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Assessment of heavy metal pollution of the snow cover of the Severodvinsk industrial district (Russia, the Arkhangelsk region).

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Abstract.

In the present work, the content of heavy metals in soluble and insoluble forms of snow was investigated, as well as in waters from wells near the Severodvinsk industrial centre (Russia, the Arkhangelsk region). The content of heavy metals (Al, Fe, Ti, Mn, Zn, Cr, Cu, Ni, Pb, Co, V, As, and Cd) in the melt snow filtrate and solid residue was measured using the ICP-MS method. On the territory of the city, there are high concentrations of metals, high values of electrical conductivity and low pH values in objects most susceptible to anthropogenic pollution. It was determined that in urban conditions the share of the solid-phase component increases, which indicates an increase in dust fallout. Statistical analysis showed that most of the metals in the snow cover are linked with each other by a strong correlation ($r > 0.9$), and also made it possible to identify sources of pollution, namely thermal power plants and shipbuilding enterprises. The calculation of various indices for assessing the snow quality and natural waters determined the level of heavy metal pollution in the Severodvinsk industrial district as extremely high and dangerous for public health. The radiation parameters of water from wells were studied and a satisfactory radioecological characterisation of the research area was established.

Keywords: snow cover, physicochemical properties, heavy metals, water quality indices, radioactive pollution, the Arkhangelsk region.

Declarations:

Ethics approval and consent to participate: Not applicable. This work does not report on or involve the use of any animal or human data or tissue.

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1. Introduction

One of the most important environmental problems at present is the deterioration of the urban environment. In cities, there is a high concentration of sources of pollutants, as well as a varied composition of pollutants. This determines the need for monitoring the ecological environment, which directly affects the health of the population, especially atmospheric air. One such monitoring method is the study of the chemical composition of snow, which has the ability to accumulate various pollutants. Compared to rainfall, snow is more efficient at absorbing pollutants and washing them out of the atmosphere due to the larger surface area and porosity of snowflakes compared to rain drops (Franz and Eisenreich 1998). Thus, the snow cover is a natural, informative and convenient object of monitoring in northern cities (Yanchenko et al. 2015; Talovskaya et al. 2018; Moghadas et al. 2015).

The main urban sources of pollutants for snow cover are emissions from the fuel and energy complex, industry and vehicles (Elik 2002). These sources emit various substances into the environment in the form of poisonous particles and dust, which in the first period of being in the air directly affect the organisms of animals and humans (Kurbakov et al. 2020). The main components of pollutants are toxic heavy metals. For example,

anthropogenic impact in Moscow has led to a significant increase in the dust load (2–7 times), the concentration of heavy metals in the snow cover (2–5 times) and the salinity of melt water (5–18 times) compared with the background level. Urban snow contains Sn, Ti, Bi, Al, W, Fe, Pb, V, Cr, Rb, Mo, Mn, As, Co, Cu, Ba, Sb, and Mg (Vlasov et al. 2020). With active snow melting, significant amounts of heavy metals and other components enter the surface and underground waters, pollute the soil cover, and have a negative impact on the biota.

Exposure to metals has become increasingly important in recent years. For example, heavy metals in urban areas are known to seriously affect human health (Hough et al. 2004). Air contaminated with heavy metals is responsible for about 80% of premature deaths (Vlasov et al. 2020). Heavy metals cause coronary heart disease, stroke, lung cancer and chronic obstructive pulmonary disease, and increase the risk of adenocarcinoma (WHO 2014; Raaschou-Nielsen et al. 2013; Veremchuk et al. 2018). So the effects of lead are manifested in an increase in blood pressure, disorders of the nervous system, liver and kidneys. Cadmium salts have mutagenic and carcinogenic properties, affect the liver, kidneys, and pancreas, and can cause emphysema and lung cancer. Chromium compounds have toxic effects, affecting the gastrointestinal tract and kidneys. Zinc has a general toxic, irritant, carcinogenic effect on humans, causing weakening of memory, anaemia and pulmonary oedema.

Dissolved heavy metals can be incorporated into the biogeochemical cycle, while insoluble solid forms of metals accumulate in the surface horizons of urban soils and road dust (Gustaytis et al. 2018; Chen et al. 2018). The ratios of the two forms of metals in the snow are also informative indicators of various sources of pollution (Telloli 2014; Huber et al. 2016; Vijayan et al. 2019). In some works, it was noted that, according to the ratios of soluble forms, elements can be divided into three groups: high (Mn, Zn, and Pb), moderate (Ni and Cu), and insoluble (Al, Ti, Fe and As) (Bacardit and Camarero 1989; Colin and Jaffrezo 1990; Millet et al. 1995). The solubility of various elements may be related to the origin and mechanisms of aerosol formation. The most dangerous for living organisms are metals that are readily soluble in water in comparison with complex or poorly soluble compounds.

Severodvinsk is located in the north of the East European Platform on the White Sea coast, 30 km west of Arkhangelsk. Severodvinsk city is the largest industrial centre of the Russian Navy in the north-west of Russia (Ostrochenko and Ostrochenko 2008). Since its formation in the late 1930s, this city, with its technogenic influence, has formed a specific spectrum of pollution around itself. The city's industry is represented by enterprises of the military-industrial complex, where the construction, repair and disposal of nuclear submarines takes place, as well as enterprises of the heat and energy complex (Ostrochenko and Ostrochenko 2008). These enterprises are potential sources of emissions of pollutants into the atmosphere, including heavy metals. Also near the industrial complex is a coal-fired thermal power station. All of the above factors indicate the need for constant environmental monitoring of this area.

In connection with the above, the purpose of the presented study was to identify the main sources of technogenic pollution of the atmosphere and surface waters in the Severodvinsk industrial region and to assess the ecological and geochemical situation based on the analysis of the content of metals in the water-soluble and insoluble components of the snow.

