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A regional approach for the calculation of characteristic toxicity factors using the USEtox model

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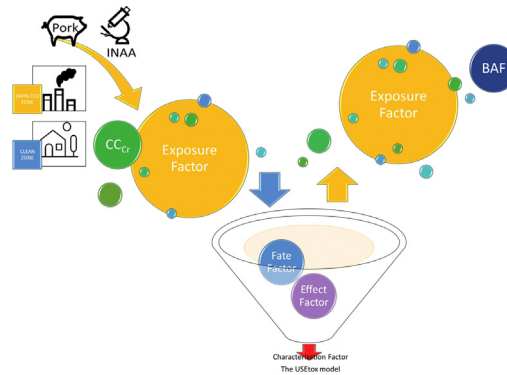
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HIGHLIGHTS

- Estimation of heavy metal content in biosubstances in the zones with mixed ecological conditions
- Experimental and regional approaches in characterization factors calculation
- The description of different levels of human health risks in areas with different environmental tension

GRAPHICAL ABSTRACT



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LCIA
Human exposure
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Heavy metals
Characterization factor

ABSTRACT

The lack of the spatial coverage as one of the main limitations of the Life-cycle impact assessment (LCIA) models leads to disagreement between their results. The USEtox model is only model that provides 8 continental and 17 subcontinental zones but does not consider the wind and water transfers affected areas around the source of pollution. Current investigation proposes the way to reduce this limitation by using the results of chemical analysis (instrumental neutron activation analysis “INAA”) of pork meat as a regional indicator of anthropogenic influence. The concentration coefficient of Cr by replacing the Bioaccumulation factor (BAF) is extrapolated into the calculation of Exposure factor (XF) to modify Characterization factor (CF). Impacted and clean areas of Tomsk district (Russia) placed around Northern industrial hub (Seversk city) are studied. Neither area is located directly in the industrial hub, but the impacted area is under an anthropogenic influence due to air and water transfer of pollution. Results of our investigation present the difference between results of own investigation and default values of USEtox. Probably the model can minimize the impact because of lack of experiment data in the database. The database can be extended more with other analytical results for wide range of metals and geographical locations.

1. Introduction

The Life-Cycle Assessment method is widely used in Europe, as the process of ecological monitoring of ecosystems for chemical elements influences assessment. Today, LCA is one of the leading instruments of

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environmental management in the European Union, based on a series of ISO standards (ISO 14040 “Environmental management”. Life cycle assessment. Principles and framework and ISO 14044 with all requirements “LCA - Requirements and Guidelines”) (Standards, 1991a; The International Standards Organisation, 2006). A universal method of LCA is used in almost all branches of industry: in machine building; construction; electronics; traditional and alternative energy; polymer production; food products; product design and waste disposal (Standards, 1991b).

Life-cycle impact assessment (LCIA), as vital phase of LCA (Fig. 1) is widely used for human health and ecosystems impact assessment of organic emissions, metals and nanoparticles (Ortiz de García et al., 2017; Peña et al., 2018; Pini et al., 2017; Pizzol et al., 2011).

Existing LCIA methods (CML 1992, Eco-Indicator 95, IMPACT 2002+, TRACI, USEtox, etc.) allow the calculation of the metals' negative impact, but there are still a lot of uncertainties connected to it (Monteiro and Freire, 2012; Pizzol et al., 2011). These disagreements in LCIA results are mainly connected to differences in the characterization model such as spatial and time scales or substance coverage (Dreyer et al., 2003).

The most of models can provide information about impact on the environment in the global or continental levels. However, the industrial influence often affects not just the areas where products are produced, but near-border zones also with wind and water transfer. Thus, the correct evaluation of impacts is impossible without consideration of regional aspect.

Among all LCIA methods, USEtox recommended by the European commission is the only LCIA model having such a parameter as geographic separation. The model includes 8 continental and 17 subcontinental zones, each of which is characterized by various climatic, hydrological, geographic-economic and other parameters (Fantke et al., 2017; Rosenbaum et al., 2008). Also, the USEtox contains the database with 28 metals for those with non-carcinogenic effect and 17 for carcinogenic effect calculation, that is representative in comparison with other models (Rosenbaum et al., 2011). Despite all the advantages of using the USEtox model in Life cycle impact assessment, this model does not include a high level of spatial resolution and metal database coverage (Fantke et al., 2017).

