Colored and Chromophoric Dissolved Organic Matter in Natural Waters

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1 Introduction

Chromophoric and colored dissolved organic matter (CDOM) is the opitically active components of bulk dissolved organic matter (DOM) composing of a complex mixture of organic compounds of both allochthonous and autochthonous origin, which absorb light at both ultraviolet (UV) and visible wavelengths (Bricaud et al. 1981; Arrigo and Brown 1996; Nelson et al. 1998; Nelson et al. 2004; Warnock et al. 1999; Nelson and Siegel 2002; Vähätalo and Wetzel 2004; Coble 2007; Zhang et al. 2009; Mostofa et al. 2009). Allochthonous organic substances

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are generally derived from terrestrial plant materials in soil, while autochthonous organic substances are produced mostly from algae, phytoplankton and bacteria within the water column. Allochthonous CDOM (mostly fulvic and humic acids) originating in terrestrial environments flows through rivers and estuaries onto coastal shelves and then reaches the open ocean. During such transport it experiences large changes in ionic composition and physicochemical environment (Mostofa et al. 2009; Malcolm 1985; Malcolm 1990; Wetzel 1992; Nakane et al. 1997; Uchida et al. 2000; Vodacek et al. 1997; Mitra et al. 2000; Fahey et al. 2005; Murphy et al. 2008). Autochthonous CDOM of algal, phytoplankton and bacterial origin is generally composed of autochthonous fulvic acids, carbohydrates, amino acids, proteins, lipids, organic acids and so on (Nelson et al. 2004; Coble 2007; Zhang et al. 2009; Coble 1996; Tanoue 2000; Jennings and Steinberg 1994; Rochelle-Newall and Fisher 2002; Yamashita and Tanoue 2003; Yamashita and Tanoue 2004; Yamashita and Tanoue 2008; Stedmon and Markager 2005; Stedmon et al. 2007; Stedmon et al. 2007; Wada et al. 2007; Helms et al. 2008; Hulatt et al. 2009; Ortega-Retuerta et al. 2009; Mostofa et al. 2009). Phytoplankton is capable of releasing 10–60 % of the carbon and 15–50 % of the nitrogen assimilated during photosynthesis (Sundh 1992; Bronk et al. 1994; Braven et al. 1995; Malinsky-Rushansky and Legrand 1996; Slawyk et al. 1998; Slawyk et al. 2000). As a consequence, CDOM levels are significantly increased after phytoplankton blooms in natural waters (Billen and Fontigny 1987; Ittekkot 1982). The contributions of allocthonous fulvic and humic acids, autochthonous fulvic acids, carbohydrates, amino acids, proteins, lipids and organic acids are significantly different among rivers, lakes and oceans. Such differences are discussed in detail in the DOM chapter.

The absorption and scattering coefficients of water were determined firstly by the aquatic scientist Gamburtsev in 1924 (cited in Kozlyaninov (MV 1972)) and then in other studies (Clarke and James 1939; Duntley 1942; Yentsch 1960; Preisendorfer 1961; Latimer 1963; Sullivan 1963; Kozlyaninov and Pelevin 1966; Jerlov 1968). Jerlov (1968) has been the first to hypothesize that CDOM in natural waters absorbs maximally in the blue region of the spectrum and that its absorption decreases exponentially with increasing wavelength, up to the photosynthetically available radiation (PAR) waveband. Since then a lot of research studies have been conducted on the absorption and scattering properties of CDOM, chlorophyll and particulate material in natural waters (Kozlyaninov and Pelevin 1966; Clarke et al. 1970; Tyler and Smith 1970; Lorenzen 1972; Petzold 1972; Morel 1973; Arvesen et al. 1973; Clarke and Ewing 1974; Duntley et al. 1974; Gordon et al. 1975; Maul and Gordon 1976; Prieur 1976; Morel and Prieur 1977; Bukata et al. 1979; Kirk 1981; Gordon and Morel 1983; Kirk 1988).

High molecular weight DOM, and particularly allochthonous fulvic and humic acids and autochthonous fulvic acids, absorb radiation along a broad spectrum from 250 to 800 nm (Warnock et al. 1999; Zhang et al. 2009; Jennings and Steinberg 1994; Rochelle-Newall and Fisher 2002; Wada et al. 2007; Helms et al. 2008; Hulatt et al. 2009; Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; Davies-Colley and Vant 1987; Zhang et al.

2009; Du et al. 2010). The humic substances (fulvic and humic acids) of allochthonous origin and autochthonous fulvic acids of algal origin are colored as they absorb visible light. They can thus be denoted both as colored and as chromophoric DOM (CDOM). Other terminology includes *yellow substances* or *Gelbstoff* in natural waters. On the other hand, low molecular weight CDOM also shows absorption at shorter wavelengths, such as formaldehyde at 207–250 nm, acetaldehyde at 208–224 nm, acetate at 204–270 nm, glyoxal at <240 nm, malonate at 225–240 nm, and pyruvate at 200–227 nm (Strome and Miller 1978; Mopper and Stahovec 1986; Kieber et al. 1990; Mopper et al. 1991; Wetzel et al. 1995; Dahlén et al. 1996). These LMW organic substances do not show any color as they do not absorb in the visible, and they can thus be denoted as CDOM but not as colored DOM.

Different variables such as the occurrence of allochthonous fulvic and humic acids and/or autochthonous fulvic acids, particulate materials (e.g. algae), chlorophyll *a* or phytoplankton, as well as incident light wavelengths and solar zenith angle, snow, ice, and water itself are responsible for the absorption and scattering of UV and PAR radiation, which may affect deeper waters in lakes and oceans (Smith and Baker 1981; Kirk 1984, 1991, 1994; Carder et al. 1989; McKnight et al. 1994; Scully and Lean 1994; Morris et al. 1995; Vernet and Whitehead 1996; Laurion et al. 1997; Sommaruga and Psenner 1997; Kahru and Mitchell 1998; Laurion et al. 2000; Markager and Vincent 2000; Belzile et al. 2000; Shank et al. 2005; Warren et al. 2006; Green et al. 2008; Hayakawa and Sugiyama 2008; Zhao et al. 2009; Shank et al. 2010).

CDOM absorption properties are significantly affected by several factors: DOM content and its chemical nature (Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; McKnight et al. 1994; Dubach et al. 1964; Zanardi-Lamardo et al. 2004; Singh et al. 2010; Singh et al. 2010; Minero et al. 2007; Minero et al. 2007; Vione et al. 2010), occurrence and types of suspended particulate matter (Zhang et al. 2009; Dupouy et al. 2010; Uusikivi et al. 2010; Gregg and Casey 2009; O'Donnell et al. 2010), photoinduced and microbial processes in freshwater and marine environments (Shank et al. 2010; Boehme and Coble 2000; Moran et al. 2000; Winch et al. 2002; Hernes and Benner 2003; Pullin et al. 2004; Scully et al. 2004; Winter et al. 2007; Zhang et al. 2007; Zhang and Qin 2007; Ma and Green 2004), and salinity (Singh et al. 2010; Singh et al. 2010; Hernes and Benner 2003; del Vecchio and Blough 2004; Blough and del Vecchio 2002; Fournier 2007). The CDOM plays several key roles in biogeochemical processes such as photoinduced reactions and biological processes, and it minimizes the deleterious consequences of UV radiation exposure for phytoplankton and other organisms in natural waters (Wetzel et al. 1995; Schindler et al. 1996; Williamson et al. 1996; Yan et al. 1996; Moran and Zepp 1997; Thomas-Smith and Blough 2001; Mostofa and Sakugawa 2009). Several reviews have been carried out on CDOM including its optical and chemical properties, sinks and distribution, relationship of CDOM with salinity, DOC and fluorescence, photochemistry, ocean color, and finally remote sensing applications in natural waters (Coble 2007; del Vecchio and Blough 2004; Blough and del Vecchio 2002; del Castillo 2005).

This chapter will give a general overview on the sources and nature of colored and CDOM, biogeochemical functions of CDOM, optical variables, chromophores in the CDOM, and the theory of CDOM absorbance in natural waters. Moreover, the controlling factors of CDOM that are most important in natural waters will be discussed. Two key factors such as photoinduced and microbial changes in CDOM are also discussed. Finally, a discussion is devoted to the importance of CDOM in natural waters.

1.1 Key Biogeochemical Functions of CDOM

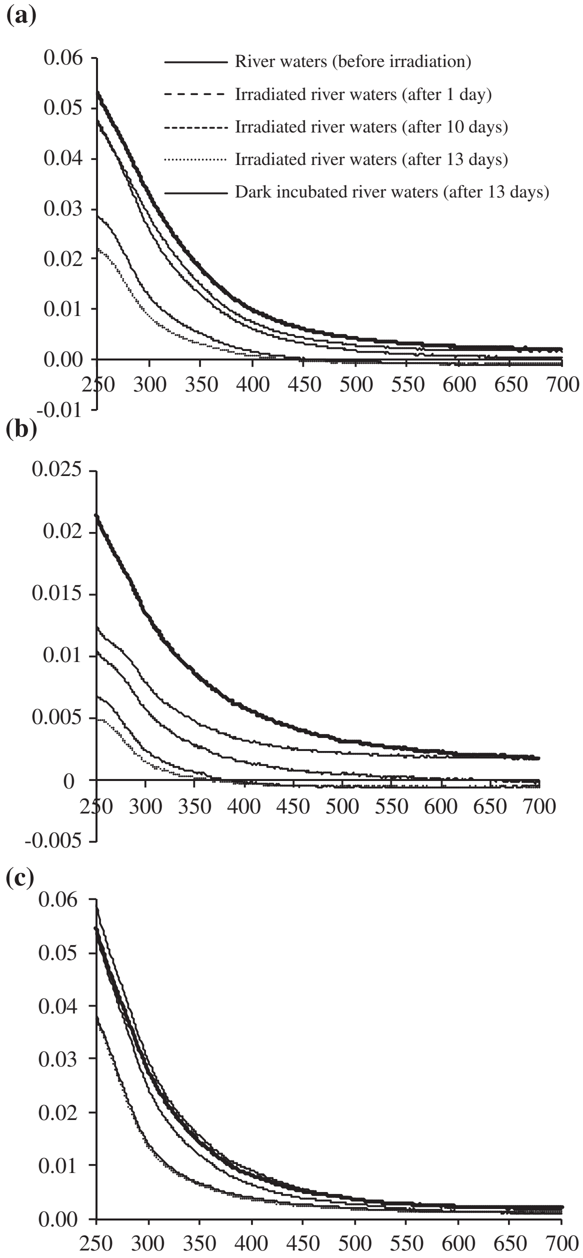
The biogeochemical functions of DOM (discussed in chapter "Dissolved Organic Matter in Natural Waters") have many similarities with those of CDOM, because DOM contains both colored and chromophoric DOM. The key biogeochemical functions of CDOM associated with natural waters are reported as follows. (i) Distribution of colored DOM in the ocean could be a useful indicator for better understanding DOC contents and distribution (del Vecchio and Blough 2004; Hayase et al. 1987; Vodacek et al. 1995; Ferrari et al. 1996; Stabenau and Zika 2004; Prahl and Coble 1994), salinity distribution (Laane and Kramer 1990; Dorsch and Bidleman 1982; Willey and Atkinson 1982; Carder et al. 1993), water mass mixing (Hujerslev et al. 1996; Nieke et al. 1997) and pollutant dispersion (Laane and Kramer 1990). (ii) Colored DOM (mostly allochthonous fulvic and humic acids, and autocthonous fulvic acids) is responsible for the photoinduced generation of hydrogen peroxide (H_2O_2) and organic peroxides (ROOH) and for initiation of photo-oxidation processes in natural waters upon sunlight absorption (Mostofa and Sakugawa 2009; Moore et al. 1993; Vione et al. 2006; Richard et al. 2007; al Housari et al. 2010; Clark et al. 2009; Minella et al. 2011). (iii) Colored DOM is partially responsible for the photoinduced generation of hydroxyl radicals ($HO^•$), strong oxidizing agents that may decompose the colored CDOM itself and other organic substances in aqueous media (Minero et al. 2007; Vione et al. 2006). The production of $HO^•$ by CDOM could take place upon water oxidation by the triplet states $^3CDOM^*$ or through formation of H_2O_2, either upon direct photoinduced dissociation ($H_2O_2 + h\upsilon \rightarrow 2HO^•$) or through Fenton or photo-Fenton decomposition (Zepp et al. 1992; Legrini et al. 1993; Von Sonntag et al. 1993; Takeda et al. 2004; Nakatani et al. 2007). (iv) The absorption of light by chromophores in CDOM initiates several important processes in natural waters such as the release of heat (Kirk 1988), the production of luminescence (Coble 1996; Vodacek et al. 1995; del Castillo and Miller 2008; Blough and Green 1995) and the formation of numerous photo products (Ma and Green 2004; Moran and Zepp 1997; Corin et al. 1996; Miller 1998; Blough and Zepp 1995). (v) Photodegradation of CDOM in surface waters results in the direct loss of absorption and fluorescence that lead to a decrease in the number of the chromophores of CDOM, thereby causing a decline of CDOM's absorption properties and photoinduced nature (Moran et al. 2000; Skoog et al. 1996; Reche et al. 1999; Whitehead and Vernet 2000; Twardowski and Donaghay 2001; del Vecchio and Blough 2002; Mostofa et al.

2007; Mostofa et al. 2007; Patsayeva et al. 1991; Kouassi and Zika 1990; Kouassi et al. 1990). (vi) CDOM absorbs light of both ultraviolet and visible wavelengths, which reduces the amount of PAR for photosynthesis (Bricaud et al. 1981; Arrigo and Brown 1996; Kirk 1994; Kalle 1966) and influences ocean color as determined by remote sensing (del Castillo and Miller 2008; Carder et al. 1991; Carder et al. 1999; Hoge et al. 1995; Hoge et al. 2001). (vii) Phytoplankton usually absorbs strongly in the blue and weakly in the green part of the spectrum. This is applied for the estimation of the chlorophyll *a* concentration within the upper layer of the water column, followed by empirical algorithms of ocean color based on blue-to-green ratios (McClain et al. 2004; Coble et al. 1998; O'Reilly et al. 1998). (viii) CDOM absorbance could be used for determining the chemical composition and structure of CDOM (González-Vila et al. 2001; Hedges et al. 2000; Osburn et al. 2001; Stabenau et al. 2004; Bracchini et al. 2010). (ix) The spectral slope (S) is often used as a proxy for CDOM composition, including the ratio of fulvic to humic acids and molecular weight (Twardowski et al. 2004). Salinity is negatively related to the spectral slopes from 275 to 295 nm, which might be considered a good proxy for DOM molecular weight (Ortega-Retuerta et al. 2010). (x) Finally, CDOM is responsible for the global carbon cycle and for biogeochemical processes through production, distribution, transport and decomposition of organic compounds in natural waters (Moran et al. 2000; Ma and Green 2004; Mostofa et al. 2007; Hedges et al. 2000; Hedges 1992; Amon and Benner 1994; Takahashi et al. 1995; Vähätalo et al. 2000; Ogawa and Tanoue 2003; Medina-Sánchez et al. 2006).

