Characterization of the organic vs. inorganic fraction of suspended particulate matter in coastal waters based on ocean color radiometry remote sensing

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Abstract. Knowledge of the organic and inorganic particulate fractions of suspended material in coastal waters is essential for the study of particle dynamics and biogeochemical cycles in these complex and highly variable environments. Thanks to the availability of appropriate spatial sensors, and to the considerable improvements of algorithms dedicated to the satellite observation of coastal waters from ocean color radiometry (OCR) achieved these two last decades, various optical and biogeochemical parameters can now routinely be monitored over coastal waters. Here we show that a Proxy of Particulate Composition (PPC) can be estimated from OCR observations. The present algorithm, based on Neural Network approach, has been validated using a broad range of biogeochemical data collected in various contrasted coastal waters, and applied to MERIS observations over the global coastal ocean at a 1km×1km spatial resolution from 2002 to 2012. The relevance of the temporal occurrence of PPC at a given water pixel has been illustrated over the global coastal ocean, and its pertinence has been deeply discussed over the English Channel and southern North Sea which are characterized by a well-documented variability of suspended particulate matter composition. The present algorithm can directly be applied to all OCR sensors.

1 Introduction

Water quality parameters display large spatio-temporal variability in coastal waters as these areas are the location of strong coupling between aquatic and terrestrial systems and are under the pressure of large natural and anthropogenic forcing. The characterization of the spatio-temporal distribution of biological, biogeochemical, and physical parameters in coastal waters is of a fundamental importance for a variety of applications dedicated to coastal management, which often contains economic interests, and to improve our understanding of the dynamics of the coastal ecosystems and their associated biogeochemical cycles. Among the large set of water quality parameters sampled in coastal waters, the concentration of the suspended particulate

matter (SPM), or equivalent parameters (i.e. the turbidity), has been intensively studied by the scientific community as a key parameter to understand the sediment transport, downstream sedimentation, and coastal geo-morphological processes (Velegrakis et al., 1997; Lahet and Stramski, 2010; Vantrepotte et al., 2012; Loisel et al., 2014; Anthony et al., 2015; Marchesiello et al., 2019). For instance, combining SPM spatio-temporal products with waves data allows resuspension areas to be identified (Loisel et al., 2014). While the SPM spatio-temporal patterns provide relevant information on the suspended particulate dynamics, the variation of the composition (i.e. chemical nature) of SPM may disclose relevant information on the complex chemical, physical, and biological processes occurring in coastal waters. For instance, the portion of particulate organic matter (POM) of SPM is of particular interest when investigating absorption of trace elements on particles, abrupt changes of water quality due to pelagic or benthic blooms, or fate of suspended matter between the water column and the sediments. Modeling the particulate transport requires specific modules coupled with hydrodynamical models. For example, in the SUBSTANCE module (Mengual et al., 2017), which can be coupled with CROCO, a French code built upon the well-known Regional Oceanic Modeling System (Shchepetkin and McWilliams, 2005), conservative and non-conservative (i.e. biological) particulate substances has to be defined with their own density and/or settling velocity, depending on the SPM nature (mineral vs. organic). The settling and flocculation processes, tightly linked to the organic fraction of SPM, to the transport of particulate variables in water column are also considered in these modules. The particulate organic carbon (POC), encompasses living (phytoplankton, heterotrophic bacteria, and virus) and non-living (i.e. detritus) organic particles in suspension. Information on the SPM and POC variability represents crucial inputs for the initiation and validation of sediment transport models (Douillet et al., 2001; Ford and Fox, 2014; Wu et al., 2020) and biogeochemical models (Aumont et al., 2015). While the analysis of the variation of the absolute values of SPM and POC concentrations bring relevant information for our understanding of carbon cycle and marine particulate dynamics in coastal waters, the dimensionless POC/SPM ratio is used in many studies to describe the seasonal variability in the particulate matter pool composition and origin (Coynel et al., 2005; Emmerton et al., 2008; Doxaran et al., 2012a, 2015; Ehn et al., 2019), which is often relate to variation in the regional environmental forcing (e.g. water discharge, phytoplankton bloom dynamics), but also to better interpret the in situ optical measurements (Babin et al., 2003a; Loisel et al., 2007; Woźniak et al., 2011; Doxaran et al., 2012b; Neukermans et al., 2016; Reynolds et al., 2016; Reynolds and Stramski, 2019) and satellite ocean color radiometry (OCR) observations (Vantrepotte et al., 2011). From in situ data collected in the near-shore marine environment at Imperial Beach in California, Woźniak et al. (2010) set threshold values of the POC/SPM ratio to identify changes in the particulate assemblage from the dominance of mineral particles (POC/SPM<0.06) to the dominance of organic particles (POC/SPM >0.25), and mixed particulate assemblages (0.25< POC/SPM <0.6). Neukermans et al. (2016) and Reynolds et al. (2016) followed the approach of Woźniak et al. (2010) to partition their Arctic seawater datasets into these three broad compositional classes.

