Relative Content of Black Carbon in Submicron Aerosol as a Sign of the Effect of Forest Fire Smokes

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Introduction

Biomass burning occurs often in regions containing vast forest tracts and peat-bogs. These processes are accompanied by the emission of aerosol particles and crystal carbon (black carbon [BC], soot).

BC is the predominant source of solar absorption in atmospheric aerosol, which impacts climate. (Jacobson 2001; Rozenberg 1982). In this paper, we analyze the results of laboratory and field investigations that focused on the relative content of BC in aerosol particles. Main attention is given to the study of possibility using this parameter as an informative sign for estimating the effect of remote forest fire smokes on the near-ground aerosol composition.

Theoretical Approach and Previous Laboratory Experiments

Any informative sign for revealing and select the effect of an aerosol should be based on the stable peculiarities of the object of investigation, which would distinguish the appearance of forest fire smoke on the background of a natural aerosol. Finding such signs is possible only by using purposeful laboratory and field measurements (Kozlov et al. 1992, 1996, 1997, 1999, 2002, 2004; Rakhimov et al. 2003; Veretennikov et al. 1980).

Aerosol generation occurs when there is incomplete oxidation of gaseous products of combustion. Carbon in gaseous products at low stages of oxidation is mainly in a combined state of very light hydrocarbon compounds. The high-temperature regime is the catalyst accelerating the oxidizing reactions in the gas phase. Intensive generation of BC (and heavy hydrocarbons) in smoke aerosol is characteristic of the high stages of oxidation during the combustion of fuels. The growth of the BC content in aerosol increases the absorption ability (absorption index) and optical hardness (refractive index) of the smoke particulate matter.

The qualitative diagram showing the formation of the chemical composition of smoke aerosol is shown in Figure 1. It describes the variability of the absorption and refractive indices of the particulate matter being impacted by the physical processes. The data derived from the kinetic theory of BC formation and from experiments are generalized in the diagram. As the degree of oxidation increases (i.e., the transition of low-temperature regime of flameless pyrolysis to high-temperature regime of combustion with flame along the ordinate axis), the BC content in particles increases, and hence, the values of the

