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Retrieval of the Atmospheric Compounds Using a Spectral Optical Thickness Information

Andrey A. Ioltukhovski

Abstract—A spectral inversion technique for retrieval of the atmospheric gases and aerosols contents is proposed. This technique based upon the preliminary measurement or retrieval of the spectral optical thickness. The existence of a priori information about the spectral cross sections for some of the atmospheric components allows to retrieve the relative contents of these components in the atmosphere. Method of “smooth” filtration makes possible to estimate contents of atmospheric aerosols with known cross sections and to filter out other aerosols; this is done independently from their relative contribution to the optical thickness.

I. Introduction

ONE of the main applications of the remote sensing from satellite is the monitoring of the atmosphere. In the solar spectrum, all sensors measure radiances passing through the atmosphere. This signal depends on the properties of the medium, primarily on the gaseous absorption and the scattering by molecules and aerosols. The interpretation of the remote sensing data is complicated due to the unknown contribution of the different atmospheric components to the radiation transport process in the atmosphere. The radiation field depends on the optical characteristics of the gases and particles such as the scattering and absorption volumetric coefficients and scattering phase functions. The spatial distribution of these characteristics completely determines the optical properties of the atmosphere. For the direct modeling of the radiative transfer, in the case of an inhomogeneous atmosphere bounded by an inhomogeneous surface, it is necessary to take into account thin structure of the atmosphere. But in the case of the inverse problem solution, we really deal with averaging information and on the whole we can use integral or effective optical characteristics of the media like optical thickness, asymmetrical factor, effective radius of the particles and so on. Most of these characteristics are quite “artificial” and can represent the natural properties of the atmosphere with very good supporting a priori information. Practically, the only characteristic, which does not depend on the thin structure of the atmosphere is the optical thickness. Moreover, when we deal with optical thickness we know exactly what it means, because there is the unique optical thickness at a given point

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The author is with the Laboratoire de Meteorologie Physique, Universite Blaise Pascal, F-63177 Aubiere, France on leave from the Keldysh Institute of the Applied Mathematics, Russian Academy of Sciences, 125047 Moscow A-47, Russia.

and for a given direction. Spectral optical thickness is very informative characteristic of the atmosphere and it can be measured or retrieved relatively easily in comparison with other parameters.

In this paper, we study the possibility to retrieve a maximum of information about the different atmospheric constituents from the spectral optical thickness.

II. Inversion Technique

The idea of using spectral inversion technique is not new and was used successfully for the occultation [1] and ground [2]-[6] measurements. The main basic aspects of this inversion method are as follows: Let us assume that the atmosphere is composed in $I + J$ different components (gases and aerosols). Let the extinction cross section of each gas be denoted by $agi(X)$ and number concentration be denoted by $p_i(r)$, $i = 1, \dots, I$, and let the volumetric cross section of the aerosols be denoted by $\langle rai(A) \rangle$, corresponding number of particles be denoted by $kj(r)$, $j = 1, \dots, J$, where A is wavelength and $r = \{x, y, z\}$ is vector of the spatial coordinates. Optical thickness along the trajectory l is

$$\begin{aligned} r(A) &= rg(X) + rp(X) \\ &= \sum_{i=1}^I \int_1^l VgiWPits'lds + \sum_{j=1}^J \int_1^l aai(X)kj(s)ds \\ &= \sum_{i=1}^I \sigma_{gi}(\lambda)Pi + \sum_{j=1}^J \sigma_{aj}(\lambda)Kj \end{aligned} \quad (1)$$

where Pi is the integral (column) number concentration

$$Pi = \int_1^l Pi(s)ds$$

and Kj is the integral (column) number of particles along the ray trajectory:

$$Kj = \int_1^l kj(s)ds$$

here s is the point of the trajectory and ds is the element of the trajectory. The optical thickness is dimensionless and it does not matter that the different units are used for the gas and aerosol components.

With measurements at different wavelengths Xn , $n = 1, \dots, N$ and under the condition that all cross section $\langle rgi(A) \rangle$

and $\text{traj}(A)$ are known, we have a system of linear equations:

$$\sum_{i=1}^I \sigma_{gi}(\lambda_n) P_i + \sum_{j=1}^J \sigma_{aj}(\lambda_n) K_j = r(X_n),$$

where P_i and K_j are unknowns.

