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Title: "Optimal Aerosol Parameterization for Remote Sensing Retrievals"

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Abstract

We have developed a new algorithm for the retrieval of aerosol and gases from SAGE III solar transmission measurements. This algorithm improves upon the NASA operational algorithm in several key aspects, including solving the problem non-linearly and incorporating a new methodology for separating the contribution of aerosols and gases. In order to extract aerosol information we have built a huge database of aerosol models for both stratospheric and tropospheric aerosols, and polar stratospheric cloud particles. This set of models allows us to calculate a vast range of possible extinction spectra for aerosols, and from these, derive a set of eigenvectors which then provide the basis set used in our inversion algorithm. Our aerosol algorithm and retrievals are described in several articles (listed in References Section) published under this grant. In particular they allow us to analyze the spectral properties of aerosols and PSCs and ultimately derive their microphysical properties. We have found some considerable differences between our spectra and the ones derived from the SAGE III operational algorithm. These are interesting as they provide an independent check on the validity of published aerosol data and, in particular, on their associated uncertainties. In order to understand these differences, we are assembling independent aerosol data from other sources with which to make comparisons.

We have carried out extensive comparisons of our ozone retrievals with both SAGE III and independent lidar, ozonesonde, and satellite measurements (Polyakov et al., 2004). These show very good agreement throughout the stratosphere and help to quantify differences which can be attributed to natural variation in ozone versus that produced by algorithmic differences. In the mid - upper stratosphere, agreement with independent data was generally within 5 - 20%, but in the lower stratosphere the differences were considerably larger. We believe that a large proportion of this discrepancy in the lower stratosphere is attributable to natural variation, and is also seen in comparisons between lidar and ozonesonde measurements.

NO_2 profiles obtained with our algorithm were compared to those obtained through the SAGE III operational algorithm and exhibited differences of 20 - 40%. Our retrieved profiles agree with the HALOE NO_2 measurements significantly better than those of the operational retrieval.

In other work (described below), we are extending our aerosol retrievals into the infrared regime and plan to perform retrievals from combined uv-visible-infrared spectra. This work will allow us to use the spectra to derive the size and composition of aerosols, and we plan to employ our algorithms in the analysis of PSC spectra. We are presently also developing a limb-scattering algorithm to retrieve aerosol data from limb measurements of solar scattered radiation.

Objectives

The objectives of the proposed work are:

- to improve the retrieval of both aerosol and gas information from SAGE II and SAGE III especially in the presence of volcanic aerosols and polar stratospheric cloud (PSC) particles
- to develop a methodology that can be consistently applied to many solar occultation experiments over a wide spectral range from UV to IR and to investigate alternative schemes for extracting simultaneous aerosol and gas information from experiments with very high spectral resolution such as those employing Fourier Transform Infrared Spectroscopy (e.g. ATMOS, MarkIV) and linear arrays of detectors (e.g. SAGE III)
- to facilitate the retrieval of aerosol optical and microphysical information, especially in the troposphere, from combined UV-visible-IR experiments
- to develop a methodology using the proposed parameterization scheme for the retrieval of gas and aerosol information from limb scattering satellite experiments

Description of work

1. A new algorithm for the retrieval of vertical profiles of ozone and nitrogen dioxide from SAGE III solar occultation measurements

Our new algorithm, developed under this grant, for the retrieval of atmospheric gas concentrations and aerosol extinction from the SAGE III transmission data is described by Polyakov et al. (2004). This algorithm uses an aerosol extinction parameterization based upon an expansion using the eigenvectors of the aerosol extinction correlation matrix. This matrix was constructed via numerical simulation for a large database of models of stratospheric and tropospheric aerosol, also developed under this grant (Timofeyev et al., 2003).