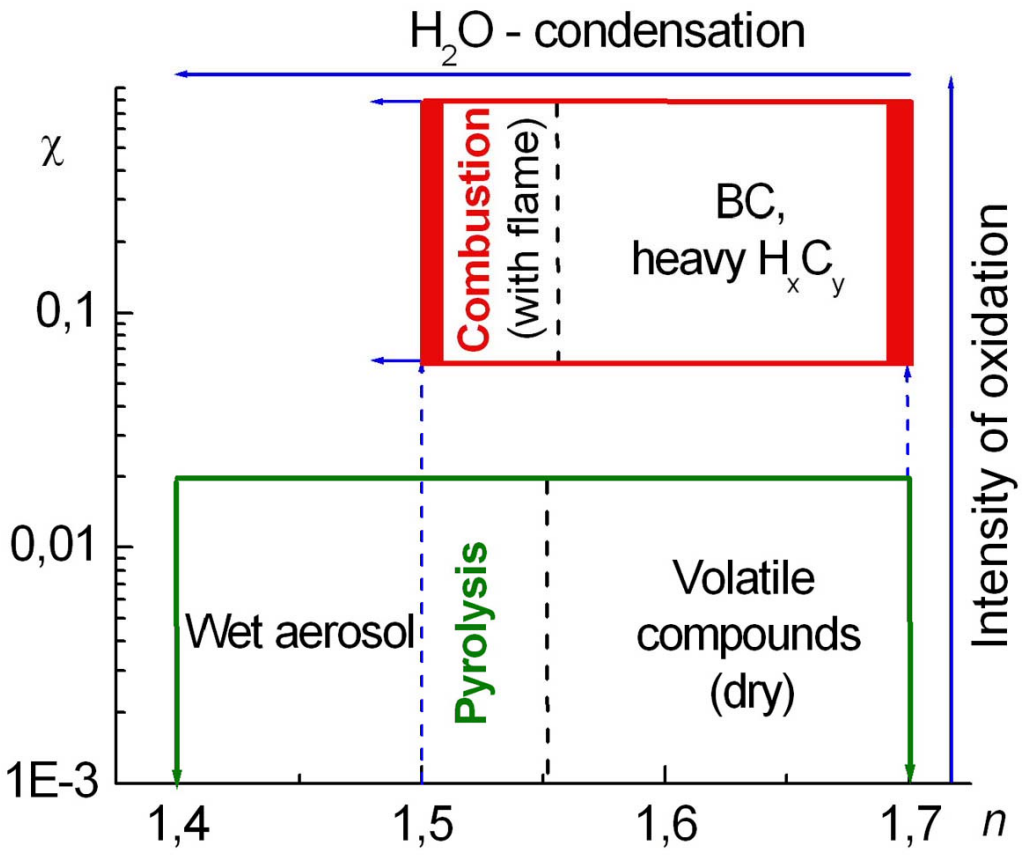

Figure 1. Formation of chemical composition of smoke aerosol.

absorption index of particles increase. Pyrolysis particles have weak absorption, while particles generated during flame combustion are strongly absorbing. Condensation humidification of particles (motion to the left along the abscissa axis) leads to the decrease of the optical hardness of particles; that is, the value of the refractive index n decreases.

Laboratory experiments carried out in chambers at different volumes (1800 and 0.1 m^3) revealed that the regime of smoke generation really is the factor that determines the formation of absorption properties of smoke particles (Kozlov et al. 1992, 1996; Rakhimov et al. 2003; Veretennikov et al. 1980). Smoke generated in two regimes of burning wood was studied. The first regime was pyrolysis low-temperature decomposition at 500°C without a flame (smoldering). The second regime was combustion high-temperature burning at 900°C with a flame having free access to oxygen. The hygrograms and thermograms of the directed aerosol scattering coefficient at an angle of 45° and a wavelength of 0.52 μm were measured using an active flow nephelometer (Zuev et al. 1992). The values of the single-scattering albedo, ω, the parameter of burning-out of particles $\eta = \mu(25°C)/\mu(350°C)$, and the parameter of condensation activity γ (Hanel 1976) were estimated using data the measurements.

A diagram showing the relationship of the parameter of condensation activity γ to the parameter of burn out of particles η (Figure 2) illustrates the stable tendency of variability of these parameters depending

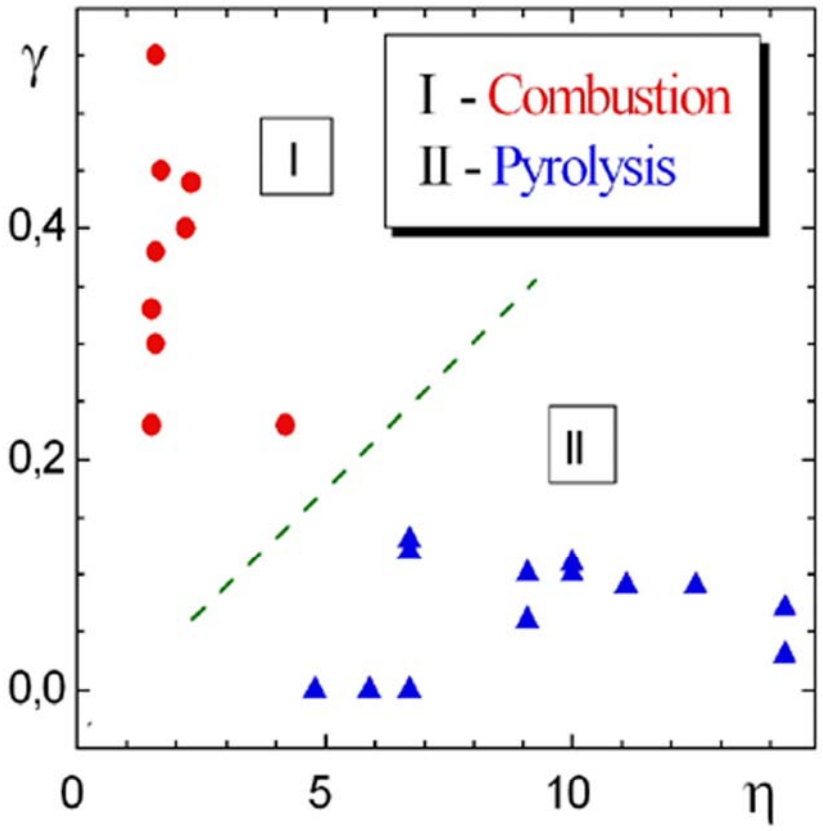

Figure 2. Relationship between combustion and pyrolysis.

