# High-spectral resolution simulation of polarization of skylight: Sensitivity to aerosol vertical profile

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[1]   A vector radiative transfer model was used in conjunction with the line-by-line radiative transfer model and the database of high-resolution transmission (HITRAN) molecular absorption to simulate the degree of linear polarization of skylight in cloud-free conditions. Differences between simulated and measured polarization data in high-spectral resolution are found to be within 1% after aerosol scattering and gas absorptions are carefully considered. Limiting experiments are conducted at wavelengths around $0.760-0.765$ $\mu$m $O_2$-A absorption band for the same columnar aerosol optical thickness but different aerosol profiles. Results showed that the degree of linear polarization of skylight at surface varies strongly and is sensitive to the vertical change of tropospheric aerosol mass (or extinction) as the wavelengths approach to the edge of $O_2$-A absorption band. However, such sensitivity is minimal at all wavelengths when the aerosol composition or single scattering properties are vertically homogeneous. This study suggests that the polarization data can be used together with radiance data to constrain the simulation of vertical distribution of aerosol composition in chemistry transport models. **Citation:** Zeng, J., Q. Han, and J. Wang (2008), High-spectral resolution simulation of polarization of skylight: Sensitivity to aerosol vertical profile, *Geophys. Res. Lett.*, *35*, L20801, doi:10.1029/2008GL035645.

## 1.   Introduction

[2]   Atmospheric aerosols affect the climate directly by absorbing and scattering radiation and indirectly by serving as cloud condensation nuclei and changing cloud microphysical properties. The *Intergovernmental Panel on Climate Change* [2007] reported that the aerosol effect is one of the largest uncertainty sources in climate prediction, partially because the aerosol radiative properties are highly variable in space and time.

[3]   Satellite remote sensing is the only observation-based approach to characterize the *global* distribution of aerosols. To date, aerosol optical thickness (AOT) data are retrieved routinely with unprecedented accuracy from radiance data collected by MODIS and MISR on polar- orbiting satellites [*Kaufman et al.*, 2002; *Kahn et al.*, 2004]. Attempts were also made to retrieve regional AOT with high-temporal resolution from geostationary satellites, although the improvement of such retrievals is hampered by the lack of accurate calibration of sensors [*Wang et al.*, 2003]. Recently, the vertical distributions of aerosols are monitored globally from the active remote sensing sensors such as CALIPSO, but the data availability is limited by the narrow ground track of these sensors [*Winker et al.*, 2007]. *Yu et al.* [2006] reviewed the last decade's progress by the satellite observations in the estimate of aerosol forcing and concluded that further improvement requires new techniques to retrieve with higher accuracy more aerosol properties from both ground and satellite remote sensing data.

[4]   The motivation of this study is to explore the potential of using high-spectral resolution polarization data to retrieve aerosol vertical profile. Early studies on the remote sensing of atmosphere in Venus [*Hansen and Travis*, 1974] and recent investigations [*Mishchenko and Travis*, 1997] showed that aerosol scattering signature in the radiative transfer process cannot be fully characterized by the radiance (or intensity **I**) alone and rich information is contained in other three Stokes parameters **Q**, **U** and **V**. Hence, measurements of **I**, **Q**, **U**, and **V** or their derivatives provide more constraints for remote sensing of aerosols. Using **I** and degree of linear polarization (DOLP = $(\mathbf{Q}^2 + \mathbf{U}^2)^{0.5}/\mathbf{I}$ or simplified form $-\mathbf{Q}/\mathbf{I}$ in case of using scattering plane as the reference plane for singly scattered light) data from the space-borne POLDER instrument, *Deuzé et al.* [2001] retrieved AOT, Ångström exponent and refractive indices of aerosols. Similar set of aerosol properties was also retrieved by *Chowdhary et al.* [2002] from the polarization data collected by sub-orbital instrument. Other efforts in using DOLP to derive aerosol optical properties included retrieval of aerosol phase function [*Vermeulen et al.*, 2000] and aerosol effective radius [*Mishchenko and Travis*, 1997]. All aforementioned DOLP data however were collected only at several atmospheric window channels such as 0.47 $\mu$m and 0.87 $\mu$m.

