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Modeling of atmospheric radiative transfer with polarization and its application to the remote sensing of tropospheric ozone

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Abstract

Light reflected or transmitted by a planetary atmosphere contains information about particles and molecules in the atmosphere. Therefore, accurate modeling of the radiation field may be used to retrieve information on atmospheric composition. In this paper, a multi-layer model for a vertically inhomogeneous atmosphere is implemented by using the doubling-adding method for a plane-parallel atmosphere. By studying the degree of linear polarization of the transmitted and reflected solar light in the Huggins bands, we find significant differences between tropospheric ozone and stratospheric ozone. The effects of tropospheric ozone change on the linear polarization are 10 times more than that of the same amount of stratospheric ozone change. We also show the aerosol effect on the linear polarization, but this effect is wavelength independent as compared to that caused by the tropospheric ozone change. The results provide a theoretical basis for the retrieval of tropospheric ozone from measurement of linear polarization of the scattered sunlight both from the ground and from a satellite.

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1. Introduction

Previous studies have demonstrated the feasibility and high accuracy of the measurement of linear polarization from ground-based and spacecraft polarimetric remote sensing of planetary atmospheres

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(e.g., [1–7]). Mishchenko and Travis [7] have provided accurate retrieval algorithms of aerosol properties over the ocean by utilizing radiance and linear polarization measurements in the near-infrared region. However, such studies of linear polarization have not been applied to retrieval of tropospheric ozone.

Since the tropospheric column ozone is only about 10% of that in the stratosphere, the signal from tropospheric ozone is usually overwhelmed by the contribution from the much larger stratospheric layer. Therefore, the measurement of the tropospheric ozone by remote sensing from both ground and space-based instruments has always been a daunting challenge. In this study, we will investigate the possibility of detecting tropospheric ozone for the linear polarization signal in the Huggins bands from 310 to 345 nm, taking advantage of an extremely sensitive instrument that has very high spectral resolution.

O_3 and SO_2 are the only important molecules that absorb sunlight in the Huggins bands in the Earth's atmosphere. Since the concentration of SO_2 is much less than that of O_3, the absorption of solar radiation by SO_2 can be neglected in this study, except during or immediately after volcanic eruptions. The Huggins bands consist of a series of alternating absorption minima and maxima. The temperature-dependent ozone absorption cross-sections were determined by Malicet et al. [8] with resolution of 0.01 nm at 5 temperatures (218, 228, 243, 273, and 295 K). In particular, the temperature sensitivities appear to be different for the maxima and the minima, with the minima being more strongly dependent on the temperature. In addition, radiation in the troposphere is multiply scattered, whereas in the stratosphere single scattering dominates. These differences in the optical characteristics of ozone in the stratosphere and troposphere give rise to the possibility for distinguishing the signal of tropospheric ozone from stratospheric ozone [9].

The radiation field in a planetary atmosphere is generally polarized. The errors in the reflected intensity resulting from the neglect of polarization were examined by Hansen [10] and Lacis et al. [11] on the basis of accurate adding-doubling calculations of multiple scattering. They concluded that in most cases the errors in the scalar approximation should be less than or of the order of 1% for light reflected by a cloud of spherical particles with sizes of the order of or larger than the wavelength of light, which makes the scalar approximation applicable for radiance calculations for cloud and aerosol layers. On the other hand, it has been known that the errors can be much larger in the case of a semi-infinite atmosphere with pure Rayleigh scattering. For light reflected by finite Rayleigh atmospheres Adams and Kattawar found errors up to 11.7%. Similar large errors are seen in Table 43 of van de Hulst [12]. Furthermore, polarization of light contains more information which cannot be retrieved just from intensity, such as the aerosol size distribution and atmospheric composition. Since the linear polarization can be measured to an accuracy of $\sim 0.1\%$, the information related to the polarization can be readily observed.

Jiang et al. [9] have pointed out a new method for detecting tropospheric ozone through remote sensing of the atmosphere by measuring the transmitted and diffuse solar radiation. But the signal is relatively small. Mishchenko and Travis [7] showed the high accuracy of the retrieval of aerosol properties from single-wavelength multiple-viewing-angle polarimetry alone. Although the high-precision polarization measurements of the Earth's atmosphere are currently unavailable, we will discuss in a model study of the characteristics of polarization of scattered sunlight in order to investigate the possibilities of detecting tropospheric ozone from both ground and space based instruments.

