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Long-Term Investigations of the Spatiotemporal Variability of Black Carbon and Aerosol Concentrations in the Troposphere of West Siberia and Russian Subarctic

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Abstract

The results of aircraft and near-ground investigations of the spatiotemporal variability of aerosol and black carbon (BC) concentrations in the troposphere for the last two decades are reviewed. Since 1999, the airborne sensing of vertical profiles of BC and aerosol concentrations in troposphere up to a height of 7 km in the regions of West Siberia and Russian Subarctic had been carried out onboard flying laboratories. Since 1997, the Aerosol Station in Tomsk conducts monitoring measurements of the aerosol and BC concentrations in the surface layer.

The data of airborne sensing were used for analysis of common and distinctive features of the spatial variability of vertical profiles of aerosol and BC concentrations, BC fraction, single scattering albedo (SSA) in the visible. The integral BC concentration and aerosol optical thickness in the column of the atmosphere were obtained for the first time. The classifications of typical optical and microphysical states of the subarctic troposphere have been performed for: 1) high transparency of air in the polar latitudes; 2) strong impact of Siberian forest fires in warm season; 3) conditions of regional average background atmosphere in the middle latitudes. As the latitude increases in the range of 55–75.2°N, the near-ground aerosol and BC concentrations decrease threefold, on average. In the subpolar latitudes, a tendency to decrease of the concentrations in the direction from the west (Kara Sea) to the east (Eastern Subarctic) is observed. The generalized empirical model of the tropospheric aerosol in middle latitudes of West Siberia for cloudless atmosphere had been developed for the first time and allows calculating the seasonal average vertical profiles of SSA needed for estimation of the direct climate impact of aerosol.

The main features of diurnal, seasonal, and inter-annual dynamics of BC and aerosol concentrations, and BC fraction have been studied for the near-ground measurements and parameterization of these dependences are performed.

Key words: aerosol, black carbon, spatial-temporal variability, troposphere, vertical profiles, empirical model of tropospheric aerosol, Russian Subarctic

INTRODUCTION

The urgency of comprehensive investigations of spatial-temporal variability of the aerosol and black carbon (BC) concentrations in the troposphere has increased sharply in recent years in connection with the significant effect exerted by the carbonaceous aerosol coming to the Arctic as a result of the long-range atmospheric air mass transport on the ecology and global climate change [1, 2].

According to the Resolution of the Intergovernmental Panel on Climate Change (IPCC), the lowest level of scientific understanding when estimating the radiative forcing of various atmospheric components is currently assigned to the aerosol and BC [1]. A particular problem is that the non-absorbing aerosol and BC (as main absorbing substance in the aerosol composition), depending on their sources, are emitted into the atmosphere in different amounts. Then in the process of their physicochemical evolution and long transport in the atmosphere they interact with each other and transformed into other structures that can differ strongly from the initial ones.

The detailed data on BC and aerosol in background and poorly studied regions allow to determine BC fraction (the mass ratio of the absorbing to non-absorbing substances), single scattering albedo (SSA) of aerosol in the visible wavelength range and parameters of radiation forcing used for assessment of climate change, particularly global warming. Exactly SSA determines the role of the aerosols in the atmosphere as a cooling or warming factor. The value of albedo depends strongly on BC fraction for the submicron particles in the real atmosphere, which determines the aerosol radiation forcing [3, 4].

Different remote methods are applied to investigation of aerosol properties: remote sensing using the multi-wavelength satellite images [5], sun-photometer measurements at the AERONET network [6, 7], laser sensing of the atmosphere [8]. Among local methods for investigation of atmospheric aerosol, the following techniques are commonly used: airborne measurements of aerosol parameters in the troposphere at different heights [9–15] and comprehensive long-term observations in the near-ground air layer at the stationary and mobile stations [16–19].

The data of such measurements are usually used for determination of the aerosol and BC mass concentrations, size distribution and complex refractive index of particulate matter, for calculation of different radiation-relevant characteristics (SSA, upward and downward solar radiative fluxes) [3, 4, 20, 21]. The most complicated problem is connected with uncertainties in estimates of the single scattering albedo of carbonaceous aerosol and its seasonal variations for different regions.

Black carbon is a tracer of anthropogenic and natural combustion that can be analyzed to identify the potential remote sources of Arctic BC-polluted air in northern and central Siberia [22–24]. The correct prediction of the "Siberian contribution" to BC and climate change in the Arctic requires quantification of major pollution sources and regions of Siberia with respect to the mean daily, average seasonal and annual BC and aerosol concentrations, vertical profiles, the annual variability of a BC fraction, SSA, and radiative forcing of carbonaceous aerosol [3, 4, 9, 10, 25, 26]. Such complex data are important for development of the empirical aerosol models in central and northern Siberia both from the ground and the airborne sensing method [4], and can be included into the models of long-range transport of BC pollutions to the Arctic and forecast of the climate changes [27, 28].

The main purpose of this paper is to review our investigations of the spatiotemporal variability of the aerosol and BC concentrations in the near-ground air layer and in the troposphere of West Siberia and Russian Subarctic in 1997–2016. The data of airborne sensing of the troposphere up to a height of 7 km in middle and subpolar latitudes were used to analyze the spatial variability of vertical profiles of aerosol and BC concentrations, BC fraction, SSA, and column-integrated concentrations. The generalized empirical model of optical and microphysical characteristics of tropospheric aerosol in West Siberia under conditions of the cloudless atmosphere has been developed. This model accounts for the absorbing and hygroscopic properties of particles. Many-year monitoring measurements at the stationary station has allowed us to reveal stable peculiarities in the diurnal, seasonal, and year-to-year dynamics of the aerosol and BC concentrations in the near-ground air level, and this fact has opened up the possibility of their analytical parameterization.

AIRCRAFT SENSING OF THE BC AND AEROSOL CONCENTRATIONS IN THE TROPOSPHERE OF WEST SIBERIA AND RUSSIAN SUBARCTIC

Since 1999, the Zuev Institute of Atmospheric Optics of the SB RAS carries out the airborne sensing of vertical profiles of BC and aerosol concentrations in the troposphere up to a height of 7 km in the regions of West Siberia (172 flights) and Russian Subarctic (31 flights) (55.0–75.2°N, 61.3–171°E) [9, 10]. Black carbon and aerosol concentrations were measured simultaneously with an MDA-02 four-wave differential aethalometer [29] and FAN-M angular nephelometer [30] (developed in IAO of the SB RAS) from onboard Optik AN-30 and Optik TU-134 flying laboratories [13]. A total of 172 flights in the southern Novosibirsk Region (West Siberia) [9] and 31 flights in subpolar subarctic regions of Siberia (within the POLARCAT-2008 Program and the YAK-

Aerosib Russian-French Project) [10] have been carried out. The routes of subarctic flights included several segments conditioned by the need of aircraft landing and refuelling in midpoint airports. To obtain vertical and horizontal cross sections of the atmosphere, the sensing was carried out with the flight height varying from the minimal possible level (0.5 km above ground level and 0.1 km in airport zones) to 8.5 km. During the flight between cities, the aircraft descended and ascended several times. As a result, at all segments of the routes, we have obtained horizontally spaced vertical cross sections of the troposphere up to a height of 8.5 km.

The data of airborne sensing have been used to study peculiarities in the spatial variability of vertical profiles of the mass concentrations of BC – M_{BC} (μg/m^3) and aerosol – M_A(μg/m^3), BC fraction $P = M_{BC}/M_A$, complex refractive index of particles, and SSA in the visible spectrum, as well as the integral BC concentration – M_{BC} (col) (mg/m^2)) and the aerosol optical thickness of scattering – AOT at a wavelength of 0.53 μm over the atmospheric column up to a height of 7 km. The ranges of variability of the differential aerosol and BC mass concentrations and the integral parameters are summarized in Table 1.

