Mass fraction of Black Carbon in submicron aerosol as an indicator of influence of smokes from remote forest fires

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Based on many-year round-the-clock measurements at the Aerosol Station of the IAO SB RAS of the scattering coefficient of the dry base of submicron aerosol and Black Carbon (BC) concentration in particles, the influence of forest fire smokes on the variability of the BC fraction in the near-ground aerosol have been analyzed. Stable regularity has been revealed that BC concentration (1–4.5%) in smokes of remote forest fires is lower than in the background aerosol. This causes a decrease of its values, when smoke mists invade into the atmosphere over the measurement site (*P*-effect). It was found from the *in situ* measurements and laboratory experiments that this effect is stable in our region being caused by the decisive contribution of the pyrolysis of forest combustible materials to the formation of the smoke mist particles. It is proposed to take the BC fraction in the submicron aerosol as an information index (*P*-criterion) for distinguishing the state of the ground atmospheric layer affected by the forest fires far removed from the observation site.

Introduction

The single scattering albedo equal to the ratio of the scattering coefficient to the extinction coefficient is an important aerosol characteristic, which takes into account the ratio of scattering and absorption in the radiation transfer. When estimating the role of aerosol in the radiative budget of the atmosphere, the most complicated problems appear because of big uncertainty in the knowledge of the single scattering albedo. Radiative calculations show[1] that if the aerosol single scattering albedo in the visible range exceeds 0.85, then aerosol particles are a cooling factor. In the case of albedo lower than this value, aerosol is a warming factor, like greenhouse gases.

The problem of correctly setting the albedo in radiative calculations is important for a number of serious reasons. Among those there are extremely strong spatiotemporal variability of the aerosol microphysical characteristics and its chemical composition under the effect of natural and anthropogenic factors, highly sophisticated instrumentation needed and, hence, obviously insufficient number of the measurement sites[2] available that causes poor regularity of observations even in the near-ground layer of the atmosphere.

Since 1996, we have been carrying out comprehensive measurements of aerosol characteristics at the Aerosol Monitoring Station of IAO SB RAS in order to study the priority processes determining the formation of the atmospheric aerosol composition, revealing the role of natural and anthropogenic constituents, and the study of the specific sources. One of the factors, which mainly determine the aerosol weather[3] at least on the regional scale, is considered in this paper.

Biomass burning like, for example, forest and peatbog fires[4,5] is one of the main sources of aerosol in warm season, which essentially affects its composition in the regions containing vast woodlands and peatbog areas. These processes cause emission of huge amount of aerosol particles and Black Carbon (BC), or crystal carbon, into the atmosphere.

It is known that black carbon is the main aerosol constituent, which absorbs in the visible range.[6,7] It is also known that smokes of forest and peatbog fires contain relatively small amount of black carbon compared to that in the industrial smokes.[8] But often, keeping in mind the fact of additional emission of black carbon, researchers assume that the value of single scattering albedo should decrease in the atmosphere over vast territories undergone the effect of natural fires. At the same time, measurements at the Aerosol Station show that quite reversed situation is observed in our region in the overwhelming majority of cases.

Obviously, the value of the single scattering albedo of particles depends, first of all, on the ratio of the mass of absorbing substance to the total aerosol mass. It is shown in this paper based on analysis of a great number of smoke events observed that this ratio decreases in the cases when forest fire smokes come to the observation site in contrast to its value characteristic of natural hazes. Moreover, we suggest considering the fact of decreased BC fraction in the total mass of submicron aerosol particles below a certain value, as an indicator of the fact that the atmosphere at this time undergoes the effect of remote natural fires.

Theoretical approach and results of laboratory experiments

Any information index should be based on a stable peculiarity of an object under study providing a preset reliability of its application. In our case, the problem was stated to reveal the aerosol characteristics in the atmosphere, which enable one to isolate the effect of natural fires against the background of all the variety of the states of near-ground aerosol directly for the region under study.

In order to justify using value of the BC fraction in submicron aerosol as a criterion, we made use of the results of our laboratory experiments,[9–12] as well as the data of observations in the near-ground layer of the atmosphere since 1997 until 2005.[13–18]

We have not managed to justify *a priori* the possibility of using this parameter, because its value essentially depends on the regime of burning.

