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Light scattering by marine algae: two-layer spherical and nonspherical models

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Abstract

Light scattering properties of algae-like particles are modeled using the T-matrix for coated scatterers. Two basic geometries have been considered: off-centered coated spheres and centered spheroids. Extinction, scattering and absorption efficiencies, plus scattering in the backward plane, are compared to simpler models like homogeneous (Mie) and coated (Aden–Kerker) models. The anomalous diffraction approximation (ADA), of widespread use in the oceanographic light-scattering community, has also been used as a first approximation, for both homogeneous and coated spheres. T-matrix calculations show that some light scattering values, such as extinction and scattering efficiencies, have little dependence on particle shape, thus reinforcing the view that simpler (Mie, Aden–Kerker) models can be applied to infer refractive index (RI) data from absorption curves. The backscattering efficiency, on the other hand, is quite sensitive to shape. This calls into question the use of light scattering techniques where the phase function plays a pivotal role, and can help explain the observed discrepancy between theoretical and experimental values of the backscattering coefficient in observed in oceanic studies.

Keywords: Light scattering; T-matrix; Nonspherical particles

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1. Introduction

Understanding the angular scattering and absorbing properties of algae is fundamental to the ability to describe light propagation through the ocean, one of the primary goals of optical oceanography. The development of such an ability is essential to the effective use of ocean colour remote sensing and primary production algorithms. Propagation of light through the sea is described by the equations of radiative transfer, relating the structure of the submarine light field to the inherent optical properties or IOPS [1]—properties of the hydrosol independent of the structure of the light field. The most important of these are the volume scattering function $\beta(\lambda)$, and the attenuation, scattering, and absorption coefficients $c(\lambda), b(\lambda), a(\lambda)$. An additional parameter of importance to ocean colour remote sensing is the backscattering coefficient $b_b(\lambda)$ [2], the integral of the volume scattering function over the backward hemisphere.

The current understanding of phytoplankton optical properties is based on a combination of the interpretation of direct measurements and the application of electromagnetic scattering models, predominantly using the formulations of Mie [3]. The first applications of Mie theory to explain oceanic particulate properties were made in the 1970s [4–6]. The anomalous diffraction approximation [10] and Mie theory were applied to derive spectral refractive indices by matching the measured and theoretical estimates of absorption and beam attenuation coefficients for a range of phytoplankton species cultured in the laboratory [7–9]. Mie theory has since been used widely in optical oceanography, e.g. in the analysis of relative importance of oceanic particulate groups to total scattering and backscattering coefficients [11,12], to establish angular constraints for in situ backscattering instruments [13], and to establish phase functions for radiative transfer models [14].

However, the assumption of spherical homogeneity that is the attractive simplicity of Mie theory is also a shortcoming as concerns application to algal optics. In reality eukaryotic phytoplankton are likely to possess heterogeneous intracellular refractive indices, associated with a variety of complex internal structures. These include silicate, cellulose or calcite cellular coatings or plates, absorbing chloroplasts containing the protein bound pigment complexes necessary for light harvesting and photosynthesis, other membrane bound organelles such as the nucleus and mitochondria, and regulatory/storage devices such as gas vacuoles and starch granules. In addition, phytoplankton are taxonomically diverse organisms, with over 5000 marine species reported in the most cosmopolitan class of *Bacillariophyceae* (diatoms) alone [15]. Associated with such diversity are a wide range of cellular shapes, many of which deviate significantly from the spherical.

While Mie theory, and the anomalous diffraction approximation, can adequately describe the observed attenuation, absorption and total scattering of algal cells [7], it is often unable to reproduce measured angular scattering data made with the chlorophyte *Chlorella* [16] and a variety of cultured algal species [17]. Experimental methods of analysing the influence of internal structure upon angular scattering data for *Chlorella vulgaris* similarly shows the strong influence of cellular heterogeneity on the volume scattering function [18,19]. These studies have also reported variable deviations from unity in the diagonal elements of the Mueller matrix, indicating potential additional departures from Mie simulations due to spherical asymmetry.

