**Comments from Reviewer 1 in italicized font.**

**Author response to Reviewer 1 in regular font.**

**General Comment**

The authors have carried out a computationally detailed analysis of the sensitivity of space borne limb radiance measurements at ultraviolet, visible, and near IR wavelengths. The authors use the OSIRIS sensor as reference for spectral coverage and viewing geometry characteristics. The methodological approach is sound. The paper is very well written, and the results discussed in detail. The main deficiency of the paper, however, is the inaccurate model representation of several aerosol types in the analysis.

We thank the reviewer for a careful review. We have addressed the concerns about choice of aerosol types as described in detail below.

**Aerosol Model Assumptions**

Desert dust aerosols are known to be non-spherical particles. Although the scattering phase function of non-spherical particles resembles that of spherical particles over a narrow range of scattering angles, (~150°), there are significant differences at the range of scattering angles (60°-120°) of the OSIRIS observations. The authors' treatment of desert dust particles as spherical elements is not justified given the range of scattering angles considered in the analysis. The same criticism applies to the modelling of ice particles which are most certainly not spherical.

Regarding ice particles, we only describe what would be considered very small ice particles (D ~1 µm) of exactly the same size as our dust particles, hence the need for non-spherical phase function calculations is not as critical as in the case of more typical ice crystals, i.e., D ~100 µm, when non-sphericity is crucial to the particles' radiative effects.

We agree that dust non-sphericity effects are important at scattering angles >~90°, except at ~150°, and include Figure 3 from Dubovik et al. [2006] for ease of reference regarding the angular dependence of the scattering phase function. We note, however, the decreasing importance of non-sphericity for high multiple-scattering scenarios and increasing particle absorption in the near-UV [e.g., Torres et al., 1998, Section 2.4].

While the full range of scattering angles observed by OSIRIS is indeed 60°-120°, as noted in the manuscript, about half of spring (MAM) and summer (JJA) observations occur at solar scattering angle (SSA) < 90°. More than two thirds of fall (SON) and winter (DJF) observations occur at SSA < 90°.

To assess the effect of non-spherical scattering phase functions across all angles of scattering encountered in our limb-view geometry we have obtained spheroid mixture scattering phase functions corresponding exactly to our particles' size distribution and refractive index as a function of wavelength, but corresponding to a reasonable distribution of spheroid shapes, which resulted in the flattened
scattering phase function shape for angles greater than 90°, similar to the above figure. The simulations in our study correspond to SSA ~68°, at which angle the non-spherical scattering phase functions are very similar (within 8% - 17% in our case) to the spherical scattering phase functions. As such, non-spherical dust has only a small modifying effect on the single-scattered radiance and does not alter the Rayleigh shielding effect discussed in our paper. The multiple-scattered radiance involves all angles of scattering and our new non-spherical simulations show that multiple scattering increases within the layer (by ~15%) but also above the layer (by ~5%). This also leads to a net (total radiance) scene brightening at all heights in the atmosphere, as in the original manuscript. We have noted the results of non-sphericity in Sections 2.1 and 3.2.

The assumed values of single scattering albedo are extremely low. A single scattering albedo value of 0.78 at 452 nm is significantly lower than any measurement at an equivalent wavelength reported by AERONET at any of the sites over arid and semi-arid regions. The Muller et al. [2009] values of imaginary refractive index, on which the authors have based their SSA calculations, may not be representative of the actual column atmosphere which is what is actually needed in the interpretation and analysis of satellite observations. The authors may refer to the Müller et al [JGR, 2012] paper that shows large discrepancies in single aerosol absorption parameters derived by different techniques.

We agree with the reviewer that AERONET retrievals yield the Single Scatter Albedo (SSA) relevant to the overall column atmosphere, i.e., an average over more and less absorbing constituents. In our theoretical study we purposely chose to use index of refraction values representative of pure dust particles obtained by ground-based in-situ methods as opposed to remotely sensed column-average aerosol mixture retrievals in order to isolate the radiative transfer effect of dust alone. We used the complex index of refraction values that were slightly higher than – but fit well within the error bars – of those reported by Thomas et al. 2009 (their Table 2, pure dust mode data, corrected for the presence of soot). This choice was based on our interest in exploring the maximum possible effect of mineral dust absorption in limb-scatter spectra. Since our main conclusion is that absorption effects are small in this viewing geometry, increasing the SSA in our simulations via a lower complex refractive index and smaller particle size will not change it.

