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Assessment and Zoning of the Urban Area According to The Level of Heavy Metal Pollution.

Stepanova N.V*., Fomina S.F., and Valeeva E.R.

Kazan (Volga Region) Federal University, Institute of Fundamental Medicine and Biology, Kazan, Russia (420008, Kazan, street K.Marx, 74).

ABSTRACT

Urban area ranking according to the level of heavy metal pollution with identification of four areas in the territory of the city of Kazan was carried out: the 1st - Derbyshki; the 2 nd - Teplocontrol; the 3 rd – Gorki; the 4th – the Kirovsky district. Pollution of snow cover in the city territory was determined by pollution coefficients calculated with application of Maximum Permissible Concentration (MPC) of chemical substances in ambient waters for household and amenity water use. The entire territory of the city of Kazan is mildly polluted by Mn, as far as other metals (Cd, Pb, Cu, Zn, Co, Ni, Cr) are concerned, the soil pollution category varies with areas. Metal content in the hair of the children living in these areas is an informative additional method to assess the present ecological situation in certain territories.

Keywords: heavy metals, snow cover, soil, urban environment, biological monitoring.

Corresponding author

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INTRODUCTION

Kazan is a large industrial center with dozens of industrial enterprises of machine-building, energy and chemical profile located in its territory and a developed intra-urban heavy traffic network. Heavy metals (HM) consistently range with the city pollutants.

Currently, ranking of the urban territory according to the level of pollution with heavy metals involves a certain difficulty. The data bank of the software product "Atmospheric Air Protection" of the Ministry of Ecology and Natural Resources of the RT, which is available in the Republic, allows analyzing consolidated statements of statistical reports on Form 2-TP (air) as far as releases of specific substances including HM in the city of Kazan for the last 10 years. However this approach is artificial as it combines relatively ecologically clean sites of the urban environment and the neglected ones into one mechanically made massif. The available stationary sites for the atmospheric air pollution surveillance are located not in all city districts, for the most part along the motor roads, and assess mainly local air pollution with motor transport, this fact prevents from giving an objective assessment of the situation. Lately, the results of snow chemical survey are used actively enough for the city territory zoning, because snow is considered to be a reliable pollution indicator preserving almost the entire volume of the atmospheric fallout for the winter period. According to observation results [1-2], concentration of pollutants found in snow appears to be 2 – 3 orders of magnitude greater than in atmospheric air. However the assessment of man-made load according to these data is considered so far to be problematic due to absence of reliable reference marks for determining background contents. According to Vasić MV[1], Mihailović A, Kozmidis-Luburić U, Nemes T, Ninkov J, Zeremski-Škorić T, Antić B [10], the major influence on the fine particle volume (mass) in the snow melt was concluded to be due to the elements from anthropogenic sources. This conclusion was based on the significant positive correlation between Fe, Zn and Al and the fine particle volume based distribution parameters. This work presents the first results from winter field campaigns focusing on trace metals and metalloid chemistry in the snow cover from an urbanized region in central Poland [11]. A large inter-seasonal variability depending on anthropogenic emission, depositional processes, and meteorological conditions was observed. The highest concentration (in μg L (-1)) was reported for Pb (34.90), followed by Ni (31.37), Zn (31.00), Cu (13.71), Cr (2.36), As (1.58), and Cd (0.25). It was stated that elevated concentrations of some trace metals in snow samples were associated with frequent occurrence of south and southeast advection of highly polluted air masses toward the sampling site, suggesting a large impact of regional urban/industrial pollution plumes. In most cases the study of the snow cover pollution with HM is limited to determination of their concentration in the snow melt [3-4].

The aim of the study is to carry out ranking of the urban territory according to the level of the snow cover and soil pollution with heavy metals and to study the metal content in the hair of children living in the identified city areas.

MATERIALS AND METHODS

The urban pollution with HM was studied by means of snow chemical survey. Investigations in the city of Kazan were carried out according to the results of expeditionary data of the Institute for the Ecological Problems and Subsurface Resources Management under the Academy of Sciences of the Republic of Tatarstan in accordance with available regulations [5]. The snow cover sampling was carried out at the sites without traces of natural occurrence disturbance at 100-400 sites (depending on the year) on the territory of 200 km^2 at the end of March – the beginning of April over the period from 2007 to 2013. Analysis of snow samples for of heavy metals content was carried out by means of flame and electrothermal atomization atomic absorption spectrometry (AAC) at the Central Certified Specialized Inspection for Analytical Control under the Ministry of Ecology and Natural Resources of the RT. The snow cover pollution of the urban territory was determined from pollution coefficients calculated with the use of regulations for household and drinking water and water for amenity needs. Summation of pollution factors can give the total pollution coefficient characterizing the overall pollution rate of the territory at the sampling site, and it allows identifying districts with different levels of pollution with HMs in the city territory [6]. Assessment of the soil pollution was carried out in keeping with SanPiN "Sanitary and Epidemiological Soil-Quality Requirements" (2003), Hygienic Standard 2.1.7.2041-06 "Maximum Allowable Concentration (MAC) of Chemicals in Soil" and Recommended Practices "Hygienic Assessment of Soil Quality in Populated Areas" (1999) according to concentration factors of certain metals (C_c) and total pollution coefficients (Z_c). Hair sampling was performed in accordance with uniform techniques in 110 children aged 8-10, placed under the 1st and the 2nd health groups and living in the identified city areas.

