



ELSEVIER

Journal of Quantitative Spectroscopy &  
Radiative Transfer ■ (■■■■) ■■■–■■■Journal of  
Quantitative  
Spectroscopy &  
Radiative  
Transfer[www.elsevier.com/locate/jqsrt](http://www.elsevier.com/locate/jqsrt)

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

Arturo Quirantes<sup>a,\*</sup>, Stewart Bernard<sup>b</sup><sup>a</sup>*Departamento de Física Aplicada, Facultad de Ciencias, Universidad de Granada, 18071 Granada, Spain*<sup>b</sup>*Department of Oceanography, University of Cape Town, Private Bag, Rondebosch 7700 Cape Town, South Africa*

## 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.

© 2004 Published by Elsevier Ltd.

*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

\*Corresponding author.

*E-mail addresses:* [aquiran@ugr.es](mailto:aquiran@ugr.es) (A. Quirantes), [bstewart@ocean.uct.ac.za](mailto:bstewart@ocean.uct.ac.za) (S. Bernard).

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  $q$ .

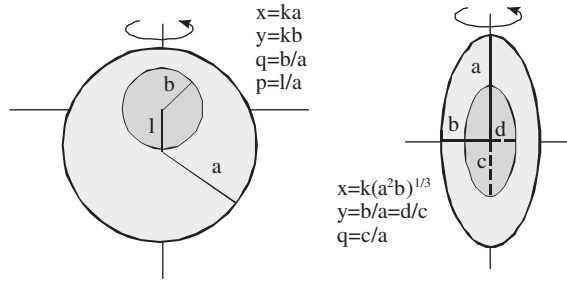


Fig. 1. Particle shapes.

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

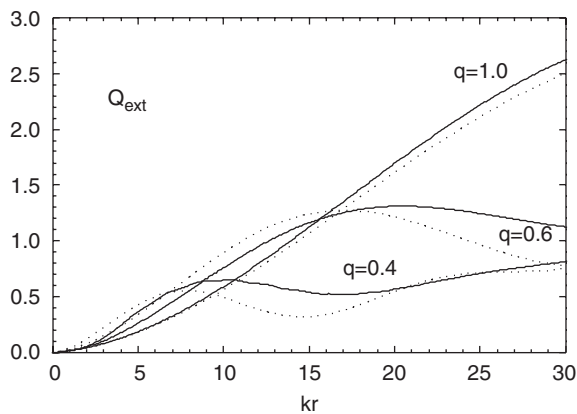


Fig. 2. Extinction efficiency values for coated, spherical particles as a function of particle size. Full line: exact (Aden-Kerker), dotted line: anomalous diffraction approximation.

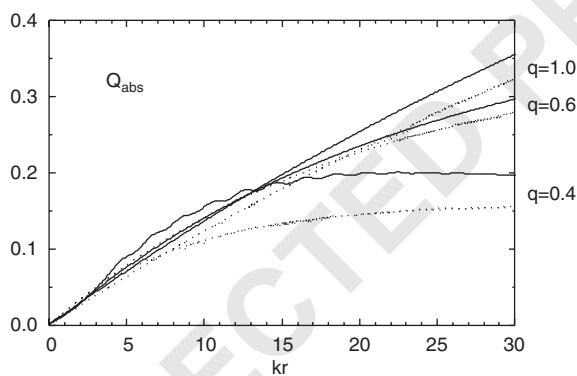


Fig. 3. Absorption efficiency values for coated, spherical particles as a function of particle size. Full line: exact (Aden-Kerker), dotted line: anomalous diffraction approximation.

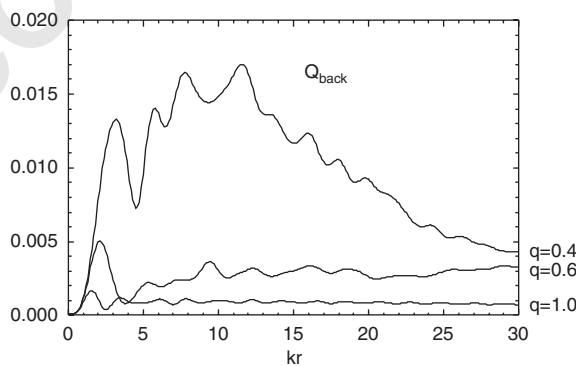


Fig. 4. Backscattering coefficient values for coated, spherical particles as a function of particle size.

1 Table 1  
Core index of refraction

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

9  
11 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  
13  $m_h = 1.05 + i0.005$ , a value usually taken as representative of some biological samples such as  
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  
absorption efficiency curves. This can be done by iteratively using the ADA, since the ADA-  
17 calculated  $Q_{\text{abs}}$  function is monotonous, increasing from 0 to 1 as the particle sizes grows from 0  
to  $\infty$ . 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  
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,  
while not perfect, compares better than the diffraction approximation for homogeneous spheres  
23 ( $q = 1$ ).

