Optical and microphysical characteristics of forest fire smokes in Moscow region in July-September, 2002

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The results of studying smoke aerosols from forest and peatbog fires in Moscow region in summer and fall of 2002 are presented. Spectropolarimetric and nephelometric measurements were carried out at the Zvenigorod scientific station. The spectropolarimeter recorded the polarized components of the near-ground aerosol scattering phase function at three scattering angles $\varphi = 45^\circ$, 90°, and 135° in the wavelength range 0.4–0.76 µm with the spectral resolution $\Delta\lambda \approx 10$ nm. The use of a low-temperature air heater and record of its temperature allow estimating the aerosol condensation activity. The spectropolarimeter provided obtaining data sufficient for solution of the inverse problem of reconstructing the particle size distribution and estimating the complex refractive index of a particulate matter. The automated nephelometer conducted round-the-clock measurements of the directed scattering coefficient $D_{11}(\varphi = 45^{\circ}, \lambda = 0.54 \,\mu\text{m})$ of wet aerosol and its dry matter. More than ten smoke emissions were observed in July - September of 2002, during which the mass concentration of the submicron aerosol was 10 to 50 times higher than the mean background level. Particle size distributions were reconstructed for these episodes and complex refractive indices of a particulate matter were estimated. The characteristic particle size and distribution width of the fire smokes turned to be similar to the pyrolysis smoke studied earlier under laboratory conditions; the real part of the refractive index for natural smokes was noticeably lower ($n \sim 1.4 - 1.5$). Imaginary part of the refractive index in all cases was lower than the resolution threshold, i.e., < 0.005. Contrary to the laboratory smokes, the condensation activity (the Hanel parameter) of natural smokes was high and varied in the range 0.15–0.3, that is rather characteristic of summer aerosol. The particle size distribution in some powerful emissions on September 6–10 and September 17 was bimodal, the characteristic size of the coarse mode was $r = 0.3 - 0.4 \mu m$. The condensation growth factor curve for some smokes was also bimodal with maxima at $r \sim 0.2$ and 0.6–0.8 µm.

Introduction

Anomalously dry and hot summer of 2002 in central Russia and associated forest fires once again underlined the importance of studying the problems of ecological safety in order that, at least, the negative effect of speculations on the problems in mass media will be weakened. At the time of the fires in Moscow region in summer of 1972, almost forgotten by now, the present-day approaches to investigation of trace atmospheric components only have made their appearance, and the instrumentation park was far behind the modern conceptions of organization of observations, so the fires of 1972 turned to be studied only in fragments.

The complex smoke experiments carried out at the Institute of Atmospheric Optics SB RAS (IAO) and the Institute of Atmospheric Physics RAS (IAP) seriously stimulated the study of smoke aerosols as a climate-forming and ecological factor. In recent years, the observations of trace atmospheric components at IAP RAS have been carried out in the monitoring regime. This job, which have been done together with scientists of other institutes,¹ resulted in quite detailed studying of the summer fire smokes of 2002. Favorable for investigations weather conditions well helped to its progress, i.e., numerous smoke emissions in the vicinity of the Zvenigorod Scientific Station and frequent occurrence of anticyclonic sunny days allowed much room for atmospheric observations. About ten strong smoke emissions both in Zvenigorod and Moscow were thoroughly investigated.¹

This paper is devoted to a specific aspect of this activity - a study of smoke aerosol optics and microphysics using the spectropolarimeter and nephelometers designed at IAP. Our earlier experiments (1985-1988) have been conducted in the framework of the "nuclear winter" program, and the character of smokes was dictated by conditions of the Some results were published.^{2–7} problem. А classification of smokes was proposed on the basis of, first of all, analysis of their optical characteristics.³ All smokes were classified into three groups: weakly absorbing (1), coarse and fine dispersed (2), and strongly absorbing (3). Almost all pyrolysis (smoldering) smokes are related to the first group. The second group mainly includes smokes of combustion of wood, peat, etc.; and the third group is presented by oil and rubber combustion smokes ("black" or "blackening").