TABLE 1

Differential and integral characteristics of aerosol and BC over the atmospheric column up to 7 km from the results of airborne sensing of the troposphere in northeastern regions of Siberia in 1999–2016

No.	Flight routes	Dates	M_A, μg/m^3	M_{BC}, μg/m^3	Integral parameters over atmospheric column up to 7 km			
					M_{BC}(col), mg/m^2	AOT	P(col)	ω(col)
1	**POLARCAT** (56–72°N)	7–12.07.2008	0.06–300	0.01–1				
	Novosibirsk–Salekhard	*High air transparency in subpolar latitudes*			0.139	0.043	0.012	0.98
	Salekhard–Khatanga				0.249	0.049	0.019	0.97
	Khatanga–Chokurdakh				0.153	0.018	0.032	0.96
	Chokurdakh–Pevek–Chok				0.161	0.019	0.032	0.96
	Chokurdakh–Yakutsk			*Wildfires*	1.72	1.52	0.004	0.99
	Yakutsk–Mirny				1.31	0.64	0.008	0.99
	Mirny–Novosibirsk				0.370	0.038	0.036	0.95
2	**YAK-Aerosib** (66–75.2°N)	15–17.10.2014	0.8–6	0.02–1	*High air transparency in subpolar latitudes*			
	Salekhard–Dikson				0.310	0.049	0.026	
	Salekhard–Novaya Zemlya				0.364	0.063	0.030	
3	**YAK-Aerosib** (56–62°N)	31.07–01.08.2012	2–295	0.02–6.5	*Strong influence of the fast Siberian forest fires*			
	Novosibirsk–Tomsk				4.25	0.511	0.031	0.96
	Tomsk–Mirny				6.19	1.32	0.017	0.97
	Mirny–Yakutsk				9.96	2.77	0.014	0.98
	Yakutsk–Bratsk				4.23	0.461	0.034	0.95
	Bratsk–Novosibirsk				5.36	1.41	0.014	0.98
4	**YAK-Aerosib** (56–62°N)	15–18.04.2010	0.7–9	0.01–3	*Background cloudless conditions*			
	Novosibirsk–Mirny				0.836	0.054	0.058	0.92
	Mirny–Yakutsk–Lensk				0.352	0.086	0.015	0.98
	Lensk–Bratsk				0.501	0.017	0.110	0.86
	Bratsk–Novosibirsk				1.820	0.137	0.049	0.93
5	**West Siberia** (54.5°N),	1999–2011	0.5–50	0.02–5	***Average seasonal background***			
	Novosibirsk Region	*Spring*			2.64	0.180	0.034	0.92
		Summer			0.78	0.101	0.029	0.96

The analysis has shown that the ranges of variability of aerosol and BC mass concentrations in the troposphere of the studied subarctic region are very wide: 0.06–300 μg/m^3 and 0.01–6.5 μg/m^3. The ranges of variability of the integral BC concentration and the aerosol scattering optical thickness over the atmospheric column up to 7 km were also wide being about two orders of magnitude: M_{BC}(col) = 0.14–9.96 mg/m^2 and AOT = 0.02–2.8. The so wide spatiotemporal variability of the aerosol and BC content in the troposphere is caused by various factors: the dynamics of vertical profiles of the studied characteristics, the action of factors associated with the processes of generation of aerosol particles, their physicochemical transformation, and long-range transport by air masses. An important purpose of the airborne sensing was the study of general and distinctive features of vertical profiles and spatial variations of the aerosol and BC concentrations, as well as classification of flights with allowance for the typical states of the subarctic troposphere. The data of airborne sensing have been classified for the first time by the integral characteristics of atmospheric column: AOT (0.53 μm), M_{BC} (col), P (col), and SSA (col). The detailed analysis of the data has allowed us to divide the flight series into three groups of "optical weather" depending on the degree of aerosol turbidity of air and reflecting the characteristic states of subarctic aerosol. The first group corresponds to the conditions of low turbidity of the troposphere (high transparency of air) in the subarctic latitudes for flight cycles Nos. 1 and 2 (see Table 1) under POLARCAT-2008 (Eastern Subarctic) and YAK-Aerosib-2014 (Kara Sea). The second group of flights in the mid-latitudes corresponded to the conditions of extremely strong (cycle No. 3) or moderate influence of smokes from Siberian forest fires on the composition of tropospheric aerosol. The influence of forest fires also manifested itself in cycle No. 1 in the flights in the mid-latitudes at the route segments from Yakutsk to Novosibirsk. The third group of flights characterizes the "regional mean aerosol background" and corresponds to the levels of moderate aero-

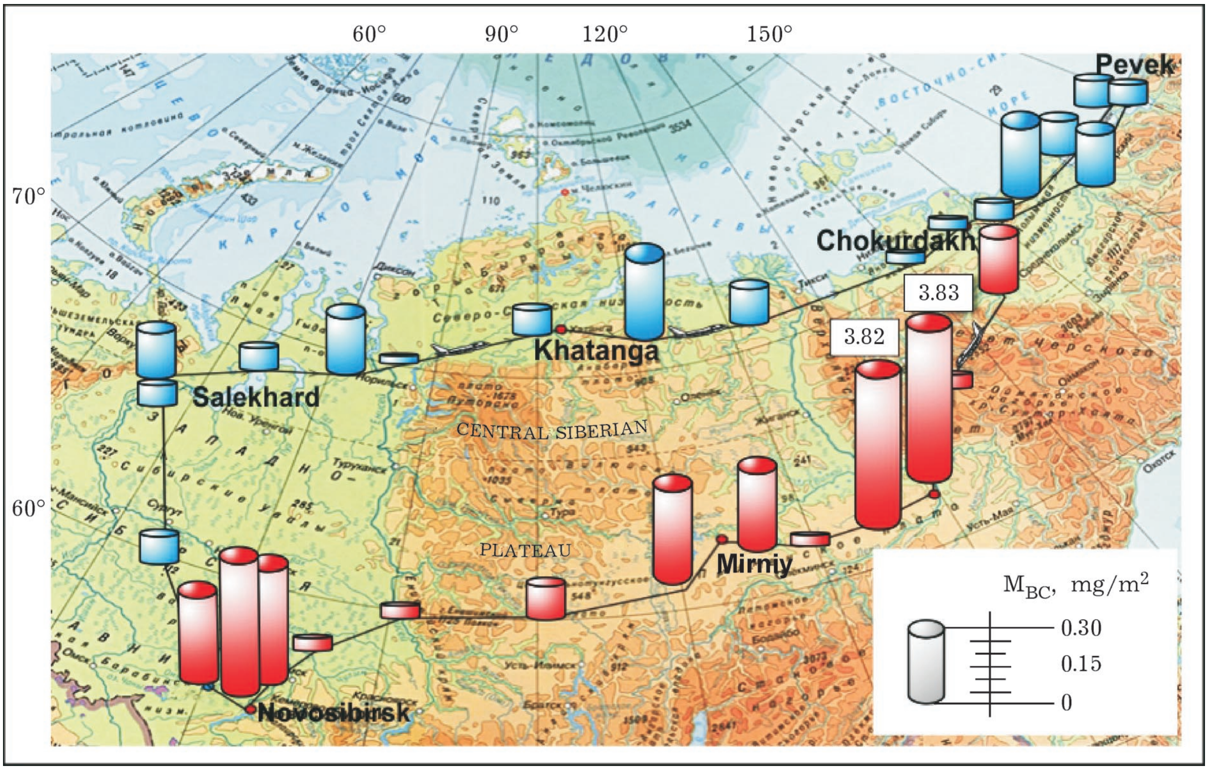

Fig. 1. Integral BC concentrations in polar and middle latitudes during the airborne sensing campaign POLARCAT (7–12.07.2008, 56–72°N).

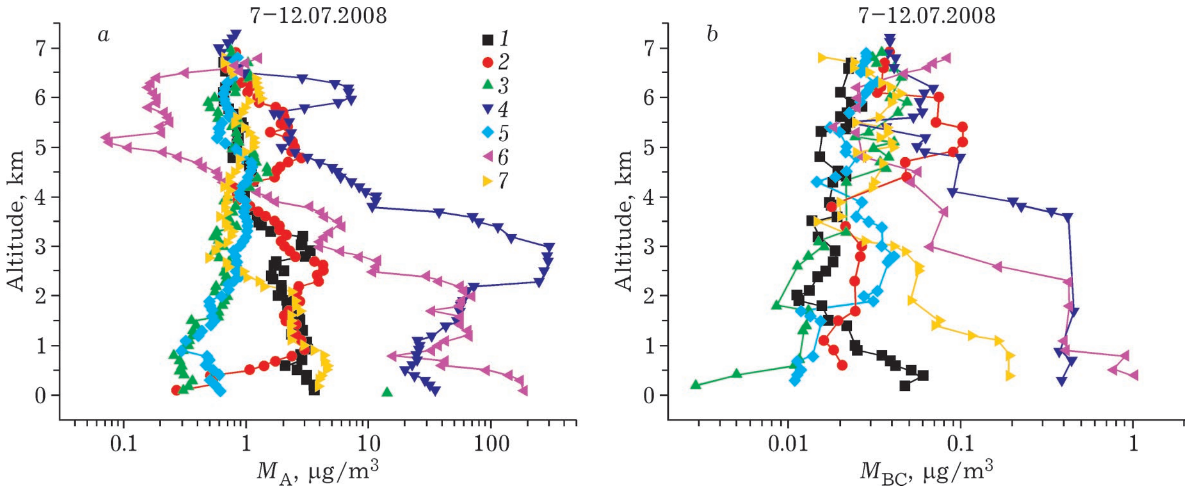

Fig. 2. Vertical profiles of aerosol (*a*) and BC (*b*) concentrations in the cycle of route flights in subarctic regions of Siberia in 2008 within the framework of POLARCAT: *1* – Novosibirsk–Salekhard, *2* – Salekhard–Khatanga, *3* – Khatanga–Chokurdakh, *4* – Chokurdakh–Yakutsk, *5* – Chokurdakh–Pevek, *6* – Yakutsk–Mirny, *7* – Mirny–Novosibirsk.

sol turbidity of air in the mid-latitudes: cycles Nos. 5 and 6. Taking into account the proposed classification of aerosol states of the troposphere, we have considered peculiarities in the dynamics of aerosol and BC content in the subpolar and mid-latitudinal regions of Siberia.