on the regime of burning. The areas of the values γ and η corresponding to different regimes are well separated in the diagram. Estimates of the single-scattering albedo have shown that the pyrolysis particles are characterized by the height of its values (ω≈0.95-1), while the absorbing ability of combustion particles of with flame significantly uncreases (ω≈0.4-0.5).

It follows that the particles generated at pyrolysis of wood materials contain little BC and principally consist of volatile resinous compounds, which actively burn out when the particles are artificially heated. However, a high degree of oxidation in combustion smokes with flame leads to the generation of particles with a high content of BC and a small contribution of volatile species.

Results of In Situ Measurements

Fires are common during the warm season in Siberia, which contains great areas of forests and peat-bogs. According to data from ground-based and satellite observations, the forest fire smoke plumes can cover vast areas of the regional scale (Kozlov et al. 1999; Grishin 1992; Sukhinin 2003). They also participate in remote transfer of smoke aerosol (Isakov 2003).

Since 1997, the Institute of Atmospheric Optics (IAO) has conducted round-the-clock aerosol monitoring measurements of the directed scattering coefficient of dry aerosol particles at a 45° angle, a

wavelength of 0.52 μm (nephelometer) and a mass concentration of BC (aethalometer) in the near-ground layer of the atmosphere. The current data are available via internet (http://www.aerosol1.iao.ru). The value of the relative content of BC in the dry matter of submicron aerosol P was determined as the ratio of the mass concentrations of BC and aerosol with mean error of about 20%. The data arrays of annual, seasonal, monthly, and daily mean values were formed from the measurement data, and the peculiarities of the temporal dynamics of the aerosol characteristics were analyzed.

Consideration of the total data arrays for 1997–2004 has shown that the observed hourly values of the mass concentrations of aerosol Ma, crystal carbon MBC and the relative content of BC vary in the limits 3-570 μg/m^3, 0.1-25 μg/m^3 and 1-20%, respectively. The ranges of variations of monthly mean values narrow to the values 12 -95 μg/m^3, 0.7-3 μg/m^3 and 3-14%.

The 8-year mean annual behaviors of the considered aerosol characteristics are shown in Figure 3. The results of averaging of full data (with fires) are marked by triangles. The data arrays, where fires have been removed, are marked by black circles. Annual behaviors of the aerosol characteristics in the smokeless atmosphere are characterized by winter maximum and summer minimum. The amplitude of variations from season to season is 2.5–3 times.

