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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 would like to kindly thank Dr. Stramski for his comments on our manuscript. In the following text, specific comments by the reviewer are shown in "*italic*", while our responses are shown in black.

Responses to General Comments

We understand Dr. Stramski concerns about our work and realize that the data presented in this study may be controversial since they challenge some of the current hypotheses on the optical backscattering budget in the ocean (Morel and Ahn, 1991; Stramski and Kiefer, 1991). However, we must also realize that such budgets are based on a seemingly inappropriate model for the backscattering coefficient of marine

particles (the homogeneous sphere) and on an as yet untested hypothesis regarding the particle size distributions (PSDs) in the ocean (i.e., that PSD measurements can be extrapolated from 1um down to 0.002um). Thus, as the reviewer acknowledges, it may not be surprising that those predictions may eventually reveal themselves as inaccurate.

The reviewer states that "*the weakness of this result in this study stems from limitations of the fractionation approach*". We have not claimed that fractionation by means of filtration is the ideal method for partitioning the PSD in the ocean. We have simply adopted it as the best method currently available and shown, through a sensitivity analysis, that our calculations and thus probably our measurements, can tolerate relatively large uncertainties in the cut-off sizes of filters. We have been as ethical as possible in presenting all the data collected, striving to show all the limitations of our study (e.g., Figure 10). The reviewer's conclusions on our work appear to be mostly based on Fig. 10, where some PSDs are problematic. However, we have also shown several pieces of evidence that support our conclusions. We would have hoped that the reviewer had also carefully evaluated these additional pieces of evidence. Our fractionated particulate scattering measurements and hypothesis regarding the particle size distribution are consistent with theory, even when large uncertainties in the filter cut off are accounted for (see the closure achieved when modeling c_p data in Fig. 8a and Figs. 9b,d,f, as well as results of the sensitivity analysis in Figs. 9b,d,f). We have validated our fractionated chlorophyll-a measurements by means of independent HPLC data (Fig. 7c) and shown that they are consistent with our size-fractionated ACs-based chl-a, when the latter are available (Fig. 7d). Thus, with all due respect, we disagree with the reviewer that "*it is difficult to argue that this approach may have some usefulness*" and that "*the fractionation with filters is not quantitative and dependable*". We have provided multiple lines of evidence in support of our conclusions. We believe we have thoroughly discussed the limitations of such method and presented data that support and, at times, challenge it. We assume full responsibility for such limitations. However, we firmly believe that these observations that question existing hypotheses

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should be presented, even if not all the necessary information is available.

Regarding his concern that "*special laboratory tests with fractionation and Coulter measurements were not done*": One of the strengths of our fractionation approach is that, to minimize particle aggregation, modification of the particle size distribution, filter clogging, and retention of significant amounts of particle smaller than the nominal pore size, we filtered in the field large volumes of water through very-large-area filters. In addition, we presented several pieces of evidence in support of such fractionations (see above). Thus we believe that "special laboratory experiments" are out of the scope of this study. However, experiments are currently being planned for future cruises.

Regarding his reservations on using flow-through measurements instead of in-situ ones: we have presented several pieces of evidence to demonstrate the consistency of our data with other independent measurements as well as their consistency with existing bio-optical relationships. We have achieved closure within less than $10 - 20\%$ with radiometric data collected in-situ. [The unmeasured and, thus, assumed absorption by colored dissolved organic matter represents no more than $5 - 10\%$ of the total absorption coefficient in surface waters of the Equatorial Pacific water sampled (see answers to specific comments). Thus, this assumption has only a minor importance in the modeled radiometric data.] We have now also compared and found consistency between our c_p data from the Cstar transmissometer to a similar, although not as well characterized, instrument mounted on the CTD. We have shown that our ACs-based chlorophyll-a concentration (chla) measurements do not show a significant bias with respect to independently determined HPLC estimates and show very good precision (10%, pg. 301 lines 5-14). Finally, we have shown general consistency with existing bio-optical models (Figs. 5 and 6) and demonstrated that our particulate backscattering ratios are well within the ranges reported in the literature (pg. 313 lines 9-12). Therefore, as also acknowledged by the other reviewer, we have presented ample evidence to demonstrate that our flow-through data are of high quality.