A comprehensive studies of smokes in a special chamber were carried out at the IAO SB RAS.^{6–8,20} Optical characteristics of smokes produced in different burning regimes were studied in detail by different methods, among them thermooptical. The

resulting classification of smokes agreed, in principle, with the conclusions of Ref. 3. Estimation of the condensation activity (the Hanel parameter χ) has shown that χ for the pyrolysis smokes is quite low, $\chi < 0.05$. In other words, practically hydrophobias smokes were realized under laboratory conditions. Some results of recent laboratory smoke experiments conducted with the help of a spectropolarimeter of new generation, ideologically and methodically close to the spectropolarimeter designed at IAP were presented in Ref. 20. In addition, the results of laboratory experiments with the pyrolysis smokes, in particular, particle size distributions quite close by principal parameters to those given in Ref. 15 were reported.

Instrumentation

Recall briefly principal parameters of the instruments. The spectropolarimeter records polarized components of the scattering phase function of the near-ground aerosol at three scattering angles ($\varphi = 45^{\circ}$, 90°, and 135°) in the wavelength range from 0.4 to 0.76 μm with the spectral resolution grid monochromator $\Delta\lambda \approx 10 \text{ nm}.$ The based illuminator of the device allows quasi-continuous scanning of the spectrum with a controllable step. The working volume at the airflow velocity of 0.5 m/s and digital averaging for about a second is approximately 1 liter. A low-temperature heater of the sampled air and the control for its temperature permit estimating the aerosol condensation activity. The control for the instrument and data storage are computerized. The spectropolarimeter provided for information sufficient for setting the complex inverse problem of reconstruction of the particle size distribution (the most visual representation is the particle dV(r)/drvolume distribution and estimation of the real and imaginary parts of the refractive index of a particulate matter. The length of one measurement cycle of the device is about 15 minutes, it usually operates at day time. Up to 15 series of records per day were made depending on the development of situation during smoke emissions. The automated nephelometer recorded hourly values of the directed scattering coefficient of dry and wet aerosol at $\lambda = 0.54 \,\mu\text{m}$ and the scattering angle $\varphi = 45^{\circ}$. This parameter enables one to determine the mass concentration M of the submicron aerosol. From the beginning of September, the measurements with automated nephelometer, similar an to the Zvenigorod one, were renewed at the meteorological observatory of the Geographical Faculty of Moscow State University at Vorobjevy Gory.

In connection with participation of IAP in many-year monitoring programs, the task of calibration of the instrumentation developed in the Institute became urgent. Since most devices and methods were unique (the prototypes that can be considered as standard were absent), a partial solution of the problem could be the calibration of one or two basic characteristics of some device through their comparison in one or another way with the same characteristics of a similar certified device. Just in this way, the nephelometers and spectropolarimeter were calibrated by a FAN production-type nephelometer at $\lambda = 0.52 \ \mu m$ and $\varphi = 45^{\circ}$. The length of the cycle of simultaneous measurements was about a month, the range of variations of the measured values was D = 0.003 - $0.3 \text{ km}^{-1} \cdot \text{sr}^{-1}$, the standard deviation from the calculated regression line was 3%. In fact, it was the maximal accuracy that could be expected from synchronous measurements with the two nephelometers. Reaching a higher accuracy was of the impossible because spatial-temporal inhomogeneity of aerosol. Spectral calibration of the spectropolarimeter was performed using a well-known standard screen covered through deposition with a fresh thick layer of magnesium oxide.

Methods of solving the inverse problem for the optical characteristics, presented in this paper, have been formed gradually, and their different aspects were described in Refs. 12, 13, and 21. A set of 120 values (20 wavelengths × 6 polarized components) resulted from the measurements for each realization. For convenience of solving the inverse problem, a smoothed curve of 8 points in the wavelength range of $0.4-0.75 \,\mu\text{m}$ with a step of $0.05 \,\mu\text{m}$ was constructed by the least square method for each polarized component. A surplus of input data leads only to disturbance in the solution, ¹⁹ therefore, the final vector of input data was represented only by 24 informative components.

