Dear Reviewer, we are grateful to you for your comments and suggestions, which have helped to improve our contribution. Below you can find our answers to all your comments, addressing the modifications performed in the paper.

Reviewer 2 (RC C9713):

Overall comment

Comment: The biggest weakness of this paper is that it presents relatively complex concepts and models in a manner that needs a better organization. In particular, a summarizing flowchart or table is necessary that shows the inputs used for each model/algorithm and in each test case, the PSD, the type of particle assumed, etc., as well as the assumptions of the model, the equations used (refer to equations in this paper or elsewhere). Such a flowchart/table will greatly help the reader be able to follow how each model is applied, in a forward or inverse manner. Also, the units are sometimes not given, please give units consistently everywhere, including captions/axes labels.

Answer: We have tried to clarify the descriptions provided along the paper by adding new Fig. 1, Fig. 2 (both in the Introduction) and Table 1 (in Section 3):

- Fig. 1 shows some phytoplankton particles and the axially symmetric figure that best characterizes their shape. It also shows the type of shape characterization that can be used with Lorenz-Mie (only spheres) and *T*-Matrix methods (more complex shapes).
- Fig. 2 shows the three steps followed to analyse each method: First, the forward models (basically, Lorenz-Mie and *T*-Matrix) are used to obtain the inherent optical properties (IOPs) of a selected configuration (using as inputs the wavelength-dependent refractive index, $m(\lambda)$, the PSD and the particle shape). Then, the inverse models (described above) are used to estimate the initial refractive index using the IOPs obtained in the first step. Finally, the estimated refractive index along with the assumed refractive index are used to analyze the accuracy of the inverse model.
- Table 1 summarizes the inputs used for each model, their outputs, the type of particle assumed, the assumptions of the model and the equations used.

The description of each figure and table has been introduced in the appropriate place of the text (usually, above each figure and table). Units to all equations and variables have also been added at the end of the equations.

Comment: I recommend the addition of a table of variables, symbols and units used. In many cases you discuss methodology mixed with the results. You even introduce new concepts such as BFGS later in the paper. All these are better placed in methods, and/or in a table/flow-chart such as I suggest. Admittedly, sometimes text flows better if you do introduce some of these methods later where you do, so this comment does not always apply.

Answer: As stated in the previous answer, several improvements have been added in the form of figures, flowcharts and tables, to clarify some parts of the paper. All units have also been added to the equations and some of the Figures (a table has not been added only for the units since there are only a few).

The reason for not introducing BFGS in more detail, which is another optimization method as it is the Genetic Algorithm, is because its performance is much worse than the Genetic Algorithm when applied on the examples described in the manuscript. The results were only added in order to state that other optimization algorithms were also tested (a part from BFGS we also tried with

Nelder-Mead, Conjugated Gradient, etc.), but still the Genetic Algorithm provided the best solutions. We did not find necessary to make the paper longer with the introduction of a new method that does not present any meaningful improvement. As explained in the text, the Genetic Algorithm presents some disadvantages, being the convergence time the most important one, but its accuracy level is not achievable by any other algorithm.

In order to clarify the reason for not introducing in more detail the BFGS algorithm in the paper, the following text has been added in Section 4.1.3:

"Other optimization algorithms were also applied to determine if similar results can be obtained with a significant reduction of the computation time. **However, since none of them led to any meaningful improvement, they are not introduced here**. As an example, Fig. 7b shows the results…"

Comment: In the real world, given IOPs of a whole seawater sample, an average complex index of refraction would be retrieved by the presented methods. This average is weighted according to the PSD and the indices of refraction of the individual types of particles present. You should discuss this clearly somewhere and preferably also derive this weighting and state what is actually retrieved. See Eq. 8 in Boss et al. (2001) (see below), and refs. therein. This would be very useful to the ocean color research community.

Answer: The major goal of the paper is to point out:

1. the need to use alternative approaches for complex-shaped particles to those based on Lorenz-Mie (i.e. those based on *T*-matrix), and
2. to demonstrate that it is possible to get accurate retrievals of the refractive index with inverse methods based on these alternative methods, using as input parameters *in situ* measurements (IOPs, PSD and shape) that can be obtained routinely with existing oceanographic instrumentation.