Chemicals in children hair were determined by means of ICP-AES (Inductively coupled plasma atomic emission spectroscopy) and ICP-MS (Inductively coupled plasma mass spectrometry) at ANO “Center for Biotic Medicine”.

RESULTS

According to the results of snow surveys performed by the “Kazan” Weather Station over the period from 1937 to 2012 the average maximum stock, which is sometimes referred to as the “background” snow storage, derived on the basis of long-term observations, makes 170 mm or 170 kg/m^2 for Kazan. According to such approach, element concentrations in the snow melt characterize snow purity, but prevent from estimating pollution dynamics of the urban landscape and determine its level of potential danger for humans and ecosystems. To assess the HM content (Cd, Pb, Cu, Zn, Co, Ni, and Cr) in the soluble fraction and the solid precipitation of the snow the technogenic concentration (C c) coefficients were used as compared to the background ones [6-8]:

$$C\,c = C_i / C\,c1,$$

where C c – concentration coefficient; C_i – element concentration coefficients in the solid precipitation of snow sampled within the city limits; C c1 – element concentration in the solid precipitation of snow of the background territory (C1). Background sites are selected in the territories, which are not exposed to pollution or subjected to its minimum. To determine the polyelemental pollution rate of the snow cover the total pollution index was used:

$$Z\,c = \Sigma\, C\,ci - (n-1),$$

where Z c – the total pollution index; C ci – element concentration coefficients, n – the number of determined HMs. However, the background approach to assessment of the pollution rate of the snow cover suffers from a number of shortcomings, since the sampling site selection is random; the values of background indices depend on the pattern of weather conditions and change annually due to complete change of the snow cover (in our studies the background indices of certain metals varied by several times in different years); the possibility to determine the actual amount of pollutant per unit of territory is not available [6-12]. The fact that the elemental toxicity class is left out of account when calculating the total coefficient of HM concentrations, is important as well. Therefore we consider that the background technique for assessing the “pure” form is of no practical importance when studying the snow cover pollution. Determination of the pollutant mass per unit area for a certain period of time appears to be more informative resulting in minimum of uncertainties caused by spatial and temporal variability of the snow cover. The element content in the snow mass located on 1m^2 of the territory and effluence on the entire territory under study (200 km^2) during 5 months of the winter period are determined from the element concentration in the snow melt (µg/l), the snow cover depth and its density on the given area, (тab. 1).

However it is impossible to determine the danger level for ecosystems by means of this method of assessing the results of the analysis for HMs in the snow cover. It is more efficient to use the excess regulation index specified for the site concerned when assessing the snow cover pollution. However at present, the Maximum Permissible Concentration (MPC) of chemicals in the snow is not determined. At the same time, snow is the pollution indicator both of atmospheric air and subsequent pollution of surface, ground waters, soils and vegetation. Thus, snow-melt waters coming into rivers during spring flood make up to 80% of the gathering ground for most of the rivers of the Republic of Tatarstan. This fact allows using MPC for any of the listed sites. Due to the fact that the amount of water and snow is expressed in identical units it is handier to use MPC for water. At the same time a direct use of MPC for assessment of the snow cover pollution rate is impractical, since recording of the snow mass and the index characterizing the pollutant fallout per unit area in a definite time is required. An attempt to combine the index showing the substance toxicity with the unit of area, on which it comes, was made. MPI (maximum permissible ingress of a substance to the snow cover), which shows the amount of substance coming on a particular area per unit time in concentrations not exceeding the established regulations, was suggested as a composite index. It is calculated according to a formula:

MPI = MPC x m_{avg} x time, where m_{avg} – average snow mass per unit area surveyed by the time of sampling. Thereafter pollution coefficients were calculated. MPC of the metals for household and drinking water and water for amenity needs is known to be determined not only according to sanitary-toxicological, but also organoleptic indicators, therefore MPI coefficients obtained with the use of MPC for these waters are designated as pollution coefficients. Mean coefficient for each element is determined (тab.2). The total pollution coefficient acquires the form of the total of element concentrations ratios in each sample to corresponding MPC:

$\sum C = C1/ MPC\ 1 + C2/ MPC\ 2 + ... Cn/ MPC\ n$, as an average mass per $1m^2$ of the territory is used when calculating MPI.

