Journal: AMT
Title: An empirical model of optical and radiative characteristics of the tropospheric aerosol over West Siberia in summer
Author(s): M.V. Panchenko et al.
MS No.: amt-2011-145
MS Type: Research Article
Special Issue: Remote sensing of aerosols and clouds (EGU 2011)

Reply to the comments made by Review#2

Authors are very grateful to the reviewer for so thorough analysis of the text. All general comments are taken into account in the revised manuscript The structure of the manuscript is changed, and all necessary additions and explanations are made.

General Comments:

1. The grammar needs to cleaned up considerably prior to publication. There were too many individual grammatical errors to be listed in this review.

We agree with this remark. Before final submission, we will send the text to http://languageediting.nature.com/ for the language check.

2. The abstract should be expanded to include some specific results and details. Currently, it is difficult to glean any real information from the abstract.

The abstract will be expanded.

3. *I'm not sure what is meant in the last line of the abstract that the regional features of West Siberia are taken into account. This should be clarified or removed.*

First, the empirical model of the vertical profiles of the aerosol scattering coefficient is based on the data of airborne sensing in West Siberian region (Tomsk, Novosibirsk, Kolpashevo, Kargasok, Omsk, Kemerovo). The total data array consists of more than 600 vertical profiles (the summer subarray includes more than 150 profiles).
Nevertheless, the main bulk of data analyzed in this paper were obtained during 2.5 years at different frequency of observations for different synoptic and meteorological situations. Therefore, before making analysis of the aerosol characteristics observed, it is necessary to assess the geophysical significance and the quality of the experimental data. Since this is a key question while there are no data of long–term observations of vertical profiles of aerosol characteristics for this region, let us try to assess representativeness of the bulk of data on hand from the standpoint of the standard meteorological and synoptic parameters.
Within each season the data were classified with respect to the type of air mass, pressure field, and weather conditions. The frequency of occurrence of different air masses and pressure field types were compared with the long-term average data (Koshinskii, S.D., Trifonova, L.I., and Shver, T.A. (eds.) Climate of Tomsk. Gidrometeoizdat, Leningrad, 1982)
Analogously we have compared vertical profiles of measured meteorological parameters with the average long–term data of aerological sensing averaged over the stations situated in the region under investigation (Aleksandrovskoe, Barabinsk, Novosibirsk, and Omsk).
Good agreement of the analyzed synoptic and meteorological characteristics of the atmosphere during the period of our observations with the climatic average for our region allows us to hope that the bulk of data obtained on the optical parameters of submicron aerosol and the factors of its

variability discussed in the papers reflect the most general features that are characteristic of the region as a whole.

4. The introduction should be expanded to include a greater review of the current literature regarding the subject.

The "*Introduction*" part will be expanded, and brief review of other publications on aerosol vertical profiles will be added.

5. *Even though references are given for the methods, it would be nice to have some information – even if it's just a list of the instrumentation and measurements, etc. in a table.*

Brief description of the instrumentation is added to text.

6. The first paragraph of section 2 refers to methods (although this should be expanded – see above comment), but the rest of this section should be combined with the introduction section.

The structure of the manuscript is changed.

7. *The discussion of the results of the radiative flux simulations should be expanded. It would be nice to see summertime radiative forcing calculations using the empirical model input parameters and how these compare to current estimates over this region.*