[5]   The high-spectral resolution measurement of DOLP was shown to be valuable for retrieving wavelength-dependent optical properties of atmospheric aerosol and gas constituents [*Aben et al.*,1999]. *Aben et al.* [1999] further found from their model simulation a significant difference of DOLP of skylight (near the surface) at wavelengths close to the $O_2$-A band between two cases that have the same total AOT but distributed differently in the troposphere and stratosphere. They attributed the difference to two factors: (a) within the $O_2$-A band, multiple scattering is suppressed by $O_2$ absorption and therefore the polarization is dominated by the single scattering of stratospheric aerosols and mol-

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**Table 1.** Optical Properties of Sulfate and Dust-Like Aerosols[a]

|         | $r_{eff}$ ($\mu$m) | $v_{eff}$ | $m_r$ | $m_i$ |
|---------|--------------------|-----------|-------|-------|
| Sulfate | 0.5                | 0.45      | 1.428 | 2.05E-8 |
| Dust    | 1.0                | 0.45      | 1.530 | 8.0E-3 |

[a]$r_{eff}$, $v_{eff}$, $m_r$, and $m_i$ respectively denote effective radius, effective variance, real and imaginary part of refractive indices (at 0.70 $\mu$m) of aerosols. Wavelength-dependence of $m_r$, and $m_i$ used for high-spectral simulations follows the data compiled by *d'Almeida et al.* [1991].

ecules; (b) outside the $O_2$-A band, the polarization is dominated by the multiple scattering of tropospheric aerosols. Hence, the variation of DOLP at $0.755-0.775$ $\mu$m contains information of aerosol vertical profile.

[6] Inspired by *Aben et al.* [1999], this paper is to further investigate two important yet unclear issues. (1) Can current model simulate the polarization data with high-spectral resolution? The simulation shown in Figure 2 of *Aben et al.* [1999], because of low-spectral resolution, missed the fine structure in the spectral variation of their observed polarization data; (2) *Aben et al.* [1999] used in the simulation the dust-like scattering properties for tropospheric aerosols and the sulfate scattering properties for stratospheric aerosols; thus the sensitivity of DOLP near the surface remains unclear to the change of *tropspheric* aerosol profiles including vertical variation of aerosol mass, extinction, and aerosol single scattering properties (e.g., single scattering albedo and phase matrix). We address these two issues by first validating a base-case simulation of polarization from a high-spectral vector radiative transfer model (HSV-RTM) in section 2, and then by using this model to conduct sensitivity studies in section 3. The results are summarized and discussed in section 4.

## 2. Data, HSV-RTM, and Base-Case Simulation

[7] We use the skylight zenith DOLP data collected from the ground-based Global Ozone Monitoring Experiment Bread Board Model by *Aben et al.* [1999]. The data were measured at a coastal site in the Netherlands (52.1°N, 5.2°E) on the morning of April, 7, 1997, covering $0.305-0.794$ $\mu$m spectrum with an interval of 0.33 nm and a uncertainty of ~1% [*Aben et al.*, 1999].

[8] The HSV-RTM is built upon a vector RTM by *Evans and Stephens* [1991], and uses the Line by Line radiative transfer model (LBLRTM) and database of high resolution transmission (HITRAN) [*Clough et al.*, 2005] to compute the gas absorption and Rayleigh scattering. The single scattering properties of aerosols are calculated through Mie theory assuming that aerosols are spherical.

