

Environmental Assessment of Coal Ash Ponds of Thermal Power Plants in the South of the Russian Far East

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Abstract—The results of environmental assessment of ash ponds of thermal power plants in Vladivostok and Khabarovsk are given. High radioactivity of coal in the Russian Far East is responsible for the accumulation of radionuclides in the fly and bottom ashes, which leads to pollution of atmosphere, lithosphere, biosphere, and hydrosphere with toxic and radioactive elements. Runoff carries these elements to the Ussuri Bay of the Sea of Japan. Harmful effect of these wastes on human health is noted, and possible ways of their utilization have been recommended.

Keywords: Fly ash, bottom ash, ash pond, thermal power plant, radioactive elements, slurry.

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INTRODUCTION

Thermal power plants (TPPs) are integral parts of the urban structure of Vladivostok and Khabarovsk and are among the main environmental pollution sources. Therefore, particular attention should be given to ash and slag waste disposal with a view to ensure their environmental and social safety. This raises the problem of environmental monitoring, which is important for the Russian Far East. Availability of detailed and reliable data on the ecological situation could make it possible to not only reveal and estimate hazardous pollution levels but also determine trends in their variation and predict the variation rate. Taking into account the above stated, the present study was aimed at estimating the impact of TPP ash ponds on the ecosystem and exploring the dynamics of heavy metal accumulation in soils and vegetation in the TPP-affected zones (in Primorsky Krai and Khabarovsk Krai) to reduce their negative effect on the environment. The following tasks were formulated:

- (1) Examine pollutant distribution in snow cover;
- (2) Estimate the effect of TPP-2 (Vladivostok) and

TPP-3 (Khabarovsk) on the land cover and surface water and the degree of their pollution;

- (3) Develop actions to reduce the impact of TPPs on the environment and human health.

SUBJECTS AND METHODS

Methodologically, our study was based on Vernadsky's biosphere and noosphere theory [1] and principal provisions of the "Program and Procedure for Biogeocenological Studies" [2]. Integral and systemic approaches were applied. Reconnaissance and experimental studies were performed in the TPP-affected zones with the aid of modern instrumental and traditional physicochemical and chemical methods. At each sampling site a profile pit was excavated, and complete morphological soil profile description was made with sample withdrawal from each soil horizon. Mixed soil samples (1.5–2 kg) were annually withdrawn with a coring tube using the envelope technique, and their agrochemical parameters and the concentration of radionuclides and heavy metals therein were determined. Mixed vegetation samples were collected during the technical maturity phase (2 kg for crops,

legumes, and grasses and 5–8 kg for tilled crops, vegetables, and fruits. The gamma-ray background was measured with the aid of an SRP 68-01P instrument. Sampling sites in the TPP-3-affected zone were laid by 8 rhumbs with a radius of 1.5 km and by 10–12 rhumbs with a radius of 3 km according to [3–5]. The snow cover was analyzed according to Vasilenko [6].

Environmental samples were analyzed for agrochemical and radiochemical parameters and heavy metals. Samples were prepared according to the procedure described in [3]. The concentration of natural radionuclides (^{40}K , ^{232}Th , ^{226}Ra) was determined using a GAMMA-PLYuS spectrometer [4]. Radiological analyses were carried out according to methodical recommendations for comprehensive agrochemical analysis of farmland soils [5]. Sampling sites were defined with account taken of soil properties, climatic conditions, wind rose, typical crops, crop management practice, and their position relative to potential pollution sources.

Ash samples from the TPP-2 ash pond (120 samples by area and by depth) were studied by X-ray analysis and IR spectroscopy. During the sampling procedure, coal ash was transferred from one pond to another with excavation to a depth of 5–7 m, so that it was possible to obtain samples of deeper horizons. The coal ash is mainly a grey or dark grey fine soft free-flowing powder, sometimes including dense interlayers which crumbled to dust upon slight touch. The ash samples were subjected to semiquantitative spectral analysis. Their radioactivity was determined using an SRP-68-01 instrument. Infiltration water was also sampled near the TPP-2 ash pond, and the water samples were analyzed for Ca, Na, Al, Ba, Mg, Fe, Mn, V, Cr, Ni, Co, Ag, Cu, Pb, Zn, Cd, Li, Sr, Se, Ga, Hg, and B by atomic emission spectroscopy using a Plasmequant-110 spectrometer.

The obtained data were statistically processed with search for correlation relationships.

