



Research Paper

First investigation of the temporal distribution of neurotoxin β -N-methylamino-L-alanine (BMAA) and the candidate causative microalgae along the South Sea Coast of Korea

Sea-Yong Kim^a, Mungi Kim^b, Young Kyun Lim^c, Seung Ho Baek^c, Ji Yoon Kim^d, Kwang-Guk An^d, Seongjin Hong^{a,b,*}

^a Department of Marine Environmental Sciences & Institute of Marine Environmental Sciences, Chungnam National University, Daejeon 34134, Republic of Korea

^b Department of Earth, Environmental & Space Sciences, Chungnam National University, Daejeon 34134, Republic of Korea

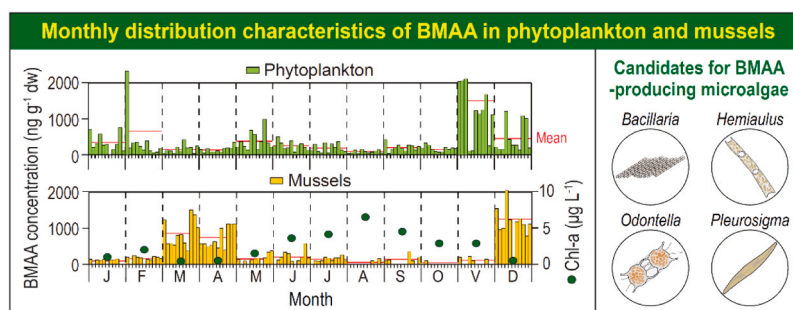
^c Ecological Risk Research Department, Korea Institute of Ocean Science and Technology, Geoje 53201, Republic of Korea

^d Department of Bioscience and Biotechnology, Chungnam National University, Daejeon 34134, Republic of Korea

HIGHLIGHTS

- Neurotoxin BMAA was first detected in phytoplankton and mussels on the Korean coasts.
- Elevated concentrations of BMAA were observed from late autumn to spring.
- Phase lags were observed between phytoplankton and mussels in BMAA concentrations.
- Chl. a and BMAA accumulation exhibited a negative correlation throughout the year.
- Four genera of diatoms are proposed as BMAA-producing microalgae on the Korean coast.

GRAPHICAL ABSTRACT



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ABSTRACT

The neurotoxin β -N-methylamino-L-alanine (BMAA), produced by cyanobacteria and diatoms, has been implicated as an environmental risk factor for neurodegenerative diseases. This study first investigated the occurrence and monthly distributions of BMAA and its isomers, 2,4-diaminobutyric acid (DAB) and N-2-aminoethylglycine (AEG), in phytoplankton and mussels from 11 sites along the South Sea Coast of Korea throughout 2021. These toxins were quantified using LC-MS/MS, revealing elevated BMAA concentrations from late autumn to spring, with phase lags observed between phytoplankton and mussels. The highest concentration of BMAA in phytoplankton was detected in November (mean: 1490 ng g⁻¹ dry weight (dw)), while in mussels, it peaked in December (mean: 1240 ng g⁻¹ dw). DAB was detected in phytoplankton but was absent in mussels, indicating limited bioaccumulation potential. In February, the peak mean DAB concentration in phytoplankton was 89 ng g⁻¹ dw. AEG was not detected in any samples. Chlorophyll-a concentrations consistently showed an inverse correlation with BMAA concentrations in mussels throughout the year. Through correlation analysis, four diatom genera, *Bacillaria*, *Hemiaulus*, *Odontella*, and *Pleurosigma*, were identified as potential causative microalgae of

* Correspondence to: Department of Marine Environmental Sciences, Chungnam National University, Daejeon 34134, Republic of Korea.

E-mail address: hongseongjin@cnu.ac.kr (S. Hong).

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BMAA. This study offers insights into identifying the causative microalgae for BMAA and informs future regulatory efforts regarding unmanaged biotoxins.

1. Introduction

Algal biotoxins, naturally synthesized by microalgae, pose significant hazards to ecosystems and human health. The rapid proliferation of these microalgae, termed harmful algal blooms (HABs), has become increasingly prevalent worldwide, attributed to factors such as global climate change and excessive nutrient inputs [1,2]. Various microalgal groups, including cyanobacteria, diatoms, and dinoflagellates, have been identified as producers of biotoxins, each categorized by their specific harmful effects [3-7]. For instance, hepatotoxins, such as cyanobacterial microcystins and nodularins, are notorious for causing liver damage [8,9]. Neurotoxic compounds like β -N-methylamino-L-alanine (BMAA), 2,4-diaminobutyric acid (DAB), and domoic acid (DA) primarily induce neurotoxicity via excitotoxicity mechanisms [3,10-14]. Cyanobacteria and diatoms have been identified as primary producers of BMAA and DAB, whereas DA is associated with diatom blooms [4,6,15,16]. Notably, BMAA has garnered attention as a secondary metabolite with potential neurodegenerative implications, including its association with disorders such as amyotrophic lateral sclerosis-parkinsonism dementia complex (ALS-PDC) and Alzheimer's disease [17,18].

Bioaccumulation and biomagnification of BMAA have been documented globally. Bioaccumulation of BMAA has been observed in zooplankton and various invertebrate and vertebrate species inhabiting diverse ecosystems across multiple countries [17,19-36]. BMAA was detected in filter feeders, herbivores, insectivores, omnivores, piscivores, and planktivores originating from aquatic ecosystems in China, South Africa, Sweden, the USA, France, Norway, and Portugal [20,22,27,29,31-37]. The phenomenon of BMAA biomagnification was initially identified in the terrestrial ecosystem of Guam, particularly within the food web of the indigenous Chamorro population [17]. Subsequently, instances of BMAA biomagnification within aquatic environments were observed in the Baltic Sea [26], followed by the Yellow Sea [35]. Notably, concentrations of BMAA in organisms were higher in the Yellow Sea, characterized by diatom dominance, compared to the Baltic Sea, where cyanobacteria dominate. However, because of the initial finding of cyanobacteria as BMAA producers [17], studies on diatom-dominated ecosystems are relatively limited. Distributions of BMAA were reported in diatom-abundant aquatic ecosystems in China [35] and France [33], where BMAA was commonly found being accumulated in mussels, as well as in Australia [38]. Chinese and French studies focused on marine ecosystems, while the Australian study examined freshwater ecosystems.

Diatoms are an abundant plankton group that thrives in marine ecosystems, including the South Sea Coast (SSC) of Korea [39-41]. The SSC consistently exhibits diatom dominance, with specific genera such as *Asterionellopsis*, *Chaetoceros*, *Eucampia*, *Pseudo-nitzschia*, *Skeletonema*, and *Thalassiosira* being the most prevalent [39,40]. Among these genera, *Chaetoceros*, *Pseudo-nitzschia*, *Skeletonema*, and *Thalassiosira* have been identified as producers of BMAA [28,35,42,43]. Considering that SSC serves as a central hub for aquaculture in South Korea [44], regular biotoxin monitoring is imperative to mitigate the risks associated with the occurrence of HABs [40,41]. However, the current regulatory and monitoring efforts for microalgal biotoxins, overseen by the National Institute of Fisheries Science (NIFS) in Korea (<https://www.nifs.go.kr/main.do>), are limited to saxitoxin causing paralytic shellfish poisoning (PSP), and okadaic acid (OA) and dinophysistoxins (DTXs) causing diarrhetic shellfish poisoning (DSP). Other marine biotoxins such as DA and its isomers, ciguatoxin, brevetoxin, azaspiracids (AZAs), pectenotoxins (PTXs), yessotoxins (YTXs), BMAA, DAB, and N-2-aminoethylglycine (AEG) have not yet been subject to regulation and monitoring. However, a recent study has revealed a spatiotemporal

pattern of AZAs, PTXs, and YTXs on the SSC, highlighting the urgency of researching unregulated and unmonitored toxins [45].

The primary objective of this study is to investigate the temporal distribution of unregulated biotoxins, such as BMAA, DAB, and AEG, in Korean marine ecosystems, focusing on the SSC. Additionally, the study aims to identify potential causative microalgae associated with these toxins. Specifically, the objectives include assessing the concentrations of these toxins in phytoplankton and mussels collected from the SSC throughout all months of 2021 and identifying specific microalgae correlated with toxin concentrations. It is noteworthy that DAB and AEG are structural isomers of BMAA; no research has been conducted on these toxins in Korean marine ecosystems. Thus, this study fills a critical knowledge gap in understanding the occurrence of these toxins in the region. Furthermore, another aim of the study is to elucidate the environmental factors that may be correlated with toxin concentrations in phytoplankton and mussels. This holistic approach provides valuable insights into toxin production and accumulation dynamics in Korean marine ecosystems.

2. Materials and methods

2.1. Sample collections

Mussels ($n = 118$, *Mytilus galloprovincialis*) and phytoplankton ($n = 132$, 20–200 μm suspended particulate matter, SPM) were collected monthly at 11 sites (S1–S11) along SSC of South Korea in 2021 (Fig. 1). Detailed information on the sampling campaign was presented in our previous study [45]. At each sampling site, 10–15 wild mussels were collected using landing nets to achieve an average wet weight of approximately 50 g after deshelling and pooling. These mussels were not intended for consumption. In the laboratory, the pooled mussels were homogenized in a blender each month to ensure the representativeness of samples for each site. The samples were stored at -20°C until analysis. Seawater was first filtered through a 200 μm mesh size net to remove zooplankton, followed by filtration through a 20 μm nylon net

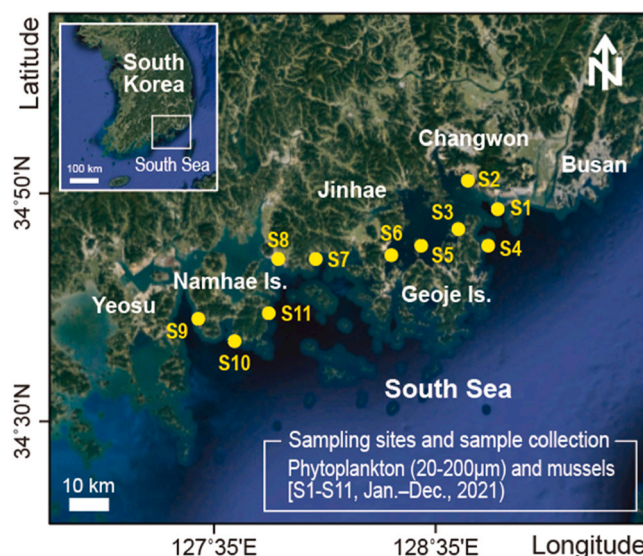


Fig. 1. Map showing the sampling sites of phytoplankton (20–200 μm suspended particulate matter, SPM) and mussels. Phytoplankton and mussels were collected monthly from sites S1–S11 along the South Sea Coast of Korea from January to December 2021.

filter (Millipore, Merck, Darmstadt, Germany) to collect phytoplankton samples. The net was towed until a sufficient amount of 20–200 μm SPM (approximately 0.1–0.5 g wet weight) was collected for BMAA analysis. The collected samples were stored at $-20\text{ }^{\circ}\text{C}$ until toxin extraction and analysis.

2.2. Identification of phytoplankton and measurement of water quality

Optical microscopy was used to identify phytoplankton at the genus level and to quantify their abundance based on their morphological features, as outlined in the phytoplankton identification guides by Omura et al. [46]. A collected 500 mL of seawater was fixed with 3% Lugol's solution and then concentrated to 50 mL. The Sedgewick-Rafter Chamber was utilized to distinguish the morphology of each phytoplankton genus and count their number. Water quality data, such as temperature, salinity, pH, dissolved oxygen (DO), and nutrients, were reported previously [45].

2.3. Extraction of neurotoxins

Extraction procedures were conducted based on previous methodologies [47,48] with minor modifications (Fig. S1). Briefly, for mussels, 50 mg (dry weight, dw) was dissolved in 3 mL of 0.1 mol L^{-1} trichloroacetic acid (TCA). Phytoplankton samples, freeze-dried on nylon net filters, were treated with 3 mL of TCA, followed by vortexing for 30 s to detach the plankton from the filter. This process was repeated three times. Subsequently, the same procedures were applied to both mussel and plankton samples. The samples, kept in an ice-water bath, were sonicated to minimize protein degradation, utilizing an ultrasonic homogenizer (Sonics & Material Inc., Newtown, CT) for 2 min at AMPL 30% and pulse 07 03. After incubating for 48 h at $4\text{ }^{\circ}\text{C}$ and lyophilization, the samples were hydrolyzed with 1.2 mL of 6 M HCl for 20 h at $110\text{ }^{\circ}\text{C}$. The samples were filtered using an Ultrafree-MC centrifugal filter (Merck Millipore, Billerica, MA) for 1 min at 10,000 g. The filtered samples were lyophilized again and reconstituted with 20 mM HCl for LC-MS/MS analysis.

2.4. LC-MS/MS analysis

The AccQ-Tag method using a WAT052880 AccQ-Tag kit (Waters, Milford, MA) was employed to derivatize the samples. BMAA and DAB standard materials were purchased from Sigma-Aldrich (St. Louis, MO) and AEG from Tokyo Chemical Industry (TCI, Tokyo, Japan). The target neurotoxins BMAA, DAB, and AEG were analyzed based on methodologies described by Faassen et al. [49] and Kim et al. [42] with minor modifications, using an Agilent 1290 infinity II LC system (Agilent Technologies, Santa Clara, CA) coupled with an Agilent 6470 triple quadrupole mass spectrometer (Agilent Technologies). Chromatographic separation was achieved using an AccQ-Tag Ultra C18 column ($100 \times 2.1\text{ mm}$, $1.7\text{ }\mu\text{m}$ particle size, Waters), with the column temperature maintained at $40\text{ }^{\circ}\text{C}$. The mobile phases consisted of 0.1% formic acid in (A) water and (B) methanol. The injection volume was 20 μL . Detailed LC-MS/MS conditions are provided in Table S1. For identification of BMAA, DAB, and AEG, one general transition ($459.18 > 119.08$) and three diagnostic transitions ($459.18 > 258.09$, $459.18 > 188.1$, and $459.18 > 214.1$) were monitored.

2.5. Quality control

Several studies have assessed the reliability of BMAA analysis by examining linearity, the limit of detection (LOD), the limit of quantification (LOQ), and matrix spike tests [18,27,35,50,51] (Table S2). Previous BMAA studies have recommended using different dilution ratios as an effective strategy to mitigate matrix effects [47,52]. In this study, 20 randomly selected extracted samples were diluted in different ratios (1:2, 1:4, and 1:8) and screened for the presence of a BMAA peak.

