DETERMINATION OF THE INTEGRAL CONTENT OF ATMOSPHERIC TRACE GASES FROM ZENITH MEASUREMENTS OF THE SPECTRAL SKY BRIGHTNESS

V.N. Marichev

Institute of Atmospheric Optics,
Siberian Branch of the Russian Academy of Sciences, Tomsk
Received September 18, 1994

A technique is proposed for determination of the integral content of atmospheric trace gases (ATG) from zenith observations of scattered solar radiation. The technique is based on measurements at two close wavelengths, one of which is on the ATG absorption line (band), and two solar zenith angles. Calibration of scattered solar radiation measurements against the direct solar radiation is not required.

Measurements of the total ozone content (TOC) performed at ozonometric network are well known in the literature. They employ Gushchin's M–83 and M–124 devices[1,2] and are based on measurements of differential absorption of the solar UV radiation by atmospheric ozone. These TOC observations use measurements of direct solar radiation and that scattered in the zenith direction. Specifically, direct solar radiation measurements are used as reference one. Signals of direct and scattered solar radiation components are compared in order to construct an empirical (calibration) curve, from which the TOC value is then derived with the use of zenith observations knowing the value of insolation at the top of the atmosphere.

Despite the two TOC measurement techniques agree fairly well, scattered radiation measurements require large volume of statistics to obtain the reliable empirical curve. This may cause no difficulties in standard ozone network measurements with the M–124 device operating in specified spectral intervals; however, this may become problematic in broadband measurements of sky brightness due to the lack of spectral measurements of direct solar radiation, abundance of employed spectral bands, ambiguous relations among spectral signals, etc.

In the present paper, a differential method is proposed of measurement and retrieval of the total ATG content using measured values of scattered solar radiation.

Let us consider the measurement scheme of zenith observations of spectral sky brightness (Fig. 1). Light signal is received in a vertical cone, passing through the entire depth of the atmosphere, formed by a receiver field of view. Let us take its volume element at a height H

$$dV = \pi \tan^2\varphi \, H^2 \, dH , \tag{1}$$

where 2φ is the plane angle of the receiver field of view.

At a solar zenith angle θ, the spectral signal $dJ_{\lambda\theta}$ from the elementary volume dV is written as

$$dJ_{\lambda\theta}=J^0_\lambda q_\lambda S T^0_{\lambda\theta}(H)\beta^\Sigma_{\lambda\theta}(H)T_{\lambda\theta}(H)\pi\tan^2\varphi \, dH , \tag{2}$$

where J^0_λ is the spectral insolation, q_λ is the instrument spectral sensitivity, $T^0_{\lambda\theta}(H)$ is the spectral transmission from the atmospheric top to the scattering layer at height H, $T_{\lambda\theta}(H)$ is the spectral transmission from H to the instrument altitude, S is the receiving aperture, and β^R_{kH} is the spectral scattering coefficient in the direction of observation at solar zenith angle θ,

$$T^0_{\lambda\theta} = T^{0a}_{\lambda\theta} \, T^{0m}_{\lambda\theta} \, T^{0g}_{\lambda\theta} , \tag{3}$$

$$T_{\lambda\theta} = T^a_{\lambda\theta} \, T^m_{\lambda\theta} \, T^g_{\lambda\theta} , \tag{4}$$

$$\beta^\Sigma_{\lambda\theta} = \beta^a_{\lambda\theta} + \beta^m_{\lambda\theta} . \tag{5}$$

Here the superscripts a, m, and g stand for aerosol and molecular scattering and ATG absorption, respectively.