In order to assess the potential accuracy of the retrieval of atmospheric parameters by our algorithm, closed–loop numerical experiments were performed taking into account the SAGE III random measurement noise. These simulations produced errors in retrieving ozone of ~ 5 % on average in the altitude range of 12 – 70 km, increasing outside this range. The mean errors in retrieving NO_2 concentration were 6 – 8 % at altitudes of 20 – 45 km. The optimal number of eigenvectors in the expansion of the aerosol extinction coefficient was determined to be 4. Errors in the assignment of tangent height have a strong influence on the errors in retrieving the atmospheric parameters, and can increase the error by two to three times for a height error of 0.5 km.

Extensive comparisons of our retrievals with those from other satellites, ozonesondes, and lidar were made. In addition, the results from our algorithm were compared with those of the SAGE III operational algorithm. Comparison of ozone profiles with ozonesonde measurements (collocated in time (within 24 hours) and space (within 550 km)) demonstrated good agreement (better than 10 – 20 %) in the middle and upper stratosphere . In the lower stratosphere and upper troposphere, the disagreement was greater and could be 40 % or more. At altitudes of 25 –35 km the rms differences between the satellite and the ozonesonde measurements were two to three times less than ozone natural variability. However, in the lower stratosphere and upper troposphere, the rms difference was almost as great as the natural variability.

In most cases, our retrieved ozone data were found to be in good agreement with the lidar measurements (within 5 – 20 % at altitudes of 17 – 40 km). Below 17 km, the differences increased. Satellite measurements recovered the fine structure in the ozone vertical profiles well. At altitudes of 15 –35 km the rms satellite–lidar differences were 1.5 to 3 times the natural ozone variability. At lower altitudes these values were essentially equal. (See Figure 1.)

Comparisons of retrieved ozone with the HALOE data demonstrated good agreement. On average, differences between individual retrievals did not exceed 10 – 20 %. Typical differences were less than 10 %, well within the sum of the estimated errors in both measurements.

Because ozone concentrations from SAGE III showed large differences from ozonesonde and lidar measurements in the lowest part of the stratosphere, it was instructive to investigate how well these independent lidar and ozonesonde measurements agreed with each other. We compared these using the same collocation criteria (within 550 km and 24 hours) and found that mean and rms differences were 3 – 6 % at 17 – 32 km. Below 17 km differences increased up to 20 – 60 % for the mean and 80 % for the rms. Thus it appears that large differences in ozone observed below 15 - 17 km are not necessarily due to the satellite retrieval but exist between other sets of measurements also.

Our retrievals of NO_2 vertical profiles demonstrated spatial–temporal variations that are in good agreement with NO_2 seasonal and latitudinal models. Comparison of our NO_2 retrievals with HALOE measurements showed good agreement in general, but in a number of cases the differences are considerable. Mean differences between the two satellite measurements were 5 – 20 % and rms differences, less than 20 % above 18 km. In the lower stratosphere, mean and rms differences increased up to 30 % and 45 %, respectively. SAGE III NO_2 retrievals using the operational algorithm differ systematically from those using our algorithm with mean and rms differences of 20 – 40 %. (See Figure 2.) The NO_2 profiles obtained with our algorithm agree with HALOE measurements significantly better than the operational retrieval profiles.

2. The retrieval of aerosol extinction spectra from SAGE III data and their comparison with the aerosol measurements of HALOE

Aerosol extinction coefficients were retrieved using the parameterization scheme described in Timofeyev et al. (2003) in which an optimal expansion in the eigenvectors of the aerosol extinction correlation matrix is employed to describe the spectral variation. In our case, this parameterization makes it possible to determine aerosol extinction as a function of wavelength in the 0.29–1.55 μm spectral range. Retrieval results were compared with the aerosol extinction spectra obtained through the NASA operational algorithm and with HALOE data.

A statistical comparison of the results of retrieving the aerosol extinction by our algorithm and by the NASA operational algorithm at different wavelengths has been shown that NASA coefficients are larger than ours. Mean and r.m.s. differences range from 10–20% and 20–40%, respectively, for long-wave channels, to 20–40% and 40–80% for short-wave ones. In the 0.8-1.2 μm spectral range the coefficients obtained by the two methods are similar.

