

A novel method to estimate DOC concentrations from CDOM absorption coefficients in coastal waters

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[1] A novel method to accurately retrieve DOC concentrations ($\pm 4\%$) from CDOM absorption coefficients, $a_g(\lambda)$, at $\lambda = 275$ and 295 nm is presented. By using these two wavelengths, the method exploits useful information about the ratio of $a_g(\lambda)$ to [DOC] contained in the 275–295 nm spectral slope coefficient, $S_{275-295}$. This approach was developed using data ($n = 222$) collected on a seasonal basis in surface waters of the Northern Gulf of Mexico. The approach is demonstrated to accurately and consistently estimate [DOC] for all seasons and for a broad [DOC] range of 63–611 μM . Application to coastal waters of the Beaufort Sea ($n = 33$) demonstrated that similar performance can be expected in other river-influenced ocean margins after parameterization using local data. Applicability to other marine environments remains to be tested but such assessment can be pursued immediately where appropriate data have already been collected. **Citation:** Fichot, C. G., and R. Benner (2011), A novel method to estimate DOC concentrations from CDOM absorption coefficients in coastal waters, *Geophys. Res. Lett.*, 38, L03610, doi:10.1029/2010GL046152.

1. Introduction

[2] Conventional methods for the analysis of Dissolved Organic Carbon (DOC) are restricted to measurements of discrete samples and are limited to providing synoptic coverage on relatively small spatial scales. The estimation of DOC concentrations, [DOC], through measurement of the optical properties of dissolved organic matter (DOM) (absorption and fluorescence) therefore represents a compelling alternative. Under optimal conditions and with proper instrumentation, the optical properties of DOM can be rapidly and continuously acquired *in situ* [Vodacek et al., 1997; Hitchcock et al., 2004].

[3] The relationship between [DOC] and DOM absorption (Chromophoric Dissolved Organic Matter, CDOM) has been investigated in a variety of coastal systems [Ferrari et al., 1996; Del Vecchio and Blough, 2004a; Guéguen et al., 2005]. Although strong positive correlations have been observed between CDOM absorption coefficients, $a_g(\lambda)$, and [DOC], the relationship varies among geographical regions and seasons [Blough and Del Vecchio, 2002]. For example, the ratio of $a_g(\lambda)$ to [DOC] varies seasonally by more than 25-fold in the Middle Atlantic Bight alone [Del Vecchio and Blough, 2004a]. Such variability in this ratio sets a limit on our capability to predict [DOC] from simple linear relationships between DOC and CDOM.

[4] The spectral characteristics of $a_g(\lambda)$ are representative of the types of chromophores present in DOM [Del Vecchio and Blough, 2004b]. A single exponential fit (equation 1) is typically used to describe the spectral dependency of $a_g(\lambda)$:

$$a_g(\lambda) = a_g(\lambda_0)e^{-S(\lambda-\lambda_0)} \quad (1)$$

where $\lambda_0 < \lambda$ and S is the spectral slope coefficient in the λ_0 to λ nm spectral range. Several studies have related the spectral characteristics of DOM absorption to the chemical and structural nature of DOM such as molecular weight and aromaticity [Chin et al., 1994; Helms et al., 2008]. Other studies utilized molar absorptivity or carbon-specific UV absorbance as indicators of the molecular weight and aromatic content of organic matter isolates [Chin et al., 1994; Weishaar et al., 2003]. A linkage between the spectral characteristics of $a_g(\lambda)$ and the ratio $a_g(\lambda)/[\text{DOC}]$ is therefore possible. If such a connection exists for DOM in the marine environment it could be exploited to improve predictions of [DOC] from measurements of $a_g(\lambda)$.

[5] In this study, we explore the relationship between S and the ratio $a_g(\lambda)/[\text{DOC}]$ in surface waters of the Northern Gulf of Mexico (NGoM), with the intent of testing this hypothesis. Following recommendations by Helms et al. [2008] on the use of S in the 275–295 nm spectral range, a strong relationship between $S_{275-295}$ and $a_g(\lambda)/[\text{DOC}]$ was discovered. This connection is exploited in a method to accurately estimate [DOC] from simple *in situ* measurements of $a_g(275)$ and $a_g(295)$ in coastal areas. Its applicability in other marine systems is discussed.