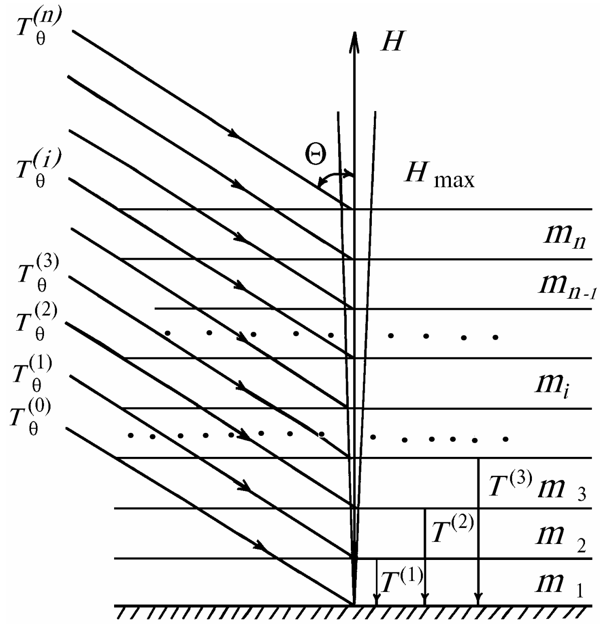

FIG. 1. Measurement scheme illustrating the derivation of formulas for signals of scattered solar radiation.

The total spectral signal from the entire atmospheric depth is given as

$$J_{\lambda\theta} = J^0_\lambda \, q_\lambda \, S \, \pi \, \tan^2\varphi \int\limits_0^{H_{max}} T^0_{\lambda\theta}(h) \, \beta^\Sigma_{\lambda\theta}(h) \, T_{\lambda\theta}(h) \, dh , \tag{6}$$

0235-6880/95/08 615-03 $02.00

where H_{max} is the altitude starting from which the atmospheric attenuation becomes important.

Let us return now to Eqs. (3) and (4) and assume the atmosphere to be plane–layered with optical thicknesses of the layers m_1, m_2, m_3, etc. (Fig. 1). Then, transmission of the layer m_i at solar zenith angle θ is $T^0_{\lambda\theta}(m_i) = \exp[-m_i/\cos\theta]$. Accordingly, the solar radiation transmission from atmospheric top to the Earth's surface and to the top of layers m_1, m_2, m_3, etc. is written as

$$T^{0(0)}_{\lambda\theta} = \exp\left[-\frac{m_0}{\cos\theta}\right], \quad T^{0(1)}_{\lambda\theta} = \exp\left[-\frac{(m_0 - m_1)}{\cos\theta}\right],$$

$$T^{0(2)}_{\lambda\theta} = \exp\left[-\frac{(m_0 - m_1 - m_2)}{\cos\theta}\right],$$

$$T^{0(3)}_{\lambda\theta} = \exp\left[-\frac{(m_0 - m_1 - m_2 - m_3)}{\cos\theta}\right],$$

$$T^{0(i)}_{\lambda\theta} = \exp\left[-\frac{(m_0 - m_1 - m_2 - m_3 - \ldots - m_i)}{\cos\theta}\right]. \quad (7)$$

Here, $T^{0(i)}_{\lambda\theta}$ denotes the slant atmospheric transmission to the ith layer, m_0 is the optical depth of the entire atmosphere.

Similarly, the vertical transmission from the ground to the top of layers m_1, m_2, m_3, etc. is defined as

$$T^{(0)}_{\lambda} = 1,$$

$$T^{(1)}_{\lambda} = \exp[-m_1],$$

$$T^{(2)}_{\lambda} = \exp[-(m_1 + m_2)],$$

$$T^{(3)}_{\lambda} = \exp[-(m_1 + m_2 + m_3)],$$

$$T^{(i)}_{\lambda} = \exp[-(m_1 + m_2 + m_3 + \ldots + m_i)]. \quad (8)$$

Writing the optical thickness for atmospheric layer of height H as

$$m(H) = \int\limits_0^H \mathrm{d}m \quad (9)$$

(and, hence, $m_0 = \int\limits_0^{H_{max}} \mathrm{d}m$, $m_1 = \int\limits_0^{H_1} \mathrm{d}m$, $m_2 = \int\limits_{H_1}^{H_2} \mathrm{d}m$, $m_3 = \int\limits_{H_2}^{H_3} \mathrm{d}m$, ..., $m_i = \int\limits_{H_{i-1}}^{H_i} \mathrm{d}m$), slant and vertical transmission for observations made from the altitude H can be written as