Due to the high variability of the physical and biogeochemical processes occurring in coastal waters, traditional approaches as oceanographic cruises and in situ time series, although essential, are very time-consuming, expensive and sometimes uncertain to yield meaningful results on the studied phenomena. Satellite observation of OCR is now well recognized as a powerful tool to monitor the spatio-temporal distribution of biogeochemical and optical parameters in coastal waters (IOCCG, 2000; Loisel et al., 2013; IOCCG, 2018; Groom et al., 2019). Over the past decades, various OCR bio-optical algorithms have shown

that SPM (Ahn et al., 2006; Nechad et al., 2010; Feng et al., 2014; Han et al., 2016; Balasubramanian et al., 2020; Pahlevan et al., 2022) and POC concentrations (Liu et al., 2015; Hu et al., 2016; Woźniak et al., 2016; Le et al., 2017; Tran et al., 2019) can be estimated over coastal waters.

In this paper we will show how a proxy of particulate composition (PPC), classifying SPM into its organic, mineral, or mix fractions, can be estimated from OCR space observations. For that purpose, we will re-examine the relevance of the POC/SPM threshold values of Woźniak et al. (2010) using an extensive in situ data set covering a wider range of optical and biogeochemical variability than the previous dataset used by Woźniak et al. (2010). The relevance of the new threshold values will be discussed as a function of the particulate backscattering to attenuation ratio in the red, an optical proxy of the chemical nature (i.e., refractive index) of the bulk particulate matter (Twardowski et al., 2001; Loisel et al., 2007). Different algorithms (empirical and Neural Network) will be developed and validated against a large in situ data set collected in various coastal environments to assess PPC from OCR. The selected algorithm will then be applied to the MERIS observations (2002-2012) over the global coastal waters at 1 km^2 of spatial resolution to discuss the significance of this new product. A specific focus will be done on the English Channel and southern North Sea for which the temporal occurrence of the organic vs. inorganic fraction at a given pixel, built from 10-year temporal series, will be discussed.

2 Materials and methods

2.1 Dataset description

2.1.1 In situ datasets

In situ measurements were collected between 1998 and 2014 in southeastern Beaufort Sea (Bélanger et al., 2008), French Guiana (Vantrepotte et al., 2012, 2015), European coastal waters (Baltic Sea, English Channel, North Sea, Bay of Biscay) (Babin et al., 2003a, b; Lubac and Loisel, 2007; Lubac et al., 2008; Neukermans et al., 2012; Bonato et al., 2016; Novoa et al., 2017), and Vietnam East Sea (Loisel et al., 2014, 2017). The sampling strategies, the field measurement protocols and the data processing are described in related papers. Measurements include concentrations of POC and SPM (μg l^{-1}), remote-sensing reflectances (R_{rs}, sr^{-1}), particulate backscattering coefficient (b_{bp}), and particulate attenuation coefficient (c_p) (m^{-1}) at 650 nm. The samples cover a wide range of biogeochemical variability as POC and SPM concentrations span 4 and 3 orders of magnitude, respectively. The first in situ database, named DS0, includes 300 coincident POC, SPM, b_{bp}, and c_p measurements. In section 2.3, DS0 will be used to examine the empirical relationship between b_{bp}/c_p and POC/SPM. The POC/SPM ratio ranges between 1.1×10^{-3} and 8.7×10^{-1}. In comparison, Woźniak et al. (2010) collected 44 samples in Imperial Beach near-shore (California) with POC/SPM values between 2.3×10^{-2} and 4.2×10^{-1}. The second in situ database, referenced as DS, is made of 325 coincident POC, SPM, R_{rs} measurements with $3.9 \times 10^{-4} \leq$ POC/SPM $\leq 5.6 \times 10^{-1}$. It will be used to test the performance of POC/SPM estimates from the neural network algorithm (section 3.1).