Kinetic theory of formation of black carbon at thermal decomposition of hydrocarbons in the gas phase is developed yet insufficiently due to its complexity. It is known that in this case the rate of BC formation significantly depends on the chemical composition of the material (the C/H ratio), temperature of burning, conditions of mixing the burnt products (surplus of oxygen, turbulence of mixing), and other factors.[19]

Overall, the chains of chemical reactions of oxidation of smoke-generating gases are realized at thermal decomposition of hydrocarbon materials. Smoke aerosol is related to aerosols of condensation origin and is formed at cooling the smoke-generating gases.[20] Chemical reactions in ideal process of burning of hydrocarbon materials (high temperature, oxygen supercharge, recirculation of smoke gases to the area of burning) should lead to complete oxidation of gases with emission into the air of only carbon dioxide and water.

Generation of particles is a sign of incomplete oxidation of gaseous products of burning that usually occurs in real natural and anthropogenic processes. Carbon in gaseous products at the stages of low oxidation is in a bound state in the form of light hydrocarbon compounds. High-temperature regime accelerates the reactions of oxidation in gas phase. Intense generation of crystal carbon in smoke aerosol is characteristic of the stages of high oxidation at burning fuels.

Then, in discussing the experimental data, let us have in mind the assumption that the dynamics of smoke formation at burning natural combustible materials (NCM) agrees with the principles of the theory of generation of BC at thermal decomposition of hydrocarbons in gas phase considered above. In this case it is quite natural to expect that the chemical composition of smoke particles (and the content of BC) even for one type of material changes depending on the degree of oxidation of smoke-generating gases reached in real process, and the degree of oxidation is significantly determined by the temperature regime of burning.

In optical manifestations, the increase of BC fraction in aerosol increases both the absorption coefficient and the refractive index of the smoke particulate matter.

Laboratory investigations carried out in aerosol chambers of different volume (1800 and 0.1 m^3) enabled us to reveal that the regime of burning really is the determining factor of formation of the absorbing properties of smoke particles.[9–12]

Smokes generated while burning wood (pine, birch, needles) were experimentally studied in two regimes: 1) pyrolysis — low-temperature decomposition at 500°C without flame (smoldering); and 2) combustion with flame — high-temperature burning at 900°C with free access of oxygen.

The first part of the experiments was carried out in Big Aerosol Chamber of IAO SB RAS (the volume is 1800 m^3). The following parameters were measured here: scattering coefficients of smoke aerosol at the wavelengths of 0.41, 0.51, and 0.63 μm, and the aerosol extinction coefficient along a 40-m long path at the wavelength of 0.63 μm.[9,10] The results obtained from synchronic measurements enabled us to study the variability of the single scattering albedo of smoke aerosol in the visible region as a function of the regime of burning. Approximate estimates of ω have shown that its value is about 1 for particles generated in the regime of pyrolysis, and about 0.4 for particles generated due to combustion with flame.

More detailed laboratory experiments have been carried out in a special chamber of small volume[11] of 0.1 m^3. Use of a chamber of such a volume enabled us to study the peculiarities of variability of the disperse composition and optical constants of particles depending on the regime of burning. To do this, two orthogonally polarized components of radiation at 10 wavelengths in the range from 0.44 to 0.69 μm scattered at the angles of 15, 45, 110, 135, and 165° were measured by means of a polarization spectronephelometer, and then the inverse problem was solved. The results of investigations have shown that optical constants of the particulate matter increase at passage from pyrolysis to burning with flame. It is observed as an increase of the values of the refractive index of particles *n* from 1.48–1.55 to 1.65. The values of the absorption index of particles χ also essentially increase, from 0.004 to ≈0.05, in passing to burning with flame.

Thus, weakly absorbing particles are generated in the regime of pyrolysis of wood materials, and strongly absorbing particles are generated at combustion with flame. It is clear that these differences in the optical characteristics of smokes of different regimes of burning are caused by difference in the chemical composition and, partially, by the morphology of particles.