The errors in the reconstruction present a particular problem. In most cases the measurement errors are always a combination of noise deviations and strongly correlated errors in calibration of the devices. Their relative contributions (at least, for optical investigations of the atmosphere) depend on particular situation and are practically a inseparable. Therefore, even in the optimal situation (the accuracy of measurements is 5-10%) we hardly can expect an accuracy greater than 15-20% in reconstruction of distributions even with $r \sim 0.2-0.5 \ \mu m$, and it is impossible to point out the error for an individual realization.

The dependence of information capacity of input data on the particle size is determined by the product of slowly changing sensitivity of the equation kernel by the quickly decreasing sought distribution, and the decrease rate can strongly change (from three to five orders of magnitude for a size decade). In other words, the range of the solution applicability can change 2-3 times for some device depending on a particular situation.

When improving the procedure of inversion, a method was found¹³ of automated semi-quantitative estimation of the boundary $r_{\rm max}$ in each situation. It is as follows. The boundaries of the area of definition must be selected in such a way that the obtained solution tends to zero. Of course, this does not mean

(relative to aerosol) the absence of such particles, but only points to the absence of information on the particles in input data, which allows us to consider their concentration negligible without any loss in the accuracy of reconstruction. To check this condition, the iteration correction at each step is multiplied by the function equal to unit in practically all range of definition except for the areas close to its boundaries, where it smoothly decreases to $\delta \sim 0.95$.

Thus, if there is no noticeable contribution of particles with sizes close to the boundary of the range in the input data, the obtained solution after fifty iterations shows a well pronounced tendency to zero. was checked mathematically, that if the It contribution of large particles exceeds the noise, it is impossible to tend the solution to zero by such a "forcing." In other words, if noticeable concentrations of large particles result from application of the procedure, they are really present in the working chamber of the device. Analysis of a distributions reconstructed for great number of various situations has shown that in the majority of events the dV(r)/dr solution can not be decreased to zero at sizes up to $r = 2 \mu m$, but the stability of solution at $r > 1.5 \ \mu m$ is low, i.e., the device sees such particles, but the received information is very noised. This also confirms that the maximal working wavelength of the device is not so important for the inverse problem solution as a statistically great body of large particles in the working volume during the time of averaging (digitizing).

Generally, the algorithm of solving the problem is as follows. The grid of the integral equation kernels is calculated for a set of values of real and imaginary parts of the refractive index. The inverse problem is solved for each kernel by the iteration method, and the most likely values of real and imaginary parts of the refractive index are determined from the minimal residual for reconstructed optics. When developing the solving procedure, we took the following parameters: the grid consisted of $m \times k = 45$ kernels, m was equal to 9 values of the real part of the refractive index in the range n = 1.35 - 1.59 with $\Delta n = 0.03$ at k equal to 5 values of imaginary part in the range x = 0-0.02with the step of 0.005. The range of r variation was from 0.05 to 2 µm (twenty points). Taking into account all above-said, the actual range was a bit less, i.e., in practice, the reconstructed distributions usually did not exceed $r = 1.5 \,\mu\text{m}$.

Discussion of results

Observations of smoke emissions in Moscow region were carried out at Zvenigorod Scientific Station (ZSS) of IAP RAS in July–September of 2002. The distance from the emission sources to Zvenigorod was ~ 100–200 km, and, taking into account small wind velocity in anticyclone, the lifetime of smokes at the moment of measurements could be estimated as one day or more, that turned to be important upon two counts. First, one day is quite sufficient for good mixing of smokes, so fluctuations of the recorded signals were quite acceptable, 5-10%. Second, due to condensation of aerosol-producing vapor, smoke particles had a time to be covered with a "shirt" of soluble substances characteristic of usual aerosol, that affected the values of the refractive index of the matter of particles and their condensation activity.

