Retrieving composition and sizes of oceanic particle subpopulations from the volume scattering function

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For a particle population with known size, composition, structure, and shape distributions, its volume scattering function (VSF) can be estimated from first principles through a governing relationship, the Fredholm linear integral equation of the first kind. Inverting the Fredholm equation to derive the composition and size distribution of particles from measured VSFs remains challenging because 1) the solution depends on the kernel function, and 2) the kernel function needs to be constructed to avoid singularity. In this study, a thorough review of the earlier and current inversion techniques is provided. An inversion method based on nonnegative least squares is presented and evaluated using the VSFs measured by a prototype volume scattering meter at the LEO-15 site off the New Jersey coast. The kernel function was built by a compilation of individual subpopulations, each of which follows a lognormal size distribution and whose characteristic size and refractive index altogether cover the entire ranges of natural variability of potential marine particles of the region. Sensitivity analyses were conducted to ensure the kernel function being constructed is neither singular nor pathological. A total of 126 potential subpopulations were identified, among which 11 are common in more than half of the inversions and only five consistently present (>90% of measurements). These five subpopulations can be interpreted as small colloidal type particles of sizes around 0.02 μm, submicrometer detritus-type particles (n_r = 1.02, r_mode = 0.2 μm), two micrometer-sized subpopulations with one relatively soft (n_r = 1.04 and r_mode = 1.6 μm) and the other relatively refringent (n_r = 1.10 and r_mode = 3.2 μm), and bubbles of relatively large sizes (n_r = 0.75 and r_mode = 10 μm). Reconstructed PSDs feature a bimodal shape, with the smaller peak dominated by the colloidal subpopulations and the larger particles closely approximated by a power-law function. The Junge-type slope averages −4.0 ± 0.2, in close agreement with the well-known mean value of −4.0 over the global ocean. The distribution of the refractive index suggested a dominance of particles of higher water content, also in agreement with earlier results based on the backscattering ratio and attenuation coefficients at the same area. Surprisingly, the colloidal-type subpopulations, which have often been operationally classified as “dissolved” and neglected for their scattering, exhibit significant backscattering with contributions of up to 40% over the entire backward angles. © 2011 Optical Society of America

1. Introduction

One of the motivations for optical observations in the ocean has been the quest for a quantitative relationship between the light scattering and the physical properties of the particles suspended in the aqueous medium. It is theoretically possible to calculate from first principles all of the aspects of scattering from a sample of water with known distributions of size, shape, internal structure, and the refractive index for particles contained. As an inherent optical property (IOP), the volume scattering function (VSF, ) at a scattering angle of θ can be calculated as a sum
of contributions from the particle groups (or subpopulations):

\[
\beta(\theta) = \sum_{i=1}^{M} b_i(n(r), F(r), G(r), S(r)) \beta_i(n(r), F(r), G(r), S(r), \theta).
\]  

(1a)

Here, we have assumed that there are \( M \) particle subpopulations, and for each of them, the total scattering coefficient \( (b_i) \) and the VSF \( (b_i, \beta_i) \) or the phase function \( (\beta_i) \) are determined by the distributions of particle refractive index \( (n(r)) \), concentration \( (F(r)) \), internal structure \( (G(r)) \), and shape \( (S(r)) \), each of which, in turn, is a function of particle size \( (r) \).

It remains challenging to determine index–shape–structure–number–size spectra for marine particles in their natural environment. Inverting Eq. (1) to determine particle properties from light scattering offers promising potential in tackling this challenge, even though it is much more complex than the forward process.

Equation (1), written in integral form,

\[
\beta(\theta) = \int_{r_{\text{min}}}^{r_{\text{max}}} b(n(r), F(r), G(r), S(r)) \beta(n(r), F(r), G(r), S(r), \theta) dr,
\]  

(1b)

is the classic Fredholm linear integral equation of the first kind and \( \beta \) is the kernel function \( [1] \). To derive the optical and size properties of particles is to invert Eq. (1) using the measured VSF, \( \beta \). The common method of inversion is to estimate the kernel function, i.e., a number of phase functions \( \beta_i \) in this case, and to find a set of values of \( b_i \) that provides the closest fit to the observed \( \beta \) \([2]\). The index of refraction \( (n) \) and the particle size distribution (PSD) corresponding to the resulting VSF, \( b_i\beta_i \), are assumed to be representative for the particle subpopulation that exhibits this specific \( b_i\beta_i \). To accommodate the theoretical computation of the kernel phase functions, several simplified assumptions have to be made regarding the characteristics of oceanic particles. For example, the size continuum has to be segmented and bounded using a minimum and maximum radii \( (r_{\text{min}} \text{ and } r_{\text{max}}, \text{ respectively}) \), and the shape of the size distribution of the subfraction is often assumed to be known \( a \text{ priori} \). Also, in order to use Mie theory, particles are assumed to be uniform and spherical.

Depending on the assumptions being made, solutions could differ from each other even though the same measurements were used. Therefore, independent verifications are highly desirable. Because of this challenge, as well as the scarcity of data, only a few studies have been conducted attempting to retrieve particle properties from the measurements of the VSF in the upper ocean.

2. Basic Definitions

For a given wavelength \( (\lambda) \) of incident light, a particle’s size \( (r) \), structure \( (G) \), shape \( (S) \), and refractive index relative to the medium \( (n) \) uniquely determine the scattering and absorption properties of the particle. The total scattering cross section \( (C_{\text{sca}}, \text{ m}^2) \), is defined as

\[
C_{\text{sca}} = \int_{4\pi} C_{\text{ang}}(\Omega) d\Omega = 2\pi \int_0^{\pi} C_{\text{ang}}(\theta) \sin \theta d\theta,
\]  

(2)

where \( C_{\text{ang}} \) \( (\text{m}^2 \text{ sr}^{-1}) \) is the angular scattering cross section and \( \Omega \) is the solid angle indicating the direction of scattering. Under azimuthal symmetry, \( \Omega \) depends only on \( \theta \), the angle between the directions of scattered and incident light. The backscattering cross section \( (C_{\text{bsc}}, \text{ m}^2) \) and the backscattering ratio \( (\bar{b}_b) \) are defined as

\[
C_{\text{bsc}} = 2\pi \int_{\pi/2}^{\pi} C_{\text{ang}}(\theta) \sin \theta d\theta,
\]  

(3)

\[
\bar{b}_b = \frac{C_{\text{bsc}}}{C_{\text{sca}}},
\]  

(4)

respectively. The backscattering ratio is often used as an indicator of the overall shape of the angular scattering. Let \( F(r) dr \) \( (\text{m}^{-3}) \) denote the PSD with sizes between \( r \) and \( r + dr \). The phase function for such a particle population,

\[
\beta(\theta, n, G, S, F) = \frac{\beta(\theta, n, G, S, F)}{b(\theta, n, G, S, F)},
\]  

(5)

where the VSF \( \beta (\text{m}^{-1} \text{ sr}^{-1}) \) and the total scattering coefficient \( b (\text{m}^{-1}) \) can be calculated, respectively, as

\[
\beta(\theta, n, G, S, F) = \int_{r_{\text{min}}}^{r_{\text{max}}} C_{\text{ang}}(\theta, r, n, S) F(r) dr,
\]  

(6)

\[
b(n, G, S, F) = \int_{r_{\text{min}}}^{r_{\text{max}}} C_{\text{sca}}(r, n, S) F(r) dr.
\]  

(7)

The minimum and the maximum radii values have to be introduced in a practical sense for computation. However, their values should be chosen such that the integrations in Eqs. (6) and (7) do not depend on them, because the VSF and the total scattering coefficient are physical quantities with finite values.

Operationally, the VSF for a sample of volume \( dV \) is defined as

\[
\beta(\theta) = \frac{dI(\theta)}{EdV},
\]  

(8)

where \( dI(\theta) (\text{sr}^{-1}) \) is the radiant intensity of light scattered at angle \( \theta \) given the incident irradiance.
$E$ (W m$^{-2}$). Note that, by definition, both $I$ and $E$ are unpolarized.

In general, two types of PSDs are considered here: one where the concentration of particles continues to increase with decreasing sizes and another that peaks at a certain size. We represent the former with the commonly used power-law function:

$$F(r) = N_0 \frac{1 - s}{r_{\text{max}} - r_{\text{min}}} r^{-s},$$

(9)

where $N_0$ is the total number of particles of sizes between $r_{\text{min}}$ and $r_{\text{max}}$ in a unit volume and $s$ is the slope or Junge exponent for the power distribution. Equation (9) is sometimes referred to as Junge-type distribution. We represent the peaked distribution with the commonly used lognormal function:

$$F(r) = N_0 \frac{\exp \left( -\frac{r^2}{2} \right)}{\sqrt{2\pi\sigma^2}r_{\text{mode}}} \exp \left( -\frac{(\ln(r) - \ln(r_{\text{mode}}))^2}{2\sigma^2} \right),$$

(10)

where $r_{\text{mode}}$ represents the location of the mode of the distribution and $\sigma$ is the standard deviation. For both distributions, $\int_{r_{\text{min}}}^{r_{\text{max}}} F(r) dr = N_0$. Phase functions do not depend on $N_0$.

