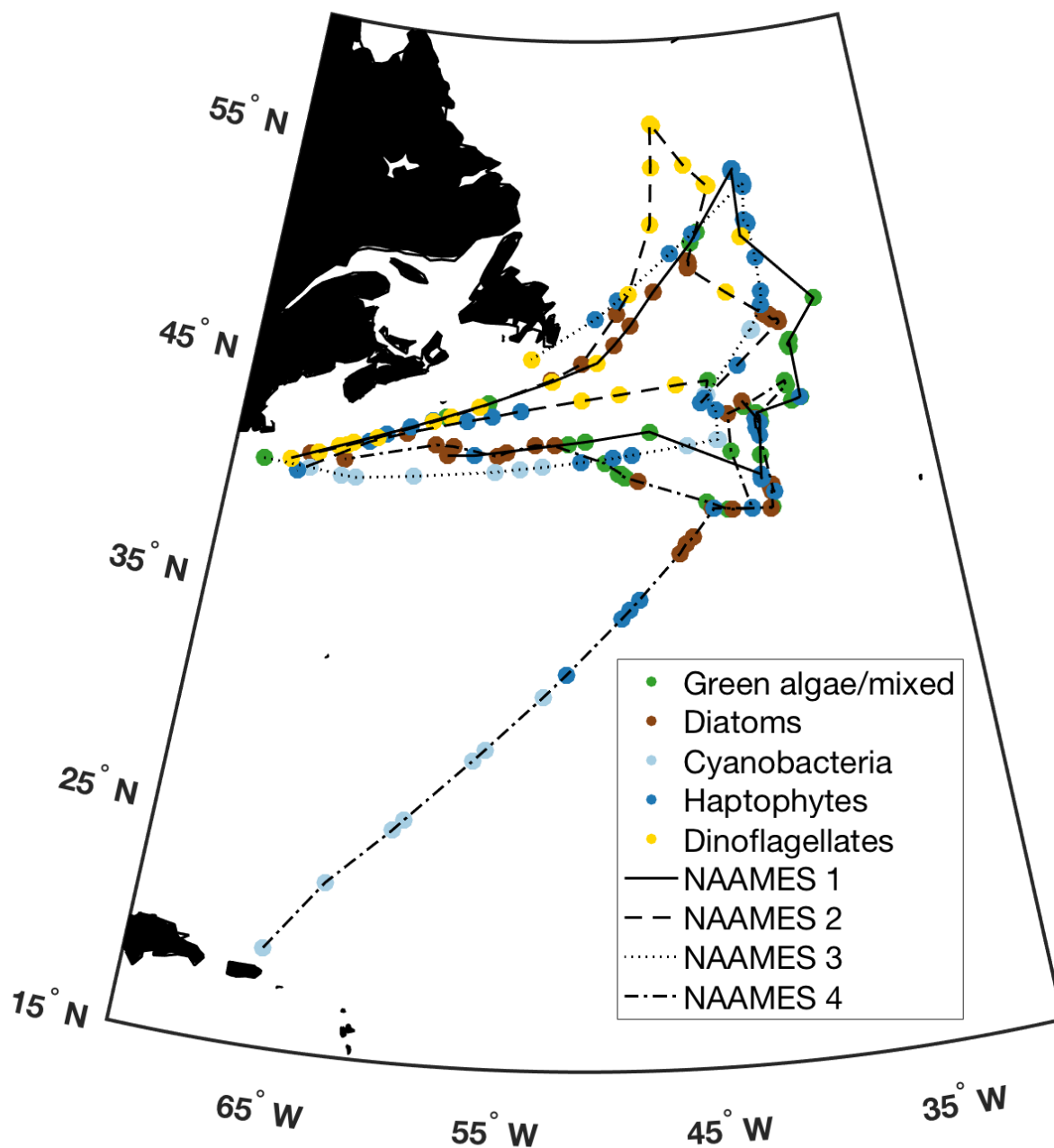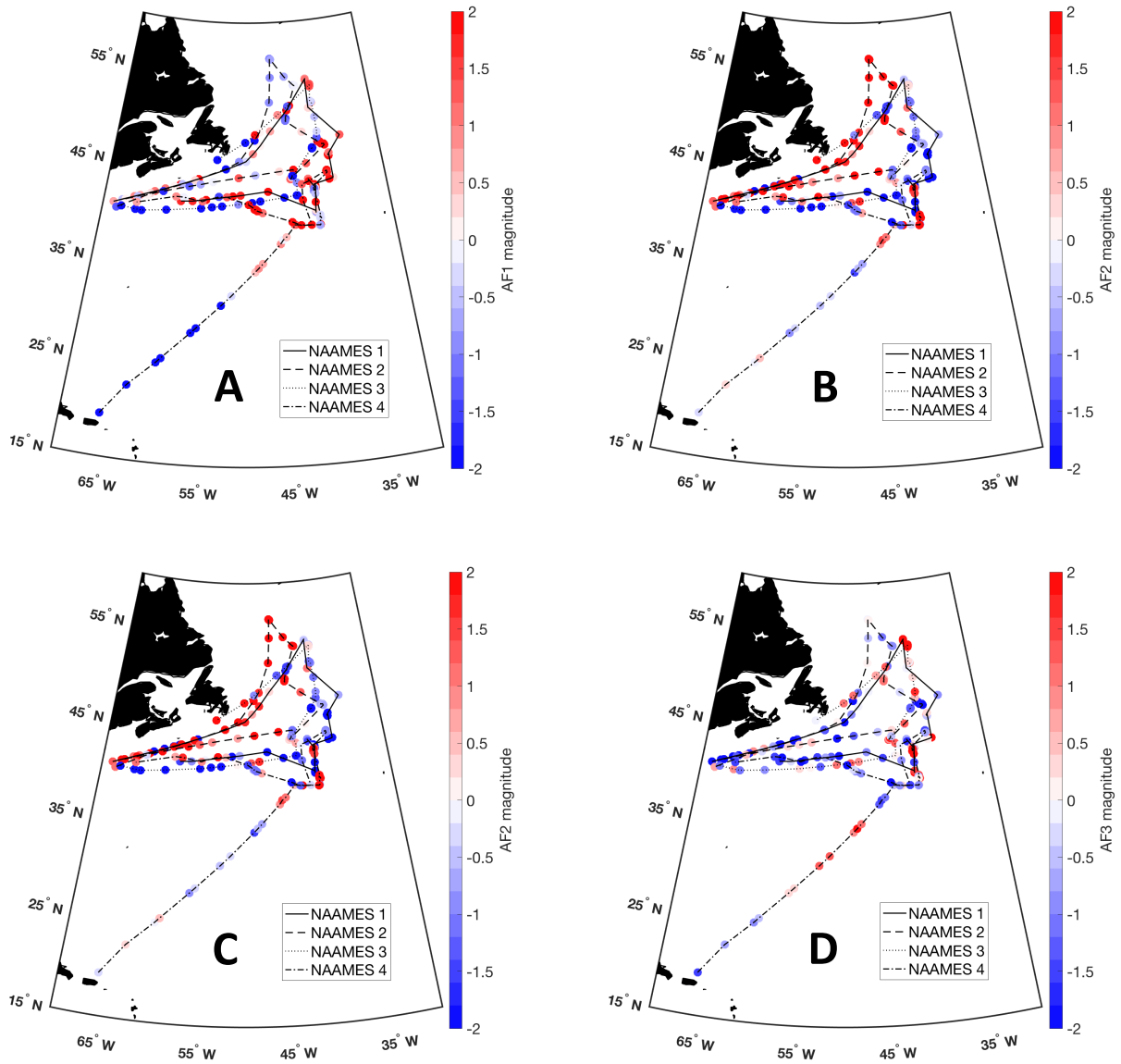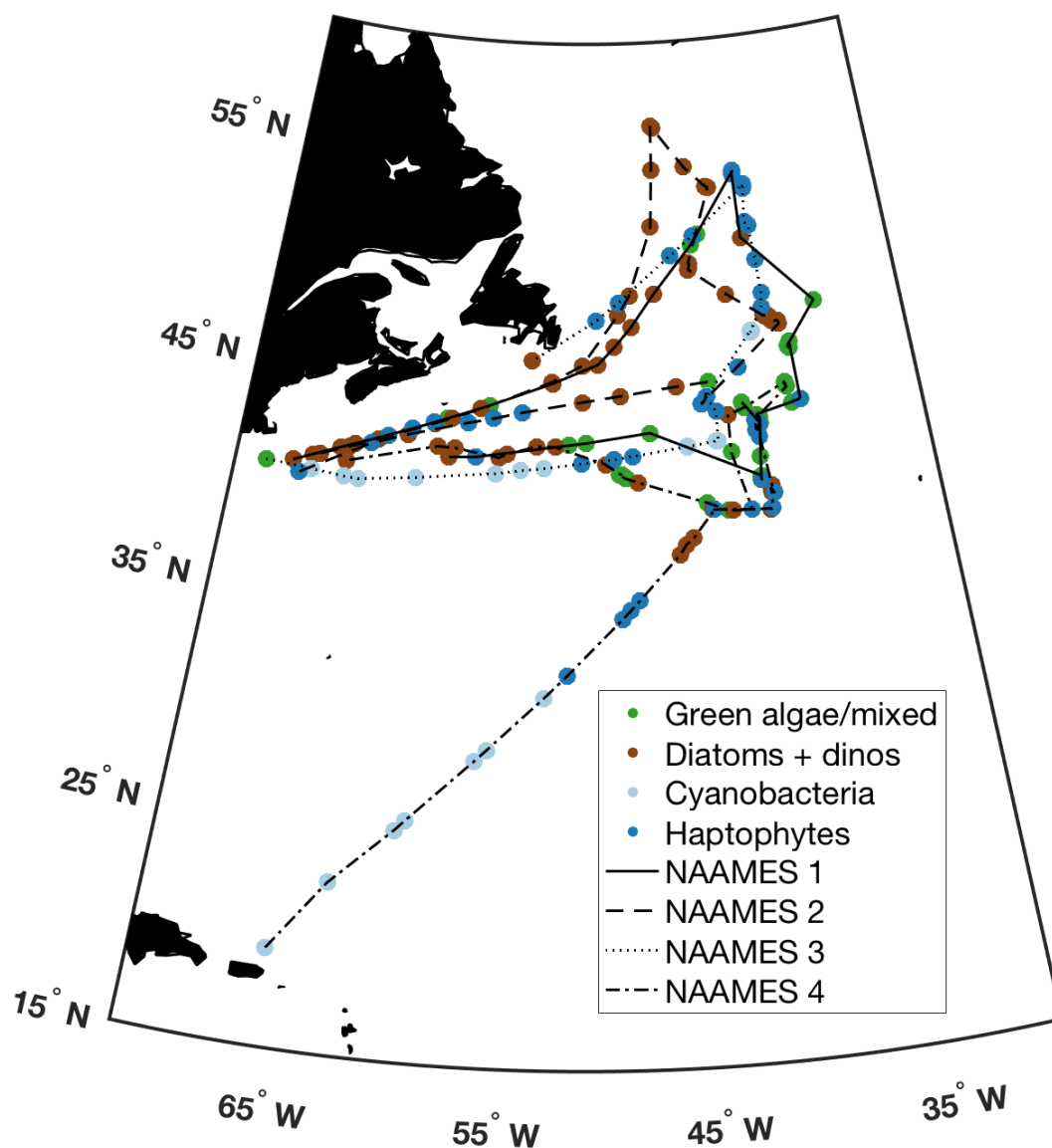**Supplementary Figure 1.** Pearson's correlation coefficient between all pigments: absolute concentration (upper right portion), normalized to total chlorophyll-*a* concentration (lower left portion), and with total chlorophyll-*a* (top row). Warm colors indicate positive correlation, cool colors indicate negative correlation.

Supplementary Figure 2. Spatial distribution of surface samples on NAAMES colored by the cluster to which that sample was assigned (light blue = cyanobacteria, dark blue = haptophytes, green = green algae/mixed, brown = diatoms, gold = dinoflagellates).
