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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 modelled 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.

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Keywords: Light scattering; T-matrix; Nonspherical particles

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

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1 structure of the light field. The most important of these are the absorption coefficient $a(\lambda)$ and the
2 volume scattering function $\beta(\lambda)$, from which the attenuation coefficient $c(\lambda)$ and the total
3 scattering coefficient $b(\lambda)$ can be derived. An additional parameter of importance to ocean colour
4 remote sensing is the backscattering coefficient $b_b(\lambda)$ [2], the integral of the volume scattering
5 coefficient over the backward hemisphere.

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

16 However, the assumption of spherical homogeneity that is the attractive simplicity of Mie
17 theory is also a shortcoming as concerns application to algal optics. In reality eukaryotic
18 phytoplankton are likely to possess heterogeneous intracellular refractive indices, associated with
19 a variety of complex internal structures. These include silicate, cellulose or calcite cellular coatings
20 or plates, absorbing chloroplasts containing the protein bound pigment complexes necessary for
21 light harvesting and photosynthesis, other membrane bound organelles such as the nucleus and
22 mitochondria, and regulatory/storage devices such as gas vacuoles and starch granules. In
23 addition, phytoplankton are taxonomically diverse organisms, with over 5000 marine species
24 reported in the most cosmopolitan class of *Bacillariophyceae* (diatoms) alone [14]. Associated with
25 such diversity are a wide range of cellular shapes, many of which deviate significantly from the
26 spherical.

27 While Mie theory, and the derived anomalous diffraction approximation, can adequately
28 describe the observed attenuation, absorption and total scattering of algal cells [7], it appears
29 unable to reproduce measured angular scattering data made with the chlorophyte *Chlorella* [15]
30 and a variety of cultured algal species [16]. Experimental methods of analysing the influence of
31 internal structure upon angular scattering data for *Chlorella vulgaris* similarly shows the strong
32 influence of cellular heterogeneity on the volume scattering function [17,18]. These studies have
33 also reported variable deviations from unity in the diagonal elements of the Mueller matrix,
34 indicating potential additional departures from Mie simulations due to spherical asymmetry.

35 It thus appears that Mie theory is not adequate for the simulation of algal angular scattering.
36 The next most simple particle geometry proven capable of reproducing measured algal angular
37 scattering is a two layered sphere with chloroplast as core [15]. Faithful to the Bohren–Singham
38 criterion of not modelling with inadequate methods [19], this paper attempts to model the light-
39 scattering behaviour of algal-like particles with more realistic models. The T-matrix method [20]
40 allows for the calculation of light scattering properties of nonspherical particles, including the
41 added complexity of layered bodies [21–23]. A comparison is made between current methods used
42 for simple (Mie, ADA) and composite (Aden–Kerker, ADA-extended, T-matrix) scatterers.

2. Theory

Efficiency factors for extinction, scattering, and absorption ($Q_{\text{ext}}, Q_{\text{sca}}, Q_{\text{abs}}$) have been calculated using different 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 [24]. 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 a 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 Q efficiencies:

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

$$Q_{\text{abs}} = 1 + 2 \frac{ze^{-\rho'_1 z}}{\rho'_1} + 2 \frac{e^{-\rho'_1 z} - 1}{(\rho'_1)^2} + 2 \frac{e^{-\rho'_2 z} - ze^{-\rho'_2 z}}{\rho'_2} + 2 \frac{e^{-\rho'_2 z} - e^{-\rho'_2 z}}{(\rho'_2)^2}, \quad (2)$$

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 particles, the T-matrix method is used, as adapted for layered particles [22]. 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 ($a = \text{revolution axis}$), equivalent-size parameter $x = (ab^2)^{1/3}$ and eccentricity $\varepsilon = b/a$. The coating/particle size parameter ratio is given as q .

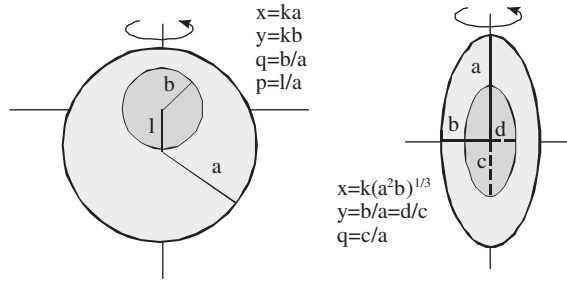


Fig. 1. Particle shapes.

Both offset spheres and coated spheroids are axisymmetrical geometries, so the calculation of the T-matrix is simplified and Mishchenko's averaging scheme [25] can be applied. In addition, centered spheroids have a plane geometry, which allows for a further simplification [26].