Among them, five samples with no BMAA peak detected were selected and spiked with BMAA at a concentration of 5 ng mL^{-1} to check the deviation of peak size among the samples and for each sample across three test runs, as well as to evaluate the background noise of the chromatogram. The dilution ratio of 1:4 exhibited a deviation of 6%, whereas the others showed deviations exceeding 6%. Thus, the 1:4 dilution ratio was selected for BMAA analysis and quality control. A blank sample spiked with 5 ng mL^{-1} of BMAA was run every ten samples to ensure consistent peak size and retention time, and comparisons were made between samples and between batches. The LOD for BMAA was determined as 3.143 times the standard deviation (SD) of each matrix (i. e., phytoplankton and mussels) spiked with a standard concentration of 1 ng mL^{-1} ($n = 7$). The LOQ was set at 10 times the SD of each matrix spiked with the standard. The LOD of BMAA was found to be $7.2\text{ ng g}^{-1}\text{ dw}$ in phytoplankton and $24.7\text{ ng g}^{-1}\text{ dw}$ in mussels. The LOQ of BMAA was determined to be $23.0\text{ ng g}^{-1}\text{ dw}$ in phytoplankton and $78.6\text{ ng g}^{-1}\text{ dw}$ in mussels. The LOD and LOQ of DAB were calculated using the same approach applied for BMAA, with phytoplankton as the representative matrix. The LOD and LOQ of DAB were determined to be 20.6 and $65.6\text{ ng g}^{-1}\text{ dw}$, respectively. Spike recovery tests were performed using mussels as a representative matrix. Twenty ng of BMAA and DAB were spiked into 50 mg of freeze-dried BMAA-free mussels ($n = 3$). The analytical method was performed in the same manner as described above. The recovery rates were $87 \pm 4\%$ for BMAA and $84 \pm 5\%$ for DAB, both within satisfactory ranges. Quality control was not performed on AEG due to the absence of detectable levels of AEG in all samples.

2.6. Statistical analysis

Spearman correlation analysis was employed to assess the relationship between the concentrations of BMAA in phytoplankton and mussels and environmental factors (i.e., temperature, salinity, pH, DO, Chl. a, and nutrients), given the non-normally distributed data. Additionally, the correlation between the concentrations of BMAA in phytoplankton and the cell densities of each observed microalgae was investigated, with a significance level set at $p < 0.05$. ANOVA was performed to assess the influence of temporal changes (i.e., month) and site on BMAA concentrations. Alpha and beta diversity were evaluated using the Shannon index and Bray-Curtis dissimilarity, respectively. R software (version 4.2.3) was used for the statistical analyses, and Microsoft Excel and Adobe Illustrator were used for visualization of results and rearrangement of figures. For statistical analysis, concentrations in samples below the LOD, below the LOQ (but \geq LOD), and with no detected peak were treated as LOD/2, LOQ/2, and "0", respectively.

3. Results and discussion

3.1. Distributions of BMAA in phytoplankton

BMAA was detected in phytoplankton samples, with concentrations ranging from $<\text{LOD}$ to $5130\text{ ng g}^{-1}\text{ dw}$ (Fig. 2a, Table 1, and Table S3). Bacillariophyceae dominated the phytoplankton community throughout the year and was more dominant in seasons with high BMAA concentrations (Fig. S2a and Table S5). No significant difference in the genus richness of phytoplankton was observed between the months ($p > 0.05$), but the composition varied, explaining 50% of the variance (Fig. S2b and S2c). Cyanobacteria have been identified as BMAA producers since 2003 [17], while diatoms were identified as producers in 2014 [6], with research on them remaining relatively limited. In addition, studies investigating the year-round temporal distribution of BMAA in diatom-dominated marine ecosystems are scarce [32,33,35], and are discussed for the first time in the present study. A relatively elevated concentration of BMAA was observed during late autumn to winter, with the highest monthly mean concentration detected in November ($1490\text{ ng g}^{-1}\text{ dw}$), followed by February ($650\text{ ng g}^{-1}\text{ dw}$) and December ($463\text{ ng g}^{-1}\text{ dw}$). The concentration of BMAA in phytoplankton along the

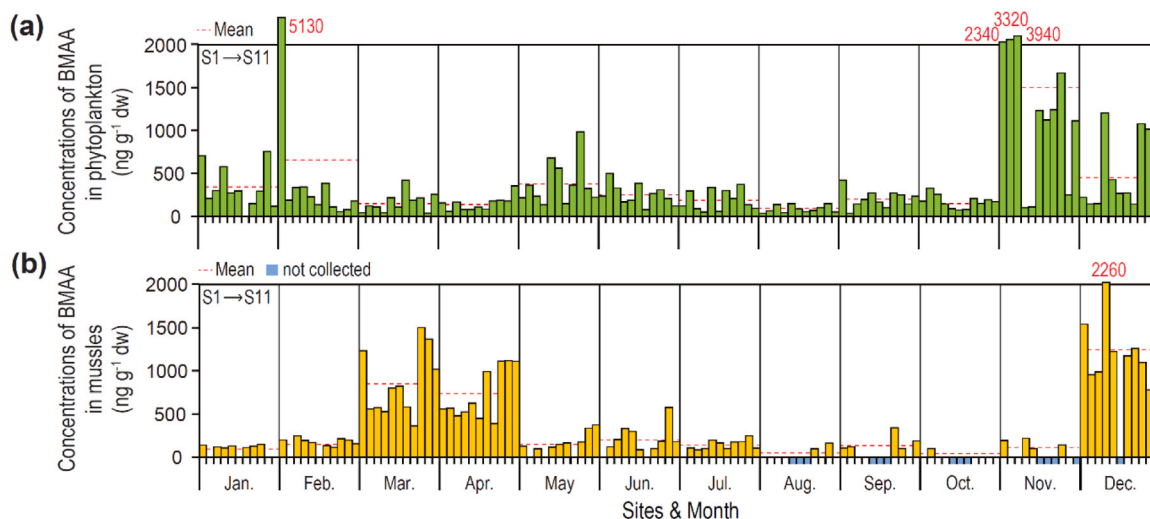


Fig. 2. Concentrations of BMAA in (a) phytoplankton (20–200 μm SPM) and (b) mussels collected from 11 sites (S1–S11) along the South Sea Coast of Korea from January to December 2021. The numbers above the y-axis have been rounded to the nearest whole number.

SSC differed greatly from site to site, which appears to be due to the heterogeneous distribution of causative microalgae. Comparing the findings with those from China [35], the highest mean concentration of BMAA in phytoplankton in Korea surpassed that of China, albeit showing a similar seasonal pattern. In Jiaozhou Bay, China, BMAA concentrations in phytoplankton ranged from $<\text{LOD}$ to $1000 \text{ ng g}^{-1} \text{ dw}$, with the highest mean concentration of $470 \text{ ng g}^{-1} \text{ dw}$ in January, representing winter [35]. In contrast, the highest concentration of BMAA in phytoplankton in Thau Lagoon, France, was approximately $1000 \text{ ng g}^{-1} \text{ dw}$, observed in September, representing late summer or early autumn [33]. Despite the distinct seasonal patterns observed for BMAA, diatoms were consistently identified as the dominant phytoplankton groups in all three countries, with diatoms isolated from Chinese and French studies identified as BMAA producers [33,35]. The research outcomes reveal variations in both the concentration and temporal distribution of BMAA in phytoplankton, particularly with diatoms serving as the causative microalgae across different countries. Remarkably, phytoplankton along the SSC of South Korea demonstrates higher concentrations of BMAA than those in Jiaozhou Bay, China and Thau Lagoon, France.

3.2. Bioaccumulation of BMAA in mussels

BMAA was detected in mussels, with concentrations ranging from $<\text{LOD}$ to $2260 \text{ ng g}^{-1} \text{ dw}$ (Fig. 2b, Table 1, and Table S3). The variability in BMAA concentrations among sites in mussels was lower compared to the variability of BMAA observed in phytoplankton. The highest monthly mean concentration was observed in December ($1240 \text{ ng g}^{-1} \text{ dw}$) during winter, followed by March ($850 \text{ ng g}^{-1} \text{ dw}$) and April ($720 \text{ ng g}^{-1} \text{ dw}$) during spring (Table 1). BMAA concentrations in mussels in other months remained relatively low. In contrast to Korea, the seasonal pattern of BMAA accumulation in *M. galloprovincialis* was similar in the Yellow Sea and the Mediterranean Sea, with the highest BMAA concentration accumulating in September (Table 1), representing later summer or autumn in both countries [32,33,35]. The highest BMAA concentration accumulated in *M. galloprovincialis* in China and France was $6650 \text{ ng g}^{-1} \text{ dw}$ ($1330 \text{ ng g}^{-1} \text{ ww}$) and $14,400 \text{ ng g}^{-1} \text{ dw}$, respectively, indicating potential variations in both the concentration and seasonal pattern of BMAA accumulation among countries [32,33,35]. The study conducted in China utilized wet weight to quantify the concentration of BMAA in bivalves, crustaceans, and gastropods, with a conversion factor of 5:1 between wet and dry weights [35]. The same conversion factor was applied to mussels in this study (Table S3). Considering the observed seasonal pattern of BMAA production and its

accumulation in mussels, BMAA produced by phytoplankton in France may exhibit a higher propensity to accumulate at elevated levels in *M. galloprovincialis* than in China and Korea.

The bioaccumulation of BMAA has been documented across trophic levels in both marine and freshwater ecosystems [26,35,36]. Higher concentrations of BMAA have been observed in higher trophic-level organisms, including zooplankton, fish, bivalves, crustaceans, and gastropods. However, some studies, including this study, have presented controversial results regarding BMAA bioaccumulation, reporting lower concentrations of BMAA in marine animals compared to phytoplankton [53,54]. Further studies are needed to evaluate BMAA concentrations across different trophic-level organisms and to assess its bioaccumulation and biomagnification potentials.

The concentrations of BMAA tended to increase first in phytoplankton and then increase in mussels (Fig. 2 and Fig. S3). A phase lag in BMAA concentrations was observed between phytoplankton and mussel samples. This phenomenon was evident at specific sites (i.e., sites 1, 4, 7, and 11 from January to April, and sites 1, 2, 3, 7, 8, 9, and 11 from November to December), as depicted in Fig. S3, and in the average concentrations across all sites along the South Sea Coast of Korea, as shown in Fig. 2. The initial increase in production and accumulation of BMAA occurred in February and March, followed by a second increase in November and December, respectively. To elaborate, while BMAA concentration in phytoplankton peaked in February and November, mussels exhibited higher concentrations in March and December, with an approximate one-month time difference. In addition, although small, the production of BMAA in phytoplankton showed a slight increase in May, which appeared to be reflected in mussels in June. This observation can be attributed to the biological half-life of BMAA in mussels. The biological half-life of an exogenous substance is the time required for half of the substance to be eliminated by biological processes [55]. This is related to the metabolic and excretion capabilities of the organism and is generally species-specific and compound-specific. The phase lag phenomenon has been documented in previous studies investigating lipophilic marine biotoxins (LMTs) within the same region (i.e., SSC) as the present study [45,56]. The determination of the phase lag relied on the half-life of biotoxins: no phase lag was observed for PTXs [45], with a known half-life of 2.9 d [57], whereas a phase lag was evident for YTXs during summer [45], with reported half-lives ranging from 20 to 24 d [58]. The reported half-life of BMAA in bivalves ranges between 9.8 and 20.4 d [55], demonstrating a duration comparable to that of YTXs. Given that this study collected samples from natural environments rather than controlled environmental experiments, various confounding

Table 1
Concentrations of BMAA in phytoplankton and mussels collected along the South Sea Coasts in Korea obtained from this study and previously reported data from China and France.

Country & Organisms		Concentration (ng g ⁻¹ dw)												References
		Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.	
Korea														
Phytoplankton (20–200 μm)	Range	<LOQ ^a -754	53– 5130	35– 419	55– 353	135– 983	77– 499	47– 371	32– 145	32– 420	74– 327	102– 3990	139– 1200	This study ^b
	Mean	334	650	159	147	384	252	186	84	202	169	1490	463	
Mussels	Range	<LOQ– 148	<LOQ– 245	362– 1500	387– 1120	<LOD ^c – 372	<LOQ–576 196	<LOQ– 250	<LOD– 162	<LOQ– 340	<LOD– 98	<LOD– 217	777– 2260	
	Mean	94	155	850	720	146	196	136	55	121	36	110	1240	
China														
Phytoplankton (20–200 μm)	Range	30– 1000		ND ^d – 150			ND– 110			ND– 190				Wang et al. [35] ^e
	Mean	470		30			20			50				
Mussels	Range	2250 ^d (450) ^e		1900 ^f (380) ^g						6650 ^f (1330) ^g				
France^h														
Phytoplankton (20–125 μm)	Range							400–600	180	1000	ND– 250	ND		Réveillon et al. [32,33] ⁱ
Mussels	Range	3500–3800			1000–9000					4000–14,400				

^a <LOQ: below the limit of quantification.

^b Reverse-phase liquid chromatography (RPLC)-MS/MS with AQC (6-aminoquinolyl-N-hydroxysuccinimidyl carbamate) derivatization was used for BMAA analysis.

^c <LOD: below the limit of detection.

^d ND: not detected.

^e Hydrophilic-interaction chromatography (HILIC)-MS/MS without derivatization was used for BMAA analysis.

^f One sample was used in the previous study (on a dry weight basis, converted from wet weight assuming 80% water content).

^g Original data (wet weight basis concentration).

^h Raw data is not available. This table provides approximate concentrations of BMAA.

ⁱ HILIC-MS/MS without derivatization was used for BMAA analysis.

factors may have influenced the results, making it challenging to identify any absolute trend. Nevertheless, the phase lag was observed both in BMAA concentrations at specific sites and in the average concentrations across all sites. Altogether, these findings suggest that the half-life of microalgal biotoxins is a critical factor affecting the accumulation period in shellfish.

