Emission of soot particles from the combustion of various fuels in boilers

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Abstract. According to modern estimates, the largest contribution to global warming is made by carbon dioxide, while its share is 60–70%. However, studies of the last decade also indicate a large impact on the planet's climate of "short-lived climate pollutants", which mean forcing substances with "lifetimes" in the atmosphere from several days to several years. These pollutants include black carbon. In the Russian Federation, black carbon refers to the substance "carbon (soot)", which is a dispersed carbon product of incomplete combustion or thermal decomposition of hydrocarbons, consisting of particles of various shapes and having a black color. Soot is a strong carcinogen. It is the 2nd class harmful substance and carried by winds for thousands of kilometres. It has been found that black carbon retains several hundred times more heat than carbon dioxide does. Therefore, reducing black carbon emissions is one of the most important challenges in the fight against climate change. Therefore, the task arises of a comprehensive study of all aspects related to black carbon emissions. Based on this, experimental studies of the emissions of soot particles formed in the combustion chambers of boilers of various capacities and designs were carried out during the combustion of various solid, liquid and gaseous fuels. The structure, size and quantitative composition of soot particles, as well as their constituent elements, were studied using scanning electron microscopy. The research results made it possible to propose a more extended approach for choosing the values of specific soot emissions from the combustion of various fuels in heat generators, which should be used in the inventory of emissions of this pollutant.

1. Introduction
The development of modern civilization leads to increasing consumption of energy. The world problems are aggravated caused by the limited reserves of fossil fuels, their extreme uneven distribution across the regions of the world and the environmental deterioration.

Power plants are the main air pollutants, sulfur and nitrogen oxides, benzopyrene, particulate matter, including soot. The contribution of power plants to carbon dioxide emissions, which causes the "greenhouse" effect, can be considered the main one.

The climate of our planet is constantly changing. Long periods of cold snap are followed by periods of warming. However, since the mid-1970s, there has been a steady upward trend in global surface air temperature, which is a matter of serious concern. According to the World Meteorological Organization, 2020 became one of the warmest year in the history of instrumental observations and is characterized by exceptionally high global surface air temperatures, an intense reduction in the area of sea and floating ice, and record high sea levels [1].
The Arctic region turned out to be the most sensitive to climate change. It should be borne in mind that in the Arctic, the rate of temperature rise is 2 times higher than that in the rest of the world [2–4]. Studies of the last decade also indicate a large impact on the planet's climate of "short-lived climate pollutants", which mean climate forcing substances with "lifetimes" in the atmosphere from several days to several years [5–9]. The results of these studies showed an objective need for urgent measures to protect the climate, and a number of countries have taken an initiative to limit anthropogenic impact on the climate system by reducing emissions of black carbon, methane and hydrofluorocarbons.

The amount of soot particles contained in the exhaust flue gases of power plants is a criterion for the efficiency of the organization of the combustion. However, currently used in the Russian Federation and many other countries, furnace and burner devices for burning various types of fuel do not have sufficient energy and environmental performance, which leads to irrational use of energy resources and significant environmental pollution. This has a particularly strong effect on the ecological situation of the subarctic territories and the Arctic [10]. Based on this, strict control over the completeness of fuel combustion is required to maintain a clean environment.

Soot is a dispersed carbon product of incomplete combustion or thermal decomposition of hydrocarbons, consisting of particles of various sizes, shapes and black color. From the standpoint of transboundary transport, the dominant negative role belongs to small particles (2.5 µm or less). Therefore, when estimating soot emissions, the methodology proposed in [11] is most often used. Therefore, when assessing soot emissions, the methodology proposed in [11] is most often used, according to which black carbon fractions with a size of 2.5 µm or less (PM2.5) have a decisive influence on transboundary transport and pollution.

Soot is a strong carcinogen. It is 2nd class harmful substance with maximum permissible concentration (MPC) of 0.15 mg/m³ and MPC daily average of 0.05 mg/m³. As dispersed dust, soot is carried by winds for thousands of kilometers and has bigger negative factor than carbon dioxide [7]. Some scientist gave black carbon the second place after carbon dioxide in term of its contribution (15-30 %) to climate warming over the past decades [12, 13]. Global estimates of black carbon emissions are 7.8 million tons per year [14].

This close focus on black carbon underscores the unique role it can play in combating global warming, especially in the Arctic, and the critical importance of conducting scientific research to develop recommendations to reduce black carbon emissions and its impact on climate.

