Thermal sources of low power and their caricena hazard to the atmosphere of the cities of the Irkutsk region

L I Belykh and M A Maksimova

Irkutsk National Research Technical University, Russia

E-mail: belariv2000@yandex.ru, marinamaksimova@outlook.com

Abstract. In the article, we presented the results of experiments in the stratified burning of solid fuels in “low” power boiler houses and household furnaces. We also studied the dynamics in which the products of incomplete combustion of coal and wood, i.e. carbon monoxide, soot, benzo(a)pyrene and other polycyclic aromatic hydrocarbons (PAHs), are emitted. The paper shows the non-stationarity of the combustion process taking into account the proposed method for calculating emissions of hazardous substances. The article also determines their specific emissions during the combustion of coal and firewood in the Eastern Siberia region. The paper contains the characteristics of “low” power heat sources and the assessment of their incomplete combustion products for the atmosphere of the Irkutsk region cities. It also proposes measures to reduce emissions of hazardous substances into the environment.

1. Introduction

Monitoring of the atmospheric air state in the cities of Russia [1] reveals the systematic exceedance of the maximum allowable concentrations (MAC) of many hazardous substances. Among them, a significant proportion belongs to carcinogenic benz(a)pyrene (B(a)P), an indicator of the priority persistent organic pollutants of the polycyclic aromatic hydrocarbons (PAH) group. These substances are formed in the processes of incomplete combustion and pyrolysis of organic materials [2-4], in particular, when burning fuel in various heat sources. Therefore, air pollution is more characteristic of the heating season. This is particularly true for cities of Siberia and the Russian Far East where there is a high proportion of boiler and individual heating with coal and firewood. For example, in the cities of the Irkutsk region only about 60% of the population lives in conditions with modern amenities and central heating. The rest is provided with heat using “low” power heat sources. In cities using such a type of heating, the maximum mean monthly concentrations of B(a)P in the air exceed MAC by ten folds [5]. Given this level of atmospheric pollution, the control of carcinogenic PAHs is in an unsatisfactory state.

Despite many studies of the formation of these substances in combustion processes [6-8], there are no exact methods for calculating the mass of PAH emissions from furnace installations. In a number of industrialized countries, such assessments are carried out [9-12], but their results have significant variations, for example, in terms of their specific values. Therefore, it seems relevant to study the process of burning solid fuels in “low” power heat sources such as boiler houses and household furnaces as sources of emissions of carcinogenic substances into the atmosphere using the example of the Irkutsk region cities.
2. Study objects and methods

The study objects were “low” power heat sources with a stratified periodic (cyclic) mode of burning solid fuel used in the Irkutsk region and in other regions of Siberia. This is a hot water boiler with a fixed grate, a thermal capacity of 0.8 MW and manual coal loading. Boiler type - NRS-18, with a grate area of 1.8 m$^2$, furnace volume of 5.8 m$^3$, dimensions – length: height – 3.14 : 2.39 : 2.60 m. For comparison, we took a 1 MW oil-burning boiler with mechanical atomizers. The brick household furnace was of a standard design with a thermal capacity of about 35 kW, intended for burning firewood and coal. Furnace dimensions 1.0 : 0.5 : 2.1 m, wall thickness 0.12 m, gas passage 0.12: 0.25 m. A distinct feature of the solid fuel stratified burning is the supply of air under the fuel strata through the grate and the rapid decrease in excess air, which does not provide sufficient oxygen.

The properties of the main hazardous incomplete combustion products are given in Table 1.