3.3. DAB and AEG in phytoplankton and mussels

DAB was detected in phytoplankton samples, as shown in Fig. S4 and Table S3. No AEG was detected in any of the phytoplankton samples. The detection rate of DAB was 45% (59 out of 132), and the highest concentrations reached $386 \text{ ng g}^{-1} \text{ dw}$ (Table S3). The monthly mean concentration of DAB in phytoplankton mostly remained below the LOQ. Nevertheless, mean concentrations surpassing LOQ were observed in February ($89 \text{ ng g}^{-1} \text{ dw}$), November ($86 \text{ ng g}^{-1} \text{ dw}$), and December ($82 \text{ ng g}^{-1} \text{ dw}$). DAB was also detected in phytoplankton collected in studies in China and France; the concentrations were approximately $480 \text{ ng g}^{-1} \text{ dw}$ and $1440 \text{ ng g}^{-1} \text{ dw}$, respectively [33,35], which were

higher than those in the SSC of Korea. In the SSC of Korea, the seasonal distribution trend of DAB in phytoplankton appeared similar to that of BMAA, suggesting that the causative microalgae of DAB might be the same or similar to those of BMAA.

Microalgae known to produce DAB include the diatoms *Halamphora coffeaeformis*, *Asterionellopsis glacialis*, *Pseudo-nitzschia delicatissima*, and *Odontella aurita* [59]. Among them, *O. aurita* showed the highest DAB production, with $5100 \text{ ng g}^{-1} \text{ dw}$ of DAB detected. *Odontella* has also occurred in the SSC of Korea (Table S5), and it is believed to be a potential causative microalgae of DAB. In Australia, freshwater diatoms such as *Aulacoseira*, *Cyclotella*, *Fragilaria*, *Navicula*, and *Tabellaria* have been identified as producers of DAB, with concentrations of 103, 283, 259, 5274, and $395 \text{ ng g}^{-1} \text{ dw}$, respectively [38]. Further research is needed to assess the DAB production potential of species from these diatom genera isolated from marine and freshwater ecosystems in Korea. Meanwhile, phytoplankton abundance and Chl. a concentrations in the SSC varied depending on season and water depth [60,61], suggesting that relatively high DAB and AEG concentrations may exist in phytoplankton communities living below the surface layer. The

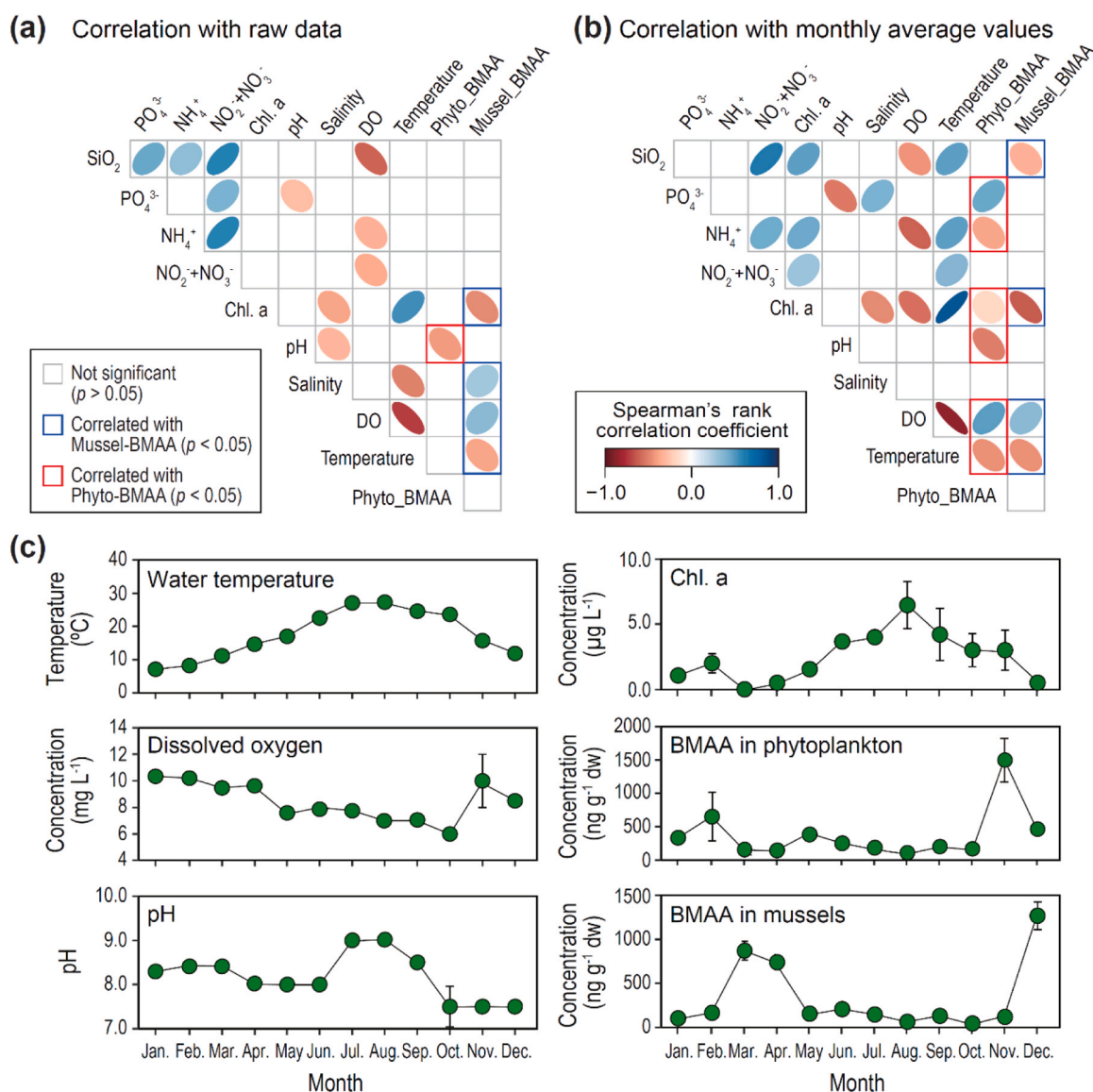


Fig. 3. Relationships between environmental factors and concentrations of BMAA in phytoplankton and mussels. Spearman's rank correlations using (a) transformed raw data and (b) transformed monthly average values. (c) Monthly distributions of environmental factors and BMAA concentrations in phytoplankton and mussels. Environmental factors that showed significance in the correlation analysis were selectively highlighted (e.g., water temperature, dissolved oxygen, pH, and Chl. a). ANOVA results for BMAA concentrations are presented in Table S4.

bacterial origin of DAB was demonstrated through the absence of DAB detection in diatom cultures treated with antibiotics [62] and the presence of DAB in bacteria isolated from diatom cultures [43]. Further research on DAB and AEG is imperative, particularly considering the contribution of symbiotic bacteria and water depth-stratified sampling of plankton.

In the present study, DAB and AEG were not detected in mussels (*M. galloprovincialis*) of the SSC of Korea. The bioaccumulation potential of AEG could not be assessed because AEG was not detected in phytoplankton. DAB did not exhibit bioaccumulation in shellfish, suggesting that DAB may have a shorter biological half-life than BMAA. In a previous study, DAB was detected in *M. galloprovincialis* in China, and AEG was not detected [35]. Wang et al. [35] proposed that marine animals (i. e., crustaceans, gastropods, and mollusks) in Jiaozhou Bay can effectively metabolize DAB. They observed that DAB did not biomagnify in the food web, and the concentrations of DAB were similar among marine animals. Future studies are needed to investigate the biomagnification potential and species-specific biological half-life of BMAA and DAB in coastal ecosystems.

3.4. Correlation between BMAA in phytoplankton and mussels and environmental factors

The relationship between BMAA concentrations in biological samples and environmental factors (temperature, salinity, pH, DO, Chl. a, and nutrients) was determined through correlation analysis (Fig. 3). Results indicated that pH was consistently correlated with BMAA concentration in phytoplankton when using transformed raw data and monthly mean values (Fig. 3a and b). However, considering the monthly distribution of pH and BMAA in phytoplankton using raw data revealed no correlation throughout the year (Fig. 3c). Previous studies showed that the production of BMAA is influenced by nitrogen availability in both cyanobacteria [63,64] and diatoms [28]. However, the results of this study did not show a significant correlation between the concentrations of dissolved inorganic nitrogen and BMAA, suggesting that additional research is necessary to assess the influence of multiple environmental factors on BMAA production in the causative microalgae. Chl. a, DO, and water temperature were consistently correlated with BMAA concentrations in mussels based on the correlation analyses (Fig. 3a and b). However, only Chl. a showed a year-round correlation with BMAA accumulation (Fig. 3c). Since no study has investigated the relationship between different forms of Chl. a (e.g., Chl. a in non-visible live microalgae via microscopy, Chl. a emitted from deceased microalgae, and Chl. a in herbivorous zooplanktons) and BMAA accumulation in mussels, further studies are necessary to understand the underlying reasons and mechanisms for their association. Additionally, the elevated BMAA concentrations in mussels during periods of low Chl. a concentrations necessitates further investigation. One possible explanation is that microalgae might be stimulated by copepodamides released by starved herbivorous copepods due to food scarcity (i.e., low Chl. a), potentially leading to increased BMAA production as a defense mechanism. While the correlation between copepodamides and enhanced PSP and DA production is reported [65,66], its association with BMAA production remains unclear.

3.5. Identification of causative microalgae of BMAA

Cyanobacteria and diatoms have been previously identified as BMAA producers [4,6]. A total of 30 genera of diatoms were identified in this study (Table 2 and Table S5), including *Amphora*, *Asterionellopsis*, *Bacillaria*, *Bacteriastrium*, *Cerataulina*, *Chaetoceros*, *Coscinodiscus*, *Cylindrotheca*, *Dactyliosolen*, *Detonula*, *Ditylum*, *Entomoneis*, *Eucampia*, *Guinardia*, *Gyrosigma*, *Hemiaulus*, *Leptocylindrus*, *Licmophora*, *Navicula*, *Nitzschia*, *Melosira*, *Odontella*, *Pleurosigma*, *Pseudo-nitzschia*, *Rhizosolenia*, *Skeletonema*, *Stephanopyxis*, *Thalassionema*, *Thalassiosira*, and *Lauderia*. Among the observed genera, *Chaetoceros*, *Pseudo-nitzschia*, *Skeletonema*,

Table 2

Diatom genera observed in this study, and the species reported as the producer of BMAA with the reported concentrations.

Genus in this study	Species reported as BMAA producer	Concentrations (ng BMAA g ⁻¹ dry weight)	References
<i>Amphora</i> *	-		
<i>Asterionellopsis</i>	-		
<i>Bacillaria</i>	-		
<i>Bacteriastrium</i>	-		
<i>Cerataulina</i>	-		
<i>Chaetoceros</i>	<i>C. calcitrans</i>	320	Réveillon et al. [33]
	<i>C. calcitrans</i>	560–1800	Réveillon et al. [43]
	<i>C. decipiens</i>	110	Réveillon et al. [33]
	<i>C. diadema</i>	260	Wang et al. [35]
	<i>C. sp.</i>	580	Réveillon et al. [33]
	<i>C. sp.</i>	260–1600	Réveillon et al. [43]
<i>Coscinodiscus</i>	-		
<i>Cylindrotheca</i>	-		
<i>Dactyliosolen</i>	-		
<i>Detonula</i>	-		
<i>Ditylum</i>	-		
<i>Entomoneis</i>	-		
<i>Eucampia</i>	-		
<i>Guinardia</i>	-		
<i>Gyrosigma</i>	-		
<i>Hemiaulus</i> *	-		
<i>Leptocylindrus</i>	-		
<i>Licmophora</i>	-		
<i>Navicula</i>	-		
<i>Nitzschia</i>	-		
<i>Melosira</i>	-		
<i>Odontella</i> *	-		
<i>Pleurosigma</i> *	-		
<i>Pseudo-nitzschia</i>	<i>P. bipertita</i>	170	Wang et al. [35]
	<i>P. caciantha</i>	300	Wang et al. [35]
	<i>P. delicatissima</i>	3050	Wang et al. [35]
	<i>P. fraudulenta</i>	270	Wang et al. [35]
	<i>P. lundholmiae</i>	1240	Wang et al. [35]
	<i>P. multiseriis</i>	410–1350	Wang et al. [35]
	<i>P. simulans</i>	840	Wang et al. [35]
<i>Rhizosolenia</i>	-		
<i>Skeletonema</i>	<i>S. marinoi</i>	1.07–1.1	Jiang et al. [6]
<i>Stephanopyxis</i>	-		
<i>Thalassionema</i>	-		
<i>Thalassiosira</i>	<i>T. pseudonana</i>	~0.2	Lage et al. [28]
	<i>T. pseudonana</i>	750	Réveillon et al. [33]
	<i>T. pseudonana</i>	170–280	Réveillon et al. [43]
	<i>T. sp.</i>	3.28	Jiang et al. [6]
	<i>T. weissflogii</i>	~50	Lage et al. [28]
<i>Lauderia</i>	-		

* Candidates for the production of BMAA proposed in the present study.

and *Thalassiosira* have been reported to produce BMAA, with specific species within each genus [6,28,33,43,35]. BMAA concentrations were specific to the genus as well as the species (Table 2). For species within the genus *Chaetoceros*, including *calcitrans*, *decipiens*, *diadema*, and others, BMAA concentrations ranged from 110 to 1800 ng g⁻¹ dw [33, 35,43]. Similarly, for species within the genus *Pseudo-nitzschia*, such as *bipertita*, *caciantha*, *delicatissima*, *fraudulenta*, *lundholmiae*, *multiseriis*, and *simulans*, BMAA concentrations ranged from 270 to 3050 ng g⁻¹ dw [35]. *Thalassiosira* species, including *pseudonana*, *weissflogii*, and others, exhibited BMAA concentrations ranging from 2 to 750 ng g⁻¹ dw [6,28, 33,43]. *Skeletonema marinoi* was also documented to produce BMAA, with concentrations reaching up to 1.1 ng g⁻¹ dw [6]. Future research on BMAA should evaluate the potential of the aforementioned diatom genera and species indigenous to Korea for BMAA production.