An indirect confirmation of the physicochemical differences in the particle properties at different regimes of burning follows from the results of our experiments with active forcing.[9] The impacts were carried out by means of controlled artificial change of

meteorological parameters (humidity, temperature) in airflow containing the aerosol to be analyzed and moving through the scattering volume of the nephelometer. Let us below, in describing these processes, use the terms of direct humidity or temperature impact on aerosol. The idea of the methods of active impacts applied is as follows:

1. One of the ways of actively affecting the aerosol enables one to measure the parameter of condensation activity of particles. To do this, relative humidity of air was gradually increased in the airflow during optical measurements from 20–30 to 95%. The measured optical parameters of submicron aerosol as functions of relative humidity (measurement of a hygrogram) are quite well described, in the first approximation, by the Kasten–Hanel empirical formula[21]:

$$\mu(RH) = \mu(RH = 0)(1 - RH)^{-\gamma},$$

where μ is the coefficient of directed aerosol scattering at the angle of 45° at the wavelength of 0.51 μm, RH is the relative humidity of air, γ is the parameter of condensation activity of particles.

2. Another way for an active impact is the so-called thermooptical method (measurement of thermograms). This method enables one to qualitatively estimate the relative content of substances of different volatility in the aerosol substance[10] while performing controlled heating of the aerosol particles in the temperature range from the temperature of ambient air up to $T = 350$°C.

To realize the regime of active impacts, a FAN nephelometer was equipped with the devices for artificial humidification of aerosol under study up to the values of relative humidity of air of 95% or heating it up to 350°C (Ref. 22). The parameters of condensation activity γ and volatility $\eta =$ $= \mu(25$°C$)/\mu(350$°C$)$ were estimated from the measurement data on hygrograms and thermograms of the coefficient of directed scattering. The selected parameters indirectly characterize the chemical composition of particles, describing their activity in the condensation process and the fraction of volatile species in aerosol.

Analysis has shown that the change of these parameters of smoke aerosol is determined by the regime of burning. The condensation activity of pyrolytic particles is comparatively low, $\gamma = 0$–0.1, and that of particles of flaming combustion is greater, 0.2–0.6. At the same time, strong "burnout" is characteristics of particles of the smoke of pyrolysis at heating up to 350°C, $\eta = 5$–15, while particles produced at burning with flame "burnout" weaker: $\eta < 4$. It follows from these estimates, that pyrolytic smokes contain large fraction (more than 80%) of highly volatile species, and the contribution of low volatile species (more than 60%) is significant in particles of smokes of combustion.

The diagram of correlation of the parameter of condensation activity γ with the parameter of volatility η for the wooden smokes shown in Fig. 1

illustrates the aforementioned stable tendencies in the variability of these parameters depending on the regime of burning. The areas of the values γ and η corresponding to different regimes are distinctly seen in the diagram. Particles of pyrolysis are characterized by high values of the parameter $\eta > 5$, that is evidence of the high content of highly volatile species, and low condensation activity $\gamma = 0$–0.1 (area 2 in Fig. 1).
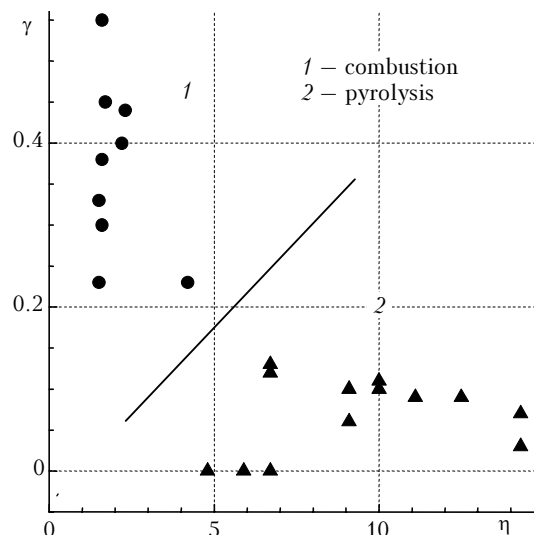

Fig. 1. Diagram of correlation between the parameter of condensation activity of smoke particles γ and the parameter of volatility η for the regimes of combustion and pyrolysis.

