Multi-Spectral Atmospheric Column Extinction Analysis of Multi-Filter Rotating Shadowband Radiometer Measurements

A.A. Lacis and B.E. Carlson
National Aeronautics and Space Administration/Goddard Institute for Space Studies
New York, New York

B. Cairns
Columbia University
National Aeronautics and Space Administration/Goddard Institute for Space Studies
New York, New York

Abstract

Multi-spectral Atmospheric Column Extinction (MACE) analysis of multi-filter rotating shadowband radiometer (MFRSR) measurements yield detailed time series information on the variations of the column amounts of atmospheric NO_2 and ozone and of aerosol optical depth, including the effective particle size and variance of the aerosol size distribution. Retrieval of such detailed information is possible because the MFRSR makes precise measurements of atmospheric extinction of the direct solar beam simultaneously at six wavelengths at 1-minute intervals throughout the day. MACE analysis retrieves relative variations in column ozone to the order of 1 Dobson Unit and NO_2 variations to about 1 ppt. The time series variability in aerosol optical depth is strongly correlated with variations in NO_2, and is anti-correlated with variations in ozone. Periodic fluctuations with a period on the of order 30 minutes are seen in the data. These fluctuations appear to be related to changes in aerosol optical depth and/or particle size that are probably associated with boundary layer gravity waves. While mono-modal aerosol size distributions yield a close fit to MFRSR data, the remaining wavelength dependent residuals in column extinction point to the need of at least a bi-modal size distribution in order to fit the time series variations of atmospheric extinction. Results of the MACE analysis show the MFRSR to be a very capable remote sensing instrument, in the same class as polarimeters and interferometers, which rely on precise relative measurements for their information content. Thus, precise absolute calibration, which would, nevertheless, be of great importance for improving MFRSR data analysis, is not actually a first order limitation for effective information retrieval for the MFRSR.

MACE Analysis of MFRSR Measurements

The MFRSR makes precise measurements of atmospheric (column) extinction of the direct solar beam simultaneously at six wavelengths (nominally, 415, 500,615, 670, 840, and 940 nm) at 1-minute intervals throughout the day. As the atmospheric airmass traversed by the solar direct beam changes during the course of the day, the MFRSR records the corresponding changes in atmospheric extinction in the form

$$I_{in} = c_n \, I_n^o \exp(-T_{in} / \mu_i) \quad (1)$$

where μ_i is the solar zenith angle at the i^{th} measurement time step, I_{in} are the six spectral intensities measured by the MFRSR at the i^{th} time step, and T_{in} are the corresponding atmospheric column extinctions at the MFRSR wavelengths at the i^{th} time step. I_n^o are the top of the atmosphere (TOA) solar radiation intensities at the MFRSR wavelengths and c_n are the respective calibration coefficients that convert measured detector voltages to intensities in Wm^{-2}.

It follows then directly from (1) that the atmospheric column extinction optical depths, T_{in} are given by

$$T_{i,n} = \mu_i \log (I_n^o / I_{i,n}) + \mu_i \log (c_n) \quad (2)$$

The atmospheric contributors to the extinction optical depth (shown in Figure 1) are Rayleigh scattering, NO_2, O_3, aerosol, cloud (when present), and H_2O (affecting only the 940-nm channel). Since the spectral dependence of Rayleigh scattering is accurately known,

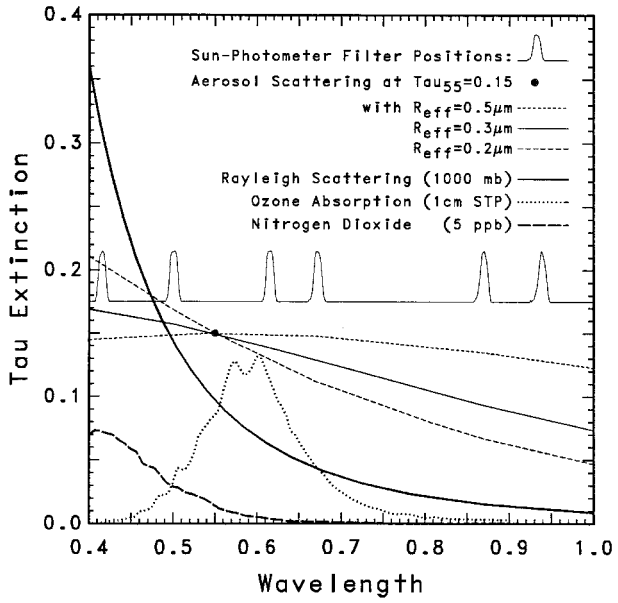

Figure 1. Atmospheric extinction.