2. Radiation transfer model with polarization

The possibility that the polarization of the diffuse light may be effectively used for determining the tropospheric ozone change was suggested by Jiang et al. [9]. In this paper, we will report model results for the Huggins bands. We have included aerosol scattering and absorption in the current model. The vertical profiles of temperature, number density and ozone concentration of the 1976 US Standard Atmosphere are used in the model. We define the tropopause based on the temperature inversion at 16 km. From this definition, the total column ozone density in the atmosphere is 347.22 DU, with 310.34 DU in the stratosphere and 36.88 DU in the troposphere. The aerosol vertical profile and the size distribution are taken from Demerjian et al. [13]. The maximum radius of this distribution and the complex refractive index of the particles are 0.07 μm and 1.5–0.1i, respectively. The Earth's surface albedo is set to 0.1 [14]. Details are found in Jiang et al. [9].

We make computations for the reflection and transmission of solar radiation in a plane–parallel model atmosphere. The atmosphere is vertically inhomogeneous and is divided into 21 layers from 0 to 80 km. Each layer is approximated as homogeneous. The monochromatic scattering properties of each homogeneous layer are determined by its optical thickness $d\tau$, single scattering albedo ω, and phase matrix $\mathbf{P_{ij}}(\alpha)$, where α is the scattering angle. The phase matrix $\mathbf{P_{ij}}(\alpha)$ is weighted by the optical depth fractions of Rayleigh and aerosol scattering. The single scattering albedo is about 0.1 in the lower stratosphere due to the absorption of ozone, but it is almost 1 in the troposphere due to high concentration of atmospheric molecules and aerosol. The other relevant radiation quantities in the model can be found in Jiang et al. [9].

3. Tropospheric and stratospheric ozone

Fig. 1 shows the polar plots of transmitted and reflected solar light polarization at solar zenith angles (SZA) from 3° to 82.3°. The maximum degree of polarization in the transmitted light is less than 70%, but it can reach 100% in the reflected light. The maximum values of polarization are reached around 90° azimuth angle. The azimuthal angle is zero at the 12 o'clock position. The azimuthal angle for incident sunlight is zero. The neutral points (Babinet and Brewster points) are apparent at azimuth angle 0° in the transmitted light.

In order to study the sensitivity of polarization to ozone change in the troposphere and stratosphere, we computed the differences in polarization between the reference atmosphere and the perturbed atmosphere when either stratospheric ozone or tropospheric ozone was decreased by 10 DU as shown in Figs. 2 and 3. By comparing Figs. 2 and 3, we can conclude that the tropospheric ozone decrease causes polarization changes that are almost ten times as large as those caused by the same change in stratospheric ozone in both the transmitted and reflected light. Figs. 2 and 3 not only show the different behavior of tropospheric ozone and stratospheric ozone; they also provide a wealth of information in the polarization change as a function of incident SZA, viewing angle and azimuth angle. When the column ozone is decreased, light propagating in the atmosphere will experience more scattering by atmospheric molecules and aerosols. Therefore, the polarization will be smoothed out due to multiple scattering in the atmosphere. This occurs preferentially in the troposphere, where the concentrations of both gas and aerosol are high. In contrast, light undergoes only single scattering in the stratosphere. By decreasing the tropospheric ozone, the polarization has