2 CDOM in Natural Waters

Colored and chromophoric dissolved organic matter (CDOM) is defined as a optically active component of bulk DOM of both allochthonous and autochthonous origin that absorbs light over a broad range of ultra-violet (UV) and visible wavelengths (200–800 nm), exhibiting absorbance spectra that generally decrease with increasing wavelength for freshwater and marine water (Fig. 1) (Bricaud et al. 1981; Arrigo and Brown 1996; Nelson et al. 1998; Warnock et al. 1999; Nelson and Siegel 2002; Vähätalo and Wetzel 2004; Coble 2007; Zhang et al. 2009). The absorbance of CDOM is useful for the knowledge of the contents of the materials present and for the identification of absorbance changes due to physical, photoinduced and biological processes in a variety of waters (Vähätalo and Wetzel 2004; Coble 2007; Vodacek et al. 1997; del Vecchio and Blough 2004; del Vecchio and Blough 2002; Vähätalo et al. 2000). It has been reported that there are differences in levels and optical properties between freshwater and marine water CDOM. Extreme enrichment in CDOM is usually observed in freshwater environments (Fig. 1) (Vähätalo and Wetzel 2004; del Vecchio and Blough 2004; del Vecchio and Blough 2002; Conmy et al. 2004; Kowalczuk et al. 2003). CDOM shows absorption at wavelengths 450–800 nm in freshwater (Kowalczuk et al. 2003), but usually not in marine waters. Carbon-specific absorption coefficients for riverine

Fig. 1 Absorption spectra of upstream CDOM (**a**, **b** Nishi-Mattaya and Higashi-Mataya, Lake Biwa watershed, Japan), and downstream CDOM (**c** Yasu River, Lake Biwa watershed, Japan). *Data source* Mostofa KMG et al. (unpublished data)



sources of CDOM are 10–150 times higher than those of marine CDOM, whilst coastal waters that receive riverine inputs typically possess higher CDOM absorption coefficients than open ocean waters (Arrigo and Brown 1996; Vähätalo and Wetzel 2004; Carder et al. 1989; del Vecchio and Blough 2004; del Vecchio and Blough 2002; Kowalczuk et al. 2003; Green and Blough 1994). This might be due to the large quantities of humic substances in freshwaters, which usually absorb radiation at $\lambda > 450$ nm. However, riverine input of chromophores contained in freshwater CDOM to the coastal marine environment are decomposed by photodegradation in the coastal area (Vähätalo and Wetzel 2004; del Vecchio and Blough 2002; Vähätalo et al. 2000). The combination of photodegradation and dilution prevents large amount of freshwater CDOM to reach the open ocean. Autochthonous CDOM usually increases relative to chlorophyll *a* concentration in the surface waters of lakes and oceans (Zhang et al. 2009; Sasaki et al. 2005; Mostofa et al. 2011). It is therefore suggested that phytoplankton degradation begins after the spring bloom in surface waters.

2.1 Sources and Nature of CDOM in Natural Waters

CDOM is generally originated from two key sources, allochthonous and autochthonous. The key sources of allochthonous DOM that includes fulvic and humic acids (humic substances) are vascular plant material and particulate detrital pools in terrestrial soil ecosystems. On the other hand, the key contributors for autochthonous DOM in natural waters and in sediment pore waters are phytoplankton or algal biomass, bacteria, coral, coral reef, submerged aquatic vegetation, seagrass and marsh- and mangrove forest. The sources and nature of CDOM are very similar to those of DOM, which have been explained in detail in chapter "Dissolved Organic Matter in Natural Waters".

2.2 Chromophores in CDOM

A chromophore is defined as a part of an organic molecule (functional group) with or without electron-donating heteroatoms such as N, O, and S, as a functional group with a highly unsaturated aliphatic carbon chain, or as a molecule with a structure that can hold up an electron or has extensive π-electron systems, which can absorb photons (light energy) with significant efficiency, causing the promotion from the ground state to an excited one. The key chromophores in a molecule or DOM in natural waters are $–OH$, $–CH{=}O$, $–C{=}O$, $–COOH$, $–COOCH_3$, $–OCH_3$, $–NH_2$, $–NH–$, $–CH{=}CH–$, $–CH{=}CH–COOH$, $–OCH_3$, $–(NH_2)CH–COOH$, S-, O- or N-containing aromatic compounds, Schiff-base derivatives ($–N{=}C–C{=}C–N–$) and so on (Mostofa et al. 2009a; Malcolm 1985; Strome and Miller 1978; Mopper and Stahovec 1986; Kieber et al. 1990; Mopper et al. 1991; Wetzel et al. 1995; Mostofa and Sakugawa 2009; Corin et al. 1996; Senesi 1990; Leenheer and Croue 2003; Peña-Méndez et al. 2005; Seitzinger et al. 2005; Zhang et al. 2005). Fulvic and humic acids of vascular plant origin have a molecular structure containing carboxy- and methoxy- benzenes and phenolic groups, carboxyl groups, alcoholic OH, carbohydrate OH, –C=C–, hydroxycoumarin-like structures, chromone, xanthone, quinoline, and O-, N-, S- and P-containing functional groups. They include aromatic carbon (17–30) and aliphatic carbon (47–63 %) (Malcolm 1985; Senesi 1990; Leenheer and Croue 2003; Steelink 2002). All of these functional groups can be considered as key chromophores in fulvic and humic acids in natural waters.

2.3 Theory of CDOM Absorbance

Photon absorption by a CDOM chromophore in aqueous solution firstly induces the excitation of an electron from its ground state to an excited one (Fig. 2), (Mostofa et al. 2009; Senesi 1990). Three types of electronic transitions occur with the CDOM chromophores in natural waters due to the absorption of UV or visible radiation: (i) transitions involving π, σ, and *n* electrons; (ii) transitions involving charge-transfer electrons, and (iii) transitions involving *d*- and *f*- orbital electrons in metals. The CDOM chromophores (e.g. fulvic acid, humic acid and tryptophan) mostly undergo transitions involving *n* or π electrons to the π^* excited state or charge-transfer electrons, and excitation of unpaired electrons in *d*- and *f*- orbitals (e.g. fulvic acid complexes with transition metals such as Cu(II) and Fe(III), having unpaired electrons) (Senesi 1990; Schulman 1985; Voelker and Sulzberger 1996; Senesi 1990; Fox 1990; Morales et al. 1997; Grabowski et al. 2003). The chromophores of CDOM absorb radiation in the wavelength range 200–700 nm and control the penetration along the water column of UV-B (280–320 nm), UV-A (320–400 nm) and total UV radiation (280–400 nm), as

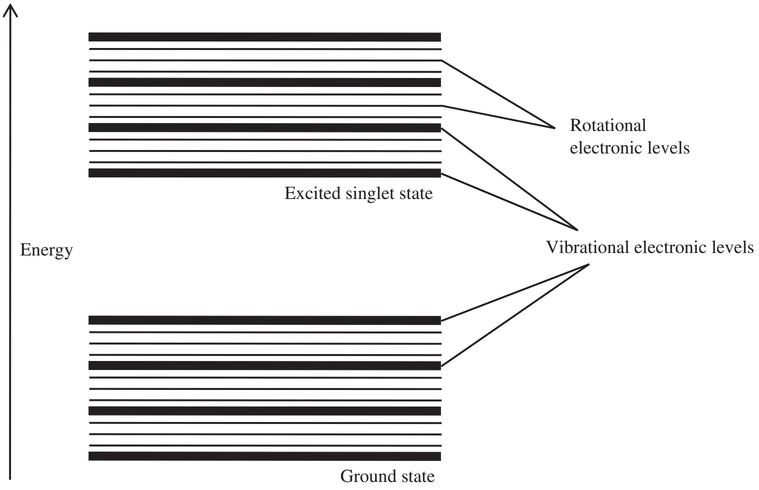


Fig. 2 A schematic energy level diagram for an organic molecule showing their rotational and vibrational electronic levels

well as of photosynthetically active radiation (PAR, 400–700 nm). CDOM absorbance in aqueous solution is directly proportional to the path length (l) and the concentration of the absorbing CDOM species (c). The *Beer's Law* states that $A = \boldsymbol{\varepsilon l c}$, where $\boldsymbol{\varepsilon}$ is a constant of proportionality called the molar absorption coefficient. The attenuation of radiation in solution increases with increasing CDOM absorbance.

As far as PAR radiation is concerned, CDOM in natural waters absorbs maximally in the blue region of the spectrum and its absorption decreases exponentially with increasing wavelength. CDOM spectra have been fitted to an exponential function by Bricaud and his co-workers (Bricaud et al. 1981) as described below (Eq. 2.1):

$$a_{\text{CDOM}}(\lambda) = a_{\text{CDOM}}(\lambda_{\text{o}})\text{e}^{-S(\lambda-\lambda\text{o})} \tag{2.1}$$

where $a_{\text{CDOM}}(\lambda)$ and $a_{\text{CDOM}}(\lambda_{\text{o}})$ are the absorption coefficients at a specific wavelength (λ) and at a reference wavelength (λ_{o}), respectively. S is the slope of the absorption spectrum, determined using either a nonlinear least-squares fitting routine over the wavelength range 290–700 nm or a linear least-squares fit of the log-transformed absorption data, over the range from 290 nm to the instrument detection limit (Vodacek et al. 1997). It is shown that the S value is significantly affected by the presence of different organic substances. Humic acids have a lower S value than fulvic acids (Zepp and Schlotzhauer 1981; Carder et al. 1989; Markager and Vincent 2000. Jerlov (1968) suggested an S value of 15 μm^{-1} whilst Bricaud et al. (Bricaud et al. 1981) showed S to vary between 10 and 20 μm^{-1} with a mean value of 14 μm^{-1}. The absorption coefficient is determined using the following equation

$$a_{\text{CDOM}}(\lambda) = 2.303 \times A_{\text{CDOM}}(\lambda)/l \tag{2.2}$$

where $A_{\text{CDOM}}(\lambda)$ is the absorbance ($\log I_0/I$), and l is the path length (in meters). The spectral slope of the absorption spectrum is widely used in remote sensing in coastal and marine environments (Vodacek et al. 1995; Hoge et al. 1995).

2.4 Absorption Coefficients of CDOM and Particulate Matter in Natural Waters

CDOM of both autochthonous and allochthonous origin generally exhibits UV and visible absorption spectra with low absorbance at longer wavelengths, and the absorbance increases with decreasing wavelength from 700 to 200 nm (Fig. 1) (Rochelle-Newall and Fisher 2002; Wada et al. 2007; Helms et al. 2008; Hulatt et al. 2009; Hayase and Tsubota 1985; Davies-Colley and Vant 1987; Zhang et al. 2009; Du et al. 2010; Odriozola et al. 2007; Bricaud et al. 2010). Allochthonous fulvic acids generally exhibit monotonous spectra whilst allochthonous humic acids have a shoulder around 400 nm in aqueous media (Fig. 3) (Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985). Dissolved mycosporine based amino acids (MAAs), released by dinoflagellates,

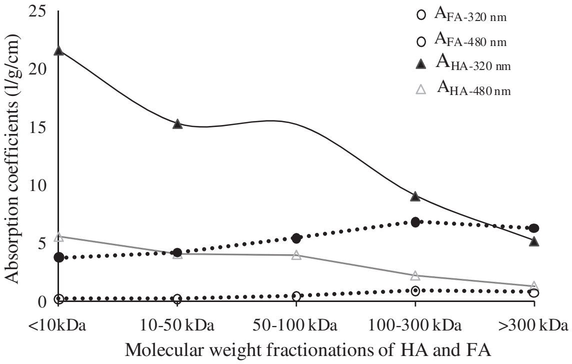


Fig. 3 Relationship between absorption coefficients examined at 320 and 480 nm and molecular weight fractionations of extracted humic acid (HA) and fulvic acid (FA) in sediment samples collected from Tokyo Bay using Amicon Diaflo Ultrafilters. Figure produced from *Data source* Hayase and Tsubota (1985)

may have a large influence on the absorption of UV light through the water column (Vernet and Whitehead 1996; Kahru and Mitchell 1998; Whitehead and Vernet 2000; Vernet et al. 1989). The MAAs are frequently identified in a wide phyletic assortment of marine organisms and can provide partial photoprotection from UV radiation (Karentz et al. 1991; Bandaranayake 1998; Jeffrey et al. 1999). It is reported that *Lingulodinium polyedrum* (formerly known as *Gonyaulax polyedra* (Dodge 1989)) produces seven types of MAAs with absorption maxima ranging between 310 and 364 nm (Vernet and Whitehead 1996). It is shown that radiation absorption by dissolved MAAs, assessed from the measured MAAs concentrations, significantly correlates with DOM UV absorption ($r^2 = 0.77$) and accounts for up to 10 % of the total DOM absorption at 330 nm (Whitehead and Vernet 2000). The chlorophyll-specific absorption coefficient for phytoplankton is high during the summer season and low during the spring bloom season. This might be the consequence of the occurrence of large diatoms during the spring bloom, while small phytoplankton dominates during the summer period (Sasaki et al. 2005). In addition, the autochthonous CDOM originated from algal or phytoplankton biomass shows strong absorption at 250–700 nm (Zhang et al. 2009; Jennings and Steinberg 1994; Rochelle-Newall and Fisher 2002; Wada et al. 2007; Helms et al. 2008; Hulatt et al. 2009). A study of Bay waters has shown that the relative contribution of CDOM absorption to the total absorption coefficient is >50 % (Sasaki et al. 2005).

Particulate material (phytoplankton or algae and detritus) have absorption spectra in the UV and visible region with several shoulders (Fig. 4a) (Zhang et al. 2009; Odriozola et al. 2007; Smith et al. 2004; Vantrepotte et al. 2007). Phytoplankton shows a strong absorption around 440 and a relatively weak absorption around 680 nm in aqueous media (Fig. 4a), (Zhang et al. 2009). The absorption coefficients of phytoplankton at 440 nm are significantly higher (63–88 %) than those of detritus studied during dry and wet seasons in seawater (Odriozola et al. 2007).

Blue light attenuation in the water column is often dominated by CDOM rather than by phytoplankton absorption. For instance, average CDOM absorption in

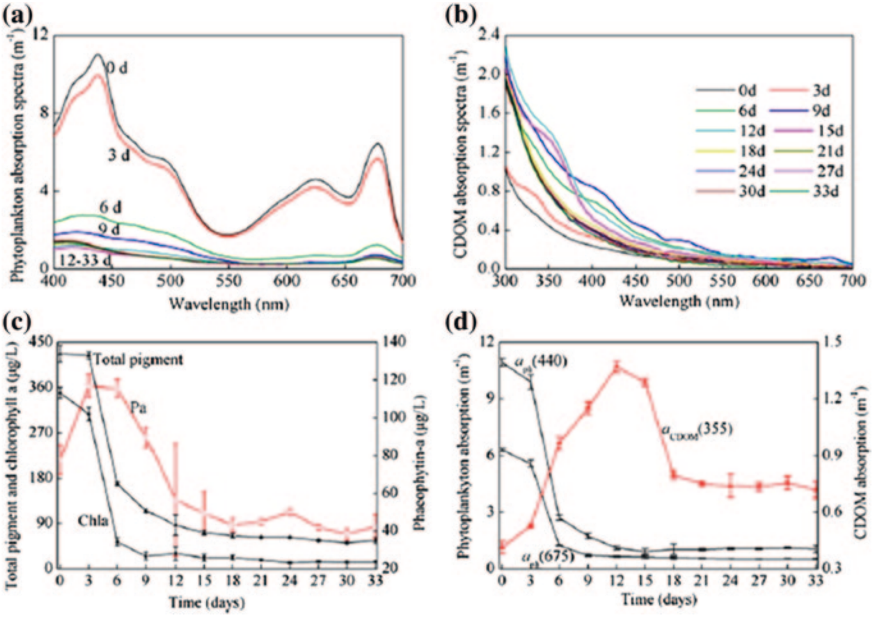


Fig. 4 Changes in the mean absorption spectra (n = 3) of (**a**) phytoplankton pigment absorption [$a_{\mathrm{ph}}(\lambda)$], (**b**) CDOM absorption [$a_{\mathrm{CDOM}}(\lambda)$], (**c**) The concentrations of total pigment, chlorophyll *a* and Phaeophytin-*a* (P_a, and (**d**) phytoplankton pigment absorption at the Chl *a* absorption maxima at 440 and 675 nm, and CDOM absorption at 355 nm during the degradation experiment period (0–33 days). Error bar indicates the means and standard deviations (n = 3). *Data source* Zhang et al. (2009)

Tampa Bay at 443 nm is about five times higher in June and about ten times higher in October than phytoplankton pigment absorption at 443 nm (Chen et al. 2007). It has been shown that the impact of the suspended and dissolved matter on lake-water optical quality is influenced by wind resuspension of particulate matter: the relative role of dissolved matter in the absorption of UV and visible radiation prevails for low wind speed conditions (<2.2 m s^{-1}), while at high wind speed (3.9 m s^{-1}) particulate matter resuspension can strongly influence the attenuation and the extinction measurements (Bracchini et al. 2005).