Months with relatively high concentrations of phytoplankton were initially selected to identify the microalgal genus responsible for elevated BMAA production along the SSC. Subsequently, the similarity of environmental conditions among these selected months (January, February, November, and December) and adjacent months (March and April) was assessed. Disparities in environmental conditions between seasons were observed during periods of heightened BMAA concentrations in phytoplankton from January to February (Jan-Feb) and from November to December (Nov-Dec) (Fig. S5). These findings facilitated the identification of BMAA-producing microalgae in a season-specific manner (Fig. 4). In Jan-Feb, seven genera of diatoms (*Amphora*, *Coscinodiscus*, *Dactyliosolen*, *Eucampia*, *Gyrosigma*, *Nitzschia*, and *Stephanopyxis*), eight genera of dinoflagellates (*Alexandrium*, *Ceratium*, *Gonyaulax*, *Gymnodinium*, *Gyrodinium*, *Katodinium*, *Prorocentrum*, and *Protoperidinium*), and one genus of cryptomonad (*Cryptomonas*) was found to be correlated with the concentrations of BMAA in phytoplankton (Fig. 4a). Given the absence of dinoflagellates and cryptomonads but the presence of diatoms, which are reported BMAA producers, it is suspected that biotic stresses could be contributory factors for the elevated BMAA concentration in Jan-Feb. Biotic stresses (e.g., predation and competition) between different plankton species have been shown to stimulate the production of microalgal biotoxins [42,65,67], including BMAA [62].

Eight genera of diatoms were found to be correlated with the concentration of BMAA in phytoplankton in Nov-Dec (Fig. 4b): *Amphora*, *Bacillaria*, *Dactyliosolen*, *Hemiaulus*, *Odontella*, *Pleurosigma*, *Pseudo-nitzschia*, and *Skeletonema*. Among these genera, the increase in cell densities of *Amphora*, *Bacillaria*, *Hemiaulus*, *Odontella*, *Pleurosigma*, and *Skeletonema* corresponded with the pattern in BMAA concentration (Fig. 5). Specifically, higher cell densities of *Amphora* and a higher concentration of BMAA in phytoplankton were observed at site S1 in November compared to other sites, as well as across all sites in December. Similarly, the density of *Bacillaria* and the concentration of BMAA at sites S7 to S9 in November surpassed those at other sites in November and across all sites in December. Relatively high cell densities of *Hemiaulus* and a high concentration of BMAA were observed at site S2 in November. The cell densities of *Odontella* at sites S1, S7, and S8, as well as those of *Pleurosigma* at sites S1, S6, and S9 in November, were higher than those at the other sites, accompanied by elevated concentrations of BMAA, respectively. In terms of *Skeletonema*, higher cell densities and a greater concentration of BMAA were observed at site S8

in November compared to other sites in November and all sites in December. Among these six genera, only *Bacillaria*, *Hemiaulus*, *Odontella*, and *Pleurosigma* exhibited higher cell density in November or December compared to April, all under similar environmental conditions (Figs. S5 and S6). The average cell densities of *Bacillaria*, *Hemiaulus*, *Odontella*, and *Pleurosigma* in November were higher than in April (Fig. S6). Additionally, the average densities of *Odontella* in December were higher than in April. These findings suggest that these four diatom genera can be considered candidates for causative microalgae of BMAA in the SSC, Korea. None of these candidates have been reported as BMAA producers to date. Therefore, further investigations are necessary to evaluate the BMAA production capability of isolated diatom genera in Korea.

4. Conclusions

The present study represents the first comprehensive investigation of the occurrence and monthly distribution of the neurotoxin BMAA and the identification of its putative causative microalgae in Korean marine ecosystems. This study mainly focused on BMAA in phytoplankton and mussels in the diatom-dominated marine ecosystems. Notably, high concentrations of BMAA were detected during late autumn and spring, with phase lags observed between BMAA concentrations in phytoplankton and mussels. Furthermore, a significant negative correlation was observed between the concentration of Chl. a and BMAA in mussels. Four diatom genera, namely *Bacillaria*, *Hemiaulus*, *Odontella*, and *Pleurosigma*, were identified as candidates for BMAA-producing microalgae. In this study, we could not evaluate the production of BMAA for these candidate species; additional research is needed to isolate and culture candidate species, confirm the production of toxins, and identify the major factors for producing BMAA. Despite these limitations, this study provides a solid foundation for further BMAA research in Korea, such as the BMAA production capacity of selected diatom genera. Considering all the aforementioned findings, further studies are imperative to elucidate the underlying reasons and relevance between Chl. a and BMAA accumulation, explore allelopathic compounds that might influence BMAA production as putative defensive metabolites, understand the multimedia fate of BMAA in the environments, and assess the biological half-life of DAB.

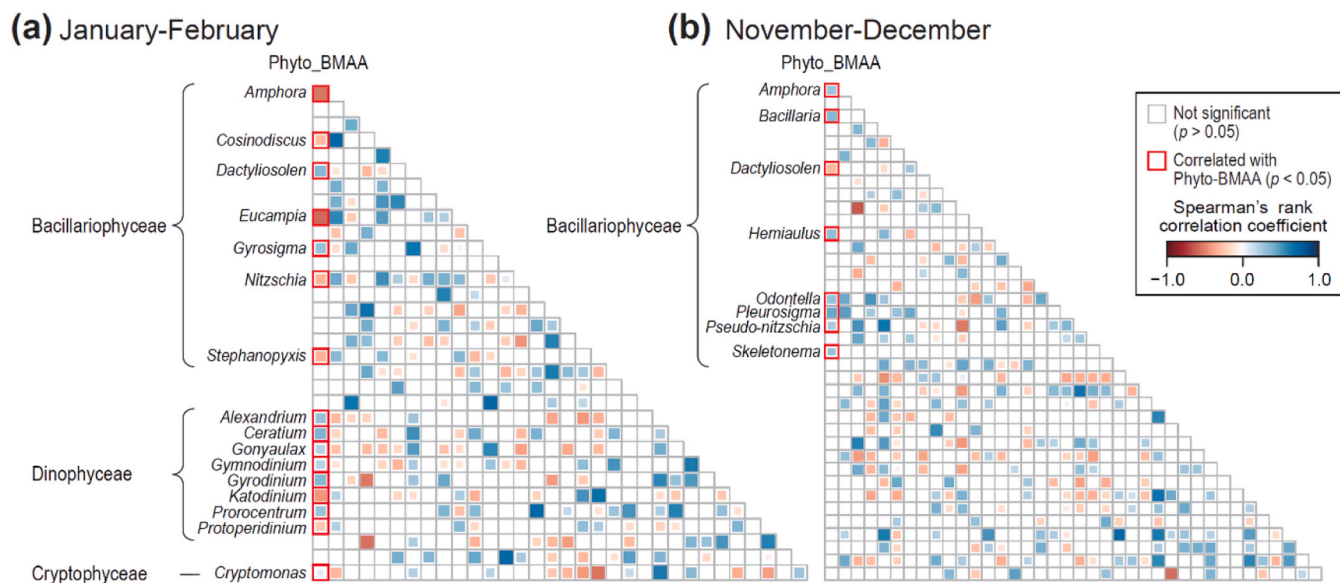


Fig. 4. Relationships between the density of microalgae identified at genus level and BMAA concentrations in phytoplankton (a) from January to February and (b) from November to December 2021.

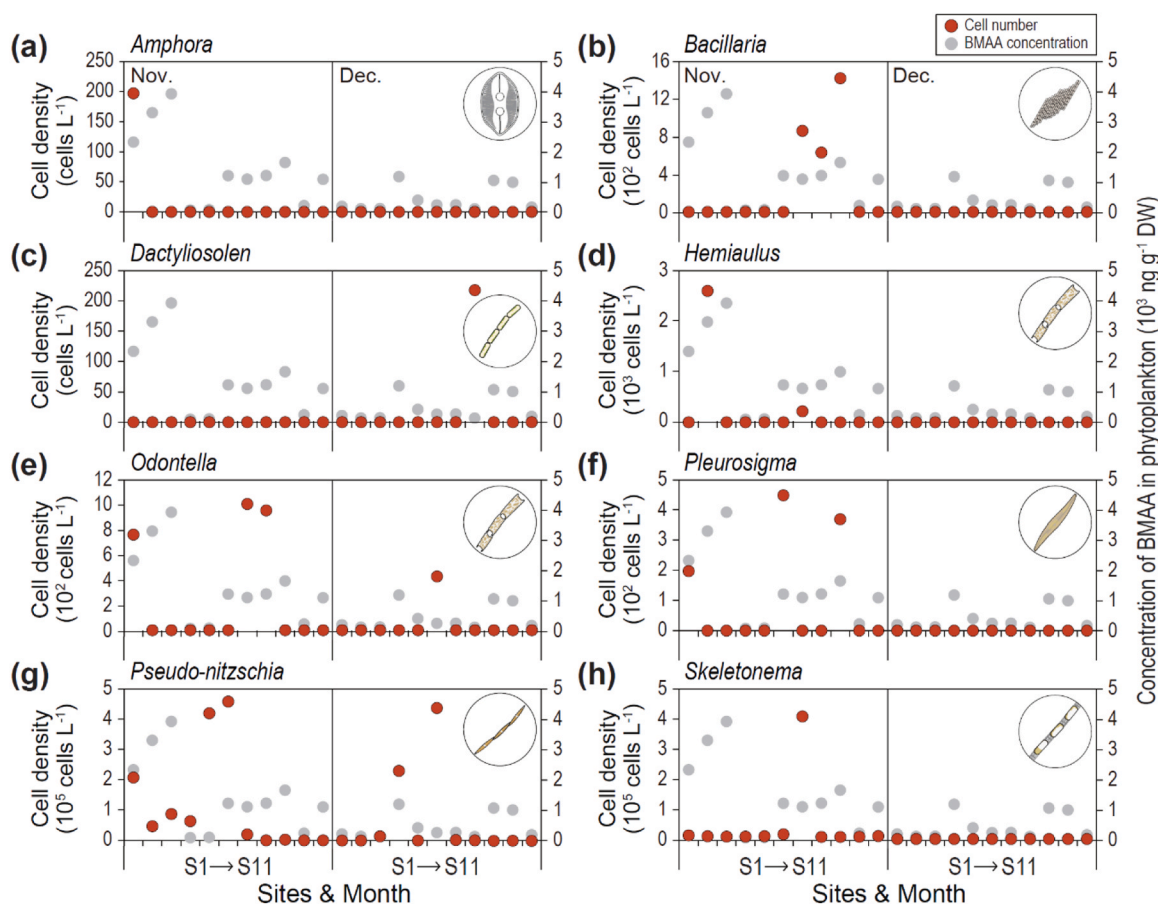


Fig. 5. The cell densities of eight diatom genera [(a) *Amphora*, (b) *Bacillaria*, (c) *Dactyliosolen*, (d) *Hemiaulus*, (e) *Odontella*, (f) *Pleurosigma*, (g) *Pseudo-nitzschia*, and (h) *Skeletonema*] and concentrations of BMAA in phytoplankton at 11 sites along the South Sea Coast of Korea in November and December 2021.

Environmental implication

The biotoxin β -N-methylamino-L-alanine (BMAA), which is associated with neurodegenerative diseases, is known to be produced by diverse microorganisms across various ecosystems worldwide. However, its presence has not been previously documented in the Korean coastal waters. This study represents the first observation of the occurrence and seasonal variation of BMAA along the South Sea Coast of Korea, thereby suggesting potential causative microalgae. The findings are anticipated to establish a baseline for predicting the seasonal distribution of BMAA across all Korean coasts, including the South Sea, and offer valuable insights into confirming the microalgae suspected of contributing to high BMAA production.

CRedit authorship contribution statement

Sea-Yong Kim: Writing – original draft, Visualization, Methodology, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Mungi Kim:** Writing – review & editing, Investigation. **Young Kyun Lim:** Investigation, Data curation. **Seung Ho Baek:** Writing – review & editing, Investigation. **Ji Yoon Kim:** Writing – review & editing, Investigation. **Kwang-Guk An:** Writing – review & editing, Investigation. **Seongjin Hong:** Writing – review & editing, Visualization, Supervision, Project administration, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at [doi:10.1016/j.jhazmat.2024.135486](https://doi.org/10.1016/j.jhazmat.2024.135486).

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Supplementary materials for

First investigation of the temporal distribution of neurotoxin β -N-methylamino-L-alanine (BMAA) and the candidate causative microalgae along the South Sea Coast of Korea

Sea-Yong Kim, Mungi Kim, Young Kyun Lim, Seung Ho Baek, Ji Yoon Kim,
Kwang-Guk An, Seongjin Hong *

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*Corresponding author. *E-mail address:* hongseongjin@cnu.ac.kr (S. Hong).

Supplementary Tables

Table S1. Instrumental conditions for analyzing neurotoxins in biological samples using LC-MS/MS.

Instrument	LC: Agilent Infinity 1290 II		
	MS/MS: Agilent 6470 triple quadrupole mass spectrometer		
Column	Waters AccQ-Tag Ultra C18, 2.1 mm × 100 mm, 1.7 μm		
Column temperature	40 °C		
Mobile phase	(A): 0.1% formic acid in water; (B): 0.1% formic acid in methanol		
Mobile phase gradient		Mobile phase	
	Time (min)	A (%)	B (%)
	0.0	99	1
	0.5	99	1
	1.5	95	5
	22.0	40	60
	23.5	95	5
	24.0	99	1
	25.0	99	1
Injection volume	20 μL		
Flow rate	0.4 mL min ⁻¹		
Ion source	ESI (electrospray ionization)		
Polarity	Positive		
Ion spray voltage	2500 V		
Gas temperature	300 °C		
Sheath gas temperature	400 °C		
Nebulizer gas	N ₂ (45 psi)		

Table S2. Linear range, coefficient of determination (R^2), limit of detection (LOD), limit of quantification (LOQ), and spike test recovery of BMAA and DAB analyzed using LC-MS/MS.

Compounds	Matrix	Linear range (ng mL⁻¹)	R²	LOD (ng g⁻¹ dw)	LOQ (ng g⁻¹ dw)
BMAA	Phytoplankton	2–120 (60–3600) ^a	0.99	7.2	23.0
BMAA	Mussel	2–100 (80–4000)	0.99	24.7	78.6
DAB	Phytoplankton	2–200 (60–6000)	0.99	20.6	65.6

^a: Linear range in ng g⁻¹

Table S3. Concentrations of BMAA and DAB in phytoplankton and mussels along the South Sea Coast of Korea from January to December 2021.