2.1.2 Matchup dataset

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In situ data of POC and SPM, collected in the framework of the French Coastal Monitoring Network SOMLIT (Service d'Observation en Milieu Littoral), were used for the extraction of the collocated satellite R_{rs}. The SOMLIT network gathers data along the French coastline (https://www.somlit.fr). MERIS level 1 data ($\sim$ 1 km pixel resolution) over the 2002-2012 period were processed using the polymer atmospheric correction algorithm (Steinmetz et al., 2011; Steinmetz and Ramon,

90 2018), which was adapted for coastal waters in the frame of the GlobCoast project. Only pixels presenting a distance to the coast lower than 200 km and with a bottom depth not deeper than 4000 m are selected (Loisel et al., 2017). The criteria considered for the matchup selection are described in (Bailey and Werdell, 2006). After the application of these criteria, the final matchup dataset, named DSM, is composed of 91 matched points. The DSM dataset is characterized by lower POC/SPM values than DS0 and DS. The POC/SPM mean is of 0.0782 for DSM instead of 0.1136 and 0.1043, for DS0 and DS, respectively.

2.2 Algorithm development

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Two different approaches have been tested to assess PPC. The algorithms developed by Tran et al. (2019) (referenced as TR19) and Han et al. (2016) (HA16) are used to retrieve the POC and SPM concentrations, respectively, from R_{rs}. Once POC and SPM are derived, the POC/SPM ratio is calculated and PPC estimated using the different threshold values (see section 2.3). However, Tran et al. (2019) showed that POC concentration can be overestimated in presence of mineral waters. It results that

100 the POC/SPM ratio is also overestimated when mineral-dominated waters dominate (not shown). Moreover, the errors on both POC and SPM estimations are additive when the POC/SPM ratio is finally calculated. To limit this error propagation, we focus on the development of a single algorithm to derive in one step the POC/SPM ratio from R_{rs}. For that purpose, a neural network approach has been selected.

2.3 Determination of the organic, mineral, and mineral-organic mixed fractions

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As previously explained, the POC/SPM ratio is an indicator of the particle assemblage and is used to partition in situ data into three water types as defined by Woźniak et al. (2010). In addition, some studies showed that the variability of the ratio b_{bp}/b_p can be related to the particle composition (Boss et al., 2004; Twardowski et al., 2001; Loisel et al., 2007; Duforêt-Gaurier et al., 2018). Low b_{bp}/b_p values corresponds to high POC/SPM and are observed for a particle population dominated by low refractive index material such as phytoplankton. In contrast, high b_{bp}/b_p, corresponding to small POC/SPM are generally observed in the

110 presence of relatively high concentration of inorganic particles. At 650 nm, c_p is dominated by the scattering and the optical ratio b_{bp}/c_p can be used instead of b_{bp}/b_p (Loisel et al., 2007).

The objective here is to re-examine the pertinence of the POC/SPM threshold values of Woźniak et al. (2010) on a larger in situ coastal dataset (DS0) through the examination of the POC/SPM vs. b_{bp}/c_p relationship. As expected, b_{bp}/c_p decreases when POC/SPM increases that is when we move from a mineral-dominated to an organic-dominated environment (Fig. 1). A rapid