2. Materials and methods

2.1. Study area

The object of research was the territories immediately adjacent to the Severodvinsk industrial region. Water sampling from wells and snow was carried out in March 2020, during the period of maximum snow accumulation, shortly before the period of snow melting in open areas. Water samples were taken at summer cottages from shallow wells with a depth of 3 to 8 m. The thickness of the snow cover at sampling points varied from 45 to 55 cm. In total, 10 snow samples were taken (marked as C) and 5 water samples from wells (marked as CB).

The relief type of the Severodvinsk territory is a water-glacial accumulative plain (Byzova and Premalina 2001). The climate of the territory is maritime, cold with a strong influence of Arctic air masses. The average annual air temperature in the Severodvinsk region is about 0 °C. Precipitation is greater than evaporation. Due to the excessive moisture, a dense river and lake network and numerous swamps have formed on the territory.

Figure 1. Location of sampling points: 1 - water samples from wells; 2 - snow samples; 3 - specially protected natural recreation complex 'Pine Forest of Yagry Island'; 4 - suburban garden plots; 5 - territories of engineering and ship repair enterprises; 6 - combined heat and power plant.

2.2. Sample collection and preparation

When taking samples of the snow cover, we were guided by the following regulatory documents (GOST 17.1.5.05–85 1985; MR 5174-90 1990; GD 52.04.186–89 1991). Snow samples were taken to the depth of the profile in 11 L plastic buckets with a lid and collected using a polypropylene scoop. Water samples were taken with a polyethylene bucket. Samples were taken at the maximum distance from the nearest busy road.

The snow was melted at temperatures up to 23 °C in the laboratory. After that, the samples were filtered through a previously prepared "blue ribbon" filter (insoluble form of snow). The filter with the precipitate was dried in an oven at 105 °C. The soluble form of snow was obtained by passing melted snow through a Minisart PES 0.45 filter with a pore diameter of 0.45 µm. Samples for analysis of metal content were preserved with nitric acid to pH 1.

2.3 Analysis of physical and chemical parameters, and heavy metals

Further, salinity was measured in melt water by direct potentiometry using a Mettler Toledo Five Go F3 conductometer (Pilecka et al. 2017). The analysis of pH and Eh was carried out with a portable multiparameter pH/ORP meter HI 9126 (pH/ORP/T) (USA, Hanna Instruments) with a replaceable electrode for measuring pH and ORP.

The analysis of samples (soluble and insoluble forms of snow, water from wells) for metal content was carried out using the ICP-MS method (Aurora Elite, Bruker Daltonics, Inc.). The absolute error of the measurement method was $\pm 0.005 \text{ mg kg}^{-1}$.

2.4 Analysis of radioactivity

Determination of uranium was carried out according to the methodology (Methods for measuring the volumetric activity of uranium isotopes... 2013). As a result of radiochemical preparation, uranium isotopes in the form of a thin film are deposited on a steel substrate. The substrate was placed in the measuring chamber of the Progress-alpha spectrometer (Russia, RPE Doza) and the semiconductor alpha-spectrometer MultiRad-AS (Russia, RTC Amplitude). The determination and calculation of the volumetric activity was carried out relative to the introduced tracer ^{232}U in the 'Progress 5.10' program.

Preparation of counting samples for measuring the total alpha activity was carried out according to the methodology (Radiation monitoring technique 2013). The total alpha activity was measured on a semiconductor alpha spectrometer MultiRad-AS (Russia, RTC Amplitude) and an alpha-beta radiometer Abelia (Russia, NTC Amplitude).

2.5 Calculation of water quality indices

2.5.1 Heavy metal evaluation index (HMEI)

HMEI provides information about overall water quality as regards to heavy metals. The HMEI was calculated using the following Eq. (1):

$$HMEI = \sum_{i=1}^n \frac{HM_{Conc}}{HM_{MPC}} \quad (1)$$

where HM_{Conc} is the monitored concentration of a particular heavy metal and HM_{MPC} is the maximum permissible concentration of the same heavy metal (Zakir et al. 2020). To assess heavy metal contamination level in water, the threshold value was set at 1.0, which means HMEI value <1.0 is rated as 'Fit' and the value > 1.0 is considered as 'Unfit' for domestic usage (Zakir et al. 2020). A more extended classification of water pollution levels is also used (Caerio et al. 2005; Haque et al. 2019), presented in Table 2.

Table 2 – Classification of water depending on the value of the HMEI (Haque et al. 2019).

Water classification	HMEI
Very pure	$<0,3$
Pure	$0,3 - 1,0$
Slightly affected	$1,0 - 2,0$
Moderately affected	$2,0 - 4,0$
Strongly affected	$4,0 - 6,0$
Seriously affected	$> 6,0$

2.5.2 Heavy metal pollution index (HMPI)

The heavy metal pollution index (HMPI) is a rating model that provides the composite influence of individual heavy metals on overall water quality. The HMPI model is given by Eq. (2):

$$HMPI = \frac{\sum_{i=1}^n (Q_i \times W_i)}{\sum_{i=1}^n W_i} \quad (2)$$

where, Q_i is the sub-index of the i th heavy metal parameter, is the unit weight of the i th parameter reflecting its relative importance, and n is the number of parameters considered (Qu et al. 2018). The sub-index (Q_i) is calculated by Eq. (3)

$$Q_i = \frac{C_i}{S_i} \times 100 \quad (3)$$

where C_i is the concentration value of the i th heavy metal parameter ($\mu\text{g L}^{-1}$), and S_i is the highest standard permissible value of the i th parameter. We selected the World Health Organization (WHO) guidelines for drinking-water quality (WHO 2017) as the source of the highest standard permissible level (Table 3).

