

# The Backscattering Enigma in Natural Waters

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## LONG TERM GOALS

One of the fundamental problems in ocean optics over the past several decades has been a lack of understanding of the source of backscattering in the ocean. Because of experimental limitations and the limitations in available theoretical models, our knowledge of the causative agents for backscattering remains poor. Experimentally, we have been limited by a lack of scattering sensor instrumentation and a methodology for routine measurement of the submicron particle size distribution. Theoretically, most models have used Mie theory with the hope that natural particles of complex shape and structure can be approximated well by homogeneous spheres. For the Navy, poorly parameterized backscattering compromises applications involving the interpretation of passive and active optical detection methods. This is particularly true in coastal regions where current inversion models fail because the effects of changing particle composition are not adequately understood. Our long term goal is to better understand the sources and distribution of backscattering in natural waters. Related to this specific project goal, my larger research goals are to better understand the physical, biological and chemical mechanisms that control the spatial-temporal distribution of phytoplankton populations and dynamics, and how these same mechanisms affect the optical structure and fine-structure (e.g. thin-layers) in natural ecosystems.

## OBJECTIVE

The apparent enigma of oceanic backscattering lies in the fact that available experimental and theoretical results do not agree. Stramski and Kiefer (1991) modeled the backscattering of representative populations of microorganisms in the ocean with Mie theory and concluded that microorganisms could only account for ~20% of the backscattering necessary to explain the upwelling radiance observed by satellites and in situ measurements. Our objective is to evaluate three hypotheses that have emerged to explain the “missing backscattering.”

We call the first hypothesis the *detrital hypothesis*. Stramski and Kiefer (1991) postulated that this missing agent is high concentrations of submicron detrital particles with high refractive index. Furthermore, the authors postulated that these detrital particles should covary with chlorophyll containing particles to satisfactorily explain the observation that satellite ocean color measurements are reasonably successful in determining chlorophyll concentrations from upwelled radiance.

We call the second hypothesis the *complex particle hypothesis*. Zaneveld et al. (1974), Kitchen and Zaneveld (1992), and Zaneveld and Kitchen (1995) asserted that the bulk refractive index (and the relative amount of backscattering) for phytoplankton would be higher than that typically used in models

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based on Mie theory because these organisms usually, particularly in coastal regions, have a hard shell (silicious frustule in the case of diatoms). It is also possible that the scattering properties of phytoplankton particles are poorly modeled as perfect spheres. Thus, the hypothesis here is that the “missing backscattering” is due, at least in part, to deficiencies in modeling efforts.

We call the third hypothesis the *mineral hypothesis*. Twardowski et al. (2001) suggested that the unexplained backscattering could result from a background population of small inorganic particles. These particles are present throughout the ocean, primarily from biogenic and Aeolian sources, and exhibit strong relative backscatter due to their high refractive index. In this case, the “missing backscattering” is due to an inorganic mineral background, with a fluctuating phytoplankton population superimposed on that background.

## APPROACH

### *Testing the complex particle hypothesis*

A key focus over the last year has been determining the scattering properties of phytoplankton populations and understanding how best to model these properties in a bulk sense. Once we have reasonable success with the forward problem, we will then be able to attempt to carry out the inverse problem. Work has been carried out in the laboratory on various cultures of phytoplankton. Field data has also been used to test this hypothesis under conditions where phytoplankton specific scattering data may be reasonably approximated. Phytoplankton for culturing are being isolated from Narragansett Bay and surrounding waters to avoid using perpetually cultured organisms that can develop morphological inconsistencies with their naturally occurring cousins. Table 1 lists the measurements being made.

**Table 1. Measurements to test backscattering hypotheses.**

| Physical parameter                                                     | Instrument                            | Specific                                                                  |
|------------------------------------------------------------------------|---------------------------------------|---------------------------------------------------------------------------|
| Volume Scattering Function, $\beta$ ( $\text{m}^{-1} \text{sr}^{-1}$ ) | WET Labs ECO sensors                  | $\beta_p$ ([100°, 117°, 125°, 150°], [450, 532, 650 nm])                  |
|                                                                        | Goniometer                            | $\beta_p$ ([~15°-155° @ 1 resolution], [532 nm])                          |
| Attenuation, absorption, scattering coefficients ( $\text{m}^{-1}$ )   | WET Labs AC9 and ACS instruments      | $a_{pg}$ , $a_g$ , $a_p$ , $b_p$ , $c_{pg}$ , and $c_p$ at 84 wavelengths |
| Particle Size Distributions (PSDs)                                     | LISST-100                             | Sizes ranging from ~1.3-250 $\mu\text{m}$                                 |
|                                                                        | Electrical resistance particle sizing | Sizes ranging from ~0.6-12 $\mu\text{m}$ and ~2.4-48 $\mu\text{m}$        |
|                                                                        | Light Microscopy                      | Sizes ranging from ~1-250 $\mu\text{m}$                                   |
|                                                                        | SEM                                   | Sizes ranging from ~0.01-100 $\mu\text{m}$                                |
| Bulk refractive index                                                  | Goniometer                            | Index-matching immersion method                                           |

Closure between measured particle characteristics and optical measurements including backscattering is being tested with theoretical models to help understand and interpret results in the context of the backscattering enigma.

