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7 **Shift in algal blooms from micro- to macroalgae around China with increasing eutrophication**
8 **and climate change**

9 Yuan Feng¹, Yonglong Xiong¹, Jason M. Hall-Spencer², Kailin Liu³, John Beardall^{1,4,5}, Kunshan Gao¹,
10 Jingke Ge¹, Juntian Xu⁶, Guang Gao^{1,*}

11 ¹State Key Laboratory of Marine Environmental Science & College of Ocean and Earth Sciences,
12 Xiamen University, Xiamen 361005, China

13 ²Marine Institute, University of Plymouth, PL4 8AA, UK and Shimoda Marine Research Center,
14 Tsukuba University, Japan

15 ³College of the Environment & Ecology, Xiamen University, Xiamen 361102, China

16 ⁴School of Biological Sciences, Monash University, Clayton, VI 3800, Australia

17 ⁵Faculty of Applied Sciences, UCSI University, Kuala Lumpur, Malaysia.

18 ⁶Jiangsu Key Laboratory for Marine Bioresources and Environment, Jiangsu Ocean University,
19 Lianyungang 222005, China

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21 Running title: Algal blooms shift from micro- to macroalgae

22 **Abstract:** Blooms of microalgal red tides and macroalgae (e.g. green and golden tides caused by
23 *Ulva* and *Sargassum*) have caused widespread problems around China in recent years, but there is
24 uncertainty around what triggers these blooms and how they interact. Here, we use 30 years of
25 monitoring data to help answer these questions, focusing on the four main species of microalgae
26 (*Prorocentrum donghaiense*, *Karenia mikimotoi*, *Noctiluca scintillans* and *Skeletonema costatum*)
27 associated with red tides in the region. The frequency of red tides increased from 1991–2003 and
28 then decreased until 2020, with *S. costatum* red tides exhibiting the highest rate of decrease. Green
29 tides started to occur around China in 1999 and the frequency of green tides has since been on the
30 increase. Golden tides were first reported to occur around China in 2012. The frequency of
31 macroalgal blooms has a negative linear relationship with the frequency and coverage of red tides
32 around China, and a positive correlation with total nitrogen and phosphorus loads as well as with
33 atmospheric CO₂ and sea-surface temperature (SST). Increased outbreaks of macroalgal blooms are
34 very likely due to worsening levels of eutrophication, combined with rising CO₂ and SST, which
35 contribute to the reduced frequency of red tides. The increasing grazing rate of microzooplankton
36 also results in the decline in areas affected by red tides. This study shows a clear shift of algal
37 blooms from microalgae to macroalgae around China over the past 30 years driven by the
38 combination of eutrophication, climate change, and grazing stress, indicating a fundamental change
39 in coastal systems in the region.

40 **KEYWORDS:** CO₂; eutrophication; golden tides; green tides; red tides; warming

41 **1 INTRODUCTION**

42 Red tides are a type of harmful algal bloom that redden seawater due to the rapid multiplication of
43 phytoplankton including dinoflagellates, diatoms, and cyanobacteria (Anderson, 2009). Toxins
44 produced by red tides, even at relatively low cell densities, can cause illness and mortality in fish,
45 seabirds, and marine mammals (Hallegraeff, 2010). People can also be poisoned if they eat
46 seafood contaminated by red tide toxins. Marine algal toxins are responsible for > 60,000 human

47 toxicity incidents worldwide per year with a mortality rate of 1.5%, associated with the
48 consumption of contaminated seafood and exposure to aerosols of algal toxins (Bourne et al., 2010).
49 Economic losses in the hotel sector at the county-level can reach 15% a month when red tides occur
50 (Bechard, 2019). Therefore, red tides can be harmful to aquatic ecosystems, human health, and the
51 economy. Meanwhile, red tides caused by nontoxic species can also lead to fish kills through
52 oxygen deprivation in the water column as they decay (Xiao et al., 2019). Here we consider blooms
53 as involving cell numbers above a given value, dependent on cell size, as defined in the Methods
54 section (see below). Red tides have a long history, having been recorded in the fossil record, as well
55 as being referenced in the Bible (Anderson, 1997). However, several decades ago red tide algal
56 blooms were infrequent but nowadays wide areas of coastal waters are often impacted by several
57 red tide toxic algal species (Gu et al., 2022; Wang et al., 2023). Reasons for this global expansion of
58 red tides include increased eutrophication, algal transport by ballast water in ships, and the multiple
59 effects of global climate change (Glibert, 2020; Dai et al., 2023).

60 The recent spread of macroalgal blooms worldwide is causing widespread concern (Lapointe et
61 al. 2021; Bermejo et al., 2023). These extensive seaweed blooms are dominated by the genera *Ulva*
62 and *Sargassum*, which cause green tides and golden tides, respectively (Bermejo et al., 2023). The
63 biggest green tide ever recorded occurred in the Yellow Sea of China in 2009, and covered an area
64 of 2100 km² (Xiao et al., 2021). Since 2011, there have been enormous *Sargassum* spp. blooms in
65 the Atlantic Ocean basin causing serious ecological problems and badly affecting tourism (Lapointe
66 et al., 2021). Although bloom-forming macroalgae are not as toxic as red tides, their rapid uptake of
67 nutrients from seawater can cause nutrient limitation for other photosynthetic organisms (Van
68 Alstyne et al., 2015). In addition, hypoxic conditions can be generated by smothering heaps of
69 rotting macroalgal biomass, which is fatal for many marine animals (Young et al., 2022). The poor

70 water quality that results from masses of rotting macroalgae can cause substantial aquaculture
71 losses. For instance, a green tide in China in 2008 caused economic losses of 760–860 million RMB
72 (104–118 million Euros) for just three aquafarms in Shandong province (Ye et al., 2011).

