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Modelling submicron aerosol formation in the atmosphere of mining regions

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Abstract

This article is concerned with the urgent problem of atmospheric air pollution in the post-industrial phase of the development of mining regions. Methodological approaches to the hygienic diagnostics of local aerosol pollution of the atmosphere have been elaborated. The modeling object was the Ursk dump of barite-pyrite loose material, formed in the 1930s after gold extraction by cyanidation in Ursk settlement, the Kemerovo Region. The field model of the dump was formed by the samples of sulphide-containing wastes collected from the object in summer. At the first stage, a theoretical analysis of the information presented in the literature on the physicochemical and toxic properties of aerosol particles in the submicron range, which pose risks to the environment and public health in mining areas, was carried out. At the stage of hazard identification, the application of the field modeling of atmospheric migration processes of target particles and mercury vapour from a sulphide-containing waste disposal facility was scientifically justified. At the modelling stage, which consisted of heating the waste samples to a temperature of 28.2±0.1, 33.0±0.1 and 50.0±0.3 °C, the trends of air concentrations of aerosol particles of various ranges and their correlations were estimated. At the design stage, the probability of accumulation of submicron particles and mercury vapour in the air basin, which poses a significant danger to the environment and public health, was determined. The data obtained from the results of simulation are intended for information provision of planning and implementation of measures for land reclamation and environmental improvement.

Keywords: sulphide-containing waste disposal facility, atmospheric migration of aerosol particles, field modeling, health hazard of aerosol particles

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INTRODUCTION

Specificity of atmospheric air pollution in the post-industrial phase of the development of mining regions is determined by the enormous accumulated amounts of mining wastes exceeding 100.109 tonnes [1, 2]. The group of most dangerous wastes includes those from mining and beneficiation of the sulphide ores of nonferrous and precious metals, which are accommodated as dumps and tailings if no technologies for re-extraction of valuable components are available [3, 4]. Even after withdrawal from operation on completion of ore deposit development, sulphide-containing waste disposal facilities remain the sources of atmospheric pollution with multicomponent gas-aerosol mixtures [5]. The leading role in the genesis of atmogeochemical anomalies in the post-industrial phase is transferred to the natural-technogenic processes, which are a set of environmental

responses to the technological environmental impact [6].

On the basis of the results of plume monitoring (measurements of impurity concentrations under the axis of the plume of pollutant release from the source), it is reasonable to consider dimethylsulphoxide and dimethylsulphide, along with the vapour phase, as the priority components of local aerosol pollution of the surface atmospheric layer in the regions where sulphide-containing tailing dumps are emplaced [7]. At the sites of waste storage after processing gold-containing ores, in particular by cyanidation and amalgamation, a possible component of man-made aerosol pollution of air is mercury vapour, increasingly emitted under the action of solar radiation and heating of the surface of waste disposal sites [8]. This causes pollution of natural ecosystems and residential territories (areas intended for housing and social objects) with mercury, its compounds being rated as the 1st hazard category. The dangerous properties of mercury compounds include high volatility, increasing with an increase in ambient temperature; long-term stability in ecosystems; solubility of mercury vapour in atmospheric precipitation; ability to be sorbed in soil and adsorbed on suspended particles in aquatic systems [8]. In spite of substantial species-specific differences in the sensitivity to mercury in animals, experimental studies have revealed correlations between exposure to mercury and endocrine disorders [9], hepatic and renal irritation [10], embryotoxicity [11], neurotoxicity [12] and fertility disorders [13]. The toxic effect of mercury can spread over a broad range of organisms, which determines the necessity to solve the problem of environmental pollution with this substance. From the viewpoint of hygienic practices, mercury is classified as one of the dangerous and toxic environmental pollutants, creating a high risk for human health [14].

According to the fundamental regularities known in human ecology and toxicology, the negative effect of atmospheric pollutants on the organism depends not only on the chemical composition but also on the size of particles in air suspension [4]. The specific effect of aerosol particles within the submicron size range is due to their ability to penetrate into the lower airways, followed by distribution over many organs and tissues [15]. Polytropic influence of submicron aerosol on human organisms has been established in [16, 17]. In particular, changes in the functions of immune and central nervous systems, damage of respiratory organs and blood circulatory system, an increase in the rate of neoplasms in the groups under exposure, the diseases of urogenital system, digestive organs and locomotor system have been diagnosed [18–21]. Aerosol particles formed from automobile emissions belong to the risk factors of increased mortality in adults and the incidence of chronic bronchitis in children [22–25].