RESULTS AND DISCUSSION

Far East hard coal and coal wastes utilized in electricity generation in Primorsky Krai and Khabarovsk Krai are sources of ecosystems. All coal varieties contain radionuclides belonging to the uranium and thorium series. The average specific activity of natural radionuclides in Far East coals exceeds the world-average value by a factor of 1.3–2 [7–11]. Nonvolatile

Table 1. Concentration of radionuclides in coal and ash, Bq/kg [11, 12]

Isotope	Coal	Bottom ash	Fly ash
U^{238}	9–31	56–185	70–370
Ra^{226}	7–25	20–166	85–281
Th^{232}	9–19	59	81–174
K^{40}	26–130	230–962	233–740

components formed as a result of coal combustion reside in the ash which thus accumulates radionuclides (Table 1).

Operation of thermal power plants and municipal and industrial boilers in Vladivostok and Khabarovsk utilizes solid fuels (black and brown coals, peat, shale), which leads to accumulation of a considerable amount of wastes as fly ash and bottom ash disposed in ash ponds. Ash ponds are engineered structures whose construction and operation are subject to stringent requirements. The capacity of the existing ash ponds is periodically exhausted, and the problem of ash disposal becomes critical; this situation is now typical of TPP-2 and TPP-3 in Vladivostok and Khabarovsk. Inappropriate ash disposal creates risks of environmental pollution [9].

Ash is discharged with the aid of both freshwater and seawater (in TPP-2); infiltration water from the settling pond (Fig. 2) flows to the Promezhutochnaya Bay of the Sea of Japan. Pollution of surface and ground water, as well as of coastal seawater, as a result of operation of coal-fired power plants is the main factor responsible for environmental problems of water industry in Vladivostok. Coal ash constitutes more than 90% of total wastes produced by thermal power plants.

From the environmental viewpoint, the use of gas fuel is more desirable; however, gas transport is very expensive, so that application of most advanced technologies for solid fuel combustion remains virtually the only and most appropriate way under insufficient funding [13].

Unfortunately, the problem of closed ash pond reclamation remains unsolved. There are no waste treatment facilities at power plants in Vladivostok (e.g., at *Energo* joint stock company). An exception is *Dal'energo* public corporation which possesses an RIFEI-4 processing line for the manufacture of wall blocks from slag concrete. Its efficiency amounts to ca. 416 m³ per annum, which is equivalent to 1200–1900 t of ash, i.e., a minor part of waste ash [14].



Fig. 1. Coal ash pond: (a) section 1, (b) section 2.

It should be noted that urban sprawl often leads to inclusion of ash ponds into city boundaries. Attempts to relocate or terminate them are not always successful. For example, old ash pond in the center of Vladivostok was transformed into a parking, which improved the appearance of this territory but did not solve the environmental problem. Lands occupied by ash and slag wastes are irreparably withdrawn from beneficial land use. Accidental embankment dam breaches are accompanied by discharge of large volumes of waste ash and highly mineralized water from settling ponds.

Ash ponds at TPP-2 include three sections with areas of 21, 36, and 47 ha (Fig. 1a, b), filled with 3.704, 6.045, and 5.623 million tons, respectively, and two sections under design (36.5 and 41 ha). TPP-2 and TPP-3 utilize black and brown coals from 17 coal deposits (Raichikhinskoe, Rettikhovskoe, Pavlovskoe, etc.). The chemical composition of fly and bottom ash obtained after coal combustion is as follows, %: SiO_2 , 10–58; Al_2O_3 , 10–30; Fe_2O_3 , 2–20; CaO , 2–60; MgO , 0–10; R_2O , 0–5 (here, R_2O is the sum of TiO_2 , FeO ,

MnO , and K_2O) [14, 15]. In addition, minor impurities of various elements are present. The major mineral components of coals are clay minerals containing small amounts of iron oxides, pyrite, and siderite. These components undergo thermal transformations during the combustion process. Most of them melt and then solidify as glassy particles. Sulfur partly reacts with calcium to give CaSO_4 . A considerable amount of carbon (unburned carbon) was detected in fly and bottom ash. Provided that the coal contains minor elements (Mn, As, Hg, Pb, Sb, Cd, etc.), their concentration in the coal ash is significantly higher than the background values, which creates increased risk to the environment.