Month	Sites	Concentration of BMAA (ng g ⁻¹ dw ^a)		Concentration of DAB (ng g ⁻¹ dw)	
		Phytoplankton	Mussels	Phytoplankton	Mussels
Jan.	S1	706	139 (28) ^b	<LOQ ^d	<LOD ^c
	S2	209	<LOQ	<LOQ	<LOD
	S3	301	117 (23)	69	<LOD
	S4	575	107 (21)	80	<LOD
	S5	270	131 (26)	<LOQ	<LOD
	S6	294	<LOQ	<LOQ	<LOD
	S7	<LOQ	110 (22)	98	<LOD
	S8	147	126 (25)	<LOQ	<LOD
	S9	293	148 (30)	<LOQ	<LOD
	S10	754	<LOQ	80	<LOD
	S11	117	<LOQ	<LOQ	<LOD
Feb.	S1	5127	200 (40)	386	<LOD
	S2	186	<LOQ	<LOQ	<LOD
	S3	337	245 (49)	107	<LOD
	S4	339	194 (39)	80	<LOD
	S5	226	172 (34)	<LOQ	<LOD
	S6	134	<LOQ	<LOQ	<LOD
	S7	384	134 (27)	198	<LOD
	S8	108	114 (23)	<LOQ	<LOD
	S9	53	215 (43)	<LOD	<LOD
	S10	76	196 (39)	<LOQ	<LOD
	S11	179	158 (32)	<LOQ	<LOD
Mar.	S1	41	1232 (246)	<LOQ	<LOD
	S2	120	561 (112)	<LOQ	<LOD
	S3	109	575 (115)	<LOQ	<LOD
	S4	44	526 (105)	<LOQ	<LOD
	S5	217	800 (160)	<LOQ	<LOD
	S6	105	823 (165)	<LOQ	<LOD
	S7	419	582 (116)	154	<LOD
	S8	184	362 (72)	67	<LOD
	S9	214	1500 (300)	<LOQ	<LOD
	S10	35	1365 (273)	<LOD	<LOD
	S11	257	1020 (204)	85	<LOD
Apr.	S1	154	559 (112)	<LOQ	<LOD
	S2	55	568 (114)	72	<LOD
	S3	165	478 (96)	<LOQ	<LOD
	S4	80	523 (105)	<LOQ	<LOD
	S5	81	625 (125)	<LOQ	<LOD
	S6	107	451 (90)	<LOQ	<LOD
	S7	81	994 (199)	<LOQ	<LOD
	S8	178	387 (77)	79	<LOD
	S9	183	1110 (222)	<LOQ	<LOD
	S10	179	1119 (224)	<LOQ	<LOD
	S11	353	1109 (222)	<LOQ	<LOD
May.	S1	218	130 (26)	<LOQ	<LOD
	S2	363	<LOD	<LOQ	<LOD
	S3	234	94 (19)	71	<LOD
	S4	135	<LOD	67	<LOD
	S5	678	115 (23)	83	<LOD
	S6	560	147 (29)	76	<LOD
	S7	146	167 (33)	55	<LOD

	S8	365	<LOQ	69	<LOD
	S9	983	178 (36)	<LOQ	<LOD
	S10	323	335 (67)	69	<LOD
	S11	221	372 (74)	<LOQ	<LOD
Jun.	S1	236	<LOQ	<LOQ	<LOD
	S2	499	122 (24)	<LOQ	<LOD
	S3	330	201 (40)	<LOQ	<LOD
	S4	166	332 (66)	<LOQ	<LOD
	S5	183	296 (59)	<LOQ	<LOD
	S6	385	87 (17)	<LOQ	<LOD
	S7	77	<LOQ	<LOQ	<LOD
	S8	262	100 (20)	73	<LOD
	S9	309	186 (37)	86	<LOD
	S10	203	576 (115)	67	<LOD
	S11	123	179 (36)	<LOQ	<LOD
Jul.	S1	122	<LOQ	<LOQ	<LOD
	S2	295	109 (22)	76	<LOD
	S3	87	83 (17)	<LOQ	<LOD
	S4	47	101 (20)	<LOQ	<LOD
	S5	334	196 (39)	129	<LOD
	S6	56	165 (33)	<LOQ	<LOD
	S7	301	98 (20)	77	<LOD
	S8	203	176 (35)	72	<LOD
	S9	371	181 (36)	101	<LOD
	S10	133	250 (50)	82	<LOD
	S11	95	103 (21)	<LOQ	<LOD
Aug.	S1	32	<LOQ	<LOD	<LOD
	S2	65	<LOD	<LOQ	<LOD
	S3	137	<LOD	<LOQ	<LOD
	S4	44	<LOQ	<LOQ	<LOD
	S5	145	- ^d	73	<LOD
	S6	87	-	69	<LOD
	S7	53	-	<LOQ	<LOD
	S8	67	96 (19)	<LOQ	<LOD
	S9	102	<LOQ	<LOQ	<LOD
	S10	145	162 (32)	80	<LOD
	S11	50	<LOQ	<LOQ	<LOD
Sep.	S1	420	105 (21)	<LOQ	<LOD
	S2	32	121 (24)	<LOD	<LOD
	S3	142	<LOQ	<LOQ	<LOD
	S4	196	<LOQ	75	<LOD
	S5	272	-	109	<LOD
	S6	163	-	109	<LOD
	S7	101	-	92	<LOD
	S8	273	340 (68)	94	<LOD
	S9	249	99 (20)	71	<LOD
	S10	141	<LOQ	<LOQ	<LOD
	S11	233	187 (37)	96	<LOD
Oct.	S1	176	<LOQ	<LOQ	<LOD
	S2	327	98 (20)	92	<LOD
	S3	258	<LOD	73	<LOD
	S4	142	<LOD	81	<LOD
	S5	87	-	74	<LOD
	S6	74	-	74	<LOD
	S7	81	-	<LOQ	<LOD

	S8	205	<LOQ	74	<LOD
	S9	148	<LOQ	<LOQ	<LOD
	S10	191	<LOD	<LOQ	<LOD
	S11	170	<LOQ	78	<LOD
Nov.	S1	2344	194 (39)	151	<LOD
	S2	3315	<LOQ	<LOQ	<LOD
	S3	3938	<LOQ	92	<LOD
	S4	102	217 (43)	143	<LOD
	S5	110	99 (20)	<LOQ	<LOD
	S6	1232	-	<LOQ	<LOD
	S7	1122	-	101	<LOD
	S8	1241	-	<LOQ	<LOD
	S9	1670	141 (28)	<LOQ	<LOD
	S10	248	<LOQ	111	<LOD
	S11	1115	-	178	<LOD
Dec.	S1	219	1541 (308)	145	<LOD
	S2	142	955 (191)	<LOQ	<LOD
	S3	146	987 (197)	<LOQ	<LOD
	S4	1204	2261 (452)	138	<LOD
	S5	425	1225 (245)	<LOQ	<LOD
	S6	267	-	<LOQ	<LOD
	S7	271	1173 (235)	116	<LOD
	S8	139	1259 (252)	76	<LOD
	S9	1079	1100 (220)	112	<LOD
	S10	1014	777 (155)	76	<LOD
	S11	192	1116 (223)	105	<LOD

^a dw: Dry weight.

^b: Concentrations in ng g⁻¹ wet weight based on a conversion factor of 5:1 between wet weight and dry weight.

^c < LOD: Below limit of detection.

^d < LOQ: Below limit of quantification.

^e -: Not collected.

Table S4. ANOVA results for BMAA concentrations in phytoplankton and mussels along the South Sea Coast of Korea from January to December 2021.

	Month			Site		
	df	F	P	df	F	P
Phyto-BMAA	11	4.76	<0.001	10	1.09	0.38
Mussel-BMAA	11	37.45	<0.001	10	1.36	0.21

Table S5. Density of phytoplankton in the South Sea Coast of Korea from January to December 2021.

Month	Genus	Density (cells L ⁻¹)										
		st.1	st.2	st.3	st.4	st.5	st.6	st.7	st.8	st.9	st.10	st.11
Jan.	<i>Amphora</i>	0	0	0	0	0	0	0	454	0	0	1615
	<i>Asterionellopsis</i>	0	0	0	0	857	433	0	0	0	0	0
	<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Bacteriastrium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chaetoceros</i>	6936	135393	10242	60242	71098	45886	15634	3631	11571	10819	18570
	<i>Cosinodiscus</i>	0	0	0	0	2	2	0	4	4	28	10
	<i>Cylindrotheca</i>	462	0	0	0	0	433	0	454	890	1664	0
	<i>Dactyliosolen</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Detonula</i>	0	0	1707	0	857	0	0	908	3560	0	3230
	<i>Ditylum</i>	462	0	0	0	0	0	0	0	0	2497	807
	<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Eucampia</i>	0	5721	0	0	0	5195	823	30406	6231	7490	31489
	<i>Guinardia</i>	462	3337	0	873	0	0	411	0	1780	0	0
	<i>Gyrosigma</i>	0	0	0	0	0	5628	0	0	0	0	0
	<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Leptocylindrus</i>	0	0	0	0	0	0	0	0	5341	0	0
	<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Nitzschia</i>	0	0	5121	0	0	0	823	0	1780	2497	2422
	<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Odontella</i>	925	0	0	0	0	0	0	0	0	0	807
	<i>Pleurosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pseudo-nitzschia</i>	5086	10965	8535	43653	191022	22077	2057	4992	890	0	13726
	<i>Rhizosolenia</i>	92	953	0	0	1713	0	411	908	0	832	4844
	<i>Skeletonema</i>	1850	4291	0	315106	558461	32033	1646	4992	0	0	3230
	<i>Stephanopyxis</i>	0	0	0	0	857	0	0	0	0	0	0
	<i>Thalassionema</i>	18958	73417	10242	6111	10279	1299	823	2723	890	832	3230
	<i>Thalassiosira</i>	925	0	0	0	0	433	0	0	890	0	0
	<i>Lauderia</i>	0	0	0	0	0	1732	0	0	0	0	0
	<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Alexandrium</i>	462	0	0	0	0	0	0	0	890	1664	0
	<i>Akashiwo</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Ceratium</i>	925	477	0	0	0	0	0	0	0	0	0
	<i>Dinophysis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Gonyaulax</i>	0	0	0	0	0	433	0	0	0	0	0
	<i>Gymnodinium</i>	2774	1907	1707	2619	2570	3463	823	0	890	1664	3230
	<i>Gyrodinium</i>	1387	0	0	0	0	0	0	0	0	1664	0
	<i>Heterocapsa</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	0	0	0	0	857	0	0	0	0	0	0
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	462	0	0	0	0	0	0	0	0	0	0
<i>Protoperidinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	0	0	0	0	0	0	0	0	1780	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	24969	14302	25606	63734	11992	28570	65417	21784	85450	43274	20992
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
Unidentified	0	0	0	0	0	0	0	0	0	0	0
Feb. <i>Amphora</i>	0	0	0	0	0	0	0	0	814	885	0
<i>Asterionellopsis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Bacteriastrum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chaetoceros</i>	0	0	0	0	11964	18274	0	5863	4887	7079	14158
<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	2443	885	0
<i>Cylindrotheca</i>	0	0	0	0	0	0	0	0	814	0	0
<i>Dactyliosolen</i>	817	0	0	0	0	0	0	0	0	0	0
<i>Detonula</i>	1633	3520	0	0	0	0	0	0	0	7964	0
<i>Ditylum</i>	0	0	0	0	0	0	0	0	814	0	0
<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Eucampia</i>	2450	86240	1686	0	26491	0	0	46903	534262	315025	59462
<i>Guinardia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gyrosigma</i>	4900	0	0	0	0	0	0	0	0	0	0
<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Leptocylindrus</i>	0	0	0	0	0	0	0	0	0	0	944
<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nitzschia</i>	0	0	0	0	0	0	0	977	1629	2655	0
<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Pleurosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pseudo-nitzschia</i>	2450	0	0	2769	34183	11313	1545	2931	1629	0	2832
<i>Rhizosolenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Skeletonema</i>	3267	0	0	44301	193986	185352	9270	1954	30134	10619	4719
<i>Stephanopyxis</i>	0	0	843	0	0	0	0	0	3258	1770	1888
<i>Thalassionema</i>	1633	4400	0	923	22219	6962	0	0	814	1770	0
<i>Thalassiosira</i>	0	0	0	0	0	0	0	0	0	885	0
<i>Lauderia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Alexandrium</i>	817	1760	0	0	0	870	0	0	0	0	0
<i>Akashiwo</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Ceratium</i>	817	0	0	0	0	0	0	0	0	0	0
<i>Dinophysis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gonyaulax</i>	2450	880	0	1846	0	870	0	0	0	0	0
<i>Gymnodinium</i>	10616	17600	843	23996	17946	5221	773	0	814	3540	1888
<i>Gyrodinium</i>	817	5280	0	1846	0	0	0	0	814	0	0
<i>Heterocapsa</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	0	0	0	0	855	870	0	0	814	885	944
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Protoperidinium</i>	0	0	0	0	855	0	0	0	814	0	0
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	817	1760	1686	923	1709	0	0	0	2443	0	0
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	68597	337920	19388	14767	16237	271501	23175	78171	13031	23007	16989
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
Unidentified	0	0	0	0	0	0	0	0	0	0	0
Mar. <i>Amphora</i>	845	0	0	855	0	0	0	0	0	921	0
<i>Asterionellopsis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Bacteriastrum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chaetoceros</i>	4227	1784	4157	0	15609	0	0	0	0	6450	0
<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cylindrotheca</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dactyliosolen</i>	0	892	0	0	0	0	0	0	0	921	0
<i>Detonula</i>	3382	0	0	0	976	0	0	0	1712	2764	0
<i>Ditylum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Eucampia</i>	342381	53520	37412	238571	58532	2946	0	0	100994	121621	0
<i>Guinardia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gyrosigma</i>	0	0	0	0	2927	0	0	0	0	0	0
<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Leptocylindrus</i>	0	0	0	0	4878	4909	0	0	0	0	0
<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nitzschia</i>	845	892	0	2565	976	0	820	1659	856	2764	1033
<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pleurosigma</i>	0	0	0	0	976	0	0	0	0	0	0
<i>Pseudo-nitzschia</i>	0	3568	2494	0	4878	1964	2460	0	0	1843	0
<i>Rhizosolenia</i>	0	0	0	0	0	0	0	830	856	2764	0
<i>Skeletonema</i>	0	0	0	0	26339	0	0	0	0	0	0
<i>Stephanopyxis</i>	0	0	0	0	0	0	0	0	0	1843	0
<i>Thalassionema</i>	0	0	0	855	0	0	0	0	0	0	0
<i>Thalassiosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Lauderia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Alexandrium</i>	0	0	831	855	0	0	0	0	0	921	1033
<i>Akashiwo</i>	3382	0	0	0	0	0	0	0	0	0	0
<i>Ceratium</i>	0	0	0	0	0	0	0	0	0	0	1033
<i>Dinophysis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gonyaulax</i>	0	0	831	2565	0	0	0	0	0	0	0
<i>Gymnodinium</i>	0	3568	9145	2565	16584	7855	9840	0	3424	10135	8264
<i>Gyrodinium</i>	1691	2676	0	0	1951	0	2460	0	1712	921	0
<i>Heterocapsa</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	0	0	0	0	4878	0	3280	0	0	921	1033
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	0	0	0	0	0	0	1640	830	0	0	0
<i>Protoperidinium</i>	0	0	3325	0	0	0	0	830	0	2764	0