Conditions of high transparency of air in subpolar latitudes

In July 2008 (International Polar Year) within the framework of POLARCAT Project, a cycle of route aircraft sensing of the concentrations of BC and submicron aerosol in the troposphere in the subpolar latitudes along the route Novosibirsk–Salekhard–Khatanga–Chokurdakh–Pevek (Chukotka)–Chokurdakh–Yakutsk–Mirny–Novosibirsk was carried out (cycle No. 1 in Table 1). The region of the flights lied in the latitude range of 56–72°N and the longitude range 77–171°E (Fig. 1). At the route segments Khatanga–Chokurdakh and Chokurdakh–Pevek–Chokurdakh, the flights were carried out in the subpolar latitudes (70–72°N).

Figure 2 shows the vertical profiles of aerosol (*a*) and BC (*b*) concentrations for various route segments. In the flights, the coordinated variations of the shape of the aerosol and BC vertical profiles were observed. Aerosol and BC concentrations varied in wide ranges from 0.06 to 300 μg/m³ and from 0.003 to 1 μg/m³, respectively. It should be noted that the flights along the subpolar Khatanga–Chokurdakh and Chokurdakh–Pevek–Chokurdakh route segments took place under conditions of the extremely high transparency of air at all the heights in the troposphere (see Figs. 2, 3). In the subpolar latitudes, the extremely low concentration levels were observed: lower than 1 μg/m³ for aerosol (see Fig. 2, *a*) and lower than 0.05 μg/m³ for BC (*b*).

Figure 3 illustrates the dynamics of the vertical profiles of the aerosol and BC concentrations in the subpolar latitudes. The profiles for every segment were obtained at the significant horizontal averaging over the subpolar path from Khatanga to Pevek about 2400 km long. It can be seen that for aerosol and for BC the

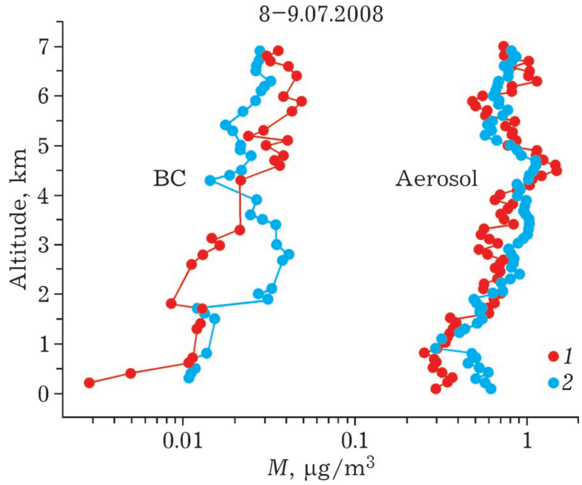

Fig. 3. Inverse vertical profiles of the aerosol and BC concentrations in the subpolar latitudes (70–72°N) obtained on July 8–9 of 2008 in the flights under POLARCAT: *1* – Khatanga–Chokurdakh, *2* – Chokurdakh–Pevek.

average vertical profiles for the considered segments are close to each other both in the shape and in the range of variation of the characteristics. This circumstance is indicative of the homogeneous composition of aerosol in the troposphere of the studied subpolar region.

The aerosol and BC concentrations at the subpolar paths took extremely low values and varied in narrow ranges of 0.3–1.5 μg/m^3 and 2–40 ng/m^3 with height. These levels of the differential concentrations corresponded to the conditions of high transparency of air with the extremely low values of AOT ~0.018 and the low values of the column-integral BC concentration ~0.15 mg/m^2 (see Table 1 and Fig. 1). The low values of the BC concentrations obtained in these flights in the near-ground layer are close to the background summer levels of the BC content in the arctic aerosol under conditions of the low effect of long-range transport of anthropogenic pollutants [14, 15, 31, 32]. Thus, according to the data of measurements in 1998–2007 at the Zeppelin station (78.9°N, 11.9°E, 475 m above ground level), the monthly average BC concentrations in February–March (in the period of Arctic haze) achieved maximal values 80 ng/m^3 and decreased down to minimal values 010 ng/m^3 in June–September [31]. The so low average summer BC concentrations (10 ng/m^3) were observed in the measurements of 1989–2003 at the Barrow (Alaska, the USA) and Alert (Canada) polar stations [32]. The measured data demonstrate the year-to-year trend to decrease of the average BC concentrations, which is likely caused by the decrease of BC emissions in high latitudes and in regions of Northern Eurasia [23].

An important feature of the subpolar atmosphere was that the shape of the vertical profiles of aerosol and BC became inverse, showing an increase of the concentrations with height (see Fig. 3). It should be noted that similar inverse profiles of BC concentration were observed by American scientists in 2008 in the flights near the Barrow research station on Alaska within the framework of the POLARCAT Program [14, 15]. The obtained inverse concentration profiles differ qualitatively from the vertical profiles usually observed in flights in mid-latitudes of 45–65°N, in which the aerosol and BC concentrations decrease with height (see, for example, [8–11] and the route segments Novosibirsk–Salekhard and Mirny–Novosibirsk in Fig. 2). The aerosol and BC concentrations can increase, on average, four to five times with height. It should be noted that a local maximum of the BC concentration was observed at heights of 2–3 km in the Chokurdakh–Pevek segment.

In October 15–17, 2014, four flights of the flying laboratory were carried out in Russian Subarctic to the Kara Sea (55.0–74.8°N, 61.3–82.9°E): flight No. 1 Novosibirsk–Salekhard, No. 2 Salekhard–Kara Sea (eastern sector, north

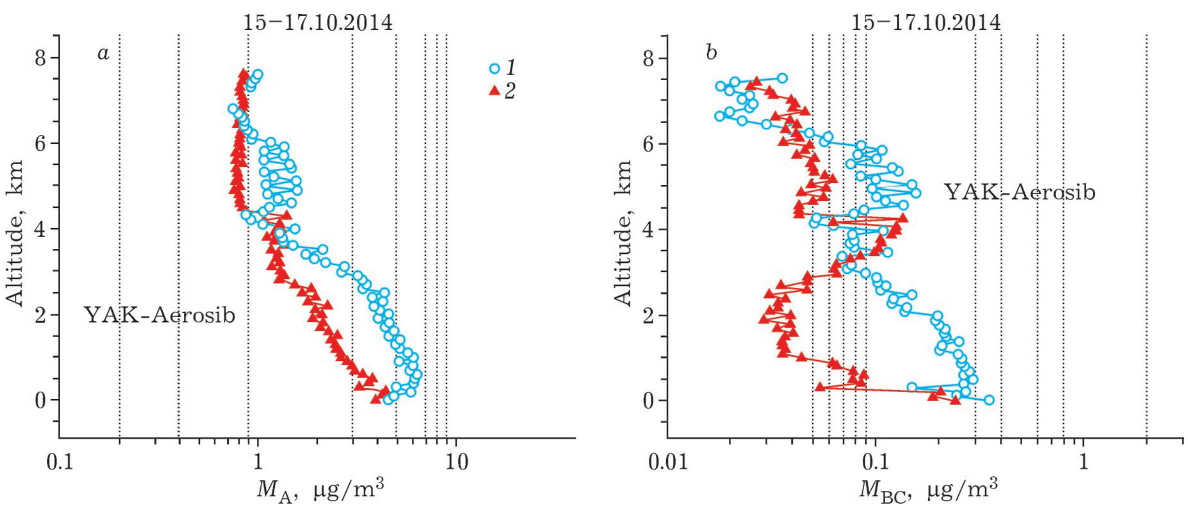

Fig. 4. Vertical profiles of the aerosol (*a*) and BC (*b*) concentrations in the cycle of route flights in October 15–17 of 2014 in Russian western Subarctic (region of the Kara Sea): *1* – Salekhard–Novaya Zemlya, *2* – Salekhard–Dikson.

of Dikson)–Salekhard, No. 3 Salekhard–Kara Sea (western sector, south of Novaya Zemlya)–Salekhard, No. 4 Salekhard–Novosibirsk. For the first time, 14 spatial vertical cross sections of the troposphere up to a height of 7 km were obtained for the region under study. In the subpolar latitudes, the shape of the vertical profiles of the aerosol and BC concentrations (Fig. 4), in contrast to those observed in 2008 in Eastern Subarctic (see Fig. 3), was characterized by a decrease of the concentrations with height and appearance of local maxima at heights of 0.5–2 km and 4–6 km. At flights in the mid-latitudes (Novosibirsk–Salekhard and Salekhard–Novosibirsk) and the subpolar latitudes, the aerosol and BC concentrations M_A and M_{BC} in the vertical profiles varied within 0.25–20 and 0.02–1.0 μg/m^3, respectively. In the subpolar latitudes (71–74.8°N) north of Salekhard (66.6°N), the ranges of variability of the concentrations become narrower: 0.8–6 μg/m^3 for aerosol and 0.02–0.3 μg/m^3 for BC. It follows from the aforesaid that in the flights Novosibirsk–Salekhard and Salekhard–Novosibirsk the latitudinal dependence manifested itself as a decrease in the value of the near-ground concentrations, on average, three times, as the latitude increased from 56 to 74.8°N (see Table 1).

