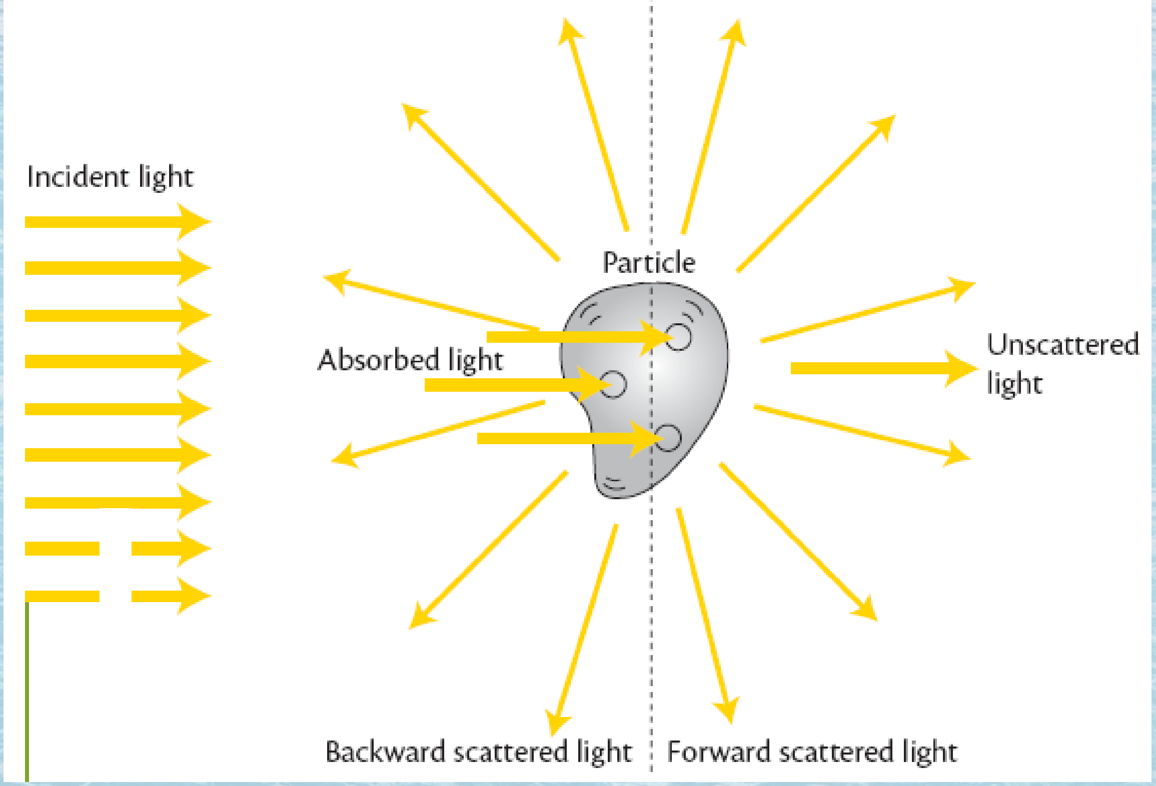
The link between particle properties and their IOPs or How do we model oceanic particles?

To be able to use optical measurements to study oceanic particles (and dissolved materials) we need to develop an understanding of how light interacts with matter.

Corollary: If optical properties of particles did not vary as a consequence of interaction with oceanic particles it would be useless to use optics as a tool to study them.

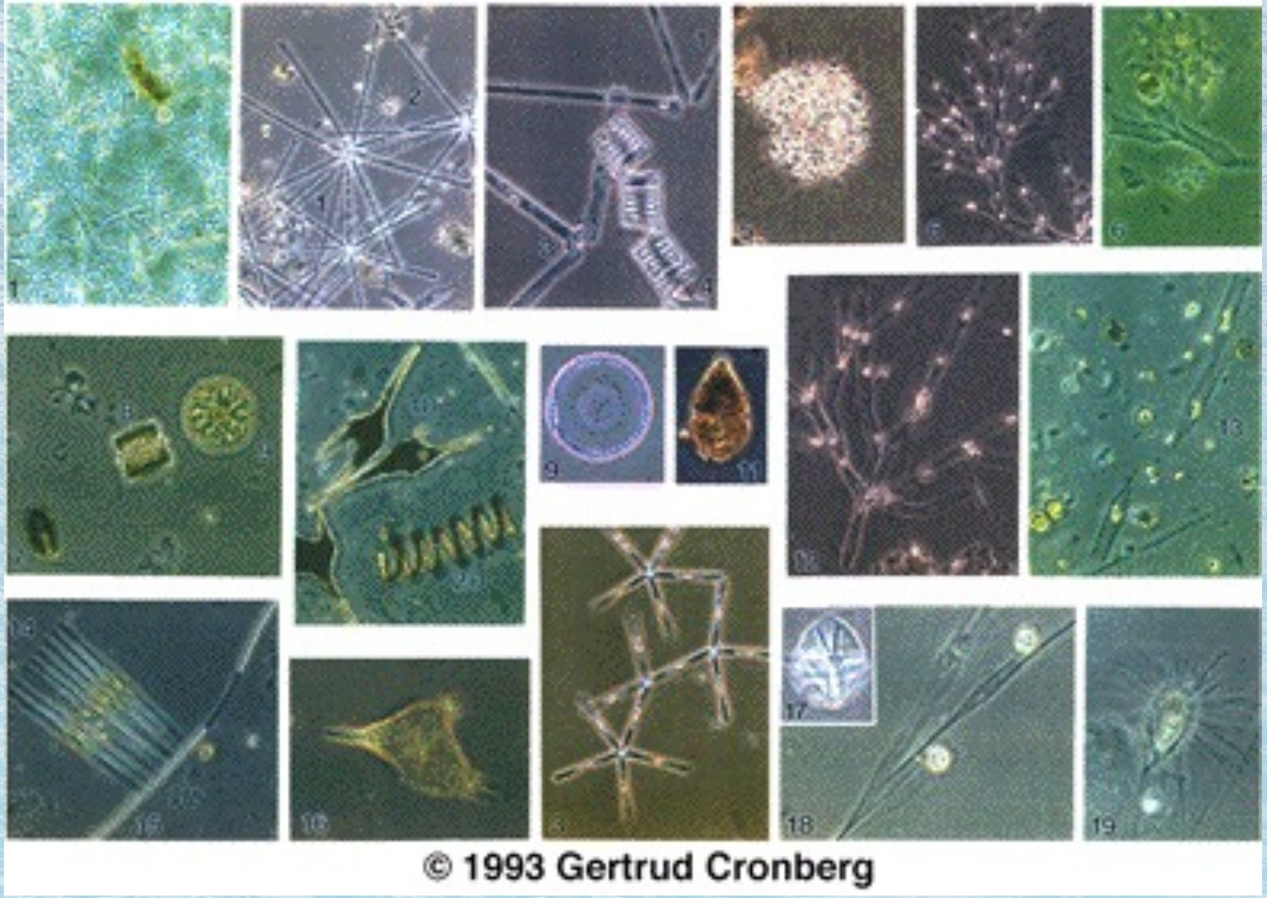
The problem (Bohren and Huffman, 1983):

Given a particle of a specified size, shape and optical properties that is illuminated by an arbitrarily polarized monochromatic wave, determine the electromagnetic field at all points in the particles and at all points of the homogeneous medium in which it is embedded.

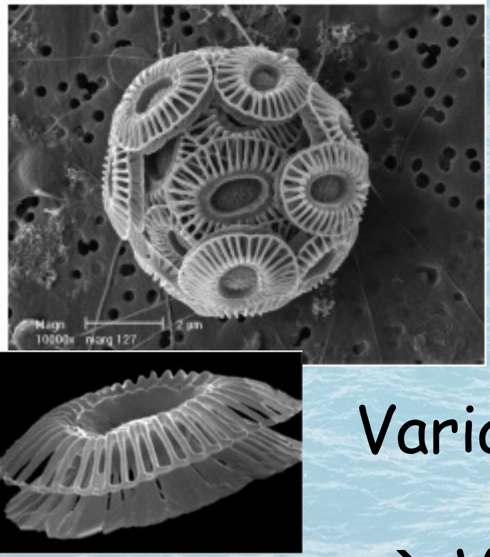


What particles scatter/absorb light in the ocean?

Phytoplankton:



© 1993 Gertrud Cronberg

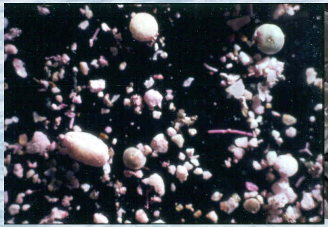


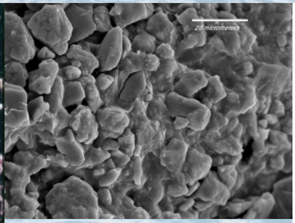
Variable in shape, size and pigment composition.

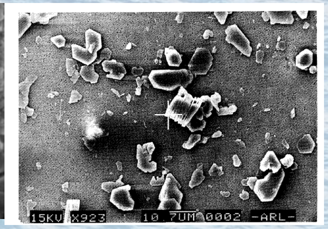
→ Variable in scattering and absorption properties

What particles scatter/absorb light in the ocean?

Non-algal particles: Organic and inorganic.


Sand

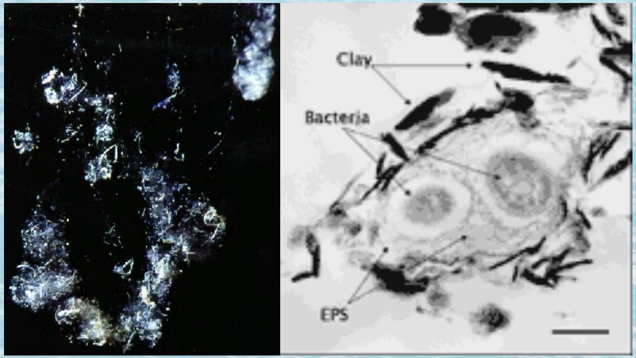

Silt



clay


http://www.aad.gov.au/default.asp

Aggregates:



→ Variable in scattering and absorption properties

What cell properties are likely to affect light?

Size – characteristic length scale of particle?

composition – characterized by the bulk index of refraction of the particle. How different is the light speed in it compared to ambient water?

Shape – departure from sphere – macro/ how smooth –micro (cocolithophores).

internal structure – inhomogeneities within the particle (cell-wall, cytoplasm, chloroplast, vacuole).

'Packing' – How 'solid' is the particle. Ratio of interstitial water volume to total volume.

Why may we care about optical theory and Mie's solution?

These solutions provide a calibration to our sensors (LISST, b_b, flow-cytometers).

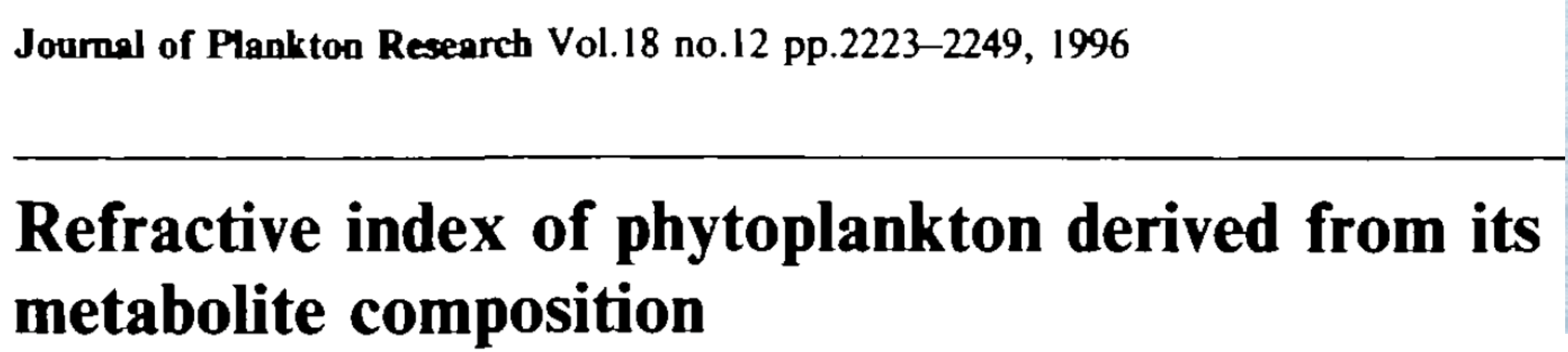
In addition, for a given concentration of particles of a given *size/wavelength* ratio and *index of refraction* we expect a given signal (some are shape independent).

Rebecca Green – used Mie theory to analyze flow-cytometer data, assigning size based on forward and side scattering of single cells.

Giorgio Dall'Olmo – used Mie theory to analyze diel cycles in optical properties.

Tiho Kostadinov – used Mie theory to look at effects of changes of population PSD on Rrs.

