# 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 Rewiever 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 Subareas 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.

#### 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 clarify.

#### 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.

# 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.

The principal factor affecting the variability of the inherent optical properties of Baltic waters in the euphotic zone is the seasonal cycle of biological activity (Sagan, 1991; Olszewski et al., 1992; Kowalczuk et al., 1999, 2005; Meler et al., 2016 a,b). This cycle is governed by physical, biological and chemical processes, which cause the biomass and species composition to vary with time. As a consequence there are three main phytoplankton blooms: a spring bloom of cryophilous diatoms, which then transforms into a bloom of dinoflagellates; a summer bloom of cyanobacteria; and an autumn bloom of thermophilous diatoms (Thamm et al. 2004, Wasmund et al. 2001, Witek and Pliński 1998). The spring blooms can take place from early March to May, the summer ones in July/ August and the autumn ones from September to October (Wasmund et al. 1996, Thamm et al. 2004, Wasmund and Uhlig 2003). In winter, biological activity is minimal. The maximum runoff of river waters occurs at the turn of April and May, and it often coincides with the spring bloom of phytoplankton initiated by an increase in air and water temperature and more sunlight. River waters carry large amounts of organic dissolved substances (DOM) and nutrients that enhance phytoplankton blooms. The increased amount of phytoplankton in the surface water layer reduces the transparency of the water. In summer there are periodic floods following very heavy rainfall which together with strong winds can effect in upwelling, which causes cooler water to rise up from the deep layers of the sea. Such events affect the optical properties of waters in the coastal zone and Gulf of Gdańsk - see Olszewski et al. (1992), Kowalczuk (1999) and Sagan (2008).

Locations of sampling stations in our study were selected to obtain the greatest possible diversity of waters in optical terms. The research was carried out mainly in the Gulf of Gdańsk, but also in open waters and coastal waters outside the Bay of Gdańsk, as the weather permits.

The analyzed data set was divided due to the sampling area: the Gulf of Gdańsk (GG), and extracted from the GG - Sopot Pier (SF) (which shows time variability over 10 months and is the only one that takes into account the summer season), open and coastal waters (OCW) In addition, the data were divided due to the season of sampling. In the case of cruise data, the division is as follows: February - winter, April- spring, September - autumn. In the case of data from Sopot Pier, the data are presented as 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 20). For the 14 samples (collected during cruises in February and April), it was not possible to separate the ultra and picoplankton fractions, because the amount of suspension of 2-5  $\mu$ m clogged the membrane filters.

## 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 shown in Figures 2-3 and 5-8, the division into seasons and sampling areas.

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.

Descriptions to corrected figures 5-7 have been extended.

An extended description for Figure 8 is presented in text. Table 3, presenting the contributions of particles from different size classes to the total light absorption by all particles, detritus and phytoplankton, has been modified by sampling regions.

New Figure 2

In winter, when there is minimal biological activity in the Baltic Sea, it can be seen that the largest contribution in SPM (> 50%) had particles < 5  $\mu$ m, which can be seen in both GG and OCW. In the spring, this trend continued in OCW, while the contribution of micro and nano particles increased in GG. In the autumn period, particles < 5  $\mu$ m again had the largest contribution in the GG, with a predominance of ultra particles. In the case of OCW, the contribution of small particles decreased and nano and micro particles (> 5  $\mu$ m) > 60% dominated. The Sopot Pier dataset shows temporary variability over 10 months and is the only one that takes into account the summer season. It can be seen that in most cases, in winter, spring and autumn, the contribution of small (< 5  $\mu$ m) and medium and large (> 5  $\mu$ m) particles in the SPM is comparable, except for SF06. In summer (SF08-SF12 and SF15) particles < 5  $\mu$ m contributed the most to SPM, with pico particles predominating, only during phytoplankton blooms, where large algae gathered at the beach, samples were dominated by micro particles, and the proportion of small particles was below 20 %.

The right panel of Figure 2 shows the proportion of Chla in individual size fractions.

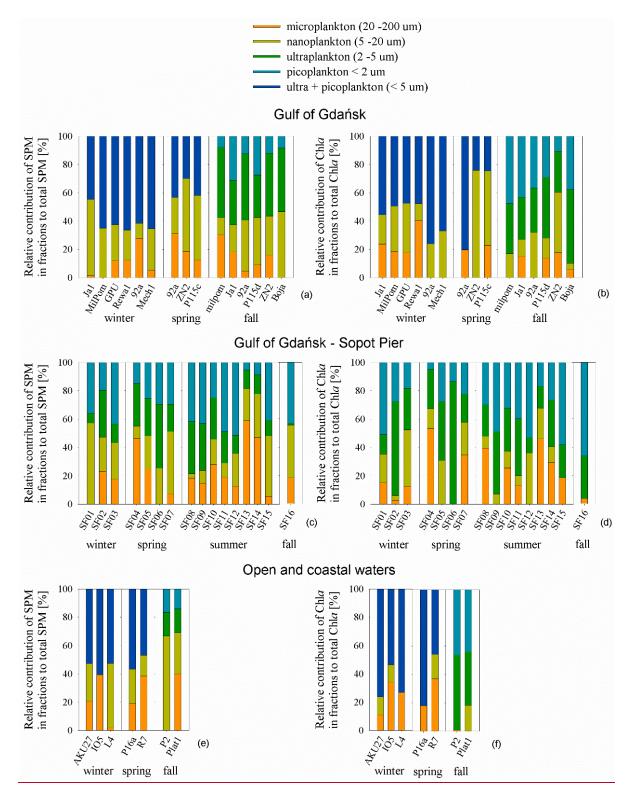


Figure 2: Relative contribution of SPM in the selected fraction to the total SPM – left panel (a, c, e), and Chl*a* in the selected fractions to the total Chl*a* – right panel (b, d, f), divided due to region (Gulf of Gdańsk, Sopot Pier, Open and coastal waters) and seasons