2. Study Object
The Arkhangelsk region is a large industrial region with developed mechanical engineering, woodworking, pulp and paper and fish industries, diamond, bauxite mining, etc. Centralized power supply covers about 70% of the territory and 90% of the region's population. The power system is part of the national grid of the North-West of Russia and includes two large power centers - Arkhangelsk and Kotlas. Arkhangelsk CHPP, two CHPPs in Severodvinsk, CHPP of Arkhangelsk pulp and paper mill operate in the Arkhangelsk power center. The CHPP of the branch of Ilim Group and the Velsk branch of GT-Energo operate in the Kotlas load center. The Arkhangelsk power center is energy-excessive, and the Kotlas power center is power-hungry. However, due to the lack of transmission capacity of power transmission lines, the flow from north to south is difficult, therefore, electricity is transmitted from the center of the country and from the power system of the Komi Republic.

Centralized heat supply to consumers is carried out from the aforementioned CHPPs, industrial enterprises, as well as from local boiler houses. The total number of local sources of heat supply is about 700.

For more than four decades in the fuel and energy balance of the region, the first position was occupied by fuel oils, and the second - by coal. In recent years, significant changes have occurred in the fuel and energy balance of the region, which is associated with two determining factors:

• conversion of large power plant facilities (Arkhangelsk CHPP, Severodvinsk CHPP-2) to natural gas combustion (2011) and further gasification of the municipal energy sector;
• implementation of a program to replace imported fuels with biofuels produced at the enterprises of the region.

Under the influence of these factors, gaseous fuels took the first position in the fuel and energy balance of the region, biofuels - the second, and liquid - the third. The most environmentally "dirty" fuel – coal – moved to the fourth position.

Changes in the fuel and energy balance of the region have had a beneficial effect on the environmental situation. Thus, the total emissions of harmful substances from stationary sources have significantly decreased since 2011 (Fig. 1). The structure of gross emissions of harmful substances has also changed (Fig. 2), the largest values are for emissions of particulate matter and carbon monoxide, while emissions of sulfur dioxide have moved from the first position to the fourth.

In the course of the implementation of the program to replace imported fuels with biofuels, not only the modernization of existing energy sources was carried out, but also the construction of new modular fully automated boiler houses operating on refined environmentally friendly biofuels. So, since 2010, more than 100 energy facilities were put into operation, modernized or converted to use solid biofuels on the territory of the Arkhangelsk Region.

Thus, the total installed capacity of six new boiler houses of OOO Arkhbioenergo, operating on wood pellets, amounted to 46 MW, and the achieved gross efficiency of hot water boilers is more than 90.0%. At the same time, emissions of carbon monoxide, soot particles and nitrogen oxides were minimal. The achieved gross efficiency of biofuel boilers corresponds to the efficiency of fuel oil boilers in good technical condition. The construction and commissioning of boiler houses operating on refined biofuel made it possible to form a stable domestic market for wood pellets. This fact has provided additional guarantees for the internal stability of regional producers of pelleted fuel.

3. Materials and Methods
The experimental study of the mass concentration of soot in emissions was carried out in accordance with [15]. In this case, the following was carried out:

• sampling of a dust-laden flow in a stationary mode of operation of a heat-generating unit, taking into account the distribution of velocities over the cross-section of the gas duct in front of the chimney;
• separation of the particulate matter with filters using the method of internal or external filtration;
• calculation of the concentration of PM in the gas flow;
• the mass content of soot was determined by calcining in a furnace (at a temperature of 900–950 °C) in the air of a filter with trapped dust particles.

A Zeiss SIGMA VP (Carl Zeiss) scanning electron microscope was used to study the structure, size, and quantitative composition of soot particles, as well as to determine their constituent elements. The
setup parameters used to obtain the images: InLens and SE 2 detectors, accelerating the electron gun voltage from 5 to 20 kV.

If it was not possible to carry out detailed studies of the size of soot particles, then the results of [16] were used to determine the emissions of soot PM2.5.

4. Results and Discussion
Experimental studies were carried out on heat-generating plants with a capacity of 60 kW to 380 MW, having a different design and operating on different types of fuel. Summarizing the results obtained, it should be noted that the level of emissions of soot particles is determined by the quality of fuel, the design of furnace and burner devices, the method of combustion, the technical level and level of adjustment of the automation system, the presence and type of gas cleaning devices, as well as the technical condition and quality of maintenance of heat generating plants [10, 17].

To perform a calculated analysis of PM 2.5 emissions of soot particles according to the fuel balance, the following values of specific emissions from combustion of various fuels are recommended in [16], g/GJ:

- coal – 6.8; fuel wood – 11.0 (with capacity less than 1 MW);
- heavy oil – 0.3; natural gas – 0.3.