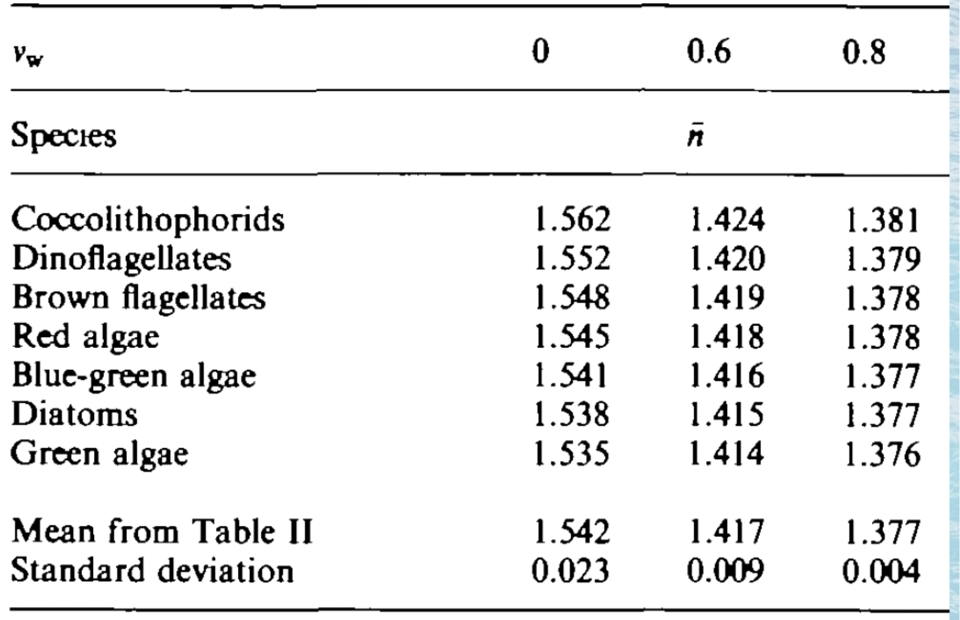
The forward problem



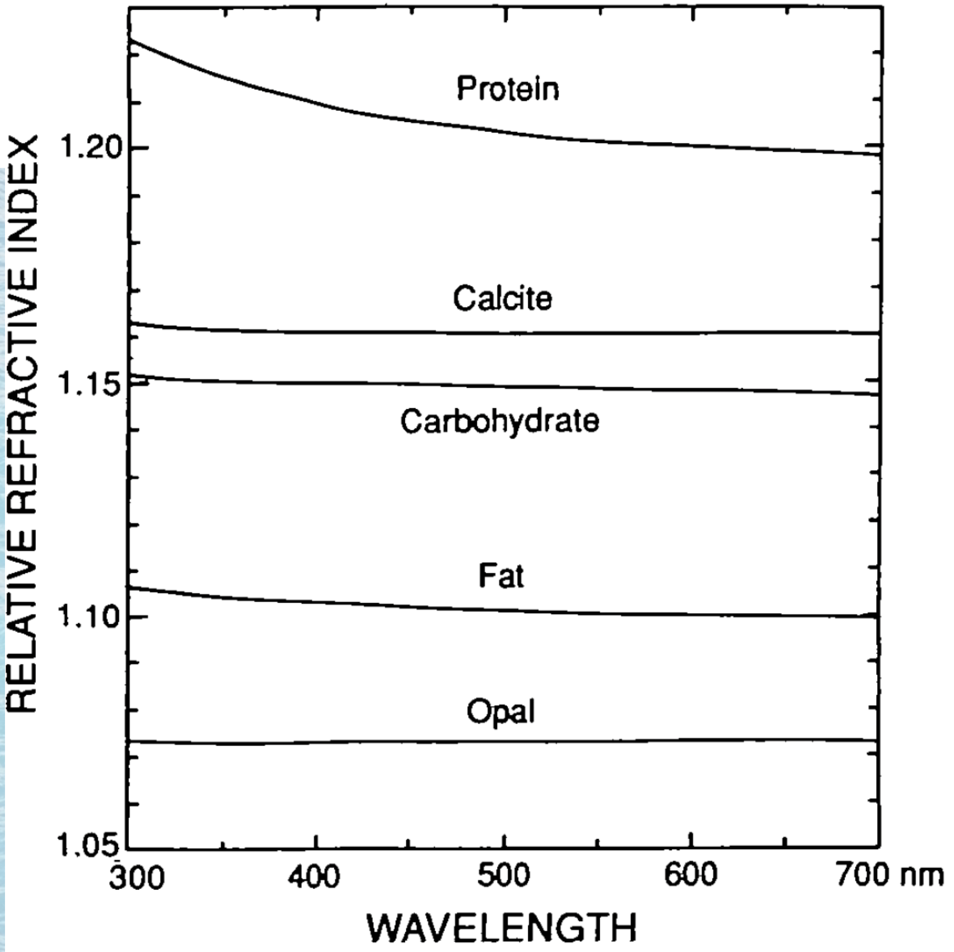
Journal of Plankton Research Vol.18 no.12 pp.2223–2249, 1996

Refractive index of phytoplankton derived from its metabolite composition

Eyvind Aas



v_w	0	0.6	0.8
Species		$\bar{n}$	
Coccolithophorids	1.562	1.424	1.381
Dinoflagellates	1.552	1.420	1.379
Brown flagellates	1.548	1.419	1.378
Red algae	1.545	1.418	1.378
Blue-green algae	1.541	1.416	1.377
Diatoms	1.538	1.415	1.377
Green algae	1.535	1.414	1.376
Mean from Table II	1.542	1.417	1.377
Standard deviation	0.023	0.009	0.004

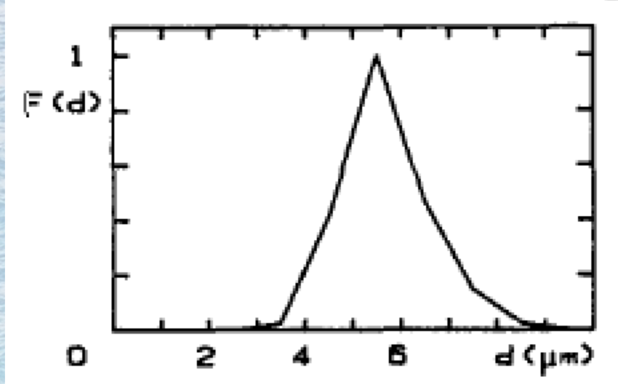


→ Water content (80+/-10%) is biggest driver. n_{phyto}=1.06+/-0.04

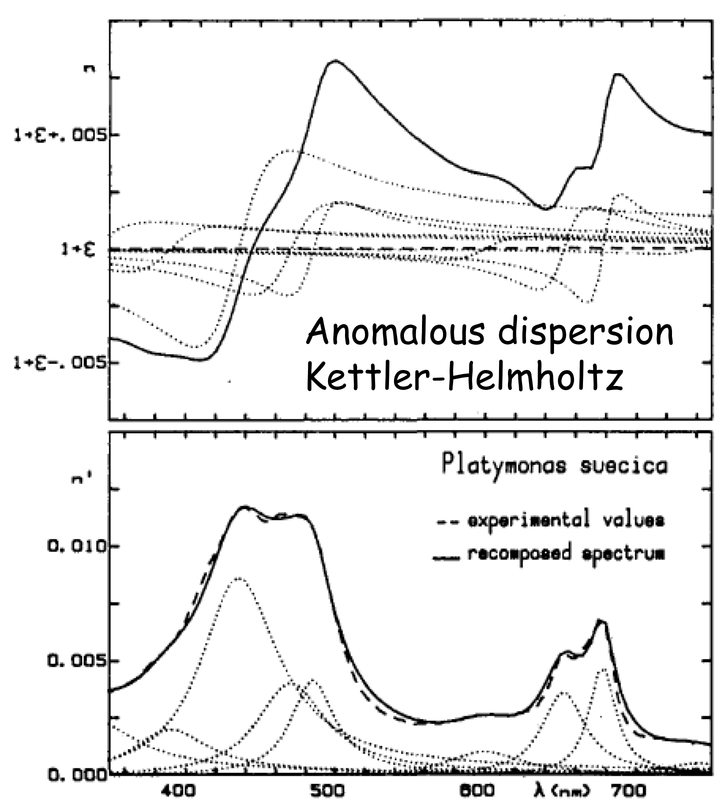
The inverse problem

Find the optical constants consistent with absorption measurements of a phytoplankton culture (Bricaud and Morel, 1986)

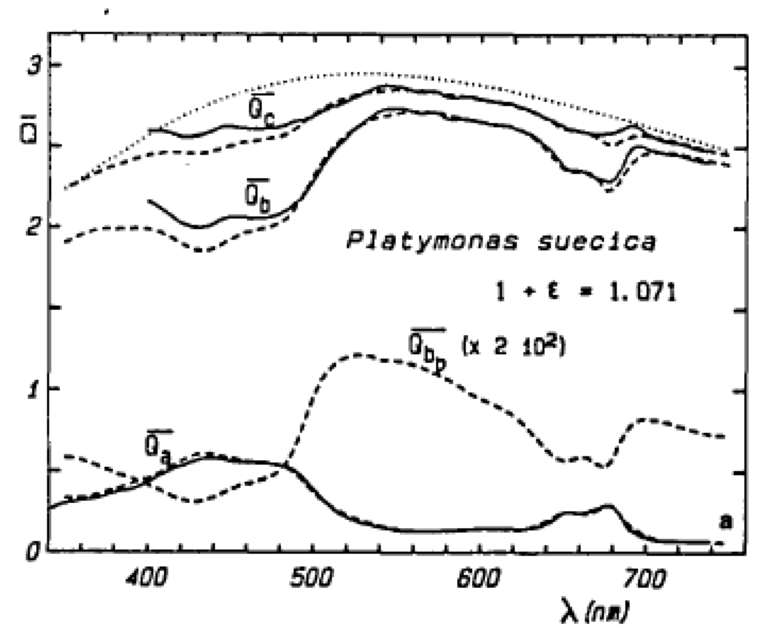
Size distribution:



Absorption measurements:

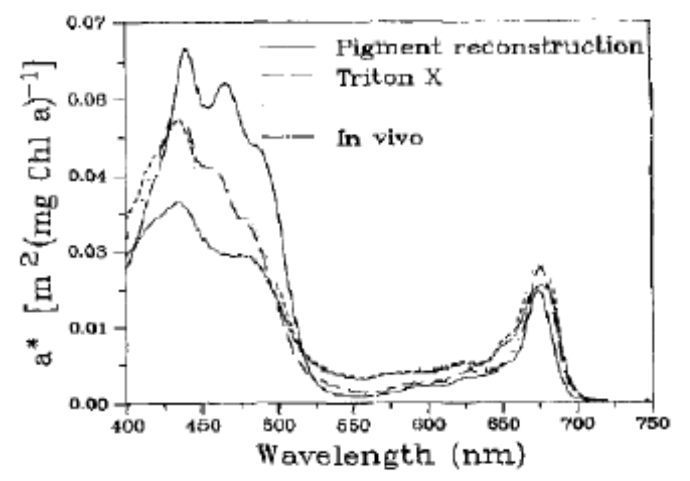

Anomalous dispersion
Kettler-Helmholtz

Results:

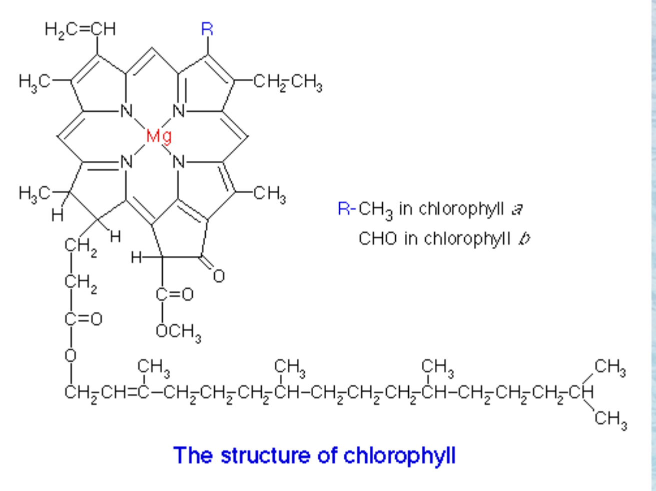


Size effect on absorption – pigment packaging

a_ϕ vs [Chlorophyll]

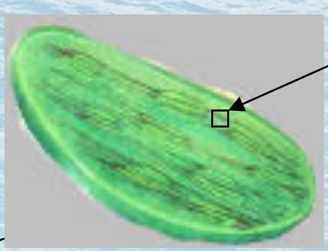


Sosik & Mitchell 1991



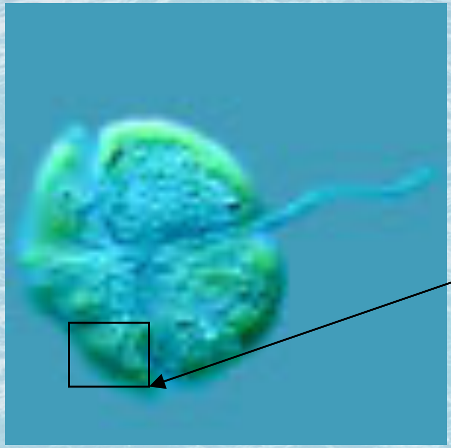
The structure of chlorophyll

http://chaitanya1.wordpress.com/2007/07/09/strawberries/



chloroplast

chlorophyll



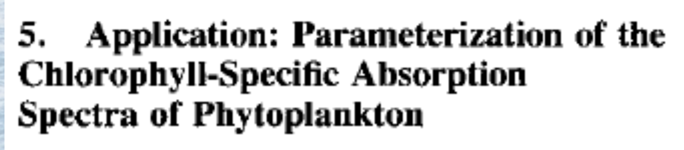
cell

Packaging: $a^*=a/[\mathrm{chl}]$ is function of size and [chl].

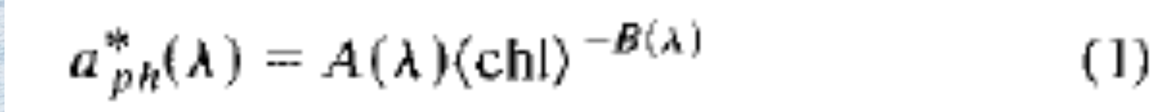
Same is true for other pigments. Duysens (1956)

Slide from Collin

Large, more packaged cells, tend to occur where [chl] is higher.

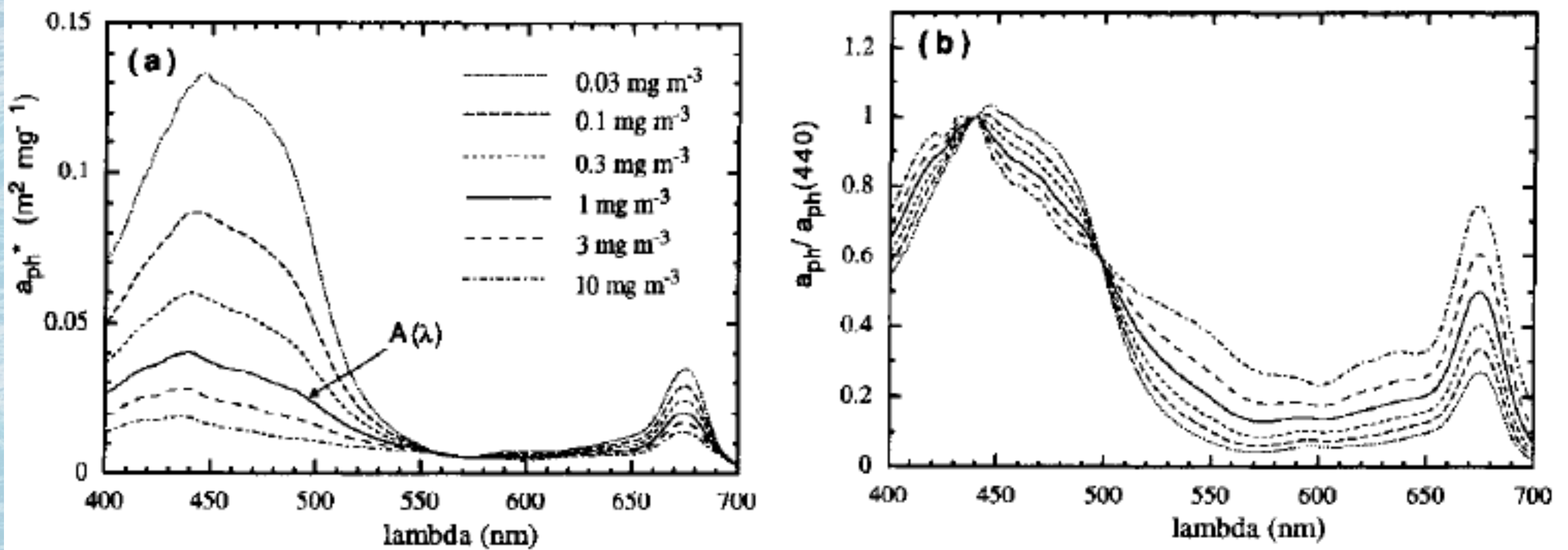


5. Application: Parameterization of the Chlorophyll-Specific Absorption Spectra of Phytoplankton



$$a^*_{ph}(\lambda) = A(\lambda)\langle\mathrm{chl}\rangle^{-B(\lambda)} \qquad (1)$$

