Light Scattering by Marine Particles: Modeling with Non-spherical Shapes

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LONG-TERM GOALS

The long-term scientific goal of my research is to better understand the distribution of phytoplankton in the world's oceans through remote sensing their influence on the optical properties of the water. An associated goal is the understanding of the absorption and backscattering properties of marine particles in terms of the distributions of their size, shape, and composition.

OBJECTIVES

The inherent optical properties (IOPs) of marine particles are most-often modeled and interpreted by assuming that they are homogeneous spheres. Although this approach has been fruitful, the next logical step in modeling the IOPs of marine particles is to abandon the normally-employed spherical approximation and use more realistic idealizations of their shape. The advent of computer codes capable of handling more complex shapes, and the increased computational speeds now available, suggest that particle modeling employing simple non-spherical shapes, e.g., disks, rods, etc., could become routine. For example, Gordon and Du (2001) used a two-disk model to try to reproduce the backscattering by coccoliths detached from *E. huxleyi*; however, they found that, while the resulting spectral variation of the backscattering cross section agreed with experiment, its magnitude was low by a factor of 2-3. As simple shapes are still at best poor approximations to real particles, i.e., real particles display significant inhomogeneity, one is led to examine the influence of such inhomogeneities.

It is often assumed that inhomogeneous particles can be replaced by homogeneous particles along with employing an "effective" refractive index; however, my study of thin disks showed the importance of periodic angular fine scale structure on backscattering and demonstrated that if the inhomogeneities are too large, the effective refractive index assumption is invalid and the backscattering increases significantly over that expected based on the effective index hypothesis. I also found that a random perturbation of the purely periodic structure, forming an aperiodic angular fine structure, did not significantly change the backscattering unless the perturbation was severe. Thus, in attempting to model with shapes other than spherical, there are situations in which fine structure, as well as gross morphology, must be considered. The objective of this study is to continue the development of an understanding of how deviations from simple shapes or how non-homogeneity of composition, affect the scattering and absorption of light. The particle considered for verification of the application of complex particle modeling on backscattering is coccoliths detached from the coccolithophorid *E. huxleyi*.

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APPROACH

I use detached coccoliths from the coccolithophorid *E. huxleyi* (Figure 1) as a case study for applying non-spherical shapes to the computation of backscattering. This particle was chosen because (1) its composition is known and homogeneous (refractive index $m \approx 1.20$), (2) its gross shape is known (it resembles a disk or two parallel disks), (3) its optical properties are known, and (4) it has a complex, quasi-periodic internal structure on a sub-visible-wavelength scale. Gordon (2006) focused on the quasi-periodic structure of the top disk (in Figure 1) and on the influence of the curvature of the lower plate. The influence of the curvature was found to be small and will not be discussed here.

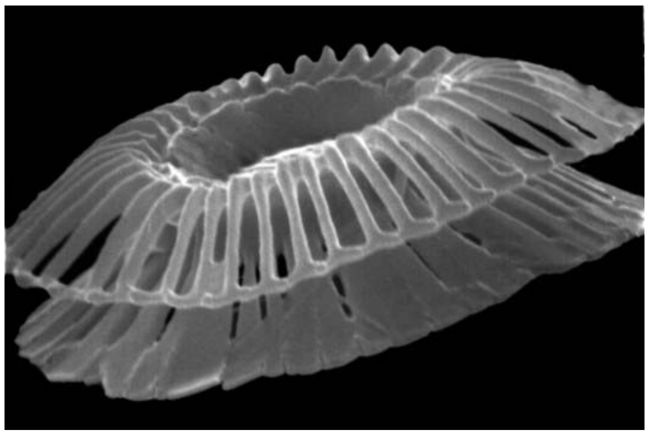

Figure 1: SEM image of coccolith a detached from E. huxleyi showing a structure grossly resembling two parallel disk-like plates. The upper plate (the "distal" shield) resembles a wheel with spokes. The spoke separation is 50 to 150 nm.