The negative effect of sulphide-containing waste disposal facilities has been under thorough investigation for many years by researchers in our country [5, 26] and abroad [27, 28]. By present, a connection between the temperature of surface layers of the sulphide-containing tailings and atmospheric emission of sulphur-containing compounds has been confirmed [5]. However, the issues related to the local aerosol pollution of the surface atmospheric layer in mining regions remain almost absent from publications, which hinders their environmental and hygienic assessment and scientific substantiation of the measures aimed at amelioration and improvement of the environment.

The goal of the present work is to develop methodological approaches to the hygienic diagnostics of local aerosol pollution of the atmosphere in the emplacement sites of the wastes from processing sulphide ores of nonferrous and precious metals, for information support of planning and implementation of the measures aimed at amelioration and improvement of the environment in mining regions.

The major objectives of the investigation are: evaluation of the natural model of a dump with sulphide-containing wastes, assessment of the dependence of submicron particles emission amount on the temperature of heated waste samples, and prediction of the distribution of their concentrations in the surface layer of the atmosphere during summer.

EXPERIMENTAL

The modelling object was the Ursk damp emplaced in Ursk settlement of the Kemerovo Region (barite-pyrite bulks from the oxidation zone of the pyrite Novo-Urskoye deposit after gold recovery by cyanidation) formed in the 1930s. The samples of sulphide-containing wastes, a reduced copy of the dump, collected in summer were used as the natural model of the dump.

The modelling procedure was as follows: the samples of sulphide-containing wastes 1 kg in

TABLE 1

Dynamics of the averaged concentrations of submicron aerosol particles (0.2–0.5 μm) in chambers at different temperatures of waste samples

Measurement	Particle concentration, 10^4 units/L		
	Chamber 1 (50.0 °C)	Chamber 2 (33.0 °C)	Chamber 3 (28.2 °C)
Series 1	15.7±0.7#	14.1±0.6#	22.2±1.0
Series 2	63.4± 3.6*	28.5±2.2*	24.6± 1.7
Series 3	30.2±1.9#*	17.6±0.9#*	14.7±0.6#*

Note. Here and in Tables 2 and 3: series 1 – before heating; series 2 – during heating; series 3 – after heating the sample.

* The difference of data from series 1 is significant for $p < 0.0001$.

\# The difference of data from series 2 ($p < 0.0001$).

TABLE 2

Dynamics of the averaged concentrations of mercury vapour in the chambers over a day during heating the samples of wastes

Measurement	Concentration of mercury vapour in the air of chamber, ng/m^3		
	Chamber 1 (50.0 °C)	Chamber 2 (33.0 °C)	Chamber 3 (28.2 °C)
Series 1	3.4±0.3	4.0±0.2	3.4±0.5
Series 2	5.6±0.6*	5.4±0.8	2.6±0.2
Series 3	3.6±0.3#	3.7±0.3	3.1±0.2

* Difference of the data from series 1 is significant for $p < 0.02$.

\# Difference of the data from series 2 ($p < 0.05$).

mass were placed on the hotplates in airtight chambers 200 L in volume, and heated to a temperature of 28.2±0.1, 33.0±0.1 and 50.0±0.3 °C in chambers 3, 2 and 1, respectively. The modes of thermal action imitating variable intensity of solar radiation were chosen on the basis of statistical data on the seasonal soil temperature variations for the Kemerovo Region [29]. The trends in temperature-induced changes of the amounts of emitted aerosol with different particle size distribution were determined by measuring the gas-phase particle concentrations in chambers every day in three series of measurements (series 1 – before heating, series 2 – during heating, and series 3 – after heating the waste samples), carried out at 9, 12 and 16 h, which corresponds to measurement series 1, 2, 3 in Tables 1, 2. The total number of air samples was 99 per series; particle concentrations in these air samples were determined within the size range of 0.2–10.0 μm using a multifunctional aerosol particle spectrometer SAChM 4801 (AeroNanoTech, Russia). The concentration of mercury vapour released from the waste samples was measured in the chambers using a multipurpose portable mercury-measuring complex UKR-1MTs (EkON, Russia), based on the flameless atomic absorption within the range of 10–50000 ng/m^3.