The comparison of the vertical profiles over the Kara Sea (see Fig. 4) shows that the western part of the troposphere in the region of Novaya Zemlya is more "polluted" with aerosol and BC than the eastern part near Dikson settlement. At heights up to 3 km and 4–6 km, the excess of the aerosol concentration was about two times, and for BC the excess was two to ten times. The excess of the integral concentrations (see Table 1) was not so pronounced (on average, 1.3 times): BC = 0.36–0.31 mg/m^2, AOT = 0.063–0.049.

The data of airborne sensing of the troposphere in the subpolar latitudes in fall of 2014 and in summer of 2008 (see Table 1) demonstrate the significant decrease of both differential and integral aerosol and BC concentrations in the direction from the west (the Kara Sea) to the east (Eastern Subarctic) (see Table 1). As to the aerosol concentration, this decrease corresponds to the ranges from 0.8–6 to 0.3–1 μg/m^3, *i. e.*, 2.7 to 6 times. The corresponding BC profiles vary from 0.02–0.3 to 0.01–0.05 μg/m^3, *i. e.*, 2 to 6 times. The mean integral BC concentration and AOT within the ranges of 0.34–0.16 mg/m^2 and 0.056–0.02, respectively, 2.1–2.8 times. The obtained estimates show a tendency to decrease of the aerosol and BC concentrations in the subpolar latitudes in the west-to-east direction.

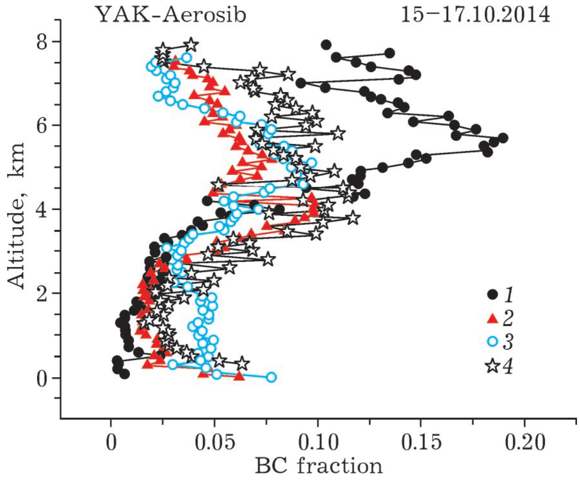

Fig. 5. Vertical profiles of BC fraction during flight campaign to Kara Sea in 2014 (56–75.2°N): *1* – Novosibirsk–Salekhard, *2* – Salekhard–Dikson, *3* – Dikson–Novaya Zemlya, *4* – Salekhard–Novosibirsk.

For all route segments of the flights to the Kara Sea, the vertical profiles of the BC fraction P value have maxima at the heights of 4–6 km, whose values can achieve 0.10–0.18 for the flights in the mid-latitudes (Fig. 5). The P values are reduced to 0.10 for the western sector of the sea and to 0.07 for the eastern sector. The revealed maxima of the elevated content of BC (P values) in the subpolar and middle latitudes evidences that in the measurements periods at the heights of 4–6 km the long-range transport of BC-containing aerosols occurred.

Influence of smokes from forest fires in the warm season

The airborne sensing of the atmosphere on July 31, 2012 and July 19, 2013 was carried out under conditions of extensive Siberian forest fires in the Tomsk Region, Krasnoyarsk Territory and Yakutia, which caused the heavy smoke pollution of the troposphere all over the tropospheric thickness at all segments of the

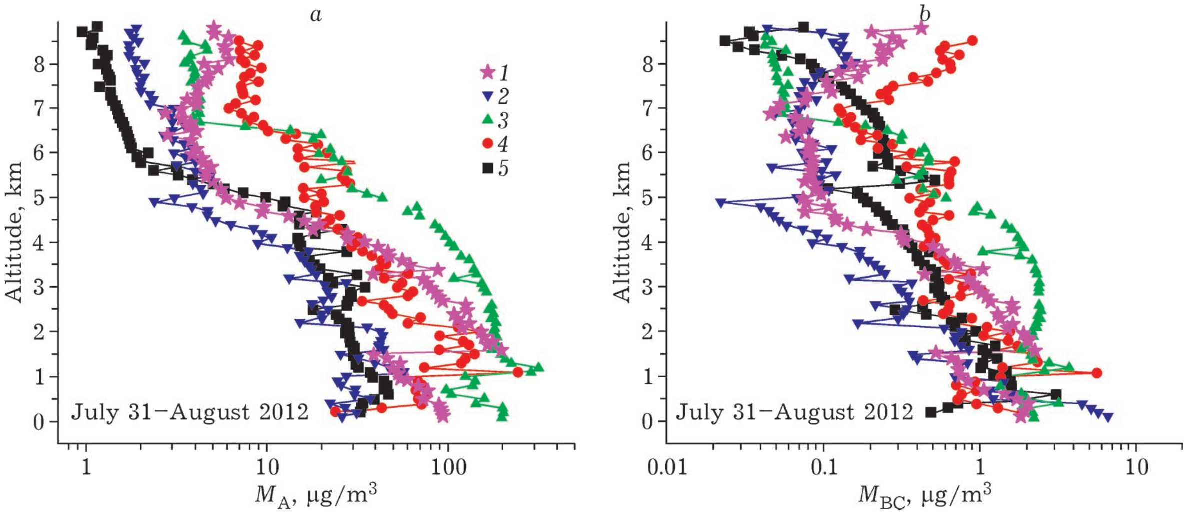

Fig. 6. Vertical profiles of aerosol (*a*) and BC (*b*) concentrations in the cycle of route flights by the YAK-Aerosib Program in the period of Siberian forest fires in July–August of 2012: *1* – Bratsk–Novosibirsk, *2* – Yakutsk–Bratsk, *3* – Mirny–Yakutsk, *4* – Tomsk–Mirny, *5* – Novosibirsk–Tomsk.

flight route (Fig. 6, see Table 1). The shape of the aerosol and BC profiles is characterized by the maxima at heights of 3, 5.5, and 8 km. The variations of the concentration with height were 30–91 times for aerosol and 4–120 times for BC. In the horizontal cross sections, the aerosol and BC concentrations varied 3–19 times, showing the maxima at heights of 0–1, 3–5, and 8 km. The flights in 2012 were subject to the extremely strong effect of smokes from Siberian forest fires. The high concentrations of aerosol and BC were achieved: up to 300 µg/m^3 and up to 6.5 µg/m^3, respectively.

The maximal smoke pollution of air was observed at the segments Mirny–Yakutsk and Tomsk–Mirny. The atmospheric optical thickness AOT at a wavelength of 0.53 µm and the integral BC concentration M_{BC}(col) were characterized by the extremely high values of 0.46–2.77 and 4.23–18.7 mg/m^2 (Fig. 7) all over the flight route. The data of Table 1 and Fig. 1 demonstrate that the flights in the middle latitudes under the POLARCAT Program in July 2008 at the segments from Yakutsk to Novosibirsk also took place under conditions of the increased influence of smoke from forest fires.

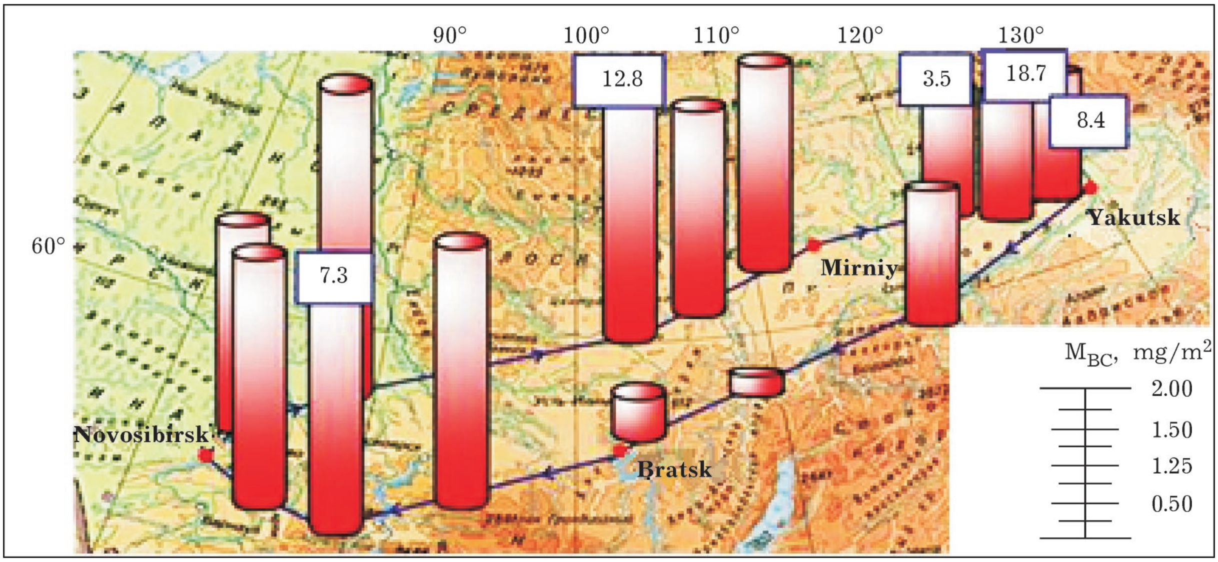

Fig. 7. Integral BC concentrations during airborne sensing campaign in the middle latitudes in July–August, 2012.