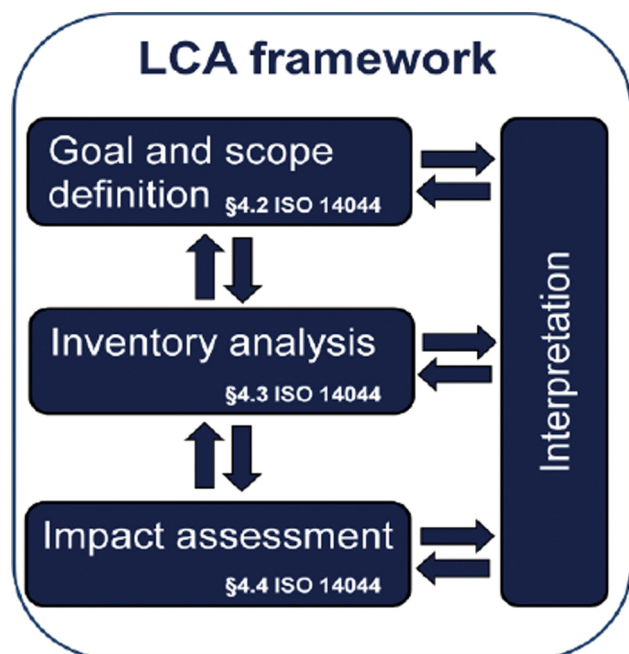


Fig. 1. LCA framework.

We propose the way to reduce those limitations using on of methods of environmental monitoring - bioindication. The method of bioindication is based on analysis of the biological substrates such as animal and human tissues, plants or microorganisms. For example, the studies of chemical content in bones are relevant for the evaluation of chronic and long-term environmental exposure to metals (Beeby, 2001; Budis et al., 2013).

Bioindication provides information about the direct reaction of organisms, communities or ecosystems to natural or anthropogenic changes (Durkalec et al., 2018), since the biota reacts even to minor changes in external conditions and can even predict the geographical origin (Denisova et al., 2008; Franke et al., 2005; Gauthier-Lafaye et al., 2008; Huang et al., 2017; Ilyinskikh et al., 1998; Sheppard, 2011). The concentration of metals in the body of animals and human beings depends, not only on anthropogenic activity (Durkalec et al., 2018), but also on type of diet (Wang et al., 2014; Zhao et al., 2016), physiological specification of organs and tissues (Carpenè et al., 2017), and genetic characteristics of organism (Demirezen and Uruç, 2006).

Bioindication includes a system for monitoring the state of the environment, assessing and forecasting changes in the medias (soil, water, air, biota) under the influence of natural and anthropogenic factors (“EcoLife”, 2018).

Thus, results of bioindication expressed by concentration of metals in the biological material sampled in different geographical areas can be extrapolated into Life-cycle assessment model (e.g. the USEtox) to extend the spatial differentiation.

In the current study methods of bioindication are used to obtain the concentrations of chemicals in the pork meat as a polluted medium. Pork is one of the most widely eaten meats in the world, accounting for about 38% of meat production worldwide. Pork meat is widely studied as the main source of macro- and microelements essential for normal homeostasis. However, besides vital elements, meat contains toxic chemicals (Adei and Forson-Adaboh, 2008; Falandysz, 1993), such as heavy metals and As, which do not have essential biological functions but are transferred through the food chain (Eisler, 1989; Nikolic et al., 2017; Wu et al., 2016). Samples of pork meat are studied as an example of food which is widely consumed by the population of studied areas. The chemical content of pork meat as indicator of the environment condition and methods of metals analysis is being actively studied by Russian and international scientists (Demirezen and Uruç, 2006; Huang et al., 2017; Meurens et al., 2012; Nikolic et al., 2017; Wu et al., 2016; Zhao et al., 2016).

Metals in general as industrial pollutants are considered important in life cycle impact assessment because of their toxicity. Among the chemicals, the heavy metal Cr is chosen from the wide spectrum of analyzed chemicals. Chromium demonstrates toxic effects on living organisms via metabolic interference and mutagenesis (Fantke et al., 2017; Guertin, 2005; Zhao et al., 2016). Cr is also an indicator of specific industrial activity such as thermal electric power stations and chemical industries. Pork meat as an organic source of Cr has a greater bioavailability than inorganic sources (NRC, 1997; Zhao et al., 2016). The normal content of chromium in pork meat is 2–3 mg/kg (“Chromium Content of Meats”, 2018).

Results of bioindication are used to indicate the difference in impact of Cr between two areas of one region with different anthropogenic influence. We suppose that the impact of chromium can be completely different even in the case of two areas only 45 km apart.

1.1. Limitations of investigation

1. The temporal aspect as life time of chemicals in the environment and the total time of Cr absorption by living organisms were not taken into account in this investigation. Also, the data analyzed by INAA express only the midpoint as increasing of Cr concentration in organs of pork and does not provide an endpoint of cause-effect chain of chromium absorption.