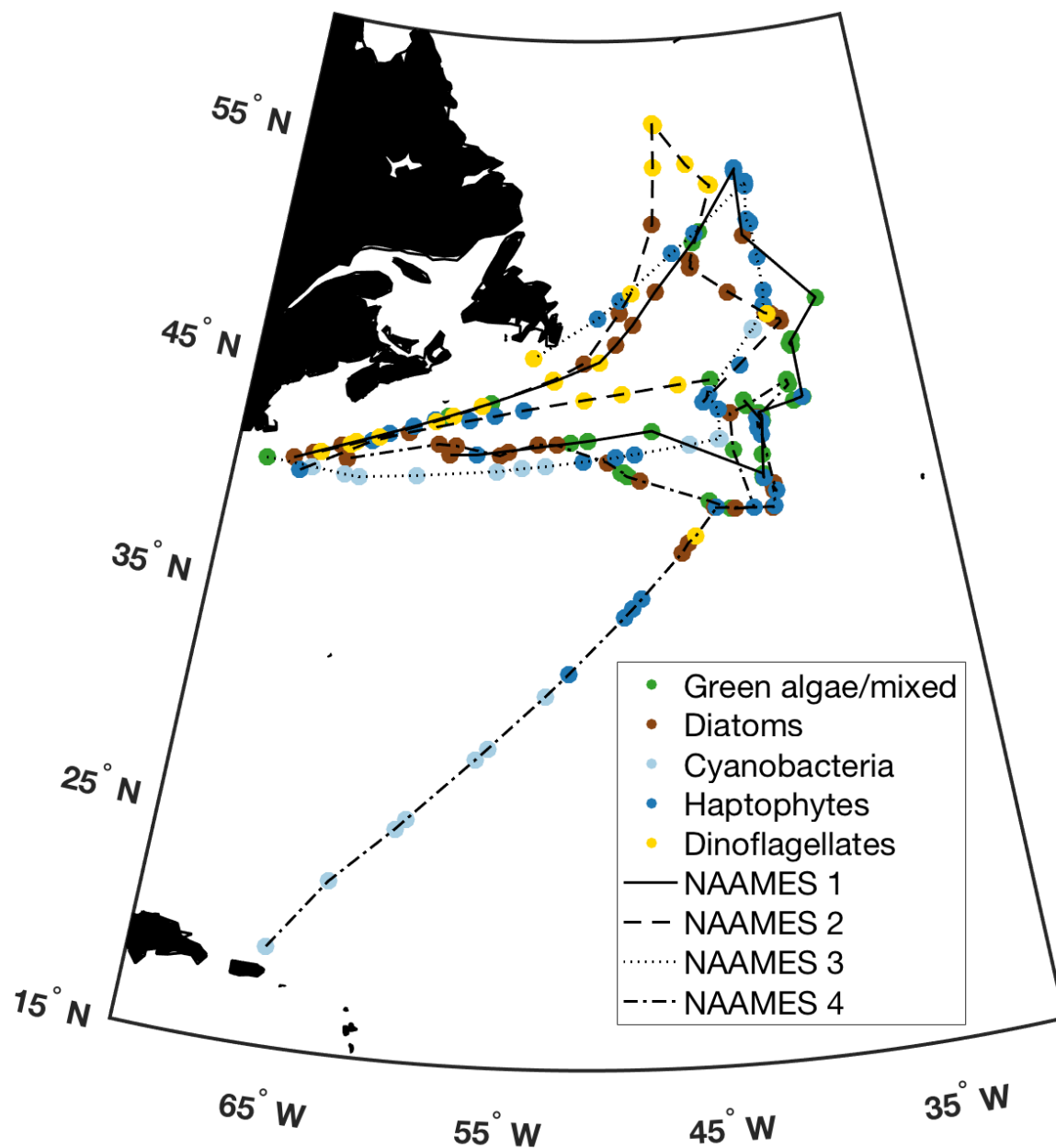
Supplementary Figure 3. Spatial distribution of amplitude functions for EOF Modes (A) 1, (B) 2, (C) 3, and (D) 4, calculated for phytoplankton pigment ratios to total chlorophyll-*a* concentration. Amplitude function magnitude is indicated as positive (red) or negative (blue) for each sample on NAAMES 1 (solid line), NAAMES 2 (dashed line), NAAMES 3 (dotted line), and NAAMES 4 (dash-dot line).