The average contribution of chlorophyll *a* in a given size class to the total concentration of Chl*a* for all data is the highest for pico-particles (35 %) and ultra-particles (35 %), while the average contribution of Chl*a* in nano and micro-particle classes is about 15 % each. The range of variability of the contributions of individual size classes in the total Chl*a* changed as follows:

micro-particles from 0 to 53 %, nano particles from 0 to 76 %, ultra-particles from 11 to 86 % and pico-particles from 5 to 66 %. (second part of Table 2). In the GG in the winter, Chla in the ultra+pico particles class had about 50% share, the rest was for medium and large particles, and it can be seen that despite the small contribution of micro-particles in SPM, the proportion of Chla in this size class was about 20% for stations Ja1, Milpom and GPU, and for Rewa1 about 40% (probably the particles lifted from the bottom contained a lot of organic detritus). In turn, for example, for station 92a, the contribution of micro-particles was about 30%, and the proportion of Chla for the same station is close to zero, which means that these particles were inorganic. In autumn, the contribution of  $Chl \alpha$  for particles < 5 um (except for the ZN2 station located closest to the mouth of the Vistula River) was on average about 80%. In the case of OCW waters, an average of > 60% of Chl $\alpha$  in the classes of small particles and about 20% of micro-particles was observed in winter and spring. In autumn, small particles accounted for more than 80% of Chla in OCW waters. The micro and nano particles in these waters were mostly inorganic. This also results from SPM analyses. At the Plat1 station, no Chla contribution was observed for the micro particle class and < 20% Chla contribution in the nano-particle fraction. At the Sopot Pier station, the proportion of Chla in the classes of small particles, < 5  $\mu$ m, in winter was > 60% with a maximum value of 95% observed on SF02. In the spring, the contribution of Chla in the class of small particles for SF04 and SF07 was about 30%, for SF05 about 65%, and for SF06 100%. In the summer season, the contribution of small particles  $< 5 \mu m$  was still dominant, except for experiments SF13 and SF14, during which phytoplankton blooms were observed and the contribution of medium and large particles was > 50%. In early autumn (SF16), despite a significant contribution of medium and large particle classes in the SPM, it can be seen that the share of Chla for these classes was negligible, while ultra and pico particles had the largest contribution.

#### New Table 2

|                            | all data       | Gulf of Gdańsk  | Sopot Pier     | Open and coastal waters |
|----------------------------|----------------|-----------------|----------------|-------------------------|
| SPM <sub>micro</sub> /SPM  | 17.2 %± 14.3 % | 13.3 %± 10.1 %  | 21.3 %± 16.3 % | 22.8 %± 16%             |
|                            | 0-58.9%        | 0-31.2%         | 0-58.9 %       | 0-39.9 %                |
|                            |                |                 |                |                         |
| SPM nano/SPM               | 28.3% ± 15.6 % | 31.6 %± 13 %    | 25.4 %± 14.1 % | 29.5 %± 19.9 %          |
|                            | 0-66.7 %       | 10.8 % - 53.9 % | 3.1 % - 57.3 % | 0- 66.7 %               |
|                            |                |                 |                |                         |
| SPM <sub>ultra</sub> / SPM | 26 % ± 13.9 %  | 41.2 %± 7.7 %   | 21.7 %± 11.8 % | -                       |
|                            | 1.1 % -49.9 %  | 30 %-49.9 %     | 1.1 % - 45.2 % | -                       |
|                            |                |                 |                |                         |
| SPM pico/ SPM              | 27 % ± 14.3 %  | 16.6 %± 9.3 %   | 31.6 %± 13.8 % | -                       |
|                            | 5.1 %-51.3 %   | 7.8 % - 31.3 %  | 5.1 % - 51.3 % | -                       |
|                            |                |                 |                |                         |
| SPM pico+ultra/ SPM        | 50 % ± 16.8 %  | 53.4 %± 12.8 %  | -              | 46.9 %± 4.6 %           |
|                            | 0-66.5 %       | 30 % -66.5 %    | -              | 45.9 % - 60.7 %         |
|                            |                |                 |                |                         |

Table 2: Contributions of particles from different size classes (micro, nano+ultra, nano, ultra, and pico\_ to the total SPM and total Chla (n=38). The mean values ± standard deviation (SD) and the variability range are given for all data and in division on sampling area.