The unit weight (W_i) of the parameter is calculated by Eq. (4):

$$W_i = \frac{k}{S_i} \quad (4)$$

where k is a proportionality constant, we set k to 1 as Wanda et al. (2012). Generally, the critical HMPI value for drinking water is 100 (Prasad and Bose 2001). A modified scale using three classes is often used to better characterize moderate levels of heavy metal pollution: low (HMPI values < 15), medium (HMPI values within 15–30) and high (HMPI values > 30) (Edet and Offiong 2002).

Table 3 – Values of parameters of heavy metals required for calculating water pollution indices.

Element	$C_i, \mu\text{g L}^{-1}$	HIS_i	K_p	RfD_{ing}	RfD_{derm}	SF, mg L^{-1}
Al	200	685	$1 \cdot 10^{-3}$	1.3	0.07	-
Cr	50	895	$2 \cdot 10^{-3}$	3	0.08	0.5
Mn	500	797	$1 \cdot 10^{-3}$	24	0.96	-
Co	100	1011	$4 \cdot 10^{-4}$	0.3	0.06	-
Ni	20	993	$2 \cdot 10^{-4}$	20	0.8	1.7
Cu	2000	805	$1 \cdot 10^{-3}$	40	8	-
Zn	3000	913	$6 \cdot 10^{-4}$	300	60	-
Cd	3	1318	$1 \cdot 10^{-3}$	0.5	0.03	15
Pb	10	1531	$1 \cdot 10^{-4}$	1.4	0.42	0.0085
V	-	648	$1 \cdot 10^{-3}$	1	0.01	-
As	10	1676	$1 \cdot 10^{-3}$	0.3	0.12	1.5
Fe	300	0	$1 \cdot 10^{-3}$	700	140	-

2.5.3 Heavy metal toxicity load (HMTL)

The HMTL index assesses the content of heavy metals in water that affect human health (Saha and Paul 2018; Zakir et al. 2020). It is calculated by multiplying the studied content of metals in water ($C_i, \text{mg kg}^{-1}$) by their total hazard score (HIS_i , Table 3) and is calculated using the Eq. (5):

$$HMTL = \sum_{i=1}^n C_i \times HIS_i \quad (5)$$

The overall hazard score of the i-th heavy metal assigned by the Toxicological Profiles of the Priority List of Hazardous Substances prepared by the Agency for Toxic Substances and Disease Registry (ATSDR 2019). The HIS parameter is distributed based on the frequency of occurrence of the heavy metal, the level of toxicity of these substances and the likelihood of contact with humans. The maximum HIS for heavy metals is 1800. Within the framework of the conducted studies, the following classification of surface waters was introduced depending on the values of the metal toxicity index: 0–100 - low toxicity; 100–300, moderate toxicity; 300–500, high toxicity; 500–1000, very high toxicity; above 1000, extremely high toxicity.

2.6 Assessment of human health risk

2.6.1 Non-carcinogenic health risk (HI_{ing} , HI_{derm})

Major routes of exposure to metals include direct ingestion, inhalation and dermal absorption. The absorbed dose in humans is calculated on the basis of chronic daily intake (ADD) and refers to the dose of a pollutant per kilogramme of body weight per day that is absorbed through direct ingestion / skin absorption / inhalation (Kumar et al. 2019). In this paper, exposure to metals has been determined using the pathways for water ingestion and absorption through the skin, as these are two important pathways for exposure to heavy metals from the aquatic ecosystem (USEPA 2004). The exposure assessment for each heavy metal was calculated based on the risk assessment methodology recommended by the United States Environmental Protection Agency (USEPA) using Eqs. 6 and 7:

$$ADD_{ing} = \frac{C_i \times IR \times EF \times ED}{BW \times AT} \quad (6)$$

$$ADD_{derm} = \frac{C_i \times SA \times K_p \times ET \times EF \times ED \times CF}{BW \times AT} \quad (7)$$

where ADD_{ing} ($\mu\text{g kg}^{-1} \text{ day}^{-1}$) and ADD_{derm} ($\mu\text{g kg}^{-1} \text{ day}^{-1}$) are the average daily doses through ingestion and dermal absorption of water, respectively (Kumar et al. 2019; Saha and Paul 2018). In equations (6) and (7) C_i is the concentration of the HMs ($\mu\text{g L}^{-1}$), IR is the ingestion rate (2.0 L day^{-1}), EF represents exposure frequency (350 days), ED is exposure duration (30 years), BW indicates body weight (70 kg), AT is the average time (10950 days), SA represents exposed skin area (18000 cm^2), K_p is the skin adherence factor (Table 3), ET represents exposure time (0.58 h day^{-1}), CF is the conversion factor (0.001).

The non-carcinogenic risks were determined by applying the hazard quotient (HQ) of USEPA (2004) and calculated by Eq. (8):

$$HQ_{ing/derm} = \frac{ADD_{ing/derm}}{RfD_{ing/derm}} \quad (8)$$

where RfD_{ing} and RfD_{derm} are the ingestion and oral/dermal reference doses ($\mu\text{g kg}^{-1} \text{ day}^{-1}$), respectively (Table 3) (Kumar et al. 2019; Naveedullah et al. 2014; Iqbal and Shah 2013; Wu et al. 2009), HQ_{ing} is the hazard quotient through ingestion, and HQ_{derm} is the hazard quotient through dermal absorption.

The HI is the total potential non-carcinogenic health risks caused by different heavy metals present in waters. It was also calculated according to the USEPA guideline for ingestion and dermal adsorption of waters for the people of the study area using the following Eq. (9) (Zakir et al. 2020):

$$HI_{ing/derm} = \sum_{i=0}^n HQ_{ing/derm} \quad (9)$$

To assess potential non-carcinogenic health risks caused by different heavy metals present in waters, the threshold value was set at 1.0, which means HI value 1.0 means the possibilities of non-carcinogenic health risks may occur to the local people (Mohammadi et al. 2019).

