The correlation between aerosol optical parameters in near IR molecular absorption bands

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Several regional statistical ensembles of tropospheric (0–12 km) aerosol optical properties (extinction – AEC, scattering – ASC, and asymmetry parameter – AP) have been modeled based on up-to-date experimental data. The covariance matrices of optical parameters for continental and marine ensembles have been calculated. The spectral correlation in molecular absorption bands: O_2 (0.76 μm), CO_2 (1.61 and 2.06 μm) between AEC, ASC, and AP has been studied. These bands are planned to be used in OCO (Orbital Carbon Observatory) Project for determination of CO_2 total content with high accuracy. It is shown that a significant correlation (0.7–1.0) takes place between AEC and ASC in all channels. The correlation between AEC and PA is lower (0.05–0.55). The optimal and Angström parameterization methods were applied to spectral approximation of AEC, ASC, and PA. It has been shown that the mean error in case of the optimal parameterization totals 0.02–1.5%, in case of the Angström parameterization it was 1.2–9.9% for continental and marine aerosol models.

Introduction

The problem of climate change stimulated intensive research of the global circulation of carbon.[1,2] This is connected with the fact that the atmospheric CO_2 is one of the most important greenhouse gases with significant anthropogenic sources. Programs of satellite measurements of total CO_2 were developed to determine the intensity of CO_2 sources and sinks, as well as their geographical distribution. The purpose of these measurements is the obtaining of global maps of CO_2 content and the solution of the corresponding inverse problem on their base, namely, the determination of the intensity of CO_2 sources and sinks.[3,4]

It is proposed to use measurements of outgoing radiation in near IR spectral range in the planned satellite experiments. Thus, in OCO (Orbital Carbon Observatory) Project, the measurement of the reflected and dispersed solar radiation in O_2 (0.76 μm) and CO_2 (1.61 and 2.06 μm) absorption bands is planned.

Based on the preliminary analysis of errors in total CO_2 content determination, we have come to a conclusion that measurements in 1.61 μm band have the maximum sensitivity to CO_2 variations near the surface, while measurements in oxygen band and CO_2 2.06 μm band allow us to retrieve the necessary information about surface pressure, albedo, atmospheric temperature, water vapor content, as well as aerosol and cloud characteristics.[3,4]

The measurements should be performed with a high spectral resolution (not less than 17000) in order to effectively separate the influence of different atmospheric parameters and surface on the outgoing radiation. Taking into account relatively small spatial and temporal variations of CO_2, the required accuracy of satellite measurements should be very high (0.1–0.5%). Thus, it is necessary to develop a high-quality physical-mathematical model of the satellite experiment and the optimal methods for solving the inverse problem, namely, the determination of CO_2 total content.

Generally, the magnitude of the intensity I of the outgoing, reflected, and scattered solar radiation at the λ wavelength is determined by a series of atmospheric and surface parameters:

$$I(\lambda)=I(\lambda,q_i(z),\alpha_r(z,\lambda),\chi_r(z,\gamma,\lambda),\alpha_a(z,\lambda),\sigma_a(z,\lambda),\chi_a(z,\gamma,\lambda),A,\ldots), \quad (1)$$

where q_i is the ratio of the mixture of *i-th* absorbing gas; α_r, χ_r are the extinction coefficient and the Rayleigh scattering phase function, determined by the vertical profile of the atmospheric density; α_a, σ_a, χ_a are aerosol extinction coefficient (AEC), aerosol scattering coefficient (ASC), and the scattering phase function; A defines optical characteristics of the surface (albedo in the simplest case). The aerosol optical characteristics, in turn, are determined by parameters of aerosol microstructure and their complex refractive index.[5]

Thus, functionals of outgoing radiation, measured from satellites, are determined by a great number of atmospheric and surface parameters, which should be retrieved during the solution of the inverse problem or their influence should be excluded in some way or another. Atmospheric aerosol is one of the most variable atmospheric parameter. Its contribution to formation of outgoing radiation in the near IR spectral range varies. Thus, the aerosol optical characteristics appear to be the functions of the wavelength and altitude (ASP and AEC), as well

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as the angle of scattering (phase function). Therefore, a large number of aerosol parameters should be retrieved.

That is why it is reasonable to use methods of statistical analysis when studying the influence of aerosol component.[6] The recent approach to building of statistical aerosol models, as well as different ways of their use in direct and inverse problems of atmospheric optics are discussed and illustrated in Refs. 7–9. In this article, that approach is used for analysis of correlations between aerosol optical characteristics in the near IR spectral range. The magnitude of these correlations substantially determines the efficiency of accounting for the aerosol influence or its elimination from solution of the inverse problem of carbon dioxide content retrieval.

This is connected with the fact that irrespectively of the approach used for CO_2 determination (solution of linearized analog of radiation transfer equation, DOAS method, the use of optical path conception) the basic aerosol optical characteristics should be specified (ASP and AEC, scattering phase function) in O_2 and CO_2 absorption bands in order to solve the direct problem of radiation transfer. An efficient estimate of aerosol optical parameters from measurements of outgoing radiation in all spectral ranges under consideration is possible only under condition that there is a sufficiently close spectral correlation bonds between them.

1. Statistical modeling of the tropospheric aerosol

In statistical modeling of variability of the tropospheric aerosol optical characteristics we based on the data of OPAC (Optical Properties of Aerosols and Clouds)[10] model. These data are formed from a series of field measurements (see http://www.lrz-muenchen.de/~uh234an/www/radaer/opac.html for details). This model contains aerosol optical characteristics (ASP, AEC, and asymmetry parameter of the phase function) from a range of 0.25–40 μm calculated per one particle for the following 10 tropospheric fractions: WASO (water-soluble aerosol consisting of different sulfates, nitrates, and other organic and inorganic compounds), INSO (water-insoluble particles being the mixture of dust, soil, and different insoluble particles of inorganic origin), SOOT (soot particles), MINM, MIAM, MICM, MITR (mineral quartz and clay particles), SSAM, SSCM (particles of marine salt), and SUSO (stratospheric sulfate particle, 75% of which is the sulfuric acid).The optical properties of such fractions as WASO, SSAM, SSCM, and SUSO depend mostly on the atmospheric humidity. The number of particles of SSAM and SSCM fractions per unit volume depends on the wind speed.

Thus, when constructing the statistical ensembles of optical properties of the global tropospheric aerosol, we vary 13 independent parameters: particle concentration of 10 aerosol fractions, relative atmospheric humidity, altitude of the aerosol layer under study (with a vertical resolution of 1 km) and wind speed for SSAM and SSCM fractions.

To build the global and local statistical models, we found mean values and root-mean-square deviations (RMSD) for the quantity of particles of described fractions, taking into account ratios between the quantity of different particles, realization frequencies of aerosol types throughout the Earth atmosphere, as well as the aerosol altitude distribution, wind speed, and atmospheric humidity. Since the quantity of particles in different fractions differ by orders of magnitude, not aerosol concentrations themselves, but their natural logarithms were used in all model calculations. Based on the received variances of the particle quantity for each fraction in troposphere, model covariance matrices with correlation radius between atmospheric levels of 2 km were built. Mean values and covariance matrices were used for construction of altitude profiles of the particle quantity in different fractions (with the help of random number generator). From the obtained realizations of particle quantities, the ASP, AEC, AP, and their spectral dependences were calculated. The peculiarities of statistical modeling of the tropospheric aerosol are described in Ref. 8 in more detail, where a good statistical agreement between statistical properties of the modeled and experimental ensembles is also shown.

As it was mentioned before, besides the global ensemble of tropospheric aerosol, several local ensembles of the same dimension were also built by us in a similar way. Local ensembles were related to zones of the desert, ocean and continent. Table 1 presents some characteristics of two local ensembles of the tropospheric aerosol (continental and marine), which exemplify connections between optical characteristics in the OCO measurement channels.