73 Red tides have a long history in China, and scientific reports date back to 1933 (Liang, 2012).
74 Macroalgal blooms, on the other hand have only been recorded as occurring over the past two
75 decades in China. At different times, both increasing and decreasing trends of red tides are apparent
76 (Xiao et al., 2019; Zeng et al., 2019). The reasons that drive the changes have been attributed to
77 water quality and warming (Xiao et al., 2019; Zeng et al., 2019). Previous studies have focused on
78 single types (micro or macro) of algal blooms (Xiao et al., 2019; Bermejo et al., 2023; Dai et al.,
79 2023), ignoring their co-evolution and interactions. However, we assume that red tides and
80 macroalgal blooms may co-evolve and interact with each other along with the changing
81 environment. To test this hypothesis, in this study we compiled data on micro- and macroalgal
82 blooms during 1991–2020 and examined long-term patterns in their frequency, distribution, and
83 duration. A tipping point for algal blooms was identified, a correlation of micro- and macroalgal
84 blooms was found and potential drivers were explored. To the best of our knowledge, this is the first
85 study to investigate the interaction between algal blooms and the shift between micro- and
86 macroalgae over a long period, making a solid contribution to our understanding of algal bloom
87 trends in the Anthropocene.

88 **2 MATERIALS AND METHODS**

89 **2.1 Algal bloom data**

90 Data on red tides were acquired from the Bulletin of Chinese Marine Disasters for 1991–2020
91 issued by the Ministry of Natural Resources of China (former State Oceanic Administration of
92 China) that is publicly available, albeit in Chinese

93 (<https://www.mnr.gov.cn/sj/sjfw/hy/gb/gg/zghyzhgb/>), and from the literature (Liang, 2012; Guo et
94 al., 2015; Li et al., 2023). In addition to the overall trends in red tides, the patterns of four main red
95 tide species, *Prorocentrum donghaiense*, *Karenia mikimotoi*, *Noctiluca scintillans* and *Skeletonema*
96 *costatum*, which contribute about 70% of red tide occurrences in China, were also analyzed (Li et
97 al., 2023). Systematic monitoring of HABs has been conducted in China since 1964 following the
98 establishment of China's State Oceanic Administration. There are 34 monitoring areas for red tides
99 across the country. The HABs monitoring standard in these monitoring areas involve routine
100 monitoring, including four surveys per month during high-risk seasons and one to two surveys per
101 month during other seasons, while contingent monitoring includes surveys every 3 days when
102 harmful algal blooms were expected to occur, and tracking monitoring to be conducted daily when
103 blooms were detected. The same methods have been used annually within these monitoring areas
104 since 1970 (Xiao et al., 2019). All monitoring tasks included sample collection to determine
105 phytoplankton species density. The occurrence of a bloom is determined to be when cell density
106 exceeds size-dependent thresholds set in the standard procedure, which is $> 10^7$, $> 10^6$, $> 2 \times 10^5$, $>$
107 10^5 and $> 3 \times 10^3$ cells dm^{-3} for algae with cell lengths of < 10 , 10–29, 30–99, 100–299 and 300–1000
108 μm , respectively (HY/T 069-2005). We have to point out that red tide events that occur outside of
109 the monitoring areas are not included in data used in this study. This may lead to the
110 underestimation of red tide occurrence. However, the monitoring areas cover the coastal waters
111 across China where red tides usually occur, based on historical records. Therefore, the
112 underestimation should be limited and would not impact the variation trend of red tides.

113 Data on transregional and local green tides were collated from the Bulletin of Chinese Marine
114 Disasters 2008–2020 and the published literature (Liu, 2008; Ma et al., 2010; Wei et al., 2011; Xing
115 et al., 2016; He et al., 2019; Lei et al., 2022; Zeng et al., 2023). Data on golden tides were acquired

116 from the following published works: Qi et al. (2017), Xing et al. (2017), Ding et al. (2019), Liu et
117 al. (2021a), Song et al. (2022b), and Zheng et al. (2022). After integrating the thresholds in China
118 and abroad, the occurrence of a local green tide is determined to be when coverage exceeds 10, 000
119 m² and coverage percentage (the ratio of coverage to distribution) exceeds 1%. Hereafter,
120 "frequency" means the number of algal outbreaks during a particular period, "distribution" is the
121 regional area over which blooms spread, while "coverage" is the size of the blooms themselves and
122 excludes bloom-free areas in the regional distribution.

123 **2.2 Environmental data**

124 Nutrient loads from rivers were obtained from Wang et al. (2021) who used an Integrated Model
125 to Assess Global Environment-Global Nutrient Model (IMAGE-GNM) (0.5° × 0.5° resolution).
126 Nutrient load from mariculture includes that from fish, prawn and crab farms. The nutrient load
127 from fish farms was calculated according to Xiong et al. (2022) and that from prawn and crab farms
128 was calculated according to the N and P budget (Ban, 2015). The total nitrogen (TN) and total
129 phosphorus (TP) loads were used to calculate the molar N:P each year for riverine and mariculture
130 inputs. The production of fish, prawns and crabs was obtained from the China Fishery Statistical
131 Yearbook for the years 1991–2020 (Table S1). The ratios of dissolved silicate (DSi) to dissolved
132 organic nitrogen (DIN) were obtained from Liu et al. (2022) and Zhang et al. (2022). Carbon
133 dioxide data were acquired from China Greenhouse Gas Bulletin for the years 1991–2020, issued
134 by the China Meteorological Administration. Atmospheric CO₂ concentrations were measured at
135 Mount Waliguan, the only station of the World Meteorological Organization/Global Atmosphere
136 Watch in Eurasia. Sea surface temperature (SST) data (1991–2020) were derived from the Met
137 Office Marine Data Bank (Table S2)
138 (<http://www.metoffice.gov.uk/hadobs/hadisst/data/download.html>), which has a resolution of 1.0° ×

139 1.0°. Monitoring data for SST in all 11 coastal administrative areas of China were used to calculate
140 annual means in coastal areas (Table S2).