The comparison with the data of flights in the slightly turbid (background) atmosphere (April 15–18, 2010, see Table 1) has shown that the forest fires lead to an increase of the aerosol concentration at heights of 2, 4, and 6 km by 80, 33, and 12 times, respectively. The values of P at heights up to 7 km becomes low – lower than 0.03, while the smokes are characterized by high values of the single scattering albedo in the visible spectral region (0.96–0.98), manifesting itself in the radiative-climate forcing as a cooling factor [3, 10, 33, 34].

Conditions of the regional mean aerosol background in Siberian mid-latitudes

Flight cycles Nos. 4 and 5 in the middle latitudes, which were characterized by the moderate levels of the aerosol and BC concentrations in the vertical profiles (see Table 1), were classified as conditions of the regional mean aerosol background in the troposphere.

Thus, in the flights under YAK-Aerosib in April 2010 by the route Novosibirsk–Mirny–Yakutsk–Lensk–Bratsk–Novosibirsk (56–62°N), the following concentrations were achieved: 0.7–9 $\mu g/m^3$ for aerosol and 0.01–3 $\mu g/m^3$ for BC. The aerosol and BC measurements in the troposphere over the Zav'yalovo village (54.5°N, West Siberia) of the Novosibirsk Region in cloudless atmosphere once every month from the Optik-E AN-30 flying laboratory in 1999–2011 [8] were also classified as background. These data had allowed us to estimate the empirical average seasonal vertical profiles of the concentrations, BC fraction, and SSA for the dry matter of aerosol particles and to study their relation with the temperature stratification in the troposphere.

In this case, the range of variability of the concentrations with height were, on average, from 0.02 to 5 $\mu g/m^3$ for BC and from 0.5 to 50 $\mu g/m^3$ for aerosol, that is, the variations exceeded two orders of magnitude. Against the background of monotonic decrease with height, local maxima of the concentrations were observed quite often at different heights. An important feature is the closeness of the shapes of vertical profiles of aerosol and BC. The shape of the vertical profiles is characterized by the significant seasonal dynamics closely connected with variations of the temperature stratification of the tropospheric boundary layer. In the cold season, temperature inversions in the height range of 0.5–1.5 km, which are characteristic of the Siberian region, lead to the most pronounced dynamics of the vertical profile of BC. The largest gradient of variation is usually observed in the subinversion layer: from 3–5 to 0.1 $\mu g/m^3$. Above the inversion layer, the further, less pronounced decrease of BC down to 20–10 ng/m^3 occurs with height. In the warm season, the daytime rare temperature inversion in combination with the seasonal decrease of the intensity of BC sources lead to a decrease of the concentrations in the near-ground layer. As a result, the range of variability becomes narrower and the dynamics of the vertical profiles becomes smooth.

The estimates of the column-integral aerosol and BC concentrations from the results of airborne sensing (see Table 1) corresponding to the aerosol background of Siberia in the middle latitudes can be used for verification of data obtained by the methods of sun photometry and satellite sensing, as well as developed models of the optical characteristics of tropospheric aerosol.

Aerosol model of cloudless troposphere of West Siberia

Based on the long-term airborne measurements under background conditions [9] the generalized dynamic model of tropospheric aerosol in West Siberia under conditions of the cloudless atmosphere has been developed for the first time [4]. The model takes into account the absorbing and hygroscopic properties of particles and uses as main input parameters the seasonal-average vertical profiles of the aerosol scattering coefficient, particle size distribution, BC mass concentration, parameter of condensation activity of particles, and relative air humidity for different seasons and air masses.

The atmospheric aerosol is modelled as a sum of the fine (submicron sizes) and coarse fractions of particles. Each fraction was represented in the form of lognormal particle size distribution. Their parameters (mean radii and half-widths of the distributions) were set according to the model [4] for different seasons. The volume filling factor of each fraction was selected so that the angular scattering coefficient at 45° and $\lambda = 0.51$ μm of the aerosol dry matter calculated by the Mie theory formulas coincided with the seasonal average value [9, 35] the respective altitude. The volume concentration of BC in each fraction was determined so that its total mass concentration at each altitude coincided with the experimental seasonal average value BC [9]. Refractive index of the dry non-absorbing aerosol was set $n = 1.5$. Then the refractive index of absorbing aerosol was calculated taking into account the data on the BC concentration. Based on experimental data [16, 36, 37], the variant was chosen for detailed analysis, when 90 % of the absorbing substance (BC) is concentrated in the submicron fraction and 10 % is in the coarse one. It was assumed that in each fraction BC is homogeneously distributed inside the particles. The complex refractive index of particles for each fraction was calculated using the well known homogeneous volume internal mixture rule. At the final stage, the values of the fine fraction parameters were reduced to the seasonal average relative humidity of air [38] using the Hanel formula and seasonal average value of the parameter of condensation activity at each altitude [39]. Coarse fraction was not humidified, because it was shown [40] that, for more adequate description of the optical properties of particles, one should ascribe the greatest intensity of the condensation growth to the particles with the sizes 0.2–0.5 μm. The vertical profiles of SSA for the humidified absorbing aerosol at 7 wavelengths in the visible and near IR were calculated for different seasons using Mie theory.

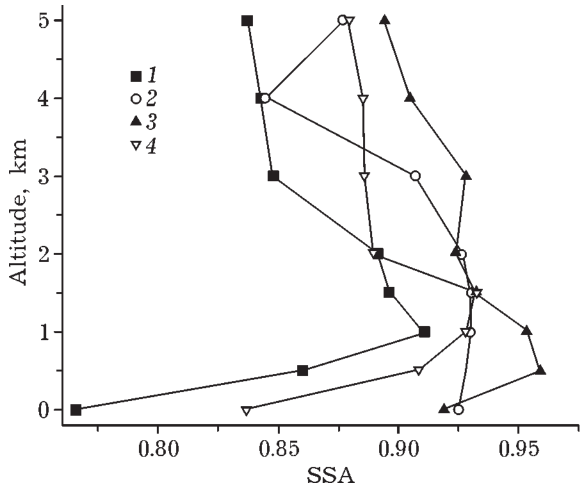

Fig. 8. Seasonal average vertical profiles of SSA at $\lambda = 0.55$ μm in different seasons.

The characteristic peculiarity in all seasons is low values of SSA in the near-ground air layer (Fig. 8). The maximum is observed at the altitudes of 0.5–1.5 km, then the values of SSA decrease with altitude. The altitude of 4 km is distinguished in spring. The extremely low value of the albedo is observed here, which is caused by the enhanced concentration of BC at this altitude [9]. Perhaps, it is effect of remote transfer of aerosol from the regions of the European part of Russia, where peatbog fires can be observed in April and May.

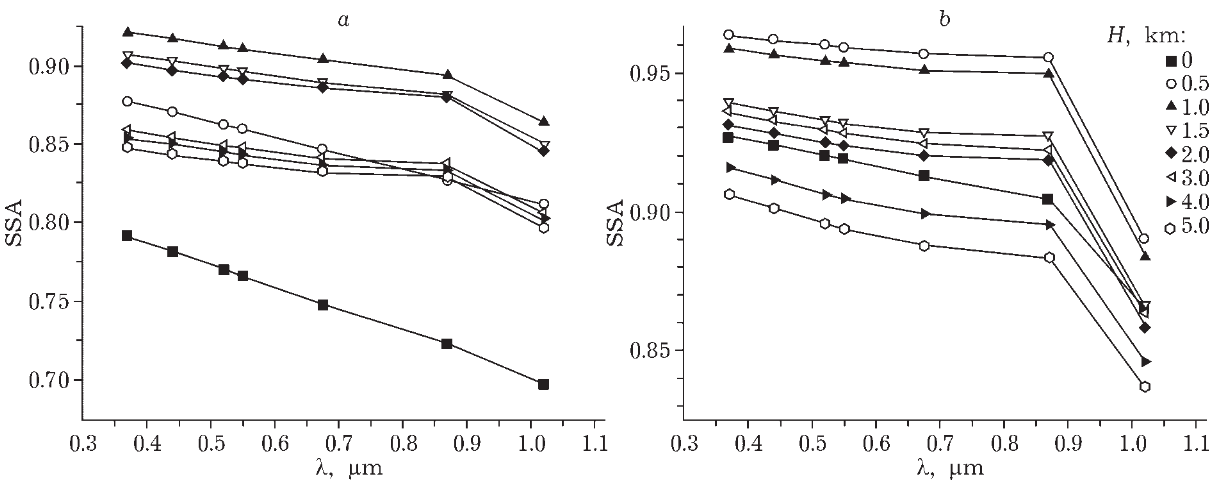

Fig. 9. Average spectral dependences of SSA at different altitudes for contrast seasons: a – winter and b – summer.