115 decrease is observed for POC/SPM values smaller than the first threshold value of Woźniak et al. (2010) (POC/SPM<0.06) corresponding to b_{bp}/c_p values higher than 0.013. Based on our knowledge of the in-situ data points, this first threshold value
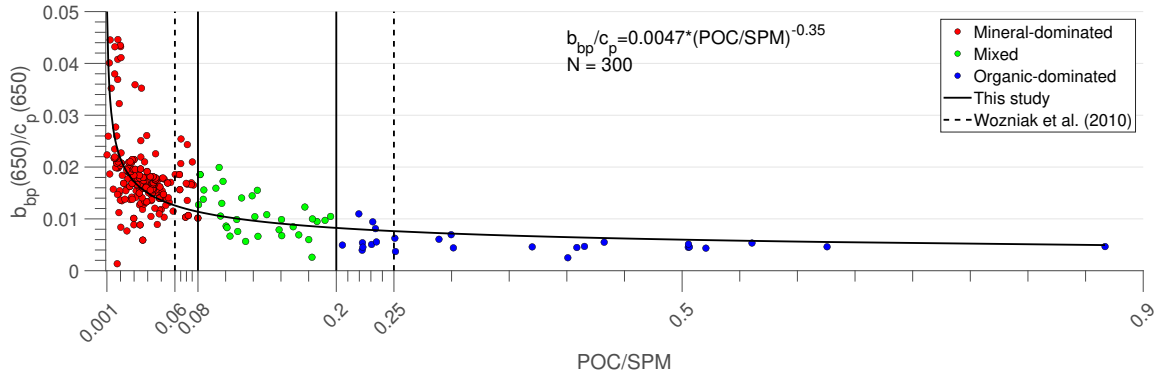
Figure 1. Relationships between b_{bp}/c_p (650 nm) and POC/SPM from DS0. The black curve stands for the power regression. The vertical dashed lines represent the thresholds delimiting the water types according to Woźniak et al. (2010). The vertical solid lines show the new thresholds defined in the present study.

has been shifted to 0.08 (corresponding to a b_{bp}/c_p value of 0.012) to encompass data points collected in mineral dominated environments (close to river mouths). An asymptote is reached at high POC/SPM values (above 0.2) and concerns data points with low b_{bp}/c_p (below 0.075) values typical to phytoplankton dominated environment. The new threshold value of POC/SPM

120 for organic-dominated waters, is set to 0.2 (i.e. $b_{bp}/c_p < 0.075$) to encompass in situ data points collected during bloom events. This value is however very similar to the value previously obtained by Woźniak et al. (2010) (= 0.25). Data points located along the asymptote are therefore associated with organic-dominated waters.

The thresholds are applied to monthly POC/SPM values derived from MERIS data, and the frequency of dominance is computed for each pixel of the scene over the ten years (2002-2012) as detailed below. A pixel geographically located at a

125 given latitude and longitude is named k pixel (k=1, S_{tot}). S_{tot} is the total number of pixels over the selected geographical area. For each k pixel, we computed the number of valid pixels (N_{tot}^k) over the period that corresponds to 120 months ($N_{tot}^k \leq 120$). The term "valid pixel" means a non-flag pixel for which the R_{rs} value is provided to derive POC/SPM with the neural network. Then, for the k pixel, we calculate how many time the class i is identified over N_{tot}^k: n_i^k is the class occurrence, i=1, 2 or 3 with 1 for mineral-dominated, 2 for organic-dominated, and 3 for mixed waters. The frequency of dominance is defined as the

130 ratio of the occurrence to the number of valid pixels:

$$D^k = \frac{maximum(n_1^k, n_2^k, n_3^k)}{N_{tot}^k} \times 100(\%) \tag{1}$$

The Equation 1 is repeated S_{tot} times to compute the frequency of dominance over the whole scene and provide maps in section 3.2.
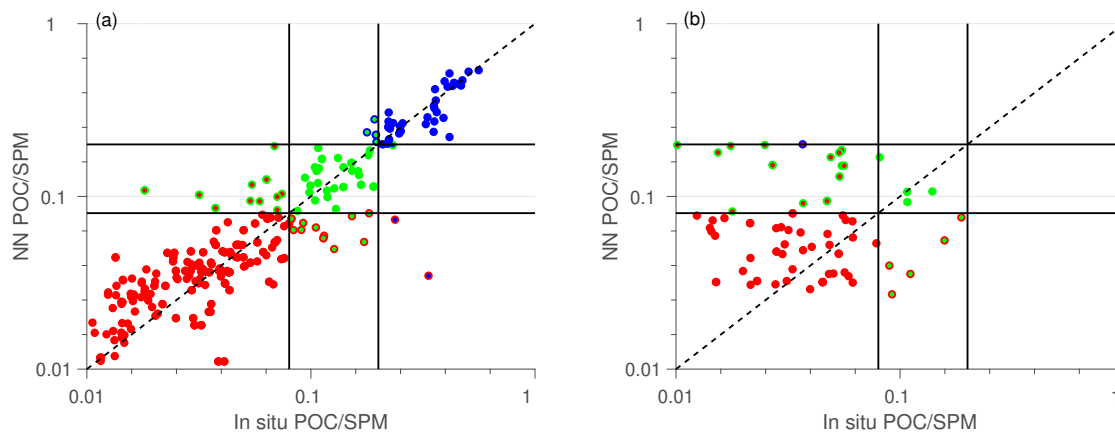
Figure 2. Comparison of in situ and derived POC/SPM from neural network algorithm (NN POC/SPM) (a) for DS (i.e., in situ R_{rs} are used to derive NN POC/SPM) (b) for DSM (i.e., satellite R_{rs} are used to derive NN POC/SPM). The dashed line is the 1:1 line. Red, blue and green color are for mineral-dominated, organic-dominated and mixed waters, respectively. For both panels, the vertical and horizontal solid lines indicate the thresholds (=0.08 and 0.2) used to partition the water types. Dots are color-labeled according to the values of the in situ POC/SPM, whereas the circles are color-labeled according to the values of NN POC/SPM. When circles and dots are of the same color that means that NN POC/SPM values conduct to the same classification as in situ POC/SPM.

3 Results

3.1 Validation of the classification

In situ measurements of R_{rs} from DS are used as input to the neural network algorithm to test the performance of the estimation of algorithm-derived values, named NN POC/SPM (Fig. 2a). The NN algorithm achieves a good performance over the whole range of POC/SPM. The median absolute percentage difference is 24% and the mean ratio MR is 1.06. The slope of the type II (log-transformed) regression is 0.90. The same classification is obtained for 88.5% of data points between the in situ and model derived values. About 10.9% of data point are misclassified in the adjacent group and only 0.62% are misclassified in a non-adjacent group. When NN POC/SPM is derived from satellite R_{rs}, data are much more scattered (Fig. 2b). It is because matchups accounts for inaccuracies related to the estimation of R_{rs} from space observations. Organic and mixed waters are underrepresented in DSM. Consequently, we do not discuss the performance of the classification for this class of waters. Regarding DSM, the use of the thresholds allows us to classify properly the mineral-dominated waters. Indeed, 62.75 % of data are well-classified and 35.29% are misclassified in the mixed-water group. It means as even if inaccuracies occurs on NN POC/SPM, it does not prevent to obtain a right PPC in most cases.