2.6.2 Carcinogenic health risk

Incremental lifetime cancer risk ($CR_{ing/derm}$) was calculated due to exposure to a potential carcinogen (in this study: Cr, Ni, Cd, As, Pb). Potential carcinogenic risk possibilities that an individual may develop cancer over a lifetime of exposure are calculated by multiplying the ADD and slope factor (SF, $\text{mg kg}^{-1} \text{ day}^{-1}$, Table 3) together (USEPA 2004; Kumar et al. 2019). The equation used to calculate the CR for each carcinogen was as follows (Eq.10):

$$CR_{ing/derm} = ADD_{ing/derm} \times SF \quad (10)$$

In the present study, the values of SF for the above-mentioned carcinogens were introduced from the California Office of Environmental Health Hazard Assessment (OEHHA 2020). The CR of oral and dermal exposure of the above-mentioned carcinogens was considered in the calculation of total CR (ΣCR) for surface water. According to USEPA, the acceptable range for ΣCR is 1.0×10^{-6} to 1.0×10^{-4} for a single carcinogenic element and/or multi-element carcinogens, and values $> 10^{-4}$ indicate a high human risk of cancer (Mohammadi et al. 2019).

3. Results and discussion

3.1 Major physicochemical properties

The data obtained when studying the physicochemical parameters of the soluble form of snow and water from wells are shown in Figure 2. The pH parameter is an integral indicator of the ratio of acidifying and alkalinizing compounds and reflects the effect of these compounds on carbonate equilibrium (Vlasov et al. 2020). The acidity of atmospheric precipitation is the most important indicator, since acidic precipitation can significantly change the modes of functioning of natural components. The pH values for the soluble form of the investigated snow vary in the range of 5.21–6.24, but this interval does not fall within the range of pH values (6.5–8.5) recommended by the WHO for drinking water. The decrease in the acidity of winter precipitation is probably due to the presence in the atmosphere of a significant content of ash from urban boiler houses, solid fractions of burnt fuel, metal oxides, and ammonia (Saltan 2015). According to literature data, the acidity index of the snow cover in the Arkhangelsk area (35 km from Severodvinsk) is in the range of pH values from 5.74 to 7.48, which is close to the pH values in a neutral or slightly acidic medium (Chagina et al. 2016). Also, the pH value in the background area in the Middle Ob region is 5.5, which corresponds to the usual weak acidity of winter precipitation, while alkalization is observed in the city - the pH values varied from 6.8 to 7.4 (Pozhitkov et al. 2018). The water in the investigated shallow wells fed by melted snow and rainwater is characterized by higher pH values (from 6.40 to 7.39). The water in the wells (C-1 and C-2) farthest from the influence of factories and the heat and power station has the most acceptable pH values for drinking water.

For all the samples taken, positive values of the redox potential are observed, which indicates the occurrence of oxidative processes. Average values of ORP for the soluble form of snow (70.4 mV) are higher than for water from wells (40.8 mV), which is probably due to the infusion of groundwater with negative ORP into shallow wells.

The content of soluble salts in thawed filtered snow varies from 3.5 to 24.1 mg L⁻¹; moreover the highest values are observed at sampling sites (C-99 and C-100) located near the thermal power plant. The work (Pozhitkov et al. 2018) also describes a multiple (on average 7.8 times) increase in the salinity of thawed city snow. Mineralization of water from wells is in a wide range from 57 to 598 mg L⁻¹, which, however, does not exceed the permissible value of the salt content (above 1000 mg L⁻¹) in drinking water, established by the WHO (WHO 2017). The highest values of mineralization are found in wells C-3 to C-5, located near the industrial centre and thermal power station.

Figure 2. Physicochemical characteristics of the investigated water samples from wells and soluble form of snow.

3.2 Heavy metal concentration

The content of metals in snow and water from wells in the Severodvinsk area is presented in Table 3. It was revealed that the qualitative composition of snow precipitation in the study area is represented by metals of the 1st to

4th hazard classes. When studying the content of metals in the soluble form of snow, a high content of Fe (up to 2843 MPC) was found, but the concentrations for other metals do not exceed the MPC values for drinking water: Cr (up to 0.16 MPC), Ni (up to 0.13 MPC), As (up to 0.05 MPC), Pb (up to 0.04 MPC), Mo (up to 0.02 MPC), Cd (up to 0.02 MPC), Mn (up to 0.01 MPC), Co (0.00 MPC), Cu (0.00 MPC), Zn (0.00 MPC), as well as V (up to 2.23 $\mu\text{g L}^{-1}$) and Ti (up to 0.43 $\mu\text{g L}^{-1}$).

