Retrieval of Aerosol Optical Depth, Aerosol Size Distribution Parameters, Ozone and Nitrogen Dioxide Column Amounts from Multifilter Rotating Shadowband Radiometer Data

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Introduction

The MultiFilter Rotating Shadowband Radiometer (MFRSR) (Harrison and Michalsky 1994a, 1994b) measures atmospheric column extinction of the direct solar beam and the diffuse radiation intensity at six wavelengths. Located at the National Aeronautics and Space Administration's Goddard Institute for Space Studies in New York City, the instrument has six spectral channels at 415, 501, 616, 672, 870, and 940 nm. The measurements are made at 1-minute intervals throughout the day.

The atmospheric column extinction optical depth corresponding to the ith channel can be expressed in terms of the parameters measured by MFRSR as

$$\tau_{i} = -\ln\left(\frac{I_{i}}{I_{i}^{0}}\right) \cdot \mu + c_{i}\mu, \qquad (1)$$

where μ is the cosine of the solar zenith angle, I_i^0 are the top of the atmosphere solar radiation intensities, and c_i are the natural logarithms of the respective calibration coefficients that convert measured detector voltages to intensities in Wm⁻². Below, we will use the name "calibration coefficients" for the coefficients c_i .

The atmospheric contributions to the extinction optical depth are as follows: Rayleigh scattering, NO_2 and O_3 absorption, aerosol and clouds scattering and absorption, and water vapor absorption (affecting only the 940-nm channel). The contribution of the Rayleigh scattering is accurately known and is subtracted out in all six channels before the data analysis starts. Water vapor is not a subject of the present study, so we do not consider the 940-nm channel, the only one affected by the water vapor absorption. The data obtained by means of the MFRSR allows us to retrieve the aerosol optical depth (at least for clear and partially clear days), some parameters of the aerosol size distribution, and the column amounts of ozone and NO_2 . However, accurate retrieval of these quantities is not easy.

First of all, there is no simple and reliable way to calibrate the instrument, while the physical quantities retrieved, say, by a direct least squares fit from MFRSR data are very sensitive to errors in calibration. It seems that the necessary absolute accuracy of knowing the calibration coefficients c_i is about 0.01-0.02. Calibration of the instrument is especially problematic because of the unstable behavior of the calibration coefficients in time: systematic drift (which for some filters can produce a difference in c_i about 1.2 in a 1-year period) and day-to-day fluctuations of the order up to 0.1 in c_i .

Another problem is to find parameters of the aerosol size distribution that can be reliably determined from the MFRSR data.

We present an original procedure that overcomes these difficulties and allows us to accurately retrieve both the physical quantities and the calibration coefficients. This procedure consists of the following steps: 1) determine the calibration coefficient (and therefore the aerosol optical depth) in the fifth channel (870 nm) using the direct to diffuse ratios, 2) separate the aerosol extinction from other factors and determine the aerosol size distribution parameters (and therefore, the aerosol optical depths in all channels), 3) retrieve NO₂ and ozone column amounts (together with the calibration coefficients in the first two channels), 4) determine the remaining calibration coefficients.

Step 1. To model the diffuse flux, multiple scattering calculations were performed using the adding and dubbing method. We have to restrict ourselves to consideration of the

fifth (870 nm) channel only because this channel is not affected by NO_2 and ozone absorption, and therefore the results of computations do not depend on the unknown vertical distribution of these gases.

Fortunately, the dependence of the direct to diffuse ratios $\Phi = I_{dir}/I_{dif}$ on the surface albedo is analytical, so it can be easily expressed in terms of the other quantities by the formula

$$A = \frac{1}{s} \left(\frac{1 - (\Phi/\Phi_0)}{\mu \Phi + 1} \right)$$
(2)

where

A is the surface albedo

 Φ and Φ_0 are the direct to diffuse ratios corresponding to this albedo and to zero surface reflectivity, respectively

 $s=s(\tau_5)$ is the reflection function integrated over angles (King and Herman 1979).

The surface albedo is an unknown quantity, however; for clear days, its dependence (2) on the aerosol optical depth is so sharp that any natural assumption like 0 < A < 50% gives the value of the optical depth, or, equivalently, of the calibration coefficient c_5 , with an accuracy ± 0.01 . Our calculations also showed that the result practically does not depend on the preassumed aerosol particle size used in the model.