"*This paper also emphasizes a correlation between the particulate beam attenuation*

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and particulate backscattering coefficients. The authors suggest that owing to this correlation the backscattering coefficient derived from satellite data could serve as a proxy for phytoplankton abundance or biomass, similarly to information provided by particulate beam attenuation. It is not clear what is meant by "abundance" or "biomass". Is the actual number concentration of phytoplankton cells or the amount of phytoplankton carbon implicitly implied in these terms? I think it would be highly problematic to claim that these concrete measures of abundance and biomass can be derived from these optical properties. Therefore, some comments would be in order on how the inherent optical property can be interpreted as phytoplankton biomass in the context of specific biological questions."

We begin our answer to this comment by considering that relatively conserved first order relationships between particulate organic carbon (POC) and b_p (or b_{bp}) have been verified in the field (e.g., Claustre et al., 1999; Gardner et al., 2006; Stramski et al., 1999; Stramski et al., 2008). One way to interpret the observed covariation of POC with b_p (or b_{bp}) is that the relationship between the (back-)scattering cross-sections and the carbon content of the "average particle" is, to first order, constant. This constancy of the (organic) carbon-specific (back-)scattering cross-sections is not expected a-priori, because the particles contributing to POC and the (back-)scattering coefficients vary over a large range of sizes and may have different compositions (e.g., Stramski et al., 2008).

On the other hand, numerous laboratory studies have shown that the phytoplankton carbon-specific scattering cross-section is constrained between about 2 and 4 m^2/gC for cells belonging to different groups (from cyanobacteria to diatoms) and grown under different conditions (see DuRand et al., 2002) for a summary of most of the existing data sets). Thus, based on the laboratory data currently available and on the particulate vs. phytoplankton scattering proportionality (see our argument in the introduction of the discussion paper), it should be less surprising that the b_p vs. phytoplankton-carbon relationship exists than a b_p vs. POC relationship. From an observational perspec-

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tive, relatively strong relationships between POC and b_p (or b_{bp}) are repeatedly demonstrated in the open ocean and the range of POC-specific scattering cross sections (1.5-3.8 $m^2/gPOC$) is in agreement to that measured in the lab for phytoplankton carbon despite differences in geographic areas, protocols, and instrumentation (Behrenfeld and Boss, 2006; Claustre et al., 1999; Gardner et al., 2006; Stramski et al., 1999; Stramski et al., 2008). Our explanation for these findings is that either phytoplankton contribute most of POC and b_p (or b_{bp}), or that the other particles affecting POC and b_p (or b_{bp}) covary with phytoplankton and, possibly, have similar carbon-specific scattering cross-sections. Unfortunately, the direct measurement of phytoplankton carbon biomass is rare relative to POC measurements, so a similar analysis has not been conducted. Nevertheless, substantial indirect evidence for a b_p (or b_{bp}) vs. phytoplankton-carbon relationship does exists (Behrenfeld and Boss, 2006; Behrenfeld and Boss, 2003; Behrenfeld et al., 2005; Huot et al., 2007; Westberry et al., 2008).

Finally, the variability in the carbon-specific scattering cross-section of phytoplankton will introduce uncertainties in the conversion of b_p to phytoplankton carbon. However, one needs to compare these uncertainties in scattering-based phytoplankton carbon biomass to the variability in chlorophyll-based phytoplankton biomass. It is indeed well documented that the ratio of chlorophyll-a to phytoplankton carbon can vary by almost two orders of magnitude (e.g., MacIntyre et al., 2002; Behrenfeld et al., 2005). Thus, we believe that scattering measurements represent a reliable alternative to chl-a for estimating phytoplankton carbon biomass in the open ocean.

In the revised manuscript, we have expanded our treatment of this subject by adding a new sub-section “Phytoplankton carbon from scattering coefficients” in the discussion.