Determination of the specific activity of coals from the Neryungrinskoe, Chegdomynskoe, Kharanorskoe, Raichikhinskoe, Urgal'skoe, and Luchegorskoe deposits, utilized in TPP-3, gave the following results, Bq/kg: ^{40}K , 24 to 171; ^{232}Th , 4.39 to 46.25; ^{226}Ra , 7.85–55.75. The specific activity of the fly ash was estimated at, Bq/kg: ^{40}K , 75–427.2; ^{232}Th , 51.24 to 190; ^{226}Ra , 65.8 to 153.3. The specific activity of the bottom ash was, Bq/kg: ^{40}K , 89.76 to 510.1; ^{232}Th , 15.46 to 125.1; ^{226}Ra , 81.55 to 165.3. The concentration of ^{40}K and ^{226}Ra in the fly ash is higher than in the coal by a factor of 2 to 8, and of ^{232}Th , by a factor of 3–8; the corresponding values for the bottom ash are: ^{40}K , 2 to 7; ^{232}Th , 3–9; ^{226}Ra , 3–8. It should be noted that the coal from the Neryungrinskoe deposit is enriched in ^{232}Th , and from the Chegdomynskoe deposit, in ^{40}K and ^{226}Ra . After coal combustion, these elements accumulate in the fly and bottom ashes.

No toxic chemical elements were detected by spectral methods in the bottom ash samples from TPP-2 ash ponds, where the bottom ash constitutes no more



Fig. 2. Settling pond of TPP-2.

than 5 wt % of the total wastes. Spectral analysis of the fly ash revealed the following elements, %: Sn and Rb, 0.0001–0.0004; Zn, 0.003–0.005; Be, 0.0001–0.0005; Cu, 0.0007–0.005; Ge, 0.0001–0.0002; Co – 0.0005–0.6; Ni, 0.0003–0.008; Zr, 0.002–0.008. The results obtained for different samples were fairly similar despite the use of coals from different deposits. The concentration of As, Cd, Hg, Sb, Bi, Au, Pt, and In in the examined samples was lower than the detection limit (0.01). Our data were in agreement with those published in the literature and Clarke values [16], and for some elements were lower even by an order of magnitude. Therefore, the fly ash from TPP-2 may be regarded as environmentally safe.

The radioactivity of the fly ash from TPP-2 was estimated at 17–21 $\mu\text{R/h}$. According to [17], an activity of 4–8 $\mu\text{R/h}$ is believed to be low, 9–20 $\mu\text{R/h}$, normal, and 21–27 $\mu\text{R/h}$, increased. Therefore, our results correspond to normal or slightly increased radioactivity level. The radioactivity of fly ash samples was measured in summer under fairly moist conditions which are known to favor reduced activity values. According to the data of the *Primorgeologiya* Production Geophysical Association Ecological Center, the activity of the fly ash from TPP-2 may reach 40 $\mu\text{R/h}$ [18].

Radiation monitoring of the TPP-2 coal ash pond, performed by Molev [17, 18], showed variation of the gamma-ray radiation from 24 to 30 $\mu\text{R/h}$, which exceeds the background value by a factor of 2 to 3. The radioactivity of the ash pond in drought seasons increases to 30–33 $\mu\text{R/h}$ and decreases to 18–25 $\mu\text{R/h}$ in rainy and soil thawing periods. Fine fraction of the fly ash is more active than the coarse fraction approximately by 10%. Spectrometric analysis revealed reduced concentration of potassium and increased concentration of uranium and thorium in the fly ash; this suggests uranium and thorium origin of its radioactivity. The concentration of potassium and uranium in the bottom is somewhat lower than in the upper layers of the ash pond. The opposite relation was observed for thorium whose concentration increased from 19×10^{-4} to $28 \times 10^{-4}\%$ downright. Presumably, this is determined by migration and removal of K and U and accumulation of less mobile Th in the bottom layers.

The infiltration water from TPP-2 had a pH value of 6.7 to 9.3. As follows from the data in Table 2, water samples no. 9 (clarified pond) and no. 10 (brook flowing into the Promezhutochnaya Bay) are charac-