Levels of the snow cover pollution with HM with application of pollution coefficients (for household and drinking water and water for amenity needs) allow assessing contribution of a chemical element to overall pollution depending on its amount (mass) and quality (MPC). Analysis of the city territory pollution according to the values of average and total pollution coefficients of the snow cover for the years 2007 -2008 showed that the greatest contribution falls on cadmium, lead, chromium and nickel (refer to тab.2). In the winter period of the years 2009 – 2011, zinc, manganese, nickel and lead dominated in the territory pollution. Total pollution for this period was below hygienic regulations, with rise in the year of 2010 (due to a considerable increase of nickel, lead and chromium fractions). Pollution of the territory in the city of Kazan with heavy metals in winter periods of 2007 - 2013 increased ($C_{avg.\ total}$ in 2013 made 6.25). Awareness of total pollution coefficients of the snow allows identifying territories with different pollution levels. The following four areas: the 1st **-** Derbyshki (C_{total} =13.4); the 2nd - Teplocontrol (C_{total} =10.6); the 3rd – Gorki (C_{total} =6.3); the 4th – the Kirovsky district (C_{total} =8.5) differing in location of enterprises, production plants and vehicle density were identified on the basis of total pollution index values of the snow cover as far as HM in the territory of the city of Kazan for the period of 2009 – 2013. Thus, the Kirovsky district (the 4th area) and the area of Teplocontrol in the Privolzhsky district of the city (the 2nd area) are old industrial centers, where large enterprises and a number of motor vehicle fleet are located. And Gorki (the 3rd area) belongs to the most ecologically favorable commune of residence. Assessment of chemical pollution of mobile forms of metals in the certain city areas showed that the permissible level of pollution ($Z_c < 16$) is registered in the 3rd area [14]. As far as the value of total coefficients (Z_c) is concerned, the 2nd and the 4th areas (the area of Teplocontrol and the Kirovsky district) appeared to be the most polluted ones. Pollution levels of the 1st and 3rd areas were practically always half as high. The level of technogenic pollution of soils is known to be characterized by sufficiently great stability observed for many years and centuries, and this fact is likely to explain the higher pollution level in the 2nd and the 4th areas, which were always characterized by a high level of the industry and production development, factories and plants of the region being concentrated there since the XIX century. As far as total index of the soil pollution with mobile forms of metals, the soils of the 1st area refer to category with permissible pollution level (Z_c - 11.3), the soils of the 3rd and the 4th areas – to moderately hazardous ones ($Z_{c.}$ - 23.4 and -27.1), and the soils of the 2nd area of Teplocontrol refer to dangerous ones according to the estimation scale ($Z_{c.}$ - 41.2) (Fig.1).

Linear regression equations between the HM content in the solid phase of the snow and the total form in soils were calculated on the basis of long-term observations, and this allows predicting ecological situation. We obtained statistical models reflecting dependence of the total content of chromium, copper and zinc in the soil from the content of corresponding metals in the solid (insoluble) phase of the snow (Figs. 2 and 3). Overall regression equation of association between the copper content in the dust and its total content in the soil is as follows: $y = 101.2*x – 159.7$, where x – the total content of copper in the soil (mg/kg), with $F=49.6$, $r =0.82$, $p < 0.001$. Regression equation for chromium: $y=87.9*x – 403.2$, where x – the total content of chromium in the soil (mg/kg), with $F=8.1$, $r =0.67$, $p < 0.001$; for zinc: $y = 0.00028*x + 0.03$, ($F=7.37$, $r =0.53$, $p < 0.001$).

The statistical models, which we obtained for chromium, copper and zinc, confirm the existence of well-defined soil anomalies in the industrial areas of the city in these HMs. As to the rest of metals (lead and cadmium) such association is not determined, this fact being indicative of their uniform distribution (diffusion) as part of the solid phase of the snow cover and the sources of their emission within the city territory, and to a certain extent implies a greater role of motor transport in their origin. The revealed association of the copper, chromium and zinc content in the dust and in the soil allows calculating the total content of the studied metals at any site of the city territory and identify the sources of pollution.

