Wastes from the processing of tin ore as a potential source of pollution of ecosystems and a reduction in the risk of environmental disasters in the Primorsky krai

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Abstract. The article presents the results of long-term experimental studies of the environmental hazard of mining waste for environmental objects in the Primorsky krai. The aim of the work is to develop the fundamental basis for an integrated assessment of the impact on the ecosystem of waste from the processing of mineral raw materials to improve the environmental situation in the study area. A high ecological toxicity is revealed, which, obviously, can contribute to contamination of environmental objects, as evidenced by acidity indicators (pH 3-4), which determine the high mobility of heavy metal compounds. It has been experimentally proved that the compounds of toxic chemical elements contained in its waste are actively involved in the geochemical cycle within the boundaries of the influence of the tailing dump. A comprehensive assessment of the impact of waste on ecosystems has made it possible to propose ways to ensure their environmental and social security, the novelty of which is confirmed by the patents of the Russian Federation received by the authors of the article.

1. Introduction
The intensive economic development, because of the steady increase of science and technology progress, has led to an inevitable increase of mineral consumption. In this regard, the increase in the mineral production in the last century and the sharp increase of the exploitation of the Earth's subsoil have contributed to the accumulation of mining waste and to the technogenic pollution of ecosystems. Despite the obvious benefits of mining for good of the human being, it is also a potential source of environmental hazard for the environment and people. It is known that many chemical elements, contained in the processing waste, except industrial value, are also characterized by different degrees of toxic impact on the biosphere - vegetation, microorganisms, and another living organisms, including human beings. Entering the animal and human body with food, water, and air, they cause many illnesses, functional disorders and, even, death. The environmental situation has deteriorated significantly, because, since the USSR collapse, at the end of the last century, many large tin-ore mining enterprises have failed to cope with the economic difficulties of the transition period and...
ceased their operations. Unfortunately, the large quantities of toxic wastes, stockpiled in tailing ponds, have remained uncontrolled, and their conservation and reclamation have not been carried out. An intensive pollution of the biosphere components due to their long-term environmental impact occurred. The increased technogenic environment impact has led to a high-stress environmental situation in the vicinity of the mining enterprises of the Primorsky krai, which, evidently, contributed to the emergence of environmental diseases of the population in mining townships.

In this regard, the research goal was to develop the fundamental foundations for a comprehensive assessment of the impact on the ecosystem of waste from the processing of mineral raw materials to reduce the risk of environmental disasters and improve the environmental situation in the study area.

Based on the purpose of the study, the research has the following tasks: (1) to analyze, summarize and systematize literary sources and materials of patent search on the named problem; (2) to estimate tailing dumps as real objects of potential threat of ecological catastrophes; (3) to comprehensively assess the impact of toxic waste on the processing of tin ore on ecosystems; (4) to propose measures to reduce the risk of man-made disasters.

2. Objects and Methods of Research
The objects of research were the natural-mining technogenic systems formed in the process of tin materials development by mining companies:

– “Dalpolimetall” and “Khrustalnensky MPP.” They include tailing wastes in ponds in the form of pulp, landslides, piles of unstable ores and empty rocks, as well as environmental objects.

Modern instrumental, traditional physicochemical and biological methods, as well as atomic emission spectroscopy, cartographic modeling, scientific prediction, systematization and scientific classification, zoning (metallogenic, landscapegeochemical), statistical data processing using software.