Table 1. Characteristics of tropospheric aerosol local models (layer thickness, altitude of homogeneous atmosphere H, aerosol fractions, and variance of their surface concentrations N_0)

Model	Layer, km	H, km	Fraction (variance N_0)
Continent	0–2	8	WASO (906–114385), INSO (0.04–3.9), SOOT (78–2096980)
Marine	0–2	1	WASO (25–11890), SOOT (1–88492), SSAM (0.8–56), SSCM (0.0001–0.007)
Open troposphere	2–12	8	WASO (34–1045), INSO (0.02–0.14), SOOT (26–735)

The second column of Table 1 shows layer boundaries of the troposphere division in the process of modeling. The third column contains the altitude of aerosol homogeneous atmosphere, according to

which the vertical behavior of the particle content is calculated exponentially. The forth column contains the fractions, which are taken into account during the modeling, as well as the values of the surface aerosol concentration in each fraction. It is seen that both in the continental and marine models aerosol particles of such fractions as WASO and SOOT are present; INSO particles are additionally present in the open atmosphere and the continental boundary layer. The SSAM and SSCM fractions are also present in the boundary layer of the marine model.

For better understanding of the spectral behavior of optical characteristic of the tropospheric aerosol ensembles (types), we preliminary have analyzed the spectral behavior of individual aerosol fractions as parts of the ensembles of interest. The analysis of imaginary part of the complex refractive index, which substantially determines the absorption of atmospheric aerosol, has shown that in the spectral range under consideration, wavelength dependence of the refractive index differs for various tropospheric aerosol fractions, with the highest value for particles of the SOOT fraction. The analysis of AEC and ASC spectral behavior per one particle has shown that the significant difference between AEC and ASC is typical for SOOT particles. As for other aerosol fractions, the main attenuation of radiation is due to the scattering. It was also found that the scattering phase function has a different form for the particles of different fractions. The AP values, close to zero (i.e., isotropic scattering), especially in the long-wave region, are observed for the SOOT fraction (AP = 0.1–0.3). For WASO fraction, the AP is slightly higher (0.45–0.65). For INSO, SSAM, and SSCM aerosol fractions, the scattering phase function has the most protruding form (AP = 0.8–0.9).

Thus, based on modeled ensembles of particle quantity (500 vertical profiles of the number of particles of different aerosol fractions, each at 25 altitudes of troposphere, 0–12 km), we calculated ensembles of aerosol optical characteristics in three spectral regions and built covariance matrices of parameters under study. These matrices can be further used in solution of inverse problems as *a priori* information or in the regression approach to determination of the sought parameters.[8,9,11] A more detailed information about the models, as well as the models themselves will be accessible on the site of the Remote Sensing Laboratory of the SpbSU Atmospheric Physics Department (http://troll.phys.spbu.ru) in near future.

Based on the covariance matrices, the following issues have been studied in this work:

– the spectral correlation of optical characteristics,

– the correlations between different optical parameters,

– the accuracy of optimal parameterization of optical aerosol characteristics for three wave lengths as an element of constructing the methodology of determination of the aerosol type and the solution of the inverse problem.

2. Statistical characteristics of the built optical ensemble

Let us analyze the optical thickness of tropospheric aerosol in the ensembles under study in order to estimate them quantitatively. Figure 1 presents mean values and RMSD of aerosol optical thickness (further – optical thickness) in three spectral regions for tropospheric models of the continental and marine aerosol.

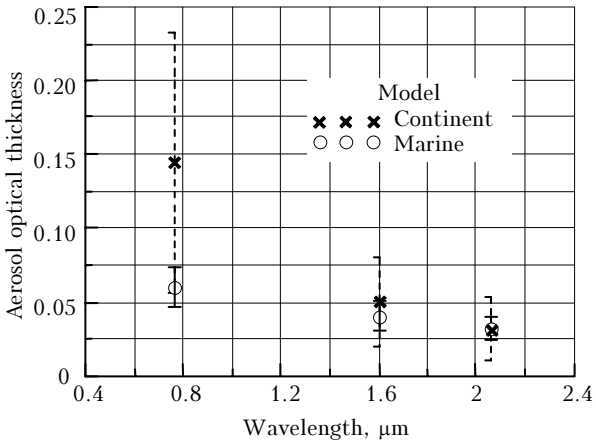

Fig. 1. Mean values and RMSD of tropospheric aerosol optical thickness in different measurement channels for different models.