A component of our approach is the development of a new microphotometric method for determining the backscattering properties of individual particles. This technique allows evaluation of backscatter from specific structural components of a particle, providing extraordinary insight into the causative agents of bulk backscattering.

### *Testing the detrital and mineral hypothesis*

The detrital and mineral hypotheses must be tested in samples from natural waters, preferably case 1 type waters without terrigenous influence. The measurements listed in Table 1 are being made in the field and the presence of detrital material and inorganic material is being determined directly (e.g., PSD and SEM analyses) and indirectly (e.g., inversion of optical measurements). Backscattering budgets will be carried out to determine significant sources.

Discrete samples will also be sequentially filtered and the VSFs will be measured in the different size fractions with the goniometer. Samples will also be filtered in-situ with real-time instrumentation including the AC9 by attaching capsule filters to the intakes of the sensors. Isolating the scattering characteristics of individual size fractions, especially in the submicron range, will allow discrimination of the PSD component(s) thought to be responsible for backscattering under the detrital hypothesis.

Standard gravimetric determinations of TSM are also being made to determine relative and absolute concentrations of minerals in samples. Concentrations and isolated scattering characteristics of the mineral fraction will allow an assessment of the overall role of mineral particles in the oceanic backscattering process. Particles collected on filters for TSM analyses are also being subjected to SEM and elemental analysis with a mass spectrometer integrated in an SEM microscope. This analysis can directly determine the type and concentration of individual particles on the filters. Elemental analyses leave virtually no doubt as to mineral type and consequently the refractive index of that particle group (from published tables in CRC). An SEM image analysis method was developed to obtain PSDs from particles collected on a filter.

## **WORK COMPLETED**

- A microphotometric method was developed to assess the backscattering properties of individual particles and internal structures
- Phytoplankton isolations, large volume phytoplankton culturing and optical sampling methods for laboratory experiments continued to be developed and conducted at URI during 2006. Five lab experiments have been conducted to assess the complex particle hypothesis using cultures of the diatom *Chaetoceros socialis*, the dinoflagellate *Gyrodinium instriatum*, the diatom *Thalassiosira weissflogii*, the diatom *Chaetoceros teres*, and the diatom *Stephanopyxis turris*. The experiment with the latter organism was carried out through a full phytoplankton growth cycle.
- To further enhance the field sampling effort for the Enigma project, I, with the help and input of the Co-PIs on the project, submitted a proposal to the Rhode Island Endeavor Program titled “Blue water optical studies in support of Rhode Island private industry, URI academics, RI Public Schools and the U. S. Navy” and successfully received funding for 5 days of ship time on the UNOLS vessel *R/V Endeavor*. This cruise took place during October 2005 and was specifically designed to allow all of the project PIs to participate in a coastal to open ocean (blue water) transect cruise to jointly collect a

comprehensive data set for the project (example results from this cruise are shown in **Fig 5**). After the success of this first cruise, we again sought funding for ship time for the Enigma program. I, in conjunction with two other URI faculty members, submitted a second proposal to the Rhode Island Endeavor Program titled “An examination of phytoplankton biodiversity, physiology and optical characteristics from coastal to open ocean ecosystems: implications to remote sensing, biogeochemical cycles and environmental change.” This proposal was recently funded and can provide the Enigma project with 8 days of blue water ship time in the North Atlantic Ocean during March of 2007.

