Responses to comments on the manuscript "Inversion of the volume scattering function and spectral absorption in coastal waters with biogeochemical implications" by Dr. G. Fournier

Dear Dr. Fournier,

We appreciate your positive evaluation of our manuscript. In the following, we respond to the two general comments you made regarding the VSF-inversion methodology.

Sincerely,

Xiaodong, Yannick, Deric, Alan and Joseph

----- Comments by Dr. Fournier

General comments.

I have found this paper to be innovative and quite thorough in its description and analysis of a remarkably complete experiment in terms of the instrumentation used. It is certainly worthy of publication.

Thank you.

I will therefore restrict my comments to some general caveats about the approach that amplify some of the comments of the authors themselves. A first specific caveat concerns the use of Log Normal distributions. Even though they are convenient mathematically and a reasonably good fit the size distribution one must be really quite careful particularly with the zero order Log Normal when using it to evaluate the cross-sectional area or the volume of the particles as this distribution has a very slowly decaying asymptote and the peak of the second and higher moments can in fact in some cases lie several sigma's away from where the parameters of the distribution where measured. An offset Gamma distribution may in fact be a better fit and has the benefit of a well behaved asymptote which restricts the dispersion of the moments. It also has a basis in physics as it accounts for the distribution of crushed minerals (see Rosin-Rammler or Weibull distribution ) and would possibly be a good fit to the mineral part of the NAP.

Mathematically, you are absolutely correct. The left figure exemplifies this case, where three "similar" distributions at zero-order have dramatically different second order moments (area) and third order moments (volume). The right figure shows a case, where three distributions have the same effective size and variance [*Mishchenko and Travis*, 1994]. In this case, all the three distributions have similar estimates of area and volume. Therefore, a key question is how we measure/define the similarity of different distributions. Please note that all these distributions shown in the figure would generate very similar VSFs.

All of these distributions have been proposed to represent the particle size function. Zhang et al. [2011] discussed in details the rationale behind log-normal that we chose. Probably in the future, we should evaluate again the effect of using Gamma or other functions.



Three distributions with similar modal size. The two values are mean area  $(\mu m^2)$  and volume  $(\mu m^3)$ , respectively.



Three distributions have the same effective size and effective variance.

My other caveats are more of a general nature and concern the fact that when solving an undetermined inverse problem one must be extremely careful in assembling the constraints to make sure that the assignation itself does not imply the results. I.e. As in the case of LISST if we assume N populations with Log Normal distributions and use a Mie solution we will obtain a fit to any data the instrument collects. We have merely found the parameters of our model that fit the data. There is nothing that guarantees us that the model is anywhere close to the reality.

Choosing candidate particle populations is not trivial. As explained in Zhang et al. [2011], the kernel function was built through a rigorous sensitivity analysis. The parameters used to define the candidates are all based on the published observations. The results [*Czerski et al.*, 2011; *Zhang et al.*, 2011; *Twardowski et al.*, 2012; *Zhang et al.*, 2012] from our inversion method have been validated well against the independent measurements on particles and bubbles in the ocean.

Even though the authors are not using a Mie solution and have in fact wisely chosen a non-spherical model for the particles, it is still very simple when compared to real structures which have shells and inclusions with varying indices and transparencies. The effect of complex structure is in fact dominant in the back-scattering hemisphere as it is the part of the phase function for large particle that is controlled almost entirely by reflection from the envelope and internal components. Simply assuming a single shell will almost double the backscattering from any particle, never mind more complex internal structures. The assignation of a very small particle component which will obviously have a Rayleigh or Rayleigh-Gans distribution with very large proportion of backscattering will always fill any gap left between the theory for large particles and the experimental data in the backscattering hemisphere. This component may however in a very large part be there just to compensate for the inadequacy of the model used for the structures of the large particles. To my mind this is an open point at this time and is the one significant but unavoidable weakness of the approach presented by the authors. There may be ways of carefully teasing out weather those very small particles are really there or not from other correlations in the data from the various instruments. The correlation would obviously need to be strong for the argument to hold.

In summary the paper is excellent. The VSF inversion is reasonable as an approach but a great deal of care must be applied as in the case of all underdetermined inverse problems where the actual constraints imposed on the solution may dictate the results. As always, one has to be very careful in solving inverse problems that you not simply get back what you put in. In this respect the one significant but unavoidable weakness of the approach presented by the authors is the assumption of the large particle phase function and whether its inadequacy to account for internal structures and other complex features of real particles is what creates a gap between theory and data that they need to fill with a large very small particle component.

This is a very good point, and another reviewer, Dr. Dall'Olmo, also had a similar comment. Below is what we discussed in the text regarding this question.

"...On the other hand, theoretical simulations make unrealistic assumptions about particles. For example, the assumption of homogeneous spheres is known to lead to lowered estimates of backscattering than equivalent non-spherical (e.g., Clavano et al., 2007) or non-homogeneous particles (Kitchen and Zaneveld, 1992; Zhang et al., 1998; Quirantes and Bernard, 2006). In computing the kernel function for the VSF-inversion, particles are represented by homogeneous asymmetrical hexahedral. While an asymmetrical hexahedral is nearly an extreme morphological opposite to a sphere, the assumption of homogeneity might have led to an underestimation of backscattering, particularly for relatively large (i.e., non-VSP) particles, which in turn may lead to an artificial elevation of the retrieved backscattering contribution by VSP particles. We do not know the uncertainty associated with this homogeneous particle assumption. However, our results have shown [Chl] estimated for phytoplankton particles, which are known to have a variety of internal and "shell" structure among different species, agreed well with the independent HPLC data."

## References

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