Table 1. Properties of incomplete combustion products.

| Substance (CAS) | Formula | Chemical structure | Molecular weight | MAC* (hazard class) | Temperature °C |
|----------------|---------|-------------------|------------------|---------------------|----------------|
| Benzo(a)pyrene (50-32-8) | C_{20}H_{12} | | 252 | MAC$_{md}$ 1 ng/m$^3$ 1 haz. cl. | | 179 | 310 |
| Soot (carbon black, 1333-86-4) | C$_n$ | C=C-C= | 138 | MAC$_{md}$ 0.05 mg/m$^3$ MAC$_{ms}$ 0.15 mg/m$^3$ 3 haz. cl. | | 156 | 313 |
| Carbon oxide (630-08-0) | CO | C=O | 28 | MAC$_{md}$ 0.3 mg/m$^3$ MAC$_{ms}$ 0.3 mg/m$^3$ 4 haz. cl. | | -205 | -192 |

MAC – maximum allowable concentration for atmospheric air in populated areas; MAC$_{md}$ – mean daily and MAC$_{ms}$ – maxim single

In addition to B(a)P adopted in Russia as an indicator of the PAH group, another 10 of its analogues were determined. They are included in the list of 6 and 16 PAH compounds with different physical-chemical and carcinogenic properties and correspond to two existing international standards [6, 9].

The fuel under study is represented by black and brown coal typical for the region of Eastern Siberia, fuel oil for the fuel oil boiler house and firewood for the blast furnace. Their main properties are given in Table 2.

The combustion technology is characterized by periodic manual fuel throwing with a burning cycle of 25-30 minutes. The process goes through three stages: ignition and heating of the fuel, release of volatile substances and their combustion, burning of the coke residue. In this mode, and in the presence of natural draft, thermal and gas-dynamic parameters in the furnace and in smoke emissions change with time.

The experimental procedure included feeding a certain amount of the fuel under study into the furnace (20 kg for boiler house and 5 kg for household furnace), its burning and measuring the emissions in the chimney behind the boiler or behind the furnace over the entire combustion cycle. We measured emission parameters (temperature, pressure, velocity, volumetric gas flow rate), gases (O$_2$, CO, CO$_2$), particulate matter and aerosols, soot, B(a)P, and other PAHs.

We used a KM9006 QUINTOX combustion analyzer to measure the parameters of gas and dust flows and combustion products; gas analyzers to measure concentrations of O2 (TEMOX) and CO (TEMCO); micromanometer to measure static pressure in the gas passage and Pitot tube to measure dynamic pressure; anemometer to measure the flow rate. Sampling of flue gases to find solid particles, soot and PAHs in them was carried out using aerosol filters (AFA-VP-20 and AFA-RFA-20) by the
method of external filtration under isokinetic conditions. Samples were taken within 1 minute with an interval of 2 minutes for the entire combustion cycle of one solid fuel load. Such experimental conditions are due to the non-stationarity of the combustion process and the need to study the dynamics of PAH emissions over the entire combustion cycle of a fuel portion. Measurements of parameters and sampling were carried out in the center of the gas passage. Sampling started from the moment of ignition of a fuel portion and continued until the complete cessation of combustion. The experiments were repeated several times after the furnace was completely cooled and a new portion of the fuel was loaded.

Table 2. Main characteristics of the fuel under study.

| Fuel                  | Fuel composition | Lower heat value, Q^\text{f}, MJ/kg |
|-----------------------|------------------|-------------------------------------|
|                       | humidity, W^\text{f}, % | ash content, A^\text{f}, % | sulfur, S^\text{f}, % |                               |
| Black coal            |                  |                                    |                        |                                |
| Cheremkhovsky         | 15^*             | 34                                  | 1.2                    | 16.4                           |
|                       | (12.0-13.3)**    | (25.0-33.0)                         | (1.0-1.5)              | (16.6-20.1)                    |
| Tugnuysky             | 7.5              | 23                                  | 0.5                    | 21.0                           |
|                       | (8.5-9.5)        | (18.2-30.0)                         | (0.36-1.14)            | (20.1-21.2)                    |
| Brown coal            |                  |                                    |                        |                                |
| Azeysky               | 25               | 22                                  | 0.6                    | 16.0                           |
|                       | (24.9-26.0)      | (18.9-29.8)                         | (0.48-0.54)            | (14.2-16.8)                    |
| Mugunsky              | 22               | 20                                  | 1.2                    | 17.3                           |
|                       | (18.9-22.0)      | (18.1-21.3)                         | (0.70-0.85)            | (18.3-19.4)                    |
| Borodinsky            | 33               | 11                                  | 0.3                    | 15.3                           |
|                       | (11.0-15.0)      | (0.3-0.4)                           |                         | (14.4-15.3)                    |
| Fuel oil (M-100)      | 2                | 0.06                                | 1.4                    | 41.0                           |
|                       | (1.6-2.3)        | (0.04-0.14)                         | (1.2-1.6)              | (40.5-41.5)                    |
| Firewood (pine)       | 40               | 0.6                                 | 0                      | 10.2                           |