Qualitative diagram of formation of the properties of smoke aerosol is shown in Fig. 2 in the form of the diagram describing variability of the refractive index and absorption coefficient depending on the physical processes. In this diagram we have generalized the data of the aforementioned laboratory experiments and the data obtained using kinetic theory of BC generation.
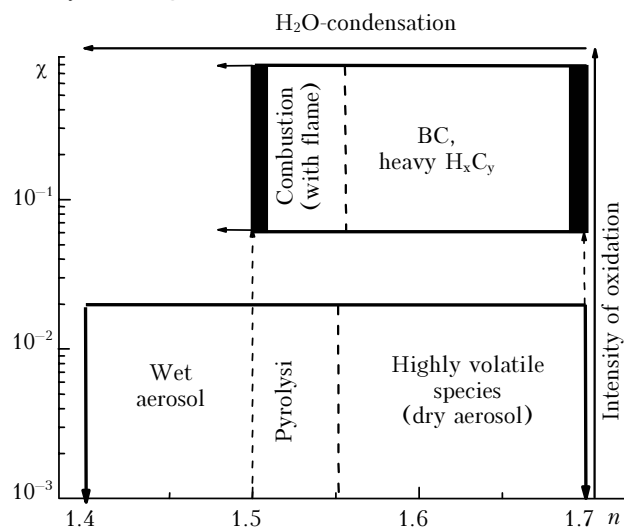

Fig. 2. Diagram of variability of the optical constants of the matter of smoke particles and their relation with physical processes.

Moving along the ordinate axis from low-temperature regimes of flameless pyrolysis to high-temperature combustion with flame (increase of the degree of oxidation) leads to an increase of the BC fraction in particles and, hence, to the increase in the values of the absorption coefficient of particles χ. Condensation growth of particles in the field of humidity (moving to the left along the abscissa axis) leads to a decrease of the refractive index n.

Overall, the obtained results are evidence of the fact that different regimes of burning generate smoke particles of qualitatively different chemical composition. Smoke particles at pyrolysis of wooden materials contain small amount of BC and consist mainly of highly volatile resinous compounds, which actively "burn out" at artificial heating of particles. On the contrary, high degree of oxidation in smokes of combustion with flame leads to generation of particles with high content of BC and small contribution of highly volatile species.

Results of field measurements

Often forest and peatbog fires in Siberia are an important factor determining the composition of atmospheric aerosol in warm season. According to the data of ground-based and satellite measurements, their plumes can cover vast territories. To analyze the effect of natural fires, we used the results of measurements at the Aerosol Monitoring Station of IAO located in the suburbs of Tomsk.[13]

Round-the-clock hourly measurements of the coefficient of directed scattering of the dry matter of submicron aerosol at the angle of $45°$ and wavelength of 0.52 μm with a FAN nephelometer and the mass concentration of BC in particles with the aethalometer[23] are being carried out here in the near-ground layer of atmosphere. Current data of the station are available via Internet at the http://aerosol1.iao.ru web site. The mass concentration of submicron aerosol M_a (μg/m^3) = $= 2400\mu$ was estimated using the data on the coefficient of directed aerosol scattering μ (km$^{-1} \cdot$ sr^{-1}) (using the one-parameter model of atmospheric hazes,[24] for the particle substance density of 1.5 g/cm^3). The value of the BC fraction of the dry matter of submicron aerosol P was determined as the ratio of the mass concentrations of BC and aerosol with the mean error of about 25%.

Analysis of the total data arrays of 1997–2005 has shown that the observed individual values of the mass concentrations of aerosol, BC, and BC fraction varied within the limits 3–570 μg/m^3, 0.1–25 μg/m^3 and 1–20%, respectively. The respective ranges of variations of monthly mean data narrow to the values of 12–95 μg/m^3, 0.7–3 μg/m^3, and 3–14%.