The digital data obtained in the measurements are represented as $M \pm m$, where M is arithmetic mean, m is the error of arithmetic mean (see Tables 1, 2). The differences were considered to be significant for $p < 0.05$, when the probability of differences is 95 % and more. Plotting and editing with database building and subsequent data output were performed with a PC using the standard applications Statistica 10.0.

RESULTS AND DISCUSSION

The results obtained in the 12 days long approbation of the natural model of Ursk dump with barite-pyrite bulks have confirmed that the inferred pollutants of the surface air layer above the dump, for heating the object up to a temperature of 50.0±0.3 °C, include aerosol particles within the size range of 0.2–0.5 μm: their concentration in the air of chamber 1 in the second series of measurements increases by a factor of 4 with respect to the initial one (see Table 1).

The studies have shown that after reaching the peak concentration at a temperature of 44 °C, a trend to decrease arises, which is depicted in Fig. 1. However, even with a decrease in the temperature of waste sample to 27.4±0.1 °C, 4 h later after completion of thermal action, the concentration of these particles reliably, by a factor of 1.9, exceeds the level detected before heating (see Table 1). This allows assuming that the lifetime of submicron aerosol in summer is also dependent on the extent of heating the surface of the emission source.

The maxima of particle concentrations within the indicated size range in chamber 2, achieved at 30 and 27.5 °C in the third series of measurements are shown in Fig. 2; these values are higher by a factor of 2.1 and 2 than the initial level. At the same time, these maxima, compared to the concentrations achieved by heating the sample to 44 °C (the second series of measurements), are 1.7 and 3.8 time lower, respectively. Concentration oscillations, distinguished by the decaying behaviour, reached their minimum at a temperature of 33 °C. After cooling to 27 °C, in the third series of measurements, the concentration of aerosol particles 0.2–0.5 μm in size reliably exceeded by a fac-

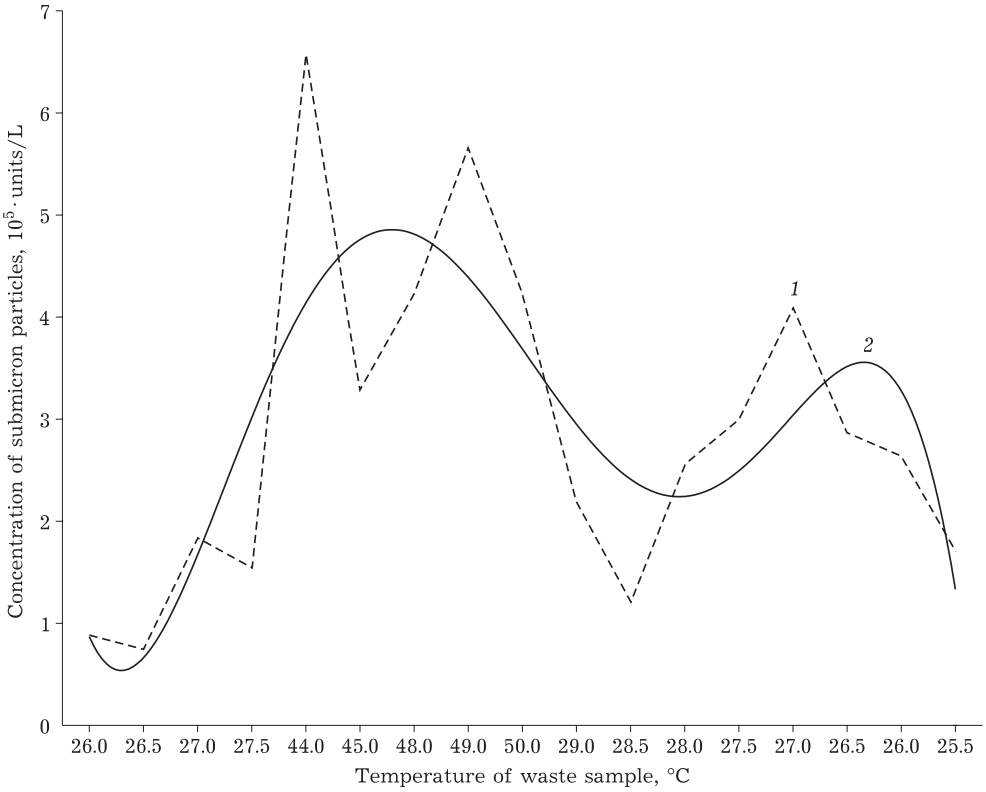

Fig. 1. Dependence of the concentration of submicron aerosol particles (0.2–0.5 μm) in chamber 1 on the temperature of the heated waste sample. Here and in Fig. 2–6: comparison of actual experimental values (*1*) and the approximating curve (*2*).