| Chla <sub>micro</sub> /Chla                 | 15.8 % ± 14.9 %<br>0-53.4 %       | 13 %± 11.5 %<br>0-40.5 %           | 18.4%± 17.3 %<br>0-53.4 %         | 18.2 %± 14 %<br>0-36.9 %         |
|---|-----------------------------------|------------------------------------|-----------------------------------|----------------------------------|
| Chla <sub>nano</sub> / Chla                 | 18% ± 6.6 %<br>0-75.8 %           | 27.2 %± 19 %<br>0.2 % - 75.8 %     | 14.7 %± 12.4 %<br>0- 40%          | 8.6 %± 7.8 %<br>0-18.3 %         |
| Chl <i>a</i> <sub>ultra</sub> /Chl <i>a</i> | 35.3 % ± 16.6 %<br>10.7 % -86.5 % | 36.8 %± 8.3 %<br>29.1 % - 52.2 %   | 33.4 %± 19.1 %<br>10.7 % - 86.5 % | -                                |
| Chl <i>a</i> <sub>pico</sub> / Chl <i>a</i> | 34.7 % ± 15.4 %<br>5 % - 66.1 %   | 34.2 %± 12 %<br>10.7 % - 47.7 %    | 33.5 %± 17 %<br>5 % - 66.1 %      | -                                |
| Chla pico+ultra/Chla                        | 53.6 % ± 22.9 %<br>0-82.1 %       | 52.5 % ± 18.8 %<br>24.2 % - 80.2 % | -                                 | 73.2 % ± 14 %<br>45.8 % - 82.1 % |

#### New Figure 3

In Figure 3, for all data, it can be seen that the winter season, regardless of the place where POM/SPM samples were taken, assumes the lowest values from SPM. No trends were observed between the remaining seasons and sampling sites. In the case of micro particles, most of the GG samples in autumn are dominated by inorganic particles (POM/SPM < 25%). For nanoparticles, in the winter, inorganic matter dominated, and in the autumn, organic matter dominated. For ultra particles, no seasonal and spatial dependencies of POM/SPM vs SPM are visible. On the other hand, for pico particles, we observed that POM/SPM increases with the increase in SPM: in winter POM/SPM had the lowest values, then in spring and autumn it was on average 65% and the highest values reached in summer on average 80%.

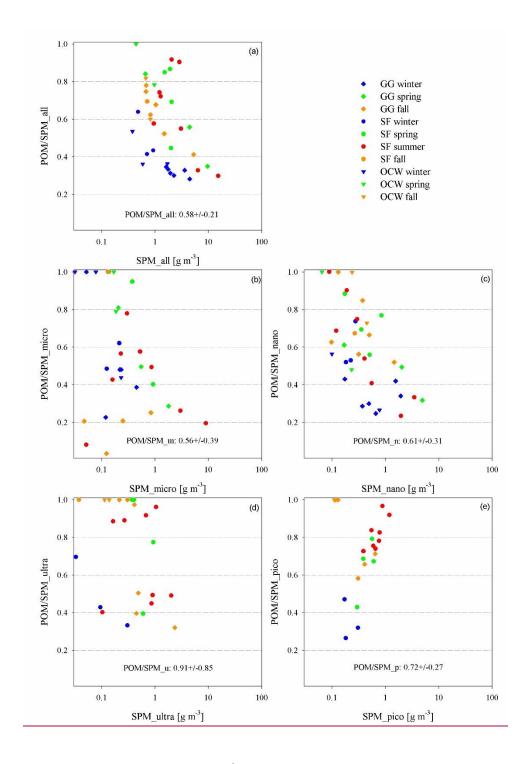


Figure 3: Relationships between the POM/SPM ratio and the SPM concentration for the original water samples and the size classes: micro, nano, ultra and pico. Mean values ± standard deviation are shown in the graph. Markers shapes and colours distinguish the season and sampling site (GG - Gulf of Gdańsk, SF - Sopot Pier, OCW - open and coastal waters).



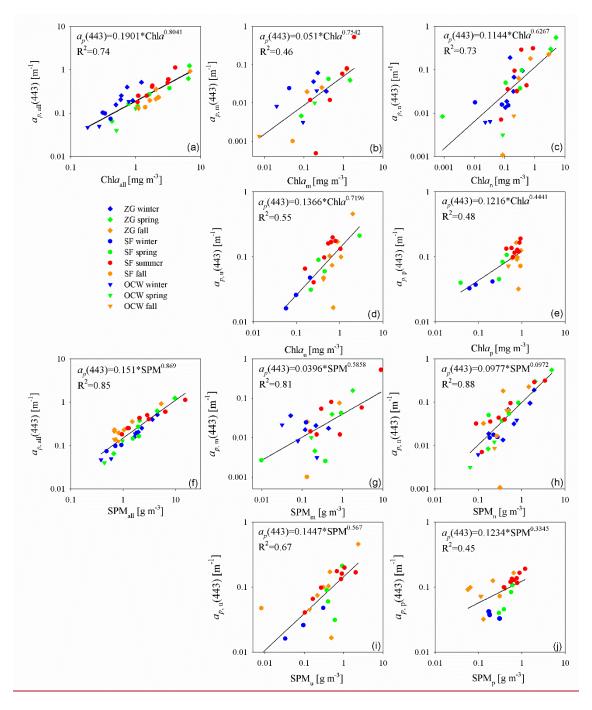


Figure 5: Relationships of the light absorption coefficients by all unfractionated particles (a, f) and in size classes: micro (b, g), nano (c, h), ultra (d, i) and pico (e, j) from the Chl*a* (a-e) and SPM (f-j), for the selected wavelength of 443 nm. Note that graphs have different axis scales.

Figure 5 presents the light absorption coefficients of all particles suspended in seawater for a wavelength of 443 nm depending on the concentration of Chl*a* and SPM, for the original (unfractionated) seawater samples and for the micro, nano, ultra and pico-size classes in log-log scale. The data was divided due to the season and place of sampling. Approximations are shown for all samples in power form (y=Ax<sup>B</sup>).