- As a Co-PI in the ONR sponsored DRI: Layered Organization in the Coastal Ocean (LOCO), I recently completed a three week field experiment (July 7 - July 30, 2006) in Monterey Bay, CA. The primary goal of this project was to examine and understand the dynamics of thin layers in the coastal ocean. Thin layers normally consist of dense concentrations of phytoplankton that have a significant impact on the underwater optical field and backscattering signal. Fine-scale optical measurements made during the LOCO field experiment that are pertinent to the Enigma project included hyper-spectral absorption and attenuation (both particulate and dissolved), angular and spectral backscattering, and phytoplankton and particle identifications within layers and the surrounding water column. An example of these recent field data are shown in **Fig. 1**. The combined data from the 2005 and 2006 LOCO field program represents over 2000 vertical profiles of optical information that will be analyzed and applied towards Enigma project goals where appropriate.
- A new method to examine the scattering characteristics of different in-situ particle size fractions was tested during the 2006 LOCO field experiment.
- A manuscript is in preparation for publication, entitled “The Contribution of Phytoplankton to Backscattering in Coastal Waters” with Twardowski and Sullivan leads.
- A manuscript is in preparation for publication, entitled “Sources of Backscattering in the Southeast Pacific” with Twardowski lead.
- A manuscript is in preparation for publication, entitled “Relative Influences of Spectral Absorption and Backscattering on Remote Sensing of Chlorophyll” with Dierssen, lead.
- A manuscript is in preparation for publication, entitled “Optical Complexity in the Coastal Ocean” with Sullivan and Twardowski leads.

## RESULTS

*Previous measurements of phytoplankton scattering in the lab have critical errors.* In culture work, we found that the contribution of the background particle population to scattering, particularly at large angles, is substantial. By gently filtering the phytoplankton particles out of the culture solutions with a 10  $\mu\text{m}$  nitex screen, we were able to quantify the optical properties of the background. The background particle population had dramatically different relative (e.g., backscattering ratio) and absolute scattering properties. No previous study has considered the background solution in cultures.

*A new method to examine the scattering characteristics of different in-situ particle size fractions was assessed.*

During the 2006 LOCO field experiment, two hyperspectral AC-S were deployed, one unfiltered and one with a 5  $\mu\text{m}$  pre-filter concurrent with two AC-9s, one unfiltered and one with a 0.2  $\mu\text{m}$  pre-filter. This allowed us to assess the scattering (and modeled backscattering ratio, Sullivan et al. 2005) of the  $<5$   $\mu\text{m}$  and  $>5$   $\mu\text{m}$  particulate fractions. In the coastal waters of Monterey Bay, the  $<5$   $\mu\text{m}$  fraction accounted for  $\sim 10$  to 30% of the bulk chlorophyll, generally had similar or higher backscattering ratio than the total fraction (**Fig. 2**) and could account for  $\sim 30$  - 40% of the total particulate absorption and scattering (**Fig. 3**).

*Individual particle backscattering can be imaged with a new microphotometry technique.* A new backscattering imaging method was developed to better assess where the backscattering from single particles was originating. At this time, the method is qualitative, but has proven highly valuable in helping us to better understand sources of particulate backscattering and develop a strategy for better addressing the hypotheses presented on the previous page. Results from this work have indicated that a hard coat is the primary source of backscattering for a phytoplankton cell. This was not necessarily the case for all cells, however (e.g., *Stephanopyxis*). And some phytoplankton are nearly invisible in the backscattering image (e.g., *Chaetoceros*). Additionally, the backscattering from detrital material, in particular the so-called Transparent Exo-Polymers (TEP), had far higher relative backscattering than previously expected based on analyses of the interstitial water content and subsequent refractive index of these particles (Twardowski et al. 2001).

*Coastal phytoplankton have significantly higher backscattering than previously thought from Mie theory calculations.* From an integrated analysis that included field measurements, lab measurements and theoretical considerations, we have demonstrated that the complex morphology of coastal phytoplankton substantially increases their relative backscattering. The higher backscattering is primarily due to their hard coats. This is clear from the new microscope laser images of single particle backscattering and is also supported by the consistency with expectations based on coated sphere Mie theory modeling (**Fig. 4**). Overall, a good rule of thumb backscattering ratio for coastal phytoplankton is  $\sim 0.5\%$ , also supported by extensive field measurements. An approximate backscattering ratio expected from homogeneous sphere Mie theory calculations is  $\sim 0.02$ - $0.05\%$  (more than an order of magnitude lower than actual) for coastal phytoplankton species (Stramski et al. 2000).