- For Characterization factor calculations were taken the default values of the Effect factor and the Fate factor from the USEtox model, only Exposure factor was modified.
- During the investigation were studied and compared two small districts (2 settlements in Tomsk oblast). The chemical components of only two species only were sampled to characterize the ecological situation of studied regions, however, the total sampling include >100 samples and can be representative.
- The content of Cr can vary in dependence of physiological functions of organ or tissue, that is why the average concentration of Cr in whole body of pork is used.
- As one of the limitations of this method, the clark of Cr concentration in biosphere does not reflect the difference between the polluted mediums (soil, air, water or vegetation) of the chemical intake. Clarke of concentration shows the average abundance of chemical in all components of biosphere.

2. Material and methods

2.1. Studied areas

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 Plain on both sides of the Ob river (Fig. 2). The oblast consists of 16 municipal districts, the administrative center is Tomsk. Potential anthropogenic sources of the element's entry into the environment are noted on the territory of the region. There are significant concentrations throughout the whole region which are connected principally with the exploitation of mineral deposits. The flow of pollutants (ashes, heavy metals, etc.) into the air can also be associated with the use of biomass, coals, gas, etc., used for heating purposes.

According to the investigations of L. P. Rikhvanov et al. (Mezhibor, 2011; Rikhvanov et al., 2008), the anthropogenic sources of air pollution

in Tomsk oblast comes from the thermal power industry, petrochemical industry, oil industry and vehicles.

Among all the areas of the Tomsk the main oblast of interest is the Tomsk district (Fig. 3). Moreover, the Tomsk district is of interest because this area contains many industrial sites, for example the Northern industrial unit is located close to Seversk city. More than thirty industrial complexes are located within the Northern industrial unit, including the world's largest nuclear fuel cycle enterprise - the Siberian Chemical Plant (Gauthier-Lafaye et al., 2008; Ilyinskikh et al., 1998; Mezhibor, 2011), and the largest oil and gas processing plant in Russia - Sibur (formerly Tomsk Petrochemical Plant).

The negative anthropogenic influence on its population has been studied over many years (Mezhibor, 2011; Rikhvanov et al., 2008; Ильинских et al., 2001; Москвитина, 1999). The fact of industrial impact on pollution in this region is highlighted in the article of Mezhibor et al. (Mezhibor, 2011). According to the investigations of Moskvitina (Москвитина, 1999), Ilinskih (Ильинских et al., 2001) pathologies of ontogenesis of animals and human were found out in the area around Northern industrial unit.

Thus, the set of natural and anthropogenic conditions of the territory of the Tomsk region (Russia) is characterized by sufficient heterogeneity. This heterogeneity contributes to the formation of various geochemical conditions and factors. The features of accumulation, distribution, and migration patterns of chemical elements in the environment of Tomsk region are determined by combination of natural and anthropogenic factors.

2.2. Materials

Pork meat (*Sus scrofa domestica*) is sampled by the Department of Geocology and Geochemistry of Tomsk Polytechnic University, from private farms in the 2 settlements in Tomsk district of Tomsk oblast of Tomsk region Russia (Fig. 4).