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

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  
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  
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  
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  
on the real refractive index of the core.

41 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  
43 coating. In Figs. 6, 7 efficiency values are shown for a coated sphere, an offset sphere (offset

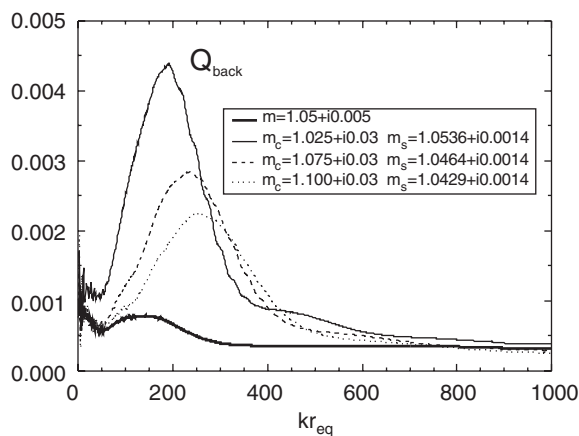


Fig. 5. Backscattering coefficient values for coated particles with core/particle ratio  $q = 0.5$  and several refractive indices within the VA.

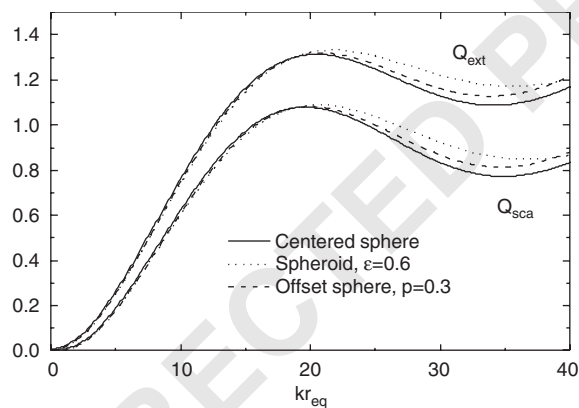


Fig. 6. Extinction and scattering efficiency values for coated particles with core/particle ratio  $q = 0.6$ : center spheres, offset spheres ( $p = 0.3$ ) and centered spheroids ( $\epsilon = 0.6$ ).

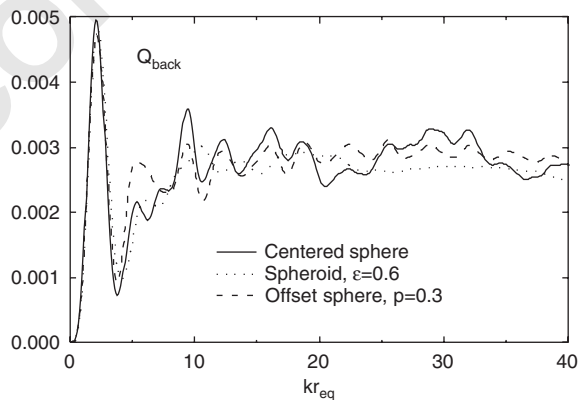


Fig. 7. Same as Fig. 6 for backscattering coefficient values.

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.

## Acknowledgements

Financial support from projects MAT2001-3803 (Ministry of Science and Technology, Spain; FEDER funds, EU) and INTAS (EU) 99-00510 is gratefully acknowledged.

## References

- [1] Preisendorfer RW. Hydrologic optics, vol 5, properties. USA: U.S. Environmental Research Laboratories; 1976.
- [2] Morel A. In-water and remote measurements of ocean color. *Boundary-Layer Meteorol* 1980;18:177–202.
- [3] Mie G. Beiträge zur Optik trüber Medien speziell kolloidaler Metallösungen. *Ann d Phys* 1908;4:377–445.
- [4] Brown OB, Gordon HR. The size-refractive index distribution of clear coastal water particulates from light scattering. *Appl Opt* 1974;13:2874–81.
- [5] Carder KL, Tomlinson RD, Beardsley GF. A technique for the estimation of indices of refraction of marine phytoplankters. *Limnol Oceanogr* 1972;17(6):833–9.
- [6] Zaneveld JRV, Roach DM, Pak H. The determination of the index of refraction distribution of oceanic particulates. *J Geophys Res* 1974;79(27):4091–5.
- [7] Bricaud A, Morel A. Light attenuation and scattering by phytoplanktonic cells: a theoretical modeling. *Appl Opt* 1986;25:571–80.
- [8] Bricaud A, Bédhomme AL, Morel A. Optical properties of diverse phytoplanktonic species: experimental results and theoretical interpretation. *J Plankton Res* 1988;10(5):851–73.
- [9] Stramski D, Morel A, Bricaud A. Modeling the light attenuation and scattering by spherical phytoplanktonic cells: a retrieval of the bulk refractive index. *Appl Opt* 1988;27(19):3954–7.
- [10] van de Hulst HC. *Light scattering by small particles*. New York: Wiley; 1957.
- [11] Morel A, Ahn YH. Optics of heterotrophic nanoflagellates and ciliates: a tentative assessment of their scattering role in oceanic waters compared to those of bacterial and algal cells. *J Mar Res* 1991;49:177–202.
- [12] Maffione RD, Dana DR. Instruments and methods for measuring the backward-scattering coefficient of ocean waters. *Appl Opt* 1997;36:6057–67.
- [13] Mobley CD, Sundman LK, Boss E. Phase function effects on oceanic light fields. *Appl Opt* 2002;41:1035–50.
- [14] Falkowski PG, Raven J. *Aquatic photosynthesis*. Oxford: Blackwell; 1997. p. 375.