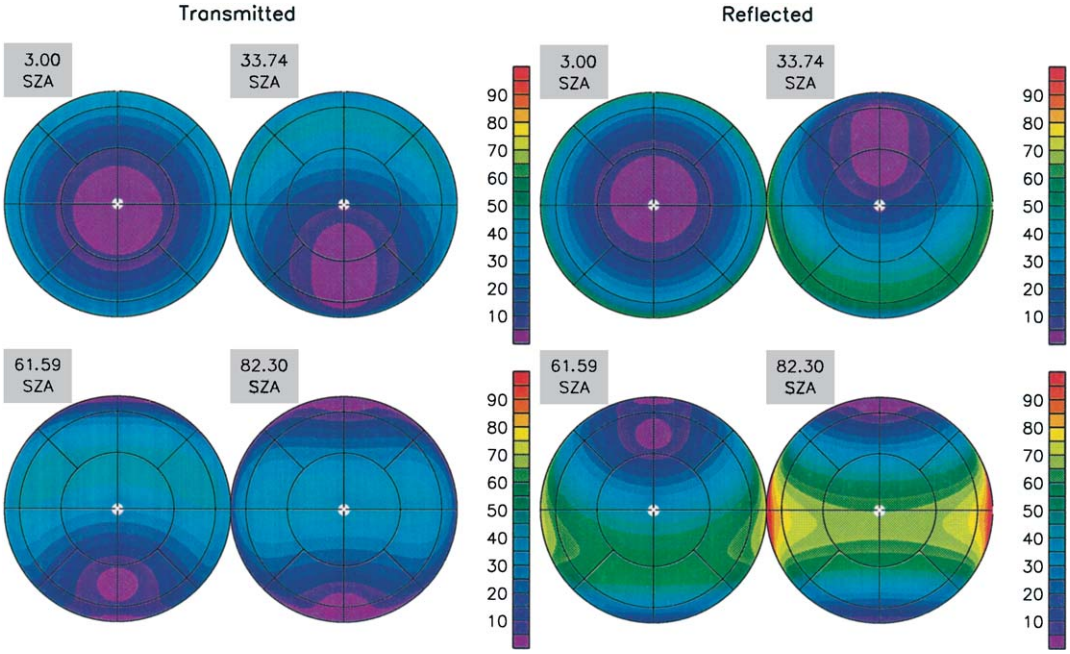

Fig. 1. Degree of polarization of the transmitted (left panel) and reflected (right panel) sunlight at wavelength 312 nm at solar zenith angles from 3.0° to 82.3°. The azimuth angle of the incident light is 0° (at the 12 o'clock position). The viewing angle (dashed circle) ranges from 0° at zenith to 90° at horizon.

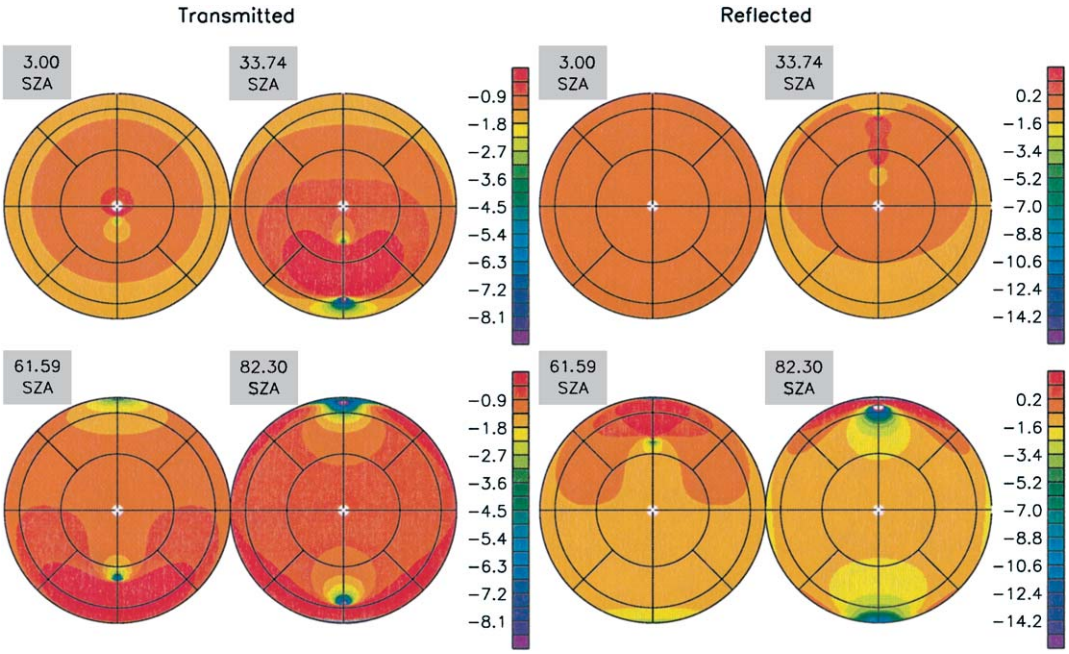

Fig. 2. Percentage change of the polarization of transmitted and reflected light as compared to the reference atmosphere when tropospheric column ozone was decreased by 10 DU.