terized by similar compositions. The concentrations of Ag, Cd, Cu, Ga, Hg, Li, Ni, Pb, Se, V, and Zn in sample no. 11 (Promezhutochnaya Bay) approached those in nos. 9 and 10, while the concentrations of other elements were lower. The amounts of Ag, Ba, Cd, Co, Cr, Cu, Fe, and Ni in these samples did not exceed the corresponding maximum allowed concentrations (MAC) for seawater [19, 20]. The concentrations of Al, B, Li, Mn, Pb, and Se exceeded MACs by an order of magnitude, and of Sr and V, by two orders of magnitude. Published data are available only for Fe, Mn, Cd, and Cu [21]. According to our results, the concentration of Fe and Mn is higher by an order of magnitude, and the data for Cd and Cu differ insignificantly. The concentrations of Cu, Zn, Cd, Pb, Ni, and Zn determined by us in seawater from the Promezhutochnaya Bay differ only slightly from the data for Ussuri Bay (water was sampled at a distance of up to 15 km from the coast). These findings indicate accumulation of chemical elements in the seawater over a long period of TPP-2 operation. Unfortunately, there were no reference data to compare with our data for rare elements such as Ga, Sr, Hg, Se, and V.

We can conclude that the water used to carry ash away from the power plant should be settled down for a long time and treated before being discharged into the sea, for the concentrations of heavy metals therein approach those typical of hydrothermal solutions. Infiltration water transfers a considerable amount of the above listed elements which accumulate in seawater, sediments, flora, and fauna and then enter into human organism by trophic chains. Furthermore, infiltration water from the ash pond trickles down into ground water thus creating risks of pollution of the underground hydrosphere, spring and drinking water.

We did not analyze the infiltration water from TPP-2 for radioactivity; the relevant data were given in [17, 18]. Mobile uranium is transported by infiltration water which is characterized by considerable radioactivity (7–17 $\mu\text{R/h}$). The concentrations of uranium and thorium in the brook mouth (Promezhutochnaya Bay) exceed the background values by 20–30%, indicating accumulation of these elements in seawater. The background radiation in the Promezhutochnaya Bay amounts to 15 $\mu\text{R/h}$.

We have revealed a relation in the coal/fly ash and bottom ash/snow/soil/vegetation distributions of natural radionuclides in the TPP-3-affected zone (Khabarovsk), namely the concentration of radionuclides decreases as the distance from the pollution source

Table 2. Concentration of elements in hydrochemical samples, mg/L

Element	MAC ^a	Sample no.		
		9	10	11
Ag	0.0059	<DL ^b	<DL ^b	<DL ^b
Al	0.040	0.238	0.248	0.145
C	0.10	4.460	4.540	3.790
Ca	0.740	0.259	0.284	0.077
Cd	0.005	<DL ^b	0.001	<DL
Co	0.010	0.028	0.035	0.014
Cr	0.02	0.013	0.018	<DL
Cu	0.001	<DL ^b	<DL ^b	<DL ^b
Fe	0.100	0.122	0.123	0.084
Ga	– ^c	<DL ^b	<DL	<DL ^b
Hg	– ^c	<DL ^b	<DL ^b	<DL ^b
Li	0.08	0.266	0.266	0.279
Mn	0.01	0.217	0.302	0.045
Ni	0.01	0.012	0.024	0.022
Pb	0.006	<DL ^b	0.016	<DL ^b
Se	0.002	<DL ^b	<DL ^b	<DL ^b
Sr	0.400	11.14	12.75	7.760
V	0.001	0.239	0.268	0.230
Zn	0.010	<DL ^b	<DL ^b	<DL ^b

^a Maximum allowable concentration. ^b DL stands for detection limit. ^c No data available.

increases. The specific activity of snow sampled at a distance of 1.5 to 3 km from TPP-3 changes as follows, Bq/L: ⁴⁰K, from 45.29 to 22.48–28 and 35–22.23, respectively; ²²⁶Ra, to 9.08–4.04 and 7.75–4.97; ²³²Th, to 7.78–3.37 and 4.38–3.27. The concentration of heavy metals was (mg/L): for Cd 0.003 (1.5 km) and 0.002–0.0006 (3 km) and for for Pd 0.027–0.006 (1.5 km) and 0.018–0.003 (3 km). The areas located 1.5 km northeast and 3 km southwest of TPP-3 were characterized by the highest heavy metal load of snow cover. This means that the snow cover accumulates airborne pollutants originating from coal combustion and wind transport of ash.

Soils also accumulate anthropogenic pollutants. The results of our radioecological studies showed that contamination of soil cover with radionuclides (⁹⁰Sr, ⁴⁰K, ²³²Th, ²²⁶Ra) exceeds average values for Russia (Fig. 3). The concentrations of Cu, Pb, Hg, Zn, Cd, Ni,

and other heavy metals do not exceed the natural background values and approach MACs.