I use the term "gross morphology" to indicate a smooth homogeneous particle having approximately the same overall shape as the particle in question (e.g., a single disk or two parallel disks as a model for a detached coccolith). I use "fine-scale structure" to indicate deviations from the gross morphology (e.g., the coccolith's periodic radial structures resembling the openings between the spokes of a wagon wheel). My goal is to understand how the fine-scale structure can induce deviations in the backscattering characteristic of a given gross morphology. Because the physical example of interest is a coccolith, I have, for the most part, limited the examination to particles with the gross morphology of a disk or a combination of two disks.

To investigate the influence of periodic fine structure in a disk-like object on backscattering, I started with a homogeneous disk and removed sectors. Specifically, the disk was divided into equal-angle sectors of angle $\Delta\alpha$, and alternate sectors were removed. The angle $\Delta\alpha$ was given by $\Delta\alpha = 2\pi/2^n$, where n is an integer. I refer to these objects as "pinwheels." If we let s be the arc length of the open (or closed) regions at the perimeter of the pinwheel, then $s = D\Delta\alpha/2$, where D is the diameter of the disk . The values of s for the various cases that I examined were such that at a wavelength (λ) of 400 nm in vacuum (300 nm in water, λ_{Water}), as n progresses from 4 to 7, s takes on the values λ_{Water}, $\lambda_{\mathrm{Water}}/2$, $\lambda_{\mathrm{Water}}/4$, and $\lambda_{\mathrm{Water}}/8$.

I computed the backscattering cross section σ_b of pinwheels using the discrete-dipole approximation (Draine,1988; Draine and Flatau, 1994). I found that the behavior of the backscattering was determined by the ratio $s/\lambda_{\mathrm{Water}}$. The backscattering cross section was nearly identical to that of a homogeneous disk of similar size (but with m reduced from 1.20 to $m_{\mathrm{eff}} = 1.10$) as long as $s/\lambda_{\mathrm{Water}} < 0.25$. In addition, pinwheels satisfying this criterion also scattered in the same manner as disks having half of the mass removed from random locations within the disk. Thus, when $s/\lambda_{\mathrm{Water}} < 0.25$, there is essentially no difference in backscattering between the periodic structure and a structure of small random voids (in disk-like particles) with the same total mass. In this regime the backscattering is totally governed by the particle's gross morphology and effective index (m_{eff}). For $s/\lambda_{\mathrm{Water}} > 0.25$ departures from a homogeneous disk are observed and manifest as a significant increase (many times) in backscattering: the fine-scale structure is as important as the gross morphology in this regime. The spokes of the distal shield of the coccolith in Figure 1 have spacing that would place the shield in the $s/\lambda_{\mathrm{Water}} > 0.25$ regime.

I then extended (Gordon, 2007b) the computations to sectorized disks with *aperiodic* structure to try to understand the influence of non-uniform spacing of the spokes in the distal shield of the coccolith (Figure 1). This was effected through a perturbation of the purely periodic pinwheel effected in the following manner. First, as before, the disk is divided into purely periodic sectors, the angular boundaries of which are designated by the 2^n angles α_P. The individual boundary angles are then perturbed to α_I according to

$$\alpha_I = \alpha_P + \varepsilon \frac{2\pi}{2^n} \rho ,$$

where $0 \leq \varepsilon \leq 1$ is a constant and $-1/2 \leq \rho \leq 1/2$ is a random number with a uniform probability density. Then, the material of the disk is removed from every other sector, yielding a pinwheel with a quasi-periodic structure. Defining σ_I to be the standard deviation in the angle α_I, $\sigma_I = 12^{-1/2} \varepsilon \Delta\alpha \approx 0.3\varepsilon \Delta\alpha$, where $\Delta\alpha = 2\pi/2^n$. Likewise defining $\sigma_{\Delta\alpha}$ to be the standard deviation of the removed (or occupied) sector angles, $\sigma_{\Delta\alpha} = 6^{-1/2} \varepsilon \Delta\alpha \approx 0.4\varepsilon \Delta\alpha$. Thus, for $\varepsilon = 0.5$ and 1.0, the relative standard deviation in angle of the removed (or occupied) sectors is 20 and 40%, respectively. The computations revealed that, for coccolith-sized disks, as long as $\varepsilon \leq 1$ there was only a small difference (few %) between the aperiodic and periodic pinwheels.