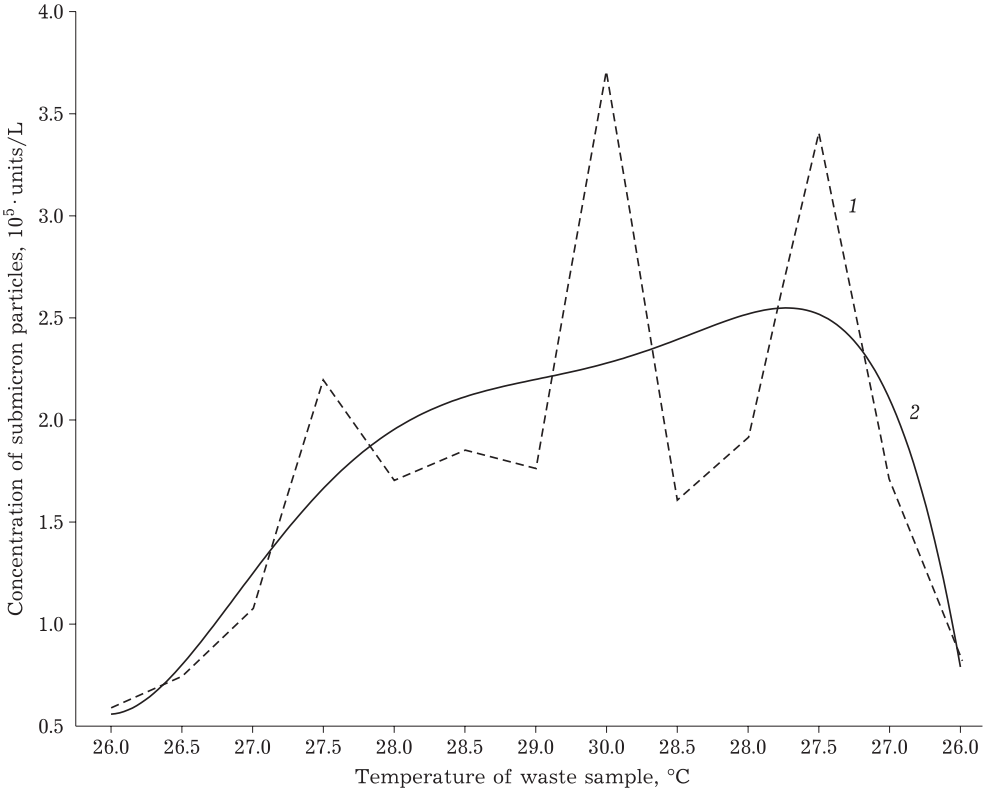

Fig. 2. Dependence of the concentration of submicron aerosol particles (0.2–0.5 μm) in chamber 2 on the temperature of the heated waste sample. For designations, see Fig. 1.

