Partitioning total spectral absorption in phytoplankton and colored detrital material contributions

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Abstract

A method based on spectral information is used to derive spectral absorption coefficients of phytoplankton $a_\varphi(\lambda)$ and colored detrital material, CDM, which includes non-algal particle and colored dissolved organic matter, $a_{CDM}(\lambda)$, from total minus water absorption coefficients measurements. This method is first validated over a dataset of more than 300 simultaneous measurements of phytoplankton, non-algal particle, and colored dissolved organic matter absorption coefficients spectra acquired with a laboratory spectrophotometer in various oceanic and coastal European waters. The validation is presented for measurements made with a high spectral resolution (hyper-spectral case), and a limited spectral resolution (multi-spectral case) – the case of most devices routinely used for in situ profiling, such as the WETLabs ac-9. In order to examine the various sources of error in the method, we test its performance considering various levels of a priori knowledge of phytoplankton absorption properties over the study area: for each site, over each region, or over a global dataset only. When the method is applied to the multi-spectral case without introducing any "local" information on phytoplankton absorption properties, we obtain a good performance with a relative Root Mean Square Error equal to 17.8%, 14.6%, and 40.7% for $a_{CDM}(412)$, the CDM exponential slope, and $a_\varphi(440)$, respectively. Finally, the partitioning method is directly applied to in situ profiles of total minus water spectral absorption coefficient measured with an ac-9 in various oceanic Mediterranean waters, allowing the in situ description of CDM and phytoplankton absorption coefficients with a high spatial resolution.

Changes in the propagation of light in seawater are related directly to its content in dissolved and suspended materials. Photons either are scattered or absorbed by optically significant constituents. The total absorption coefficient, $a_t(\lambda)$ (λ is the wavelength) is the result of the additive contributions of pure water (w), phytoplankton (φ), colored dissolved organic material (CDOM), and non-algal particles (NAP; biogenous detritus, heterotrophs, and minerals):

$$a_t(\lambda) = a_w(\lambda) + a_\varphi(\lambda) + a_{CDM}(\lambda) \qquad \text{(Eq. 1a)}$$

in which

$$a_{CDM}(\lambda) = a_{CDOM}(\lambda) + a_{NAP}(\lambda) \qquad \text{(Eq. 1b)}$$

These constituents are characterized by different spectral absorption signatures and are present in the ocean in variable proportions. The separation of their relative contributions to the total absorption coefficient measured in situ is of main interest for bio-optical and remote sensing studies (e.g., $a_\varphi(\lambda)$ for primary productivity models or for the discrimination of phytoplanktonic groups, $a_{CDOM}(\lambda)$ for CDOM impact on light availability and the dissolved organic carbon cycle, e.g., Siegel et al. 2002 and references therein). Colored dissolved organic material and non-algal particles often have been described as a sole compartment (colored detrital material or CDM; e.g., Siegel and Michaels 1996; Siegel et al. 2002), and can be characterized by similar spectral dependencies, usually modeled according to an exponential decrease with increasing wavelength (Babin et al. 2003 and references therein):

$$a_{NAP/CDOM}(\lambda) = a_{NAP/CDOM}(\lambda_o)\, e^{(-S_{NAP/CDOM}(\lambda - \lambda_o))} \qquad \text{(Eq. 2a)}$$

or

$$a_{NAP/CDOM}(\lambda) = A e^{-S(\lambda - \lambda_o)} \qquad \text{(Eq. 2b)}$$

where A is the absorption coefficient at a reference wavelength (λ_o) and S is the slope of the exponential model. S_{CDOM} generally

is higher than S_{NAP} (for oceanic waters: 0.014 nm^{-1} for S_{CDOM} and 0.011 nm^{-1} for S_{NAP} on average, Bricaud et al. 1981, Davies-Colley 1992, Bricaud et al. 1998; and for coastal waters: ~0.018 nm^{-1} for S_{CDOM} and 0.012 nm^{-1} for S_{NAP}, Babin et al. 2003). Variations in S_{CDOM} have been attributed to changes in the material composition of terrestrial or oceanic origin and/or CDOM photo-oxidation under high light intensities in stratified conditions (e.g., Twardowski and Donaghay 2001). Variations in S_{NAP} also are likely the result of changes in material composition, although more studies are required on this topic. Phytoplankton absorption coefficients vary as a result of changes in pigment packaging effect due to changes in algal cell size and internal pigment concentration and, to a lesser extent, in pigment composition (Bricaud et al. 2004 and references therein). The contribution of the optically significant constituents to the total minus water absorption coefficient of seawater measured in situ can be separated by experimental and numerical methods ranging from seawater filtration for the determination of dissolved absorption coefficient (Prieur and Sathyendranath 1981; Twardowski and Donaghay 2001), to chemical bleaching of the material retained on the filters (Kishino et al. 1985; Ferrari and Tassan 1999) or the use of numerical methods (Morrow et al. 1989; Bricaud and Stramski 1990) to partition phytoplankton and non-algal particle absorption coefficient, to the use of empirical relationships to discriminate between phytoplankton and colored detrital matter absorption coefficients (Roesler et al. 1989; Claustre et al. 2000).

The recent widespread usage of in situ spectral absorption-attenuation meters (WETLabs ac-9 or more recently ac-s; Moore 1994; Zaneveld et al. 2004), particularly as part of various observing systems (moorings, autonomous underwater vehicles; Dickey 2003), enlarges the range of space and time scales at which absorption coefficients can be measured. This reinforces the need for alternative (new) methods that allow the retrieval of individual constituent contributions from bulk measurements. Chang and Dickey (1999) applied the method implemented by Roesler et al. (1989) to time series of total minus water spectral absorption coefficient measured with a high temporal resolution (ac-9 meters on moorings), and ultimately discriminated CDOM from non-algal particle contributions based on discrete CDOM samples taken on a regular basis. Claustre et al. (2000) used statistical relationships derived from discrete measurements performed in the same area to partition surface horizontal profiles of total spectral absorption coefficient into CDOM, algal, and non-algal contributions. Recently, Gallegos and Neale (2002) introduced a method relying on normalized absorption cross-sectional areas of the absorbing components and a matrix inversion method, and Schofield et al. (2004) implemented an optical inversion-optimization method to extract optical weights of various constituents in the bulk signal.

The objective of the new method we propose here is to derive phytoplankton and colored detrital material absorption coefficients from total minus water absorption coefficient measurements. This method is adapted from the Bricaud and Stramski (1990) numerical method developed for partitioning particulate absorption coefficients into algal and non-algal components. First, the numerical method we propose is tested on a large database (more than 300 determinations) of concurrent hyper-spectral measurements of CDOM, phytoplankton, and non-algal particle absorption coefficients acquired over a large range of water types from oceanic to coastal waters. Second, we modify it for application to multi-spectral measurements of total minus water absorption coefficient acquired with commercially available in situ profilers such as the WETLabs ac-9. Finally, this method is applied to ac-9 data for various conditions/areas in the Mediterranean Sea. The present method has the advantage of being at the same time simple and "spectral", i.e., phytoplankton and CDM absorption coefficients are estimated over the whole spectrum and can be applied easily to in situ ac-9 profiles.