# (...)

The relationships  $a_p(443)$  vs Chla allow to distinguish the winter season from the summer season (blue and red points). In the winter season, the absorption values increase much faster with the increase of Chla than in other seasons. Taking into account the relationships between  $a_p(443)$  and Chla in individual size classes, the values are slightly dispersed, but there is also a difference between seasons. In the case of dependence on SPM, a division into seasons is also visible. For all particles in the winter and spring season abs coefficients increase slightly faster with increasing SPM than in summer and autumn. Individual size classes also show seasonal trends, however, further research is necessary to draw clear conclusions. As for the sampling area, as expected, OCW are characterized by lower concentrations of Chla and SPM than GG, and the related lower values of absorption coefficients.

## New Figure 6

The Baltic Sea is characterized by a large influence of anthropogenic factors on the optical properties of its waters, including the inflow of a large amount of dissolved and suspended organic substances with river waters into its catchment area (especially the Gulf of Gdańsk, which is strongly influenced by the waters of the Vistula). These are waters with complex optical properties that do not depend solely on Chl*a*, especially in the case of detritus. However, in order to compare, we showed how  $a_d$  vs Chl*a* dependencies look like. (...)

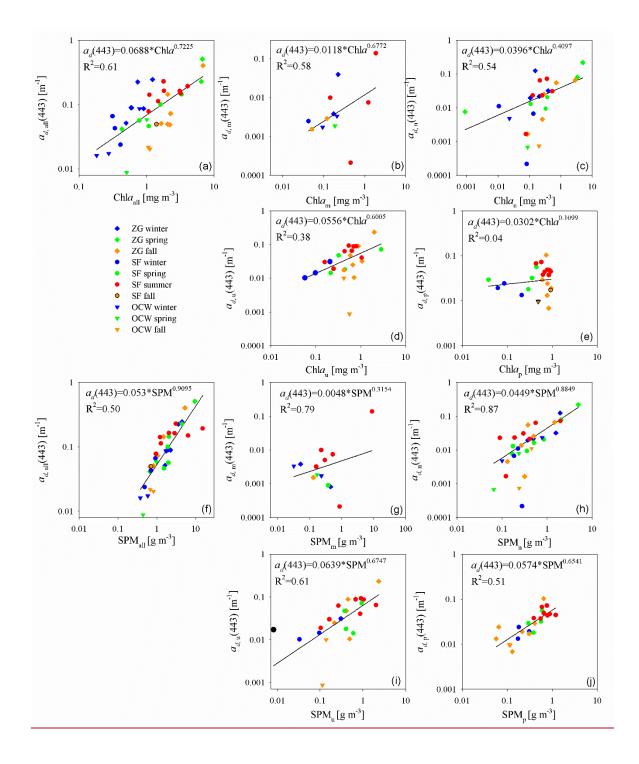
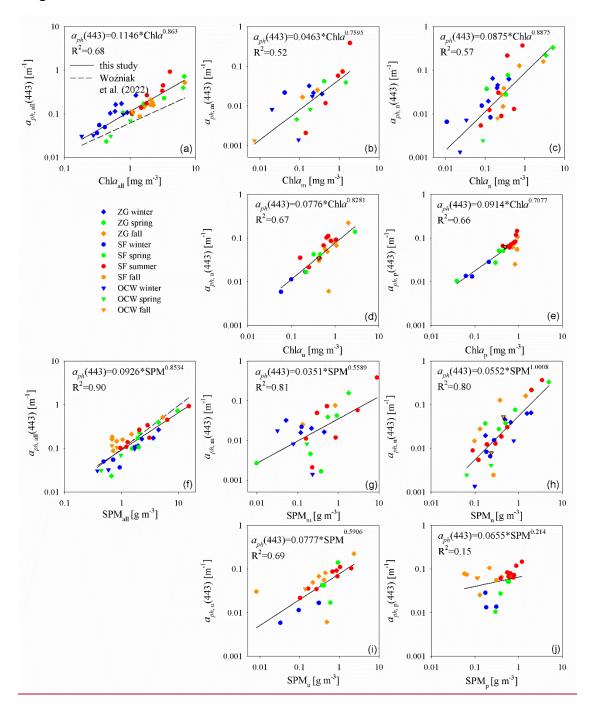


Figure 6: Relationships of the light absorption coefficients by all unfractionated detritus (a, f) and in size classes: micro (b, g), nano (c, h), ultra (d, i) and pico (e, j) from the Chla (a-e) and SPM (f-j), for the selected wavelength of 443 nm. Note that graphs have different axis scales.

New Figure 7



# Figure 7: Relationships of the light absorption coefficients by all unfractionated phytoplankton (a, f) and in size classes: micro (b, g), nano (c, h), ultra (d, i) and pico (e, j) from the Chla (a-e) and SPM (f-j), for the selected wavelength of 443 nm. Note that graphs have different axis scales.

As in the case of light absorption by all particles, seasonal variation can be observed, both for Chla and SPM dependence. The winter season is characterized by low Chla values and relatively low  $a_{ph}(443)$ . The spring season is characterized by the greatest range of Chla variability. Summer and autumn are characterized by Chla values >1 and  $a_{ph}(443)$  values highest in a year. Similar trends are visible in the case of division into size classes, with a large

sample dispersion for micro particles. The dependence of  $a_{ph}(443)$  on SPM is characterized by a high coefficient of determination  $R^2 = 0.9$ . We can distinguish the winter and spring seasons, which in most cases lie under the approximation curve, while the data collected in the summer and autumn seasons lie above this curve (see Figure 7).

