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Radiance and flux simulations for mineral dust aerosols: Assessing the error due to using spherical or spheroidal model particles

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[1] The use of simple model particles to represent mineral dust aerosols may introduce errors in radiative transfer calculation. A modeling study has been performed to assess these errors. At a wavelength of 550 nm it is found that spheres can cause errors in the diffuse spectral radiance between −18% and 26% at the bottom of the atmosphere (BOA) and between −16% and 115% at the top of the atmosphere (TOA). These errors correspond to an uncertainty in the extinction optical depth τ between 0.7τ and 1.5τ (BOA) and (disregarding extreme errors in the solar nadir direction) between 0.5τ and 2τ (TOA). When using spheroidal model particles, the spectral radiance errors are reduced by roughly a factor of 2 (BOA) and are almost zero at the TOA over a large angular range. For a latitude of 20° we find, as a conservative estimate, that spherical model particles overestimate the yearly averaged TOA spectral net flux by almost 10 W m^{-2} nm^{-1}. This error is mainly due to the misrepresentation of the aerosol phase function by the spheres. A rough estimate using Mie calculations indicates that this error is 5 times higher than (50% of) the error due to the uncertainty in the real (imaginary) part of the refractive index. Spheroids, on the other hand, only differ by 1.3 W m^{-2} nm^{-1} from the reference case. This indicates that using spherical model particles may cause significant positive errors in radiative forcing simulations for dust aerosols and that using simple spheroids can substantially improve the results. *INDEX TERMS:* 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0360 Atmospheric Composition and Structure: Transmission and scattering of radiation; 0669 Electromagnetics: Scattering and diffraction; 3359 Meteorology and Atmospheric Dynamics: Radiative processes; *KEYWORDS:* mineral dust, radiative forcing, remote sensing

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1. Introduction

[2] Mineral dust aerosols have received growing attention both in climate research and in air pollution monitoring. Developing a predictive capability for size- and composition-resolved dust particle concentrations in the atmosphere can be considered one of the major outstanding problems in quantifying the spatial and temporal variabilities of dust aerosols and in assessing their radiative impact [*Sokolik et al.*, 2001]. Remote sensing observations of mineral dust can contribute useful information for developing more reliable parameterizations of mineral dust concentrations that can be used in dynamic aerosol models. Optical observations can be useful for validating such models. Also, mineral dust aerosols can have a strong local effect on the radiative balance, particularly in arid regions [*Harshvardhan and Cess*, 1978; *Carlson and Benjamin*, 1980; *Haywood et al.*, 1999, 2001]. Anthropogenically induced changes in land vegetation and precipitation have contributed to an increase in arid areas, and to a corresponding increase in mineral dust aerosol loads in and near arid regions. In these areas radiative forcing due to mineral dust can be the largest of all anthropogenically caused forcing mechanisms [*Myhre and Stordal*, 2001].

[3] Our ability to interpret remote sensing data on mineral aerosols as well as our ability to predict radiative forcing due to mineral aerosols depend on accurate radiance and flux computations in radiative transfer simulations, and thus on a knowledge of the aerosols' single scattering optical properties and optical depth. Those, in turn, are determined by the physical and chemical properties of the aerosols. Both the mineral composition of aerosols and the internal or external mixing of mineral aerosols with other aerosols is often uncertain [*Sokolik and Toon*, 1999]. Also, uncertainties in the number concentration and the size distribution of aerosols contribute to the uncertainties in the single scattering optical properties and optical depth. Information on size-resolved mineralogical and chemical composition of dust particles over a wide size range are still not sufficient for developing reliable parameterizations of the radiative impact of aerosols in terms of their physical and chemical

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properties [*Sokolik et al.*, 2001]. Another source of uncertainty in radiative transfer simulations is the spatial distribution of aerosols in the atmosphere and its temporal variability. In a recent study, *Myhre and Stordal* [2001] simulated the optical properties of dust aerosols by using Mie theory, and they varied physical and chemical aerosol properties as well as their vertical distribution in the atmosphere. They concluded that uncertainties in the refractive index and size distribution make the dominant contribution to the uncertainties in radiative forcing due to mineral aerosols.

