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5 authors, including:

Zhao Dongzhi
National Marine Environmental Monitoring Center

Xiaogang Xing
Second Institute of Oceanography MNR

Yuguang Liu
Ocean University of China

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The relation of chlorophyll-*a* concentration with the reflectance peak near 700 nm in algae-dominated waters and sensitivity of fluorescence algorithms for detecting algal bloom

DONGZHI ZHAO*†, XIAOGANG XING‡, YUGUANG LIU§, JIANHONG YANG† and LIN WANG†

†National Marine Environmental Monitoring Center, SOA, Dalian, 116023, PR China
‡Optics and remote sensing group, Laboratoire d'Océanographie de Villefranche, Villefranche-sur-Mer, 06230, France
§Physical Oceanography Laboratory, Ocean University of China, Qingdao, 266100, PR China

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In order to investigate the relation of chlorophyll-*a* concentration with the reflectance peak near 700 nm, reflectance spectra of harmful algal bloom (HAB) species and non-HAB algae were obtained based on *in situ* measurements in the oceans and cultural tank data. It is found that the fluorescence line heights (FLH) of reflectance spectra and the concentrations of seawater chlorophyll-*a* have good correlation; their coefficients of determination are larger than 0.86, excepting *Ceratium furca* and *Heterosigma akashiwo*. It is specially noted that for some algae, such as *Dicrateria zhanjiangensis Hu.*, *Pyramimonas* sp. and *Nitzschia closterium*, the corresponding coefficients of determination exceed 0.95. In addition, the various satellite fluorescence algorithms were compared, and the sensitivity of fluorescence algorithms was investigated. It is found that the designed bands of Medium Resolution Imaging Spectrometer (MERIS) are more reasonable than those of Moderate Resolution Imaging Spectroradiometer (MODIS) for detecting algal bloom.

1. Introduction

Oceanic satellite observations in the visible and near-infrared bands allow the measurements of varieties of ocean colour information, mainly including phytoplankton chlorophyll-*a*, CDOM (coloured dissolved organic matter) and suspended sediments. Since the 1970s work has been carried out on the determination and retrieval of chlorophyll-*a* concentration. First- and second-generation sensors, i.e. Coastal Zone Colour Sensor (CZCS) and Sea-viewing Wide Field-of-view Sensor (SeaWiFS), adopted blue-green ratio algorithms. However, many new algorithms based on *in vivo* chlorophyll fluorescence for retrieving chlorophyll-*a* concentration have been explored for the third generation sensors, i.e. the Moderate Resolution Imaging Spectroradiometer (MODIS) and the Medium Resolution Imaging Spectrometer (MERIS),

In Case I waters of the open ocean, there is little CDOM and suspended sediments. Phytoplankton with its accompanying and covarying retinue of biological origin material is the principal agent responsible for variations in optical properties of the

*Corresponding author. Email: dzzhao@nmemc.gov.cn

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water. Blue-green ratio algorithms have been confirmed to work well. Nevertheless, CDOM and chlorophyll-*a* concentrations are not correlated in coastal Case II waters. CDOM absorbs light very strongly at shorter wavelengths. In extreme cases, the variation in concentration of phytoplankton has no effect on water reflectance spectra at all as it is totally dominated by CDOM. In such cases, most satellite sensors are not sensitive enough at blue wavelengths to detect changes in water reflectance due to varying CDOM (Kutser *et al*. 2005), thus the changes in CDOM concentration are interpreted as changes in chlorophyll. Therefore, the traditional blue-green ratio algorithms become less efficient, whereas the fluorescence-based algorithms have been demonstrated to be more effective.