'Mean' a_ϕ as function of [chl]

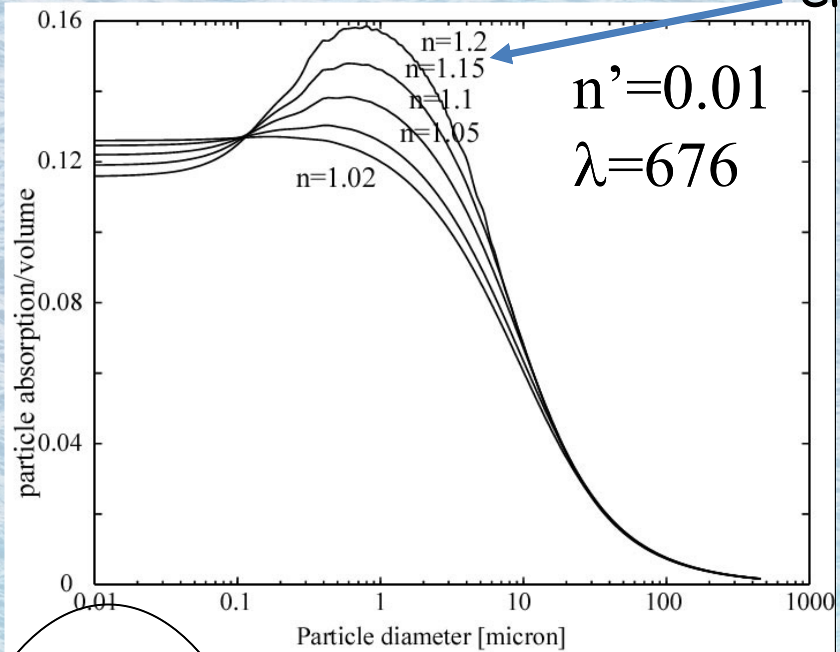


Bricaud et al., 1995

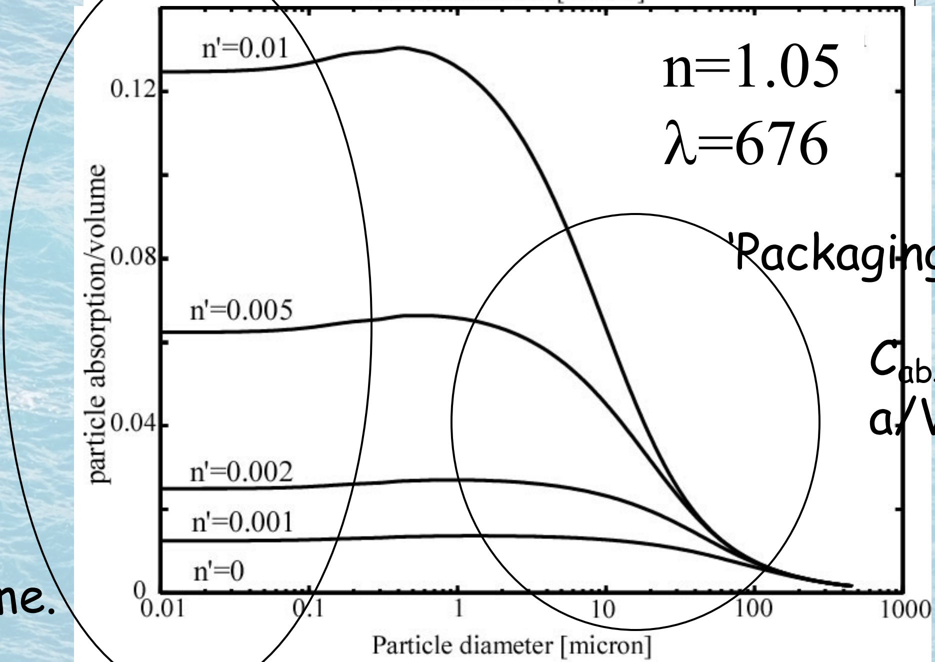
Dependence of IOP on properties of particles

Mie calculations reveal pigment packaging: Absorption per cell volume.

$n' = a_{pure}\lambda/4\pi$



enhancement



'Packaging'

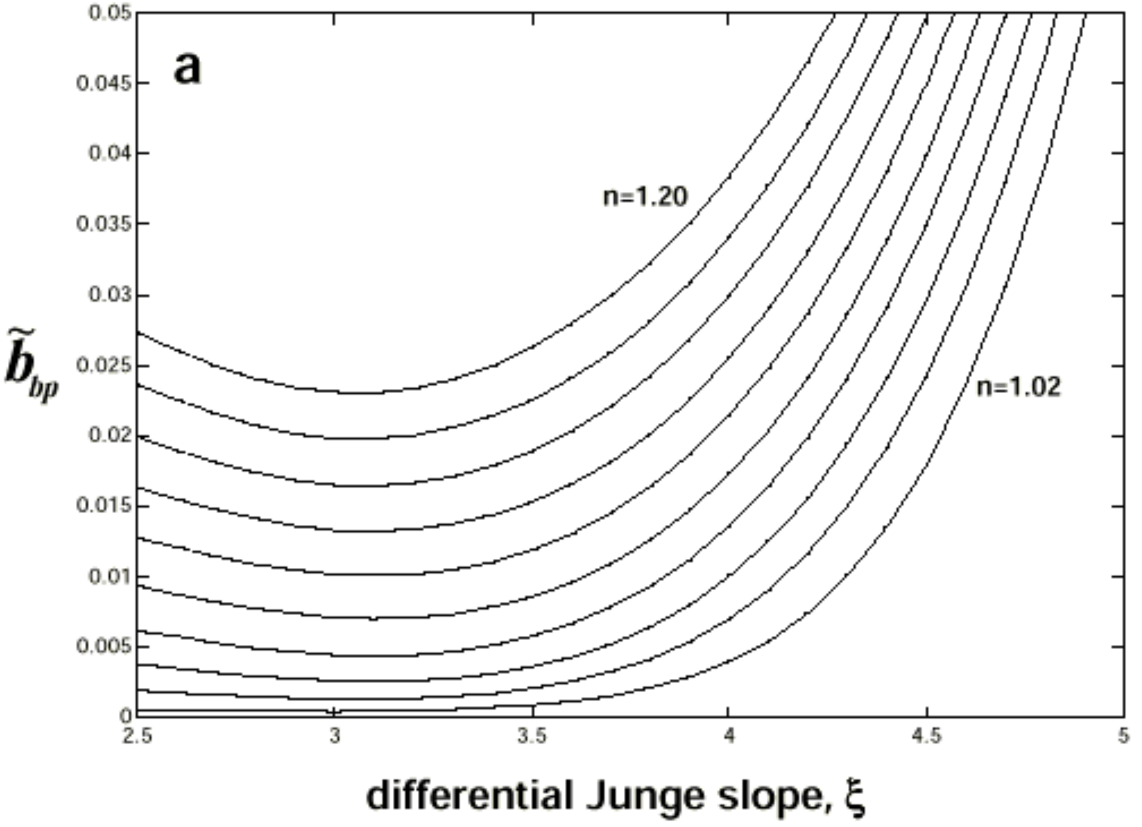
C_{abs} = area→ $a/V \propto 1/D$

Molecular absorption ∝ volume.

Backscattering ratio- sensitivity to composition and size

backscattering ratio $\left(\tilde{b}_{bp} \equiv b_{bp}/b_p\right)$ depends on:

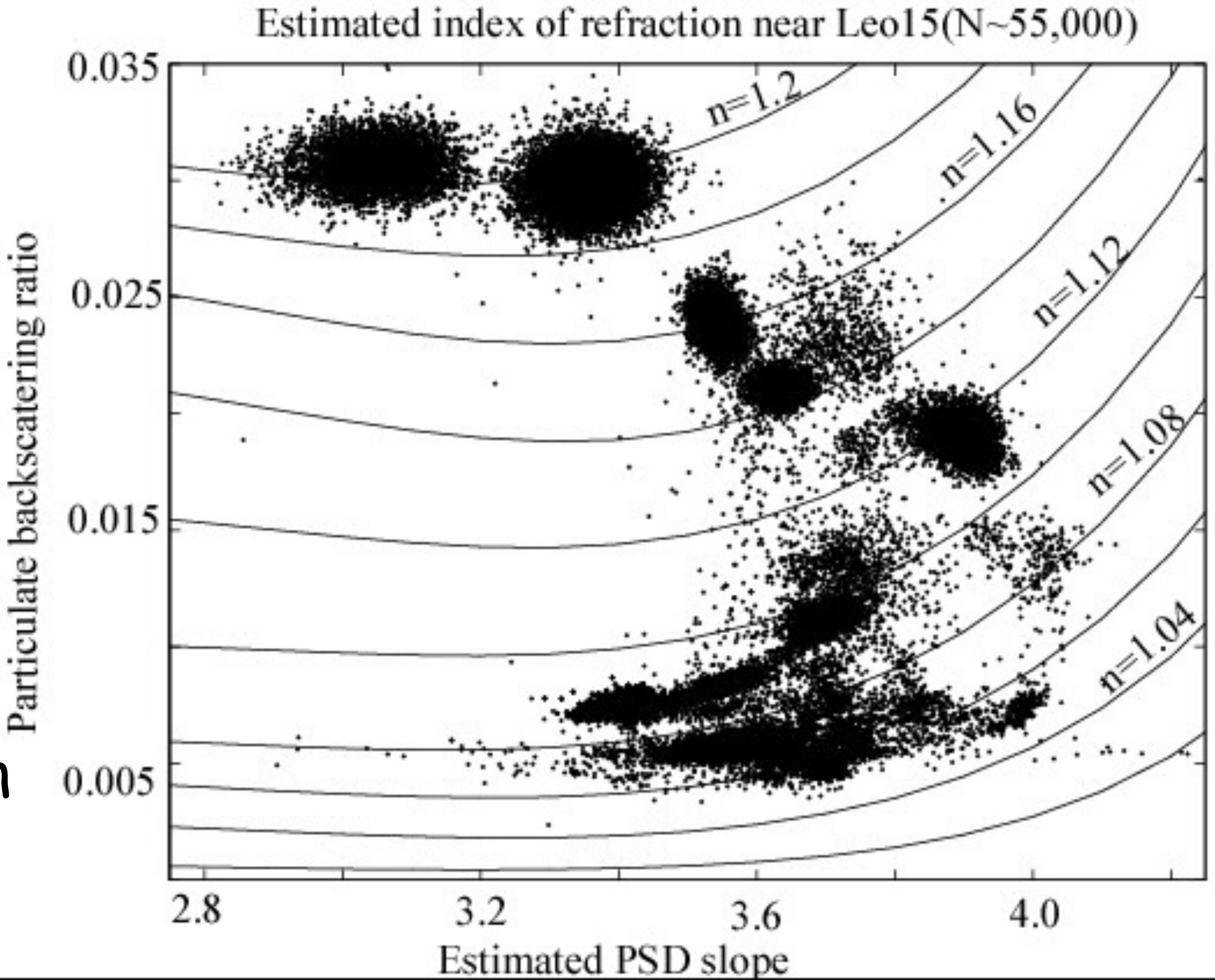
1. Index of refraction (*n*)
2. Slope of PSD (ξ)



Twardowski et al., 2001

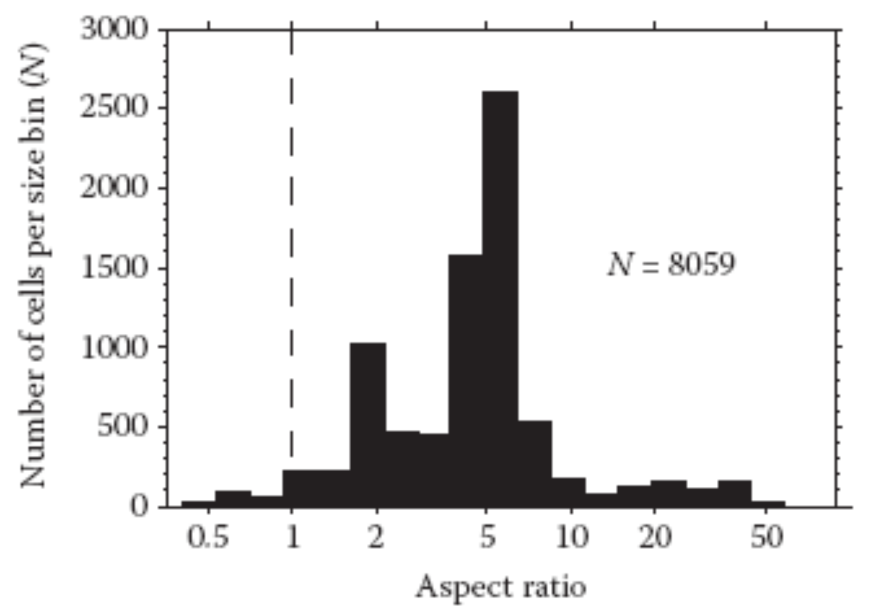
Backscattering ratio (55,000 observations from NJ shelf): consistent with theoretical prediction.

