\begin{array}{l} Mo_{5,6}\text{-}Mn_{5,1}\text{-}Tl_{3,9}\text{-}Cs_{3,4}\text{-}C\\ V_{2,5}=Bi\text{-}Fe_{2,4}\text{-}Pb_{2,1}\text{-}Rb_{2}\text{-}C\\ Zn_{1,6}=In\text{-}Cu_{1,4}=Sb\text{-}Ni_{1,3}\text{-}i_{1,4}\text{-}i_{$	$c_{03,2}=Sn-Li_3-Ti_{2,7}-Cr_{1,9}-As_{1,7}=W=U-Sc_{1,2}-Hg_{1,1}=Th-Cd_1$	
5		$Ni_{2,8}=Cu-Cd_{1,8}-Co_{1,7}-Cr_{1}$ V _{1,4} =Ir=Hg=Th-Sc _{1,3} -Al ₁	,6=Bi-Re1,5- ,2=Fe=In=Sn-Li1,1	
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 V _{2,2} -As ₂ -Fe _{1,9} -Sn _{1,8} -Ti _{1,7} = Pt _{1,4} -Bi _{1,3} -Co _{1,2} -Mo _{1,1} =Co		
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	$Cd_{3,4}-Cr_{2,6}=Cu-Sb_{2,1}-Hg_{2}$ Al_{1,4}=V=Co=Ga=Ir=Bi-S As_{1,1}=Au-Pb_{1}	2= Ni-In _{1,8} -Fe _{1,6} - Sn _{1,3} -Sc _{1,2} =Os-	
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	$\begin{array}{l} Rb_{1,7} = Tl - Co_{1,6} - Ni_{1,5} - \\ Cu_{1,4} - Ba_{1,3} - Sc_{1,2} = Ti - \\ Li_{1,1} = V = Cr = Mn = Fe = Hg - \\ Al_1 \end{array}$
10	Pb2-Bi1,2- Cd1,1	$In_{2,7}-Au_{2,6}-Ti_{2,5}-Fe_{2,3}-V_{2,2}-Cr_{2,1}-Hg_{2}-Sc_{1,8}=Sn-Cs_{1,4}-Al_{1,3}=Tl-Co_{1,2}=Ga=Rb-Li_{1,1}-Cd=Lu-Mn_{1}$	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	I,1=V=Ag=Cs-Co1	
	1	The Back	ground area	
11		$Li_{1,4}$ -Fe _{1,3} -Ti _{1,2} =V- Sn _{1,1} -In ₁ =Bi	Li _{1,1}	
1		Na15-Sr5.6-K4.1-B4-Pd3.8-H	Be3-Hf2.8-As2.6=Ba-	

	Concentration coefficient							
Ν	A2	E	C					
		B1	B2					
		Zr2.5-Ca2.4-La2.3=Nd-Ce2.						
		Sm1.9=U-Ho1.8=Gd=Tb=						
		Pt _{1.6} =Yb=Er-Au _{1.5} =Tm-Z						
		Ta1.3=Ga=Ge=Nb-Cs1.2-S						
	Sn _{1,2}	Fe _{2,3} -In _{2,1} -V _{1,9} -	Li _{1,9} -Sc _{1,4} =In-Fe _{1,3} -	Li _{1,2} -Th _{1,1}				
13		Li1,7=Al=Sc=Ti-Pb1,5-	Al1,2-Ti1=Cr					
		Cr _{1,3} =Th-Ga _{1,2} =Hg-Bi ₁						
		Sr8.4-Sb8.3-I6.5-Be6.1-W5.7-						
		Nb4.6-M04.5-C84.3-Ga3.7-R						
		La _{3.1} =Hf-Rb ₃ =Zn=Ru-Na						
4		Cl _{2.6} -Ce _{2.3} =Pr-Pt _{2.2} -As _{2.1} =						
		Pb1.8=Ge-Eu1.7=Gd=K=T						
		Er1.5=Dy=Au=Ho=Tm-M						
		Cu _{0.3} -Ni _{0.1}						
		Zr21-Hf20=Nb-Ta17=Eu-P						
		U15=Gd=Tb=Dy=Ho=Ce						
3		Ag13=Pt=Ge-Sr12=Lu-Be						
5		As _{8.3} -Ba ₈ -B _{6.1} Ga _{5.3} -Mn _{4.9}						
		$Pb_{4,2}=Zn-P_{4,1}=Cs-Rb_{3,5}-A$						
		$Sn_{1.9}=B_1=L_1=C_1=Se-Ir_{1.8}=$	= 11 - 511.6 - 111.5 = Au - 11.4 - 52 - 52 - 52 - 52 - 52 - 52 - 52 - 5					
		$1111.2 = \Pi g = 1 M g - C01.1 = C u =$	-re-sc-s0411 NI 0.4					

25

26

27 Table 5. Mann-Whitney U Test

	Rank	Rank	U	Ζ	p-value	Ζ	2*1sided
	Sum I	Sum B					
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, $p_{0.05} = 14$							

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

29

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

N	N of sampl	ing The distance [km]	Height of the soil horizon [m]		
	point				
		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	l		
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		

32 Table 7. Ni and Cu emissions in the studied area

Area Impact a		ea Background		und	Deferrer ees	
Element	Ni	Cu	Ni	Cu	References	
Emission, ton/year	245	462			(Barkan and Lyanguzova, 2018)	

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	























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.

Declaration of interests

 \boxtimes 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: