

Investigations of the Absorption Properties of Near-Ground Aerosol by the Methods of Optical-Acoustic Spectrometry and Diffuse Extinction

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Introduction

Aerosol absorption is an important factor in the formation of non-selective radiation extinction in the visible wavelength range, and plays a great role in solving many radiative and climatic problems. The principal absorbing substance in atmospheric aerosol is soot (crystal carbon), which strongly affects the atmospheric transparency, albedo of clouds, and snow cover. The non-selective absorption by finely dispersed soot aerosol is considered to be one of the most plausible reasons for the appearance of so-called “surplus absorption” of the shortwave (SW) optical radiation in clouds and cloudless atmosphere. Investigations of the spectral dependencies of the efficiency factor of absorption of the SW optical radiation by soot particles is of significant interest (Moosmüller et al. 1998; Bergstrom et al. 2002). The instruments designed for investigating the aerosol absorption properties and the results of synchronic measurements of the spectral values of the aerosol absorption coefficient and the mass concentration of soot in the near-ground air layer are discussed in this paper.

Methods and Instrumentation for Measurements

Both the optical acoustic spectrometry method and the diffuse extinction method of light by a layer of particles at their sampling on an aerosol filter are considered now as a perspective for highly accurate real-time measurements of the characteristics of absorbing aerosol.

The pulse optoacoustic spectrometer with a big-size cell was created for measurements of the aerosol absorption coefficients in the visible wavelength range (Tikhomirov et al. 2001). The block-diagram of the device is shown in Figure 1. The spectrometer contains the pulse ruby and Nd:Yag lasers, optoacoustic detector with blocks for amplification and recording the signal shape, the calorimeter for measuring the laser pulse energy, the system for measuring the laser radiation spectrum, the vacuum post for pumping out the detector cell, and the pump for sampling atmospheric air.

The use of an absorbing big-size cell in the optoacoustic detector (30 cm long, 20-cm-diameter cylinder) makes it possible to determine spatial-temporal resolution of optoacoustic signals of different origin. So, the time-resolved optoacoustic (TROA) cell allows you to exclude the contribution of adjacent noises, molecular and aerosol scattering into the optoacoustic absorption signal. Finally, the signal of aerosol absorption or the signal of absorption by the gas under study is reliably selected and detected.

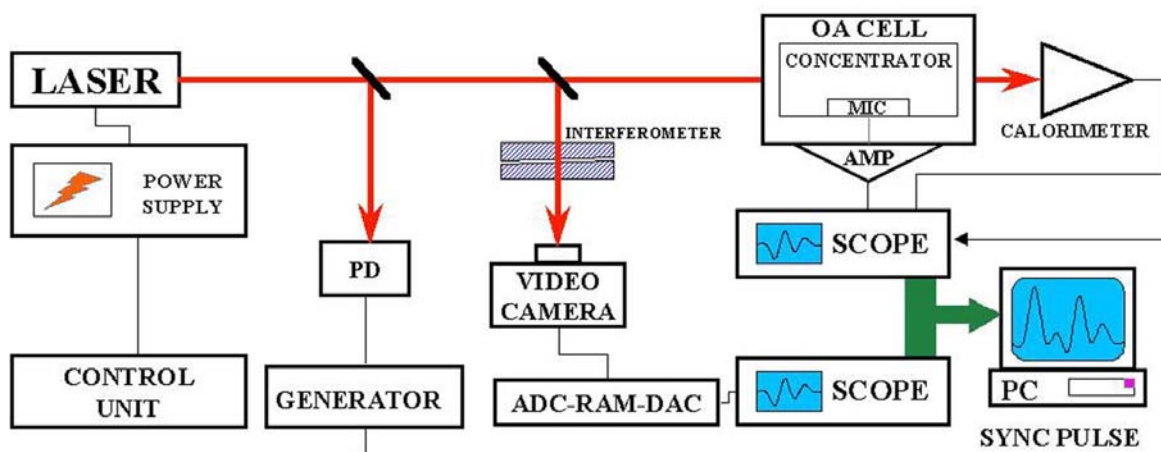


Figure 1. Block-diagram of the TROA spectrometer.