I examined the deviation in the angular spacing of the "spokes" in the distal shield of the individual coccoliths provided in Fig. 2 of Gordon (2006). For this particular coccolith, $\sigma_{\Delta\alpha}/\Delta\alpha \sim 0.27$, and there were 40 open angular sectors. This coccolith shield is similar in size and shape to a 2.75 μm pinwheel I examined ($n = 6$, 32 open sectors). As the computations for that pinwheel showed that for the purpose of computing backscattering, the periodic pinwheel is a good approximation to aperiodic pinwheel as long as $\sigma_{\Delta\alpha}/\Delta\alpha \leq 0.4$ ($\varepsilon \leq 1$), they suggest that replacing the aperiodic fine structure of the distal shield of *E. huxleyi* coccoliths with a strictly periodic fine structure will not degrade the modeling of their backscattering.

WORK COMPLETED

The approach described above sets the stage for a refined physical model of a detached coccolith for the purpose of computing light scattering properties, based on the initial work of Gordon and Du (2001). I set out to make an improved light scattering model of a detached coccolith. My previous work (Gordon, 2006) suggested that the open spaces in the proximal shield (that near the interior of the

E. huxleyi coccolithphore) of a coccolith can be handled by assigning an effective (smaller) refractive index than would be assumed for a solid disk. In addition, the computations in Gordon (2007b) suggest that, although the spacing between the "spokes" of the distal shield is aperiodic, this shield can be modeled as a pinwheel with purely periodic spacing. A schematic of the model coccolith and the associated parameters is provided in Figure 2.

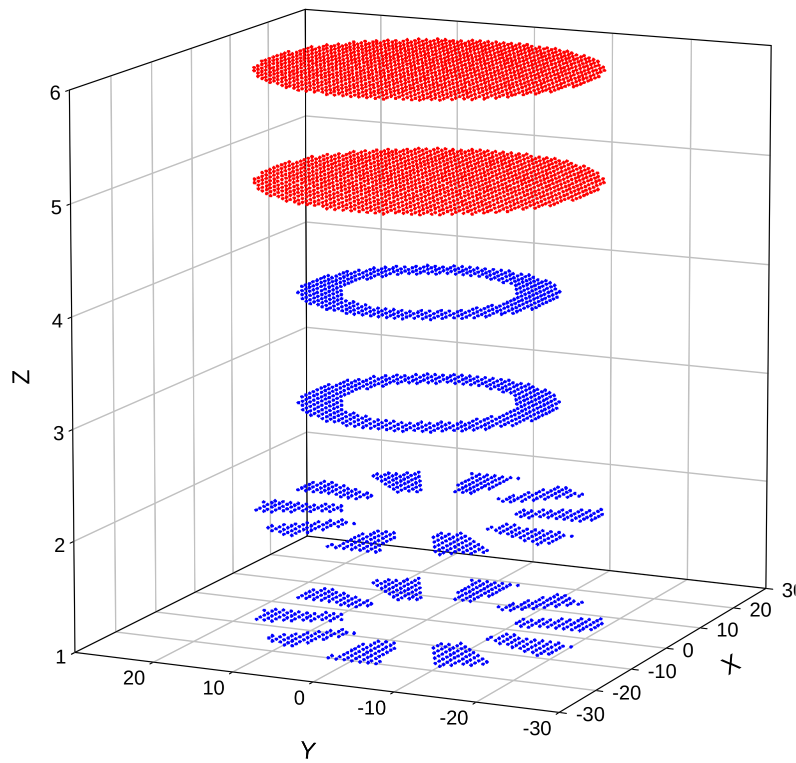

Figure 2: Schematic of the model of the position of the individual dipoles comprising the body of a detached coccolith. The top (red) disks are the proximal shield. They have a diameter of 3.5 μm. The thickness of this disk ranges from 0.04 to 0.06 μm. The two central "washer-shaped" disks represent the cylinder joining the proximal and distal shields. This cylinder has an inner diameter of 1.38 μm and an outer diameter of 1.58 μm. The "pinwheel" at the bottom represents the distal shield, and is shown here with 10 "vanes." In the actual coccolith model, the distal shield has 40 "vanes." The separation between the distal and proximal shields is 0.30 to 0.35 μm. In the figure, the blue objects have a refractive index of 1.20, while the red objects have an index of 1.19. The maximum number of layers (6 are shown here) of dipoles in the actual coccolith model is up to 24, corresponding to a layer spacing of ~ 0.02 μm.