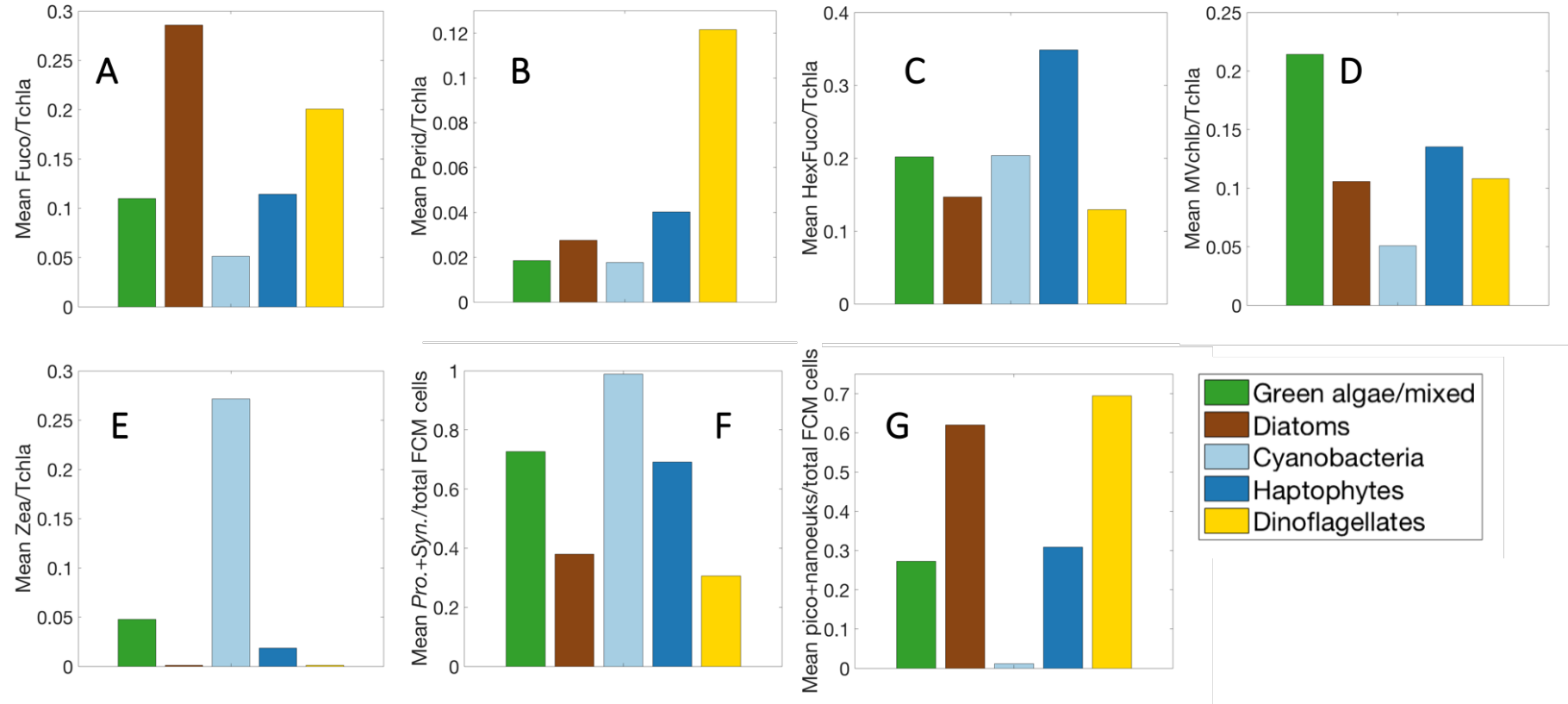**Supplementary Figure 4.** Spatial results of network-based community detection on NAAMES 1 (solid line), NAAMES 2 (dashed line), NAAMES 3 (dotted line), and NAAMES 4 (dash-dot line). Samples are colored based on the dominant community determined from the community detection analysis: light blue (cyanobacteria), dark blue (haptophytes), green (green algae), brown (diatoms and dinoflagellates).