Responses to Specific comments

(1) The title of the paper: I find ‘Direct’ in the title confusing.

We have changed the title to: “Significant contribution of large particles to optical backscattering in the open ocean”

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(2) p. 295, line 3 Replace 'phytoplankton-like particles' with 'low refractive index homogeneous spherical particles'

We have changed it to "low refractive index (phytoplankton-like) homogeneous spherical particles"

(3) p. 297, line 13 and everywhere else in the manuscript where relevant 'Palmer' should be 'Parmer'.

Corrected, thank you.

(4) p. 297, line 20 Are you suggesting that by making a measurement of the wavelength corresponding to the emission maximum of the light source, your determination of "effective" wavelength of beam attenuation measurement is actually correct? This "effective" wavelength is a function of both the light source and the spectral response function of the detector. The same question applies to your determinations of 'effective' wavelengths for the ECO-BB3 instrument.

The detector of the WetLabs C-star transmissometer is a silicon photodiode (UDT) that is characterized by a relatively flat spectral response over the spectral range of the red LED emission. This small spectral variation in detector response (about 0.8% over the spectral width at the half maximum of the peak emission) causes a negligible shift in the emission spectrum of the LED. Thus, it is accurate to estimate the effective wavelength of the C-star by measuring the spectral emission of its LED.

With respect to the ECO-BB3 sensor, the spectral responses of the detectors are similarly featureless. However, the spectral transmittance of the interference filters placed in front of these detectors need also to be taken into account. We had done so in the discussion paper, even though we did not mention it. We have now added the following explanation: ", after accounting also for the spectral responses of their detector and interference filters." to the relevant sentence.

(5) p. 298, line 10 Wouldn't it be desirable to have the internal surfaces black mat

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rather than black glossy to minimize reflection?

By using a black glossy surface we minimize back-reflection of photons. Even though part of the light is reflected *forward* (i.e., away from the instrument), the increasing number of interactions between these photons and the baffle, as they travel towards the side of chamber opposite to the instrument, ensures that most of these forward reflected photons are eventually absorbed. Thus, the part of the chamber opposite to the instrument effectively acts as light trap (we have added a reference in the text), as confirmed also by the relatively low values of $b_{b,wall}$ that were measured in the laboratory. Another practical advantage of using a glossy surface is that it is easier to clean.

(6) p. 300, lines 10-16 For the Coulter measurements it is critical to provide sample volumes that were examined and address possible limitations in particle counts if sample volumes were too small. The statement that triplicate measurements were taken is insufficient. If each replicate measurement was taken on a very small sample volume (which is normally the case with the Coulter technique), then the particle counts are usually noisy over a large range of particle sizes, perhaps with the exception of small sizes where particle abundance is high enough. Therefore, for typical oceanic particle concentrations, it is necessary to make many more than three replicate runs and accumulate the data from these runs to achieve a high enough total volume examined and high enough particle counts over a broad size range examined. This is a highly demanding but necessary procedure to get good Coulter data on open ocean samples. It appears that this procedure has not been followed in this study as the Coulter data presented in Figure 10 are very noisy across the entire size range examined, including even the small-sized particles.

We believe that the reviewer inaccurately concluded that the quality of our Coulter counter data is not "good" because he focused only on Fig. 10. The data presented in Fig. 10 of the discussion paper, refer only to the last fractionation experiment which was conducted in the clearest waters sampled during the entire cruise (see Fig. 1 of discussion paper). In addition, in Fig.10 we have presented 8211;on a linear scale-

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all the replicated PSD measurements without accumulating the number of particles per bin for each replicate and then normalizing by the cumulative volume of the three replicates (this was stated in the caption of Fig 10). Thus, one should a-priori expect a noisier plot, than when replicates are accumulated and presented on a log-log plot (as is typically done). We have now improved this figure.

