DIFFERENCES in seasonal average concentrations of aerosol and Black Carbon and particle size distributions from the data of monitoring in Tomsk and under background conditions in 2014-2015

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ABSTRACT

The mass concentrations of the dry basis of aerosol and Black Carbon (November 2013 – December 2015) and the volume particle size distributions (2015) measured in the monitoring mode under background conditions (Fonovaya (Background) Observatory, 60 km west of Tomsk) and under urban conditions (Akademgorodok District of Tomsk) have been used to determine the average seasonal contribution of the city to the aerosol characteristics. It has been found that the annual profile of the seasonal average city contribution to the aerosol and Black Carbon (BC) contents, as well as the volume fill factor of the submicron aerosol, has a maximum in winter and a minimum in summer. In the coarse size range, the maximal excess of the volume fill factor was observed in the spring period. The observed differences between the city and the background conditions for the relative content of Black Carbon in aerosol and the absorption coefficient of the particulate matter were largest in summer and smallest in winter. Under the background conditions, the annual behavior had an untypical feature, namely, the seasonal average aerosol concentrations in summer exceeded the values obtained for the spring and autumn seasons. Analytical parameterization of the annual profile of monthly average difference of the urban and background mass concentrations of aerosol and Black Carbon in the form of parabolic dependence has been proposed.

Keywords: surface layer, submicron aerosol, Black Carbon concentration, absorption coefficient, particle size distribution.

1. INTRODUCTION

The problem of anthropogenic impact of a city on regional average background values of atmospheric aerosol parameters is actively studied in recent years^{1.4}. The Aerosol Station and TOR Station of IAO SB RAS (Akademgorodok, southeast suburbs of Tomsk) conduct monitoring measurements of optico-microphysical characteristics of aerosol in the atmospheric surface layer for many years. To determine the city contribution to the concentrations of submicron aerosol and Black Carbon measured at the Aerosol Station of IAO, we have carried out simultaneous expedition aerosol measurements at the Fonovaya Observatory located in the forest zone 60 km west of Tomsk in different months of 2001, 2003, 2011⁵. According to the wind rose, the prevailing wind directions in this area are southwestern and southern. Thus, this observation site can be considered as background with respect to Tomsk. In November of 2013, the Observatory was equipped with instrumentation for background aerosol measurements in the monitoring mode.

In this paper, we analyze the data of simultaneous two-site measurements and estimate the city contribution to: (1) seasonal average concentrations of aerosol and Black Carbon (BC) in 2014-2015; (2) relative mass content of Black Carbon in aerosol and the absorption coefficient of the dry basis of particulate matter; (3) seasonal average volume fill factors of submicron and coarse particles measured at the TOR station and in the Fonovaya Observatory in 2015. The obtained monitoring results will be used for development of approaches to parameterization of the seasonal profile of anthropogenic impact of the city.

2. INSTRUMENTATION AND PARAMETERS UNDER STUDY

At the Aerosol Station (Tomsk), the angular aerosol scattering coefficient of the dry basis of particles μ (45°) (Mm⁻¹sr⁻¹) is measured round-the-clock by the FAN-A nephelometer at a wavelength 0.51 μ m since 1997. The data on the angular

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scattering coefficient are used to determine the mass concentration of submicron aerosol M_A (µg/m³)⁶. Simultaneously, the Black Carbon concentration M_{BC} (µm/m³) is measured by the MDA-02 aethalometer developed by IAO SB RAS⁷, whose operating principle is analogous to that described in⁸.

In the Fonovaya Observatory, the aerosol concentration was measured by the FAN-M nephelometer⁹, while the Black Carbon concentration was measured by the MAAP 5012 aethalometer (Thermoscientific Fisher Inc., USA). The particle size distribution was recorded every hour at the TOR Station with the GRIMM optical counter of particles (model 1.109, Germany), which provides measurements in the size range 0.25-32 μ m, and at the Fonovaya Observatory by an optical counter (model 1.108) in the range 0.3-20 μ m. For all the measured characteristics, the seasonal average and monthly average values were analyzed.

Realizations affected by plumes from forest fires were rejected from the data arrays. This approach is caused by the fact that forest fires are powerful sources of particles, whose optical-microphysical properties differ widely from the properties of the background natural and anthropogenic aerosols¹⁰.

In addition to the measured parameters, the relative BC mass content in aerosol $P = M_{BC} / M_A$ and the absorption coefficient of the dry particulate matter $\chi = P_{V*}\chi_{BC}$, where P_V is the relative BC volume content in aerosol and $\chi_{BC} = 0.74$ is the absorption coefficient of the BC matter, were analyzed¹¹. The seasonal average volume fill factors of submicron and coarse particles were calculated and compared with each other.