Neville and Gower (1977) detected *in vivo* fluorescence signal in Saanich Inlet by using an airborne multichannel spectrometer, and first brought forward an assumption of using fluorescence line height (FLH) to determine chlorophyll-*a* concentration. Afterwards Gower (1980) again demonstrated the possibility of using a shipborne spectroradiometer. Pan *et al*. (1989) first discussed the atmospheric effect on phytoplankton fluorescence, and proposed two suits of reasonable band combinations for fluorescence remote sensing to avoid atmospheric influence. In 1990, Fischer and co-workers (Fischer and Kronfeld 1990, Fischer and Schlüssel 1990) made several studies of oceanic and atmospheric effects on fluorescence line height, such as phytoplankton taxonomy, physiological status, chlorophyll absorption, fluorescence quantum yield, solar zenith angle, yellow substance, suspended matter and atmospheric composition. Gower *et al*. (1999) and Hoge *et al*. (2003) validated FLH algorithms of MERIS and MODIS sensors, respectively. Furthermore, some workers found the ratio between the reflectance peak near 700 nm and another reflectance peak (near 560 nm) or absorption trough (near 670 nm) was considerably correlated with chlorophyll-*a* concentration in mesotrophic and eutrophic waters, and established another method for estimating chlorophyll-*a* concentration (Vasikov and Kopelevich 1982, Vos *et al*. 1986, Kishino *et al*. 1986, Gitelson and Kondrat'ev 1991, Mittenzwey *et al*. 1991, 1992, Gitelson 1992, 1993, Yacobi *et al*. 1995, Gitelson *et al*. 1996, 2000).

2. Methods for investigation

The reflectance spectra of the harmful algal bloom (HAB) species and non-HAB algae were obtained based on *in situ* observation of the HABs in Bohai Sea and measurement of the cultured algae in laboratory tank, respectively. In order to investigate the relations of the reflectance peak near 700 nm with chlorophyll-*a* concentration for different algal species, we chose a typical eight species of algae: the HAB algae *Gymnodinium* sp., *Heterosigma akashiwo* and *Ceratium furca*, and the non-HAB algae *Nitzschia closterium*, *Dicrateria zhanjiangensis Hu.*, *Pyramimonas* sp., *Platymonas* sp. and *Chlorella* sp.

The VF-921B spectroradiometer (Optical Precise Mechanism Institute, Chinese Academy of Science, Anhui, PR China) used in radiance measurements has a working range from 400 to 1040 nm with a spectral resolution of about 5.4 nm and a wavelength precision of 1.0 nm. The reflectance spectra of the cultured algae in the laboratory tank were measured by using a custom-made bucket, with 40 cm depth and 30 cm diameter, covered with dark paint inside. Dense algal liquid was mixed with filtered natural seawater in container. Radiance investigations in Bohai Sea and in the laboratory were taken according to above-water measurement methods recommended by *NASA Ocean Optics Protocols for Satellite Ocean Color Validation, Vol. III* (Mueller *et al.*

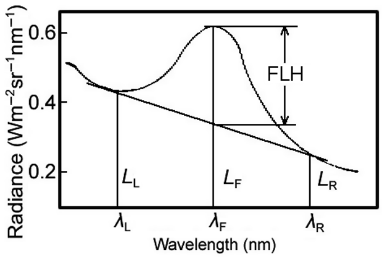

Figure 1. The scheme of fluorescence line height. Here λ_F, λ_L, λ_R are respectively the central wavelength of fluorescence peak and two baseline wavelengths; L_F, L_L, L_R are respectively the radiances in fluorescence peak and two baseline bands.

2003). Simultaneously, seawater of surface layers was sampled and the fluorometric chlorophyll-*a* concentration of seawater samples was determined by using a Turner Designs TD-700 Laboratory Fluorometer and following the method recommended by *NASA Ocean Optics Protocols for Satellite Ocean Color Validation, Vol. 5* (Trees *et al.* 2003). The spectroradiometer and the fluorometer were both strictly calibrated before the experiment.