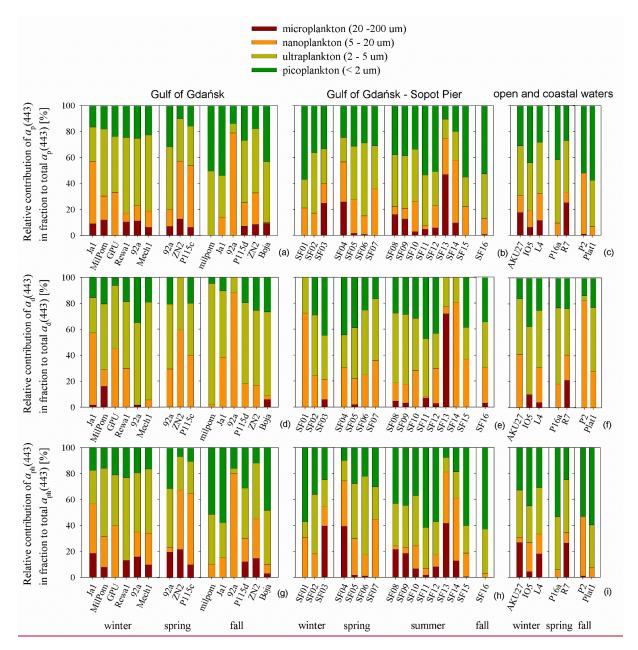
Table 3 contains such contributions in our dataset (all data, and divided for sampling area: GG, SF, OCW) divided into 4 size classes according to Ciotti et al. (2002): micro i.e. large particles ( $20-200 \mu m$ ), nano-particles ( $-5-20 \mu m$ ), ultra-particles ( $2-5 \mu m$ ) and pico-particles ( $<2 \mu m$ ).

#### New Table 3

Table 3: Contributions of particles from different size classes (micro, nano+ultra, nano, ultra, and pico) to the total light absorption by particles, detritus and phytoplankton for a wavelength of 443 nm (n=38). The mean values ± standard deviation (SD) and the variability range are given. for all data and in division on sampling area.

|                                      | all data        | Gulf of Gdańsk  | Sopot Pier      | Open and coastal waters |
|--------------------------------------|-----------------|-----------------|-----------------|-------------------------|
| $a_{\rm p,micro}/a_{\rm p}$          | 8.2 %± 9.6 %    | 6.6 %± 4.4 %    | 9.5 %± 12.7 %   | 8.8 %± 9.1 %            |
|                                      | 0-46.9%         | 0-12.7%         | 0-45.9 %        | 0-25.3 %                |
| $a_{\rm p,nano}/a_{\rm p}$           | 21.6 % ± 16.4 % | 24.8 %± 21%     | 20.6 %± 11.2 %  | 16.8 %± 13.1 %          |
|                                      | 0-78.9 %        | 0-78.9 %        | 2.8 % - 48.2 %  | 6.8 % - 47 %            |
| $a_{\rm p,ultra}/a_{\rm p}$          | 36.8 % ± 13.5 % | 42.3 %± 13.5 %  | 32.7 %± 11%     | 34.3 %± 14.6 %          |
|                                      | 0-58.6 %        | 7.2 %-58.6 %    | 15 % - 56.3 %   | 0-49.1 %                |
| $a_{\rm p,pico}/a_{\rm p}$           | 33.4 % ± 13.9 % | 26.3 %± 12.8 %  | 37.2 %± 13.1 %  | 40.1 %± 11.1 %          |
|                                      | 10 %-57.6 %     | 10 % - 54 %     | 10.3 % - 56.8 % | 27 % - 57.6 %           |
| $a_{\rm d,micro}/a_{\rm d}$          | 4.2 % ± 12 %    | 1.7 %± 4.1 %    | 6.3 %± 17.1 %   | 5 %± 7.4 %              |
|                                      | 0-71.9 %        | 0-16.1 %        | 0-71.9 %        | 0-21.1 %                |
| $a_{\rm d,nano}/a_{\rm d}$           | 29.5 % ± 22.7 % | 29.6%± 24.3 %   | 28.7 %± 20.5 %  | 30.8 %± 23.9 %          |
|                                      | 0-87.9 %        | 0.1 % – 87.9 %  | 1.2 % - 80.8 %  | 0.1 % - 82.4 %          |
| a <sub>d,ultra</sub> /a <sub>d</sub> | 44.9 % ± 16.2 % | 52.9 %± 18.1 %  | 39 %± 9.7 %     | 41.1 %± 16.6 %          |
|                                      | 4 %-93.4 %      | 14.5 % - 93.4 % | 24.4 % - 54 %   | 4 % - 58.7 %            |
| $a_{\rm d,pico}/a_{\rm d}$           | 33.1 % ± 10.9 % | 26.6 %± 9.7 %   | 38 %± 9.6 %     | 35.7 %± 8.5 %           |
|                                      | 9.4 %-55.9 %    | 9.4 % - 47.5 %  | 22.7 % - 55.9 % | 25.1 % - 47 %           |
|                                      | 8.8 % ± 11.5 %  | 9.7 %± 7.4 %    | 12.1 %± 15 %    | 10.9 %± 11.4 %          |
|                                      | 0-41.7 %        | 0-21.5 %        | 0-41.7 %        | 0-26.6 %                |
| $a_{\rm ph,nano}/a_{\rm ph}$         | 18.2.% ± 18.3 % | 27.4 %± 20.9 %  | 21 %± 14.6 %    | 15.6 %± 13.6 %          |
|                                      | 0-80.3 %        | 0-80.3 %        | 2.7 % - 48.2 %  | 4.4 % - 45.5 %          |
| $a_{\rm ph,ultra}/a_{\rm ph}$        | 26.6 % ± 18 %   | 37.6 %± 14%     | 29.4 %± 12.8 %  | 30.8 %± 13.2 %          |
|                                      | 0-63.7 %        | 3.8 % - 63.7 %  | 11.1 % - 60.4 % | 0-41.1 %                |
| $a_{\rm ph,pico}/a_{\rm ph}$         | 27.2 % ± 20.5 % | 25.3 %± 15.1 %  | 37.5 %± 17.9 %  | 42.7 %± 12.4 %          |
|                                      | 0-62.9 %        | 6.8 % - 57.9 %  | 7.7 % - 62.9 %  | 24.6 % - 59.6%          |
|                                      |                 |                 |                 |                         |

