Features of the formation, intake and distribution of the iron-containing component in the water suspension of the Dnieper river within Zaporizhzhia city

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The article presents the results of research devoted to one of the aspects of environmental issues, namely: the determination of the distribution features of the solid component of the river suspension containing iron oxide; investigation of the morphological characteristics of iron-containing particles; and the discovering of their genetic affiliation to various technological processes of metallurgical production.

The results of many years of continuous comprehensive research on the distribution of iron in the suspended matter of the Dnieper water within Zaporizhzhia city are presented. Field observations were carried out by scientists of the Institute of Geological Sciences of the National Academy of Sciences of Ukraine (IGS) in cooperation with specialists from the State Institution "Scientific Hydrophysical Center of the National Academy of Sciences of Ukraine" (SHC). Analytical studies of the solid phase of the suspended matter were done in the Center for Collective Use of Scientific Equipment of the National Academy of Sciences of Ukraine and the Educational and Scientific Institute "Institute of Geology" of Taras Shevchenko Kyiv National University.

The publication considers the seasonal features of the distribution of the total iron content in suspension and the relationship of its monthly changes with the distribution of concentrations of such heavy metals as copper, chromium, nickel, zinc. Considerable attention is paid to the examination of iron-containing particles present in suspension, their chemical and trace element composition and the allocation of their individual categories in accordance with the technogenic processes during which they were formed.

Comprehensive analysis of suspension samples made it possible to identify and classify a group of typical iron-bearing formations that are inherent in the territory of Zaporizhzhia city. Based on a set of characteristics, three main genetic categories have been identified: spherical mineral aggregates represented by iron oxide; clastic fragments of Fe2O3, and aluminosilicate spherules (formation, the main or essential component of which is aluminum oxide with an admixture of iron).

The obtained data showed a significant influence of the metallurgical industry on the chemical composition of the Dnieper suspension within Zaporizhzhia. Comparison of iron-containing particles observed in the Dnieper suspended matter with the substance filtered out by gas purification systems of various parts of metallurgical production and slag deposits showed that the most active emission to the environment occurs during the preparation and smelting of iron ore.

Keywords: ecology; river suspension; iron oxides; metallurgy; Zaporizhzhia city; Ukraine.


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Introduction

Long-term field observations of the distribution of pollutants in the environment are considered effective mechanisms for obtaining new knowledge in the field of environmental research. Such knowledge provides a reliable environmental monitoring system and allows detecting the appearance of negative changes. A significant number of relevant publications on this subject are devoted to the distribution of metals in the atmospheric environment, surface waters, land soils, bottom sediments, and river suspension (Boboko et al., 2007; Kruopiene, 2007; Ali et al., 2022; Zhang et al., 2022).

Zaporizhzhia city occupies a leading position in Ukraine among industrially loaded metallurgical centers. The city territory can be considered as a full-scale test site for studying the distribution of emissions of the metal-containing component as a result of the corresponding production processes. A significant concentration of enterprises of ferrous, non-ferrous metallurgy and mechanical engineering causes in some areas of the city an intensive emission of a number of microelements into the environment, including iron and heavy metals. The distribution of pollutants depends on a complex of natural factors, such as the strength and direction of winds, the hydrodynamic and hydrochemical regimes of surface watercourses, the features of the local relief, and other indicators.

Taking into account the above natural factors during the research, we can conclude that pollutants belong to certain technological processes, trace the sequence of their distribution in natural conditions, temporal transformation and interaction with natural objects.

The comprehensive studies envisaged by the scientific task included monthly observations of quantitative and qualitative changes in the composition of the river suspension, atmospheric aerosol, the state of land soils and bottom sediments of the Dnieper. The continuity and long period of observations allowed to determine the features of the distribution of ferrous and heavy metals in transit flows of sedimentary matter and in accumulation areas, to discover and evaluate the patterns of redistribution of their forms and concentrations depending on natural and anthropogenic factors (Fig. 1).