141 **2.3 Grazing rate**

142 The grazing pressure on phytoplankton is mainly from microzooplankton that are defined as
143 heterotrophic and mixotrophic organisms with a size range of 20–200 μm . The microzooplankton
144 consists of ciliates, heterotrophic and mixotrophic dinoflagellates, as well as mesozooplankton
145 nauplii (Sieburth et al., 1978; Liu et al., 2021b). It has been reported that microzooplankton consume
146 about 59–75% of daily primary production (Calbet & Landry, 2004; Calbet, 2008). As such, we used
147 their grazing rates to evaluate the top-down controls on phytoplankton by grazers. The
148 microzooplankton grazing rate was estimated using a machine-learning technique known as a
149 ‘Boosted Regression Trees’ model, which has been increasingly used in ecological studies (Elith &
150 Leathwick 2017). The BRT model was derived based on a published data set consisting of
151 microzooplankton grazing rate and related environmental parameters in China’s marginal seas,
152 including the South China Sea, the East China Sea, and the Yellow Sea (Liu et al., 2020). As
153 temperature and Chl *a* concentrations have been reported as the primary factors determining
154 microzooplankton grazing rate, we used them as predictors for the BRT model. The BRT was
155 implemented using the function ‘gbm.step’ in the R package ‘dismo’. The tree complexity was set as
156 1 and the learning rate was set as 0.001 to allow the model to approach the best performance slowly
157 with more trees to reduce error. Other settings remained as the defaults in ‘gbm’ (Elith & Leathwick,
158 2017). To evaluate the prediction accuracy of the model, 20% of data from the data set were randomly
159 selected as testing data and excluded from the model fitting. The final fitted BRT models for the
160 training data had 3700 trees, enabling the model to approach the plateau very slowly (Fig. S1). The
161 model performs well, and Pearson’s correlation coefficient (r) for the model predictions and the

162 testing data is 0.52 ($p < 0.001$, Fig. S1). We then used this model to predict the microzooplankton
163 grazing rate of each Province in each year (1997–2020). The sea surface temperature and Chl *a* in all
164 11 coastal administrative areas of China were input to the model as predictors. Sea surface
165 temperature (Table S2) and Chl *a* (Table S3) data were derived from the Met Office Marine Data
166 Bank and NOAA, respectively. Microzooplankton grazing rate (d^{-1}) in each coastal administrative
167 area (Table S4) was weighed based on their area proportion to calculate annual means. The data after
168 1997 were presented since Chl *a* data were not monitored before 1997.

169 **2.4 Statistical analysis**

170 The method used to identify trends in red tide coverage around China followed Ban et al.
171 (2022). We used the Mann–Kendall trend test to analyze whether trends changed significantly ($p <$
172 0.05) due to a sustained increasing trend switching abruptly to a decreasing trend, i.e. a tipping
173 point.

174 Linear and nonlinear regressions were used for our time series analysis. Pearson correlations of
175 multiple variables was used to analyze potential relationships between microalgal and macroalgal
176 blooms and the environmental factors including total nitrogen, total phosphorus, atmospheric CO₂,
177 annual average sea surface temperature and grazing rate. Adjusted R-squared was used to avoid the
178 false improvement of the model due to increased independent variables.

179 **3 RESULTS**

180 Figure 1 shows the widespread distributions of micro and macroalgal blooms around China in
181 1991–2020. Red tides have occurred off all coasts of China, with the East China Sea having the
182 highest frequency (58%) and the Yellow Sea having the lowest frequency (10%) during the period
183 1991–2020. Since 2006, huge transregional green tides occurred in the Yellow Sea while golden
184 tides have affected large areas in the East China Sea and the Yellow Sea. More localised green tides

185 have occurred in coastal waters of the Bohai Sea (Yingkou, Qinhuangdao, Tangshan), the Yellow
186 Sea (Yantai, Nantong), the East China Sea (Ningbo, Xiamen) and the South China Sea (Shantou,
187 Shenzhen, Zhanjiang, Beihai, Lingshui, Sanya) where red tides also occurred.

188 To show more detail on red tides, the distribution of red tides caused by the four main species
189 in China is shown (Fig. 2). The occurrence of red tides caused by *Prorocentrum donghaiense* was
190 confined to the coastline of the East China Sea (Fig. 2a). *Karenia mikimotoi* dominated red tides
191 also occurred mainly in the coastal waters of the East China Sea but could also be found in the
192 Bohai Sea and the South China Sea (Fig. 2b). In contrast, *Noctiluca scintillans* red tides mainly
193 occurred in the Bohai Sea, although they could be detected in the other three seas (the Yellow Sea,
194 the East China Sea and the South China Sea) (Fig. 2c). *Skeletonema costatum* red tides had a
195 relatively even distribution along the coastline of China (Fig. 2d) and could be detected in the
196 southernmost location of Hainan province.