3.2 Global and regional PPC patterns
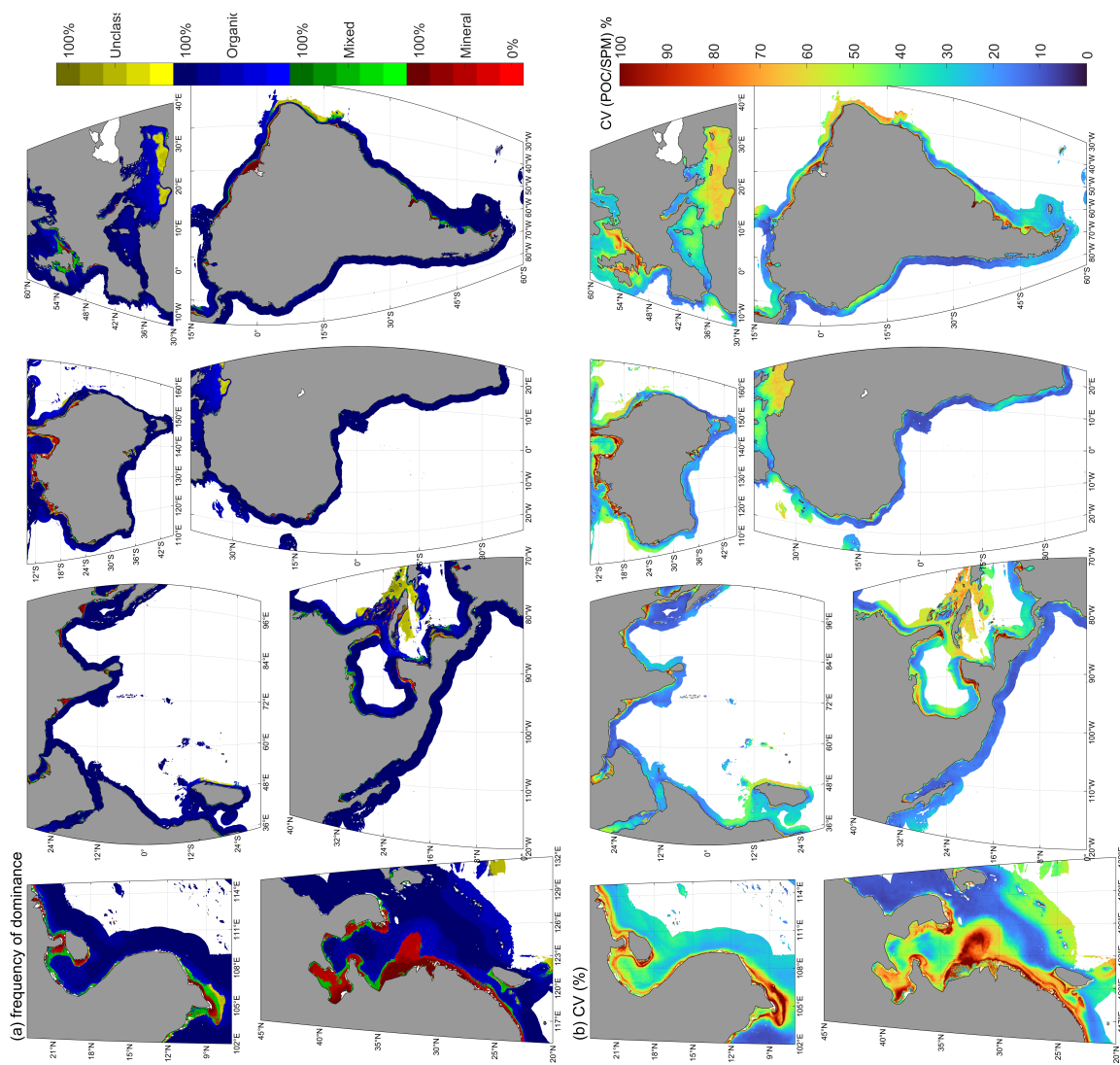
Figure 3. (a) Frequency (%) of dominant water-type (red for mineral-dominated/green for mixed/ blue for organic-dominated) over the MERIS time period 2002-2012. The yellow color corresponds to unclassified pixels that means pixels for which the retrieved value of POC/SPM is non-valid (i.e., negative) (b) Coefficient of variation of POC/SPM over the MERIS period 2002-2012