[4] In view of this large number of error sources, little attention is paid in flux simulations to possible errors due to simulating optical properties of nonspherical dust aerosols by using Mie theory. Shape effects in flux computations are usually believed to be small, although only few quantitative studies exist that actually quantify the significance of nonspherical particle shapes in flux simulations. *Mishchenko et al.* [1995] have compared simulations for spheres and spheroids and found that the single scattering albedo, the asymmetry parameter, and the local albedo of spheres and spheroids are rather close, thus indicating that spheres may be suitable for radiative flux simulations. However, the error in flux computations due to using spherical model particles still needs to be quantified by performing accurate radiative transfer simulations.

[5] Even for radiance simulations spherical model particles are still widely used, although it becomes more and more recognized that they perform poorly for this purpose. Several studies indicate that spheroids may be much better model particles [*Mishchenko et al.*, 1997; *Kahnert et al.*, 2002a, 2002b; *Nousiainen and Vermeulen*, 2003; *Kahnert*, 2004], and retrieval algorithms for remote sensing observations are developed that are based on using spheroids instead of spheres [*Dubovik et al.*, 2002]. Nevertheless, also spheroids have their limitations [*Veihelmann et al.*, 2003; *Kahnert*, 2004].

[6] In this paper the errors in spectral radiance and spectral flux simulations due to using spherical or spheroidal model particles will be quantified. This study builds on a recent comparison of the single scattering optical properties of dust-like particles, spheroids, and spheres [*Kahnert*, 2004].

2. Approach

[7] There are in principle two possible approaches for checking the suitability of simple model particles for simulating optical properties of complex-shaped particles. One can either use experimental data for the optical properties of complex-shaped particles, or one can try to compute them. Subsequently, one attempts to fit the measured or computed optical properties by using simple model particles. The former approach has the obvious advantage of using realistic results as a reference case. However, the disadvantage of this approach is that one often does not have the full information about all optical properties, thus introducing additional uncertainties. For instance, one may have results for the phase function of an ensemble of aerosols, but the refractive index and the single scattering albedo are not known exactly. Also, the physical properties of the particles are ofte accurately known. This makes it difficult to address the question if it is possible to retrieve size or shape information from fitting optical properties by using simple model particles. The latter approach, on the other hand, has the disadvantage that one cannot know for sure if the modeled reference case gives a good representation of realistic aerosol ensembles. However, this approach has the advantage that one has full control and thus complete information about all physical and optical properties of the reference case. This makes it possible to address the retrieval question, and to separately assess the contribution of different error sources. For instance, one can investigate how much of the error in radiance or flux simulations using simple model particles is due to a misrepresentation of the phase function by the model particles, and how much of the error is due to an error in the single scattering albedo.

[8] Clearly, the two approaches described above are supplementary to each other. Comparisons of measured phase functions with simulated phase functions of model particles have been conducted, e.g., by *Nousiainen and Vermeulen* [2003] and by *Kokhanovsky* [2003]. Comparisons of computed single scattering optical properties of particles with different degrees of shape complexity have been reported by *Lumme and Rahola* [1998], *Kahnert et al.* [2002a, 2002b], and *Kahnert* [2004].

[9] In this paper, we will use the modeling approach; that is, we will use as a reference case computed optical properties for complex-shaped particles. The focus will be on how errors in simulated single scattering optical properties affect computed radiances and fluxes in radiative transfer simulations. We use as a proxy for dust-like aerosols a size-shape mixture of non-axisymmetric, randomly oriented particles with low symmetries, consisting of 110 different sizes, aspect ratios, and geometries. The distribution of aspect ratios is assumed to be equi-probable. We used 11 quadrature points to represent the size distribution, 5 different aspect ratios, and 2 different geometries (bricks and pentahedral prisms). The maximum size parameter used in the computations was $kD = 52$, where k denotes the wave number in vacuo, and where D represents the size of the particle along its maximum extent.