Method description

The numerical method developed by Bricaud and Stramski (1990) relies on the information contained in the shape of the particulate absorption coefficient spectrum, and is recalled briefly here. In some spectral domains, the variability in the shape of the phytoplankton absorption coefficient spectrum is low whereas the variability in the corresponding particulate absorption coefficient is large. The difference, therefore, is attributed to the variability in non-algal particle absorption coefficient. Two pairs of wavelengths (505/380 and 580/692 or λ_2/λ_1 and λ_3/λ_4) have been identified for which absorption coefficient ratios are relatively stable in phytoplankton, because they are only weakly affected by the spectral variations due to pigment composition (i.e., minimal absorption by accessory pigments) and the package effect (ratios must be close to 1.0). The phytoplankton absorption ratios at these two pairs of wavelengths ($r\varphi_1$ and $r\varphi_2$) are determined from measurements on samples collected in situ, on which the phytoplankton absorption coefficient is determined using a chemical bleaching method, and usually is averaged over an area or a dataset. The particulate absorption ratios at the same two pairs of wavelengths, determined on a given sample, are then used as inputs in a system of two equations with two unknowns (Eq. 5 in Bricaud and Stramski 1990, given below in a more general expression) to extract S_{NAP} (or S) and $a_{NAP}(\lambda_o)$ (or A), the parameters required to describe $a_{NAP}(\lambda)$ (see Eq. 2):

$$r\varphi_1 A e^{(-\lambda_1 S)} - A e^{(-\lambda_2 S)} = r\varphi_1 a_p(\lambda_1) - a_p(\lambda_2) \qquad \text{Eq. 3a}$$

$$A e^{(-\lambda_3 S)} - r\varphi_2 A e^{(-\lambda_4 S)} = a_p(\lambda_3) - r\varphi_2 a_p(\lambda_4) \qquad \text{Eq. 3b}$$

Phytoplankton absorption is finally computed by difference between the particulate and non-algal particle fractions (Bricaud and Stramski 1990 provides more details).

Here, we investigate the possibility of using the Bricaud and Stramski (1990) method to retrieve, from total minus water absorption coefficient measurements ($a_t(\lambda) - a_w(\lambda)$), the respective contributions of phytoplankton ($a_\varphi(\lambda)$) and CDM

Table 1. List of campaigns, study areas, type of waters, measurements, devices used, and corresponding papers relevant for this study.

Campaign	Location	Type of waters	Measurements used in this study	Reference
Coastlooc	European waters and Atlantic Ocean	Mainly coastal (some oceanic) waters	– a_{nap} (laboratory spectrophotometer) – a_{φ} (laboratory spectrophotometer) – a_{CDOM} (laboratory spectrophotometer)	Babin et al. 2003
Prosope	Moroccan upwelling - Mediterranean Sea	Oceanic waters	– a_p (laboratory spectrophotometer) – a_{CDOM} (WETLabs filtered ac-9) – a_t (WETLabs ac-9)	Oubelkheir et al. 2005
Almofront II	Alboran sea (Frontal area)	Oceanic waters	– a_t (WETLabs ac-9)	Claustre et al. 2000

($a_{CDM}(\lambda)$). An "average" exponential slope (S_{CDM}), describing the spectral dependency of CDM absorption coefficient, is estimated following the same principle as above. This method will be tested on total minus water absorption coefficient measurements acquired with a high spectral resolution (hyperspectral case) – using the same pairs of wavelengths as Bricaud and Stramski (1990) – and with a limited spectral resolution (multi-spectral case), which corresponds to the case of most commercially available in situ absorption-meters, such as the WETLabs ac-9 (nine wavelengths in the visible range). In this last case, the choice of the two pairs of wavelengths is constrained by the available channels in standard ac-9 sensors (412, 440, 488, 510, 532, 555, 630 or 650, 676, and 715 nm), but must still satisfy, as closely as possible, the same conditions as those above for the Bricaud and Stramski (1990) method. Based on these criteria, the two new pairs of wavelengths chosen are 510/412 nm and 555/630 nm.

Experimental methods and observations

Total, dissolved, and particle (phytoplankton and non-algal) spectral absorption coefficients were determined in various oceanic and coastal waters around Europe and in the Mediterranean as part of several cruises (Table 1) using different experimental protocols. A short description of the regional variability in absorption properties for each study area also is included, so as to provide a bio-optical "context" for the method validation/application.
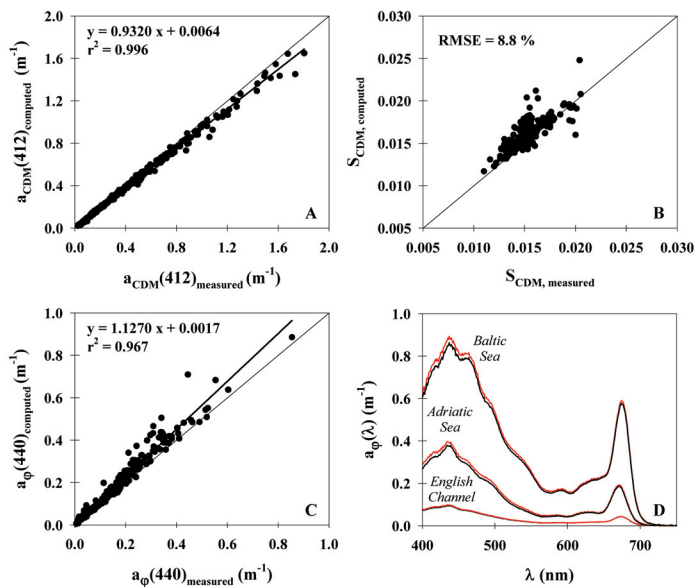
COASTLOOC campaigns—Various coastal European waters were investigated during the COASTLOOC survey: the Adriatic Sea, Baltic Sea, English Channel, Mediterranean Sea, and North Sea (*see* table 1 in Babin et al. 2003). Additionally, some open ocean waters in the North Atlantic were studied. Spectral absorption coefficients of the individual constituents were determined on water samples taken at the surface (Babin et al. 2003). Briefly, particle and CDOM absorption coefficients were measured in the 380-750 nm and 350-750 nm spectral range, respectively, with a 1-nm spectral resolution using Perkin Elmer dual beam spectrophotometers (equipped with an integrating sphere for particle absorption determinations). Non-algal particle absorption coefficients were deter-

mined after bleaching the material retained on the filter by sodium hypochlorite (Ferrari and Tassan 1999). Phytoplankton absorption coefficients were computed by difference between particle and non-algal particle absorption coefficients. For full details on the measurement protocols, see Babin et al. (2003).