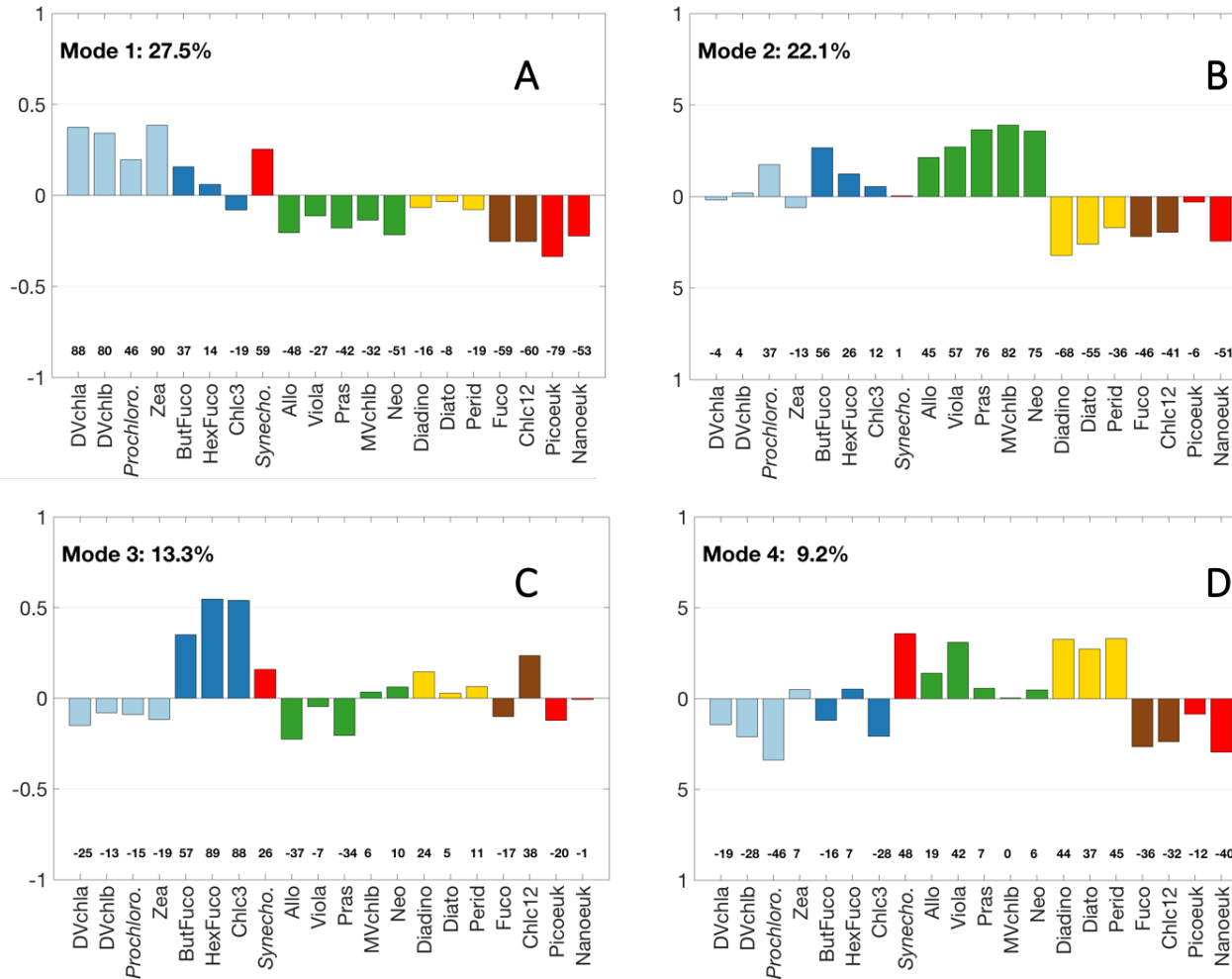**Supplementary Figure 5.** Spatial distribution of all five communities identified using network-based community detection and EOF regression on NAAMES 1 (solid line), NAAMES 2 (dashed line), NAAMES 3 (dotted line), and NAAMES 4 (dash-dot line). Samples are colored by the dominant community (light blue = cyanobacteria, dark blue = haptophytes, green = green algae/mixed, brown = diatoms, gold = dinoflagellates).