197 The frequency (number of blooms in a given year) of red tides first decreased and then
198 exponentially increased during 1991–2003 ($F_{(3, 12)}=92.142, p<0.001, r^2=0.907$), with an overall
199 increasing rate of 7 times yr^{-1} (Fig. 3a). After 2003, their frequency decreased yearly until 2020 at a
200 decreasing rate of 5 times yr^{-1} ($F_{(1, 16)}=46.825, p<0.001, r^2=0.729$). Red tide coverage (Fig. 3b)
201 increased by 1896 $\text{km}^2 \text{yr}^{-1}$ during 1991–2005 ($F_{(3, 14)}=101.754, p<0.001, r^2=0.918$) then decreased
202 by 1687 $\text{km}^2 \text{yr}^{-1}$ from 2005–2020 ($F_{(1, 14)}=51.204, p<0.001, r^2=0.770$). The Mann-Kendall-Tau test
203 ($Z=-4.187, p <0.001$) shows that a tipping point in the trend of red tides occurred in 2005.

204 No macroalgal blooms were recorded in China until 1999 when transregional green tides
205 started to occur in the Yellow Sea (Fig. 3c). Thereafter, the frequency of macroalgal blooms
206 increased linearly ($F_{(1, 21)}=596.319, p<0.001, r^2=0.964$) and reached 13 blooms per year in 2020.
207 Local green tides have occurred annually since 2003, while transregional golden tides have

208 occurred annually since 2012, stretching across the East China Sea and the Yellow Sea. The
209 increasing frequency of macroalgal blooms with year is mainly due to local green tides because no
210 local golden tides have been reported. When analyzing outbreaks of micro- and macroalgal blooms
211 (Fig. 3d), a significant negative relationship was found for the frequency between green+golden and
212 red tides during 2005–2020 ($F_{(1, 14)}=15.426, p=0.002 < 0.05, r^2=0.490$). The increase in the
213 frequency of green+golden tides was accompanied by a decrease in red tide coverage during 2005–
214 2020 ($F_{(1, 14)}=42.916, p<0.001, r^2=0.736$). These results suggest competition between micro- and
215 macroalgal blooms.

216 The frequency of red tides caused by the four main species during 1991-2020 was further
217 analyzed (Fig. 4). Red tide events caused by *Prorocentrum donghaiense* increased exponentially
218 during 1999–2004 ($F_{(3, 13)}=29.286, p<0.001, r^2=0.831$), with an overall increase rate of 6 times yr⁻¹
219 (Fig. 4a). After 2004, its frequency shows a decreased trend ($F_{(1, 15)}=6.211, p=0.025, r^2=0.246$). The
220 pattern for *Karenia mikimotoi* is similar to *P. donghaiense* although the decrease after the peak in
221 2006 was not linear ($F_{(1, 13)}=4.298, p=0.059, r^2=0.191$, Fig. 4b). The frequency of red tides caused
222 by *Noctiluca scintillans* decreased first during 1990-1996 and then increased to a peak in 2003 ($F_{(3, 12)}$
223 $=34.153, p<0.001, r^2=0.821$, Fig. 4c). Thereafter, it also shows a decreasing trend in spite of large
224 fluctuations ($F_{(1, 16)}=3.380, p=0.085, r^2=0.123$). In terms of *Skeletonema costatum* (Fig. 4d), its red
225 tides increased during 1997-2006 ($F_{(3, 15)}=40.468, p<0.001, r^2=0.831$) but decreased linearly after
226 that ($F_{(1, 13)}=22.803, p<0.001, r^2=0.609$). Frequencies of red tides caused by the four species all
227 experienced two stages, with an increase in the first stage but a decrease in the second stage. In the
228 second stage, *S. costatum* had the highest rate of decrease while *N. scintillans* had the lowest
229 decrease rate based on the slope values.

230 The coverage and distribution of green tides in China are shown in Figure 5a. The coverage of

231 green tides during 1999-2007 was small, ranging from 1.83–57 km². The area affected by green
232 tides increased to a coverage of 653 km² in 2008 and then reached a peak of 2,103 km² in 2009.
233 Afterwards it has since fluctuated between 198–793 km² during 2010–2020. The regional
234 distribution of green tides was 82 times larger than the coverage of the blooms themselves. Golden
235 tides were first recorded off the Zhejiang coast in the East China Sea in 2012, with a coverage of
236 33.5 km² (Fig. 5b). These golden tides had a coverage of 534 km² in Chinese waters in 2017 and are
237 now an annual problem, with a coverage of 75.16 km² in 2020. The regional distribution of these
238 golden tides was, on average, 1,284 times larger than the coverage of the blooms themselves.

239 Seasonal characteristics of algal bloom occurrences were also analyzed (Fig. 5c&d). Red tides
240 have occurred in all months of a year (Fig. 5c). May had the highest frequency (442) of red tides in
241 China during 1991–2020, followed by June (334), while December had the lowest frequency (4),
242 followed by November (21). The seasonal pattern of coverage is similar to that of bloom frequency,
243 being highest in May (105,898 km²) and lowest in December (936 km²). Unlike red tides,
244 transregional green tides occurred in specific seasons (Fig. 5d). They usually began in March or
245 April and ended in July or August. This pattern lasted until September in 2019. Compared to
246 transregional green tides, local green tides had a shorter duration, usually 2–3 months. Due to their
247 wide distribution, from Liaoning province in the North of China to Hainan province in the South of
248 China, the overall duration of all local green tides is very long, with a narrow temporal gap in
249 October and November. Golden tides usually began in March and end in June, but sometimes occur
250 in winter (Fig. 5d).