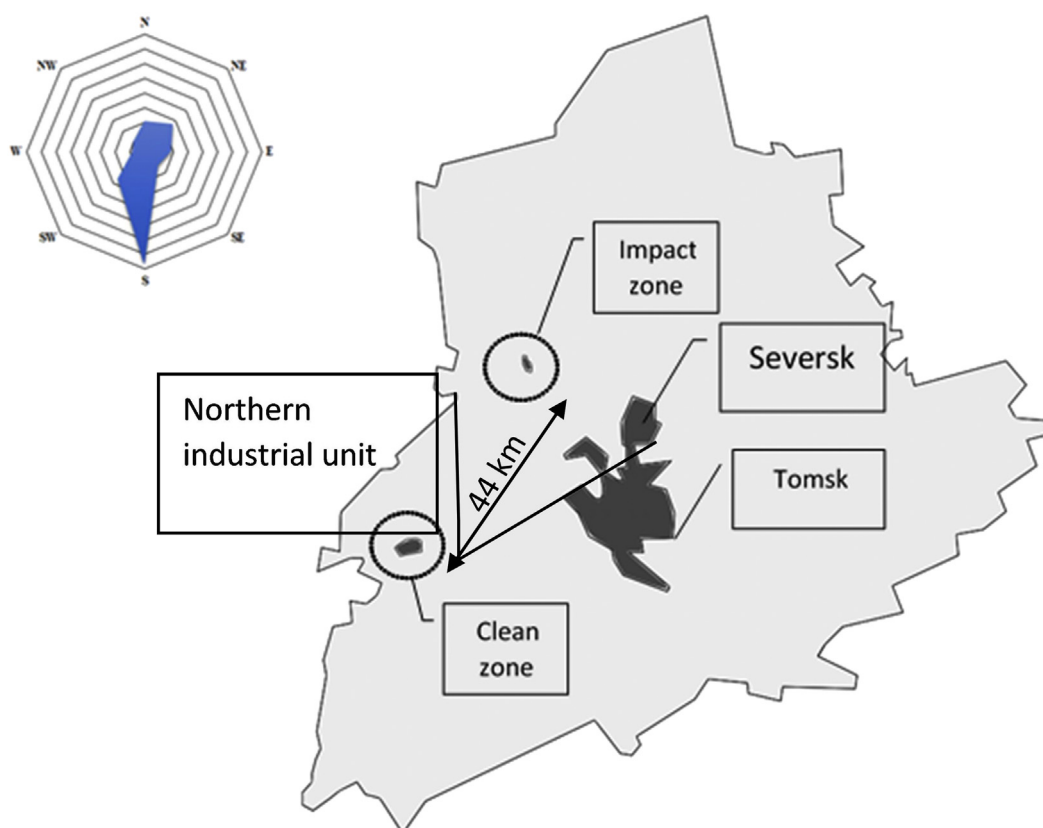


Fig. 2. Sampling map and wind destination of Tomsk district.

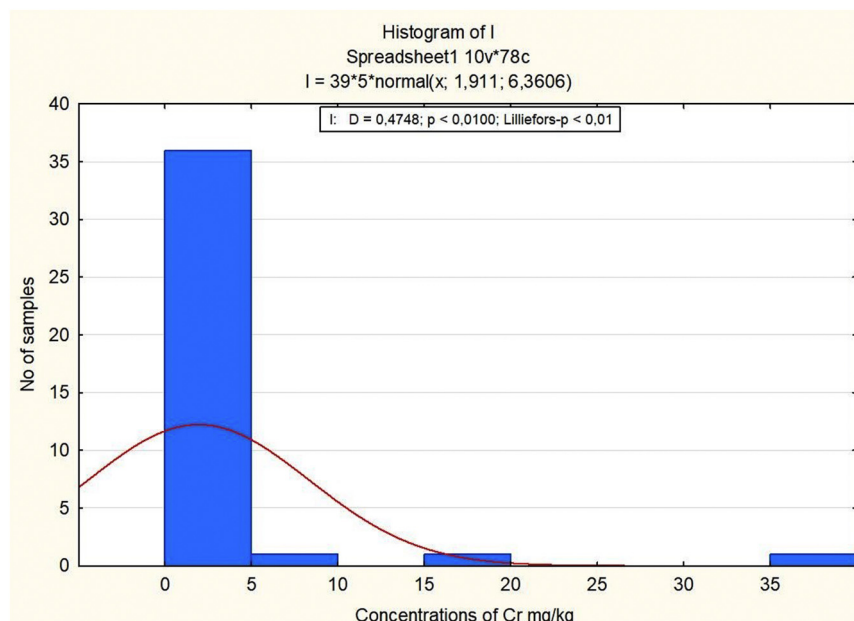


Fig. 3. Diagram of distribution of statistical samples, Impacted area, 39 samples.

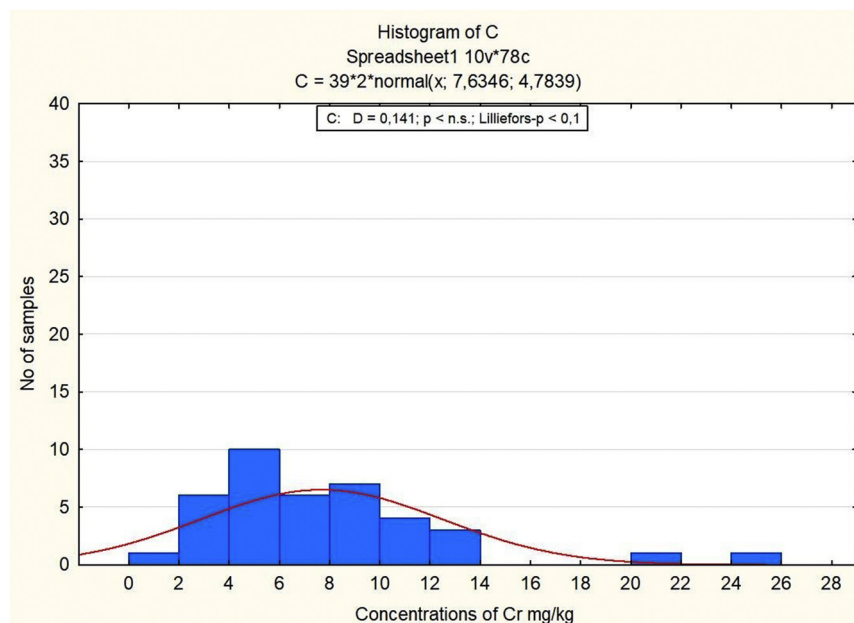


Fig. 4. Diagram of distribution of statistical samples, Clean area, 39 samples.