Assessment of the city soils pollution rate with the account of the hazard class of the element, its MPC and maximum permissible level of the metal content (Кmax) showed that the entire territory of Kazan, which we investigated, is slightly polluted with manganese, as for other HMs, the soil pollution category varies with areas. The 1st area (the Derbyshki settlement), was heavily polluted only with copper; the 2nd area (Teplocontrol) was heavily and very heavily polluted with nickel, lead, cobalt, copper and zinc; the 3rd area (Gorki) – with nickel and cobalt; the 4th area – with cadmium and copper. The second area, which is nominally designated as "Teplocontrol" and belongs to heavily and very heavily polluted one because of high concentrations of lead, cobalt, the mobile forms of copper, nickel and zinc, as compared with MPC, claims particular attention. Wide variability of the metals pointwise values, "outbursts" in the enterprise sanitary protection areas in particular (e.g., Zn in Derbyshki), confirm their technogenic origin. This being the case, at the next step, we carried out analysis for the HMs content in the hair of the children living in the identified areas. Up to date, one of the approaches to identify actual chemical load and assess the ratio of negative impact on the health and safety of the population is determination of chemical compounds in human biological media. However identification of the regional levels with the account of a complex of ecological and hygienic factors in the territory under study remains a key aspect [8-14]. The results of the hair analysis in children living in identified areas of the city of Kazan showed that concentrations of lead, cadmium, nickel, zinc, manganese and copper exhibited a large scale of absolute values. Check of a distribution pattern of absolute values of HMs concentrations showed that statistical concentration distribution of just one biogenic metal – zinc (according to Kolmogorov–Smirnov test) follows the normal distribution law (Fig. 4).

As for the rest of metals (toxic and nominally essential ones), the sample schedule was asymmetric, with shift to the right. However following a traditional approach, at the preliminary stage, we compared arithmetic mean values of the HMs content in the hair of the children living in previously identified four city areas (Fig. 5).

Thus, lead content in the hair ($p<0.01$) was significantly lower in the children living in the 1st area (Derbyshki) and the 4th area (the Kirovsky district), in comparison with the children from the 2nd area (Teplocontrol). The highest average content of cadmium (0.51µg/g) in the hair was observed in the children of the 3rd area. The HMs content in the hair of the children from the 3rd area had no significant differences from the similar ones in other areas, although the area of Gorki is traditionally considered to be the ecologically favorable city district. However we paid attention to a number of factors: first, some of the children live at the boundary of two opposite areas as far as ecological situation (Teplocontrol and Gorki); second, there is higher percentage of children with increased toxic HMs content in the hair among the children living in the 2nd area, than in the area of Gorki, third, an opposite pattern was revealed with respect to essential HMs – the percentage of children with high values of Cu and Zn, appeared to be less in the technogenic area, than in the 3rd area (Fig. 6). Thus, the most significant differences in certain metals content in the hair, particularly of lead and cadmium, were revealed in the children living in the technogenically polluted area (Teplocontrol).

The occurrence of pronounced right-sided asymmetry in distribution of the toxic and conditionally essential metals concentrations determined it necessary to use the centile method of analysis for developing standard scales based on the results of assessing the heavy metals' content in children hair. The carried out investigations resulted in defining the boundaries of standard centile intervals for the child population of the city. We considered the interval from the 25-th to the 75-th centile as a norm due to its correspondence to average values of this chemical element concentration in the hair of the child population of the city of Kazan (тab. 3).

The median of centile distribution (the 50-th centile) of the lead content in the hair in all studied areas of the city did not exceed the established permissible lead level in the hair ranging from 2.02 to 5.27 µg/mg. The upper boundaries of cadmium exceeding the permissible values were observed in two areas, but in the 2nd area, cadmium content exceeded the recommended level already at the level of the 75-th centile. Nickel and zinc concentrations in over 50% of children hair exceeded the permissible levels in all city areas, apart from Gorki. The upper boundary of increased concentrations exceeded the permissible level for lead reaching 11.03 µg /mg, for nickel (up to 23.25 µg /mg), for zinc (up to 232.4 µg /mg), for cadmium (up to 2.39 µg /mg), for copper (up to 75.7 µg /mg), for manganese (up to 17.2 µg /mg) and for chromium (up to 6.62 µg /mg) in the hair of urban children. In conclusion it should be pointed out that the increased content of zinc, copper and nickel proved to be typical for the hair of children from the 1st area; that of zinc and copper - for the hair of children from the 2nd area. Thus, the area of Teplocontrol was again defined as the hazard area (imbalance of essential microelements and increased HMs content). We obtained linear statistical relationship between the hair content and the total content of lead and mobile forms of cadmium in the soil (95% CI), regression coefficient r = 0.878, r^2 = 0.772. Therefore, the content of essential and conditionally essential microelements (zinc and copper), as a rule, in the hair of children living in ecologically unfavorable areas of the city was lower, as compared with ecologically favorable area. At the same time, concentrations of toxic elements (cadmium, lead, nickel, chromium and manganese) were higher in the hair of children from the area of Teplocontrol than the levels permissible for our city. Application of the centile method for assessment of the HMs content in the children hair allows validating and introducing definiteness into the notion of “a norm” with the account of local – regional peculiarities and carrying proper selection of children into risk groups of developing diseases associated with deficiency or excess of microelements. So far, methods for determination of chemical compounds in biological media alongside with clinical-diagnostic, epidemiological, statistical and other survey methods allow solving problems on the study of common mechanisms of the human body interaction with environmental chemical factors and revealing health risks at low levels of various contaminants exposure in biological media as a whole [9].