Determination of the regression relationship between chlorophyll-*a* concentration and fluorescence line height (FLH) is the key process for the chlorophyll fluorescence algorithm. Figure 1 shows a scheme for estimating FLH. At first, a baseline is drawn by a linear connection between radiances of two baseline bands, and then the FLH is obtained by subtracting baseline-determined radiance from fluorescence peak radiance. The expression of FLH is shown as follows.

$$\mathrm{FLH} = L_F - \left[L_R + \frac{\lambda_R - \lambda_F}{\lambda_R - \lambda_L}(L_L - L_R)\right], \tag{1}$$

where λ_F, λ_L, λ_R are respectively the central wavelength of fluorescence peak and two baseline wavelengths. L_F, L_L, L_R are respectively the radiances in fluorescence peak and two baseline bands.

3. Relation between the chlorophyll-*a* concentration and the spectral reflectance

3.1 *Spectral characteristics in the red and NIR bands*

Figure 2 shows different characteristics of spectral reflectance of eight algal species with increasing chlorophyll-*a* concentration. There are three types of distinct spectral shape of the reflectance between 600 and 900 nm. The first is the single-peak type, where there is only a single reflectance peak in the red and near-infrared (NIR) bands, and the peak signal is located at 680–750 nm, such as with the cases of *Heterosigma akashiwo* and *Ceratium furca*. The second is the double-peak type, where there are two distinct reflectance peaks with a main peak around 700 nm and an accessory peak around 800 nm, as in the case of *Gymnodinium* sp. The third is the wide-peak type, such as in the cases of *Platymonas* sp., *Nitzschia closterium* and *Chlorella* sp.; there is a quite wide reflectance peak covering the NIR spectrum from 680 to 900 nm. Since different spectral configurations for certain algal species have a direct effect on the baseline for calculating FLH, the relationships between FLH and chlorophyll-*a* concentration may not be uniform.