Earlier, it has been shown¹⁸ that the spectral dependences of $D_{11}(\varphi = 45^{\circ})$ of natural aerosol in the wavelength range of 0.4–0.65 µm are well approximated by the inverse power law (Angström formula) with the coefficient α called the Angström parameter. This approximation is applicable to almost all events of smoke emissions. Only in the most powerful emissions on September 8-10 and September 17 the spectral dependences $D_{11}(\varphi = 45^{\circ})$ turned to be convex, i.e., the range of applicability of the approximation was somewhat less. Variations of α were closely related to $D_{11}(\varphi = 45^{\circ})$. The correlation coefficient R of these parameters for the entire period was equal to 0.7, the value of α for smokes changed in the range 0.5-1.7. A steep at times spectral behavior of D was first of all due to narrow limits of the smoke particle size distribution.

The results of reconstruction of microphysical characteristics of some smokes studied in the late 80s at the ZSS IAP during smoke laboratory experiments are presented in Ref. 15. The smokes simulate peatbog, forest, and urban fires. It is assumed that the main mechanism of smoke formation (from the standpoint of aerosol generation) in forest and peatbog fires is pyrolysis. Basic characteristics of the pyrolysis smokes are the following: the principal part of the distribution is well approximated by the narrow one-mode distribution with a maximum in the size range 0.2-0.25 µm and the variance of about 0.1; real part of the refractive index $n \sim 1.6-1.73$, imaginary part is close to zero. Note that forest fires are common in central Russia. Before analyzing in detail the results of 2002, we consider only one episode. In August 1999, there was a forest fire in Tver Region, and the smoke emission was observed in Zvenigorod on August 6. The power of the emission was similar to the power of the first fire episode in July 2002. That episode could not be representative because of its short length. The resulting one-mode distribution, as well as parameters of the approximating lognormal distribution: $r = 0.19 \,\mu\text{m}$, variance of 0.2, absorption less than minimally resolved, and the refractive index n = 1.53 well met the scheme.¹⁵

During the summer of 2002, we had a possibility to study forest fires *in situ* for a long time and to compare laboratory and field investigations, having in hand a great body of data. A series of intermediate peaks of a less amplitude are seen in the time dependence of the near-ground aerosol mass concentration (Fig. 1) between most powerful emissions. These results were determined from the automated nephelometer data. Note that hourly readings not always show maximal values of the directed scattering coefficient.

Primary analysis of the data²¹ was carried out immediately after the smoke emissions, therefore only most powerful of them have been analyzed. Based on the episodes of the most strong smoke-screens, a conclusion was drawn that the particle second mode appears during the most vigorous of them, because just the coagulation and particle sizes determine the rate of Brownian coagulation. Somewhat more thorough analysis of the period from July 20 to September 18 made us to revise the conception. It is not necessary to prove that variations of natural aerosol are significantly controlled by warm and cold fronts of cyclones, because a front passing results in a change of air mass and, hence, of aerosol. So, they dictated the regime of smoke-screening in Moscow region to a large extent, and not only through a change of wind direction from the smoke source. The moments of front passing were determined by the character of changing pressure, wind direction, type of cloudiness, and so on. The process of evolution of strong emissions usually was the same: warm front of the cyclone – peak of smoke – cold front – purification of air. The moments of passing of atmospheric fronts are shown in Fig. 1 by arrows. The scattering level in the morning maxima of D_{11} between the most powerful peaks changed within the range $0.05-0.12 \text{ km}^{-1} \cdot \text{sr}^{-1}$. A well pronounced diurnal behavior of the mass concentration Mconnected with the nighttime smoke accumulation under the near-ground temperature inversion and noon convective emission was observed; the relative humidity retained low.



Fig. 1. Temporal behavior of mass concentration of the submicron aerosol from July 23 to September 25, 2002. Arrows in the figure mark the moments of passing the warm (upward arrows) and cold (downward arrows) cyclonic fronts.

The results of reconstruction of microphysical characteristics of smokes observed in summer of 2002 are shown in Figs. 2a-d.

Let us analyze data in Table. In the beginning of the period July 30 – September 5, the modal radii and variances of approximating lognormal distributions as well as almost zero values of imaginary part of the refractive index are close to the laboratory values. The real part of the refractive index of natural smokes turned to be noticeably less: n = 1.4-1.5. Figure 2*a* presents volume distributions dV(r)/dr for two days prior to the first emission for July 23 and 24 (left axis), which can be considered as a background, and those of the first series of emissions for July, 30 - 31(right axis). Transformation of distributions at transition from background to smoke occurs first of all due to increase of the median size and narrowing the distribution. In the background situation $r_{\rm m} = 0.11$ and variance is 0.3. In the first period of smoke emissions except for August, 25 (i.e., ten days of smoke haze until September 6), all distributions are one-mode, there is only a hint at the second mode (see Figs. 2a and b).