The article presents the research results of features of the distribution of the solid component of the river suspension containing iron oxide, the morphological characteristics of these particles and the elucidating of their genetic affiliation to various technological processes of metallurgical production. In the environmental sphere, these works are also important because such particles can be carriers of heavy metals, quite often including significant concentrations of such metals (Glagoleva, 1959; Dobrovol’skiy, 1979; Mitropol’skii et al., 1982).
Methods

The study of water suspension is one of the reliable tools for studying ecological problems of water bodies, because suspended particles accumulate and store information about the course and intensity of the processes of anthropogenic influence on the water environment (Allan, 1986; Galas et al., 2006; Schubert, 2012). The growing man-made pressing and the expansion of the spectrum of pollutants entering the environment actualize the following areas of research and the development of analytical methods for the study of sedimentary matter and modern sedimentation processes.

Sedimentation traps are the main tools of water suspension research. Conducting field studies of organic, mineral, chemical composition, planar distribution and intensity of suspended matter flows using sedimentation traps is an established practice of researchers (Schloss et al., 1999; Barbizzi et al., 2008; Masson et al., 2018; Kim et al., 2019).

The technique for sampling suspended matter is based both on methodological recommendations (Moroz et al., 1990; Cockburn et al., 1999; Zajączkowski, 2002) and on our own experience gained during such work in marine areas (Nasedkin et al., 2013).

Monitoring observations of the Dnieper suspension were carried out near the berth of the SHC (Zaporizhzhia city, lower reaches of the Dnieper). Observations included continuous accumulation of suspended matter in vertical sedimentation glasses developed in IGS, and monthly sampling of this matter for further laboratory research. The high information content and efficiency of using such sediment traps is confirmed by our many years of practical experience (Nasedkin et al., 2013).

The selected suspension was settled and dried. To study the composition of the suspension, the methods of mineral, granulometric and chemical analyzes were used. The mineral, macro- and microcomponent, chemical composition of the samples of sedimentary matter was determined using electron microscopy, as well as energy and wave dispersive analysis in the Center for Collective Use of Scientific Equipment of the National Academy of Sciences of Ukraine.

Natural and technogenic conditions within the study area

The total length of the Dnieper river within Zaporizhzhia city is 24.2 km, the average long-term runoff volume is 53 km³. Dnieper and Kakhovka reservoirs
influence on the hydrochemical regime of the city area. The hydrographic network of Zaporizhzhia city includes Dnieper, small rivers, such as Mokra Moskovka, Sukha Moskovka, Verkhnya Khortytsya, Serednya Khortytsya, Nyzhnya Khortytsya, Kabytsya, a lot of streams and arroyos. The total length of small rivers within Zaporizhzhia city is 26 km, streams is 11.5 km, arroyos with watercourses is 22.3 km (Fig. 2).

According to (Regional…, 2018; Strategic…, 2019) the most significant factors that have the greatest impact on the ecological state of water bodies within Zaporizhzhia are:

- discharge of polluted and insufficiently treated return waters due to inefficient operation of treatment facilities or their absence at all;
- flushing of pollutants from urban areas;
- discharges of drainage water when protecting irrigated agricultural land and settlements from flooding;
- violation of the economic activity regime within the coastal and water protective zones.

Outdated treatment technology, deterioration of equipment, untimely repair and overhaul are the main reasons of poor-quality treatment of return and waste water. The largest polluter of water bodies within Zaporizhzhia city is the metallurgical industry, in particular, Zaporizhzhia metallurgical plant PJSC “Zaporizhstal” and a number of other industrial enterprises located in the same area of the city, discharging polluted wastewater into the sludge collector of “Zaporizhstal”, such as electrometallurgical plant PJSC “Dniprospetsstal”, PJSC “Zaporizhzhia Aluminum Production Plant”, PJSC “Zaporizhzhia Steel Rolling Plant”, CJSC “Zaporizhzhia Plant of Metal Structures” and some others. The volume of return water discharge from these enterprises reaches almost half of the total volume of water discharge from all enterprises in Zaporizhzhia (Strategic…, 2019), and the percentage of PJSC “Zaporizhstal” in the total volume of discharge of polluted return waters in the whole region is 80.8 % (annually about 5 \times 10^7 m^3) (Regional…, 2018; Strategic…, 2019).