The subarray of data obtained during the periods when the atmosphere underwent the effect of smokes of natural fires was selected from the total data array for further analysis. For convenience of statement, let us call the data obtained in situations

when one can suppose the absence of the effect of the smoke component with high degree of reliability "background" or "natural."[25]

The 9-year mean annual behaviors of the considered aerosol parameters are shown in Fig. 3. The annual behaviors under background conditions are characterized by winter maximum and summer minimum. The amplitude of seasonal variations of the aerosol and BC concentrations are, in average, 2–3 times.
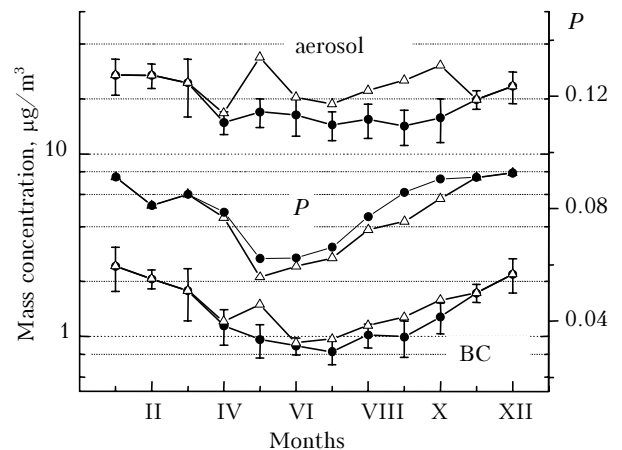

Fig. 3. Mean annual behaviors of the concentrations of aerosol, BC, and BC fraction (1997–2005; (Δ) taking into account fires, ($\bullet$) background aerosol).

Intrusions of air masses with smoke to the region of measurements occurred at fires in Tomsk, Novosibirsk, Omsk and Krasnoyarsk Regions. Long-distance transfer of smokes from fires in Moscow region was observed in September 2002.[26–28] The strongest effect of forest and peatbog fire smokes on the aerosol characteristics was observed in 1997 (September–October), 1999 (July–August), and 2003 (May–October). Increase of the annual mean values of the aerosol mass concentration was 1.6, 1.2, and 1.4 times due to fires relative to the "background" conditions in these years, respectively. The differences in seasonal and daily mean values are essentially greater. The example of the strong effect of fires on the aerosol composition of air is the dense smoke haze of 1997 (with a decrease of the visible range down to 1–2 km), when the monthly mean concentrations of aerosol and BC increased by 7.5 and 1.7 times, respectively.

The effect of fires is usually observed during the period from April until October and reaches its maxima in spring and fall.

It follows from analysis of annual and daily mean data, that the stable tendency toward BC fraction decrease in particles is observed in passing from the "background" to smoke conditions (Figs. 3 and 6).