*reference data; **data of manufacturers

The mass of particulate matter collected on the filter was determined by the gravimetric method with an error of determination of ± 30% at a confidence level of P=95%. The samples were tested for soot and concentrations of 11 PAHs using the chemical method. After extracting PAHs with n-hexane from filters, they were identified by high performance liquid chromatography on a Milichrome A-02 chromatograph (Russia) with a column and with Nucleosil 5-C18 PAH sorbent. Qualitatively, PAH peaks in chromatograms were identified by retention time and spectral ratios of the peak areas of individual compounds. Quantitatively, PAHs were determined by external standardization using Supelco Co standards (USA). The maximum error in the chromatographic determination of PAHs did not exceed ± 10% at a confidence level of P=95%. Taking into account the sampling stage, the total error increased to ±30% at P=95%.

3. The results of the study and their discussion

The main parameters of fuel combustion are shown using a typical example of burning coal in a furnace stratum during a cycle (Table 3).

The initial stage of combustion is characterized by low temperatures and low CO_2 content of the complete combustion product. At this stage of the process, there is a peak of emissions of incomplete combustion products - CO and soot - although at this time there is enough oxygen in the furnace. Low rate of oxidation of substances is due to low temperature. At the stage of intensive fuel burning, the temperature, having reached a maximum, stabilizes. At this time, a small increase in the CO content is recorded despite the sufficient amount of oxygen until the end of the fuel combustion. At this stage of
combustion, the oxidation process is limited not by the diffusion of oxygen, but by the kinetics of the reactions, i.e. by the temperature. In separate experiments, it was shown that the CO concentration decreases by a factor of 3-4 when using fine fractions of coal, a small amount of fines, a small fuel load, and when using dry, not wet, firewood.

Table 3. Parameter dynamics in the flue emissions from stratified burning of coal per cycle.

| Parameter               | Burning time, min |
|-------------------------|-------------------|
|                         | 1                 | 4                 | 7                 | 10                | 13                | 16                | 19                | 22                |
| Temperature, oC         | 143               | 182               | 220               | 232               | 237               | 237               | 235               | 232               |
| O₂ content,%            | 17.6              | 16.9              | 16.2              | 15.8              | 15.5              | 15.4              | 15.4              | 15.5              |
| CO₂ content,%           | 2.70              | 3.95              | 4.80              | 5.10              | 4.85              | 4.55              | 4.25              | 4.20              |
| CO content, ppm         | 3870              | 2940              | 710               | 1000              | 930               | 870               | 820               | 790               |
| Soot content, g/m³      | 1.64              | 0.40              | 0.45              | 0.30              | 0.75              | 0.35              | 0.23              | 0.24              |
| Particulate matter content, g/m³ | 2.61 | 0.65 | 0.72 | 0.46 | 0.47 | 0.40 | 0.37 | 0.38 |

Soot, similar to CO, is also formed at the initial stage of combustion, being a product of oxidative pyrolysis. Significant amounts of soot are found on particulate matter and on unburned fuel particles. Soot is a carrier of PAHs, many of which are not only toxic, but also carcinogenic, especially B(a)P.

The content dynamics of 11 PAHs in the combustion cycle showed that the main part of the substances, like CO and soot, are formed at the initial stage of the combustion process, namely at the stage of pyrolysis. Figure 1 shows typical examples of identification of some PAHs.

