

Dear Editor,

Thank you for looking after our manuscript “Variability of light absorption coefficients by different size fractions of suspensions in the southern Baltic Sea” by Justyna Meler, Dagmara Litwicka and Monika Zabłocka.

All suggestions and amendments proposed by the Reviewers were taken into account by the authors.

Please find our detailed response for Reviews.

**Reviewer#1:**

Reviewer's comment:

**General comments:**

*Meler et al. present a descriptive study documenting measurements of particulate, phytoplankton, and detrital absorption coefficients obtained from size-fractionated water samples collected from nearshore and offshore waters of the Baltic Sea. The water samples comprise a significant range in biogeochemical properties based on SPM and Chla observations, although the sampling locations were confined to a southern sub-region of the Baltic sea. The dataset contains 38 elements. The results do not indicate significant differences between size fractionated samples in terms of absorption properties. Further, the results indicate differences in mass-specific (but not Chla-specific) light absorption coefficients between larger (micro) versus smaller (nano and pico) organic and inorganic particles.*

Author's response:

We thank the Reviewer for insightful critical comments on our work.

*Although the general topic could potentially be compliant with the journal's scope, the manuscript does not satisfy the journal's criteria to merit publication, as follows:*

*Scientific Significance: The study is not sufficiently comprehensive based on insufficient number of observations (n=38) and small spatial extent of the sampling area, compared with the variety of oceanic conditions, physical forcings, biological conditions, and the terrestrial, riverine, and anthropogenic inputs to the Baltic Sea. The key-finding (that Chla-specific absorption properties of size-fractionated samples are not significantly different from each other within the authors' n=38 dataset from southern Baltic Sea waters near Poland) would be more compelling if the study was more expansive, or if the authors could better establish the significance of their null results. The study may also not be generalizable outside of the Baltic Sea, and because the observations span a small geographical sub-region of the Baltic sea (the southern waters around Poland) the results may also not be representative of the optical properties elsewhere in the Baltic Sea;*

Author's response:

The research results presented in the manuscript include 38 observations collected during 1 year of research conducted during the implementation of a small scientific project. The implementation of 1 measuring station was very time-consuming. Water intake, size fractionation and then filtration of the original water samples and fractions took from 4 to 7 hours, depending on the "purity" of the water. The data was collected during 3 several-day cruises on the Baltic Sea, covering waters with different optical properties, and periodically conducted research at Sopot Pier (monitoring of temporal variability). The research was focused mainly on the Gulf of Gdańsk, where the ranges of variability of optically active components

are representative for the southern part of the Baltic Sea, which was shown in earlier works by Meler et al. (2016a and b, 2017). However, 1/3 of the observations during cruises also included stations outside the Gulf of Gdańsk, i.e. open and coastal waters without direct influence of river waters. In the Gulf of Gdańsk, we conducted research at stations located in the plume of the Vistula River (the main, large river that has a large impact on optically active components in the waters of the Gulf) and at stations distant from the mouth of the Vistula River. Based on literature data on other Baltic regions, in particular off the coasts of Sweden, Finland and Latvia, the results presented in our manuscript are rather not representative of these areas, however, knowledge about the diversity of the Baltic waters in the aspects we study is still limited.

The presented results may not be of global interest, however, the Baltic Sea is an important element of the ecosystem for 11 countries of the Baltic Region under the Monitoring and Assessment Strategy of the Helsinki Commission (HELCOM, 2013). The HELCOM strategy is intended to provide assessment and monitoring of data that can be used both for international assessment by HELCOM and for monitoring at the national level. The strategy is designed to ensure both data production and dissemination of information by contracting parties of EU Member States and meeting the requirements of several EU Directives such as the Marine Strategy Framework Directive (MSFD), the Water Framework Directive (WFD), Habitats and Birds Directives, EU Strategy for the Baltic Sea Region (EUSBSR) and EU Integrated Maritime Policy (HELCOM, 2013). First of all, the Strategy aims to support ecosystem-based maritime spatial planning (MSP) in the Baltic Sea by enabling high-quality spatial data and assessment tools for MSP purposes. For the purpose of regional assessment, HELCOM divides the Baltic Sea into different waters. These basins have been described in the document "HELCOM Sub-areas of the Baltic Sea" (Annex 4; HELCOM, 2013), according to separate hierarchical division levels, depending on management needs.

Author's response:

In the new version of the manuscript, we have modified selected parts of the text. We have supplemented the Introduction section with the above information (Lines 95-102, 110-112)

Reviewer's comment:

*Presentation Quality: Comprehension of the manuscript is inhibited by low presentation quality. In particular, the authors' combination of the Results and Discussion materials into a single section significantly detracts from the presentation of each, and at times made comprehension of the manuscript difficult, or resulted in ambiguity in elements of the methods or results. I suggest that the authors separate the results and discussion in order to add clarity.*

Author's response:

We agree with the Reviewer that the first draft of the manuscript may have been difficult to read. The manuscript was reorganized following the Reviewer suggestion. We have separated the Results section (lines 233-600) and the Discussion section (lines 601-718).

