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Influence of local geological data and geographical parameters to assess regional health impact in LCA. Tomsk oblast, Russian Federation application case

Alexandra Belyanovskaya^{1,2}  · Bulat Soktoev¹ · Bertrand Laratte³ · Elena Ageeva¹ · Natalia Baranovskaya¹ · Natalia Korogod⁴

Abstract

The research paper is aimed to modify the human health impact assessment of Cr in soils. The current article presents the input of several critical parameters for the human health Impact Score (IS_{hum}) assessment in soils. The modification of the IS_{hum} is derived using geological data — results of neutron activation analysis of soils are used in the IS_{hum} calculation; research area is divided using the watersheds and population size and density. Watersheds reflect the local environmental conditions of the territory unlike the administrative units (geographical areas of the studied region) due to their geological independence. The calculations of the characterization factor value underestimate the influence of the population size and density on the final result. Default characterization factor values cannot be considered during the assessment of the potential human health impact for the big sparsely inhabited areas. In case of very low population density, the result will be overrated and underestimated in the opposite case. The current approach demonstrates that the geographical separation in the USEtox model should be specified. The same approach can be utilized for other geo zones due to the accessibility of this information (area size, population size, and density, geological, and landscape features).

Keywords LCIA · Heavy metals · USEtox · Cr · Regional impact assessment · Impact assessment models

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Introduction

Human health impact from the soils assessment attracts a particular scientific attention (Trujillo-González et al. 2016a; Tarasova et al. 2018; Wang et al. 2020). Heavy metals input from the soils due to their direct and indirect long-term effects (Carr et al. 2008) is investigated worldwide (Yang et al. 2018; Islam et al. 2018; Jafari et al. 2019; Li et al. 2019; Tepanosyan et al. 2020).

A significant number of studies have been devoted to the role of soil as one of the factors contributing to disease occurrence (Alloway 2013; Li et al. 2019; Sevbitov et al. 2020; Tepanosyan et al. 2020). Chemical elements entering the human body through food chains originate, to a large extent, from soils (Brevik and Sauer 2015; Trujillo-gonzález et al. 2016b; He et al. 2021). It undoubtedly requires careful study of soils' elemental composition (Zhang et al. 2021) and the potential negative effects caused by chemical elements.

Cr as a heavy metal could have a serious negative impact on human health as cancer, skin diseases (Khitrov and Jaeger 2001; Guertin 2005; Balachandar et al. 2010; Danesh Miah

et al. 2010; Chatterjee et al. 2015; Sharma and Bhattacharya 2017; Sall et al. 2020; Sinha et al. 2022). The most widespread forms of the element in soils are Cr^{3+} , which is very stable in reducing terrain (Vodyanicky 2008; Ashraf et al. 2017), and Cr^{6+} , which is very unstable, easily mobilized, extremely toxic to living organisms (Brasili et al. 2020). Sources of Cr soil pollution can be both natural and man-made (Wuana and Okieimen 2011; Islam et al. 2018). The accumulation of Cr in soils depends on the local conditions. The main natural source of Cr to the environment is rock weathering (Ni et al. 2009; Tian et al. 2021). Major anthropogenic inputs of Cr into the environment include the emissions and wastes from chemical (pigments, metal finishing, leather tanning) refractory and metallurgical industries (Sheikhupura et al. 2016; Bashkin et al. 2019; Xia et al. 2019). These conditions should be considered in the processes of environmental monitoring. The importance of the local data determines the necessity of the spatially oriented investigations.

Assessing impact on human health (the Impact score - IS_{hum} calculation) based on the elemental composition of soils is one widespread approach (Senesil et al. 1999; Vithanage et al. 2019; Adimalla 2020), showing its relevance and importance for human health studies. In such studies, the gross values (mass or the concentration) of heavy metals in soils are commonly utilized (Bratec et al. 2019; Sharma et al. 2021). This approach is based on the multiplication of the gross values of the pollutant in the soils on the substance specific characterization factor — CF. The characterization factor is derived by the impact assessment models. CF represents the potential toxicity of a substance (Fantke et al. 2017, 2021). CF is used as weighting factors to aggregate life cycle emissions into scores for human health damage and ecosystem health damage (de Schryver et al. 2009).

Life cycle impact assessment models provide the human health impact characterization factor for organic and non-organic pollutants. IMPACT World + provides impact on human health, the regional separation is modeled using the USEtox regional archetypes (Bulle et al. 2019). ReCiPe provides the CF for the human non- and carcinogenic toxicity in country- or region-specific levels (Huijbregts et al. 2017). LC-IMPACT includes spatial aspects for the human health toxicity in country, continental, and a global level (Hauschild 2006; Huijbregts et al. 2016; Verones et al. 2020a).

All these models in case of the regionalized human health impact are based on the USEtox methodology. 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; Fantke et al. 2021). The USEtox model, recommended by the European Commission, has already proved its efficiency for the coupling of environmental and geochemical studies (Bratec et al. 2019) and was, therefore, chosen for our study.

USEtox model provides midpoint and endpoint characterization factors for human toxicological and freshwater ecotoxicological impacts of chemical emissions in life cycle assessment (European Commission. Joint Research Centre. Institute for Environment and Sustainability. 2011; Fantke et al. 2017). The USEtox model is constructed to provide characterization factors for human health and freshwater ecological damage for contaminant emissions to indoor air, urban air, rural air, freshwater, and agricultural soil (Fantke et al. 2017). The human health CF is given in the USEtox model is widely used in the Life cycle assessment (Walser et al. 2014; Liang et al. 2019; Luo et al. 2021; Alejandrino et al. 2021).

One of the main advantages of the USEtox is that the spatial aspect is considered at different scale (17 sub-continental regions); however, there is a lack of the local environmental information in the calculations (Shaked 2011). The LCIA is usually carried out in the regional scale (Hauschild and Potting 2000). At the same time, the environmental studies are performed at the local level. The model includes sub-continental regions, as “Central Asia,” which is heterogeneous from the geological point of view. The regional information in the USEtox about these areas is rather generic.

At the same time, previous investigations show that the environmental fate and the exposure of the chemicals at the local level strongly depends on the geological structure of the area (river flows, maternal rocks compositions, etc.) and the anthropogenic tension (Franke et al. 2005; Mirmiran et al. 2009; Kim et al. 2017; He et al. 2019; Verones et al. 2020b). These parameters are local, and they affect the environmental conditions of the research area, and consequently influence the population health.

The other spatial aspect of the impact modeling is the division of the geo areas. As it was previously mentioned, the USEtox model includes 17 sub-continental regions. The separation is organized based on the level of the administrative units. The previous investigations of the research group (Belyanovskaya et al. 2019, 2020) were also based on the administrative borders. However, such division of the regions in the impact assessment usually ignores natural conditions. Meanwhile, the watersheds reflect the landscape and the geological structure (Solntsev 2001; Kvasnikova 2003). Thus, we propose using the watersheds to reduce the regional limitation, mentioned above.

The other underestimated parameter of the model is a population density (PD). In the USEtox model, the population density of the area is rather generic. Population density depends on the type of the area, as rural or urbanized zones. For example, in Russia, the PD varies from 4956 person/km² in Moscow to 0.07 person/km² in Chukotka Autonomous Region (Federal State Statistics Service [Rosstat]). For the comparison, in the USEtox model in the continental scale, the PD of the “Central Asia” is 14 person/km² (Fantke et al. 2017). We suppose that overpopulated areas and the regions

with a low level of urbanization should be considered differently unlike it is performed in the model dataset.

The approach also considers the exposure of Cr with meat products through the food chain. In the article, the modification of the indirect exposure factor ($XF_{indirect}$) is developed. The paper is the continuation of the previous investigations of the research group. The indirect human exposure factor calculation modification was already developed earlier (Belyanovskaya et al. 2019, 2020).

The previous works were extended with the following:

- 1) The CF_{hum} calculated for the geologically oriented zones (watersheds) of Tomsk oblast' — to highlight the importance of the geological parameter in the LCIA, we compared the administratively divided zones with the interfluvies;
- 2) 78 samples of organs and tissues of the domestic pig were analyzed with the instrumental neutron activation analysis — INAA (in the previous research 18 samples analyzed with the inductive coupled plasma spectrometry - ICP-MS were proceeded);
- 3) Exposure factor $indirect$ modified using the local population density for each researched zone of the oblast'.

