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PREDICTION OF TRANSBOUNDARY POLLUTION OF AIR AND SOIL IN ODESA REGION BY FLY ASH FROM MOLDAVIAN SDPP

О. Бутенко, С. Мельник, А. Карамушко, К. Васютинська, В. Лєсніков, В. Жерденко. **Прогнозування транскордонного забруднення атмосферного повітря і ґрунтів Одеської області летючою золою Молдавської ДРЕС.** Оскільки у даний час через брак газу велике енергетичне підприємство Молдавська ДРЕС повернулося до використання твердого палива (кам'яного вугілля), проблема забруднення летючою золою території Одеської області стала актуальною. У статті запропонувано метод розрахунку розсіювання по поверхні ґрунту твердих частинок за формою близькою до сферичної на прикладі транскордонного забруднення Одеської області летючою золою енергетичних котлів Молдавської ДРЕС. На відміну від діючого нормативу ОНД-86, метод використовує загальні закони аеродинаміки і полягає у визначення швидкості осідання твердих частинок. Показано, що хоч летюча зола пиловугільних котлів і складається з великої кількості різних речовин, основну її масу складають діоксид кремнію, оксиди алюмінію, заліза та кальцію. Вони відрізняються за фракційним складом і густиною матеріалу. Запропонований метод враховує цю відмінність та дозволяє прогнозувати координати випадіння частинок летючої золи того чи іншого матеріалу та певного розміру на ґрунт. Розрахунки засвідчили, що хімічний склад забруднення ґрунту змінюється вздовж лінії димового факелу – у північній частині Одеського району переважають оксиди заліза, а у безпосередній близькості та у місті Одеса – діоксид кремнію. Через брак достовірних даних про фракційний склад летючої золи Молдавської ДРЕС можливою виявилася тільки приблизна оцінка розсіювання. Але навіть такі оціночні розрахунки дозволили зробити висновок про те, що золові частинки поширюються значно глибше у територію Одеської області ніж за результатами ОНД-86. Це є підставою для перегляду цієї методики у частині розрахунку розсіювання летючої золи теплових електростанцій.
Ключові слова: летюча зола, транскордонне забруднення, забруднення ґрунту, фракційний склад

O. Butenko, S. Melnyk, A. Karamushko, K. Vasiutynska, V. Lesnikov, V. Zherdenko. **Prediction of transboundary pollution of air and soil in Odesa region by fly ash from Moldavian SDPP.** Since at present, due to the lack of gas, a large energy enterprise Moldavian SDPP has returned to the use of solid fuel (coal), the problem of fly ash pollution of the territory of Odesa region has become relevant again. The article proposes a method for calculating the dispersion of solid particles with a shape close to spherical on the soil surface on the example of transboundary pollution of Odesa region by fly ash from power boilers of the Moldavian SDPP. In contrast to the current UND-86 standard, the method uses the general laws of aerodynamics and is based on determining the settling velocity of solid particles. It has been shown that, although fly ash from pulverised coal boilers consists of a large number of different substances, the bulk of it is silica, aluminium, iron and calcium oxides. They differ in fractional composition and material density. The proposed method takes this difference into account and allows predicting the coordinates of fly ash particles of a particular material and a certain size falling on the soil. The calculations showed that the chemical composition of soil pollution varies along the smoke plume line, with iron oxides predominating in the northern part of Odesa district and silicon dioxide in the immediate vicinity and in Odesa city. Due to the lack of reliable data on the fractional composition of fly ash from Moldavian SDPP, only an approximate estimate of dispersion was possible. However, even such estimates allowed us to conclude that ash particles spread much deeper into the territory of Odesa Oblast than according to the results of the UND-86. This is the basis for revising this methodology in terms of calculating the dispersion of fly ash from thermal power plants.
Keywords: fly ash, transboundary pollution, soil pollution, fractional composition

Introduction

According to the World Coal Association, coal combustion in power plants accounts for approximately 37% of global electricity consumption, and coal ash is the main waste product of solid fuel combustion in the energy sector. A certain portion of coal ash, called fly ash or fly ash, is emitted into the atmosphere along with flue gases. Despite modern and fairly sophisticated flue gas cleaning systems, the total amount of ash that pollutes the air and soil is very significant. About 32% of Ukraine's land is contaminated by TPP emissions.