To compare aerosol extinction coefficients from SAGE III and HALOE measurements, we employed regression to extrapolate the HALOE IR data to the SAGE III visible and near-IR spectral range using our statistical models of stratospheric aerosols. Theoretical errors of such an extrapolation from the HALOE channels to the SAGE III channels are 10–40% at 10-30 km under the assumption that the errors in the HALOE measurements are less than 15%.

The best agreement between SAGE III and HALOE aerosol data is observed at altitudes of 17-21 km and at the SAGE III channels between 0.4 and 1.1 μm. In some channels and at some altitudes, the agreement is better for aerosol extinction data retrieved by our algorithm, in other cases - for data retrieved by NASA operative algorithm. The small number of comparisons (limited to 45 collocated SAGE III and HALOE measurements) and extrapolation errors are such that we cannot evaluate the performance of one algorithm over another for interpreting the SAGE III aerosol retrievals. Thus it is necessary to obtain additional independent information for testing the quality of the aerosol retrievals. This is important given that the aerosol spectral data are further employed to solve another inverse problem – the retrieval of the microphysical characteristics of stratospheric aerosol.

In this next stage, aerosol extinction spectra were used to solve the inverse problem with respect to the retrieval of the size distribution function of the particles. In this problem, we have developed a regression method based on our statistical model of stratospheric aerosols. Our retrieved size distributions are in good agreement with climatological data for background stratospheric aerosols.

3. A new method for interpreting the limb measurements of solar scattered radiation

Using our large statistical database of microphysical characteristics of stratospheric aerosols, calculations of the optical properties of these aerosols were carried out for a large range of loading conditions. Aerosol properties, and the stratospheric temperature and humidity (which influence the composition and thus the refractive index of the aerosols) were varied within wide limits. Using these results, we constructed covariance and cross-covariance spectral and altitude matrices of the principal optical characteristics of aerosol – extinction and scattering coefficients, and scattering phase function. There are very strong covariance relationships for aerosol extinction, scattering coefficient, and phase function (correlation coefficients are 0.99–0.96) at different wavelengths in the 0.29–1.53 μm spectral range. The cross-covariance relationships between different optical characteristics are also significant and produce correlation coefficients up to 0.6–0.7. These relationships were then used as a priori information in solving the inverse problem with respect to the retrieval of aerosol optical characteristics from limb measurements of solar scattered radiation.

Using the radiative transfer code we developed for the spherical atmosphere and our statistical models of aerosol optical characteristics, we carried out numerical simulations of the LORA, SOLSE and SAGE III satellite experiments. We used these simulations to analyze the information content of limb measurements of scattered solar radiation with respect to aerosol optical characteristics using a variety of approaches in solving the inverse problem. We have found that by employing a retrieval method which uses *a priori* statistical information on spectral relationships between the different optical aerosol characteristics we can improve the accuracy of the retrievals relative to traditional approaches which do not use such statistical information.

4. The retrieval of aerosol microphysical properties from simulated IR spectra

Using simulated transmission data over the infrared spectral region between 800 cm^{-1} and 4750 cm^{-1} and errors derived from ATMOS measurements we have used optimal estimation theory to retrieve three parameters—composition, effective radius, and volume—associated with atmospheric aerosols. The covariance matrix generated from these simulations has been used to estimate the accuracy to which these parameters can be retrieved from real measurements. These error estimates are dependent on the size and composition of the aerosols, the total loading, and the spectral range employed.

For moderate loading conditions and the high signal to noise ratio afforded by these high-resolution infrared transmission measurements these aerosol parameters can be retrieved to accuracies of better than 3% for the range of normal stratospheric conditions when employing the full spectral range. However the errors increase substantially when the spectral range is more limited. Composition can still be retrieved to within a15% accuracy for the limited spectral range between 820 and 1240 cm^{-1}, but effective radius is very poorly retrieved if the spectral range does not extend beyond 4000 cm^{-1}.

Additional systematic error due to the use of an incorrect size distribution model or size distribution width are likely to results in larger errors — particularly in the case of the size parameters. Sulfuric acid weight percent could be retrieved to better than 2% regardless of the aerosol size parameters, but for the range of size distribution widths tested there was an error of as much as 50% in effective radius from the use of an incorrect size distribution model. Resulting errors in total volume were less than 10%.

The spectral range was much more important in determining accuracy than the number of data points within that range. This suggests that our results may be much improved by ultimately combining the UV-visible data from the MAESTRO experiment with the ACE infrared data.

It is clear from our study that given a correct form and composition model for stratospheric aerosols, their sulfuric acid weight percent, effective radius and total volume can be accurately retrieved. The use of actual ATMOS transmission spectra showed that the general shape of the residual spectra (spectra after the removal of the absorbing gases) is well-matched by typical aerosol representations and produces reasonable size and composition results. However there are some fairly large differences between the fitted and measured spectra which cannot be resolved by varying the three aerosol parameters applied in our study. This suggests that the real state of the stratospheric aerosol (viewed along the path of a solar occultation instrument) may not be modeled accurately by a single-composition, unimodal size distribution model. Additionally, spectroscopic errors from both the gas and the aerosol fitting may contribute to the disagreement. However, to determine whether systematic differences do exist between our models and the actual spectra will require high-resolution, calibrated transmission spectra over the full infrared spectral range such as those afforded by the ACE instrument.

Summary

We have developed and tested a new inversion algorithm for the interpretation of SAGE III solar occultation measurements. The ozone and nitrogen dioxide vertical profiles obtained have been compared to those obtained through the NASA operational algorithm and to ones from ozonesonde, lidar and other satellite experiments (see Section 1 above). Our new algorithm differs from the NASA operational algorithm in several key regards – (i) the algorithm takes into account the finite altitude and spectral resolution of the measurements by integrating over the width of the viewing window both spatially and spectrally; (ii) the problem is solved non-linearly using optimal estimation; (iii) the algorithm employs transmittance rather than optical density; (iv) the aerosol extinction is

parameterized by an optimal expansion using the eigenvectors of the aerosol extinction correlation matrix. This matrix was constructed through numerical simulation for a large database of models of stratospheric and tropospheric aerosol (see Timofeyev et al., 2003). Our work is described in an article submitted to *JGR* (Polyakov et al., 2004 – under review), which demonstrates our technique and presents our retrievals of ozone and NO_2. These show good agreement with independent measurements.

A new method has been developed for the retrieval of aerosol extinction spectra from SAGE III solar occultation measurements using optimal expansion as described in Timofeyev et al. (2003). These spectra have been compared to those from HALOE using regression to extrapolate the IR data to the visible and near-IR spectral range employed by SAGE III (see Section 2 above). In this work, we employed our statistical models of the stratospheric aerosol that were developed under the first year of funding. A regression method for retrieving the microphysical characteristics of the stratospheric aerosol from SAGE III aerosol extinction spectra has also been developed (see Section 2 above). This method uses our huge database of statistical models of tropospheric and stratospheric aerosol, and PSCs. The retrieved characteristics of the aerosols show good agreement with climatological data.

A new method for interpreting limb measurements of solar scattered radiation with respect to the retrieval of atmospheric aerosol characteristics has been developed (see Section 3 above). The key aspect of the method is the use of *a priori* information on the statistical relations between different aerosol optical characteristics: extinction and scattering coefficients, and phase function. To realize this, we have carried out extensive calculations of stratospheric aerosol optical characteristics and constructed covariance and cross-covariance matrices for extinction and scattering coefficients, and scattering phase function. An analysis of the information content of this method for obtaining the aerosol optical characteristics from limb scattering measurements has shown its advantage in comparison with the traditional approach.

We have also investigated the use of optimal estimation theory in the retrieval of aerosol microphysical properties from high-resolution infrared transmission spectra (see Section 4 above). This method has been employed to retrieve aerosol effective radius, composition and loading from simulated ACE (Atmospheric Chemistry Experiment) measurements. We plan to combine our different regression methods to extend them to the entire UV-visible-IR spectral range.