There are other types of mathematical representations proposed for PSDs, such as, for example, exponential functions for the monotonic distribution or Gamma (or Weibull) functions for the peaked distribution. However, their effects are secondary as compared to the major differences of these two groups.

3. Review

Compared to the other IOPs, measurements of the VSFs have been scarce and mostly were conducted during the 1960s and 1970s. A detailed analysis of these early experiments can be found in Morel [3]. Recently, the development of a custom volume scattering meter (VSM) [4], a custom Multi-Angle Scattering Optical Tool (MASCOT) (5, 6), and other commercial VSF sensors, such as Laser In Situ Scattering and Transmissiometry (LISST-100X, Sequoia Scientific, Inc.), [7] has led to a renewed interest in observing angular scattering in the upper ocean [8–12]. Only a few studies have been conducted attempting to retrieve particle properties from measurements of the VSF and Table 1 summarizes the results of these efforts. A common feature of these analysis is that a power-law relationship [Eq. (9)] was assumed (or used) to represent the PSD.

Kullenberg [13] measured VSFs in the Sargasso Sea using two separate instruments: a laser-based device measuring perpendicularly polarized scattering at 1°, 2.5°, 3.5°, 25°, 30°, 45°, 60°, and 90° at 632.8 nm, and a “β-meter” measuring unpolarized scattering from 10° to 165° with an interval of 15° at two wavelengths of 460 and 655 nm. The measurements by the laser instrument were made from the surface to a 400 m depth, whereas the β-meter was lowered to only one depth of 10–15 m. Despite a wider angular range for the β-meter, which covered important backward angles, the laser measurements were used in all the studies that are referred to as

<table>
<thead>
<tr>
<th>Study</th>
<th>VSFs</th>
<th>Index ($n$)</th>
<th>PSD</th>
<th>$b_p$, $b_{\text{sub}}$, $b_{\text{io}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Kullenberg [14]</td>
<td>Sargasso Sea</td>
<td>1.20</td>
<td>$r^{-3}$, 1.5 r ≤ 20</td>
<td>0.032, 0, 1.0</td>
</tr>
<tr>
<td>Kullenberg [14]</td>
<td>Baltic Sea</td>
<td>1.04–1.06</td>
<td>$r^{-2.25}$, 1 ≤ r ≤ 20</td>
<td>N/A</td>
</tr>
<tr>
<td>Kullenberg [14]</td>
<td>Mediterranean</td>
<td>1.20</td>
<td>$r^{-2.25}$, 1 ≤ r ≤ 20</td>
<td>N/A</td>
</tr>
<tr>
<td>Gordon and Brown  [15]</td>
<td>Sargasso Sea</td>
<td>1.05 ± 0.017</td>
<td>$r^{-4}$, 0.04 ≤ r ≤ 5</td>
<td>0.023, 0.38, 0</td>
</tr>
<tr>
<td>Brown and Gordon  [16]</td>
<td>Sargasso Sea</td>
<td>1.01 ± 0.017</td>
<td>$r^{-4}$, 0.05 ≤ r ≤ 1.3</td>
<td>0.025, 0.04, 0.95</td>
</tr>
<tr>
<td>Brown and Gordon  [22]</td>
<td>Tongue Of The Ocean (TOTO)</td>
<td>1.01</td>
<td>$r^{-5}$, 0.006 ≤ r ≤ 0.33</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.01</td>
<td>$r^{-7.5}$, 0.33 ≤ r ≤ 0.63</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.15 &amp; 1.01</td>
<td>$r^{-4}$, 0.63 ≤ r ≤ 1.9</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.03 ± 0.017</td>
<td>$r^{-7.5}$, 1.9 ≤ r ≤ 9.0</td>
<td>N/A</td>
</tr>
<tr>
<td>Jonasz and Prandke [23]</td>
<td>Baltic Sea (winter)</td>
<td>1.1</td>
<td>$r^{-4.1}$, 0.05 ≤ r ≤ 1</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.1</td>
<td>$r^{-3.41}$, 1 ≤ r ≤ 4.3</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.1</td>
<td>$r^{-4.86}$, 4.3 ≤ r ≤ 16.5</td>
<td>N/A</td>
</tr>
<tr>
<td>Jonasz and Prandke [23]</td>
<td>Baltic Sea (summer)</td>
<td>1.1</td>
<td>$r^{-4.1}$, 0.05 ≤ r ≤ 1</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.05 ± 0.005</td>
<td>$r^{-2.73}$, 1.0 ≤ r ≤ 4.3</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.03 ± 0.017</td>
<td>$r^{-4.86}$, 4.3 ≤ r ≤ 16.5</td>
<td>N/A</td>
</tr>
<tr>
<td>Zaneveld et al. [26]</td>
<td>Sargasso Sea</td>
<td>1.05</td>
<td>$r^{-3.5}$, 0.02 ≤ r ≤ 20</td>
<td>0.028, 0.54, 0.62</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1.15</td>
<td>$r^{-3.5}$, 0.02 ≤ r ≤ 20</td>
<td>N/A</td>
</tr>
<tr>
<td>Morel [3]</td>
<td>Global mean VSF</td>
<td>1.05</td>
<td>$r^{-4}$, 0.02 ≤ r ≤ 20</td>
<td>N/A</td>
</tr>
<tr>
<td>Schoonmaker et al. [24]</td>
<td>San Diego Bay</td>
<td>1.2 + 0.015</td>
<td>$r^{-3.1}$, 0.05 ≤ r ≤ 50</td>
<td>N/A</td>
</tr>
<tr>
<td>Zhang et al. 2002 [9]</td>
<td>Coastal water off New Jersey</td>
<td>1.05 ± 0.002</td>
<td>$r^{-4}$, 0.4 ≤ r ≤ 325</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0.75</td>
<td>$r^{-4}$, 0.1 ≤ r ≤ 325</td>
<td>N/A</td>
</tr>
</tbody>
</table>

*In the last column, $b_p$, total particulate scattering coefficient (m$^{-1}$); $b_{\text{sub}}$, fractional contribution by submicrometer particles; and $b_{\text{io}}$, fractional contribution by inorganic particles, were estimated only for studies using Sargasso Sea VSF data.
“Sargasso Sea” in Table 1, probably because of measurements of scattering at small angles associated with the laser measurements. The total particulate scattering measured for the Sargasso Sea was 0.023 m⁻¹. We should point out that Kullenberg’s VSFs measured by the laser were not true VSFs by definition, because the incident irradiance $E$ [Eq. (8)] was polarized. But because the measurements with the laser were made in the dominant scattering polarization plane, the overall shapes of the VSFs by the two instruments are fairly consistent (Fig. 1). We can expect the laser scattering results to be only 10%–20% less than the actual unpolarized VSF near the region of greatest depolarization around 90° and exactly 50% less near 0 and 180°, where scattering in both the parallel and perpendicular planes will be equivalent. The overall shape is thus slightly flatter than the true VSF, although this effect is minor considering a maximum error of a factor of 2 for a single VSF spanning about 4 orders of magnitude. Thus, the results based on the laser scattering VSFs by Kullenberg [14], Gordon and Brown [15], Brown and Gordon [16], and Zaneveld and Pak [17], and particularly their respective methodology and the intercomparison of their results, should still be considered valid.

By fixing the sizes of particles between 1 and 20 μm and assuming spherical shapes (i.e., Mie theory applicable), Kullenberg [14] found the best fit with a population of inorganic particles ($n = 1.20$) with $s \leq 3$ to his Sargasso Sea laser measurements. The exponent, $s$, determined for Sargasso Sea was less steep than the Coulter Counter determinations (3.4–4.2) in the same area [15]. Kullenberg [14] also applied the same approach to the measurements from the Baltic and Mediterranean seas and obtained similar values for $s$ (∼2.2) for both locations. Overall, the derived PSDs suggested the presence of a significant portion of large particles. Any $s < 3$ implies that the calculated optical properties (including VSF) would depend on the value of $r_{max}$ [3]. For example, with the same PSD that Kullenberg derived, but increasing the terminal size from 20 to 40 μm, the scattering coefficient would increase from 0.032 to 0.039 m⁻¹, an increase of 20%. It is not clear how Kullenberg’s upper limit was determined.

Using the same laser measurements from the Sargasso Sea but constraining $s$ to values between 3.4 and 4.2 and concentrations of particles larger than 1 μm ranging from $12 \times 10^3$ to $20 \times 10^3$ m⁻¹, both of which were inferred from Coulter Counter sizing of particles, Gordon and Brown [15] arrived at a different conclusion with $n = 1.05 + 0.01i$ and $s = 4$. It should be pointed out that the deployments of the laser device and the Coulter Counter were two years apart. Their results suggested the existence of a large component of organic particles (presumably of lower index due to watery protoplasm) in the suspension, consistent with a study by Manheim et al. [18], who found that, in the offshore surface waters along the east coast of the United States, suspended matter is chiefly organic particles. Using Gordon and Brown’s results, we estimate that about 38% of total scattering was due to submicrometer particles, in agreement with inferences from an earlier field observation [19] that a significant fraction (∼1/2) of the total scattering appears to be derived from particles smaller than 1 μm and with theoretical prediction based on Mie theory [20]. But recent measurements in oligotrophic north Pacific gyre showed that submicrometer particles can only account for ∼15% of total scattering [21].