	<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	
	<i>Scrippsiella</i>	4227	1784	2494	1710	976	1964	0	830	856	2764	3099
	<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Blixia</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cryptomonas</i>	65095	21408	27435	14537	7804	11783	4920	6637	12838	0	21694
	<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Tetraseimis</i>	0	0	0	0	0	0	0	0	0	0	0
	Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
	Unidentified	0	0	0	0	0	0	0	0	0	0	0
Apr.	<i>Amphora</i>	488	0	0	937	0	0	0	0	0	194	0
	<i>Asterionellopsis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Bacteriastrium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chaetoceros</i>	11069	195594	56554	27179	33646	1917	960067	3717	0	0	33480
	<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cylindrotheca</i>	0	0	0	937	0	0	0	0	398	0	0
	<i>Dactyliosolen</i>	163	0	0	0	0	0	0	0	0	0	182
	<i>Detonula</i>	977	186	0	0	0	0	0	0	0	0	0
	<i>Ditylum</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Eucampia</i>	29952	2235	21208	14058	0	0	0	0	0	0	0
	<i>Guinardia</i>	0	0	9089	0	0	0	0	186	0	388	0
	<i>Gyrosigma</i>	0	0	0	0	0	0	2137	0	0	0	0
	<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Leptocylindrus</i>	0	0	82811	8435	0	1917	0	0	64522	6006	0
	<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Nitzschia</i>	326	0	0	0	0	192	178	0	0	0	0
	<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pleurosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pseudo-nitzschia</i>	2930	43962	19188	0	3542	1150	1069	7061	112317	25575	80426
	<i>Rhizosolenia</i>	488	186	2020	937	885	383	0	0	0	1356	0
	<i>Skeletonema</i>	6837	1118	51505	0	0	0	0	48686	35647	112956	324615
	<i>Stephanopyxis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Thalassionema</i>	977	3726	2020	937	885	0	1425	5203	10156	7944	0

<i>Thalassiosira</i>	0	0	0	937	0	0	0	557	0	0	0
<i>Lauderia</i>	0	0	0	0	0	0	0	743	0	0	546
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	326	373	0	0	0	192	0	0	0	0	0
<i>Alexandrium</i>	0	186	0	0	0	0	178	0	0	1163	182
<i>Akashiwo</i>	0	745	1010	0	1771	27995	0	0	0	194	364
<i>Ceratium</i>	0	0	0	187	0	0	0	0	398	3100	0
<i>Dinophysis</i>	0	0	0	0	0	0	0	186	0	2519	546
<i>Gonyaulax</i>	326	186	0	0	0	192	356	372	80	581	364
<i>Gymnodinium</i>	5535	83826	13129	2812	7969	5561	2672	372	597	15500	546
<i>Gyrodinium</i>	326	2980	5049	0	6198	1534	534	372	398	10656	182
<i>Heterocapsa</i>	163	0	0	0	0	0	178	0	0	581	0
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	651	186	11109	937	6198	2301	6590	0	398	581	182
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	326	0	0	937	0	0	178	0	398	11044	0
<i>Protoperidinium</i>	163	186	404	187	0	0	0	372	11550	1550	364
<i>Pyrophacus</i>	163	186	0	0	0	0	0	0	0	194	0
<i>Scrippsiella</i>	1139	13412	11109	7498	11510	5752	1781	6318	1593	5619	2911
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	163	186	1010	937	1771	192	178	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixa</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	67067	24962	38376	101218	244375	666121	6412	37537	113113	25575	4731
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nano-plankton</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Unidentified</i>	0	0	0	0	0	0	0	0	0	0	0
May											
<i>Amphora</i>	0	0	0	0	0	828	0	0	0	0	0
<i>Asterionellopsis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Bacillaria</i>	0	0	0	0	0	0	20145	0	0	0	0
<i>Bacteriastrum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chaetoceros</i>	22331	3242	5490	7387	15220	0	26591	31892	14163	72385	161258
<i>Cosinodiscus</i>	0	0	0	0	0	0	161	189	0	0	0
<i>Cylindrotheca</i>	859	0	0	0	0	0	0	0	0	883	0
<i>Dactyliosolen</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Detonula</i>	1718	811	0	1477	0	828	0	0	0	0	0
<i>Ditylum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Eucampia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Guinardia</i>	0	0	0	0	0	0	0	0	0	883	0
<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	1765	0
<i>Leptocylindrus</i>	0	0	0	0	895	0	2417	0	0	0	0
<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nitzschia</i>	12883	2432	1569	3693	10744	2483	2417	778	0	1765	0
<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pleurosigma</i>	0	0	784	739	0	0	0	0	0	0	0
<i>Pseudo-nitzschia</i>	5153	0	3137	5909	1791	0	8058	0	0	3531	0
<i>Rhizosolenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Skeletonema</i>	8589	0	0	0	0	0	40290	0	7967	37075	24682
<i>Stephanopyxis</i>	0	0	0	0	0	0	0	0	0	0	2468
<i>Thalassionema</i>	0	811	0	0	2686	828	0	0	885	3531	823
<i>Thalassiosira</i>	4294	2432	0	6648	0	828	0	0	0	0	0
<i>Lauderia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Alexandrium</i>	258	162	0	0	0	166	0	156	0	0	0
<i>Akashiwo</i>	859	102941	32939	2955	895	0	0	0	0	0	0
<i>Ceratium</i>	0	0	0	0	0	0	0	0	1770	177	823
<i>Dinophysis</i>	0	811	0	0	358	0	0	156	1770	177	823
<i>Gonyaulax</i>	0	162	784	148	895	497	2417	156	885	1059	4114
<i>Gymnodinium</i>	3436	0	3137	2216	8953	8278	5641	3889	9737	6179	15632
<i>Gyrodinium</i>	859	4863	0	1477	895	0	806	778	9737	5296	2468
<i>Heterocapsa</i>	0	0	0	0	0	0	0	1556	0	883	0
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	859	8916	4706	0	895	828	4029	1556	4426	2648	6582
<i>Oxytoxum</i>	0	0	0	0	895	0	0	0	885	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	0	0	0	0	0	0	3223	3111	0	3531	8227
<i>Protoperidinium</i>	859	4053	784	0	1791	0	0	0	2656	1765	0
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	0	6484	3921	8864	20592	4139	3223	1556	0	10593	26328
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	1718	3242	784	0	0	0	0	0	885	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0

	<i>Blixa</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cryptomonas</i>	16319	12158	12548	8125	8058	278141	62852	35781	175270	358395	36201
	<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
	Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
	Unidentified	0	0	0	0	0	0	0	0	0	0	0
Jun.	<i>Amphora</i>	1117	0	442	0	0	0	0	0	545	0	0
	<i>Asterionellopsis</i>	0	0	0	0	0	0	0	0	0	0	563
	<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Bacteriastrum</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cerataulina</i>	1117	0	0	0	0	0	0	0	0	0	0
	<i>Chaetoceros</i>	228904	405538	182157	106888	50047	22858	465821	110729	6538	90049	369878
	<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cylindrotheca</i>	0	0	0	0	0	0	0	543	0	0	0
	<i>Dactyliosolen</i>	1675	0	442	0	801	0	0	0	0	1133	0
	<i>Detonula</i>	5583	0	0	2620	0	0	1499	0	0	0	0
	<i>Ditylum</i>	3350	4030	0	0	0	0	0	0	0	0	0
	<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Eucampia</i>	1675	0	0	0	0	0	0	0	0	0	0
	<i>Guinardia</i>	0	0	442	0	0	0	0	0	0	566	0
	<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Hemiaulus</i>	0	0	0	524	0	0	0	0	0	0	0
	<i>Leptocylindrus</i>	7258	0	4863	129943	8008	0	37985	0	4358	13592	57424
	<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Navicular</i>	0	1008	884	0	0	0	0	0	0	0	0
	<i>Nitzschia</i>	558	0	0	1572	0	544	0	0	0	0	563
	<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Odontella</i>	2792	0	0	0	0	0	0	0	0	0	0
	<i>Pleurosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pseudo-nitzschia</i>	101053	36272	41560	377253	34032	56600	57978	20626	32688	44741	385079
	<i>Rhizosolenia</i>	1117	0	0	0	0	0	0	0	0	0	0
	<i>Skeletonema</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Stephanopyxis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Thalassionema</i>	3908	0	30065	6288	93288	0	21992	13027	7627	6796	114848
	<i>Thalassiosira</i>	0	0	0	0	0	0	0	1086	0	0	0
	<i>Lauderia</i>	558	0	0	0	801	0	500	0	545	0	0
	<i>Meuniera</i>	0	0	0	0	400	0	0	0	0	1133	0
	<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Amphidinium</i>	0	504	0	1572	0	0	0	0	0	0	0
	<i>Alexandrium</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Akashiwo</i>	0	0	0	524	0	0	0	0	545	0	2252
<i>Ceratium</i>	9491	5038	1769	524	1602	11429	1000	3800	2179	2832	16889
<i>Dinophysis</i>	0	7557	0	5240	0	479467	0	0	0	0	0
<i>Gonyaulax</i>	2233	504	0	524	0	5442	1000	10856	1634	566	2815
<i>Gymnodinium</i>	2233	5038	0	1048	0	0	1000	543	0	0	1689
<i>Gyrodinium</i>	19541	22670	6190	16767	12011	3265	8497	1628	11986	18123	9008
<i>Heterocapsa</i>	4466	1511	0	2620	3603	9796	0	0	2179	566	563
<i>Karenia</i>	558	1008	0	0	0	0	0	0	0	0	563
<i>Katodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Oxytoxum</i>	0	1008	0	1048	1201	0	1000	1086	1634	2265	1126
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	1117	2015	442	0	400	1088	1499	1628	2724	2832	6756
<i>Protoperdinium</i>	1117	1511	884	5240	2002	1633	4498	6513	1090	5097	2252
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	1117	3023	884	2096	0	8163	1999	4885	1090	566	1689
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixa</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	1086	1090	2832	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	207688	485638	26528	11527	22822	117554	138947	162837	148188	453643	1240810
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
Unidentified	0	0	0	0	0	0	0	0	0	0	0
Jul. <i>Amphora</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asterionellopsis</i>	0	382	0	0	0	0	0	0	0	0	0
<i>Bacillaria</i>	0	0	0	0	0	0	0	0	886	0	0
<i>Bacteriastrum</i>	0	0	0	446	0	0	0	0	0	0	0
<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chaetoceros</i>	128056	74090	147553	360951	17356	82440	11259	299501	44321	122936	66251
<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cylindrotheca</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dactyliosolen</i>	0	1146	0	0	0	0	0	0	0	0	0
<i>Detonula</i>	0	1910	0	0	0	0	0	1234	3102	0	0
<i>Ditylum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Entomoneis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Eucampia</i>	0	0	0	891	0	0	0	0	0	3771	0
<i>Guinardia</i>	902	0	0	0	868	0	0	0	0	0	381
<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Hemiaulus</i>	0	0	0	1782	0	0	0	0	0	754	0
<i>Leptocylindrus</i>	36072	245949	227802	86896	180504	34350	0	2468	2659	9050	0
<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	377	0
<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nitzschia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pleurosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pseudo-nitzschia</i>	250700	420100	870651	1234365	67689	75570	34211	309786	499495	47515	22084
<i>Rhizosolenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Skeletonema</i>	73948	4583	18121	213897	42523	0	0	81458	9307	22626	15230
<i>Stephanopyxis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Thalassionema</i>	0	0	0	891	0	0	0	1234	886	1886	0
<i>Thalassiosira</i>	0	0	863	446	0	0	0	0	0	0	0
<i>Lauderia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	377	0
<i>Alexandrium</i>	902	0	431	0	0	382	0	0	443	0	381
<i>Akashiwo</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Ceratium</i>	451	382	431	0	0	0	433	1234	0	377	1904
<i>Dinophysis</i>	0	0	0	0	434	0	0	0	0	0	762
<i>Gonyaulax</i>	451	764	1294	0	1302	0	8228	6171	0	754	762
<i>Gymnodinium</i>	9469	50794	54362	20053	15621	5343	16456	83103	2216	1886	48356
<i>Gyrodinium</i>	2705	4201	3452	2228	0	0	0	823	0	0	4569
<i>Heterocapsa</i>	0	0	0	0	0	0	0	0	0	0	381
<i>Karenia</i>	0	9548	3020	0	434	0	1732	0	0	377	0
<i>Katodinium</i>	2705	2673	0	0	434	382	0	0	0	754	1142
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	0	8402	4746	0	1736	291975	118224	823	5318	754	762
<i>Protoperidinium</i>	2705	3819	1294	1337	6509	1145	0	4937	7091	377	762
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	7665	4201	15532	2228	24299	0	0	4525	3102	5279	33506
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	1146	0	0	0	0	0	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	0	0	10786	0	0	0	0	0	0	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	381
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	15331	266191	157045	16042	9546	116027	11693	53071	33241	236821	31603
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0

