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========== **GEOPHYSICS**==========

STUDY OF THE COMPOSITION OF THE ATMOSPHERIC SMOKE SCREEN OVER THE MOSCOW REGION

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In the summer-autumn of 2002, peat bog fires led to long-term large-scale smoke in the Central Region of the Russian Federation. The smoke caused noticeable changes in the radiation budget of the atmosphere and the deterioration of the ecological situation in the region.

In July-September 2002 we carried out complex studies of the dispersed and chemical composition of the smoke aerosols in the surface air layer and in the atmospheric column and measured optical characteristics of the aerosols. The microstructure of the aerosols and the refractive index are determined by light scattering methods. It is shown that peat bogs fires produce smokes with low specific absorption. Survival probability of the quantum for the visible range of the spectrum was 0.95-0.98. Two types of smoke formation were identified in the peat bog fires. Along with the typical for the aerosols formed during the combustion of biomass, the narrow unimodal particle size distributions are relatively wide, including bimodal particle distributions in the range of radii 0.1-0.6 μm. The peat bog smoke is characterized by the relatively high condensation activity of the submicron fraction of the aerosols. Elemental composition of smoke aerosols was determined, including the content of soot and heavy metals, which is substantially transformed by the transfer of the smoked air mass. High concentrations of heavy metals in the smoky atmosphere of Moscow are explained by the effect of urban sources of air pollution.

The purpose of this paper is to study the features of the microstructure (the particle size distribution function) and the absorptivity of peat bog smoke.

The smokiness of the surface atmosphere was determined by the nephelometric method. As shown in [1], the mass concentration M of the submicron aerosols, which dominates in the formation of the radiation properties of smoke [2], is proportional to the scattering coefficient σ. As an example, Fig. 1 presents the results of synchronous measurement of the mass

concentration of a dry part of submicron aerosols in Moscow on the Meteorological Observatory of the Moscow State University and in the Moscow Region at the Zvenigorod Scientific Station (ZSS) of the Institute of Atmospheric Physics (IAP) from 28.08.02 to 10.09.01. The maximum values of M (greater than 2 mg/m^3) were 50-100 times higher than the corresponding background concentrations for the summer season in the Moscow region.

During periods of heavy smoke in July-August, the visibility range $L = 3.9/\varepsilon$, where ε - the extinction coefficient, decreased down to average to 3 km, and in some cases down to 1 km. In September visibility decreased, as a rule, down to about 1 km, and sometimes down to 0.1-0.3 km. At high values of relative air humidity the scattering coefficient σ of the natural aerosols was measured, which determines the value of L and the scattering coefficient σ_d used in the calculation of M.

Aerosol particle size distribution in the surface atmosphere $g(r)$, where r - the particle radius was determined using the light scattering method [3, 4] by measurements of the spectral dependences of the two components of the light scattering matrix [4]. The aerosol particle size distribution function $h(r)$ in the atmospheric column was determined from measurements of spectral transparency [5]. An example of a simultaneous measurement of the aerosol particle volume distributions by size