The ⁹⁰Sr radioactivity of vegetation in the south of Khabarovsk Krai ranges from 13.4 Bq/kg in soya straw to 1.35 Bq/kg in pears, which is considerably higher than in Omsk oblast [22]. The buildup factor for ⁹⁰Sr changes from 1.64 in perennial grasses to 0.09 in wheat corns. Thus, ⁹⁰Sr accumulates mainly in vegetative parts of oat and soya, while reproductive organs of plants contain minor amounts of that isotope.

Vladivostok and other cities in Primorsky Krai (Artem, Spassk, Dal'negorsk, Ussuriisk, Fokino, and Nakhodka) constitute a critical group by environmental stress parameters [23]. Khabarovsk may also be included in that group.

Analysis of biological samples (hair of children in Luchegorsk) performed in the present study revealed considerable levels of toxic elements as compared to other regions in Russia. The concentration of lead in the hair of children in Luchegorsk was 1.4 to 2.7 times higher than in Central Chernozem, Non-chernozem area, and Crimea, and the concentrations of zinc, copper, and cadmium were, respectively, 1.4 to 1.5, 1.2 to 1.7, and 1.8 to 2.2 times higher (Table 3).

According to the data of other authors [24], biological samples (hair of children in Vladivostok) also contained higher amounts of toxic elements as compared to the above listed regions of Russia. The concentrations of lead, zinc, and cadmium in the hair of children in Vladivostok were higher by factors of 1.2 to 2.5, 1.1 to 1.3, and 1.2 to 2.3, respectively (Table 3).

It should be noted that Luchegorsk is located near coal pits and that Vladivostok is exposed to the effect of coal ash ponds; however, the exceedance values for the above elements in biological samples (hair of children) in these cities are fairly similar and considerably higher than those found for central regions of Russia.

The results of our studies and published data showed a strong anthropogenic impact of TPP operation and waste ash on the environment. The population of the affected areas suffers from a broad spectrum of diseases, including bronchial asthma, allergic bronchitis, and allergic rhinitis. In most cases, these diseases are induced or aggravated by inhalation of dust, specifically of fly ash from TPPs. Finely dispersed ash dumped in huge amounts at TPP-2 and TPP-3 is spread over large areas due to strong winds typical of Primorskii Krai and Outer Manchuria