[10] A lognormal size distribution was used with $\langle r \rangle$ = 0.7 μm and $\sigma = 1.9$ [*d'Almeida*, 1987], and for the refractive index we choose m = 1.65 + 0.005i [*Sokolik et al.*, 1993; *Ivlev and Popova*, 1973] at a wavelength of 550 nm. This is a typical size distribution for background concentrations of desert dust aerosols. We note that ultrafine dust particles contribute little to the optical depth, and coarse mode particles only contribute significantly during dust storm events.

3. Single Scattering Optical Properties

[11] We used our own T matrix code based on the null-field method [*Kahnert et al.*, 2001] for computing the single-scattering optical properties of the proxy of dust aerosols. The code exploits particle symmetries [*Schulz et al.*, 1999] and analytical averaging over particle orientations [*Mishchenko*, 1991; *Khlebtsov*, 1993]. Computations for the proxy of dust aerosols took a week on an ordinary work station. On the basis of run-time intercomparisons with a discrete dipole code, we estimate that the same computation

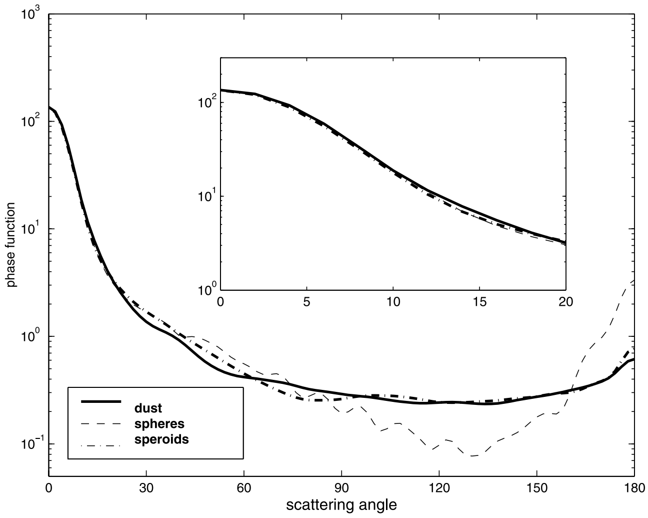

Figure 1. Phase functions of the proxy of dust aerosols (solid line), spherical model particles (dashed line), and spheroidal model particles (dash-dotted line).

would have taken several years with a code not exploiting particle symmetries and analytical orientational averaging.

[12] The resulting phase function for the reference case of dust-like aerosols is shown in Figure 1 (solid line). The single scattering albedo is $a = 0.885$, which deviates by less than 1.7% from results obtained by in situ nephelometer and particle soot absorption photometer measurements [*Haywood et al.*, 2001].

[13] For assessing the performance of spherical model particles in radiance and flux simulations, we wish to consider a "best-case scenario" by employing as input to the radiative transfer computations a phase function of spherical model particles that fits the reference phase function of the dust-like particles as closely as possible. Therefore we first attempt to find a size distribution of spheres that yields a best fit of the reference phase function.