Exposure factor $indirect$ (XF) reflects the intake of chemicals through the meat products via soils (Rosenbaum et al. 2011; Fantke et al. 2017). The chemical composition of food, such as meat or dairy products, reflects the state of the environment (Baroni et al. 2011; Kim et al. 2017; Pu et al. 2017; Rikhvanov et al. 2019). Based on the biomagnification principle through the food chain, meat products reflect soil compositions as well. Thus, concentration of heavy metals in meat products can indicate the exposure of living organisms to them from polluted mediums (Kim et al. 2017; Panichev et al. 2021). This modification is

expressed in the article by exposure factor modification (Fig. 1).

To reflect the heterogeneity of geological conditions inside one geo zone, Tomsk oblast' in Russia was chosen for the case study. The method is based on indicators that are customized for each area without reference to a specific territory. Tomsk oblast' is used as an example of the current application case.

The region is an area with complex environmental conditions. It includes natural anomalies. There are manifestations of brown coal deposits, more than 100 hydrocarbon deposits and about 18% of Russian peat reserves (Arbuzov et al. 1999; Arbuzov and Ershov 2007). The main oil and gas districts of the region are Aleksandrovsky, Kargasoksky, and Parabelsky. There are also mineral ore deposits: iron ore Bakcharsky deposit (Nikolaeva 1967); zirconilmenite deposits (Rikhvanov et al. 2001) bauxite, zinc, gold, antimony, uranium, and many more (Evseeva 2001).

At the same time, the research results of A.M. Mezhibor (Mezhibor 2009) showed that the accumulation of elements in peat is influenced not only by natural factors but also by the impact of technogenic factors (Baranovskaya, 2011). The infrastructure of the Tomsk region is formed by more than 200 large and medium-sized industrial enterprises. The main sources of large-scale pollution in the Tomsk region are the Tomsk Petrochemical Plant (the largest in the Russian Federation), the Siberian Chemical Plant (in Russian SKHK), agro-industrial complexes (Mezheninovskaya, Tuganskaya, Tomsk pig farms), as well as industrial landfills and household waste (Krivov et al. 2019) ash dumps, quarries, treatment facilities, and others. Tomsk oblast' includes urbanized regions with high population density and sparsely inhabited zones.

The proposed approach is applicable for the further investigations with no direct connection with the research area.

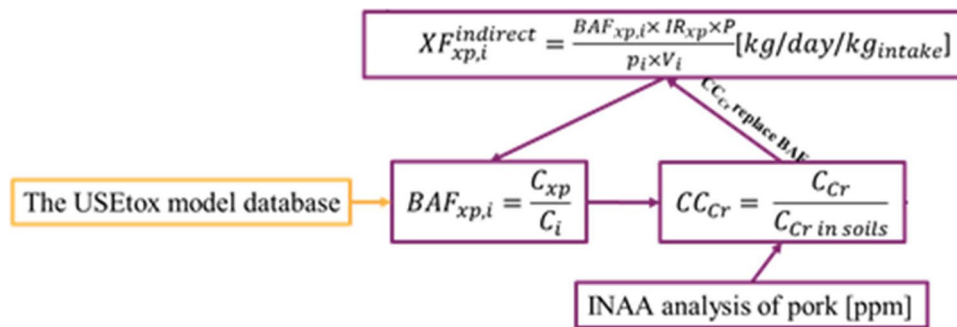


Fig. 1 The indirect exposure factor calculation (Fantke et al. 2017) and modification, (Belyanovskaya et al. 2019, 2020). $XF_{indirect\ xp,i}$, exposure factor; $BAF_{xp,i}$, bioaccumulation factor; IR , individual ingestion rate of a food substrate corresponding to exposure pathway xp ; P , population head count; p_i , bulk density of medium i [kg/m^3], V_i , volume of medium i linked to the exposure pathway xp $BAF_{xp,i}$,

bioaccumulation factor; C_{xp} , concentration of an element in the food substrate corresponding to the xp exposure pathway; C_i , concentration in the environment (soil, air); CC_{Cr} , concentration coefficient; C_{Cr} , concentration of Cr (C_{Cr}) [mg/kg_{xp}] in pork meat (according to the results of chemical analysis); $CC_{Cr \text{ in soils}}$, average concentration of Cr in soils; INAA, instrumental neutron activation analysis

The analytical data depend on the local conditions of the environment and the type of the chemical analysis. Different chemical analysis can provide a wide range of chemical elements in the natural environments under study. Geographical indicators such as population density and area size are available for each area.

The available analytical data allows us to characterize the degree of impact on human health, combining different methods of assessing the ecological condition of urban areas (Bratec et al. 2019): (1) geological approach and (2) impact assessment, which reflects the novel character of the study.

Study area

Tomsk oblast' is a region of the Russian Federation with a total area of 316.9 thousand km², located in the southeastern part of the Western-Siberian Plain on both sides of the Ob River (The official internet portal of the Administration of the Tomsk region, <https://tomsk.gov.ru/>). The climate is strong continental with a prevalent South-West wind destination (Fig. 2). The region includes a large sparsely inhabited area with non-developed infrastructure (e.g. the northern part). The southern part (Tomsk region - N 14, Shegarsky region – N 16, Kozhevnikovskiy region N 7 in Fig. 2) of Tomsk oblast' is an industrial center where the bulk of the population lives.

There are 6 types of soil in the Tomsk region. The North and Northeast of the region are composed of podzolic

and boggy soils. West and southeast are represented by sod-podzolic and swampy soils. The southern part of the region is formed by grey forest and chernozem soils. Floodplain soils form the area around the Ob, Chulym, and Tom rivers (Soil map of the Tomsk region, 1989).

Studies showed that the territory of Tomsk oblast' is characterized by significant geochemical heterogeneity due to both natural and technogenic factors (Rikhvanov et al. 2006; Rikhvanov et al. 2011). Another factor influencing the potential accumulation of chemical elements is cross-border transport carried by a dense river flows located in the region. Water masses move from the south to the north. Transit flow from the Kemerovo region (the rivers Tom, Yaya, and Kiya) and the Krasnoyarsk Territory (Chulym, the upper Ket, Cheti, and Tyma) make up 50% of the water flow.

This heterogeneity contributes to the accumulation of chemical elements by organisms living in this territory.

The investigation combines two environmental impact assessment approaches for spatial orientation: administrative units of Tomsk region and watershed areas. An interfluvial area is the territory situated between streams of rivers. These two types of spatial differentiation can be equally used in the impact assessment investigations. Administrative areas reflect the dominative type of industries developed in each district, meanwhile the interfluvial areas are comparable with soil types and geology. In addition, all the settlements of Tomsk oblast' are situated along rivers.