$$T_{Ray} = 0.008569\ \lambda^{-4}(1 + 0.0113\ \lambda^{-2} + 0.00013\ \lambda^{-4})\ P/P_o \qquad (3)$$

where λ is the wavelength in μm, and P/P_O is the atmospheric pressure relative to 1013.25 mb, the column extinction due to Rayleigh scattering is determined with high precision by a barometric measurement of surface pressure and knowledge of the solar zenith angle (Hansen and Travis 1974). Accordingly, the Rayleigh extinction given by (3) is subtracted out in all six channels in (2). The 940-nm channel, which is the only channel affected by water vapor extinction, is analyzed separately.

The standard approach to interpreting the MFRSR extinction measurements has been to construct Langley regression plots of log ($I°_n/I_{i,n}$) versus Airmass ($1/\mu$) to back out the column extinction and the TOA solar intensity. This yields the twice-daily determinations of mean column extinction in the six MFRSR spectral channels which then can be inverted to extract the mean NO_2, O_3, and aerosol column amounts. However, each spectral set of MFRSR measurements is individually a precise measure of the instantaneous column extinction; a great deal of information is contained in the time series of daily MFRSR measurements. This information can be extracted by first renormalizing the calibration coefficients c_n using MFRSR data from the clearest day available and performing least-squares inversions of MFRSR data as described below.

MACE Analysis

MACE is the Multi-spectral Atmospheric Column Extinction analysis performed using the MFRSR time series measurements limited to filters 1 through 5 (see Figure 2). Note that the only contributors of atmospheric extinction in filters 1 through 5 are NO_2, O_3, and aerosol (and cloud, when present). We can thus write the time series of MFRSR extinction measurements in matrix form as

$$T_{in} = \delta_{ik}\, a_{kn} \qquad (4)$$

where T is a five-column matrix with the index n (n = 1, ... 5) referencing the five 'aerosol' channels of the MFRSR, with I denoting the i^{th} time step. Similarly δ is a three-column matrix (k = 1,2,3) depicting the time variations in column NO_2 amount, column O_3, and column aerosol optical depth referenced at λ = 0.55 μm. The solar spectrum weighted absorption coefficients for the respective filters and extinction contributors are given by

$$a_{kn} = \begin{bmatrix} a_{11} & a_{12} & a_{13} & a_{14} & a_{15} \\ a_{21} & a_{22} & a_{23} & a_{24} & a_{25} \\ a_{31} & a_{32} & a_{33} & a_{34} & a_{35} \end{bmatrix} \qquad (5)$$

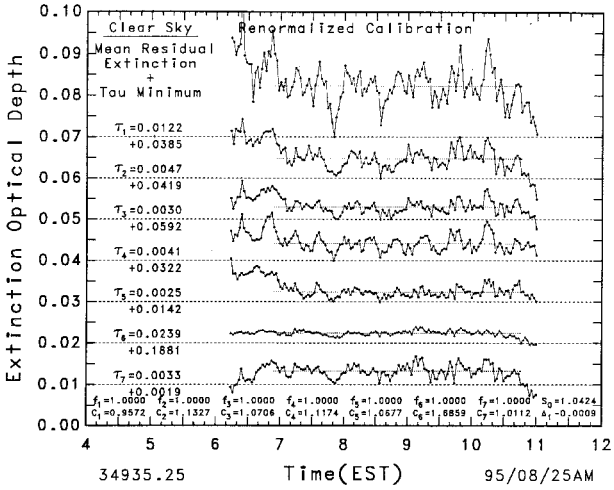

Figure 2. MACE daily time series (IDN = 1).

