**Biogeosciences
Discussions**

Interactive comment on "Direct contribution of phytoplankton-sized particles to optical backscattering in the open ocean" *by* G. Dall'Olmo et al.

G. Dall'Olmo et al.

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We kindly thank Dr. Twardowski for his constructive review: we believed it allowed us to significantly improve our manuscript.

Below are our detailed answers to his comments.

1) Why were fitted Junge-distributions used in the simulations instead of the directly measured PSDs? Fitting a power law will remove noise, but it will also remove any fine structure in the distributions; there are potentially better ways to remove noise while retaining PSD structure. For the fractionated PSD data plotted in Fig. 10, some of those distributions will not be described well by a single slope. For the purposes of extrapolation into size regions where there is no data, I can understand the fit, you do

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not have a choice, but it is better to use measured data where available.

We agree. PSDs used for simulations are now measured PSDs within the size range 1.4-8um. As explained in detail in the revised text, these limits have been established based on the precision of the data and on peaks often present below 2um. Outside this size range, PSDs have now been extrapolated to from 1.4um to 0.3um and from 8um to 100um using the power law slopes fitted to the measured PSDs.

2) Why was the imaginary n set at zero? A more reasonable value would have been 0.001 or at least 0.0005. If the real refractive index is nonzero, i.e., the particle scatters light, then its imaginary n must also not be zero (e.g. Bohren and Huffman 1983). The difference in Mie results from varying imaginary n at different small values usually has little impact, but the difference in results when setting the imaginary n at zero versus a small value can be significant in some cases. Even a small imag n can dampen refractive oscillations in Mie phase functions so they are more characteristic of phase functions for more realistic particles.

In the simulations presented in the discussion paper we had set the imaginary part of the refractive index to zero because absorption at the adopted wavelength (526 nm) is usually relatively low. In addition, oscillations in Mie phase functions were already smoothed because we reported results for polydispersions (i.e., the single particle phase functions were weighted using the PSDs). However, we have now repeated our simulations using a nominal imaginary part of the refractive index (n_i) equal to 0.0005. Sensitivity analyses using different values for n_i showed no major variations from the overall conclusion of this exercise. We have incorporated this information in the revised text.

3) Why was Dmax varied? Dmax should be set at a sufficiently high value to include the effective particle size range sampled (at least 100um). The argument could be made that a Dmax is chosen based on a low frequency threshold in measured abundance at that particular D, but then setting Dmax at any higher D will provide the same

result anyway because of the negligible influence of the larger particles. Setting Dmax at these low values (e.g., 6um) seems justified only if multiple filters were used in series to provide effective Dmax cutoffs. I see no justification for artificially truncating a measured PSD, effectively dismissing particles that will potentially contribute to the bulk optical properties. 4) How were n and Dmax iteratively solved so that the results matched a measured c_p value? There is clearly no 1 solution here and the results will be very sensitive to both. Something was assumed.

We will provide a single answer to the above questions 4) and 5), since they are related.

We have now presented in Appendix A of the revised text a more detailed and clearer explanation of how we matched our in situ c_p measurements with modeled values based on PSD measurements. The simulations were repeated for an n_i value of 0.0005 for the bulk and fractionated data and former Figures 8 and 9 have been updated. Importantly, even though the optimum set of n_r and Dmax has slightly changed in these new simulations, the main result (i.e., that two n_r values are needed to reproduce measured c_p and b_{bp}) is entirely consistent with that presented in the discussion paper.

5) Why were the n and Dmax results from matching a c_p value then used for b_{bp}?

Our objective here is to test whether we can reproduce measured b_{bp} values using the same parameters (n_r, PSD, Dmax, and Dmin) that allow us to successfully reproduce measured c_p values. This approach is an extension of studies carried out in the laboratory where these optical parameters are derived from c_p measurements and direct measurements of PSD and then used to simulate measured b_{bp} values (e.g., Vaillancourt et al., 2004). We show that the n_r needed to match measured b_{bp} values using the same PSD parameters is significantly higher that that needed to match measured c_p values. Thus, Mie theory suggests that different particle populations (characterized by different refractive indices) are responsible for the backscattering and scattering coefficients as shown in previous studies of this kind (e.g., Brown and Gordon, 1974). An alternative conclusion could be that b_{bp} and c_p are responding to different characteris-

tics of the same particle population as suggested by previous theoretical studies (e.g., Meyer, 1979). We have expanded our discussion to better explain this concept:

"Our results are in agreement with the latter studies: our Mie simulations could not simultaneously reproduce the measured c_p and b_{bp} (Figs. 9 and 10). This is likely because the shape of the volume scattering function (but not the total scattering) is sensitive to the internal structure and nonsphericity of natural particles (Meyer, 1979; Kitchen and Zaneveld, 1992; Quirantes and Bernard, 2004; Clavano et al., 2007). In other words, the relative amount of light scattered in the backward direction is higher for a microorganism that contains internal organelles and membranes than for a homogeneous sphere with the same average refractive index.