[14] The reference phase function was fitted by using spheres with the same refractive index by applying a nonlinear least-squares minimization routine for optimizing the size distribution of the spheres (see *Kahnert* [2004] for details). The resulting phase function of the spheres is represented in Figure 1 by the dashed line. In the integration routines for computing averaged optical properties of ensembles of particles we replaced the crude rectangular integration scheme used earlier with a trapezoidal one and obtained slightly more accurate results than those reported earlier [*Kahnert*, 2004]. Higher order integration schemes did not give further improvements. For the single scattering albedo of the spherical model particles we find $a = 0.878$, which deviates from the reference case by less than 1%. The effective radius and effective variance of the size distribution of dust-like particles are $r_{\rm eff} = 1.19$ μm and $\nu_{\rm eff} = 0.14$. The corresponding results for the optimized size distribution of spheres are $r_{\rm eff} = 1.18$ μm and $\nu_{\rm eff} = 0.15$. Thus the "retrieval" errors for $r_{\rm eff}$ and $\nu_{\rm eff}$ are 1% and 7%, respectively. (The effective radius is the ratio of the 3rd to the 2nd moment of the size distribution and can be interpreted as the average size contributing to the optical cross sections and thus to the optical depth effective variance is a measure for how strongly particle sizes contributing to the optical depth are spread about the effective radius.) The phase function of the spherical model particles shows a characteristic overestimation of the reference phase function between 20° and 70°, an underestimation at side scattering angles between 80° and 160°, and a strong overestimation at backscattering angles between 160° and 180°.

[15] Next the phase function of the dust-like aerosols was fitted with an ensemble of randomly oriented spheroids by using for the spheroids the same refractive index, the optimized size distribution obtained by using spherical model particles, and by optimizing the distribution of spheroidal aspect ratios. Again, the optimization was performed by using a nonlinear least-squares error minimization method as described earlier [*Kahnert*, 2004]. No specific form of the shape distribution function (e.g., equiprobable, Gaussian, etc.) was prescribed in the fitting procedure. The resulting shape distribution is strongly peaked at the edges of the shape parameter interval, thus indicating that more elongated prolate and more flattened oblate spheroids are more useful for obtaining a good fit of the phase function than mildly aspherical spheroids. The resulting phase function of the spheroids is represented by the dash-dotted line in Figure 1. Clearly, using spheroids instead of spheres improves the fit of the reference case significantly. The single scattering albedo of the ensemble of spheroids is 0.877, which deviates from the reference case by less than 1%.

4. Radiative Transfer Simulations

[16] The single scattering albedo and the phase function of the dust-like aerosols, the spheres, and the ensemble of spheroids were used as input to the uvspec model [*Kylling et al.*, 1998; *Mayer et al.*, 1997]. The uvspec model is part of the libRadtran radiative transfer package which is available from www.libradtran.org. The DISORT algorithm was used as the radiative transfer solver [*Stamnes et al.*, 1988]. A standard tropical atmosphere with 49 layers was used for molecular scattering [*Anderson et al.*, 1986]. The aerosol optical depth profile is given in Table 1 and corresponds to measurements of scattering coefficients reported by *Myhre et al.* [2003]. The cumulative optical depth is 0.5 and corresponds well with the number densities reported by *d'Almeida* [1987] for background concentrations of dust aerosols. A Lambertian surface with an albedo of 0.1 was used, which is a typical value for ocean surfaces. With the uvspec model the differences in irradiances and radiances between the reference case (dust-like aerosols) and the results obtained with the spherical and spheroidal model particles were computed.

[17] The upper row in Figure 2 shows the downwelling diffuse spectral radiance I^- (left) at the bottom of the atmosphere (BOA) and the upwelling diffuse spectral radiance I^+ (right) at the top of the atmosphere (TOA). Results are shown for a solar zenith angle of 20°. The most prominent feature is a bright region around the solar zenith direction, which is due to the strong forward scattering peak in the phase function of the dust aerosols.

[18] The middle row in Figure 2 shows the error in diffuse spectral radiance in percent due to using spherical model particles. Maximum and minimum errors are listed in

Table 1. Vertical Profile of the Optical Depth of Dust-Like Aerosols[a]

Height, km	Extinction Optical Depth
4–5	0.043
3–4	0.124
2–3	0.123
1–2	0.133
0–1	0.082

[a]The same phase function and the same single scattering albedo were used for all layers.