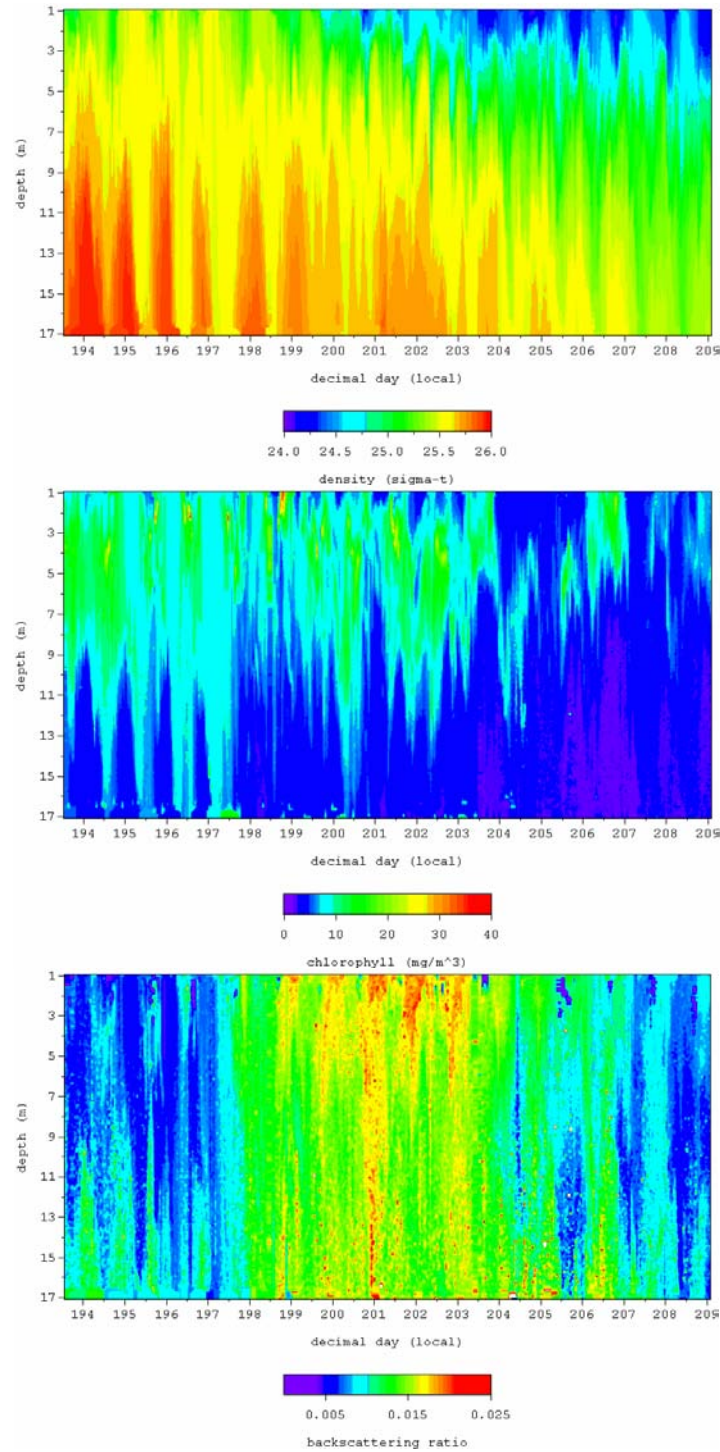
*Colloidal material accounted for  $\sim 50\%$  of the backscattering in the Northwest Atlantic.* The VSF of the colloidal fraction was measured in a sample from Northwest Atlantic surface water co-located with concurrent in situ ECO-VSF backscattering measurements (**Fig. 5**). The bench top goniometer was brought on the R/V Endeavor for these measurements. The backscattering from the colloidal fraction was about half that obtained with the real-time, in situ ECO-VSF sensor in the whole sample. This is the first time the contribution of colloids to backscattering in the ocean has ever been measured directly.

*Conventional remote sensing chlorophyll algorithms generally work because of selective filtering of light (i.e., spectral absorption) rather than selective backscattering, except when chlorophyll concentrations exceed  $\sim 1 \text{ mg m}^{-3}$ .* Radiative transfer modeling of remote sensing reflectance from the IOPs has shown that phytoplankton backscattering does not need to dominate a water-leaving radiance signal for chlorophyll algorithms to work. The influence of backscattering from phytoplankton only starts to exceed the influence from spectral absorption when chlorophyll concentrations are greater than about  $1 \text{ mg m}^{-3}$ . These modeling results are consistent with experimental conclusions from the field and lab.