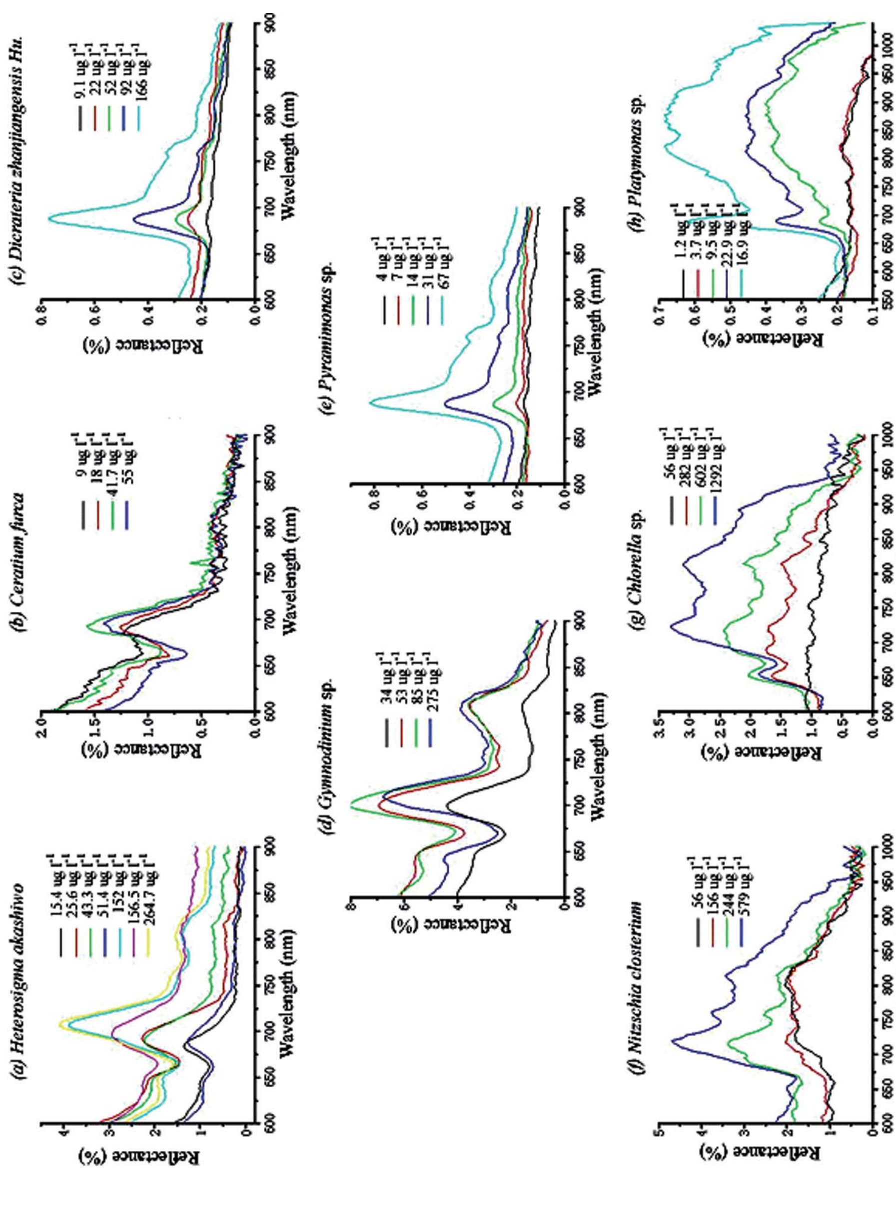

Figure 2. Spectral reflectance characteristics of eight algae species in the red and NIR bands.

The chlorophyll fluorescence occurs only in the wavelength range from 650 to 720 nm. Note that the intensity and position of reflectance peaks and spectral characteristics in NIR are affected by both the phytoplankton reflectance and the exponentially increasing absorption of water molecules (Dall'olmo and Gitelson 2006, Dekker *et al.* 2001, Kutser 2004). The higher the concentration of phytoplankton and/or the closer to the seawater surface it is, the stronger the reflectance peak. The shift of the peak towards longer wavelengths, called the 'red shift' phenomenon, is also caused by the same effect. The reflectance of pure phytoplankton reaches its maximum around 740–750 nm, but in this range the absorption of seawater molecules is so strong that the reflectance of seawater and phytoplankton mixture decreases. For these extremely high chlorophyll concentrations (Chla > 200 mg m^{-3}), we found the phytoplankton forms surface scum and the corresponding reflectance around 750 nm is raised; the spectra are similar to terrestrial plants due to the relative decrease of the seawater effect (Kutser 2004).

3.2 *Relation of chlorophyll-a concentration with the peak near 700 nm*

Figure 3 shows the FLH calculated from equation (1) for three satellite radiometers, MERIS, GLI and MODIS, based on observations of eight species of dominant algae versus the measured data of chlorophyll-*a* concentration. At present, all of the third generation ocean colour radiometers, including MODIS, MERIS and Global Image (GLI), are equipped with sensors of fluorescence bands and two baseline bands. The detailed specifications are listed in table 1. However, the calculated FLH exhibits some difference due to the differences of sensor bands. As shown in figure 3, for some species three FLH values are very close to one another, e.g. for *Platymonas* sp., *Dicrateria zhanjiangensis Hu.* and *Chlorella* sp. However, for some algae, there exist considerable differences among three FLH values, e.g. for *Heterosigma akashiwo* and *Ceratium furca*.

Generally, there is a linear relationship between FLH and chlorophyll-*a* concentration, expressed as:

$$\mathrm{FLH} = a + b([\mathrm{Chla}]) \tag{2}$$

where a denotes the intercept, b denotes the slope, Chla denotes the chlorophyll-*a* concentration with a unit of μg l^{-1}, and FLH denotes the fluorescence line height with a unite of W m^{-2} sr^{-1} nm^{-1}. Based on the observation data, a linear regression analysis for eight algal species and three sensor bands was carried out. Table 2 shows the intercept (a), slope (b) and coefficient of determination (R^2) obtained by the linear regression. Here R^2 denotes the correlation between the FLH calculated from chlorophyll-*a* concentration by using equation (2) and that obtained from spectral reflectance.