Two adult pigs (about 10-months-old) are chosen as a source of 78 samples of organs and tissues (Table 1, supportive information), all types of organs are selected in the territory with a variety of ecological situations. Both animals have the same type of feeding with mixed pig feed.

- 1) 39 samples of biological material are taken at the farm of the village of Kizhirovo located 12 km along the prevailing wind direction and impacted by the city and the Northern industrial unit. In addition, a natural geochemical anomaly associated with the presence of the Tugan zircon-ilmenite deposit is found in this territory. The village of Kizhirovo is part of the closed administrative-territorial formation of Seversk, 40 km from Tomsk city.
- 2) 39 samples were taken from a farm located in an area with insignificant territorial technogenic changes in the water intake, but which

experiences periodic load from the city when the wind direction changes. The background area is located on the leeward side located in the interfluvium of the Tom and Ob rivers in the Tomsk region in the village of Verkhnee Sechenovo, 55 km from Tomsk city.

Table 1
Statistical analysis of results of INAA (mg/kg) of biomaterials of *Sus scrofa domestica* 78 samples in total.

No samples	Area of Tomsk district	Mean	Standard deviation	Min	Max
1. 78	Tomsk district (all samples)	5,0	1,0	0,2	36,2
2. 39	Impact area	11,1	5,6	0,2	36,2
3. 39	Clean area	7,6	0,8	0,3	24,2

2.3. Methods of analysis

All studied samples are taken immediately after the death of the animal, packed in plastic bags and frozen. Blood samples are taken from the carotid artery and packaged in eppendorf vessels. Tools (scissors, scalpel) used in the sample preparation process are made of medical steel. Samples of biological material are pre-dried at a temperature of 100–120 °C and prepared for neutron-actinic analysis according to the instructions (Australian Nuclear Science and Technology, 2016).

The samples used for investigation are analyzed by method of instrumental neutron-activation analysis in the Nuclear Research Reactor-Tomsk (IRT-T) in the nuclear geochemical laboratory of the Department of Geoecology and Geochemistry of the National Research Tomsk Polytechnic University.

The method of INAA is based on the irradiation of samples and standards in a reactor neutron flux and the measurement of the induced radioactivity using high-resolution gamma-ray spectrometry with a high level of sensitivity (0.1–10 ppm) (Australian Nuclear Science and Technology, 2016).

The detection limit of Cr by instrumental neutron activation analysis according to Suduko A. presented in the habilitation thesis of Baranovskaya N. is 0,1 [mg/kg] (Барановская, 2011). Methods of descriptive statistical analysis (arithmetic mean, maximum and minimum) are used to identify the samples with the highest concentrations of chromium.

The hypothesis of the normal distribution law of the sample is tested by the Kolmogorov-Smirnov test. The high specificity of the biological material, and the fact that most of the samples under investigation obeyed the log-normal distribution law, necessitated the use of a non-parametric criterion to test the equality of means among several samples.

Samples with values are below the detection limits are not considered to avoid artificial understatement of the average values of sample variety. In current database all values are under 0,5 mg/kg were not detected. Detection limits of Nuclear Research Reactor-Tomsk (IRT-T) is 0,2 mg/kg [52].

3. Calculation

According to the USEtox model support documentation (Fantke et al., 2017) the USEtox model is currently constructed to provide characterization factors (CFs) for human health and freshwater ecological damage for contaminant emissions to indoor air, urban air, rural air, freshwater and agricultural soil.

Combined with emitted mass, the CFs thus enable the derivation of an Impact Score (IS) for all compounds for each product or product system in a comparative LCA-setting. The final impact scores used to select the preferred product or product system. Human health damage includes carcinogenic impacts, non-carcinogenic impacts, and total impacts (carcinogenic and non-carcinogenic). Ecological damage addressed in USEtox is freshwater ecotoxicity for a range of aquatic species.

The resulting characterization factor (CF) that is required for the impact score for either human health or ecological impacts is generally defined as the combination of these three factors (Eq. (1)):

$$CF = EF \times XF \times FF \quad (1)$$

Characterization factor calculation, [CTU_h]

This formula covers two major aspects, related to the environmental fate and behavior of chemicals (FF and XF), and related to human or ecological effects (EF).