Recognizing that assigning the same refractive index to all the particles in the suspension, while simple, is not realistic, Brown and Gordon [16] proposed a two-component model but with the same constraints derived from the Coulter Counter observations. They used the two-component model to explain their early one-component model results as a mixture of smaller organic particles ($n = 1.01 + 0.01i$, $r < 1.3 \mu m$) and larger inorganic particles ($n = 1.15$, $r > 1.3 \mu m$). This particle distribution led to a predominance of scattering by large inorganic particles (95%), even though approximately 70% of the volume of particles is organic. They recognized that a good fit can be obtained even if the contribution of the low index organic particles of less than 1 μm is completely ignored, which accounts for only 4% of particulate scattering. This contradicts their earlier one-component model result that found about 38% of scattering was due to submicrometer particles. Also, the midpoint size separating organic and mineral particles is hard to explain.

Brown and Gordon [22] improved over their earlier studies by using a three-component model with simultaneous measurements of PSDs and VSFs in the Tongue of the Ocean, Bahama Islands. The Coulter Counter measurements indicated that the particles

![Fig. 1. Kullenberg’s Sargasso Sea particulate (SSP) VSFs at 632.8 nm (SSP 632.8 nm) by the laser instrument and at 460 nm (SSP 460 nm) and 655 nm (SSP 655 nm) by the $β$-meter and the derived VSFs by Gordon and Brown (GB1972) [15], Brown and Gordon (BG1973) [16], Zaneveld et al. (Z1974) [26], and Kullenberg (K1974) [14].](image-url)
followed a three-fraction distribution (0.33–0.63, 0.63–1.9, and 1.9–9.0 μm), with steep slopes (s ~ 7.5) for the small and large fractions and moderate slope (s ~ 4) for the middle section. Also measured were the linear polarized components of the VSFs, the total attenuation, and the absorption coefficients, all at 488 nm. The angular scattering at 45° was measured at two additional wavelengths of 436 and 546 nm.

By assuming each size fraction was represented by one type of particle, they found a best fit could be obtained by n = 1.01 for the small fraction, n = 1.15 for the middle fraction, and any index for the large fraction as long as n < 1.15. They chose n = 1.03 + 0.01i because it provided increased scattering for angles <2° without contributing strongly to the total scattering. Similar to the two-component model, most scattering except at angles <5° was due to scattering by minerals in the midsize section. While this three-component model could reproduce the measured VSF within 20%, it did not give the correct spectral variation of scattering at 45°, which had a ratio of 1.20 (436 versus 546 nm). And the only way to reproduce the observed spectral variation was to include smaller particles. In an effort to extend size below 0.33 μm, they found multiple solutions were possible. The best one is organic particles (n = 1.01) following a Junge slope of −5 to 0.006 μm, which not only reproduced the spectral variation observed for scattering at 45° but also agreed with measured Mueller matrix component M_33.

Similar to Brown and Gordon [22], Jonasz and Prandke [23] applied a three-component model to the inversion of the VSF measurements at angles from 5° to 165° in the Baltic Sea. The Coulter Counter measurements provided the estimate of PSD for the midfraction (1 < r < 4.3 μm) and the large fraction (4.3 < r < 16.5 μm) and they assumed a presence of small fraction (0.05 < r < 1 μm). All fractions followed Junge distribution. Jonasz and Prandke [23] derived these parameters from the inversion of the measured VSFs: n and s for the small fraction and n for the midfraction and large fraction. They found s = 4.1 for the small fraction in both winter and summer, n = 1.1 for all fractions in winter and the small fraction in summer, n = 1.05 + 0.005i for the midfraction and 1.03 + 0.01i for the large fraction in summer. The derived variations in particles seemed to suggest more organic particles in summer than in winter, in agreement with concurrent chlorophyll a data. The results also suggested that particles with r < 0.25 μm contribute significantly to the VSFs at angles greater than 10°: about 55% at 45° and up to ~80% at 150°.

Morel [3] applied a one-component model to the averaged values of VSFs measured in the open oceans and found the best fit was by a particle population with n = 1.05, s = 4, and r = 0.02–20 μm, a result very similar to the Gordon and Brown [15] one-component model with n = 1.05 + 0.01i, s = 4, and r = 0.04–5 μm. Morel [3] also commented that the agreement would hold well for the neighboring values of refractive index and exponent.

A one-component model was also used by Schoonmaker et al. [24] to fit Petzold’s measurements of the VSF in San Diego Bay [25]. In contrast to the open ocean cases of a one-component model, where particles were of organic origin with n = 1.05 [3, 15], the particles that gave the best fit for the turbid coastal water are of inorganic origin with n = 1.2 + 0.015i, s = 3.1, and r = 0.05–50 μm. Schoonmaker et al. [24] noted that the agreement was better at the forward angles because the scattering phase functions computed from Mie theory (assuming spherical particles) exhibited rainbow effects in the backward angles, which were not observed in Petzold’s VSF measurements.

So far, the inversion methods mentioned above were effected via trial and error. Zaneveld et al. [26] employed a rapidly converging minimization algorithm [27], which improved both efficiency and accuracy of the inversion. Against the same Sargasso Sea dataset, they estimated that the average error was reduced from about 20% with the trial-and-error method to about 13% with the minimization method. A total of 40 particle components or subpopulations were used as candidates, all following Junge distributions with s = 3.1, 3.3, 3.5, 3.7, 3.9, 4.2, 4.5, and 5.0 within the same size range of r = 0.02–20 μm, and varying refractive index of n = 1.02, 1.05, 1.075, 1.10, and 1.15. The particles are assumed to be nonabsorbing (no imaginary component in the indices).

Fitting against Kullenberg’s laser measurements, Zaneveld et al. [26] found that seven particle subpopulations contributed significantly to the observed VSF even though all 40 were found to be present numerically. These seven components are particles with n = 1.05 and s = 3.5, 3.7, and 3.9; particles with n = 1.075 and s = 3.7 and 3.9; and particles with n = 1.15 and s = 3.5 and 3.7. Because particles with n = 1.075 contribute little as compared to the other two particle groups, only particles with n = 1.05 and 1.15 are listed in Table 1. Particles with n = 1.02 and 1.10 were essentially not present.

Using a newly designed VSM measuring scattering at over 600 angles [4], Zhang et al. [9] measured VSFs in coastal water off New Jersey under both calm and windy conditions. In wind-roughened water with injected bubbles, they assumed that there were four subpopulations (n = 1.03, 1.05 + 0.002i, 1.15, and 0.75) that could potentially contribute to the observed VSFs. For each subpopulation, they further assumed that the particle distribution followed a Junge-type with a fixed s = 4, a fixed r_max = 300 μm, and a varying r_min from 0.1 to 10 μm. By applying a minimization algorithm based on nonnegative least squares, Zhang et al. [9] found that the enhanced VSF in the rough sea relative to the calm sea could be largely reproduced within 6% by three subpopulations, presumably mineral particles with n = 1.15 and r_min = 0.15, phytoplankton with
For a two-parameter model consisting of particles with $r < 1.3 \, \mu m$ and with $n = 1.15$, and particles with $r > 1.3 \, \mu m$ and with $n = 1.03$. It differed from the other studies mentioned above in two ways. First, the inversion returned volume concentrations of the respective small and large particles, which are not measured routinely and not easily accessible; and second, only the VSF at two angles ($1^\circ$ and $45^\circ$) are used in the inversion through an empirical relationship. Mobley [29] showed that the model underestimated Petzold's measurement in the coastal water at small scattering angles and overestimated at large angles. Berthon et al. [10] found that the model could reproduce observed VSF's in the coastal northern Adriatic Sea with average root mean square percentage differences between 8% and 17%. Shifrin [30] also showed that the two-parameter model fit well with the observation data. There are other similar models, such as Fournier and Forand [31]. However, most of these studies seemed to focus on the forward modeling of the VSF using a few measurements of scattering.

Deriving PSDs from the laser diffraction technique has been well established theoretically (e.g., [32]) and experimentally [33–35] with developed commercial products (e.g., [3]). The basis of inversion is still Eq. (1), where kernel functions are estimated based on Mie theory for spherical particles, but measurements of the VSFs are restricted to small forward angles where diffraction dominates. Because the technique relies on the diffraction theory, it has several limits: 1) it is not very sensitive to the refractive index of the particles and, therefore, only the number–size spectra can be derived; 2) only particles significantly larger than the light wavelength can generate diffraction patterns and, consequently, the size ranges that can be resolved are typically 2–200 $\mu m$. As a result, the composition of particles (i.e., the refractive index) and particles of sizes less than 1 $\mu m$ cannot be derived from the laser diffraction method [36]. Despite these limits, the applications of the technique (e.g., LISST-100X) have led to reasonable estimates of the size distribution for particles in laboratory phytoplankton cultures [37], lakes and rivers [38,39], marine bottom boundary layer [40,41], and surface coastal waters [36,42].

Among the three general approaches reviewed here, Kopelevich’s [28] two-parameter model and the laser diffraction approach only utilize a subset of full VSF and therefore the information on particles that can be inverted is limited. For example, only PSD for particles of micrometer sizes or larger can be derived from the laser diffraction approach. On the other hand, inversion of the VSF measurements at full angular ranges has the potential to derive both size and composition information of particles over wide ranges of sizes and the refractive index, and only those studies based on this approach are listed in Table 1.