[9] Following *Aben et al.* [1999], we use in the HSV-RTM (a) the standard mid-latitude summer atmospheric profile; (b) AOT of 0.08 at 0.5 $\mu$m; (c) solar zenith angle of 58°; (d) the Lambertian surface spectral reflectance of grassland (http://speclib.jpl.nasa.gov/). Aerosols are homogenously distributed in bottom 4 km because the observed low AOT implies a profile of aerosol mixture in lower troposphere from local sources. Since the aerosol composition data were not available to this study, *Zeng* [2006] conducted simulations for various set of aerosol properties [*d'Almeida et al.*, 1991], from which aerosol optical prop-

erties of sulfate with consideration of its high hygroscopicity (Table 1) at the coast are found to give best match with the observed DOLP and hence are used in our base-case simulation. Sensitivity simulation using dust-like aerosol properties (Table 1) is described in section 3.

[10] Figure 1 shows the comparison between the HSV-RTM simulated and measured DOLP. Also shown is the simulated DOLP using database of low resolution transmission (LOWTRAN) [*Clough et al.*, 2005] to illustrate the importance of using LBLRTM and HITRAN. The simulation with HITRAN is in good agreement with the observation in capturing the major absorption lines in the atmosphere, especially in the $O_2$-A band. However, our calculated DOLP at *some* of $O_2$ absorption lines are larger than the observed, because the spectral resolution (0.2 nm) in our calculations is finer than that of measurement (~0.33 nm) to describe the peaks of $O_2$ absorption intensity. In contrast, the LOWTRAN simulation, because of its limited number of spectral wavelengths, misses the fine structure of DOLP in $O_2$-A band and significantly overestimates the DOLP transition from 0.75 to 0.76 $\mu$m. Quantitatively, the Chi-square ($\chi2$) of 978 observation-simulated pairs of DOLP data is 0.79, much lower than 2 that is considered as an acceptable fit by *Chowdhary et al.* [2002]. The absolute difference between HSV-RTM simulated and measured DOLP data is generally within the measurement uncertainty of 1%. We emphasized that the high-spectral simulation of DOLP is challenged by the lack of spectrally-matched measurements of surface and aerosol optical properties including particle shape. Nevertheless, Figure 1 can be taken as base-case results in the following
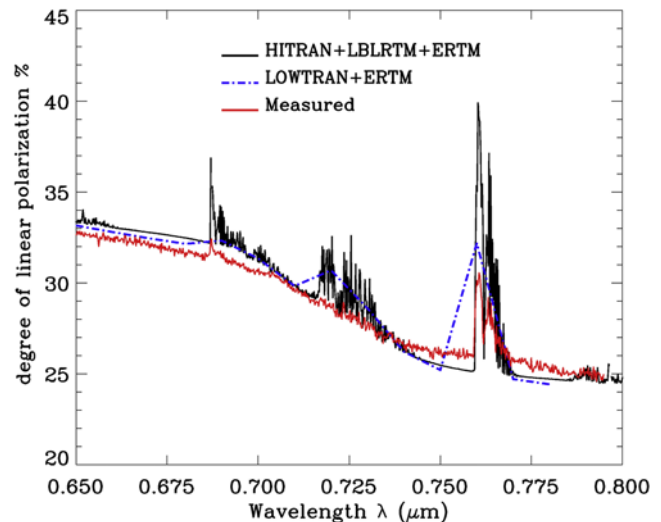