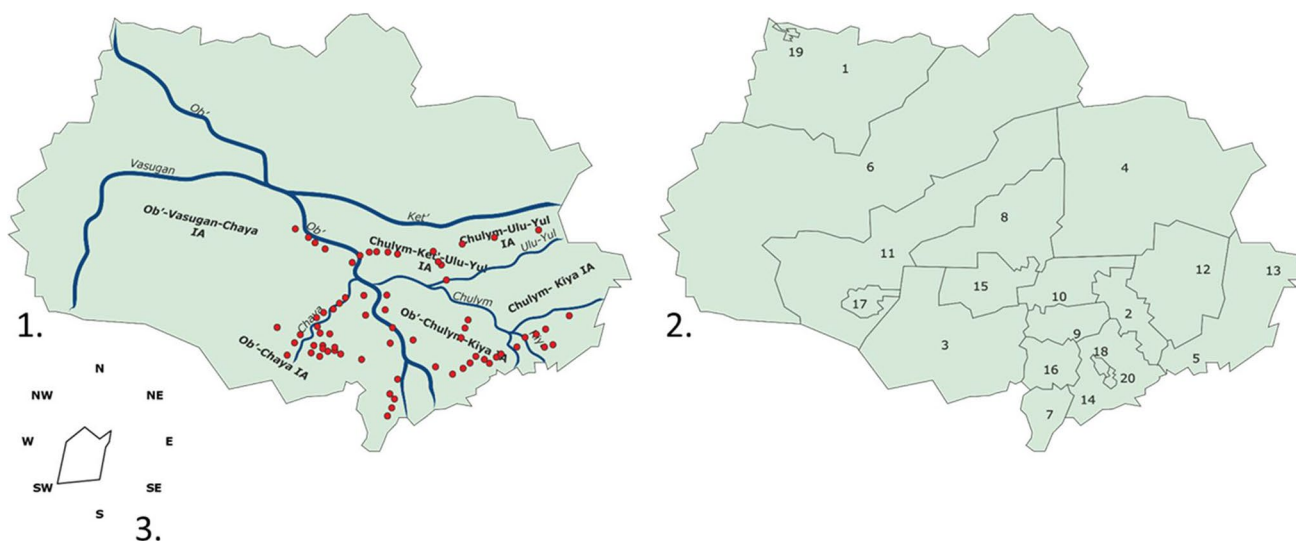


Fig. 2 The map of Tomsk oblast' with interfluvial areas (1), administrative map (2), and wind destination (3) ("Archive of weather in Tomsk," 2020) Note: Districts: 1. Aleksandrovsky; 2. Asinovsky; 3. Bakcharsky; 4. Verkhneketsky; 5. Zyryansky; 6. Kargasoksky; 7. Kozhevnikovskiy; 8. Kolpashevskiy; 9. Krivosheinskoy; 10. Mol-

chanovsky; 11. Parabelsky; 12. Pervomaisky; 13. Teguldetsky; 14. Tomsky; 15. Chainsky; 16. Shegarsky; 17. Kedrovyy; 18. Closed city Seversk; 19. Strezhevoy city; 20. Tomsk city Map 1: red points, sampling points; blue lines, river flows; map 2: administrative regions; 3: wind destination of the Tomsk oblast'

Material and methods

Soil sampling

Soil sampling was carried out in the last decade of April — the first 10 days of May, by the “envelope” method of five points, the depth of which was about 10 cm, i.e., the upper fertile layer. During the sampling, the presence or absence of organic fertilizer use was registered. Spot samples taken at the same sampling site were combined, thereby achieving their mixing and homogenization. Sample preparation included preliminary drying of the soil, removal of foreign matter. The dataset includes a total of 189 samples of soils of Tomsk oblast’.

For the investigation, the settlements of the Tomsk region located within the interfluvium of the Ob River and its large tributaries are considered. There are 6 inter-stream areas in Tomsk oblast’ and 47 settlements are included in the research area.

In the following table, the list of the districts included in the interfluvium is given, information about the number of settlements in each administrative district is given in the annex (Table 1).

As can be seen from Table 1, some interfluvium areas include settlements included administratively in the same district, others overlap administrative districts. We exclude the double-counting of the same districts by using the concentration of Cr from different villages.

Meat sampling

The sampling was carried out by the researchers from the Division for Geology of Tomsk Polytechnic University. The total quantity of samples is 78 samples of organs and tissues of 2 animals. Organs and tissues of 2 domestic pigs *Sus scrofa domestica* were taken from private farms in Tomsk region of Tomsk oblast’ (Russia).

The samples of organs and tissues of the domestic pig represent the whole organism of the animal and all parts that can be consumed as food. The food samples were taken on

private farms, raising animals for sale to the native population in the local markets.

The representability of biomaterials of domestic pigs as a sufficient environmental indicator was already presented in a previous investigation by the research group of Tomsk Polytechnic University (Baranovskaya and Rikhvanov, 2011; Belyanovskaya et al. 2019). Results of the chemical analysis are presented in the corresponding table, under the ‘Annex’ section (Annex, Table 2).

Sample preparation and analysis

Soil and pork samples were analyzed by Instrumental Neutron Activation Analysis (INAA). The INAA analysis has an advantage as a direct non-destructive analysis without chemical decomposition of samples. In the published papers, the accuracy of the analysis is proven (Arbuzov et al. 2016, 2019; Arbuzov 2017).

Sample preparation for the INAA takes place in several stages: a package of aluminum foil (size 3 cm x 3 cm), its pretreatment with an alcohol, a bag formation with tweezers, then weighing the foil bag (mg) on an electronic balance. A sample code is affixed to the bag, the sample is poured into bags on electronic scales to determine the weight of the sample (ideally 100 mg) and the total weight.

The analysis of the samples is carried out at the “IRT-T” research nuclear reactor in the nuclear geochemical laboratory of the Division for Geology of the National Research Tomsk Polytechnic University (accreditation certificate RA.RU.21AB27 of 04/08/2015). Analysis was carried out by A.F. Sudyko and L.F. Bogutskaya according to the instructions of NSAM VIMS No. 410-YAF.

The thermal neutron flux density in the irradiation channel is $2 \cdot 10^{13}$ neutrons (cm²·s), and the duration of sample irradiation is 20 hours. The measurements were carried out on a gamma spectrometer with a germanium-lithium detector DGDК-63A. The detection limit of Cr in soils by the INAA is 0.2 ppm.

Table 1 The list of regions settled in each interfluvium area

	Interfluvium area	Districts of Tomsk oblast	N
1	Vasyugan-Chaya	Bakcharsky, Kolpashevsky, Parabelsky	4
2	Ob'-Chaya	Bakcharsky, Kozhevnikovskiy, Molchanovskiy, Chainskiy, Shegarskiy	31
3	Chulym-Ulu-Yul	Pervomaiskiy	1
4	Chulym-Ket'-Ulu-Yul	Verhneketskiy, Kolpashevsky, Molchanovskiy	12
5	Ob'-Chulym-Kiya	Zuryanskiy, Asinovskiy, Tomskiy	11
6	Chulym-Kiya	Zuryanskiy, Teguldetskiy	10

N, the total number of settlements included in the interfluvium area

Calculation

Processing and generalization of the obtained analytical data was carried out on a personal computer using the office suite Microsoft Office (Excel, Word 2013) and the program “Statistica 7”. To build the graphic material, the software “Surfer 10” and “CorelDraw” were used. Samples were organized for Tomsk oblast, using both the administrative approach of spatial recognition and by considering the divisions according to the interfluvial areas to which they belong.

The statistical analysis

Statistical processing of data (with a reliability level of 95%) is carried out.

When calculating the average contents of elements from the total sample, “hurricane samples” were removed, but they are shown in the scatter of values. When some elements were present in concentrations below the detection limit of the analysis, half of the threshold value is used in the calculation (Mikhalechuk and Iazikov 2014). Regardless of the nature of the distribution of elements, we took the

arithmetic mean values of the sample as average levels of content, which, with both normal and asymmetric distribution, gives the most consistent estimate of the concentration values (Tkachev and Iudovich 1975).

The significance of the differences in the sample sets is estimated using the Kolmogorov-Smirnov statistical non-parametric analysis method. The differences were considered significant at a p -level $p < 0.001$.

Methodology for assessing the toxicity of elements

The basics of the USEtox model developed on the Microsoft Excel platform (Fig. 1, Annex).

According to the USEtox, Impact Score (IS_{hum}) (Fig. 3, formula I) is a LCIA impact score used for characterizing human toxicity that is expressed as a number of cancer or non-cancer disease cases at midpoint level and as a number of disability-adjusted life years [DALY] at endpoint level (Fantke et al. 2017).

Midpoint indicators focus on single environmental problems in the case-effect chain, endpoint indicators show the environmental impact on higher aggregation levels (Bare et al. 2000; Huijbregts et al. 2016).