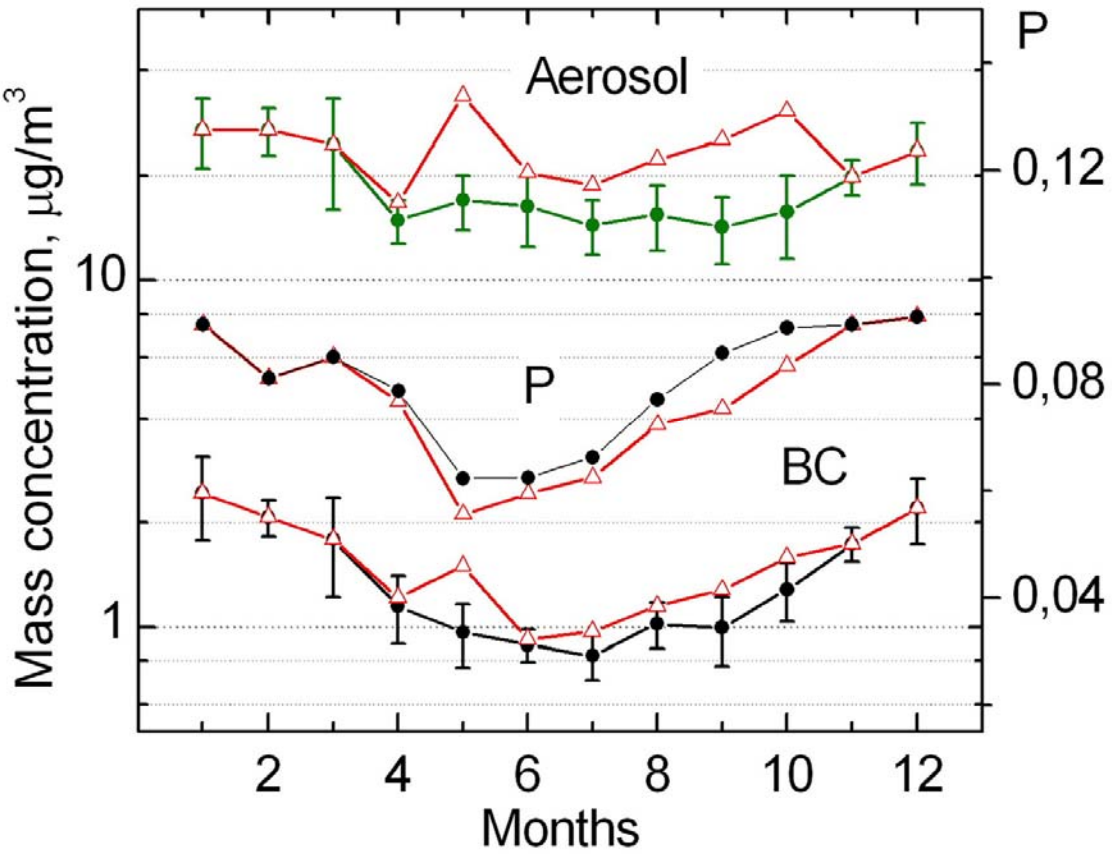

Figure 3. The 8-year mean annual behaviors of the considered aerosol characteristics. The triangles represent the results of averaging of full data (with fires), and black circles represent the data arrays where fires have been removed.

The 8-year mean values P for natural aerosol vary from 6 to 9% (Figure 3). Annual behavior of P is caused by the fact that seasonal dynamics of BC is better pronounced than the aerosol variations, which is determined by seasonal differences of the intensities of the principal processes responsible for the emission of submicron aerosol and BC.

The effect of fires on the contents of aerosol and BC at our measurement site was observed during all years. Intrusions of air masses with smoke to the region of measurements were observed at forest fires occurring in the Tomsk, Novosibirsk, Omsk, and Krasnoyarsk regions. Remote transfer of smoke from fires in the Moscow region was observed in September 2002 (Isakov 2003). The greatest effect of forest and peat-bog fires on aerosol characteristics was observed in 1997 (September–October), 1999 (July–August), and 2003 (May–October). The increase of annual mean values of the aerosol concentration due to fires relatively to natural conditions in these years was 1.6, 1.2, and 1.4 times, respectively. The disturbances of seasonal and monthly mean values are essentially greater. The example of strong effect of fires on the aerosol composition of air was dense smoke haze in 1997 (with the decrease of meteorological visibility range to 1–2 km), when the monthly mean concentrations of aerosol and BC had increased by 7.5 and 1.7 times, respectively.

The effect of fires was observed from April until October, and is maximal in the spring and fall seasons. It follows from the analysis of annual and daily mean data that the stable tendency of decrease of the relative content of BC particles is observed when passing from natural conditions to smoke.

P- effect

The experimentally established feature of the decrease of the relative content of BC in smoke is conventionally called “P-effect.”

The image of the Tomsk region obtained during the strong smoke in the atmosphere on May 16, 2004, by MODIS is shown in Figure 4. A smoke plume from a forest fire covered a significant part of the region. Analysis has shown (Kozlov et al. 2004) that coming of strongly smoked air mass to the region of measurements was accompanied by a significant increase in submicron aerosol and BC concentrations in the near-ground layer (Figure 5). Three strong intrusions 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 $\mu g/m^3$) concentrations significantly exceeded the respective values of the concentrations (7-16 $\mu g/m^3$ and 1-1.4 $\mu g/m^3$) observed under conditions of weak effect of smokes (on May 6–9). The maximum hourly values of the concentrations in smoke increased to 460 $\mu g/m^3$ (aerosol) and 9 $\mu g/m^3$ (BC). The most powerful smoke haze was observed between May 13 and 23. The stages of coming (2 days), stable stay (5 days) and disintegration (4 days) of the haze are well seen in the figure. The most important peculiarity of the dynamics of the aerosol composition is strong difference of the rates of increase of the concentrations of aerosol and BC. As is seen in Figure 5, the aerosol concentration increases, in average, by 10 times, while the BC concentration increases only by 3 times.