This system is well-posed and has a unique solution in the least-squares sense providing that all of the n -components vectors $\{\langle zgi(A_n) \rangle\}$ and $\{\langle \text{craj}(A_n) \rangle\}$ are linearly independent. The necessary conditions for that are

- 1) $N \geq I + J$,
- 2) All of the spectral curves are linearly independent.

The first condition means that the number of spectral measurements must be more than the number of the atmospheric components to be retrieved.

It is theoretically needed to check *a priori* the linear independence of the vector $\{\langle \text{ogi}(X_n) \rangle\}$ and $\{\langle \text{craj}(A_n) \rangle\}$ for every set of the selected wavelengths for the measurements and for the inversion procedure. However, in practice, the cross sections of the different gases are absolutely independent. The main difficulties in data inversion based upon this inversion technique are due to the dependence of the gas attenuation cross section with respect to the atmospheric temperature and the uncertainty in sampling of the basic aerosol's cross sections.

In the first case the expression for the optical thickness is given by

$$\tau_g W = \int \hat{J} \hat{A} \backslash \hat{T} \hat{p} \hat{dS}. \quad (2)$$

Taking into account the fact that density is essentially positive ($p_i(A) \geq 0$) we can use mean-value theorem:

$$\tau_g(A) = \sum_{i=1}^I \sigma_i(\lambda, T^*) \int_1 P \hat{dS} = \sum_{i=1}^I \sigma_i^*(\lambda) P_i. \quad (3)$$

The main objective of this approach is to calculate the effective cross section $\langle \tau^*(A) \rangle$ for each gas constituents with an acceptable accuracy that determines the accuracy of the column density restoration. When the cross section does not depend or weakly depends on the temperature, then the integral density can be accurately retrieved. A more complicated situation is encountered when the cross section is strongly depending on temperature. In that case, $\sigma_i^*(X)$ can be calculated for every spectral interval involving real/modeled temperature and density distribution for a given point of the Earth and at a given time.

There are two factors supporting the effectiveness of the approach involving calculation of the effective cross section:

- 1) A relatively weak dependency of the basic gases cross sections on the temperature in the visible spectral range. (For example for the ozone cross section such dependency is observed in the Huggins band only.)
- 2) The relative stability of the atmospheric temperature field. In fact the seasonal temperature variations for the given Earth's point and given altitude usually does not exceed 30-40°C.

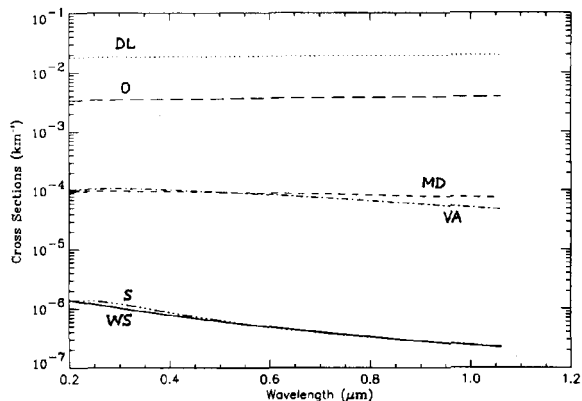


Fig. 1. Volumetric cross sections of the different aerosols.

III. Estimation of the Aerosol Contents

If we know *a priori* all types of particles which are present in the atmosphere, their size distributions and refractive coefficients, cross sections can be calculated using Mie theory [7]. This knowledge is obviously not available, because, in the general case, the atmosphere contains solid and liquid particles of different nature and variable size. Moreover the definition of the aerosol's model is more complex due to a great variability in the occurrence of aerosols as well as in their distributions in time and space [8]–[10].

Consider model which based upon the main types of aerosol according to the classification proposed in [11], [12]:

- water-soluble (WS) —particles
- dust-like (DL) —particles
- soot (S) —particles
- oceanic (O) —particles
- 75% H₂SO₄ —droplets
- volcanic ash (VA) —particles
- meteoric dust (DL) —particles

From the possible mixtures of these components six aerosol type of models have been selected: Continental, Urban/Industrial, Maritime, Background Stratospheric, Volcanic, Meteoric. Although it constitutes very useful standard for modeling the radiative transfer in the atmosphere, these aerosol types can not describe all atmospheric natural conditions. Spectral extinction volumetric cross sections of the different aerosol's components are presented in Fig. 1 [11]–[13]. All cross sections were calculated for the total number of particles per unit volume is 1 sm⁻³.