251 Total Nitrogen (TN) load discharged from rivers into coastal waters increased from 5.06 Tg in
252 1991 to 11.03 Tg in 2018 and levelled-off to 10.89 Tg in 2020 (Fig. 6a). In contrast, the TN load
253 from mariculture has continued to increase during 1991–2020 and reached 0.33 Tg in 2020 (Fig.

254 6b). The pattern of Total Phosphorus (TP) load was similar to TN although that from mariculture
255 had a higher percentage of the total load (0.19–8.47%) compared to TN (0.03–2.92%). The N:P
256 ratio of riverine inputs decreased from 30.16 in 1991 to 28.57 in 2020 (Fig. 6c). The total N:P
257 (average from all sources) also showed a decreasing trend, from 29.66 in 1991 to 26.94 in 2020.
258 The N:P ratio from mariculture effluents was much lower than that of rivers, fluctuating from 6.80
259 to 11.28 during 1991–2020. In 2005, when coverage of red algal blooms switched abruptly from an
260 increasing to a decreasing trend, the TN, TP and N:P ratio were 8.61 Tg yr⁻¹, 0.76 Tg yr⁻¹ and 29.15
261 respectively.

262 The variation trends of DSi/DIN were also analyzed (Fig. 7). The DSi/DIN ratio in the Bohai
263 Sea linearly decreased from 0.59 to 0.53 during 2003-2020 ($F_{(1, 16)}=7.539, p=0.014, r^2=0.278$, Fig.
264 7a). While the DSi/DIN ratio in the Yellow Sea did not show a significant decrease trend ($F_{(1, 16)}=0.004, p=0.951, r^2=-0.062$, Fig. 7b), it decreased from 0.69 in 2003 to 0.64 in 2020 in the
265 Changjiang River estuary ($F_{(1, 16)}=8.039, p=0.012, r^2=0.293$, Fig. 7c) and also decreased from 3.69
266 in 1990 and to 0.88 in 2016 in the Pearl River estuary ($F_{(1, 16)}=50.101, p<0.001, r^2=0.743$, Fig. 7d).
267 As show in Fig. 1, the Changjiang River estuary and Pearl River estuary are two areas that suffer
268 from severe red tides.
269

270 Atmospheric CO₂ concentrations increased from 354.7 ± 0.10 ppm in 1991 to 414.53 ± 0.20
271 ppm in 2020, with a rising rate of 1.99 ppm yr⁻¹ ($F_{(1, 28)}=7637.835, p<0.001, r^2=0.996$) and was at
272 379.7 ± 0.08 ppm when red tide coverage started to decline (Fig. 8a). The mean sea surface
273 temperature for the coastal waters of China during 1991–2020 ($F_{(1, 28)}=6.008, p=0.021, r^2=0.147$)
274 increased from 18.63 °C in 1991 to 19.13 °C in 2020, in spite of fluctuations (Fig. 8b). The mean
275 SST of all Chinese coastal waters combined was 18.68 °C in 2005 when the coverage of red tides
276 started to fall and be replaced by nuisance macroalgal blooms.

277 In addition to abiotic factors, biotic factors were also analyzed (Fig. 9). Chl *a* concentration of
278 coastal waters around China increased from 3.87 in 1997 to 4.83 mg m⁻³ in 2020 ($F_{(1, 22)}=15.051$,
279 $p<0.001$, $r^2=0.379$, Fig. 9a). Similar to Chl *a* concentration, zooplankton grazing rate of coastal
280 waters around China also increased from 0.548 to 0.583 d⁻¹ during the period of 1997-2020 ($F_{(1,$
281 $22)}=45.415$, $p<0.001$, $r^2=0.659$, Fig. 9b). The Chl *a* concentration and grazing rate were 3.78 mg m⁻³
282 and 0.523 d⁻¹, respectively when the coverage of red tides started to fall.

283 We used Pearson correlation analysis for multiple variables from 2005–2020, the period over
284 which red tides have been diminishing whereas macroalgal blooms have increased (Fig. 10). The
285 coverage of red tides had significant negative correlations with the frequency of macroalgal blooms,
286 TN, TP, CO₂, and zooplankton grazing rate ($p<0.01$) over this time period. Among them, TN (-
287 0.926) and TP (-0.923) had a very high correlation coefficient with the coverage of red tides,
288 followed by CO₂ (-0.879) and macroalgal blooms (-0.868). *Prorocentrum donghaiense* or *Noctiluca*
289 *scintillans* did not have significant correlations with biotic or abiotic factors, while *Karenia*
290 *mikimotoi* (-0.613) and *Skeletonema costatum* (-0.826) had significant negative correlations with the
291 frequency of macroalgal blooms. In contrast, the frequency of macroalgal blooms positively
292 correlated with TN, TP, CO₂, SST and zooplankton grazing rate ($p<0.05$ for SST and $p<0.01$ for the
293 others). Among them, CO₂ (0.962), TP (0.955) and TN (0.950) had very high correlation
294 coefficients with the frequency of macroalgal blooms. Sea surface temperature positively correlated
295 with rising atmospheric CO₂ levels at the 0.01 level of statistical significance and with TN and TP
296 at the 0.05 level of statistical significance. Zooplankton grazing rate positively correlated with TN,
297 TP, and CO₂ ($p<0.01$).