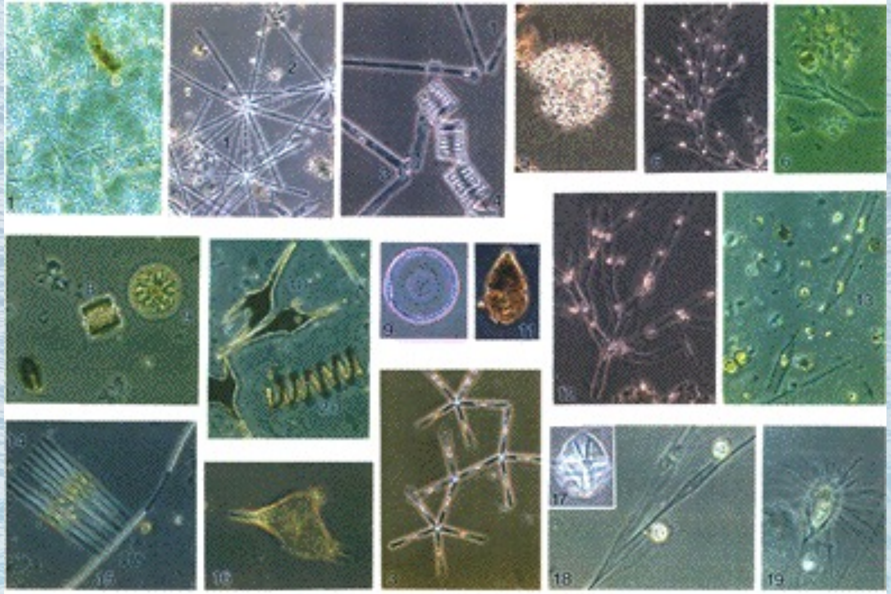
bulk suspension

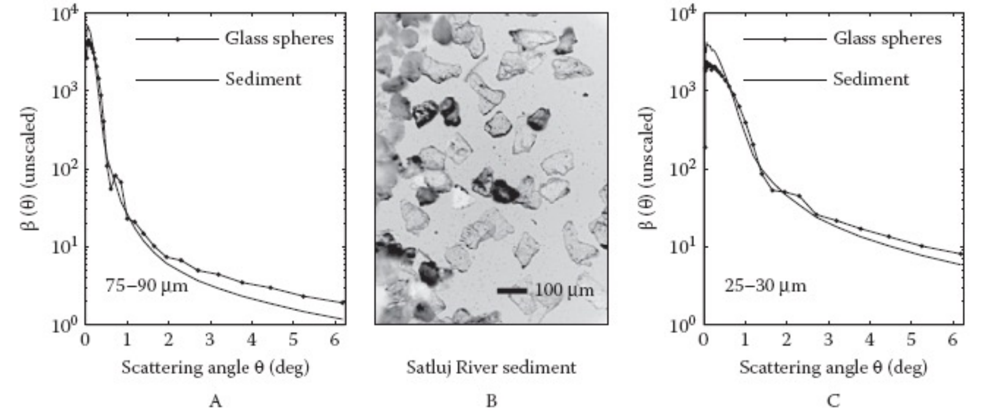


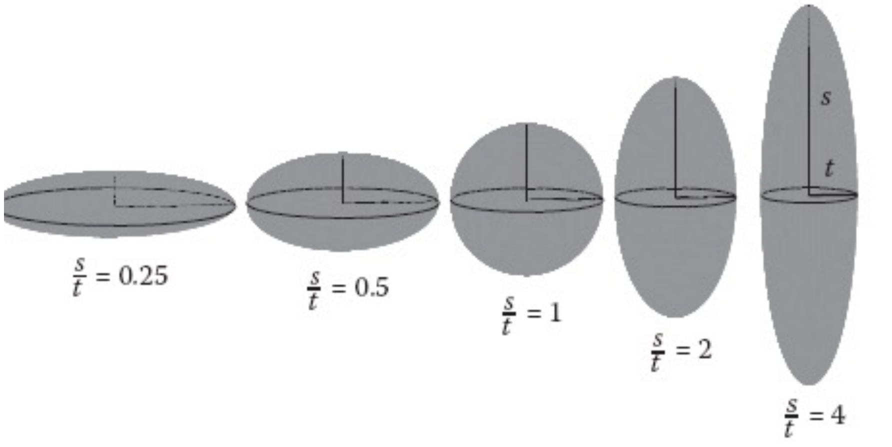
Varies from: phytoplankton → inorganic particles.

Shape consideration



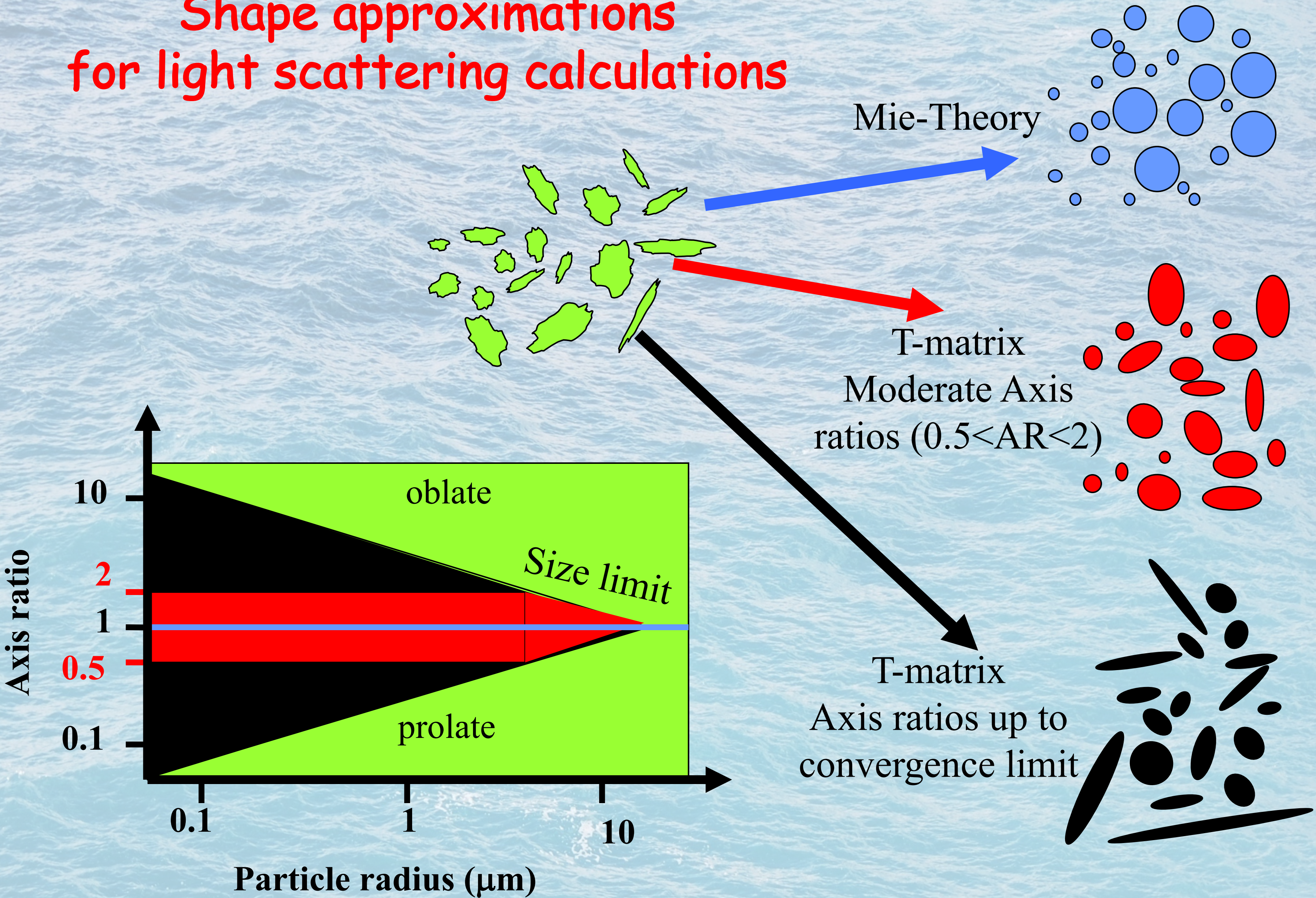

© 1993 Gertrud Cronberg





Clavano et al., 2007

Shape approximations for light scattering calculations



Mie-Theory

T-matrix
Moderate Axis
ratios (0.5<AR<2)

T-matrix
Axis ratios up to
convergence limit

Slide From Volten

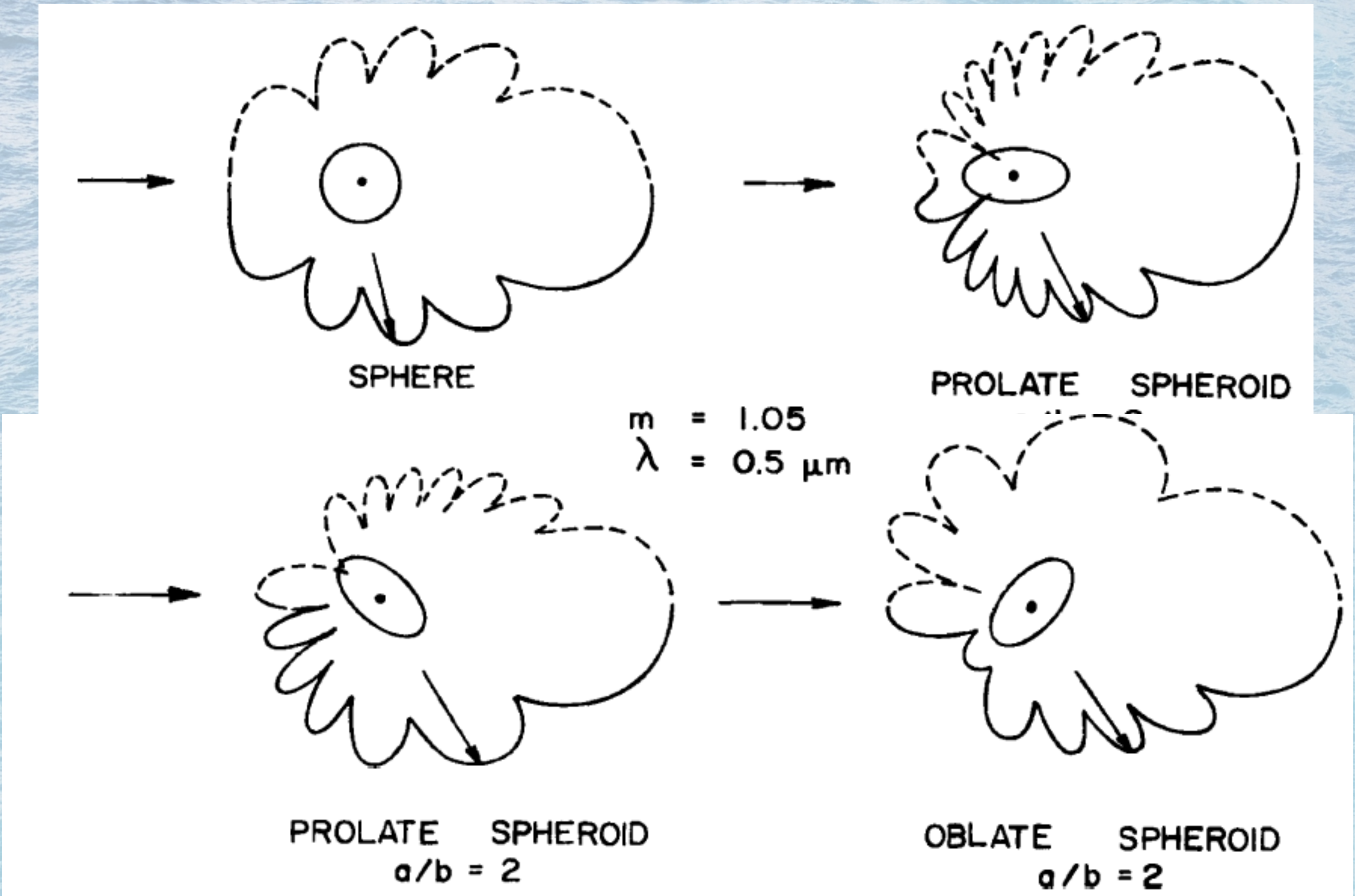
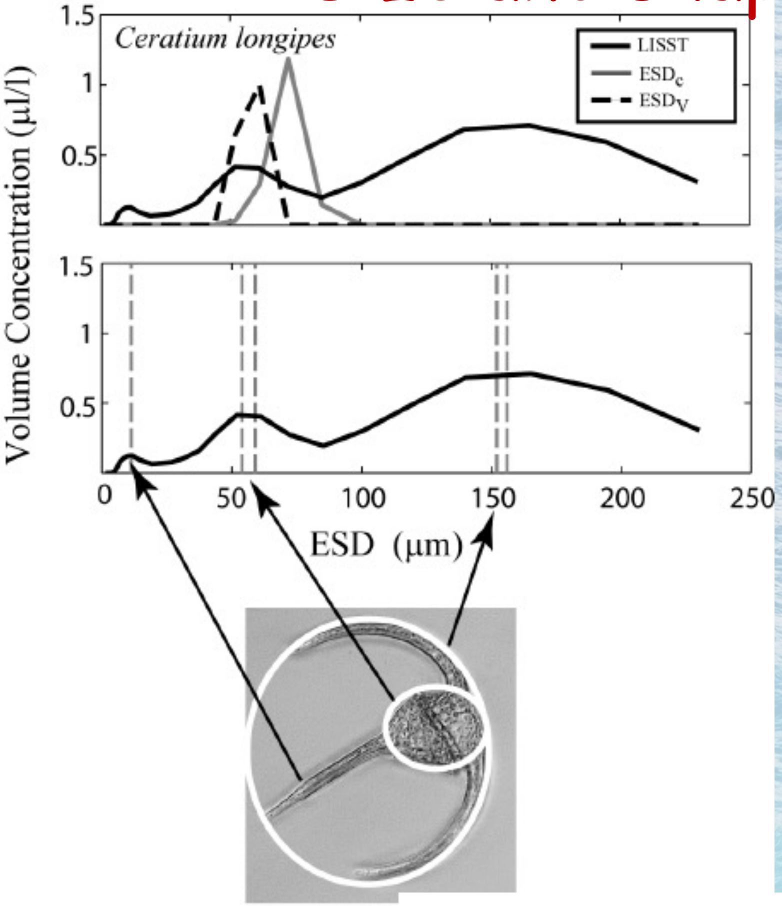
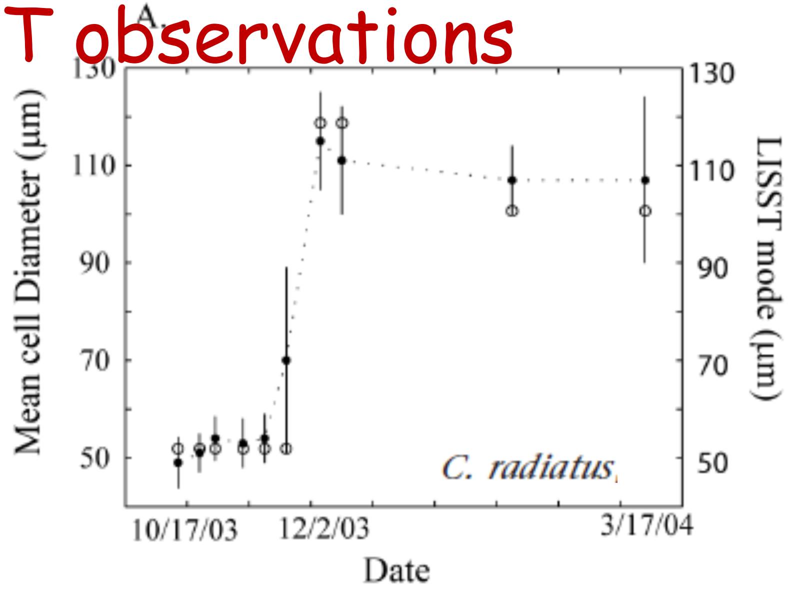


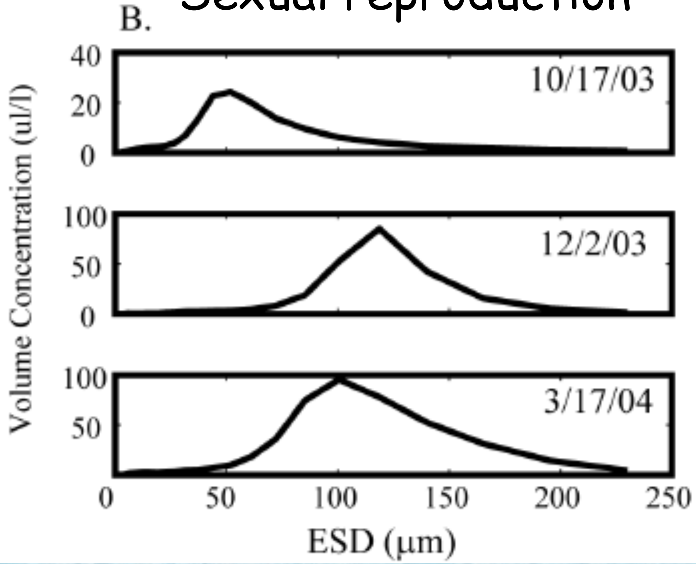
Figure 13.6 Polar scattering diagrams for equal-volume spheroids. The incident light is unpolarized. From Latimer et al. (1978).