Reference [9] measured the aerosol optical thickness in the wavelength range 0.36-1.05 μm . The spectral behavior of their curves shows a significant difference from the curves shown in Fig. 1, and it means that the existing model is not able to cover the full range of possible situations. Of course, it is possible to improve this model by adding new spectral curves, but our inversion technique imposed some restrictions. First of all, the number of components (including gases) cannot exceed the number of spectral measurements. This problem can be technically solved by using spectral instruments offering a good spectral resolution, such as, the

CCD detectors for the GOMOS (Global Ozone Monitoring by Occultation of Stars) experiment have a resolution of 0.6 nm in visible (250-675 nm) and of 0.06 nm in near IR (926-952 nm) which corresponds to about 1 500 spectral values in one occultation recording [15]. The second restriction is the linear independence of the spectral curves. This restriction is more serious than the previous one because the dependence of the aerosol's cross sections on wavelength at visible wavelengths is very smooth. We can estimate a priori a maximal number of basic aerosol types for our inversion method if we know the accuracy of the measurements. Let $[A_i, A_2]$ be an spectral interval of our measurements and let $Ch_k[(2A-A_i-A_2)/(A_2-A_i)]$, $k = 1, \dots, K$ are the Chebyshev polynomials of the degree k . These polynomials are orthogonal in the interval $[A_i, A_2]$ and we can expand each spectral curve into the Chebyshev-series with an accuracy better then the accuracy of the measurements. The maximum number of the polynomials for approximating of all spectral curves with given accuracy corresponds to the maximum number of the linear independent (with the same accuracy) spectral curves. A more detailed description of this approach will be given later in Section V.

IV. Filtration Method

In practice, we have quite exact information about the gas constituents in the atmosphere and very approximate information about the particles. Any model of the atmospheric aerosol can not be complete due to an infinity of possible variations. Hence, the idea of the complete retrieval of the atmospheric structure based upon the inversion method described above is not quite realistic. Therefore it is needed to revise an objective. Let us now try to retrieve 1) all constituents for which an information is available (e.g. O₃, NO₂, NO₃ etc.) and 2) all components which presence can be occasionally assumed. According to Section II, $\{c_{9i}(A_n)\}$ are the gases extinction cross sections and $\{a_{aj}(A_n)\}$ are the cross sections of particles. The optical thickness due to these components is

$$T_i(A) = \sum_{i=1}^I C_{Tg} i l X^i P_i + \sum_{j=1}^J \sigma_{aj}(\lambda_n) K_j. \quad (4)$$

Let $rm(A)$ be a measured or a retrieved spectral optical thickness of the media. Then $r_{,,}(A) = rm(X) - t_6(A)$ is the optical thickness of the unknown components. Our task is therefore to separate the useful part of the information and to retrieve the integral densities P_i and K_j .

Since the specific spectral behavior of the optical thickness in visible wave range is most commonly connected with gases constituents in the atmosphere it is assumed, that $ru(X)$ smoothly depends on wavelength. Thus, a function $ru(A)$ can be easily represented by the Chebyshev polynomials expansion:

$$TuW = \sum_{k=1}^K c_k Ch_k. \quad (5)$$

TABLE I
Contribution of Each Atmospheric Component to the Optical Thickness

| Component | Optical thickness | Wavelength (pm) |
|--------------------|-------------------|-----------------|
| O ₃ | 1.455 | 0.3 |
| NO ₃ | 0.05 | 0.4 |
| NO ₃ | 0.0012 | 0.625 |
| Water-soluble (WS) | 0.05 | 0.55 |
| Dust-like (DL) | 0.1 | 0.55 |
| Volcanic ash (VA) | 0.2 | 0.55 |

The initial system of the equations can be substituted by the new following system

$$T^i(A_n) = \sum_{i=1}^I \hat{\sigma}_{gi}(\lambda_n) P_i + \sum_{j=1}^J \hat{\sigma}_{aj}(\lambda_n) K_j + \sum_{k=0}^K c_k Ch_k(\lambda_n), \quad (6)$$

under the conditions

$$P_i \geq 0, \quad K_j \geq 0$$

and without any restrictions for the Chebyshev's coefficients C_k .