2. Sampling and Methods

[6] Surface water from the NGoM was collected and filtered for DOC analysis and CDOM absorbance measurements. A total of 222 stations ($n = 222$) were sampled during five research cruises (January, April, July, October/November 2009 and March 2010) as part of the GULFCARBON project. About 50 stations were sampled per cruise (Figure 1a) with the exception of January 2009 when 24 stations were sampled. Representing a salinity range of 0–37 psu, these samples include most water types typically encountered in river-dominated ocean margins. DOC analysis was done by High Temperature Combustion (HTC) and CDOM absorbance, $A(\lambda)$, was measured using a dual-beam spectrophotometer. Absorbances were converted to absorption coefficients, $a_g(\lambda)$, and spectral slope coefficients, S , were calculated using linear fits of log-linearized $a_g(\lambda)$. The carbon-specific absorption coefficients of DOM were calculated as the ratio of $a_g(\lambda)$ to DOC concentration and are denoted here as $a_g^*(\lambda)$, with units of $\text{m}^{-1} \mu\text{M}^{-1}$. The value of $a_g^*(\lambda)$ at $\lambda = 355$ nm was calculated for consistency with previous studies [Vodacek

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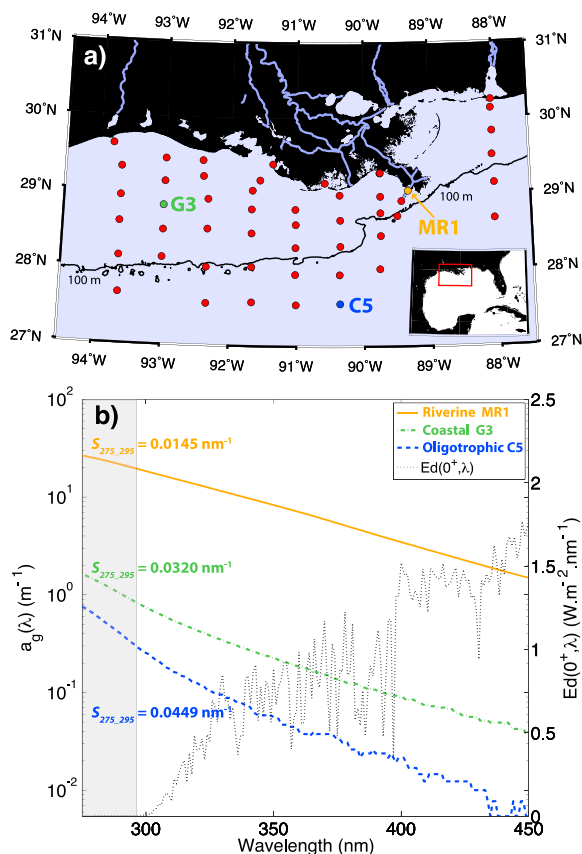


Figure 1. (a) Station locations in the Northern Gulf of Mexico. (b) CDOM absorption coefficient spectra, $a_g(\lambda)$, and corresponding spectral slope coefficient, $S_{275-295}$, for three contrasting water samples. The y-axis for $a_g(\lambda)$ is a log scale. $S_{275-295}$ is typically low in river water and increases in coastal and oligotrophic waters. A modeled, downward plane irradiance spectrum just above the sea surface, $E_d(0^+, \lambda)$, is overlaid and illustrates the absence of photons at wavelengths (λ) < 295 nm. $E_d(0^+, \lambda)$ was modeled for June 21st, 12:00 p.m., at latitude 28°N, with 300 DU of ozone, and a clear sky. These conditions correspond to the maximal incident irradiance expected in the Northern Gulf of Mexico.

et al., 1997; Del Vecchio and Blough, 2004a]. Detailed sampling and methods are provided in the auxiliary material.¹

[7] Surface water from the Beaufort Sea was sampled in August 2009 as part of the MALINA project. A total of 33 stations ($n = 33$) were sampled across a salinity gradient of 0 to 30 psu and [DOC] and absorbance were measured as described above.

3. Dynamics of DOC and CDOM in the Northern Gulf of Mexico

[8] The measured range of DOC concentrations, ([DOC]: 63–611 μM) spanned an order of magnitude over the salinity range of 0 to 37 psu. The strong relationship between salinity and [DOC] ($r^2 = 0.83$) indicates that DOM dynamics in surface waters were, to a first degree, domi-

¹Auxiliary materials are available in the HTML. doi:10.1029/2010GL046152.