As for pollution from the public utilities sector (volumes of pollution entering water bodies), the storm sewerage of the city, together with rain and melt water, are the main pollutant of the Dnieper, especially during periods of intense precipitation or snow-melt. Pollution significantly increases due to the annual discharge of more than ten million m^3 of polluted wash water into the Dnieper, which is the result of natural water treatment at the Dnieper waterworks No. 1 and No. 2. (Regional…, 2018; Strategic…, 2019).

According to the results of monitoring the ecological state of surface waters in the region, carried out by the Zaporizhzhia Regional Department of Water Resources in the upstream and downstream of the Dnieper HPS and outlets of the central treatment facilities, the iron content in water reaches an average of 0.25 g/m^3, exceeding the fishery safe exposure levels approximately 2.5 times (Regional…, 2018; Strategic…, 2019).

Results and discussion

The carried out field and analytical studies, as well as the generalization of the information obtained during the observation period, made it possible to identify and evaluate the contribution to the complex composition of suspended matter of the component that with high probability can be considered anthropogenic. Comparison of the total iron content in the composition of the river suspension within experimental site, located 10 km downstream from the area of metallurgical industry concentration, and the corresponding average values for the waters of the Dnieper (Mitropolskii et al., 1982) showed significant excesses of the iron oxide content, reaching 13 % of macrocomponents composition of dry matter of suspension samples. The results of SEM investigations and chemical analysis of samples also indicate a significant role of metal-containing particles (of aleu-ritic dimension) in the formation of the main phase of the mineral component of suspended matter in the study area. In the conditional distribution of components by volume, the metal-containing component occupies the third place after mixed-layered formations of chlorite-illite-montmorillonite and skeletal remains of siliceous microalgae. Iron is the predominant chemical element in the metal-containing component. According to the results of the analyses the formation of such particles is genetically associated with the technological processes of grinding iron ores and their high-temperature processing.

The seasonal features of the distribution of the iron-containing component in the composition of suspended matter were studied; in addition, we analyzed the influence of hydrometeorological factors on the formation and change in the intensity of flows of metallurgical products within the studied water area. A summary of the data obtained over a five-year research period and averaging the distribution
of the main components of suspended matter by months showed that the second place in the total amount of mineral matter, after silica, is "shared" by iron oxide and calcium oxide (13% of each in the total content of basic oxides) (Fig. 3). Insignificant fluctuations in the content of the metal-containing component were detected in different periods of natural substance sampling, the minimum was observed in the spring. The correlation of recorded natural and anthropogenic factors with the distribution of iron depends on a significant increase of organic silica content, and to a lesser extent on changes in the intensity of winds from land and the features of the hydrological regime. In turn, the increase of SiO₂ content during spring periods is probably associated with the peak of siliceous phytoplankton productivity (phytoplankton valves present in large numbers in the suspended matter during these periods). Simultaneously with a decrease in the concentration of iron oxide, a reducing the concentrations of alumina and calcium oxides occurs, in other periods of the year their concentrations increase.

At the same time, persistent associations of a number of these components are observed. In particular, the correlation between the time distribution of Al₂O₃ and Fe₂O₃ reaches 0.75, between Al₂O₃ and MnO exceeds 0.6; a significant negative correlation coefficient is observed between SiO₂ and Fe₂O₃ (-0.84) and SiO₂ and Al₂O₃ (-0.77). Perhaps, such ratios should be considered in the context of the distribution of the factors of the anthropogenic component (Fe, Al, Mn) input from the atmospheric environment into surface waters.

The iron-containing components of the river suspended matter can be divided into several categories according to morphological characteristics, the distribution of microelements and the dimension.