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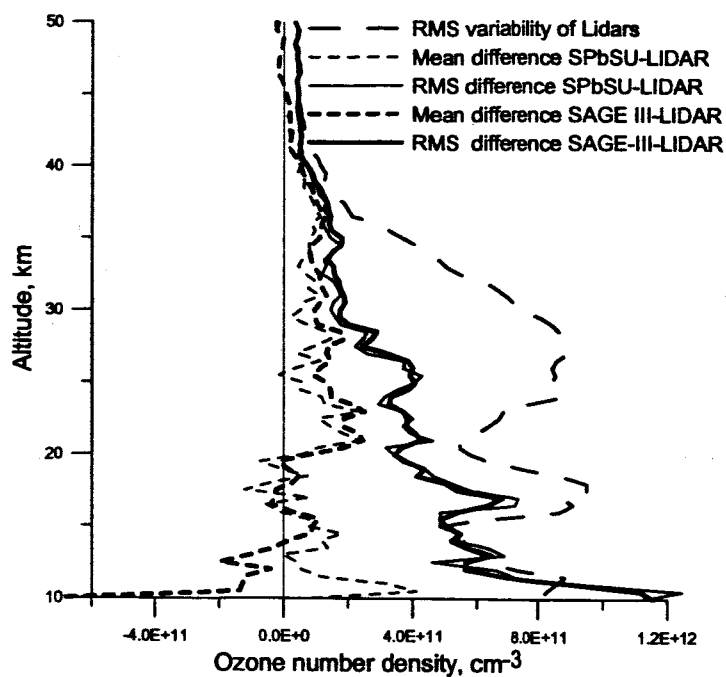
Figure 1. The statistics of the differences between the SAGE III ozone profiles retrieved by our (SPbSU) algorithm, the SAGE III operational algorithm and the lidar measurements
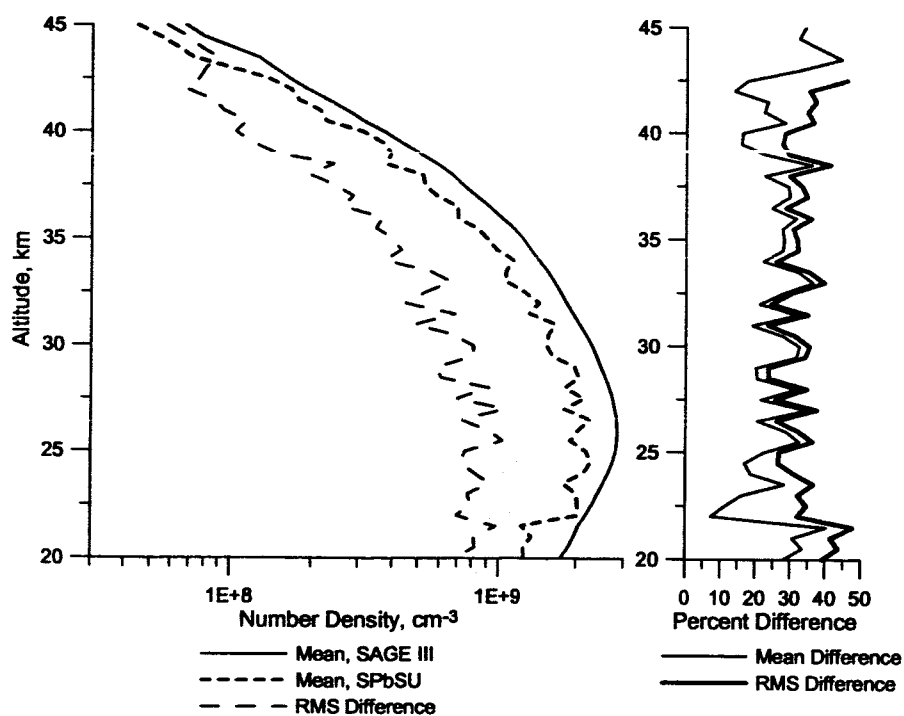

Figure 2. Comparisons of NO_2 profiles retrieved from SAGE III measurements using our (SPbSU) algorithm with those using the operational algorithm.