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Supporting Information for

Drawdown of Atmospheric pCO_2 via Variable Particle Flux Stoichiometry in the Ocean Twilight Zone

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Introduction

This Supplementary Information contains POC:POP flux measurements used in Figure 1 (separate file), model description, model parameters, and model results. Model codes are archived in the Zenodo (https://doi.org/10.5281/zenodo.4960404) and model input/output are publicly archived in Dryad (https://doi.org/10.5061/dryad.70rxwdbx).

Text S1: Model Equations for Biogeochemical model MOPS

S1.1 Source-minus-sink terms in MOPS (see Kriest and Oschlies, 2015 for full details)

9 state variables are: PO_4, phytoplankton biomass in P (PHY), zooplankton biomass in P (ZOO), dissolved organic phosphorus (DOP), particulate organic phosphorus (POP), dissolved O_2, NO_3, DIC, and alkalinity (constant). Notations and parameters follow the original model description paper (Kriest & Oschlies, 2015) except for POP, which is denoted as "Detritus (DET)" in the original paper. In the original model, C:P for phytoplankton ($r_{C:P}^{PHY}$) and zooplankton ($r_{C:P}^{ZOO}$) are fixed at 117:1.

$$S(PO_4) = (-PP + \lambda_{ZOO} ZOO)\mathcal{H}_e(k) + S_{PO_4}^R + S_{PO_4}^D \tag{1}$$

$$S(PHY) = (PP - G - \lambda_{PHY} PHY)\mathcal{H}_e(k) - S_{PHY}^M \tag{2}$$

$$S(ZOO) = (\epsilon_{ZOO} G - \lambda_{ZOO} ZOO - \kappa_{ZOO} ZOO^2)\mathcal{H}_e(k) - S_{ZOO}^M \tag{3}$$

$$S(DOP) = \sigma_{DOP}[(1-\epsilon_{ZOO})G + \kappa_{ZOO} ZOO^2 + \lambda_{PHY} PHY]\mathcal{H}_e(k) + \\ S_{PHY}^M + S_{ZOO}^M - S_{DOP}^R - S_{DOP}^D \tag{4}$$

$$S(POP) = (1-\sigma_{DOP})[(1-\epsilon_{ZOO})G + \kappa_{ZOO} ZOO^2 + \lambda_{PHY} PHY]\mathcal{H}_e(k) - S_{POP}^R - S_{POP}^D + \frac{\partial wPOP^*}{\partial z} \\ POP^* = \max(0, POP - P_{min}) \tag{5}$$

$$S(O_2) = R_{-O_2:P}(PP - \lambda_{ZOO} ZOO)\mathcal{H}_e(k) - S_{O_2}^R \tag{6}$$

$$S(NO_3) = d(-PP + \lambda_{ZOO} ZOO)\mathcal{H}_e(k) + S_{NO_3}^{NFix} + S_{NO_3}^R - S_{NO_3}^D \tag{7}$$

$$S(DIC) = (-PP \cdot r_{C:P}^{PHY} + \lambda_{ZOO} ZOO \cdot r_{C:P}^{ZOO})\mathcal{H}_e(k) + S_{DIC}^R + S_{DIC}^D \tag{8}$$

S1.2. New source-minus-sink terms for organic carbon state variables

We added 4 new state variables related to organic carbon: phytoplankton biomass in C (PHY_C), zooplankton biomass in C (ZOO_C), dissolved organic carbon C (DOC), and particulate organic carbon (POC). Kinetic parameters such as rate constants (λ) for remineralization for various source-minus-sink terms of POC and DOC are identical to those of POP and DOP, respectively. Details on computing phytoplankton C:P ratio ($r_{C:P}^{PHY}$) and zooplankton C:P ratio ($r_{C:P}^{ZOO}$) are described in the main text.