Because the same laser measurements in the Sargasso Sea had been used in several studies, we compare their results in Fig. 1. The laser operated at 632.8 nm and measured scattering at eight angles between $1^\circ$ and $90^\circ$. The measurement by the $p$-meter at both 460 and 655 nm were also shown as baselines for extrapolating the laser measurement into the backward angles. If we assume that the shape of the VSF at 632.8 nm in the backward angles would fall somewhere between that at 460 nm and that at 655 nm, all the studies would have failed to reproduce the dip between $100^\circ$ and $120^\circ$ as observed by the $p$-meter. This is presumably because the modeled phase functions were unpolarized. Actually, three of them predict a bump in the VSF around these angles. Overall, it seems that the simple one-component model by Gordon and Brown [15] agreed best with the observations.

Rigorously, the minimization of Eq. (1), either by trial and error or by optimization, will be likely to converge for any given set of the kernel functions as long as the kernel functions are not singular. From Table 1 and Fig. 1, it is apparent that results of the inversion depend on the assumptions made regarding the index–size distribution of particles. Take the results for the Sargasso Sea as an example. Assuming a one-component system, Kullenberg [14] found the scattering can be entirely due to inorganic particles ($n = 1.20$), while Gordon and Brown [15] found that it can be accounted for by organic particles ($n = 1.05 + 0.01i$). In addition, inorganic particles ($n = 1.15$) were found to account for 95% of scattering with a two-component model [16], but only 62% with a multicomponent model [26]. The recent advent of newly designed VSF instruments, such as the VSM [4] and MASCOT [5,6], provide an opportunity to revisit this challenging task of inverting VSF data to characterize particles. Particles in the ocean exist in a continuum in the index–size domain; therefore, the fewer assumptions made regarding the index or size for particles, the more realistic the results would be. And yet, to model the volume scattering, assumptions have to be made. In the next section, we will evaluate the effects of different assumptions on modeling the volume scattering by a particle population.

4. Modeling Volume Scattering Functions

To facilitate the modeling of the volume scattering by particles in the ocean using Eq. (6), we assume that these particles can be partitioned into subpopulations, each can be characterized by a unique refractive index, and each has a PSD that can be analytically described. The one-component, two-component, or multicomponent models listed in Table 1 are specific examples of this assumption.

A. Junge Distribution

A common assumption of early studies listed in Table 1 is that particle subpopulations all follow
Junge distributions, which might be applicable to the simplified one-component model. For example, assuming a single representative particle population following a Junge-type size distribution, both Gordon and Brown [15] and Morel [3] found a slope of −4 fitted best to the measured VSFs, consistent with the well-known shape proposed by Sheldon et al. [43] on the average PSD based on the Coulter Counter measurements of surface and deep water of the Atlantic and Pacific oceans. The approximate abundances and sizes of major groups of microorganisms of sizes between 0.2 and 100 μm in the open ocean also seem to follow the Junge distribution [20]. From steady-state theory of the energy flow within the ecosystem and the total community metabolism, Platt and Denman [44] derived that total biomass \( m(w) \) in a given weight class \( w \) can be approximated as \( m(w) \sim w^\alpha \), where \( \alpha \) has a value varying from 0.22 for heterotomers to 0.18 for unicells [45]. From this and assuming roughly equal body density for different species in the pelagic ecosystem, one can derive that the Junge slope would have values between −3.46 and −4.66.

The truncations of the Junge distribution, however, are hard to interpret. Morel [3] did a thorough analysis on the effects of \( r_{\min} \), \( r_{\max} \), and \( s \) of Eq. (9) on the estimates of phase functions for a subpopulation following a Junge distribution. He found that if the slope satisfies the inequality, \( 2.7 < s < 7 \), the estimates of phase functions (except at near 0° and 180°) would be finite and not dependent on the exact values of \( r_{\min} \) and \( r_{\max} \) as long as \( r_{\min} < -0.2 \mu m \) and \( r_{\max} > -50 \mu m \). Stramski and Kiefer [20] found that predictions from Eq. (7) are virtually insensitive to the choice of \( r_{\min} \) as long as it is kept smaller than 0.015 μm, a value roughly corresponding to the sizes of smallest viruses. This value is also consistent with the calculation by Ulloa et al. [46], who found that the backscattering ratio for a particle population with \( s = 4 \) reaches an asymptotic value (0.75) at all visible wavelengths when \( r_{\min} < 0.02 \mu m \).

B. Lognormal Distribution

It is well known that phytoplankton species, one of the most optically significant and sometimes dominant constituents in the ocean, follow a lognormal distribution in their number–size spectra [47]. The lognormal distribution has also been used to characterize the other living or nonliving individual populations, including microbes [20], detritus [48–51], and mineral particles [52,53]. Theoretically, the natural processes of breakage [54], coagulation [55], or cell division [56] would render a population of particles following the lognormal distribution. Taking particle breakage as an example, if a particle of initial size \( r_0 \) is repeatedly broken into pieces of random proportions after \( m \) steps in the process, the size is \( r_n = r_0 \prod_{i=1}^{m} x_i \), where \( x_i \) take values between 0 and 1 representing the random proportions. Then, for large \( m \), the distribution of \( \ln(r_m) \) will, by the central limit theorem, be approximately normal. Intuitively, we also know that, at some small size, particles transition into the dissolved phase, so having a model that reflects this is appealing. As will also be shown, this PSD model is not incompatible with the Junge-type model when assessed over specific size ranges.

For phytoplankton species, \( r_{\text{mode}} \) ranges from submicrometer for picoplankton to several tens of micrometers for nanoplanckton to several hundreds of micrometers for microplanckton. The standard deviation of the logarithmic distribution, \( \sigma \), on the other hand, has been found to vary in a small range from 0.07 to 0.11 for monospecific laboratory cultures [57] and from 0.16 to 0.26 for natural populations over a wide range of oceanic regimes [47,58]. For detritus and colloids, the reported values of \( r_{\text{mode}} \) range from 0.02 to 2 μm and \( \sigma \) ranges from 0.16 to 1.6 [48–51]. For mineral particles, measurements show they follow lognormal distributions with a limited range of \( r_{\text{mode}} \) between 0.5 and 0.7 μm and \( \sigma \) between 0.5 and 0.7 [52]. Table 2 summarizes the measured or estimated values of \( r_{\text{mode}} \) and \( \sigma \) for particles following lognormal size distribution.

### Table 2. Measured or Estimated Values of \( r_{\text{mode}} \) and \( \sigma \) for Particles Following Lognormal Size Distribution

<table>
<thead>
<tr>
<th>Ref.</th>
<th>( r_{\text{mode}} ) (μm)</th>
<th>( \sigma )</th>
<th>Range of Sizes (μm)</th>
<th>Notes</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>[48]</td>
<td>0.25</td>
<td>0.17</td>
<td>0.1–1</td>
<td>nonliving organic matter</td>
<td>Notes</td>
</tr>
<tr>
<td>[49]</td>
<td>0.025 to 0.35</td>
<td>0.47–0.53</td>
<td>0.005 &lt; 5</td>
<td>colloids</td>
<td>particles of biological origin</td>
</tr>
<tr>
<td>[51]</td>
<td>0.28–0.3</td>
<td>0.16–0.22</td>
<td>0.2–0.5</td>
<td>three classes of particles: organic (living and detritus), inorganic (e.g., iron colloids), and clay minerals</td>
<td>mineral particles</td>
</tr>
<tr>
<td>[50]</td>
<td>—</td>
<td>1.6</td>
<td>&lt;0.12</td>
<td>natural phytoplankton population</td>
<td>natural phytoplankton population</td>
</tr>
<tr>
<td>[52]</td>
<td>1.0</td>
<td>0.5–0.7</td>
<td>&gt;0.1</td>
<td>cultured monospecific phytoplankton population</td>
<td>natural phytoplankton population</td>
</tr>
<tr>
<td>[57]</td>
<td>—</td>
<td>0.07–0.11</td>
<td>&gt;0.2</td>
<td>natural phytoplankton population</td>
<td>partition measured PSDs as a sum of log-normal functions</td>
</tr>
<tr>
<td>[58]</td>
<td>—</td>
<td>0.16–0.26</td>
<td>&gt;0.2</td>
<td>natural phytoplankton population</td>
<td></td>
</tr>
<tr>
<td>[55]</td>
<td>—</td>
<td>0.64 ± 0.3</td>
<td>0.2–100</td>
<td>nonliving organic matter</td>
<td>Notes</td>
</tr>
</tbody>
</table>

In addition to sizes, the optical properties of particles also depend on the refractive index, \( n = n_r + i n_i \). While absorption is due to the imaginary part only, scattering is controlled by both the real and imaginary parts of the refractive index [59]. In the visible wavelengths, the imaginary part of the refractive index of oceanic particles (living or nonliving) is of the order of \( 10^{-4}–10^{-2} \); for example, \( n_i \) for phytoplankton at its absorption peak at 430 nm is about 0.005–0.01 [60]. Therefore, for oceanic particles, scattering is primarily controlled by \( n_r \), which ranges between 1
Oceanic particles are seldom perfectly spherical or uniform. It has been shown that nonsphericity can alter the angular scattering significantly, especially for large particles and in the backward directions \[63,64\]. Based on the calculations of scattering for spheroids of various aspect ratios, Clavano et al. \[63\] concluded that assumption of sphericity would underestimate the attenuation, absorption, and scattering coefficients, but within a factor of 3, for particles with polydispersal distribution. They pointed out that modeling the VSF for nonspherical particles remains a challenge because no numerical solution is available for particles of intermediate sizes \(2 < r < 20 \mu m\). In this study, we will assume that oceanic particles are spheres, or more specifically, that the VSF of oceanic particle subpopulations can to first order be approximated using Mie theory. Even for spherical particles, internal structures impact the pattern of scattering. Many oceanic particles or organisms have a layered structure. Diatoms have a silicate shell; bubbles have an organic film on their surface. It has been shown that a shell or coating would enhance backscattering \[65–67\]. We will consider layered spheres for bubbles in our analysis.