The values of SSA decrease with wavelength in all seasons (Fig. 9). The most strong decrease is observed in infrared range ($\lambda = 1.06$ μm). Near to neutral spectral behaviour is observed in spring at the altitudes above 3 km in the visible wavelength range. In summer, selectivity of the spectral behaviour of SSA in the visible range is lower than in other seasons.

The developed empirical model of tropospheric aerosol in West Siberia provides good recovery of the vertical profiles of seasonal average values of SSA needed to calculate the radiation balance of the atmosphere in the visible region of the spectrum. It was established that in the wavelength range 0.44–0.87 μm the average values of SSA and asymmetry factor for the column of the atmosphere, calculated using the empirical model, are in good agreement with the respective average values obtained according AERONET network in Tomsk for 2004–2009 [4]. Successful testing of the developed methodological approach allows us to take it as the basis for the subsequent development of an empirical model of tropospheric aerosol in the Arctic region.

TEMPORAL DYNAMICS OF BC AND AEROSOL CONCENTRATIONS IN THE NEAR-GROUND AIR LAYER

Since 1997, the Aerosol Station of IAO of the SB RAS located on the southeastern periphery of the city of Tomsk conducts round-the-clock every-hour monitoring measurements of the angular scattering coefficient of the dry matter of submicron aerosol at a wavelength of 0.51 μm with the FAN-A angular nephelometer and the BC mass concentration M_{BC} (in μg/m^3) with the MDA-02 four-wave aethalometer developed by IAO, SB RAS [3, 17, 29, 30, 33]. The data on the angular scattering coefficient at an angle of 45° are used to calculate the mass concentration of dry submicron aerosol M_A (in μg/m^3) based on the one-parameter model of atmospheric haze [41]. The measurements of the aerosol and BC content are used for calculation of the important radiation-significant parameter – BC fraction P in particles, which is determined as a ratio of the mass concentrations of BC and submicron aerosol with a mean error of about 15 %. The results of monitoring measurements are then organized into arrays of annual average, seasonal average, and diurnal average values of aerosol parameters.

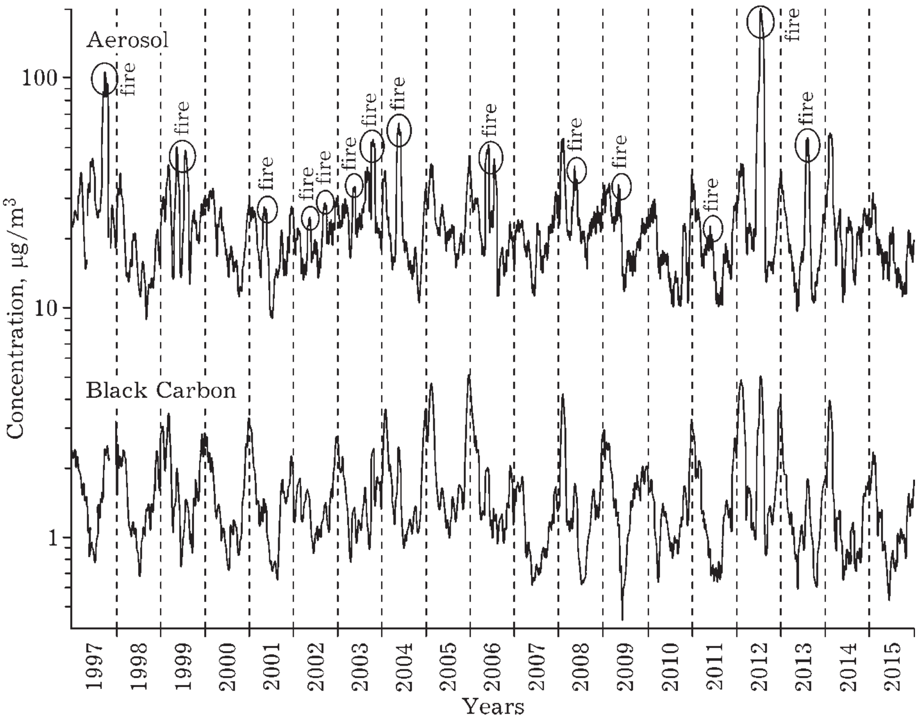

Fig. 10. Temporal dynamics of the mass concentrations of submicron aerosol and BC (30-day smoothing average of diurnal average values).

Multi-year regular observations have revealed the stable season-to-season variability of the mass concentrations of submicron aerosol and BC with a minimum in the summer period and a maximum in winter (Fig. 10). A similar dependence is also characteristic of the BC fraction *P*. Monthly average values of *P* vary from 3 to 15 %.

The annual dynamics of *P* is caused by the fact that the seasonal dynamics of BC concentration is more pronounced than variations of the aerosol concentration, which is determined by seasonal differences in the intensity of the major processes, responsible for emission of submicron aerosol and BC. Actually, the processes of photochemical generation of aerosol particles become significantly more intense upon the transition to the warm season. At the same time, the intensity of BC generation decreases with termination of the heating season. These seasonal differences in the processes of aerosol emission and BC sink favour the formation of the summer minimum in the annual profile of the BC fraction.

Forest fires, various by their scales often appear at the territory of Siberia in the warm period. As a smoke plume of forest fires comes to a measurement site, the BC and aerosol concentrations increase drastically. For a half of these realizations the mass concentrations of submicron aerosol exceed the high winter peaks of the aerosol content caused by the heating season and temperature inversions that are often observed in Siberia in winter. These situations were observed in 1997, 1999, 2003, 2004, 2006, 2013, and, especially, 2012. For the BC values, the excess was observed only in 2012. The level of the summer minimum of the BC fraction *P* under these conditions becomes even lower, since the value of *P* decreases in smokes of forest fires [3, 33]. In summer of 2012, the smoke haze from Siberian forest fires was extremely dense for the last 15 years. A characteristic feature of smokes is a sharp decrease of the soot fraction to 0.02–0.03 [33].

For the analysis of temporal variability of aerosol characteristics, arrays, freed of the forest fires influence, were created (Fig. 11).

For the entire period of observations, the stability of the annual behaviour of aerosol characteristics in the near-ground atmosphere is observed. The weak effect of smokes from spring grassland fires and summer-fall forest fires shows itself for only 1/3 of year realizations. Under these conditions, the annual behaviour of the BC and aerosol concentrations and the relative content of BC has a pronounced winter maximum and summer minimum of concentrations.

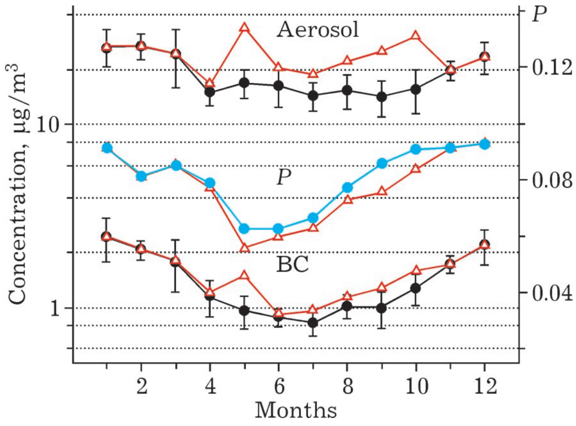

Fig. 11. Mean annual behaviour of the mass concentrations of aerosol, black carbon and BC fraction (red lines – taking into account fires).

In addition, the year-to-year variability of these aerosol characteristics is observed (Fig. 12). For the entire period, statistically significant negative trends for M_{BC} (−2.4 % a year) and *P* (−2.9 % a year) were observed, as well as the statistically insignificant trend of M_A (−0.2 % a year). One can see that the temporal profiles of the annual average values of aerosol parameters under study have local extremes. Straight lines in the figure illustrate these trends. The significant negative trend of *P* indicates that the decrease

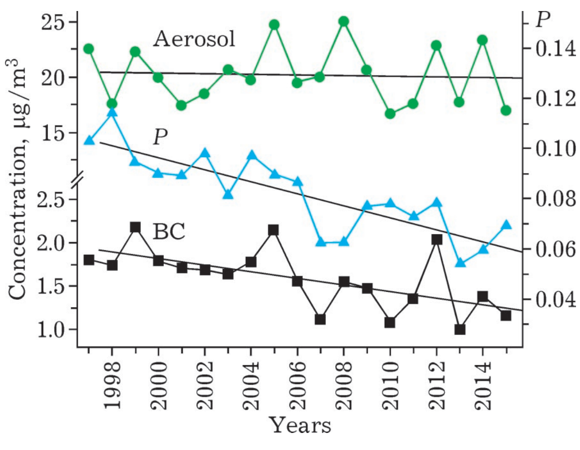

Fig. 12. Temporal course of the annual average mass concentrations of submicron aerosol and soot and the relative content of soot and their linear trends.

of the absorbance of aerosol particles observed for the last 19 years still continues.

Using the results of measurements at the Aerosol Station of the Institute of Atmospheric Optics of the SB RAS, peculiarities of the diurnal dynamics of the aerosol characteristics in the surface air layer were analyzed [17].

For the influence study of the cloudiness on the form of diurnal courses, alongside with complete data arrays, subarrays for the radiative type of weather: clear atmosphere (cloud amount of 0–2) and cloudy atmosphere were considered (cloudiness amount of 8–10).