Hence, the regularity experimentally revealed in our region of decreased fraction of absorbing substance in submicron aerosol at appearing smoke component can be used as an indicator. This value is conventionally called P-criterion.
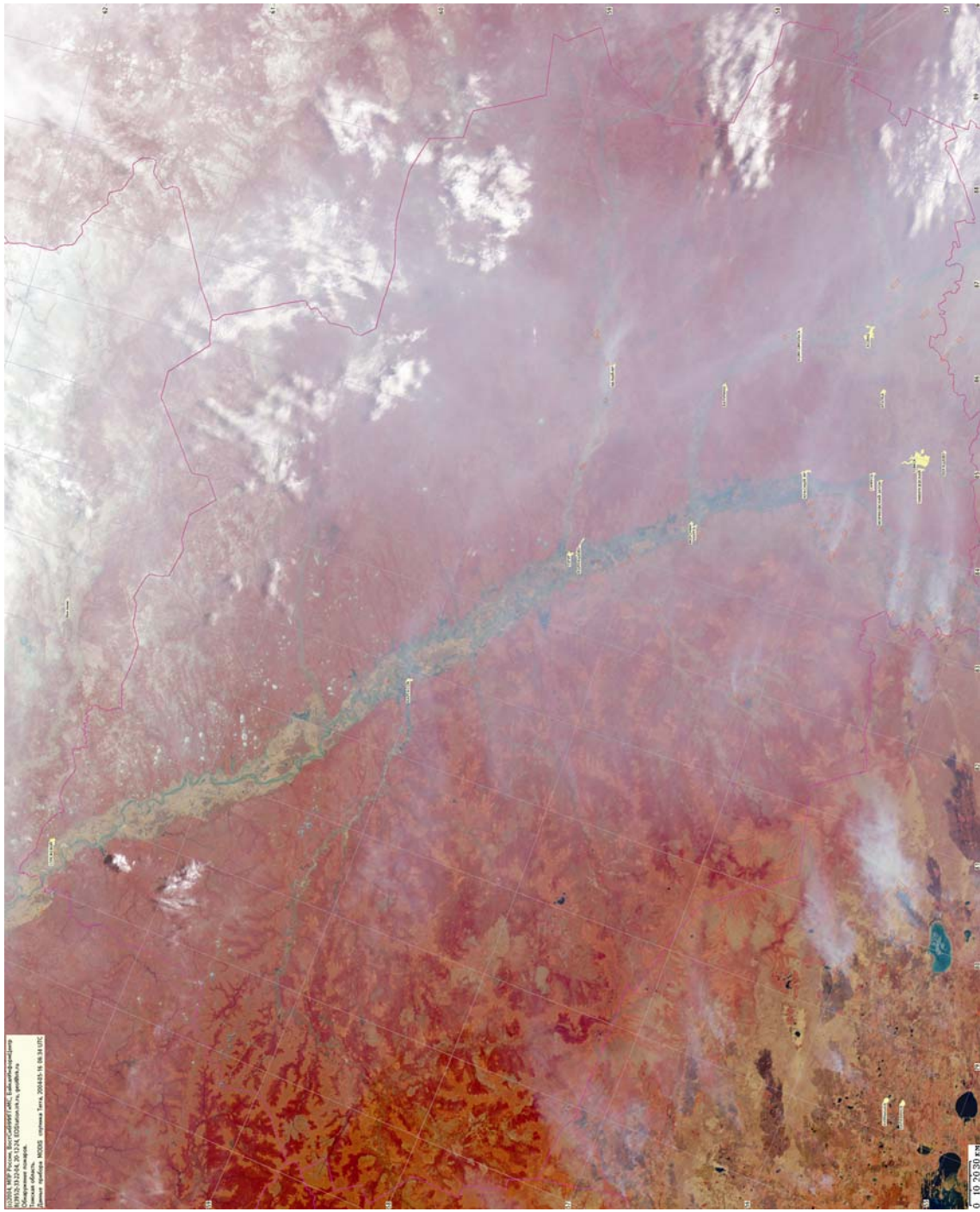
Fig. 4. **Satellite picture of smoke plumes of forest fires (May 16, 2004, Tomsk, MODIS)**

P-criterion

The photo of Tomsk Region obtained from onboard a MODIS satellite in the period of strongly smoked atmosphere on May 16, 2004 is shown in Fig. 4.

It is seen from this photo that a significant part of the region is covered with smoke plumes of forest fires.

Analysis has shown[16] that inflow of air mass heavy smoked to the region of measurements was accompanied by a significant increase of the concentrations of submicron aerosol and BC in the near-ground layer (Fig. 5).
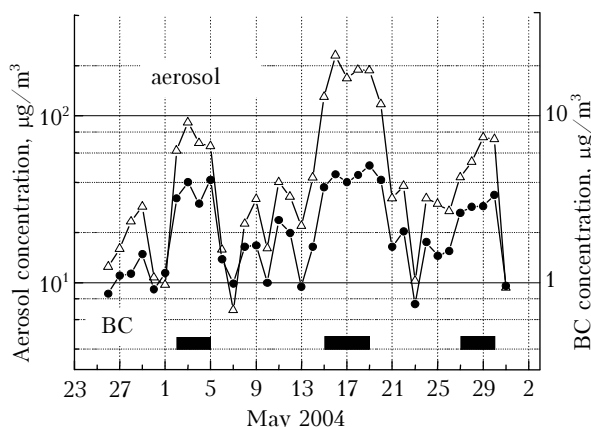
Fig. 5. Temporal behavior of daily mean values of the concentrations of aerosol and BC (smoke haze episodes are marked by lines in the temporal scale).