Size and shape – LISST observations

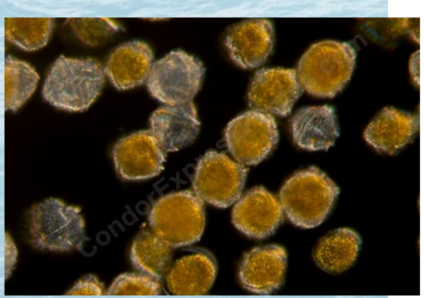


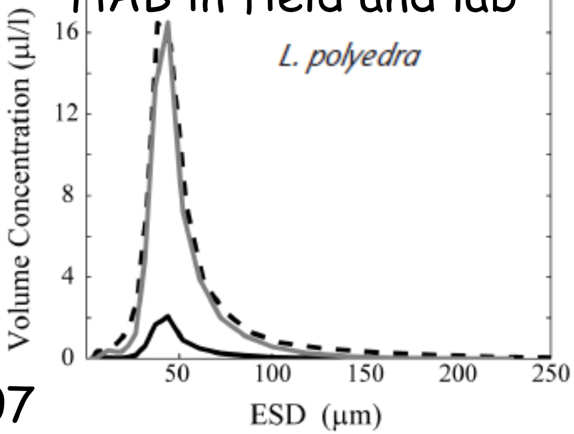


Sexual reproduction



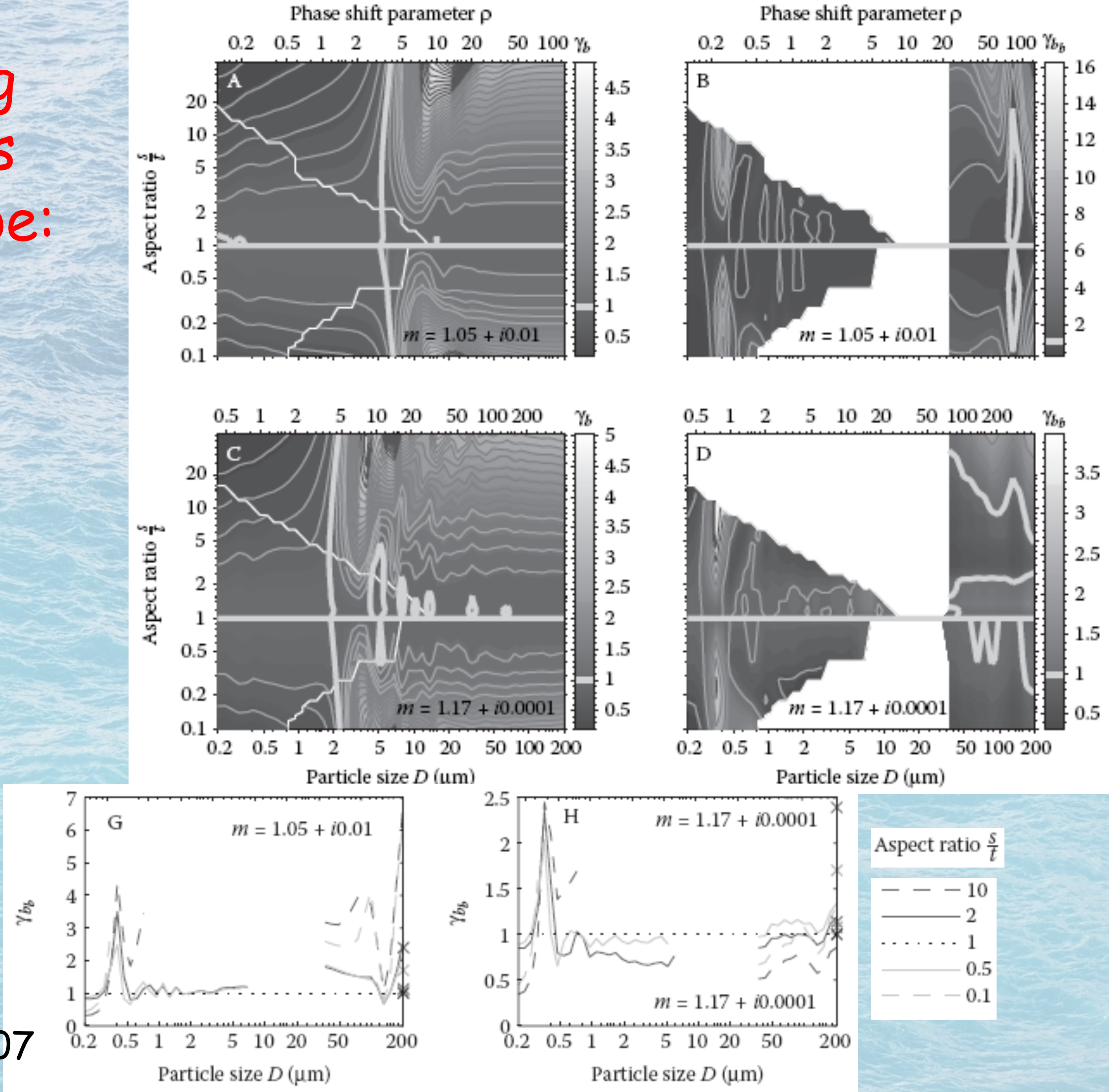
HAB in field and lab





Karp-Boss et al., 2007

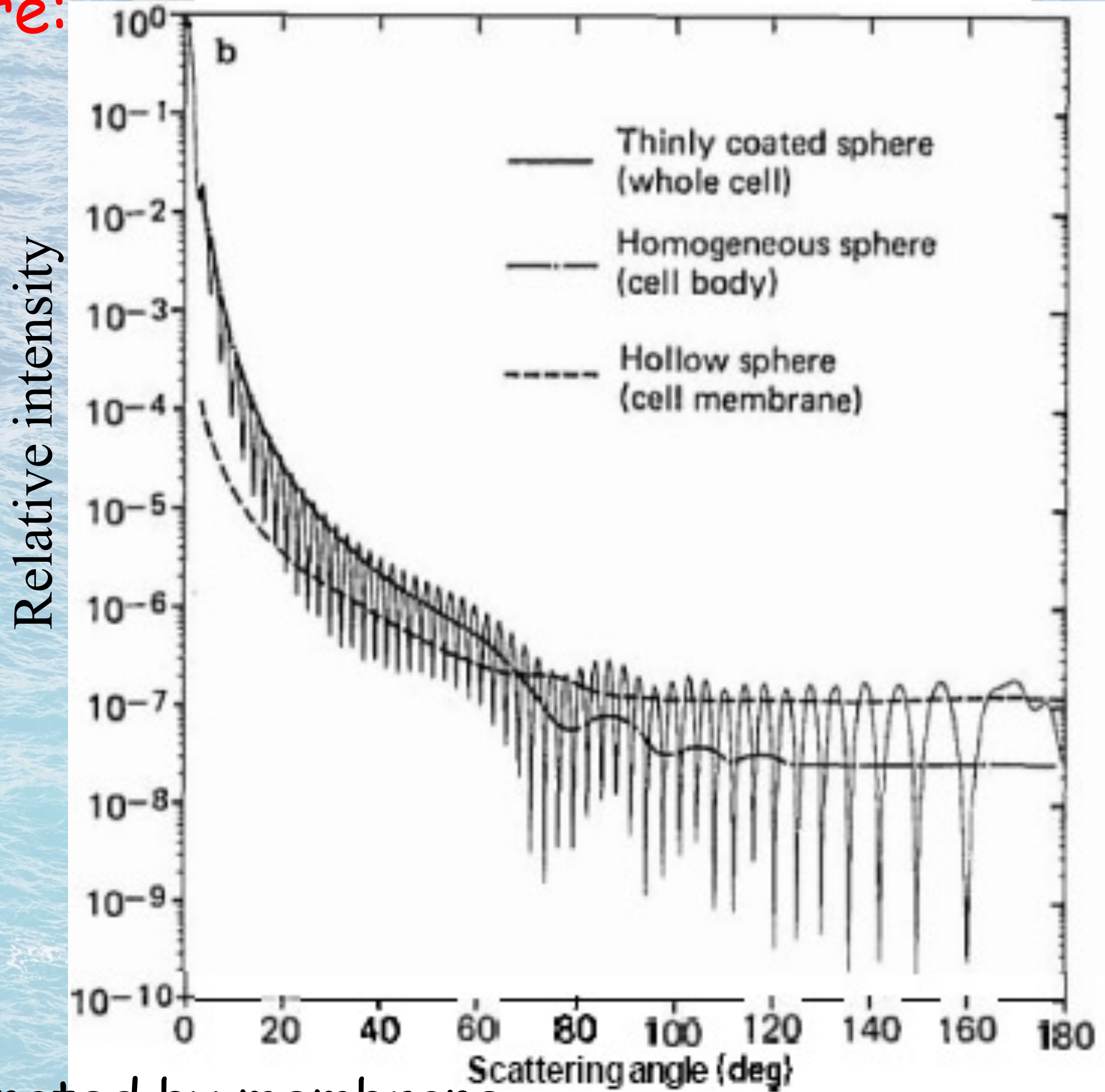
Quantifying differences due to shape:



Clavano et al., 2007

Internal structure:

10-µm diameter cell and a 100-Å thick membrane.

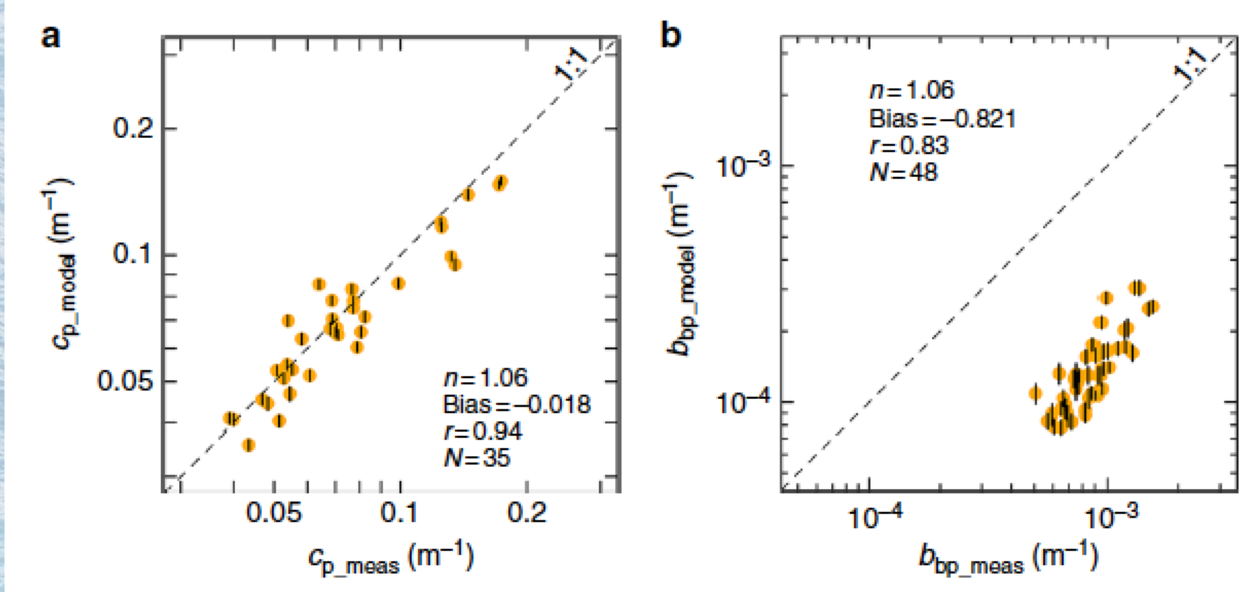


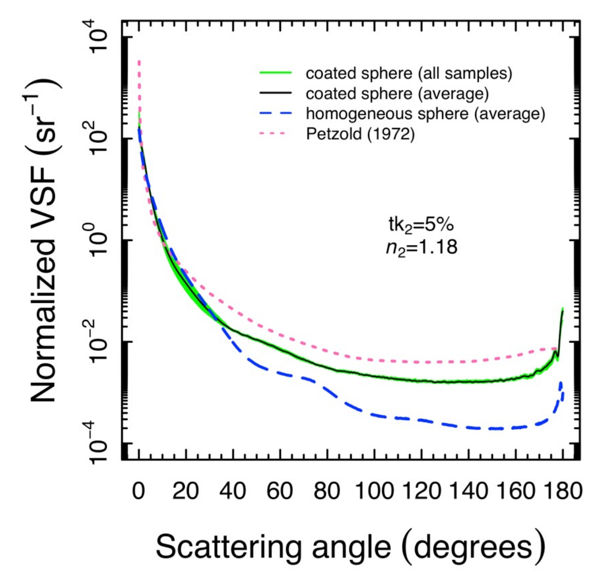
Backscattering dominated by membrane.

