2.2.2 T-matrix method (TMM)

We use the extended precision version of the code described by Mishchenko and Travis (1998) for modeling optical properties of spheroids. To improve the coverage of the particle spectrum (x, ϵ_m, and m), internal parameter values of the TMM code, which primarily determine the limits of the convergence procedures, were increased (NPN1 = 290, NPNG1 = 870, NPN4 = 260) as discussed by Mishchenko and Travis (1998). Though, in general, the TMM provides exact solutions for scattering problems, non-physical results might be obtained due to numerical problems. To reduce the probability of non-physical results and to increase the accuracy of the results, the parameter DDELT, i.e. the absolute accuracy of computing the expansion coefficients, was set to 10^{-6} (default 10^{-3}). In non-converging cases, which happened near the upper limit of the covered size range, the requirements were relaxed to DDELT = 10^{-3}. Cases that did not converge even with the relaxed DDELT were not included in the data set. Nevertheless, some non-physical results were obtained by this approach, for example, ~~$\omega_0 > 1$~~$\omega_0 \geq 1$, or outliers of otherwise smooth $\omega_0(x)$ or $g(x)$ curves. Thus, for plausibility checks for each particle shape and refractive index, single scattering albedos ω_0 and asymmetry parameters g were plotted over size parameter x and outliers were recalculated with slightly modified size parameters. Recalculations with non-physical results were not included in the data set, which reduces the upper limit of the covered size range for that particular particle shape and refractive index.

2.2.3 Improved geometric optics method (IGOM)

Optical properties of large spheroids were calculated with the improved geometric optics method (IGOM) code provided by ~~Bi et al. (2009); Yang et al. (2007)~~Yang et al. (2007); Bi et al. (2009). In general, this approximation is most accurate if the particle and its structures are large compared to the wavelength. In addition to reflection, refraction, and diffraction by the particle, which are considered by classical geometric optics codes, IGOM also considers the so-called edge effect contribution to the extinction efficiency q_{ext} (Bi et al., 2009). Classical geometric optics results in ~~$q_{ext}=2$~~$q_{ext} = 2$, whereas q_{ext} is variable in case of IGOM. The default settings of the code were used. The minimum size parameter was selected depending on the maximum size ~~achieved~~ calculated with TMM.

2.2.4 Discrete dipole approximation code ADDA

Natural non-spherical aerosol particles, such as desert dust particles, comprise practically an infinite number of particle shapes, thus it is impossible to cover the full range of shapes in aerosol models. Moreover, the shape of each individual particle is never known under realistic atmospheric conditions. Consequently, typical irregularities such as flat surfaces, deformations or aggregation of particles, can be considered only in an approximating way. To enable the user of MOPSMAP to investigate the effects of such irregularities the properties of six exemplary irregular particle shapes, as introduced by Gasteiger et al. (2011b), are provided. The geometric shapes were constructed using the object modeling language Hyperfun (Valery et al., 1999). The first three shapes are prolate spheroids with varying aspect ratios (A: ~~$\epsilon'=1.4$~~$\epsilon' = 1.4$, B: ~~$\epsilon'=1.8$~~$\epsilon' = 1.8$, C: ~~$\epsilon'=2.4$~~$\epsilon' = 2.4$) and

surface deformations according to Gardner (1984). Shape D is an aggregate composed of ten overlapping oblate and prolate spheroids; surface deformations were applied as for shapes A-C. Shape E and F are edged particles with flat surfaces and varying aspect ratio.

The optical properties were calculated with the discrete dipole approximation code ADDA (Yurkin and Hoekstra, 2011). A large number of particle orientations needs to be considered for the determination of orientation-averaged properties. ADDA provides an optional built-in orientation averaging scheme in which the calculations for the required number of orientations is done within a single run. An individual ADDA run using this scheme requires approximately the time for one orientation multiplied with the number of orientations (typically a few hundreds), which can result in computation times of several weeks for large x. Because of the long computation times we split them up and performed independent ADDA runs for each orientation. The orientation-averaged properties are calculated in a subsequent step using the ADDA results for the individual orientations (see below).