Reviewer's comment:

***Additional (general) comments:***

*The authors do not adequately demonstrate the other dimensions of variability in their dataset, e.g., due to seasonal factors, site-specific differences like onshore vs offshore, biomass, or total particle content. One way that the authors could have helped with this would be to color the markers in the scatter plots to indicate other parameters, e.g., by seasons or by whether the site was nearshore or offshore.*

Author's response:

As suggested by the Reviewer, in Figures 2-8, we marked the season and the sampling area with colors and various markers. In the case of cruise data, the division is as follows: February-winter, April-spring, September-autumn, and spatial division into samples from the Gulf of Gdańsk and open and coastal waters. In the case of data from Sopot Pier, the data is presented as a separate group covering all 4 seasons: winter (December 21 - March 20), spring (March 21 - June 22), summer (June 23 - September 20) and autumn (September 21 - December 20th).

We supplemented the Material and Methods section with a description of the sampling area, where we presented the seasonal cycle of biological activity in the Baltic Sea, as well as a division into regions and a description of hydrological conditions (lines 119-135, 138-141).

Reviewer's comments:

*The authors did not identify differences in Chla-specific optical properties between size fractionated samples. I'd suggest that the authors investigate or discuss what other factors (e.g., distance from shore, biomass, wind-driven mixing, contribution of inorganic particles) may have been associated with the variability in observed Chla-specific absorption properties within size fractions.*

Author's response:

The average Chla-specific absorptions (as well as mass-specific absorptions) presented by us were determined only for samples in which a given fraction was dominant, and not for all samples for a given fraction. This was to try to show the average absorption spectra for a given particle size class, similar to what Ciotti et al. (2002) for waters that were optically less complex than the Baltic Sea we studied. As suggested by the reviewer, we divided the data set into seasons and sampling areas.

As suggested by the Reviewer, we have supplemented the Results section with the division of the study area into sampling areas: the Gulf of Gdańsk, the separated Sopot Pier, and open and coastal waters. We also made a division into the sampling season (lines 241-246). All of these modifications we have shown also in new Figures 2-3 and 5-8.

We have modified Figure 2 and its description (lines 255-272, 283-302)).

We have added Table 2 showing the proportions of SPM and Chla in size classes (micro, nano, ultra, pico, or ultra+pico) in total SPM and Chla, for all data and divided by regions (Gulf of Gdańsk, Sopot Pier and open and coastal waters).

We have also extended the POM/SPM description to seasonal and spatial division (modified Figure 3 and relevant description in lines 360-367).

We have modified Figures 5-7 and extended their description (for Figure 5: lines 428-436, for Figure 6: lines 437-441 and for Figure 7: lines 454-460).

We have modified Figure 8 and Table 3 (in old version of manuscript it was Table 2, and we deleted old Table 3). In new Table 3 we presenting the contributions of particles from different size classes to the total light absorption by all particles, detritus and phytoplankton in sampling regions (relevant description in lines 482-485, 492-508).

Reviewer's comments:

*Comparing the overlap in mean +/- std between data points is most useful when uncertainties due to environmental or methodological variability are well described (uncertain measurements of moderately dissimilar parameters can easily overlap). The authors do not convey uncertainty in their absorption, Chla, or SPM measurements, which would help to identify the extent to which overlap in absorption properties is or is not meaningful.*

Author's response:

In the case of the analyzed data set, the precision of the measurements of the light absorption coefficients and the concentration of chlorophyll a was not checked, because no duplicate samples were made. Checking the precision of the measurements of these parameters previously performed on a different dataset yielded the following results.

The precision of the measurement of light absorption coefficients using the IS method for 3 different filters from the same station was 4.96% +/- 2.91%. When measuring the concentration of chlorophyll a for duplicate seawater samples, the measurement precision was 5.3% +/- 1.5%.

In the case of SPM, according to the methodology, 3 subsamples are always taken and the measurement precision for 95% of the triplets was below 15%, and for all cases the average was 5.83% +/- 4.40%.

In the Materials and Methods section, appropriate descriptions of measurement precision have been added (lines 211, 220-221, 233).

Reviewer's comments:

***Minor (Specific) Comments:***

*Table 1: Is the section "Nano+ultra particles (2-20um)" intended to be Pico + nano particles (based on the sampling difficulty of the first 14 samples; L200-202)?*

Author's response:

Nano+ultra particles refers to the classic division into size classes according to Sieburth et al. (1978), where particles with a size of 2-5  $\mu\text{m}$  were still treated as nanoplankton and did not constitute a separate size class. In the case of pico + ultra particles, due to too much of these particles, the membrane filters were clogged and had to be replaced too often, and it was not possible to filter enough water volumes for filtration to obtain SPM and Chla, due to limited funds and time.