Meyer, 1979

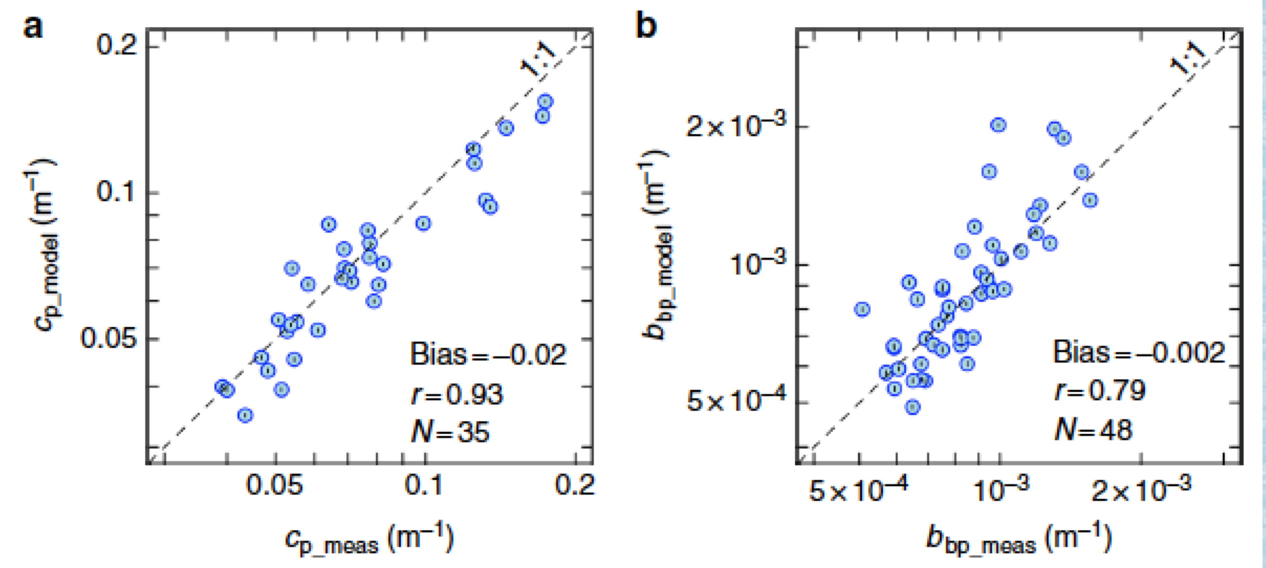
We get better agreement with observation if we account for internal structure:

Mie





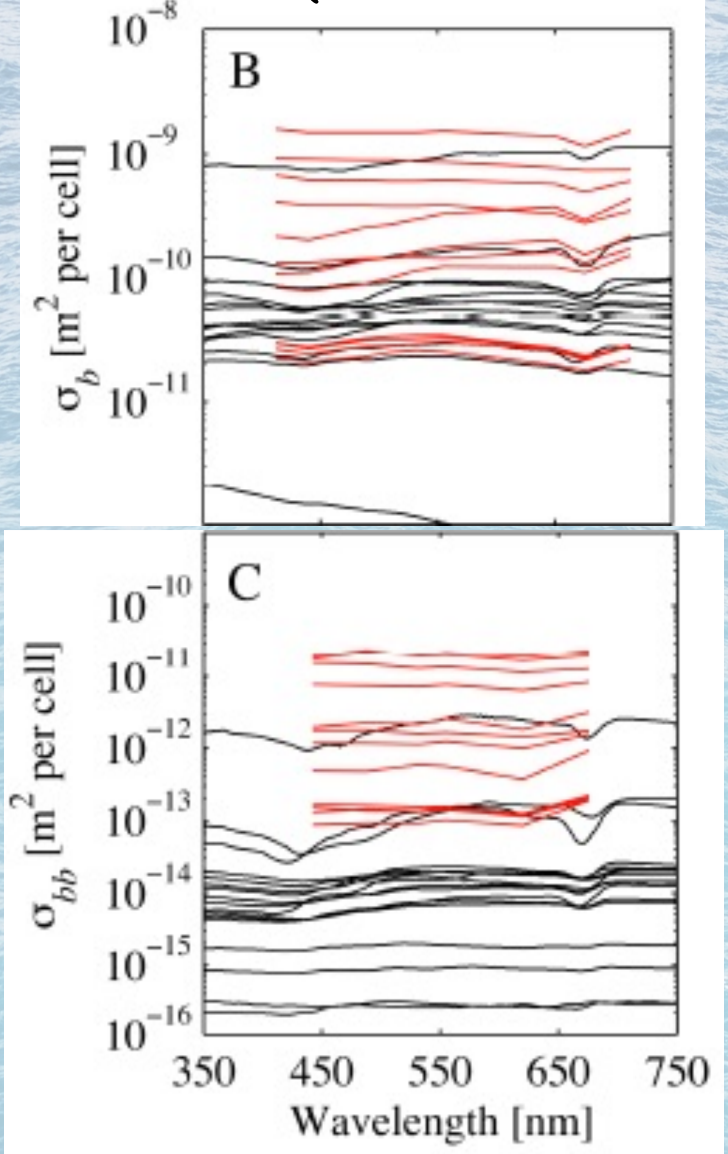
Coated spheres (inside=1.06, shell=1.13) :



Organelli et al., 2018

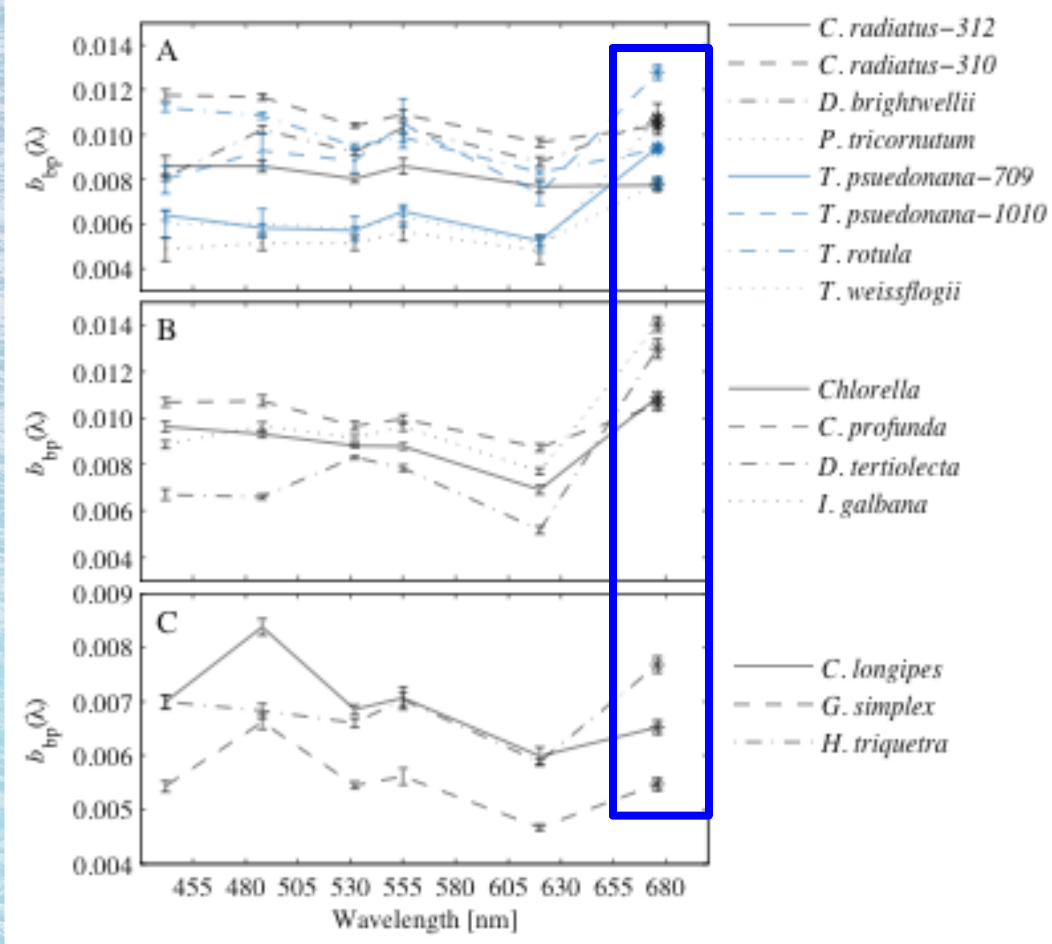
Scattering and backscattering by phytoplankton

In cultures (watch out for NAP)



Comparison with Mie theory of Stramski et al., 2001

b_b+F_{chl}



Whitmire et al., 2010

Backscattering of light from disklike particles: is fine-scale structure or gross morphology more important?

Howard R. Gordon

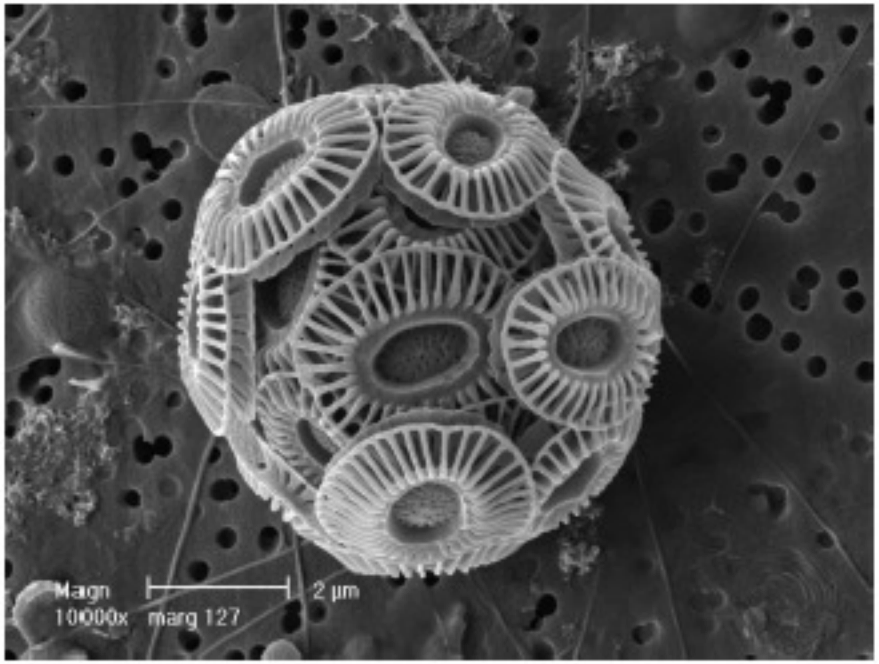
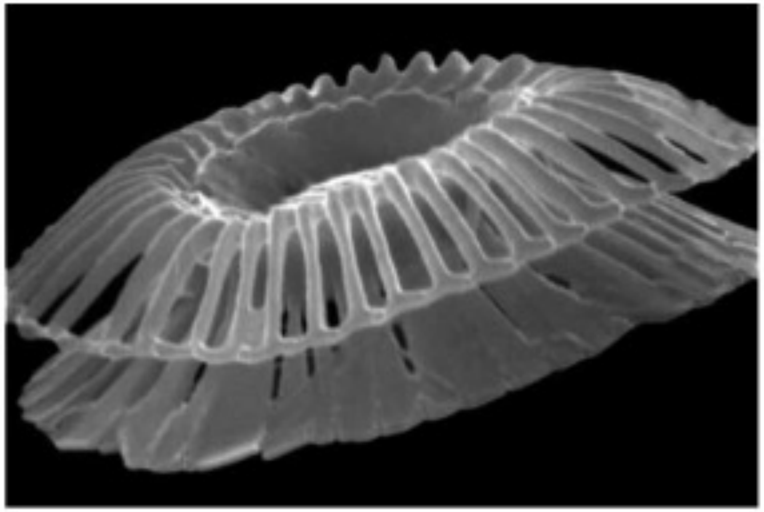
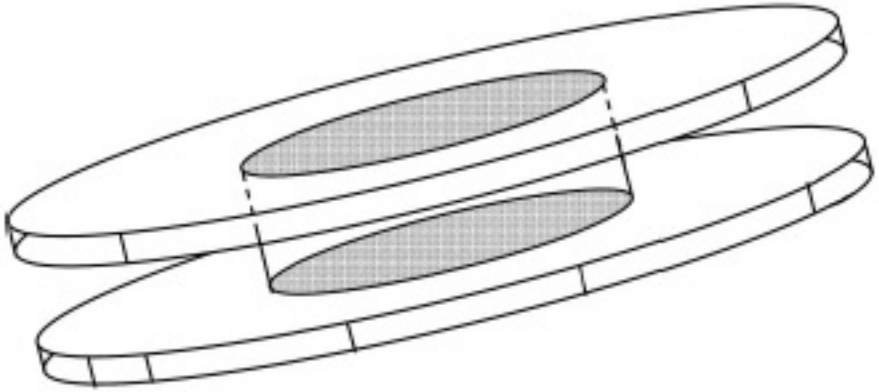


Fig. 1. Scanning electron micrograph image of a single cell of *E. Huxleyi* (the spherically shaped object) resting on a filter pad. The individual disklike structures covering the cell are the coccoliths (Fig. 2.). The pore size of the filter (small holes in the background) is 0.2 µm. The horizontal white bar in the lower left has a length of 2 µm. (Photo courtesy of Jeremy Young, The Natural History Museum, London.)





Bottom line:

The goal of the study was to obtain some understanding of the differences between the backscattering of a collection of such objects in random orientation and a collection of randomly oriented homogeneous disks of the same size.

In this regime the backscattering is totally governed by the particle's gross morphology and effective index **of refraction** .

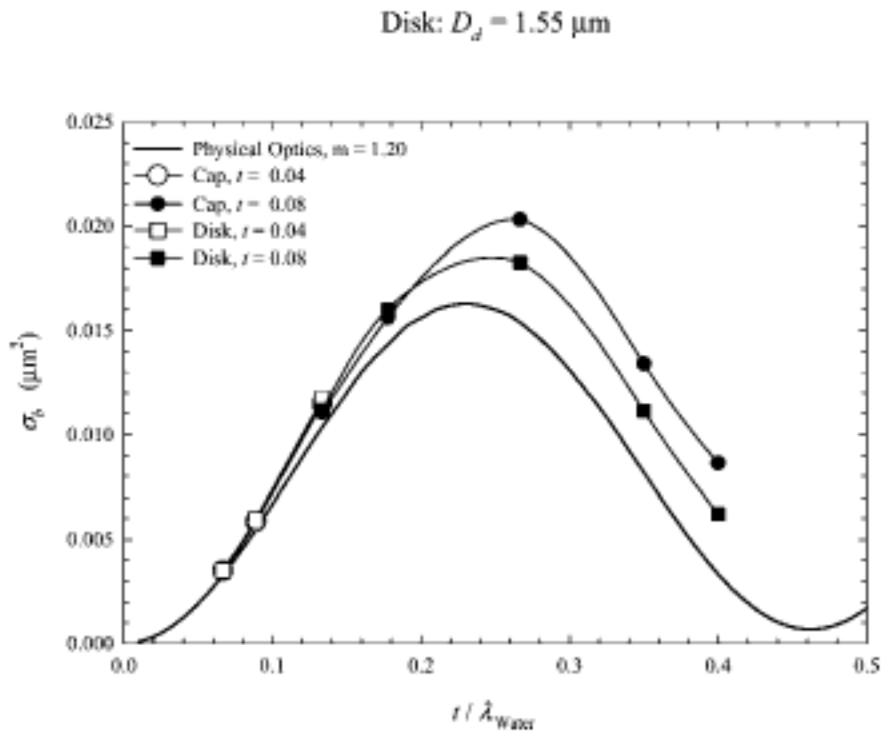


Fig. 6. Comparison of the backscattering coefficients of a disk and the associated spherical cap as a function of the thickness of the disk. Also shown are the results of the physical optics model of Gordon and Du (Ref. 7) for $m = 1.20$.