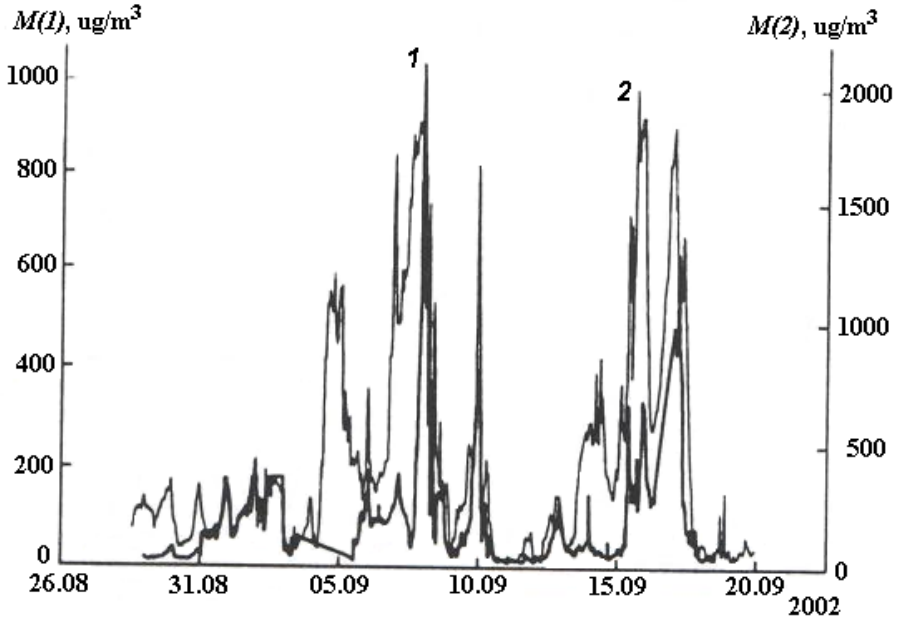

Fig. 1. Time variation of mass concentration of submicron aerosols in Moscow (1) and in the Moscow region (2) in the period from 28.08.02 to 20.09.02.

$v(r) = 4/3\pi r^3 g(r)$ in the surface layer of the atmosphere and the aerosol particle volume distribution by size $w(r) = 4/3\pi r^3 h(r)$ in the atmospheric column in July 30, 2002 shown on Fig. 2. This example indicates the proximity of the microstructure of the submicron smoke aerosols in the surface layer and in the atmospheric column. The microstructure of the submicron aerosols observed in July-August is satisfactorily approximated by the lognormal distribution

$$v(r) = V(2\pi)^{-0.5}(vr)^{-1}\exp\left[\frac{(\ln(r)-\ln(r_0))^2}{2v^2}\right]$$

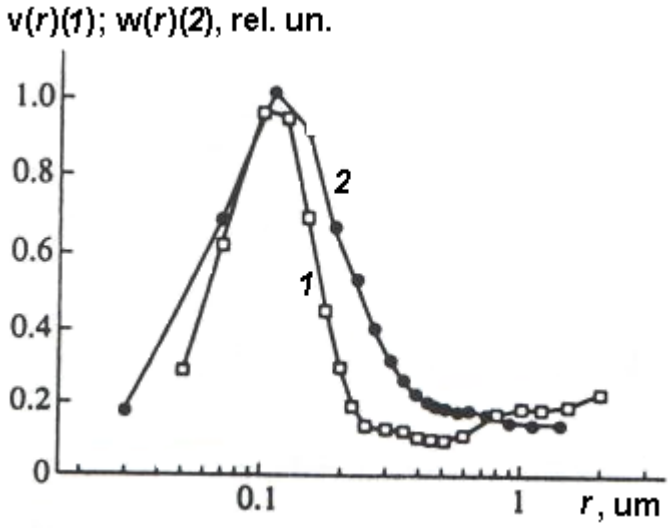

Fig. 2. Volume distribution of aerosols in the surface layer (1) and in the atmospheric column (2) according to the measurements on July 30, 2002.

where r_0 is the median radius of the distribution, V is the total volume of the particles, and v is the parameter characterizing the width of the distribution. Calculations showed that the particle size distribution of smoke aerosols in July-August 2002 turned out to be rather narrow (v = 0.40-0.45). The median distribution radius r_0 varied in the range from 0.05 to 0.12 μm.

The results of determining the microstructure parameters of the submicron smoke aerosols, including (for volume scattering) r_0, v, the refractive index n (real part), which is determined by solving the inverse light scattering problem and the survival probability of the quantum $\Lambda = \sigma/\varepsilon$ are shown in Table 1. In addition to data on the range of measurements of these microphysical parameters of peat bog smokes in July-August 2002 the results of the determination of the parameters of smoke aerosols [6] formed during the burning of some combustible materials [7] (according to complex experiments performed under G.S. Golitsyn in connection with the problem of "nuclear winter"), as well as microphysical parameters of aerosol in forest fires in the Amazon, boreal forests of North America [8] and in savannah fires registered in the network of AERONET stations [9] are shown in Table 1. It can be seen that the microphysical parameters of submicron smoke aerosols generated during peat bog fires (July-August 2002) are quite comparable with the published results for different types of smokes.

The aerosol' absorptivity has a large effect on the radiation effects of the aerosols. It is characterized by the survival probability of the quantum $\Lambda = \sigma(\sigma + \alpha)^{-1}$, where α is the absorption coefficient.

