Emissions of soot particles from heat generators

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Abstract. «Soot carbon» or «Soot» - incomplete combustion or thermal decomposition particulate carbon product of hydrocarbons consisting of particles of various shapes and sizes. Soot particles are harmful substances Class 2 and like a dust dispersed by wind for thousands of kilometers. Soot have more powerful negative factor than carbon dioxide. Therefore, more strict requirements on ecological and economical performance for energy facilities at Arctic areas have to be developed to protect fragile Arctic ecosystems and global climate change from degradation and destruction. Quantity of soot particles in the flue gases of energy facilities is a criterion of effectiveness for organization of the burning process. Some of heat generators do not provide the required energy and environmental efficiency which results in irrational use of energy resources and acute pollution of environment. The paper summarizes the results of experimental study of solid particles emission from wide range of capacity boilers burning different organic fuels (natural gas, fuel oil, coal and biofuels). Special attention is paid to environmental and energy performance of the biofuels combustion. Emissions of soot particles PM2.5 are listed. Structure, composition and dimensions of entrained particles with the use of electronic scanning microscope Zeiss SIGMA VP were also studied. The results reveal an impact of several factors on soot particles emission.

1. Introduction
Quantity of soot particles in the flue gases of energy facilities is a criterion of burning process effectiveness. Many of the existing furnace and burner devices do not provide the required energy and environmental efficiency, which results in irrational use of energy resources and acute pollution of environment. This in particular affects environment of the Arctic territories. Soot is a dispersed carbon product of incomplete combustion or thermal decomposition of hydrocarbons, consisting of particles of various shapes and sizes. Emission of soot particles can lead to big losses of fuel. Soot is a powerful carcinogen classified as a class 2 hazardous substance with maximum permissible concentration limit 0.15 mg/m³, and can be dispersed by wind as a dust for thousands of kilometers. The negative impact of soot is much (680 times) higher than for carbon dioxide.

2. Main part
2.1 Residual fuel oil and natural gas
High sulfur fuel oil ranked first in the fuel balance of the Arkhangelsk region for a long period. This exerted a strong influence on the ecological situation in the region, which is part of the Arctic zone of the Russian Federation. The transfer of large energy facilities carried out in 2011 in Arkhangelsk and
Severodvinsk from fuel oil to natural gas allowed to reduce the total emissions of pollutants into the atmosphere by more than 50 thousand tons per year.

Survey of boiler units (320-380 MW) of large power plants using natural gas (the proportion of 98% methane) revealed [1] that the experimental measurements carried out by the method [2] provide results within the method’s permissible tolerance. These boiler units operating on grade 100 residual fuel oil provide PM 2.5 emission factor of 0.104-0.110 g/GJ.

2.2 Coal
The second position in the fuel and energy balance of the region is occupied by coals. The results of the test measurements performed at boiler units (128-160 MW) equipped with tangential burners and wet scrubbers revealed that PM 2.5 emission factor during burning of high reactivity black coal was 0.25-1.05 g/GJ.

For the small power boilers with high physical depreciation operating at black coal PM 2.5 emission factor values is within a very wide range 2.0-26 g/GJ. In accordance with the investigations [1] the level of soot particles emissions depends on the fuel quality, burner unit design, combustion method and settings of the automation system, gas cleaning installation type, technical state and maintenance factor of the heat producing units.

2.3 Biofuel
Increase of the bioenergetics share in the energy balance of many European countries and forest regions of Russia is an upcoming trend to reduce technological environmental impact of the power industry facilities. It is particularly important to involve by-products of timber industry for energy production.

2.3.1 Arkhangelsk plywood factory
A large amount of by-products is formed in the cycle of plywood production, which from economic and environmental points of view should be used as energy fuel. In 2015 ZAO «Arkhangelsk plywood factory» was installed and put into operation boiler PRD 22000 Austrian company «Polytechnik Luft- und Feuerungstechnik GmbH». This boiler produces saturated steam at a temperature of 192 °C, its nominal capacity is 22 MW. Fuel for boiler is the mixture consists of chip plywood, pitch bark, veneer, wood sanding dust, sawdust of the plywood processing. The components of the fuel mixture significantly differ in thermotechnical characteristics and technological parameters, but especially in size composition [3]. Particle dimensions in the fuel mixture differ by more than a thousand times which makes it «unique» and very difficult to ensure the effective and non-explosive use of energy. However, the boiler design and automation system provided cost-effective energy use of the fuel mixture with high environmental performance. Gross efficiency in the nominal capacity range 89-96 % was 89.4-90.26 %; NOx emission was 62-67; carbon monoxide emission was 11-12; soot particles emission factor was 7.99-8.80 g/GJ. The PM 2.5 emission factor was 1.62-1.79 g/GJ, which is a good result.