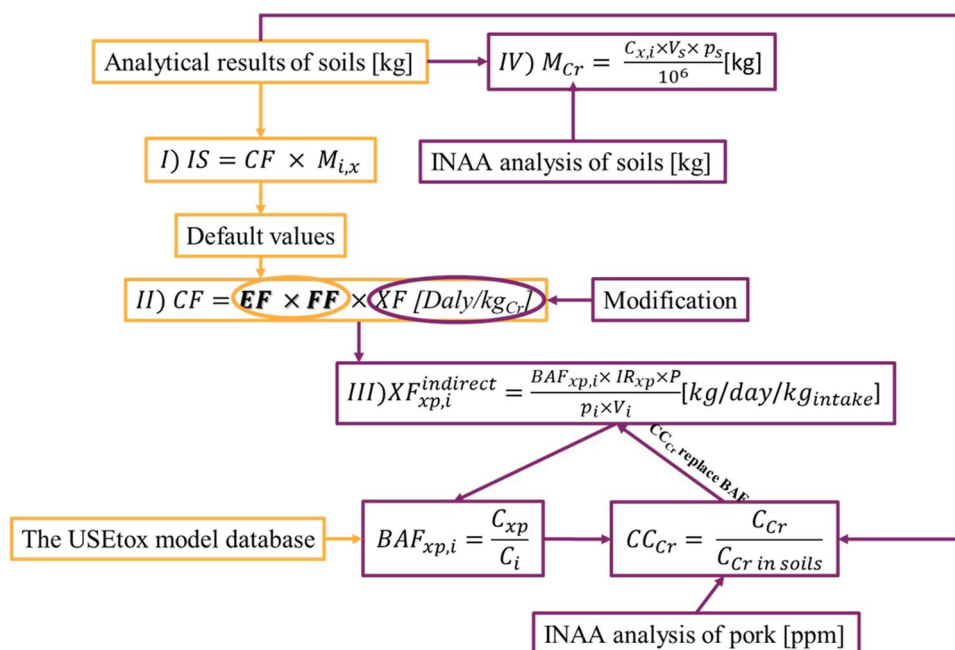


Fig. 3 The framework of the investigation, according to (Fantke et al. 2017; Belyanovskaya et al. 2019; Bratec et al. 2019). Roman numbers on the figure refer to formulas. (I) IS, impact score; CF, characterization factor, $M_{i,x}$, the total mass of the element. (II) CF, characterization factor; EF, effect factor; XF, exposure factor. (III) $XF_{indirect xp,i}$, exposure factor; $BAF_{xp,i}$, bioaccumulation factor; IR, individual ingestion rate of a food substrate corresponding to exposure pathway xp; 4) P, population head count; 4) p_i , bulk density of medium i [kg/m^3], V_i , volume of medium i linked to the exposure pathway xp; 5)

$BAF_{xp,i}$, bioaccumulation factor; 6) C_{xp} , concentration of an element in the food substrate corresponding to the xp exposure pathway; 7) C_i , concentration in the environment (soil, air); 8) CC_{Cr} , concentration coefficient; 9) C_{Cr} , concentration of Cr (C_{Cr}) [mg/kg_{xp}] in pork meat (according to the results of chemical analysis); 10) $CC_{Cr \text{ in soils}}$, average concentration of Cr in soils. (IV) M_{Cr} , total mass of the element; $C_{x,i}$, Cr concentration in agricultural soils; V_s , soils volume; 4) p_s , bulk density of soils [kg/m^3]

The IS_{hum} for potential impacts of Cr is calculated using a weighted summation of pollutants released from potential pollution sources and characterization factors for the damage (Fig. 3, formula I). The characterization factor needed for the impact score calculation was derived using the USEtox model dataset.

For the modification of the characterization factor and the Impact score, two types of local samples are used.

- 1) Tomsk oblast' soils. These are used for the characterization factors and the impact score calculation and modification (Fig. 3, formulas I, II);
- 2) Pork sampled in two settlements of Tomsk oblast'. This data is extrapolated into the exposure factor modification (Fig. 3, formula III).

The total mass of the element in soils (M_{Cr}) is calculated according to the formula developed by (Bratec et al. 2019) (Fig. 3, formula IV).

Where:

- $C_{x,i}$ is the concentration of Cr in agricultural soils in each studied area. $C_{x,i}$ is taken from the own analytical results;
- p_s is the bulk density of soils, which is the table value taken from the USEtox documentation [kg_{soil}/m^3_{soil}];
- V_s is the volume of soils of each considered region [m^3].

The characterization factor (CF) is calculated by USEtox documentation. Characterization factor for the potential human health damages at the endpoint is expressed in DALY/kg_{emitted} – disability adjusted years per kg_{emitted}. The CF is derived as the multiplication of three factors:

Effect factor (EF) [kg_{intake}/day] reflects the impact on human health due to the arrival of a chemical element substance in the living organism in various ways (through air, water, soil, or food).

Fate factor (FF) [$kg_{in\ compartment} \text{ per } kg_{emitted}/day$] represents the persistence of a chemical in the environment (e.g., in days) as well as the relative distribution.

Exposure factor (XF) [$kg_{intake}/day \text{ per } kg_{in\ compartment}$] describes the effective human intake of a specific environmental medium– soil – through ingestion.

In the current investigation, we use the default values of effect and fate factors, but with the modification of exposure factor using the analytical data of the concentration of Cr in the pork meat (Belyanovskaya et al. 2019). This reflects the effective intake of elements via soil or air into the human body when eating meat products.

The exposure factor is calculated according to the formula given in the framework of the investigation (Fig. 3, formula 5), where the bulk density (p_i) of the soils and the individual consumption rate (IR) are tabular values and are taken from

the model for calculation. The volume of soils ($V_i [m^3]$) is calculated by the following formula:

$$V_i = h_i [m] \times S_i [m^2]$$

Formula 1: The volume of medium i calculation. Where, $h_i [m]$ is the height of medium i (continental and global air, or soil), the table value presented in the model, and $S_i [m^2]$ is the area of agricultural soils, depending on the geographical features of the studied region.

In order to take into account the environmental features of the region, the bio-transfer factor (BTF) is replaced by the ratio of the concentration of Cr (C_{Cr}) [mg/kg_{xp}] in pork (according to the results of chemical analysis), and the concentration of Cr in soils of each studied region (Fig. 3, formula 6) [mg/kg]. Where, the bio-transfer factor BTF [$days/kg_{substrate}$] is the steady-state ratio between the concentration $C_{substrate}$ in meat or milk respectively and the intake i of a chemical (Cr) by the animal.

In previous investigations, the concentration ratio was normalized to the percent abundance (Glazovsky clarke of Cr in biosphere (Glazovsky 1982)). In the current investigation, the Cr concentration in soils is taken for the ratio.

Official data is used for the total square footage and population calculation of the administrative areas and interfluves (2020). The area between the biggest rivers of Tomsk oblast, forming the interfluves, is used for the square footage calculation. The geographical information obtained is given in the annex section.

Results

Statistical analysis results

Results of statistical analysis demonstrate abnormal distribution of Cr concentration in soils of Tomsk oblast, according to the variation coefficient (V) (Table 2), where $V=70\%$. In most samples, Cr concentration ranges from 65 to 116 ppm.

The literature analysis shows that the average concentration of Cr in Tomsk region topsoil is above most of the literature values (Fig. 4).

The database was divided into groups — interstream areas. The maximum values of Cr content are detected in the soils of (the Southeast part of Tomsk oblast) — the Chulym-Kiya interfluve area (Table 3, Fig. 5). The Chulym-Ulu-Yul interfluve is highlighted as an area with the lowest Cr content. However, it can be noted, that only one settlement and 5 samples from these zones were studied. Topsoil of the South-West part of Tomsk oblast' (Ob'-Vasugan-Chaya and Ob'Chaya interfluve zones) also contains low Cr concentrations.