Over the COASTLOOC dataset, the relative contributions of CDOM, phytoplankton, and non-algal particles to total minus water absorption coefficient are highly variable, thus providing a large range of conditions for a proper validation of the partitioning method presented here. As shown by Babin et al. (2003), for 73% of the samples, the relative contributions of each of these constituents at 440 nm vary between 40 and 80% for CDOM, 20 and 60% for phytoplankton, and 10 and 40% for non-algal particle. Average phytoplankton contribution to the absorption coefficient at 440 nm was maximal in the Adriatic Sea (52%) and minimal in the North Sea (28%) (Table 2). Average CDOM contribution to CDM at 440 nm was minimal in the Adriatic and Mediterranean Sea (59%) and maximal in the Atlantic Ocean (82%). Measured S_{NAP} and

Table 2. Average contribution of phytoplankton absorption to total minus water absorption coefficient and CDOM absorption to CDM absorption coefficient at 440 nm over the COASTLOOC dataset. The corresponding standard deviation is given in italic.

	a_{φ} to $a_t - a_w$(440)	a_{CDOM} to a_{CDM}(440)	*n* samples
Adriatic Sea	0.52	0.59	39
	0.10	*0.12*	
Atlantic Ocean	0.33	0.82	21
	0.14	*0.11*	
Baltic Sea	0.32	0.70	54
	0.07	*0.06*	
English Channel	0.42	0.68	72
	0.10	*0.15*	
Mediterranean Sea (Case 2)	0.35	0.59	43
	0.17	*0.17*	
North Sea	0.28	0.61	85
	0.10	*0.17*	

Fig. 1. Relationship between absorption parameters measured and computed from total minus water absorption coefficient measurements, for the "site by site" analysis in the hyper-spectral case (the partitioning method is applied to total minus water absorption data acquired with a full spectral resolution). A. Colored Detrital Material absorption coefficient at 412 nm ($a_{CDM}(412)$, in m^{-1}), B. exponential slope of CDM absorption coefficient (S_{CDM}, in nm^{-1}), C. phytoplankton absorption coefficient at 440 nm ($a_{\varphi}(440)$, in m^{-1}). D. Three examples of computed (red) and measured (black) spectra in the English Channel, Adriatic Sea, and Baltic Sea (Sta. C6024000, C3011000, and C6148000, respectively).

S_{CDOM} varied between 0.009–0.018 nm^{-1} and 0.011–0.025 nm^{-1}, respectively, probably as a result of changes in material composition, and photo-oxidation state for CDOM (Twardowski and Donaghay 2001) over the variety of water types and seasons explored.

For testing the present partitioning method, total spectral absorption coefficients (the water contribution is not included) were computed as the sum of measured CDOM, phytoplankton, and non-algal particle absorption coefficients; and CDM spectral absorption coefficients were computed as the sum of CDOM and non-algal particle absorption coefficients. The slope of measured CDM absorption spectrum (sum of CDOM and NAP; characterized by S_{CDM}) was estimated by fitting an exponential model to the data over the 350–500 nm spectral range, as for S_{CDOM}.

PROSOPE campaign—Various Mediterranean oceanic waters were investigated during late summer as part of the PROSOPE campaign (September 1999). Eleven stations were sampled: the MIO site in the Ionian Sea, the DYF site in the North-Western Mediterranean and stations 1–9 in various regions of the Mediterranean (for station locations see fig. 1 in Oubelkheir et al. 2005). Trophic conditions were contrasted, from the eutrophic waters of the Malaga Upwelling (Sta. 1) and the frontal area in the Alboran Sea (Sta. 2) to the oligotrophic/ultra-oligotrophic conditions encountered

over most of the Mediterranean. Vertical profiles of total and dissolved absorption coefficients were determined in situ at nine wavelengths with a WETLabs ac-9 (without or with a 0.2μm filter at the intake of the measuring tubes). Ac-9 data acquisition and analyses are detailed in Oubelkheir et al. (2005). Water samples also were collected for particle absorption coefficient measurements, $a_p(\lambda)$, following a protocol explained in more detail below, as it is not described elsewhere. After filtration of seawater through Whatman GF/F glass-fiber filters, $a_p(\lambda)$ was measured over the 370–750 nm spectral range with 2 nm increments, using a LI-1800UW LICOR radiometer equipped with an integrating sphere. The path length amplification effect by the filter was corrected by using the algorithm of Bricaud and Stramski (1990). Background correction was applied by subtracting the average value measured at 750 nm from all the measured spectral values. Phytoplankton and non-algal particle absorption coefficients were finally computed from particle absorption measurements using the numerical partitioning method of Bricaud and Stramski (1990).

Over the Mediterranean during the PROSOPE campaign, CDOM absorption at the maximum depth ranges between 0.06 and 0.10 m^{-1}, whereas the corresponding a_{NAP} values are around 0.005 m^{-1}; CDM is mainly CDOM (a_{NAP} <20% a_{CDM} at all sites). At the chlorophyll maximum depth, phytoplankton absorption at 440 nm ($a_{\varphi}(440)$) contributes from 40% (MIO, Ionian Sea) to 60% (Sta. 1, Malaga upwelling) of total minus water absorption coefficient. The partitioning method is applied here to total minus water absorption coefficient spectra measured along depth (0–400 m) with a high spatial resolution with the WETLabs ac-9.

ALMOFRONT II campaign—The Almeria-Oran frontal system, resulting from the abutting of less dense Atlantic waters against dense Mediterranean waters in the Western Mediterranean Sea, was investigated during winter (December 1997–January 1998) as part of the ALMOFRONT II campaign. The horizontal distribution of the total spectral absorption and attenuation coefficients was determined in the near-surface waters at high spatial resolution using an underway WETLabs ac-9 along a transect crossing the frontal area several times. A detailed description of the data acquisition and analysis is presented in Claustre et al. (2000), together with a complete bio-optical characterization of the near-surface waters in the Almeria-Oran frontal system.

Our numerical partitioning method is applied here to the underway ac-9 measurements of total minus water absorption coefficient. CDM absorption coefficients computed using the present method are compared with the values estimated at 412 nm over the same area and on the same dataset using a different method published by Claustre et al. (2000) and briefly recalled here. In the method presented by Claustre et al. (2000), $a_t(676) - a_w(676)$ is assumed to equal $a_{\varphi}(676)$, and a statistical relationship is used to estimate $a_{\varphi}(412)$ from $a_{\varphi}(676)$. Then, $a_{CDM}(412)$ is derived by subtracting $a_{\varphi}(412)$ from $a_t(412)$

– $a_w(412)$ (for more details refer to Claustre et al. 2000). In the near-surface waters over the frontal area, CDM computed using the method of Claustre et al. (2000) contributed for 55 to 75% of total minus water absorption coefficient at 412 nm (their fig. 6).