Before the start of large-scale Russian aggression, about 40% of Ukraine's electricity was generated by thermal power plants. There was not and is not a single coal-fired thermal power plant in Odesa

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region. However, Odesa region, in particular Rozdilnyanskyi, Bilhorod-Dnistrovskyi, and Odesa districts, was subject to air and soil pollution from pulverized coal boilers of the Moldavian SDPP, which is located on the border of Moldova (unrecognized Transnistria) and Odesa region.

The power plant has 8 pulverized-coal units of 200 MW each, of which 5 are in operation. They can burn more than 350 tons of fuel equivalent every hour. With an ash content of more than 20% and a declared flue gas cleaning efficiency of 97%, more than 4 tons of pollution can be emitted into the atmosphere every hour. This defines the problem of transboundary pollution of Odesa region with fly ash.

The Soviet methodology UND-86 is still used in Ukraine to determine air pollution from industrial enterprises, including thermal power plants. It is designed to calculate ground-level concentrations of pollutants at a distance of up to 100 km from the source. One of the drawbacks of the methodology is that it does not actually take into account the properties of the dust mixture, which is the fly ash from pulverized coal boilers of thermal power plants, namely its fractional composition and physical properties of each element. Meanwhile, these factors, together with meteorological conditions, are decisive for the dispersion of fly ash in the environment. Therefore, the task of determining the peculiarities of the distribution of solid particles with a size of 1 to 100 microns of various physical properties in the atmospheric air is relevant.

Analysis of publications on the topic of research

Solid microscopic particles can penetrate the deepest structures of living organisms and cause a wide range of acute and chronic diseases. Fine dust, which includes fly ash, includes particles of various sizes that may contain chemically active, toxic, and carcinogenic substances, which causes serious respiratory and cardiovascular diseases in humans, causes allergic reactions, and increases carcinogenic risks [1 – 4]. According to the WHO, particulate matter is the most dangerous pollutant because, among other harms, it can carry chemicals and bacteria [5, 6]. For example, study [7] assessed the potential concentrations of toxic heavy metals and the associated environmental and health risks for residents of the surrounding areas near the Bokaro thermal power plant, Jharkhand, India. It was noted that fly ash containing heavy metals is emitted from the thermal power plant and contaminates the soil of nearby agricultural land, roadside areas and residential areas. This causes an increase in environmental risk, as well as carcinogenic effects on human health. Residents are exposed to heavy metals in a variety of ways – through direct ingestion, dermal contact, and inhalation. The study concluded that the adult population is at high risk of non-carcinogenic health effects due to exceeding the permissible limits for Cd, Pb and Cr in vegetables and cereals. The target carcinogenic risk for Pb and Cr was also high, indicating the existence of a cancer risk in the adult population.

Numerous studies clearly indicate that the fly ash released into the environment during coal combustion consists of SiO_2, Al_2O_3, Fe_2O_3, CaO, MgO, Na_2O, K_2O, TiO_2, P_2O_5 and a small amount of MnO, BaO and SO_3 [8, 9]. Fly ash may also contain a small number of heavy metals. In general, the chemical and fractional composition depends on the place of coal extraction, combustion method and type of dust preparation.

Fly ash collected at thermal power plants can be useful in the construction industry as a raw material for Portland cement clinker [10] and white sulfoaluminate clinkers [11], as a pozzolanic additive for cement and concrete, bricks [12], glass ceramics [13], etc. Thanks to new green transformation technologies, the scope of ash use has expanded significantly. For example, it can be used as a source of carbon for the synthesis of nanomaterials (e.g., carbon nanotubes, which are promising sorbents for hydrogen [14]), aerogels, geopolymers, and rare earth elements [15]. Fly ash has demonstrated good qualities for geotechnical structures, such as in paths, embankments, etc. [16].