As it is seen, the highest mean values of optical thickness are in the ensemble of the continental model, the lowest – in the ensemble of the marine model. For continental model of the tropospheric aerosol, significant changes of optical thickness are observed when passing from 0.76 μm to 2.06 μm channel. Similar changes are typical for atmospheric conditions of moderate latitudes, which are close to background ones.[12] The mean values themselves for continental models are in good agreement with mean data of long-term observations,[12] in particular, for the clear atmosphere.

The highest variability of optical thickness occurs in the 0.76 μm channel. For the ensemble of the continental model the RMSD value is about 60% of the mean one. The mean value of optical thickness in the marine model is close to the mean measurement data obtained in the central zone of the Atlantic, beyond the influence of the African continent, in the southern part of the Indian ocean, and in the South ocean,[12] i.e., in the regions with relatively high atmospheric transparency.

3. Correlations between different aerosol optical parameters

The study of correlations between different parameters allows us to estimate the possibility of determination of some parameters from the others.[8,9] In the context of the OCO Project,[3,4] we plan to obtain the main information about aerosol

parameters, based on CO_2 concentration measurements, from the oxygen band of 0.76 μm and extrapolate it to the bands of carbon dioxide absorption of 1.61 and 2.06 μm channels. Let us consider spectral statistical bounds between aerosol optical parameters using the built ensembles as an example.

Consider three individual ensembles: continental boundary, marine boundary, and open troposphere (see Table 1), simply referring to them as to continental, marine, and open troposphere models, respectively. In order to simplify the calculations and the analysis we mix all values of optical parameters inside the models, i.e., we study statistical bonds independently of the altitude of the troposphere level, considering all realizations of optical parameters as random variables. Thus, ensembles of continental and marine models consist of 2500 realizations and the open troposphere ensemble consists of 10000 realizations of optical parameters of the tropospheric aerosol.

Figure 2 shows a significant spectral correlation between AEC at 0.76 μm (0.85–0.99) and AEC, ASC, AP (solid, dotted, and dot-and-dash lines, respectively) in all channels for above three models of the tropospheric aerosol.

An ensemble of continental model is an exception, in which correlations with ASC decrease from 0.8 in 0.76 μm channel to 0.55 in 2.06 μm channel. Correlations between AEC and AP are significantly lower and do not depend on the wavelength. Thus, for the ensemble of marine model the correlations are close to zero; for the ensemble of the open troposphere they are equal to 0.40–0.45; for the ensemble of continental aerosol they are equal to 0.55 (by the absolute value).

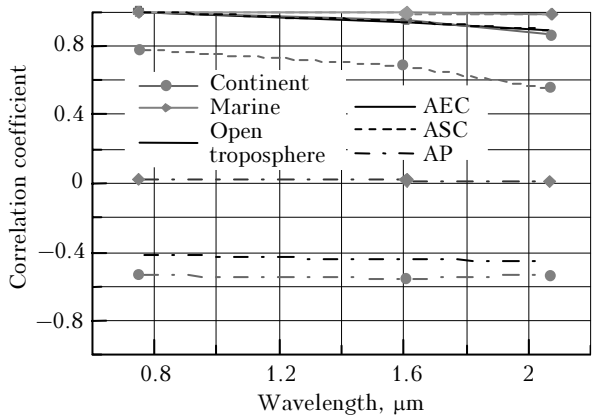

Fig. 2. The correlation between AEC in 0.76 μm channel and other optical parameters in different channels for different ensembles of the tropospheric aerosol.

Note that using a statistical relation between natural logarithms of AEC and ASC (not between AEC and ASC values themselves) for the continental model ensemble, we can slightly improve the correlations between them. As for other ensembles, the correlation magnitude is independent of the fact whether we use the values of their coefficients or the logarithms.