Step 2. The measured quantities and the quantities to be determined satisfy the following system of five equations:

$$\tau_{1} = q_{1}\tau_{a} + \beta_{1}x_{NO_{2}} + c_{1}\mu$$

$$\tau_{2} = q_{2}\tau_{a} + \beta_{2}x_{NO_{2}} + \gamma_{2}x_{O_{3}} + c_{2}\mu$$

$$\tau_{3} = q_{3}\tau_{a} + \beta_{3}x_{NO_{2}} + \gamma_{3}x_{O_{3}} + c_{3}\mu$$

$$\tau_{4} = q_{4}\tau_{a} + \beta_{4}x_{NO_{2}} + \gamma_{4}x_{O_{3}} + c_{4}\mu$$

$$\tau_{5} = \tau_{a} + c_{5}\mu$$

where

 τ_i is the measured optical depths in the i^{th} channel

 $q_i = Q_{ext}^i/Q_{ext}^5$ are extinction ratios normalized to the fifth channel ($q_5 = 1$)

 β_i are NO₂ spectral absorption coefficients ((B₅ = 0))

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 γ_i are O₃ spectral absorption coefficients ($\gamma_1 = \gamma_5 = 0$)

 x_{NO_2} and x_{O_2} are NO₂ and ozone amounts, respectively

 τ_a is the aerosol optical depth in the fifth channel

c_i are calibration coefficients.

By means of a simple row reduction, one can exclude x_{NO_2} , x_{O_3} , and τ_a from the system, which then (after division by μ) takes the form:

$$F_3 = B_3(x - c_5) + A_3, \quad F_4 = B_4(x - c_5) + A_4$$
 (3)

where $x = \tau_5/\mu$

$$F_{k} = \frac{1}{\mu} \left(\tau_{k} - \frac{\beta_{k}}{\beta_{1}} \tau_{1} - \frac{\gamma_{k}}{\gamma_{2}} \left(\tau_{2} - \frac{\beta_{2}}{\beta_{1}} \tau_{1} \right) \right)$$

$$A_{k} = c_{k} - \frac{\beta_{k}}{\beta_{1}} c_{1} - \frac{\gamma_{k}}{\gamma_{2}} \left(c_{2} - \frac{\beta_{2}}{\beta_{1}} c_{1} \right)$$

$$(4)$$

$$\mathbf{B}_{k} = \mathbf{q}_{k} - \frac{\beta_{k}}{\beta_{1}} \mathbf{q}_{1} - \frac{\gamma_{k}}{\gamma_{2}} \left(\mathbf{q}_{2} - \frac{\beta_{2}}{\beta_{1}} \mathbf{q}_{1} \right)$$
(5)

k = 3,4

It is easy to see that all points on the plot of $F_{\nu}(x)$ corresponding to the same aerosol size distribution belong to a straight line with the slope B_k passing through the "calibration point" with the coordinates (c_5, A_1) . This point must be located strictly to the left of the curve; otherwise, the aerosol optical depth in the fifth channel would be negative. Actually, we do not know which points belong to the same mode, so the calibration point cannot be found as the intersection of all such lines. However, the shape of the curves $F_3(x)$ and $F_4(x)$ is such that it "points" to the calibration point. Rigorously speaking, this means that if we know the x-coordinate c_5 (obtained in step 1), we can determine the values of A_k by the condition of the minimal mean deviation of the corresponding slopes B_k from their mean values. Once the calibration points are fixed, the values of B_k can be determined from (3).

The method described is very similar to the Langley calibration. The only difference is that now we deal with the variability of the aerosol size distribution, which appears to be much more stable then the aerosol optical depth.

The coefficients B_3 and B_4 depend only on the (unknown) aerosol extinction ratios q_i . Therefore, formally they can be used to determine the parameters r_{eff} and v_{eff} of a preassumed aerosol size distribution by solving numerically the system of equations (5) (k=3,4), where $q_i = q_i$ (r_{eff} , v_{eff}) are known functions of r_{eff} and v_{eff} obtained, e.g., from Mie theory using this model aerosol size distribution. Solutions to each of these equations constitute a (level) curve in the (r_{eff} , v_{eff})-plane, and the solution of the system is the point of intersection of these level curves.