In this paper we did not have an object to completely study the radiative effects of aerosol taking into account regional peculiarities of West Siberia on the basis of the proposed model. We planned to show the possibilities of the use of the model of the vertical profiles for calculation of the broadband fluxes of solar radiation. In this connection, we carried out several numerical experiments, the results of which are briefly described in section 4.2. In addition, the estimates of the aerosol direct radiative effect are obtained. The daily average values of the short-wave component of the direct radiative effect (DRE) of aerosol for the parameters described in section 4.2 are as follows: (-15.1) W/m^2 and (-6.8) W/m^2 at the bottom and top of the atmosphere, respectively.
The "Discussion" section will be added by discussion of how problematic is comparison of the aerosol radiative effect obtained using our model with current estimates over this region.
Comparison of the aerosol radiative effects obtained using other models is rather problematic for the territory of West Siberia. One of few sources of the data on the columnar aerosol optical characteristics is the data obtained using the Dubovik-King algorithm [King and Dubovik, 2000] for the AERONET site located in the city of Tomsk (http://aeronet.gsfc.nasa.gov). However, the data array collected since 2002 (in particular, the complex refractive index and the single scattering albedo) at the Level 2.0 characterizes the aerosol state only under conditions of enhanced turbidity of the atmosphere ($\tau_a(0.44\ \mu m) \geq 0.4$), caused mainly by forest fires.
Taking into account that the use of AERONET data for the typical summer conditions of West Siberia ($\tau_a(0.5\ \mu m) \sim 0.16$) requires additional study, at this stage we have considered 3 models of continental aerosol: OPAC clean, OPAC average and OPAC polluted, [Hess et al., 1998]. The wide range of variations of the single scattering albedo and the asymmetry factor presented in these models makes it possible to describe the sufficiently large number of situations which can be observed in the atmosphere. Calculations of daily average values DRE performed for $\tau_a(0.5\ \mu m)=0.16$ have shown that the range of DRE at the use of three aforementioned models is (-16.5) - (-14.6) W/m^2 and (-7.8) – (-5.0) W/m^2 at the bottom and top of the atmosphere, respectively.
The estimates of DRE obtained using our empirical model and OPAC continental aerosol model are in good agreement. At the same time, one should note that we compared the aerosol radiative

effects only at the boundaries of the atmosphere. The majority of aerosol models, and the data obtained by AERONET, on the contrary to the model of the vertical profiles of the aerosol characteristics, do not enable to obtain the data on the change of the upwelling and downwelling radiative fluxes inside the atmosphere and to more adequately estimate, for example, the change of the cooling rate.

Specific Comments:
Items 1, 6, and 7 concern technical corrections of the text, and are taken into account in the revised manuscript.

2. *I don't think that just because the scattering profiles for the two time periods agree (if they even do – see comment above), the aerosol absorption properties necessarily would as well, as I believe is being stated starting on line 11, section 3.1, pg. 141. Also, where does the value Maer come from without mass concentration measurements of the aerosols?*

The aerosol mass concentration M_{aer} was calculated from the data on the angular aerosol scattering coefficient. Existence of close correlation between the volume concentration and the scattering coefficient in atmospheric hazes was revealed for the first time from experimental data. Then, corresponding numerical calculations were carried out for the wide set of lognormal distributions, which well describe the haze microstructure.
Physical justification of the existence of this correlation are the following properties of scattering particles.
a) if consider real submicron particle size distributions in atmospheric hazes, one will see that the main part of the number concentration lies in the radius range less than 0.5 μm.
b) hence, for the wavelength of 0.5 μm, all these particles are located to the left from the first diffraction maximum of the scattering efficiency of particles with the refractive index from 1.33 to 1.5.
c) let us pay attention to the fact that the strongest changes of the particle properties at the change of the size or the wavelength of the incident radiation are characteristics in the vicinity of the first maximum. For the particles of size less than the wavelength, one can select the range in the curve $K(\rho, m)$ where $\rho = 2\pi r/\lambda$, $K(\rho, m)$ is directly proportional to ρ (i.e. r, or λ^{-1}). For particles with ρ from 1 to 4, the linear dependence describes the behavior of $K(\rho)$ with high accuracy.
Hence, for such particles at the certain wavelength of radiation $K_p(\rho) \sim r$, and the formula for the scattering coefficient can be written as follows:

$$\mu \approx \int_{\rho=1}^{\rho=4} r\pi r^2 f(r)dr \sim V \quad ,$$

where V is the volume of scattering particles in a unit volume of the medium. The presence of linear correlation between the angular scattering coefficient μ and the volume of scattering particles V makes it possible to use it for various estimates.
In the present paper, in order to illustrate the ratio between the mass concentrations of black carbon and aerosol, we calculated the so-called BC fraction $P = M_{BC}/M_{aer}$. Here M_{aer} is conditional mass concentration of dry aerosol, M_{aer} is proportional to the angular scattering coefficient: $M_{aer}=\mu(45°)\times K\times\rho_{aer}$, where K is an empirical coefficient connecting the angular scattering coefficient and the volume concentration of aerosol particles, ρ_{aer} is the density of aerosol matter. We assume $\rho_{aer}=1.5 g/cm^3$.
One should note that this approach did not lead to the errors in calculating the mass of black carbon used for subsequent calculations of the optical constants, because only the measured parameter μ(45°) was used for calculations.
But, in practically all the publications, we have to use terms "volume concentration" and "mass concentration", because the "language" of scattering coefficients does not enable us to bring our

conclusions to specialists in the atmospheric sciences and to give them the possibility to compare our results with the results of other (not optical) measurements.

3. *Section 3.1, line 23, pg. 141: what does 0.05-0.7 μm represent? The aerosol diameter? This should be clarified.*

It is the aerosol radius. It will be indicated in the text.

4. *Section 3.1, line 5, pg. 142: what is the meaning of the following, 0.2 (0.3)–5 μm? Why is 0.3 in parentheses?*

Depending on the problem, the solar radiation fluxes can be calculated for different wavelength ranges (0.2 - 5 μm), (0.3 - 5 μm), (0.3 - 2.8 μm), etc. In this paper we simulate the fluxes in the range (0.2- 5 μm). The text will be changed, (0.2(0.3)-5.0 μm) will be replaced by (0.2-5.0 μm).

5. *What aerosol diameters are capable of being sampled through the aircraft aerosol inlet? Again, this information should be tabulated and included in this paper. It is briefly mentioned in the last paragraph of section 3.1 that larger particles are not sampled as well. What is the collection efficiency with size? Has this been measured?*

Samples to be analyzed were collected continuously by direct airflow. Here the principle technique moment is to satisfy the isokinetic condition. To do this, it is necessary to satisfy a number of requirements to the geometry of the collecting device. The sampling device we used was made of a thin–walled tube with inner diameter of 10 mm placed in the forepart of the aircraft in front of the propellers at a distance about 30 cm from the fuselage, i.e., in the undisturbed zone. The radius of the collecting tube curvature was chosen so that to avoid the inertial sedimentation of particles with the size $d \leq 10$ μm on its walls.
Possible changes of the particles number concentration when moving along the collection path are connected firstly with their diffusion and sedimentation on the walls, and secondly with the inertial settling on the wall bends that are not completely avoided in the construction of the air transport path. Estimates made according to [Fuchs, 1955; Green and Lane, 1964] show that the loss of particles of submicron size did not exceed 1% but it can reach ten percent for the particles with the size $d \geq 10$ μm.
The particle size distribution function was measured with a AZ-5 photoelectric aerosol counter in the radius range 0.2 to 5 μm.
The mass concentration of absorbing substance (black carbon) was measured by an aethalometer, in which the method is realized of measuring the diffuse extinction of light by a layer of particles during their sedimentation from airflow to a filter. The threshold of detection of the instrument is about 0.01 $\mu g/m^3$. The instrument was calibrated in absolute units by means of comparison of the data of synchronic optical and gravimetric measurements of the concentration of black carbon. The black carbon particles were produced by a special pyrolysis generator.

Fuchs, N.A.: The Mechanics of Aerosols. Pergamon, Oxford, 1964.
Green, H.L., and Lane, W.H.: Particulate Clouds: Dusts, Smokes, and Mists. Spon., London, 1964; Van Nostrand, Princenton–New York, 1964.

8. *The literature on which the assumption is based that 90% of black carbon is in the submicron fraction and 10% is in the coarse fraction seems rather limited. I have a hard time believing that assumption based on such limited references.*
The next discussion item is related with our choice of the relationship between BC fraction in submicron and coarse range.