The computational demand of DDA calculations increases strongly with size parameter x, typically with about x^5 to x^6. Thus, when aiming for large x, which is required for mineral dust in the visible wavelength range, it is necessary to find code parameters and an orientation averaging approach that provide a compromise between computation speed and accuracy.

The ~~orientation sampling and averaging is described in Appendix ??. The~~ ADDA code allows mainly the following code parameters to be optimized:

- DDA formulation
- Stopping criterion of the iterative solver
- Number of dipoles per wavelength

We estimate the accuracy of the ADDA results by comparing orientation-averaged q_{ext}, q_{sca}, $a_1(0°)$, $a_1(180°)$, and $a_2(180°)/a_1(180°)$ with results obtained using more strict calculation parameters. Accuracy tests are performed for shapes B and C~~(Fig. 1 of Gasteiger et al. (2011b))~~, for size parameters ~~x_v=10.0~~$x_v = 10.0$, 12.0, 14.4, 17.3, 19.0, and 20.8, and for refractive index ~~m = 1.52 + 0.0043i~~$m = 1.52 + 0.0043i$, i.e. 12 single particle cases are considered in total. By comparing the different DDA formulations available in ADDA, it was found that the filtered coupled-dipole technique (ADDA command line parameter ~~"~~'-pol fcd -int fcd~~"~~'), as introduced by Piller and Martin (1998) and applied by Yurkin et al. (2010), offers the best compromise between computation speed and accuracy of modeled optical properties. Using a stopping criterion for the iterative solver of 10^{-4} instead of 10^{-3} ~~,~~has only negligible influence on ~~orientation-averaged~~ optical properties (~~< 0.1~~< 0.1 %) but requires approximately 30 % more computation time; thus, we used 10^{-3} for the ADDA calculations to create our data set. The extinction efficiency q_{ext} and the scattering efficiency q_{sca} change in all cases by less than 0.3 % if a grid density of 16 dipoles per wavelength is used instead of 11. The maximum relative changes due to the change in dipole density are 0.2 % for $a_1(0°)$, 1.7 % for $a_1(180°)$, and 1.9 % for $a_2(180°)/a_1(180°)$. Because of the large difference in computation time, which is about a factor 3-4, and the low loss in accuracy, about 11 dipoles per wavelength were selected for the MOPSMAP data set. For

$x_v < 10$ we use the same dipole set as for $x_v = 10$ so that the number of dipoles per wavelength increases with decreasing x_v, being about $110/x_v$.

The particle orientation is specified by three Euler angles (α_e, β_e, γ_e) as described by Yurkin and Hoekstra (2011) and basically a step size of 15° is applied for β_e and γ_e resulting in 206 independent ADDA runs for each irregular particle. The orientation sampling and averaging is described in detail in Sect. S1.1 of the Supplement.

To test the accuracy of the selected orientation averaging scheme, orientation-averaged optical properties were ~~calculated using a denser grid of orientation grid points (β_e and γ_e, as defined in Appendix ??, but with a step size~~ compared to results using a much smaller step of 5°~~); these~~ for β_e and γ_e. These calculations consider about 12 times more orientations than the calculations ~~for our data set~~used for MOPSMAP. Details are presented in Sect. S1.2 of the Supplement. Maximum deviations of ~~0.6~~less than 1 % are found for q_{ext}~~and~~ , q_{sca}~~are found.~~ , and $a_1(0°)$. For backscatter properties, $a_1(180°)$ ~~,~~and $a_2(180°)/a_1(180°)$~~change by not more than 0.9%, 6.5%, and 12%, respectively~~typical deviations are of the order of a few percent (max. 14 %). Moreover, in Sect. S1.3 of the Supplement, the selected orientation averaging scheme is applied to spheroids and their optical properties are compared to reference TMM results. These deviations are comparable to those given in Sect. S1.2.

In summary, ADDA with the filtered coupled-dipole technique, at least 11 dipoles per wavelength, and a stopping criterion for the iterative solver of 10^{-3} was used for optical modeling of the irregularly-shaped particles in our data set together with the orientation averaging scheme ~~described in Appendix ??~~combining 206 ADDA runs. Tests demonstrate that the modeling accuracy is mainly determined by the applied orientation averaging scheme.