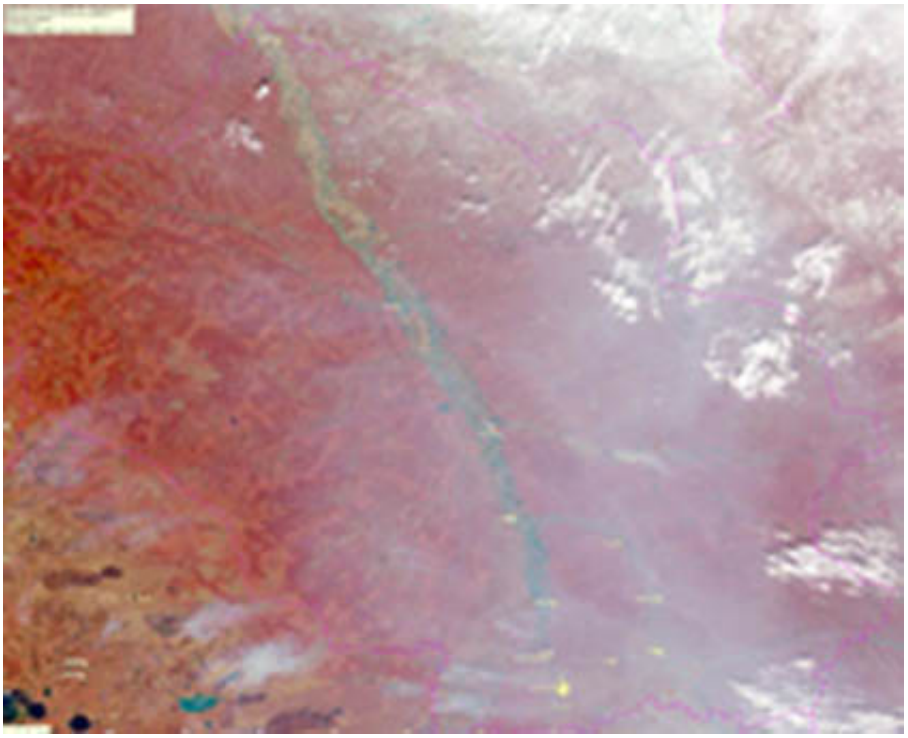

Figure 4. Need Caption.