3 Apparent and Inherent Optical Properties of Natural Waters on Solar Altitude

The nature of the light field in a water body under a given incident solar irradiance is a function of inherent optical properties such as the absorption coefficient, the scattering coefficient and the normalized volume scattering function (which characterizes the angular distribution of single-event scattering). Sometimes the volume scattering function is referred to as the scattering phase function of the aquatic medium (Preisendorfer 1961; Kirk 1991). Optical properties of water at any given point in the medium are dependent on the irradiance distribution at that point (Preisendorfer 1961). The apparent optical properties of water bodies (e.g. vertical attenuation coefficient, irradiance and reflectance) are significantly dependent on the inherent optical properties (e.g. absorption, scattering and backscattering coefficients), which depend on the shape of the volume scattering function (Kirk 1984, 1991, 1994; Belzile et al. 2002). Kirk (1984, 1991, 1994) derived an empirical relationship between the apparent and inherent optical properties of natural waters, depending on the angle of incidence of the photons on the surface, based on field observation data followed by Monte Carlo modeling of the underwater light field. He stated that the vertical attenuation coefficient for downward irradiance at the midpoint of the euphotic zone, K_d, can be expressed as a function of the absorption coefficient, the scattering coefficient, and the solar altitude in accordance with (Eq. 2.3):

$$K_d = \frac{1}{\mu_0}\left[a^2 + G(\mu_0)\, ab\right]^{1/2} \tag{2.3}$$

where a is the absorption coefficient, b is the scattering coefficient, μ_0 is the cosine of the zenith angle of refracted solar photons (direct beam) just beneath the surface (calculated from the incident zenith angle using Snell's Law). $G(\mu_0)$ is a coefficient function that specifies the relative contribution of scattering to the vertical attenuation of irradiance. Its value is determined by the shape of the volume scattering function $\beta(\theta)$ and by μ_0. $G(\mu_0)$ is a linear function of μ_0 that can be expressed as (Eq. 2.4):

$$G(\mu_0) = g_1\mu_0 - g_2 \tag{2.4}$$

where g_1 and g_2 are numerical constants that depend on the volume scattering function used in the calculations.

In case of irradiance reflectance, Gordon et al. (1975) fit their data to another power series that has been simplified by Jerlov (1976) to (Eq. 2.5):

$$R(0) = \text{Constant}\,\frac{b_b}{a + b_b} \tag{2.5}$$

where $R(0)$ is the irradiance reflectance just below the surface, b_b is the backscattering coefficient, and the constant has the value 0.32 for zenith sun and 0.37 for an overcast sky. The irradiance reflectance can be expressed as (Eq. 2.6) (Kirk 1981)

$$R(0) = 0.328 b_b/a \tag{2.6}$$

This equation is in good agreement with the result obtained by Prieur (1976) using a different calculation procedure (Eq. 2.7):

$$R(0) = 0.33 b_b/a \tag{2.7}$$

These results show that the irradiance reflectance just below the surface is proportional to the backscattering coefficient and inversely proportional to the absorption coefficient in accordance with (Eq. 2.8)

$$R\,(0)\ = C\,(\mu_0)\, b_b/a \tag{2.8}$$

where $C(\mu_0)$ is a function of the angular distribution of the incident light flux that is constant for a given angular distribution. Other studies also show similar results where the reflectance increases as solar altitude decreases, indicating that $C(\mu_0)$ is a function of μ_0 (Gordon et al. 1975; Kirk 1981). Kirk's equation has been widely used to determine *a* and *b* in earlier studies (Belzile et al. 2002; Weidemann and Bannister 1986; Shooter et al. 1998).

3.1 Optical Variables for the Attenuation of UV and Photosynthetically Available Radiation

The absorption and scattering coefficients of different optical variables to UV and PAR are identified in natural waters (Smith and Baker 1981, Kirk 1984, 1991, 1994; Carder et al. 1989; McKnight et al. 1994; Scully and Lean 1994; Morris et al. 1995; Vernet and Whitehead 1996; Laurion et al. 1997; Sommaruga and Psenner 1997; Kahru and Mitchell 1998; Laurion et al. 2000; Markager and Vincent 2000; Belzile et al. 2000; O'Donnell et al. 2010; Belzile et al. 2002; Pierson et al. 2008; Pérez et al. 2010; Morris 2009). Reflectance, scattering and absorption of light occur in any surface as a function of latitude. Once light penetrates the air–water interface, it can either be scattered or absorbed by the constituents present in natural waters. The various optical variables in natural waters can be discriminated as below: (1) Content of CDOM, (2) Nature and molecular weight fractions of DOM, (3) Absorptivity (a_{CDOM}) and fluorescence (F_{CDOM}) of CDOM at specific wavelengths, (4) Effect of variation in incident light wavelengths and solar zenith angle, (5) Particulate materials, (6) Chlorophyll *a* concentration, (7) Water, and (8) Ice in Arctic and Antarctic regions.

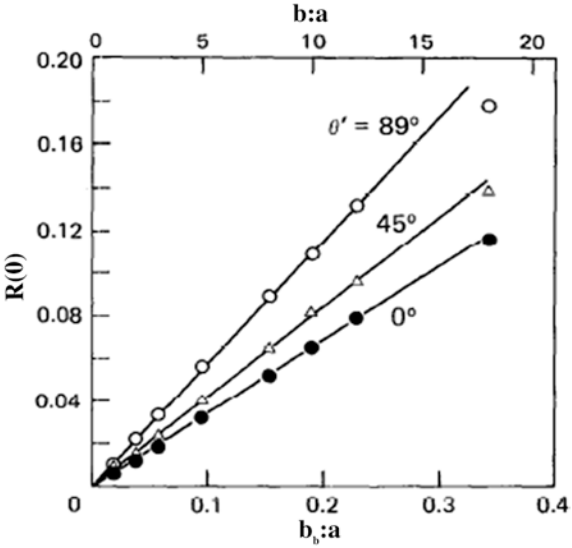
3.1.1 Contents of CDOM

CDOM is one of the key factors explaining the role of absorption and scattering to the attenuation of UV and photosynthetically available radiation in natural waters (Vodacek et al. 1997; Scully and Lean 1994; Morris et al. 1995; Laurion et al. 1997; Hayakawa and Sugiyama 2008; Smith et al. 2004; Belzile et al. 2002; Pierson et al. 2008; Smith and Baker 1979; Morris and Hargreaves 1997; Vincent et al. 1998; Pienitz and Vincent 2000; Kratzer et al. 2008; Devlin et al. 2009). DOM controls the downward irradiance flux through the water column of UV-B (280–320 nm), UV-A (320–400 nm), total UV (280–400 nm) and photosynthetically active radiation (PAR, 400–700 nm) (Markager and Vincent 2000; Morris and Hargreaves 1997; Tranvik and Kokalj 1998; Bertilsson and Tranvik 2000). Post bloom increases in DOM concentration from phytoplankton biomass cause increased DOM absorption, which in turn decreases the UV transmission through the water column (Whitehead and Vernet 2000). Mineralization of DOM by photoinduced and microbial processes can decompose the chromophores in CDOM, thereby reducing the absorption of UV and visible radiation. The consequence is an increase of UV transparency of surface waters (Nelson et al. 1998; Vodacek et al. 1997; Kieber et al. 1990; Moran et al. 2000; Skoog et al. 1996; Reche et al. 1999; Whitehead and Vernet 2000; Twardowski and Donaghay 2001; del Vecchio and Blough 2002; Mostofa et al. 2007; Mostofa et al. 2007; Patsayeva et al. 1991; Kouassi and Zika 1990; Kouassi et al. 1990; Morris and Hargreaves 1997; Allard et al. 1994; Zepp et al. 1995). CDOM accounts on average for 17–98 % of the total light attenuation coefficient in lake, estuarine and coastal seawaters (Odriozola et al. 2007; Lund-Hansen 2004; Obrador and Pretus 2008; Effler et al. 2010). CDOM shows variable and important contributions in summer (10–90 %) along the Patagonia shelf-break front (Ferreira et al. 2009). Absorption by CDOM and water together contributes to 88–94 % of UV radiation attenuation in tributaries, but only 37–77 % in lakes (Smith et al. 2004). These studies show that light attenuation by CDOM significantly depends on the occurrence of suspended particulate matter and phytoplankton with high contents of Chl *a* and water. The total content of DOC could be a useful estimate of UV transparency in natural surface waters.

3.1.2 Nature and Molecular Weight Fractions of CDOM

The absorption and scattering coefficients for UV and PAR are significantly dependent on the chemical nature and variability of organic compounds (Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; McKnight et al. 1994; Dubach et al. 1964; Zanardi-Lamardo et al. 2004; Singh et al. 2010). Fulvic and humic acids are typically a mixture of organic compounds with molecular weights ranging from <100 to >300,000 Daltons in natural waters (Hayase and Tsubota 1985; Ghassemi and Christman 1968; Thurman 1985; Ma and Ali 2009). Absorption coefficients of allochthonous fulvic and humic acids are significantly dependent on the molecular weight fractions (Fig. 3) (Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; Dubach et al. 1964; Ghassemi and Christman 1968). In addition, autochthonous CDOM of algal origin showed efficient absorption over the entire spectrum at 250–700 nm during 33 days of dark incubation (Fig. 4) (Zhang et al. 2009). This suggests that autochthonous CDOM has a similar absorption pattern as allochthonous fulvic acids. The absorption coefficients (l/g/cm) of fulvic acid extracted from sediment pore waters are much higher at 320 nm than at 480 nm in all the molecular weight fractions. However, absorption is low in low molecular weight fractions (<10 kDa) and generally increases with increasing molecular weight up to >300 kDa (Fig. 3) (Hayase and Tsubota 1985).

Fig. 5 Linear relationship between irradiance reflectance and the ratio of the backscattering coefficient (b_b) to the absorption coefficient at three different angles of incidence. *Data source* Kirk (1984)

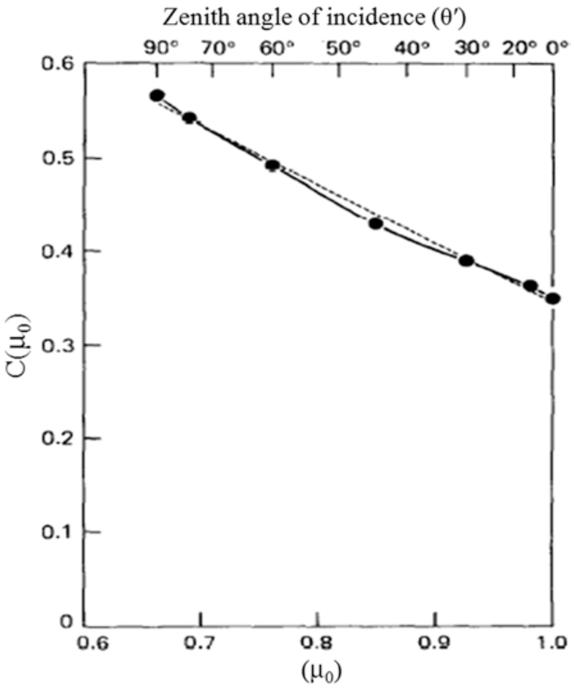


On the other hand, the absorption coefficients (l/g/cm) of whole humic acid extracted from sediment pore waters are much higher at 320 than at 480 nm, but absorption is much higher in low molecular weight fractions (<10 kDa) and typically decreases with increasing molecular weight up to >300 kDa (Fig. 3) (Hayase and Tsubota 1985). A small peak shoulder is observed for molecular weight fractions of humic acid of 50–100 kDa (Fig. 3), which might account for the shoulder detected in whole humic acid absorbance spectra (Ishiwatari 1973; Hayase and Tsubota 1985). Therefore, the absorption coefficient (l/g/cm) of humic acid decreases whilst that of fulvic acid increases with increasing molecular weight. Similarly, soil fulvic acid shows a parallel increase in absorption coefficients with molecular weight (Dubach et al. 1964). Such opposite trends of the absorption coefficients between fulvic and humic acids may result from the difference in their molecular structures (Figs. 5 and 6).

3.1.3 Absorption (a_{CDOM}) and Fluorescence (F_{CDOM}) of CDOM at Specific Wavelengths

The variations of the absorption (a_{CDOM}) and fluorescence (F_{CDOM}) of CDOM at specific wavelengths are significant depending on the nature and sources of CDOM in waters (Zhang et al. 2009; Coble 1996; Hayase and Tsubota 1985; Moran et al. 2000; Winter et al. 2007; del Vecchio and Blough 2004; del Vecchio and Blough 2002; Belzile et al. 2002; Mostofa KMG et al. unpublished data). Fluorescence spectroscopy has been applied to identify the fulvic and humic acids of allochthonous origin, autochthonous fulvic acids (also denoted as marine humic-like) of algal origin, aromatic amino acids (tryptophan, tyrosine and phenylalanine),

Fig. 6 Relationship between $C(\mu_0)$ with μ_0 with zenith angle of incidence (θ'). The continuous curve is drawn to join all the points where the broken line is the best straight line that can be drawn through the points. *Data source* Kirk (1984)



proteins and some anthropogenic compounds (such as fluorescent whitening agents and detergents) in natural waters (Coble 2007; Coble 1996; Yamashita and Tanoue 2003; Mostofa and Sakugawa 2009; Mostofa et al. 2010). These substances have fluorescence peaks at specific excitation-emission maxima, namely in the C-, A-, T- and T_{UV}-regions. Allochthounous fulvic and humic acids show absorption over the entire spectrum (250–700 nm), with a small peak shoulder for humic acid of 50–100 kDa (Fig. 3) (Ishiwatari 1973; Hayase and Tsubota 1985). The absorption coefficient (l/g/cm) of extracted humic acid typically decreases whilst that of fulvic acid increases with increasing molecular weight, as already reported. In addition, allochthonous fulvic acids of upstream and downstream waters generally show relatively high absorption at longer wavelengths (500–700 nm, see Fig. 1). Such absorption is either greatly decreased or not detectable in lake and marine waters (Moran et al. 2000; Winter et al. 2007; del Vecchio and Blough 2004; del Vecchio and Blough 2002; Belzile et al. 2002). Autochthonous CDOM of algal origin shows strong absorption over the entire 250–700 nm spectrum, and the fluorescence excitation-emission maxima resemble those of allochthonous fulvic acids in water (Fig. 4) (Zhang et al. 2009; Mostofa et al. 2009; Zhang et al. 2009). The absorption of both allochthonous and autochthonous fluorescent substances is very variable at specific wavelengths because these components can be significantly degraded by both photoinduced and microbial processes (Zhang et al. 2009; Moran et al. 2000; Winter et al. 2007; del Vecchio and Blough 2004; del Vecchio and Blough 2002; Mostofa et al. 2007;

Mostofa et al. 2010). As a consequence, the absorption of CDOM at specific wavelengths spectrum is found to considerably change for an array of waters. CDOM absorption contributed in average to 66 and 40 % of the UV diffuse attenuation coefficients in clear and turbid water, respectively (Belzile et al. 2002).

3.1.4 Effect of Variation in Incident Light Wavelengths and Solar Zenith Angle

The inherent and the apparent optical properties of natural waters are significantly dependent on the angle of the light flux incident on the water surface. They vary with solar altitude and with the proportion of diffuse and direct solar radiation (Kirk 1984, 1991, 1994; Morris 2009; Morel and Bélanger 2006). Remembering the a and b coefficients of Eqs. (2.5–2.8), when the solar zenith angle increases from 0° to 45° and finally to 89°, the ratio of the vertical attenuation coefficient to the absorption coefficient (K_d,$(z_\mathrm{m})/a$) increases by 15 and 41 %, respectively, when $b{:}a = 1$, by 8 and 22 % when $b{:}a = 5$, and by 5 and 12 % when $b{:}a = 10$ (Kirk 1984). K_d is thus rather insensitive to solar altitude in highly scattering waters (high b), but a considerable effect of solar altitude is observed in clear oceanic waters with low values of $b{:}a$. For all natural waters the shape of the volume scattering function is such that there is much more scattering in a forward than in a backward direction (Kirk 1984). When the incident beam moves away from the vertical, an increasing proportion of the more intense forward scattering becomes upward rather than downward scattering, thereby increasing the irradiance reflectance with decreasing solar altitude (increasing zenith angle) (Kirk 1984).

3.1.5 Suspended Particulate Matter

Absorption and scattering play a major role in UV and PAR attenuation by suspended particulate matter (SPM) such as phytoplankton pigments, algae, living heterotrophs, mineral sediments and detritus (organic, inorganic and mineral constituents) (Zhang et al. 2009, Kirk 1984, 1991, 1994; Laurion et al. 2000; Hayakawa and Sugiyama 2008; Dupouy et al. 2010; Uusikivi et al. 2010; Odriozola et al. 2007; Vantrepotte et al. 2007; Belzile et al. 2002; Pierson et al. 2008; Kratzer et al. 2008; Devlin et al. 2009; Hodoki and Watanabe 1998; Smith et al. 1999; Stambler 2005; Bowers and Binding 2006; Binding et al. 2008; Devlin et al. 2008; Foden et al. 2008). The spectral absorption coefficients of particulate matter (PM) are about twice higher in UV than in PAR wavelengths in the Baltic Sea ice (Uusikivi et al. 2010). Particulate absorption coefficients are appreciable in magnitude, averaging up to 103 % of a_CDOM at 380 nm and reflecting significant influence of both algal and detrital particles in lake and tributary waters (Smith et al. 2004). PM absorption spectra can include significant contribution by mycosporine-like amino acids between 320 and 345 nm (Uusikivi et al. 2010).