#### **New Figure 8**



# Figure 8: Contribution of particles from size classes in the total light absorption by all particles, detritus and phytoplankton according division into 4 size classes according to Ciotti et al. (2002) (micro-, nano-, ultra- and pico-particles) in GG, SF and OCW in for seasons.

For all data it can be seen that in the total light absorption by particles, detritus or phytoplankton, particles with sizes < 5  $\mu$ m - i.e. pico-particles and ultra-particles, had the largest contributions (on average about 38% and 31%). Particles with a size of 5-20  $\mu$ m accounted for approximately 20% of all particles and phytoplankton, and 29% of the detritus. The rest was due to the contribution of large particles, about 5-10% on average. A significant contribution of micro particles to the absorption is observed only for the summer phytoplankton bloom (SF13), and for SF03 and SF04 (end of winter and spring) and autumn bloom at station R7. Graphically, the contribution of size classes to total absorption is

illustrated in Figure 8 with division into sampling areas: the Gulf of Gdańsk and a separate set of data from Sopot Pier, as well as open and coastal waters outside the GG. In addition, the data were compiled according to the sampling season. The average contribution in  $a_p(443)$ from GG of micro particles was 6.6%, nano particles: 24.8%, ultra particles: 42.3% and pico particles: 26.3%. The average proportion of  $a_p(443)$  from Sopot Pier was respectively for micro: 9.5%, for nano: 20.5%, for ultra: 32.8% and pico: 37.2%. For OCW waters, the average contributions in  $a_p(443)$  of particles from the micro, nano, ultra and pico size classes were: 8.7%, 16.9%, 34.2% and 40.1%, respectively. In the case of  $a_d$ (443) for micro particles, the smallest contribution was recorded in GG (average 1.7%), while in Sopot Pier and OCW it was 6.3% and 7.4%, respectively. The proportion of nano particles was higher on average 30% in all regions. In  $a_d$ (443), ultra particles dominated in each area of the study: in GG an average of 53%, in Sopot Pier 39% and OCW 41%, and pico particles: in Sopot Pier 38% and OCW 35.7%, and in GG 26.6%. There is greater regional variation in uptake by phytoplankton. The smallest contribution of micro particles was observed in GG 9.7%, and in OCW 11%, in Sopot Pier 12%. Nano particles had the greatest contribution in  $a_{ph}(443)$  in GG (27.4%) and the smallest in OCW (15.6%). Ultra particles also had the highest contribution in  $a_{ph}(443)$  in GG (37.6%) and about 30% in Sopot Pier and OCW. In turn, the largest contribution of pico particles was observed in OCW (42.7%) and the smallest in GG (25.3%).

As suggested by the Reviewer, we have divided the Results and Discussion sections into two separate sections. In its current form, the Discussion section is as follows.

#### **4 Discussion**

We have carried out a number of analyzes of the variability of the mass of suspended particles, Chl*a* concentrations and the absorption properties of suspended particles for various size classes in the waters of the southern part of the Baltic Sea. So far, such analyzes have not been carried out empirically. No similar studies have been reported in the available literature so far. Fractionation based on the size of the particles suspended in the water column was carried out for completely different particle size ranges (<50  $\mu$ m and >50  $\mu$ m) and was most often associated with geotrace studies, in which the chemical composition of the suspension was determined (e.g., Lam et al. , 2015, 2018; Xiang and Lam, 2020; Yigiterhan et al., 2020). However, in the work of Mohammadpour et al. (2017) optical properties of SPM size fractions in littoral waters of Quebec were studied, but for size classes 0.2-0.4  $\mu$ m, 0.4-0.7  $\mu$ m, 0.7-10  $\mu$ m and > 10  $\mu$ m. The contribution of particles with sizes > 10  $\mu$ m did not exceed 17 %.