In this first paper we have selected simplified synthetic scenarios that demonstrate these ideas. The practical question about how to apply these new ideas in real (and much more complex) scenarios implies many questions to consider, assuming the need to take into account complex-shaped particles: is it reasonable (or useful) to try to get a single averaged value in this case? Since complex particles cannot be characterised with a single volumetric parameter (the radius of the sphere) and there could be several refractive indices associated to each particle, which could be the best averaging approach? Maybe there will not be a single answer to this last question and the averaging methods would depend on the "dominant particle community". All these questions are very important to the ocean color research community, but will require a dedicated effort to try to answer them. Probably they are topics for developing new research lines in the future.

Comment: You need to discuss the applicability of these models to remote sensing data. Is it feasible for them to be applied to IOPs derived from ocean color remote-sensing reflectance? The problem with this may be that many operational remote-sensing inversion models for IOPs have in them an implicit or explicit assumption about the index of refraction when they were developed, so it would become a circular reasoning. Retrieving the index of refraction from space would improve our ability to distinguish sources of backscattering from each other in the ocean, so a paragraph in the discussion about that would be really important.

Answer: The proposed new inversion method use as input parameter in situ measurements (IOPs, PSD and shape) that can be obtained routinely with existing oceanographic instrumentation. We hope that these approaches will also have further applicability with other type of optical measurements (one case will be remote-sensing reflectance), but this topic is beyond the scope of the paper, as it implies a much more complex inversion scheme.

Nevertheless, we have mentioned this option in the final discussion:

"Besides, all these approaches can also have further applicability with other type of optical measurements, as for instance with remote-sensing reflectance. As an example, Fig. 19 shows the spectral backscattering of the three test cases, the homogeneous sphere, the coated sphere and the homogeneous cylinder Many operational remote-sensing inversion models for IOPs use an implicit or explicit assumption about the refractive index, and, in combination with these methods, could be severely improved. Retrieving the index of refraction from space would improve the ability to distinguish sources of backscattering from each other in the ocean. To this end, a much more complex inversion scheme should be developed".

Comment: You discuss the limitations of having limited degrees of freedom in one instance (Sect. 4.2.3). The same applies to multispectral sensors of several wavelengths only. What is the feasibility of retrieval of the bulk m value from space with the advent of hyperspectral sensors such as the planned NASA PACE mission?

Answer: In principle, the inverse method is applied iteratively to a single wavelength, so it will be possible to apply with both multi-spectral and hyperspectral in situ instrumentation. Regarding its use with remote sensing instrumentation, this is a two-step inversion problem. First, it is necessary to estimate the IOPs and PSD from the AOPs (RS) measurements (which, as it is mentioned in the previous comment, goes beyond the scope of the paper). Once the IOPs and PSD have been obtained, it is possible to apply the proposed method in the paper. Here, the main constrain is that the inverse method is sensible to inaccuracies of the input parameters, so the quality of the refractive index retrievals will depend strongly on the accuracy of the estimated IOPs and PSD in the first inversion step.

<u>More specific comments</u>

Comment: Title needs revision of word order and, more importantly, it needs to better reflect that this paper refers to aquatic optics.

Answer: Title has been modified as suggested here and in the pdf to: "Methods to Retrieve the Complex Refractive Index of Oceanic Particles: going beyond simple shapes"

Comment: Abstract: Needs major revision. Several sentences need to be added to set the context (aquatic optics), state that complex index of refraction determines IOPs and as such is input to forward models. Refractive indices are not easy to measure, thus are often assumed or retrieved with inverse modeling, etc.

Answer: Abstract has been completely rewritten to put the addressed questions and results in context, as well as the first part of the Introduction.

Comment: Introduction: You need to state more clearly how this test procedure works. I.e. do you start with particle(s) with known complex refractive indices, and then do a forward model (Mie, T-matrix, specify), than pass the IOPs to the inversion models and compare the results to the known inputs. Is this the scheme of your tests in this paper? It does not become very clear.

Answer: Yes, your description is correct. For the forward model, Lorenz-Mie or T-Matrix can be used. Lorenz-Mie, only for homogeneous or coated spheres, and T-Matrix, for the same kind of particles and also for more complex shapes.