Results and discussion

First, the performance of the partitioning method is tested on the COASTLOOC dataset (simultaneous measurements of phytoplankton, CDOM, and non-algal particle absorption coefficients) by using the same pairs of wavelengths as in Bricaud and Stramski (1990) (hyper-spectral case, 505/380 nm and 580/692 nm) and by using new pairs of wavelengths corresponding to ac-9 measurements (multi-spectral case, 510/412 nm and 555/630 nm). In the hyper-spectral case, we investigate three different types of data analysis (site by site, on a regional scale, and on a global scale, *see* below), depending on the a priori knowledge of phytoplankton absorption properties. Second, the partitioning method is applied to multi-spectral absorption coefficient measurements made in situ with a high resolution on the vertical scale (PROSPOPE) or horizontal scale (ALMOFRONT II) using an ac-9.

Method validation: COASTLOOC Campaigns

Hyper-spectral measurements: In the hyper-spectral case, phytoplankton absorption ratios used as inputs of the model ($r\varphi_1$ and $r\varphi_2$, Eq. 3) are determined following three different ways in order to determine the various sources of errors in computed parameters, as described hereafter: (1) The "site by site" case: $r\varphi_1$ and $r\varphi_2$ are determined for each sample based on measured phytoplankton absorption spectra ("true" values), in order to test the first source of error originating from the computation of a unique set of parameters to describe CDOM and NAP absorption properties as a whole compartment (CDM), (2) The "region by region" case: $r\varphi_1$ and $r\varphi_2$ are averaged over each region explored during the COASTLOOC survey (the Adriatic Sea, the Atlantic Ocean, the Baltic Sea, the English Channel, the Mediterranean Sea, and the North Sea), in order to examine the second source of error introduced by using "regionally averaged" phytoplankton absorption ratios rather than "true" values, (3) The "global" case: $r\varphi_1$ and $r\varphi_2$ are averaged over the entire COASTLOOC dataset, to test the applicability of the method in a given area or at a given depth without any specific knowledge of the absorption properties of phytoplankton. $r\varphi_1$ and $r\varphi_2$ are given in Table 3a for cases 2 and 3. The performance of the method is quantified by the relative Root Mean Square Error (RMSE) between computed and measured absorption coefficients. The RMSE is calculated in relative value so as to give equal weights to all measurements and estimated as follows:

$$\text{RMSE (\%)} = 100 * \sqrt{\frac{\sum_{i=1}^{n}\left(\frac{x_{i\,computed} - x_{i\,measured}}{x_{i\,measured}}\right)^2}{n}} \qquad \text{Eq. 4}$$

Where x is the variable and n is the number of observations.

Table 3a. Average phytoplankton absorption coefficient ratios computed at two key pairs of wavelengths (*see* text) for the various datasets used in this study (in the top 100 meters of the water column), for the hyper-spectral case. The corresponding standard deviations are given in italic.

	505/ 380 nm	580/ 692 nm	*n* samples
Adriatic Sea	0.78	1.58	39
	0.20	*0.50*	
Atlantic Ocean	0.61	1.27	21
	0.08	*0.30*	
Baltic Sea	0.88	1.02	54
	0.29	*0.12*	
English Channel	0.56	1.18	72
	0.13	*0.19*	
Mediterranean Sea (Case 2)	0.49	1.24	43
	0.15	*0.42*	
North Sea	0.61	1.28	85
	0.21	*0.32*	
COASTLOOC Global dataset	0.64	1.25	314
	0.23	*0.34*	

"Site by site" case: Computed and measured CDM absorption coefficients at 412 nm are linked by a highly significant linear relationship ($r^2 = 0.996$, Table 4), with a slope close to 1.0 (0.93) (Fig. 1A, Table 4). Computed and measured phytoplankton absorption coefficients are in good agreement as shown at a given wavelength (Table 4, e.g., computed and measured values are linked by highly significant linear relationships with an r^2 equal to 0.967 and a slope of 1.13 at 440 nm, Fig. 1C) and spectrally (Fig. 1D, three example spectra are presented). The partitioning method estimates CDM and phytoplankton absorption from total minus water absorption coefficient measurements with a good accuracy: RMSE equal to 9.5%, 8.8%, and 24.6% for CDM absorption at 412 nm, S_{CDM}, and phytoplankton absorption at 440 nm, respectively (Fig. 1, Table 4), despite the fact that individual constituents are present in highly variable proportions over the COASTLOOC dataset and that CDOM and NAP can be characterized by highly different S values (*see* Experimental methods and observations).

In order to further understand the various sources of error, we performed a method sensitivity analysis under known/

Table 3b. Same as Table 3a, multi-spectral case.

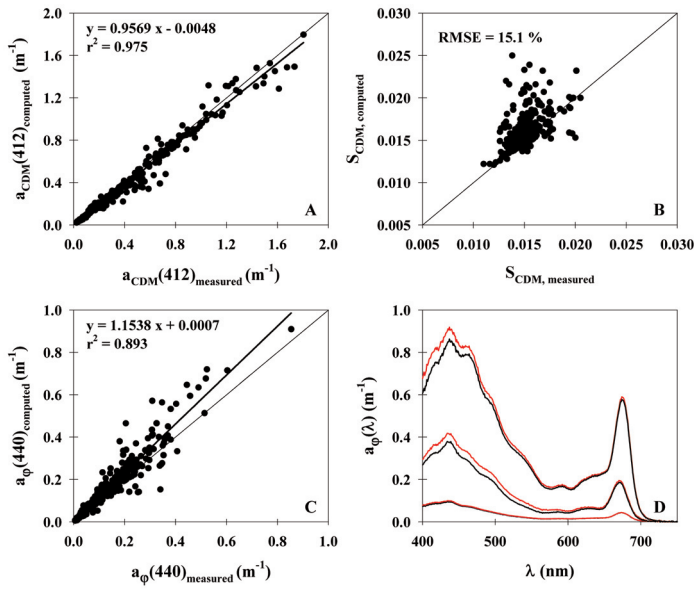|	510/412 nm	555/630[1] nm	
COASTLOOC global	0.43	0.99	314
	0.08	*0.24*	
PROSOPE global	0.44	0.97	268
	0.07	*0.26*	

[1]The average phytoplankton absorption ratio at 555/650 nm also is given for information for the COASTLOOC dataset as for most ac-9s, the wavelength 650 nm replaces 630 nm: $a_\varphi(555)/a_\varphi(650) = 0.92$ (SD: 0.28)

Table 4. Statistical parameters of the relationships between measured and computed absorption coefficients (colored detrital material, a_{CDM}, and phytoplankton, a_φ) derived from total minus water hyper-spectral absorption measurements (ratios: 505/380 and 580/692) and multi-spectral absorption measurements (ratios: 510/412 and 555/630 nm). The model applied is of the type: $y = y_0 + a\,x$. The number of samples of the relationship between computed and measured values slightly varies from one case to another (site by site, region by region, and global cases) because computed CDM slopes out of the range of the measured NAP and CDOM slopes are discarded.