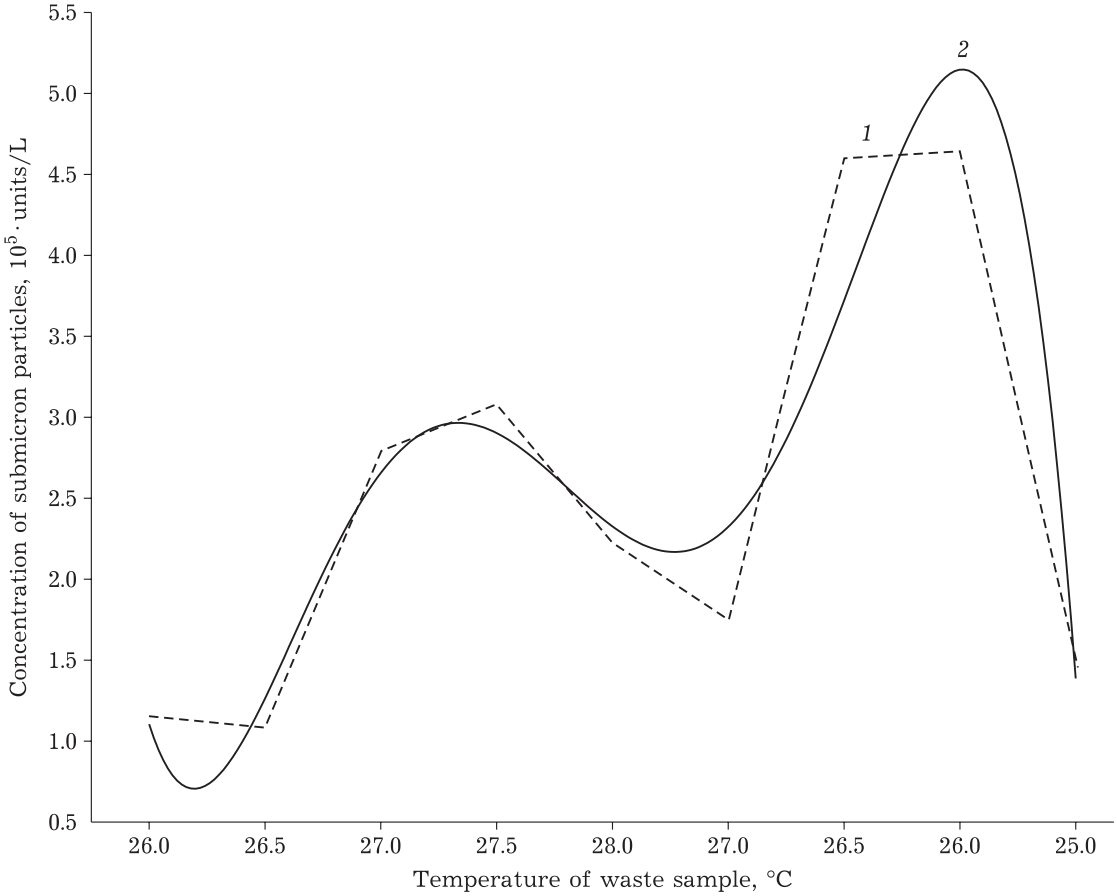

Fig. 3. Dependence of the concentration of submicron aerosol particles (0.2–0.5 μm) in chamber 3 on the temperature of the heated waste sample. For designations, see Fig. 1.

tor of 1.6 the level of the first series of measurements at the same temperature.

Distribution of the concentrations of aerosol particles with particle size 0.2–0.5 μm in chamber 3 is presented in Fig. 3 as a fluctuating curve, with the minimum ($10.8 \cdot 10^4$ units/L) is recorded at the waste matter temperature of 26.5 °C in the first series, the maximum ($46 \cdot 10^4$ units/L) – at a temperature of 26 °C in the third series of measurements. The levels of aerosol particle concentrations in the air of this chamber over a span of three series of measurements increased by a factor of 4.3. However, the visual analysis of the plot revealed that the slope of trend line with respect to the abscissa axis was substantially smaller than in other chambers.

Results of modelling the atmospheric emission of mercury vapour (see Table 2) show that its intensity depends on sample heating temperature and, to a smaller extent, on the measurement timeframe. One can see that the averaged concentrations of mercury vapour in air at heating the waste sample to the average temperature of 50.0±0.3 °C (chamber 1) demonstrate a certain dependence on heating extent. In particular, it is shown in Fig. 4 that at the waste sample temperature of 48 °C the concentration of mercury vapour in the air increases by a factor of 3.1 and decreases back to the initial level after cooling to 28 °C. Calculations of Spearman rank correlation coefficients confirmed a significant correlation between the concentration of aerosol particles within the size range of 0.2–0.5 μm and mercury vapour ($r_S = 0.7$).