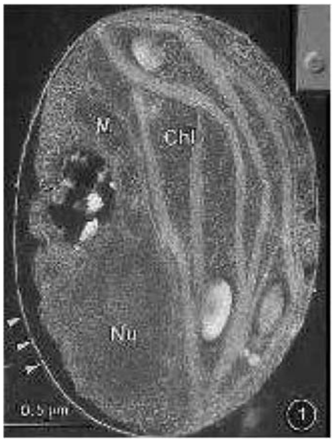

Fig. 1. A real-life oceanic scatterer: *Aerococcus anophagefferens*.

It thus appears that Mie theory is not adequate for the simulation of algal angular scattering [20,21]. The next most simple particle geometry proven capable of reproducing measured algal angular scattering is a two layered sphere with chloroplast as core [16]. But even this is a first approximation to real shapes of algal particles such as *Aerococcus anophagefferens* (see Fig. 1), whose shapes can be more accurately modeled as coated spheroids.

Faithful to the Bohren–Singham criterion of not modeling with inadequate methods [22], this paper attempts to model the light-scattering behavior of algal-like particles with more realistic models. The T-matrix method [23] allows for the calculation of light scattering properties of nonspherical particles, including the added complexity of layered bodies [24–26]. A comparison is made between current methods used for simple (Mie, ADA) and composite (Aden–Kerker, ADA-extended, T-matrix) scatterers.

2. Theory

Efficiency factors for extinction, scattering, and absorption ($Q_{ext}, Q_{sca}, Q_{abs}$) have been calculated by the authors using various methods; these factors, in turn, can be used to obtain the IOP of a particle suspension: attenuation, scattering, and absorption coefficients: c, b, a [7].

For coated spherical particles, the Aden–Kerker theory has been used [27]. Light scattering properties depend on the complex refractive index of core ($m_1 = n_1 + in'_1$) and coating ($m_2 = n_2 + in'_2$) relative to that of the surrounding medium. The dimensionless outer size parameter $x = ka$ and the core/particle ratio $q = b/a$ are used to describe the particle size and structure.

Assuming that the complex index of refraction of an homogeneous particle m is close to unity, efficiency factors can be calculated with the anomalous diffraction approximation (ADA). Values for spherical particles are well known [10]. When coated spherical particles are to be taken into account, the ADA can still be modified to yield optical efficiency factors:

$$
\begin{aligned}
Q_{\mathrm{ext}} = {} & 2 - 4z\mathrm{e}^{-z\rho_1 tg\beta_1}\,\frac{\cos\,\beta_1}{\rho_1}\,\sin(z\rho_1-\beta_1)\\
& - 4\left(\frac{\cos\,\beta_1}{\rho_1}\right)^2 \mathrm{e}^{-z\rho_1 tg\beta_1}\,\cos(z\rho_1-2\beta_1) + 4\left(\frac{\cos\,\beta_1}{\rho_1}\right)^2 \cos(2\beta_1)\\
& - 4\,\frac{\cos\,\beta_2}{\rho_2}\,\mathrm{e}^{-\rho_2 tg\beta_2}\,\sin(\rho_2-\beta_2) + 4\,\frac{\cos\,\beta_2}{\rho_2}\,z\mathrm{e}^{-z\rho_2 tg\beta_2}\,\sin(z\rho_2-\beta_2)\\
& - 4\left(\frac{\cos\,\beta_2}{\rho_2}\right)^2 \mathrm{e}^{-\rho_2 tg\beta_2}\,\sin(\rho_2-2\beta_2) + 4\left(\frac{\cos\,\beta_2}{\rho_2}\right)^2 z\mathrm{e}^{-z\rho_2 tg\beta_2}\,\sin(z\rho_2-2\beta_2),
\end{aligned} \tag{1}
$$

$$
Q_{\mathrm{abs}} = 1 + 2\,\frac{z\mathrm{e}^{-\rho_1' z}}{\rho_1'} + 2\,\frac{\mathrm{e}^{-\rho_1' z}-1}{(\rho_1')^2} + 2\,\frac{\mathrm{e}^{-\rho_2'} - z\mathrm{e}^{-\rho_2' z}}{\rho_2'} + 2\,\frac{\mathrm{e}^{-\rho_2'} - \mathrm{e}^{-\rho_2' z}}{(\rho_2')^2}, \tag{2}
$$

where $z = (1-q^2)^{1/2}$ and

$$
\begin{aligned}
&\rho_1 = 2x(n_2-1), \quad \rho_2 = 2x[qn_1 + (1-q)n_2 - 1], \quad tg\beta_1 = \frac{n_2'}{n_2-1},\\
&\rho_1' = 4xn_2', \quad \rho_2' = 4x[qn_1' + (1-q)n_2'], \quad tg\beta_2 = \frac{qn_1' + (1-q)n_2'}{qn_1 + (1-q)n_2 - 1}.
\end{aligned}
$$

In order to determine efficiency factors for nonspherical—or spherical, nonconcentric-particles, the T-matrix method is used, as adapted for layered particles [25]. Two particle geometries have been considered (Fig. 1):

- A sphere with an off-centered spherical inclusion (offset sphere) of inner and outer size parameters a, b, respectively. The inner layer is centered on the origin of coordinates, while the origin of the outer layer is displaced by a distance l. The size parameters $x = ka$, $q = b/a$ and $p = l/a$ are used.
- A centered, coated spheroid with axes a, b ($a =$ revolution axis), equivalent-size parameter $x = (ab^2)^{1/3}$ and eccentricity $\varepsilon = b/a$. The coating/particle size parameter ratio is given as q.

Both offset spheres and coated spheroids are axisymmetrical geometries, so the calculation of the T-matrix is simplified and Mishchenko's averaging scheme [28] can be applied. In addition, centered spheroids have a plane geometry, which allows for a further simplification [29].

In addition to extinction, absorption, and scattering efficiencies, the backscattering coefficient, defined as

$$
Q_{\mathrm{back}} = \frac{\int_{\pi/2}^{\pi} F_{11}(\vartheta)\sin\,\vartheta\,\mathrm{d}\vartheta}{\int_0^{\pi} F_{11}(\vartheta)\sin\,\vartheta\,\mathrm{d}\vartheta} \tag{3}
$$

is calculated, where $F_{11}(\vartheta)$ is the phase function for a scattering angle ϑ. This parameter (not to be mistaken as the backscattering cross section or the backscattered fraction for isotropically incident radiation) cannot be expressed in an explicit form in either the ADA or the full (Aden–Kerker) frameworks, but the angle integration can be easily calculated in the T-matrix formulation. Following the well-known expansion of the phase function as

$$F_{11}(\vartheta) = \sum_{s=0}^{\infty} a_1 P_s(\cos\,\vartheta) \tag{4}$$

and using the integration properties of the Legendre polynomials, Eq. (3) can be expressed as

$$Q_{\text{back}} = \frac{1}{2}\left(a_0 + \sum_{\substack{s=1\\ s\ \text{odd}}}^{\infty} a_s I_s\right) \quad \text{where } I_1 = -\frac{1}{2},\ \ I_s = -\frac{s-2}{s+1} I_{s-2}\ \ (s\ \text{odd}). \tag{5}$$

3. Results and discussion

Extinction, absorption and backscattering efficiency values for coated, concentric spheres are displayed in Figs. 2–4 as a function of size parameter. The shell refractive index is fixed at $m_2 = 1.02$, while the core refractive index (RI) has been chosen so that the volume-averaged RI index of the composite particle is the same as that of an "equivalent" homogeneous scatterer, a scheme known as volumetric approach (VA). In our case, the volume-averaged equivalent RI is set as $m_h = 1.05 + i0.005$, a value usually taken as representative of some biological samples such as planktonic cells [30–32]. Core RI values are given in Table 1 for several core/particle q ratios.