298 **4 DISCUSSION**

299 **4.1 Trends of red tides and the associated reasons**

300 The frequency of red tides increased rapidly around China during 1995–2003, which has been
301 attributed to the rising discharge of inorganic nutrients from agriculture, sewage and other land-
302 based sources (Wang et al., 2021). The frequency and coverage of red tides have decreased since
303 2003 and 2005 respectively, despite a continuing increase in nutrient loads. This decreasing trend
304 was also reported in previous study (He et al., 2021; Sakamoto et al., 2021). A large number of
305 studies have shown that eutrophication and warming can stimulate the occurrence of red tides
306 (Heisler et al., 2007; Xiao et al., 2019). The observed increases in total nitrogen, total phosphorus
307 and temperature would not, therefore, be expected to lead to declines in the frequency and coverage
308 of red tides unless some other factor has been driving the negative correlations between red tides
309 and these environmental factors over the past 16 years.

310 Coverage of red tides was significantly negatively correlated with macroalgal bloom frequency,
311 suggesting that the shrinkage of red tides since 2005 may be caused by increased outbreaks of
312 macroalgal blooms around China, particularly because red tides and green tides overlap in both
313 their distribution and seasonal occurrence. Microalgae and macroalgae are known to compete for
314 nutrients, light, and space in coastal waters (Besterman & Pace, 2018; Gao et al., 2021). Moreover,
315 *Ulva* species (commonly responsible for green tides) can inhibit photosynthesis and growth of red
316 tide microalgae via allelopathic effects (Sun et al., 2016; Gao et al., 2019). The inhibitory effects of
317 *Ulva* species on red tide microalgae have been reported, based on both laboratory and field work
318 (Gao et al., 2019; Imai et al., 2021; Li et al., 2022). The fastest decrease of *S. costatum* dominated
319 red tides may be related to its distribution. Compared to *P. donghaiense* and *K. mikimotoi*, *S.*
320 *costatum* red tides have a wider distribution, from the southernmost to the northernmost provinces.
321 The wider distribution suggests that there is a greater chance that they would be inhibited by
322 macroalgal blooms. Although *N. scintillans* red tides are also distributed from South to North of

323 China seas, *N. scintillans* is a heterotrophic dinoflagellate that does not compete with *Ulva* and
324 *Sargassum* species for light and inorganic nutrients. Therefore, it is less likely to be inhibited by
325 macroalgal blooms, showing the lowest decrease rate. In addition, *N. scintillans* feeds on diatoms
326 and the increased temperature could stimulate its grazing rate and thus growth (McLeod et al.,
327 2012; Sheng et al., 2022). Allelopathic inhibition cannot explain the differential decreases of red
328 tides caused by the four microalgal species since allelochemicals from *U. prolifera* have lower
329 inhibition rates on *S. costatum* compared to *P. donghaiense* (Sun et al., 2019).

330 **4.2 Trends of macroalgal blooms and the associated reasons**

331 In contrast to the situation with red tides, the frequency of macroalgal blooms has increased
332 each year since they first began around China in 2006. The primary reason for this can be attributed
333 to increases in nutrient load. Large biomass macroalgal blooms require exceptionally high levels of
334 nutrients in the surface seawater. That is a key reason why most macroalgae are restricted to coastal
335 areas where nutrient levels are usually higher than in surface waters of the open ocean. Although
336 microalgae have much higher growth rates than macroalgae under nutrient-limiting conditions,
337 macroalgae can outcompete phytoplankton at high levels of nutrients (Fong & Zedler, 1993; Nan &
338 Dong, 2004). Microalgae can acclimate to nutrient-limiting conditions better than macroalgae
339 because their larger surface area to volume ratio enhances nutrient uptake.

340 Increased CO₂ may also contribute to outbreaks of macroalgal blooms around China. Carbon
341 dioxide can limit the photosynthesis and growth of algae due to its low concentrations and slow
342 diffusion rates in seawaters (Cornwall et al. 2017; Raven et al., 2017). This limitation is particularly
343 serious for macroalgae and larger microalgae because they have thicker diffusive boundary layers
344 which hinder CO₂ uptake by their cells (Finkel et al., 2010; Noisette & Hurd, 2018). Therefore,
345 macroalgae or larger microalgae could benefit more from increased CO₂ (Wu et al., 2014).

346 However, more dissolved CO₂ dissolved leads to ocean acidification, which has a range of effects
347 on seawater carbonate chemistry that can negatively affect some algae (Flynn et al., 2012; Hong et
348 al., 2017; Shi et al., 2019) but stimulate the growth of others, triggering major shifts in coastal algal
349 communities (Cornwall et al. 2017; Harvey et al. 2019). In the coastal waters of China, where
350 chronic and increasing eutrophication has now combined with rising CO₂ levels, blooms of fast-
351 growing and opportunistic macroalgae such as *Ulva* spp. have replaced microalgal red tide blooms,
352 i.e., one problem has been replaced by another. In terms of red tide species, it has been reported that
353 CO₂ enrichment increases the maximum population density and carrying capacity of the
354 raphidophyte alga *Heterosigma akashiwo* but decreases these parameters for *S. costatum* (Zheng et
355 al., 2016). Furthermore, growth of *P. donghaiense* is accelerated more strongly than the diatom
356 *Conticribra weissflogii* by simultaneous acidification and eutrophication (Xu et al., 2010). These
357 findings indicate that flagellate algae may be more adaptable to increased acidification and
358 eutrophication than diatoms, which can explain the slower decrease of dinoflagellate red tides in
359 frequency compared to diatom red tides shown in this study.