Combined with organic fertilizers, fly ash can be used to improve degraded or marginal soils [17, 18]. The combined effect of fly ash and organic matter improves soil structure, increases nutrient concentration, mass moisture, porosity, and fine-grained mineral content, and improves biological activity in the soil [19].

However, fly ash that has passed through flue gas cleaning devices and entered the atmosphere through the chimney of a thermal power plant is definitely a pollutant that negatively affects air and soil quality.

The purpose and objectives of the study

The aim of the study is to develop a simple and reasonable method for calculating the distribution of TPP fly ash particles, taking into account its chemical and fractional composition, as well as to assess the nature of air and soil pollution in Odesa region by fly ash from Moldavian SDPP.

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Research results

The decisive role in the dispersion of fly ash is played by the value of the wind speed u_{air} and the deposition rate of solid particles u_{dep}. Since fly ash mainly consists of spherical solid particles ranging in size from 0.1 μm to 100 μm [20] (Fig. 1), the general laws of aerodynamics can be used to calculate u_{dep}.

According to these laws, a balance of forces can be compiled for particles (Fig. 2), including the Archimedean force F_{arch}, the force of gravity F_{grav}, and the force of drag F_x [22].

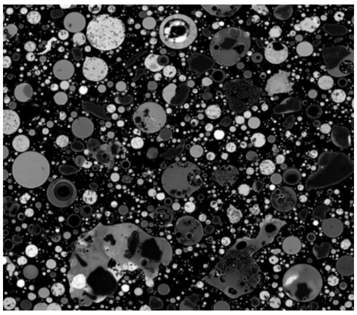

Fig. 1. Fly ash from coal combustion [21]

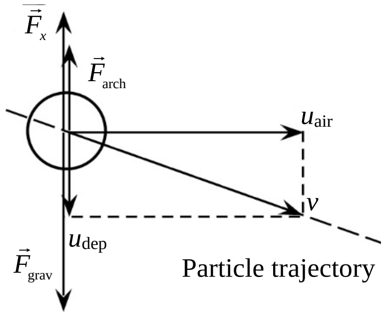

Fig. 2. Balance of forces for an ash particle

$$F_{arch} = \rho_{air} g V = \rho_{air} g \frac{\pi d^3}{6}, \tag{1}$$

$$F_{grav} = mg = \rho_{par} g \frac{\pi d^3}{6}, \tag{2}$$

$$F_x = C_x A \frac{\rho_{air} u_{dep}^2}{2} = C_x \frac{\pi d^2}{4} \frac{\rho_{air} u_{dep}^2}{2}, \tag{3}$$

where: ρ_{air} and ρ_{par} are the densities of air and ash particle material, kg/m^3;

g is the acceleration of free fall, m/s^2;

d is the diameter of the ash particle, m;

C_x – coefficient of drag.

The balance of these forces:

$$F_x = F_{grav} - F_{arch}. \tag{4}$$

Where the deposition rate:

$$u_{dep} = \frac{1.155}{\sqrt{C_x}} \sqrt{gd \frac{(\rho_{par} - \rho_{air})}{\rho_{air}}}. \tag{5}$$

In general, the drag coefficient is a complex function of the Reynolds number and the shape of the particle, a graph of the dependence is presented, for example, in [22]. Reynolds number for the problem under consideration:

$$\mathrm{Re} = \frac{d u_{dep}}{\nu_{air}}, \tag{6}$$

where ν_{air} the kinematic viscosity of air, m^2/s.

Considering that to determine u_{dep}, it is necessary to first determine the drag coefficient, which in turn depends on u_{dep}, the problem of calculating the deposition rate should be solved by the method of successive approximations. This method involves performing several calculation cycles. At the beginning of the first cycle, the deposition rate is arbitrarily set in the first approximation u_{dep1}, after which Re_1, C_{x1} and from (5) u_{dep2} are determined. If with the accepted accuracy $u_{dep1} \neq u_{dep2}$ then the calculations are repeated $u_{dep2} \rightarrow \mathrm{Re}_2 \rightarrow C_{x1} \rightarrow u_{dep3}$ and so on until the condition $u_{dep(n-1)} = u_{dep(n)}$ is fulfilled.