The VA-averaged imaginary part of the RI for a homogeneous sphere is usually obtained from absorption efficiency curves. This can be done by iteratively using the ADA, since the ADA-calculated Q_{abs} function is monotonous, increasing from 0 to 1 as the particle sizes grows from 0 to ∞. Both exact (Aden–Kerker) and approximate (ADA) calculations for coated particles show that the $Q_{\text{abs}} - kr$ ceases to be a monotonous curve. Absorption efficiency values reach a local maximum, before falling to their large particle size limit q^2 (valid only for a nonabsorbing shell, $n_2' = 0$). The results (dotted curves) are also shown in Fig. 3. It can be seen that the coated-sphere ADA, while not perfect, compares better to exact (Aden–Kerker) results than the ADA for homogeneous spheres (dotted curve, $q = 1$).

The comparison of homogeneous vs. coated spheres (Fig. 4) shows that the assumption of homogeneity ($q = 1$) can yield backscattering values up to an order of magnitude lower than those of coated particles. Results for smaller core ratios might be questioned, as the VA forces

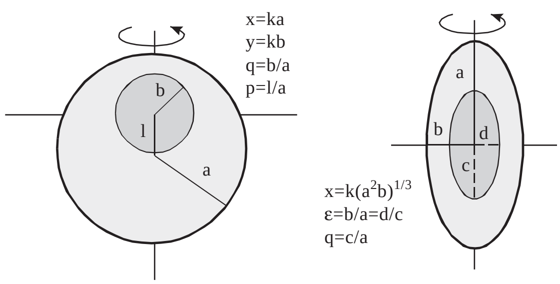

Fig. 2. Particle shapes.

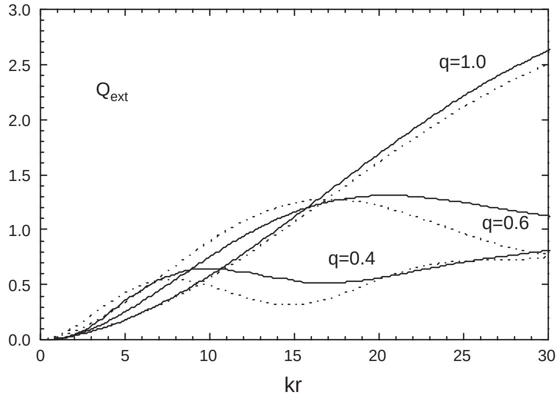

Fig. 3. Extinction efficiency values for coated, spherical particles as a function of particle size. Full line: exact (Aden–Kerker), dotted line: anomalous diffraction approximation.

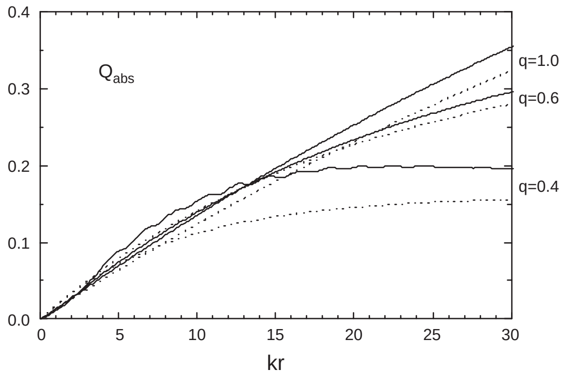

Fig. 4. Absorption efficiency values for coated, concentric spherical particles as a function of particle size. Full line: exact (Aden–Kerker), dotted line: anomalous diffraction approximation.