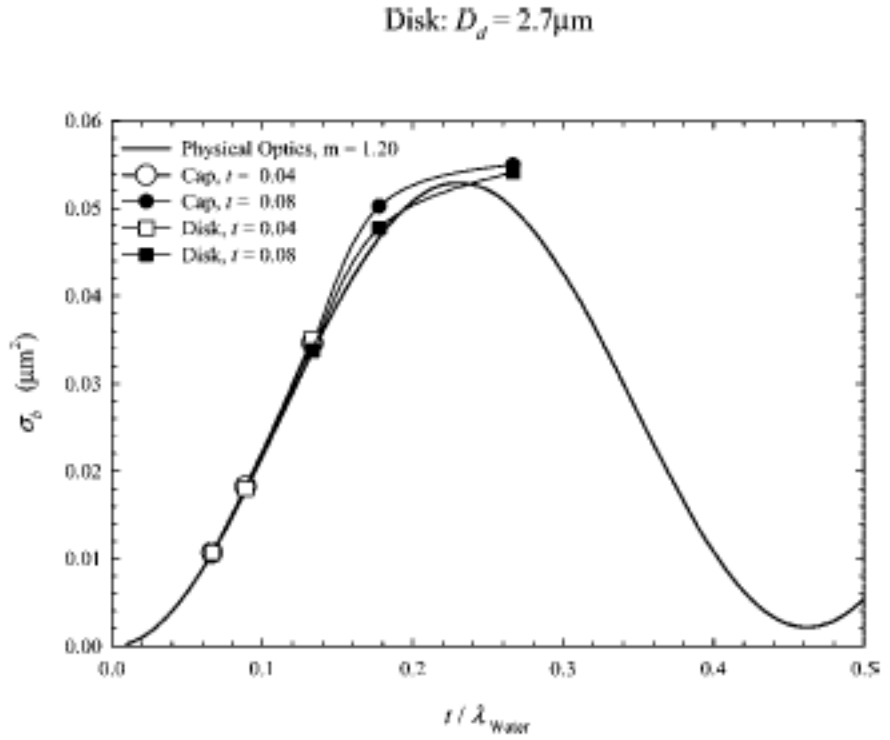


Fig. 8. Comparison of the backscattering coefficients of a disk (D_d = 2.7 μm) and the associated spherical cap as a function of the thickness of the disk. Also shown are the results of the physical optics model of Gordon and Du (Ref. 7) for $m = 1.20$.

Aggregation (packing) in the marine environment

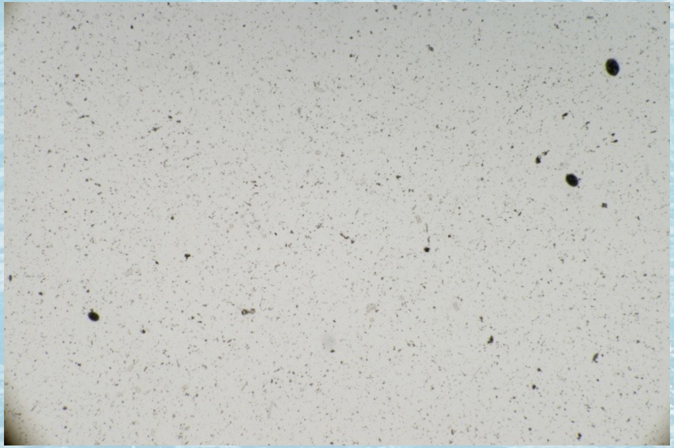
Aggregation is a [concentration]^2 phenomena.

Mechanisms for encounter: Brownian motion, differential settling, and turbulent shear.

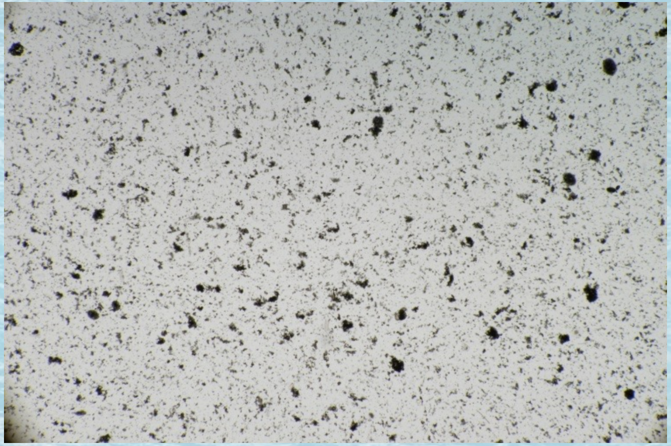
Aggregate sink faster than their component particles.

Aggregates break when shear is too high.

Camera pictures at 1mab at a 12m deep site within 1day:

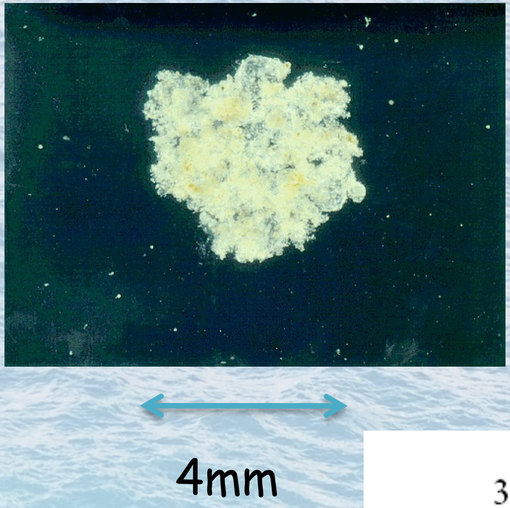


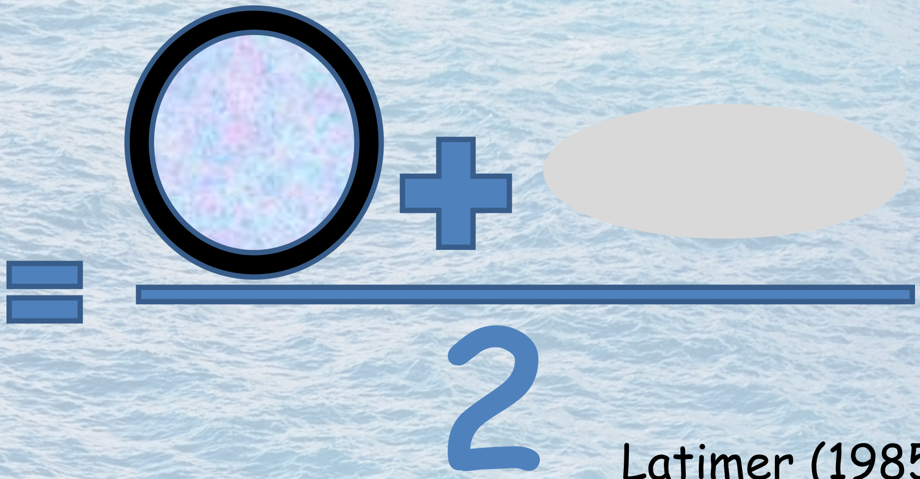
Dominated by <100um particles



Dominated by >1000um particles

Aggregate modeling:

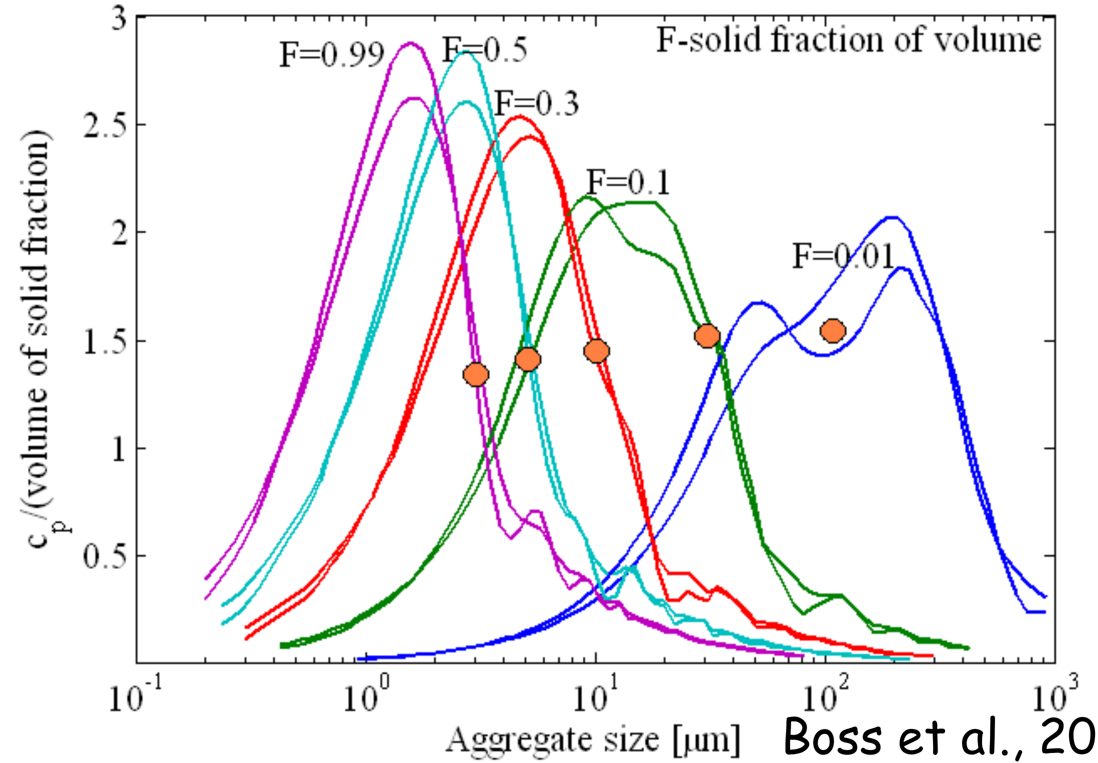




Latimer (1985)

For marine aggregates size and solid fraction correlate.

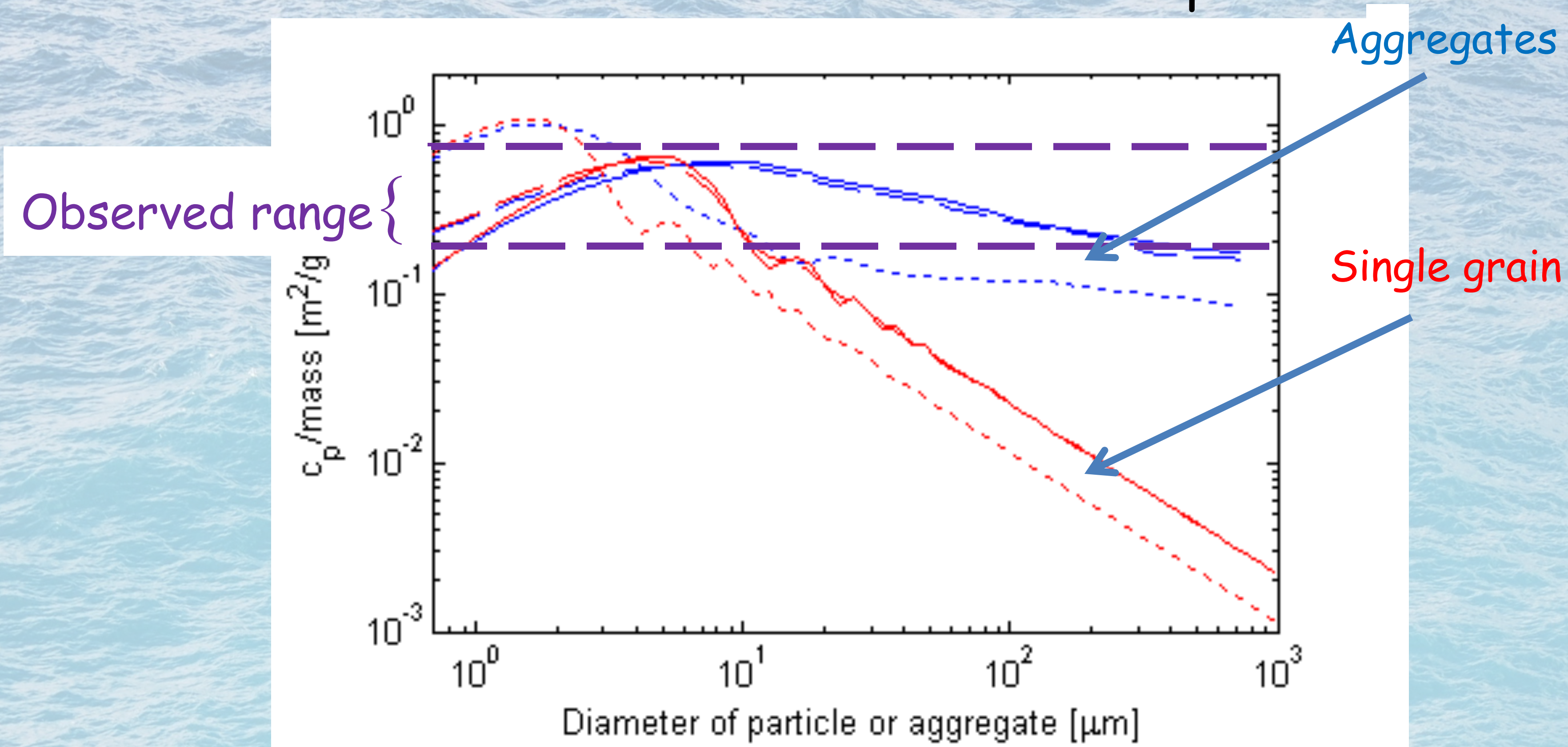
● -points having size-F as in Maggi, 2007, or Khelifa and Hill, 2006.