Such calculations were performed for the four main components of fly ash – silica, aluminum, iron, and calcium oxides. Their density and expected fractional composition are presented in Table 1.

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Table 1

Density and fractional composition of the main elements of fly ash

Particle size, μm	SiO_2, %. (ρ_{par} = 2650 kg/m^3)	Al_2O_3, %. (ρ_{par} = 3970 kg/m^3)	Fe_2O_3, %. (ρ_{par} = 5240 kg/m^3)	CaO, %. (ρ_{par} = 3340 kg/m^3)
< 5	35…50	30…45	25…40	20…30
5…10	20…30	20…30	15…25	15…25
10…30	10…20	15…25	20…30	20…30
30…50	5…10	5…10	10…15	15…20
50…80	3…5	3…7	5…10	10…15
80…120	< 3	< 5	< 5	5…10
> 120	0	0	< 2	< 5

The dependence of the deposition rate on the size of the ash particle is shown in Figure 3.

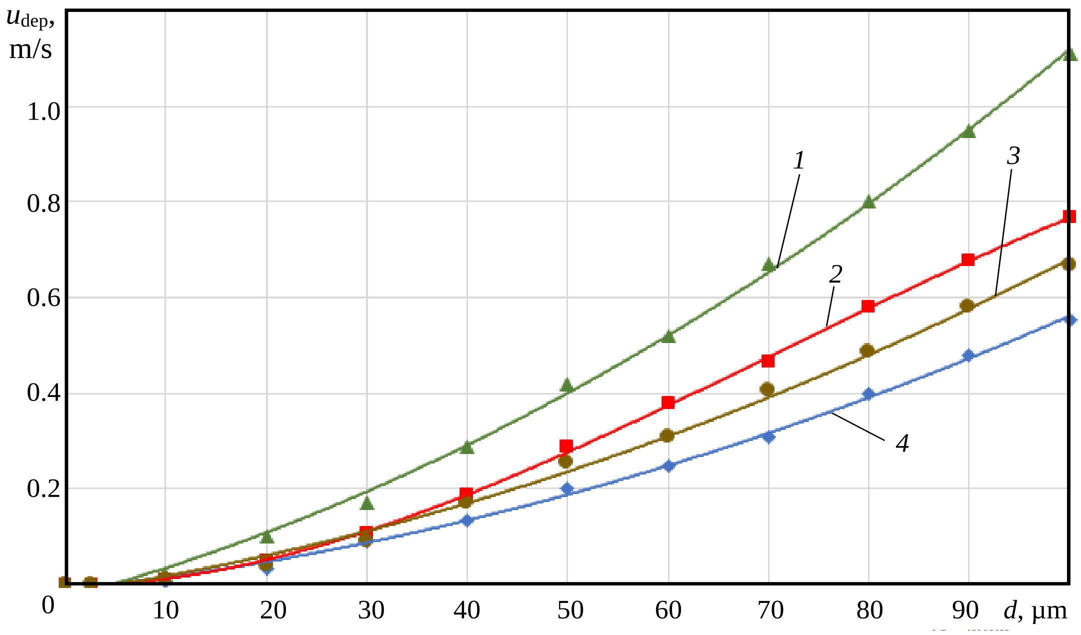

Fig. 3. Dependence of the deposition rate on the size of the ash particle: *1* – Fe_2O_3, *2* – Al_2O_3, *3* – CaO, *4* – SiO_2

Given that the ash particle is in motion for a while:

$$t = \frac{H}{u_{dep}}, \quad (7)$$

the distance from the chimney at which it will fall to the ground:

$$L = u_{air}\frac{H}{u_{dep}}, \quad (8)$$

where H is the height of the chimney, m.

As an example, Figure 4 shows graphs of the dependence of L on d for silica at a chimney height of H = 180 m (Moldavian SDPP) at different wind speeds.