Relationship	n	r^2	Coefficients	Std. Err.	P	RMSE (%)
Hyper-spectral measurements						
Site by site case						
$a_{CDM}(412)_m$ versus $a_{CDM}(412)_c$	306	0.996	$y_0 = 0.0064$	0.0021	0.0026	9.5
			$a = 0.9320$	0.0036	< 0.0001	
$a_\varphi(440)_m$ versus $a_\varphi(440)_c$	306	0.967	0.0017	0.0024	0.4678	24.6
			1.1270	0.0120	< 0.0001	
Region by region case						
$a_{CDM}(412)_m$ versus $a_{CDM}(412)_c$	292	0.975	– 0.0048	0.0053	0.3690	14.1
			0.9569	0.0090	< 0.0001	
$a_\varphi(440)_m$ versus $a_\varphi(440)_c$	292	0.893	0.0007	0.0045	0.8829	37.4
			1.1538	0.0234	< 0.0001	
Global case						
$a_{CDM}(412)_m$ versus $a_{CDM}(412)_c$	294	0.951	0.0077	0.0070	0.2762	21.4
			0.8896	0.0118	< 0.0001	
$a_\varphi(412)_m$ versus $a_\varphi(412)_c$	294	0.795	- 0.0060	0.0073	0.4069	72.4
			1.3524	0.0397	< 0.0001	
$a_\varphi(440)_m$ versus $a_\varphi(440)_c$	294	0.886	- 0.0151	0.0055	0.0067	55.8
			1.3554	0.0285	< 0.0001	
$a_\varphi(510)_m$ versus $a_\varphi(510)_c$	294	0.868	- 0.0039	0.0026	0.1337	65.7
			1.3702	0.0308	< 0.0001	
Multi-spectral (ac-9) measurements						
Global case						
$a_{CDM}(412)_m$ versus $a_{CDM}(412)_c$	300	0.957	- 0.0084	0.0072	0.2401	17.8
			0.9830	0.0120	< 0.0001	
$a_\varphi(412)_m$ versus $a_\varphi(412)_c$	300	0.743	0.0016	0.0070	0.8240	57.8
			1.1109	0.0379	< 0.0001	
$a_\varphi(440)_m$ versus $a_\varphi(440)_c$	300	0.872	- 0.0109	0.0051	0.0337	40.7
			1.1563	0.0257	< 0.0001	
$a_\varphi(510)_m$ versus $a_\varphi(510)_c$	300	0.809	0.0016	0.0025	0.5316	46.3
			1.0425	0.0293	< 0.0001	

constrained conditions. In the "site by site" case, the factors expected to affect the method performance are highly different contributions of phytoplankton and CDM to total minus water absorption coefficient, the compartment with the lower contribution is expected to be retrieved with a lower accuracy, and/or highly different S_{NAP} and S_{CDOM} values (as the computed S_{CDM} describes the spectral dependency of CDOM and NAP as a whole). These two sources of error have been examined successively here by applying the partitioning method to total minus water absorption spectra reconstructed as the sum of a fixed phytoplankton absorption spectrum (simulated using the algorithm of Bricaud et al. 1998, for a chlorophyll concentration equal to 1 mg m^{-3}) and different CDM absorption spectra simulated as detailed below. First, we simulate CDM absorption spectra assuming a fixed slope S (equal to 0.013 nm$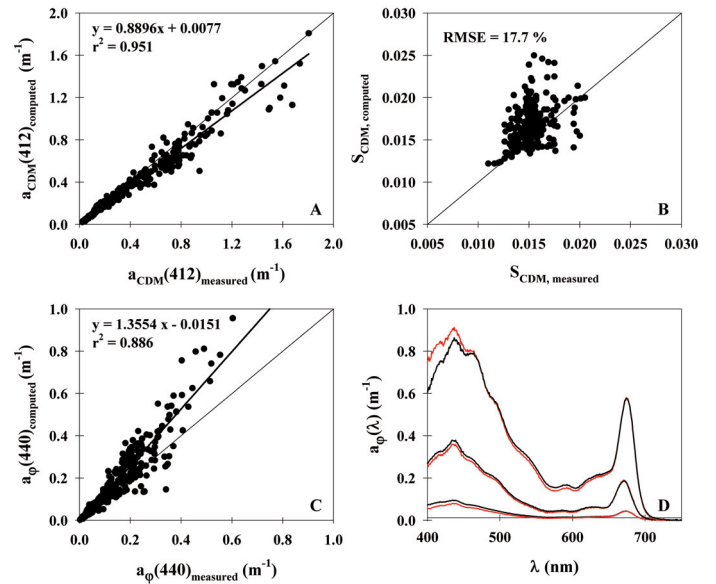^{-1}$, intermediate value between CDOM and NAP) but variable amplitudes A, in order to obtain a CDM contribution to total minus water absorption coefficient ranging from 10 to 90% at 440 nm. This analysis shows that the method performance is not affected by differences in the contributions of phytoplankton and CDM to total minus water absorption coefficient alone. Indeed, on the COASTLOOC dataset, no clear trend was observed in the method performance (i.e., estimates of a_{CDM}, S, or a_φ) in relation to the relative contribution of individual constituents to the absorption coefficient at 440 nm. Second, we simulate various NAP and CDOM absorption spectra assuming increasingly different slopes, ranging from 0.005 to 0.011 nm^{-1} for S_{NAP} and from 0.014 to 0.020 nm^{-1} for S_{CDOM}, and, for each of these cases, increasingly different amplitudes A (so that CDM equaled 10 to 90% of total absorption at 440 nm, as above). The lowest method performance was observed when S_{NAP} and S_{CDOM} were highly different (0.005 and

Fig. 2. Same as in Fig. 1, for the "region by region" analysis in the hyper-spectral case.

Fig. 3. Relationship between absorption parameters measured and computed from total minus water absorption coefficient measurements, for the "global" analysis in the hyper-spectral case. A. Colored Detrital Material absorption coefficient at 412 nm ($a_{CDM}(412)$, in m^{-1}), B. exponential slope of CDM absorption coefficient (S_{CDM}, in nm^{-1}), C. phytoplankton absorption coefficient at 440 nm (in m^{-1}). D. Three examples of computed (red) and measured (black) spectra in the English Channel, Adriatic Sea, and Baltic Sea (Sta. C6024000, C3011000, and C6148000, respectively).

0.020 nm^{-1}, respectively) and the relative contribution of CDM to total absorption was high (e.g., 90%). Interestingly, errors in computed values were low when the contribution of CDM to total absorption was low (10%) even for highly different values of S_{NAP} and S_{CDOM} (0.005 and 0.020 nm^{-1}, respectively). As a conclusion, the first source of error in computed parameters using the present partitioning method comes from a combined effect of large differences between S_{NAP} and S_{CDOM} and in the relative contribution of the individual constituents to total minus water absorption coefficient, but not of this last factor alone.