2.3 Optical data set

Using the codes with the settings described above, a data set of modeled optical properties of single particles in random orientation was created. ~~The complete data set requires about 42 gigabytes of storage capacity.~~ For spheres we stored, instead of single particle properties, averages over narrow size bins as described above. An overview over the wide range of sizes, shapes, and refractive indices of the particles in the data set is given in Tables 1 and 2. For each combination of refractive index and shape a separate netcdf file was created, e.g., 'spheroid_0.500_1.5200_0.008600.nc' for spheroids with $\epsilon_m = 0.5$ (prolate with $\epsilon' = 2.0$) and $m = 1.52 + 0.0086i$. Each file contains the optical properties on a grid of size parameters. The complete data set requires about 42 gigabytes of storage capacity.

For spheres and spheroids the minimum size parameter is set to 10^{-6}, and the maximum size parameter is set to ~~$x \approx 1005$ to cover $r_c = 40$ μm at $\lambda = 250$~~ $x \approx 1005$ to cover, e.g., $r_c = 80$ μm at $\lambda = 500$ nm. The size increment is 1 % (i.e. $x_{i+1}/x_i = 1.01$) in case of spheres, 5 % in case of TMM spheroids, and 10 % for IGOM spheroids. In case of spheroids having refractive indices most relevant for atmospheric studies, the TMM is applied up to (or close to) the largest possible size parameter with the approach described in Sect. 2.2.2. The maximum size parameter ~~for TMM~~ of the TMM calculations is reduced for less relevant refractive indices. An overview is given in Sect. S2 of the Supplement and a detailed list of the maximum size parameters for

S1 Orientation averaging of irregularly-shaped particles and accuracy assessment

S1.1 Orientation averaging scheme

The particle orientation is specified by three Euler angles (α_e, β_e, γ_e) as described by Yurkin and Hoekstra (2011). Averaging over β_e is done with a step width of 15°, and for each β_e up to 24 γ_e are used for averaging (see details in Table S1). The averaging over α_e is done within a single ADDA computation because rotation over α_e is equivalent to the rotation of the scattering plane and is computationally cheap. The optical properties are averaged over 32 α_e. In total, for a single particle 206 individual ADDA runs are performed and, if the averaging over α_e is considered, 6592 orientations are evaluated. For the numerical calculation of orientation averages of extensive optical properties $\zeta \in \{C_{ext}, C_{sca}, C_{sca} \cdot \mathbf{F}\}$, the following steps were applied.

Table S1: β_e and γ_e grid points for irregularly-shaped particles in MOPSMAP data set.

β_e	γ_e
0°, 180°	0°
15°, 165°	0°, 60°, 120°, ... , 300°
30°, 150°	0°, 30°, 60°, ... , 330°
45°, 60°, 75°, 90°, 105°, 120°, 135°	0°, 15°, 30°, ... , 345°

Averaging over α_e is done by ADDA for each β_e-γ_e-pair using the option '-phi_integr 1'. The α_e-averaged quantity is denoted as $\zeta_\alpha(\beta_e, \gamma_e)$.

As the next step, averaging over γ_e is done (outside ADDA) for each β_e using

$$\zeta_{\alpha,\gamma}(\beta_e) = \frac{1}{N_\gamma} \sum_{i_\gamma=1}^{N_\gamma} \zeta_\alpha(\beta_e, \gamma_e[i_\gamma]). \tag{S1}$$

where N_γ is the number of equidistant γ_e grid points for the given β_e (Table S1).

Finally, the orientation-averaged ζ is obtained by averaging over β_e using

$$\zeta = \frac{1}{2} \int_{0°}^{180°} \widetilde{\zeta}_{\alpha,\gamma}(\beta_e) \cdot \sin\beta_e \cdot d\beta_e \tag{S2}$$

where $\widetilde{\zeta}_{\alpha,\gamma}(\beta_e)$ is linearly interpolated between the available $\zeta_{\alpha,\gamma}(\beta_e)$ grid points (Eq. S1, Table S1). Numerical integration of Eq. S2 is performed using a step width of $\Delta\beta_e = 0.15°$.