The extinction coefficients for the gaseous absorbers, NO_2 and O_3, are fixed values that are obtained by computing the spectrally weighted averages of NO_2 and O_3 absorption with respect to MFRSR filter functions and the solar spectrum. Thus,

$$a_{kn} = \int_0^\infty K_k(\lambda)\, F_n(\lambda)\, S(\lambda)\, d\lambda \Big/ \int_0^\infty F_n(\lambda)\, S(\lambda)\, d\lambda \qquad (6)$$

For aerosol, the spectral extinction coefficients a_{3n} are derived from Mie scattering calculations covering the full range of particle sizes, r_{eff}, and variances, v_{eff}, that are likely to be encountered, with specific values obtained in the process of least-squares fitting to the observed extinctions.

A first estimate of the column extinction contributions by NO_2, O_3, and aerosol, including also the mean particle size, r_{eff}, and variance, v_{eff}, of the aerosol, is obtained by taking the mean values of the time series extinction given in (4) and performing a brute force numerical least-squares fit (shown in Figure 3) to

$$\bar{T}_n = \bar{\delta}_k\, a_{kn} \qquad (7)$$

This generally yields a close fit to the time-mean spectral dependence of extinction. We next assume that the first order time series residuals,

$$T'_{in} = T_{in} - \bar{\delta}_k\, a_{kn} \qquad (8)$$

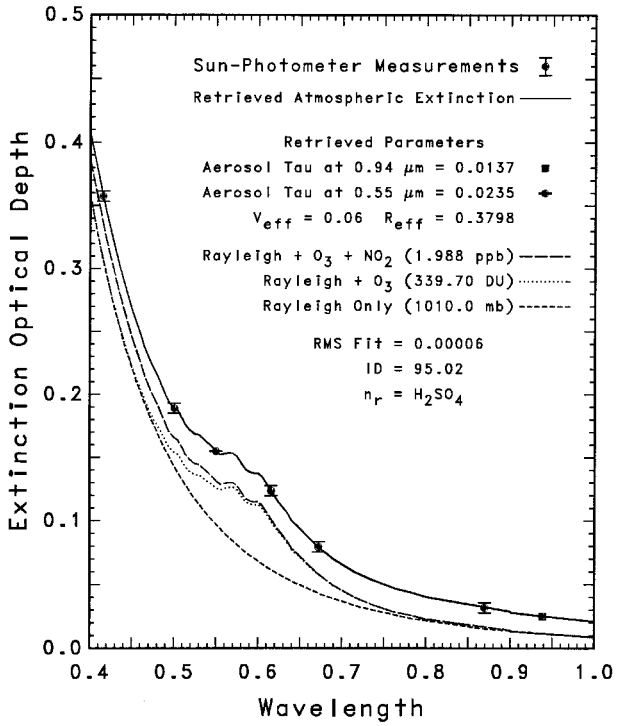

Figure 3. MACE retrieved parameters.

can be explained in terms of column amount perturbations, $\delta'_{k'}$ of NO_2, O_3, and aerosol optical depth (with fixed r_{eff} and v_{eff} as determined above). Thus,

$$T'_{in} = \delta'_{ik}\, a_{kn} \qquad (9)$$

which solve by means of a least-squares fit. This is accomplished by multiplying (9) from the right by the transpose of a_{kn} so that

$$T'_{in}\, \tilde{a}_{nk} = \delta'_{ij}\, a_{jn}\, \tilde{a}_{nk} \qquad (10)$$

We then construct the inverse matrix a_{jk}^{-1} of $a_{jn}\, \tilde{a}_{nk}$ and obtain the time series variations about the mean extinction values by

$$\delta'_{ik} = T'_{in}\, \tilde{a}_{nj}\, a_{jk}^{-1} \qquad (11)$$

The time series $\delta_{ik} = \bar{\delta}_k + \delta'_{ik}$ in Figure 4 shows the minute-to-minute fluctuations that take place in the column amounts of NO_2 and O_3, and aerosol optical depth. This accounts for most of the extinction variability observed in the MFRSR measurements. Thus far the retrieval analysis has assumed that r_{eff} and v_{eff} do not vary. If indeed the aerosol size and variance of the size distribution were to remain constant, we would then expect the second order extinction residuals

$$T''_{in} = T_{in} - (\bar{\delta}_k + \delta'_{ik})\, a_{kn} \qquad (12)$$

that remain after the least-squares fitting to be nothing but random noise. That, however, is not the case, as periodic