A well pronounced second mode is observed for the first time on August 25 in the warm sector of cyclone. Starting from September 6, it is seen in all distributions dV(r)/dr (Figs. 2c and d). Sometimes its amplitude is comparable with that of the first mode. The position of its maximum varies in the range 0.3–0.4 µm, sometimes both modes are combined into one wide mode. And, finally, the last episode on September 16-17 (Fig. 2d) was observed once again in the warm sector of the cyclone with rain. It may be concluded that the factors, at least, accompanying the appearance of the second mode, were the occurrence of Moscow region in the warm sector of the cyclone with rains and high relative humidity of air.

The Hanel formula is known already for 30 years. It links together the scattering coefficient σ of the atmospheric aerosol and the relative humidity of air *Rh*:

$$\sigma = \sigma_0 \left(1 - Rh \right)^{-\chi}, \tag{1}$$

where σ_0 is the scattering coefficient of dry particles, χ is the Hanel parameter interpreted as the parameter of aerosol condensation activity. When applying a spectropolarimeter, we have to use the "short" method for estimating χ from two *Rh* values. If $D_{1,2}$ were measured at the relative humidities *Rh*_{1,2}, then

$$\chi = \ln(D_1/D_2) / \ln[(1 - Rh_2) / (1 - Rh_1)]. \quad (2)$$

Relative humidity of air was low due to drought (30–50%), therefore, χ could be obtained only for situations when the initial humidity *Rh* exceeded 60%. Air was heated by approximately 6–8°C, so in conditions of high summer temperatures the aerosol drying was only partial. The conclusion about a low condensation activity of pyrolysis smokes ($\chi < 0.05$) was drawn from the results of complex investigations of smokes carried out at IAO SB RAS (Refs. 6–8). On the contrary, summer smokes of 2002 (Table) turned to be rather hygroscopic, the value of the Hanel parameter for them corresponds sooner to natural summer aerosol.¹⁷



Fig. 2. Particle volume distributions dV(r)/dr observed in Zvenigorod in July – September 2002. One-mode distributions: situation close to background on July 23–24 (curves 1,2 left ordinate), the first (July 30, curve 3), and the second (July 31, curve 4, right ordinate) forest fire smoke emissions (*a*); volume distributions dV(r)/dr in the dense smoke emission on September 2 (*b*). Bimodal distributions: emission on September 10 (*c*), emission on September 17 (*d*). The time of record shown in the parenthesis enables one to estimate the characteristic length of development of situations.

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Date	$D_{ m max},\ { m km}^1\cdot{ m sr}^{-1}$	< < >	Rh	$r_{\rm max}, \ \mu { m m}$	<n></n>	II mode	<χ>
07.30	0.08	1.7	0.4	0.13	1.59	-	_
07.31	0.125	1.2	0.5	0.18	1.47	—	—
08.15	0.2	1.5	0.5	0.2	1.41	—	—
08.25	0.08	1.1	0.92	0.23	1.38	+	0.2
08.30	0.1	1.2	0.6	0.18	1.47	—	—
09.01	0.05	1.5	0.4	0.18	1.47	—	—
09.02	0.16	1.3	0.7	0.2	1.47	_	—
09.03	0.11	1.35	0.8	0.15	1.47	—	0.17
09.04	0.09	1.5	0.6	0.15	1.53	_	0.2
09.05	0.17	1.15	0.55	0.18	1.47	—	—
09.06	0.12	0.7	0.84	0.18	1.41	+	0.18
09.07	0.07	0.9	0.6	0.18	1.41	+	0.1
09.08	0.24	0.9	0.8	0.18	1.41	+	0.1
09.09	0.05	1.2	0.75	0.16	1.44	+	0.32
09.10	0.45	0.6	0.8	0.22	1.41	+	0.19
09.17	0.25	0.45	0.95	0.22	1.38	+	0.17