S1.2 Accuracy assessment using dense Euler angle grid

Here we present a comparison of single particle properties calculated either with the orientation averaging scheme used in the MOPSMAP data set (Sect. S1.1) or using a much denser grid of orientation angles (5° step size for β_e and γ_e). This comparison provides an estimation of the accuracy of the former using the latter as reference.

Irregular shapes B, C, and F with $m = 1.52 + 0.0043i$ at six size parameters x_v from 10.0 to 20.8 are considered, i.e., 18 different single particles. In the following, tables are shown for the extinction efficiency q_{ext} (Table S2), scattering efficiency q_{sca} (Table S3), forward scattering phase function $a_1(0°)$ (Table S4), backscattering phase function $a_1(180°)$ (Table S5), and the normalized 2,2-element of the scattering matrix at backward direction $a_2(180°)/a_1(180°)$ (Table S6), where each line corresponds to a size parameter and the three values separated by slashes correspond to the different irregular shapes B/C/F.

In case of q_{ext} and q_{sca}, i.e. properties integrated over all scattering angles, the deviation between MOPSMAP and the reference is virtually zero (Tables S2, S3). The same is true for forward scattering (Table S4). Larger deviations typically in the order of a few percent (max. 14%) occur in case of backscattering. It is well known that scattering under 180° is very sensitive to various parameters of a scattering problem, here the particle orientation. The effect on atmospheric aerosols is however reduced as under realistic conditions over- and underestimates partly compensate according to the ensemble of different particles.

Table S2: Extinction efficiency q_{ext} from the MOPSMAP data set compared to results obtained using a dense grid of step size 5° for β_e and γ_e. The three values separated by slashes correspond to shapes B, C, and F. The relative deviation of q_{ext} of MOPSMAP from the reference is rounded to full percent values.

size parameter	MOPSMAP data set	dense β_e and γ_e grid	rel. deviation in %
x_v=10.0	2.269/2.064/2.069	2.267/2.053/2.076	0/+1/0
x_v=12.0	2.521/2.442/2.279	2.530/2.430/2.075	0/+1/0
x_v=14.4	2.180/2.367/2.272	2.184/2.371/2.280	0/0/0
x_v=17.3	2.203/2.097/2.090	2.205/2.100/2.101	0/0/-1
x_v=19.0	2.303/2.200/2.106	2.306/2.200/2.114	0/0/0
x_v=20.8	2.229/2.289/2.195	2.232/2.297/2.181	0/0/+1

Table S3: Scattering efficiency q_{sca} from the MOPSMAP data set compared to results obtained using a dense grid of step size 5° for β_e and γ_e. The three values separated by slashes correspond to shapes B, C, and F. The relative deviation of q_{sca} of MOPSMAP from the reference is rounded to full percent values.

size parameter	MOPSMAP data set	dense β_e and γ_e grid	rel. deviation in %
x_v=10.0	2.076/1.881/1.904	2.074/1.870/1.910	0/+1/0
x_v=12.0	2.295/2.226/2.082	2.303/2.214/2.075	0/+1/0
x_v=14.4	1.916/2.114/2.042	1.920/2.118/2.049	0/0/0
x_v=17.3	1.898/1.803/1.822	1.899/1.807/1.832	0/0/-1
x_v=19.0	1.975/1.885/1.816	1.977/1.886/1.824	0/0/0
x_v=20.8	1.879/1.954/1.883	1.881/1.961/1.869	0/0/+1

Table S4: Forward scattering phase function $a_1(0°)$ from the MOPSMAP data set compared to results obtained using a dense grid of step size 5° for β_e and γ_e. The three values separated by slashes correspond to shapes B, C, and F. The relative deviation of $a_1(0°)$ of MOPSMAP from the reference is rounded to full percent values.

size parameter	MOPSMAP data set	dense β_e and γ_e grid	rel. deviation in %
x_v=10.0	74.00/70.20/78.66	73.94/69.62/78.62	0/+1/0
x_v=12.0	119.8/121.9/126.7	120.3/121.6/126.8	0/0/0
x_v=14.4	154.7/176.6/188.7	154.8/176.8/188.8	0/0/0
x_v=17.3	224.8/225.4/253.3	224.8/225.5/253.2	0/0/0
x_v=19.0	288.1/288.4/309.4	288.0/288.2/309.6	0/0/0
x_v=20.8	340.4/366.8/391.0	340.7/367.2/389.5	0/0/0