It is not easy to see all the points presented in the right side of Figure 3 of Detlef Müller et al. 2012, however, our near-UV complex index of refraction (~0.017 at ~357 nm if we average over our spectral points at 337 and 377 nm) falls in the range of the reported measurements on 3 out of 4 observation days, always closer to the values obtained by the in-situ SOAP (Spectral Optical Absorption Photometer) technique of T. Müller et al. 2009, as expected. D. Müller et al. 2012 discuss in detail the discrepancies in the complex index of refraction obtained by different techniques (AERONET sunphotometer, laboratory analysis by scanning electron microscopy, ground-based and air-craft-based in-situ analysis by inversion of spectral absorption coefficient measurements) and conclude that whether or not the imaginary part measured by one method can be verified with another method is an open question, with treatment of particle shape being only one factor to consider among several. At short wavelengths, AERONET values are systematically lower than those derived from mineralogical analyses of single particles, however, D. Müller et al. 2012 do not point to any fundamental problems with the analysis presented by T. Müller et al. 2009.

Nevertheless, our chosen complex index of refraction and particle size leads to lower than average single scatter albedo values, which we have noted in the manuscript (Section 2.1). That is, our SSA_{452nm} = 0.78 as compared to the SOAP campaign-average SSA_{450nm} = 0.88 and again our SSA_{377nm} = 0.65 as compared to the SOAP campaign-average SSA_{350nm} = 0.78.
The dust particle mode radius assumed in this analysis is much larger than AERONET’s reported values. The Wiacek et al (2010) reference in support of the assumed particle size distribution parameter does not provide any information on the subject. I suspect that the extremely low SSA values assumed in this analysis are a consequence of the large values of imaginary refractive index and mode radii used in the analysis.

Reference to Wiacek et al (2010) should have included “and references therein”, however, we have included in the manuscript the explicit reference to Tegen (2003, Quat Sci Rev), who states that “It has been shown that for far-traveled dust the size distribution can be represented by a single mode with a mean radius around 1–1.5 µm.” Table 1 of Tegen (2003) shows a number of dust modeling schemes with a variable size range. Additionally, Figure 5 of Tegen (2003) illustrates the size-dependence of the single scatter albedo at 550 nm for a particular index of refraction. Indeed, there is a pronounced drop in SSA for a particle radius increase from 0.5 µm to 1.0 µm.

In our lognormal particle size distribution, a mode radius of $r = 1$ µm combined with a mode width of $\sigma = 1.6$ µm yields an effective radius of $r_{eff} = r \times \exp \left( \frac{5+10\sigma^2}{2} \right) = 1.74$ µm. This value compares very well with a recent dust climatology based on IASI measurements [Peyridieu et al., 2013, ACP, http://www.atmos-chem-phys.net/13/6065/2013/acp-13-6065-2013.pdf], which in turn compares favourably to AERONET measurements. The AERONET effective radii presented in this paper generally fall around 1.6 µm in the peak of the African dust season in JJA.

Nevertheless, we agree that upper tropospheric particles are likely smaller than those used in our study, though measurements are sparse and aerodynamic sampling issues present challenges to interpretation. Therefore, we calculated limb-scatter radiances for particles with size distribution mode radii of 0.5 µm and found the short-wavelength results to be qualitatively similar: a phase-function driven reduction in single scattered radiance, an increase in multiple scatter radiance, and in this case a combination of total radiance increases above the layer height and total radiance decreases within and below the layer height, similar to what we described in Figure 6 pertaining to 1-km layers of dust and ice. An increase in total radiance at all heights in the atmosphere could be achieved if the particle number density was adjusted to maintain a constant optical thickness of the dust layer as compared to the 1 µm mode radius particles.

Carbonaceous aerosols contain both black carbon (BC) and organic carbon (OC). As shown by observations based on filter collected samples during SAFARI 2000 [Kirchstetter et al, 2004 JGR], and by OMI satellite observations [Jethva and Torres, 2011], OC in carbonaceous aerosols is responsible for the spectral dependence of aerosol absorption observed at wavelengths shorter than about 420 nm. The smoke aerosol model assumed in this paper assumes BC as the only absorbing component and, therefore, ignores OC absorption effects in the UV. AERONET retrieved absorption parameters are largely insensitive to OC because their inversion algorithm does not use radiance measurements at wavelengths shorter than 440 nm.