3. Results and Discussion
A number of conclusions were drawn based on the analysis, compilation, and systematization of literary and patent data for creation of the fundamental framework for assessment of the present state of the technogenic system. (1) In the course of mineral development, the heavy metals compounds coming into natural ecosystems from mineral-processing wastes are actively involved in the cycle of substances and participate in metabolic processes of living organisms. They migrate by food chains, accumulate in natural environment objects [1-4], and constitute a threat to human life [5-8]. (2) The specificity of the biogeochemical landscape and ecosystem situation stipulates the behavior of toxic chemical elements in the components of the biosphere [9-11]. (3) At present, the main patterns of the migration of pollutants in the environment are not sufficiently studied. (4) The technogenic load on landscapes, caused by a negative impact of mineral wastes on human health, contributes to the emergence of environmentally induced diseases [7, 8, 10]. Children are the most sensitive to technogenic pollution, they require special attention in solving the complex problem: “Technogenic pollution from wastes – Environment – Health of Nation” to ensure environmental and social security of mining wastes. The tin-sulphide deposits in Komsomol’sky, Kavaleryovsky and Dal’negorsky districts of the Far East region were developed by the open and underground mining. In Komsomol’sky district the mining industry existed from 1957 to 2005. In the district, there are two factories and three tailing dumps. Two of them have been dried, and the third one is in part covered with a slime lake. The tailing dumps occupy the area of 80.8 ha, and the tailings mass is 41.5 million tons. They are composed of (%): veined quartz – 37.5, tourmaline – 12.1, hornfels-sedimentary rocks – 45, and sulphides (pyrite, pyrrhotite, arsenopyrite, chalcopyrite, galena and sphalerite) – 3.8. They contain the following valuable minerals (g/t): Sn – 0.2, Cu – 0.46, Zn – 0.094, Pb – 0.123, Ag – 1.227, Bi – 0.03, As – 0.629.
The objects of the Kavalerovsky district are six mines and four concentrating mills that existed from 1941 to 2001. In the district, there are five tailing dumps with a total area of 17.7 ha, where 37.72 million tons of the tailings have been accumulated, which are composed of pyrite, pyrrhotite, galena, sphalerite, arsenopyrite, chalcopyrite, quartz, fluorite, tourmaline, chlorite, and other minerals. The quantitative and semiquantitative spectral analyses of the samples showed that the ore elements content in them ranges as follows: Sn – 0.04-0.10; Cu – 0.0062-0.2600; Pb – 0.0039-0.0760; Zn – 0.081.00; As – 0.01-0.05; Ni – 0.0014-0.0033; Co – 0.0002-0.0009; Cr – 0.0019-0.0030; V – 0.00430.0100; Ag – 0.0003-0.0030; Ga – 0.0011-0.0016; B – 0.01-0.05; Bi – 0.0001-0.0003; Sr – to 0.01, Ca – to 0.1.

The tin-sulphide ores of Krasnorechenskoe deposit of Dal’negorsky district were processed from 1956 to 1995 at the same-named concentrating mill (KCM). The concentration wastes were stored at the old and new tailing dumps of the KCM of the area of 300 thousands m², and their mass is 6.8 million tons. The mineral composition of the concentration tailings is represented by ore minerals as follows: pyrrhotite, pyrite, chalcopyrite, galena, sphalerite, and others. Among the non-metalliferous minerals are quartz, calcite, fluorite, chlorite, and other minerals. The chemical composition of the tailings is (%): Zn – 0.27-0.29; Pb – 0.11-0.18; Cu – 0.01-0.03; Fe – 4.37-4.60; Ag – 5-6 (g/t).

The development of the mining industry in the study districts brought into existence the mining technogenic systems in them, in which as a result of the increase of the contact surface of sulphides with weathering agents (water, oxygen, carbon dioxide, and so on) the hypergene processes activated. They are based on the reactions of oxidation and hydrolysis. These result in the formation of mine, slime, and drainage technogenic waters. Most of them are highly concentrated, and the technogenic minerals are crystallized from them the technogenic minerals containing Cu, Pb, Zn, Fe, and other elements from the class of oxides and hydroxides, sulfates, carbonates, arsenates, silicates, and so on. The technogenic minerals such as chalcocite, brochantite, posnjakite, wroewolfeite, woodwardite, glockerite, hisingerite, and others are found universally in quarries, on the walls and roof of mining workings (adits), and on the surface and banks of the tailing dumps [12, 13].