The PPC algorithm has been applied to MERIS monthly $R_{rs}(\lambda)$ data over the 2002-2012 time period, and the maximum
of occurrence of PPC has been calculated over this 10-years time period as described in section 2.3 (Fig. 3a). As expected,
150   we observe a well-marked near-shore off-shore gradient from mineral-dominated waters to organic-dominated waters with a
transition zone corresponding to mixed particulate assemblages. Large deltas and estuaries characterized by intense sediment
discharge such as the Amazon estuary, Mekong delta, Huanghe (Yellow River) delta, Yangtze estuary, or Ganges Brahmaputra
delta, present very high occurrence of mineral-dominated waters (70-90%). These areas can extend much further from the river
outlet, depending on the bathymetry and the surface currents, as for instance in the central part of the Yellow Sea in front of
155   the Yangtze estuary. Values of POC/SPM of 0.12-0.15, depicting mix-dominated particulate assemblages have been measured
in the Eastern China Sea region (124-126°E; 30-32°N) in great coherence with our present finding (Hung et al., 2007). The
dominance of mineral dominated waters is generally persistent all over the considered time period as depicted by the relatively
low (20-30%) coefficient of variation observed at the outlet of these different estuaries and deltas (Fig. 3b). In contrast, in front
of the Amazon and Yangtze estuaries and Huanghe delta some off-shore areas of high (70%) coefficient of variation depict the
160   impact of seasonal and inter-annual interactions between the continental (river and sediment discharges) and oceanic forcing
(tide, wind, surface current, sub-mesoscale structures) and the bathymetry. For example, tight interaction between the Amazon
plume dominated by mineral particles and the retroflection of the North Brazil Current carrying oceanic particulate organic
matter occur in region in front of the Amazon estuary where the slope in the continental shelf drastically changes (Gensac et al.,
2016; Varona et al., 2019), causing a significant temporal variation of the PPC. In contrast to the previously mentioned rivers,
165   and despite the Congo river is the world's second largest river in terms both of drainage area and water discharge, no PPC
signature can be noticed at the outlet of its estuary (Fig. 3a), except in the coefficient of variation (Fig. 3b). This is coherent
with the fact that the sediment discharge of the Congo river is very low, in contrast to its dissolved part, and that most of the
particulate assemble is organic (Coynel et al., 2005). A specific zoom on this area reveal the dominance of organic matter even
at the outlet of the estuary, in good agreement with in situ data reported in Coynel et al. (2005).

170   While an extensive investigation of the different spatio-temporal patterns presented in Fig. 3 are out of the scoop of the
present paper, a specific focus illustrating the pertinence of the PPC product is conducted over the eastern English Channel
(EEC) and southern North Sea (SNS) for which the biogeochemical and physical processes driving the suspended particulate
matter composition are relatively well-documented. The eastern English Channel and southern North Sea coastal region is
known for its strong hydrodynamics marked by intense tidal currents. This shallow water region, therefore largely impacted by
175   resuspension effects, is also largely influenced by freshwater discharges from different rivers, la Seine and la Somme being the
most important ones along the French coast of EEC, and Thames and Scheldt along the SNS. Strong spring and early summer
phytoplankton blooms of diatoms and Phaeocystis globosa occurred in this region at different time of the year, depending on
the nutrient and light (i.e. season and turbidity level) availability (Breton et al., 2000; Antajan et al., 2004; Sazhin et al., 2007).
For these different reasons, the composition of the suspended particulate matter in this area is strongly variable, as already
180   stressed from different field studies based on optical and biogeochemical measurements (Loisel et al., 2007; Vantrepotte et al.,
2007; Lubac et al., 2008). The monthly occurrence of each PPC class is in excellent agreement with our present knowledge
of this area (Fig. 4). Between mid-fall and winter (from October to February), the particulate matter pool of EEC and SNS
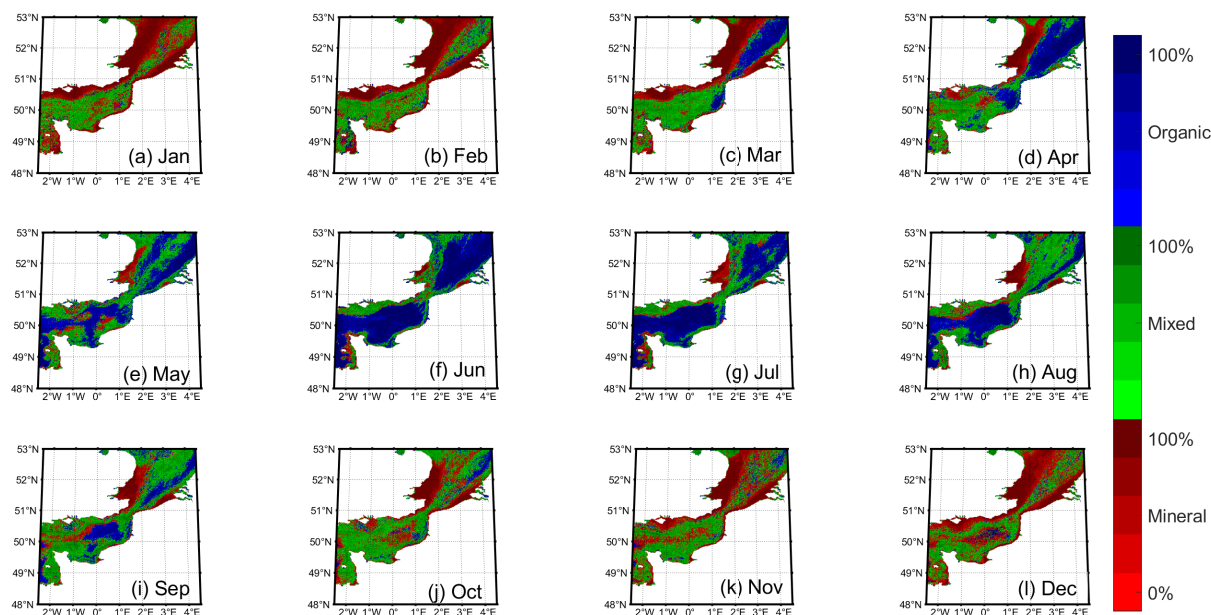
Figure 4. Frequency of dominance (%) for mineral-dominated (red), mixed (green) or organic-dominated waters (blue) over the MERIS period 2002-2012 in the eastern English Channel and southern North Sea.