Our photoacoustic measurements [Tikhomirov et al, 2005] of the spectral dependences of the aerosol light absorption in the near-ground layer of the atmosphere showed that, with quite high accuracy, this dependence on wavelength is close to λ^{-1} and, hence, also is evidence of the fact that the main mass of absorbing substance is concentrated in fine particles.
At the same time, when accepting the ratio of black carbon content in submicron and coarse aerosol as 90 to 10%, respectively, first of all, we followed the data of Gelencsér, 2004, Hitzenberger and Tohno, 2001; Höler et al, 2002; Delene and Ogren, 2002], [Höler et al, 2002].
Taking into account the methodical remarks, let us note that the main assumptions made when developing the empirical model do not contradict to the data on the aerosol properties in the troposphere.

Delene, D. J. and Ogren, J. A.: Variability of aerosol optical properties at four North American surface monitoring sites, J. Atmos. Sci., 59, 1135–1150, 2002.
Gelencsér, A. Carbonaceous particles (Springer, 2004)
Hitzenberger, R., and Tohno, S.: Comparison of black carbon (BC) aerosols in two urban areas - concentrations and size distributions, Atmos. Environ., 35, 2153-2167, 2001.
Höler, R., Tohno, S., Kasahara, M., Hitzenberger, R.: Long - term characterization of carboneous aerosol in Uji, Japan, Atmos. Environ., 36, 1267-1275, 2002.
Tikhomirov A.B., Firsov K.M., Kozlov V.S., Panchenko M.V., Ponomarev Y.N., Tikhomirov B.A. Investigation of spectral dependence of shortwave radiation absorption by ambient aerosol using time-resolved photoacoustic technique. Optical Engineering. 2005. V.44. №7. P.071203 1–11.

9. *Section 3.3.1, second paragraph: The Terpugova, 2005 reference is for a conference abstract. This is an important parameter, the mean seasonal values of the black carbon mass concentration, and there should be a proper, peer reviewed reference for these data.*

The reference is replaced with:
Kozlov, V.S., Shmargunov, V.P., and Panchenko, M.V.: Seasonal variability of the vertical profiles of absorption parameters of submicron aerosol in the troposphere, Atmospheric and Oceanic Optics, 22, 635-642, 2009.

10. *Page 145, line 9: Why was the activity parameter different at the surface and above the surface? Were you encountering different aerosols at the surface and aloft?*
The differences in the parameter of condensation activity at the ground (z=0) and other heights are observed in the experiment (Panchenko et al, 1996). The data on the parameter of condensation activity at the heights up to 5 km were obtained from measurements onboard an aircraft-laboratory in 1986-1988 (more than 200 realizations). The differences between the values γ at z=0 and other heights are more than 95% reliable according to Student criterion.
In summer, the portion of particles generated and emitted into the atmosphere immediately in the region of observations should be large in the near ground layer. The increase of the parameter of condensation activity with height is indicative of the increase in the specific content of low volatile hygroscopic or well soluble components in the aerosol particle composition with height. It can be caused by the contribution from a source of such a kind of particles that is situated quite far from the region of observation. Therefore, relative contribution coming from particles transported from the remote regions into the total scattering coefficient should decrease when approaching to the underlying surface (i.e. as the height decreases). Particularly, it can not be excluded that the observed tendency in the behavior of thermo and hygrooptical characteristics with height can be caused by the effect of remote sea and oceanic areas.

11. *Figure 3: Why are the Angstrom exponent values listed on the AOD plot? Also, please be consistent with using either Angstrom exponent or Angstrom parameter throughout.*

We agree with the remark, the Angstrom exponent values are removed from the figure.

12. *Figure 3: Were the samples dried prior to your nephelometer measurements? It appears that there may be a strong RH dependence in the single scattering albedo measurements as the altitude approaches the atmospheric boundary layer. Large changes in RH (as near the boundary layer) will cause a strong dependence in the scattering coefficient values if the samples are not dried. For example, in the Andrews et al. (2011) paper cited, as you noted, the single scattering albedo does not have a large increase near the boundary layer. That is because their samples are dried prior to measurement.*

The data presented in Fig. 3 are recalculated for the seasonal (summer) average values of the relative humidity of air.