The use of elliptic and parabolic reflectors for focusing of the pressure pulses (energy concentrators) and the specially designed microphone allowed us to increase the energy potential of the spectrometer by more than one order of magnitude and to measure the aerosol absorption coefficient by means of only one laser pulse. A sensitivity to the absorption coefficient of about $10^{-6} \text{ km}^{-1} \text{ J}^{-1}$ was reached. The spectrometer was calibrated using the known resonance absorption of the ruby laser radiation by water vapor in the center of the absorption line 694.38 nm. Comparison of the data of laboratory measurements of the absorption coefficient at the wavelength of $\lambda=694.30 \text{ nm}$ by the binary gas mixture of H_2O and N_2 with the results of calculation of the molecular absorption using the database of spectral data HITRAN-96 has shown their good agreement and then has confirmed the correctness of the calibration of the device. The developed TROA spectrometer is capable of measuring the spectral values of the aerosol absorption coefficient at the wavelengths of the used pulse lasers with a mean error of about 20%.

The mass concentration of soot in the composition of aerosol particles M_S ($\mu\text{g}/\text{m}^3$) was recorded in real time by the aethalometer based on the method for measuring the diffuse attenuation of light by a layer of aerosol particles during their settling on an aerosol filter analogously to Hansen et al. 1984. The developed system for amplification of processing the signals using the pulse mode of operation with accumulation of signals and transforming them into frequency, subtracting of dark levels, etc., provides for the increased sensitivity and stable measurements. The device was calibrated with a mean error of about 20% (Baklanov et al. 1998).

Measurements and Results

Synchronic measurements of the aerosol absorption coefficient $k_\lambda(\text{cm}^{-1})$ at the wavelengths of 532, 694, and 1064 nm and the mass concentration of soot M_S ($\mu\text{g}/\text{m}^3$) were carried out in winter 2000-2002 in the near-ground layer of air by means of the TROA spectrometer and the aethalometer. Instrumentation was installed in the working room of the institute building at the eastern outskirts of the city of Tomsk. Atmospheric air was delivered to the cells of the devices through the hoses of the samplers situated at a height of 3 m above the ground surface. Measurement cycles were carried out with a mean periodicity of about 20 minutes.

The temporal behavior of the aerosol absorption coefficient at a wavelength of 532 nm and the mass concentration of soot are shown in Figure 2. The results of measurements show that the data on the absorption coefficient and the mass concentration of soot obtained by two different methods are in good agreement between each other and synchronically follow the fine peculiarities of the variations of absorbing aerosol. High correlation between investigated characteristics at each from three wavelengths is observed; the values of the correlation coefficients are more than 0.90 (Figure 3). The close

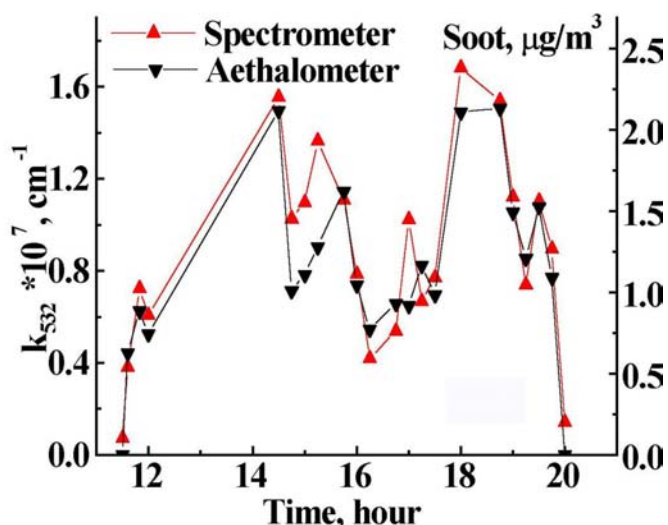


Figure 2. Temporal behavior of the aerosol absorption coefficient at 532 nm and of the mass concentration of soot (near-ground layer, winter - spring seasons).

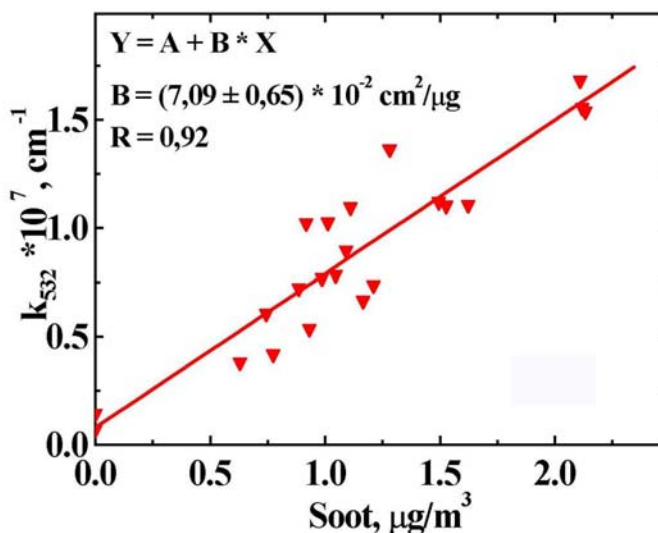


Figure 3. Correlation between the values of the mass concentration of soot and of the aerosol absorption coefficient at 532 nm.