**Figure 1.** Comparison of the high-spectral resolution skylight zenith DOLP from the measurements (red line) and the simulations with gas absorption based upon LOWTRAN (blue line) and HITRAN (black line) respectively. Aerosols are assumed to be sulfate with size information and refractive indices listed in Table 1. ERTM denotes the vector radiative transfer model by *Evans and Stephens* [1991].
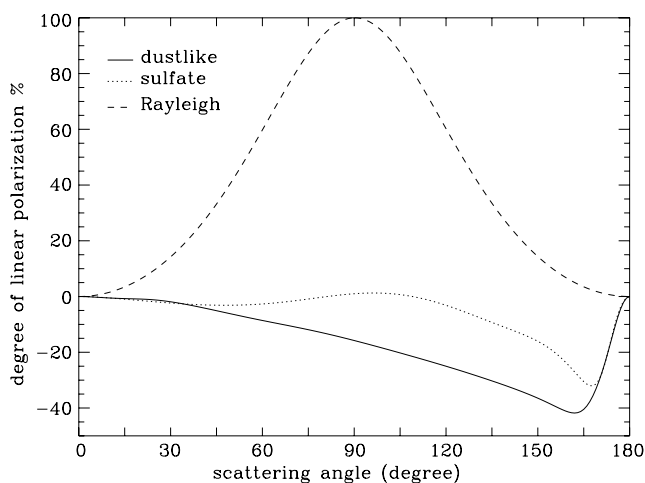
**Figure 2.** Degree of linear polarization (DOLP) as a function of scattering angle for light singly scattered respectively by dust-like aerosols (solid line), sulfate aerosols (dotted line), and molecular (Rayleigh scattering, dashed line). DOLP is calculated as $-P_{12}/P_{11}$ where $P_{11}$ and $P_{12}$ follow the traditional denotation of $4 \times 4$ scattering phase matrix [*Hansen and Travis*, 1974].

use of HSV-RTM to study the sensitivity of DOLP to the tropospheric aerosol vertical profile.

## 3. Sensitivity Experiments

[11] We first study the sensitivity of skylight zenith DOLP to vertical variations in the aerosol mass. We adopt for this study the same base case simulation as in section 2 except for (a) using dust-like aerosols, and (b) varying the vertical distribution of AOT or extinction as follows. In Experiment-1 we specify 70% of total dust AOT in $0-2$ km (hereafter layer-1) and 30% dust AOT in $2-4$ km (hereafter layer-2). In Experiment-2, we distribute the AOT equally among the two layers. Both experiments were conducted at $0.755-0.775$ $\mu$m (similar to *Aben et al.* [1999]).

[12] Dust-like aerosols, because of larger particle size and real-part of refractive indices (Table 1), generally result in more negative DOLP than sulfate aerosols in the single-scattered light (Figure 2). Consequently, the simulated DOLP values at surface in both experiments (Figure 3) are generally smaller than their counterparts in base case (Figure 1). However, within the $O_2$-A band (centered at 0.761 and 0.764 $\mu$m), the absorption of $O_2$ dominates the Rayleigh and aerosol scattering, diminishing the effect of aerosol multiple scattering on DOLP and leading to more stronger positive polarization. As a result, the DOLP values within $O_2$-A band in Experiment-1 and Experiment-2 are nearly the same at all altitudes as their counterparts in base case (Figure 1). In contrast, when the wavelengths depart from the center of $O_2$-A band, DOLP values at the bottom two altitudes in both Experiments start to exhibit differences because of weaker absorption of $O_2$ and larger effect of aerosols on polarization. The differences increase as the wavelengths depart further from $O_2$-A band and become virtually constant at the wavelengths 0.01 $\mu$m away from the $O_2$-A band. Such fine structure of spectral DOLP was

not reported previously and can only be examined via the high-spectral resolution simulation.

[13] Figure 3 also shows two interesting features regarding the effect of aerosol vertical profile on DOLP: (i) between $0.755-0.775$ $\mu$m, the DOLPs *at surface* in both experiments are the same; (ii) the DOLP at 2 km is smaller in Experiment-2 than in Experiment-1 because larger AOT in layer-2 results in smaller DOLP. Figure 3 therefore suggests that DOLP of skylight near the surface is not sensitive to the change of aerosol mass vertical distribution provided that the aerosol composition (or single scattering properties) and total optical depth are the same.