Experimental studies on phytoplankton biomass showed that phytoplankton has a strong absorption capacity at 400–700 nm and that its absorption decreases during 33 days of dark incubation (Fig. 4) (Zhang et al. 2009). An increase in UV absorption by PM may result from additional algal biomass during the phytoplankton bloom (Whitehead and Vernet 2000). The algal community composition in term of dominant cell size and, therefore, of pigment packaging is the key factor driving the phytoplankton specific absorption in the water column (Vantrepotte et al. 2007). The scattering coefficient of particulate materials increases approximately linearly with decreasing wavelength where suspended sediments dominate the optical signal in natural waters (Belzile et al. 2002; Morel 1988; Ahn et al. 1992; Roesler and Zaneveld 2258; Haltrin 1999; Pegau et al. 1999; Roesler 1998; Morel and Loisel 1998). The experimental and theoretical efficiency factor for scattering by the picocyanobacteria *Synechococcus* sp., *Synechocystis* sp. and *Anacystis marina* increases with decreasing wavelength (Ahn et al. 1992). However, an increase of the scattering efficiency with decreasing wavelength is not systematically detected in larger algal species (Ahn et al. 1992). The spectral slope coefficients (300–700 nm) of CDOM samples increase by as much as 20 % after mixing with 10 g L^{-1} sediment and by 5 % after mixing with 1 g L^{-1} sediment (Shank et al. 2005). This suggests that sorption to particles has the potential to significantly alter the optical properties of CDOM in the water column of turbid shallow environments or in areas of high benthic exchange (Shank et al. 2005).

Phytoplankton is the key driver of the spatial–temporal variations of the light attenuation coefficient, which accounts on average for 44 % of the total light attenuation (Obrador and Pretus 2008). The backscattering coefficient is highly correlated with turbidity and suspended matter ($R^2 = 0.98$), but it is poorly correlated to chlorophyll ($R^2 = 0.42$) (Dupouy et al. 2010), suggesting the importance of the inherent optical properties of PM in waters. The concentration-specific absorption coefficient of mineral particles is generally found to decrease exponentially with wavelength towards a constant non-zero value in the red (Bowers and Binding 2006). Specific scattering coefficients of mineral particles show a tendency to decrease from the open ocean into energetic shelf seas and estuaries, but then to increase again within shelf seas as turbulent energy increases (Bowers and Binding 2006). Light attenuation and scattering by particles can account for 11–52 % of the total attenuation/scattering in a variety of waters (Smith et al. 2004; Belzile et al. 2002; Lund-Hansen 2004; Smith et al. 1999). PM can contribute an estimated 25–90 % of the attenuation coefficients for the first-year sea ice at wavelengths <500 nm (Fritsen et al. 2011). The total particulate absorption coefficients at 300 nm are 0.1–0.3 m^{-1} in Southern Ocean waters (Holm-Hansen et al. 1993). Specific absorption coefficients for Antarctic phytoplankton is 0.1 m^2 (mg chl $a)^{-1}$ within the UV range (Mitchell et al. 1989; Arrigo 1994). Bacterial attenuation at 390 nm ranges from 0.002 m^{-1} for *Micrococcus* sp. to 2.80 m^{-1} for *Moraxella* sp. at concentrations of 1012 cells m^{-3}, and increases markedly at shorter wavelength (Kopelevich et al. 1987). These studies show that light attenuation by suspended particles is very variable depending on the water (clear or turbid) and the particle loading.

3.1.6 Total Content of Chlorophyll *a*

Chloropigments (chlorophyll *a* and carotenoids) could be important determinants of UV attenuation in natural waters (Zhang et al. 2009; Dupouy et al. 2010; Williamson et al. 1996; Belzile et al. 2002; Kratzer et al. 2008; Devlin et al. 2009; Lund-Hansen 2004; Morel and Bélanger 2006; Smith et al. 1999; Stambler 2005; Baker and Smith 1982; Gallegos and Bergstrom 2005). Chlorophyll *a* (Chl *a*) or phytoplankton biomass shows an absorption maximum around 440 nm (Fig. 4a, c, d) (Zhang et al. 2009; Bowers et al. 2000). Phytoplankton absorption is maximal when Chl *a* and phaeophytin-*a* are detected at the highest levels (Fig. 4a, c, d) (Zhang et al. 2009). Extraordinary spring blooms of the dinoflagellate *Prorocentrum minimum* can produce very high concentrations of chlorophyll, which increase for instance light attenuation in the upper Chesapeake Bay (Gallegos and Bergstrom 2005). Chlorophyll *a* specific absorption coefficients for both UV and PAR domains are representative of the dominant picophytoplankton in the Red Sea (Stambler 2005).

The study showed a deep chlorophyll maximum at about 50–60 m, with ~1 × 10^8 cells L^{-1} dominated by high concentrations of *Prochlorococcus* (~75 %), whereas in the Gulf of Eilat (Aqaba) ~4 × 10^7 cells L^{-1} have been reported. Eukaryotic algae (~20 %), cyanobacteria (*Synechococcus*) (~50 %) and *Prochlorococcus* (~25 %), are distributed throughout the water column in the Red Sea (Stambler 2005). Microbial degradation of phytoplankton or chlorophyll a are responsible for the decrease in Chl *a* or phytoplankton absorption in waters (Zhang et al. 2009). The share of light attenuation by phytoplankton is on average 32 % and reaches up to 74 % at high Chl *a* concentrations in estuarine-coastal waters (Lund-Hansen 2004). Phytoplankton absorption is the dominant optical component of light absorption (60–85 %) in spring along the Patagonia shelf-break front (Ferreira et al. 2009). Therefore the CDOM absorption is significantly dependent on the contents of phytoplankton or total contents of Chl *a* in natural waters.

3.1.7 Water

Light absorption by natural waters depends on the water quality (clear or turbid, presence of particulate matter, and CDOM content) (Kirk 1984; Hayakawa and Sugiyama 2008; Gregg and Casey 2009; Fournier 2007; Belzile et al. 2002; Pérez et al. 2010; Morris 2009; Lund-Hansen 2004; Effler et al. 2010). Recent studies show that water even in its purest form exhibits a complex absorption spectrum and a significant amount of scattering caused by refractive index fluctuations (Fournier 2007). The optical properties of natural waters are typically function of the underwater irradiance and of either the vertical attenuation coefficient for downward irradiance (K_d) or the irradiance reflectance. It is $R = E_u/E_d$, where E_u and E_d are the upward and downward irradiance at a given depth (Kirk 1984). These optical properties significantly depend on the nature of the light field within the water body and vary with depth and solar altitude (Kirk 1984; Belzile et al. 2002). The vertical attenuation coefficient (K_d PAR) has been found to vary from 0.40 to 47 m^{-1} in sixteen Argentinean shallow lakes. High K_d PAR values (>13 m^{-1}) have been detected in highly turbid lakes, medium K_d PAR values (<10 m^{-1}) in clear-vegetated lakes, and very low K_d PAR values in Patagonian lakes (<2.5 m^{-1}) (Pérez et al. 2010). Depending on the occurrence of key absorbance variables such as high CDOM, particulate material and chlorophyll, the absorption of water can vary considerably. Light attenuation by water contributes on average 0.3–9 % in UV and PAR, although it is highly variable between clear and turbid waters (Belzile et al. 2002; Lund-Hansen 2004).

3.1.8 Snow and Ice in Arctic and Antarctic Regions

Absorption and scattering by snow and ice significantly affect the UV and PAR attenuation, particularly in the Arctic and Antarctic region (Belzile et al. 2000; Warren et al. 2006; Grenfell and Perovich 1984; Buckley and Trodahl 1987; Perovich 1993; Trodahl and Buckley 1990; Arrigo et al. 1991; Perovich et al. 1998; Norman et al. 2011). Snow is a scattering-dominated medium, the scattering of which is independent of wavelength between 350 and 600 nm. The attenuation of solar radiation in snow can be used to infer the spectral absorption coefficient of pure ice, by reference to a known value at 600 nm (Warren et al. 2006). The spectral downwelling diffuse attenuation coefficient is caused by both scattering and absorption within the medium. Scattering by snow depends on grain size, snow density and water content, whilst scattering by ice depends on ice structure and particle back-scattering (Buckley and Trodahl 1987; Trodahl and Buckley 1990; Arrigo et al. 1991; Perovich et al. 1998).

It has been shown that the UV-B transmittance through 1.7 m–thick first-year ice decreases from 2–1 to 0.2–0.1 % from the end of October to mid-November in McMurdo Sound (Trodahl and Buckley 1990). The decrease in transmittance is the effect of the formation of a highly scattering layer, subsequent to ice-surface drainage. UV-B transmittance at 320 nm for 1.6 m of snow-covered first-year ice also decreases by an order of magnitude from 0.3 % in April to 0.03 % in June in the Chukchi Sea (Perovich et al. 1998). A bloom of ice algae at the bottom of the ice can also reduce the UV radiation transmittance (Perovich et al. 1998). Belzile et al. (2000) report that about 2–13 % of incident UV-B irradiance is transmitted through snow, ice and ice algae biomass, whilst transmittance increases to 5–19 % for UV-A and to 5–12 % for PAR. An influence of ice algae on PAR transmission is also observed (Belzile et al. 2000; Arrigo et al. 1991; Palmisano et al. 1987). The absorption of irradiance depends on the absoprtion by pure ice and brine, CDOM and particulate organic matter (POM) (Belzile et al. 2000; Uusikivi et al. 2010; Fritsen et al. 2011; Grenfell and Perovich 1984; Perovich et al. 1998; Norman et al. 2011; Warren et al. 1993). In Baltic Sea ice organic matter, both particulate and dissolved, influences the optical properties of sea ice and strongly modifies the UV radiation exposure of biological communities in and under snow-free sea ice (Uusikivi et al. 2010). Note that transmittance, $T(\lambda)$, is the ratio of the downwelling irradiance at the lower surface of the ice, $E_d(z_{ice}, \lambda)$, to the incident irradiance, $E_d(0, \lambda)$ (Perovich 1993). The transmittance depends on the spectral reflection coefficient, $\alpha(\lambda)$—that is, the fraction of $E_d(0, \lambda)$ that is reflected—and on the attenuation of irradiance by snow and ice, according to Beer's Law.

4 Factors Affecting Absorption of Radiation by CDOM

CDOM absorption differs considerably in a variety of natural waters and depends on several factors, which can be distinguished as follows: (i) Contents and molecular nature of CDOM; (ii) Occurrence and type of sediments; (iii) Photoinduced degradation; (iv) Microbial degradation; and (v) Salinity.

4.1.1 Contents and Molecular Nature of DOM

The CDOM absorption depends on total DOM contents and on its molecular nature (Fig. 7) (Vodacek et al. 1997; Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; McKnight et al. 1994; Dubach et al. 1964; del Vecchio and Blough 2004; Belzile et al. 2002; Vincent et al. 1998; Pienitz and Vincent 2000). DOC contents are very much correlated with CDOM absorption (a_{CDOM}) in natural waters, with the exception of surface waters during the summer stratification period (Rochelle-Newall and Fisher 2002; del Vecchio and Blough 2004; Vodacek et al. 1995; Ferrari et al. 1996; Ferrari 2000; Klinkhammer et al. 2000; Chen et al. 2002; Kowalczuk et al. 2010). The results typically suggest that the CDOM fraction often increases linearly with the DOC content, whilst the non-CDOM fraction of DOC remains relatively constant at approximately 50–100 μM C (Fig. 7). It has been shown that humic-like CDOM components with excitation maxima at longer wavelengths have significantly higher non-absorbing DOC compared to humic-like CDOM components with excitation maxima at shorter wavelengths (Kowalczuk et al. 2010). The relationship between the DOC concentration and the intensity of one of the protein-like components can result in significantly reduced non-absorbing DOC fraction (Kowalczuk et al. 2010). This study suggests that the relative proportion of humic-like CDOM components (characterized by excitation maximum at longer wavelengths) and the main protein-like component have the highest impact on the absorption at 350 nm (Kowalczuk et al. 2010). Moreover, two phenomena are responsible for the observed differences in CDOM absorption in surface waters. First, CDOM properties (chromophores in CDOM) are significantly altered by exposure to natural sunlight in surface waters, which reduces the CDOM absorption. Second, new CDOM is produced from algal or phytoplankton biomass under

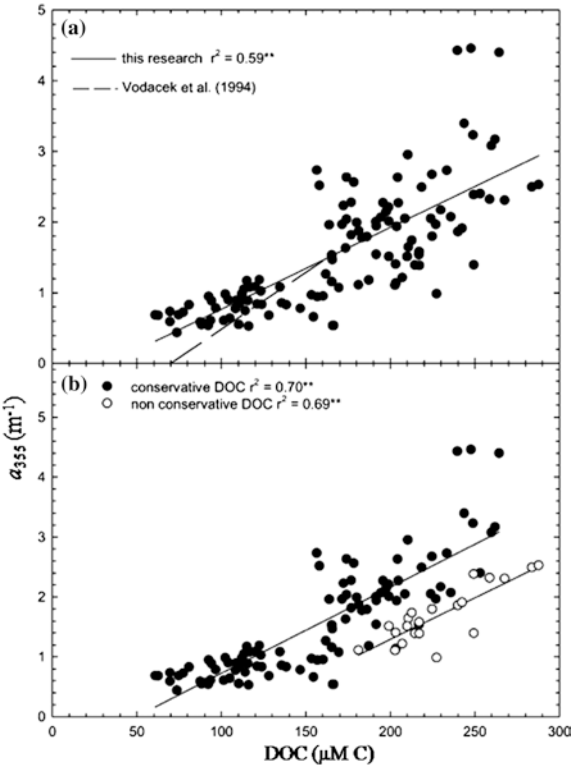


Fig. 7 Relationship between CDOM absorption (a_{355}, m^{-1}) and DOC concentrations in Chesapeake Bay. *Panel* (**a**) shows DOC and CDOM absorption for all cruises, and in *Panel* (**b**) DOC concentrations are separated into conservative and non-conservative groups. *Dashed line* on *Panel* (**a**) represents data of (Vodacek et al. 1995). *Data source* Rochelle-Newall and Fisher (2002)

both photoinduced and microbial assimilations during the summer stratification period in surface waters (Zhang et al. 2009; Rochelle-Newall and Fisher 2002; Yamashita and Tanoue 2004; Yamashita and Tanoue 2008; Mostofa et al. 2009; Rochelle-Newall 1999; Mostofa et al. 2005; Parlanti et al. 2000). This material does not show strong CDOM absorption. CDOM absorption is strongly dependent on the molecular nature of DOM (Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; Ghassemi and Christman 1968; Thurman 1985; Ma and Ali 2009), fulvic and humic acids depending on their sources and molecular weight are significantly different as far as radiation absorption is concerned.

4.2 *Occurrence and Nature of Sediments*

DOM composition depends significantly on the sources and nature of sediments, such as vascular plant material and algal biomass in soil or sediment pore waters (Zhang et al. 2009; Mostofa et al. 2009; Malcolm 1985; Zhao et al. 2009; Parlanti et al. 2000; Fu et al. 2010; Li W et al. unpublished data). Vascular plant material of terrestrial origin in waters or pore waters is mostly responsible for microbial production of allochthonous humic substances (fulvic and humic acids), carbohydrates, amino acids and so on (Mostofa et al. 2009; Malcolm 1985; Ittekkot et al. 1985; Guéguen et al. 2006). The terrestrial run-off through rivers may bring the plant material to many surface waters, such transport depending on precipitation and on the type and density of terrestrial plants. On the other hand, algal biomass and microbes which develop in the photic zone may release autochthonous fulvic acids, amino acids, carbohydrates, proteins, fatty acids, peptides, organic acids and other compounds (Zhang et al. 2009; McCarthy et al. 1996; Biddanda and Benner 1997; Wakeham et al. 1997; Rosenstock et al. 2005; Hama and Handa 1992). Sometimes such a process is photolytically enhanced. Major seasonal differences in the spectral slope values show that phytoplankton degradation is one of the important sources of CDOM in summer, whereas in other seasons CDOM mainly reaches lake water from river input (Zhang et al. 2009; Zhang and Qin 2007). At the same time, sediment-trap studies demonstrate that only 1–35 % of the organic carbon (viz. algae) synthesized in the photic zone reaches the sediment surface in marine and lacustrine waters (Bernasconi et al. 1997; Hernes et al. 2001; Lehmann et al. 2002). Such algal material releases the same autochthonous organic substances in pore waters as it does in the upper parts of the water column (Burdige et al. 2004; Li W et al., unpublished data). Some DOM components from the pore-water sediment surface can mix up with surface waters during the vertical mixing (overturn) period in lakes or oceans. Allochthonous and autochthous DOM has very variable absorption properties (Zhang et al. 2009; Vodacek et al. 1997; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; McKnight et al. 1994; del Vecchio and Blough 2004; Vodacek et al. 1995; Belzile et al. 2002; Vincent et al. 1998; Pienitz and Vincent 2000). Therefore, DOM composition also depends on the occurrence and nature of sediments in natural waters.