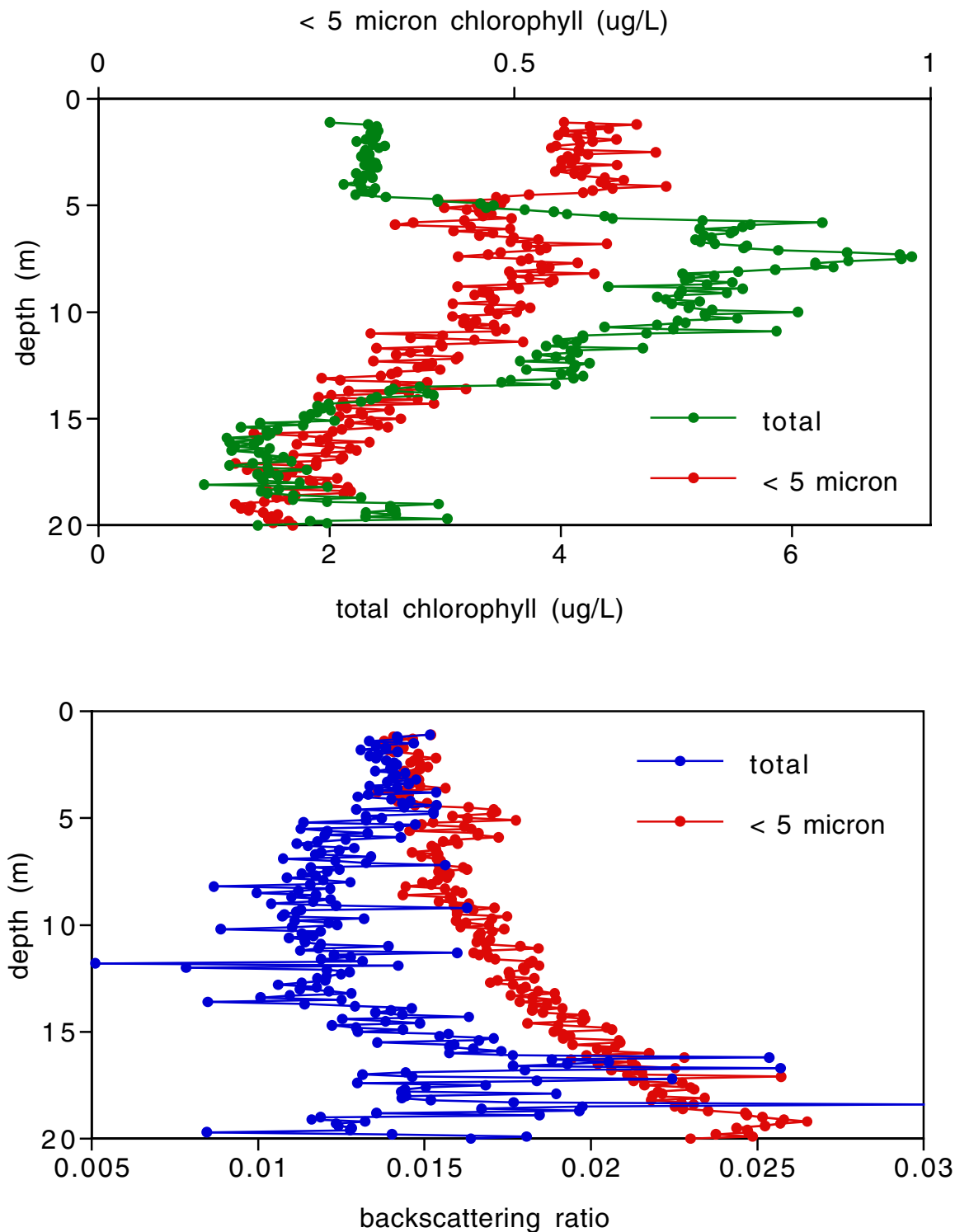
For additional results related to this project, refer to the annual reports of co-I's Mike Twardowski and Heidi Dierssen.