5. Sensitivity Analyses

For sensitivity analyses, particles are assumed to be uniform and spherical, for which Mie scattering theory can be applied \[68\]. We calculated the angular scattering cross sections, \(C_{\text{ang}}\), for particles of refractive indices from 1.01 to 1.20 for \(n_r\) and from 0 to 0.02 for \(n_i\), and of radii from 0.001 to 300 \(\mu m\). The wavelength was fixed at 532 nm. An example of angular variation of \(C_{\text{ang}}\) is shown in Fig. 2, where the backscattering ratios for single particles of four different indices are plotted as a function of particle radius. Because we are interested in individual subpopulations, we use the lognormal to approximate their size distributions, with \(r_{\text{mode}}\) varying from 0.01 to 10 \(\mu m\) and \(\sigma\) from 0.1 to 1.6. These ranges of values would encompass the majority of particle distributions in the global ocean (Table 2). Also shown in Fig. 2 are examples of PSDs for \(r_{\text{mode}} = 0.1 \mu m\) and \(\sigma = 0.3\), and \(r_{\text{mode}} = 10 \mu m\) and \(\sigma = 1.0\), representing subpopulations that are dominated by particles with sizes optically equivalent to \(<0.5\) and \(>10\mu m\). Equations (5)–(7) were used to compute the phase functions for subpopulations. The phase functions thus estimated are diverse and cover over 4 orders of magnitude of variation at each angle. To evaluate the sensitivity of phase functions with respect to the change in \(r_{\text{mode}}, \sigma, n_r, \) or \(n_i\), we use the following parameter:

\[
R_{\text{VSP}}(\theta, x) = \frac{\max(\beta(\theta, x, Y))}{\min(\beta(\theta, x, Y))},
\]

where \(x\) represents the changing variable, and \(Y\) the rest of the variables that are averaged over. For example, \(R_{\text{VSP}}(\theta; \Delta n_r)\) gives the maximum variation of the VSF for a change of \(\Delta n_r\) in \(n_r\), averaged for \(r_{\text{mode}}, \sigma, \) and \(n_i\) over their respective ranges. The parameter \(R_{\text{VSP}}\) gives an estimate of the maximum variation in phase functions in response to a prescribed change in one of the parameters in the particle-size–index domain. In the natural environment and considering all possible particle subpopulations, \(R_{\text{VSP}}(\theta)\) is expected to vary over 3–4 orders of magnitude.

A. Particles of Extreme Sizes

Phase functions calculated for various PSDs and indices form clusters, indicating that the sensitivity of the angular scattering by particles is not uniformly spaced in the size–index domain. The first group of clusters, shown in Fig. 3(a), is for subpopulations dominated by small particles of sizes \(<0.1 \mu m\), which are practically dissolved and referred to as very small particles (VSPs) hereafter. In this group, the phase functions are primarily determined by the PSD, with weak effect by the refractive index of...
particles. This can be explained by the fact that the shapes of $C_\text{ang}$ (represented by $b_0$) for small particles are almost solely determined by particle sizes, regardless of its refractive index (Fig. 2). For particles of further smaller sizes (<0.02 μm), even the size does not matter anymore, with the phase functions resembling those of molecules, converging to classic Rayleigh scattering theory. Therefore, it can be expected that factors normally determining the phase function of PSDs are minor details for VSPs. Within each cluster shown in Fig. 3(a), $R_{\text{VSF}}(\theta, n) < 1.5$.

The second group of clusters, shown in Fig. 3(b), is for subpopulations dominated by large particles, referred to as LPs. In this group, the phase functions are largely determined by the refractive index, with smaller effect by PSD. Also note that the distinction between different clusters diminishes for particles of higher values of $n_r$, presumably of inorganic origin. Again this can be explained that $b_0$ for particles >20 μm seems to reach asymptotic values dependent only on the values of particle refractive index and these asymptotic values are closer to each other among more refringent particles (Fig. 2). Within each cluster shown in Fig. 3(b), $R_{\text{VSF}}(\theta, \text{PSD}) < 2$ on average.

![Fig. 3. Phase functions for particles of lognormal distributions and of various refractive indexes. In (a), legend entries indicate the values of $r_{\text{mode}}$ and $\sigma$. Different curves within each group are for different $n = 1.01$ to 1.20 + 0.001i; in (b), legend entries indicate the values of $n_r$. Different curves within each group are for different size distributions with $r_{\text{mode}} > 10 \mu m$ and $\sigma > 1.0$.](image)

B. Particles of Intermediate Sizes

For subpopulations of intermediate sizes that are most significant optically and of most interest, both PSD and the refractive index are equally important in determining the phase function. The sensitivities of phase functions with respect to $r_{\text{mode}}$, $n_r$, and $n_i$ are evaluated in Figs. 4(a)–4(d), respectively. Overall, the changes in phase functions increase rapidly with increasing differences in each of these parameters. On average, phase functions vary by less than 50% ($R_{\text{VSF}} < 1.5$) if $r_{\text{mode}}$ changes by <50%, $\sigma$ by <0.1, or $n_r$ by <0.01. On the other hand, phase functions would differ by 1 order of magnitude ($R_{\text{VSF}} > 10$) if $r_{\text{mode}}$ differs by a factor of 5 or greater, $\sigma$ by 0.5, or $n_r$ by 0.04. Because of the relatively constrained variation in $n_i$ for particles at 532 nm or other longer wavelengths outside of the absorption band of chlorophyll, the variations in phase functions due to $n_i$ are small in general, typically $R_{\text{VSF}} < 2$ for $n_i = 0–0.01$.

C. Similar Size Distributions

In addition to lognormal, other functions of similar shape have been proposed to represent the PSDs, such as Gamma [69,70] or Weibull [71]. Simulations with spheres or spheroids have shown that different size distributions with the same effective radius and effective variance (both weighted by area) would produce similar optical properties (e.g., [72]). A typical phytoplankton population with a peak located at 1.6 μm and a width of dispersion of ~2 μm was approximated by the three functions of lognormal, Gamma, and Weibull in Fig. 5(a) and their corresponding phase functions were shown in Fig. 5(b). The three mathematical functions differ in their detailed representations of the size distribution, but the resulting phase functions are similar to each other within 50% in general ($R_{\text{VSF}} < 1.5$).

D. Summary

Relatively and within their respective ranges of natural variability, $r_{\text{mode}}$ and $n_r$ seem to exert the largest influence on phase functions, with $\sigma$ having a lesser but still significant impact. For a given peak size, slight differences in the shape of the PSD, as represented by different mathematical functions, have a minor effect on the phase functions, as also the exact values of $n_i$. For VSP subpopulations, the phase functions are primarily determined by PSD and, for LP subpopulations, the phase functions are primarily determined by $n$.

6. Method and Data

During the Hyperspectral Coastal Ocean Dynamics Experiment (HyCODE) at the LEO-15 site off the New Jersey coast [73] from 24 July to 4 August 2001, a prototype VSM [4] was deployed on board the R/V Endeavor along with an AC-9 (WET Labs), conductivity–temperature–depth device, and other instruments, in an in-line configuration for continuous measurements during the cruise. Water was
taken from the ship’s intake (~5 m). During the experiment, there were a few high wind events with wind speeds reaching 15 m/s. VSFs were measured at a nominal wavelength of 532 nm from 0°–177°3′ at an interval of 0°3′. The scattering by pure seawater, estimated for the temperature and salinity [74, 75], was subtracted from the VSF measurements. One scan of the VSM consists of a complete rotation of 360° and the output VSF is the mean values between 0°–180° and 360°–180°. Often, several runs of measurement are taken (~1.5 min one run) for one sample. Assuming the VSF of a sample does not change over a short measurement period, we found that the instrument uncertainty (estimated as the coefficient of variation) of the VSM varies between 3% and 15% across the angles, with a mean of ~10%. The design of the VSM allows an output of scattering at 606 angles and the measurements at the LEO-15 are shown in Fig. 6(a).