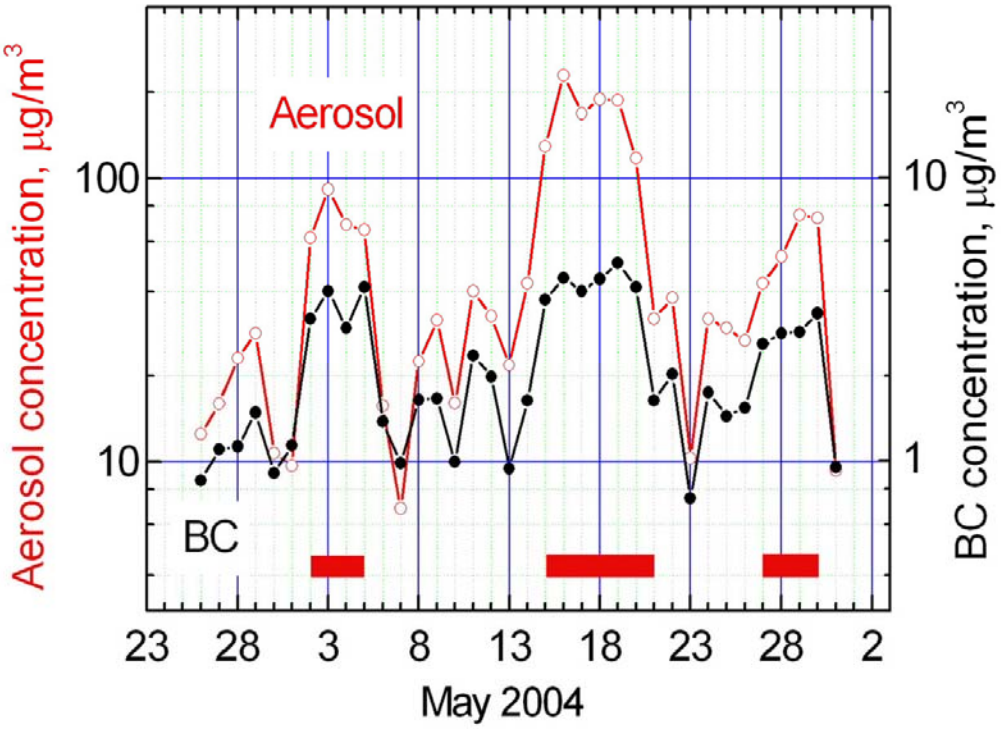

Figure 5. Need Caption.

As a result of this peculiarity, which is right for all observed events of smoke haze, the effect of the decrease of the relative content of BC in particles is observed under the effect of smokes (Kozlov et al. 1997, 2002). All stages of the development of smoke haze are well seen in Figure 6. As the considered haze is set, the values of P decrease, in average, from 7 to 2%. The reached lowest value P is the characteristic of the content of BC directly in smoke haze.

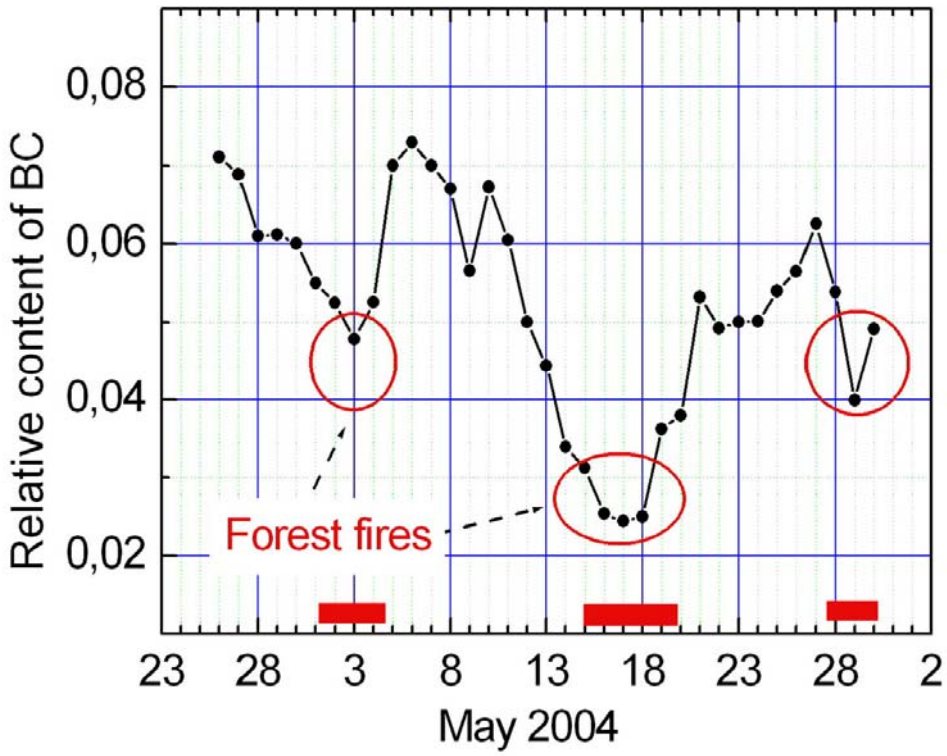

Figure 6. Need Caption.

Analysis of the total 8-year data array has shown that the daily mean values P in warm season in smokeless atmosphere vary in the limits 5–10% at the concentrations of aerosol and soot, in average, 10–20 and 1–2 μg/m^3, respectively. The considered results allow us to conclude for the first time, that the forest fire smokes are characterized by very low relative content of BC in submicron particles, essentially less (more than by 3 times) than its values in natural aerosol.