Boss et al., 2009

Aggregation (packaging) and IOPs

Theoretical calculations: monodispersion



Aggregates

Observed range {

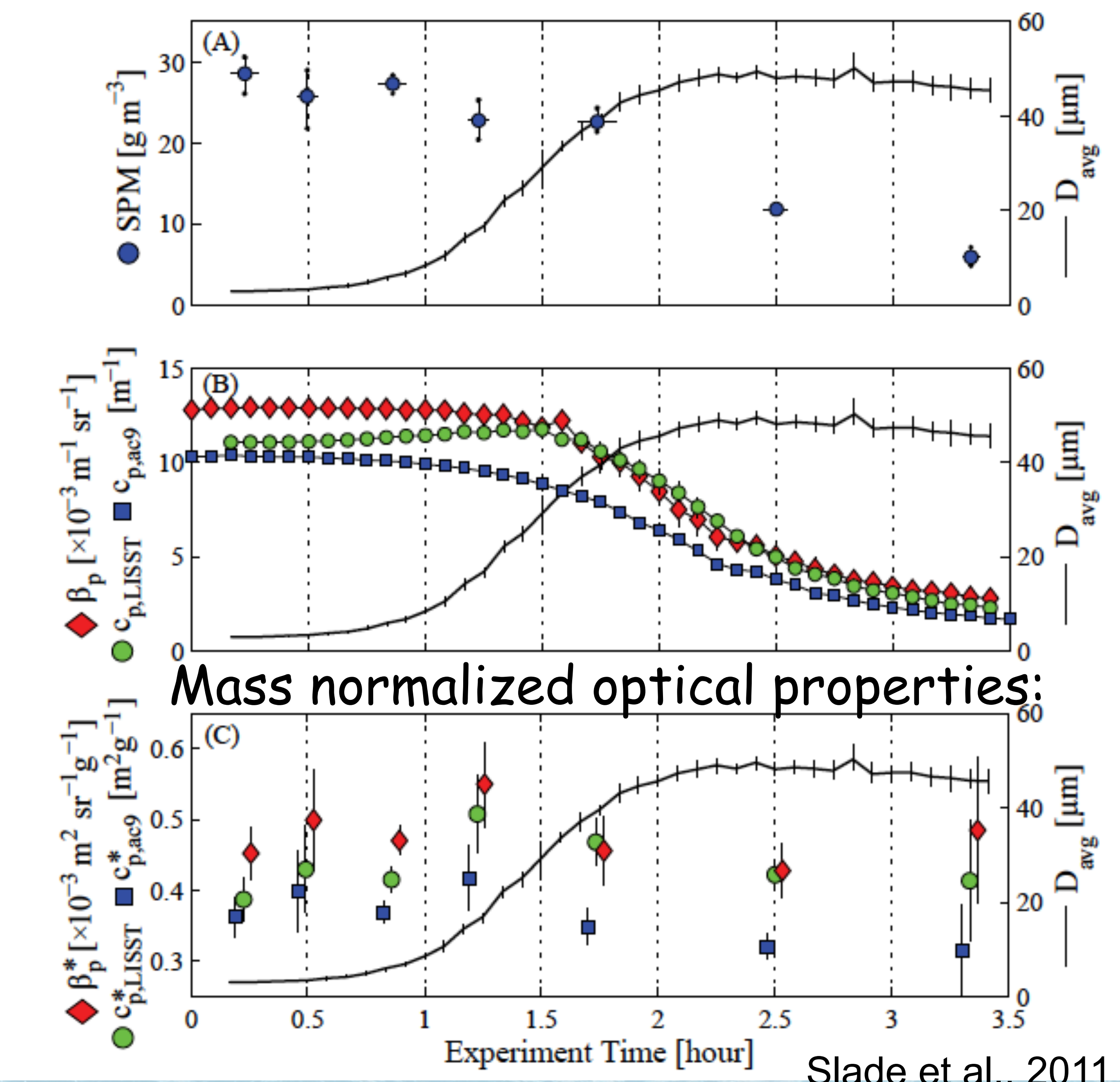
Single grain

Boss et al., 2009, OE

Water fraction as in Kehlifa and Hill, 2006

Aggregation approximately 'conserves' *area* not *volume*

Effect of aggregation on the mass-specific attenuation - lab experiment



Mass normalized optical properties:

Slade et al., 2011

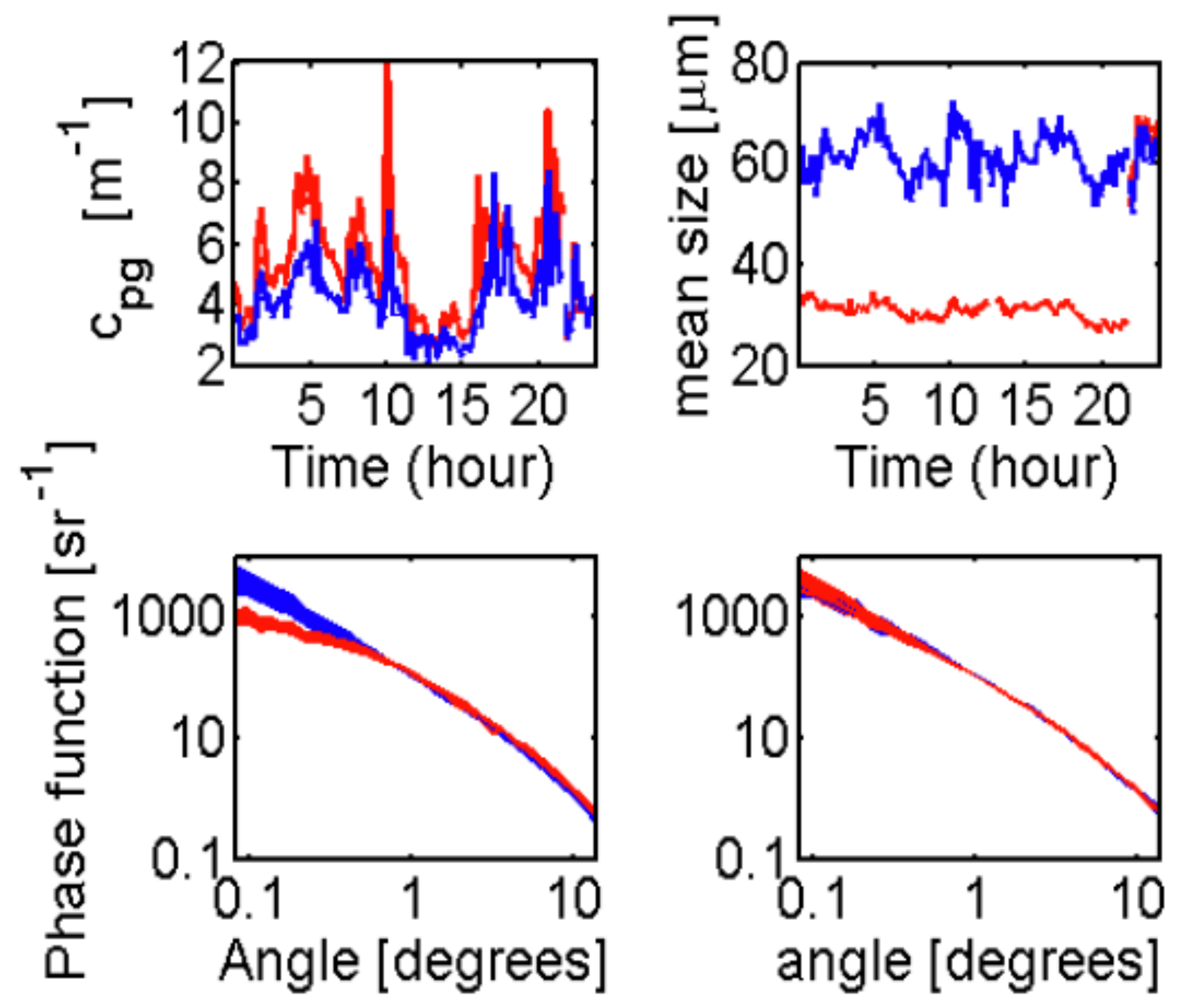
→It is of important that we consider aggregation when dealing with particle suspensions.

For aggregates we cannot simply assume:

$$OP \neq \int_{D_{\min}}^{D_{\max}} C_{OP,Mie}(D)N(D)dD$$

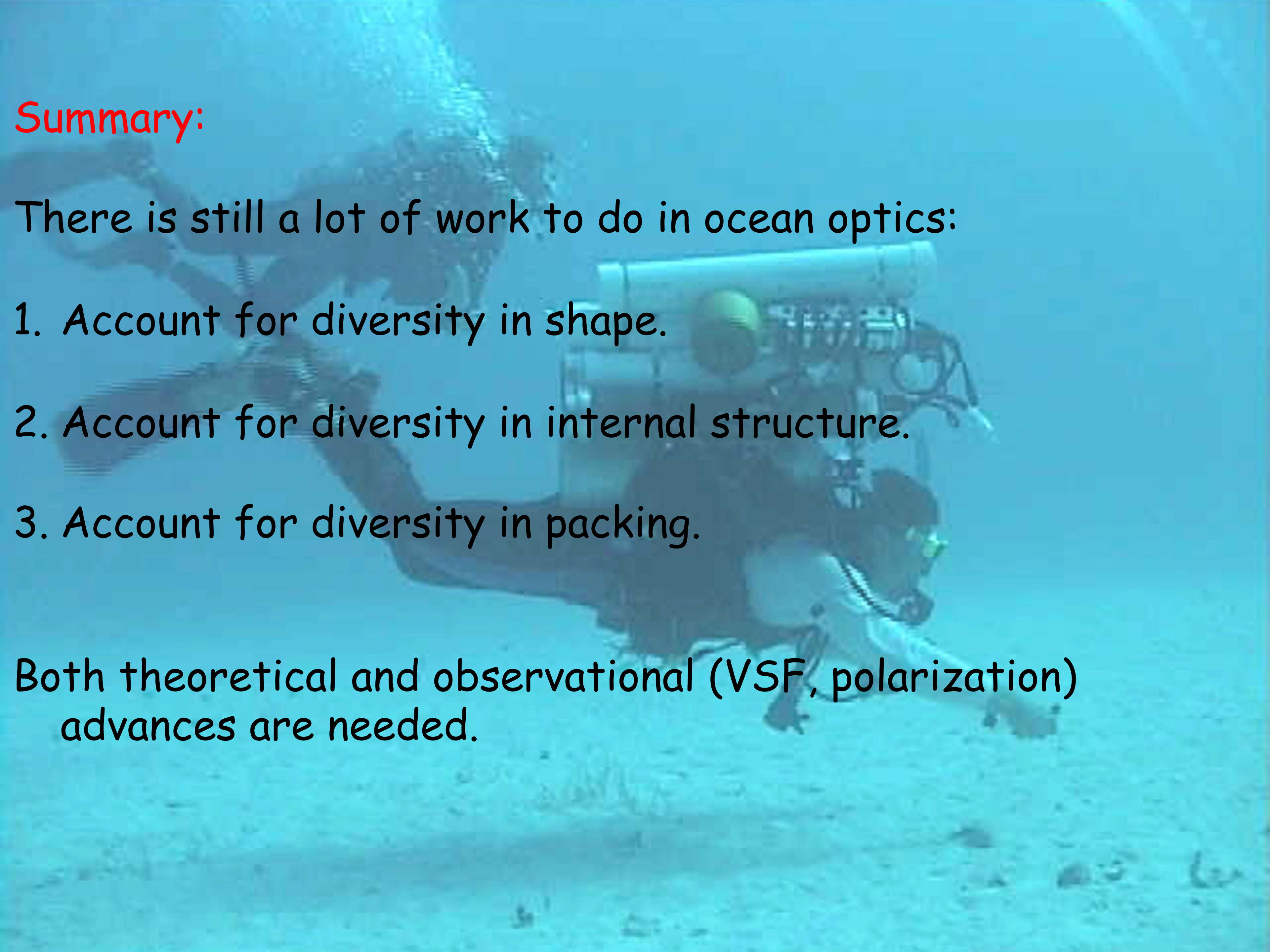
Such suspensions occur in open ocean as well as coastal areas (can be tested, see below).

How do we test that aggregation is important in-situ?



Boss et al., 2009, OE

Small effect on c_p large effect on near-forward β



Summary:

There is still a lot of work to do in ocean optics:

1. Account for diversity in shape.
2. Account for diversity in internal structure.
3. Account for diversity in packing.

Both theoretical and observational (VSF, polarization) advances are needed.

The way of the future: the AERONET inversion for aerosols

Figure 8: Cimel performing a GOSUN procedure

Measurements of cloud free day angular distribution of sky radiance + AOD + RT calculations are used to obtain:

Particulate size distribution
Index of refraction (real and imaginary)
Spectral single scattering albedo

Requires consideration of three main components:

1. Gaseous absorption (avoided by choice of λ, and use of climatologies).
2. Molecular scattering (calculated for given Pressure).
3. Aerosol absorption and scattering.

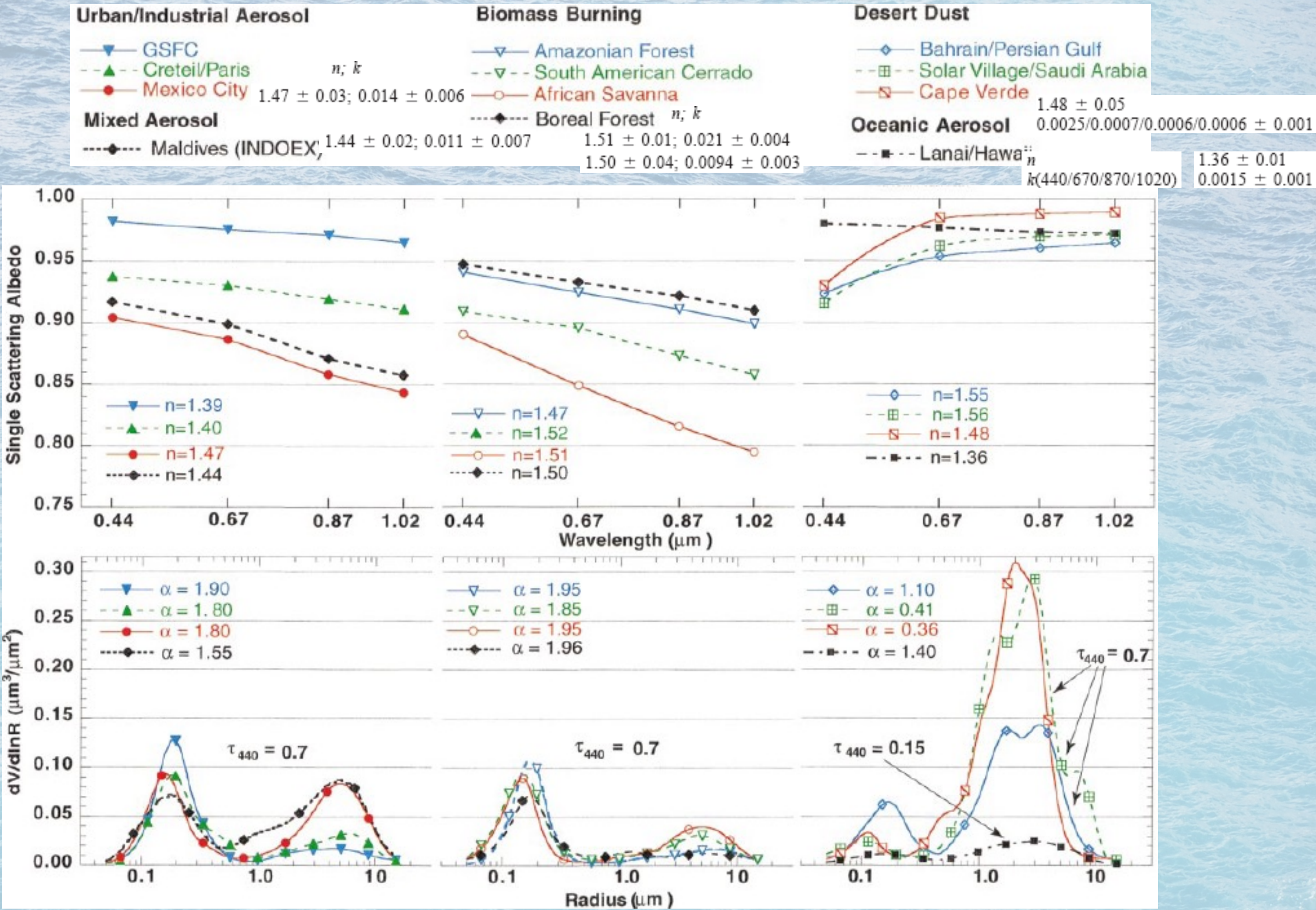
Minor (ignored) components: ground albedo, stratification

Use libraries of single particles optical properties (Mie or other)

Needs: RT model
Optimum inversion scheme

Bottom line: the more *information* we have the more properties we can constrain.

Summary of aerosol optical properties retrieved from worldwide AERONET network of ground-based radiometers.



α - Angstrom Exponent at 440 and 870 nm; n - index of refraction