[14] Since the aerosol scattering properties or compositions are vertically inhomogeneous in the real atmosphere, we conducted another two experiments to test the sensitivity of DOLP near the surface to such vertical inhomogeneity (Figure 4). In Experiment-3, we specify all aerosols as dust, and calculate DOLP as a function of AOT ratio between layer-1 and layer 2. The Experiment-4 is similar as Experiment-3 except aerosols in layer-2 is specified as sulfate. We found that DOLP values in these two experiments are visually same within the $O_2$-A band, and hence only results at 0.755 $\mu$m are analyzed.

[15] Features consistent with Figure 3 are found in Figure 4 for Experiment-3 that DOLP at surface doesn't vary with the change of AOT distributions between layer-1 and layer-2. However, contrasting results are revealed in
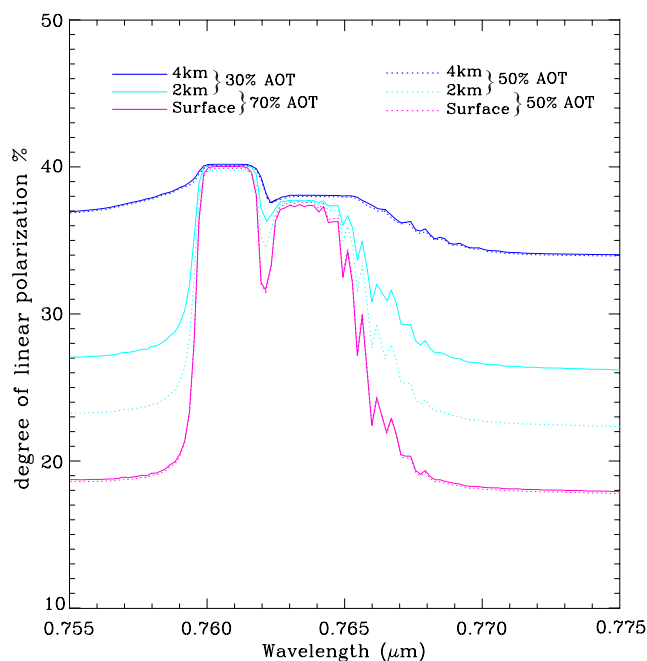


**Figure 3.** Skylight zenith DOLP near $O_2$-A band at the surface (pink lines), 2 km above the surface (green), and 4 km above surface (blue). Results at solar zenith angle of 58° are shown for two cases that assume the same (dust-like) aerosol properties for total AOT = 0.08 but with different vertical distributions: (1) 70% and 30% of total AOT distributed respectively in surface$-2$ km and $2-4$ km (solid lines); (2) same (50% of total) AOT in surface$-2$ km and $2-4$ km (dotted lines). Noted that the dot lines and solid lines are indistinguishable at surface and 4 km.
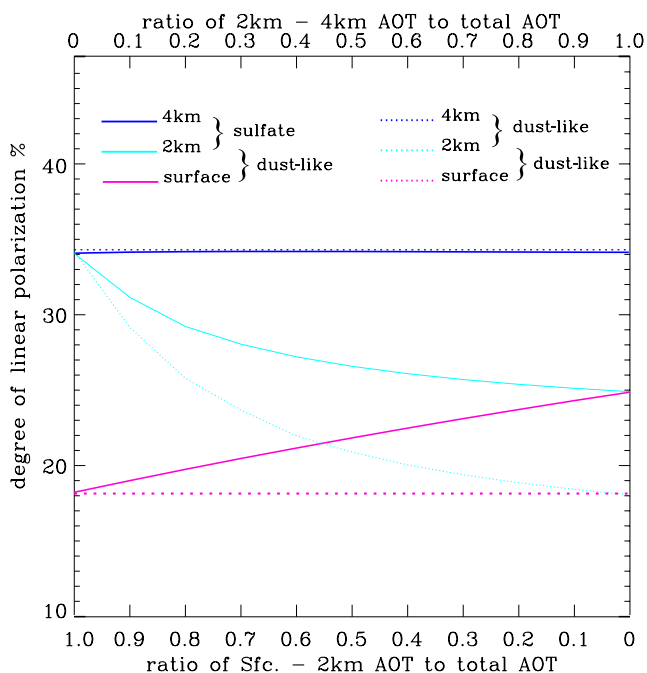
**Figure 4.** Skylight zenith DOLP at $0.775$ $\mu$m at different altitudes (surface, pink lines; 2 km, green lines; and 4 km, blue lines) as a function of the ratio of AOT in surface$-$2 km (and 2$-$4 km) to total AOT of 0.2. Results at solar zenith angle of $58°$ are shown for two cases with different vertical distribution of aerosol optical properties. The first case (solid lines) assumes dust-like aerosols in both surface$-$2 km and 2$-$4 km, while the second case (dot lines) assumes dust-like aerosols in 0$-$2 km and sulfate aerosols in 2$-$4 km.