We agree with the reviewer that sample volumes should be provided because they determine the precision of the measurements. In the revised version of the manuscript we have reported these volumes in the expanded PSD method section (3replicates X 1 ml=3 ml total volume sampled). We note that accumulation of the number of particles per bin over the replicates and normalization to the total volume sampled is mathematically equivalent in this study to computing the mean number of particles per bin per volume of the three replicates. In the discussion paper we computed *median* values of the volume-normalized replicates and not their means. We have now corrected this imprecision and computed the means of the replicates and recalculated the total number of particles per ml and the PSD slopes. Since the mean and median of these data do not differ considerably, the new results are entirely consistent with the older ones (new:3.51 ± 0.34 vs. old:3.57 ± 0.47).

To further improve the precision of our measurements, we have also accumulated data over several bins and generated new bins with significantly improved precision. A detailed explanation of this procedure is provided in the revised version of the manuscript. Briefly, we generated a reduced set of bins from the original 256 raw bins measured. The precision of the number of particles per bin (N/bin) can be computed from their variance, which follows Poisson statistics and, thus, is equal to N/bin. Thus, the precision of the newly generated bins are considerably higher than those in the raw data. Regardless, the statistics derived from these data do not differ considerably from those reported in the discussion paper (new:3.46 ± 0.42 vs. old:3.57 ± 0.47). This is likely because the information content of the noisier, but more numerous raw data is approximately the same of that of the fewer, but more precise data accumulated over larger bin

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sizes. Importantly, all these newly derived and more precise PSDs have coefficients of variation (CVs) of at most 30% and typically $< 15\%$ at the $8\,\mu m$ bin. CVs are considerably lower for bins corresponding to ESD$< 8\,\mu m$ (i.e., 10%). We consider this level of precision sufficient for the purpose of our study.

We have also added an additional consistency check where we computed the efficiency factor for attenuation of the average particle using bulk c_p data and total geometric cross-sections derived from the PSD measurements. The results are in agreement with theoretical predictions and indicate consistency between PSDs and c_p measurements, further supporting our statements about the high quality of our data.

(7) p. 302, line 12-13 I think that if the calibration method for scattering instruments is based on standard beads, at least 2 or 3 different particle sizes should be used. Generally the calibration is not a trivial task. The nearly-monodisperse populations of standard beads exhibit a complex shape of volume scattering function with multiple maxima and minima at different scattering angles. The presence of these peaks and their amplitude and angular width can affect the calibration of the scattering detector that has some angular response over a finite range of scattering angles. This is why the use of one particle size does not appear to me like a robust approach for calibration.

We agree with the reviewer that by comparing calibration results from beads of different size one could increase the confidence in the derived calibration coefficients. However, we used the calibration technique suggested by the manufacturer. This is the standard way of calibrating ECO-BB3 meters (e.g., (Twardowski et al., 2007). Particulate backscattering data obtained using this calibration method were found to be consistent within $< 10\%$ to other data calibrated by means of reference plaques (e.g., (Boss et al., 2007; Twardowski et al., 2007).

(8) p. 306-307, Section on IOP validation using radiometric data I suggest removing this section. This type of closure exercise has several sources of difference, in particular not all the inputs needed to run radiative transfer simulations were measured, so

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some assumptions had to be invoked. I do not see how this exercise can serve as a robust quantitative tool for validating your IOP measurements. All it shows is that the modeling results and measurements agree to within 10-20

(see also our answer to the general comments).

To the best of our knowledge, this sentence is referring to the missing aCDOM data (possibly also to sky conditions). We note that aCDOM in the surface waters of the Equatorial Pacific was only a minor component of the total absorption coefficient needed for our validation exercise ($5 - 10\%$). These small contributions influence negligibly the Rrs values calculated. Therefore, we do not agree with the reviewer that this section should be removed. Within the stated uncertainties it does contribute to the quantitative assessment of our IOP measurements.

(9) p. 307-308, Section on Bulk measurements What is the correlation coefficient for the plots shown in Figure 5? The relationships involving chlorophyll-a in Figure 6 are poor. What is the correlation coefficient? Clearly, your data support the notion that correlating the scattering properties with chlorophyll-a does not lead to good results.