Our studies have confirmed the real ecologicalgeochemical hazards associated with the development of the deposit and accumulated mining wastes (sludge and tailing dumps, piles of lean ores and overburden). It is the wastes, often finely fragmented high-dispersed soils, intensively oxidizing in the aeration zone and containing remnants of chemical reagents of flotation ore enrichment, that are the sources of toxic chemical elements and their compounds, polluting the environment. The tails (waste) are represented by a fine gray mass, sometimes in different shades of brown by iron hydroxides, formed by sulphides oxidation. The results of our calculations show that the hazard class of the wastes of the Solnechny (Khabarovsky territory) and Khrustalnensky (Primorye territory) closed enterprises is 2. A detailed study of the specific nature of the technogenic geochemical flows formed around tailing ponds made it possible to establish: (1) the identity of mineral deposit geochemical spectrum and the generated technogenic flows in different media; (2) the relationship between sulphides in ore and their composition variations determines the nature of the negative impact on the environment components; (3) the impact of local environmental conditions on the chemical elements flow range and on generation of secondary sources of pollution. Sulphides in hypergene conditions are the most volatile compounds and easily destroyed, which leads to the formation of mobile compounds of metal salts and sulfuric acid, which contributes to the chemical destruction of both sulphides and other minerals.

Despite the fact that mining enterprises are closed (such as “Solnechny MPP,” “Khrustalnensky MPP”), the formation of air and water technogenic geochemical objects now (after their bankruptcy) continues to intensify. Our studies are confirmed by Bubnova’s data (2007) that indicates that the migration of the substance is appropriate-directed and sub-coordinate to natural mechanisms in the watersheds (denudation), from high to low marks along the associated water body channels and in accordance with the dominant wind direction, duration, and intensity.

The nature, intensity, and morphology of environmental pollution is largely determined by the location of the tailing dump in the catchment basin. The location in the estuary part of the basin is the
most environmentally friendly, and it is the worst for the environment within this basin in the near-watershed part, since the role of area geochemical pollution in the latter case is much greater [14].

A description of the generated technogenic flows according to [12, 13] and others is given below.

Air flows. A few studies have been carried out on the special research of this problem, considering the Primorsky krai mining enterprises of non-ferrous metallurgy. It has been established that the air near these objects is determined by dust and aerosol content in the surface atmosphere layer. Seventy per cent of technogenic load in the mining areas fall on the soil. It is the soils which are central in relationship of the biosphere components and are able to accumulate information on contemporary and past technical genesis processes.

In the Solnechny settlement, the content of heavy metal compounds in soils changes within the following limits (per cent): Pb – 0.040-0.006, Zn – 0.073-0.085, Cu – 0.004-0.104, Cd – to 0.0003, which exceeds the background for Pb by 30-45 times, Cu – 5-50, Cd – 30, and Zn – 10.

Water technogenic flows in the Komsomolsk mining area are studied much better than air flows, since the result of this type of pollution is more obvious. The analysis of the mineral composition of hypogene and technogenic minerals shows that the following compounds of heavy metals are brought into the natural waters within the boundaries of the tailings of the former mining enterprise – Solnechny MPP: Zn, Cu, Sn, Pb, Mn, Fe, Ag, as, Co, Ni, Bi, Cr, Sb, Cd, W, B, Li, Sr, Al, Ca, Mg, K, Na, S, P, Si, Ga, and Ge.

To date, the most complete information on ore drainage and slime waters of tin deposits is available in the works [12, 13].

The analyzed samples of rainfall, snow, and vegetation in the tin-ore region have shown that the highest concentrations of heavy metal compounds are associated with the initial phase of drizzling precipitation, where the Zn, Pb, Fe content may reach 100-200 mg/l and Cu, Mn, Al – 30-60 mg/l [12, 13].