Particularly insightful with this respect is the study by Meyer (1979), who demonstrated that the scattering intensity of a coated sphere can be approximated by the sum of the scattering intensities due to two simpler particles (Fig. 11). The first of these particles accounts for most of the forward part of the scattering intensity and is the homogeneous core of the coated sphere. The other particle contributes most of the backscattering and is the hollow-sphere that constitutes the shell of the coated sphere. Thus, the scattering intensity of a complex coated sphere can be approximately predicted by employing two different and simpler particles that separately contribute most of the forward and most of the backward scattering, respectively. This theoretical finding could likely be the reason for why two (or more) particle populations are needed when trying to reproduce volume scattering functions measured in-situ using the homogeneous spherical model (Figs. 9 and 10; see also Brown and Gordon, 1974; Kitchen and Zaneveld, 1992). Moreover, oceanic microorganisms modeled as coated spheres can contribute up to one order of magnitude more backscattering than when modeled as homogeneous spheres (Kitchen and Zaneveld, 1992; Quirantes and Bernard, 2006; Bernard et al., 2009). Therefore, the coated spherical model could help resolving the "backscattering enigma" (Stramski et al., 2004) and, at the same time, explain the strong correlation we found between c_p and b_{bp} (Fig. 5)."

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Why not proceed with b_{bp} independently in the same manner as with c_p (although not sure what that really was)? These optical properties have different sensitivities to these input variables, e.g., b_{bp} is more sensitive to n than c_p. Why not vary n and Dmax until the b_{bp}/c_p value matches the measurement, i.e., use all the information you have?

See above.

Incidently, looking at Mobley et al. 2002, a Junge slope of 3.5 intersects the Mobley et al. dashed regression in Fig. 2 right at a b_{bp}/b_p of 0.01 - very close to the values measured here - and corresponds to a bulk refractive index of 1.1. The Mobley et al. algorithm is based on the Fournier-Forand phase function model and has no inherent specificities to particle shape. Note this bulk refractive index is close to what you would expect from Mie theory if b_{bp} was addressed independently of the constraints developed from the c_p fitting. So which is correct?

To address this comment, we have now added the following paragraph to the discussion:

"It is also noteworthy that the models proposed by Twardowski et al. (2001) and Mobley et al. 19 (2002) predict that the average particle in our study should have a value of the real refractive index close to 1.1, when using as inputs for the model the median $b_{bp}:b_p$ ratio at 526nm and the median slope of the PSD derived in this study (0.010 and -3.5, respectively). Thus, these models predict that a single particle population simultaneously contribute to c_p and b_{bp}. This prediction disagrees with our results that indicate that Mie theory was unable to simultaneously reproduce the measured c_p and b_{bp} using a single population of particles (Figs. 9 and 10). The likely explanation for this disagreement is that both the above models assume that very small and large particles contribute significantly to the measured optical properties (0.006-73 um, in Twardowski et al., 2001 and 0-∞um for the Fournier-Forand phase functions used by Mobley et al., 2002). Our negligible b_{bp}(<0.2 um) values are however at odds with this assumption and the cumulative seawater sample over which our data are binned is

too small to measure particles larger than about 40-100 um. Furthermore, the finite acceptance angles of our transmissometers also act as filters for the signals generated by large particles (Boss et al., 2009). Instead, very large particles have been shown to be important in Mie theory simulations when the PSD exponent is -3.5 and when the finite acceptance angle is not accounted for. For example, Stramski and Kiefer (1991) needed to increase their maximum diameter to 1000um to achieve a saturation in their cumulative scattering contribution when the PSD exponent was set at -3.5. In addition, the refractive index of 1.1 appears to be rather large for open ocean waters as the surface Equatorial Pacific notoriously deficient in atmospheric dust deposition (Mahowald et al., 1999) and thus likely dominated by organic particles. We recognize that a value of n_r = 1.1 is on the theoretical upper range for phytoplankton (1.06 ± 0.04; Aas,1996), but it is also significantly higher than values expected for "soft" organic particles typical of open ocean waters (1.02–1.05, Zaneveld and Pak, 1973; Carder et al., 1972). Thus, the n_r = 1.1 derived from our median backscattering ratio and median PSD exponent using the models by Twardowski et al. (2001) and Mobley et al. (2002) could be overestimating the actual average n_r."