However, the approach proposed in [16] should be considered too simplistic. The results of the studies performed with the determination of the content of soot particles according to the method [15] made it possible to propose a more extended approach for choosing the values of specific emissions during combustion of various fuels, g/GJ.

Biofuel includes the following:

- raw firewood when burning in sectional cast iron hot water boilers with manual operation («Universal», «Tula», «Energiya» etc.);
- a mixture of chips with sawdust during combustion in mechanized boilers with a capacity of up to 2 MW, equipped with inertial ash collectors;
- a mixture of chips with sawdust during combustion in mechanized boilers with a capacity of up to 6 MW, equipped with inertial ash collectors and in operation for up to 15 years;
- a mixture of chips with sawdust during combustion in boilers of the DKVr and KE brands, equipped with high-speed combustion pre-furnaces and snail ash catchers;
- boiler units with a capacity of up to 4 MW, operating on wood pellets, equipped with inertial ash collectors;
- boiler units operating on bark and wood waste, equipped with an adiabatic furnace with three-way gas movement in the furnace volume, equipped with a pneumatic system for cleaning heating surfaces and inertial ash collectors;
- boiler units operating on waste plywood production, equipped with an adiabatic furnace with a two-way movement of gases in the furnace volume, equipped with a pneumatic system for cleaning heating surfaces and inertial ash collectors.

Peat briquettes are used in:

- steel hot water boilers KVr-0.4K with manual fuel loading, equipped with inertial ash collectors;
- steel hot water mechanized boilers KVm-2.0, equipped with fireboxes with a rustling bar and inertial ash collectors.

Reactive coal is used in:

- fire-tube boilers (two-furnace and single-furnace) with manual operation with a capacity of up to 2 MW;
- raw firewood when burning in sectional cast iron hot water boilers with manual operation («Universal», «Tula», «Energiya» etc.);
- steel hot water mechanized and steam boilers with a capacity of up to 2 MW with manual fuel loading equipped with inertial ash; 6.6
- mechanized boilers of the DKVr and KE brands, equipped with inertial ash collectors 5.1
- coal dust flare boiler units with a nominal capacity of 128–160 MW, equipped with angular slot burners with a tangential arrangement, and equipped with wet-type ash collectors. 0.7

Liquid fuels are used in:
- boilers of the DKVr, KE and DE brands when operating on heavy oil of the brand 100; 3.4
- modern waste oil boilers with a capacity of up to 5 MW; 0.3
- high-pressure steam boilers operating on heavy fuel oil, with a capacity of 320-380 MW. 0.11

Natural gas is used in:
- high pressure steam boilers with a capacity of 128 MW and more; 0.01
- low-capacity boilers. 0.05

Microscopic studies have shown that soot particles can have different shapes. Thus, in the samples taken during the combustion of peat pellets in the firebox of the Firematic 60 boiler of the Austrian company "Herz Energietechnik GmbH", three main types of particles were identified: spherical; with an amorphous structure and crystalline particles (Fig. 3).

The experimentally obtained specific emissions of PM 2.5 soot particles during the combustion of various fuels in heat-generating plants of various capacities and design made it possible to quantify the emissions of black carbon in the Arkhangelsk region for 2016. Partial financing of this work was carried out by the Center for Environmental Investments.

The total emissions of soot were calculated by multiplying the energy potential of the combusted fuel by the value of the specific emission, taking into account the type of fuel burned and the heat generating equipment used. When carrying out the calculations, the data of official statistics on the use of various types of fuel were used, taking into account the previously given values of PM 2.5 emissions obtained experimentally. The performed calculations showed that the total emissions of black carbon (PM2.5) in the territory of the Arkhangelsk region (w/out NAD) in 2016 amounted to 539.213 tons.

5. Conclusion
The level of emissions of soot particles is determined by the quality of the fuel, the design of furnace and burner devices, the combustion method, the degree of perfection and the level of adjustment of the automation system, the presence and type of gas cleaning devices, the technical condition and quality of service of heat generating plants. To determine the composition and size of soot particles, as well as the quantitative and weight content in them of particles with a size of 2.5 microns and less, the method of electron microscopy should be used.

The relevance of the task of a comprehensive study of all aspects related to emissions of black carbon, its distribution in the atmospheric air and the impact on human health and environmental components is increasing every day. The first step towards its solution should be an inventory of soot emissions. To carry out the inventory, the values of the specific emissions of soot particles PM2.5 should be used when various fuels are burned in heat generating plants of various capacities and designs.
Figure 3. The structure of solid particles produced by burning peat pellets: 
a – spherical; b – amorphous; c – crystalline.

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