As discussed on pg. 8, lines 7-8, “we applied the 440nm values available from this AERONET inversion, noting that carbonaceous aerosols may be more absorbing than this.” Our complex index of refraction at 452 nm, 377 nm and 337 nm was thus 0.02677 (Table 2). The complex index of refraction reported by Dinar et al. [2008, Faraday Discuss.] at 390 nm is between 0.089 and 0.031 depending on the fraction of humic-like substances (HULIS) extracted from smoke aerosols in an internal mixture with 2% soot and the remainder consisting of ammonium sulfate (AS). Thus the index of refraction at our wavelength of
377 nm could be ~3X higher than the value that we used for an internal mixture with 70% HULIS, 28% AS and 2% soot. At 337 nm, the factor may even be higher, given the spectral dependence of light attenuation down to 300 nm observed by Kirchstetter et al. [2004]. We re-defined the complex index of refraction of our smoke particles at 337 and 377 nm to include a spectral dependence following both Dinar et al. [2008] and Jethva and Torres [2011]. This has increased the scene darkening introduced by smoke particles but not affected our overall conclusions. Figures 7 and 8 were affected (radiance ratios w.r.t. air only at 337 and 377 nm), as was Figure 11 (low vs. high SZA for D1, soot, smoke). The sensitivity to carbonaceous particles was also affected (Table 5, smoke values), bringing the scene reduction to -4.5% under extreme particle number concentrations, but still comparable (0.49%) to OSIRIS measurement precision (0.1%) for more normal particle number concentrations. Changes have been tracked in the manuscript, however, the overall conclusions about absorption effects have not been altered.

Regarding the optical properties of sulfate aerosols, the authors rely on the measurement of Palmer and Williams [1975] that are more relevant to stratospheric sulfate aerosol production at low temperatures in the aftermath of volcanic eruptions. Although I do not expect major differences, the authors are advised to consult the more recent work of Beyer et al [JGR, 1996] that performed measurements at different temperature conditions.

The results of Palmer and Williams [1975] were used for wavelengths longer than 360 nm, while the reported radiative transfer modeling parameters of Torres et al. [1995, JGR] were the basis for the near UV, which has now been noted correctly in the paper. As such, they agree well with values reported by Beyer et al. [1996], who also compare their inferred low-temperature (224 K) result of 1.48 ± 0.01 at 313 nm for a 75 wt % solution to the work of Bhartia et al. [1993] who use 1.47 at the same wavelength. We use 1.472 at 313, in excellent agreement with both Beyer et al. and Bhartia et al. Finally, the lower stratospheric temperatures are more appropriate to our study of the upper troposphere than the room temperature measurements of Beyer et al.

Recommendation

Because of the large uncertainty in the validity of the desert dust aerosol model, ice particles, and smoke model, any conclusion on the significance of the results of the modeled radiative transfer interaction is equally uncertain. The authors are encouraged to revisit the aerosol models and submit a revised version of their analysis.

We have explored the effects of dust particle non-sphericity and of the complex index of refraction and its spectral dependence for both dust and smoke. We have revised the manuscript to reflect these additional calculations, which strengthen our work but do not lead to changes to the main conclusions.

Other comments

Pg 1903

Line 12. Is SASKTRAN a vector code?

OSIRIS measures scalar radiance, which has now been indicated in the manuscript.
Line 24. Scattering and surface reflection are two entirely different processes.

The manuscript has been revised to describe reflection, not scattering, off a Lambertian surface.

Pg 1909

Line 10 This result is not surprising as it is well known that the sensitivity to refractive index is significantly lower at side-scattering angles than at nadir viewing configurations. That is the reason satellite-based aerosol optical depth retrievals are more accurate at off-nadir viewing condition that a near-nadir [Chylek et al., 2003 GRL].

Chylek et al. (2003) report more accurate AOD retrievals from the Multispectral Thermal Imager at smaller solar scattering angles, which are produced with the off-nadir view (60° back along track). The RMS error is reduced from 0.11 to 0.03 (65% to 20% of an average AOD) by switching from the near-nadir to the off-nadir view. This is attributed to 1) a longer aerosol path at smaller scattering angles in the off-nadir view, increasing aerosol signal relative to surface signal, 2) a decreased relative signal of instrumental effects like stray light, dark current and digitization step in the longer path length, and 3) smaller effect of uncertainty in the real and complex index of refraction (as well as particle shape) on the scattering phase function at the smaller scattering angles in the off-nadir view (Figure 5).

We added some text about this topic to the manuscript, "The small effect of absorption is consistent with Chylek et al. (2003) who demonstrate the small sensitivity of the scattering phase function to the complex refractive index at scattering angles < 120°."

Pg 1915

Line 4 Angular dependent scattering is fundamentally a single scattering property

We agree.