The analysis of experimental data has allowed revealing the major peculiarities in the time dynamics of the diurnal profiles, normalized with the values of daily average concentrations. The BC diurnal profile is close to that of aerosol in its shape. The shape of the diurnal profiles is identical for all seasons and has two maxima (morning and evening-night ones) and two minima (nighttime and daytime) (Fig. 13). The positions of the maxima in the diurnal profiles of aerosol demonstrate seasonal variability, which manifests itself in the closer positions of the maxima in winter (morning maximum at 8–9 Local Time and evening maximum at 21–22 Local Time) and the separation in summer (6 and 23 Local Time). For the diurnal profile of soot, the corresponding positions of the maxima are 11–12 and 20–21 LT in winter and 9 and 22 LT in summer. In the spring-summer period, the extremes of the diurnal profiles are most pronounced in amplitude. In fall and winter, the diurnal profiles are less contrast.

At the radiative type of weather, all the extremes of the diurnal profiles of soot and aerosol are clearly pronounced and statistically significant. Under overcast conditions, the seasonal profiles of aerosol and BC smooth out. The amplitude expressiveness of the extremes may decrease by 10–20 % for aerosol and by 10–40 % for soot. In this case, all the extremes of the BC diurnal profile remain statistically significant. However, for the aerosol, the amplitudes of the morning maximum and the night minimum strongly level out, and, as a result, for all seasons the difference between these extremes becomes statistically insignificant.

This allows making the conclusion that the continuous cloudiness is an important geophysical factor that affects strongly peculiarities of the diurnal profile of aerosol and soot. A probable mechanism of cloud influence on the diurnal profiles is its limiting effect of clouds on the processes of aerosol removal to the upper tropospheric layers and on the intensity of the aerosol formation in the surface layer.

The availability of stable peculiarities of the seasonal variability of the normalized diurnal profiles of the aerosol and BC concentrations favours their approximation. This is an important stage in development of aerosol models. We have proposed an approximation of the seasonal average diurnal profiles

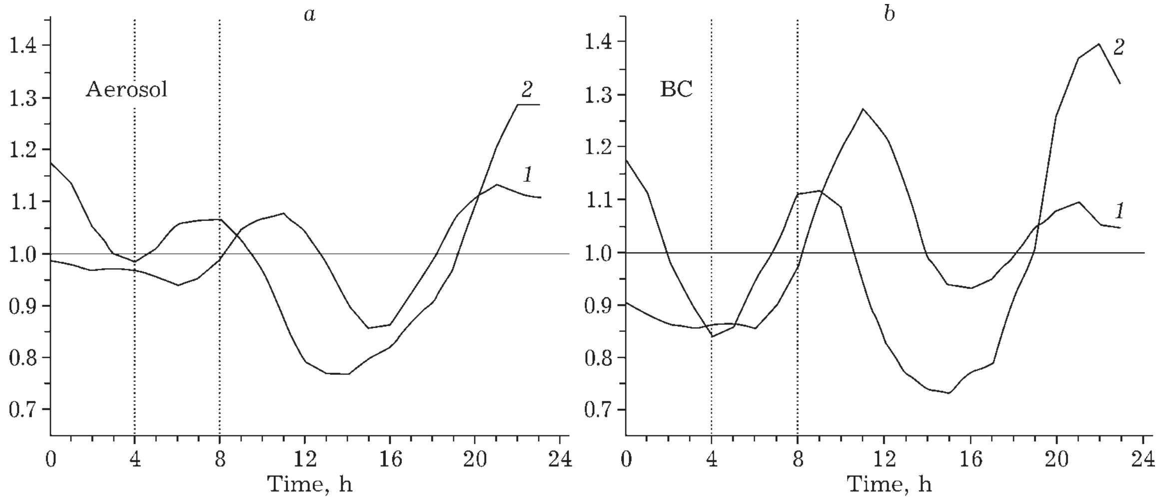

Fig. 13. Seasonal-average normalized diurnal variations of the aerosol (*a*) and BC (*b*) concentrations at the Aerosol station of IAO of the SB RAS in cloudless atmosphere in 1997–2008: *1* – winter, *2* – summer.

TABLE 2

Parameters of diurnal behaviour of the seasonal-average aerosol and BC concentrations for the complete data arrays

Parameters	Aerosol				Black carbon			
	Winter	Spring	Summer	Autumn	Winter	Spring	Summer	Autumn
A_1	0.048	0.069	0.15	0.105	0.08	0.168	0.2	0.21
φ_1	7.43	7.97	5	7.045	7.05	18.2	7	6.28
T_1	10.8	10.5	13.4	10.6	11.1	12.4	12	11.2
A_{02}	0.987	0.97	1.33	0.966	0.995	1.01	1.04	0.995
A_2	0.057	0.19	0.5	0.145	0.08	0.068	0.1	0.135
φ_2	−1.42	−1.54	26.1	−2.5	10.09	−8.22	−18.1	10.8
T_2	19.1	20.4	50.8	18.6	25.6	28.6	36.6	26.3

of aerosol and BC as a product of two sine functions:

$$y = (1 + A_1 \sin(2\pi \frac{t - \varphi_1}{T_1}))(A_{02} + A_2 \sin(2\pi \frac{t - \varphi_2}{T_2}))$$

where A_1 and A_2 are the amplitudes; φ_1 and φ_2 are the phases; T_1 and T_2 are the oscillation periods (Table 2) [17].

The period of oscillations in the first sine function corresponds to the time interval between the daytime and nighttime maxima. The second sine function follows the amplitude difference of the diurnal extremes. The estimates have shown that the proposed approximation reflects the main peculiarities in the shape of seasonal average diurnal profiles of the aerosol and BC concentrations and provides their quantitative description with a mean error of about 5 % (Fig. 14).

In the most part of observations in our region, the important role of smokes from fires in boreal forests of Siberia has manifested itself in the dynamics of concentrations and the chemical composition of aerosol in the surface air layer. A weak influence of smokes from grass fires and forest fires was observed only in 1/3 of year realizations. The largest invasion of smokes from forest fires was observed in summer of 2012.

Extensive forest fires took place in the Siberian region in summer of 2012 under conditions of the low-gradient baric field of high pressure. The most intense forest fires were detected at the Aerosol Station occurred from late June to mid-August (Fig. 15). For the period of measurements, four prominent episodes of invasion of smoke haze to the region of measurements were observed: June 17–July 6,

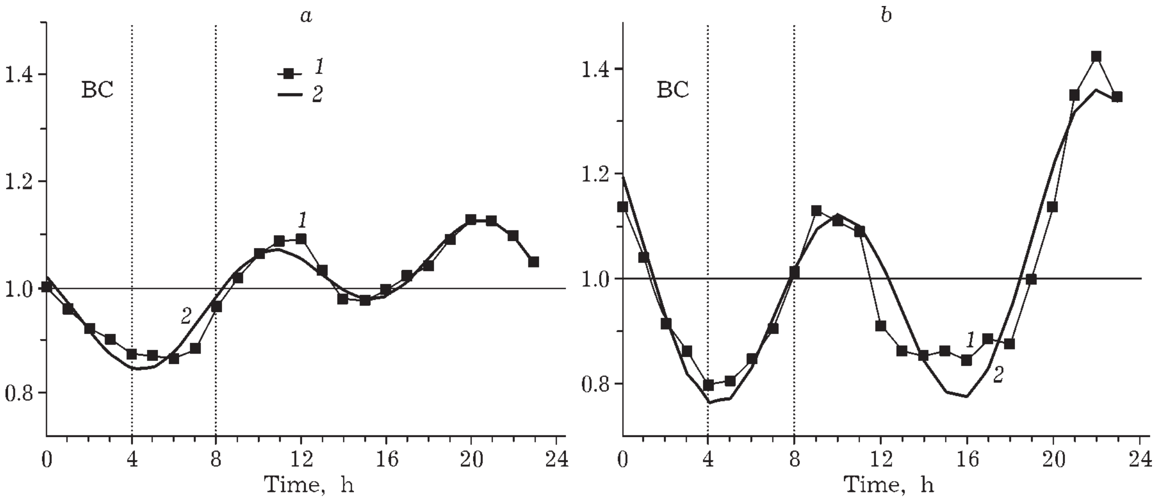

Fig. 14. Normalized seasonal average winter (*a*) and summer (*b*) diurnal profiles of the BC concentrations for 1997–2004 and their analytical approximation: *1* – measurement, *2* – approximation.

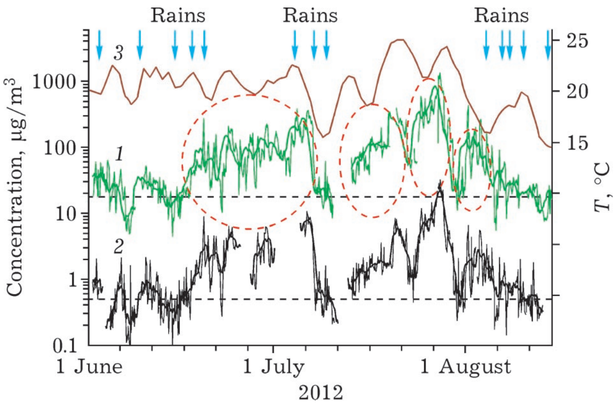

Fig. 15. Temporal behaviour of the submicron aerosol (*1*) and BC (*2*) and their smoothed over 30 hourly points, temperature (*3*) at the Fonovaya observatory. Dash straight lines show background levels (August 10–31).