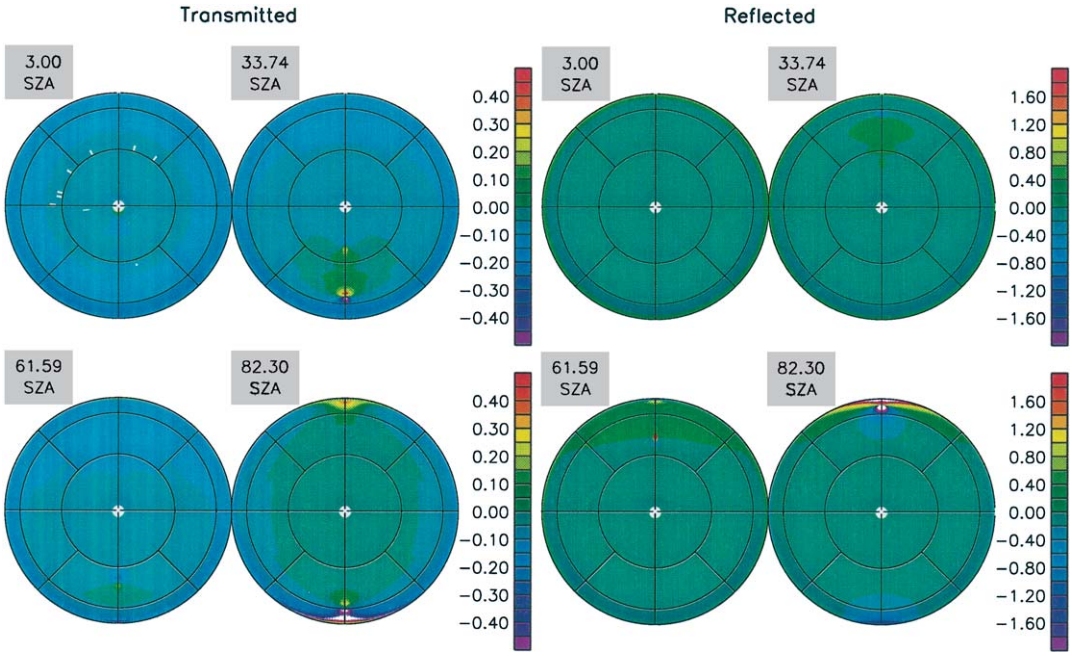

Fig. 3. Percentage change of the polarization of transmitted and reflected light as compared to the reference atmosphere when stratospheric column ozone was decreased by 10 DU.

decreased in most directions except in the direction of the sun, in both the transmitted and diffuse light.

To give a detailed view of the polarization percentage change due to both tropospheric and stratospheric ozone changes, we present in Fig. 4 the polarization change as a function of viewing angle at different azimuth angles when SZA is fixed as 41.1°. The neutral points (Babinet and Brewster points) can be seen around 30° and 60° at azimuth angle 0° (solid line) in the transmitted light. It should be pointed out that the change of polarization is positive between the neutral points while it is negative outside this region. The changes of polarization in the stratosphere are negligible. The changes of polarization in both reflection and transmission are comparable.

Fig. 5 shows the percentage change of polarization from a different perspective by fixing the viewing angle. In general, the difference shows the same shape between the tropospheric and stratospheric ozone change, but the amplitude is ten times larger when the change is in the troposphere. The difference in the transmitted light is as big as 2% at SZA 33.7° when the tropospheric ozone is decreased by 10 DU, while it is only 0.005% when the stratospheric ozone is decreased by 10 DU. This percentage difference is comparable in the reflected light, where the change is as larger as 0.5% at SZA 33.7° when the tropospheric ozone is decreased by 10 DU.

In order to show the different reactions of intensity and polarization to the decrease of ozone in the atmosphere and to compare the results with that shown by Jiang et al. [9], Fig. 6 gives the larger and opposite effects of column ozone on polarization changes than on the intensity changes. The reason is that the decrease of column ozone amplifies the effects of multiple scattering such that intensity increases but polarization decreases.