We do not believe that the correlation coefficient is the appropriate metric for providing a quantitative understanding of the goodness of these relationships. That is why we reported the bias and precision with which the data can be reproduced by our relationships. However, for completeness, we now report also the correlation coefficients.

To facilitate the comparison with Fig. 5, we now report such metrics also for the Chl-based relationships.

The statement on p. 308 (lines 5-6) about the relatively constant average values for the $b_{bp} : c_p$ and b_{bp}:Chl-a ratios presented in Fig. 4c can be misleading. To me, the bottom line is that these ratios are highly variable. When one looks at selected portions of these plots, they show differences in the 'average' values. So the statement about the relative constancy of average values is just based on arbitrary choice of the portion of

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data used in averaging.

Had there not been a relatively constant $b_{bp} : c_p$ ratio, we would not have observed the strong first order relationship between these two variables that instead we have presented. We have now modified the text as follows:

"At 526 nm, the $b_{bp} : c_p$ ratio exhibited a relatively constant median value along the cruise track (0.0112 ± 0.0013). On the other hand, the and b_{bp}:chla ratio was more variable ($0.0066 \pm 0.0020\,\mathrm{m^2 mg^{-1}}$; Fig.4c). Both these ratios displayed diel variations, which were absent form the bulk properties."

(10) p. 308, Section on Along track size-fractionated IOPs As explained in my general comments I have little confidence in quantitative data derived from fractionation experiments.

As stated above (see our answer to the general comments), we have presented several pieces of evidence that support our fractionation experiments. In addition, we have shown through a sensitivity analysis that our calculation, and thus the scattering properties, can tolerate relatively large uncertainties in the filter cut-off sizes. Thus, although we respect the reviewer's opinion, we would have hoped that our work had not been dismissed without a thorough evaluation of the data presented.

(11) p. 309, line 11 The lower limit of your Coulter measurements is 1.4 um. Is it reasonable to suggest the presence of peak centered at 1.5 um, which is so close to the lower limit of detection? The prudent practice of experimentalists is to ignore data from several bins near the lower limit of Coulter detection.

Although we are aware of potential problems with the bin(s) corresponding to the lowest size(s), the data collected by our instrument did not show an anomalous behavior at the lowest size bin(s) where the particle number is typically high. Thus, we believe that the peaks observed by our Coulter counter data around 1.5um are real. In addition, all fits to the data have been carried out for bin sizes larger than 2um and smaller than

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8um.

(12) p. 310, Section on the 3rd fractionation experiment The data presented in Figure 10 suggest that the protocol to collect the particle size data was inadequate to obtain large enough particle counts and to minimize noise due to counting statistics. Significantly larger volumes (i.e., more replicate runs) should have been accumulated to obtain reasonably good PSDs.

This issue has been addressed above in our 'Response to General Comments', as well in the response to reviewer's comment (6).

In addition, as the authors point out and as discussed in my general comments, the Coulter data clearly show that the approach to fractionate samples with filters is not quantitative and can be unreliable (for example, higher particle counts were here obtained for the <1 um fraction than the <3 um fraction). (13) p. 313, lines 13-14

The Coulter counter also showed that the data collected for the <5um and <0.7um size fractions were consistent with a successful fractionation. We therefore disagree with this comment. See also 'Response to General Comments' response to comment (6) above.

(13) p. 313, lines 13-14 Support the statement about tight correlation in Figure 5 with the values of the correlation coefficient.

We have done so in the revised text that now reads: "b_{bp} and c_p (or b_p) were tightly correlated both at 470 and at 526 nm ($r = 0.87$ and $r = 0.90$ for the c_p vs. b_{bp} relationships at 470 and 526 nm, respectively) and the fitted..."

(14) p. 313, discussion of diel effects Previous studies of diel optical variability in phytoplankton cultures showed that changes in refractive index (in addition to cell size) can be an important source of variations in optical cross-sections on time scales of a few hours. It seems possible that the diel variability in cellular refractive index may induce a quantitatively different effect in b_{bp} and c_p.

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We agree. We have added the words “and/or compositional (Stramski and Reynolds, 1993)” to the relevant sentence.

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