A similar pattern of dependencies was observed for other compounds studied in the stratified boiler and the household furnace for all types of coal and firewood. The curves show that the largest fraction of the PAH mass is released in the first 10-15% of the combustion cycle time. Significant irregularity of substance emissions is associated with the non-stationarity of physical and chemical processes in the stratum. In the initial period, the stratum volume contains zones with a lack of oxygen where thermal destruction (pyrolysis) takes place. As the temperature increases, the substances formed burn out, as evidenced by a significant decrease in PAH concentrations. When feeding a new portion of fuel, the dynamics of emissions recur. For such an uneven process of emissions, the results of their single measurements do not provide accurate estimates of the mass of substances formed. Therefore,
based on the results of experiments, it was proposed to calculate the total quantities of emitted substances over the entire combustion cycle.

The method for calculating specific emissions of incomplete combustion products was developed based on the measurements of the concentration of substances in flue gases, taking into account the amount of fuel burned in the furnace. We carried out discrete sampling of 10-12 samples in 25-30 minutes of one cycle (portion) of fuel combustion. Independent experiments were repeated at least three times. The results allowed for calculating the average values.

To determine the specific emissions (M_{sp}), first, we calculated an instantaneous mass emission, kg/s, using the measurement data and the formula:

\[ m_{ij} (g/s) = C_{ij} \cdot V_i \cdot 10^{-3}, \]  

where \( C_{ij} \) is the concentration of the \( j \)-component (g/m\(^3\)) taken for the \( i \)-measurement number; \( V_i \) is the volumetric flow rate of flue gases (m\(^3\)/s) which can be determined by instrumental or computational methods using appropriate methodologies; \( 10^{-3} \) is the coefficient of the content of recalculation, g to kg. When expressing concentrations in units of mg/m\(^3\) or µg/m\(^3\), conversion factors amount to \( 10^{-6} \) or \( 10^{-9} \), respectively.

The obtained values of instantaneous mass emissions for each component are summed over the entire combustion cycle. They are necessary to determine the specific substance emission per GJ unit of heat of the burned fuel. Specific emissions (\( M_{sp,j} \), kg/GJ) are calculated using the formula:

\[ M_{sp,j} = \frac{1000}{B} \cdot \frac{Q'}{l} \sum_{i=1}^{n-1} (m_{ij} \cdot \Delta t_i), \]  

where \( B \) is the mass of fuel burned during the combustion cycle, kg; \( Q'/l \) is the lower heat value of the fuel, MJ/kg; \( i \) is the ordinal number of the measurement, \( n \) is the number of measurements; \( \Delta t_i \) is the time interval between the \( i \)-th and \( i+1 \) measurements, sec.

Specific emissions of hazardous substances are used to calculate gross emissions (\( M_{G,j} \), t/year) using the formula:

\[ M_{G,j} = M_{sp,j} \cdot Q'/l \cdot B_B \cdot 10^{-3}, \]  

where \( B_B \) is the amount of fuel consumed per year, t/year.

The results of determining specific emissions of the main incomplete combustion products in a boiler house and a household furnace are given in Tables 4 and 5, respectively. The analysis of the data obtained for the stratified boiler shows that the values of specific emissions of particulate matter, CO, soot and B(a)P significantly depend on the type of fuel and, accordingly, the method of burning (stratified and flare) in coal and fuel oil boilers. The difference in this case may amount to several orders of magnitude, especially for B(a)P. Whereby, specific emissions of substances are essentially independent of the coal type. The results are comparable when burning both black and brown coals under the same conditions.

The scatter of the specific values is characterized by a variation coefficient from 7.4 to 47%, which is close to the errors in determining emission indicators. Therefore, it is possible to propose to average specific emissions for all types of coal studied and applied in the region. It can be assumed that other types and grades of coal will have similar emission characteristics.

The behavior considered for heating boilers is retained for the study of emissions from the household furnace, for which the variation in specific emissions of hazardous substances varies from 14 to 64% and allows averaging data for different types and grades of coal. For such type of fuel as firewood, specific emissions of substances are 3-20 times less than for coal. Therefore, their averaging is impossible.