Whether this high angular resolution leads to data redundancy depends on 1) the inherent relationship between scattering at different angles and 2) the uncertainty associated with the measurements. In other words, can the scattering at one particular angle be predicted from the scattering at other angles within the experimental uncertainty? From a statistical point of view, however, the inversion does not assume any “redundant” or “useless” measurements. Therefore, the VSFs at additional angles simply serve the purpose of lowering the uncertainty by a factor of \( N_{\text{add}}^{-1/2} \), where \( N_{\text{add}} \) is the number of

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Fig. 4. Sensitivity of phase functions (RVSF) with respect to (a) \( r_{\text{mode}} \), (b) \( \sigma \), (c) \( n_r \), and (d) \( n_i \). In (a), the change in \( r_{\text{mode}} \) between two values is evaluated as a ratio (\( R_{r_{\text{mode}}} \)), and the curves from bottom to top correspond to \( R_{r_{\text{mode}}} = 1.5, 2.0, 3.0, 4.0, \) and 5.0, respectively; in (b), the change in \( \sigma \) is evaluated as a difference (\( \Delta \sigma \)), and the curves from bottom to top correspond to \( \Delta \sigma = 0.05, 0.1, 0.2, 0.3, 0.4, \) and 0.5, respectively; in (c), the change in \( n_r \) is evaluated as a difference (\( \Delta n_r \)), and the curves from bottom to top correspond to \( \Delta n_r = 0.01, 0.02, 0.03, \) and 0.04, respectively; in (d), the change in \( n_i \) is evaluated against \( n_i = 0.001 \). Note the ordinate scale in (d) is different from (a)–(c).

---

Fig. 5. (a) Typical phytoplankton PSD is represented by three different functions: Weibull, Lognormal, and Gamma; all have the same peak size and similar measures of spread. (b) Corresponding phase functions (left y axis) and RVSF (right y axis).
redundant angles \[^76\]. The number of independent angular VSFs in the presence of relative error of measurement \(\varepsilon\) is determined by \[1\]
\[
\sqrt{\Lambda_{\text{min}}} < N_{\text{add}}^{-1/2} |\varepsilon|,
\]
where \(\Lambda\) represents the eigenvalues of the VSFs. Figure 6(b) shows the variation of the square root of the eigenvalues for the VSFs as a function of number of angles at which the VSFs were measured. With the experiment error of 10\%, the number of independent angles is estimated to be 40 [see Fig. 6(b)] and, if the uncertainty is to be reduced to 1\%, the independent angles would be 140. The VSM used in this study measures scattering at 606 angles; a relative error of 10\% means that there are 170 independent angles \((N_{\text{add}} = 566)\), i.e., that more than 2/3 of VSM angles are redundant. However, in this study, all of the VSM angles are used in the inversion recognizing that the redundancy was treated as a statistically independent repetition of the similar measurement in an attempt to lower the inversion uncertainty. Note that the angular redundancy in the measured VSFs will not affect the stability of the inversion, but the singularity of the kernel functions will, which is addressed in the following.

To derive the size and index information of particle subpopulations from measurements of the VSFs, the Fredholm linear integral equation [Eq. (1)] needs to be inverted. For the inversion, a pool of phase functions for the potential subpopulations (the kernel functions) needs to be constructed first. As shown in Fig. 1 and Table 1, the results of inversions depend on the initial assumptions made regarding the size–index spectra of particles. In principle, the assumptions should be based on \textit{a priori} knowledge of the potential subpopulations that may exist in the environment under any given condition. Also, for the results of inversion to be statistically representative and stable, the candidate phase functions should be constructed such that they distribute relatively evenly at all angles without clustering in order for the scattering kernels to be neither singular nor pathological \[^77\].

Based on the results of sensitivity analyses presented above, we prescribed a pool of candidate subpopulations, the details of which are listed in Table 3. In determining these candidate subpopulations, we followed these guidelines. First, the subpopulations follow a size distribution with a dominant size, and the lognormal function is used to represent this size distribution. Second, each subpopulation is uniquely decided by \(r_{\text{mode}}, \sigma, \) and \(n(= n_r + i n_i)\). Third, the values of these variables are chosen so that the corresponding phase functions differ from each other.

![Figure 6](image-url)
Table 3. Values of Parameters Defining the Size–Index Spectra for the Subpopulations Used in Inversion

<table>
<thead>
<tr>
<th>VSP</th>
<th>Particles of Intermediate Sizes</th>
<th>LP</th>
</tr>
</thead>
<tbody>
<tr>
<td>PSD</td>
<td>Lognormal</td>
<td>Lognormal</td>
</tr>
<tr>
<td>$r_{\text{mode}}$ ($\mu$m)</td>
<td>0.02, 0.04, 0.08</td>
<td>0.2, 0.4, 0.8, 1.6, 3.2</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>0.3:0:2; 0.7$^a$</td>
<td>0.1:0:2:0.9$^a$</td>
</tr>
<tr>
<td>$n_r$</td>
<td>1.05$^b$</td>
<td>0.75, 1.02, 1.04, 1.06, 1.10, 1.15, 1.20</td>
</tr>
<tr>
<td>$n_i$</td>
<td>0.001</td>
<td>0.001</td>
</tr>
</tbody>
</table>

$^a$The three values indicate start, increment, and end, respectively.
$^b$The phase functions for VSP do not depend on the exact values of $n_r$.
$^c$For bubbles.

by at least a factor of 2. Since phase functions typically vary by 3–4 orders of magnitude at any given angle over the range of possible changes in PSD and index of particles, a factor of 2 difference should be able to provide sufficient details covering the entire range of variations without causing clusters in the kernel functions. Based on Figs. 3 and 4, for a factor of 2 differences in phase functions, the change in $r_{\text{mode}}$ (as measured by a ratio $R^r_{\text{mode}}$), with everything else the same, needs to be at least 2 ($R^r_{\text{mode}} = 2$). Similarly, the change in $\sigma$ (as measured by a difference) needs to be at least 0.2 ($\Delta \sigma = 0.2$); $\Delta n_r$ needs to be 0.02. Because the phase functions are not very sensitive to $n_i$, $n_i$ was fixed at 0.001.

We used Mie theory for spheres and modified Mie theory for coated spheres [68] to calculate the phase functions for particle and bubble subpopulations ($\vec{b}_i$ for $i = 1 \ldots M$), respectively, for a given set of values of $r_{\text{mode}}$, $\sigma$, and $n$ from Table 3. Visual observation showed that the dissolution of natural bubbles is consistent with that of microbubbles coated with short-chain and unsaturated fatty acids [78] and we assumed that the surfactant film on natural bubbles is a monolayer of lipid type with a constant thickness of 2 nm [79] and with an index of refraction of 1.20. Because the phase function depends in a nonlinear manner on $n$, $r_{\text{mode}}$, and $\sigma$, it is possible that different particle subpopulations produce a similar phase function. For example, the calculated phase function based on Mie theory for the subpopulation of $n_r = 1.04$, $r_{\text{mode}} = 0.2 \mu$m, and $\sigma = 0.5$ is very similar (within 10%) to that of $n_r = 1.06$, $r_{\text{mode}} = 0.4 \mu$m, and $\sigma = 0.3$. In other words, within the uncertainty of the measurements of 10%, the inversion cannot discern between these two subpopulations. Therefore, the similar phase functions were eliminated from the kernel functions. From this “similarity elimination” and the sensitivity analysis presented in Section 5 (Figs. 3 and 4), we estimated that, for an average of 3%–15% uncertainty in the measured VSFs, the inversion uncertainties are ±0.005 for $n_r$, ±10% for $r_{\text{mode}}$, and ±0.04 for $\sigma$.

The candidate phase functions and their corresponding backscattering ratios are shown in Figs. 6(c) and 6(d), respectively. As can be seen, they cover a wide range of variations. The total number of candidate phase functions, and, hence, of subpopulations, is 126. For a given VSF measurement, we used the linear least squares algorithm with nonnegative variables [80, 81] to solve Eq. (1) in order to derive a set of scattering coefficients ($\vec{b}_i$) for each candidate $\vec{b}_i$. Because the variables to be determined from Eq. (1) are scattering coefficients, the constraint of nonnegativity is important to ensure physical soundness of the solution. If one of the derived $b_i$ is zero, then the subpopulation associated with the corresponding phase function is assumed to be not present or have insignificant contribution to the measured VSF.

There are 606 angles (or 170 removing redundancy) between 0° and 180° for the VSM as compared to 126 candidate phase functions; this gives overdetermined kernel functions of Eq. (1), which always lead to a unique solution mathematically. But sometimes, the kernel functions are underdetermined; for example, Zaneveld et al. [26] used 40 candidate phase functions for inversion from the VSF measurements at nine angles. In these cases, the solution has to satisfy the following conditions in addition to the nonnegativity constraint: 1) has the nearest Euclidean distance to the vector space representing the solutions without the nonnegativity constraint; and 2) has the most nonzero components. To further test the stability and uniqueness of the solution, the inversion was run twice: first, with full candidate phase functions and, second, with only those candidate phase functions corresponding to which the derived scattering coefficients are nonzero from the first run. For the LEO-15 VSF data, the inversion results are exactly the same between the two runs, suggesting that the method is stable and the solution obtained is unique. This also suggests that the inclusion of “nonrealistic” subpopulations will not distort the results.