The plots of mercury vapour concentrations in the air of chamber 2 at sample temperature 26–33 °C are shown in Fig. 5 as decaying oscillations. During waste sample heating within the temperature range from 28 to 33 °C, oscillations of mercury vapour concentrations attain the forced nature (see Fig. 5). The line of their trend is depicted as a fluctuating curve, with its minima 5.8 and 3.7 ng/m^3 appearing at 31 and 33 °C, respectively, and the maximum at 8.4 ng/m^3 – at 32 °C. At the same time, changes of mercury vapour concentrations at these temperature modes

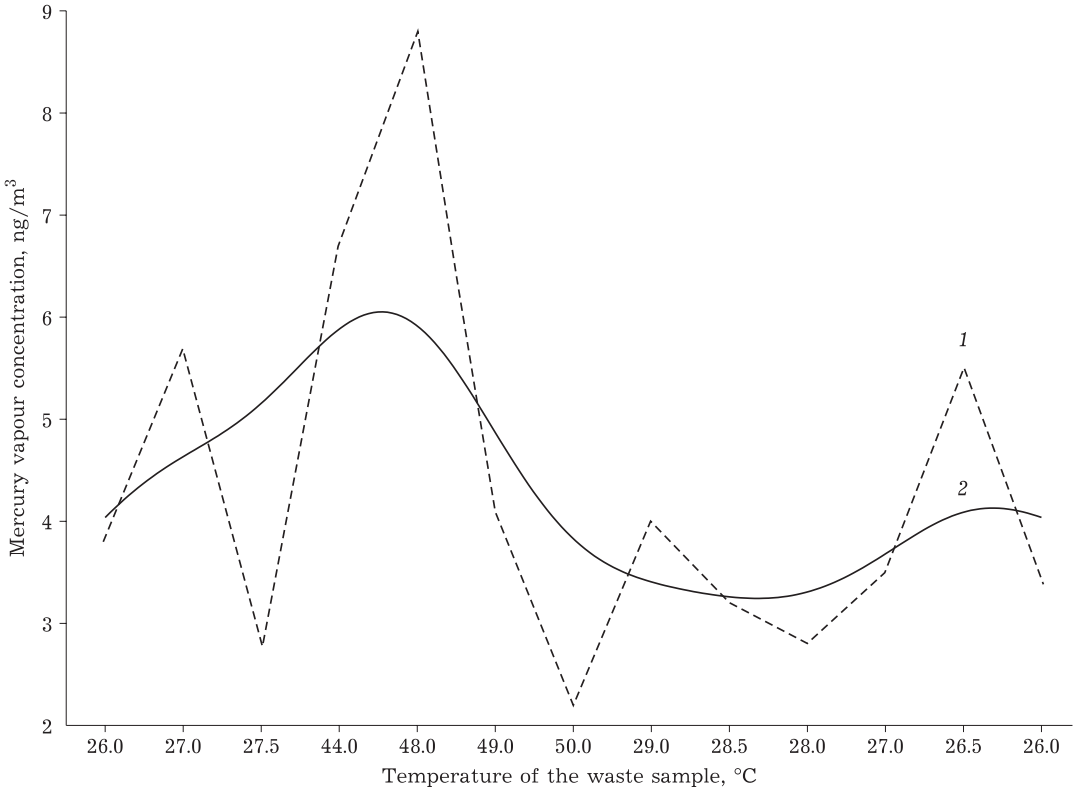

Fig. 4. Dependence of mercury vapour concentration in the air of chamber 1 on the temperature of the heated waste sample. For designations, see Fig. 1.