"Region by region" case: The linear relationship between computed and measured CDM absorption at 412 nm is highly significant ($r^2 = 0.975$), but with a larger dispersion than in the "site by site" case (Fig. 2A, Table 4). S_{CDM} tends to be overestimated (Fig. 2B). Computed and measured phytoplankton absorption coefficients are still in good agreement at a given wavelength (Table 4, e.g., computed and measured values are linked by a highly significant linear relationship with an r^2 equal to 0.893 and a slope of 1.15 at 440 nm, Fig. 2C) as well as spectrally (Fig. 2D). Phytoplankton absorption coefficient tends to be slightly overestimated in the blue region (e.g., by 15% at 440 nm; Table 4, Fig. 2C). However, the overall method performance is still satisfying: RMSE are equal to 14.1%, 15.1%, and 37.4% for CDM absorption at 412 nm, S_{CDM} and phytoplankton absorption at 440 nm, respectively, (Table 4), despite input phytoplankton absorption ratios significantly different from 1.0 (Table 3A), and thus affected by the package effect. The decrease in method performance relative to the "site by site" case is likely the result of the variability of phytoplankton absorption properties within each region, as depicted by the standard deviation of the absorption ratios averaged over each region (Table 3a). The strongest decrease in performance relatively to the "site by site" case is observed for the Adriatic Sea, where the standard deviation of the average phytoplankton absorption ratios is high (Table 3a).

"Global" case: Computed and measured CDM absorption coefficients are in relatively good agreement at 412 nm (linked by a linear relationship with an r^2 equal to 0.951, RMSE equal to 21.4%, Fig. 3A, Table 4). The method performance slightly decreases relatively to the previous cases for CDM retrieval, with measured values at 412 nm being underestimated by ~11% (Fig. 3A) and S_{CDM} being overestimated (Fig. 3B; RMSE equal to 17.7%). Computed and measured phytoplankton absorption coefficients are linked by a highly significant linear relationship (r^2 equal to 0.886 at 440), with a clear tendency for an overestimation of the measured values in the blue region as shown at given wavelengths (e.g., by ~35% at 440 nm, Fig. 3C and Table 4) and spectrally (Fig. 3D).

The increased errors in computed parameters (a_{CDM}, S_{CDM}, and a_φ) relative to the "region by region" case is likely the result of differences in absorption properties between regions, as shown by phytoplankton absorption ratios averaged over each region relatively to the ones computed on the global dataset (Table 3a). Indeed, the COASTLOOC dataset covers a large variety of water types (Babin et al. 2003) and using one set of average phytoplankton absorption ratios computed on the whole dataset does not take into account regional specificities. Inter-
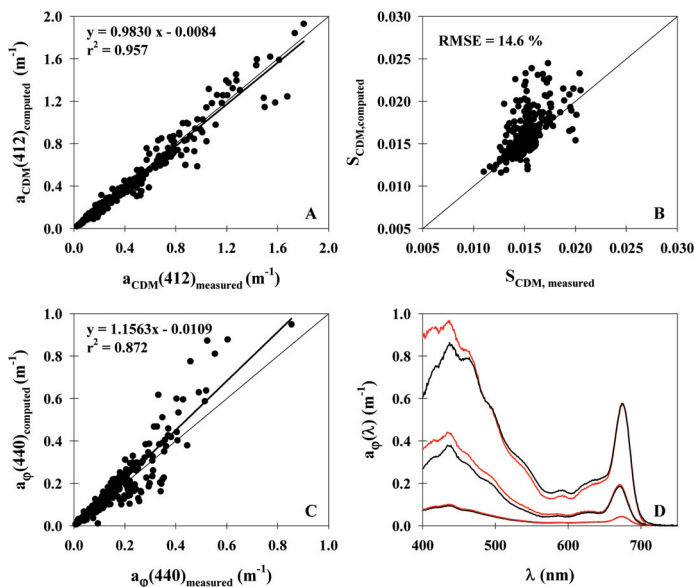
Fig. 4. Same as in Fig. 3, for the "global" analysis in the multi-spectral case (the partitioning method is applied to total minus water absorption data acquired with a limited spectral resolution, here with a WETLabs ac-9).

estingly, the lowest method performance is again observed in the case of the Adriatic Sea, which is characterized not only by a high variability in phytoplankton absorption ratios (see the standard deviation within region) but also strong differences

relatively to the global dataset (Table 3a). Note that the method will perform better in the "global case" for a region where a large number of samples is used to compute the globally averaged phytoplankton absorption ratios used as input. This is the case of the North Sea where the variability in phytoplankton absorption ratios is relatively low and the number of samples used in the global dataset is high (Table 3a).

In conclusion, phytoplankton and CDM absorption spectra can be derived from hyper-spectral measurements of total minus water absorption coefficient using the present partitioning method with a good accuracy, and this accuracy can be significantly increased by an a priori knowledge of the absorption properties of the phytoplankton species occurring in the study area.