7. Results and Discussion

A total of 385 measurements of the VSF at different locations and times were analyzed. An example of inversion results is shown in Fig. 7, where the VSFs by individual subpopulations that were detected to be present through inversion [namely, having $b_i > 0$ as in Eq. (1)] are compared with the measured VSF in Fig. 7(a) and the corresponding PSDs for the subpopulations are plotted in Fig. 7(b). For this particular measurement, where wind speed was 9.5 m/s, only 21 of the initial 126 subpopulations were identified to be present. The total scattering coefficient estimated from subpopulations (0.725 m$^{-1}$) was almost identical to the estimate from the measured
VSF (0.727 m\(^{-1}\)), suggesting that identified subpopulations can account for the observed angular scattering variations, particularly in the forward direction. The size distributions for the subpopulations show a wide range of variations in both sizes and shapes. There were six subpopulations identified for particles of index 1.02, four subpopulations for bubbles, three subpopulations for particles of index 1.04, two subpopulations for particles of indices 1.1 and 1.2, and only one subpopulation for particles of indices 1.06 and 1.15. It is interesting to note that the cumulative size distribution for all the particles can be closely approximated by a power-law function. The Junge slope for this particular example is -3.9 and the total particle concentration of sizes between 0.2 and 200 \(\mu\)m is \(1.0 \times 10^{13}\) m\(^{-3}\).

The subpopulations shown in Fig. 7 are grouped together by the refractive index (\(n\)) and the dominant size (\(r_{\text{mode}}\)), and the results are shown in Figs. 8 and 9, respectively. The observed shape of the VSF for angles <60° is largely due to particles with high water content (\(n_r = 1.02\)) and for angles >90° due to more refringent particles (\(n_r = 1.10\) and 1.15) [Fig. 8(a)]. As can be expected, bubbles (\(n_r = 0.75\)) contributed most around critical angles (60°–80°). On the other hand, particles of different size ranges also contributed differently to the angular distribution of the VSF. For example, the VSF at angles >90°, as shown in Fig. 9(a), is mostly due to particles with dominant sizes of 3.2 \(\mu\)m, and for angles <5°–10° mostly due to particles of sizes 0.8–1.6\(\mu\)m. For a given particle size range, there are particles of different indices present, particularly between 1 and 100 \(\mu\)m, where no single particle subpopulation seems to dominate numerically [Fig. 8(b)].
Boss et al. [8] estimated the variations of the refractive index and the Junge slope for particles over the LEO-15 site using the measurements of the backscattering ratio and the attenuation coefficient. The backscattering ratios were measured by three different instruments, Hydroscat-6 (HOBILabs), Eco-VSF (WET Labs), and the VSM; the attenuation coefficients were measured by AC-9 (WET Labs). Their results are shown in Fig. 10(a), in which the slope, estimated for sizes between 0.5 and 10 μm, varied between 3.3 and 4.1 with a mean of around 3.7. Reynolds et al. [36] examined LISST-100 field measurements from coastal regions of California and northern Europe and found that the average Junge slope for particles larger than 3 μm ranges from 3.0 to 4.0 with a mean of 3.5. Estimated over 0.5–10 μm, our slope values are 4.1 ± 0.4 and steeper by 0.4 than those reported by Boss et al.; over 3.2–200 μm, our values are 3.8 ± 0.2 and steeper by 0.3 than those of Reynolds et al.. All three methods are based on the assumption of spherical particles, but differ in the inversion techniques. Boss et al. [8] derived the slopes from the spectral slopes of the attenuation coefficient, assuming particles following a Junge distribution with a global mean refractive index. The laser diffraction technique in LISST-100 is not sensitive to the refractive index. Therefore, in both of these methods, the refractive index is not an input parameter, while we estimated the Junge slope from summation of individual subpopulations, each with different size distribution and different refractive index. The other reason could be because the 2001 VSF data were measured more offshore than the 2000 data that Boss et al. used, therefore, potentially fewer larger particles could be retrieved. Apparently, the value of the Junge slope is sensitive to the size ranges over which it is estimated, because changes in individual species or particle population often distort the mean state (e.g., [36,82]). Over the broader range from 0.2 to 200 μm, the mean value of the Junge slope from our method is 4.0 ± 0.2 [Fig. 10(b)], in close agreement with the global mean value of 4.0.

On the other hand, the distributions of the refractive index by Boss et al. [8] and our method exhibit similarity. Both the index histogram from the VSF inversion [Fig. 10(c)] and the index estimated from the backscattering ratio using the model by Twardowski et al. [12] indicate that particles of lower index appeared more often than particles of higher index, suggesting that particle populations at LEO-15 are dominated by particles of high water content. The VSF inversion also indicates a consistent presence of bubbles. This could not be resolved based on the backscattering ratio method alone, which was developed for particles with relative $n > 1$. In the Twardowski et al. [12] model, a size distribution of bubbles with Junge slope around
−4 is falsely assigned a refractive index value of around 1.10.

Figure 10(d) shows that subpopulations of sizes 0.4−3.2 μm are dominant at the LEO-15 site. It is also interesting to note that, for the VSP groups, subpopulations with dominant sizes of 0.04 and 0.08 μm have never shown up in the inversion.

Among 126 candidate subpopulations, there are 56 that were not identified as being present over the entire LEO-15 database. There are 11 subpopulations that were common in more than half of the inversions; however, only five subpopulations were consistently present (>90% of measurements). These five subpopulations, summarized in Table 4, can be interpreted as representing VSPs ($r_{\text{mode}} = 0.02$ μm, $\sigma = 0.3$), whose phase function is effectively the same as that of molecules; submicrometer detritus particles with higher water content ($n_r = 1.02$, $r_{\text{mode}} = 0.2$ μm); two micrometer-sized subpopulations with one relatively soft ($n_r = 1.04$ and $r_{\text{mode}} = 1.6$ μm) and the other relatively refringent ($n_r = 1.10$ and $r_{\text{mode}} = 3.2$ μm); and bubbles of relatively large sizes ($n_r = 0.75$ and $r_{\text{mode}} = 10$ μm). Their presence is consistent with histograms shown in Figs. 10(c) and 10(d). The LEO-15 site is located in a coastal area with influences of river runoff and tide-induced sediment resuspension [73]. Also several strong wind events with wind speeds reaching 15 ms$^{-1}$ during the experiment were expected to cause further mixing in addition to injecting air bubbles. Therefore, it is reasonable to assume all these five subpopulations were present at LEO-15 during the experiment and have significant contributions to the signals measured by the VSM.

Figures 7 and 8 show that particles of different index and size contribute differently to the angular distribution of scattering and to the total PSD of one particular measurement. Figure 11 summarizes the average percentage contributions by the five persistent subpopulations to the observed VSFs [Fig. 11(a)] and to the total PSDs [Fig. 11(b)] over the entire LEO-15 site. The five subpopulations (Table 4) can explain between 25% and 70%, depending on angle, of the observed VSFs. At angles of less than 10°, micrometer-sized soft particles ($n_r = 1.04$, $r_{\text{mode}} = 1.6$) can account for between 10% and 50% of the volume scattering. Submicrometer detritus-type particles ($n_r = 1.02$, $r_{\text{mode}} = 0.4$) have the most significant contribution of 25% at angles of around 10°. On the other hand, micrometer-sized relatively refringent particles ($n_r = 1.1$, $r_{\text{mode}} = 3.2$) contribute almost equally (10%−20%) at angles greater than 90°, with less than 5% contribution at smaller angles. As can be expected, scattering contribution due to bubbles is restricted to around the critical angle (60°−80°) and only amount to 5%. It is surprising to notice that VSP particles, contributing negligibly to the forward and, hence, total scattering, remain as an important source of backscattering (accounting for between 20% and 40%) in such a turbid environment as LEO-15. Dall’Olmo et al. [21] measured the backscattering and attenuation coefficients (a proxy for particulate scattering or forward scattering) for particles at different size fractions and they found (from their Fig. 10) that particles of <0.1 μm have an insignificant contribution to bulk forward scattering (at 526 nm), which is consistent with our VSP results, but for backscattering, their contributions range from negligible for oligotrophic water to ∼7%−25% for mesotrophic water in open ocean. The latter, in combination with our result that VSP can account for 20%−40% backscattering in a coastal water, may also indicate that contribution by VSPs to backward

Table 4. Persistent Subpopulations

<table>
<thead>
<tr>
<th>No.</th>
<th>Subpopulation</th>
<th>$n_r$</th>
<th>$r_{\text{mode}}$ (μm)</th>
<th>$\sigma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>VSP</td>
<td>0.02</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>1.02</td>
<td>0.4</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>1.04</td>
<td>1.6</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>1.10</td>
<td>3.2</td>
<td>0.3</td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.75</td>
<td>10.0</td>
<td>1.1</td>
<td></td>
</tr>
</tbody>
</table>

![Graph](image-url)
scattering might increase progressively with trophic state.

The contributions by the five persistent subpopulations to the total PSDs can be easily discerned [Fig. 11(b)]: they are significant (up to 90%) in their respective size ranges, with the exception of bubbles. It is interesting to note that between 10 and 100 μm, none of these five persistent subpopulations dominate in the PSD, even though the dominant size for the bubble subpopulation is 10 μm. This also implies that particles of sizes 10–100 μm might have varied significantly in composition at the LEO-15 site during the experiment. The dominance of the bubble subpopulation over sizes greater than 100 μm can be explained by two factors: continuous injection of bubbles at high winds and their subsequent stabilization [83], and/or some artificial generation of bubbles due to pumping of water through the ship’s intake, which we cannot categorically rule out.