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

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

is calculated, where $F_{11}(\theta)$ is the phase function. 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

$$F_{11}(\vartheta) = \sum_{s=0}^{\infty} a_s P_s(\cos \vartheta) \quad (4)$$

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

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

3. Result and discussion

Extinction and absorption efficiency, plus backscattering coefficient, values for coated 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 RI has been chosen so that the volume-averaged RI index of the

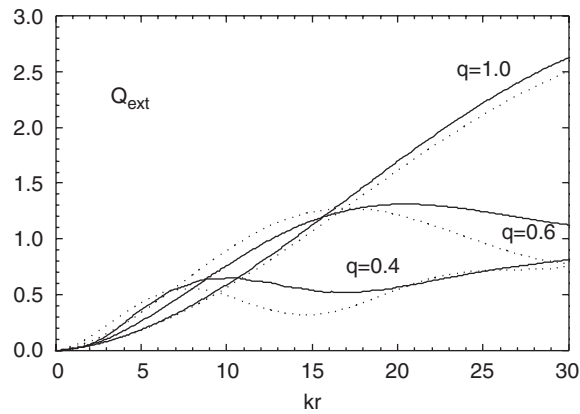


Fig. 2. Extinction efficiency values for coated, spherical particles as a function of particle size. Full line: exact (Aden-Kerker), dotted line: anomalous diffraction approximation.

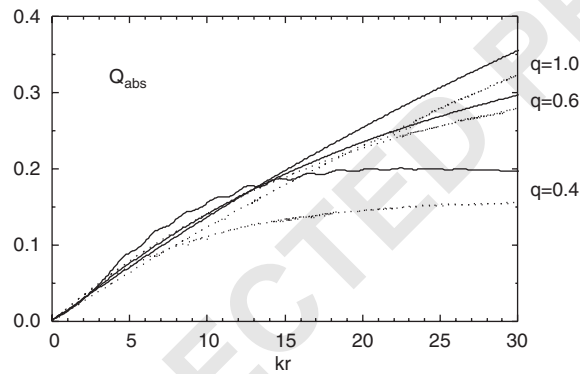


Fig. 3. Absorption efficiency values for coated, spherical particles as a function of particle size. Full line: exact (Aden-Kerker), dotted line: anomalous diffraction approximation.

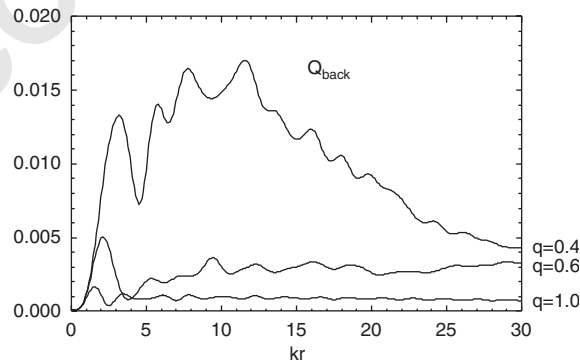


Fig. 4. Backscattering coefficient values for coated, spherical particles as a function of particle size.

1 Table 1
 2 Core index of refraction

3 q	4 m_1
5 0.4	1.489 + i0.078
6 0.6	1.156 + i0.023
7 0.8	1.079 + i0.010
8 1.0	1.050 + i0.005

9
 11 composite particle is the same as that of an “equivalent” homogeneous scatterer, a scheme known
 12 as volumetric approach (VA). In our case, the volume-averaged equivalent RI is set as
 13 $m_h = 1.05 + i0.005$, a value usually taken as representative of some biological samples such as
 14 planktonic cells [27–29]. Core RI values are given in Table 1 for several core/particle q ratios.

15 The VA-averaged imaginary part of the RI for a homogeneous sphere is usually obtained from
 16 absorption efficiency curves. This can be done by iteratively using the ADA, since the ADA-
 17 calculated Q_{abs} function is monotonous, increasing from 0 to 1 as the particle sizes grows from 0
 18 to ∞ . Both exact (Aden–Kerker) and approximate (ADA) calculations for coated particles show
 19 that the $Q_{\text{abs}} - kr$ ceases to be a monotonous curve. Absorption efficiency values reach a local
 20 maximum, before falling to their large-size limit q^2 (valid only for a nonabsorbing shell, $n'_2 = 0$).
 21 The results (dotted curves) are also shown in Fig. 3. It can be seen that the coated-sphere ADA,
 22 while not perfect, compares better than the diffraction approximation for homogeneous spheres
 23 ($q = 1$).