In a smoke the absorption of light in the visible region of the spectrum is due to the soot component of the smoke aerosols. The mass concentration of soot aerosol M_S is regularly measured in Moscow and at the ZSS IAP [10]. An example of the results of day-and-night measurements of M_S in Moscow is shown in Fig. 3. On the example the diurnal course of the M_S clearly manifested itself. Knowing M_S it is easy to estimate the absorption coefficient $\alpha = \zeta \times M_S$, where ζ is the specific absorption of soot aerosol. In the calculation of α, the values of $\zeta = 10$ m^2/g are usually taken.

According to measurements at the ZSS IAP during periods of smoke, the ratio of M_S to the total concentration of submicron aerosols averaged 1.5%. In Moscow, due to additional sources of soot aerosol, this ratio reached about 3%. The relatively low concentrations of soot aerosol suggest that in peat bog fires, the generation of smoke is associated with pyrolysis, and not with burning in the open air.

An estimate of the survival probability of the quantum $\alpha/(\sigma + \alpha)$ yields $\Lambda_1 = 0.95\text{-}0.96$, and the determination of Λ for the entire column of the smoked atmosphere by ratio of direct and diffuse solar radiation fluxes yields the value $\Lambda_2 = 0.97\text{-}0.98$. Note that the difference between Λ_1 and Λ_2 is partly due to the additional soot pollution of the surface layer of the atmosphere by the emissions of automobile engines. Our estimates of Λ in the smoky atmosphere of the Moscow region differ significantly in the values of Λ for the urban aerosol of Beijing [11] (0.65-0.85) and for the smoke aerosols produced by the combustion of biomass [2, 8, 9]. In particular, according to [9], for tropical and boreal forests fires, as well as in savannah fires, Λ varies from 0.86 to 0.94. So, peat bog smoke on average differs by large values of Λ, which leads to a noticeable change in the radiation budget of the smoky atmosphere.

Analysis of the results of a complex experiment on the study of smokes in the Moscow region showed that in September 2002 a change in the smoke production regime occurred so the aerosol particle size distribution function is noticeably broadened. In many cases, the distributions of $g(r)$ and $v(r)$ turned out to be bimodal (Fig. 4), in contrast to unimodal

Table 1. Microphysical parameters of smoke aerosols

Aerosols	r_0, μm	v	n	Λ
Smoke of peat bog fires, summer 2002	0.11-0.20	0.40-0.45	1.40-1.45	0.96
Combustion of biomass:				
tropical forests	0.11-0.17	0.35-0.46	1.44-1.53	0.91
savannah	0.09-0.15	0.45-0.63	1.50-1.52	0.84
boreal forests	0.14-0.18	0.44-0.52	1.46-1.54	0.94
Combustion of material:				
Coniferous wood	0.19-0.25	0.35-0.30	1.50-1.66	0.79
Wood, mixture	0.24-0.27	0.35-0.55	1.62-1.70	0.76
Peat, pyrolysis	0.20-0.25	0.45-0.55	1.68-1.70	0.76

distributions in July-August (Fig. 2). The right-hand side (according to Fig. 1 and Fig. 4) of the distribution v(r) moved to a range of sizes 0.5-0.6 μm and went beyond the submicron range. The change in the smoke generation regime could be affected both by an increase in the relative humidity of air and by change of the mixing regime in the surface layer of the atmosphere.

In most cases, the microstructure of the smoke aerosols in October 2002 is satisfactorily approximated by the sum of two lognormal distributions $v^{(1)}(r)$ and $v^{(2)}(r)$. In particular, for the distribution in Fig. 4, we have $r^{(1)}_0 = 0.2$ μm, $r^{(2)}_0 = 0.4$ μm, $v^{(1)} = 0.40$ and $v^{(2)} = 0.45$. In general, in September 2002, the peat bog smokes differed from the smokes of July-August 2002 and from the typical aerosols produced during the burning of biomass by a significantly larger effective particle size (about 0.3 μm), significantly more wide distribution (v = 0.5-0.6) and large volume

