Reviewer #1

Main comments

The manuscript has been re-organized and significantly improved since the first version. Some valuable work analyses are presented and discussed, based on shipborne measurements made on water samples collected in complex estuarine environments. At least part of the results obtained are (potentially) interesting for a better understanding of relationships between the optical, physical and biogeochemical properties of particles in suspension in natural waters. The discussion at the end of the study is rather speculative, notably due t the lack of complementary measurements (organic carbon, phytoplankton pigments, iron) and/or theoretical computations.
I still have many detailed comments concerning:
1. the section 'Data and methods' in which (i) the different parameters must be better defined, with appropriate physical units, (ii) the processing of optical and biogeo-chemical measurements must be clarified (see detailed comments below)
2. the section 'Results' in which could be partly re-organized for better logics
3. the section 'Discussion' which too speculative and could be shortened.

We addressed the concerns of points 1-3. Also, the analysis of mass-normalized optical coefficients for PIM and POM was removed given the relatively small number of samples and large error of estimates.

Therefore, I recommend a major revision of the manuscript.

Detailed comments

Page 5 lines 20-25
The absorption-beam attenuation meter (ac-s, WetLabs) is already calibrated by Wetlabs in pure water, so that it measures light absorption and attenuation on top of pure water contributions. Therefore, you do not have to subtract pure water absorption and attenuation coefficients from your measurements. Please correct in the text.
we clarified that pure seawater contributions are removed during the calibration of the ac-s instrument

Also why not applying the "proportional" method recommended by Wetlabs, that developed by: J. R. V. Zaneveld, J. C. Kitchen, and C. C. Moore, "Scattering error correction of reflecting tube absorption meters," Proc. SPIE 2258, 44-55 (1994).

this point was explained in previous corrections. See below
Zaneveld et al. (1994) proposed the residual scattering correction method proportional to wavelength. This is analogous to the flat baseline correction but it assumes a spectral dependency on scattering errors. This approximation is in debate. Some studies have reported a spectral dependency on volume scattering functions or particulate backscattering ratios (Chami et al., 2006; McKnee et al.,

2009;McKnee et al., 2013). But there is doubt regarding if this assumption can be generalized (McKnee et al., 2013).

Page 6, line 16
You should introduce/define here the Junge exponent often used to describe the size distribution of marine particles

it was defined before since it needed to be discussed in introduction

Why 35.17um?

because it is the midpoint of the logarithmic size range

Page 6, Line 27
'the spectral slope of the particulate attenuation coefficient is negatively related with the mean particle size for particles smaller than 20 mm.'
What is the reference for that??

sorry but we couldn't find this sentence along the text

Page 7, lines 4-6
Untrue: if SPM includes a significant contribution of phytoplankton (chlorophyll-a) to light absorption, which kas absorption peaks at440 nm and 675 nm.
Your Equation (5) only applies in the case of non-algal particles.
Please correct this.
done

Equations (6-7): units?
this is explicit in the list of acronyms, however we added units too before describing the equations

Lines 10-15: totally unclear whereas you define the mass-specific absorption and scattering coefficients (in m2 /g) of the mass-specific absorption cross sections for mineral and organic fractions of SPM??? And what is the difference between these coefficients. Please clarify these definitions and always provide the appropriate physical units.

the definition of these sigmas or cross sections are in section 2.7
the units again are in table 1 but we placed units too along the text

where m is the mass in g m-3 for each size class i….? g m-3 is a mass per unit of volume, which means a concentration of SPM…… not surprising I do not understand the definitions of your parameters!!

sorry, good catch. now missing text added

>> Please rewrite sections 2.5, 2.6 and 2.7 and clarify the definitions of the parameters you compute then use to describe how the SPM size and chemical composition influence the SPM optical properties. As it I found it very confusing.
these sections were clarified in terms of units and re-organized

Page 8, section 3.1
Please use either decimal numbers (e.g., 0.0111) or percentages (11%) to report the contributions f small/large and organic-rich/poor particles on the measured optical properties. This should make the text easier to read.

done

Line 10
'The uncertainty of x calculations, as estimated from 2 standard errors, varied between 1.6 and 10.2%'
>> To be computed and reported in the section 'Data and Methods'

done

page 8
'3.1 Spatial variability of microphysical properties of SPM'
What do you mean exactly by 'microphysical'?

microphysical properties is a term also used in atmospheric optics for aerosols and it refers to physical properties at the microscopic level (e.g., size, shape).
The opposite would be macroscopic level or macrophysical properties (e.g., clouds in in atmospheric sciences). In oceanography, an aggregate should be considered as a macrophysical phenomenon.

Overall I do not clearly understand the logics with the other two Results sections entitled: '3.2 Mass-specific optical properties of SPM' and '3.4 Optical proxies' (should be 3.3)
I would rather suggest to
1) report the mass-specific coefficients and corresponding spectral slopes, analyse their spatial variations and compare them to values already published in similar environments,

we appreciate your suggestion but the way we organized the data makes sense for us. Mass-normalized optical properties and optical proxies are two different topics in the publication that are clearly separated.
We can not perform spatial analysis of mass-normalized optical properties with respect to other pubs because we dont have really have resolution for chemical fractions (only 2 regions) and for size fractions there are NO studies in the literature other than ours for comparing. We compared geographic variations on mass-normalized optical coefficients in Table 4

2) analyse the influence of SPM size on the mass-specific absorption and scattering coefficients, then
the size and particle composition effects are discussed when talking about the mass-normalized optical properties for different size fractions of SPM in Table 2. We cant with chemical fractions
the focus of this study is examine the influence of PSD and particle composition on optical properties of SPM fractions and not total SPM.