Reviewer's comments:

*Lines 303-312 and figures 5-7: I'd suggest that log scale R2 values are reported as well. These datasets are mostly log-normally distributed in both axes, and R2 calculated on the linear axes is strongly influenced by the points in the upper-right corner of the plot. For example, consider the high R2 despite low association of points in Fig 6 panel G.*

Author's response:

Figures 5-7 show the dependence of the light absorption coefficients of all particles, detritus and phytoplankton at 443 nm on the Chla and SPM concentrations on the log-log scale. The presented approximations are a power function  $y=A*y^B$  and the coefficients  $R^2$  correspond to these approximations.

We added this information in section 3.3 (lines 422-423).

## Reviewer#2:

### Reviewer's comment:

*The manuscript by Meler et al. investigated the size-fractionated absorption spectra of particles, phytoplankton, and non-algae particles (NAP) in the southern Baltic Sea. They also conducted the measurement of total and size-fractionated suspended particulate matter (SPM) and Chlorophyll (Chl) a concentrations and then examined the relationships between the absorption coefficients, SPM, and Chl a concentrations for each size fraction. They found that the SPM-specific absorption coefficients are a useful parameter to distinguish between large and small plus medium particle fractions. The data presented in this study is informative. However, this manuscript requires considerable alteration along the lines I have suggested below.*

### Author's response:

We thank the Reviewer for insightful critical comments on our work.

We modified selected fragments of the text and figures, in accordance with most of the Reviewer's suggestions.

### Reviewer's comment:

#### **Major comments**

*The description of total and size-fractionated Chl a-specific NAP absorption needs more detail. It is possible to understand the meaning for calculating the absorption coefficients of particles ( $a_p$ ) and phytoplankton ( $a_{ph}$ ) normalized by Chl a and SPM concentrations to see the contribution of each size component to the spectral shape and magnitude. However, I am not sure the significance of the Chl a-specific NAP absorption spectra and coefficient at 443 nm as shown in Figures 6a – e, and 9c, 10c.*

### Author's response:

Figure 6 shows  $a_d(443)$  vs Chl a and vs SPM coefficients, while Figure 9c and 10c show chlorophyll-specific  $a_d(443)$ . In general, if we can express  $a_p$  and  $a_{ph}$  in the form of a Chl a-dependent function, then so can  $a_d$ , since  $a_p = a_d + a_{ph}$ . For clean ocean waters (the so-called Case1) Bricaud et al. (1998) presented Chl a-specific NAP absorption  $a_d(\text{Chl a})$  can be determined from the difference of  $a_p(\text{Chl a}) - a_{ph}(\text{Chl a})$  (Woźniak & Dera, 2007). In the case of the optically complex Baltic Sea, these relationships are not so simple, and we just wanted to illustrate what they look like. For the analyzed data set, on average, 52% of suspended matter was organic matter of both autogenic and allogenic origin, and the contribution of inorganic matter to light absorption is not significant (Woźniak and Dera, 2007). Therefore, the absolute values of  $a_d$  in the analyzed set depend on the concentration of organic matter suspended in the water, which is not phytoplankton. Chl a-specific NAP absorptions are independent of the SPM concentration, and their values and spectral distributions are determined by the absorption properties of the suspension particles themselves, i.e. they depend on the chemical and physical properties of the material they are made of (chemical composition, optical properties, sizes, shapes).

Earlier, we heard the opinion that the concept of  $a_{\text{NAP}}$ , or "particles other than algae", is too empirical, because it consists of an unknown admixture of mineral and organic detritus. The very different refractive indices of mineral and organic matter make it impossible to interpret changes in  $a_{\text{NAP}}$  in any quantitative way. Mass-specific  $a_{\text{NAP}}$  based on SPM are of no value

without the partitioning of SPM into PIM and POM (Duarte et al. 1998, Richter and Stavn, 2014). On the other hand, chlorophyll-specific  $a_{NAP}$  may be of some value. Therefore, in our work we decided to show both approaches.

We have added comment in Discussion section in lines 674-684.

Reviewer's comment:

*I agree with the author's assertion that the data obtained by this study could improve the model to retrieve the inherent optical properties (IOPs) in the Baltic Sea (Lines 464 – 465). However, it is not clear that which of the results or relationships examined in this study would contribute to the improvement of the IOPs models and how to expand the results into the models for estimating the size parameters. Given that many cases have already been reported in the literature (as cited by the authors themselves in the Conclusion section), it would be advisable to explain specific information on the improvement of IOP models.*

Author's response:

Our research results are preliminary and very limited.