There is an underlying problem in the current approach in that there is an implicit assumption that Mie theory applied to a natural nonspherical population should without question work for c_p from first principles. While there may be differing shades of gray between Mie theory's application to c_p vs b_{bp}, this is clearly not a given. From personal experience, which I know is shared by Emmanuel from our correspondences, testing closure between measured PSDs and measured c_p usually does not add up.

We have now added a paragraph that makes the hypothesis explicit and discusses the non-sphericity issue raised by the reviewer:

"Finally, so far we have been implicitly assuming that measured c_p values can be accurately reproduced by using homogeneous spheres as models of phytoplankton cells. This assumption is based on theoretical findings showing little sensitivity of the absorption and total scattering coefficients to particle inhomogeneities and shape (e.g.,

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Meyer, 1979; Clavano et al., 2007). Admittedly, a nonspherical population of parti-
cles can produce in certain cases important deviations from the optical properties of
volume-equivalent spheres (Clavano et al., 2007). However, such deviations are ex-
pected to be constrained to about 20-30% for aspect ratios ranging from 0.5 to 2. Only
at extreme aspect ratios and for non-spherical particles with ESD$>$ 10 um, the devia-
tions become very significant (Clavano et al., 2007). Since non-sphericity is an attribute
typical of large cells, and since those large cells are usually rare in the surface waters
of the Equatorial Pacific and likely undersampled by our instrumentation (see above),
we believe that our assumption is valid."

*6) Finally, I questioned the overall purpose of the Mie theory analyses. It does not
seem to add much to the central conclusions and may create a confusing diversion;
at least it did for me. One expressed purpose was essentially to test the efficacy
of using Mie theory to obtain b_{bp} from first principles, but can you really do that with
much certitude with a size distribution of limited size constraints, while guestimating n
distributions, imag n distributions, Dmax, and having to assume Mie theory is reliable
for c_p for natural particle populations? I question whether this data set could be used
well for this purpose. So I would suggest leaving the Mie modeling out. I can see
some modeling being retained if the approach was changed and the purpose was
clear. Putting aside the Mie modeling for a moment, the empirical data set is wonderful
and clearly shows conscientious attention to planning, detail and accuracy. These
measurements are not easy to make. The authors are to be commended for such fine
work.*

We disagree with the reviewer that the Mie modeling should be removed from our
manuscript. We believe that the Mie modeling part is fundamental to show the mis-
match between theoretical predictions and our data.

However, we agree that modeling sections increased the length of the manuscript and
somewhat distracted the attention of the reader. Thus, we have decided to move most
of the text related to the Mie modeling to Appendix A, while we have retained in the

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main text the presentation and discussion of its most important results.

Note also that we have now clearly discussed in the appendix that: 1) the value of n_i is of secondary importance once it is kept to a small value, n_r is rather well constrained, and Dmax can be set to a large value without affecting our results. Thus, we believe that the data available, although not perfect, are an excellent starting point for the comparison between theory and observations.

Specific Comments

Title: I do not think "phytoplankton-sized particles" is especially meaningful. I understand there is a strong underlying desire here to link b_{bp} to phytoplankton biomass, but neither phytoplankton generally nor their biomass specifically were characterized in any way that I can see except their chlorophyll content.

The title has been changed in the revised version of the manuscript to "Significant contribution of large particles to optical backscattering in the open ocean"

Well written Introduction. Thank you.

I like the use of the dye in assessing $b_{b,wall}$.

The additional measurements with the die were necessary to establish the range of variations of $b_{b,wall}$.

p. 300: For the most correct beta water values, now see: Xiaodong Zhang and Lianbo Hu, "Estimating scattering of pure water from density fluctuation of the refractive index," Opt. Express 17, 1671–1678 (2009). The Morel (1968) derived correction of 1+S/37*0.3 for salts still needs to then be applied.

The entire b_{bp} dataset has now been reprocessed using the (Zhang and Hu, 2009) model for the scattering of pure water, and the (Zhang et al., 2009) salinity correction. However, the results changed only slightly. This is because most of the difference in the β_{sw} value was due to new β_w and this difference has been "absorbed" by the

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recomputed $b_{b,\text{wall}}$ values. All figures have been updated.

Verification of the WET Labs bead calibrations after the cruise to both verify their values and assess any drift is important in these relatively clear waters.