correlation between independent data on the spectral absorption coefficients and the concentration of soot is evidence of the consistency and correctness of the experimental data obtained by each method. It is also an additional confirmation of the important role of soot in formation of absorption in the visible wavelength range.

Approximation of correlation diagram of the absorption coefficient and the mass concentration of soot enable us to determine the values of the absorption efficiency factor at three wavelengths (Figure 4) and to estimate the spectral dependence of this parameter. The values of the absorption efficiency factor obtained in the series of measurements are $\sigma \approx 7 \text{ m}^2\text{g}^{-1}$ at 532 nm, $\sigma \approx 5 \text{ m}^2\text{g}^{-1}$ at 694 nm, and $\sigma \approx 3 \text{ m}^2\text{g}^{-1}$ at 1064 nm. Spectral behavior of the mean values, σ , is well approximated by the inversely proportional dependence on the wavelength ($\sim \lambda^{-1}$) (Figure 4). The obtained estimate of the shape of spectral dependence of the absorption efficiency factor is in agreement with the data of (Bergstrom et al. 2002).

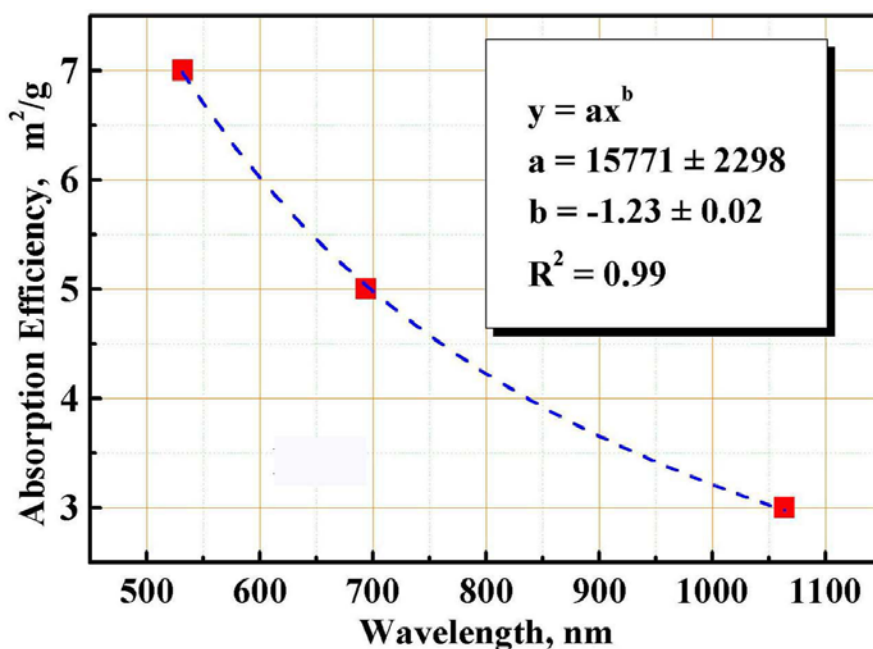


Figure 4. Estimates of the aerosol absorption efficiency factor as function of wavelength (n - measurements; curve – fitting; near-ground layer, winter).

Conclusion

Simultaneous measurements of the aerosol absorption coefficient in visible wavelength range and the mass concentration of soot were carried out in the near-ground air layer during winter-spring period by means of the TROA spectrometer and aethalometer. The results of measurements have shown that the data obtained using the two methods are in good agreement. The high correlation connection of the absorption coefficients and the soot content made it possible to calculate the spectral values of the absorption efficiency factors corresponding to three wavelengths. The spectral behavior of the obtained

mean values of absorption efficiency factors, depending on the wavelength, is described closely to λ^{-1} . The results confirm supplementary evidence of the decisive role of the soot component in formation of the aerosol absorption in the visible spectral range.

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