## **IMPACT/APPLICATIONS**

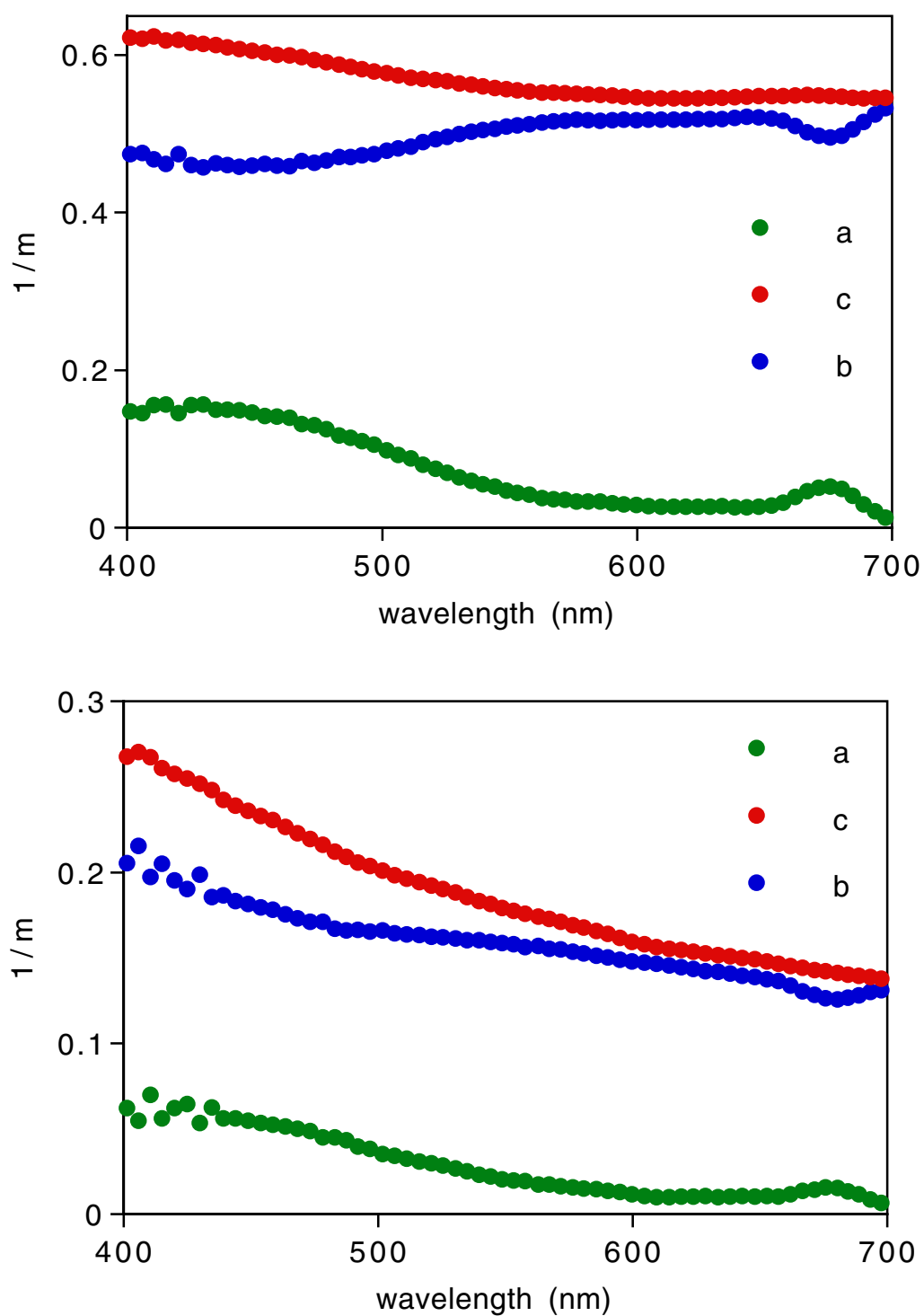
Progress and results represent important steps toward understanding the sources of backscattering in the ocean. Naval applications requiring an understanding of the optical properties of water will benefit from this work. Since the optical properties of seawater are driven by the composition of suspended materials, we must understand this link to know how the underlying biogeochemical processes influence seawater optics. Applications directly influenced by seawater backscattering include lidar, laser line scanning systems, and remote sensing. Oceanographic research implications of this work include better inversion models for estimating the composition and concentration of suspended particles from optical sensors.