Table 1
Core index of refraction

q	m_1
0.4	$1.489 + i0.078$
0.6	$1.156 + i0.023$
0.8	$1.079 + i0.010$
1.0	$1.050 + i0.005$

unrealistically high values or core RI; however, the trend remains that larger core values under the VA result in lower backscattering values. Fig. 5 shows how the use of different core and shell RI values (assumed a volume-averaged equivalent RI set as $m_h = 1.05 + i0.005$ in all cases) result in

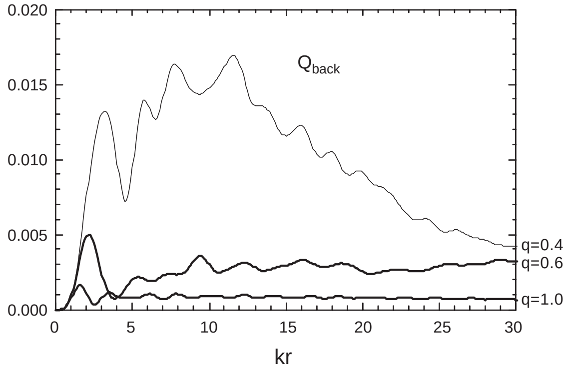

Fig. 5. Backscattering efficiency values for coated, concentric spherical particles as a function of particle size.

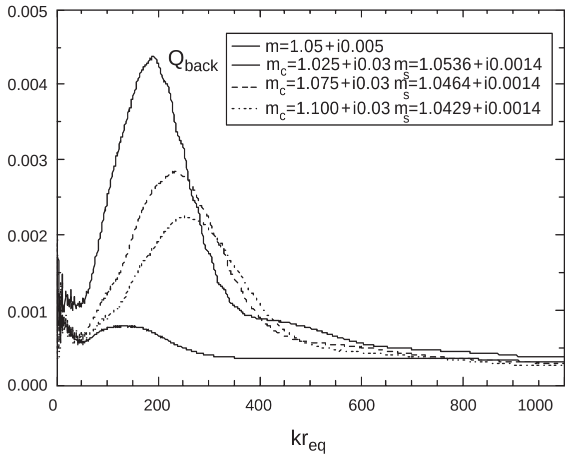

Fig. 6. Backscattering efficiency values for coated, concentric spherical particles with core/particle ratio $q = 0.5$ and several refractive indices within the VA.

higher Q_{back} values for coated ($q = 0.5$) particles, as compared to the homogeneous case, for most size parameter values.

This result falls in line with other studies employing volume equivalent refractive schemes to compare homogeneous and heterogeneous spheres. Models using two layered spheres with the chloroplast as core [33,34] or three layered spheres with the chloroplast as the central layer [34,35], have found backscattering most effected by cellular heterogeneity. The use of a heterogeneous geometry resulted in both spectral changes and an increase in magnitude of backscattering by between two and fifty times. The thickness and real refractive index of the shell appeared to have a significant effect on the magnitude of backscattering: a finding supported by Kitchen and Zaneveld [36] and Quinby-Hunt et al. [16], who also found a strong dependence of backscattering on the real refractive index of the core.

A more realistic model of marine particles requires nonsphericity to be taken into account. Two shapes have been considered: a coated spheroid and a sphere with a centered core and an offset

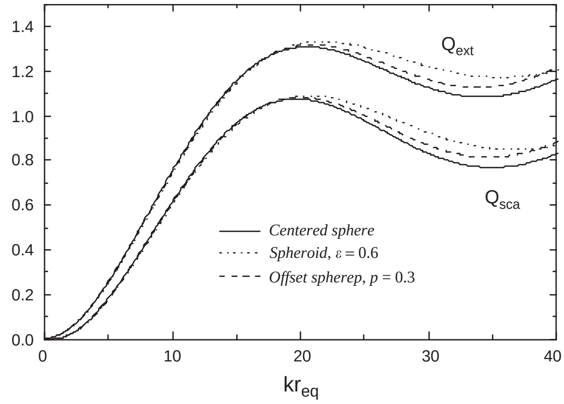

Fig. 7. Extinction and scattering efficiency values for coated particles with core/particle ratio $q = 0.6$: centered spheres, offset spheres ($p = 0.3$) and centered spheroids ($\varepsilon = 0.6$).

coating. In Figs. 6 and 7, efficiency values are shown for a coated sphere, an offset sphere (offset parameter $p = 0.3$) and a coated spheroid (axial ratio $\varepsilon = 0.6$). In all three cases, the core/particle q ratio is 0.6.