Table 6 presents specific emissions of all 11 PAHs under study in the process of fuel combustion in heating boilers and household furnaces.
Table 4. Specific emissions of fuel combustion products in stratified heating boilers.

| Fuel                   | Average specific emissions of hazardous substances, kg/GJ |                 |                  |                  |
|------------------------|----------------------------------------------------------|-----------------|-----------------|-----------------|
|                        | particulate matter | carbon oxide, CO | soot            | benzo(a)pyrene, g/GJ |
| Coal-burning boiler    |               |                 |                 |                  |
| Black coal             |               |                 |                 |                  |
| Cheremkhovsky          | 0.80          | 2.7             | 0.58            | 0.207           |
| Tugnuysky              | 0.66          | 2.5             | 0.43            | 0.066           |
| Brown coal             |               |                 |                 |                  |
| Azeysky                | 0.55          | 2.5             | 0.36            | 0.180           |
| Mugunsky               | 0.35          | 3.0             | 0.18            | 0.093           |
| Borodinsky             | 0.57          | 2.6             | 0.35            | 0.100           |
| For all coals average ± S (V, %) | 0.60±0.16 | 2.7±0.2 | 0.38±0.14 | 0.130±0.061 |
| Oil-burning boiler     |               |                 |                 |                  |
| Fuel oil M-100         | 0.06          | 0.7             | 0.038           | 0.0003          |

Note: *S* - standard deviation; *V* - coefficient of variation.

Table 5. Specific emissions of solid fuel combustion products in the household furnace.

| Fuel                   | Average specific emissions of hazardous substances, kg/GJ |                 |                  |                  |
|------------------------|----------------------------------------------------------|-----------------|-----------------|-----------------|
|                        | particulate matter | carbon oxide, CO | soot            | benzo(a)pyrene, g/GJ |
| Black coal             |               |                 |                 |                  |
| Cheremkhovsky          | 0.60          | 4.0             | 0.52            | 0.263           |
| Tugnuysky              | 0.60          | 4.0             | 0.55            | 0.929           |
| Brown coal             |               |                 |                 |                  |
| Azeysky                | 0.50          | 3.1             | 0.31            | 0.450           |
| Mugunsky               | 0.40          | 3.2             | 0.3             | 0.291           |
| Borodinsky             | 0.20          | 3.0             | -               | -               |
| For all coals average ± S (V, %) | 0.50±0.17 | 3.5±0.50 | 0.40±0.13 | 0.480±0.308 |
| Firewood (pine)        | 0.15          | 3.0             | 0.10            | 0.018           |

Note: *S* - standard deviation; *V* - variation coefficient.

Pyrene, phenanthrene, and fluoranthene predominate among individual PAHs in the composition of the combustion products. The specific emissions of indicator B(a)P vary considerably depending on the fuel type. The processes of burning fuel oil in heating boilers and burning wood in household furnaces have the lowest values. The content of B(a)P in the total amount of PAHs is 2-7% for a coal-fired boiler and 2-3% for an oil-burning boiler, 3-10% for a coal-burning household furnace, and 9-10% for burning firewood. For all PAHs and their total, the highest specific emissions are observed when fuel is burned in a household furnace. Comparison of the PAH content in flue emissions and the main incomplete fuel combustion product, CO, gives positive correlations. This demonstrates close mechanisms of the formation of PAH and CO, especially at the initial stage of the process at low temperature in the furnace stratum.

From the obtained dependencies of the PAH formation, we can conclude that in order to reduce the emissions of substances, it is necessary to accelerate the pyrolysis stage, for example, by more
frequent throwing of small portions of fuel into a hot furnace. In all experiments, approximately in the middle of the combustion cycle, at sufficiently high temperatures, there was a small peak of PAH concentrations (see Figure 1) and CO (see Table 3). This result can be explained by a decrease in the oxygen content in the stratum. It can be assumed that the emissions of PAHs and B(a)P in their composition depend more on burning conditions, and not on the coal properties. This is also confirmed by the above-noted results, according to which there is no significant effect of the coal grade on emissions of substances. The available results show that during stratified burning of coal and firewood, PAHs are more likely to be formed by synthesis of volatile substances during gas-phase oxidation, and not due to the destruction of carbon-bearing fuel substances. The predecessors of PAHs may be unsaturated organic compounds of thermal fuel destruction. By providing optimal conditions for temperature and excess oxygen in the combustion stratum by using sorted coal with its periodic mixing, it is possible to reduce the emissions of PAHs and B(a)P.