To clarify this in the paper, we have added Figure 2 and its explanation in the Introduction:

“The comparison has been done following the three steps presented in Fig. \ref{Fig0b}. First, the forward models (basically, Lorenz-Mie and *T*-Matrix) are used to obtain the inherent optical properties (IOPs) of a selected configuration (using as inputs the wavelength-dependent refractive index, m(λ), the PSD and the particle shape). Then, the inverse models (described above) are used to estimate the refractive index using the IOPs obtained in the first step. Finally, the estimated refractive index is compared with the original one to obtain the accuracy of the inverse model.”

Besides, in each example, some comments have been added. In example 1 -Section 4.1- (new comments highlighted in black):

“In particular, the BHMIE code, originally from Bohren and Huffman and modified by B.T. Draine, was used **as a Forward Model** (additional features were added, such as polydispersion and the computation of the Stokes scattering matrix).”

In example 2 (Section 4.2):

“Using this PSD with the previous refractive indices in the BART code from A. Quirantes (Quirantes, 2005) (**a Forward Model** based on the Aden-Kerker theory to calculate light-scattering properties for coated spherical particles), the absorption, scattering and extinction coefficients of Fig. 10a, and the volume scattering function of Fig. 10b were obtained.”.

And, in example 3 (Section 4.3):

“In order to find which is the most accurate model for the characterization of such complex shapes, an example considering 100 prolate cylinders per mm^3 with a diameter-to-length ratio equivalent to 0.8, the PSD of Fig. 14 (showing the radius of an equivalent volume sphere with a slope parameter \xi = 3, effective radius r_{eff}=3.2 μm and effective variance v_{eff}=0.005 resulting in R_{min} = 0.8 μm to R_{max} = 3.6 μm), and the assumed refractive index of Fig. 4b was simulated using the T-Matrix algorithm (Mischenko1996, Mischenko1998) **as a Forward Model**”.

Comment: pg. 18730, line 11, eq. 14 – this needs a better explanation because it is confusing as it is presented. Do you mean that the values in Eq. 14 are already the values relative to seawater, as you use them throughout the paper? Show the actual equation to calculate the relative index, given the complex indices of the particle and the medium.

Answer: Yes, this is what it means, as a matter of convention. We have followed the same assumption with respect to the normalization of the refractive index as in other publications on light scattering. See for instance Bernard et all 2009, Twardowski et all 2001, etc.

We have clarified the text just before Section 3.1:

“Note that this paper assumes effective refractive indices relative to seawater, which has a constant value of $m_{water} = 1.334 + i0$ (Hale et al, 1973). Absolute values can be recovered using $m_a = m \times m_{water}$.”

Comment: Eq. 15 – I believe the Twardowski model retrieves just the real part of the refractive index, not something equivalent to |m|.

Answer: It is not clear, since all along the paper only refers to it as the “bulk refractive index”. However, we have considered this suggestion (and the one related to the imaginary part, which is assumed to be 0.005), and have applied the following modification:

- Fig. 6a is shows the real part of the refractive index instead the absolute value, and also compares the imaginary part.

- Table 2 shows the error on the real and imaginary parts of the refractive index.
- Descriptions of Fig. 6a (Section 4.1.1) and Table 2 (Discussion).

Comment: pg. 18731, line 2: Fig,. 1a caption in Twardowski et al. (2001) says that k was fixed at 0.005, therefore they do not ignore/neglect k in this model, which is a fit to their Fig. 1a.

Answer: It has been corrected (see answer above).

Comment: pg. 18734, line 3 – you use inconsistent notation for the complex index of refraction – it is 'm' above, and 'n' here. Please use consistent notation everywhere.

Answer: I has been corrected.

<u>Additional comments</u>

Comment: Sect. 4.2 Why not apply the Twardowski model to these coated particle IOPs and see how the retrieved bulk n compares to the input ones? I.e., why can't the Twardowski model be applied to the cases other than homogeneous spheres?

Answer: It must be considered that the Twardowski model was developed to apply to entire particle populations that are assumed to follow a power-law size distribution, and is fundamentally different from the other models, developed to apply to isolated phytoplankton cultures which, in general, follow a different size distribution.

In our contribution, only the first example uses a power-law size distribution, precisely, to apply the Twardowski model and compare its results with the other models. The other two test cases follow particle size distributions more similar to those presented by monocultures, and therefore, the Twardowski model was not used in order to avoid unfair comparisons.