Table 2. At the BOA (left) errors are small in close proximity to the solar zenith direction. However, there is a broad band of zenith angles between 20–70° from the solar zenith direction where the spherical particle model overestimates the reference case, with maximum errors around 26%. This corresponds closely to the overestimation of the reference phase function by the spherical model particles at scattering angles between 20–70° (see Figure 1). At zenith angles more than 80 away from the solar zenith direction one observes a strong underestimation of the spectral radiance down to −18%. Again, this corresponds to an underestimation of the dust phase function by

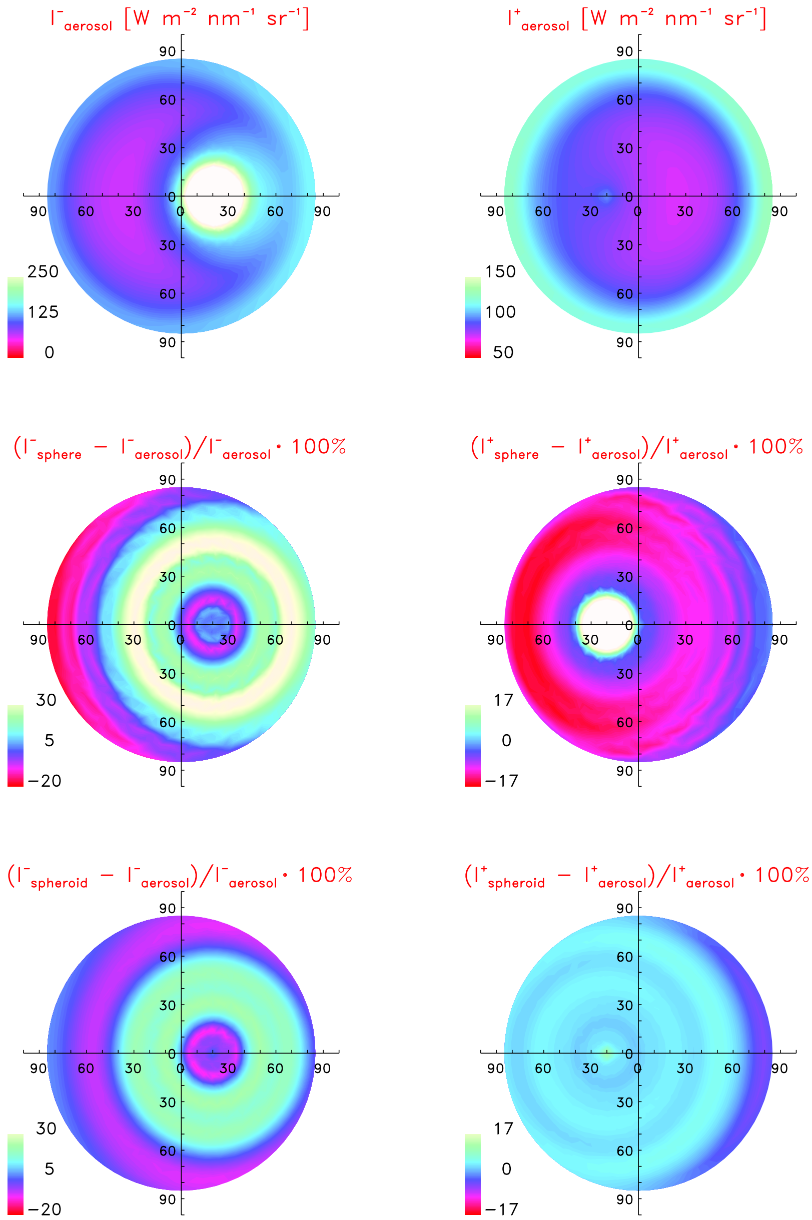

Figure 2. Results at the BOA (left column) and at the TOA (right column). The first row shows the diffuse spectral radiance computed for the dust-like aerosols (reference case). The second and third rows show the differences in percent between results computed with spherical and spheroidal model particles, respectively, a reference case.