2.3.2 Sawmill 25
Boilers PRD 9500 Austrian company «Polytechnik Luft- und Feuerungstechnik GmbH» were installed in the CHP-2 of ZAO «Sawmill 25» and produce superheated steam at a pressure of 2.4 MPa and a temperature of 425 °C. Nominal capacity of each boiler is 9.5 MW. During energy tests the boilers burned bark wastes at a moisture content \( W_r = 57.2-59.8 \% \). Gross efficiency in the capacity range 7.82-9.14 MW was 86.96-87.86 %; NOx emission was 57-71; carbon monoxide emission was 4-144 g/GJ [4]. Two sampling methods were used to determine the particles content and the efficiency of the ash collectors: internal and external filtration. Sampling was carried out at the dust-laden flow isokinetic conditions; in this case, data from preliminary calibrations was used.

The study allowed to determine soot particles emission factor which amounted in 52.72-58.61 g/GJ. Wherein PM 2.5 emission factor varies between 8.61 and 10.03 g/GJ. Study of selected particles
with use of electronic scanning microscope Zeiss SIGMA VP (Carl Zeiss) allowed to determine the structure, dimensions of particles and qualitative composition of constituent elements. PM 2.5 emission factor were 1.21-1.4 g/GJ. The size of soot particles was in the range of 0.1-10 µm [4]. The results of the experiments revealed that the inertial ash collectors (RGE 9500) do not allow effective purification of flue gas from soot particles due to their small dimensions and lower density in comparison with mineral particles. This is evidenced by the increased content of soot particles in the solid phase: before ash collector 4.6-5.5 %, after - 16.3-17.1 %. The values of the total purification degree of flue gas from the solids in the gas cleaning facilities of boilers accounted for 85.3-90.0 %, and the degree of purification of soot particles was 63.1-67.0 %.

2.3.3 Boiler rooms
Automated boilers Polytechnik Luft- und Feuerungstechnik GmbH with capacity of 4.0 MW and Arimax Bio Energy with capacity of 0.7-1.5 MW equipped with inertial ash collectors are used at small power facilities operating on pellets in the Arkhangelsk region. Three-step scheme of combustion is utilized at the 4.0 MW boilers furnaces. Two-step scheme of combustion is utilized at the 0.7-1.5 MW boilers furnaces. Additional configuration of the boilers automation system has allowed to provide high completeness of fuel combustion and stable low values of soot particles emissions [5]. PM 2.5 emission factor was 0.521-0.966 g/GJ. Maximum achieved gross efficiency of boilers with capacity 0.7 MW was 91.0 %, 1.5 MW - 92.0 % and 4.0 MW - 91.3 %; which corresponds to the efficiency of oil-fired boilers in a good technical condition. To ensure stable high economic and environmental performance of these boilers it is necessary to conduct monitoring of the pellets quality indicators including the content of dust particles. In addition, cleaning of each boiler element must be carried out promptly.

2.3.4 Scientific platform
An effective direction of comprehensive solution of energy and environmental problems in providing heating loads of the household sector consumers is the use of modern boilers for solid biofuels and peat. The scientific platform for research is the boiler Firematic 60 manufactured by Austrian company «Herz Energietechnik GmbH». The boiler is design to work on pellets and wood chips no larger than 50 mm with relative moisture less than 35 %.

The study was carried out in several stages. During the first stage, experiments were carried out with feeding boiler furnace by 8 mm in diameter pellets with uniform particle size distribution. At the second stage the boiler worked with pine bark which thermotechnical characteristics were: moisture \( W_{fr} =13.72 \% \); ash content \( A' =2.52 \% \); received fuel mass inferior calorific value \( Q_i' =17.98 \) MJ/kg.

Results showed that the boiler operating on biofuels provides high environmental and economic performance and should be used for the heating low-rise buildings, especially in the Arctic region conditions [5]. Gross efficiency of the boiler operating on pellets was 92.86-95.20 %; NO\(_x\) emission was 51-54; carbon monoxide emission was 12-21 g/GJ. For the boiler operating on pine bark which has less uniform particle size distribution, the environment and economic performance had the following values: gross efficiency was 92.7-93.2 %; NO\(_x\) emission was 57-100; carbon monoxide emission was 85-323 g/GJ.

During the third stage, experiments were carried out with applying non-project fuels: peat granules with a diameter of 10 mm and sod peat characterized by a nonuniform particle size distribution. Thermotechnical characteristics of peat granules were: \( W_{fr} =16.50 \% \); \( A' =9.95 \% \); \( S_i' =0.22 \% \); \( Q_i' =14.88 \) MJ/kg. Thermotechnical characteristics of sod peat were: \( W_{fr} =8.2 \% \); \( A' =11.76 \% \); \( S_i' =0.19 \% \); \( Q_i' =15.47 \) MJ/kg [6].