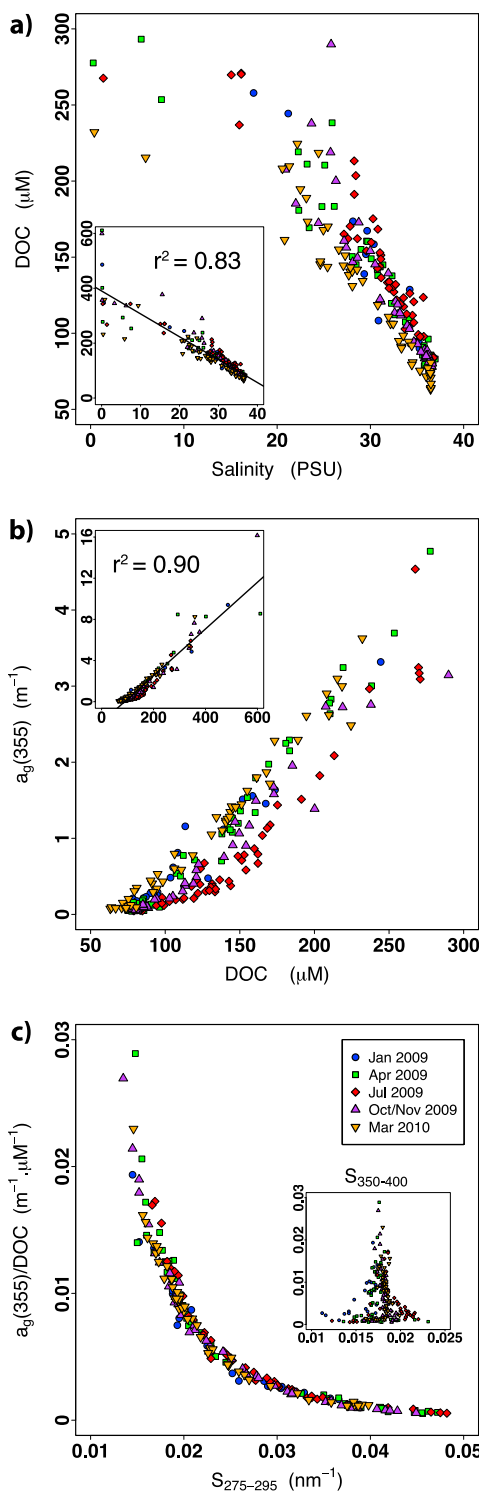


Figure 2. Relationships between different DOM properties in the Northern Gulf of Mexico: (a) [DOC] vs. salinity; (b) $a_g(355)$ vs. [DOC]; (c) $a_g^*(355) = a_g(355)/[\text{DOC}]$ vs. $S_{275-295}$. In Figures 2a and 2b, the inset plots present linear regressions for the entire range of the data whereas the main plots magnify the 50–300 μM [DOC] range. In Figure 2c, the inset plot shows the lack of relationship between $S_{350-400}$ and $a_g^*(355)$.

nated by terrigenous inputs (inset of Figure 2a). Within this general view, however, a significant seasonality and deviation from linear mixing at salinity extremes was apparent (Figure 2a). A poor correlation observed between [DOC] and salinity at salinities less than 20 psu ($r^2 = 0.18$) indicated the presence of multiple riverine sources with varying DOM properties. This observation is in agreement with earlier studies demonstrating varying mixing behavior of DOC as it transits from estuaries to the Gulf of Mexico [Guo *et al.*, 1998].