- Fate factor (FF) [kg_{in compartment} per kg_{emitted}/day] represents the persistence of a chemical in the environment (e.g. in days) as well as the relative distribution, and the exposure factor expresses the availability for human or ecosystem contact represented by the fraction

of the chemical transferred to the receptor population in a specific time period such as a day.

- Exposure factor (XF) [kg_{intake}/day per kg_{in compartment}] describes the effective human intake of a specific environmental medium – air, water, soil – through inhalation and ingestion.
- Effect factor (EF) [kg_{intake}/day] reflects the impact on human health and the state of ecosystems due to the arrival of a chemical element / substance in the living organism in various ways (through air, water, soil or food).

The human exposure assessment of a chemical emitted into the environment (indoor or outdoor) is based on a cause and effect chain linking the (time-integrated) chemical mass in the environmental compartments (estimated in the fate model) to the substance intake by the total population via various exposure pathways. Human exposure factors XF corresponding to specific pathways XP can be distinguished as direct (e.g. direct consumption of an environmental compartment such as drinking water, or inhalation of air) and indirect (e.g. via food such as meat, dairy produce, vegetables, and fish) exposure factors.

The Characterization factors (CF) are calculated according to the USEtox documentation. The default values of the fate factor (FF), effect factor (EF) and exposure factor (XF) default and modified with results of INAA analysis are used in calculation of CF.

In our investigation, we are concentrating on the calculation of human exposure factor with an indirect pathway via pork meat. This study includes an investigation of the exposure factor for chromium that enters the human body only through air and soil as a result of eating pork, not taking into account fresh water and vegetation.

XF_{xp,i} indirect can be interpreted as the equivalent intake rate of the polluted medium i via the food substrate corresponding to exposure pathway xp. Each exposure factor represents the increase in human exposure via pathway xp due to an increase in concentration in compartment/medium i (Rosenbaum et al., 2008).

The equation to calculate the human exposure factor for an indirect pathway is (Eq. (2)):

$$XF_{xp,i}^{indirect} = \frac{BAF_{xp,i} \times IR_{xp} \times P}{\rho_i \times V_i}$$

Calculation of the human exposure factor with indirect pathway, (2)
[kg_{intake}/day per kg_{in compartment}]

where ρ_i is the bulk density of medium i [kg/m³], and V_i [m³] is the volume of medium i linked to the exposure pathway xp. IR_{xp} [kg/day] is the individual ingestion rate of a food substrate corresponding to exposure pathway xp, P is the population head count, and is the bioaccumulation factor.

$$BAF_{xp,i} = \frac{C_{xp}}{C_i} \quad (3)$$

Calculation of bioaccumulation factor, [kg_{xp}/kg_i]

where C_{xp} is a concentration of Cr in the food substrate corresponding to exposure pathway xp – such as meat or milk, and C_i a specific compartment i such soil, air, water (Fantke et al., 2017; Rosenbaum et al., 2007).

In the USEtox model, the BAF purposed by the USEtox model does not show the impact of environmental conditions in the accumulation of Cr (Fantke et al., 2017). Thus, BAF was replaced by the ratio between concentration of Cr (C_{Cr}) [mg/kg_{xp}] in the pork meat (according to the INAA analysis) and clark concentration of Cr in biosphere [mg/kg_i] (Glazovsky, 1982).

The clark concentration was chosen to calculate the coefficient of concentration of Cr because this value expresses the average concentration of metal in biosphere. The clark concentration is used in the calculations of the maximum concentration limit (PDK) of metals for the

hygienic ratings of Cr in soils (Federal Law of the Russian Federation, 1999).

This concentration ratio expresses the coefficient of concentration of Cr in the pork meat according to the average concentration of this metal in the biosphere. The proposed ratio reflects the variability of metal depending on environmental condition, because of indicative capabilities of pork meat.

$$XF_{xp,i}^{indirect} = \frac{C_{Cr,pork} \times IR_{xp} \times P}{\rho_i \times V_i} \quad (4)$$

Calculation human exposure factor with indirect pathway, [kg_{intake}/day per kg_{in compartment}]

The results of the calculation represent the quantity of chromium absorbed by a human body every day through eating pork meat. These results can be useful in geoecological investigations of natural and industrial territories to assess the human health risks connected with excess chrome.

To see the difference between default USEtox values and results of measured data integration we compare XF USEtox default and XF_i (impact area), XF_C (clean area). To calculate the characteristic level of toxicity, the default data values of the FF and EF from the USEtox models for the region "Central Asia" are used.

4. Results and discussion

4.1. Analysis of Cr concentration

Results of the INAA show that the chromium content in the biological material of a domestic pig in the Tomsk region differs depending on the geographical location, and maxima and minima belong to impacted area samples (Table 1).