4.3 Photoinduced Degradation of CDOM in Natural Waters

Photoinduced processes can decompose the chromophores in CDOM and thus decrease the CDOM absorption (Vähätalo and Wetzel 2004; Zhang et al. 2009; Shank et al. 2010; Moran et al. 2000; Winter et al. 2007; del Vecchio and Blough 2002; Norman et al. 2011; Zagarese et al. 2001). A theoretical model for the photoinduced degradation of CDOM and its effects on absorption properties are discussed in this section.

4.3.1 Theoretical Model for Photoinduced Degradation of CDOM

The CDOM chromophores absorb photons and sometimes undergo degradation, while DOM undergoes partial mineralization to hydrogen peroxide (H_2O_2), CO_2, DIC (sum of dissolved CO_2, H_2CO_3, HCO_3^-, and CO_3^{2-}), COS, CO, ammonium, gaseous hydrocarbons, organic peroxides (ROOH), low molecular weight (LMW) DOM, and so on in upper surface waters (Fig. 1) (Vähätalo and Wetzel 2004; Ma and Green 2004; Moran and Zepp 1997; Mostofa and Sakugawa 2009;

del Vecchio and Blough 2002; Vähätalo et al. 2000; Bertilsson and Tranvik 2000; Allard et al. 1994; Amador et al. 1989; Fujiwara et al. 1995; Bertilsson and Allard 1996; Granéli et al. 1996; Granéli et al. 1998; Miller and Moran 1997; Clark et al. 2004; Xie et al. 2004; Johannessen et al. 2007; Fichot and Miller 2010). In surface waters, the rate of photoinduced mineralization of CDOM (pm_z, mol C m^{-3} d^{-1}), modified by Vähätalo et al. (2000) from Schwarzenbach et al. (1993) and Miller (1998), can be expressed as follows (Eq. 4.1):

$$pm_z = \int_{\lambda\text{min}}^{\lambda\text{max}} \varphi_\lambda Q_{\text{s,z},\lambda} a_{\text{CDOM},\lambda} \text{d}\lambda \tag{4.1}$$

where φ_λ is the spectrum of the apparent quantum yield for photoinduced mineralization (mol produced DIC/mol absorbed photons), $Q_{s,z,\lambda}$ is the scalar photon flux density spectrum at the depth z (also referred to as actinic flux, mol photons m^{-2} d^{-1}), and ${}_\lambda a_{\text{CDOM},\lambda}$ is the absorption spectrum of CDOM (m^{-1}). The parameters λ_{max} and λ_{min} are the minimum and maximum wavelengths contributing to photoinduced mineralization.

In the whole water column the rate of photoinduced mineralization, modified by Vähätalo et al. (2000) from Miller (1998), can be expressed as follows (Eq. 4.2):

$$pm = \int_{\lambda\text{min}}^{\lambda\text{max}} \varphi_\lambda Q_{\text{a},\lambda} (a_{\text{CDOM},\lambda}/a_{\text{tot},\lambda}) \text{d}\lambda \tag{4.2}$$

where $Q_{\text{a},\lambda}$ represents the photons absorbed by the water column (mol photons m^{-2} d^{-1}) and the $a_{\text{CDOM},\lambda}/a_{\text{tot},\lambda}$ ratio expresses how much CDOM contributes to the total absorption. In infinitely deep waters, $Q_{\text{a},\lambda}$ roughly equals the downward vector photon flux density just below the surface ($Q_{\text{d,v,0-}\lambda}$) (Sikorski and Zika 1993; Sikorski and Zika 1993).

The quantum yields related to CDOM decrease exponentially with increasing wavelength (Moran and Zepp 1997; Vähätalo et al. 2000; Sikorski and Zika 1993; Ratte et al. 1998; Gao and Zepp 1998. A generalized equation linking quantum yield and wavelength (Vähätalo et al. 2000) can be expressed as below (Eq. 4.3):

$$\varphi_\lambda = \text{c} \times 10^{-\text{d}\lambda} \tag{4.3}$$

where c (dimensionless) and d (nm^{-1}) are positive constants and λ is wavelength (nm). Different combinations of c and d can cover a wide range of exponential relationships between quantum yield and wavelength.

4.3.2 CDOM Absorption Loss in Long- and Short-Wavelengths Due to Photoinduced Degradation

Photoinduced degradation rapidly lowers the CDOM absorption coefficients across the entire spectrum, both in natural waters and in standard organic substances (Fig. 1) (Vodacek et al. 1997; Zhang et al. 2009; Shank et al. 2010; Moran et al. 2000; Hernes and Benner 2003; Winter et al. 2007; del Vecchio and Blough

2002; Ortega-Retuerta et al. 2010; Norman et al. 2011; Kowalczuk 1999; Kitidis et al. 2006). Absorption losses are likely different for a variety of natural waters (Table 1) (Zhang et al. 2009; Moran et al. 2000; Winter et al. 2007; Mostofa KMG et al., unpublished data; Norman et al. 2011). They are of order 77–97 % at 340–350 nm for upstream CDOM (Kago and Nishi-Mataya upstream, Japan) and 58–59 % at 340–350 nm for downstream CDOM (Yasu River, Japan) after 13 days of irradiation (Table 1) (Mostofa KMG et al. unpublished data). The CDOM absorption is entirely quenched at 700–444 nm for Kago upstream and 700–366 nm for Nishi-Mataya upstream, but downstream CDOM is little decomposed (14–45 %) at 600–700 nm (Fig. 1). Note that the upstream CDOM is mostly made up of fulvic acids having low DOC concentrations (99 and 38 μM C for NM upstream), whilst downstream CDOM has different origin such as autochthonous (protein-like or tryptophan-like), allochthonous (fulvic acids), and agricultural sources with relatively high levels of DOC (e.g., 194 μM C for Yasu River) (Mostofa et al. 2007; Mostofa et al. 2005). This suggests that autochthonous CDOM may originate in the river bed during the summer season and agricultural CDOM may be released from nearby agricultural fields.

The differences in CDOM absorption losses between upstream and downstream river waters suggest three issues. First, absorption losses depend on CDOM source and composition. Second, fulvic acids in upstream river waters are highly decomposed, as demonstrated by the complete loss of absorption in the longer wavelength region (from 366 to 700 nm). Such absorption losses are accompanied by high losses (72–84 % at peak C-region) in the fluorescence intensity of fulvic acids (Mostofa et al. 2007). Third, CDOM absorption losses are relatively limited in downstream river waters. A possible reason is that this CDOM may be a mixture os compounds originating from several sources such as autochthonous, allochthonous and agricultural. Because of the high decrease of the fluorescence intensity of allochthonous fulvic acids (80 % in downstream waters) and tryptophan (59 % in downstream waters) detected in earlier studies (Mostofa et al. 2007), it is suggested that the remaining autochthonous and agricultural CDOM might be recalcitrant or refractory to photoinduced degradation.

In addition, CDOM absorption losses are 55–76 % at 340 nm in the water of various lakes and ponds after 13 days irradiation (Table 1) (Winter et al. 2007). However, much lower absorption losses have been observed in the case of Lake Taihu after 12 days irradiation: 30 % at 355 nm and 21 % at 280 nm (Table 1) (Zhang et al. 2009). CDOM absorption losses are in the range of 50–64 % at 350 nm for estuarine CDOM after 70 days irradiation period (Table 1) (Moran et al. 2000). It is estimated that approximately 70 % of terrestrial CDOM is lost by photo-oxidation on the Middle Atlantic Bight shelf (Vodacek et al. 1997). In Antarctic surface waters, sea ice CDOM susceptibility to photo-bleaching in an in situ 120 h exposure showed a loss in CDOM absorption of 53 % at 280 nm, 58 % at 330 nm, and 30 % at 375 nm (Norman et al. 2011). This result suggests that Antarctic CDOM is more photosensitive than average lake or seawater CDOM. Absorption losses for standard Aldrich humic acid are 42–47 % at 340 nm in deionized water after 13 days irradiation (Table 1) (Winter et al. 2007).

Table 1 Changes in the specific absorbance coefficients of CDOM at different wavelength ranges by photoinduced degrdation on natural waters

Samples	Incubation time	Changes in the specific absorbance coefficients (SAC) at different wavelenths (%)		References
	(h or days)	Photoinduced degradation	Microbial degradation	
Rivers				(Mostofa KMG et al. unpublished data)
Upstream waters (Kago, Japan) at 340–350 nm	13	−(77–79)	+(20–22)	(Mostofa KMG et al. unpublished data)
Upstream waters (Kago, Japan) at 444–700 nm	13	−100	+(19–38)	(Mostofa KMG et al. unpublished data)
Upstream waters (Nishi-Mataya, Japan) at 340–350 nm	13	−(93–97)	+(78–81)	(Mostofa KMG et al. unpublished data)
Upstream waters (Nishi-Mataya, Japan) at 366–700 nm	13	−100	+(4–25)	(Mostofa KMG et al. unpublished data).
Downstream waters (Yasu River, Japan) at 340–350 nm	13	−(58–59)	−(6–7)	(Mostofa KMG et al. unpublished data)
Downstream waters (Yasu River, Japan) at 600–700 nm	13	−(14–45)	+(8–49)	(Mostofa KMG et al. unpublished data).
Lakes				
Lake Taihu, China at 355 nm	12	−30	nd	Zhang et al. (2009b)
Lake Taihu, China at 280 nm	12	−21	nd	Zhang et al. (2009b)
Lake Taihu, China at 280 nm	2–12 h (UV)	−(8–19)	nd	Zhang et al. (2009b)
Lake Erie water (42°N) at 340 nm	13	−(75–76)	−(12–32)	Winter et al. (2007)
Luther Marsh northeast water (43°N) at 340 nm	13	−(61–63)	+ (2–3)	Winter et al. (2007)
Mill Creek east of Cambridge water (43°N) at 340 nm	13	−(56–72)	−1, + 2	Winter et al. (2007)
Bannister Lake southwest of Cambridge water (43°N) at 340 nm	13	−(55–62)	+3, −3	Winter et al. (2007)
Sanctuary Pond water (41°N) at 340 nm	13	−(64–74)	+3, −4	Winter et al. (2007)
Aldrich humic acid in deionized water at 340 nm	13	−(42–47)	+(2–12)	Winter et al. (2007)
Estuaries and oceans				
Satilla Estuary (Georgia, USA) at 350 nm	70	−(59–64)	−(2–4)	Moran et al. (2000)
Satilla Estuary (Georgia, USA) at 350 nm	70	−(50–53)	+0.6, −7	Moran et al. (2000)
Satilla Estuary (Georgia, USA) at 250–500 nm	51	nd	−(4–11)	Moran et al. (2000)
Antarctic sea ice and oceanic water at 375 nm	120 h	−30	nd	Norman et al. (2011)
Antarctic sea ice and oceanic water at 330 nm	121 h	−58	nd	Norman et al. (2011)
Antarctic sea ice and oceanic water at 280 nm	122 h	−53	nd	Norman et al. (2011)

Irradiation time-hours (h) mentioned with each time as ‘h’ and ‘days’ mentioned as a whole digit only

nd Not detected

The decrease in CDOM absorption is relatively lower in the waters of Bay and oceans (Blough and del Vecchio 2002; del Vecchio and Blough 2002.

Generally speaking, CDOM absorption losses in lakes, estuaries, Bay and oceans are significantly lower than for upstream CDOM. The main reason might be the autochthonous sources of most of the CDOM in these waters, which make the corresponding CDOM less susceptible to photoinduced degradation. Indeed, the fraction of autochthonous CDOM is maximum (25–98 %) in lakes and oceans, whilst allochthonous humic substances (mostly fulvic acids) are 2–75 % (see also chapter "Dissolved Organic Matter in Natural Waters") (Mostofa et al. 2009; McCarthy et al. 1996; Biddanda and Benner 1997; Moran et al. 1991; Moran and Hodson 1994; Benner and Kaiser 2003). In addition, the amount of allochthonous CDOM (mostly fulvic and humic acids) is considerably decreased in the transport from rivers to lakes and oceans because of photoinduced decomposition by natural sunlight (Vähätalo and Wetzel 2004; Vodacek et al. 1997; Mopper et al. 1991; Wetzel et al. 1995; Moran et al. 2000; Skoog et al. 1996; Mostofa et al. 2007; Bertilsson and Tranvik 2000; Amon and Benner 1996; Twardowski and Donaghay 2002; Waiser and Robarts 2004; Wu et al. 2005; Brooks et al. 2007). The experimental results demonstrate the photoreactive nature of CDOM, with half-lives from 2.1 to 5.1 days due to photobleaching in the upper layer and duplication times from 4.9 to 15.7 days due to photohumification. Such results highlight the highly dynamic nature of CDOM in the Southern Ocean (Ortega-Retuerta et al. 2010). In addition, the high susceptibility to photobleaching of CDOM in Antarctic ice waters might be the effect of the presence of fresh CDOM in bulk ice samples, due to elevated in situ production (Norman et al. 2011). The fresh CDOM in Antarctic ice waters is characterized by low S and high a_{375}. In contrast, aged material present in brine and seawater samples is characterised by high S values and low a_{375} (Norman et al. 2011). Therefore, photoinduced degradation is one of the key factors that can regulate the CDOM absorption depending on its composition for a variety of natural waters.

4.3.3 Changes in Spectral Slope Due to Photoinduced Degradation

Photoinduced degradation can alter the spectral slope of CDOM (S) either in natural surface waters or in experimental observations under solar irradiation (Fig. 8) (Vodacek et al. 1997; Helms et al. 2008; Zhang et al. 2009; Shank et al. 2010; Moran et al. 2000; Zhang and Qin 2007; del Vecchio and Blough 2002; Mostofa KMG et al., unpublished data; Xie et al. 2004; Twardowski and Donaghay 2002; Tzortziou et al. 2007; Whitehead et al. 2000). Two key phenomena are generally observed: the first one is an increase of S because of CDOM photobleaching by solar radiation (Fig. 8) (Helms et al. 2008; Zhang et al. 2009; Shank et al. 2010; Moran et al. 2000; del Vecchio and Blough 2002; Xie et al. 2004; Twardowski and Donaghay 2002; Whitehead et al. 2000). It is suggested that photobleaching can be caused by the transformation of high-molecular weight CDOM complexes that absorb at longer wavelengths into smaller complexes that absorb at shorter wavelengths. The opposite effect can also be observed: in some

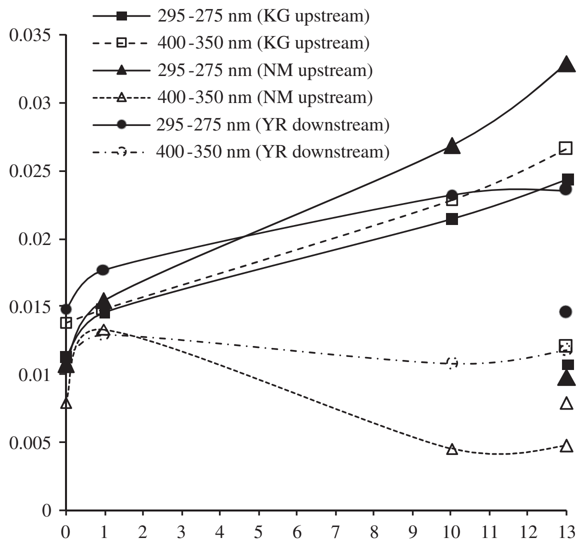


Fig. 8 Changes in the spectral slope values (*S*) in the upstream (Kago, KG and Nishi-Mataya, NM) and downstream waters (Yasu River) due to photochemical and microbial degradation of CDOM during the 13 days of irradiation and dark incubation period. Microbial degradation is presented for 13 days using the similar symbols for the respective samples. *Data source* Mostofa KMG et al. (unpublished data)

cases, CDOM photobleaching by solar radiation decreases *S* (Fig. 8) (Helms et al. 2008; Stabenau et al. 2004; Morris and Hargreaves 1997; Gao and Zepp 1998; Tzortziou et al. 2007; Miller 1994; Zepp et al. 1998; del Castillo et al. 1999). It is possible that *S* variation is caused by the different protocols employed in its calculation (linear function, LF versus non-linear function, NLF) or by the different spectral ranges adopted in the irradiation experiments (Zhang et al. 2009; del Vecchio and Blough 2002). Spectral wavelength ranges used to calculate *S* are most often 275–295 or 350–400 nm. Deviation of *S* for different spectral ranges is mainly caused by the occurrence of different chromophores in CDOM, which show variable reactivity toward photoinduced degradation.