Of great importance for determining the nature of solid technogenic emissions into the atmosphere is the study of the insoluble fraction of snow - sediment consisting of dust (construction, cement, road), ash, and soot (coal combustion products). In the zone of influence of the thermal power station, a very voluminous sediment is formed in the snow cover, which includes soot and other products of coal combustion. The insoluble form of the investigated snow contains more metals than the soluble form. Thus, there are high concentrations of Al (up to 4680 MPC), Fe (up to 2807 MPC), as well as Ni (up to 66.5 MPC), Pb (up to 44.7 MPC), Cd (up to 43.3 MPC), Cr (up to 43.2 MPC), Mn (up to 13.3 MPC), Co (up to 7.3 MPC), and As (up to 3.4 MPC). Metals such as Zn (up to 0.85 MPC), Cu (up to 0.52 MPC) and, possibly, V (up to 225 $\mu\text{g L}^{-1}$) do not exceed the MPC. It should be noted that for the insoluble form of snow, there is a large variability in the values of the metal concentrations relative to the soluble form. Calculations of the metal content in the water-soluble and solid phases of snow have shown that in urban conditions, chemical elements are contained mainly in the insoluble phase, due to the predominance of poorly soluble compounds in urban emissions. The chemical composition of the snow cover of the high mountains of the Central Pyrenees can be used as global background concentrations (Bacardit and Camarero 2010): Mn (0.50 $\mu\text{g L}^{-1}$), Fe (9.01 $\mu\text{g L}^{-1}$), Ni (0.06 $\mu\text{g L}^{-1}$), Cu (0.06 $\mu\text{g L}^{-1}$), Zn (2.72 $\mu\text{g L}^{-1}$), Pb (1.92 $\mu\text{g L}^{-1}$), Al (8.61 $\mu\text{g L}^{-1}$) and Ti (0.46 $\mu\text{g L}^{-1}$). This is due to the fact that they are relatively remote places, as a rule, free from local inputs that mask sources arriving through the air from large distances (Battarbee et al. 2002).

The results of studying the metal content in water from wells showed that there is an extreme excess of the maximum permissible concentration for Fe (up to 56213 MPC). This is due to the feeding of the well with melted snow contaminated with iron, and the subsequent accumulation of iron in the water. According to the WHO (WHO 2017), Fe does not pose a health hazard at levels causing problems with the acceptability of drinking water and thus no guideline value has been postulated, but at levels above 0.3 mg L^{-1} Fe stains linen and plumbing. For the rest of the studied metals, the concentrations do not exceed the MPC: Mn (up to 0.92 MPC), Ni (up to 0.81 MPC), Pb (up to 0.65 MPC), As (up to 0.32 MPC), Cr (up to 0.27 MPC), Cd (up to 0.08 MPC), Zn (up to 0.08 MPC), Mo (up to 0.07 MPC), Cu (up to 0.02 MPC), Co (up to 0.01 MPC), and also Ti (up to 23.68 $\mu\text{g L}^{-1}$) and V (up to 16.6 $\mu\text{g L}^{-1}$).

Based on the research results, the concentration series of heavy metals in the insoluble form of snow have been constructed: $\text{Al} < \text{Fe} \ll \text{Mn} < \text{Zn} < \text{Cr} < \text{Cu} < \text{Ni} < \text{Pb} < \text{Co} < \text{V} < \text{As} < \text{Cd}$, and in the water from the wells: $\text{Fe} \ll \text{Mn} < \text{Zn} < \text{Ti} < \text{Cu} < \text{Ni} < \text{V} < \text{Cr} < \text{Mo} < \text{Pb} < \text{As} < \text{Co} < \text{Cd}$. It should be noted that high metal concentrations are observed for all the studied samples, including at the specially protected natural recreational complex "Pine Forest of Yagry Island". When compared, according to research data on melted snow in Arkhangelsk (Chagina et al. 2016), it was found that the main pollutant elements were: Fe from 45.3 to 83.8%, Mn from 11.9 to 48.6%, and Cu from 2.4 to 4.3%, while in the composition of solid particles the maximum mass fractions of metals were 31.31% for Fe, 4.98% for Pd, 0.75% for Ni, 3.23% for Zr, 0.55% for Ti, and V - 0.19% and Pt - 9.17%.

Table 3. The content of metals in snow and water from wells in the Severodvinsk area.

Sample	Metal content in the surface waters, $\mu\text{g L}^{-1}$													
	Mo	Ti	Cr	Mn	Co	Ni	Cu	Zn	Cd	Pb	V	As	Fe	Al
	ppb												ppm	
Water from wells														
CB-1	1.12	4.40	1.15	256	1.45	16.17	2.68	14.4	0.014	0.00	2.49	0.7	200	-
CB-2	2.52	1.28	1.33	155	0.76	6.98	1.9	17.6	0.023	0.07	3.15	1.69	538	-
CB-3	1.04	23.68	13.49	402	0.32	3.72	1.73	114	0.010	0.08	16.6	0.64	16864	-
CB-4	2.37	18.48	6.10	461	1.28	9.87	39.1	246	0.236	6.46	8.59	3.15	8198	-
CB-5	4.57	1.29	1.11	93	0.50	3.04	1.83	32.4	0.042	0.35	2.08	0.73	1086	-
Soluble form of snow														
C-97	0.88	0.08	5.50	1.67	0.10	1.60	0.85	10.4	0.042	0.41	0.19	0.04	127	-
C-98	0.90	0.09	5.54	1.54	0.14	1.63	0.76	13.6	0.010	0.36	0.26	0.09	127	-
C-99	1.68	0.30	6.43	7.18	0.30	2.63	0.93	7.22	0.019	0.31	2.23	0.53	649	-
C-100	1.31	0.43	6.09	7.18	0.31	2.54	1.04	10.9	0.046	0.28	1.57	0.54	132	-
C-101	0.98	0.26	5.92	6.21	0.10	1.60	0.72	8.54	0.010	0.41	0.69	0.38	144	-
C-102	0.93	0.09	5.82	2.50	0.17	1.81	0.89	8.83	0.014	0.38	0.87	0.31	139	-
C-103	0.77	0.21	5.55	2.01	0.11	1.68	0.56	6.08	0.028	0.36	0.55	0.08	146	-
C-104	0.83	0.14	5.68	2.27	0.13	1.63	0.82	10.6	0.037	0.35	0.56	0.05	161	-
C-105	0.97	0.16	7.90	3.97	0.16	2.12	0.70	4.25	0.018	0.37	0.68	0.14	853	-
Insoluble form of snow														
C-97	-	-	26	21	0.76	12	12	38	0.242	14	7.1	0.60	1.85	1.08

C-98	-	-	33	31	1.6	24	18	28	0.520	78	14	0.93	3.58	2.70
C-99	-	-	2160	6650	731	1330	1060	2560	130.0	447	225	34	842	936
C-100	-	-	875	2600	347	566	440	1040	60.0	235	186	32	275	378
C-101	-	-	15	34	0.85	11	5.3	13	0.127	5.2	7.6	0.91	2.38	2.42
C-102	-	-	41	127	2.50	22	20	93	0.519	23	32	6.9	9.05	6.90
C-103	-	-	21	135	1.10	12	7.6	11	0.235	9.2	11.09	1.2	2.99	2.33
C-104	-	-	14	20	0.63	12	7.0	11	0.192	4.7	4.6	0.69	1.52	1.49
C-105	-	-	15	22	0.72	13	6.5	18	0.294	5.1	6.2	0.72	1.58	1.55

3.3 Multivariate statistical analysis for physicochemical properties and heavy metals

In order to identify the relationships between heavy metals and determine the sources of snow pollution (the sum of individual metals of soluble and insoluble forms), statistical analysis was carried out using the Pearson correlation coefficient (Table 4), as well as the method of principal component analysis (PCA, Table 5), which is considered to be an effective method for identifying contamination pathways (Islam et al. 2018).

Using the correlation coefficient for snow, strong positive relationships (from $r^2 = 0.9$) were revealed between Mo-Cr, Mo-Mn, Mo-Co, Mo-Ni, Mo-Cu, Mo-Zn, Mo-Cd, Mo-Pb, Mo-V, Mo-As, Mo-Al, Cr-Mn, Cr-Co, Cr-Ni, Cr-Cu, Cr-Zn, Cr-Cd, Cr-Pb, Cr-V, Cr-As, Cr-Al, Mn-Co, Mn-Ni, Mn-Zn, Mn-Cd, Mn-Pb, Mn-V, Mn-As, Mn-Al, Co-Ni, Co-Cu, Co-Zn, Co-Cd, Co-Pb, Co-V, Co-As, Co-Al, Ni-Cu, Ni-Zn, Ni-Cd, Ni-Pb, Ni-V, Ni-As, Ni-Al, Cu-Zn, Cu-Cd, Cu-Pb, Cu-V, Cu-As, Cu-Al, Zn-Cd, Zn-Pb, Zn-V, Zn-As, Zn-Al, Cd-Pb, Cd-V, Cd-As, Cd-Al, Pb-V, Pb-As, Pb-Al, V-As, V-Al, As-Al, as well as significant correlations (from $r^2 = 0.7$) between Mo-Fe, Ti-As, Ti-V, Fe-Cr, Fe-Mn, Fe-Co, Fe-Ni, Fe-Cu, Fe-Zn, Fe-Cd, Fe-Pb, Fe-V and Fe-As (Table 4). The large number of strong and significant correlations between metals indicates the presence of one main source of environmental pollution in the study area.

Table 4. Correlation matrix from the studied parameters of snow of the Severodvinsk industrial district.

	Mo	Ti	Cr	Mn	Co	Ni	Cu	Zn	Cd	Pb	V	As	Fe	Al
Mo	1													
Ti	0.671	1												
Cr	0.967	0.610	1											
Mn	0.964	0.608	1.000	1										
Co	0.973	0.654	0.998	0.997	1									
Ni	0.970	0.623	1.000	0.999	0.999	1								
Cu	0.968	0.615	1.000	1.000	0.998	1.000	1							
Zn	0.969	0.605	1.000	1.000	0.997	1.000	1.000	1						
Cd	0.972	0.647	0.998	0.997	1.000	0.999	0.999	0.998	1					
Pb	0.964	0.623	0.985	0.983	0.989	0.987	0.987	0.985	0.988	1				
V	0.949	0.760	0.940	0.936	0.960	0.946	0.943	0.941	0.956	0.960	1			
As	0.927	0.778	0.905	0.901	0.930	0.913	0.909	0.908	0.926	0.929	0.995	1		
Fe	0.835	0.403	0.838	0.840	0.822	0.835	0.835	0.836	0.826	0.786	0.702	0.656	1	
Al	0.968	0.616	1.000	1.000	0.998	1.000	1.000	1.000	0.999	0.985	0.940	0.906	0.839	1

Number of samples = 9

Correlation is significant at the 0.01 level.

The diagram of factor loads (Table 5, Fig. 3) reveals 3 groups of factors that determine the accumulation of metals in the snow of the studied Severodvinsk area. The total variance for the four factors is 99%. The first factor with the main share of dispersion of 91.62% combines Mo, Cr, Mn, Co, Ni, Cu, Zn, Cd, Pb, V, As and Al, which is caused by atmospheric deposition of soot from thermal power plants. The second factor contributes 5.46% and consists of Ti, V and As, which are metals released into the atmosphere, probably through metalworking at shipyards. The third factor includes Fe with a total variance of 1.94%. This factor is probably a long-range transport of soluble iron from the atmospheric emissions of the thermal power plant towards the natural recreational complex 'Pine Forest of Yagry Island'.

Table 5. Factor loadings matrix for data set on snow of the Severodvinsk industrial district.

Parameter	Factor		
	1	2	3

Mo	0.748	0.412	0.485
Ti	0.292	0.939	0.154
Cr	0.822	0.311	0.474
Mn	0.819	0.308	0.479
Co	0.815	0.367	0.447
Ni	0.820	0.327	0.468
Cu	0.823	0.317	0.468
Zn	0.827	0.307	0.468
Cd	0.816	0.357	0.453
Pb	0.852	0.335	0.385
V	0.793	0.530	0.274
As	0.769	0.572	0.221
Fe	0.471	0.151	0.864
Al	0.818	0.318	0.476
% of Variance	91.62	5.46	1.94

Figure 3. Plot of factor loadings for snow of the Severodvinsk industrial district.

3.4 Quality indices of surface waters and health risk of heavy metals

3.4.1 Heavy metal evaluation index (HMEI)

Most of the HMs present in the investigated emissions belong to the first and second hazard classes. Their negative effect on humans is manifested not only in the direct effect of high concentrations, but also in the long-term consequences associated with the ability of many metals to accumulate in the body.

The HMEI pollution index is calculated on the basis of the sum of the values of certain metal concentrations to their maximum permissible concentrations. In accordance with the classification given in Table 2, it can be concluded that the snow and water in wells in all the study areas is seriously affected. The highest values of the HMEI index are observed for water samples in the wells CB-3 and CB-4, which is associated with the accumulation of a large amount of iron that gets into the wells when the contaminated snow melts. These wells are located close to factories and thermal power plants and are polluted by atmospheric fallout. The strongest metal contamination is characterized by snow from the C-99 sampling point near the thermal power plant and shipyards, where high HMEI indices are observed (2164 for the soluble form and 7710 for the insoluble form of snow). The soluble form of snow in sample C-105 (HMEI = 2844) due to the transfer of Fe and the insoluble form of snow in sample C-100 (HMEI = 2908) are also characterized by high HMEI values.

3.4.2 Heavy metal pollution index (HMPI)

According to the HMPI index, contamination of only a sample of the insoluble form of snow C-104 is classified with an average level of contamination, while all the other samples have a high level of contamination. Below the critical level of pollution (HMPI<100) are five samples of the insoluble form of snow; however, in general, the level of snow pollution is 4–101 times higher than the critical level. The level of the HMPI index for

water from wells is 36–295 times higher than the critical one; moreover, the greatest contamination, similar to the HMEI index, is observed for samples CB-3 and CB-4.

3.4.3 Heavy metal toxicity load (HMTL)

The HMTL index assesses the level of toxicity of heavy metals in waters that affect public health (Kumar et al. 2019). When interpreting the results obtained, it should be noted that when calculating this index, the toxicity of iron is zero. As a result, the soluble form of snow, for which the main pollutant is Fe, has a low toxicity for all samples (HMTL values at the level of 16–28). However, the HMTL level for the insoluble form of snow is very high with extremely high toxicity (HMTL 864 to 654702) due to the high content of Al as well as Cr, Mn, Co, Ni, Cu, Zn and Pb. Similar to the HMEI and HMPI indices, the highest toxicity was determined for the C-99 and C-100 snow samples. For water from wells, the toxicity level varies from moderate to very high (the highest for CB-3 and CB-4) due to the content of Mn and Zn, but it is worth mentioning the absence of toxicity in Fe.

3.5 Assessment of human health risk

3.5.1 Non-carcinogenic health risk (HI_{ing} , HI_{derm})

When assessing the potential non-carcinogenic risks to public health caused by heavy metals present in snow, the following was noted: soluble and insoluble forms of snow have a HI_{ing} index greater than 1, which corresponds to a high non-carcinogenic risk from drinking water. The highest HI_{ing} values (up to 19881) are characteristic of the insoluble form of snow, especially for samples C-99 and C-100. When exposed to a soluble form of snow through the skin (HI_{derm}), the non-carcinogenic effects are minimal, but for the insoluble form of snow, this index is greater than 1 and reaches a value of 1916. For water from wells, the following conclusions can be drawn: when drinking water (HI_{ing}), there is a high risk of non-carcinogenic effects for all samples; it is also necessary to note the high risk of water absorption through the skin for wells CB-3 to CB-5.

3.5.2 Carcinogenic health risk

Carcinogenicity is the potential for any type of cancer to occur throughout life when exposed to carcinogenic metals (Li et al. 2014). This index is calculated for oral and dermal exposure to carcinogenic metals such as Ni, As, Cd, Cr and Pb (ATSDR 2019).

The results of determining the risk of cancer when drinking water from wells showed a high possibility of carcinogenic effects (CR_{ing} up to $75.8 \cdot 10^{-4}$). Moreover, it should be noted that the greatest value is observed for the CB-1 well, remote from the industrial centre, for which the other calculated pollution indices are minimal. This is probably due to local contamination of the well. For all water samples from wells, high values of this index are dictated by high concentrations of Ni. By absorbing this water through the skin, the risk of cancer is at an acceptable level. Similar to well water, the soluble form of snow can cause cancer when ingested, but the risk is minimal when absorbed through the skin. The insoluble form of snow, heavily contaminated with Ni Cr and Cd, shows extremely high risk values for carcinogenic effects when consumed as drinking water (CR_{ing} up to 0.7038), as well as when there is exposure to water through the skin (CR_{derm} up to $12.4 \cdot 10^{-4}$). Snow samples C-99 and C-100, containing metals from emissions from thermal power plants and industrial shipyards, have the highest carcinogenic index.

Table 6. Health risk of heavy metals and quality indices of snow and water from wells in the Severodvinsk area.