| PAH              | Average specific emissions, g·GJ-1 |
|------------------|-----------------------------------|
|                  | Heating boilers | Household furnace |
|                  | coal | fuel oil | coal | firewood |
| Phenanthrene     | 0.620 | 0.0028 | 1.970 | 0.0187 |
| Anthracene       | 0.140 | 0.0003 | 0.517 | 0.0025 |
| Fluoranthene     | 0.470 | 0.0005 | 1.650 | 0.0211 |
| Pyrene           | 0.690 | 0.0014 | 2.064 | 0.0322 |
| Benz(a)anthracene| 0.102 | 0.0005 | 0.350 | 0.0136 |
| Chrysene         | 0.132 | 0.0012 | 0.400 | 0.0143 |
| Benz(b)fluoranthene| 0.090 | 0.0009 | 0.360 | 0.0205 |
| Benz(k)fluoranthene| 0.100 | 0.0019 | 0.250 | 0.0174 |
| Benzo(a)pyrene   | **0.130** | **0.0003** | **0.480** | **0.0183** |
| Benzo(g,h,i)perylene | 0.035 | 0.0004 | 0.120 | 0.0104 |
| Indeno(1,2,3-c,d)pyrene | 0.062 | 0.0005 | 0.250 | 0.0216 |
| **Total of 11 PAHs** | **2.580** | **0.0107** | **8.418** | **0.1906** |

Note: the average is calculated for all grades of coal: black coal: Cheremkhovsky, Tugnuysky; brown coal: Azeysky, Mugunsky, Borodinsky. Firewood - pine, fuel oil M-100.

“Small” heat power engineering has a significant place in the structure of the fuel and energy sector of the Irkutsk region. The heat supply of the Irkutsk region cities for the period of 2017 was provided by various sources. These are 15 combined heat and power plants with a capacity of more than 50 MW, medium-sized boilers with a capacity from 5 to 50 MW and small boilers with a capacity of less than 5 MW. All heat sources use coal of local open-pit mines as the main fuel. Their characteristics are presented in Table 2. This heat supply covers about 60% of the population housing with modern amenities. The remaining 40% of housing lacks amenities and has different types of heating. These include 1005 boiler houses, of which 872 are communal ones. There are also about 300 electric boiler heat sources, a large number of heat recovery units (HRU) and individual heating furnaces.

The main type of fuel for household furnaces is firewood and coal, whereby firewood predominates approximately by three-fold. Using statistical reports of a number of heat supply programs in the Irkutsk region [13-15], we took into account the quantities and types of fuel consumed for “small” boiler houses and individual houses. Table 7 shows the structure of small heat sources and fuel consumed by them.

Coal (62.5%), then firewood and chips (14.6%), liquid fuel (3.3%) and gas (0.9%) predominate among the types of fuel used. Electric plants take approximately a fifth (18.8%) of the total number of boiler houses. A comparison of gross emissions of B(a)P and total PAHs in boiler houses using coal and fuel oil and the best environmental characteristics of fuel oil boiler houses shows the discrepancy
between the results of more than three orders of magnitude. At the same time, the number of fuel oil boiler houses in the region is negligible.

Table 7. Structure of communal “small” heat sources, their fuel consumption and formation of incomplete combustion products in the Irkutsk region.

| Indicator (for 2017) | Fuel-fired boiler houses | Total | Coal* | Firewood | Fuel oil | Gas | Electric |
|----------------------|--------------------------|-------|-------|----------|----------|-----|----------|
| Number of boiler houses | 1010 | 631 | 147 | 33 | 9 | 190 |
| Amount of fuel, thnd TJ | 59,844 | 33,165 | 18,784 | 7,337 | 0,558 | - |
| Amount of substance emitted, tons/year: | | | | | | |
| Dust | 20339 | 19899 | | 440 | | |
| CO (carbon oxide) | 94680 | 89545 | | 5136 | | |
| Soot | 12881,8 | 12603 | | 278,8 | | |
| Benzo(a)pyrene | 4.3132 | 4.311 | | 0.0022 | | |
| Total of 11 PAHs | 85.644 | 85.566 | | 0.0785 | | |

*Coal grades see in Table 2.