Three strong invasions of air masses with smoke were observed during this month. The daily mean values of the aerosol ($110-230$ $\mu g/m^3$) and BC ($3.7-5$ $\mu g/m^3$) mass concentrations in smoke mist significantly exceeded the relevant values ($7-16$ and $1-1.4$ $\mu g/m^3$) observed under conditions of weak smoke effect (6 to 9 of May). The maximum hourly values of the concentration in smokes increased up to 460 $\mu g/m^3$ (aerosol) and 9 $\mu g/m^3$ (BC). The strongest smoke mist was observed since May 13 until May 23. In Fig. 5, one can easily see the moments of the mist invasion (2 days), stable state (5 days), and the recovery-destruction stage (4 days). The most important peculiarity of the dynamics of aerosol composition is strong difference in the rates of increase of the concentrations of aerosol and BC. As seen in Fig. 5, the aerosol content increases, on the average, by 10 times, while the BC concentration increases, on the average, by 3 times.

Owing to this regularity, which was observed in all events of the smoke mist invasions observed, the effect of decrease of the BC fraction in particles under the effect of smokes occurred.[14–15] All stages of the development of smoke mist are well seen in Fig. 6.

Analysis of the total 9-year long series of measurements has shown that the daily mean values P in the smokeless (background) atmosphere vary

within the limits of $5-10\%$ about the mean concentrations of aerosol and BC of $10-20$ and $1-2$ $\mu g/m^3$, respectively. The results considered above enable us to conclude that forest fire smokes are characterized by very low BC fraction in submicron particles, essentially lower (by more than 3 times) than its values in the background aerosol in our region.
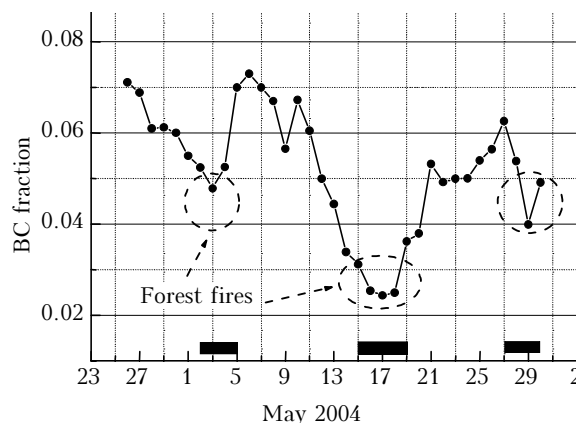
Fig. 6. Temporal behavior of daily mean values of the BC fraction (smoke mist episodes are marked by black bars on the time axis).

Figure 7 illustrates the correlation between the BC fraction of submicron aerosol with the scattering coefficient of the dry matter of particles. Two data arrays have been selected from the 5-year measurement results on P (May–September). Each of the points in Fig. 7 is the result of averaging the values P over 3 or 4 days. Formation of the samples was based on the data obtained from satellite and visual observations. The first data sample (light symbols) presents the background aerosol. The second sample (dark symbols) illustrates the reliably observed events of the effect of forest fire smokes.
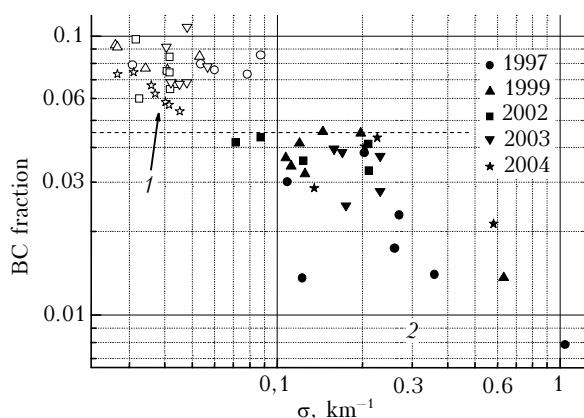
Fig. 7. Diagram of the mutual variations of the BC fraction and the scattering coefficient of the dry matter of aerosol particles under conditions of background atmosphere (*1*) and smoke (*2*) according to the data samples on 1997–2004.