Iron in river waters exists in two main forms, soluble and suspended. Soluble forms (components with sizes of up to 0.1 µm; can pass through a membrane filter) consist of a truly soluble form and of highly dispersed aggregates (colloids). The suspended form is iron existence in the composition of solid particles of different genesis and chemical forms found in the aquatic environment. According to (Mitropolskii et al., 1982), in the suspended matter of the Dnieper, as well as all rivers of the basin of the northwestern part of the Black Sea, iron exists mainly in: suspended form (fixed in the clastic component of rocks); an exchange form (hydroxides), and organic form. At the same time, a certain natural geochemical balance is maintained between the distribution of the number of elements in solution and suspended form, which depends on such external factors as the changes in the pH of the environment, the hydrodynamic and hydrochemical regime of the water area, as well as the size, composition and distribution of suspended matter density in the aquatic environment (Krasintseva et al., 1977).

In the analysed samples the main amount of Fe is associated with flows of fine suspension of aulervit-pelite size, consisting of mixed-layered formations of chlorite-illite-montmorillonite and remains of siliceous microalgae. Enrichment of the substance with iron and a number of heavy metals (Zn, Cu, Ni, Pb, etc.) represented by a finely dispersed

![Fig. 3. Average monthly distribution of the main components of the river suspension substance (%) within the experimental area (2015–2019)](image-url)
component is associated with a certain mineral composition, as well as with the natural properties of the pelitic part of the river suspension and bottom sediments to accumulate some trace elements from the aquatic environment. A visual assessment of such component is almost impossible due to the insignificant dimension of its components, as well as their aggregation in the variegated formations, which are difficult to assess and describe qualitatively.

At the same time, the composition of the river suspension contains a sufficient amount of relatively well-identified iron-containing components, which, with a high degree of probability, can be genetically linked to technogenic processes. The first category is spherical mineral aggregates (Fig. 4), represented by iron oxide Fe₂O₃ (ferruginous spherules), of varying degrees of corrosion, sometimes with microimpurities of ZnO, TiO₂, MnO, and CuO. The particle size of the main phase is on average from 0.1 to 20–30 µm, in rare cases it can reach 70 µm; objects of silty dimension are characterized by impurities of manganese and titanium. The ferruginous spherules have relatively regular spherical shape (the content of iron oxides, as a rule, exceeds 80 %). The approximate percentage of such formations in the total distribution of the identified iron-bearing component of suspended matter is significantly inferior in volume to the clastic component and reaches 20 %.

![Fig. 4. SEM images: Fe₂O₃ spherules from the Dnieper suspended matter, sampled by sedimentation traps at the monitoring point. Sampling periods: a – December–January 2019; b – December–January 2019; c – May–June 2016; d – March–April 2016](image-url)
The formation of spherules is the result of iron ores smelting, metalworking, welding and other man-made processes using high temperatures. They enter the aquatic environment from the atmosphere. Ferruginous spherules are the most common anthropogenic components in the environments of territories adjacent to metallurgical industries. They are found in the composition of bottom sediments and river coastal drifts that are under the influence of emissions from enterprises. It is believed that ferruginous spherules are particularly unstable to environmental factors and are exposed to decomposition processes in the river environment (Menshikova, Osovetskii, 2015). Oxidation and other changes in such formations can affect the ecological state of the environment, since some of the toxic elements are able to go into the ionic state, forming the composition of natural waters.

An analysis of literary sources on the issue of the genetic affiliation of iron-bearing spheres to natural geological processes indicates that their entry can occur with cosmic matter, the sources of their formation can be both endogenous processes (Yatsenko et al., 2012) and exogenous processes of ferromanganese nodule generation in the aquatic environment.

At the same time, for the study area, these factors do not have a significant effect on the formation of an iron-containing substance in comparison with the intensity of its introduction into the composition of technogenic flows.