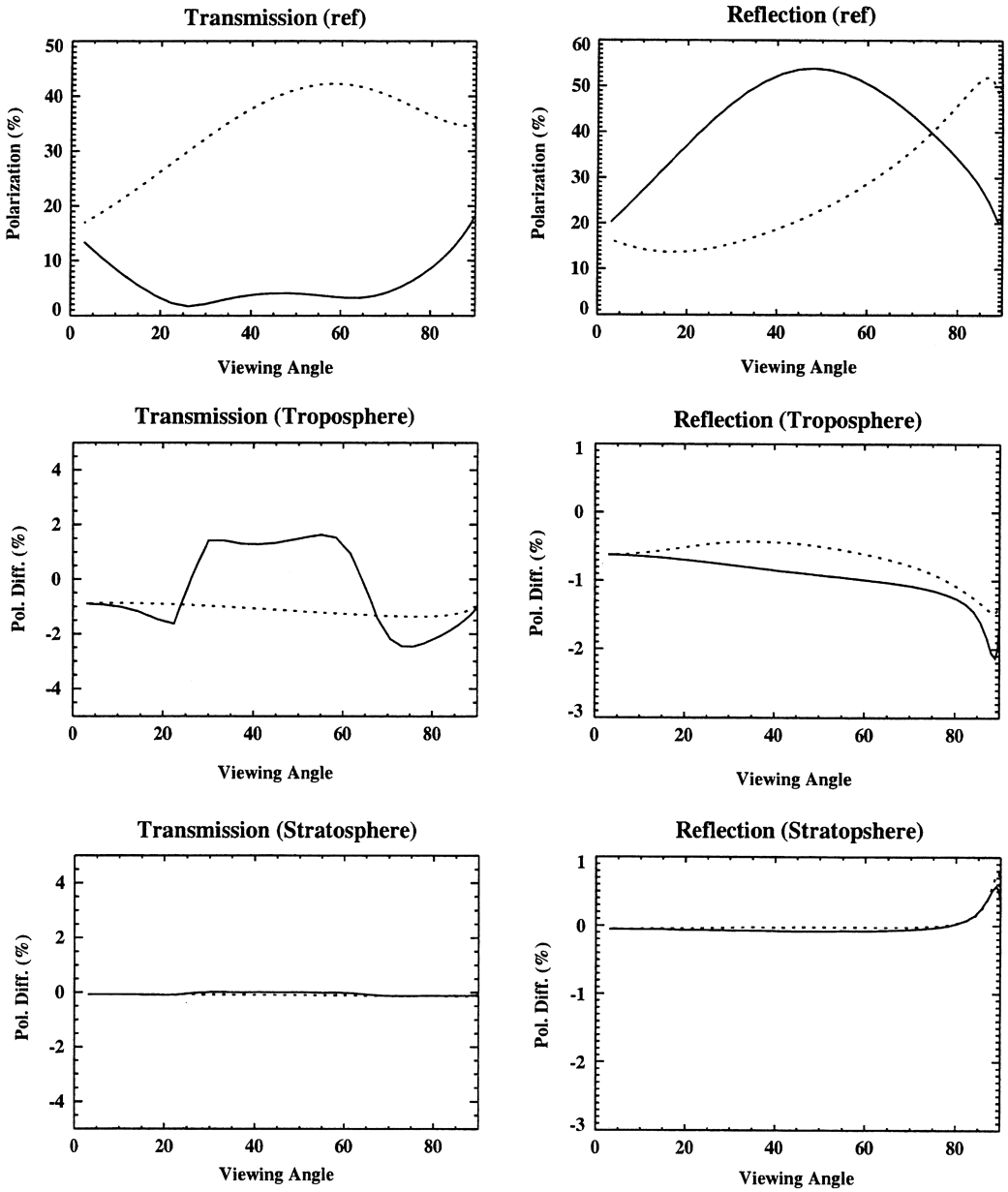

Fig. 4. Polarization change of transmitted and reflected light relative to the reference atmosphere when tropospheric column ozone was decreased by 10 DU vs. view angle at azimuth angles 10° (solid line) and 120° (dotted line). The SZA is 41.1°.

The top two plots of Fig. 7 show the transmitted and reflected linear polarization in the Huggins bands for the reference atmosphere, and for the perturbed atmosphere whose tropospheric column ozone has decreased by 10 DU. Features of the Huggins bands are clearly shown in both the