The analyzes presented by us are an introduction to further research, in which HPLC data and the actual particle size distribution should be taken into account in order to formulate an absorption model for particles in different size classes, similarly to Devred et al. (2006, 2011) and Brewin et al. (2010, 2011).

Our pilot studies on the study of the contribution of individual particle size fractions in the total SPM, Chl $a$  and related absorption properties for the southern part of the Baltic Sea indicate the need to develop this topic, especially for the remote estimation of the size structure of phytoplankton populations, the so-called PFT.

These sentences have been included in Conclusions in lines 757-762.

Reviewer's comments:

*The large part of the sentences in the Introduction reviews the previous literatures. Therefore, it seems to me that it is hard from reading the Introduction to understand why this study is needed. To better organized the introduction and objectives, I would encourage the authors to rewrite the section. Similarly, abstract and most parts of results and discussion sections, especially 3.2, 3.3, and 3.4, are not well organized. It is descriptive and is like a data report, making difficult to follow what is the new findings described in this study. However, I believe that the authors can elaborate.*

Reviewer's comments:

*Figure 2a showed the results of size-fractionated "SPM" in each sampling station. A more appropriate legend would be required for Figure 2a to better reflect the investigation of SPM.*

Author's response:

We agree with the opinion of the Reviewer. Sections Introduction, 3.2, 3.3, 3.4 have been modified. Figures and tables have been modified. The descriptions were extended with the seasonal and spatial division of the analyzed data set.

We have modified abstract (lines 11, 14-15, 22-23)

In the Introduction section, we have completed the goals of our research (lines 79-86, 95-102, 110-112). We have deleted sentence in lines 64-68.

We have supplemented the Results section with the division of the study area into sampling areas: the Gulf of Gdańsk, the separated Sopot Pier, and open and coastal waters. We also made a division into the sampling season (lines 241-246). All of these modifications we have shown also in new Figures 2-3 and 5-8.

We have modified Figure 2 and its description (lines 255-272, 283-302)).

We have added Table 2 showing the proportions of SPM and Chla in size classes (micro, nano, ultra, pico, or ultra+pico) in total SPM and Chla, for all data and divided by regions (Gulf of Gdańsk, Sopot Pier and open and coastal waters).

We have also extended the POM/SPM description to seasonal and spatial division (modified Figure 3 and description in lines 360-367).

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We have modified Figure 8 and Table 3 (in old version of manuscript it was Table 2, and we deleted old Table 3). In new Table 3 we presenting the contributions of particles from different size classes to the total light absorption by all particles, detritus and phytoplankton in sampling regions (lines 482-485, 492-508).

Reviewer's comments:

*Although average Chl a-specific absorption coefficients of phytoplankton generally decrease with increasing cell size because of self-shading, the authors showed the opposite trends as compared with previous work of Ciotti et al. (2002). Therefore, I feel that the package effect (as mentioned by the authors themselves in Line 410) may be open to further discussion.*

Author's response:

Figure 9 and 10 shows the average spectra of specific light absorption coefficients by all particles, detritus and phytoplankton for given fractions, determined for cases where a given size fraction was dominant (it is not an average for all measured coefficients for a given fraction). In the analyses, we used Chla determined by spectrophotometry, not by HPLC, so we do not know the share of individual pigment groups, so we are unable to determine the packing effect. Of course, taking into account that the largest share of micro particles was recorded for the SF13 station during phytoplankton bloom (large algae gathered at the beach in Sopot), we can assume that the packing effect occurred and was significant.

Reviewer's comments:

***Minor Comments:***

*Names of observed stations are missing in Figures 1, which make it difficult to refer to Figures 2 and 8 and SF04 and SF13 in Lines 417 – 427. The information will help readers understand the results more easily.*

Author's response:

SF01-SF16 refer to measurements on Sopot Pier and constitute a separate group showing temporal variability. We have improved the descriptions in the text. Figure 1 has been modified, station names have been added.

Reviewer's comments:

*I would suggest that the results of Figure 4, 9, or 10 be presented in a different way; for example, a box plot at satellite ocean colour bands with average spectra could be used. I think*

*that this make it easier for the readers to understand the importance of them. For example, please refer to Brunelle et al. (2012, doi: 10.1029/2011JC007345).*

Author's response:

We have presented Figure 4 as suggested by the Reviewer, as far as possible. However, Figures 9 and 10 in boxed form for selected wavelengths do not work in our case. The variability ranges of individual means and standard deviations for size classes overlap, obscuring the picture. Therefore, in the existing figures, we have bolded the average values for better visibility, and in the Table 4 we have compiled numerical values for selected wavelengths, corresponding to the ranges observed by satellite sensors such as Seawifs or OLCI (lines 595-600).

We have modified the description to Figure 4 (lines 385-394, 398, 402-407).