July 12–20, July 23–27, and July 31–August 4. The maximal (since 1997) values of the mass concentrations of submicron aerosol and BC were observed on July 27 of 2012. The mass concentration of submicron aerosol of 1505.6 μg/m^3 and the BC mass concentration of 38.8 μg/m^3 were measured at 14:00 LT. The daily average values for the same day were 1212.8 and 26.6 μg/m^3, respectively [33].

To determine the contribution of the city to the concentrations of submicron aerosol and BC measured at the Aerosol Station, we conducted simultaneous aerosol measurements at the Fonovaya (Background) observatory located in the forest area 60 km west of Tomsk in different months of 2001, 2003, and 2011 [42]. In November 2013, the Fonovaya observatory of IAO of the SB RAS has started the monitoring measurements of the aerosol concentration (FAN-M angular nephelometer) and the BC mass concentration (MAAP 5012 aethalometer, Thermo Fisher Scientific Inc., the USA) to provide for simultaneous two-site observations in the near-surface air layer with the following comparison of the dynamics of aerosol composition under urban and background conditions.

The annual profile of the seasonal average mass concentrations of submicron aerosol and BC in 2014–2015 at the Aerosol Station is quite typical – with a minimum in summer and a maximum in winter (Fig. 16). However, some non-typical peculiarities appeared under background conditions. Thus, the seasonal average concentrations of aerosol in summer turned out to be higher than those in spring and fall, while for BC the summer and fall concentrations are close. In the annual profile, the differences in absolute values of the aerosol parameters under urban and background conditions are characterized by the typical seasonal behaviour – they are maximal in winter and minimal in summer. Thus, in winter, spring, summer, and fall, their values are 11.1, 8.2, 2.1, 6.4 μg/m^3 for aerosol and 0.83, 0.45, 0.42, 0.53 μg/m^3 for BC. The presence of these stable features provides the possibility of the analytical parameterization of the seasonal variability of the city's

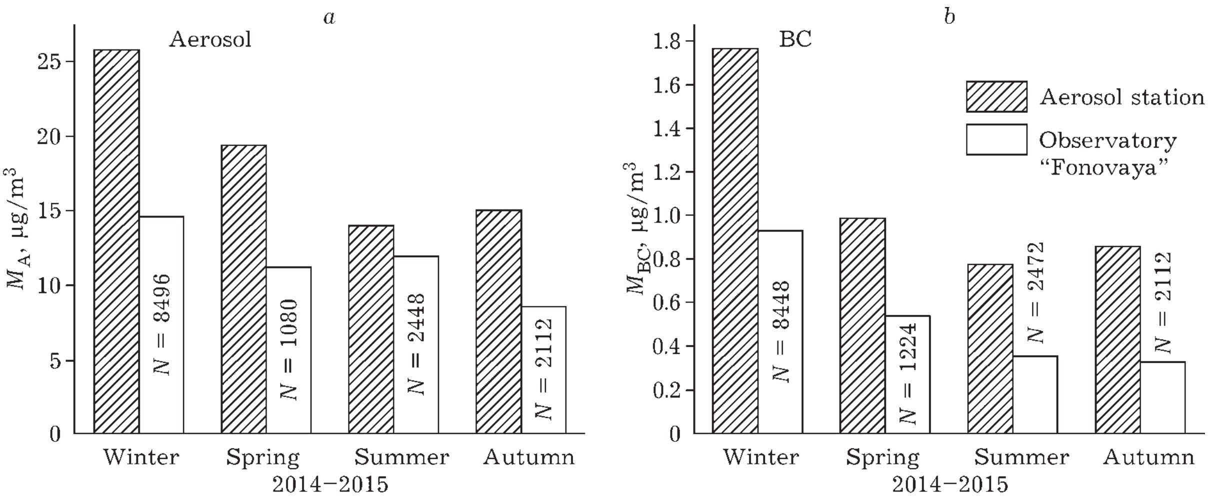

Fig. 16. Seasonal average mass concentrations of submicron aerosol (*a*) and BC (*b*) in Tomsk and under background conditions in 2014–2015.

contribution (difference concentrations of aerosol and BC).

However, relative differences in the seasonal behaviour are unstable. In the fall period, the BC content under the urban conditions is 2.6 times higher than under the background conditions, while in winter the difference is only 1.9 times.

CONCLUSIONS

This paper presents the detailed analysis of our longstanding investigations of the spatiotemporal variability of the aerosol and black carbon concentrations in the near-ground layer and in the troposphere of West Siberia and Russian Subarctic. The data of airborne sensing of the troposphere in the middle and subpolar latitudes have been used to study the ranges of variability, as well as general and distinctive features of the spatial variability of vertical profiles of the aerosol and BC concentrations, BC fraction, single scattering albedo, and concentrations integral over the atmospheric column.

Based on the results of the comprehensive analysis of the set of flights, we have proposed the classification of aerosol properties in the subarctic troposphere by the three characteristic states of the optical weather reflecting peculiarities of the spatial variability of the studied characteristics, namely, (1) high transparency of air and inverse vertical profiles in the subpolar latitudes, (2) strong or moderate effect of the smoke from forest fires on the aerosol composition in the warm season in the middle latitudes, and (3) slightly perturbed states of the regional mean aerosol background in the middle latitudes.

The latitudinal and longitudinal (west-to-east) dependences of the aerosol and BC concentrations have been revealed. It follows from the structure of the vertical profiles that in the northern regions of Siberia there are no significant sources of aerosol and Black Carbon (except for the period of forest fires) and the regional aerosol background at the vast areas of these regions is formed by the long-range transport of pollutants. The relation of the scattering and absorbing characteristics of aerosol suggests that the aerosol in the studied regions is now the cooling factor in all the seasons.

The original empirical model of vertical profiles of the optical and microphysical characteristics of tropospheric aerosol in West Siberia under conditions of the cloudless atmosphere that takes into account the absorbing and hygroscopic properties of particles in the calculation of radiation-significant characteristics of aerosol (SSA, radiative forcing) has been developed. The methodical approaches of the model can form a good basis for the development of a new model of subarctic aerosol.

Based on the many-year monitoring measurements at the stationary aerosol station, the stable peculiarities of the diurnal, seasonal, and year-to-year dynamics of aerosol and BC concentrations in the surface air have been revealed, and this opens up the possibility of their analytical parameterization.

Acknowledgements

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Многолетние исследования пространственно-временной изменчивости концентрации Black Carbon и аэрозоля в тропосфере Западной Сибири и Российской Субарктики

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Аннотация

В статье представлен обзор результатов самолетных и приземных исследований авторами пространственно-временной изменчивости концентраций аэрозоля и Black Carbon в тропосфере за последние два десятилетия. С 1999 г. в Институте оптики атмосферы (ИОА) СО РАН выполняется самолетное зондирование вертикальных профилей концентраций аэрозоля и ВС в тропосфере до высоты 7 км в регионах Западной Сибири и Российской Субарктики. С 1997 г. на Аэрозольной станции ИОА, расположенной в юго-восточной окрестности Томска, проводятся круглосуточные мониторинговые измерения концентраций аэрозоля и ВС в приземном слое воздуха.

По данным самолетного зондирования проанализированы общие и отличительные особенности пространственной изменчивости вертикальных профилей концентраций аэрозоля и ВС, относительного содержания ВС и альбедо однократного рассеяния в видимой области спектра. Впервые получены оценки интегральной концентрации ВС и аэрозольной оптической толщи рассеяния столба атмосферы до высоты 7 км. Проведена классификация характерных оптико-микрофизических состояний субарктической тропосферы для следующих условий: 1) высокой прозрачности воздуха в полярных широтах; 2) сильного воздействия дымов Сибирских лесных пожаров в теплые сезоны года; 3) среднего регионального аэрозольного фона атмосферы в средних широтах. С ростом широты в интервале 55–75.2° с.ш. происходит уменьшение приземных концентраций аэрозоля и ВС в среднем в 3 раза. В приполярных широтах проявляется тенденция убывания концентраций в направлении с запада (Карское море) на восток (Восточная Субарктика). Впервые разработана обобщенная эмпирическая модель тропосферного аэрозоля Западной Сибири в условиях безоблачной атмосферы, позволяющая с учетом поглощающих и гигроскопических свойств частиц определять среднесезонные вертикальные профили альбедо однократного рассеяния, необходимые для оценок радиационного форсинга аэрозоля.

Изучены основные особенности суточной, сезонной и межгодовой изменчивости концентраций аэрозоля и ВС, относительного содержания ВС в приземном слое воздуха и проведена параметризация этих зависимостей.

Ключевые слова: аэрозоль, Black Carbon, пространственно-временная изменчивость, тропосфера, вертикальные профили, эмпирическая модель тропосферного аэрозоля, Российская Субарктика