$$S(PHY_C) = S(PHY) \cdot r_{C:P}^{PHY} \tag{9}$$

$$S(ZOO_C) = (\epsilon_{ZOO} G \cdot r_{C:P}^{PHY} - \lambda_{ZOO} ZOO \cdot r_{C:P}^{ZOO} - \kappa_{ZOO} ZOO^2 \cdot r_{C:P}^{ZOO})\mathcal{H}_e(k) - S_{ZOO}^M \cdot r_{C:P}^{ZOO} \tag{10}$$

$$S(DOC) = \sigma_{DOP}[(1 - \epsilon_{ZOO})G \cdot r_{C:P}^{PHY} + \kappa_{ZOO} ZOO^2 \cdot r_{C:P}^{ZOO} + \lambda_{PHY} PHY \cdot r_{C:P}^{PHY}]\mathcal{H}_e(k) + S_{PHY}^M \cdot r_{C:P}^{PHY} + S_{ZOO}^M \cdot r_{C:P}^{ZOO} - S_{DOC}^R - S_{DOC}^D \tag{11}$$

$$S(DET_C) = (1 - \sigma_{DOP})[(1 - \epsilon_{ZOO})G \cdot r_{C:P}^{PHY} + \kappa_{ZOO} ZOO^2 \cdot r_{C:P}^{ZOO} + \lambda_{PHY} PHY \cdot r_{C:P}^{PHY}]\mathcal{H}_e(k) - S_{POC}^R - S_{POC}^D + \frac{\partial wPOC^*}{\partial z}$$

$$POC^* = \max(0, POC^* - P_{min} \times 117) \tag{12}$$

$$r_{C:P}^{PHY} = \frac{1}{[P:C]_{PHY}} = 1/[P:C]_{PHY,ref} \cdot \left(\frac{[PO_4]}{[PO_4]_0}\right)^{s_{PO_4}^{P:C}} \cdot \left(\frac{T}{T_0}\right)^{s_T^{P:C}} \tag{13}$$

$$r_{C:P}^{ZOO} = \frac{1}{[P:C]_{ZOO}} = 1/[P:C]_{zoo,ref}{}^{1-H}[P:C]_{PHY}{}^{H} \tag{14}$$

S1.3. Release of excess C or P by zooplankton for maintaining homeostatic C:P

When phytoplankton C:P does not equal C:P of zooplankton, zooplankton needs to release extra C or P from uptake into the particulate organic matter pool for zooplankton to maintain their homeostatic C:P. When phytoplankton $r_{C:P}^{PHY}$ is greater than $r_{C:P}^{ZOO}$ (i.e., excess C), extra C is added back to POC. The flux of excess C released E_{ZOO}^C is,

$$E_{ZOO}^C = \epsilon_{ZOO} G \times \max(0, r_{C:P}^{PHY} - r_{C:P}^{ZOO}) \tag{15}$$

The updated SMS terms for zooplankton carbon biomass and POC are:

$$S(ZOO_C)^* = S(ZOO_C) - E_{ZOO}^C$$

$$S(ZOO_C)^* = \text{Updated SMS of ZOO}_\text{C} \text{ after zooplankton C release} \tag{16}$$

$$S(POC)^* = S(POC) + E_{ZOO}^C$$

$$S(POC)^* = \text{Updated SMS of POC after zooplankton C release} \tag{17}$$

Conversely, if phytoplankton $r_{C:P}^{PHY}$ is less than $r_{C:P}^{ZOO}$ (i.e., excess P), zooplankton releases its extra P back to POP. The flux of extra P (E_{ZOO}^P):

$$E_{ZOO}^{P} = \epsilon_{ZOO} G \times \max\left(0, 1 - \frac{r_{C:P}^{PHY}}{r_{C:P}^{ZOO}}\right) \tag{18}$$

The updated SMS terms for zooplankton biomass in P and POP are:

$$S(ZOO)^{*} = S(ZOO) - E_{ZOO}^{P}$$

$$S(ZOO)^{*} = \text{Updated SMS of ZOO after zooplankton P release} \tag{19}$$

$$S(POP)^{*} = S(POP) + E_{ZOO}^{P}$$

$$S(POP)^{*} = \text{Updated SMS of POP ater zooplankton P release} \tag{20}$$

With this flux adjustment, $S(ZOO_C)^{*} = S(ZOO)^{*} \cdot r_{C:P}^{ZOO}$ and it ensures the total mass of C and P in the system are conserved.