Table 2. Minimum and Maximum Errors in the Spectral Radiance Simulations[a]

model	BOA		TOA	
	δ_{min}	δ_{max}	δ_{min}	δ_{max}
Spheres	−18.3	25.9	−16.3	115.3
Spheroids	−11.7	13.6	−7.1	5.4

[a]In percent.

the spherical particle model at scattering angles larger than 80°. At the TOA (right) the spectral radiance is strongly overestimated by the spherical particle model at backscattering directions. The errors are actually off the colour scale in the figure and go as high as 115%. The origin for these errors is the strong overestimation of the dust phase function by the spherical model particles at scattering angles larger than 160° (see Figure 1). At nadir angles more than 40° away from the solar nadir direction the spectral radiance is strongly underestimated (down to −16%). Again, this can be explained by the underestimation of the dust phase function at scattering angles between 80° and 160° (see Figure 1).

[19] The close correspondence between the errors in the single scattering phase function and the errors in spectral radiance indicate that the aerosols influence the transmitted and reflected spectral radiance primarily by single scattering. At the aerosol optical depth considered here the effect of multiple scattering is rather small.

[20] The bottom row in Figure 2 shows the error in diffuse spectral radiance in percent due to using spheroidal model particles. At the BOA the errors are considerably lower than in the case of spherical model particles. Particularly for zenith angles farther away from the solar zenith direction the spheroidal model particles produce significantly more accurate results than spherical model particles. However, in the region of 20–70° around the solar zenith direction, the spheroids also overestimate the spectral radiance, although the error is reduced by a factor of 2 as compared to the simulations with spheres. At the TOA (right) the improvements achieved with the spheroidal model particles are even more dramatic. Only around a rather narrow angular region around the solar nadir direction does the model overestimate the reference case by up to 5%, and it underestimates the reference case by down to −7% in a region farther than 90° away from the solar nadir direction. In between lies a broad angular region in which the error is almost zero. Again, a comparison with Figure 1 shows that these observation closely correspond to what we expect from comparing the phase functions of dust-like aerosols, spheres, and spheroids.

[21] Next we consider radiative net fluxes, which determine the radiative forcing in climate simulations. We computed spectral net fluxes for 550 nm at the TOA, averaged over all times of the day and all days of the year for a latitude of 20°. The direct solar flux was corrected for the varying earth-sun distance over the year. Results are shown in Table 3. Rows 1–3 show results obtained by using the phase function and single scattering albedo of the proxy of dust aerosols, spheres, and spheroids, respectively. Using spherical model particles results in a spectral flux error of 9.8 W m^{-2} nm^{-1}. By contrast, the corresponding error due to using spheroidal model particles is only 1.3 W m^{-2} nm

[22] The question arises how much the misrepresentation of the phase function by the model particles and the misrepresentation of the single scattering albedo each contribute to the total error in the spectral flux. We therefore repeated the flux computations by using the phase functions of spheres and spheroids in the simulations, but by substituting the "correct" value of the proxy of dust particles for the single scattering albedo. This allows us to isolate the error in the spectral flux simulation due to a misrepresentation of the phase function. The results are shown in rows 4 and 5 in Table 3. The spherical model particles still overestimate the flux by 8.1 W m^{-2} nm^{-1}. The corresponding error for the spheroidal model particles is −0.9 W m^{-2} nm^{-1}. As we saw earlier, the spherical and spheroidal model particles reproduce the single scattering albedo with an error of less than 1%. It is therefore not surprising to observe that the spectral flux error of the spherical model particles can be largely explained by the poor fitting of the phase function.