	<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
	Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
	Unidentified	0	0	0	0	0	0	0	0	0	0	0
Aug.	<i>Amphora</i>	0	0	0	0	0	0	0	761	0	0	0
	<i>Asterionellopsis</i>	39125	3930	1312	0	3598	5240	64242	448340	1901	5780	35123
	<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	1239	0
	<i>Bacteriastrium</i>	1276	786	0	0	0	0	2409	3291	0	0	0
	<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chaetoceros</i>	671931	536394	317508	517959	433611	161578	995743	218000	0	6606	393378
	<i>Cosinodiscus</i>	0	0	0	0	0	0	0	411	0	0	0
	<i>Cylindrotheca</i>	851	393	1749	9756	8096	3494	28106	65811	51339	7845	43904
	<i>Dactyliosolen</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Detonula</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Ditylum</i>	425	393	0	0	0	0	0	0	0	0	0
	<i>Entomoneis</i>	0	0	0	0	0	0	0	411	0	0	0
	<i>Eucampia</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Guinardia</i>	10207	1179	2624	3991	1799	3494	4015	1645	24719	14451	3073
	<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Leptocylindrus</i>	9356	0	5248	15078	24289	2620	0	102830	60846	36334	32489
	<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Navicular</i>	851	1572	0	0	0	0	0	0	0	826	0
	<i>Nitzschia</i>	13609	9824	0	0	0	873	0	12340	0	0	0
	<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pleurosigma</i>	0	786	0	0	0	0	0	0	0	0	0
	<i>Pseudo-nitzschia</i>	478432	622846	102337	62084	98057	159395	72272	518264	156267	468211	153663
	<i>Rhizosolenia</i>	0	2358	0	0	0	873	0	0	24719	5368	0
	<i>Skeletonema</i>	1435721	742700	126828	313081	51278	0	48181	185094	26620	10735	26342
	<i>Stephanopyxis</i>	0	393	0	0	0	0	402	823	0	0	0
	<i>Thalassionema</i>	2126	0	0	0	0	0	12045	8226	3803	2477	878
	<i>Thalassiosira</i>	851	3537	1312	0	0	0	0	8226	3423	413	0
	<i>Lauderia</i>	0	1572	0	0	0	0	0	823	0	0	0
	<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Alexandrium</i>	425	0	875	1330	0	0	402	0	0	0	0
	<i>Akashiwo</i>	0	0	0	443	0	0	0	0	0	0	0
	<i>Ceratium</i>	851	393	0	443	450	0	0	0	532	413	0
	<i>Dinophysis</i>	0	0	175	0	0	0	402	411	152	826	0
	<i>Gonyaulax</i>	851	0	1749	887	0	0	0	823	380	5368	878
	<i>Gymnodinium</i>	11482	4716	16182	13304	9446	17468	2409	4113	10268	5368	8342
	<i>Gyrodinium</i>	4253	1179	2624	3548	0	873	1205	3702	0	1239	2195

	<i>Heterocapsa</i>	425	0	875	887	0	4367	0	0	0	1652	0
	<i>Karenia</i>	0	0	1749	0	0	1310	0	0	0	413	0
	<i>Katodinium</i>	2126	0	3061	0	3149	6114	0	0	0	826	3512
	<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Prorocentrum</i>	7655	393	6560	4878	1799	2183	2811	0	0	0	0
	<i>Protoperidinium</i>	4253	0	3936	5765	0	437	2409	411	0	413	0
	<i>Pyrophacus</i>	0	0	1312	0	0	0	402	0	0	413	0
	<i>Scrippsiella</i>	851	393	3499	16408	0	1747	2409	2057	380	3716	3073
	<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cochlodinium</i>	0	0	1225	23060	0	0	0	823	0	0	0
	<i>Podolampas</i>	425	0	0	0	0	0	0	0	0	0	0
	<i>Blixia</i>	0	0	0	0	0	0	0	2057	0	4129	0
	<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chatonella</i>	0	0	350	0	0	0	0	0	152	0	0
	<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cryptomonas</i>	17436	21613	2187	5321	1799	3494	13651	14396	7606	53675	50050
	<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Tetrasetmis</i>	0	0	0	0	0	0	0	0	0	0	0
	Nano-plankton	8505	0	0	0	0	0	0	0	0	0	0
	Unidentified	0	0	0	0	0	0	0	0	0	0	0
Sep.	<i>Amphora</i>	0	0	0	899	0	0	0	0	0	0	0
	<i>Asterionellopsis</i>	0	0	0	0	0	0	223725	0	0	0	0
	<i>Bacillaria</i>	0	0	800	0	0	0	0	0	0	0	0
	<i>Bacteriastrum</i>	0	3365	0	0	0	0	0	0	0	0	0
	<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chaetoceros</i>	2117	93387	484800	0	1782	12249	284563	0	0	2738	166752
	<i>Cosinodiscus</i>	0	421	0	0	0	817	0	0	0	0	0
	<i>Cylindrotheca</i>	423	2524	3200	899	891	1225	29438	0	1534	2738	2338
	<i>Dactyliosolen</i>	0	0	0	0	0	1633	0	0	0	0	0
	<i>Detonula</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Ditylum</i>	0	421	2000	0	0	408	0	0	0	0	390
	<i>Entomoneis</i>	0	0	0	899	0	0	0	0	0	0	0
	<i>Eucampia</i>	1693	355879	73600	0	0	19598	9813	882	0	0	1558
	<i>Guinardia</i>	1693	40804	4400	0	0	31848	37288	0	0	0	1948
	<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Leptocylindrus</i>	0	32812	6000	0	2673	625518	70650	0	0	1825	575061
	<i>Licmphora</i>	0	0	0	449	0	0	0	0	383	456	0
	<i>Navicular</i>	1270	421	0	0	0	0	0	0	0	456	0
	<i>Nitzschia</i>	7619	15144	40800	0	0	2450	0	0	0	0	0
	<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pleurosigma</i>	0	421	0	0	0	0	0	0	0	456	0
<i>Pseudo-nitzschia</i>	28785	282684	273600	28763	0	73494	788925	5289	3067	319442	773761
<i>Rhizosolenia</i>	2540	1262	0	0	0	7349	6280	0	0	456	0
<i>Skeletonema</i>	1693	7572	60000	4944	0	11432	129525	0	0	12778	539996
<i>Stephanopyxis</i>	847	14723	800	0	0	817	393	0	0	0	0
<i>Thalassionema</i>	423	12620	0	0	0	0	43175	0	0	4563	65454
<i>Thalassiosira</i>	3810	10096	7200	3595	3119	17149	1963	0	0	0	14026
<i>Lauderia</i>	0	2945	0	0	0	4900	11775	0	0	456	0
<i>Meuniera</i>	423	0	0	0	0	817	0	0	0	0	0
<i>Asteromphalus</i>	0	421	0	0	0	408	0	0	0	0	0
<i>Amphidinium</i>	0	421	0	0	0	0	0	0	0	0	0
<i>Alexandrium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Akashiwo</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Ceratium</i>	847	0	600	0	0	0	0	0	0	0	0
<i>Dinophysis</i>	0	0	0	0	0	0	393	0	0	0	0
<i>Gonyaulax</i>	0	0	2000	0	2228	0	0	441	0	0	0
<i>Gymnodinium</i>	9736	5469	4000	3146	7129	817	1963	882	71317	9127	1948
<i>Gyrodinium</i>	2540	2524	3200	899	5793	4491	393	441	5368	1369	2727
<i>Heterocapsa</i>	3386	2945	0	1798	891	0	2355	6171	3834	3651	1169
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	1693	841	0	3146	4456	817	0	0	2301	0	0
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	423	421	1200	0	0	0	785	0	0	456	0
<i>Protoperidinium</i>	0	5469	400	2247	891	4900	1570	441	767	0	2338
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	4233	6310	1600	0	3119	1633	1178	18072	6902	913	1169
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixa</i>	0	0	400	0	0	0	0	3967	0	0	0
<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	755171	262492	75200	32358	16041	14699	17663	186445	832034	142380	4675
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nano-plankton</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Unidentified</i>	0	0	0	0	0	0	0	0	0	0	0
Oct. <i>Amphora</i>	0	0	0	0	0	0	0	0	0	450	447
<i>Asterionellopsis</i>	0	0	2367	0	5428	2712	30148	0	0	0	0

<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Bacteriastrum</i>	0	0	0	0	3619	0	0	0	0	0	0
<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chaetoceros</i>	2433	4321	97817	0	59711	131075	45222	0	0	0	2684
<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cylindrotheca</i>	1216	5041	4733	423	5428	6328	0	0	2211	7195	10736
<i>Dactyliosolen</i>	0	0	0	0	2262	0	0	0	0	0	0
<i>Detonula</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Ditylum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Entomoneis</i>	405	0	0	0	0	0	0	0	0	0	0
<i>Eucampia</i>	0	0	0	0	2714	1808	5862	0	0	0	0
<i>Guinardia</i>	0	0	5522	0	23522	15819	2094	0	0	899	0
<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Leptocylindrus</i>	2433	3601	7888	0	0	0	10049	0	0	0	895
<i>Licmphora</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Navicular</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Nitzschia</i>	0	720	789	847	5428	2260	2512	858	0	899	447
<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Odontella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pleurosigma</i>	0	0	0	0	0	452	2094	0	0	0	0
<i>Pseudo-nitzschia</i>	8109	112709	425977	4233	982511	319099	2155599	27042	38021	286912	144942
<i>Rhizosolenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Skeletonema</i>	0	12243	81251	0	127564	4520	165815	1717	0	0	0
<i>Stephanopyxis</i>	0	0	0	0	905	0	0	0	0	0	0
<i>Thalassionema</i>	811	1080	3155	423	9047	2712	196801	0	442	899	1789
<i>Thalassiosira</i>	0	0	3944	423	18094	4972	3350	429	0	899	895
<i>Lauderia</i>	0	0	8677	0	160133	19887	2512	0	0	0	0
<i>Meuniera</i>	0	0	0	0	452	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	442	0	0
<i>Alexandrium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Akashiwo</i>	0	360	0	0	0	0	0	0	0	0	0
<i>Ceratium</i>	1216	180	0	0	452	0	0	0	0	0	0
<i>Dinophysis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gonyaulax</i>	0	0	394	0	0	0	837	0	0	0	0
<i>Gymnodinium</i>	6082	1080	3550	8890	9952	4520	8793	1717	17684	11692	3579
<i>Gyrodinium</i>	2027	720	3155	1693	2714	452	4187	1717	3979	1349	2684
<i>Heterocapsa</i>	1622	0	3550	1270	452	0	6281	1717	5305	899	1342
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	10137	0	789	3387	1357	0	1675	1288	13263	5396	4026
<i>Oxytoxum</i>	0	360	0	0	452	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	2027	0	50092	423	3166	0	0	0	0	0	0

	<i>Protoperidinium</i>	405	720	394	423	0	452	1256	0	1326	0	0
	<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Scrippsiella</i>	0	360	789	1270	1357	0	23449	2575	1326	1799	3131
	<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	899	0
	<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Blixia</i>	405	0	0	0	0	0	0	0	0	0	447
	<i>Dictyocha</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cryptomonas</i>	8515	18725	4733	8890	3619	5424	13399	7726	58357	70154	13868
	<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
	Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
	Unidentified	0	0	0	0	0	0	0	0	0	0	0
Nov.	<i>Amphora</i>	197	0	0	0	0	0	0	0	0	0	0
	<i>Asterionellopsis</i>	0	0	0	0	0	18398	0	0	0	0	0
	<i>Bacillaria</i>	0	0	0	0	0	0	878	628	1478	0	0
	<i>Bacteriastrium</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Chaetoceros</i>	20517	19023	6412	13642	18827	10769	31829	628	2032	1294	2729
	<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	369	431	0
	<i>Cylindrotheca</i>	789	0	0	0	224	224	0	0	369	0	210
	<i>Dactyliosolen</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Detonula</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Ditylum</i>	197	221	0	0	0	0	0	628	185	0	210
	<i>Entomoneis</i>	0	0	0	0	0	224	220	0	185	216	210
	<i>Eucampia</i>	7497	139357	9132	21021	20172	0	3951	36002	76473	18329	47029
	<i>Guinardia</i>	0	885	777	1342	672	2917	0	0	369	216	210
	<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Hemiaulus</i>	0	2654	0	0	0	0	220	0	0	0	0
	<i>Leptocylindrus</i>	0	0	389	0	672	449	0	0	554	0	0
	<i>Licmphora</i>	0	0	0	0	224	224	0	0	0	0	0
	<i>Navicular</i>	1973	442	0	2684	448	0	0	0	4987	216	210
	<i>Nitzschia</i>	395	0	0	0	0	224	659	209	0	0	420
	<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
	<i>Odontella</i>	789	0	0	0	0	0	1098	1047	0	0	0
	<i>Pleurosigma</i>	197	0	0	0	0	449	0	0	369	0	0
	<i>Pseudo-nitzschia</i>	209903	48664	88986	65301	423390	462187	21073	1675	3879	1725	1470
	<i>Rhizosolenia</i>	0	0	0	224	0	224	0	0	0	0	0
	<i>Skeletonema</i>	4735	2212	1360	1342	2017	8077	400386	0	0	647	2729
	<i>Stephanopyxis</i>	395	885	0	224	448	0	659	0	369	0	0

<i>Thalassionema</i>	0	221	583	0	0	0	0	1047	0	0	630
<i>Thalassiosira</i>	7102	0	3303	2907	165860	5385	1976	1884	739	431	210
<i>Lauderia</i>	395	664	0	0	0	0	0	0	0	0	0
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Alexandrium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Akashiwo</i>	197	6636	289301	447	0	0	0	0	0	0	0
<i>Ceratium</i>	0	885	0	0	672	224	0	0	0	0	210
<i>Dinophysis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gonyaulax</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gymnodinium</i>	2367	1770	777	447	1121	224	1537	209	185	1078	2519
<i>Gyrodinium</i>	395	885	194	224	672	673	659	0	0	863	210
<i>Heterocapsa</i>	0	0	0	0	0	0	0	0	185	216	0
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	1184	2212	0	0	448	0	0	209	0	0	630
<i>Oxytoxum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	0	0	194	0	672	0	0	0	0	216	420
<i>Protoperidinium</i>	395	442	0	0	224	0	439	0	0	0	0
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	395	664	0	0	672	0	439	0	0	216	1470
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixa</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	4340	1770	0	0	0	0	0	0	0	0	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	24462	7742	3109	8722	24207	46667	27219	7117	4803	13585	13017
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
Unidentified	0	0	0	0	0	0	0	0	0	0	0
Dec. <i>Amphora</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asterionellopsis</i>	0	0	617	433	0	213	0	0	0	0	0
<i>Bacillaria</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Bacteriastrum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cerataulina</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chaetoceros</i>	0	2991	11526	141951	887	175684	12336	2179	3079	13972	1469
<i>Cosinodiscus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cylindrotheca</i>	658	199	206	433	0	0	0	654	660	0	0