However, in practice these two level curves either do not intersect or the point of their intersection is unreliable (it depends on the shape of the pre-assumed aerosol size distribution). This means that, in general, as far as extinction ratios are concerned, $r_{\rm eff}$ and $v_{\rm eff}$ are not universal parameters and their values make sense only if the shape of the distribution is accurately known.

However, we found that in the case of relatively small particles ($r_{eff} < 0.5 \mu m$ at $v_{eff} = 0$, which often takes place in reality), the level curves are very close to each other and can be treated as one curve. In this case, the point of intersection is certainly unreliable, but the position of the curve itself is well-defined and does not significantly depend on the preassumed aerosol size distribution shape. Actually, the position of this curve is the only information about the aerosol size distribution that we can reliably retrieve. We will characterize it by the value of r_{eff} corresponding to the point of the curve with $v_{eff} = 0$. This r_{eff} will be called "mono-distribution radius" (because it is calculated under the assumption that all aerosol particles have the same size).

The dependence of the individual extinction ratios q_i on the unknown v_{eff} along the curve should be taken into account only for the first two channels, where it can cause some uncertainty in determining aerosol optical depth, which is not very big when r_{eff} is less than 0.5 µm (about 10% to 15% for the first channel and about 3% to 5% for the second) one). Changes in the asymmetry parameter g with v_{eff} do not exceed 5%. Thus, we can say that the knowledge of the mono-distribution radius gives us quite accurate information about the aerosol extinction properties and its asymmetry parameter.

Knowing B_3 (or B_4) at each moment, we can determine the mono-distribution radius as a function of time. On average, the values obtained from B_4 coincide with the ones from B_3 ; however, they usually contain more noise, so we keep them only as a reference (i.e., we practically do not use the fourth channel in our analysis). After the mono-distribution radius is determined the aerosol optical depths in all channels can be calculated.

Step 3. These optical depths can be then subtracted from the data. The remaining optical depth in the first channel is caused by NO_2 absorption, in the second channel, by both NO_2 and ozone absorptions. Of course, calibration effects are present in both these channels.

To separate NO₂ and ozone absorption from calibration in the first channel, we use a simple least squares fit procedure similar to the Langley calibration (in the case of ozone, we analytically exclude the NO₂ contribution from the second channel first). This procedure is excellent for ozone, whose column amount almost does not change during the day. In the case of NO₂, however, it is not very accurate because of the high variability of NO₂ column amount. The situation is even more complicated because of the dependence of the retrieved NO₂ column amount values on the unknown aerosol v_{eff}, thus it can be measured only with an accuracy about 30% to 50%.

The results of the ozone retrieval are much better and are in good agreement with the Earth Probe Total Ozone Mapping Experiment Spectrometer (TOMS) measurements.

Step 4. The method described above allows us to determine the values of the calibration coefficients together with the physical quantities. In addition to the calibration coefficients determined on the previous steps, c_3 and c_4 can be obtained from A₃, A₄, c_1 and c_2 , or directly by subtracting the determined physical optical depths from the measured ones.

Figure 1 shows the results of our analysis of the clear and partially clear days data for the period from August 25, 1995, to November 27, 1996.

The top two plots show the daily mean aerosol optical depth (at 550 nm) and the daily mean aerosol mono-distribution radius for the whole observation period. The polynomial fits show seasonal variations of these parameters. We see that the mean aerosol optical depth in winter is smaller than in summer, while the aerosol particles are bigger. This effect may be related to changes in condensation and evaporation conditions; however, so far, we do not have any satisfactory explanation.

The bottom two plots show changes in NO₂ and ozone column amounts. (The plots show how the accuracy of retrieval is limited by dependence on the unknown aerosol v_{eff} .) One can see from the third plot that, despite the low accuracy of NO₂ retrieval and its high variability, the average values given by the polynomial fit show reasonable seasonal behavior (maximum in May-June, minimum in November-December) consistent with the results of other authors. The



Figure 1. Plots of data from clear and partially clear days from August 25, 1995, to November 27, 1996.

polynomial fit curve on the bottom plot shows the maximum in the ozone column amount in spring (April-May), which is typical for our latitude.

References

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