In the technogenic geosystem, the Pb content in local run-off waters is 2.5 times higher than in background conditions, Al - 3 times, and the rest metals (Cu, Cd, Zn, Fe, V, Ti) – 1.5-1.8 times. Increase of heavy metal compounds concentrations in local run-off waters indicates an apparent technogenic influence, although the concentration of heavy metal compounds is low. Mine and slime waters, including the tin-ore deposit under study, are enriched in both the major (Sn, Cu, Pb, Zn, Fe, Mn, Al, and T) and rare elements (As, Sb, Cd, in, Sr, etc).

Based on the analysis of own materials, as well as the systematization of literature data on the detailed study of the regularities in the migration of chemical elements from wastes to ecosystems (man-made soils, vegetation, lichens, microorganisms, waters, sediments, humans), the following conceptual provisions are formulated:

– The area of geochemical technogenic pollution of the various environment components from tailing ponds is always greater than any other violations (especially for regional, long-lasting, natural mining-technological systems);
– The morphology of negative impact zones depends on the way of the deposit development and local conditions (topography, forest cover, hydrological regime, predominant wind direction, etc.); the scale of impact of the closed mining enterprises (“Solnechny MPP,” “Khrustalnensky MPP,” and others) are mainly determined by the volumes and methods of the past development of a particular deposit or their group, composition of ores and rocks, technology of their production, enrichment and processing, and the specificity of the environment, including the soil ability to self-cleaning;
– In the non-ferrous metal deposits, the composition and ratio of sulphide minerals and scale, speed, and nature of chemical contamination of the environmental components in the course of these objects operation are determined.

Soils, sediments, and surface water were used as test sites when carrying out the area studies. The same objects were tested in the study of technogenic flows in mining areas. In these terms, the following picture of the study of technogenic environment pollution within the boundaries of the
impact of the closed mining enterprises “Solnechny MPP” and “Khrustalnensky MPP” has emerged. (1) Soils are the main object of the environment, depositing different types of pollution and acting as both buffer and detoxicant. (2) The bottom sediments of local watercourses emerging from active water exchange also have a high capacity to accumulate many chemical compounds. Their transformation into sediments is carried out by the mechanical deposition of suspensions, metal enriched organic sediments on the bottom of the water bodies, co-precipitation in the course of salting and sorption directly. A sharp increase of the weighted forms proportion is one of the main features of the tailing ponds impact in the studied region on the nature of the elements migration in the water systems. (3) The formation of contrasting and long lithochemical flows of ore and related chemical elements in bottom sediments is a feature of water migration in mining areas. The transition of chemical elements into water flows is due to the natural and climatic characteristics of the Primorsky krai with a humid monsoon climate. The temperature contrasts are important in predetermining the intensification of the sulphide ores oxidation in the hypergenesis zone and the formation of ionic solutions of metals.

The integral assessment of the waste impact on the environmental objects (air basin, snow cover, soil, vegetation, surface water, and groundwater) led to the conclusion that the environment conditions within the boundaries of tailings are estimated as critical at a distance of 7-8 km from tailing ponds, as unsatisfactory – at a distance of 12-13 km and partially satisfactory – at a distance of 20-22 km. It was found that the content of heavy metals and arsenic compounds exceeds the background values from 2 to 45 times, not only in water, bottom sediments, snow cover, but also in soils (both total and mobile shapes) and plants. The highest concentrations were found in environmental objects near tailing ponds (up to 7-8 km). The total pollution index (Zc) was 149-157, while the concentration coefficient (Kc) in the residential zone (up to 2-3 km from the tailings ponds) did not correspond to a permissible pollution category.