However, variation in *S* is also observed when the same spectral range is considered and the same calculation method is adopted, in a variety of natural waters and in their photobleached samples (Fig. 8). Photoinduced degradation increases *S* at 275–295 nm ($\mathrm{S}_{275–295}$) for upstream (115 % for Kago, KG and 207 % for Nishi-Mataya, NM) and downstream DOM (59 % for Yasu River, YR). In contrast, *S* at 350–400 nm ($\mathrm{S}_{350–400}$) is increased for upstream (92 % for KG) and downstream DOM (6 % for YR), but is decreased for upstream DOM (41 % for NM) during 13 days irradiation (Fig. 8). Earlier studies have shown that upstream DOM is mainly composed of fulvic acids whist downstream DOM is contributed by several sources including autochthonous, allochthonous and agricultural DOM (Mostofa et al. 2007; Mostofa et al. 2005). *S* also increases at 290-500 nm in mangrove and Sargassum CDOM after 48 h irradiation (Shank et al. 2010). The highest increase of *S* has been observed upon irradiation at shorter wavelengths, while irradiation at $\lambda > 400$ nm produced small losses in absorption and little changes in *S* in Bay waters (del Vecchio and Blough 2002). Maxima of $S_{290–350}$ and $S_{250–650}$ and minima of a_{300} have been attributed to CDOM photo-oxidation in the surface waters of the Atlantic Ocean (Kitidis et al. 2006).

The variation of S suggests two important characteristics of natural waters. The first is that CDOM chromophores are decomposed photolytically and their decomposition rates are variable depending on the chemical nature (allochthonous or autochthonous) of CDOM and on its molecular structure. The second issue is that the decomposition rates of the different CDOM chromophores affect in different ways the values of S in different spectral ranges. The overall effect depends also in this case on the CDOM origin and composition. It has been shown for instance that decomposition of CDOM fractions with higher-than-average concentrations of carboxyl-, hydroxyl- and ester-substituted aromatic rings, upon either photoinduced oxidation or chlorine addition, decreases the intensity and width of the electron-transfer and benzenoid bands (Korshin et al. 1997). Such a finding suggests a correlation between the spectral slope (S) alterations by photoinduced degradation and the modification of mean molecular size and molecular structure of CDOM. The latter have generally been found to decrease from rivers to lakes and oceans (Moran et al. 2000; Moran and Zepp 1997; Corin et al. 1996; Allard et al. 1994; Amador et al. 1989; Wu et al. 2005; Yoshioka et al. 2007; Clark et al. 2008). Moreover, the spectral slope S (Jerlov 1968) as well as the carbon-specific absorptivity could be useful indicators to examine photodegradation processes in natural waters (Vodacek et al. 1997; Twardowski and Donaghay 2001; Morris and Hargreaves 1997; Whitehead et al. 2000).

4.3.4 Effect of Monochromatic and Polychromatic Irradiation on CDOM Absorption

Monochromatic irradiation of Suwannee River Fulvic Acid (SRFA) and natural waters can result in the loss of absorption across the entire spectrum and the largest absorption losses are often observed at the irradiation wavelength, λ_{irr} (Fig. 9) (del Vecchio and Blough 2002). Outside the band of direct bleaching, the loss of absorption appears to be fairly uniform across the examined spectral range. Major secondary bands of absorption loss outside λ_{irr} are not evident in the difference spectra (Fig. 9b, d, f). The high losses of absorption at λ_{irr} can mostly be attributed to the direct photoinduced destruction of the chromophore(s) absorbing at this wavelength. The kinetics of absorption loss at both λ_{irr} and at the wavelengths outside of this band exhibit excellent fits to either a single exponential or a sum of two exponentials functions. The overall rate of absorption loss is always higher at the λ_{irr}. The smaller, indirect absorption losses observed outside λ_{irr} could be produced by two effects (del Vecchio and Blough 2002): (i) the direct photoinduced destruction of chromophore(s) having absorption bands both at λ_{irr} and outside λ_{irr}; (ii) the production of reactive oxygen species from primary photochemistry at λ_{irr} that react secondarily to destroy chromophores absorbing at wavelengths outside λ_{irr}. The uniform loss of absorption outside the primary bleaching band (e.g., away from λ_{irr}) suggests that indirect photobleaching could result from the indiscriminate destruction of chromophores by reactive oxygen species produced by the primary photochemistry. It is shown that the reactive oxygen species

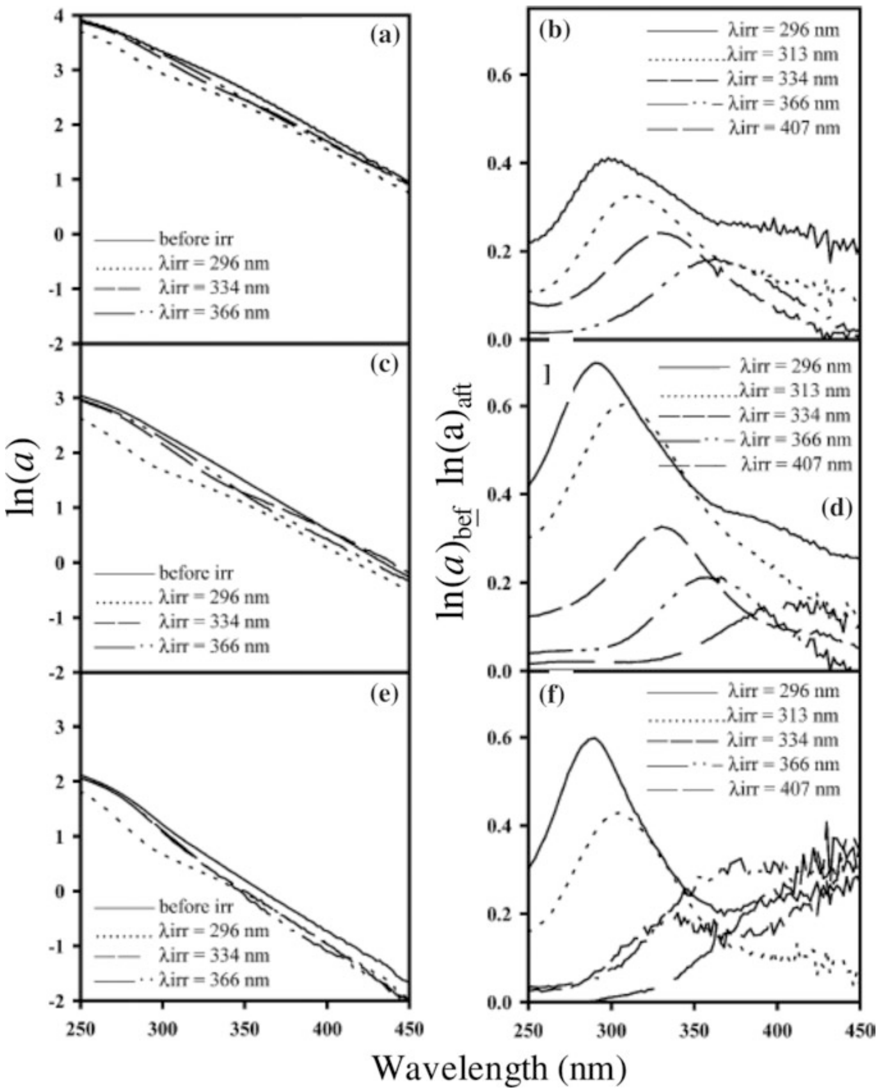


Fig. 9 Log-linearized absorption spectra obtained before and after monochromatic irradiation (10 nm band pass) (*left panels*) and difference spectra obtained by subtracting the ln (**a**) after irradiation from the original spectrum (*right panels*) for SRFA (**a** and **b**; 1 cm optical path length), Delaware Bay water (**c** and **d**; 5-cm optical path length), and Chesapeake Bay water (**e** and **f**, 5- and 10-cm optical path lengths). All spectra reported on the *left panels* are the result of independent experiments. *Data source* del Vecchio and Blough (2002)

(e.g. hydrogen peroxide, the hydroxyl radical, singlet oxygen, superoxide) are photolytically generated from CDOM in waters (Thomas-Smith and Blough 2001; Mostofa and Sakugawa 2009; al Housari et al. 2010; Minella et al. 2011; Zepp et al. 1998; Vaughan and Blough 1998; Zafiriou et al. 1998). The wavelength dependence observed for reactive intermediate production is consistent with the decrease in the efficiency of the primary photobleaching: because the efficiency of reactive intermediate production decreases with increasing wavelength, any indirect photobleaching caused by reactions with these reactive intermediates would be expected to follow the same trend (del Vecchio and Blough 2002).

Polychromatic irradiation of SRFA and natural waters can result in the loss of absorption across the entire spectrum and the bleaching is often more pronounced in the spectral region that is transmitted by the cut-off filter (Fig. 10) (del Vecchio and Blough 2002), in analogy with the results obtained for monochromatic irradiation. Coherently, absorption losses increase with decreasing λ of the cut-off filters (Fig. 9) (del Vecchio and Blough 2002). The results show that the relative loss of absorption is higher at longer wavelengths, although the efficiency of direct photobleaching decreases significantly with increasing wavelength (del Vecchio and Blough 2002). This result can be attributed to two factors (del Vecchio and Blough 2002): (i) the higher number of longer-wavelength photons produced by the light source; (ii) the higher rates of indirect absorption loss produced at longer wavelengths by the absorption of short wavelength photons. The losses of absorption at longer wavelengths lead to an increase of *S* when the spectral data are fit to a single exponential function using either linear or non-linear least squares methods (del Vecchio and Blough 2002).

Using a 320-nm filter, the spectral dependence of a solar simulator is similar to that of ground-level solar spectrum. The changes in *S* for $\lambda_{\mathrm{irr}} > 320$ nm (Fig. 10) should thus reflect the changes in an optically thin section of surface water. The results indicate that in waters where the penetration depths of the photolytically active UV-B and UV-A wavelengths are comparable to the mixed layer depth, the loss of CDOM absorption and the increase in *S* in the mixed layer will be relatively rapid. If the penetration depths are much shallower than the mixed layer depth, absorption losses and changes in *S* in the mixed layer will be very small even over extended periods of time (del Vecchio and Blough 2002).

4.3.5 Factors Controlling the Photoinduced Degradation of CDOM Absorption

Photodegradation of CDOM depends on the sources of water, CDOM concentration, optical-chemical CDOM nature, time, space, sunlight irradiance, water chemical conditions, DOM contents, mixing regime, rain or precipitation and so on (Ma and Green 2004; Reche et al. 1999; Whitehead and Vernet 2000; Gonsior et al. 2008). It has been shown that photobleaching varies significantly depending on the lamp distance from the samples. The decrease of CDOM absorption is 8–19 % at 5 cm lamp distance, but only 2–5 % when the lamp is positioned at 45 cm, during a 2–12 h irradiation period using a UV-B lamp (Zhang et al. 2009). Moreover, the key factors that affect CDOM photobleaching are: (1) Solar radiation; (2) Water temperature; (3) Effects of total dissolved Fe and photo-Fenton reaction; (4) Occurrence and quantity of NO_2^- and NO_3^- ions; (5) Molecular nature of DOM; (6) pH and alkalinity of the water; (7) Dissolved oxygen (O_2; (8) Depth

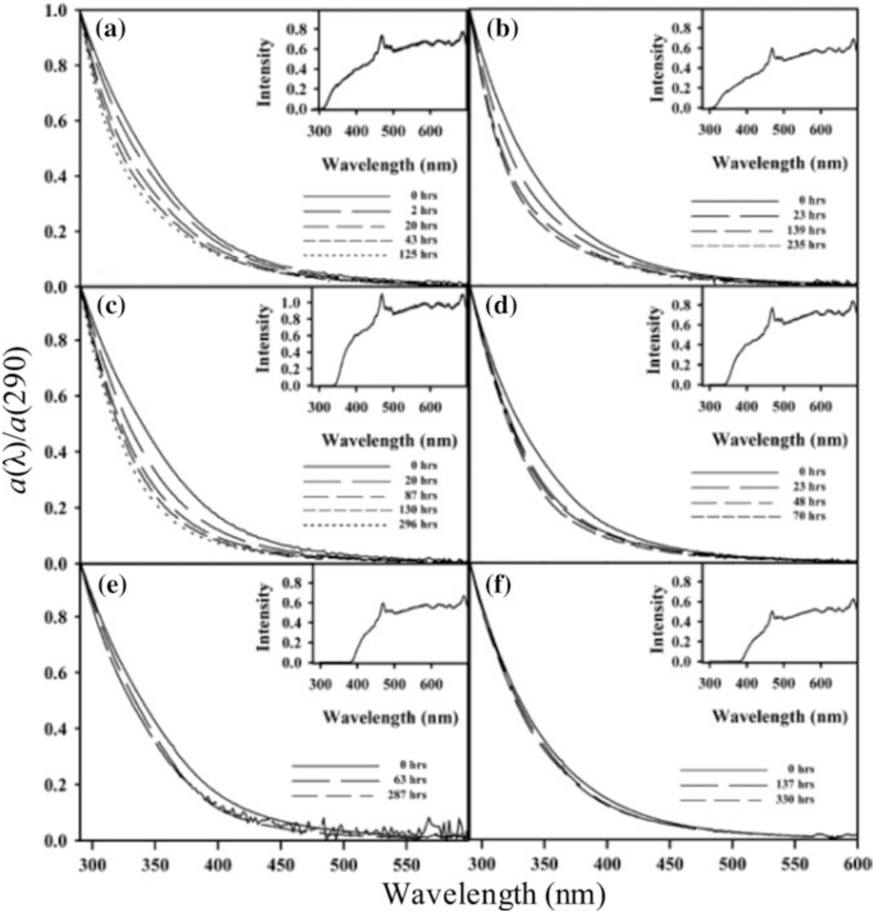


Fig. 10 Absorption spectra normalized to *a*(290) acquired during polychromatic irradiations for SRFA (*left panels*) and Delaware Bay water (*right panels*) using a cut-off filter at 320 (**a**, **b**), 360 (**c**, **d**), and 400 nm (**e**, **f**). Insets: spectral irradiance of the source in units of 10^{15} photons cm^{-2} s^{-1} nm^{-1}. Note that in the absence of an increase in the S, these spectra would be superimposable regardless of the fitting procedure. *Data source* del Vecchio and Blough (2002)

of the water; (9) Physical mixing in the surface mixing zone; (10) Increasing UV-radiation during ozone hole events; (11) Global warming and (12) Salinity. These factors are discussed in chapter "Photoinduced and Microbial Degradation of Dissolved Organic Matter in Natural Waters".

4.4 Microbial Degradation of CDOM

Microbial degradation significantly changes the CDOM absorption and spectral slope properties in natural waters (Fig. 1; Table 1) (Helms et al. 2008; Moran et al. 2000; Winter et al. 2007; Mostofa KMG et al., unpublished data; Brown 1977). After 13 days incubation, CDOM absorption has been found to increase over the entire spectrum in upstream waters. Absorption increase was 20–81 % at 340–350 nm and 4–38 % at 600–700 nm, but it was highest at 312–410 nm (77–88 %) for Nishi-Mataya upstream and at 440–570 nm (39–49 %) for Kago upstream (Fig. 1; Table 1). In downstream waters, CDOM absorption was decreased (6–7 %) at 340–350 nm but increased (8–49 %) at 600–700 nm. The maximum decrease occurred at 390–415 nm (9–11 %) (Fig. 1; Table 1). CDOM absorption was also decreased in pond (4 % at 340 nm), lakes (1–32 % at 340 nm), estuaries (2–4 % at 350 nm and 4–11 % at 250–500 nm). In other cases, very small increases have been observed in pond (3 % at 340 nm), marsh (2–3 % at 340 nm), lakes (2–3 % at 340 nm) and estuaries (1 %) (Table 1) (Moran et al. 2000; Winter et al. 2007). These results show a significant microbial effect on CDOM in natural waters (Table 1; Fig. 1).