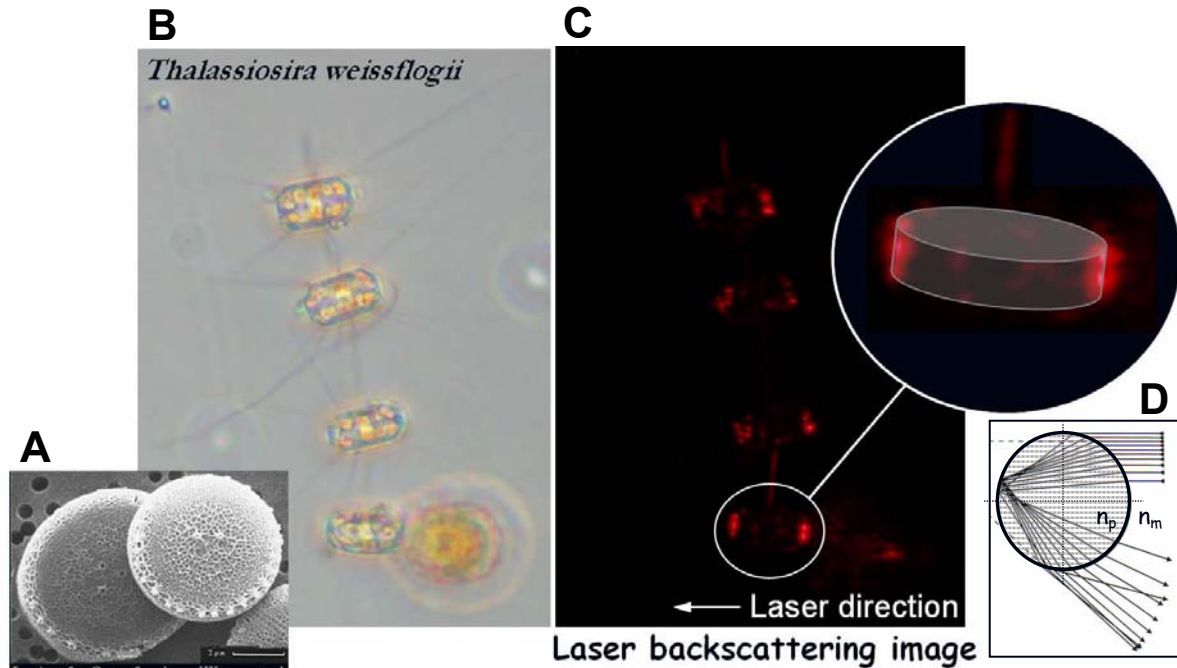
**Figure 1. Color contour plots of density (top), chlorophyll (middle) and backscattering ratio (bottom) representing over two weeks of continuous hourly data through the entire 2006 LOCO experiment in Monterey Bay, CA. Backscattering ratio varied greatly as different populations of phytoplankton and particles moved through the study area.**



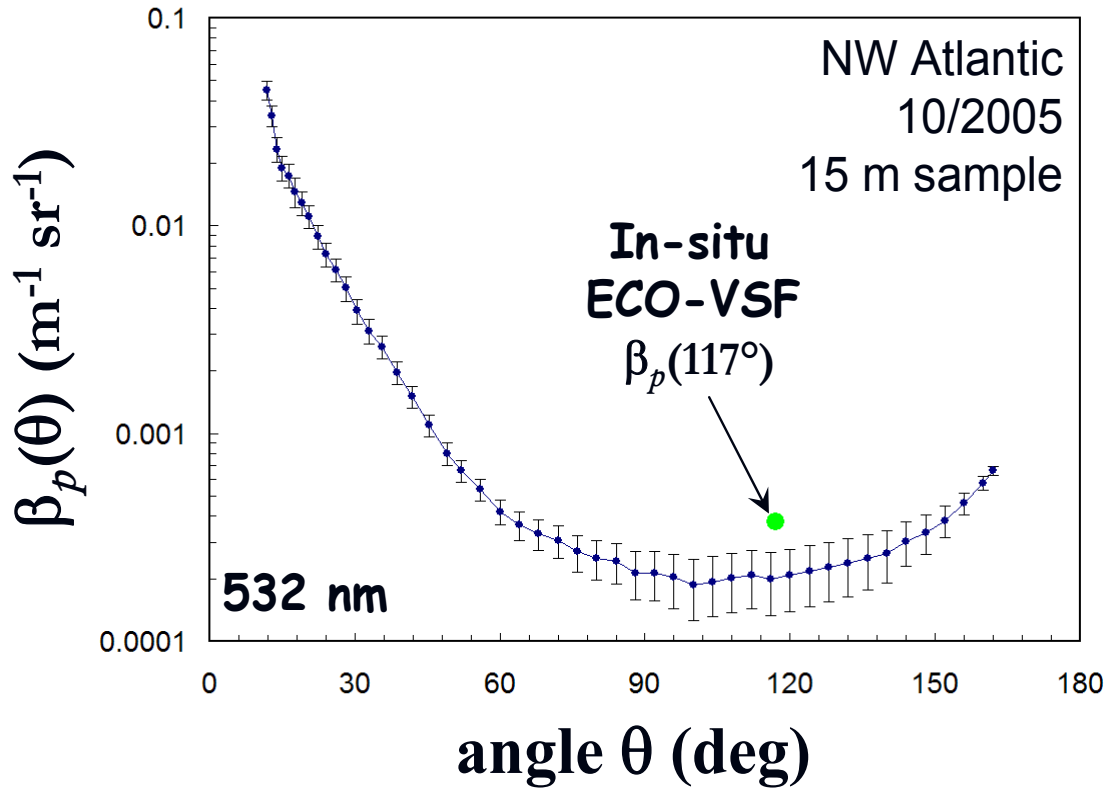
**Figure 2. Representative vertical profiles of chlorophyll a concentration (top panel) and backscattering ratio (bottom panel) for the total and <5  $\mu\text{m}$  fractions measured during LOCO 2006. Note that the chlorophyll data has different scales. These profiles show that the <5  $\mu\text{m}$  fraction composed between 10 to 30% of the total chlorophyll a and generally had similar or higher backscattering ratio than the unfiltered (total) fraction.**