The geometrical parameters of the physical model were determined based on the measurements provided in Young and Ziveri (2000). They give a mean diameter of coccoliths of 3.5 μm, and a mean volume of 0.9 μm^3. I modeled the coccoliths as two disks (one uniform and one with a pinwheel structure) separated by a hollow cylinder. I used three disk thickness (0.04, 0.05, and 0.06 μm) and three thicknesses for the gap between the disks (0.300, 0.325, and 0.350 μm, which is also the length of the "cylinder") for a total of 9 separate models. The scattering properties were determined for each model and an average over models was taken. The average coccolith volume was 0.835 μm^3, ~ 7% less than the Young and Ziveri (2000) average.

In addition to refining the coccolith model, I started a new investigation into the backscattering of cylindrical particles (finite length) in random orientation. My hypothesis is that, for a given cylinder diameter and refractive index, as the length of the cylinder increased, the backscattering probability ($\sigma_{back}/\sigma_{total}$) would approach an asymptotic value – that of an infinite cylinder. My goal was to determine the length (or the aspect ratio) above which one could use the infinitely-long cylinder computations for the backscattering probability (for which a Mie-like solution is available, e.g., van de Hulst, 1957).

RESULTS

Figure 3 compares the averaged backscattering cross section for the modeled coccoliths with measurements inferred from radiance/irradiance profiles obtained from a bloom off Plymouth, UK (Gordon, 2004).

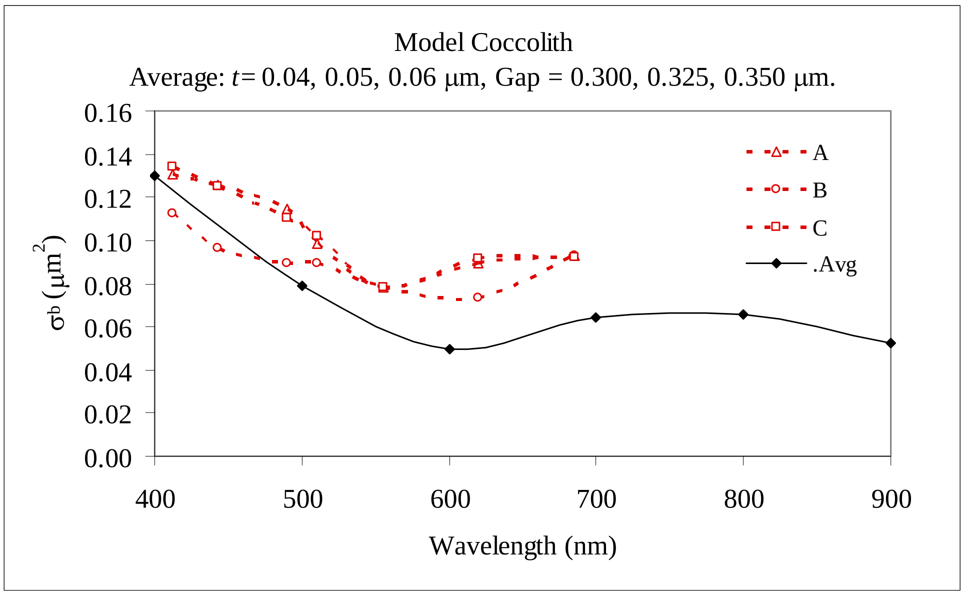

Figure 3: The average backscattering cross section of model coccoliths in random orientation. The red curves are retrieved spectra from a bloom off Plymouth, UK. They are normalized to 0.078 μm^2 per coccolith at 550 nm. This normalization was observed by regressing σ_b(550) against coccolith number as determined by microscope counts (about twice that determined by flow cytometry). "A", "B", and "C" refer to retrievals from 3 distinct stations.