This overdetermined system is well posed if all vectors $\{c_{9i}(A_n)\}$, $\{a_{aj}(A_n)\}$ and $\{Ch_k(A_n)\}$ are linearly independent. It means that the aerosols cross sections $c_{raj}(X)$ cannot be approximated by K Chebyshev polynomials with an accuracy comparable to the one accessible using our inversion procedure.

V. Results and Discussion

The basic model considered for the inversion process represent an atmosphere consisting of three gases and three aerosols types. Table I shows the maximal contribution of each gas to the optical thickness and also the optical thickness of each aerosol type at $A = 0.55 \mu\text{m}$.

For modeling the spectral optical thickness 11 and 41 wavelengths from 0.3 to 0.670 μm are used.

The first problem to investigate concerns the impact random errors on the retrieval accuracy. For the inversion algorithm we use the same basic components as for the direct modeling. Least-squares method is used for the overdetermined system (1) solution. It should be noted that for applying the inversion procedure, each of the spectral curves must be normalized, since the cross sections of the different components differ by a few size orders. We used normalization of the each spectral curve using the maximum value of this cross section in the spectral range $[A_i, A_2]$. In Table II, the initial and retrieved integral densities for the different random errors are given.

From these calculations, it appears that 1) the inversion algorithm is stable when the random errors are added to the measurements, i.e. small random errors result in small errors for the retrieval of substantial components, and 2) it is not necessary to use significantly more measurements than the number of components. The reason of instability of the retrieved NO₃ optical depth to measuremental noise is that the random errors (3 and 5%) are greater than the NO₃ relative contents (less than 1, 5%). The better accuracy for

TABLE II
Influence of the Random Errors on the Accuracy if the Inversion Algorithm

| MAX ERROR | GRID INX | Gases, Integral number concentration (lnr2) | | | Aerosols, Integral number of particles (km) | | |
|-----------|----------|---|--------------------------------------|--------------------------------------|---|--------|------------------------|
| | | O ₃ (x10 ¹⁹) | NO ₂ (x10 ¹⁹) | NO ₃ (x10 ¹⁵) | WC (x10 ⁶) | DL M01 | VA (x10 ⁶) |
| initial | - | 0.485 | 0.740 | 0.150 | 0.314 | 0.483 | 0.222 |
| 0% | 11 | 0.485 | 0.741 | 0.142 | 0.920 | 0.537 | 0.220 |
| 0% | 41 | 0.485 | 0.740 | 0.149 | 0.915 | 0.533 | 0.222 |
| ±1% | 11 | 0.486 | 0.737 | 0.313 | 0.833 | 0.510 | 0.233 |
| 11% | 41 | 0.484 | 0.683 | 0.222 | 1.001 | 0.542 | 0.215 |
| ±3% | 11 | 0.488 | 0.738 | 0.643 | 0.703 | 0.484 | 0.248 |
| *3% | 41 | 0.482 | 0.564 | 0.352 | 1.182 | 0.551 | 0.204 |
| ±5% | 11 | 0.490 | 0.732 | 0.961 | 0.546 | 0.442 | 0.269 |
| ±5% | 41 | 0.479 | 0.448 | 0.490 | 1.366 | 0.565 | 0.191 |

TABLE III
Retrieval of the Gas Integral Number concentration

| Max degree of the polynomial | Gases, Integral number concentration (sm ²) | | |
|------------------------------|---|--------------------------------------|--------------------------------------|
| | O ₃ (x10 ¹⁹) | NO ₂ (x10 ¹⁹) | NO ₃ (x10 ¹⁵) |
| initial density | 0.485 | 0.740 | 0.150 |
| 3 | 0.484 | 0.697 | 0.169 |
| 4 | 0.484 | 0.566 | 0.323 |
| 5 | 0.485 | 0.553 | 0.275 |
| 6 | 0.485 | 0.570 | 0.275 |
| 7 | 0.489 | 0.553 | 0.348 |

minor components retrieval can be obtained using observatory's measurements under clear conditions as it was reported in [6]. The next step in our approach is to retrieve the gas densities without any given information about the aerosol properties. The "smooth" filtration proposed in Section IV was applied and Table III shows that the algorithm based upon the polynomial approximation is stable with respect to the choice of the polynomial's degree.

All calculations were done with random errors of $\pm 1\%$. Fig. 2 shows the initial and retrieved optical thicknesses for the gas constituents. Clearly the "smooth" filtration algorithm allows 1) to retrieve substantial atmospheric components (O₃) with a very good accuracy and 2) to estimate contents of the minor components (NO₂, NO₃) in spite of the fact that their contribution is comparable to the random errors.