Experiment-4 that DOLP at 2 km is enhanced (relative to that in Experiment-3) and the enhancement approaches greater with more AOT distributed in layer-2. Consequently, the simulated DOLP near the surface is larger in Experiment-4 (dust and sulfate) than in Experiment-3 (all dust), and positively increases as the contribution to total AOT by sulfate (in layer-2) increases. Such accretion of DOLP with the increment of sulfate AOT can be understood by comparing aerosol single scattering properties that dust-like aerosols produce more negative polarization than sulfate, and thus decrease more positive polarization induced from Rayleigh scattering (Figure 2).

## 4. Discussion and Conclusion

[16] In this study, we presented a high-spectral vector radiative transfer model that is capable to simulate DOLP with high spectral resolution. The comparison with limited but high-quality DOLP observation demonstrated that the model can accurately simulate the atmospheric polarization caused by aerosols and gases. It captures within 1% the polarization details in the gas absorption lines. The possibility of using DOLP around $O_2$-A band to infer the tropospheric aerosol vertical profiles is studied through several limiting sensitivity experiments. The results indicated that the skylight zenith DOLP varies strongly with wavelength as one approaches the $O_2$-A band, and its sensitivity

to the vertical profile of aerosols depends on the aerosol optical thickness and composition. In an ideal environment where aerosols have the same single scattering properties, the DOLP at the surface is insensitive to the shape of vertical profile of aerosol mass or extinction even for absorbing aerosols, and therefore have the potential for simultaneous retrieval of aerosol optical thickness and absorbing properties. However, in real atmosphere where aerosol composition most often is vertically inhomogeneous, the skylight zenith DOLP is amenable to the vertical change of aerosol mass or extinction.

[17] The implication of this study is twofold. First, given the importance of the inclusion of polarization for the accurate computation of radiance [*Lacis et al.*, 1998], our model simulations here can be used for channel selection in the sensor design or be combined with real skylight zenith DOLP measurements around $O_2$-A band to provide complimentary information on aerosol vertical structure for retrievals of aerosols from space-borne polarimeters such as the Aerosol Polarimeter sensor (APS) onboard the 2009 NASA/Glory mission [*Mishchenko et al.*, 2007]. Secondly, this study also implies the value of assimilating polarization data to the chemistry transport model (CTM) to constrain the simulation of aerosol composition in the vertical dimension. In current assimilation of satellite-derived aerosol optical thickness into CTM, the optical thickness of different aerosol species in all vertical layers above the same surface grid is usually updated by scaling with a *single* factor, i.e., the ratio between satellite-derived and model-calculated total aerosol optical thickness of that grid [*Wang et al.*, 2004]. With the strongly varying sensitivity of skylight zenith DOLP around the $O_2$-A band to aerosol vertical profile and the available information of aerosol composition in CTM, one can assimilate data obtained from such polarization to infer the scaling factors for aerosol mass in different layers. Further research is needed to combine radiance (or aerosol optical thickness) data and polarization data from both active and passive remote sensing to constrain the CTM simulation of 3D distribution of aerosol mass and composition.

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