To clarify this in the text, the following sentence has been added just before Section 4.2.1:

"Note that the Twardowski model is not applied to avoid unfair comparisons with the other methods (it was designed to be used with entire particle populations that are assumed to follow a power-law size distribution)."

Comment: The Bernard (2009) reference is not given in the list of references.

Answer: It did not appear due to a problem of compilation in Latex. It has been solved.

Comment: In all figures with the output IOPs, consider showing spectral backscattering as well – would be very useful for remote sensing relevant applications.

Answer: Although it is possible to add extra plots showing spectral backscattering, we wandered up to which point it would be really relevant for remote sensing applications, taking into account that all tests cases in the paper were developed using synthetic and probably too simplistic data to compare with real remote sensing products. For the time being, we have only added the spectral backscattering of the three test cases in the Discussion Section.

Comment: Sect. 2.1: Whenever you use equations not derived by you, please give citations.

Answer: They have been added (except when it is obvious where do they come from; for instance, in Section 3.1, The Twardowski Model, the whole Section is referred to Twardowski et al., 2001. In such cases, references have been omitted).

Comment: + or – in the m = n+ik expression? Different sources list it differently. So it would be useful if you clarify this.

Answer: The sign of the complex part is a matter of convention. The notation in the paper, which is usually used by physicists, corresponds to waves with time evolution given by $e^{-i\omega t}$.

In the paper, it has been clarified in the second paragraph of Section 3 with:

"The sign of the complex part is a matter of convention (it can also be defined with the negative sign). The notation above corresponds to waves with time evolution given by $e^{-i\omega t}$".

References

Comment: Boss E., M.S. Twardowski, and S. Herring, 2001. Shape of the particulate beam attenuation spectrum and its relation to the size distribution of oceanic particles. Applied Optics, 40, 4885-4893

Answer: According to our findings, the exact name of this reference is "Shape of the particulate beam attenuation spectrum and its inversion to obtain the shape of the particulate size distribution", as seen in:

https://www.osapublishing.org/ao/abstract.cfm?uri=ao-40-27-4885

This reference already appeared in the manuscript. If it is not correct, please, tell us and we will modify it.

Comment: Please also note the supplement to this comment:

http://www.biogeosciences-discuss.net/12/C9713/2016/bgd-12-C9713-2016-supplement.pdf

Answer: All comments have been considered and applied, but some specific comments need to be answered. For instance:

- Just before equation 19, "explain what xi is". Xi was already defined in the first paragraph of Section 3.1 (The Twardowski Model).
- In Fig. 3, "say what conv. means." It is already explained in Section 3.4: "After the evaluation, the algorithm may stop if either a maximum number of generations (each generation is a new vector of solutions) or a satisfactory fitness level have been reached. If the convergence condition is not fulfilled, the best solutions are selected and separated…"
- Section 2.2, "most authors give D+dD, explain your choice". It is a matter of convention. We follow the same definition as in Bricaud and Morel, 1986, Bernard et al, 2009, etc. A reference has been added.
- First paragraph of Section 4, "how does this combination work?" This is only an introduction of the Section. Please, refer to Section 4.2.3 for further details.
- Section 4.1.3, "are these code, or algorithms? Explain the acronyms." As stated, they are frameworks for programming. Acronyms have been explained. References may provide further information for the interested readers.
- Section 4.1.3, "Isn't this a very small number of generations. I think typically many more are used. Justify your choice." That's a good point, since normally it is usual to achieve at least 100 generations. Indeed, it strongly depends on the length of the initial-population vector and cross-over and mutation percentages, among other parameters of the genetic algorithm. In this particular case, using an initial vector of 2000 solutions and 50 and 20% of probability of crossover and mutation, not significant improvements are generally found beyond the tenth generation. It has been clarified in the text at the end of the first paragraph of Section 4.1.3.

- In Section 4.2, “why a peak at 500 nm, Chl does not normally have a peak of absorption there? is it accessory proteins/pigments?”. Some examples shown in Stramski et al, 2001, and Bernard et al, 2009, exhibit similar peaks (with a lower amplitude, to be honest) around 500 nm. We tried to emulate such refractive indices in order to have assumed values close to those measured in nature.
- Section 4.1.2 and 4.2: “Explain Aden-Kerker and Hilbert Transform”. In order to not further extend the length of the paper, references have been added for both mathematical definitions.