The PSDs derived from the VSF measurements and their mean values are shown in Fig. 12. There are two salient features of the mean PSD: an overall bimodal distribution and a smooth Junge-type distribution for particles of larger sizes. Risovic [69] found that the PSDs in the global ocean can often be described by a two-component model, one for small and the other for large particles. Aerosols are often found to follow bimodal distribution [84]. Figure 12 suggests that marine particles in the LEO-15 site may follow the similar bimodal distribution and, at the same time, is consistent with earlier studies showing power-law distribution for larger particles. Jonasz and Fournier [85] were able to decompose the size distributions of marine particles of 0.25–100 μm into a sum of lognormal components. These size distributions were measured in various water bodies and could be expressed as power-law functions to the first-order approximation. Figures 7(b) and 12 show that the cumulative size distribution of particle subpopulations, each following a lognormal distribution, does produce a distribution that can be approximated by the power-law function. This further confirmed the validity of Junge-type distribution as a first-order approximation for particles in the upper ocean.

On a physical basis, the Junge-type distribution cannot extend down to infinitesimal sizes, and our results based on the inversion of the VSF showed that there are significant numbers of particles of sizes around 0.02 μm. From an optical point of view, this group of particles contributed considerably to the backscattering and hardly any to the total scattering [Fig. 11(a)]. Particles at this size range are viruses, inorganic crystalline materials, but mostly colloids [86]. From Fig. 11(a), the total number of particles of <0.1 μm vary from 1.9 to 7.7 × 10^15 m^-3 with a mean value of 3.86 × 10^15 m^-3. Wells and Goldberg [50,86] estimated that the number of small colloids (<0.12 μm) ranged from 2.6 to 4.5 × 10^15 m^-3 in the surface waters and from 1.0 to 3.5 × 10^15 m^-3 from surface to depths in the coastal water of California. On the other hand, the abundance of viruses in the marine environment ranges from 0.003 × 10^12 to 15 × 10^12 m^-3 in open ocean waters and from 0.005 × 10^12 to 0.5 × 10^15 m^-3 in coastal waters [87]. Based on these numbers, we postulated that the VSP subpopulations at LEO-15 are dominated by the colloidal-type particles. However, because the volume scattering is not sensitive to the refractive index for the VSP particles, the composition of these colloidal particles cannot be determined, even though the majority of them are believed to be organic [86].

Oceanographers have traditionally employed filters of 0.2–0.7 μm pore size to arbitrarily partition seawater into “dissolved” and “particulate” phases. And optically, for the “dissolved” part, only the absorption is accounted for. Earlier theoretical [20,46] and experimental [22,88] studies lead to a hypothesis that colloidal particles may be a dominant source of particulate backscattering in the upper ocean, although recent measurements have suggested that their contribution might have been overestimated in oligotrophic waters [21]. Stramski et al. [89] suggested that the abundance, size distribution, and composition of colloidal particles and their associated scattering properties need to be quantified to better understand the roles of submicrometer particles in ocean optics. Here, by inverting the measured VSFs, we were able to map the size distributions of the colloidal particles at the LEO-15 site, which, often discounted as operationally partitioned “dissolved” particles, can still account for up to 40% scattering in the backward angles.

8. Conclusions

Mathematically, deriving the PSDs and characteristics from measurements of the VSF involves inverting the Fredholm linear integral equation, for which the kernel function is a pool of precalculated candidate phase functions. For the solution to be stable, a kernel function should be constructed that is neither singular nor pathological. This means that the phase
functions for the candidate subpopulations should be as distinctive as possible.

The candidate phase functions were carefully chosen based on sensitivity analyses and they differ from each other by an average of a factor of 2 at every angle, which was to the first order driven by refractive index and size distribution (Fig. 4). The uncertainty for the VSM measurement was estimated to be ±10%, and the corresponding uncertainties in the inversion are ±0.005 for \( n_i \), ±10% for \( r_{\text{mode}} \) = 3.2 and ±0.04 for \( \sigma \), respectively.

Applying the inversion method to the VSF measurements at the LEO-15 site indicated five omnipresent subpopulations (Table 4): VSPs (\( r_{\text{mode}} \) = 0.02 \( \mu \)m); submicrometer particles presumably with higher water content (\( n_r = 1.02 \) and \( r_{\text{mode}} = 0.4 \mu \)m); two micrometer-sized subpopulations, relatively soft (\( n_r = 1.04 \) and \( r_{\text{mode}} = 1.6 \mu \)m) and relatively hard (\( n_r = 1.10 \) and \( r_{\text{mode}} = 3.2 \mu \)m); and bubbles of relatively large sizes (\( n_r = 0.75 \) and \( r_{\text{mode}} = 10 \mu \)m). Among 126 candidate phase functions, 56 were not present at all and only 11 were present in half of the analysis. This indicates the consistency and stability of the algorithm. The PSDs of particles over the entire size range typically follows a bimodal distribution (Fig. 12), and, for particles greater than 0.5 \( \mu \)m, these closely resemble the well-known Junge distribution with slopes of −4.0 ± 0.2 [Fig. 10(b)]. The VSPs, mostly of a colloidal type, can contribute significantly to the backscattering.

In constructing the phase functions for the potential particle subpopulations, we have assumed that particle subpopulations follow a lognormal distribution with varying peak sizes and standard deviations and that those particles are homogeneous and spherical. Individual particle subpopulations, regardless of their origin, have been found to follow lognormal distribution in their sizes either based on observations [20,48–52] or theory [54–56]. Sphericity for particles, however, is seldom observed in nature, with the probable exception for bubbles [66]. Unfortunately, nonsphericity is not a particular shape; it remains challenging to model the particle scattering taking into account the entire variation of the shapes. We acknowledge that assuming sphericity is only an approximation, useful in demonstrating the potential of deriving detailed information of particle populations from measured VSFs. The key question is that, under this assumption, what conclusions can still be valid?

Mie theory for spherical particles predicts the presence of the optical glory, an enhancement of intensity in the direction of \( \theta = 180^\circ \), which was constantly observed at the LEO-15 site (e.g., see Fig. 7) as well as in the coastal northern Adriatic Sea [10]. However, the presence of the glory is not necessarily indicative of sphericity, because, theoretically, particles of a shape with continuous curvature, such as spheroid [90], can trap the surface wave and generate the glory [59,91]. The sphericity assumption is less of a problem for small particles and for scattering at forward angles where diffraction, dependent mostly on particle cross-sectional area, has a controlling influence [92]. The largest uncertainty with this assumption is, therefore, for larger particles in the backward scattering direction [93]. Certain modeled phase function features, such as the local maxima around 90° to 120° observed for some particle subpopulations with high refractive index (see Fig. 7), are the result of spherical lensing effects and are not likely observed in nature. These drawbacks of the spherical assumption do not belie our key results, however.

As was mentioned in Section 6, the inversion result does not change whether or not the “nonrealistic” subpopulations are included in the kernel functions. In other words, the method should be able to pick up a realistic subpopulation representation so long as it is introduced. Because volume scattering by small particles (<0.2 \( \mu \)m) is modeled well with Mie theory [92], finding that the VSP subpopulation is always a significant component of backscattering is likely a robust conclusion. The finding that larger bubbles (which are, presumably, effectively modeled with Mie theory) are usually a significant contributor to scattering in coastal waters is also considered robust [9]. These two claims are corroborated by the results of a recent study [94], where the measured VSFs in a surf zone were inverted for three particle subpopulations: VSPs, coated bubbles, and mineral particles of asymmetric hexahedron shapes [95]. We found the VSP subpopulation was consistently present in the inversion results whether the mineral particles were modeled as spheres or asymmetric hexahedra, and that the mineral subpopulations were surprisingly consistent regardless of the assumed shape. Also the bubble subpopulations agree well with concurrent acoustic attenuation data [96]. Despite uncertainties in the relative amount of backscattering by larger particles, the observation of an overall bimodal PSD is not expected to suffer from biases from the spherical assumption, especially since variability in the large size fractions was relatively restricted (Fig. 12). Junge slopes computed over ranges inclusive of larger particles were consistent with theoretical predictions and expectations based on size distribution measurements that have been made in coastal waters [97]. Our largest uncertainty is likely the modeling of highly complex phytoplankton subpopulations, which may have substantial species-to-species variability in phase function shapes [98–100]. All current empirical assessments of scattering by phytoplankton, however, have not been able to account for background scattering in the culture, which undoubtedly has some contribution in the backward direction. Clearly, more research is needed to better elucidate the scattering properties of these particles.

Future work will pursue VSF inversions that can account for some of the particle shape dependencies that affect phase function shapes. We are also adapting the inversion technique to other VSFs.
measurements collected in diverse environmental conditions.

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References

6. M. S. Twardowski, C. Moore, J. Sullivan, M. Slivkov, S. Freeman, and J. R. V. Zaneveld, are preparing a manuscript to be called “Volume scattering functions for selected ocean waters: revisited.”


68. C. F. Bohren and D. R. Huffman, Absorption and Scattering of Light by Small Particles (Wiley, 1983).


79. M. L. Longo, Department of Chemical Engineering and Materials Science, University of California, Davis, Calif., USA (personal communication, 2010).


92. M. I. Mishchenko, L. D. Travis, and A. A. Lacis, Scattering, Absorption, and Emission of Light by Small Particles (Cambridge University, 2002).