The second component of technogenic origin in the river suspension is clastic fragments of Fe₂O₃.
(with microimpurities of MnO), represented by a significant range of dimensions from 5 µm to 70 µm or more in the form of acute-angled, sometimes slightly rolled grains. Metal-bearing grains contain microimpurities of a number of metals (Cu, Cr, Ni, Ti, Zn, Mn), the content of Fe2O3 according to microprobe chemical analysis can be up to 80-95 %. At the same time, in contrast to iron-bearing condensation formations, dispersive formations are present not only in the pelitic and silty components, but even in the fine psammitic (Fig. 5).

The enrichment of the dispersive component with a significant amount of iron oxide fragments of aleuritic dimension is explained by the insignificant distance to mining facilities. The concentration of iron in the ore can significantly exceed the background; when such iron-containing particles are transferred to another geochemical province as a result of human activity, they may be considered pollutants (Zhovinskii, Kuraeva, 2002). The chemical analysis carried out to identify the forms of the element in suspension showed that more than 80 % of the total iron content is a component fixed in the crystal structure of rock fragments (that directly confirms the origin of these particles).

The third category of the suspended matter component is aluminosilicate spherules consisting of aluminum oxide (Fig. 6). They are also potential results of high-temperature processes of human production activities (metallurgical and related industries, operation of thermal power plants, etc.). The mineral composition of these spherules (60–70 %) is represented by aluminosilicates with an admixture of iron and microimpurities of heavy metals (mostly, zinc and copper). Their average size is 1–20 µm, but individual spherules are 50 µm or more. As for other metal-containing components, their dimension is determined by the characteristics of emission treatment systems at industrial enterprises and thermal power facilities.

One of the features of aluminosilicate spherules (as well as the difference from ferruginous spherules) is their discontinuous structure, they are usually hollow inside. The formation of cenospheres (hollow aluminosilicate balls) according to (Zyrianov V., Zyrianov D., 2009) occurs as part of the fly ash in the furnaces of thermal power plants (TPP) and hydroelectric power plant (HEPP) during high-temperature burning of coal. During the combustion of finely divided coal particles, impurities of aluminum oxide, silica and other elements present in natural coal form composite silicates, which take a spherical shape in the molten state. Due to the gases (nitrogen, oxygen and carbon oxide) dissolved in silicates, spherical microdroplets of molten silicates blow up into microspheres (Cenospheres, 2019).

Morphological characteristics of the particles of the technogenic component in an aqueous suspension makes it possible to determine the genetic affiliation of the condensed and dispersed components to various technological processes for the preparation and processing of iron ore at the enterprises of the metallurgical complex of the city.

This is proved by the results of laboratory studies of the substance of atmospheric emissions from some metallurgical industries in Zaporizhzhia city. In particular, the dust obtained in the process of wet gas cleaning of open-hearth furnaces, as well as from the filters of electric steel-smelting furnaces, largely consists of iron-containing spherules and cenospheres, similar in chemical composition and external features to those analyzed by us in suspended matter (Nasedkin et al., 2019). The substance filtered out by the systems of wet gas cleaning of process gases of sintering machines and electrostatic precipitators of the tail section of sintering machines, in morphology, dimensions, and chemical composition, to a large extent corresponds to the detrital component of iron oxide, which is also observed by us in samples of river suspension. The dispersed iron-containing substance is also present in the sludge of the treatment facilities of the industrial zone of metallurgical facilities of Zaporizhzhia. This may indicate the entry of a certain amount of iron-containing particles by water from the territories of sludge storages, in particular, with the waters of Sukha Moskovka river.

The presence of a large number of aluminosilicate balls on the air filters of the technological capacities for smelting silicomanganese and the production of ferrosilica enterprises in Zaporizhzhia, which in composition, dimensions and external features correspond to those observed by us in the Dnieper suspension (Nasedkin et al., 2019), indicates their significant contribution to the pollution of water surfaces at the same level with objects of thermal power engineering.
Conclusions

The undertaken studies have shown that the iron content in the river suspension of the Dnieper within Zaporizhzhia city is more than twice the background indicators for the section of the middle course of the water artery. A significant iron-containing component of river suspended matter is solid particles of silty dimension, which were formed or were transferred from other territories (including mining sites), and also got into the aquatic environment due to human activities.