Multi-spectral measurements: The "average" phytoplankton absorption ratios computed at the two new pairs of wavelengths used for the multi-spectral (ac-9) case (510/412 and 555/630 nm) on the COASTLOOC dataset (as for the hyper-spectral case) are given in Table 3b. Computed and measured CDM absorption coefficients at 412 nm are linked by a highly significant linear relationship ($r^2 = 0.957$, Fig. 4A and Table 4) with a slope close to 1.0 (0.98). Computed and measured S_{CDM} values are in relatively good agreement (Fig. 4B, RMSE equal to 14.6%), with a tendency for overestimation of the measured values, as for the "global" hyper-spectral case. Computed and measured phytoplankton absorption coefficients are in good
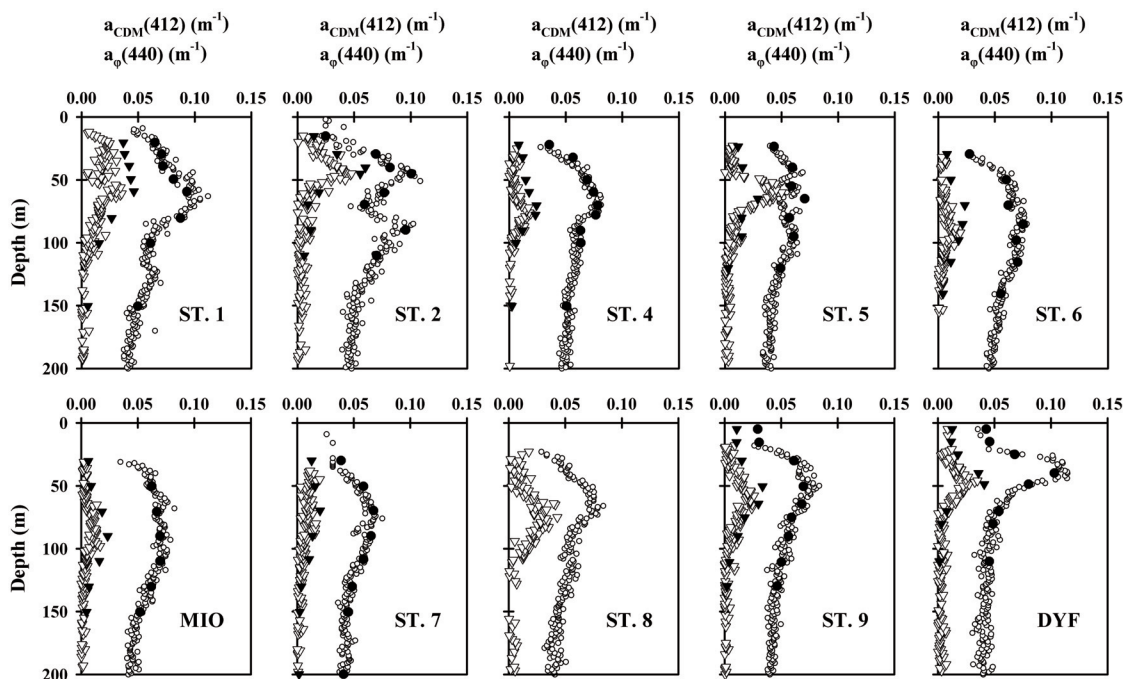
Fig. 5. Application of the partitioning method to vertical profiles of total spectral absorption coefficient measured in situ using a WETLabs ac-9 in various areas of the Mediterranean Sea (PROSOPE cruise). Comparison between computed and measured CDM absorption coefficients at 412 nm, and between computed and measured phytoplankton absorption coefficients at 440 nm. Black circles: measured CDM absorption coefficient at 412 nm ($a_{CDM}(412)$), black triangles: measured phytoplankton absorption coefficient at 440 nm ($a_{\varphi}(440)$); white corresponding symbols: computed values. Note that at Sta. 8, only computed values are presented because of the lack of direct measurements.

agreement at given wavelengths (e.g., linked by a significant linear relationship with an r^2 equal to 0.872 and a slope of 1.16 at 440 nm, respectively, Table 4, Fig. 4C) and spectrally (Fig. 4D). We observe a slight tendency for an overestimation of the true phytoplankton absorption coefficient in the blue (Table 4 and Figs. 4C, D; e.g., by 16% at 440 nm) as for the hyper-spectral "global" case. These results show that phytoplankton and CDM absorption coefficients can be retrieved from total minus water absorption coefficient measurements made with a limited spectral resolution with a good accuracy: RMSE equal to 17.8% and 40.7% for CDM absorption at 412 nm and phytoplankton absorption at 440 nm, respectively (Table 4), despite the fact that average phytoplankton absorption ratios at the new pairs of wavelength do not satisfy fully the conditions of application of the Bricaud and Stramski (1990) method (e.g., phytoplankton absorption ratio at 412/510 nm significantly different from 1.0). Interestingly, the partitioning method appears to perform better in the multi-spectral "global" case than in the hyper-spectral "global" case, likely as a result of a phytoplankton absorption ratio at 555/630 nm closer to 1.0 (0.99, Table 3b) than at 580/692 nm (1.25, Table 3a).

Application to in situ high resolution measurements acquired with an ac-9

Vertical profiles: PROSOPE Campaign: The model presented here is applied to in situ vertical profiles of total minus water spectral absorption coefficient acquired with an ac-9 over different trophic regimes in the Mediterranean Sea (PROSOPE campaign, table 1; Oubelkheir et al. 2005). The computed CDM absorption coefficient at 412 nm is compared with measurements derived from the sum of CDOM absorption coefficients measured with an ac-9 with a filter, and non-algal particle absorption coefficients determined on discrete samples (*see* Methods). The phytoplankton absorption ratios averaged over the whole PROSOPE dataset used as inputs of the partitioning method are given in Table 3b. We used the Bricaud and Stramski (1990) method here to compute both phytoplankton and

non-algal particle absorption coefficients from particle absorption coefficient measurements (using the filter-pad technique) and phytoplankton and CDM absorption coefficients from total minus water absorption coefficient measurements (made with the ac-9). Because CDM is mainly CDOM in the waters explored, this approach is considered not to be circular for the purpose of the present method validation.

Vertical profiles of computed and measured $a_{CDM}(412)$ are in good agreement over the gradient of trophic conditions sampled (Fig. 5). As an example, at Sta. 2 in the Almeria-Oran frontal area, the two measured maxima of $a_{CDM}(412)$ (at ~45 and ~90 m) are well reproduced using the partitioning method. It is worth noting that the absence of a maximum in $a_\varphi(440)$ at 90m is very well reproduced despite the presence of a significant maximum in $a_{CDM}(412)$, which illustrates the discrimination power of our method. Computed CDM exponential slopes at the CDM maximum depth (Table 5) are comprised between 0.013 and 0.017 nm^{-1}, which is in the range of values expected for oceanic waters (*see* introduction). The above results show that CDM and phytoplankton absorption coefficients can be accurately determined from in situ ac-9 measurements of total minus water spectral absorption coefficient in open ocean waters, where average phytoplankton absorption ratios used as input in the partitioning method are rather stable over various trophic conditions. The good performance of this method in oligotrophic areas, where total minus water absorption coefficients are low and thus subject to greater relative errors in absolute measurements, is a good indicator of its robustness. Note that there is a definite requirement for high quality pure water calibrations of the ac-9 in oligotrophic oceanic waters, as errors in measurements at the two pairs of wavelengths used as input to the partitioning method will introduce errors in the computed parameters.

Surface underway profiles – ALMOFRONT II campaign: The CDM determinations estimated following the present method and Claustre et al. (2000) method are in good agreement at 412

Table 5. Measured chlorophyll *a* concentration and contribution of non-algal particles to CDM absorption at 440 nm, and CDM absorption coefficient exponential slope (S_{CDM}) computed through the numerical partitioning method, in various trophic conditions in the Mediterranean, at Sta. 1-9, DYF, and MIO of the PROSOPE cruise.