The σ_b spectra show reasonable agreement with the measurements, but the modeled spectra again appear to be a little too low; however, one must keep in mind that, although detached coccoliths far outnumber other particles, they are not the only backscatterers in the water. Backscattering by other constituents will increase the apparent coccolith-specific backscattering.

Figure 4 compares the averaged phase function at 500 nm with the measurements reported in Voss et al. (1998). The estimated phase function and the model differ by no more than a factor of 2. A paper providing the details of the measurements and the model is in preparation.

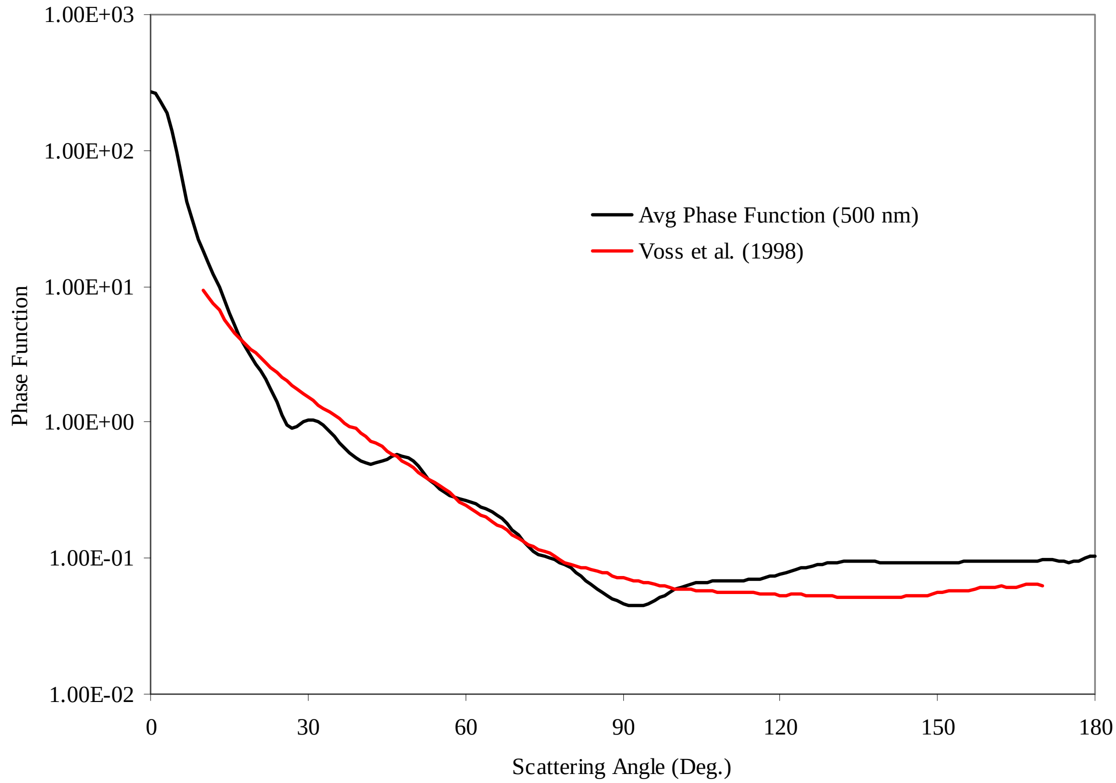

Figure 4: The computed average scattering phase function at 500 nm (black curve), compared with that estimated from the measurements of Voss et al. (1998). As Voss et al. were unable to measure the total scattering coefficient, their estimate provides only the angular shape of the phase function. Their estimate is multiplied by an arbitrary normalization for comparison of the measured shape with the model.