Table 2 The statistical parameters of chromium concentrations in soils of Tomsk oblast

Cr total	Mean	St. error	Median	Mode	St. dev.	Min	Max	V	N
	119	6	98	94	84	13	618	70	189

mean, arithmetical mean [ppm]; *St. error*, standard error of the arithmetical mean; *St. dev.*, standard deviation of the arithmetical mean; *min*, minimum value [ppm]; *max*, maximum value [ppm]; *V*, variation coefficient, [%]; *N*, total number of samples

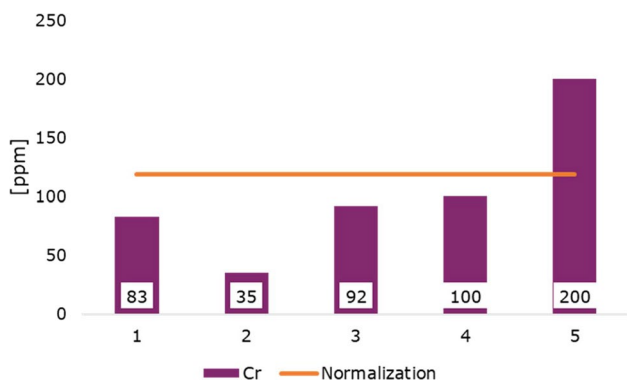


Fig. 4 Diagram of Cr distribution in soils of Tomsk oblast, 189 samples in total, [ppm]. Note: 1. Lithosphere Clarke (Vinogradov 1962); 2. Lithosphere Clarke (Taylor and McLennan 1985); 3. Lithosphere Clarke (Grigorev 2009); 4. World soils (Bowen 1979) 5. World soils, (Vinogradov 1957); Normalization is analytically obtained data

Using ranges of chemical elements, we can see that in the southeast side of Tomsk oblast, soils concentrate more Cr than samples taken in the south-western part.

Soils taken in the southern part of Tomsk oblast' accumulate more Cr than in the northern part of the oblast.

However, the maximum total concentration coefficient of chemical elements in soils is observed in samples of areas around the Ob' river and the Chulym-Ulu-Yul interstream area. Besides Cr, there is intensive accumulation of other heavy metals (As, Fe, Co), radioactive elements (U), rare earth elements (Lu, Yb, La, Ce, Eu), noble-metals (Ag, Au), and Sb, Sr, Br (Table 4).

Accumulation of elements forming significant correlations with Cr, whose accumulation is noted in most of the regions studied (Table 5). The elements having the strong positive correlation with Cr are accumulating in soils as well, and their potential toxicity should be considered for the impact assessment.

Comparative analysis of correlations between chemical elements allows the differences between elemental interactions in soils from areas around different rivers to be clearly seen. Cr forms positive and negative correlations with heavy metals whose toxicity is underestimated (Sb, Ba). In soils taken from the southeast side of Tomsk oblast, rare earth

Table 3 The statistical parameters of the chromium concentration in soils sampled in interfluvial areas of Tomsk oblast'

Interfluvial areas	S	Mean	SE	Me	Mo	SD	Min	Max	V	N
Chulym-Kiya	6608	219	27	190	N/d	144	14	619	66	29
Ob'-Vasyugan-Chaya	88780	94	9	93	N/d	27	51	144	28	8
Ob'-Chaya	26141	97	4	97	108	35	38	173	36	62
Chulym-Ulu-Yul	24391	33	5	37	N/d	11	21	44	34	5
Chulym-Ket'-Ulu-Yul	24391	137	14	129	N/d	64	42	298	47	20
Ob'-Chulym-Kiya	20327	99	7	89	N/d	53	14	619	62	65

S, square footage [km²]; *mean*, arithmetical mean [ppm], *SE*, standard error, *Me*, median, *Mo*, mode, *SD*, standard deviation, *min*, the minimum value [ppm], *max*, the maximum value [ppm], *V*, coefficient of variation [%], *N*, the number of samples, *N/d*, no data

Fig. 5 The distribution of Cr in soils sampled at the interfluvial areas of Tomsk oblast, [ppm]

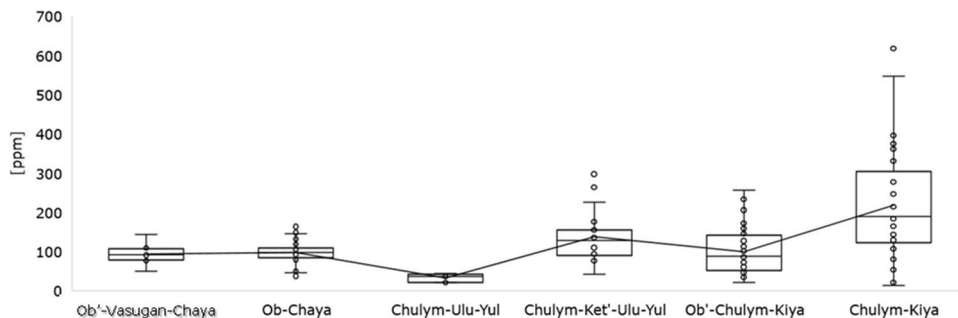


Table 4 The values of the concentration coefficients of chemical elements relative to the average content of elements in the sample

Interfluvial area	Chemical elements	$\sum CC > 1$
Chulym-Kiya	$As_{1.4}-Cr_{1.2}=Au-Ca_{1.1}-Na_1=Fe=Sb=Yb=Lu=U=Ba=Sr$	12.9
Ob'-Chulym-Kiya	$Au_{1.2}-Sb_1=Lu=Fe=Sc=Ba=U=Yb=Na=La=Cr=Ca=Rb$	12.2
Chulym-Ket'-Ulu-Yul	$Sr_{1.6}=Ag-Hf_{1.2}-Na_{1.1}=Ba-Cr_1$	7.6
Ob-Chaya	$Br_{1.7}-Ta_{1.5}-Co_{1.3}=Th=Ce=Sc_{1.2}=Fe=Rb=La=Ce=Sm=Tb=Eu-Ag_{1.1}=Yb=Lu=U=Hf-Na_1=Sb=Ba=Sr...-Cr_{0.8}$	15.5
Ob'-Vasugan-Chaya	$Ag_{1.8}-Hf_{1.6}-Eu_{1.4}-Na_{1.2}=Br=Ce=Th=Ba=Ce=Sc=Fe=Co=Sm=Tb-La_1=U=Sr-Ta...-Cr_{0.8}$	15.3
Chulym-Ulu-Yul	$Sr_{2.2}-As_{2.1}-Ca_2-Rb_{1.7}-Sb_{1.6}-Lu-Au_{1.5}-Na_{1.4}-Yb_{1.3}=Ba=Tb-Ta-U_{1.2}-La_1=Sm=Hf...-Cr_{0.3}$	19.3

$\sum CC > 1$, the sum of the concentration coefficient exceeding 1

Table 5 Correlation interactions in soils sampled in Tomsk oblast according to the Spearman method, $p_{0.05}$

N	Interfluvial area	Correlation with Cr	
		Positive correlation	Negative correlation
1	Chulym-Kiya	-	Br, Eu
2	Ob'-Chulym-Kiya	Ba, Eu, As	Na, Ca, Rb, Br, Sm, Yb, Lu, U, Au, Sr
3	Chulym-Ket'-Ulu-Yul	Sr	Au
4	Ob-Chaya	Ce, Ba, Tb, Eu, As	-
5	Ob'-Vasugan-Chaya	Sb, Au	
6	Chulym-Ulu-Yul	Tb	

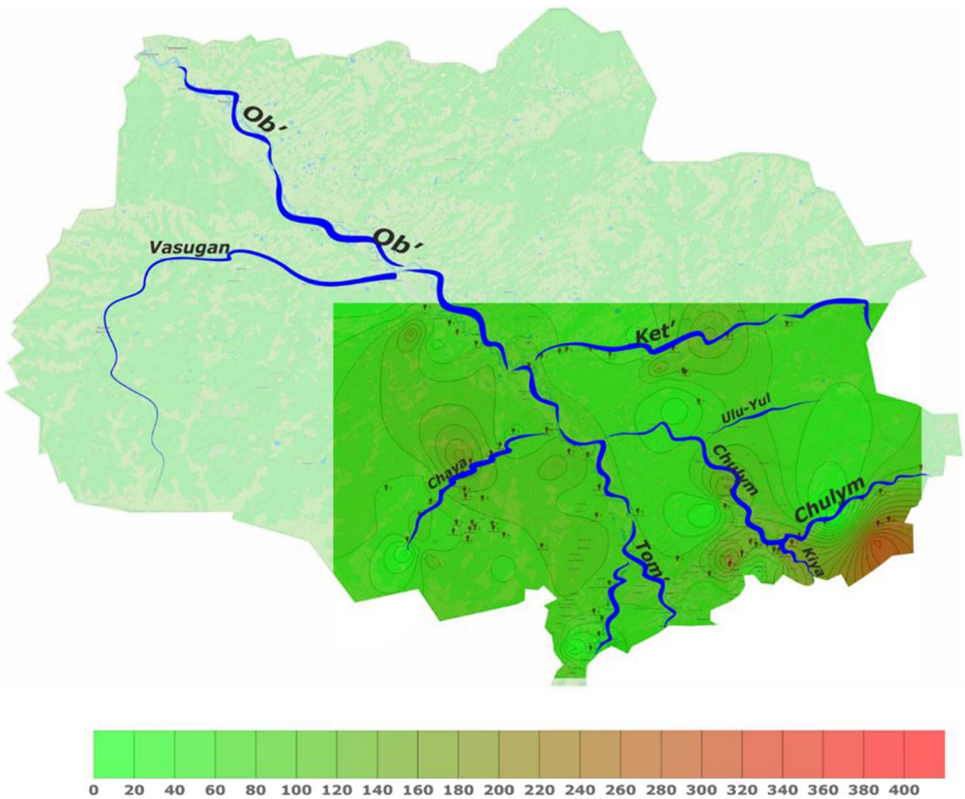
r , critical value of the Pearson correlation

elements, Au, Ba, As, and Sb create a significant positive correlation with Cr.