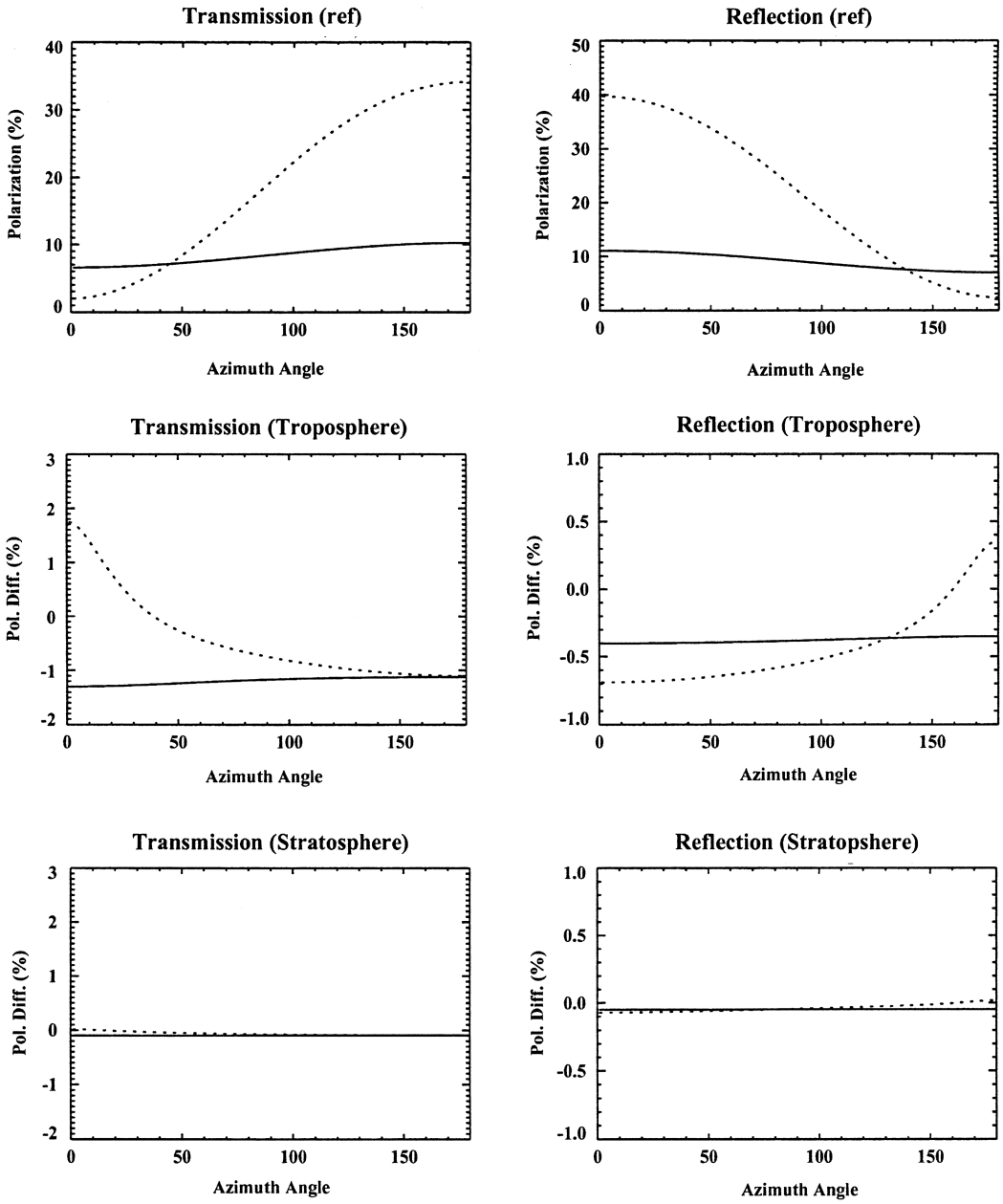

Fig. 5. Polarization change of transmitted and reflected light relative to the reference atmosphere when tropospheric column ozone was decreased by 10 DU vs. azimuth angle at SZAs 3.0° (solid line) and 33.7° (dotted line). The viewing angle is 30.0°. The upper two plots are for tropospheric ozone change and the bottom two plots are for stratospheric ozone change.

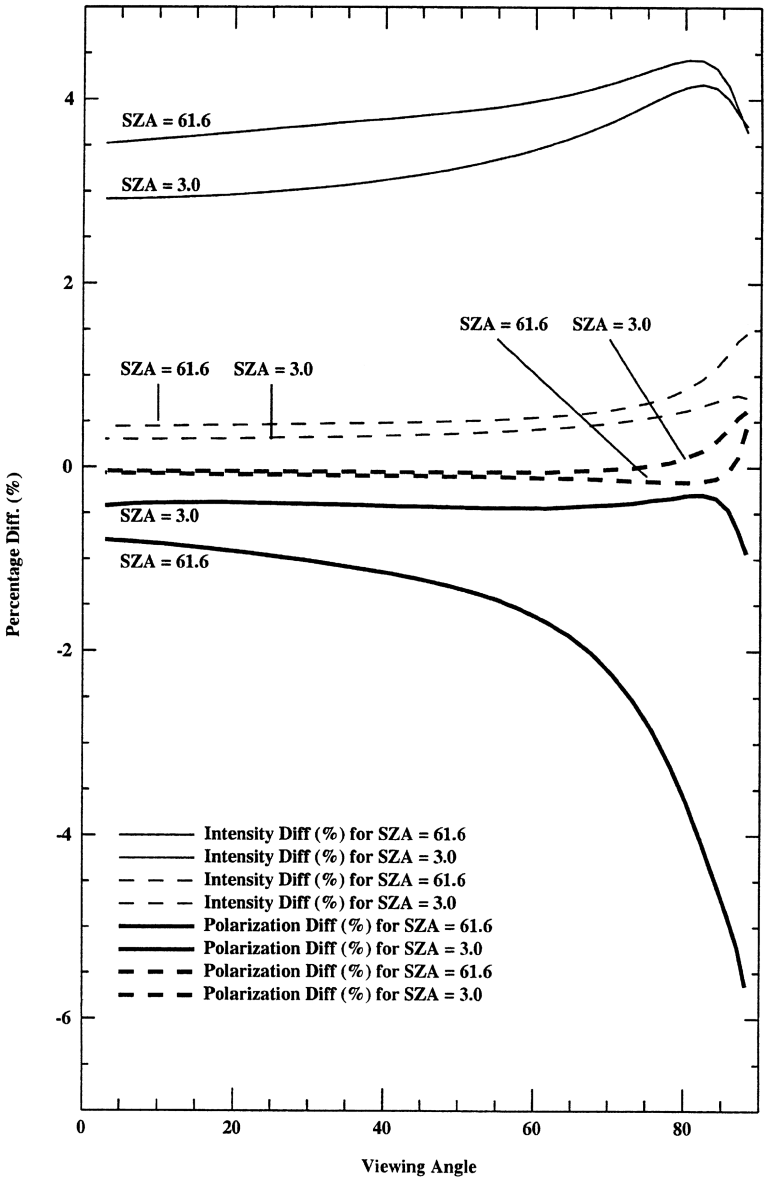

Fig. 6. Intensity and linear polarization changes of transmitted and reflected light relative to the reference atmosphere when tropospheric column ozone was decreased by 10 DU at. azimuth angle 0.0°, SZAs 3.0° and 61.6°. The thin lines represent the intensity changes and the thick lines represent the polarization changes.

transmitted and reflected spectrum. In the transmitted light, the linear polarization shows a minimum around 328 nm. The polarization of the reflected light decreases with wavelength. The polarization of the reflected light changes about 1.5% between 310 and 328 nm while it is around 3.5% for the reflected light.