In our sensitivity runs, we set a hard-bound minimum on C:P of phytoplankton so that $r_{C:P}^{PHY}$ is always greater than or equal to $r_{C:P}^{ZOO}$. This way, zooplankton do not excrete excess P into POP, which keeps phosphate distribution identical in each sensitivity run.

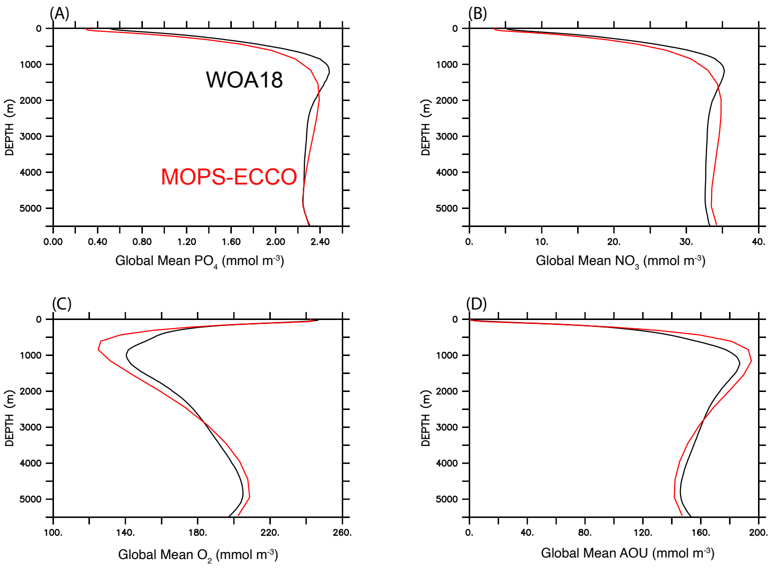

Figure S1. Global mean vertical profiles of phosphate (a), nitrate (b), oxygen (c), and AOU (d). Red lines are model outputs after 3000 years of spin-up, and black lines are observations from World Ocean Atlas 2018 (Garcia et al., 2018).

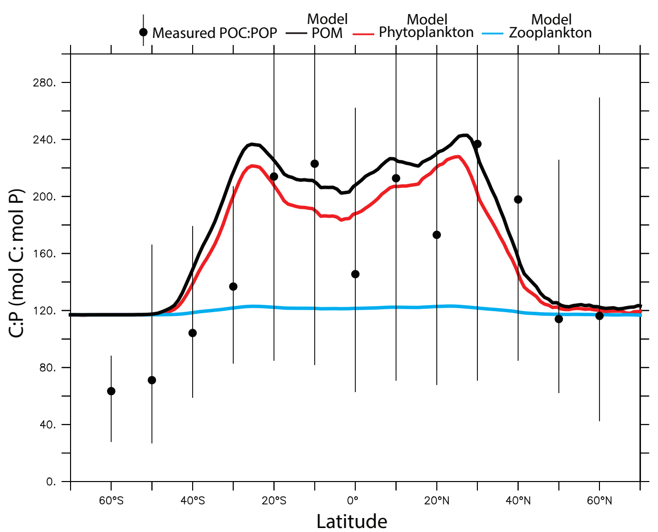

Figure S2. Zonal mean C:P of model suspended POM (black), phytoplankton (red), and zooplankton (blue) in the surface ocean after 3000 years of spin-up. Black boxes show median and the 95% confidence interval of observed suspended C:P of POM binned by latitude (Martiny et al., 2013).