[23] In Figure 1 we see that the spherical particles both overestimate and underestimate the reference phase function for different angular intervals. However, the underestimation obviously dominates in the flux simulations, since an underestimation of the diffuse flux reflected back into space results in an overestimation of the net flux. The reason for this can be seen in Figure 2 (middle right panel). The overestimation of the diffuse upwelling spectral flux at the TOA is limited to a rather narrow solid angle interval around the solar nadir direction. The underestimation, on the other hand, occurs in a fairly broad range of solid angles. When integrating over all nadir and azimuth angles, only a partial error cancellation occurs. The final result is dominated by the negative errors due to their broad distribution over nadir directions.

[24] Finally we want to estimate how these spectral net flux errors compare to corresponding errors due to uncertainties in the refractive index. Since we only want to obtain a rough estimate, we simply re-run the Mie computations for spheres with different refractive indices, use the resulting single scattering optical properties in the radiative transfer simulations, and compare the resulting net fluxes with those we reported for the spherical model particles. The imaginary part of the refractive index we used of 0.005 has an error margin of ±0.003 [*Sokolik et al.*, 1993]. More recent measurements indicate that the imaginary part is rather lower than the value we used [*Myhre et al.*, 2003]. For the real part of the refractive index we used 1.65. Measurements have been reported that yield for the real part of the refractive index at a wavelength of 550 nm a value of 1.56 [*Sokolik et al.*, 1993]. We therefore perform two

Table 3. Annual Average of the Spectral Net Flux F_λ at the TOA for the Reference Case and for Spheroidal and Spherical Model Particles

Phase Function	Single Scattering Albedo	F_λ, W m^{-2} nm^{-1}	ΔF_λ, W m^{-2} nm^{-1}
Dust-like	dust-like	963.2	reference case
Spheres	spheres	973.0	9.8
Spheroids	spheroids	964.5	1.3
Spheres	dust-like	971.3	8.1
Spheroids	dust-like	962.3	−0.9

Table 4. Annual Average of the Spectral Net Flux F_λ at the TOA for Spheres With Different Refractive Indices

Refractive Index	F_λ, W m^{-2} nm^{-1}	ΔF_λ, W m^{-2} nm^{-1}
1.65 + 0.005*i*	973.0	
1.65 + 0.002*i*	953.3	−19.7
1.56 + 0.005*i*	974.8	1.8

different flux simulations using spherical particles with a refractive index of 1.65 + 0.002*i* and of 1.56 + 0.005*i*. The results are shown in Table 4 and are compared to the results obtained for spheres with a refractive index of 1.65 + 0.005*i*.

[25] Clearly, uncertainties in the real part of the refractive index seem to be less important. The corresponding error in spectral net flux simulations amounts to less than 2 W m^{-2} nm^{-1}. The error due to using spherical model particles is more than a factor of 5 higher (see Table 3) than the error due to the uncertainty in the real part of the refractive index. However, the uncertainty in the imaginary part of the refractive index is substantial. It results in an error of almost −20 W m^{-2} nm^{-1}. Nevertheless, comparing absolute error values we see that the error due to the use of spherical model particles (almost 10 W m^{-2} nm^{-1}) amounts to 50% of the error resulting from the uncertainty in the imaginary part of the refractive index, which is quite substantial.

5. Discussion

[26] For the downwelling spectral radiance at the BOA we found that spherical model particles can cause errors between −18% and 26%. This error range corresponds to varying the extinction optical depth τ of the dust-like aerosols between approximately 0.7τ and 1.5τ. At the TOA the spherical model particles caused errors in the spectral radiance simulations between −16.3% and 115.3%. If we disregard the extreme error values in the solar nadir direction, then this error range corresponds to varying the extinction optical depth of the dust aerosols between approximately 0.5τ and 2τ. Thus simulation errors caused by using spherical model particles correspond to a rather large uncertainty range in the optical depth.