The concentration of chromium in the territory of the settlement located in the zone of influence of the Seversk city is higher than in the zone remote from the urban agglomeration. Comparing the average chromium content in the pork meat in different study areas, it can be assumed that the site in whose territory the biomaterial contains more chromium is more exposed to anthropogenic load.

Obviously, the population of the village of Kizhirovo (impacted area) receives more chrome through eating pork than the residents of the village of Verkhnee Sechenovo (clean zone).

According to the results of the Kolmogorov-Smirnov test, statistical samples from both the studied areas have the lognormal distribution (Figs. 3-4.) and the significant difference between two studied samplings ($p < 0,001$) is found.

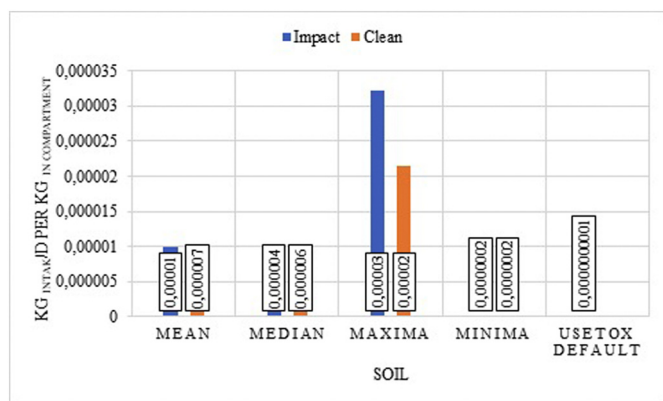


Fig. 5. Human indirect exposure via air through eating pork meat calculated for the populations of impacted and clean areas, according to the results of INAA [kg_{intake}/d per kg_{in compartment}].

4.2. USEtox model

Calculation of the exposure factor by the USEtox method confirms the increased supply of chromium in the zone close to the industrial hub. The intake of chromium through the air and soil via pork meat in the contaminated village is higher compared with the settlement located far from the industrial node (Figs. 5-8).

In the USEtox model wind transport of pollutants does not have any influence on the characterization factor calculations. The model supports "production-based" scenario, thus contamination of medias is associated with place where products were produced, but not with a place where consumers live (Fantke et al., 2017). However, in the current investigation we can see that wind transfer of elements change the characterization factor.

We compared the default data given by the model with our own results of Cr in the meat grown and consumed by the population of the impacted locations. The default values from the USEtox model for both coefficients are significantly lower than the results of own studies using the measured chromium concentrations in the samples of pork. Using the minimum value of chromium content in the sample gives the result closest to the default indicators of the USEtox model. Based on that, it can be concluded that the USEtox model can underestimate the consumption of chemicals by the population of the study areas.

For calculations of human indirect exposure of Cr, we suggest using the arithmetic mean value (mean) of studied metal in the samples. The mean is a statistically more sufficient value for human indirect exposure calculations than other statistical parameters because it considers the content of an element in of all the studied organs, without distinction of physiological factors which may have an influence on an accumulation of metals. Also, the arithmetic mean value represents the statistical sample in 38 samples for each studied area. Use of mean values permits us to avoid the minimization of obtaining results, which happens if only default USEtox values or the minima of chromium concentration are used.

Calculation of the characteristic toxicity factor for both study zones shows that the potential toxicity of chromium for residents of areas closer to the northern industrial complex is higher than for remote zones. Therefore, even within a small administrative unit (the distance between the zones is 44 km (Fig. 2), the exposure factor may vary, depending on the geographical and economic characteristics of the locality.

The characterization factors calculated for soil pollution are closer to the USEtox model default values than for air pollution. Thus, we suggest that the results of CF for air pollution provided in the model are minimized and need to be confirmed with more analytical data.

Another conclusion that can be drawn is that, even if samples of pork meat produced in the territory of Seversk or Tomsk cities were not

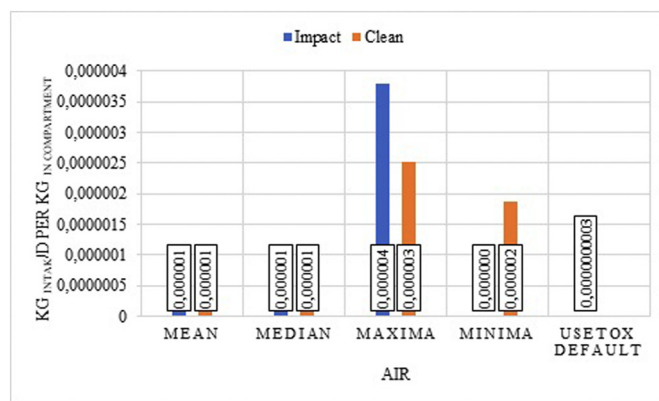


Fig. 6. Human indirect exposure via soil through eating pork meat calculated for the populations of impacted and clean areas, according to the results of INAA [kg_{intake}/d per kg_{in compartment}].