The graphical interpretation as distribution map is the common approach to estimate the potential sources of the heavy metals and soil qualities (Duan et al. 2020; Fathizad et al. 2020; Zhang et al. 2021). The Cr distribution in soils of Tomsk oblast' (Fig. 6) allows us to see the spatial distribution. We can observe the main halo of the Cr accumulation in the southeastern part of the oblast around the Ulu-Yul, Chulym and Kiya rivers. The area with the highest concentrations of Cr is a border area with Kemerovsky oblast.

Areas of elevated Cr concentration cross four border districts of Tomsk oblast': Pervomaisky, Teguldetsky,

Fig. 6 Map of the total concentration of Cr in the territory of Tomsk oblast, [ppm], scale 10 to 1



Asinovsky, and Zuryansky. Soils sampled from the northern part of Tomsk oblast' contain lower concentrations of Cr, but with some exceptions. There are halos with medium concentrations of Cr in agricultural soils above the Ket' river divided between Verkhneketsky, Kolpasevsky, and Parabelsky districts.

Points of high Cr concentration are situated on the border areas between different administrative units with no connection with internal division of Tomsk oblast, which confirms the advantages of using the interfluvial spatial organization of the research.

Comparing the distribution of Cr in soils according to the administrative divisions of Tomsk oblast', a certain heterogeneity of concentration is also seen (Fig. 7).

The central part of Tomsk oblast', Bakcharsky, Krivosheinsky, and Shegarsky districts are the areas where the lowest variation coefficient is observed. The biggest variation in the dataset is found in the Teguldetsky district.

As discussed before, the heterogeneity of given results is probably connected with the mixed environmental condition of the region, caused by the mix of anthropogenic influence and natural conditions.

The human health characterization factor the Impact score assessment

The results of our studies show that soils of Tomsk oblast' accumulate a significant number of elements, including toxic and conditionally toxic ones, such as Cr, in the impact area, 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.

The modified characterization factor varies significantly between different interfluvial areas (Fig. 8). The sequence of the factor does not correlate with the value of the Cr concentration in soils. Investigated zones are ranged in the following order according to the value of the human health CF: Chulyum-Ulu-Yul > Ob' - Chaya > Ob' - Chulyum - Kiya > Chulyum-Ket' - Ulu-Yul > Chulyum-Kiya > Ob' - Vasugan-Chaya.

The modified human health characterization factors for Cr vary from the CF proposed by the USEtox model. However, for the soils of the Ob' - Chulyum-Kiya and Ob' - Chaya interfluvial areas, the values of modified human health CF are close to the default characterization factor.

The human health impact score was calculated using the modified and default characterization factors (Fig. 9).

Fig. 7 The distribution of Cr in soils according to the administrative divisions of Tomsk oblast', [ppm]. Districts: 1. Asinovsky region, 2. Bakcharsky, 3. Verkhneketsky, 4. Zyryansky, 5. Kozhevnikovskiy, 6. Kolpashevskiy, 7. Krivosheinskoy, 8. Molchanovskiy, 9. Parabelskiy, 10. Pervomaiskiy, 11. Teguldetsky, 12. Tomskiy, 13. Chaynskiy, 14. Shegarskiy

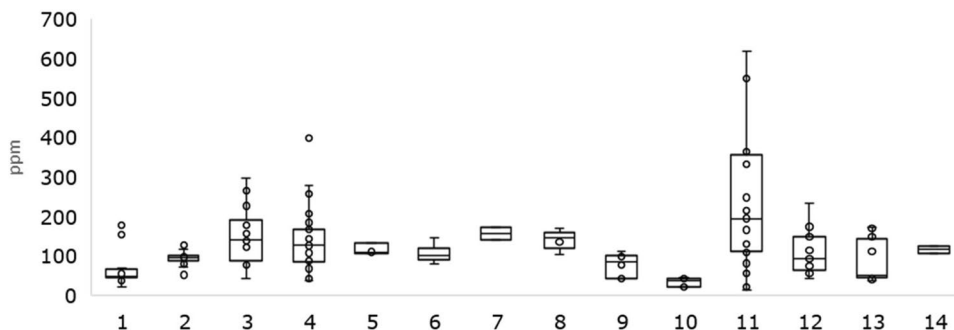


Fig. 8 The human health characterization factors - CF_{hum} (modified and default) of Cr in interfluvial areas, soils [Daly/kgCr]. CF_{hum} default is the table value provided in the USEtox model. 1. Chulyum-Kiya, 2. Ob' - Chulyum-Kiya; 3. Chulyum-Ket' - Ulu-Yul; 4. Ob' - Chaya; 5. Ob' - Vasyugan-Chaya; 6. Chulyum-Ulu-Yul

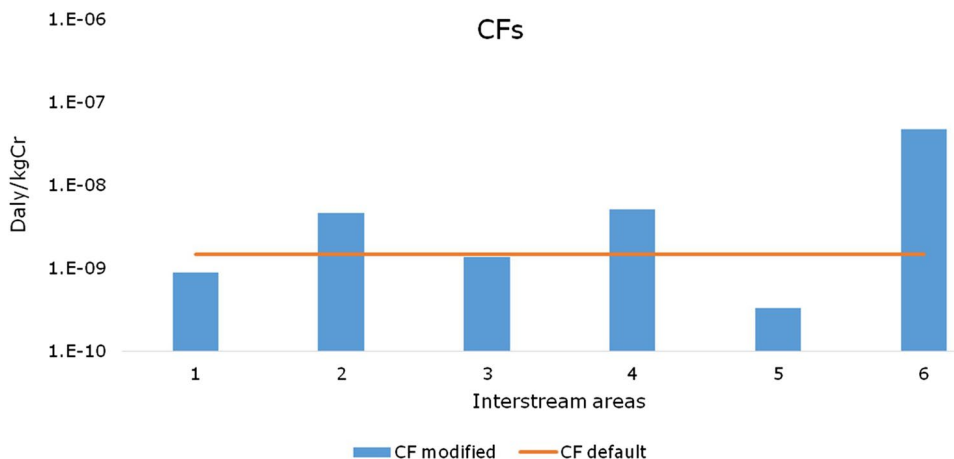
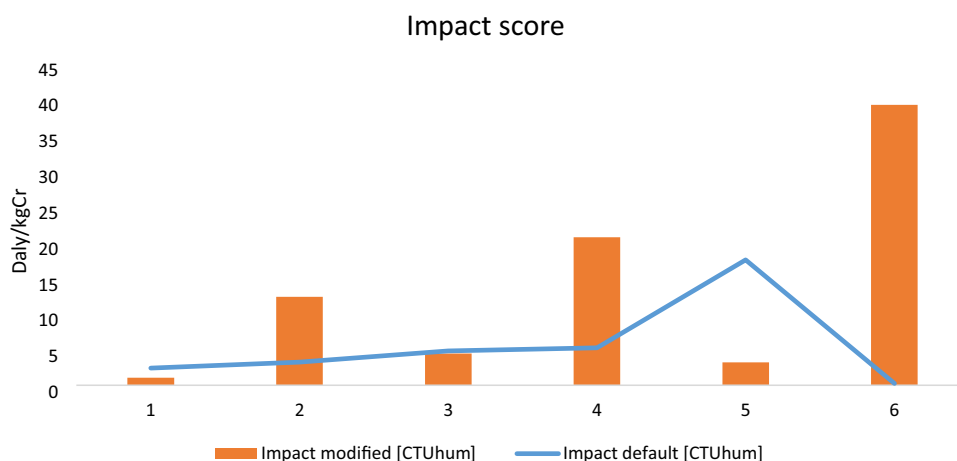