[9] The NGoM is a river-dominated system in terms of DOM optical properties [Chen and Gardner, 2004; Conmy *et al.*, 2004; D'Sa and DiMarco, 2009]. A strong, linear relationship ($r^2 = 0.90$) was observed in the present study between [DOC] and the CDOM absorption coefficient, $a_g(355)$ (inset of Figure 2b). However, this strong relationship is misleading for the purpose of retrieving [DOC] from $a_g(355)$. The ratio $a_g(355)/[\text{DOC}] = a_g^*(355)$, varies by a factor of 55, from a minimum value of $5.3 \cdot 10^{-4} \text{ m}^{-1} \mu\text{M}^{-1}$ in the most oligotrophic sample to a value of $2.9 \cdot 10^{-2} \text{ m}^{-1} \mu\text{M}^{-1}$ in freshwater. The trend, range and values of $a_g^*(355)$ observed in this study are in general agreement with those observed by Del Vecchio and Blough [2004a] in the Middle Atlantic Bight. Some of this variability can be attributed to the conservative mixing of river water and seawater along the salinity gradient and can be accounted for in a linear relationship of the form $[\text{DOC}] = \alpha + \beta a_g(355)$, where α and β are regression coefficients. However, a magnified view of the relationship between [DOC] and $a_g(355)$ (Figure 2b) also shows a strong seasonality and some non-linearity which can result from: 1) the presence of multiple riverine sources with different $a_g^*(355)$; 2) the decoupling between the autochthonous sources and sinks of DOC with those of CDOM; and 3) photobleaching, known to decrease $a_g^*(355)$ [Del Vecchio and Blough, 2004a]. This variability cannot be constrained in the simple linear model and although seasonal linear models can be derived, their implementation is always difficult.

[10] Numerous studies have investigated the dynamics of the spectral slope coefficient in aquatic environments and have concluded that it is of limited utility as a biogeochemical indicator [Blough and Del Vecchio, 2002]. However, the spectral range used among investigators has been inconsistent, generally broad (e.g., 290–700 nm) and restricted to the UV-A and visible domains. Helms *et al.* [2008] recently suggested the use of $S_{275-295}$ (UV-B domain, narrow range) as an indicator of photochemical alterations and DOM molecular weight and source in the marine environment. In light of their results, a remarkable finding of the present study is a strong relationship between $S_{275-295}$ and $a_g^*(355)$ (Figure 2c), which is best approximated by an exponential equation of the form: $a_g^*(355) = e^{(\alpha - \beta S_{275-295})} + e^{(\gamma - \delta S_{275-295})}$, where α , β , γ and δ are regression coefficients. This relationship is remarkable because a strong connection does not exist between $a_g^*(355)$ and other spectral slope coefficients such as $S_{300-350}$, $S_{300-400}$ or $S_{350-400}$ in this data set (inset of Figure 2c), thereby highlighting the unique potential of the 275–295 nm spectral range to retain information about DOM composition and the ratio of $a_g(\lambda)$ to [DOC]. The strong link between $S_{275-295}$ and $a_g^*(355)$ is indicative that the dynamics of these two DOM properties are regulated by the same processes. Although gaining a full understanding of the dynamics responsible for this rela-

tionship is beyond the scope of this manuscript, it can be inferred from a few recent studies that the processes responsible for the variabilities in $S_{275-295}$ and $a_g^*(355)$ are of the same nature [Del Vecchio and Blough, 2004a; Helms *et al.*, 2008; Ortega-Retuerta *et al.*, 2009].

[11] A unique and important aspect of $S_{275-295}$ not mentioned by Helms *et al.* [2008] is the unique position of the 275–295 nm spectral region on the outside edge of the natural solar spectrum (Figure 1b). Even under optimal conditions, very few photons of $\lambda < 295$ nm are present in the natural environment. According to the work of Del Vecchio and Blough [2002] on the photobleaching of $a_g(\lambda)$ using monochromatic irradiations, the decrease in $a_g(\lambda)$ upon absorption of photons of wavelength λ_{ex} is maximum at or near $\lambda = \lambda_{ex}$ and decreases exponentially towards other wavelengths. It is therefore expected that any natural photon absorbed would always lead to a greater change in $a_g(295)$ than in $a_g(275)$, and consequently, to an increase in $S_{275-295}$. In contrast, other spectral regions used for the determination of S tend to overlap with the photochemically-active part of the natural solar spectrum (typically 300–400 nm). A well behaved response of S to photobleaching is therefore unlikely for spectral regions other than 275–295 nm. This phenomenon can contribute to the erratic behavior of S typically observed in the marine environment.

[12] An important implication of the relationship between $S_{275-295}$ and $a_g^*(\lambda)$ in this system is the novel capability to constrain the variability in the ratio $a_g(\lambda)/[\text{DOC}]$ using information contained in the spectral shape of $a_g(\lambda)$. Because $a_g(\lambda)/[\text{DOC}]$ can vary by a large factor, exploiting this information can considerably improve the accuracy of [DOC] retrieved from $a_g(\lambda)$.