Пробы	HMEI	HMPI	HMTL	HI_{ing}	HI_{derm}	HI	$CR(ing), 10^{-4}$	$CR(derm), 10^{-6}$	$CR, 10^{-4}$
Колодцы									
CB-1	668	359	243	8,4	0,3	8,7	75,8	8,2	75,9
CB-2	1794	949	157	21,6	0,6	22,2	33,5	4,0	33,5
CB-3	56215	29569	466	661,2	17,3	678,5	19,5	3,9	19,5
CB-4	27329	14396	671	322,3	8,5	330,8	49,1	6,9	49,1
CB-5	3621	1908	115	42,8	1,1	44,0	14,8	1,9	14,8
Растворимая форма снега									
C-97	424	225	19	5,1	0,1	5,2	8,4	1,7	8,4
C-98	424	225	22	5,1	0,1	5,2	8,4	1,6	8,5
C-99	2164	1141	25	25,6	0,7	26,3	13,4	2,4	13,5
C-100	440	235	28	5,4	0,1	5,5	13,1	2,3	13,1
C-101	480	255	23	5,8	0,2	5,9	8,5	1,7	8,5
C-102	464	246	20	5,6	0,2	5,7	9,4	1,8	9,4
C-103	487	258	16	5,8	0,2	6,0	8,7	1,7	8,8
C-104	537	285	20	6,4	0,2	6,6	8,5	1,7	8,6
C-105	2844	1498	19	33,5	0,9	34,4	11,1	2,2	11,1
Нерастворимая форма снега									
C-97	14	42	864	23,7	2,2	26,0	60,7	10,2	60,8
C-98	35	165	2100	59,6	5,6	65,2	118,8	17,7	119,0

C-99	7710	9015	654702	19881,0	1916,7	21797,7	7038,6	1241,8	7051,1
C-100	2908	3787	264615	8036,8	774,2	8811,0	3015,8	535,9	3021,1
C-101	22	31	1741	51,8	5,0	56,7	54,2	8,0	54,3
C-102	70	112	5058	148,6	14,2	162,8	113,1	19,2	113,2
C-103	24	42	1775	50,3	4,8	55,1	60,2	9,6	60,3
C-104	14	26	1089	32,0	3,1	35,0	58,9	8,4	59,0
C-105	15	30	1141	33,3	3,2	36,5	64,1	9,3	64,2

In general, the assessment of various pollution indices for water from wells and snow in the Severodvinsk territory unanimously led to the conclusion that the area is heavily contaminated with heavy metals. The most affected areas are the snow sampling locations C-99 and C-100 and water from wells CB-3 and CB-4.

3.6 Radioecological characteristics of water

In the present study, the radiological characterization of well water was carried out. The following concentrations of uranium isotopes were detected in water: 0.002–0.116 Bq L⁻¹ for ²³⁸U, 0.003–0.193 Bq L⁻¹ for ²³⁴U, and 0.0002–0.0027 Bq L⁻¹ for ²³⁵U. It should be noted that the contents of ²³⁸U and ²³⁴U correlate with each other and the largest amount is observed for the CB-3 well, and the minimum for CB-4 and CB-5. The maximum concentration of ²³⁵U is determined for the CB-1 well, and the minimum, similar to ²³⁸U and ²³⁴U, for CB-4 and CB-5.

The level of total α -activity, which is used for preliminary assessment of water quality, varies from 0.01 to 0.31 Bq L⁻¹ and correlates with the isotopes ²³⁸U and ²³⁴U. According to the WHO (WHO 2017), the level of intervention for total α -activity is 0.1 Bq kg⁻¹; therefore, samples CB-1 and CB-3 are characterized by values of α -activity above the intervention level.

The total concentration of uranium in water is 0.12–9.41 μ g L⁻¹ and also correlates with the isotopes ²³⁸U and ²³⁴U. Considering that, based on chemical toxicity, the approximate value of the total uranium content in drinking water should not exceed 30 μ g L⁻¹ (WHO 2017; Nuccetelli et al. 2012), it can be concluded that the water in all wells is suitable for drinking according to this parameter.

The radioactivity of natural uranium is due to the isotope ²³⁸U and its daughter nuclide ²³⁴U, and in the rocks the specific activities of ²³⁴U and ²³⁸U are equal (Nuccetelli et al. 2012). However, a violation of the radioactive equilibrium is observed during the transition of uranium into the liquid phase from minerals, and at the same time the enrichment of natural waters with the isotope ²³⁴U is observed (Borylo and Skwarzec 2013). In the studied water from wells, the ²³⁴U/²³⁸U ratio does not vary significantly in the range from 1.31 to 2.32, with an average value of 1.78 (Fig. 2d). The greatest value of the isotope ratio is observed at the CB-5 well.

On the whole, based on the results of radioecological analysis of water from wells near the Severodvinsk industrial region, it can be concluded that the radiation situation in the area under study is satisfactory.

Figure 4. Radiological properties of well water: a) total α -activity and activity of ^{234}U and ^{238}U isotopes in the studied waters; b) activity of the ^{235}U isotope in the investigated waters; c) total concentration of uranium isotopes; d) the ratio of the activities of uranium isotopes $^{234}\text{U}/^{238}\text{U}$.

Conclusion

As a result of the studies carried out on atmospheric air pollution in the Severodvinsk industrial region, it was established that the most important factors of the anthropogenic load with heavy metals are emissions from the fuel and energy complex and the shipbuilding industry. Changes in the physical and chemical parameters of snow and water from shallow wells have been revealed. Analysis of heavy metals by atomic adsorption spectroscopy showed that the investigated soluble fractions of snow and water from wells are predominantly contaminated with non-toxic iron, while the rest of the metals are present in amounts below the MPC for drinking water. However, the insoluble form of snow contains high concentrations of Al, Fe, Ni, Pb, Cd, Cr, Mn, Co and As. The statistical analysis performed made it possible to identify sources of pollution of snow with metals, as well as to assess the contribution of sources to anthropogenic impact. The use of indices made it possible to determine the quality of snow and well water. For some samples subject to the highest metal contamination, critical values are noted for the HMEI, HMPI, HMTL, HI and CR indices. This indicates the strongest anthropogenic metal pollution of atmospheric air, and, consequently, atmospheric precipitation, soil and natural waters. An assessment of the radioactivity of natural waters has been carried out, as a result of which it can be concluded that the radiation situation in the study area is satisfactory.

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