24 The comparison of homogeneous vs. coated spheres (Fig. 4) shows that the assumption of
 25 homogeneity ($q = 1$) can yield backscattering values up to an order of magnitude lower than those
 26 of coated particles. Results for smaller core ratios might be questioned, as the VA forces
 27 unrealistically high values or core RI; however, the trend remains that larger core values under the
 28 VA result in lower backscattering values. Fig. 5 shows how the use of different core and shell RI
 29 values (assumed a volume-averaged equivalent RI set as $m_h = 1.05 + i0.005$ in all cases) result in
 30 higher Q_{back} values for coated ($q = 0.5$) particles, as compared to the homogeneous case, for most
 31 size parameter values.

32 This result falls in line with other studies employing volume equivalent refractive schemes to
 33 compare homogeneous and heterogeneous spheres. Models using two layered spheres with the
 34 chloroplast as core [30,31] or three layered spheres with the chloroplast as the central layer [31,32],
 35 have found backscattering most effected by cellular heterogeneity, The use of a heterogeneous
 36 geometry resulted in both spectral changes and an increase in magnitude of backscattering by
 37 between two and 50 times. The thickness and real refractive index of the shell appeared to have a
 38 significant effect on the magnitude of backscattering: a finding supported by Kitchen and
 39 Zaneveld [33] and Quinby-Hunt et al. [15], who also found a strong dependence of backscattering
 40 on the real refractive index of the core.

41 A more realistic model of marine particles requires nonsphericity to be taken into account. Two
 42 shapes have been considered: a coated spheroid and a sphere with a centered core and an offset
 43 coating. In Figs. 6, 7 efficiency values are shown for a coated sphere, an offset sphere (offset

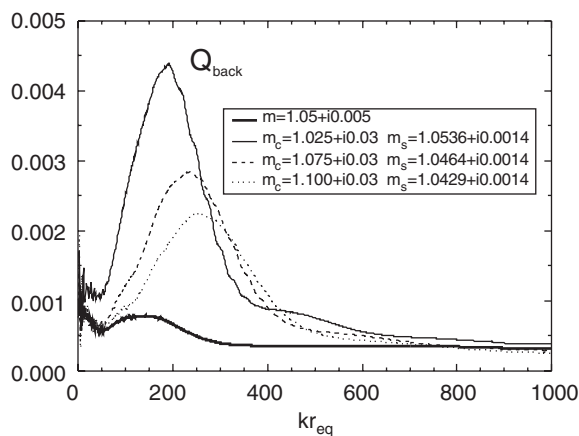


Fig. 5. Backscattering coefficient values for coated particles with core/particle ratio $q = 0.5$ and several refractive indices within the VA.

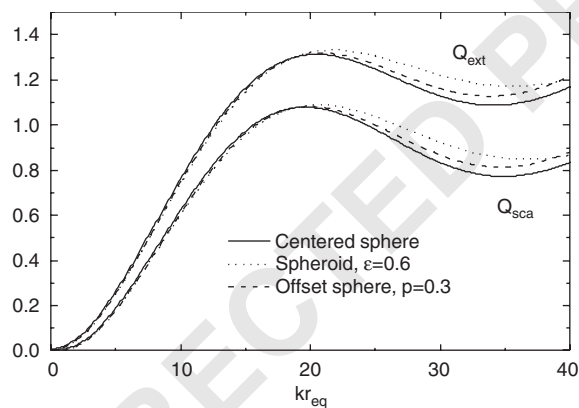


Fig. 6. Extinction and scattering efficiency values for coated particles with core/particle ratio $q = 0.6$: center spheres, offset spheres ($p = 0.3$) and centered spheroids ($\epsilon = 0.6$).

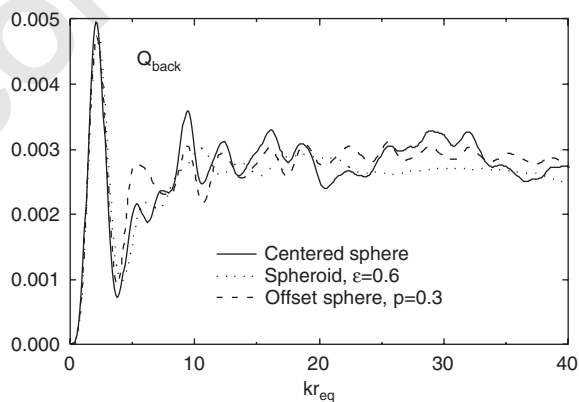


Fig. 7. Same as Fig. 6 for backscattering coefficient values.

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.

Both offset sphere and coated spheroid have extinction and absorption efficiencies very similar to that given by the coated spherical model. This suggests that the use of nonspherical models only becomes necessary if high accuracy is needed in the calculation of Q values (extinction, scattering, absorption). Only backscattering efficiency values depart significantly from the coated spherical case.

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 backscattering is found to be higher than theory predicts by an order of magnitude [11,19]. 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 models more complex (T-matrix, coated sphere ADA) are considered as a more plausible alternative for light-scattering sizing techniques.

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