The analysis of suspension samples, continuously have been taken for 5 years by sediment traps in the lower reaches of the Dnieper within the city, made it possible to identify and classify a number of typical iron-bearing formations inherent in the territory of Zaporizhzhia. According to the complex of morphological characteristics, the distribution of microelements and the dimensions of the components, they could be divided into three categories: ferruginous spherules (spherical mineral aggregates represented by iron oxide Fe₂O₃; Fe₂O₃ fragments; and aluminosilicate spherules (the main or essential component of them is aluminium oxide with an admixture of iron).

At the same time, typical for the Dnieper suspension within other urban agglomerations not associated with the metallurgical industry are Al₂O₃ spherules with an admixture of iron. Their formation is associated with the functioning of thermal power facilities, their presence is recorded in all components of the environment (unlike particles with a high iron content).

The obtained data indicated a significant influence of the metallurgical industry on the chemical composition of the Dnieper suspension within Zaporizhzhia city. Comparison of iron-containing particles observed in the Dnieper suspended matter with the substance filtered out by gas purification systems of metallurgical industries and slag industries showed that the most active emission to the environment occurred during the preparation and smelting of iron ore. Each type of process emissions at the enterprises of the metallurgical complex of the city is characterized by different types of solid particles.

The continuation and expansion of such studies, the determination of the environment state within the industrially loaded regions of Ukraine might make a significant contribution in the future to the development of recommendations on practical measures to protect human health and environment under anthropogenic pressure conditions.

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Особливості утворення, надходження і розподілу залізовмісної складової у водній зависі р. Дніпро в межах Запоріжжя

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Викладено результати досліджень, які присвячені одному з аспектів екологічної проблематики – визначенню особливостей розподілу твердої компоненти річкової зависі, що містить оксиди заліза, дослідженю морфологічних характеристик залізовмісних частинок та з’ясуванню їх генетичної належності до різних технологічних процесів металургійного виробництва.

Представлено результати багаторічних безперервних комплексних досліджень розподілу заліза у завислій речовині дніпровської води в межах м. Запоріжжя. Польові спостереження проведено науковцями Інституту геологічних наук НАН України у співробітництві з фахівцями Наукового гідрофізичного центру НАН України. Аналітичні дослідження твердої фази завислої речовини виконано в лабораторіях Центру колективного користування науковим обладнанням ІГН НАН України та Наукового гідрофізичного центру НАН України.

Розглянуто сезонні особливості розподілу загального вмісту заліза в зависі та зв’язок його змін з розподілом концентрацій таких важких металів, як мідь, хром, нікель, цинк. Значну увагу приділено висвітленню наявних у зависі залізовмісних частинок за морфологією, хімічним та мікроелементним складом та відділенню окремих їх категорій відповідно до техногенних процесів, у ході яких вони утворились.

Всебічний аналіз зразків зависі дозволив виявити і класифікувати групу типових залізовмісних утворень, властивих самій території м. Запоріжжя. За комплексом характеристик визначено три основні генетичні категорії: сферичні мінеральні агрегати, представлені оксидом заліза, уламкові фрагменти Fe 2O3 та алюмосилікатні сферули – утворення, основну чи суттєву компоненту яких складає оксид алюмінію з домішкою заліза.

Отримані дані засвідчили значний вплив металургійної індустрії на хімічний склад дніпровської зависі в межах Запоріжжя. Порівняння виявлених у завислій речовині Дніпра залізовмісних частинок з речовиною, відфільтрованою системами очистки газів різних ланок металургійних виробництв та шлаковідвалів, показали, що найбільш активна емісія в навколишнє середовище відбувається в процесі підготовки та плавлення залишкової руди.

Ключові слова: екологія; річкова завись; оксиди заліза; металургія; м. Запоріжжя; Україна.