Table 1 Heavy metals ingress (water-soluble form) to the snow cover of the city of Kazan in the years 2007–2013 (kg/200 km^2 for 5 months)

Element	2007	2008	2009	2010	2011	2012	2013
Cadmium	23.6	12	12	ND	101	331	44
Cobalt	20	14	20	ND	58	ND	613
Manganese	-	-	-	114	336	350	235
Copper	282	74	379	157	171	270	428
Nickel	57	92	209	172	1473	120	165
Lead	86	-	24	37	290	27	418
Chromium	93	64	129	59	360	-	185
Zinc	713	771	770	1823	426	884	1258
Total	1275	1027	1573	2362	3215	1682	3346

Notes:
ND – the given metal is not determined in samples;
“-’’ metal content below analytical sensitivity of determination techniques

The snow sampling was carried out at 100 – 400 sites (depending on the year) in the territory of 200 km^2 in area. Average time of the snow cover preservation on the city territory makes 5 months. Accordingly, the ingress was calculated in kg/ 200 km^2 for 5 months.

Table 2 Coefficients of the snow cover pollution with heavy metals in the city of Kazan (according to MPC for drinking and household waters)

Year	Average pollution coefficients, $К_{avg}$									Total
	Cd	Co	Mn	Cu	Ni	Pb	Cr	Zn	Fe	$К_{avg}$
2007	0.15	0.003		0.003	0.01	0.03	0.02	0.02	ND	0.24
2008	0.14	0.008	-	0.001	0.05	0.03	0.06	0.02	ND	0.303
2009	-	-	0.04	0.004	0,04	0.04	0.03	0.06	ND	0.21
2010	0.001	0.013	0.05	0.004	0.32	0.1	0.15	0.01	ND	0.748
2011	0.25	-	0.13	0,01	0.03	0.01	-	0.03	ND	0.453
2012	0.89	0.18	0.08	0.015	0.05	0.26	0.06	0.04	0.13	1.59
2013	5.43	-	0.09	0.01	0.08	0.24	0.24	0.03	0.17	6.29

Notes:
ND – the given metal is not determined in samples;
“-’’ metal content below analytical sensitivity of determination techniques

Table 3 Centile scales to assess the content of heavy metals (µg / mg) in the hair of children in zones of the city, Me (q25 ÷ q75)

Metal	Areas (zone)			
	I - Derbyshki	II - Teplocontrol	III – Gorki	IV- Kirovsky district
Zn	131.4 (111.4÷158.9)	143 (117.4÷166.9)	155.5 (122.8÷173.6)	143.25(112.8÷169.3)
Cd	0.45 (0.23÷0.94)	0.3 (0.19÷1.05)	0.32 (0.032÷0.64)	0.19 (0.017÷0.15)
Cu	11.19 (8.5÷16.4)	9.97 (9.33÷11.9)	11 (8.67÷13.4)	11.91 (8.82÷12.5)
Mn	1.12 (0.48÷2.91)	2.14(4.29÷4.79)	0.89 (0.65÷1.38)	2.25 (1.32÷2.62)
Ni	1.11(0.27÷2.25)	2.07 (9.42÷41.8)	0.68 (0.22÷1.13)	1.28 (0.14÷1.40)
Pb	2.38 (0.9÷5.1)	5.27 (15÷15.1)	2.02 (0.87÷3.71)	2.02 (0.36÷2.94)
Cr	0.27(0.091÷0.6)	0.7 (7.98÷24.41)	0.91 (0.43÷1.13)	0.68 (0.68÷4.04)