is characterized by mineral and mixed-dominated assemblages mainly due to sediment river inputs and resuspension effects due to strong physical (i.e. waves) forcing (Lesourd et al., 2003; Lancelot et al., 2007; Gohin, 2011; Renosh et al., 2014). This
185 is consistent with results of Vantrepotte et al. (2012) who observed that relative contribution of particulate matter assemblage with a strong proportion of mineral particles increases significantly during the winter time period. In March, organic-dominated pixels appear in the SNS coastal region and in front of the Somme river area in the EEC. Then, while the organic dominated particles assemblages areas are developing in SNS from March to July, similar particulate assemblages start developing from May to July in off-shore waters of EEC, and along the EEC coast from June. The timing of the development of organic matter
190 areas in the EEC and SNS is in perfect agreement with phytoplankton blooms dynamics in these regions which first develop in the Bay of Somme area and SNS followed by another bloom which developed southern in the Seine river bay area (Brunet et al., 1996; Vantrepotte et al., 2007; Joint and Pomroy, 1993; Van der Zande et al., 2012; Gohin et al., 2019). Finally, areas covered by organic-dominated pixels are maximum in June/July, and then mix assemblages start developing in the northern part of the domain to fully cover the whole region in October. Mix assemblages are more likely dominated by aggregates of mineral
195 particles and detrital matter (non-living organic particles) which developed during the senescence of the bloom (Vantrepotte et al., 2007; Loisel et al., 2007; Gohin, 2011; Grattepanche et al., 2011).

4 Concluding remarks

The PPC can be used as a proper indicator for characterizing the nature of the bulk suspended particulate matter from satellite OCR remote sensing. The MERIS spatio-temporal patterns of this new indicator are consistent with general features docu-
200   mented in the literature. Due to visible bands used, common to all OCR sensors, the PPC algorithm can be applied to all OCR satellite sensors allowing to assess the long-term trend of suspended particulate matter over the global coastal ocean. The application of this algorithm to the satellite Copernicus Marine Environment Monitoring Service (CMEMS) data is underway to assess the different temporal patterns (seasonal, trend, and irregular) using the CENSUS X-11 approach (Vantrepotte et al., 2011).

205   *Author contributions.* H.L. as the major funder proposed the idea of the work, gave permission to data in LOG, was involved in the algorithm development and data analysis, and co-wrote the manuscript. L.D.-G. was involved in the algorithm development, supervised the analysis and visualization of the data, was involved in manuscript organization and co-writing. T.K.T. analyzed and visualized the in situ and satellite data, developed and tested the algorithms, developed the codes to processed images. D. S. F. J. supported the development of the Matlab processed images, provided the final plots and statistics and, revised the manuscript. F. S. provided Polymer algorithm to perform atmospheric
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