$$T^0_{\lambda\theta}(H) = \exp\left[-\frac{m_0 - m(H)}{\cos\theta}\right] = \exp\left[-\int\limits_H^{H_{max}} \frac{\mathrm{d}\,m}{\cos\theta}\right], \quad (10)$$

$$T_{\lambda}(H) = \exp[-m(H)] = \exp\left[-\int\limits_0^{H_{max}} \mathrm{d}\,m\right]. \quad (11)$$

Product of functions (10) and (11), appearing in Eqs. (2) and (3), will take the following form:

$$T^0_{\lambda\theta}(H)T_{\lambda}(H) = \exp\left[-\frac{m_0}{\cos\theta}\right]\exp\left[\left(\frac{1}{\cos\theta} - 1\right)m(H)\right]. \quad (12)$$

Substitution of the above expression in Eq. (6) for spectral signal from the entire atmospheric depth finally gives

$$J_{\lambda\theta} = C\exp\left[-\frac{m_0}{\cos\theta}\right] \times$$

$$\times \int\limits_0^{H_{max}} \beta^{\Sigma}_{\lambda\theta}(h)\exp\left[\left(\frac{1}{\cos\theta} - 1\right)m(h)\right]\mathrm{d}\,h, \quad (13)$$

where $C = J^0_{\lambda}q_{\lambda}S\pi\tan^2\varphi$.

From Eq. (13), we see that the spectral signal of sky is directly proportional to the atmospheric vertical depth and solar zenith angle, and depends in a more complex fashion on vertical distribution of the scattering coefficient at a given θ and on the optical depth.

Now we consider the feasibility of determining ATG concentration from Eq. (13). To this end, we write out the optical depth m and the scattering coefficient $\beta^{\Sigma}_{\lambda\theta}$, both entering into Eq. (13), as

$$m(H) = \int\limits_0^H \alpha_a\,\mathrm{d}\,h + \int\limits_0^H \alpha_m\,\mathrm{d}\,h + \int\limits_0^H\left(\sum_{j=1}^{n} K_j N_j\right)\mathrm{d}\,h =$$

$$= m^a(H) + m^m(H) + m^g(H), \quad (14)$$

$$\beta^{\Sigma}_{\lambda\theta}(H) = \beta^a_{\lambda\theta}(H) + \beta^m_{\lambda\theta}(H). \quad (15)$$

Here α_a and α_m are the aerosol and molecular scattering coefficients at a wavelength λ, and K_j and N_j are the absorption coefficient at λ and concentration of the jth ATG constituent.

In simultaneous measurements (at θ = const) at two close wavelengths λ_1 and λ_2, the contributions from aerosol and molecular scattering can be considered equal, that is,

$$m^a_{\lambda_1}(H) = m^a_{\lambda_2}(H), \quad m^m_{\lambda_1}(H) = m^m_{\lambda_2}(H),$$

$$\beta^a_{\lambda_1\theta}(H) = \beta^a_{\lambda_2\theta}(H), \qquad \beta^m_{\lambda_1\theta}(H) = \beta^m_{\lambda_2\theta}(H). \quad (16)$$

With one of the sounding wavelengths on the absorption line (band) of the lth ATG constituent and the other off the absorption line (band), the two spectral signals will be differently absorbed, and their ratio will be

$$I_{\lambda_1,\lambda_2} = \frac{J_{\lambda_1}}{J_{\lambda_2}} = \exp\left[-\frac{1}{\cos\theta}\int\limits_0^{H_{max}} K_l N_l\,\mathrm{d}\,h\right] \times$$

$$\times \exp\left[\left(\frac{1}{\cos\theta} - 1\right)\int\limits_0^{H_{eff}} K_l N_l\,\mathrm{d}\,h\right]. \quad (17)$$