We also believed that and indeed found a significant drift in the red channel that was also clearly visible in the b_{bp}(<0.2um) at 656 nm. A similar drift has been also noticed in the 595nm channel of a different bb3 meter.

p. 304: This is somewhat tangential, but regarding χ_p factors, I can say from recent work in our lab that the Boss and Pegau value and Berthon et al.'s value around 1.1 looks accurate at 117 deg. Sullivan et al.'s value looks accurate as well - so let me explain. The issue is that the ECO sensors have an angular weighting function that is broader than just 117; in fact I recalculated these weighting functions for the ECO recently (Ron Z computed the original ones) and found much (much) broader functions than the ones we have been using for calibrations. Weighting a proper χ_p function in the backward direction with the more correct ECO weighting brings the χ_p value at 117 down by about 10% on average, maybe a little more. We only know this now because of the more complete VSF measurements from the MASCOT sensor. This is why Sullivan et al. found a value of 0.90 for the ECO, but that value was also affected somewhat (a few percent) by the estimates of b_{bp} from the 3-angle ECOVSF that we were calling reality at the time. Bringing down your values 10% will not affect your results much; in fact all the relative b_{bp} results will be the same. I have no problem with the values being left as they are, as we are still working on the issue and getting a paper ready for submission (if interested, we can send a preliminary draft when ready).

We thank the reviewer for this clarification. However, as the reviewer suggests, for the time being we decided to keep the χ_p factor at the value proposed by (Boss and Pegau, 2001).

But it may be worth commenting that although there may be upwards of 10% bias uncertainty in the b_{bp} estimates from an ECO at this time (which is a number used

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in several previous publications to describe the estimated level of uncertainty in bb measurements), the key sources of bias error cancel in any ratiometric analysis of the subfraction data, so that these results in particular should have much better accuracies.

We agree with the reviewer. However, this is only true for the values of the fractional b_{bp} values. For absolute b_{bp} values measured on fractionated samples the uncertainty due the χ_p factor does not cancel out.

p. 310, 3 lines from bottom: should 1.5um be 2.5um?

Yes, corrected.

Also, note in this discussion that many soft biological particles squeeze through filter pad pore sizes smaller than their ESD. This is common.

We have added text in the discussion to cover this concept.

p. 314: While the fractionation results from mesotrophic stations show that 40-50% of bulk backscattering on average was found in the <3um fraction, why not also discuss the results from the oligotrophic fractionation experiments 2 and 3? These experiments seem to show that nearly 100% of the bulk backscattering was found in the <1um fraction as well as obviously the subfractions from larger pore sized filters. Since the open ocean is dominated by oligotrophic conditions and the expressed interest here is in developing a global understanding of the sources of oceanic backscattering, I would expect these experiments to be discussed in more detail, especially considering the comparison to Stramski and Kiefer's Mie results, which I believe were specifically intended for only the oligotrophic ocean.

We agree with the reviewer that the fractionated data from the oligotrophic stations suggest an interesting scenario for the backscattering budget of the oligotrophic ocean. However, because only two such stations were available we preferred to be cautious in drawing conclusions.

Following the reviewer suggestion, we have added text to hint at the possibility that b_{bp}

may come from the 0.2 to 1um fraction in the most oligotrophic waters.

It is stated here that the fractionation results show more backscattering in larger particle size classes than predicted from Stramski and Kiefer's Mie theory results - for experiments 2 and 3 in oligotrophic waters, which are most applicable, this statement appears to be untrue, or at least not unequivocally supported by the data. Furthermore, since there was no filter used in the experiments with a pore size of exactly 1.2 um (the 50% cutoff for S+K's results), the statement is weakly supported by the data from mesotrophic regions as well. From my viewing of the results, it really does not look like S+K were too far off in this respect.

We have now removed this section

The result that essentially all the bulk backscattering in the oligotrophic samples appears to arise from the relatively narrow 0.2 to 1.0 um size class (i.e. prokaryotes) is absolutely fascinating.

We agree, but we suggest caution because only one fractionation experiment was carried out in the most oligotrophic waters.

p. 320: In the first bulleted conclusion, it would probably be good for clarity's sake to insert something like ", when considered with previous findings that bulk c_p may be used effectively to track phytoplankton biomass," after "suggesting that..."

The text has been updated to include this suggestion.

Fig. 1: if the black triangle sites were labeled 1-2-3 it would be a help to the reader in figuring out which fractionation experiments took place where.

The figure has now been updated according to the reviewer's suggestion. Thank you.

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