Table S5: Backward scattering phase function $a_1(180°)$ from the MOPSMAP data set compared to results obtained using a dense grid of step size 5° for β_e and γ_e. The three values separated by slashes correspond to shapes B, C, and F. The relative deviation of $a_1(180°)$ of MOPSMAP from the reference is rounded to full percent values.

size parameter	MOPSMAP data set	dense β_e and γ_e grid	rel. deviation in %
x_v=10.0	0.3855/0.3277/0.3184	0.3758/0.3262/0.3084	+3/0/+3
x_v=12.0	0.3445/0.2643/0.3270	0.3238/0.2557/0.3254	+6/+3/+1
x_v=14.4	0.3488/0.2409/0.4018	0.3423/0.2492/0.3844	+2/-3/+5
x_v=17.3	0.3244/0.2423/0.4634	0.3091/0.2581/0.4306	+5/-6/+8
x_v=19.0	0.2866/0.2200/0.4625	0.2702/0.2174/0.4115	+6/+1/+12
x_v=20.8	0.2882/0.2185/0.4352	0.2800/0.2170/0.3822	+3/+1/+14

Table S6: Normalized (2,2)-element of the scattering matrix at backward direction $a_2(180°)/a_1(180°)$ from the MOPSMAP data set compared to results obtained using a dense grid of step size 5° for β_e and γ_e. The three values separated by slashes correspond to shapes B, C, and F. The relative deviation of $a_2(180°)/a_1(180°)$ of MOPSMAP from the reference is rounded to full percent values.

size parameter	MOPSMAP data set	dense β_e and γ_e grid	rel. deviation in %
x_v=10.0	0.4894/0.5856/0.3342	0.5081/0.5166/0.3347	-4/+13/0
x_v=12.0	0.5398/0.5713/0.3195	0.5135/0.5407/0.3417	+5/+6/-6
x_v=14.4	0.5356/0.5489/0.3229	0.5200/0.5385/0.3396	+3/+2/-5
x_v=17.3	0.4884/0.5684/0.3274	0.4890/0.5454/0.3574	0/+4/-8
x_v=19.0	0.4799/0.5362/0.3343	0.4800/0.5159/0.3606	0/+4/-7
x_v=20.8	0.5188/0.5382/0.3294	0.4973/0.5387/0.3590	+4/0/-8

S1.3 Accuracy assessment using spheroids

Here ADDA together with the orientation averaging scheme used for the irregular shapes in the MOPSMAP data set (as described in Sect. S1.1) is applied to prolate spheroids with $m = 1.52 + 0.0043i$ and ϵ'=2.0 (ADDA option '-shape ellipsoid 1.0 2.0'). Volume-equivalent size parameters x_v=2, 4, and 10 are considered. As reference the optical properties of the same randomly-oriented particles are calculated with the TMM code of Mishchenko and Travis (1998).

The comparison again shows that the integrated parameters and the forward scattering almost perfectly agree whereas for backscattering few cases with larger deviations up to 17 % are obtained. In general, the relative deviations are of similar magnitude as those found in our tests of Sect. S1.2 though the number of independent ADDA calculations is lower for spheroids than for irregular particles because of the rotation symmetry of spheroids.

Table S7: Properties of prolate spheroids with ϵ'=2.0 and $m = 1.52 + 0.0043i$. The three values separated by slashes correspond to size parameters x_v=2, 4, and 10. The relative deviation of ADDA from TMM is rounded to full percent values.

optical parameter	ADDA + orient. avg.	TMM	rel. deviation in %
q_{ext}	1.656/3.861/2.274	1.650/3.860/2.256	0/0/+1
q_{sca}	1.621/3.780/2.068	1.615/3.778/2.048	0/0/+1
$a_1(0°)$	5.741/18.86/69.30	5.748/18.84/69.20	0/0/0
$a_1(180°)$	0.1069/0.1700/0.3597	0.0948/0.1686/0.3568	+13/+1/+1
$a_2(180°)/a_1(180°)$	0.9350/0.5175/0.4721	0.9230/0.4927/0.5678	+1/+5/-17