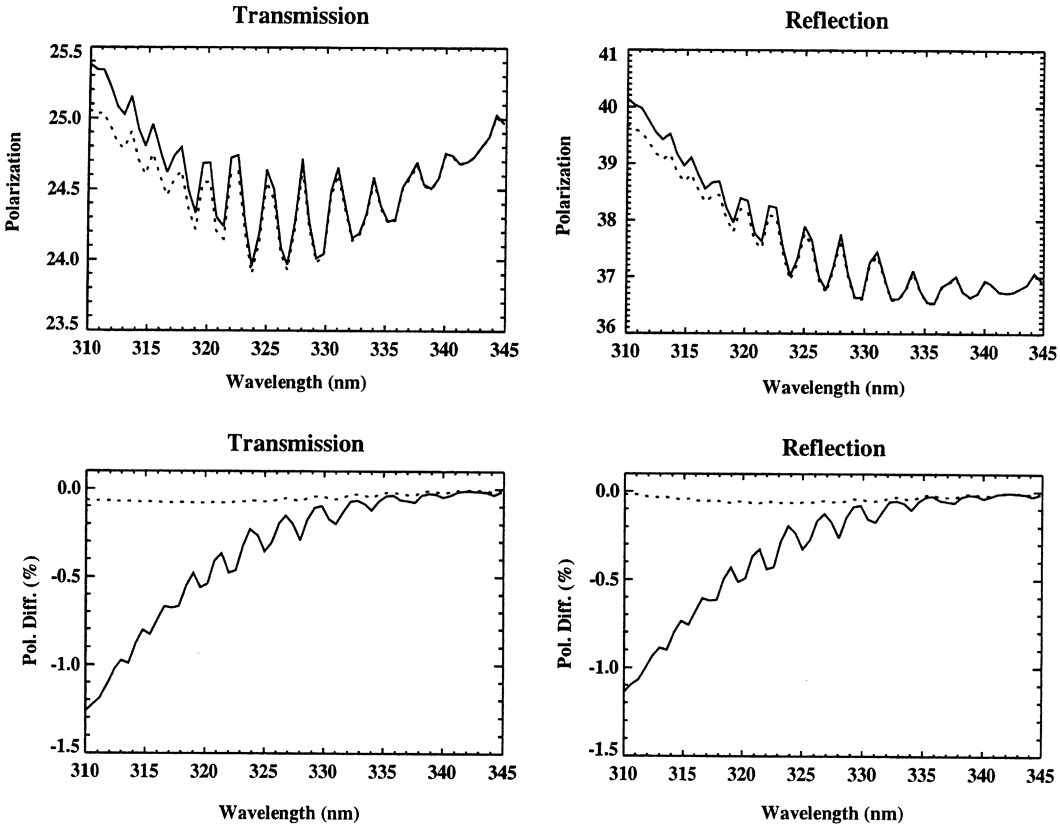

Fig. 7. The linear polarization of transmitted and reflected light for the reference (solid line) and perturbed (tropospheric column ozone was decreased by 10 DU) (dotted line) atmospheres in Huggins bands at azimuth angle 80°. The SZA and viewing angle are both 41.1°. The top two plots show the linear polarization of transmitted and reflected light, and the bottom two plots show the change of linear polarization relative to the reference atmosphere.

The bottom two plots of Fig. 7 show the transmitted and reflected linear polarization change relative to the reference atmosphere when tropospheric or stratospheric column ozone is decreased by 10 DU respectively. In both the transmitted and reflected light, the linear polarization change decreases as the wavelength increases in the Huggins bands. The tropospheric ozone decrease has a significant impact on the polarization as compared with that of the stratospheric ozone change. The change of linear polarization when stratospheric ozone is decreased by 10 DU is small and almost constant in the whole Huggins bands.