Here the second multiplier has been obtained by applying the theorem of the mean

$$\exp\left[\left(\frac{1}{\cos\theta}-1\right)\int\limits_0^{H_{\text{eff}}} K_l N_l \,\mathrm{d}\,h\right]=$$

$$=\frac{\int\limits_0^{H_{\max}} \beta^{\mathrm{R}}_{\lambda_1\theta}(h)\exp\left[\left(\frac{1}{\cos\theta}-1\right)m_{\lambda_1}(h)\right]\mathrm{d}\,h}{\int\limits_0^{H_{\max}} \beta^{\mathrm{R}}_{\lambda_2\lambda\theta}(h)\exp\left[\left(\frac{1}{\cos\theta}-1\right)m_{\lambda_2}(h)\right]\mathrm{d}\,h}, \qquad (18)$$

where the altitude H_{eff} is chosen somewhere between 0 and $H_{\max}$.

In analogy with Eqs. (9) and (14), we introduce the notation

$$\int\limits_0^{H_{\max}} K_l N_l \,\mathrm{d}\,h = m^g_{l_0}\,, \qquad \int\limits_0^{H_{\text{eff}}} K_l N_l \,\mathrm{d}\,h = m^g_{l_{\text{eff}}}\,. \qquad (19)$$

For brightness measurements at two angles θ_1 and θ_2, Eq. (17) modifies into a system of two equations. Solving this system, we obtain

$$\frac{I^{\theta_1}_{\lambda_1,\,\lambda_2}}{I^{\theta_1}_{\lambda_1,\,\lambda_2}} = \exp\left[-\left(\frac{1}{\cos\theta_1}-\frac{1}{\cos\theta_2}\right)\right]\times$$

$$\times\, m^g_{l_0}\exp\left[\left(\frac{1}{\cos\theta_1}-\frac{1}{\cos\theta_2}\right)\right] m^g_{l_{\text{eff}}} =$$

$$=\exp\left[-\left(\frac{1}{\cos\theta_1}-\frac{1}{\cos\theta_2}\right)(m^g_{l_0}-m^g_{l_{\text{eff}}})\right]=$$

$$=\exp\left[-\left(\frac{1}{\cos\theta_1}-\frac{1}{\cos\theta_2}\right)m^g_*\right], \qquad (20)$$

where

$$m^g_* = m^g_{l_0} - m^g_{l_{\text{eff}}}\,. \qquad (21)$$

Finally, from the expression for two–wavelength measurements at θ_1 (or θ_2) and m^g_* derived from Eq. (20), we obtain the integral gas content for optical depth $m^g_{l_0}$

$$I^{\theta_1}_{\lambda_1,\,\lambda_2} = \exp\left[-\,m^g_{l_0}\right]\,\exp\left[1-\frac{1}{\cos\theta_1}\right]m^g_*\,. \qquad (22)$$

We note that the optical depth $m^g_{l_{\text{eff}}}$ defined by Eqs. (18) and (19) is not strictly constant at different zenith viewing angles and may vary with the viewing angle due to the H_{eff} dependence on θ. However, these variations, in accordance with our preliminary estimates, are insignificant as compared with the optical depth $m^g_{l_0}$. In future we plan a thorough assessment of the method accuracy, including the errors introduced by $m^g_{l_{\text{eff}}}$, multiple scattering, spectral separation of sounding wavelengths, and spectral characteristics of specific ATG constituents.

Summarizing, the proposed differential technique of measuring at two close wavelengths and two solar zenith angles makes it possible to determine the integral ATG content over the entire atmospheric depth. The author is gratefully indebted to B.S. Kostin for his consultation on some mathematical problems.

REFERENCES

1. G.P. Gushchin, *Investigation of Atmospheric Ozone* (Gidrometeoizdat, Leningrad, 1963), 269 pp.

2. G.P. Gushchin and N.N. Vinogradova, *Total Ozone Content in the Atmosphere* (Gidrometeoizdat, Leningrad, 1983), 238 pp.