Supplementary Figure 6. Mean pigment ratios to total chlorophyll-*a* for five biomarker pigments: (A) fucoxanthin, (B) peridinin, (C) 19'hexanoyloxyfucoxanthin, (D) mono-vinyl chlorophyll b, (E) zeaxanthin and (F) *Prochlorococcus + Synechococcus* and (G) pico- and nanoeukaryote fractions of total cells measured by FCM for each community detected in the merged EOF + network-based community detection analysis (light blue = cyanobacteria, dark blue = haptophytes, green = green algae/mixed, brown = diatoms, gold = dinoflagellates).

Supplementary Figure 7. Empirical orthogonal functions for (A) Modes 1, (B) 2, (C) 3, and (D) 4, calculated for phytoplankton pigment ratios to total chlorophyll-*a* concentration and flow cytometry group cell counts to total cell counts. Loadings are colored based on pigment clusters (Fig. 2): light blue (cyanobacteria + *Prochlorococcus*), dark blue (haptophytes), green (green algae), brown (diatoms and dinoflagellates), red (other flow cytometry groups).

	Dvchla	DVchlb	Zea	Diadino	Diato	Perid	Fuco	Chlc12	ButFuco	HexFuco	Chlc3	Allo	MVchlb	Neo	Pras	Viola
Mode 1	-72	-58	-80	-37	-32	-5	20	26	12	8	29	67	75	85	80	42
Mode 2	-60	-62	-50	56	37	41	72	71	-62	-19	17	-5	-48	-30	-38	-40
Mode 3	-9	1	-10	12	-8	-1	-22	34	63	90	88	-44	1	2	-31	-13
Mode 4	-15	-22	11	65	62	37	-48	-19	5	18	-19	2	12	15	2	58

Supplementary Table 1. Pearson's correlation coefficient (R) values between each pigment ratio to Tchla for the 16 pigments used in this analysis and the first four EOF modes.

Community	Hierarchical clustering	Network-based community detection
Diatoms	58	64
Dinoflagellates	30	24
Haptophytes	73	72
Green algae	39	41
Cyanobacteria	29	28

Supplementary Table 2. Number of samples in each community identified by hierarchical clustering and network-based community detection analysis.

Parameter	*Prochlorococcus*	*Synechococcus*	Picoeukaryotes	Nanoeukaryotes
Fuco:Tchla	$R^2 = 0.09$ $p \ll 0.001$	$R^2 = 0.17$ $p \ll 0.001$	$R^2 = 0.24$ $p \ll 0.001$	$R^2 = 0.25$ $p \ll 0.001$
Perid:Tchla	$R^2 = 0.09$ $p \ll 0.001$	$R^2 = 0.002$ $p = 0.56$	$R^2 = 0.04$ $p = 0.01$	$R^2 = 0.02$ $p = 0.13$
HexFuco:Tchla	$R^2 \ll 0.001$ $p = 0.70$	$R^2 = 0.08$ $p \ll 0.001$	$R^2 = 0.05$ $p = 0.003$	$R^2 = 0.05$ $p = 0.003$
MVchlb:Tchla	$R^2 = 0.04$ $p = 0.01$	$R^2 = 0.06$ $p = 0.002$	$R^2 = 0.04$ $p = 0.008$	$R^2 = 0.03$ $p = 0.03$
Zea:Tchla	$R^2 = 0.11$ $p \ll 0.001$	$R^2 = 0.20$ $p \ll 0.001$	$R^2 = 0.35$ $p \ll 0.001$	$R^2 = 0.13$ $p \ll 0.001$
DVchla:Tchla	$R^2 = 0.23$ $p \ll 0.001$	$R^2 = 0.06$ $p = 0.001$	$R^2 = 0.23$ $p \ll 0.001$	$R^2 = 0.10$ $p \ll 0.001$

Supplementary Table 3. Pearson's correlation coefficient (R^2) and significance (p) between each cell group enumerated with flow cytometry and six biomarker pigment ratios to Tchla. Red values are the highest for a given parameter; blue values are the lowest. Bold values indicate significant correlations ($p<0.05$).