In addition to boilers in all cities of the Irkutsk region there is individual heating with firewood and coal. It should be noted that almost every year, many cities in the region are included in the list of priority cities in terms of air pollution. Furthermore, the excess of the B(a)P content makes a significant contribution (sometimes up to 30%) of the total amount of other hazardous substances. According to the state reports of the Ministry of Natural Resources and Environment of the Russian Federation for 2017 [1, 5], eight cities of the Irkutsk region - Angarsk, Bratsk, Zima, Irkutsk, Svirsk, Usolye-Sibirsckoye, Cheremkhovo and Shelekhov - were among the 27 cities in Siberia with a high and very high air pollution level. For the above cities, we systematized the types, the number of “small” boiler houses and individual heating furnaces, and the amount of fuel consumed. The data obtained are shown in Table 8.

The presented data makes it clear that almost all cities are on the list of cities with polluted atmosphere. Whereby, in some of them there are no “small” and individual heat sources. However, high concentrations of B(a)P are constantly observed in the atmosphere of some of these cities [1, 5]. Among them are the cities of Bratsk and Shelekhov where emissions of the aluminum production are great sources of PAHs [16]. The cities of Angarsk, Sayansk, Baikalsk with ecologically clean heating and a minimum of “small” heat sources have relatively low levels of B(a)P in the atmosphere. At the same time, the agricultural cities of Cheremkhovo and Zima, with almost 50% of private heating, reach maximum monthly concentrations of B(a)P in the atmosphere up to 30-40 MACs.

Estimation of the impact incomplete combustion products make on the atmosphere of the Irkutsk region cities was carried out based on the calculated specific emissions of substances and accounting for the amount of fuel consumed (see Table 8). The obtained gross emissions of hazardous substances into the atmosphere of the cities are given in Table 9. A comparison between the contents of incomplete combustion products showed that the concentration of B(a)P is linearly related to the values of CO and soot emissions as well as the total of 11 PAHs. Figure 2 shows an example of the high accuracy of these dependencies for boiler houses. A similar dependency was obtained for household furnaces. Their presence is due to the same nature of the dynamics of the formation of incomplete combustion products (see Table 3 and Figure 1).

Analysis of the data in Table 9 shows that the minimum gross emissions of hazardous substances from “low” power heat sources are found in small - with population of up to 20 thousand people (Sayansk, Baikalsk) - and large industrial cities with population of more than 200 thousand people (Angarsk, Bratsk, Shelekhov). Whereby, the cities of Bratsk and Shelekhov are leaders in terms of the
B(a)P content in the atmosphere, having, for example, maximum monthly average concentrations for 2015 equal to 30 and 9 MACs, respectively.

**Table 8.** Structure of “low” power heat sources and their fuel in large cities of the Irkutsk region (as of December 1, 2017).

| City             | Boiler houses, small | Household furnaces |
|------------------|----------------------|--------------------|
|                  | number | coal, thnd TJ | share of furnace heating, % | coal, thnd TJ | firewood, thnd TJ |
| Irkutsk          | 13     | 0,680         | 5                      | 0,625         | 1,875             |
| Bratsk           | 3      | 0,157         | 3                      | 0,125         | 0,375             |
| Angarsk          | 0      | 0             | 2                      | 0,087         | 0,262             |
| Usolye-Sibirskoye| 4      | 0,210         | 10                     | 0,172         | 0,517             |
| Ust-Ilimsk       | 3      | 0,157         | 2                      | 0,035         | 0,105             |
| Shelekhov        | 1      | 0,052         | 3                      | 0,025         | 0,075             |
| Zima             | 6      | 0,315         | 45                     | 0,267         | 0,800             |
| Cheremkhovo      | 10     | 0,525         | 48                     | 0,550         | 1,650             |
| Sayansk          | 0      | 0             | 0,05                   | 0,0003        | 0,0012            |
| Baykalsk         | 0      | 0             | 5                      | 0,0125        | 0,0375            |
| Total:           | 40     | 2,100         |                        | 1,888         | 5,663             |
| In the region    | 631    | 33,165        |                        | Okolo 40      | -                 |

Note. 1 - boiler houses; 2 - household furnaces.