The results of the computation of the backscattering by cylinders of finite length are provided in Figure 5. Specifically, Figure 5 provides a sample of the computations for a cylinder with a diameter of 1 μm for various refractive indices and lengths (λ = 400 nm). Focusing on the red curves ($\sigma_{back}/\sigma_{total}$) on sees that in 3 of the 4 cases, it appears that the backscattering probability reaches an asymptotic value near an aspect ratio (length/diameter) of ~ 5. This appears to be true in the fourth case as well (m = 1.05); however, for this case, as the length is increased beyond ~ 15 μm, the backscattering increases dramatically. This increase is clearly non-physical, and an error in the computations produced by the

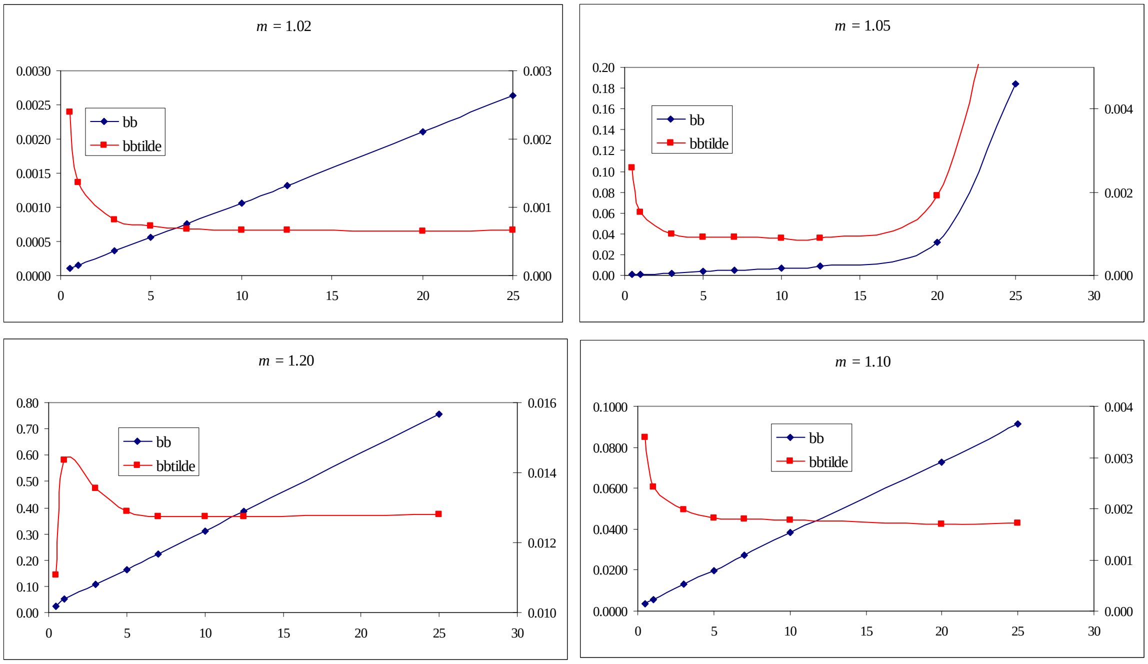

Figure 5: These figures provide the backscattering cross section (blue curves, left "y"-axis) and the backscattering probability (red curves, right "y"-axis) in μm^2 for a right circular cylinder with a diameter of 1 μm as a function of its length ("x"-axis) in μm. The individual panels are for refractive indices (clockwise from the upper left) of 1.02, 1.05, 1.10, and 1.20. Note the anomalous behavior of backscattering for length greater than about 15 μm for an index of 1.05.

discrete-dipole approximation (DDA, Draine, 1988) is believed to be the cause. I investigated the behavior of the backscattering near m = 1.05 and found that this behavior is restricted to the vicinity of this index, and furthermore, that the extinction coefficient became negative for light incident on the particle parallel to the symmetry axis. I have informed Professor Draine of this development, and plan to investigate the range of parameters more fully to try to understand the source of the problem.

IMPACT/APPLICATIONS

The questions examined in this report are of fundamental importance in light scattering modeling. The complexity of the non-spherical particle models that are required to reproduce the IOPs of particles in natural waters provides the complexity of models required for the interpretation of the IOPs in terms of particle characteristics.

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