Variations of P for the background aerosol occur within the range of $P = 6-10\%$ (area *1*). The range

of variations of P in the atmosphere with smoke corresponds to the values $P \leq 4.5-5\%$ with the probability of $90-95\%$ (area *2*). These values P can be approximately considered as the level of division between the most probable values P for background and smoke aerosols.

It is natural to assume that the reached lower estimate $P = 0.5-1.5\%$ corresponds to the "true" P value in a dense smoke mist of forest fires.

The considered data of field and laboratory investigations are evidence of the fact that particles in smoke plumes of remote forest fires are characterized by very low values of the BC fraction and, hence, low absorption coefficient of the particle substance, i.e. they are weakly absorbing and, hence, demonstrate obvious properties of particles produced in pyrolysis. Corresponding estimates have shown that high values of the single scattering albedo in the visible range $\omega > 0.95$ are reached in forest fire smokes.[17,29] Hence, forest fire smokes are the cooling factor in radiative-climatic effect.

Obviously, the prevalent role of pyrolysis in the formation of the properties of smoke aerosol is explained, first of all, by higher values of the coefficient of emission of the aerosol mass at pyrolysis compared with the regime of flaming combustion, that was established earlier in laboratory investigations.[9,30] Perhaps, significant part of biomass is oxidized and "burnt out" in gas phase in zones of the forest fire front at flaming combustion.

Additional confirmation of mainly pyrolytic origin of particles in smoke plumes of forest fires are the measured hygrograms and thermograms of submicron aerosol in smoke mist of May 2004 shown in Fig. 8.[16]
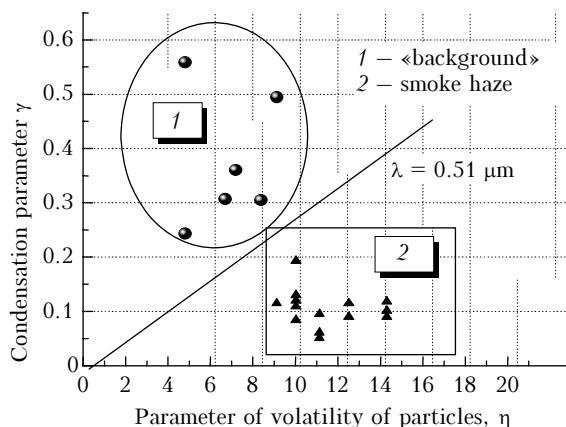

Fig. 8. Diagram of correlation between the parameter of condensation activity γ and parameter of volatility of particles, η, in the smoke mist of forest fires (May 2004).

As seen from the diagram, daily realizations of smokes are compactly grouped (area *2*) separately from realizations under background conditions (area *1*). The smoke particles are characterized by low values of the parameter of condensation activity of $0.05-0.2$ and great content of highly volatile species (the parameter of volatility at heating up to 250°C exceeds 9). These parameters under background conditions vary within the limits of $0.24-0.55$ and less than 9, respectively.

Comparison of the data obtained *in situ* (Fig. 8) with the results of laboratory experiments (see Fig. 1) has shown that, by their properties, particles of real smoke mist correspond to the area of the regime of pyrolysis.

Thus, the obtained results confirm that P-criterion really can be used, at least, in our region, for distinguishing the state of the near-ground layer of the atmosphere, which has undergone the effect of forest fire smokes.

Conclusions

The stable regularity has been revealed of lower BC fraction in the forest fire smokes (1 to 4.5%) compared to that in the background aerosol.

Long-term measurements confirmed that this effect is stably observed in our region and is caused by the dominating contribution of pyrolysis of wooden combustible materials to the formation of properties of smoke mist particles.

It is proposed to use the BC fraction of submicron aerosol as a criterion (at the level of $P \leq 4.5-5\%$) for distinguishing the state of the near-ground layer of the atmosphere, which has undergone the effect of forest fires far from the observation site.

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