The maximum amounts of B(a)P, PAHs, CO, and soot are found in the large industrial center, the city of Irkutsk, and in the small agricultural cities of Zima and Cheremkhovo. There, the maximum monthly average concentrations of B(a)P also for 2015 were about 9, 40, and 4 MACs, respectively. Thus, we could clearly see the dependency of gross emissions of incomplete combustion products on the presence and number of “small” heat power engineering sources. Analysis carried out to estimate the correlation of the results of monitoring the B(a)P concentration in the atmosphere of all the cities studied with the mass of its emissions from “small” heat power engineering sources showed a tendency in the average annual and maximum monthly average content. However, no statistically
significant dependency was found due to the fact that we took into account such cities as Bratsk, Shelekhov and Irkutsk where high concentrations of B(a)P are due to other emission sources. Without these cities, for the remaining ones, we found a close statistically direct correlation between atmospheric pollution and gross B(a)P emissions (Figure 3).

**Figure 2.** Correlations between incomplete combustion products in “small” boiler houses.

**Figure 3.** Dependencies of air pollution in the cities of Angarsk, Usolye-Sibirskoye, Ust-Ilimsk, Zima, Sayansk, Baikalsk on gross B(a)P emissions from stratified boilers and household furnaces.
The effect of emissions from “low” power heat sources on the carcinogenic hazard of B(a)P for the atmosphere of the Irkutsk region cities dictates the need to take measures to reduce this impact. It should be noted that work in this direction is being carried out. For example, over the past 20 years, the number of boiler houses has decreased on the territory of the region cities by about five times (to 1005 boiler houses in 2017) [13]. Individual furnace heating has switched to the active use of wood fuel and electric plants. At the same time, the programs [13-15] plan the need to reconstruct the “small” heat supply system by building new heat sources using wood waste (chips, bark, pellets); increase in the number of electric boiler houses and individual electrical plants; technical re-equipment of coal-fired boiler houses into the ones using fuel oil and gas; replacement of boiler houses with manual throwing of coal with modular automated coal boiler houses of Thermorobot type.

4. Conclusion
The main results of the study of stratified burning of coal and firewood in heating boilers and small power household furnaces (up to 1 MW) are the following.

1. We studied the dynamics of emissions of incomplete coal and firewood combustion products in Eastern Siberia from coal-fired and oil-fired boiler houses, and household furnaces with periodic manual throwing of fuel. We also demonstrated the impact of temperature and oxygen, type and method of combustion, design of the combustion device, and fuel composition on the ecological efficiency of combustion.

2. In the paper, we proposed methods for determining specific and gross emissions of the studied substances based on the methodical approach to determining the total emissions during the fuel combustion cycle. We calculated specific emissions of particulate matter, CO, soot, B(a)P and 10 PAHs from “small” coal and fuel oil boiler houses and household furnaces for black (Cheremkhovsky, Tugnuysky) and brown (Azeysky, Mugunsky, Borodinsky) coal, firewood (pine) and fuel oil. It was found that the difference between specific emissions is determined more by the type of fuel (coal, fuel oil, firewood) than by the grade of coal.

3. We presented the structure of boiler houses and the share of furnace heating for the Irkutsk region and its 10 major cities, as well as the types and amount of fuel consumed by them. In the study, we estimated gross emissions of incomplete combustion products in the atmosphere of the cities. It was shown that the maximum emissions of hazardous substances are found in cities with the largest share of furnace heating, and minimal - in cities with central heating, including industrial cities. Direct correlations were found between the B(a)P content in the atmosphere of a number of the cities and the amount of its gross emissions.

4. We identified possible measures to reduce emissions of hazardous and carcinogenic substances from “small” heat sources by optimizing the combustion process (fuel type, coal fractionation, high-quality air supply), upgrading small boiler houses (mechanical supply of fractionated coal), and introducing alternative heat sources and fuel (electric plants, wood waste, gas).

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