4. Estimating DOC from CDOM

[13] DOC concentrations can be retrieved from the combination of $a_g(\lambda)$ and a non-linear fit of $a_g^*(\lambda)$ versus $S_{275-295}$. However, we found through extensive testing that the most accurate [DOC] were obtained by performing multiple linear regressions (MLR) of log-linearized [DOC] against log-linearized $a_g(275)$ and $a_g(295)$, as described in equation (2):

$$\ln[\text{DOC}] = \alpha + \beta \ln[a_g(275)] + \gamma \ln[a_g(295)] \quad (2)$$

where α , β and γ are regression coefficients.

[14] This method exploits all the useful information contained in $S_{275-295}$ while being simpler, more direct and accurate. The best wavelengths for prediction of [DOC] were $\lambda = 275, 295$ nm. The use of additional variables in the MLR (e.g., $a_g(\lambda)$ at $\lambda \neq 275, 295$ nm, salinity, chlorophyll-a fluorescence) did not improve the predictive capability of the model. A MLR against $\ln[a_g(275)]$ and $\ln[a_g(295)]$ therefore represents an optimal model. In order to relieve the constraint of using a single MLR for a broad range of [DOC] (63–611 μM), the data were separated into two subsets and a specific MLR was done on each subset. The data were divided based on the cutoff value $a_g(275) = 3.5 \text{ m}^{-1}$, which corresponds to the median value in these data. The regression coefficients are provided in Table 1 and the performance of the model is evaluated in Figure 3.

[15] The performance of the model (Figure 3b) was compared to that of a single regression of $\ln[\text{DOC}]$ versus $\ln[a_g(355)]$ model (Figure 3a). A large seasonal bias and

Table 1. Parameters α , β and γ Derived From the Multiple Linear Regressions of \ln [DOC] Against \ln [$a_g(275)$] and \ln [$a_g(295)$] for the Northern Gulf of Mexico and the Beaufort Sea^a

	α	β	γ	Adjusted r^2
<i>Northern Gulf of Mexico</i>				
$a_g(275) \leq 3.5 \text{ m}^{-1}$	3.4707	1.8591	-1.2421	0.92
$a_g(275) > 3.5 \text{ m}^{-1}$	2.9031	2.7703	-2.0400	0.95
<i>Beaufort Sea</i>				
$a_g(275) \leq 3.5 \text{ m}^{-1}$	4.2952	0.1153	0.3187	0.80
$a_g(275) > 3.5 \text{ m}^{-1}$	2.3603	3.0395	-2.1298	0.96

^aSee equation (2).

poor accuracy at low and high DOC concentrations resulted from the use of the single regression, even after log-linearization of [DOC] and $a_g(355)$. An even larger bias and lower accuracy was observed if these values were not log-linearized before regression. The new approach demonstrates that the use of two carefully chosen wavelengths and their use in MLR can considerably improve the accuracy of estimated DOC concentrations. Overall, [DOC] estimated using this approach were within $\pm 4.2\%$ of the measured [DOC]. For comparison, the percent error associated with replicates of [DOC] measurements was typically $\pm 1\%$. A sensitivity analysis of the model also revealed that about half of the $\pm 4.2\%$ error could be attributed to errors in the reproducibility of the absorbance measurements. The distribution of points around the 1-to-1 line indicated that the percent error associated with the [DOC] retrieved using this approach was consistent over the entire [DOC] range (Figure 3b). The error associated with the estimate is typically $\pm 2.4 \mu\text{M}$ for a measured [DOC] value of $60 \mu\text{M}$, and $\pm 12 \mu\text{M}$ for a measured [DOC] value of $300 \mu\text{M}$.

5. Applicability of the Approach

[16] The results presented in the previous section are valid in the NGoM, for which the model was parameterized. Application of these parameters to other marine systems can therefore lead to unpredictable results. In order to test the applicability of the approach to a different coastal system, we applied it to independent data ($n = 33$) acquired in August 2009 in the Beaufort Sea, in a region influenced by the Mackenzie river outflow. Our results indicate that applying the approach to this region using the NGoM parameters results in decreased accuracy ($\pm 18\%$ of measured [DOC]) and biases in the derived [DOC]. However, excellent accuracy ($\pm 4.7\%$) and a consistent percent error over the full range of measured [DOC] ($66\text{--}458 \mu\text{M}$) is obtained after the model was re-parameterized using local data (Figure 3c and Figure 4 in the Auxiliary Material). The regression coefficients derived for the Beaufort Sea are given in Table 1.