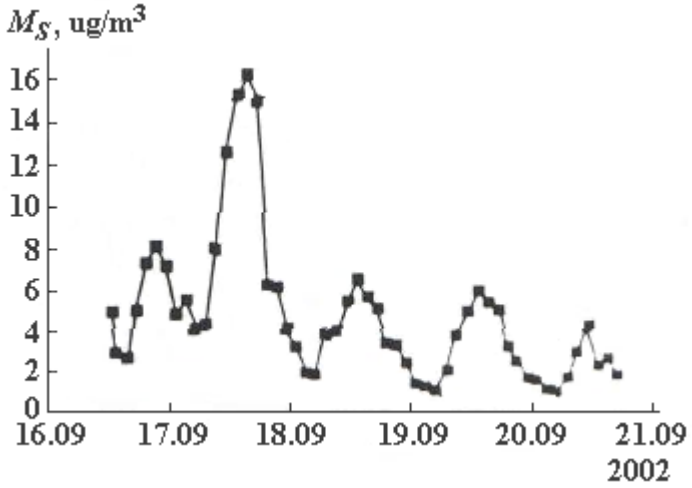

Fig. 3. The time variation of the mass concentration of soot aerosol in Moscow in the period of 16.09.02-21.09.02.

Table 2. Concentrations of chemical elements in atmospheric air (ng/m^3)

Site	Al	Fe	Zn	Cd	Pb	Ni	Co	Cr	Vn
Moscow (amc*), March 2002	510	1060	143	1.1	33	47	0.6	21	-
ZSS IAP, smoke	83	102	82	0.2	5	0.7	0.04	-	-
Shatura district, smoke	515	555	14	0.2	4	1.3	0.2	3	6
Moscow, smoke	4000	5500	690	35	108	45	3	120	25

*adverse meteorological conditions

concentrations. It should also be noted that in September 2002 the particle sizes of the natural (watered) smoke aerosols at high relative air humidity (up to 95%) significantly exceeded (up to 15 times) the particle size of the dry part of the aerosols. The parameter of condensation activity χ [12], which determines the dependence of the scattering coefficient $\sigma(f) = \sigma_d \times (1 - f)^{-\chi}$ on the relative humidity of air f, varies from about 0.2 to 0.3.

Along with the dispersed composition of the smoke aerosols, its chemical composition was studied. The results of the determination of the mass concentrations of certain chemical elements, including heavy metals: zinc, cadmium, lead, nickel, cobalt, chromium and vanadium are shown by the mass spectrometric method [13] in Table 2.

Comparison of the results of measurements of the elemental composition of the smoke aerosols in July-August 2002 in the Shatura district of the Moscow region, in the Moscow region at the ZSS IAP and in Moscow showed that elemental atmospheric air pollution in Moscow conduced mainly due to local sources. This conclusion is confirmed by the results of

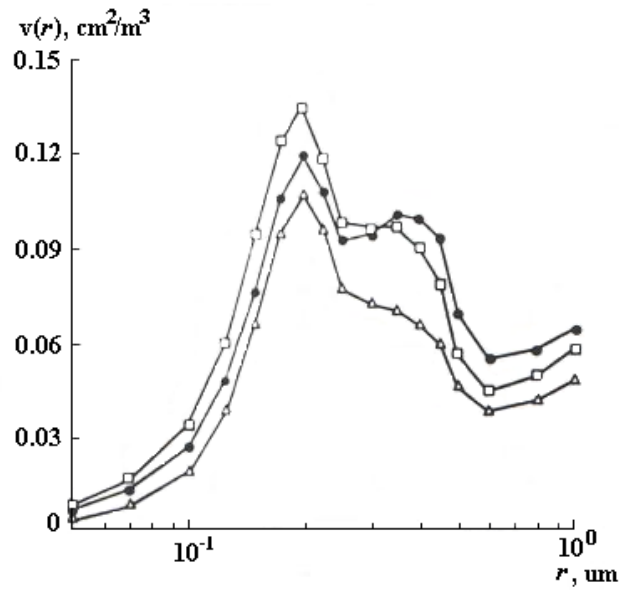

Fig. 4. Variations in the volume distribution by size of the dry part of particles of smoke aerosols in the surface layer of the atmosphere according to the measurements at the ZSS IAP 05.09.02.

measurements in the unpolluted atmosphere of Moscow in March 2002 (Table 2) under adverse meteorological conditions. It should be pointed out that adverse meteorological condition (stability of air masses, weak wind, direction of air transport, etc.) favor in many ways to the prolonged heavy smoke in atmosphere over Moscow. We also note that the mass concentrations of heavy metals given in Table 2 did not exceed its one-time maximum permissible concentrations of the elements.

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