Such an effect shows several characteristic phenomena. First, an increase in CDOM absorption over the entire spectrum in upstream waters might be due to a microbial alteration of the composition of fulvic acids. In fact, it has been known that upstream CDOM is mainly composed of fulvic acids (Mostofa et al. 2007; Mostofa et al. 2005). Second, an increase in absorption at longer wavelength and a decrease at shorter wavelength in the waters of downstream river might be due to the presence of various CDOM sources. Note that upstream water is one of the sources of the downstream one, and downstream fulvic acids (FA) might derive from upstream FA upon transformation induced by photochemistry and/or by microorganisms. The latter process is rather slow but could account for the increase of downstream CDOM absorption in the longer wavelength region, because a similar phenomenon is also observed in upstream waters. Conversely, the autochthonous and agricultural CDOM in downstream waters are likely to undergo rapid microbial degradation that, given the different nature of this kind of CDOM, might result in a decrease of CDOM absorption in the shorter wavelength region. Downstream DOM is in fact derived from several sources including autochthonous (protein-like or tryptophan-like), allochthonous (mostly fulvic acids of upstream origin) and agricultural DOM that is released from nearby agricultural fields (Mostofa et al. 2007; Mostofa et al. 2005).

CDOM absorption is also found either to decrease or to increase in ponds, lakes, marshes and estuaries that are relatively similar to downstream river environments. The CDOM in these natural waters generally consists of both allochthonous (mostly fulvic acids) and autochthonous material. Autochthonous organic substances are microbially labile and their absorption is decreased by microbial degradation, differently from allochthonous fulvic substances. This can explain the rather complex effects of microbial processing on CDOM absorption in the different wavelength ranges.

Experimental studies also show that a large amount of high molecular weight CDOM is produced during phytoplankton lysis. $S_{300–500}$ is decreased in the first 9 days when CDOM composition is changed due to increasing microbial activity, which is expected to decrease the molecular weight of organic substances (Fig. 4b) (Zhang et al. 2009; Mostofa KMG et al., unpublished data).

Alteration of autochthonous fulvic acids (C-like) changes their fluorescence intensity during a long incubation period in the dark (120 days) at room temperature, possibly because of microbial assimilation of lake algae (Mostofa KMG et al., unpublished data). The autochthonous fulvic acid (C-like) of algal origin is identified using the PARAFAC model on the EEM spectra of the samples. The aerobic microbial incubation in the dark results in a statistically significant decrease of *S* over timescales of days to weeks, due to microbial production or to selective preservation of long-wavelength absorbing substances (Helms et al. 2008).

Microbial degradation typically alters *S* in natural waters (Fig. 8) (Helms et al. 2008; Mostofa KMG et al., unpublished data; Brown 1977). It has been observed a decrease in *S* of 1–13 % at both 275–295 nm ($S_{275–295}$) and 350–400 nm ($S_{350–400}$) for upstream and downstream DOM, with the esception of 350–400 nm ($S_{350–400}$) in downstream DOM where *S* has been found to increase (~5 %) after 13 days of dark incubation (Fig. 8). Because of significant variations in CDOM composition between upstream and downstream samples, it is suggested that microbial degradation of CDOM depends on its chemical nature and on its sources.

4.4.1 Factors Controlling the Microbial Degradation of CDOM Absorbance

Microbial degradation of DOM depends on several key factors that can be listed as: (1) Occurrence and nature of microbes in waters; (2) Sources of DOM and amount of bacterial fermentation products; (3) Temperature; (4) pH; and (5) Sediment depths in pore waters. These factors are discussed in detail in chapter "Photoinduced and Microbial Degradation of Dissolved Organic Matter in Natural Waters".

4.5 Salinity

The absorption properties of CDOM are modified when the terrestrial riverine input of CDOM is mixed with costal seawaters (Singh et al. 2010; Hernes and Benner 2003; del Vecchio and Blough 2004; Blough and del Vecchio 2002; Fournier 2007; Nieke et al. 1997; Twardowski and Donaghay 2001; del Castillo et al. 1999; Gonsior et al. 2008; Uher et al. 2001; Blough et al. 1993; Sholkovitz 1976). A recent study shows that the addition of various salts, which are present in seawater, gives rise to an extra absorption in the far UV and to an increase of the amount of scattering because small variation in salt concentration can cause refractive index fluctuations (Fournier 2007). A linear inverse relationship of CDOM absorption to salinity could be a useful indicator of salinity in coastal waters strongly affected by river input (Singh et al. 2010; del Vecchio and Blough 2004; Nieke et al. 1997; Blough et al. 1993). However, at lower salinity minor deviations from linearity may result from consumption or production of CDOM in coastal waters (del Vecchio and Blough 2004; Twardowski and Donaghay 2001; del Castillo et al. 1999; Uher et al. 2001; Blough et al. 1993; Sholkovitz 1976). In addition, a nonlinear dependence may result from the conservative mixing of multiple water masses containing variable CDOM (del Vecchio and Blough 2004; Blough and del Vecchio 2002; Hujerslev et al. 1996; Chen et al. 2007; Blough et al. 1993). Photoinduced degradation can greatly decrease the CDOM absorbance in intermediate- to high-salinity surface waters under stratified conditions during the summer period (Vodacek et al. 1997; del Vecchio and Blough 2004; Chen et al. 2007; Osburn and Morris 2003; Osburn et al. 2009). Dissolved lignin phenols are significantly affected by salinity and two key phenomena are generally detected: First, a nonconservative decrease in dissolved high molecular weight (HMW) lignin phenols at salinity <25 psu is likely due to flocculation and microbial degradation. In contrast, LMW dissolved lignin phenols mix conservatively (Hernes and Benner 2003). Second, at salinity >25 psu photooxidation is a dominant factor influencing lignin composition and concentration (Hernes and Benner 2003).

CDOM photoreactivity can increase with salinity across an estuarine gradient. Shortwave CDOM absorption loss (e.g. at 280 nm) does not change with salinity, but longwave CDOM absorption loss (e.g. at 440 nm) is often decreased by 10–40 % with increasing salinity (Osburn and Morris 2003; Osburn et al. 2009). In another study, a decrease in CDOM photobleaching at 280 nm is detected when humic CDOM is added to an artificial salinity gradient used to mimic coastal mixing (Minor et al. 2006). The decrease of the absorption properties of CDOM with salinity can be accounted for by several factors: (i) Mixing of CDOM-rich riverine water with CDOM-poor coastal water (del Vecchio and Blough 2004; Gonsior et al. 2008; Blough et al. 1993); (ii) Photodegradation of chromophores present in riverine CDOM after they reach the coastal regions during the summer stratification period (Vodacek et al. 1997; Moran et al. 2000; del Vecchio and Blough 2004; Blough and del Vecchio 2002; Whitehead and Vernet 2000; del Vecchio and Blough 2002; Osburn et al. 2009); (iii) Microbial degradation, in particular of the autochthonous fraction that is the major part of CDOM in marine waters (Table 1) (Moran et al. 2000; Winter et al. 2007; Moran and Hodson 1994; Brown 1977; Opsahl and Benner 1998); (iv) Flocculation and precipitation of riverine CDOM because of increased salinity (Blough et al. 1993; Sholkovitz 1976; Sieburth and Jensen 1968; Fox 1991); and possibly (v) Enhanced CDOM photodegradation in saline waters.

The mechanism behind the latter process apparently involves two factors: first, irradiated CDOM can induce photoinduced production of hydrogen peroxide (H_2O_2) that is a $HO^{\bullet}$ source via photolysis or the photo-Fenton reaction, and the photoinduced generation of H_2O_2 is enhanced by salinity. Trace metal ions (M) in salinity or sea waters can complex with DOM (M-DOM) forming a strong π–electron bonding system between metal ions and the functional groups in DOM (see chapter "Complexation of Dissolved Organic Matter with Trace Metal Ions in Natural Waters" for in details explanation). This π–electron in M-DOM complex is rapidly excited photolytically, which is responsible for high production of aqueous electrons (e_{aq}-) and subsequently the high production of superoxide ion

($O_2^{\bullet -}$), H_2O_2 and $HO^{\bullet}$, respectively. Indeed, photogeneration of H_2O_2 from ultrafiltered river DOM is substantially increased with salinity, from 15 to 368 nM h^{-1} at circumneutral pH (Osburn et al. 2009). Salinity or NaCl salts are responsible for generating high production of aqueous electrons (e_{aq}^{-}) photolytically in aqueous media (Gopinathan et al. 1972; Assel et al. 1998) that may subsequently enhance the H_2O_2 production in waters (Mostofa and Sakugawa 2009; Moore et al. 1993; Richard et al. 2007; Fujiwara et al. 1993).

Recent studies observe that the sea-salt particulate matter extracted from coastal seawaters show substantially high $HO^{\bullet}$ production (rate: ~2778–27778 M s^{-1}), approximately 3–4 orders of magnitude greater than $HO^{\bullet}$ photoformation rates in surface seawater (Anastasio and Newberg 2007). Note that comparison of river and salinity of sea waters shows that Na^{+}, Ca^{2+}, Mg^{2+}, K^{+}, HCO_3^{-}, Cl^{-} and SO_4^{2-} are typically 1,670 times, 27 times, 330 times, 170 times, 2.4 times, 2,400 times and 245 times, respectively, higher than those in rivers (Livingstone 1963; Hem 1985). The order of the other cations is $Mg^{2+} > Ca^{2+} > K^{+} > Sr^{2+}$ and the anion Cl^{-} is approximately equal to the sum of the cations and the other anions are SO_4^{2-}, HCO_3^{-}, Br^{-}, and F^{-} (Livingstone 1963; Hem 1985; Carpenter and Manella 1973). Second, the reaction of $HO^{\bullet}$ with halide ions (X^{-}) can form reactive halogen radicals ($X_2^{\bullet -}$) that can react with electron-rich functional groups within DOM more selectively than $HO^{\bullet}$ (Zafiriou et al. 1987; Song et al. 1996; Von Gunten and Oliveras 1997; Goldstone et al. 2002; Grebel et al. 2009). The absorption of radiation by CDOM is usually increased in ionic solutions of NaCl, which might suggest an increase of CDOM absorption in coastal waters. However, such an effect is more than compensate for by the efficient photodegradation of CDOM chromophores in saline waters.

5 All Colored DOM is Chromophoric DOM, But Not All Chromophoric DOM is Colored

The main chromophores in colored or chromophoric DOM are Schiff-base derivatives (–N=C–C=C–N–) and groups such as –COOH, $–COOCH_3$, –OH, $–OCH_3$, –CH=CH–, –CH=O, –C=O, $–NH_2$, –NH–, –CH=CH–COOH, $–OCH_3$, $–CH_2–(NH_2)CH–COOH$, and S-, O- or N-containing aromatic compounds or functional groups (Mostofa et al. 2009; Malcolm 1985; Corin et al. 1996; Senesi 1990; Leenheer and Croue 2003; Peña-Méndez et al. 2005; Zhang et al. 2005; Steelink 2002; Seitzinger et al. 2005). The allochthonous fulvic and humic acids (humic substances) of vascular plant origin and the autochthonous fulvic acids of algal (or phytoplankton) origin show absorbance in a wide wavelength interval, 200–800 nm (Figs. 2, 3, 4) (Zhang et al. 2009; Ishiwatari 1973; Lawrence 1980; Zepp and Schlotzhauer 1981; Hayase and Tsubota 1985; Dubach et al. 1964), and contain the above mentioned chromophores (or functional groups) in their molecular structure. These macromolecular substances are both colored DOM (they absorb in the visible) and chromophoric DOM. On the other hand, there are a lot of organic substances including most notably low molecular weight (LMW) CDOM that are not colored, not being able to absorb radiation in the visible range. For example, acetaldehyde absorbs light at 208–224 nm (Strome and Miller 1978; Kieber et al. 1990; Mopper et al. 1991), acetate at 204–270 nm (Wetzel et al. 1995; Dahlén et al. 1996), formaldehyde at 207–250 nm (Mopper and Stahovec 1986; Kieber et al. 1990; Mopper et al. 1991), glyoxal at <240 nm (Mopper and Stahovec 1986; Mopper et al. 1991, malonate at 225–240 nm (Dahlén et al. 1996), pyruvate at 200–227 nm (Kieber et al. 1990; Mopper et al. 1991; Wetzel et al. 1995) and propanal at ~230 nm (Mopper and Stahovec 1986). These organic compounds are definitely not colored, but they contribute to the absorption of radiation by water in the relevant wavelength ranges. It is thus shown that, while all colored DOM is also chromophoric DOM, not all the chromophoric DOM is also colored.

6 Importance of CDOM Studies in Natural Waters

CDOM is a major bio-optical parameter because of its strong light-absorbing properties. These properties are involved in some very important biogeochemical processes and are very useful for detection techniques. Implications are: (i) remote sensing of CDOM in natural waters; (ii) DOM dynamics in natural waters; (iii) Photoinduced degradation of CDOM and its impact in natural waters; and (iv) Protection of microorganisms from UV radiation by CDOM.

6.1 Remote Sensing of CDOM in Natural Waters

Remote sensing is widely used to estimate the ocean color constituents such as chlorophyll *a* and algae, and also to assess primary productivity, occurrence of toxic dinoflagellate, total suspended solids (TSS), tripton (inorganic suspended particulate matter), inherent optical properties, CDOM contents, diffuse attenuation coefficients (K_d), DOC concentration and transport from rivers to lakes and oceans (Coble 2007; Del Castillo and Miller 2008; Carder et al. 1991; Carder et al. 1999; McClain et al. 2004; O'Reilly et al. 1998; Ferreira et al. 2009; Tzortziou et al. 2007; Son et al. 2011; Sathyendranath et al. 1989; Woodruff et al. 1999; Stramski et al. 2001; Volpe et al. 2011; van der Woerd et al. 2011; Le et al. 2011; Carvalho et al. 2011; Santini et al. 2010; Matthews et al. 2010; Doxaran et al. 2002; Cui et al. 2010; Zibordi et al. 2009; Werdell et al. 2009; Tomlinson et al. 2009; Friedrichs et al. 2009; Van Der Woerd and Pasterkamp 2008; Hunter et al. 2008; Brown et al. 2008; Zawada et al. 2007; Mélin et al. 2007; Tzortziou et al. 2006; Koponen et al. 2007; Kishino et al. 2005; Phinn et al. 2005; Vahtmäe et al. 2006; Zimba and Gitelson 2006). Satellite remote sensing is also used to monitor cyanobacterial blooms in natural waters, which can be detected from a small peak in reflectance spectra near 650 nm that is specific of cyanobacteria (Matthews et al. 2010; Kutser et al. 2006;

Becker et al. 2009; Hunter et al. 2010). Remote sensing is currently used to monitor broad changes in phytoplankton communities, exploiting the spectral dissimilarities of brown, green, blue-green and red algae in inland waters. This technique is an extremely useful tool for limnological research and water resource management (Hunter et al. 2008). Water quality (Secchi depth, K_d in PAR, tripton, CDOM) and substrate cover type (seagrass, algae, sand) parameters, which vary in sub-tropical and tropical coastal environments may also affect the satellite image data (Phinn et al. 2005) and can thus influence the remote sensing information. A combination of a chlorophyll anomaly (spectral shape at 490 nm) and a backscatter ratio can provide an improvement in satellite detection of the toxic dinoflagellate *Karenia brevis*. It is possible to increase the detection accuracy by 30–50 % in seawaters (Tomlinson et al. 2009; Cannizzaro et al. 2008). The remote sensing application has also been used to characterize high concentrations of suspended sediment and to map chlorophyll *a* (Chl *a*) or phytoplankton and non-phytoplankton suspended matter distribution in lakes and oceans (Ferreira et al. 2009; Cannizzaro and Carder 2006; Gons et al. 2008; Oyama et al. 2009; González Vilas et al. 2011).