3. RESULTS OF MEASUREMENTS

3.1. Mass concentrations of submicron aerosol and Black Carbon. The annual profile of the seasonal average mass concentrations of aerosol and Black Carbon measured in 2014-2015 at the Aerosol Station has a typical form with a minimum in summer and a maximum in winter (Fig. 1). However, under background conditions, some nontypical features showed themselves. Thus, the seasonal average aerosol concentrations in summer appeared to be higher than in spring and in autumn, while for BC the summer and autumn concentrations were close.

It should be noted that the relative concentration differences in the annual profile also is not stable and deviate from the typical seasonal behavior. Thus, if in the autumn period the urban BC content exceeds the background level 2.6 times, then in winter this excess decreases down to 1.9 times.

In the following, when describing differences in the aerosol parameters under urban and background conditions, we consider differences of the seasonal average mass concentrations for aerosol $\Delta M_A = M_A$ (urban) - M_A (background) and for BC $\Delta M_{BC} = M_{BC}$ (urban) - M_{BC} (background), which are conditionally referred to as the city contribution. The annual profile of the city contribution is close in its shape to the annual profile of the concentrations under the urban conditions, also having a maximum in winter and a minimum in the summer season (Fig. 2). During a year, the seasonal average concentration differences vary as $\Delta M_A = 2.1 \div 11.1 \text{ µm/m}^3$ (5.3 times) and $\Delta M_{BC} = 0.42 \div 0.83 \text{ µm/m}^3$ (2 times).



Figure 1. Seasonal average mass concentrations of submicron aerosol (a) and Black Carbon (b) in Tomsk and under background conditions in 2014-2015.



Figure 2. Difference of the seasonal average mass concentrations of submicron aerosol (a) and Black Carbon (b) in Tomsk and under background conditions in 2014-2015.

3.2. Relative BC mass content in aerosol and absorption coefficient of dry particulate matter. The annual profile of the relative BC content in aerosol is different for the two measurement sites (Fig. 3). At the both sites the maximum was observed in winter, while the minimum took place at the Aerosol Station in spring and at the Fonovaya Observatory in summer. As a result, the profiles of *P* differ most widely in summer ($\Delta P = 0.02$) and are closest in winter ($\Delta P = 0.01$). The annual profile of the absorption coefficient of the dry particulate matter χ under the urban and background conditions is analogous to the annual profile of *P* with the maximal difference between the Aerosol Station and the Fonovaya Observatory in summer ($\Delta \chi = 0.013$) and minimal in the winter period ($\Delta \chi = 0.002$). The differences of the seasonal average values of *P* and χ for all the seasons are confident by the Student's criterion with a probability of 95%.



Figure 3. Seasonal average values of the relative BC mass content in aerosol (a) and absorption coefficient of the dry particulate matter (b) in Tomsk and under background conditions in 2014-2015.

3.3. Fill factors of particles in the submicron and coarse ranges. Seasonal average particle volume size distributions at the two measurement sites in 2015 appeared to be close. Figure 4 shows the distributions for the two contrast seasons: winter and summer. The seasonal average distributions were used to calculate the seasonal average fill factors of particles in the submicron and coarse size ranges (Table 1). The largest difference of the fill factor of submicron particles between two measurement sites was observed in winter (0.69 μ m³cm⁻³), while the smallest one took place in summer (0.04 μ m³cm⁻³). In the winter period, the differences of the fill factors of submicron particles are confident by the Student's criterion with a probability of 95%, while in summer the differences are not confident. The relative differences have a different annual behavior with a maximum in autumn (1.8 times) and a minimum in the summer period (1.03 times).

Other pattern is observed for the coarse particles. In Tomsk, the fill factor for the coarse particles was maximal in the spring period, while at the Fonovaya Observatory it was maximal in summer. In the coarse range, the largest absolute $(8.4 \,\mu\text{m}^3\text{cm}^{-3})$ and relative (3.1 times) differences were obtained in spring.



Figure 4. Average volume size distributions of particles in Tomsk and under background conditions in winter (a) and in summer (b) in 2015.

Table 1. Seasonal average fill factors of aerosol particles (μm^3	cm ⁻³), their differences and ratios at the TOR station (Tomsk) and in the
Fonovaya Observatory in the submicron and coarse ranges in 2	.015.