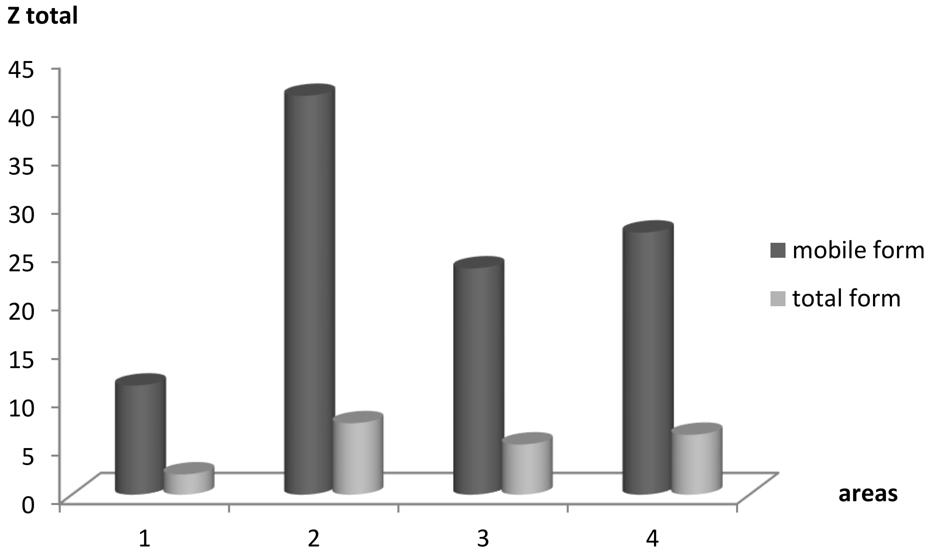

Fig.1. Average total coefficients of the soil pollution (total and mobile forms) in the areas of the city of Kazan (the 1st – Derbyshki, the 2nd – Teplocontrol, the 3rd – Gorki, the 4th – the Kirovsky district).

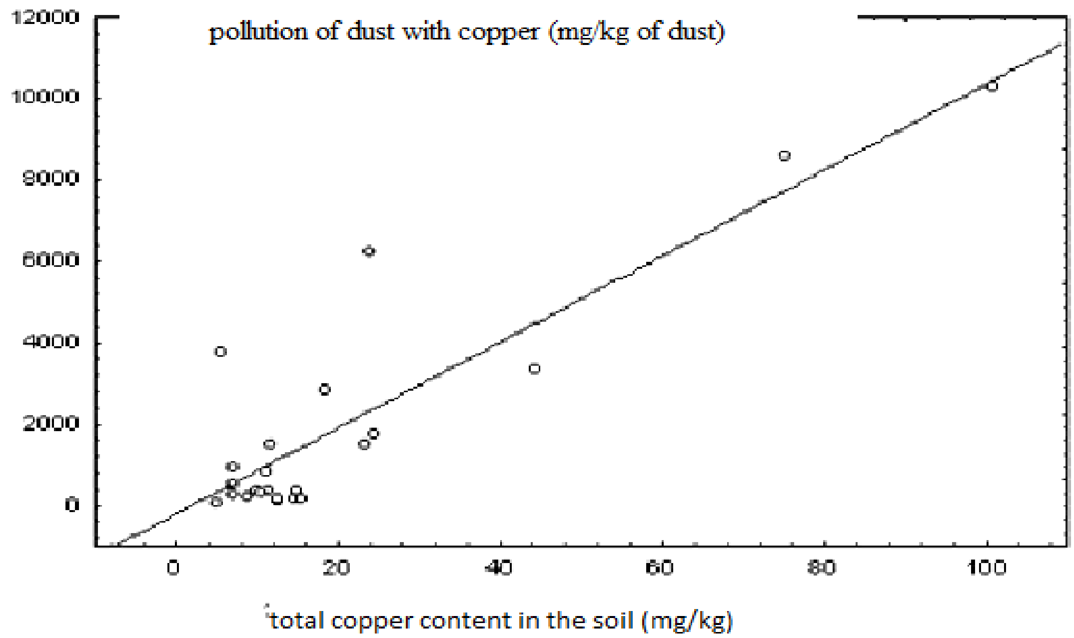

Fig.2. Diagrammatical model of the relationships between the total copper content in the soil and the solid phase of the snow