The addition of one or more aerosol type in our inversion model makes it possible to estimate the relative contribution due to these aerosols and to filter out the other components. Moreover, this complication of the model practically does not affect the accuracy of retrieval of the gas contents restoration. Fig. 3 illustrates the simultaneous retrieval of the gases and the volcanic ash contents.

The relative contribution of the volcanic ash to the optical thickness exceeds by a factor close to 5 the maximal contribution due to NO₂, but still the accuracy in the volcanic ash retrieval is the same as NO₂ (about 25%). This is due to the fact that the extinction cross section of VA smoothly depends on wavelength in the visible range and can be approximated

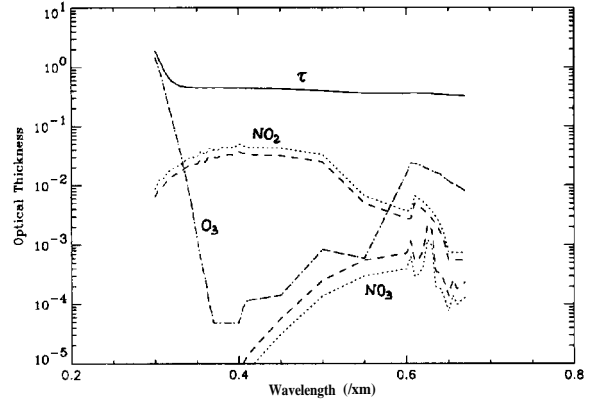


Fig. 2. Relative contribution of the gases to the optical thickness. (—) total optical thickness, (.....) initial curves, (---) retrieved curves.

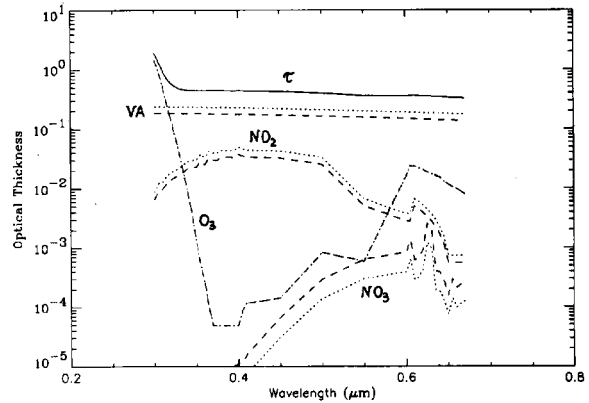


Fig. 3. Relative contribution of the gases and VA to the optical thickness. (—) total optical thickness, (.....) initial curves, (---) retrieved curves.

with a very good accuracy by very few Chebyshev polynomials (e.g. the approximation with accuracy of 1% requires only four polynomials). Hence, the more the behavior of the spectral cross section is specific, the more the accuracy for this component retrieval can be expected. It should be noted that the method of "smooth" filtration allows also to filter out the optically thin cloud's effect because scattering by large particles is not practically sensitive to wavelength in visible wave range.

VI. Conclusion

In this paper, we did not discuss the methods of the spectral optical thickness measurement or retrieval because in the first place the different problems imply the different solutions and secondly there are many well-known methods. For example, in case of the occultation measurements one can obtain the optical thickness using measure of the direct starlight or sunlight traversed through the atmosphere and outside of the atmosphere [15]. Multiangular measurements are very useful for the optical thickness restoration in case of the upward radiation measurements [16]. Spectral optical thickness is the

objective characteristic of the media and contains a lot of information about the atmospheric constituents. But really we can retrieve the component's contents if we know spectral behavior of the cross section and this behavior is quite specific. It is not necessary restrict oneself to use the visible wave range only. This method can work in wave range where it is possible to measure optical thickness and where relation (1) is fulfilled.

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Andrey A. Ioltukhovski received the Ph.D. degree in applied mathematics from the Keldysh Institute of Applied Mathematics, the Russia Academy of Sciences, Moscow, in 1986.

He has been with Keldysh Institute since 1981. Presently he is a visiting scientist at the Laboratoire de Métrologie Physique, University Blaise Pascal, France. His research interests include direct and inverse problems of the radiative transfer in the atmosphere and applications to remote sensing.