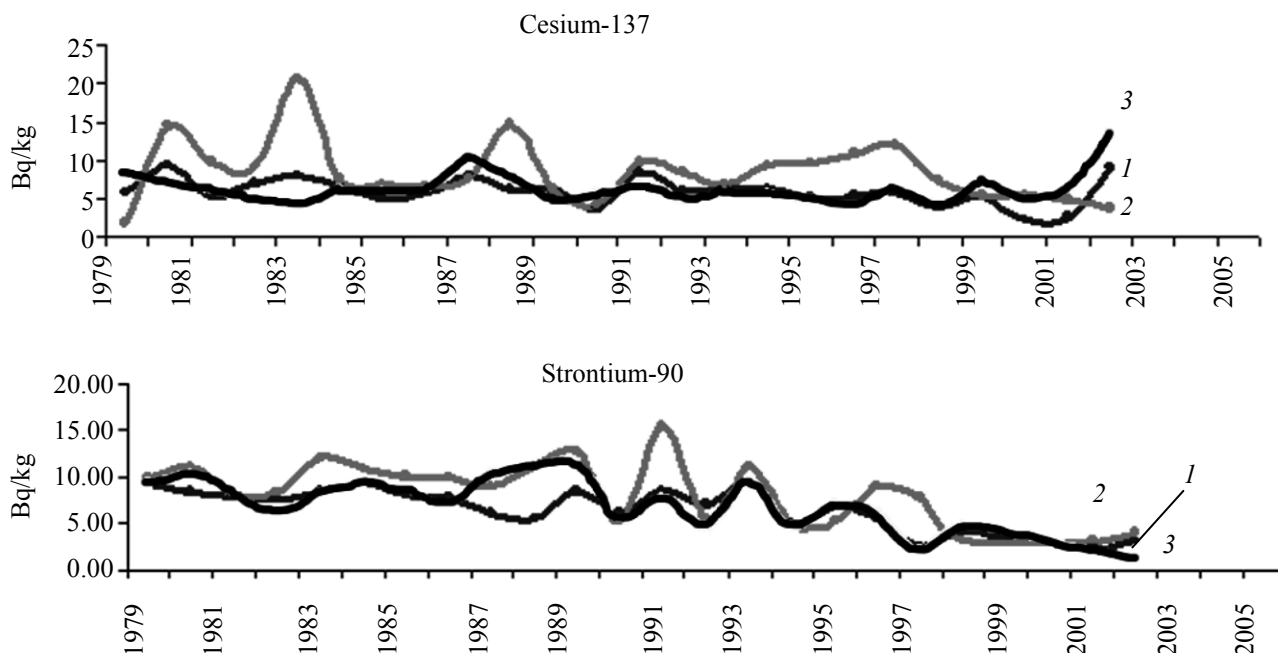


Fig. 3. Specific activity of radionuclides in different soils: (1) brown forest soil, (2) meadow brown soil, and (3) meadow gley soil.

climate (including Vladivostok and Khabarovsk), which favors propagation of the above diseases. Therefore, coal ash ponds should be relocated far beyond city boundaries. Furthermore, it is undesirable to accumulate large amounts of ash; instead, their utilization in different branches of industry is advisable.

In the winter time, when most precipitation falls as snow, coal ash from TPPs is often used in large amounts by road services for deicing/anti-icing treatment of roads and footpaths [25]. The ash is carried by wind thus contributing to pollution of the environment and adverse effect on human health and biota. Being an open system, respiratory organs constitute the first line of protection of organisms from harmful toxic, allergenic, infectious, and physical factors, including finely dispersed fly ash. Under these

conditions, children breathe harder and get sick more frequently and seriously than do adults. Taking into account the impact of fly and bottom ashes on human health, their use by road services should be prohibited.

Depending on the chemical composition, waste ash may be utilized as raw material for the manufacture of, e.g., clay bricks, but the fraction of ash in construction materials should not exceed [18]. Up to now, many procedures have been developed for utilization of coal ash in building construction, agriculture, chemical industry, metallurgy, and other branches of industry, and relevant data have been well documented in scientific literature. We believe that the scope of waste ash utilization should be more rational.

Although the coal ash from TPP-2 contains minor amounts of toxic elements, the ash/water slurry

Table 3. Concentrations of some toxic elements in hair of children in Luchegorsk (Primorskii Krai) and other regions of the UIS [24], $\mu\text{g/g}$

Region	Lead	Zinc	Copper	Cadmium	Chromium
Luchegorsk	9.81	201.34	17.23	0.67	3.22
Vladivostok	8.80	164.70	9.20	0.44	—
Non-chernozem	3.58	132.80	11.00	0.19	—
Central Chernozem	7.24	131.50	10.10	0.38	—
Crimea	4.80	147.00	14.10	0.30	—

remains enriched with these elements, including radioactive ones, even after settling. Therefore, there exists a risk of pollution of Ussuri Bay and other water basins.

Thus the thermal power plants in Vladivostok and Khabarovsk are potent sources of pollution of the atmosphere, biosphere, lithosphere, and hydrosphere with heavy metals, radioactive elements, and fly ash. Uptake of toxic elements via food chains causes a broad spectrum of serious diseases in humans. In order to reduce the impact of ash ponds on the environment and human health it is necessary to initiate ash pond reclamation and environmental monitoring.

CONCLUSIONS

The average radioactivity of natural radionuclides in the Far East coals exceeds the world average values by a factor of 1.3 to 2. The most hazardous consequences of coal combustion in the Primorskaya and Khabarovskaya thermal power plants are release of natural radionuclides from coal, discharge of fly ash containing toxic microelements and carcinogenic substances, and accumulation of huge amounts of bottom and fly ashes containing radioactive radium, thorium, and potassium isotopes.

It is necessary to maintain coal ash ponds in strict agreement with the corresponding regulations. Natural water basins and streams should never be used to dispose polluted wastewater. Pollution of natural surface and ground waters and coastal seawater as a result of operation of coal-burning power plants is among the main factors responsible for environmental problems of the water utilization system. The lack of data on the volumes of infiltration water from TPP-2 and TPP-3 is inadmissible. This parameter is equally important in addition to the pollutant concentration in infiltration water since accumulation of toxic elements is directly proportional to the discharged water volume.

It is very urgent to construct settling tanks for repeated sedimentation and purification of infiltration water from coal ash ponds. It is desirable to recycle infiltration water.

To ensure environmental safety in the TPP-affected areas it is necessary to perform timely reclamation of coal ash pond areas and organize environmental monitoring.

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