For most algal species, the coefficients of determination (R^2) are larger than 0.86, except for *Ceratium furca* and *Heterosigma akashiwo*. It is particularly noted that for some algae, such as *Dicrateria zhanjiangensis Hu.*, *Pyramimonas* sp. and *Nitzschia closterium*, the corresponding coefficients of determination exceed 0.95. However, the intercepts (a) and the slopes (b) in the regression equations are diverse for different algal species. The intercepts actually vary from -0.45×10^{-2} to 53.42×10^{-2} with a unit of W m^{-2} sr^{-1} nm^{-1}. Even for the same species, the intercepts vary intensely with different sensor bands, such as *Gymnodinium* sp. For MERIS, GLI and MODIS sensor bands, the intercepts are 5.46×10^{-2}, 37.67×10^{-2} and 39.81×10^{-2} with a unit of W m^{-2} sr^{-1} nm^{-1} per μg l^{-1}, respectively.

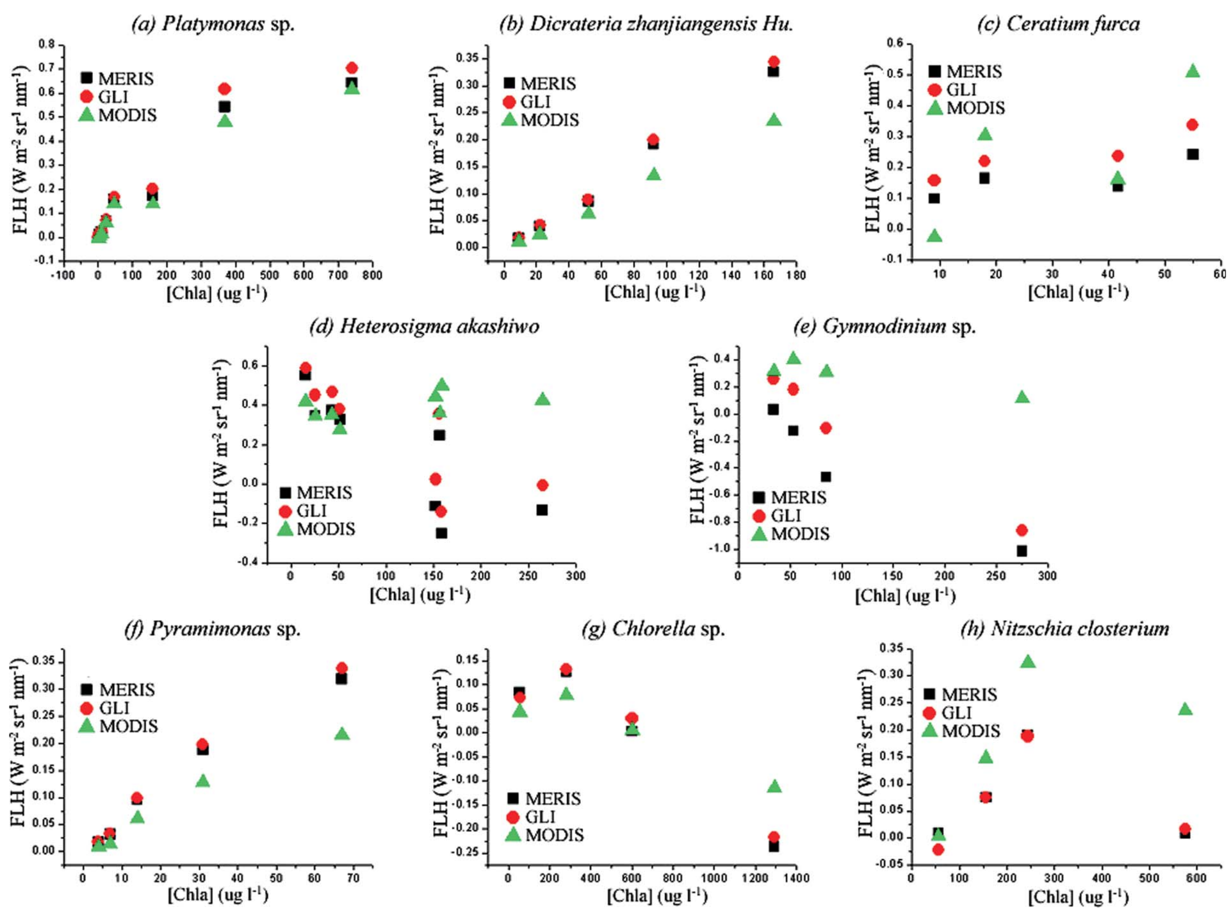

Figure 3. The FLH calculated from equation (1) for three satellite radiometers, MERIS, GLI and MODIS, based on observations in the seawater for eight species of dominant algae versus the measured data of chlorophyll-*a* concentration.

Table 1. Comparison of sensor fluorescence bands. Here NEΔL denotes noise equivalent radiance, PFT denotes proto flight test, EMT denotes engineering model test, and SPC is specification (IOCCG report 2, 1999).

Specification	Left baseline band (λ_L)	Fluorescence peak band (λ_F)	Right baseline band (λ_R)
Band centre (nm)			
MERIS	665	681.25	709
GLI	666.7	679.9	710.5
MODIS	665.1	676.7	746.4
Bandwidth (nm)			
MERIS	10	7.5	9
GLI	10	10	10
MODIS	10	10	10
NEΔL (W m^{-2} sr^{-1} μm^{-1})			
MERIS	0.013	0.014	0.011
GLI	0.015	0.014	0.012
MODIS	0.008	0.007	0.009
Signal/noise ratio			
MERIS(PFT)	708	589	631
GLI(EMT)	863	853	826
MODIS(PFT)	1163	1265	1077