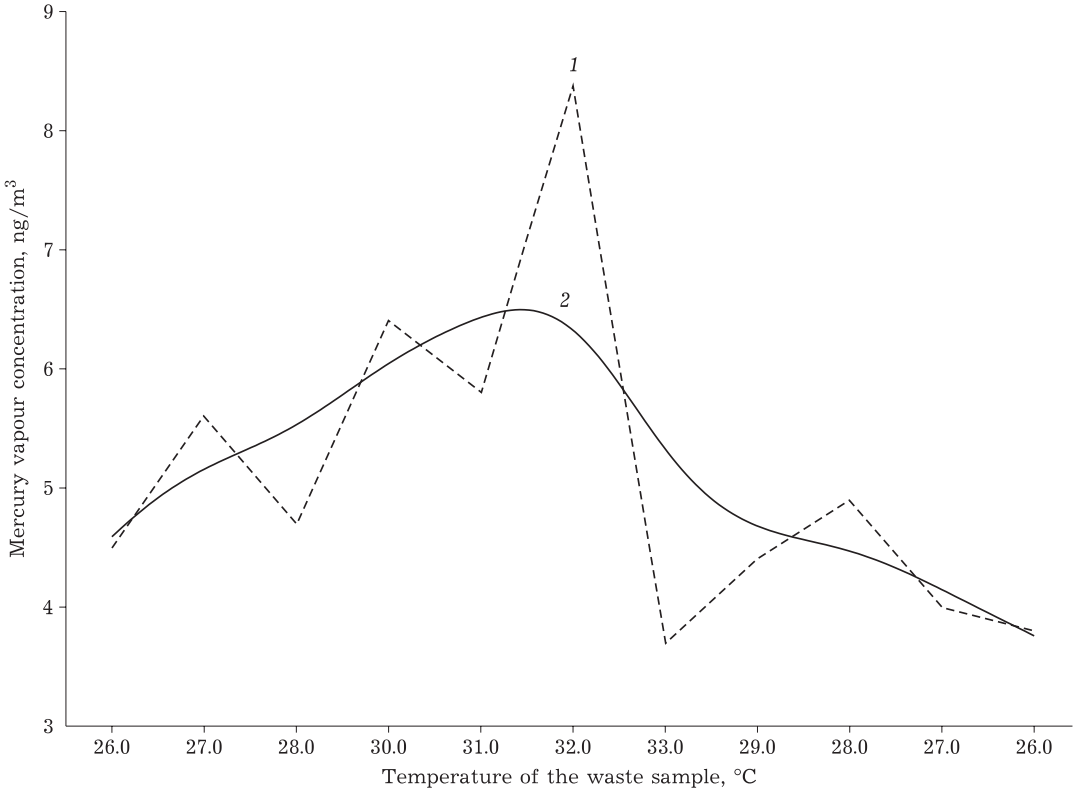

Fig. 5. Dependence of mercury vapour concentration in the air of chamber 2 on the temperature of the heated waste sample. For designations, see Fig. 1.

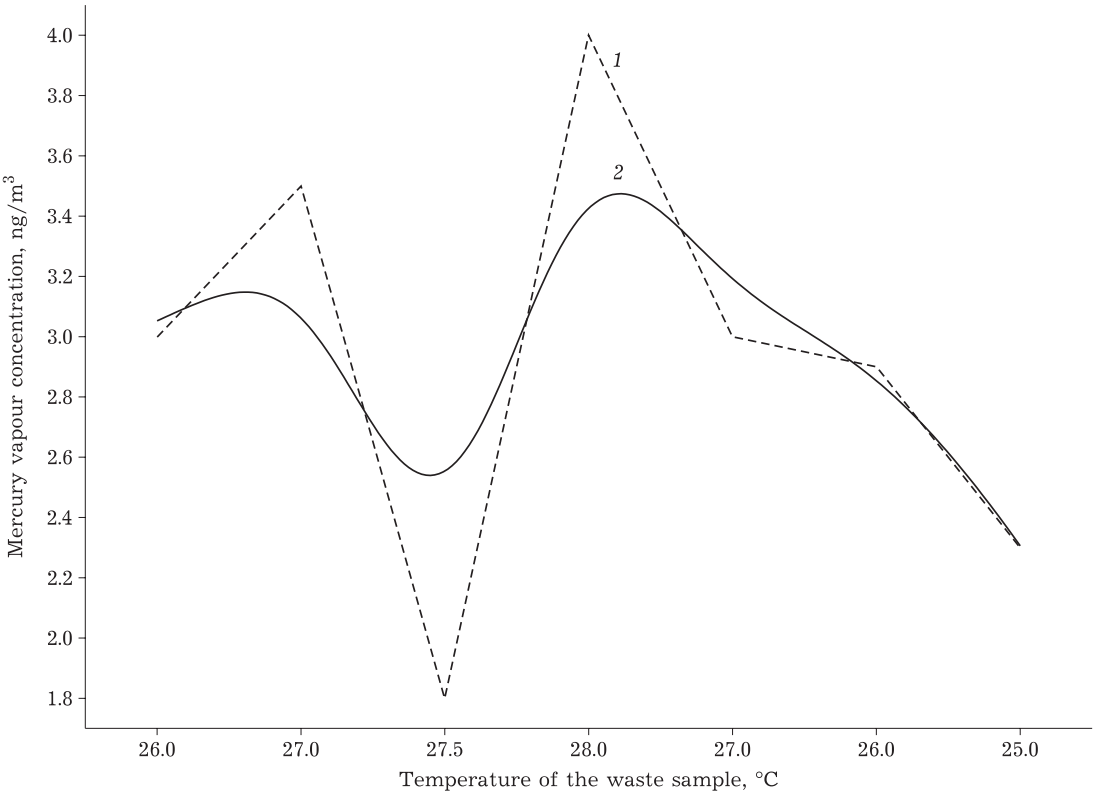

Fig. 6. Dependence of mercury vapour concentration in the air of chamber 3 on the temperature of the heated waste sample. For designations, see Fig. 1.

are not statistically significant. The Spearman coefficient between the concentrations of aerosol particles 0.2–0.5 μm in size and mercury vapour in chamber 2 (r_S = 0.7) was also statistically significant.