3) analyse the influence of SPM composition (PIM, POM) on the mass-specific absorption and scattering coefficients.

the focus of this study is examine the influence of PSD and particle composition on optical properties of SPM fractions and not total SPM.

page 8, section 3.2
Are your mass-specific optical properties of SPM in good agreement with values reported in similar environments? Please discuss this first.

this is discussed in section 4.3. In general there is an agreement with published values reported in other environments for aspm* and bspm* as derived from GF/F filtrations. However, differences were detected for optical cross sections of chemical fractions. This is also discussed.

Why finally presenting results for the 400-650 nm spectral interval?

the spectral range of ac-s measurements in this study is from 400.3 to 747.5 nm. So we don't have wavelengths below 400 nm in the UV spectral range. In the NIR, only wavelengths below 710 nm were examined due to the large error of ac-s determinations at longer wavelengths.

Line 23
Here I do not understand why you report values in 1/m. Based on the definition of this parameter (Equation 6, i.e. absorption coefficient (in 1/m) divided by a concentration of SPM (in g m-3) we would expect values in m2 g-1….??? Please check and clarify all

this.sorry about the typo. now units are corrected

Line 25
'Similar to ai*, highest bi* values (up to 5.7 m2 g-1 for l'
and here you report values in m2 g-1…..

these are mass-specific optical coefficients and the units are correct

Line 26

‘corresponded with size fractions having particles with the smallest and the largest diameter’
What do you mean?? Please rewrite

these coefficients were measured in size-fractionated samples corresponding to particle size ranges of 0.2-0.7 microns and >10 microns.

Lines 27-28
‘In general, the spectral slope of bi* was very variable in all size fractions (-6 10-5 to 6.28 10-3 nm-1) with the greatest spectral changes associated to particulates greater than 10 mm.’
??? Why suddenly talking about the spectral slope of the SPM scattering coefficient? This spectral slope was not even defined!

that's true. this result is disconnected and was part of an old manuscript version.
it is irrelevant and was deleted

Page 9, line 22
‘Also for SPM fraction having the largest particulates’
?? largest grain size?
we rewrote the senetence: 'Also for the size fraction of SPM with a grain size >10 microns'

Reviewer #2

<u>General comments</u>

Page 2, lines 1-2 of original manuscript. Algorithms based only on CSPM will never have the accuracy required for optical inversions because SPM is undefined optically, an unknown mixture of inorganic and organic matter. Therefore partition of SPM into at least major chemical composition classes (PIM and POM) and estimation of size distribution are required independently for optically-based remote sensing algorithmsof primary productivity and suspended mineral dynamics of “disappearing shorelines” etc.

we added this important point in the introduction

Estimating SPM from summing PIM and POM estimates has at least a definable and reproducible measurement error. Basically there is a confusion here between systematic errors and measurement errors. The ubiquitous (and wrongly proposed in my opinion) algorithms based on just SPM will provide precisely calculated inaccurate estimates.

also, we emphasized this point in introduction

Author's response for Page 3, lines 15-20 of original manuscript:

No, different filters were used in this study to fractionate 0.4-0.7 and 0.7-10 micron fractions This sentence was clarified as:

Size fractionation of SPM into four size classes (>10 μm, 0.7-10 μm, 0.4-0.7 μm, and 0.2-0.4 μm) was done after sequentially filtering the original samples through preweighted membranes having a diameter of 47 mm and a pore size of 10 μm (Whatman, polycarbonate), 0.7 μm (GF/F, Whatman, glass fiber), 0.4 μm (Whatman, polycarbonate), and 0.2 μm (Nucleopore, polycarbonate), respectively. Also, we add the whatman GF/F was used for PIM and POM determinations.

What is unclear here is why the glass fiber GF/F filters were used for both “size fractionation” and retrieval of “total SPM.” If the GF/F filters are removing all the SPM then sequential fractionation should not yield anything removed by filters smaller than the 70 micron nominal pore size of the GF/F filters.

this paragraph was clarified. GF/F filters were only used for SPM fractions. Total SPM was computed by adding the weight of the 4 size fractions. Thus, total SPM represent particulates > 0.2 microns

Page 7, line 19 of revised manuscript. The authors should quote Richter and Stavn (2014) which actually gives instruction on accomplishing Model II multiple regression utilizing R code
done

Page 13, lines 12 and 19, Page 14, lines 29 and 30 of revised manuscript. Don't the p values quoted in these lines indicate that the correlations quoted are not significant?

there was a typo here and the sentence was rewritten

Page 11, lines 20-21 of original manuscript. Suggested wording: These relationships will be useful in investigating local and regionally-limited relationships and properties of SPM. Without separate independent studies of true optical properties of PIM and POM, and of PSD, these relationships will remain problematical.
We added the first sentence to the end of the conclusions paragraph
I recommend both sentences at the end of the conclusions.
done