[17] Although the approach may be applied to other coastal environments, differences in DOM sources and regulatory processes make the re-parameterization of the model using local data necessary to achieve high performance. Suitable data have already been collected in other coastal systems such as the Middle Atlantic Bight [Del Vecchio and Blough, 2004a; Mannino et al., 2008] and are therefore readily available for parameterizing the model. For other marine systems where both the range in DOM properties and the

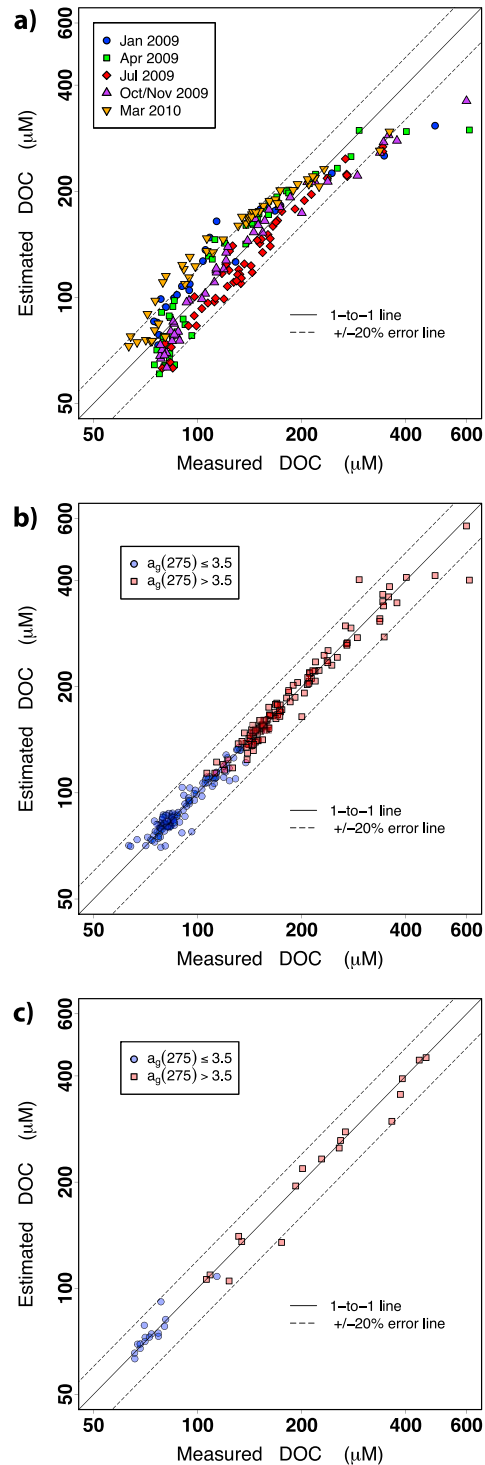


Figure 3. (a) [DOC] estimated using a single linear regression of \ln [DOC] against \ln [$a_g(355)$] in the Northern Gulf of Mexico; (b) [DOC] estimated using the new approach in the Northern Gulf of Mexico. (c) [DOC] estimated using the new approach in the Beaufort Sea, after re-parameterization of the model using local data (see Figure 4 in the Auxiliary Material for station locations). In all plots, “Estimated DOC” indicates concentrations estimated from $a_g(\lambda)$.

chemical composition of DOM is less influenced by terrigenous inputs (e.g., Sargasso Sea), the validity of the approach itself remains to be assessed.

[18] Besides its simplicity of implementation, this method presents a number of advantages that make it suitable for high-resolution and long-term *in situ* monitoring of [DOC] in coastal environments. First, absorbance measurements at only two wavelengths are required, which can be acquired at a fast rate while storing minimal amounts of data. Second, the values of $a_g(275)$ and $a_g(295)$ are high in most environments thereby making the approach less sensitive to limitations in the precision of the instrument. Third, the relationship between [DOC] and $a_g(275)$ or $a_g(295)$ should remain minimally affected by changes in inorganic ion concentrations (e.g., nitrate, nitrite, bromide and bisulfide) [Johnson and Coletti, 2002]. Finally, if the model is parameterized using representative data, the approach should be applicable to a given region for all seasons using a single parameterization.

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