In our analyses, we showed the seasonal variability of Chl*a* contributions in size classes for three separate regions of the Baltic Sea: Gulf of Gdańsk, Sopot Pier, and open and coastal waters. This division results from the relationship between the optical properties of waters and their location in relation to river mouths, distance from the shore, or bottom bathymetry. This relationship is conditioned by the changing content of optically active water components and water circulation (Sagan, 2008). Water in the Gulf of Gdańsk was strongly influenced by river waters, as evidenced by the average salinity of 6.4 (0.4 - 7.7). At Sopot Pier, the average salinity was 7.6 (7 - 8.3), and at OCW 7.6 (7.2 -8.0). Water in the Gulf of Gdańsk was strongly influenced by river waters, as evidenced by the average salinity of 6.4 (0.4 - 7.7). At Sopot Pier, the average salinity was 7.6 (7 - 8.3), and at OCW 7.6 (7.2 -8.0). The proportions of Chla obtained by us in the micro, nano, ultra and pico size classes in these regions (without division into seasons) were: GG - 13%, 27%, 37% and 34%; in SF - 18%, 15%, 33% and 34%, and in OCW - 18%, 9%, 73% (for ultra+pico, where the fraction could not be separated). The proportions of  $Chl \alpha$  in various size classes of phytoplankton were reported in the literature. However, in most cases, this type of research concerned primary production. For the Baltic Sea, such studies have not been carried out so far. Reported by Maranon et al. (2001) contributions of Chla in fractions up to the total concentration of Chla (integrated for the water column 0-200 m) in the waters of the Atlantic Ocean showed that in most data the contribution of picoplankton was 60-80 % (average 61 %), nanoplankton (including ultraplankton) was approximately 20 % to 30 % (average 29 %) (from low-production regions to temperate regions), while the contribution of microplankton varied from < 10 % to 20 % (the share was low in most cases and increased to 20 % in upwelling areas and moderate, especially in spring) (average 9 %). Deng et al. (2022) presented results for data from the southern part of the China Sea from the surface layer, where the average contribution of picoplankton was 81 %, nanoplankton (including ultraplankton) 12 % and microplankton 7 %. In turn, Decembrini et al. (2014) for the Magellan Strait investigated the seasonal variability of the contributions of fractions in total Chla integrated for a depth of 0-50 m. For the spring season, the contribution of picoplankton in total Chla was on average 12 %, and the micro+nano fraction was 88 %. In the summer season, the contribution of picoplankton was on average 60 %, micro+nano 40 %. In late summer and early autumn, the average contribution of picoplankton was 47 % and micro+nano 53 %. Saggiomo et al. (1994) for austral summer (February-March 1991), in the Strait of Magellan, reported that the most important characteristic identified for this area was the confinement of the microplankton fraction to the external parts of the Strait and the rather uniform dimensional structure of the phytoplankton communities (< 5  $\mu$ m) within the internal sectors. In particular, the nanoplankton fraction  $(2-10 \,\mu\text{m})$  comprised 33 %, while the picoplankton one (0.5-2  $\mu$ m) represented 62 % of the total. Cermeno et al. (2005) studied the photosynthetic efficiency of fractionated phytoplankton in the Ria de Vigo (Iberian Penisula). The contribution of fractions with sizes  $< 5 \,\mu$ m was on average 46 %, fractions with sizes 5-20  $\mu$ m and > 20  $\mu$ m were 27 % each, in the total Chl*a* integrated in the euphotic zone (0-20 m). The data presented in this paper do not differ from those reported in the literature. By comparison of the above data, it can be seen that the percentage contribution of individual size fractions to the total concentration of Chla is not a constant value, but changes over time and space.

The overall proportion of organic matter in the total POM/SPM suspension for the analyzed dataset averaged 58% for total particles (see Figure 3). In the works of Woźniak and Meler (2020) and Woźniak et al. (2022) on the study of the optical properties of the waters of the Baltic Sea (conducted in the period 2017-2020), the presented POM/SPM ratios were characterized by similar values. The average POM/SPM was 61% and 62%, respectively. In previous studies, the average POM/SPM for the Baltic Sea was 80% (Woźniak et al., 2011 - for the period 2006-2009) and 76% (Meler et al., 2016 a, b - for the period 2006-2012). Long-term variability from previous years indicated a higher proportion of organic particles in the total suspension, while data from 2017-2021 may indicate that the proportion of detrital and mineral particles in the particle composition increased. However, this may be due to the increased number of stations located in the coastal waters zone in the total database. This, in turn, is caused by the variability of the weather (change in climatic conditions). In recent years, optical cruises, which have been taking place more or less at similar times of the year for 20 years, are characterized by more windy weather, which results in sailing closer to the coastal zone (there is less data from open waters), and therefore there is a greater proportion of particles flowing from land and/or lifted from the bottom, as a result of stormy weather. It is also possible that during the period of our research, the influence of river waters from the Vistula was greater than in the case of previous research. Research by Grelowski and Wojewódzki (1996) shows that the flow of the Vistula waters feeding into the GG carry sedimentary material extends horizontally from 2 to 15 nautical miles from river mouth, and has a vertical range of 0.5-12 m, depending mainly on the wind speed and direction, as well as a combination of factors such as the river water discharge rate, sea level and the duration of their interactions (Matciak and Nowacki, 1995).