- 1 [15] Quinby-Hunt MS, Hunt AJ, Lofftus K, Shapiro D. Polarized-light scattering studies of marine *Chlorella*. *Limnol*  
2 *Oceanogr* 1989;34(8):1587–600.
- 3 [16] Volten H, de Haan JF, Hovenier JW, Schreurs, Vassen RW, Dekker AG, Hoogenboom HJ, Charlton F, Warts R.  
4 Laboratory measurements of angular distributions of light scattered by phytoplankton and silt. *Limnol Oceanogr*  
5 1998;43:1180–97.
- 6 [17] Witkowski K, Wolinski L, Turzynski Z, Gedziorowska D, Zielinski A. The investigation of kinetic growth of  
7 *Chlorella vulgaris* cells by the method of integral and dynamic light scattering. *Limnol Oceanogr*  
8 1993;38(7):1365–72.
- 9 [18] Witkowski K, Krol T, Zielinski A, Kuten E. A light-scattering matrix for unicellular marine phytoplankton.  
10 *Limnol. Oceanogr* 1998;43(5):859–69.
- 11 [19] Bohren CF, Singham SB. Backscattering by nonspherical particles: a review of methods and suggested new  
12 approaches. *J Geophys Res* 1991;96:5269–77.
- 13 [20] Waterman PC. Symmetry, unitarity, and geometry in electromagnetic scattering. *Phys Rev D* 1971;3:825–39.
- 14 [21] Peterson B, Ström S. T-matrix formulation of electromagnetic scattering from multilayered scatterers. *Phys Rev D*  
15 1974;10:2670–84.
- 16 [22] Mishchenko MI, Hovenier JW, Travis LD. Light scattering by nonspherical particles. San Diego: Academic Press;  
17 2000.
- 18 [23] Quirantes A, Delgado AV. Scattering cross sections of randomly oriented coated spheroids. *JQSRT*  
19 2001;70:261–72.
- 20 [24] Bohren CF, Huffman DR. Absorption and scattering of light by small particles. New York: Wiley; 1983.
- 21 [25] Mishchenko MI. Light scattering by randomly oriented axially symmetric particles. *J Opt Soc Am A*  
22 1991;8:871–82[Errata 1992;94:97].
- 23 [26] Barber PW, Hill SC. Light scattering by particles: computational methods. Singapore: World Scientific; 1990.
- 24 [27] Morel A, Bricaud A. Inherent properties of algal cells including picoplankton: theoretical and experimental results.  
25 In: Platt T, Li WKW., editors. *Photosynthetic picoplankton Canadian Bull Fish Aq Sciences*, vol. 214; 1986. p.  
26 521–59.
- 27 [28] Bernard S, Probyn TA, Barlow RG. Measured and modelled optical properties of particulate matter in the  
28 southern Benguela. *S Afr J Sci* 2001;97(9–10):410–20.
- 29 [29] Stramski D, Bricaud A, Morel A. Modeling the inherent optical properties of the ocean based on the detailed  
30 composition of planktonic community. *Appl Opt* 2001;40:2929–45.
- 31 [30] Aas E. Influence of shape and structure on light scattering by marine particles. *Univ Oslo, Inst Rep Series*, vol. 53,  
32 1984. p. 112.
- 33 [31] Zaneveld JRV, Kitchen JC. The variation in the inherent optical properties of phytoplankton near an absorption  
34 peak as determined by various models of cell structure. *J Geophys Res* 1995;100(C7):13,309–20.
- 35 [32] Bricaud A, Zaneveld JRV, Kitchen JC. Backscattering efficiency of coccolithophorids: use of a three-layered  
36 sphere model. In: Gilbert GD., editor. *Ocean Optics XI, Proceedings of SPIE*, vol. 27. New York: Wiley; 1992. p.  
37 27–33.
- 38 [33] Kitchen JC, Zaneveld JRV. A three-layered sphere model of the optical properties of phytoplankton. *Limnol*  
39 *Oceanogr* 1992;37:1680–90.
- 40
- 41
- 42
- 43