The graph shows that the smallest ash particles of the lightest component (silica), even at low wind speeds, will cover a distance of 60 km from Moldavska TPP to Odesa. If we have reliable data on the fractional composition of the fly ash from this TPP after cleaning in wet scrubbers, then the combined use of the relationship $L = f(d)$ and the differential distribution curve of particulate matter $N = f(d)$ would allow us to determine the mass distribution of a particular component along the axis of the smoke plume. Since there is no data on the fractional composition and the management of the DRES is not interested in providing such information, we can only speak of a qualitative analysis of the pollution, namely that:

1) given the prevailing north wind [23], most of the fly ash falls and settles in the Odesa region;

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2) the chemical composition of soil pollution changes along the imaginary line of the smoke plume – in the northern part of Odesa district, iron oxides prevail, and in the immediate vicinity and in the city of Odesa, silicon dioxide prevails;

3) the results of even very rough estimates by the authors allow us to conclude that ash particles spread much deeper into the territory of Odesa region than according to the results of the UND-86.

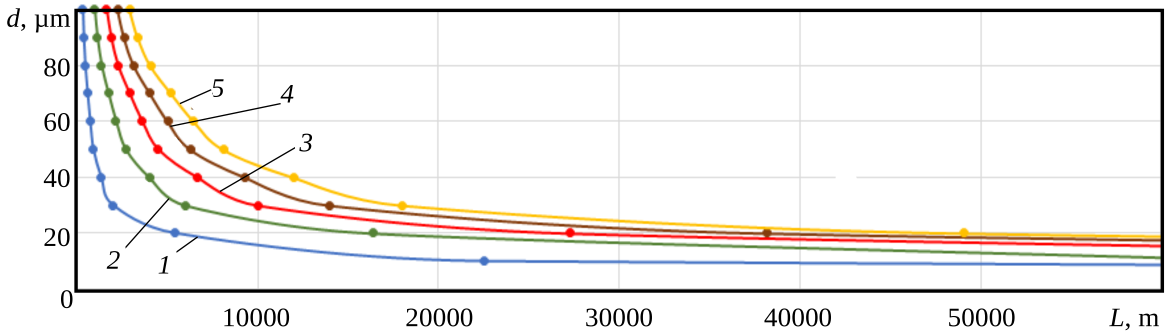

Fig. 4. Dependence of the distance L on the size of the ash particle for SiO_2:
1 – $u_{air} = 1$ m/s; *2* – $u_{air} = 3$ m/s; *3* – $u_{air} = 5$ m/s; *4* – $u_{air} = 7$ m/s; *5* – $u_{air} = 9$ m/s

It should be understood that certain simplifications had to be made in the calculations. For example, the classical experimental curve for spherical bodies $C_x = f(\mathrm{Re})$ was used, which introduces a certain error. This error increases in the zone Re << 1, which roughly corresponds to PM_{10}. The hypothesis about the sphericity of ash particles, although largely confirmed by the analysis of photographs from various literature sources, is still a simplification. Deviations from sphericity increase C_x, and thus the actual propagation distance of a non-spherical particle, ceteris paribus, will be greater. The same deviation is caused by the possible failure to fulfill the condition of particle integrity, i.e., the presence of microscopic cavities filled with flue gas. In addition, the proposed method assumes that the properties of atmospheric air and wind speed are unchanged.

Conclusions

1. Fly ash, which is not captured by flue gas cleaning devices of thermal power plants, is a dangerous pollutant of the atmospheric air and soil of the surrounding areas. Fly ash consists of a wide range of chemical elements and compounds, so its negative impact on the environment and human health can be quite significant and diverse.

2. The main components of fly ash are silica, aluminium, iron and calcium oxides. They differ in physical properties and fractional composition. This, as calculations have shown, causes their uneven distribution over the soil surface of the surrounding areas.

3. Taking into account the wind rose for Odesa region with the predominance of the north wind and the most likely wind speed of 5 to 10 m/s, fly ash pollutes the territory of the cities of Odesa, Chornomorsk, Zatoka and reaches the Tuzly Estuaries National Nature Park.

4. The UND-86 methodology does not take into account the properties of fly ash from pulverized-coal boilers of thermal power plants, namely, its fractional, chemical composition and physical properties of each element. This is the basis for revising this methodology at least in terms of calculating the dispersion of fly ash from TPPs. The revision should be in line with current EU requirements and regulations.

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