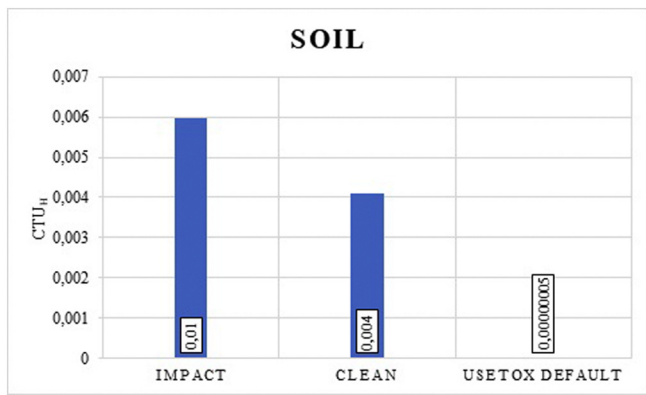


Fig. 7. The Characterization factor of chromium in pork meat via soils, [CTU_H].

considered, the characterization factors calculated for the populations living in the villages located close to the industrial unit are significantly higher than default data are presented in USEtox. Probably, the measured concentrations of chemicals in contaminated air or soil meat represent a more realistic reflection of the impact of the industries on the studied region.

5. Conclusions

We compared the default data given by the model with our own results of Cr concentration in the meat grown and consumed by the population of the impacted locations. The default values from the USEtox model for both coefficients are significantly lower than the results of our own studies using the measured chromium concentrations in the samples of pork. Using the minimum value of chromium content in the sample gives the result closest to the default indicators of the USEtox model. Based on that, it can be concluded that the USEtox model can underestimate the consumption of chemicals by the population of the study areas.

The analytical aspect of our study presents results of chemical analysis of the concentration of chromium in samples of pork meat consumed by population that are much higher in comparison with accumulation ratios derived by the USEtox model. It is possible that model is not sensitive enough to the actual amount of anthropogenic pollutants which is transferred into organisms from environmental media through the food chain.

The analytical method can be complemented by the regional aspect to specify the anthropogenic influence. We consider the regional aspect as a comparative assessment of characterization factors of different locations. Results show that the characterization factors can vary greatly within one administrative unit. Since both studied settlements do not

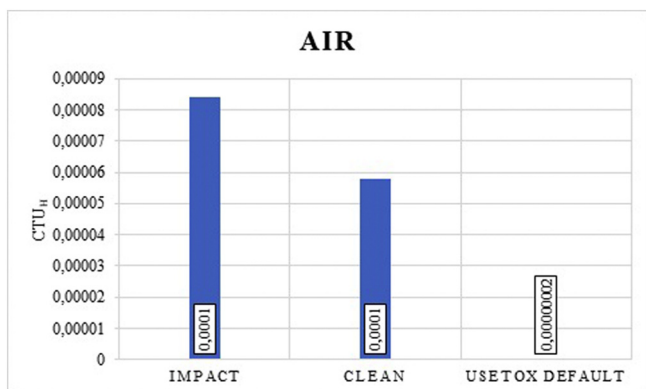


Fig. 8. The Characterization factor of chromium in pork meat via air, [CTU_H].

have industries on their territory, it is assumed that geographic conditions, such as wind transfer of pollutants, are the main difference between the environmental conditions of the study areas. Results confirmed the substantial difference between characterization factors of chromium in the impact zone and clean areas. That proves the diversity of environmental effect. Consequently, an application of geocological research methods and the USEtox model allows the environmental tension on the territory of different settlements to be compared.

The approach proposed in the current article demonstrates the uncertainties in metal impacts assessment results. As it was mentioned in the USEtox method documentation one of limitations of the model is lack of regional resolution. Probably this limitation can be reduced using more precise data about each geo zone provided by model as it was presented in the current article.

As possible extension of this studying, the model's database can be extended with empirically obtained results for wide range of metals and geographical locations.

The integration of results of biomonitoring lead to an alternative scientific approach that allows to study the impact of small localities individually because the environment tension on studied zones is not the same. This approach considers the alternative way of calculation the Exposure factor, considering local data instead of using the information from the USEtox database that is the same for each geo zone. Results are new, they present the exposure of Cr only with pork meat and show the huge influence of pork meat composition on the total impact on population.

This approach could reduce the main limitations of LCIA models: substance and spatial coverage based on the idea of extrapolation of analytical data to LCIA models.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2018.11.169>.

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