As seen in Fig. 6, both offset sphere and coated spheroid have extinction and scattering efficiency values very similar to that given by the coated spherical model. The absorption efficiency is found to be particularly shape-independent, as both particle shapes show Q_{abs} values that hardly differ from those given by the homogeneous, spherical (Mie) model. Previously mentioned studies on spherical scatterers [33–36] had already concluded that adopting a heterogeneous geometry had little effect on absorption, and variable effects on extinction and scattering. Aas [33] extended those results to nonspherical particles such as disks and cylinders. From our results, it is safe to state that the same general results applies to coated spheroids and offset spheres. Shape can then be considered as a minor contributor to absorption, which depends mainly on particle size and composition (refractive index, or indices for an heterogeneous scatterer). This result is of great importance to the oceanographic community, where refractive indices are obtained from absorption curves. Calculations of Q_{abs} by using the Aden–Kerker theory—or the ADA, still simpler—can be used for that purpose, thus obviating the need to resort to the full—T-matrix-theory for nonspherical particles.

Accurate calculations for backscattering efficiency values, on the other hand, require the use of the full nonspherical theory, as Fig. 7 shows. Aden–Kerker theory does not guarantee an accurate (or even approximate) value, so a further step is needed to simulate real particles. This also applies for the phase function (which is needed to calculate backscattering efficiency values, see Eq. (3)) (Fig. 8). In real particle systems, attention must be paid to the effect of polydispersity, which tends to smooth out the features of the phase function curves. Preliminary results on backscattering values obtained from a T-matrix simulation of a particle size distribution of polydisperse, coated spheroids [37] indicates that the smoothing out is not enough to assume that shape is not a relevant factor.

Previous assessments of the light-scattering capabilities of several seawater constituents show that their contribution is insufficient to account for the observed brightness of the ocean, where

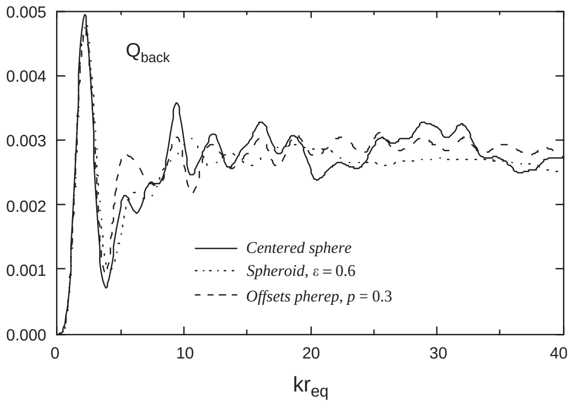

Fig. 8. Same as Fig. 6 for backscattering efficiency values.

backscattering is found to be higher than theory predicts by an order of magnitude [11,22]. Several theories have arisen to try to explain this discrepancy: bubbles in the water, wind-carried lithogenic particles, very small, poorly known particles, etc. It is also possible that the use of oversimplified light scattering models plays a role in the discrepancy. The use of more complex models (T-matrix, coated sphere ADA) are considered as a more plausible alternative for light-scattering sizing techniques where the phase function, or quantities derived from it, are used. The challenges are daunting, not least because many oceanic particles cannot be approximated as coated, axially-symmetric scatterers. Gordon and Du [38] concluded that modeling backscattering from marine particles, with their complex shapes, will be very difficult, if not impossible. While not pretending to downplay the difficulties involved in oceanic light scattering, similar problems have been encountered in other fields. Phase functions have been modeled for tropospheric particles using randomly oriented, polydisperse (in both size and shape) homogeneous spheroids [39]. An extension of that procedure to coated scatterers is well within the limits of the T-matrix method, and can be applied to particles known to have a coated spheroidal, or offset coated spherical, geometry.

Acknowledgements

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