The distribution of mercury vapour concentrations in the air of chamber 3 without heating the waste sample (the mean temperature of the model sample was 28.2±0.1 °C) was characterised by a minimum, 1.8 ng/m^3 at 27.5 °C, and maxima equal to 4 ng/m^3 at 28 °C. The detected changes in mercury vapour concentrations are insignificant, and their graphical representation appears to be free oscillations (Fig. 6).

Summarising the results of field modelling, we may state that the concentration of submicron aerosol particles (0.2–0.5 μm fraction) reaches its maximum equal to 660 211 units/L, mercury vapour concentration equal to 8.8 ng/m^3 at the sample temperature of 44 and 48 °C, respectively, which identifies a pronounced dependence of the atmospheric emission of aerosol particles on the intensity of heating the surface of the modelling object. On the basis of identification of the above-described trend, free oscillations of submicron aerosol concentrations without external thermal action on the waste matter have been stated. The detected changes of mercury vapour concentration in the air under the thermal action on the waste samples did not have significant differences from initial levels, however, the forces pattern of its oscillations in the graphical representation implies a significant effect of the temperature factor.

From the hygienic viewpoint, intensification of gas-vapour emission of aerosol particles during heating the waste disposal object is essentially dangerous for the health of the population living within the pollution scattering halos [30–33]. Toxicological studies provide evidence that if the dose is expressed through mass, ultrafine inhalable insoluble particles are generally more toxic than larger ones with similar chemical composition [34–37]. At the same time, the assumption based on experimental results has been formulated: the key factor of the negative inhalation action of aerosol on the organism is the surface area of aerosol particles [31, 34, 35, 38]. The information available at present in insufficient to determine significant hygienic parameters characterising aerosol particles (the number of particles of definite size, their surface area, mass concentration). As a consequence, there is no list of relevant procedures for ecological and hygienic eval-

uation of the action of submicron aerosol on the organism. Theoretical analysis of the literature allows us to assume that the methodical basis for the programmes aimed at the studies of the effect of submicron aerosol on human organism and the standards of diagnostics of the revealed responses in potentially exposed groups of population will be formulated in the nearest future [39].

CONCLUSION

Thus, based on the results of the studies into the natural model of a sulphide-containing dump, it appears reasonable to predict substantial danger of the aerosol pollution of the surface atmospheric layer in the post-industrial phase of mining region development under intense solar radiation. The maximum of particle size distribution is within the submicron range, which constitutes the most substantial danger for the environment and for the health of population. Summarisation of the results of modelling local aerosol pollution of the atmosphere at the sulphide-containing dump in summer provided evidence of the predominance of submicron-sized particles, thus confirming the working hypothesis concerning the high risk of the polytropic influence of the formed aerosol on human organism. The major danger of the components of atmospheric emissions from sulphide-containing waste disposal facilities is in neurotoxic effects, which bring the prime threat to the health of the child population. In addition, the levels of exposure to the components of the formed atmogeochemical anomalies, studied previously in experiments with animals, were accompanied by the dose-dependent haematotoxicity. In combination with the signs of tension of the functional systems responsible for the development of adaptation processes, this can be the basis for the formation of specific pathological states in the exposed population of mining regions. The risk of propagation of the indicated pathologies increases under the action of submicron aerosol. In the case of hot weather, because of intense insolation of the sulphide-containing waste disposal facilities, exposure to submicron aerosol of complex composition can reach the toxic concentration range, thus increasing the risk of hazardous neurotoxic and haematotoxic effects. The limiting parameter index of harm becomes resorptive action, which should be taken into account in the forthcoming development of the measures aimed at environmental amelioration and prevention of environmentally-dependent morbidity of the population of mining regions.

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