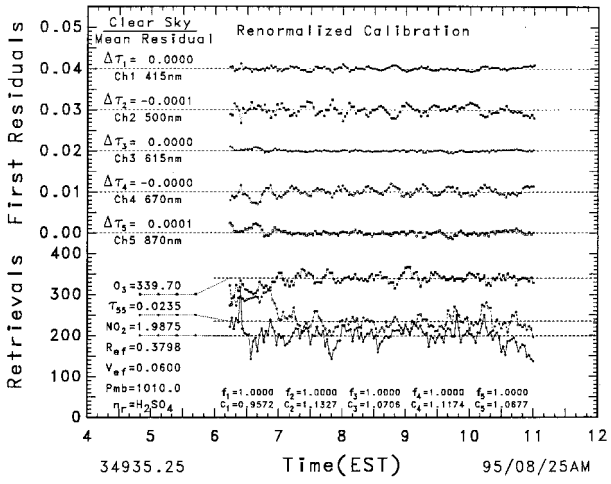

Figure 4. MACE mean retrieval results (IDN = 1).

fluctuations characteristic of gravity waves still remain in the second order residuals T''_{in}. Performing brute force least-squares fits using mono-modal size distributions at specific points along the time series cannot make the residuals disappear. This implies the need for a bi-modal aerosol size distribution in order to improve the model fit to the observed data.

Discussion

The success of the MFRSR measurements is due to the high precision with which the shadow band occultation of the sun permits isolation of the direct solar beam extinction (Wesley 1982), along with the cosine corrected response function of the instrument which permits accurate calibration of the separation of the direct and diffuse components of solar radiation (Harrison et al. 1994). In addition, the stable operation of silicon photodiodes and accurate knowledge of the solar zenith angle further assure the integrity of the MFRSR measurements of atmospheric column extinction. Once the data from a good clear day are used to establish the relative instrument calibration, each set of instantaneous spectral measurements acquired by the MFRSR can then be individually analyzed to retrieve the extinction contributions by the individual contributors.

In the five 'aerosol' channels of the MFRSR, Rayleigh scattering, aerosol scattering, and absorption by NO_2 and O_3 are the only contributors. The Rayleigh contribution, however, can be subtracted out directly if the surface pressure is known. Cloud extinction, in those cases when cloud is present, is also easily recognizable by its characteristic spectral signature and generally larger optical depths. The optical depth of cloud extinction up to an optical depth of $\tau = 4$ can also be determined. This leaves the column amounts of NO_2 and O_3, and the aerosol optical depth, particle size, and size distribution variance as the five physical parameters which can be extracted by means of brute force least-squares fitting to the observed spectral extinction. In principle, if the absorption and scattering coefficients are accurately known, no residual other than detector noise should be left following a successful model fit to the data.

The time dependence of the leftover residuals and the inability to improve the fit with a mono-modal size distribution suggest the existence of at least two different modes of aerosol sizes. Clearly, stratospheric aerosols and tropospheric aerosols are different in composition and probably in size. It is not clear whether the fluctuations in the second order residuals arise from stratospheric or tropospheric aerosols, or whether it is aerosol optical depth or particle size that varies. Possible candidates are optical depth variations, as boundary layer aerosols serve as tracers of the thinning and thickening of the boundary layer under the influence of gravity waves. It is also possible that gravity-wave-induced temperature and humidity variations drive hygroscopic sulfate aerosols to vary in size, and thus affect the extinction optical depth.

We find the time series variations in NO_2 to be similar to the variations that take place in aerosol optical depth. The variations in column O_3, on the other hand, appear to be anti-correlated with the fluctuations in NO_2 and aerosol. While most of the NO_2 and O_3 are located in the stratosphere, the bulk of the aerosol is more than likely tropospheric. This uncertainty is unlikely to be resolved through MFRSR data alone. The time series variations in NO_2 and O_3 column amounts and in aerosol optical depth appear to be robust; performing the MACE analysis using H_2SO_4 and H_2O refractive indices for the aerosol produces essentially the same variability except for small off-setting differences between the mean NO_2 and O_3 amounts and aerosol size and optical depth. This result implies, however, that use of a spectrometer rather than the five MFRSR filters would constrain the gaseous components with their unique spectral signature and would allow the MFRSR to have some capability to distinguish aerosol composition. The MACE analysis also implies that the MFRSR measurements could be used for real-time monitoring of the column amounts of O_3 and NO_2 and of the aerosol size and optical depth.

References

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