**Table 1.** Biondication of ecological tension on sterility of plant pollen-bioindicators within the limits of influence of tailing dump of the closed mining enterprise “Solnechny MPP.”

| Name of the mining enterprise / plant selection site | Name plants - bioindicators | Number of cells examined | Sterility of pollen | Impact area |
|----------------------------------------------------|-------------------------------|--------------------------|--------------------|-------------|
| 1                                                  | 2                             | 3                        | 4                  | 5           |
| Tailings Storage Facility                          | Mayweed                       | 500                      | 21.9±0.57          | 1           |
| “Solnechny MPP,” 1 zone of impact                  | Anaphalis pearl               | 500                      | 18.4±0.57          | 1           |
| Tailings Storage Facility                          | Small-leaved Cyprime          | 500                      | 19.9±0.49          | 1           |
| “Solnechny MPP,” 2 zone of impact                  | Mayweed                       | 500                      | 11.9±3.10          | 2           |
| Tailings Storage Facility                          | Anaphalis pearl               | 500                      | 9.9±0.67           | 2           |
| “Solnechny MPP,” 3 zone of impact                  | Small-leaved Cyprime          | 500                      | 7.8±0.59           | 2           |
| Tailings Storage Facility                          | Mayweed                       | 500                      | 5.9±0.57           | 3           |
| “Solnechny MPP,” 3 zone of impact                  | Anaphalis pearl               | 500                      | 4.6±0.35           | 3           |
| “Solnechny MPP,” 3 zone of impact                  | Small-leaved Cyprime          | 500                      | 5.3±0.29           | 3           |
| Control                                            | Mayweed                       | 500                      | 0.19±0.17          | 3           |
|                                                   | Anaphalis pearl               | 500                      | 0.28±0.18          | 3           |
|                                                   | Small-leaved Cyprime          | 500                      | 0.30±0.17          | 3           |

In order to evaluate the response of biota to the negative effect of waste enrichment, the test system “Pollenicity of pollen” was used (table). Studies have revealed that, as a result of mining technogenesis, the physiological state of living organisms (biota, microflora) deteriorates, and the
genetic status of the population as a whole changes. This is expressed in an increase in the sterility of the pollen grains (up to 25-38%), which entails a low germination of the seeds.

Estimating the composition of microflora, we can conclude that in the state of deep biological destruction are the soil, judging by the bacterial complex. Thus, the impact of waste from the processing of mineral raw materials stored in tailing ponds causes negative changes in the soil and vegetation cover, undermining its recovery potential.

According to our research the ecological system has been severely disrupted here. Its recovery period will be not less than 30 years after the total elimination of the adverse impact source.

It is apparent that the mining-industrial systems and the hypergene and technogene processes proceeding in them result in the pollution of all natural constituents: air, soil, water, and living organisms including humans. The study of these processes have been fulfilled using the Selector- Windows program complex, elaborated at the A.P. Vinogradov Institute of Geochemistry, Siberian Branch of RAS (Irkutsk), that is based on the thermodynamic method with the calculation of the equilibrium composition of solutions and solid phases – hypergene and technogene minerals, originated through the oxidation of sulphide ores in the mining workings and concentration tailings.

Modeling was done within the temperature interval of -25 to +45 C with different ratios in the sulphide system: pyrite, pyrrhotite, chalcopyrite, arsenopyrite, galena, and sphalerite with regard to the minerals of the cementation zone (native Cu, covellite, bornite, and chalcocite). The ratio between the sulphides and enclosing rocks in the concentration tailings is as follows: 5:95, 10:90, 20:80, 40:60, 60:80, and 80:20.

Modeling of the oxidation processes in the concentration tailings in the Komsonol'sky district showed that Eh-pH parameters of the systems vary from 0.75 to 1.15 B and 1.32-8.04. It was established that the following minerals are crystallized from the highly concentrated solutions: Fe and Al from the class of oxides and hydroxides (goethite and gibbsite), sulfates of Cu, Fe, Pb, K, Al, and Ca (antlerite, ktenasite, woodwardite, fibroferrite, plumbojarosite, jarosite, alunogen, and gypsum), arsenates of Fe, Cu, and Pb (pitticite, scorodite, olivenite, duftite, clinoclase, and bayldonite), carbonates of Ca, Mg, Fe, and Zn (calcite, magnesite, siderite, and smithsonite), phosphate of Fe – vivianite, and silicates of Al, Na and Fe (allophane and nontronite). Arsenate of Fe (scorodite) as well as Cu and Pb (bayldonite) precipitated only at the temperature of below 0C. In the modeling of the processes of the sulphide oxidation in the concentration tailings with regard to the minerals of the cementation zone the Eh-pH parameters of the systems vary from 0.75 to 1.15 B and 1.3-8.0. In them, there are the minerals of Fe and Al from the classes of oxides and hydroxides (goethite, gibbsite), sulphates of Cu, Fe, Pb, Al, Mg, and Ca (chalcanthite, woodwardite, anglesite, fibroferrite, alunogen, starkeyite, gypsum), carbonates of Ca, Mg, Zn (calcite, magnesite, smithsonite), arsenates of Cu and Pb (olivenite, bayldonite), and silicates of Na, Al, and Fe (allophane and nontronite). Sulfates of Mg and Cu (starkeyite and chalcanthite) precipitated only at the temperature of below 0C, and Pb (anglesite) – at the temperature of above 0C [12, 13].