Fig. 9 The human health impact score modified and default of Cr in interfluvial areas, [Daly/kgCr], Lg scale. 1. Chulym-Kiya; 2. Ob'-Chulym-Kiya; 3. Chulym-Ket'-Ulu-Yul; 4. Ob'-Chaya; 5. Ob'-Vasyugan-Chaya; 6. Chulym-Ulu-Yul



We can see that the Chulym-Ulu-Yul interfluvial area has the highest Impact score among all studied districts of Tomsk oblast. The impact score value is determined by the concentration of Cr in soils. The source of soil pollution in settlements below the Chulym river is situated in the south-eastern part of Tomsk oblast.

The impact score calculated with the modified characterization factor is also higher than the default value. Significant variation between default and modified results is observed for the Ob'-Vasyugan-Chaya and Chulym-Ulu-Yul interfluvial areas. These zones are characterized by the high heterogeneity of the population density. We can assume that the USEtox model underestimates the value of the characterization factors for big, sparsely inhabited areas.

Considering the limitation of the square footage of each interfluvial determination, the calculation of CF and IS according to administrative divisions was performed (Figs 11 and 12). The population density of studied areas is available in the official governmental sources.

To simplify the reading of the resulting charts, we repeat the map of Tomsk oblast' (Fig. 10).

Making a comparison between the values of the characterization factors (Fig. 11) calculated for the same region, but on the basis of administrative spatial divisions, we can observe that for Tomsky (Ob'-Chulym-Kiya interfluvial) and Verkhneketsky (Chulym-Ket'-Ulu-Yul) regions modified factors are lower than default data. Other modified values are higher.

The similar trend is observed for the impact score calculation, but for Tomsky and Pervomaisky districts (Fig. 12). These zones are situated in the eastern part of the Tomsk oblast around the Ob' and the Chulym rivers.

Proving that the administrative divisions-oriented approach to the impact assessment is too generic, we can make a conclusion. In the resulting map (Fig. 6), we see that only some parts of the regions have soils with a high concentration of Cr. Similar environmental conditions are artificially divided into 4 zones. This division affects the conclusions about the state of the environment in the whole district.

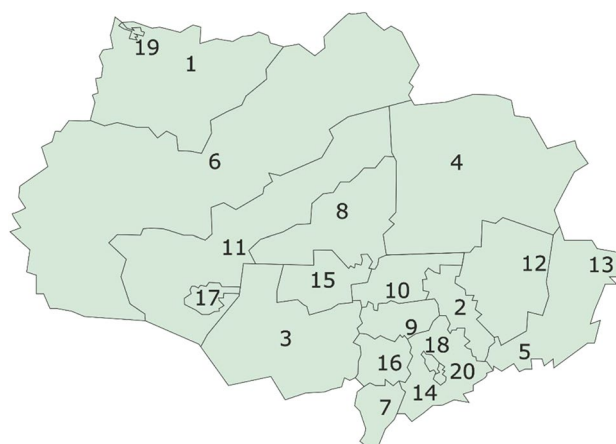


Fig. 10 The administrative map of Tomsk oblast'. Districts : 1. Aleksandrovsky; 2. Asinovsky; 3. Bakcharsky; 4. Verkhneketsky; 5. Zyryansky; 6. Kargasoksky; 7. Kozhevnikovsky; 8. Kolpashevsky; 9. Krivosheinsky; 10. Molchanovsky; 11. Parabelsky; 12. Pervomaisky; 13. Teguldetsky; 14. Tomsky; 15. Chainsky; 16. Shegarsky; 17. Kedrovyy; 18. Closed city Seversk; 19. Strezhevoy city; 20. Tomsk city

Discussions

There are plenty of factors affecting Cr as a heavy metal concentration in soils, such as use of fertilizers, atmospheric deposition, industrial emissions, and soil parent materials (Ke et al. 2017; Lv and Wang 2018; Hedberg et al. 2019).

In the current application case, we can highlight two factors as well:

1. Man-made sources. Anthropogenic sources of Cr pollution are situated mainly in the south of Tomsk oblast'—Tomsk region. Soils of the Tomsk region, as a zone of increased technogenesis, have been studied in the most detail (Zhornyyak 2009). The average Cr content of

Fig. 11 The human health characterization factors - CF_{hum} (modified and default) of Cr in areas of Tomsk oblast', soils [Daly/kgCr], Lg scale. Lg, logarithmic scale. 1. Asinovsky; 2. Bakcharsky; 3. Verkhneketsky; 4. Zyryansky; 5. Kozhevnikovsky; 6. Kolpashevsky; 7. Krivosheinsky; 8. Molchanovsky; 9. Parabelsky; 10. Pervomaisky; 11. Teguldetsky; 12. Tomsky; 13. Chainsky; 14. Shegarsky

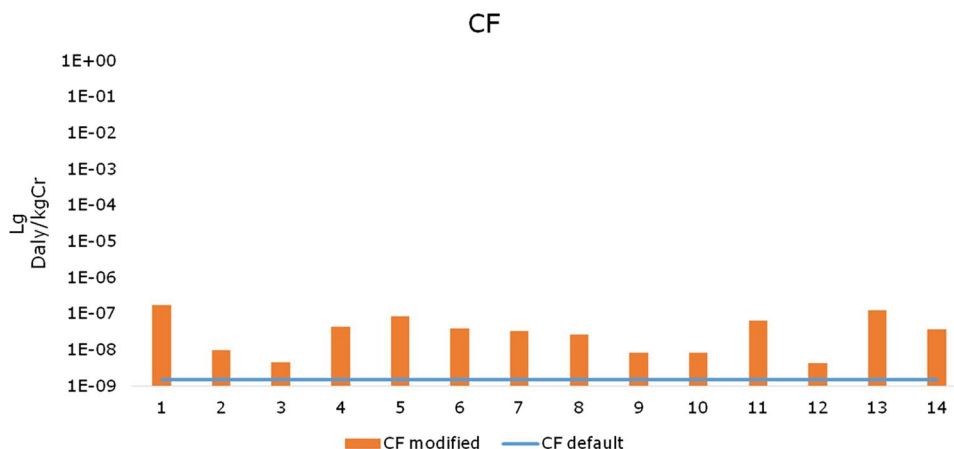
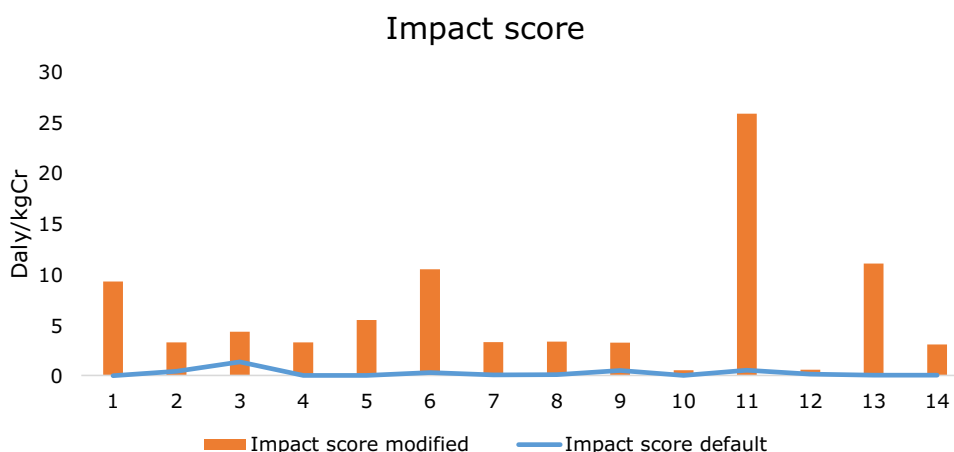