4. Aerosol effects on polarization

Fig. 8 is analogous to Fig. 7 but includes the computed degree of linear polarization when the aerosol concentration in the atmosphere was decreased by 10%. It shows the large effect of aerosol on the degree of linear polarization as compared to the effect of tropospheric ozone. The bottom two

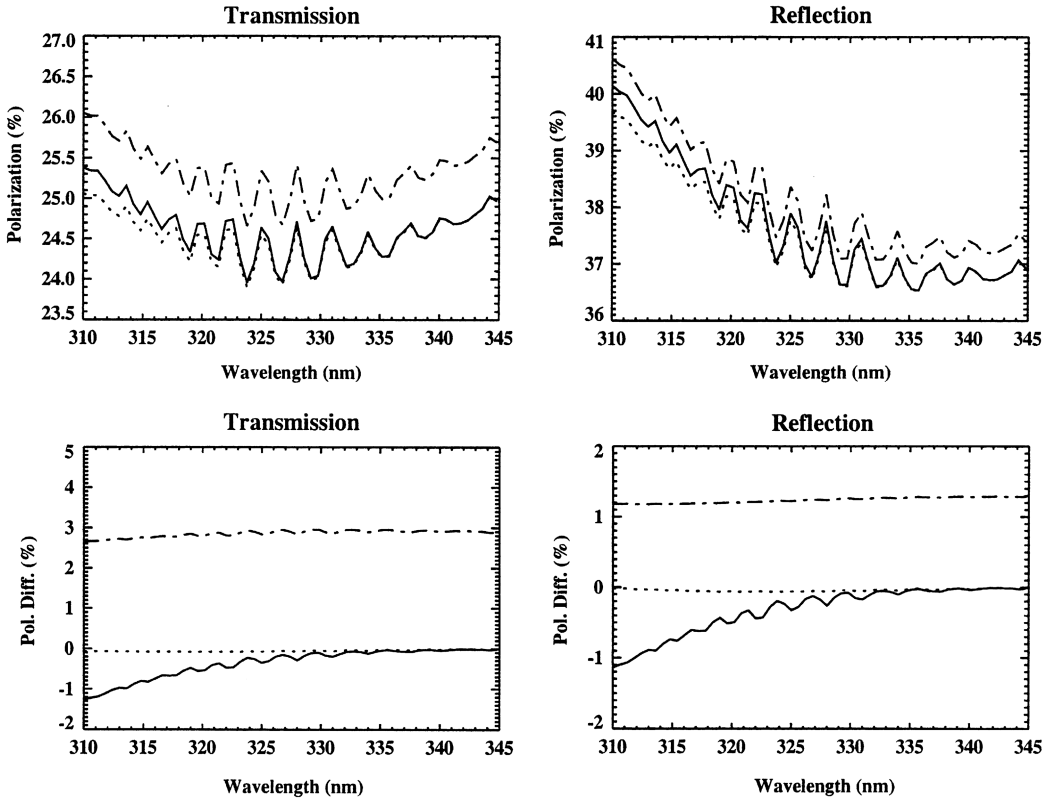

Fig. 8. The linear polarization of transmitted and reflected light relative to the reference atmosphere (solid line) when tropospheric (dotted line) was decreased by 10 DU or aerosol concentration (dash–dotted line) was decreased by 10% at azimuth angle 80°. The SZA and viewing angle are both 41.1°. The top two plots show the linear polarization of transmitted and reflected light, and the bottom two plots show the change of linear polarization relative to the reference atmosphere.

plots of Fig. 8 show the linear polarization differences as the function of wavelength in the Huggins band. The changes of aerosol and tropospheric ozone have opposite effects on the linear polarization relative to the reference atmosphere. Decreasing aerosol concentration caused much larger increasing linear polarization while tropospheric ozone decreasing causes the linear polarization to decrease. The linear polarization change seems to have a weak dependence on the wavelength when aerosol changes in the Huggins band. In the Huggins band range, the linear polarization changes only 10% from 310 to 345 nm when aerosol concentration was decreased by 10% while it changes 100% when tropospheric ozone is decreased by 10 DU.

5. Discussion and conclusions

In this paper we have described and developed a multi-layer radiative transfer model with the state of polarization fully taken into account by using the doubling-adding method. The model

is designed for passive atmospheric remote sensing applications using reflection or transmission measurements. Modeling of the Huggins bands show the distinct features of linear polarization in the transmission and reflection spectrum due to the multiple Rayleigh scattering in the atmosphere, mainly in the troposphere. The change of the polarization when the tropospheric ozone is perturbed is ten times larger than that when the stratospheric ozone is perturbed by the same amount. A model study (not shown here) further shows that the different behavior in the upper and lower tropospheric ozone, which provides us with background for the retrieval of tropospheric ozone vertical distribution profile. In the last part, we also studied the effects of aerosol on the polarization change, which in general is wavelength independent and can be removed as an averaged background value. Mishchenko and Travis' [7] retrieval of aerosol in the infrared may be combined with this study to retrieve tropospheric ozone.

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