Table 2. The intercept (a), slope (b) and coefficient of determination (R^2) obtained by the linear regression (N is the number of data).

	MERIS			GLI			MODIS			
Species	$a \times 10^{-2}$	$b \times 10^{-2}$	R^2	$a \times 10^{-2}$	$b \times 10^{-2}$	R^2	$a \times 10^{-2}$	$b \times 10^{-2}$	R^2	N
Platymonas sp.	4.88	0.09	0.905	5.43	0.10	0.894	3.60	0.09	0.93	8
Dicrateria zhanjiangensis Hu.	–0.45	0.20	0.995	–0.63	0.21	0.995	–0.47	0.14	0.997	5
Ceratium furca	9.14	0.22	0.606	13.81	0.32	0.8535	–0.68	0.79	0.548	4
Heterosigma akashiwo	46.96	–0.28	0.691	53.42	–0.25	0.6665	34.76	0.04	0.259	8
Gymnodinium sp.	5.46	–0.40	0.927	37.67	–0.46	0.984	39.81	–0.10	0.871	4
Pyramimonas sp.	1.31	0.48	0.975	1.21	0.51	0.978	0.46	0.33	0.971	5
Chlorella sp.	15.55	–0.03	0.914	45.36	–0.03	0.869	8.54	–0.01	0.881	4
Nitzschia closterium	–5.44	0.10	0.965	–8.95	0.11	0.994	–9.91	0.17	0.991	3

In table 2, we can see that the slopes of certain species may be negative, e.g. for *Heterosigma akashiwo*, *Gymnodinium* sp. and *Chlorella* sp. As plotted in figure 3, the values of FLH decreased with increasing chlorophyll-*a* concentration for these species. This demonstrates that the FLH, calculated from equation (2), cannot reflect the reflectance peak signal in the red and NIR part because of the red shift phenomenon. Generally speaking, the reflectance peak is observed near 683 nm for seas with chlorophyll-*a* concentration less than 3 μg l^{-1}. When the concentration increases to 10 μg l^{-1}, the peak position would be observed at 685 nm. If there are blooms of phytoplankton or HABs, the red shift would be more evident. It may shift to 715 nm for concentrations of more than 100 μg l^{-1}. Gitelson (1992) first investigated the red shift of *in vivo* fluorescence peak, and proposed a red shift equation as follows:

$$\text{Peakposition} = 683.51 + 0.268\ (\pm 0.0075)([\text{Chla}]), \tag{3}$$

where [Chla] denotes the chlorophyll-*a* concentration with a unit of $\mu g\ l^{-1}$, and 0.268 is the red shift velocity with a unit of nm per $\mu g\ l^{-1}$ given by Gitelson (1992). However, according to our experiments, we found the red shift velocities of fluorescence peaks for different species are likely to be variable. Although chlorophyll-*a* concentration covered a wide range in our experiments, the reflectance for some algal species such as *Dicrateria zhanjiangensis Hu.* and *Pyramimonas sp*. had a permanent peak position at 687.28 nm, which implies that their red shift velocity is zero. The red shift velocity detected for *Heterosigma akashiwo* is the fastest, reaching 0.263 nm per $\mu g\ l^{-1}$, which is very close to the value given by Gitelson (1992). The fluorescence peak position for other algae shifted to a smaller distance, and the red shift velocities were usually from 0.01 to 0.03 nm per $\mu g\ l^{-1}$.

4. Sensitivity of present satellite fluorescence algorithms

As shown in table 1, there are differences in the band position of the three sensors. For the fluorescence peak band, 676.7 nm of MODIS is the farthest from the actual fluorescence peak. The design aims to avoid the influence of the oxygen absorption band at 687 nm to the greatest extent. The band of 681.25 nm on MERIS is the closest to 685 nm in order to gain more fluorescence signal. Gower *et al*. (2004) showed that MODIS responds to only 57% of fluorescence information, and MERIS can obtain 78%. There is a strong absorption band of water vapour at 730 nm, and to avoid its influence the right baseline band on MODIS is set to its right (746.4 nm), and MERIS and GLI are designed to its left (709 nm and 710 nm). Because the red shift of the reflectance peak would reduce the sensitivity of fluorescence algorithms, Gower *et al.* (2003) noticed that the band of 709 nm on MERIS is able to detect blooms of phytoplankton. They produced a similar FLH, using two bands of 681 nm and 753 nm to obtain baseline and using 709 nm band to get the fluorescence peak, named 'Maximum Chlorophyll Index (MCI)', and demonstrated that MCI could provide considerable sensitivity in eutrophic environments and HAB waters.

The fluorescence algorithms of chlorophyll-*a* concentration for three satellite radiometers, MERIS, GLI and MODIS, can be compared based on the calculation of correlation between FLH of algorithm (2) and that obtained from spectral reflectance. The average R^2 of algorithms based on MERIS, GLI and MODIS are respectively 0.87, 0.90 and 0.81, which demonstrates that MERIS and GLI look more reasonable in band design than MODIS.

5. Conclusion

There are three types of reflectance peak in the red and NIR spectra: single-peak, double-peak and wide-peak. Different types may directly lead to differences in calculating FLH and regression algorithms, and this study plays an important role in selecting the proper position of fluorescence bands on satellite sensors.

In the above investigation, we can clearly see that there is a linear relationship between FLH and chlorophyll-*a* concentration as described by equation (2); hence this equation can be used as a fluorescence algorithm to retrieve the chlorophyll-*a* concentration. Further study indicated that the slope and the intercept of equation (2) are related with algal species, therefore different fluorescence algorithms should be proposed for particular species.

In addition, it is found that the red shift velocities of fluorescence peak for different species are likely to be variable. The fastest red shift velocity was 0.263 nm per μg l^{-1} for *Heterosigma akashiwo*, which is very close to the value given by Gitelson (1992). The fluorescence peak position for other algae shifted to a smaller distance, and the red shift velocities usually were from 0.01 to 0.03 nm per μg l^{-1}.

It is necessary to develop the FLH algorithms of chlorophyll-*a* concentration for different algal species, and this will help us to further understand phytoplankton fluorescence principles and characteristics, and to improve the precision of satellite fluorescence algorithms.

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