The ocean color depends on the optical variables (Coble 2007; Del Castillo and Miller 2008; Carder et al. 1991; Hoge et al. 1995; Hoge et al. 2001; Sathyendranath et al. 1989; Stramski et al. 2001; Brown et al. 2008; Mélin et al. 2007; Lee et al. 1994; Kahru and Mitchell 2001; Siegel et al. 2002; Nair et al. 2008; Siegel et al. 2005). The key factors are (i) phytoplankton species (or algae) and their variability; (ii) the amount of colored DOM; (iii) the amount and size of organic particles; (iv) the contents of inorganic particles (tripton); and (v) water itself. CDOM can be estimated by using ocean color with various levels of success (del Castillo and Miller 2008; Carder et al. 1999; Hoge et al. 1995; Hoge et al. 2001; Lee et al. 1994; Kahru and Mitchell 2001; Siegel et al. 2002; Siegel et al. 2005). In open ocean waters, far from the influence of terrestrial runoff that mainly affects coastal waters (case 1 waters), the spectral quality and intensity of light leaving the ocean depends first of all on the concentration of phytoplankton (Morel and Prieur 1977; Morel 1980). Empirical algorithms of ocean color based on blue-to-green ratios are used to estimate the chlorophyll *a* concentration within the upper layer of the water column (McClain et al. 2004; O'Reilly et al. 1998; Harding et al. 2005). The second-order variability for given chlorophyll levels depends on two main sources: (i) the amount of non-algal absorption, especially due to colored dissolved organic matter; and (ii) the amplitude of the backscattering coefficient of particles (Brown et al. 2008). Remote sensing of surface waters in the open ocean (case 2 waters) could be used in conjunction with the inversion of UV-blue wavelengths, to separate the contribution of non-algal particles and of colored dissolved organic matter to the total light absorption, and to monitor non-algal suspended particle concentration and distribution (Tzortziou et al. 2007). It is shown that phytoplankton typically absorbs strongly in the blue and weakly in the green. CDOM absorption thus overlaps to that of phytoplankton and non-algal particulate matter in the blue part of the visible spectrum. This issue might affect the primary productivity and the remote sensing estimation of phytoplankton biomass and of total suspended matter concentration (Zhang et al. 2009; Carder et al. 1991; Doxaran et al. 2002).

Ocean color remote sensing could be applied to estimate DOC transport in river-ocean transects, depending on the robust empirical relationships between DOC and CDOM, CDOM and salinity, and salinity and river flow (del Castillo and Miller 2008). A flow chart is depicting the method and rationale for the estimate of DOC transport (Fig. 11). The success of this approach depends on four conditions (del Castillo and Miller 2008): (1) DOC and CDOM must behave conservatively at the study site; (2) The relationship between CDOM and DOC in the river end member must remain constant; (3) One should be able to derive CDOM from satellite ocean color measurements; (4) Salinity in the study area should

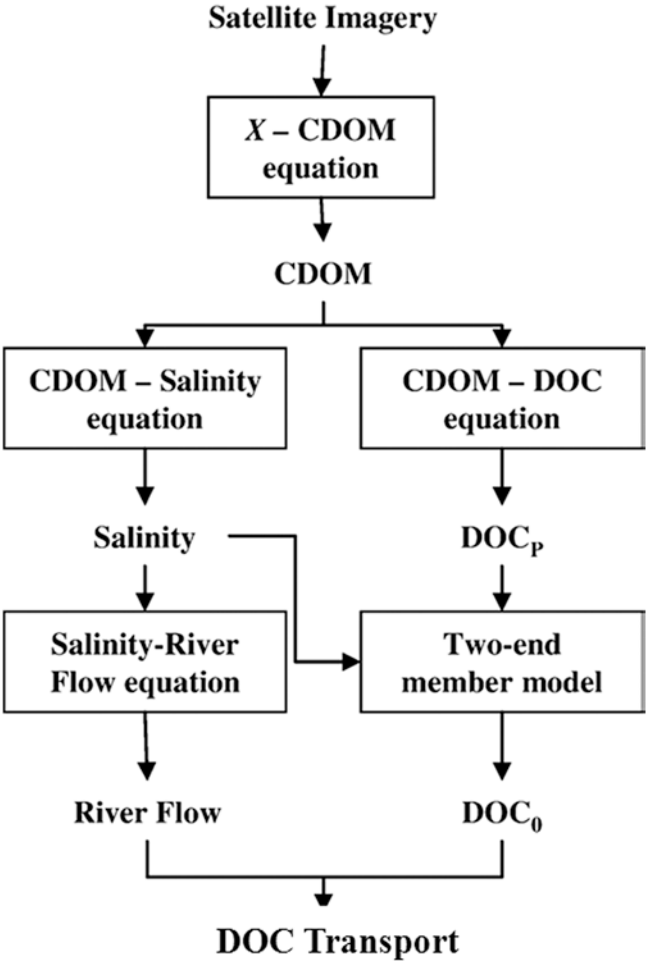


Fig. 11 Flow chart showing the method and rational of DOC transport. Boxes represent empirical relationship, plain text represent the outputs of these relationships. *X* is the remote sensing reflectance ratio used in this study, and DOC_P and DOC_0 are the concentrations of DOC in the river plume and river end-member respectively. *Data source* del Castillo and Miller (2008)

correlate with river flow. Note that the remote sensing device generally detects radiation that is reflected or backscattered from the target that initially absorbs the radiation.

6.2 DOM Dynamics in Natural Waters

Good correlations between CDOM absorption and DOM (DOC concentration) are usually characterized by a positive intercept on the DOC axis at 50–100 μM, because offshore waters contain only very small amounts of CDOM (Vodacek et al. 1997; Rochelle-Newall and Fisher 2002; del Vecchio and Blough 2004; Vodacek et al. 1995; Klinkhammer et al. 2000; Chen et al. 2002; Gueguen et al. 2011). The non-absorbing DOC fraction varies with the qualitative composition of the CDOM (Kowalczuk et al. 2010). The CDOM/DOC dependence is typically changed in surface waters during the summer stratification period, when the water samples show lower absorption to DOC ratios compared to waters from below the mixed layer or collected in other seasons (Vodacek et al. 1997). This might be the effect of high autochthonous production of non-absorbing DOM in surface waters during the summer stratification period (Mitra et al. 2000; Ogawa and Tanoue 2003; Mostofa et al. 2005; Fu et al. 2010; Yoshioka et al. 2002; Hayakawa et al. 2003; Hayakawa 2004; Ogawa and Ogura 1992), and/or of photoinduced degradation of CDOM with production of non-absorbing compounds (Coble 2007; Vodacek et al. 1997; Mostofa et al. 2005). Note that a rough estimate shows that the increase in autochthonous DOC contents during the summer stratification period is 0–88 in lakes and 0–194 % in oceans, determined by comparing the epilimnionetic DOM with that of the hypolimnion (Mostofa et al. 2009).

The predominant presence of colored DOM such as allochthonous fulvic and humic acids is responsible for the good correlation that is usually observed between CDOM absorbance and DOM contents in a variety of waters (Vione et al. 2010). The contributions of allochthonous humic substances (fulvic and humic acids) in rivers are 30–85 % (the ratio of fulvic acid to humic acid is 9:1 for lower stream DOC and it decreases to 4:1 or less for higher DOC stream), 15–60 % in lakes (30–60 % during winter and 15–40 % during the summer period), 1–75 % in shelf seawater (38 % of marsh origin and or 62 % of river origin), and 2–38 % in ocean (see chapter "Dissolved Organic Matter in Natural Waters" for detailed description) (Mostofa et al. 2009; Moran et al. 1991; Moran and Hodson 1994). The seasonal and spatial–temporal variations of the CDOM absorbance to DOC ratios are dependent on the presence of colored DOM compounds in natural waters.

A recent study demonstrates that DOC transport can be determined using ocean color remote sensing if the empirical relationships between DOC and CDOM, CDOM and salinity, and salinity and river flow are known (del Castillo and Miller 2008). It has been shown that remote sensing estimates of river flow and DOC transport are correlated well ($r^2 = \sim 0.70$) with field observation data, showing low variability in DOC concentrations in the river end-member (7–11 %), and high seasonal variability in river flow (~50 %) in the Mississippi River Plume (del Castillo and Miller 2008). This result can be influenced by several biogeochemical processes such as high DOM photodegradation, biodegradation, production and flocculation, as well as extreme precipitation caused by natural disaster (del Castillo and Miller 2008; Wright 2005). These biogeochemical processes have little or negligible effects in low salinity waters of river plumes due to the predominance of riverine CDOM (Blough and del Vecchio 2002; del Castillo and Miller 2008; del Vecchio and Blough 2002; del Castillo et al. 1999; del Castillo et al. 2000; del Castillo et al. 2001; Mantoura and Woodward 1983).

6.3 Photoinduced Degradation of CDOM and Its Impact in Natural Waters

Photoinduced degradation of CDOM by sunlight can affect its optical and chemical properties, by inducing decomposition of the CDOM chromophores and thus reducing CDOM absorptivity of UV and visible radiation (Kieber et al. 1990; Moran et al. 2000; Skoog et al. 1996; Reche et al. 1999; Whitehead and Vernet 2000; Twardowski and Donaghay 2001; del Vecchio and Blough 2002; Mostofa et al. 2007; Patsayeva et al. 1991; Kouassi and Zika 1990; Kouassi et al. 1990; Morris and Hargreaves 1997; Allard et al. 1994; Fichot and Miller 2010). The effect of the photoinduced degradation of CDOM is an increase of UV transparency in surface waters (Nelson et al. 1998; Vodacek et al. 1997; Kieber et al. 1990; Morris and Hargreaves 1997; Zepp et al. 1995). However, CDOM absorption losses by photoinduced degradation can also result in a variety of changes in CDOM composition, which can be listed as follows: (i) Formation of strong oxidants such as singlet oxygen, superoxide, hydroxyl radical, hydrogen peroxide, organic peroxides during the photodegradation of CDOM may have severe and chronic toxic effects on aquatic organisms and important ecological consequences in aquatic environments (Williamson et al. 1996; Thomas-Smith and Blough 2001; Mostofa and Sakugawa 2009; Moore et al. 1993; Zepp et al. 1998; Vaughan and Blough 1998; Zafiriou et al. 1998; Xenopoulos and Bird 1997; Palenik et al. 1991); (ii) Formation of low molecular weight organic substances, which is generally more important in lakes and oceans than in rivers (Moran and Zepp 1997; Corin et al. 1996; Biddanda and Benner 1997; Yoshioka et al. 2007); (iii) Formation of biologically labile compounds that enhance biodegradation (Wetzel et al. 1995; Moran and Zepp 1997); (iv) Photo formation of carbon-gas end photoproducts (CO_2, CO), DIC, COS and so on in natural waters (Ma and Green 2004; Bertilsson and Tranvik 2000; Miller and Moran 1997; Fichot and Miller 2010; Weiss et al. 1995); (v) Release of nitrogen compounds (e.g. NH_4^+) and phosphate, which may typically be produced by degradation of dissolved organic nitrogen (DON) and dissolved organic phosphorus (DOP) in the epilimnion of natural waters (Mostofa et al. 2011; Zagarese et al. 2001; Kim et al. 2006; Vähätalo and Järvinen 2007; Li et al. 2008; Zhang et al. 2004). These nutrients are used by algae and bacteria. (viii) Finally, releases of energy to the water ecosystem (Wetzel 1992; Hedges et al. 2000; Tranvik 1992).

The decrease of CDOM absorption over the entire spectrum, induced by photoinduced processes is accompanied by the decrease of total fluorescence intensity at peak C-, A-, T- and T_{UV}-regions of various fluorescent substances (Zhang et al. 2009; Coble 1996; Moran et al. 2000; Mostofa et al. 2007; Mostofa et al. 2010). Photoinduced DOC mineralization (decrease of DOC concentration) is also observed (Moran et al. 2000; del Vecchio and Blough 2002; Mostofa et al. 2007; Mostofa et al. 2005; Frimmel and Bauer 1987; Vione et al. 2009; de Haan 1993). As a matter of fact, the impacts of photoinduced degradation on CDOM absorption are similar to those on DOC degradation that have been explained in details in earlier chapter (see chapter "Photoinduced and Microbial Degradation of Dissolved Organic Matter in Natural Waters").

6.4 Protection of Microorganisms from UV Radiation by CDOM

Natural UV radiation (280–400 nm) is a selective and strong environmental factor that damages the cell structures including proteins, lipids, membranes, pigments and DNA. It affects the productivity of freshwater and marine organisms (Marchant et al. 1991; Vincent and Roy 1993; Bothwell et al. 1994; Banaszak and Trench 1995; Leavitt et al. 1997; Poli et al. 2004; Lesser 2006; Valko et al. 2006; Xiong et al. 1997; Teai et al. 1998). Such impacts are caused by the UV-radiation induced production of strong reactive oxygen species such as superoxide radicals, singlet oxygen, hydrogen peroxide and hydroxyl radicals. The main processes involved are the photo-Fenton reaction and the photolysis of NO_2^-, NO_3^- and DOM (see chapter "Photoinduced Generation of Hydroxyl Radical in Natural Waters"). UV attenuation by CDOM is important in minimizing the deleterious consequences of UV radiation on phytoplankton and other organisms in natural waters (Wetzel 1992; Morris et al. 1995; Schindler et al. 1996; Williamson et al. 1996; Yan et al. 1996; Smith and Baker 1979). It is shown that postbloom increases in DOM concentration induced by grazing and decomposition of phytoplankton biomass cause an increase of DOM absorption and a related decrease of UV transmission through the water column (Whitehead and Vernet 2000). The partial UV protection by autochthonous DOM has the consequence that shallow blooming phytoplankton may assist the development of a subsequent bloom in deep waters (Whitehead and Vernet 2000). On the other hand, any changes in hydrology (e.g., high temperature, drier climate) or geochemistry (e.g., increased atmospheric deposition of strong acids) due to global warming can reduce DOM concentrations in surface waters. The consequence might be an increased exposure of aquatic organisms to both UV-A and UV-B radiation (Schindler et al. 1996; Yan et al. 1996; Morris and Hargreaves 1997).

7 Scope of the Future Challenges

CDOM is generally produced from two major sources: allochthonous and autochthonous. Absorption coefficients of CDOM have often been determined jointly in earlier studies. Recent studies show that autochthonous DOM is significantly produced from photo- and microbial respiration or assimilation of algae or phytoplankton biomass, yielding compounds that are very similar to allochthonous fulvic and humic acids (Zhang et al. 2009; Mostofa et al. 2009; Yamashita and Tanoue 2004; Yamashita and Tanoue 2008; Mostofa et al. 2009). The specific absorption properties of allochthonous and autochthonous CDOM could be important to understand the physical, photoinduced and microbial changes of those two CDOM pools in natural waters. Remote sensing is widely applied to characterize various parameters as explained earlier, but a key application could be the detection of algal blooms in natural waters. The CDOM research for future challenges can be distinguished as: (i) Investigation on CDOM absorption of allochthothous fulvic and humic acids compared to autochthonous DOM originated from algae or phytoplankton biomass in natural waters. (ii) Investigation on the absorption of radiation by various amino acids or proteins originated from phytoplankton with respect to their standard substances. (iii) Study of the photoinduced and microbial changes of CDOM absorption for both allochthonous and autochthonous DOM. (iv) Investigation on the differences of photoinduced reactivity between allochthonous (fulvic and humic acids) and autochthonous DOM of algal or phytoplankton origin (termed autochthonous fulvic acid, see chapter "Fluorescent Dissolved Organic Matter in Natural Waters"). (v) Application of remote sensing in investigation of algal blooms in natural waters.

Problems

(1) Define the chromophoric dissolved organiu matter (CDOM)?
(2) Define the chromophores in CDOM. Mention the key chromophores in CDOM found in natural waters.
(3) Explain the CDOM absorbance theorem.
(4) What are the optical variables for the attenuation of UV and photosynthetically available radiation in waters? Mention the key optical variable and how it affects the absorption properties of waters.
(5) Explain the effects of suspended particulate matter and chlorophyll on absorption coefficients in waters.
(6) What are the controlling factors that affect CDOM absorption in waters? Explain the role of CDOM contents on absorption properties.

(7) Explain the effects of photoinduced and microbial processes on CDOM absorption properties in waters.
(8) Explain the effects of photoinduced and microbial degradation of CDOM on spectral slopes.
(9) What are the effects of monochromatic and polychromatic irradiation on CDOM absorption spectra?
(10) Why are the CDOM absorption losses by irradiation significantly different for a variety of natural waters?
(11) How does salinity affect CDOM absorption in oceans?
(12) ‘All colored DOM is chromophoric DOM, but not all chromophoric DOM is colored’- Explain this concept.
(13) Explain shortly the applications of remote sensing to surface waters.
(14) Explain the impacts of photoinduced degradation of CDOM in waters.

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