Fig. 12 The human health impact score (modified and default) of Cr in areas of Tomsk oblast', [Daly/kgCr]. 1. Asinovsky; 2. Bakcharsky; 3. Verkhneketsky; 4. Zyryansky; 5. Kozhevnikovsky; 6. Kolpashevsky; 7. Krivosheinsky; 8. Molchanovsky; 9. Parabelsky; 10. Pervomaisky; 11. Teguldetsky; 12. Tomsky; 13. Chainsky; 14. Shegarsky



Tomsk city soils is 104 ppm (Zhornyak 2009), the own results is 1.1 times higher. Also comparing analytical results with abundance ratios of Cr in lithosphere, we established a high level of Cr cumulation in the studied topsoil. The south-eastern halos of the concentration have also probably an anthropogenic nature. We can assume that the most polluted point in the border area with Kemerovo oblast could relate to the wind transfer of pollutant from the opencast coal mines (Arbuzov 2017) situated in the area to the south (“Weather archive in Kemerovo for months and seasons,” 2020).

2. Natural sources. There are several geological factors of Cr accumulation to be noted. According to V. Goldshmidt, Cr is correlated with Fe in geochemical processes in the environment (Goldshmidt 1952). Halos of Cr concentration in the left bank of Ob' river reflect the bedding of the iron-ore assets. The south of Tomsk oblast' is formed with the Tom'- Koluva' bow area, where magma is not covered with the platform mantle and can be a source of Cr in soils (Milanovskii 1996). As the content of clay minerals and organic matter increases in the southern part of Tomsk oblast', the content of heavy metals in the soil increases (Tu et al. 2011; Jia

et al. 2020; Qu et al. 2020). Research on the heavy metal content of forage fodder and topsoil of the southwest part of Tomsk region demonstrated low accumulations of heavy metals in grey forest soils (Nikolaeva 2020).

The results show the trend of the Cr accumulation in all parts of the studied region. Gross concentrations of elements in soils do not express the level of the impact on the public health. Although soils are a critical repository for numerous deleterious pollutants (Doyi et al. 2018), which could be transferred to plants, living organisms, and groundwaters (Alloway 2013; Nag et al. 2022). Polluted soils could be the source that poses the greatest threat to human health (Cuajungco and Lees 1998; Bare 2006; Başaran et al. 2012; Brown et al. 2016; Turbinsky and Bortnikova 2018; Wang et al. 2018; Shen et al. 2021). Correlation of Cr content in soils of Tomsk region with prevalence of sarcoidosis is proven (Denisova et al. 2016). Thus, the accumulation of Cr in soils, its monitoring, and mapping has great importance for the human health impact assessment.

Using the LCIA model allows us to evaluate and predict the impact. Analyzing obtained results, we can make several conclusions.

Use of impact assessment models often leads to disparate results, and therefore we observe the contradiction between modified and default factors. The difference between the characteristic factors of the model modified with analytical data and the default results is mentioned in other scientific studies (Adam et al. 2013; Hedberg et al. 2019; Xiao et al. 2020) including our own case studies (Belyanovskaya et al. 2019, 2020).

The obtained results differ from the model data to a greater or lesser extent. However, the impact of analytically obtained local data is undeniable. In this paper, we observe a strong disparity between the characterization coefficients modified with our data and the ones proposed in the default model. The same disparity is observed for the impact score. We assume that this dispersion highlights the contradiction between generosity of the USEtox model dataset and the high specificity of analytically obtained data.

The modified exposure factor provides information about chemical components in food consumed in the region. The possibility that the state of the environment may significantly affect the content of pollutants in meat or dairy products is underestimated in the model. The USEtox includes the different types of meat products depending on such parameters as the level of fat in meat. However, this information is rather generic. The pollutant transfer mechanisms through the trophic chain still needed to be investigated. In the USEtox dataset, the bio-transfer factor expressing the accumulation of the elements does not vary according to the region. Thus, the BTF given for the “Central Asia” region is generic for all areas.

Continental levels of CF of soils given in the USEtox cannot reflect the environmental features of local areas. The values of CFs_{default} and consequent ISs depend on many parameters like the chemical properties of elements, their effect on human organisms, and geographic features of the region. We see that the population density is the most critical parameter affecting the results. (Annex, Table 8). In this case, using the administratively organized spatial orientation is a better approach, because the square footage and the population is updated every year by the government. At the same time, this type of spatial organization does not reflect the landscape, geology, and ecology of the area. Natural, historically formed, environmental conditions apply to the zones of a given geological formation, with similar geographic conditions and climate. Applying only the administrative approach, the potential toxicity score is artificially divided between regions. In this case study, interfluves represent the naturally made locations.

The current investigation suffers from several limitations of sampling and data processing technique:

I. Limitations in representativeness of the statistical dataset:

1. One settlement from Chulum-Ulu-Yul interfluve area is taken for the investigation (Pervomaisky village);
2. Two soil samples were taken in the Krivosheinsky and Shegarsky districts, and three in Kozhevnikovskiy;
3. The indirect exposure factor modification is carried out using analytical data about Cr content in 78 organs and tissues of 2 pigs;

II. Limitations in the indirect exposure factor calculation:

1. The Cr content in the pork taken for the indirect Exposure factor modification is the same for each settlement of Tomsk oblast — it might reduce the accuracy of the local data;
2. There is uncertainty in the interfluve areas square and population density calculations, which are not precisely documented — the population density may differ.

III. Limitations in the human health impact score calculation:

1. For the calculation of the characterization factor, only the non-cancerogenic effect was considered — the human health impact calculation for cancerogenic effect is not covered in the research;
2. Default fate factor and effect factor from the USEtox model documentation are taken for the calculation of the CF -fate factor and effect factors do not describe the local conditions.

Conclusions

The current research highlights the importance of local information in environmental monitoring and the environmental impact assessment modeling. It is important to note, that the current case study has not directly correlated with the particular area; thus, the approach can be further utilized.

Analysis of Cr in soils enables its calculation in spatial soil distributions. We conclude that the mapping of Cr accumulation halos is an effective tool to divide Tomsk oblast' in particular zones. The efficiency of spatial orientation with interfluves in environmental monitoring is also visible. We see that Cr accumulation ranges follow the river and the wind destination.

Administrative separation inside one region makes a supplementary limitation for pollution monitoring. We recommend keeping to geographical and geological approaches to delimitate the region. Both approaches have certain limitations and advantages, and they are complementary to one another in the impact assessment processes.

However, in the current project, we refer to the naturally recognized locations more, because they properly reflect the geography of the region.

We particularly highlight the importance of the population density values in the characterization factor modification. Population density calculation should be based on the official governmental data, to avoid data inconsistencies. We recommend avoiding a comparison between urbanized areas with a high population density and the countryside with low-populated settlements.

Future perspectives of the work include the development of the database of the impact score of Tomsk oblast' with other mediums such as water and air. To improve the representativeness of the statistical dataset, more sampling data should be proceeded. Otherwise, outlier grades may affect the results. Using the outliers leads to the underestimation of the environmental monitoring. The capability of the neutron activation analysis allows the other non-organics from the USEtox model dataset (Zn, Ba, As, Sb) to be measured. We consider the ISs and CFs modification for four additional pollutants for our future research. The studied area can be also regionalized according to the concentration of other pollutants, and the value of their ISs and CFs.

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

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Bertrand Laratte: Conceptualization, Supervision, Methodology.

Elena V. Ageeva: Writing - original draft, Formal analysis.

Natalia V. Baranovskaya: Conceptualization, Supervision.

Natalia P. Korogod: Conceptualization.

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Data availability All relevant data generated during the study are included in the article.

Declarations

Ethics approval The authors declare that all applicable international, national, and institutional guidelines for the care and use of animals were followed. Sampling of biomaterial was carried out as part of the slaughter of the livestock in a private farm.

Consent to participate Not applicable.

Consent for publication Not applicable.

Competing interests The authors declare no competing interests.

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