The oxidation of the sulphide component of the concentration tailings of the tailing dumps in Kavalerovsky district results in the formation of the solutions with the Eh-pH parameters of 0.42 to 1.09 B and 2.7-13.3. From the modeling solutions the following minerals are crystallized: Fe, Cu, Pb, Ca, Al, and Mg from the class of oxides and hydroxides, sulphates, carbonates, arsenates, and silicates (goethite, tenorite, fibroferrite, alunogen, woodwardite, anglesite, gypsum, magnesite, duftite, bayldonite, allophane, and montmorillonite), and Zn passes in total into the solution. The elimination of arsenopyrite from the modeling systems results in the crystallization of technogene sulfates of Cu (antlerite, posnjakite, and wroewolfeite), in the systems without pyrite, sphalerite, pyrrhotite, and galena the arsenite of Cu precipitates (olivenite, duftite, and bayldonite), olivenite – only with the absence of the two latter sulphides, and sulphate of Pb – anglesite is marked in the models without pyrite, sphalerite, and arseno- and chalcopyrite [12, 13].

The oxidation of the sulphide component of the KCM tailings of the Dal’negorsky district results in the formation of the solutions with the Eh-pH parameters of 0.98 to 0.66 B and 3.7-9.7, from which the following hypergene minerals are crystallized: Fe, Pb, Zn, Sb, and Ca from the classes of sulfates,
oxides, and hydroxides (fibroferrite, plumbojarosite, anglesite, adamite, gypsum, goethite, hydrogoethite, and valentinite) [12, 13].

The characteristics of the modeling systems showed that the technogenesis results in the formation of the highly concentrated solutions containing both the elements of sulphide ores (Fe, Cu, Pb, Zn, As, Sb, Ag, and S) and enclosing rocks (K, Na, Ca, Mg, Al, Si, and others, which before and after the precipitation of technogene minerals from them, with the total mass of about 233 g) enter the surface and ground waters of the district and affect them adversely. The excess over the background indices in them reaches the tens of thousands times, which is well verified by the data of the study of hydrochemical samples of technogene waters in the study districts [12, 13].

Thus, the main source of pollution of the districts’ environment is the waste of the closed mining enterprises (Solnechny and Khrustalnensky GOKs) and the operating company (Dalpolimetall), because they have accumulated a huge amount of toxic chemical elements. Undoubtedly, they have a negative impact on the ecosphere and human.

On the basis of these studies, the methods of reducing the negative impact of toxic wastes on environmental objects have been developed, their novelty was confirmed by RF Patents [15-19]. A number of recommendations have been prepared for reducing the risk of environmental disasters in the closed mining enterprises of the Far East Federal District.

4. Conclusions
The research results indicate that the toxic wastes of reprocessing of tin materials accumulated in tailings ponds by both closed and operating mining enterprises constitute a major threat to the environment and to humans of the Far East Federal District. The recommendations are proposed to reduce the risk of environmental disasters in the area under study.

5. Acknowledgements
The reported study was funded by RFBR according to the research project № 18-35-00260.

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