360 Temperature may also contribute to the occurrence and expansion of green and golden tides.
361 Compared to many other macroalgae, *Ulva* and *Sargassum* species have higher tolerance to
362 increased temperature (Jiang et al., 2022). While other macroalgae such as kelps may die back due
363 to warming or marine heatwaves (Straub et al., 2019; Agostini et al., 2021; Gao et al., 2021),
364 increased SST can stimulate the growth of *Ulva* and *Sargassum* species (Gao et al., 2017; Sanchez-
365 Rubio et al., 2018). In addition, increased SST can result in the extension of green tides northward,
366 helping to explain the recent occurrence of green tides in the Bohai Sea, an example of coastal
367 ecosystem simplification in the face of multiple stressors (Agostini et al., 2021).

368 Unlike the continuous increase in the frequency of macroalgal blooms over time, the coverage

369 of macroalgal blooms showed large interannual variability, which could be attributed to human
370 intervention. Since 2016, *Ulva* and *Sargassum* biomass has been collected at the early stages of
371 bloom formation in the Southern Yellow Sea, limiting large-scale proliferation. Since 2019, some
372 measures have also been taken to prevent the outbreak of green tides, such as reducing the densities
373 of *Porphyra* cultivation rafts, and using antifouling chemicals to kill juvenile *Ulva* (Sun et al.,
374 2022). Competition between golden tides and green tides also contributes to annual fluctuations
375 because they overlap in their distribution (Song et al., 2022a, b). It is also notable that the golden
376 tides are less responsive to eutrophication than green tides and pelagic species of *Sargassum* are
377 known to be adapted to oligotrophic conditions (Devault et al., 2021).

378 Mean atmospheric CO₂ and sea surface temperature around China have increased by 140 ppm
379 and around 1.7 °C relative to preindustrial levels, respectively (Xiao et al., 2019). This level of
380 warming above preindustrial temperatures already lies within the lower end of five climate tipping
381 point uncertainty ranges (Armstrong McKay et al., 2022). Six climate tipping points, including
382 collapse of the Greenland and West Antarctic ice sheets, die-off of low-latitude coral reefs, and
383 widespread abrupt thawing of permafrost, become likely within the Paris Agreement range of 1.5 to
384 <2°C warming (Armstrong McKay et al., 2022). The present study indicates that it is likely that
385 increased CO₂ and temperature combined with increased nutrient load triggered a tipping point in
386 the coastal seas of China, leading to a shift from microalgal- to macroalgal-dominated eutrophic
387 blooms. Previous laboratory work has also demonstrated that eutrophication combined with
388 increasing CO₂ and temperature can stimulate the settlement, germination, and growth of
389 microscopic propagules of bloom-forming *Ulva* species, suggesting that more intense outbreaks of
390 green tides are likely to become the "new normal" around Chinese coasts (Gao et al., 2017, 2018).
391 In addition to the environmental factors mentioned above, the expansion of *Porphyra* aquaculture

392 may also contribute to the increased outbreak of floating green tides in the Yellow Sea as the
393 accumulation and disposal of *Ulva prolifera* from *Porphyra* aquaculture rafts is the most likely
394 source of the initial floating biomass (Keesing et al., 2011; Wang et al., 2015).

395 **4.3 The top-down effects on micro- and macroalgal blooms**

396 In addition to the bottom-up effect, top-down effects may also contribute to the shift of algal
397 blooms. Microzooplankton grazing rate rose with year due mainly to the increased SST and Chl *a*
398 since these two factors can influence grazing rate directly (Chen et al., 2021). In addition, our result
399 shows that microzooplankton grazing rate also correlated positively with CO₂ level. This is
400 consistent with the finding of laboratory experiments showing that increased CO₂ stimulated
401 grazing rate of zooplankton (Li & Gao, 2012). The increased grazing rate of zooplankton could
402 contribute to the decreased occurrence and coverage of red tides. The microzooplankton in this
403 study, that consists of ciliates, heterotrophic and mixotrophic dinoflagellates and mesozooplankton
404 nauplii, can graze any phytoplankton smaller than 200 μm. Their grazing activities are size-
405 dependent rather than species-dependent (Hansen et al., 1994; Zhou et al., 2015). Therefore, the
406 grazing pressure from microzooplankton may contribute little to the faster decrease of diatom-
407 dominated red tides. In terms of macroalgae, when floating on surface seawater they are
408 inaccessible to benthic grazers and benefit from reduced grazing pressure. Although some epiphytic
409 animals (e.g. gammarid species) feed on floating macroalgae, they only consume a very small
410 proportion of increased biomass of floating macroalgae (Wang et al., 2020; Miao et al., 2021). In
411 addition, the *U. prolifera* fragments resulting from gnawing by *Apothyale* sp. had a higher growth
412 rate than individual thalli, which may lead to higher floating biomass (Miao et al., 2021).