[27] The errors in the downwelling spectral radiance at the BOA simulated with spheroidal model particles are between −11.7% and 13.6%. This is roughly half as much as the corresponding errors for spherical model particles. However, these values are still rather high. The origin of the positive errors is the overestimation of the dust phase function by spheroids for scattering angles between 20° and 70°. At the TOA the improvements achieved by using spheroidal model particles become much more pronounced. The errors in the upwelling spectral radiance range from 5.4% within a rather small angular region around the solar nadir direction to −7.1% farther than 90° away from the solar nadir direction. However, for a large range of solid angles the error is close to zero.

[28] It was found earlier [e.g., *Kahnert*, 2002b; *Nousiainen and Vermeulen*, 2003; *Kahnert*, 2004] that more elongated prolate and more oblate spheroids are particularly useful in fitting the phase function of dust particles. Thus by extending the range of aspect ratios of the spheroidal model particles it may ossible to improve the fit of the phase function and thus to further reduce the errors in spectral flux simulations. For homogeneous spheres no such improvements could be made. However, it would be interesting to test the performance of inhomogeneous (coated) spheres as model particles. Another possibility would be to use different axisymmetric particles as model particles, such as finite circular cylinders. Spheroids of aspect ratios close to one have phase functions similar to spheres, which are of little use for accurately fitting phase functions of complex-shaped nonspherical particles. On the other hand, the phase functions of finite circular cylinders with aspect ratios close to one do not resemble those of spheres. Therefore the use of cylinders may not require as high a range of aspect ratios for producing better phase function fits as the use of spheroids. This would be an advantage, since all electromagnetic scattering codes based on expanding the fields in vector spherical functions often encounter numerical problems when simultaneously considering large size parameters and more extreme aspect ratios.

[29] Using spheres in the spectral net flux simulation results in a large positive error of almost 10 W m^{-2} nm^{-1}, which is mainly caused by the misrepresentation of the phase function by the spherical model particles. For several reasons, we think that this may actually be a conservative estimate. Firstly, we assumed constant number concentrations of dust aerosols typical for background situations over the entire year, thus neglecting dust storm events. During dust storms the particle size distribution is strongly shifted to larger sizes. However, for larger size parameters the misrepresentation of the phase function of nonspherical particles by spherical model particles will become even more pronounced. Secondly, the imaginary part of the refractive index used in this study is probably at the higher end of what is reasonable for dust aerosols. Recent studies indicate that it may be much lower [*Kaufman et al.*, 2001; *Myhre et al.*, 2003]. This means that for dust aerosols scattering may be even more important as compared to absorption. A misrepresentation of the scattering phase function by spherical model particles may therefore, in reality, have an even stronger effect on spectral flux simulations as was estimated here.

[30] A simulation based on using Mie theory suggests that the error in spectral net flux simulations due to using spherical model particles is about 5 times larger than the error resulting from an uncertainty in the real part of the refractive index, and about 50% of the error due to the uncertainty in the imaginary part of the refractive index. We emphasize that these figures are only based on a rough estimate, and that the error related to the use of spherical model particles has to be considered a conservative estimate. Also, as more accurate measurements for the refractive index of dust aerosols become available, the relative importance of using model particles more suitable than spheres will greatly increase.

[31] The use of spheroidal model particles only caused an error in spectral net flux simulations of 1.3 W m^{-2} nm^{-1}. This indicates that it is in principle possible to achieve dramatic improvements in spectral flux simulations by using simple axisymmetric particles instead of spheres. It may also be possible to further reduce the error by extending the range of spheroidal aspect ratios or by using finite circular cylinders as model particles.

[32] We emphasize again that the simulations reported here were performed for one wavelength (550 nm). Accurate broad band simulations of aerosol optical properties based on using non-axisymmetric particles would place very high demands on computational resources. However, the wavelength considered here is an important wavelength, as it lies near the maximum of the solar spectrum. Therefore, on the basis of our results it appears probable that the use of spherical model particles in broadband flux simulations may cause significant errors, and that spheroids or other simple axisymmetric model particles may give considerably more accurate results.

[33] **Acknowledgments.** Funding from the Norwegian Research Council is gratefully acknowledged.

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