The composition of suspension particles and organic solutes has a significant impact on their absorption properties. The absorption budget presented by us for the selected wavelength of 443 nm indicates that phytoplankton absorbed on average 29% of light, 19% was due to detritus, and 52% was due to CDOM (Figure 4h). Similar analyzes were carried out by Woźniak et al. (2011) and reported for the wavelength of 440 nm the average contribution were:  $(a_{ph}(440) + a_d(440)) - 45\%$  and  $a_{CDOM}(440) - 55\%$ . For the same wavelength, Figures 5-7 show how the absorption properties of all particles (including detritus and phytoplankton) changed with the change of Chla and SPM, i.e. the basic characteristics of the suspension. The original relationships (unfiltered samples) were slightly different in Woźniak et al. (2011). There, for the wavelength of 440 nm, the coefficient of determination for the dependence  $a_{p,all}(440)$  vs Chla was 0.73, and for the dependence  $a_{p}(440)$  vs SPM - 0.53. In the case of the coefficient  $a_{d,all}$  (443) for the Baltic Sea, it has been already shown that the dependence on SPM is better than the dependence on Chla (Woźniak et al., 2011; Meler et al., 2017). Comparing obtained by us relationships for  $a_{ph,all}(443)$  with those obtained by Woźniak et al. (2022), shown for a wavelength of 440 nm, it can be seen that also in this case the  $a_{ph}$  vs SPM relationships have higher determination coefficients  $R^2$  than the  $a_{ph}$  vs Chla relationships, 0.86 and 0.82, respectively.

The obtained average spectra for the dominant size class do not clearly explain the shape of the absorption spectrum, regardless of the division into 3 or 4 size classes (Figure 9-10). According to Ciotti et al. (2002), average chlorophyll-specific light absorption coefficients have the lowest values for microplankton, then nanoplankton, and ultraplankton, and the highest values for the specific light absorption coefficient for picoplankton. These average spectra were determined by size fractionating for the coastal waters of Oregon, the shelf waters of the Bering Sea, and the Bedford Basin (Nova Scotia, Canada). Such an arrangement of average specific absorption coefficients is related to the packing effect of pigments, where in larger particles there may be shading, and therefore less light absorption. In this work, for the Baltic data, the average chlorophyll-specific absorption coefficients are the lowest for nano-, then ultra- and picoplankton, and the highest for microplankton. Average values of  $a_{\rm ph}^{\rm (Chla)}(443)$  for micro, nano, ultra and pico-size classes by Ciotti et al. (2002) were approximately 0.012 m<sup>2</sup> mg<sup>-1</sup>, 0.03 m<sup>2</sup> mg<sup>-1</sup>, 0.042 m<sup>2</sup> mg<sup>-1</sup> and 0.068 m<sup>2</sup> mg<sup>-1</sup>, respectively. For the data analyzed in this article, the values of  $a_{ph}^{(Chla)}(443)$  for the micro, nano, ultra and pico-size classes are characterized by higher values, respectively 0.122 m<sup>2</sup> mg<sup>-1</sup>, 0.057 m<sup>2</sup> mg<sup>-1</sup> <sup>1</sup>, 0.082 m<sup>2</sup> mg<sup>-1</sup> and 0.112 m<sup>2</sup> mg<sup>-1</sup>.

The average determined for the micro-size class is probably overestimated and this applies to all absorption coefficients  $a_p$ ,  $a_d$ , and  $a_{ph}$ . This is due to the fact that the Chla\_fraction/Chla or SPM\_fraction/SPM predominance condition was met only in 2 cases, the absorption spectra of which differed significantly from each other. Both cases relate to samples collected at the pier in Sopot, one at the end of March (SF04 - Secchi disk was 5.5 m, the bottom was 6 m) and the other at the end of August (SF13 - Secchi disk was 1.5 m, the

bottom was at 6 m, there was a large wave, so there was probably mixing with the bottom particles as well). For the SF04 sample, the concentrations of Chl*a* and SPM were 0.78 mg m<sup>-3</sup> and 1.99 g m<sup>-3</sup>, respectively, for the original samples, and the light absorption coefficients of the particles were low. For the micro size class Chl*a* and SPM were 0.41 mg m<sup>-3</sup> and 0.92 g m<sup>-3</sup>, respectively. However, for the SF13 sample, Chl*a* and SPM were 4.05 mg m<sup>-3</sup> and 15.24 g m<sup>-3</sup>, respectively, for the original samples, and the light absorption coefficients of the particles were an order of magnitude higher than for SF04. For the micro size class, the Chl*a* and SPM concentrations were 1.87 mg m<sup>-3</sup> and 8.97 g m<sup>-3</sup>, respectively.

As mentioned above, Mohammadpour et al. (2017) studied the optical properties of the SPM size fractions in the littoral waters of Quebec for the size classes 0.2–0.4 µm, 0.4–0.7 µm, 0.7-10 µm and >10 µm. Among other things, mass-specific coefficients of light absorption by various size fractions of suspensions for different regions of the studied water body were presented. Due to the difference in the determination of size fractions, we can only conclude that the tested waters within Quebec are dominated by mineral suspensions, while the particles from the Baltic Sea waters analyzed in this work are dominated by organic suspensions. Mohammadpour et al. (2017) report spatial differentiation of the contribution of different size fractions of the suspension, which results in a large differentiation of light absorption coefficients. For the 0.2-0.7 µm size classes, this variability ranges from close to zero to > 0.4 mg<sup>2</sup> g<sup>-1</sup> at 400 nm wavelength. Similar differentiation was observed for the fraction >10 µm. Mohammadpour et al. (2017) did not report averaged specific mass factors  $a_p^{(SPM)}(\lambda)$ . However, their variability is similar to that presented in the Baltic Sea dataset, except that our spectra are characterized by clear chlorophyll maxima in the green (400-460 nm) and red (660-690 nm) bands.

#### 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.

# <u>Reviewer's comments:</u> *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  $\mu$ 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 Chl*a* 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.