Tabla

N o t e. Basic characteristics of smoke emissions are presented in Table: the value of the directed scattering coefficient $D(\varphi = 45^{\circ}, \lambda = 0.54 \ \mu\text{m})$ in the peak of smoke obtained from spectropolarimetric measurements, the Angström index α averaged over several realizations; the relative humidity of air Rh at the moment of measurements, the radius of the maximum of the corresponding distribution dV(r)/dr; the refractive index of dried smoke n, the presence (+) or absence (-) of the second maximum of distribution; and the Hanel parameter for this series of records.

With the particle size distributions for wet and dry smoke aerosol in hand, it becomes possible to estimate the condensation growth factor of particles using the technique proposed by A.G. Laktionov.²² Density variations of the particle size distribution $\zeta(r)$ at a change of relative humidity of air are equivalent to changes of the radius axis scale by the law r' = h(r) on retention of the total number density

$$\eta = \int_{a}^{b} \zeta(r) \mathrm{d}r = \eta' = \int_{h(a)}^{h(b)} \zeta(h) \mathrm{d}h \,. \tag{3}$$

In the process of condensation transformation particles pass from the size range [a, b] to the range [h(a), h(b)]. Considering η and η' as functions of the upper limit of integration, from the condition $\eta = \eta'$ one can determine the dependence h(r) defined in Ref. 22 as a particle growth factor. The problem is reduced to determining the upper limit of the integral r' = h(r) from its value under the condition $\eta = \eta'$. In our opinion, it is more reasonable to determine h(r) from the smoothed curves using, for example, polynomial interpolation by the least square method (LSM). Since the argument and the function in LSM are readily replaceable (constructing the inverse function), it is easy to determine h(r). Such estimates of the natural aerosol growth factor are presented in Ref. 10. It is shown there that the usual dependence of the growth factor on the particle radius has a shape of bell with the maximum at $r \sim 0.3-0.5 \,\mu\text{m}$. There is no reason to assume different chemical compositions for smoke particles of different sizes, therefore, one can expect the growth factor of smoke particles to be close to a Π -shape. Such a shape was observed in two cases, on August 28 and September 6: wide maximum in the range of $r = 0.2-0.8 \,\mu\text{m}$. However, the growth factor curve for smokes on September 17 was bimodal (Fig. 3). Perhaps, it is somehow connected with bimodality of the particle size distribution curve (Fig. 2*d*).



Fig. 3. Condensation growth factor of particles as a function of the particle radius for two first distributions shown in Fig. 2d.

Conclusions

1. Smokes of forest and peatbog fires have been studied for about two months at Zvenigorod Scientific Station in the Moscow region. Time behaviors of the Angström parameter were obtained for the directed scattering coefficient D_{11} and mass concentration of dry submicron aerosol. A high correlation between these two values was revealed (R = 0.7). During smoke haze peaks, the mass concentration in Zvenigorod was 10–50 times higher than the summer background level; in Moscow (nearby Vorobjevy Gory) it was approximately 100 times higher.

2. Optical and microphysical characteristics of real smokes of forest and peatbog fires in summer of 2002 turned to be close to those obtained under laboratory conditions, the differences are sooner due to the smoke age. They are the following:

a) median radii of approximating distributions are somewhat less and vary in the range of $r_m = 0.16-0.21 \,\mu\text{m}$; r_m usually increases as smoke turbidity increases;

b) values of real part of the refractive index of natural smoke matters are noticeably less, n = 1.41-1.51, obviously, due to condensation of vapor of the aerosol producing compounds. Upon the same reason, the condensation activity of natural smoke particles is essentially higher ($\chi \approx 0.15-0.3$) than that of laboratory ones.

The last by time smoke episodes have demonstrated a presence of the second mode in the distribution dV/dr for $r \sim 0.4 \mu m$.

Estimates of the condensation growth factor of smoke particles were obtained for several smoke episodes, for some of them the curves were bimodal.

Acknowledgments

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