413 **5 CONCLUSIONS**

414 To summarize, increasing eutrophication combined with rising CO₂ and temperature may play

415 an important role as the cause of widespread outbreaks in macroalgal blooms around China since
416 2006, with warming expanding the northward distribution of these macroalgal blooms. This increased
417 occurrence of macroalgal blooms has been accompanied by a shrinkage of red tides, likely due to
418 competition and inhibition. In addition, increasing grazing rate of microzooplankton also contributes
419 to the decreased frequency and coverage of red tides. While smaller phytoplankton will likely
420 increasingly dominate surface waters of the open ocean, due to warming and increasing nutrient
421 limitation (Falkowski & Oliver, 2007; Gao et al., 2021), this is not the case in coastal areas where
422 people are more directly affected. Around coasts already impacted by eutrophication, due to nutrient
423 inputs from land and mariculture, increasing levels of CO₂ allow macroalgae to compete strongly for
424 resources (Celis-Pla et al., 2015). Macroalgal blooms seem less harmful to human health than
425 microalgal blooms, since they have not been found to excrete toxins. However, the massive biomass
426 of seaweed blooms can have severe impacts on mariculture and tourism and they can produce toxic
427 hydrogen sulphide from their anoxic interior when decaying, leading to the death of aquatic and
428 terrestrial animals (Smetacek & Zingone, 2013). Thus, monitoring the development of local green
429 tides is very essential in future since they do not draw enough attention. In addition, we have to
430 develop strategies to reduce nutrient loads into coastal waters, as well as reducing overall CO₂
431 emissions. The improvement of agriculture and a more sustainable food system is needed since of
432 nine planetary boundaries (PBs) agriculture is the major driver of four PBs (biosphere integrity,
433 biogeochemical flows, land-system change and freshwater use) that are in the high or increasing risk
434 zones and a significant driver of climate change that is at increasing risk (Campbell et al., 2017).

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448 **COMPETING INTERESTS**

449 The authors declare no competing interests.

450 **DATA AVAILABILITY STATEMENT**

451 The data underpinning the analyses presented here are openly available from Figshare at
452 <https://doi.org/10.6084/m9.figshare.24297757> (Feng et al., 2023).

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748 **Figure legends**

749 **Fig. 1 Distribution of marine algal blooms around China during 1991–2020.** Red and green
750 circles represent localized red and green tides, respectively. Green and brown dashed lines show the
751 extent of very extensive transregional green and golden tides, respectively. The pie chart shows the
752 percentage occurrence of red tides in four seas (BS: Bohai Sea, YS: Yellow Sea, ECS: East China
753 Sea, SCS: South China Sea) during 1991–2020. CRE and PRE represent the Changjiang River
754 estuary and Pearl River estuary, respectively. Map lines delineate study areas and do not necessarily
755 depict accepted national boundaries.

756 **Fig. 2 Distribution of four types of main red tides along with macroalgal blooms around**
757 **China during 1991–2020.** (a) *Prorocentrum donghaiense* dominant red tides, navy circles; (b)
758 *Karenia mikimotoi* dominant red tides, grey circles; (c) *Noctiluca scintillans* dominant red tides,
759 pink circles; (d) *Skeletonema costatum* dominant red tides, red circles. Green circles represent
760 localized green tides, respectively. Green and brown dashed lines show very extensive transregional
761 green and golden tides, respectively. Map lines delineate study areas and do not necessarily depict
762 accepted national boundaries.

763 **Fig. 3 Frequency and coverage of red tides and their relationship with macroalgal blooms**
764 **around China.** (a) frequency of red tides; (b) coverage of red tides; (c) frequency of macroalgal
765 blooms; (d) relationships between red tide frequency (filled squares), red tide coverage (open
766 circles) and macroalgal bloom frequency.

767 **Fig. 4 Frequency of red tides caused by four main species around China during 1991–2020.** (a)
768 *Prorocentrum donghaiense*; (b) *Karenia mikimotoi*; (c) *Noctiluca scintillans*; (d) *Skeletonema*
769 *costatum*.

770 **Fig. 5 All plots based on data from the seas around China from 1991–2020;** a) coverage and
771 regional distribution of green and b) golden tides; c) seasonal frequency and coverage of red tides;

772 d) box plots of the timing of macroalgal blooms. Green tides I and II represent transregional and
773 local blooms, respectively. "Distribution" is the regional area over which blooms spread while
774 "coverage" is the actual size of blooms and excludes bloom-free areas in the regional distribution.

775 **Fig. 6 Total Nitrogen (a) and Total Phosphorus (b) contributions from riverine and**
776 **mariculture sources to coastal waters and their ratio (c) around China during 1991–2020.**

777 "Total" means the sum of river and mariculture contributions. Red dashed vertical lines indicate the
778 timing of a tipping point in red tides when they started to decrease, following a decade of increasing
779 frequency.

780 **Fig. 7 Variation of molar DSi/DIN ratios in the Bohai Sea (a), Yellow Sea (b), Changjiang**
781 **River estuary (c) and Pearl River estuary (d).**

782 **Fig. 8 Mean sea surface temperature (SST) of coastal waters around China and atmospheric**
783 **CO₂ in this region during 1991–2020.** Vertical red dashed lines indicate when red tides started to
784 become less frequent.

785 **Fig. 9 Mean Chl *a* concentration and zooplankton grazing rate in coastal waters around**
786 **China during 1997–2020.** Vertical red dashed lines indicate when red tides started to become less
787 frequent.

788 **Fig. 10 Correlation between algal blooms and environmental drivers during 2005–2020.** Micro
789 and macro represent coverage of microalgal blooms and frequency of macroalgal blooms,
790 respectively. Frequency rather than coverage of macroalgal blooms was used because coverage
791 could be affected by human intervention (e.g., clean-up removals). Proro, Karen, Nocti and Skele
792 represent *Prorocentrum donghaiense*, *Karenia mikimotoi*, *Noctiluca scintillans* and *Skeletonema*
793 *costatum* dominated red tides, respectively. "*" and "***" indicate significant correlations at the 0.05
794 (2-tailed) and 0.01 levels (2-tailed), respectively.

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