*Figure 3. A representative example of absorption, attenuation and scattering spectra for the total (top panel) and <5  $\mu\text{m}$  (bottom panel) particulate fractions from the vertical profiles shown in Figure 2. These data show that the <5  $\mu\text{m}$  fraction can account for ~ 30 - 40% of the total particulate absorption and scattering.*



**Figure 4.** The importance of the “hard coat” in backscattering by coastal phytoplankton is illustrated by this microscopic backscattering image from a chain of *Thalassiosira weissflogii* cells. An SEM image (A) shows the heavily silicified hard coat (frustule) of the diatom cells. Images (B) and (C) are of the same field-of-view, with the (B) image obtained with conventional light transmission microscopy and the (C) image obtained from laser backscattering at ~140 degrees. Backscattering “hot spots” were observed at the outer edges of the frustule from ray focusing, analogous to the effect achieved with a focusing lens (D). The observed effect results from significant refraction of the incident rays that is only possible if there is a relatively high refractive index difference between the medium and the particle (or particle shell in this case). These images provide, for the first time, experimental evidence of the importance of complex structure in particle backscattering.



**Figure 5. High-resolution VSF of the colloidal fraction of seawater collected from the NW Atlantic. The VSF was determined with a bench top goniometer using a new statistical large particle ( $> a$  few  $\mu\text{m}$ 's) rejection algorithm. The green data point was collected with an in-situ ECO-VSF sensor and represents the volume scattering at 117 degrees by the total particulate fraction. Assuming scattering at 117 degrees is a reasonable proxy for integrated backscattering (Oishi 1990; Boss and Pegau 2001), the colloidal fraction comprised about half the particulate backscattering. These results verify experimentally for the first time that colloids represent a significant, if not dominant, portion of particulate backscattering.**

## TRANSITIONS

I have continually worked with WET Labs in laboratory and field testing of the hyper-spectral absorption-attenuation sensor, the AC-S. This work included evaluating and improving the ac-s data acquisition, processing software and data analysis techniques. Using the AC-S, we determined the hyper-spectral coefficients for the temperature and salt dependent absorption of pure seawater in the visible and near-IR spectral region and this work was recently published in 2006 (see publications list). These coefficients are vital for the normalization of optical measurements using the AC-S and potentially important to other hyper-spectral instrumentation measurements. This work has resulted in the rapid inclusion of instrument improvements and distribution of correction coefficients for the commercial AC-S, thus making them available to the broader community. The coefficients are being made available to the community through WET Labs and the Applied Optics publication.

I have continued in 2006 to work with Andrew Barnard of WET labs and Percy Donaghay of URI on a Navy DoD SBIR titled “The **Miniaturized Autonomous Moored Profiler** (Mini-AMP)”. This instrument is the commercial version of the ORCAS profiler (Donaghay and Sullivan) that was used during the LOCO DRI field experiments (see **Fig 1**). This work entails consulting on design improvements and payload options, deployment issues and beta testing prototype designs. The results of this work will benefit the community with a robust and well designed commercial instrument.

Co-I Mike Twardowski has agreed to teach a short course on Observational Approaches in Ocean Optics at the next Ocean Optics conference in Montreal in 2006, I will assist Mike with this class by guest lecturing during the course on the topic of “Optical measurements from autonomous profilers”.

## RELATED PROJECTS

The LOCO DRI ‘thin-layers’ project will yield an exciting data set to examine the backscattering characteristics of dense populations of different phytoplankton species under a variety of ecosystem conditions and co-occurring particle types. As part of this project, the particle characteristics within thin-layers will be optically quantified using a variety of quantitative methods and instrumentation, including examining changes in particulate absorption and attenuation spectra and using scattering inversion techniques to estimate particle size distribution and the bulk refractive index of particles.

## PUBLICATIONS

**Sullivan, J. M., M. S. Twardowski, J. R. Zaneveld, C. Moore, A. Barnard, P. L. Donaghay & B. Rhoades** (2006) -The hyper-spectral temperature and salinity dependent absorption of pure water, salt water and heavy salt water (D<sub>2</sub>O) in the visible and near-IR wavelengths (400 - 750 nm). *Applied Optics*, 45(21): 5294-5309. [refereed]

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