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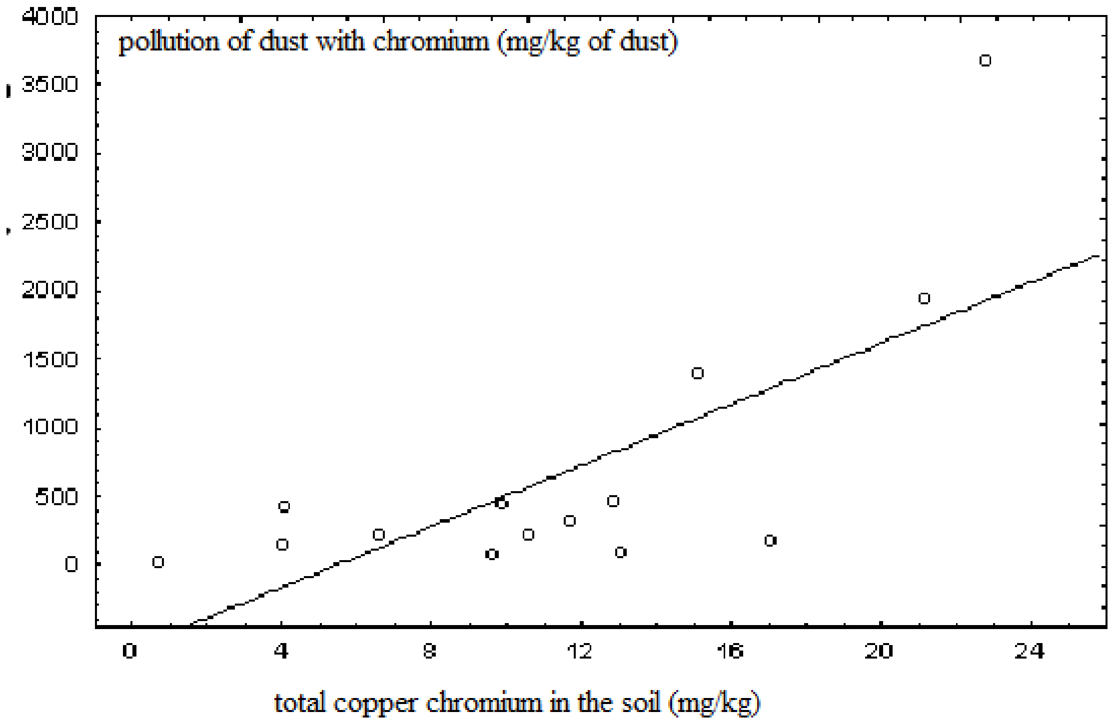

Fig.3. Diagrammatical model of the relationships between the total chromium content in the soil and the solid phase of the snow

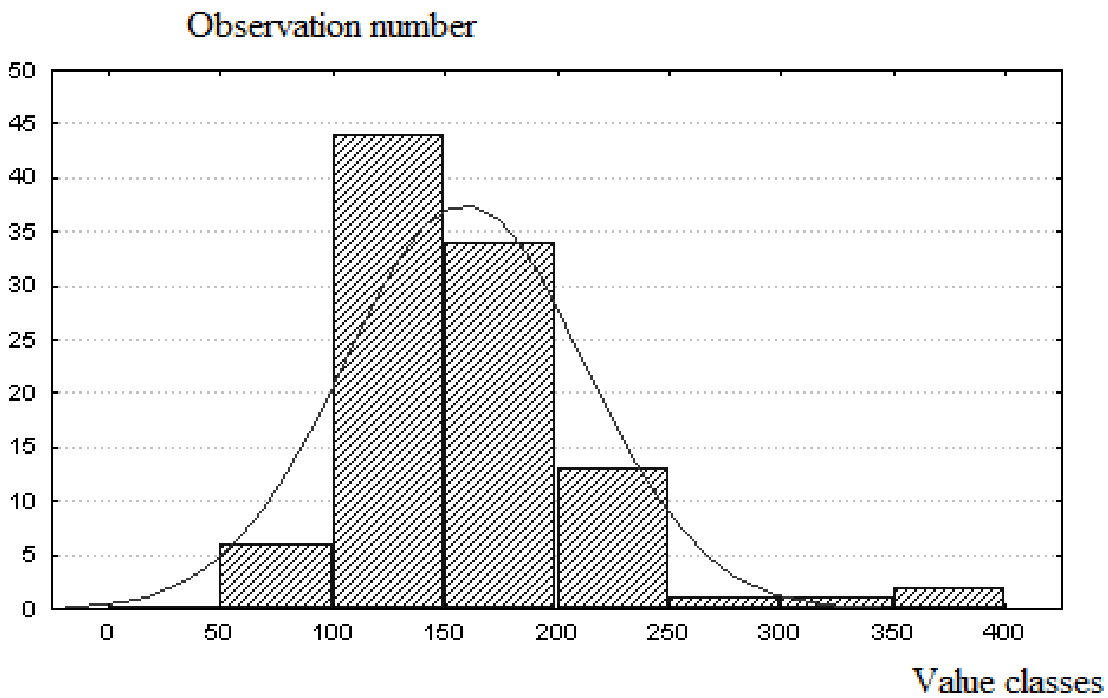

Fig.4. Distribution of the Zn content values in the hair of urban children (K-S d=0.10879)