Figure 7 illustrates the relation of the relative content of BC with the aerosol scattering coefficient of the dry matter of submicron particles. Two data samples resulted from measurements during 5 years (May – September) are shown here. Each of the points is the result of averaging the values P over 3–4 days. Sample formation was based on the satellite data and visual observations. The first sample (rectangles) corresponds to natural aerosol. The second sample (circles) illustrates the reliably recorded cases of the effect of forest fires. Separation of the data by the value of the relative content of BC is well seen in the diagram.

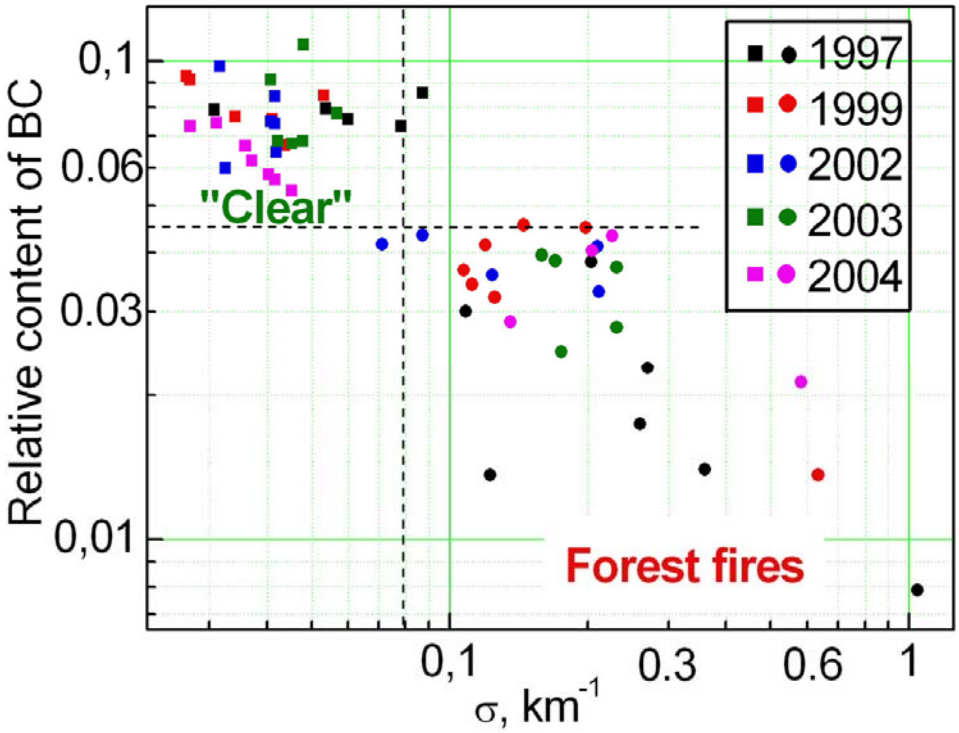

Figure 7. Needs Caption.

The values P of natural aerosol vary in the limits 6–10% (area "Clear"). The range of variations of P in the atmosphere with smoke correspond to the values P ≤ 4.5–5% with probability of about 90–95% (area "Forest fires"). These values P can be approximately considered as indication level of separation of the most probable values P of natural and smoke aerosol. The data shown in Figure 7 confirm that P-effect is the stable feature and are evidence of uniqueness of the effect of forest fire smokes on the value of the relative content of BC in dry submicron aerosol.

Conclusion

The stable feature of the low relative content of BC in forest fire smokes (from 1 to 4.5%) in comparison with natural aerosol is revealed, and it is proved that this fact is caused by the prevalent role of the pyrolysis component in the mass concentration of smoke aerosol.

The conventionally called P-effect lies in the decrease of the values P in conditions of smoke haze relatively its values in natural conditions.

Hence, intrusion of forest and peat-bog fire smokes in the region of observation leads to the increase of the single scattering albedo of submicron aerosol in the visible wavelength range.

The long-term measurements confirm that this effect is stably observed and is caused by the determining effect of the process of pyrolysis of forest combustible materials.

It is supposed to use the relative content of BC in submicron aerosol as an informative sign (by the indication level $P \leq 4.5\text{-}5\%$) for revealing and selection of the effect of remote forest fires.

Acknowledgements

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