Site-Sta. Number	Chl*a* concentration in mg m^{-3} (depth of chlorophyll maximum)	Computed S_{CDM} (depth of CDM maximum)	Max % a_{NAP} in a_{CDM}
Sta. 1–11	0.72 (49.5 m)	0.013 (63 m)	15
Sta. 2–14	0.89 (40.2 m)	0.014 (41 m)	15
Sta. 4–19	0.23 (77.9 m)	0.017 (64 m)	14
Sta. 5–23	0.96 (55.1 m)	0.017 (46 m)	15
Sta. 6–26	0.33 (70.2 m)	0.016 (85 m)	10
MIO – 31	0.23 (90 m)	0.016 (89 m)	8
Sta. 7–64	0.23 (69.7)	0.016 (73 m)	10
Sta. 8–67	0.27 (70.4)	0.017 (59 m)	*#NA*
Sta. 9–70	0.42 (50.5 m)	0.014 (50 m)	13
DYF – 94	0.56 (48.7 m)	0.015 (40 m)	11
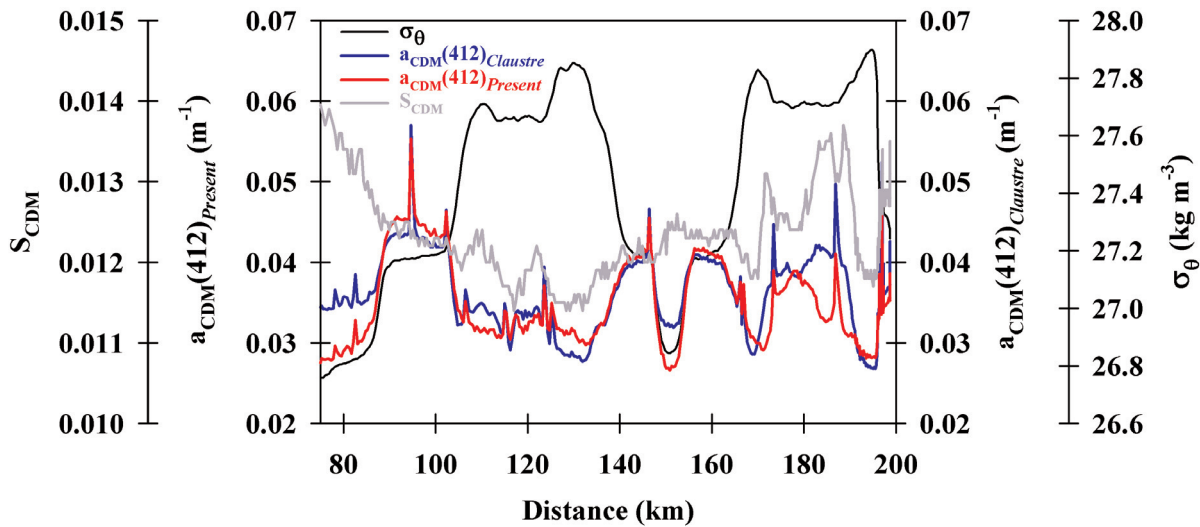
Fig. 6. Application of the partitioning method to horizontal cross sections of total minus water spectral absorption coefficient measured in the surface layer of a frontal area using a WETLabs ac-9 (ALMOFRONT II campaign). Comparison between CDM absorption coefficients at 412 nm computed using the present method (and corresponding S_{CDM} slopes) and using a method published by Claustre et al. (2000).

nm over the diversity of water types crossed in the Almeria-Oran frontal system (Fig. 6). The spatial distribution of the CDM absorption coefficient derived using both methods presented clear covariations with the surface density field over the various water types explored. In particular, higher CDM (and CDOM) absorption coefficient values were observed at the level of the density steps (~27.2 kg m^{-3}) in the areas of density gradients between Atlantic and Mediterranean waters (*see* horizontal profiles; Fig. 6). However, we can notice some areas where CDM absorption coefficients estimated through our method are slightly lower than using the method of Claustre et al. (2000), as, for example, in the dense Mediterranean waters (~180–190 km from the beginning of the transect), for reasons not fully explained based on the available in situ information.

The variability in CDM absorption spectral shape (S_{CDM}) also can be examined along the same transect using our partitioning method, providing an additional level of information on CDM; Claustre et al. (2000) method produces no information about S. Computed S_{CDM} are comprised between 0.0114 nm^{-1} (in typically Mediterranean waters) and 0.0139 nm^{-1} (in Atlantic waters crossed at the start of the transect), which is in the range observed for CDOM and NAP in oceanic waters (*see* introduction). Interestingly, the variability in S_{CDM} either reveals changes in the respective contributions of NAP and CDOM to the CDM signal or, if CDOM is dominant in the CDM signal, changes in the chemical composition and/or photo-oxidation state of CDOM (leading to associated changes in S_{CDM}). Measurements performed on discrete samples in the same area and during the same campaign (Claustre et al. 2000) show that CDOM dominates the CDM signal in all the waters explored, thus favoring the second hypothesis in the present case. This example study shows that the information contained in the CDM spectral dependency can now be accessed in situ with a high resolution

using the present partitioning method, allowing us to gain a further understanding of the factors driving CDM (and eventually CDOM) dynamics in the marine environment.

Conclusion and recommendations

The numerical method presented here allows the partitioning of total spectral absorption coefficients, measured with a high spectral resolution or with a limited spectral resolution, into hyper-spectral (or spectral) phytoplankton and colored detrital material absorption coefficients. This method has been validated here on a large dataset of in situ measurements made over a large variety of water types (from coastal to oceanic conditions) around Europe and in the Mediterranean Sea. For application to in situ continuous absorption coefficient measurements, the choice of the wavelengths was constrained by the specifications of commercially available absorption meters (WETLabs ac-9); a constraint which has been partly removed recently by the development of hyper-spectral in situ spectrophotometers (WETLabs ac-s, spectral resolution of 4 nm between 400 and 730 nm).

Interestingly, in waters where the contribution of CDOM to CDM absorption coefficient can be known (a priori or based on simultaneous discrete determinations), particulate attenuation spectra can be computed as the difference between total minus water attenuation and CDM (derived from CDM) absorption coefficients. Therefore, the partitioning method proposed here allows us to estimate the slope of the particulate spectral attenuation coefficient from simultaneous measurements of total spectral absorption and attenuation coefficients made using an ac-9 (or ac-s). Past and recent studies (Boss et al. 2001*a*, 2001*b*; Oubelkheir et al. 2005; and references therein) have shown that the slope of the particulate attenuation spectrum can be related to changes in the size of the particle assemblage. Inde-

pendently, Ciotti et al. (2002) have shown that a cell size parameter can be retrieved from phytoplankton absorption spectra. As a result, optical indices of the size of both particle assemblage and phytoplankton are accessible in situ and with a high spatial and temporal resolution from ac-9 (or ac-s) measurements when using the present partitioning method (e.g., over a diel cycle, Oubelkheir et al. in prep.).

Finally, as shown by Ciotti and Bricaud (2006) using SeaWiFS data, the same partitioning method can be applied to multi-spectral or hyper-spectral absorption coefficient measurements derived from satellite ocean color measurements and inverse radiative transfer models (e.g., Loisel and Stramski 2000; Lee and Carder 2003), in order to extract synoptic maps of phytoplankton and CDM absorption spectra on regional scales. The information derived on a large range of spatial and temporal scales, from the micro-scale (using in situ spectral absorption-attenuation meter with a high resolution) to the macro-scale (e.g., from satellite measurements) will provide us with tools for better understanding the parameters that determine the variability in phytoplankton and CDM distributions in coastal and oceanic environments.

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