When the boiler operates on the peat granules which ash content significantly larger in comparison with pine, the heat loss with mechanically incomplete combustion increased to 2.17 % and gross efficiency reduced to 91.24-91.37 %. Emissions of harmful substances were: NO\(_x\) 169-177; CO 7-23; SO\(_2\) 331-338 g/GJ.
Conversion to burning the sod peat with high content of fine particles led to a sharp increase of heat loss with mechanical incomplete combustion to 13.00%. Gross efficiency of the boiler is decrease to 81.01-81.22%. Fractional ash fly analysis revealed that the greatest amount of combustible components contained in particles of 0.5 mm or more. Analysis of the experimental data, with taking into account the mass content of different fractions, revealed that removal of unburned peat particles with a size of 0.25<δ<2 mm has significant role in the mechanical incomplete combustion heat loss.

This boiler for burning sod peat is not recommended to use without significant changes in the collection and removal of the combustion residues due to the high ash content and high heterogeneity of particle size distribution of sod peat in comparison with biofuels (pellets and wood chips). Conversion from the peat granules to burning sod peat caused approximately three-time increase of the solid particles concentration in the flue gases.

During the fourth stage, experiments were carried out with spruce granules with a diameter of 6 mm and hydrolytic lignin biochar with diameter of 8 mm. Thermotechnical characteristics of biochar had the following values: \( W'_r = 6.27 \% \); \( A'_r = 2.45 \% \); \( Q'_i = 21.34 \text{ MJ/kg} \). Transition to burning the hydrolytic lignin biochar led to increase of boiler efficiency to 92.25-92.55%; despite the higher ash content of the latter (5.56 times). In addition, with the burning of the biochar, minimal emissions of nitrogen oxides (44-46 g / GJ) and carbon monoxide (2-5 g / GJ) were achieved.

In the experiments, particulate sampling from the gas flue before chimney was carried out using the method of external filtering. The captured particles were studied by the electronic scanning microscope. Device parameters used to capture images were: InLens and SE2 detectors, the accelerating voltage of the electron gun 5 to 20 kV.

Results of the study of soot particles emissions during the combustion of pellets revealed that the average soot particles emission factor was 5.594 g/GJ and PM 2.5 emission factor was 0.834 g/GJ. During the pine bark combustion the soot particles emission factor grew up to 9.324 g/GJ and PM 2.5 emission factor up to 1.305 g/GJ.

For the boiler operating on peat granules the average soot particles emission factor was 2.477 g/GJ and PM 2.5 emission factor - 0.347 g/GJ. Transition to burning the sod peat was followed by the sharp increase of soot particles emission factor to 17.254 g/GJ and PM 2.5 emission factor to 2.416 g/GJ.

For the boiler operating on hydrolytic lignin biochar the average soot particles emission factor was 2.467 g/GJ and PM 2.5 emission factor - 0.345 g/GJ. Studies have shown that solid particles with a particle size of less than 13 μm are predominantly emitted into the atmosphere.

Three main types of particles in selected samples have been identified [7]: spherical shape, with an amorphous structure and solid crystal particles as shown in Fig. 1 and 2. The origin of spherical particles is probably due to the decomposition of plant organic matter. Influenced by heat, the sugar decompose and take on the properties of a fluid. This fluid forms spherical particles during solidification. We can assume that the particles are composed of hydroxymethylfurfurol.
**Figure 1.** The structure of the solid particles produced by burning peat granules: a - spherical; b - amorphous; c – crystalline

**Figure 2.** The structure of the solid particles produced by burning sod peat: a - spherical; b - amorphous; c – crystalline
Crystalline particles in the sample taken after peat granules combustion are likely unburned plant residues that are part of peat. The part of crystalline particles contained in the sample taken after sod peat combustion with determined amounts of silicon can be particles of sand and clay. Another part of crystalline particles is unburnt plant residues that are part of peat.

With given elemental composition data [7], particles that have amorphous structure are fragments of the peat-forming components.

Simultaneously with the study of soot particles shapes, their dimensions were determined (Table 1).

| Name of particles              | Peat granules, min/max, micron | Sod peat, min/max, micron | Hydrolytic lignin biochar, min/max, micron |
|-------------------------------|--------------------------------|---------------------------|------------------------------------------|
| Spherical shape               | 0.7 / 11                       | 0.2 / 15                  | 0.7 / 3                                  |
| With amorphous structure      | 0.1 / 15                       | 0.1 / 25                  | 0.5 / 15                                |
| Crystalline particles         | 0.1 / 20                       | 0.1 / 30                  | 1 / 15                                  |

3. Conclusion
The process of burning organic fuels is accompanied by the formation of soot particles. The amount of soot particles emissions depends on the fuel quality, burner unit design, combustion method and settings of the automation system, gas cleaning installation type, technical state and maintenance factor of the heat producing units. Small size of soot particles and the global atmospheric circulation contribute to its transfer for thousands of kilometers. Therefore, more strict requirements on energy and ecological performance for energy facilities at Arctic areas have to be developed to protect fragile Arctic ecosystems and global climate change from degradation and destruction.

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