<i>Dactyliosolen</i>	0	0	0	0	0	0	0	218	0	0	0
<i>Detonula</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Ditylum</i>	0	1994	0	433	443	0	0	0	0	205	0
<i>Entomoneis</i>	0	0	0	0	222	0	0	0	0	411	0
<i>Eucampia</i>	0	340173	0	0	5322	0	881	139041	125813	252730	52057
<i>Guinardia</i>	0	997	0	0	0	0	0	654	0	0	0
<i>Gyrosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Hemiaulus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Leptocylindrus</i>	0	0	412	0	0	426	0	0	0	205	0
<i>Licmphora</i>	219	0	0	0	222	213	220	0	0	411	0
<i>Navicular</i>	219	0	412	216	443	0	220	654	440	1027	840
<i>Nitzschia</i>	0	399	0	216	222	426	0	218	660	0	210
<i>Melosira</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Odontella</i>	0	0	0	0	0	426	0	0	0	0	0
<i>Pleurosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pseudo-nitzschia</i>	658	798	14819	231969	1109	440594	2643	1308	0	616	0
<i>Rhizosolenia</i>	0	0	0	433	0	0	0	0	220	0	0
<i>Skeletonema</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Stephanopyxis</i>	0	199	0	866	0	0	0	1090	1540	616	0
<i>Thalassionema</i>	1097	798	0	0	1774	852	220	872	220	0	0
<i>Thalassiosira</i>	658	1196	1441	4328	443	1704	0	218	880	411	210
<i>Lauderia</i>	0	199	0	0	0	426	0	0	0	1438	0
<i>Meuniera</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Asteromphalus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Amphidinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Alexandrium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Akashiwo</i>	0	199	119375	0	0	7027	0	0	0	205	0
<i>Ceratium</i>	0	7777	0	2813	665	0	0	0	0	0	0
<i>Dinophysis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Gonyaulax</i>	0	0	0	216	0	426	0	0	0	0	0
<i>Gymnodinium</i>	1974	598	412	433	1552	639	1101	0	440	1849	2309
<i>Gyrodinium</i>	219	199	1235	433	1330	1278	0	0	880	205	0
<i>Heterocapsa</i>	0	0	206	0	222	0	0	0	220	205	0
<i>Karenia</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Katodinium</i>	219	997	1852	0	443	852	0	654	880	1233	840
<i>Oxytoxum</i>	0	199	0	0	0	0	0	0	0	0	0
<i>Polykrikos</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Prorocentrum</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Protoperidinium</i>	0	399	0	216	0	213	0	0	440	411	0
<i>Pyrophacus</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Scrippsiella</i>	0	0	0	866	1330	852	0	218	220	411	210
<i>Trodinium</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Pyrocystis</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cochlodinium</i>	0	0	0	0	0	0	0	0	0	0	0

<i>Podolampas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Blixa</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Dictyocha</i>	658	598	0	0	443	0	0	0	440	205	0
<i>Chatonella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Heterosigma</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Cryptomonas</i>	16229	4387	2881	11252	23725	4259	59476	5666	30353	20547	14903
<i>Eutreptiella</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Chlamidomonas</i>	0	0	0	0	0	0	0	0	0	0	0
<i>Tetraselmis</i>	0	0	0	0	0	0	0	0	0	0	0
Nano-plankton	0	0	0	0	0	0	0	0	0	0	0
Unidentified	0	0	0	0	0	0	0	0	0	0	0

Supplementary Figures

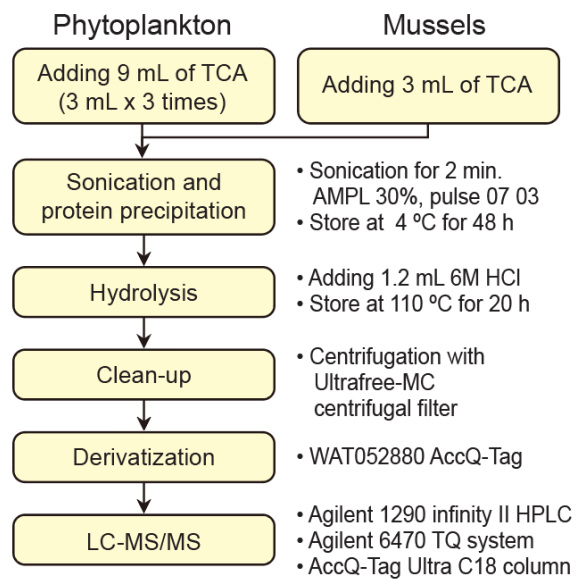


Fig. S1. Workflow of extraction, clean-up, and instrumental analysis of BMAA in phytoplankton and mussels.

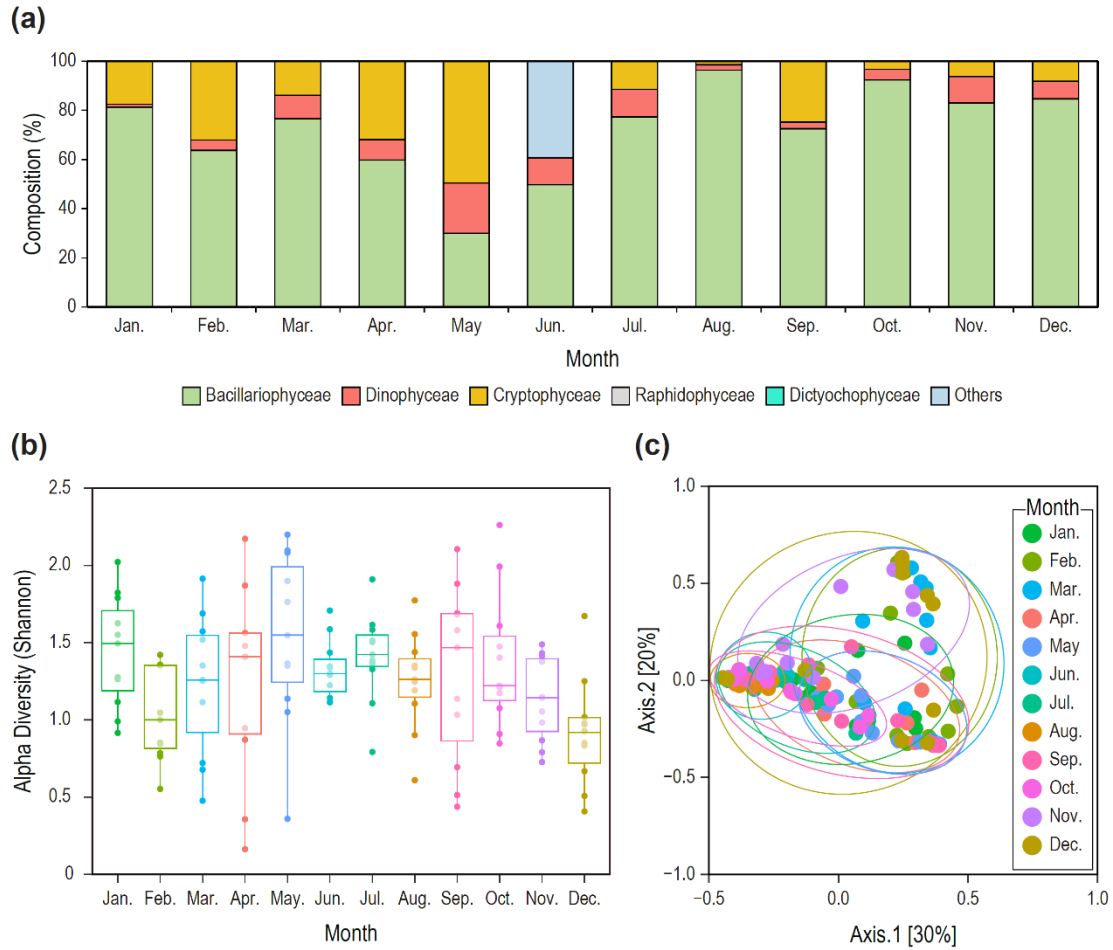


Fig. S2. (a) Phytoplankton compositions at the class level, and (b) alpha and (c) beta diversity of phytoplankton at the genus level along the South Sea Coast of Korea in 2021. (b) A box plot based on the Shannon index was used to determine phytoplankton genus richness within each month. (c) A principal coordinate analysis (PCoA) plot based on Bray-Curtis dissimilarity was used to assess the difference between months in genus composition.

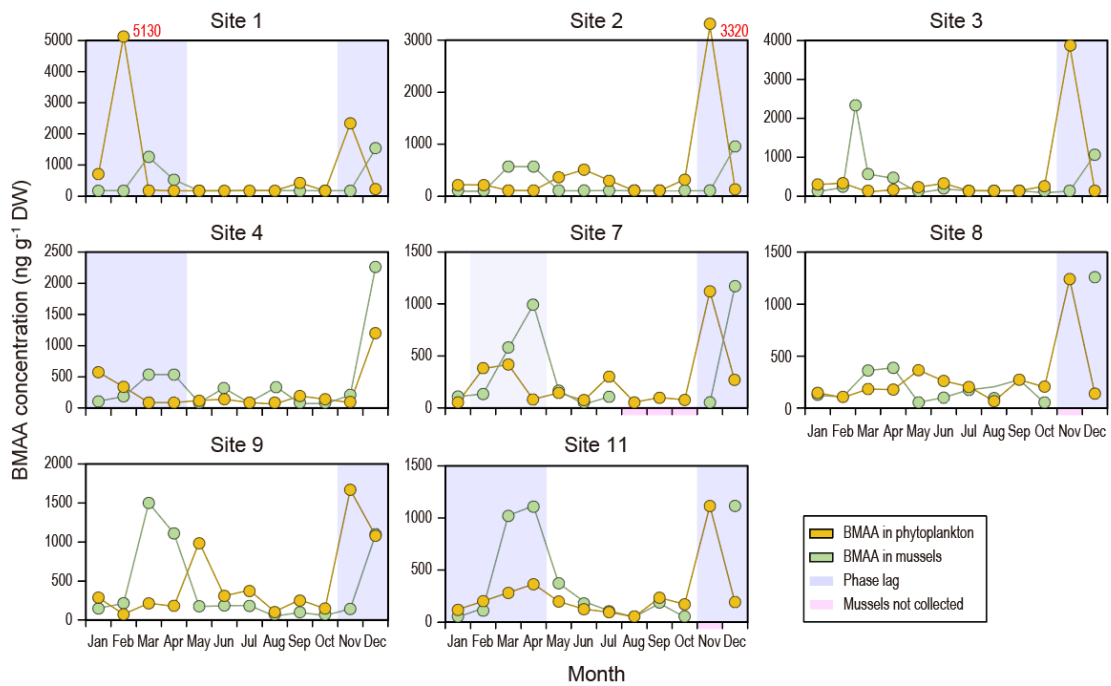


Fig. S3. Phase lag between phytoplankton and mussels in BMAA concentrations at the specific sites. The numbers above the y-axis have been rounded to the nearest whole number.

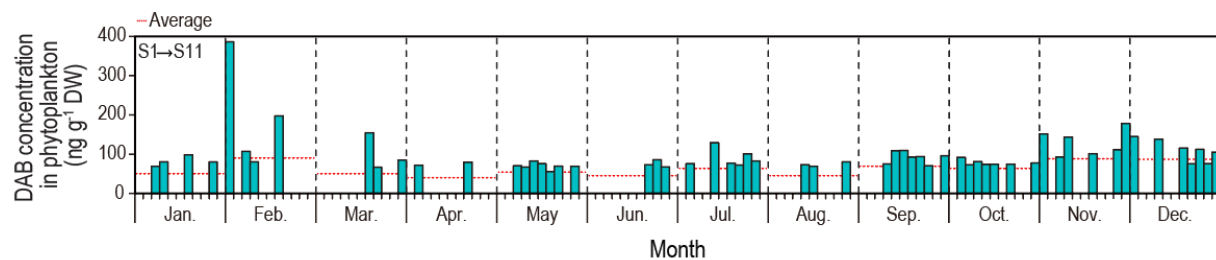


Fig. S4. Monthly distributions of DAB in phytoplankton in the South Sea Coast of Korea in 2021.

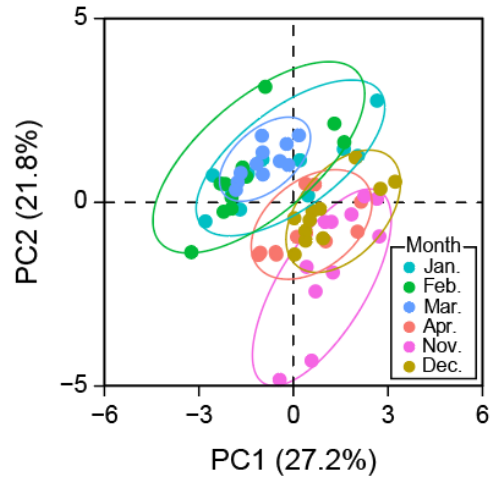


Fig. S5. Principal component analysis (PCA) using environmental factors in January, February, March, April, November, and December 2021.

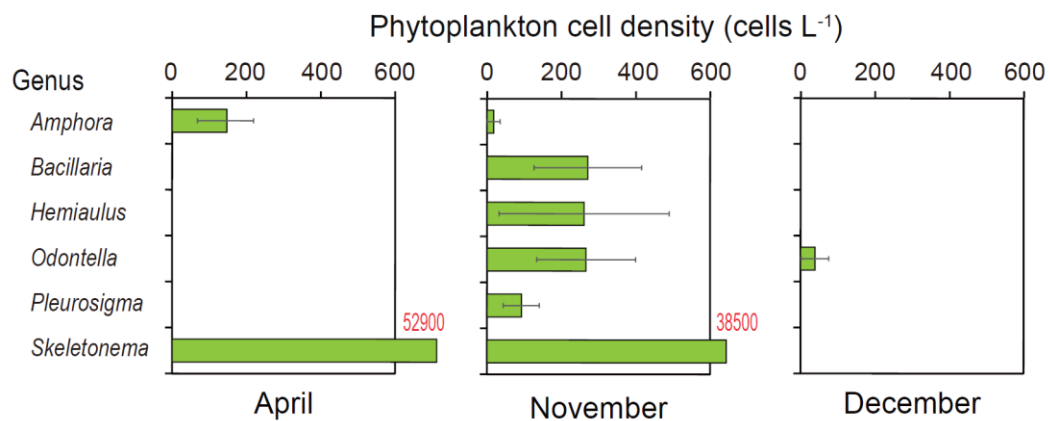


Fig. S6. Average cell numbers of six diatom genera in the South Sea Coast of Korea in April, November, and December 2021.