Ranges	Seasons	TOR station	Observatory	City- background difference	City- background ratio	Number of measurements
	winter	3.03 ± 3.46	2.34 ± 1.83	0.69	1.3	3720
Submicron range	spring	1.5 ± 0.77	1.19 ± 0.96	0.31	1.3	1440
(0.3-1 µm)	summer	1.33 ± 0.79	1.29 ± 1.14	0.04	1.03	2088
	autumn	1.29 ± 0.71	0.73 ± 0.53	0.56	1.8	1464
	winter	2.88 ± 2.23	2.15 ± 1.77	0.73	1.3	3720
Coarse range	spring	12.4 ± 37.1	4.0 ± 8.54	8.4	3.1	1440
(1-10 µm)	summer	4.71 ± 4.0	4.98 ± 4.69	-0.27	0.9	2088
	autumn	5.41 ± 5.61	2.88 ± 2.8	2.53	1.9	1464

3.4. Analytical parameterization of the annual profile of the city contribution to the aerosol and BC concentrations in the approximation of parabolic dependence. The measurements obtained in section 3.1 were used as an example for analytical parameterization of the annual profile of the city contribution. Figure 5 shows the annual profile of the "city-background" concentration differences for the monthly average BC concentration and the concentration of submicron aerosol. February 2014 was rejected from the calculations, because urban winter smog¹² with extreme values of optical characteristics was observed for the most part of this month. The figures show that the seasonal variability of the concentration differences is characterized by a maximum in winter and a minimum in the summer period. In the profiles, we can see local extremes (fine structure), which are likely caused by the influence of nonsynchronous variations of different variability factors under the urban and background conditions (precipitation, local sources of aerosol and black carbon, nonsynchronous alternations of air masses at the two observation sites, and so on).



Figure 5. Analytical parameterization of the annual profile of the city contribution to the monthly average BC concentrations and mass concentrations of the near-surface submicron aerosol from the results of measurements at the aerosol Station of IAO (Tomsk) and the Fonovaya Observatory in 2014-2015.

The figure illustrate the possibility of approximation of the obtained experimental dependences of the annual profile of the city contribution by nonlinear curves shaped as a parabola $Y=A+B*X+C*X^2$, where the number of a month N_i is used as an input parameter. The analysis has shown that the proposed approach allows us to describe the main peculiarities of the average annual profile: the presence of the summer minimum and winter maximum of the concentration differences. As can be seen, in this case, the left and right "branches" of the approximation curves are symmetric and smooth the fine structure of seasonal dependences at the sections from winter to summer and from summer to next winter.

In Fig. 5 the approximation curves are accompanied by the average values of standard deviations calculated for the difference of monthly average reconstructed and experimental concentrations for 2 years of measurements. This value of standard deviation characterizes an error of the parabolic approximation for reconstruction of values of the city contribution. As can be seen, the differences between the measured and reconstructed values of the city contribution to the aerosol and BC concentrations mostly fall within the standard deviation.

For the BC concentration, the proposed approximation allows us to obtain satisfactory results of the estimates: the standard deviation is relatively small $\sim \pm 77 \text{ ng/m}^3$, thus the relative errors of reconstruction with the approximating formula is about 8.5% for the winter concentration differences and about 19% for the summer differences.

The higher errors of reconstruction appear at the application of the parabolic approximation for the description of the average annual profile of the city contribution to the aerosol concentration (Fig. 5,b). In this case, the annual average value of the standard deviation ~ \pm 1.9 µm/m³ corresponds to the relative error of reconstruction of about 15.8% for estimates under the winter ($\Delta M_A \sim 12 \mu m/m^3$) conditions. However, in the summer period (at low $\Delta M_A \sim 3 \mu m/m^3$), the error of reconstruction increase significantly, and in July-August it can lead to the 1.6-fold excess of the reconstructed city contribution to the aerosol concentration over the measured one.

To improve the analytical approximation of the annual profile of the city contribution to the BC and aerosol concentrations, it is necessary to accumulate the longer series of monitoring measurements under the urban and background conditions, as well as, to study possible causes for the appearance and significance of the fine structure in the annual profile.

4. CONCLUSIONS

The two-year simultaneous monitoring measurements of the submicron aerosol and BC concentrations in the atmospheric surface layer under the urban (Akademgorodok of Tomsk) and mean regional background conditions (Fonovaya Observatory) have been conducted for the first time in the region of Western Siberia for all seasons. The results of two-site measurements have been used to analyze peculiarities of the annual profile of the city contribution to the aerosol concentrations.

It has been found that the annual profile of the city contribution to the aerosol and BC content and to the volume fill factor of aerosol particles in the submicron size range has a maximum in winter and a minimum in summer.

In the annual profile of the seasonal average values of the volume fill factor of coarse aerosol particles, the city influence was strongest in the spring season.

The "city-background" differences for seasonal average values of the relative BC mass content in aerosol and the absorption coefficient of the particulate matter were maximal in summer and minimal in winter.

The annual profile of the city contribution to the BC and aerosol concentrations can be well approximated by the parabolic dependence.

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