4. Optimal parameterization of spectral behavior of aerosol optical parameters

When solving direct and inverse problems of atmospheric optics, we often use the parameterization of spectral characteristics of aerosol optical parameter. The available statistical ensemble of aerosol parameters allows us for approximation of spectral behavior of one or another parameter to use the expansion of parameters in terms of eigenvectors of the corresponding covariance spectral matrix, which forms an empirical orthogonal basis.[7,13] Remind that the optimal parameterization of spectral behavior of aerosol parameters (AEC, ASC, or AP) $\sigma(\lambda_i)$ can be written as

$$\sigma(\lambda_i) = \overline{\sigma}(\lambda_i) + \sum_{p=1,n} a_p f_p(\lambda_i),$$

where $f_p(\lambda_i)$ are the vectors of spectral covariance matrix of an aerosol parameter; a_p are the corresponding expansion coefficients.

Using the built ensembles as an example, we compared the method of optimal parameterization and one more frequently used method, which is based on the particle size distribution by the Angstrom formula[5,14]: $\sigma(\lambda) = \sigma_0 \lambda^{-a}$, where σ_0 and a are some parameters. The relative error of AEC, ASC, and AP approximations, averaged over three channels, is shown in Table 2 for optimal and Angström parameterizations. Note that in both cases two-parameter parameterization was used.

Table 2. Error (%) for different types of spectral parameterization of aerosol parameters (in three channels) for different ensembles of the tropospheric aerosol

Type	Parameter	Continental 0–2 km	Marine 0–2 km	Open troposphere
Angström	AEC	5.9	3.9	2.1
	ASC	9.9	4.1	1.2
	AP	5.4	1.4	6.5
Optimal	AEC	0.3	0.03	0.02
	ASC	0.3	0.02	0.02
	AP	1.5	0.04	0.07

It is seen that the optimal parameterization has a great advantage over the parameterization based on the Angström formula. This advantage is most prominent in case of ASC parameterization in the ensemble of continental model, when the absorption by soot particles plays an important role in aerosol extinction. It is seen that the optimal parameterization yields an error of 0.02–0.3% in spectral approximation for AEC and ASC and 0.04–1.5% for AP, while the Angström approximation

error for three studied parameters and ensembles of interest is 1.2–9.9%.

We also have analyzed spectral behaviors of the relative error for optimal and Angström parameterizations. It was found that for AEC and ASC, the Angström parameterization error increases with increasing wavelength. Approximation error for ASC at 2.06 μm in the ensemble of continental aerosol reaches 22%. As for the optimal parameterization, the relative approximation error for AEC and ASC is the highest at 1.61 μm, although it does not exceed 0.5%.

Conclusion

The results of this work are the following:

1. A global statistical model, as well as a series of regional statistical models of tropospheric aerosol optical properties have been built. Ensembles of continental and marine models of tropospheric aerosol, which contain 500 realizations of altitude profiles (0–12 km) of extinction and scattering coefficients, as well as the asymmetry parameter of the Henvy–Grinstein phase function (mean scattering cosine) have been generated. The covariance matrices of the built ensembles of optical parameters have been calculated.

2. Correlations, including the spectral ones, between different aerosol parameters at 0.76, 1.61, and 2.06 μm have been studied, which turned to be significant between AEC and ASC in all above channels. Correlation coefficients between natural algorithms of these parameters are equal to 0.7–1.0 depending on the model, channel, and parameter. The correlation between AEC and AP is significantly lower (0.05–0.55) and practically does not depend on the wavelength.

3. The approximation of AEC, ASC, and AP spectral behaviors with the use of optimal and Angström parameterization methods was conducted. It was found that the method of optimal parameterization gives mean approximation errors equal to 0.02–1.5% for three channels, this value for the Angström parameterization method is 1.2–9.9%. Optimal parameterization errors in individual channels for the ensembles of marine model (0–2 km) and open troposphere model (2–12 km) do not exceed 0.1%. The Angström parameterization error for AEC and ASC increases with wavelength and reaches 22% for ASC at 2.06 μm of the continental aerosol ensemble (0–2 km).

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