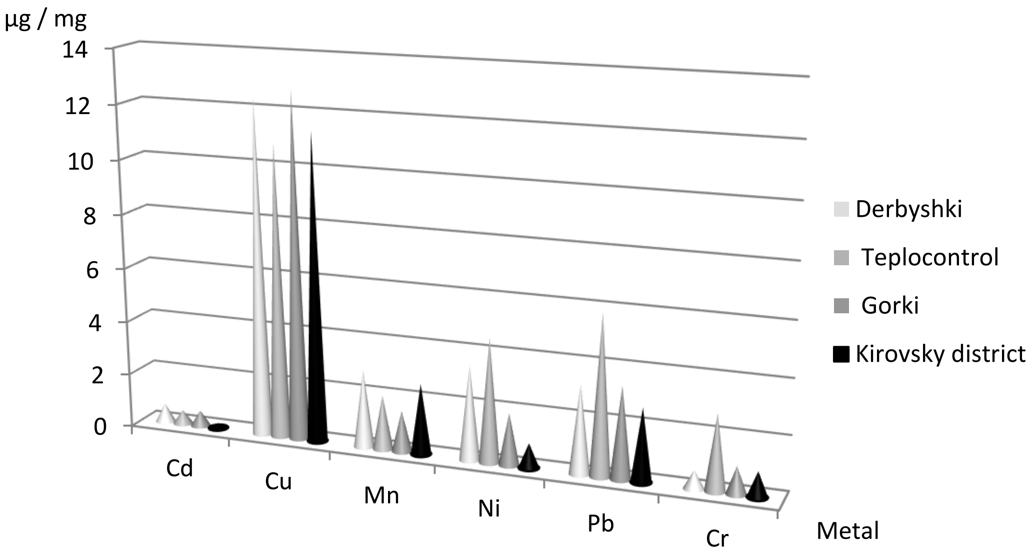

Fig.5. The average metal content in the hair of children living in different areas of the city, µg / mg

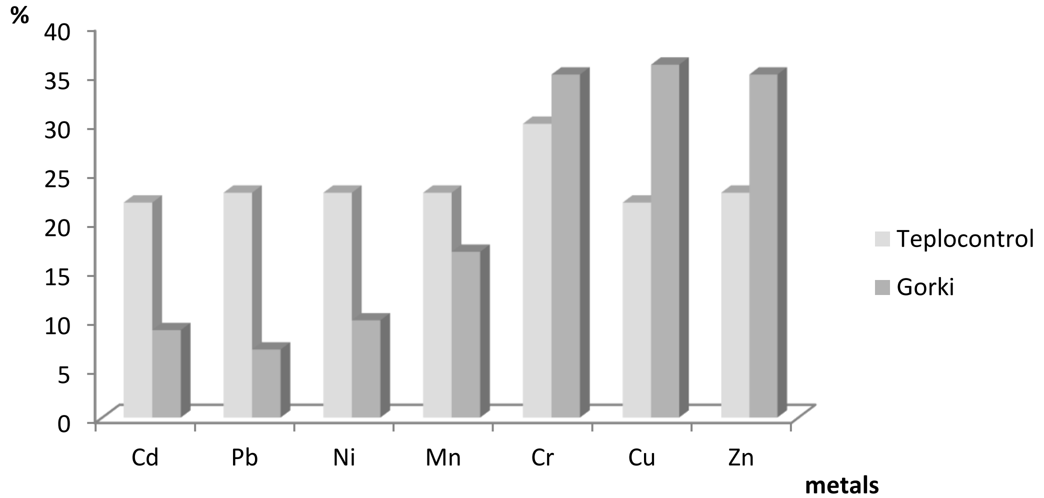

Fig.6. Percentage of children with metal content in the hair higher than the permissible level, %

CONCLUSION

- Assessment of the urban territory according to the level of the heavy metals pollution based on the data of the snow cover and soil chemical characteristics allowed identifying the most unfavorable areas of human habitation (the area of Teplocontrol and the Kirovsky district).
- When levels of the atmospheric air pollution with HM are considerably below the regulations, the environmental monitoring by means of the snow chemical survey is not only an economically justified and adequate method of assessing the urban territory pollution reflecting annual slice, but also the only technique to allow territory ranking according to HM pollution rate.
- Biological monitoring of the children hair is additional informative method of assessing ecological situation as far as heavy metals in certain city territories (the area of Teplocontrol). Biological monitoring and revealing of children with increased sensitivity to chemical agents seems to be more important than extension of programs on ecological monitoring of determining the concentrations of various chemical agents in environmental compartments.
- Revealed relationship of copper, chromium and zinc content in the solid phase of snow (dust) and in soil allows calculating the total content of the studied metals anywhere in the city territory and identify the sources of urban environmental pollution.

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