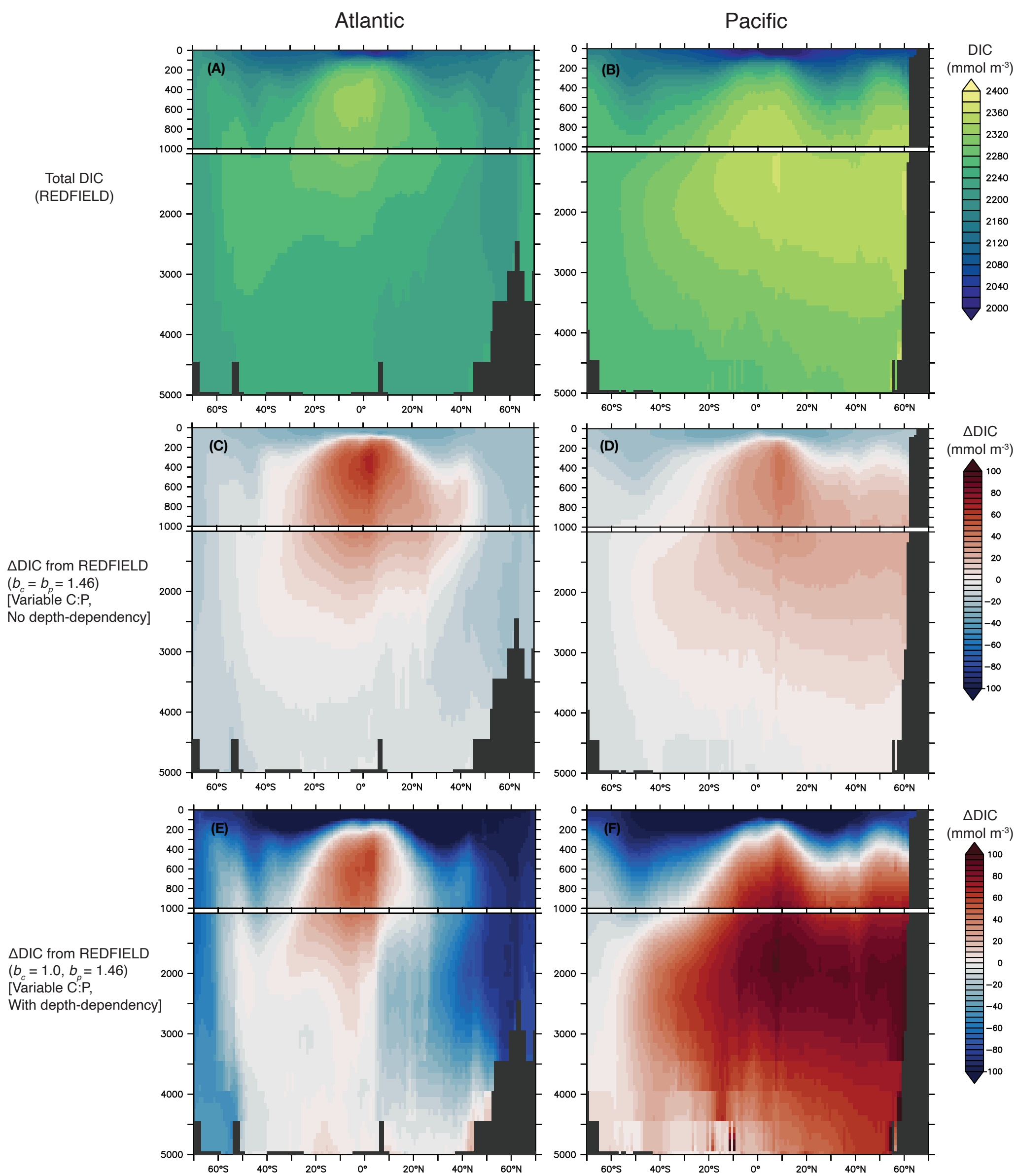

Figure S3. Zonal-averaged DIC under the Redfield run with fixed C:P, and no preferential remineralization (a-b), DIC anomaly (ΔDIC) from the Redfield run in the control run without preferential remineralization (c-d), and in a run with preferential remineralization of POP over POC (e-f).

Table S1. List of POC:POP flux measurements. Abbreviations: TA = Tropic Atlantic, TP = Tropical Pacific, STNA = Subtropical North Atlantic, STNP = Subtropical North Pacific, SNP = Subpolar North Pacific.

Table S2. Median C:P of suspended POM and sinking POM from different oceanographic regions and with different sediment trap types delineated by depth. Brackets indicate the number of samples.

Regions	**C:P of Suspended POM*** **(top 300 m)**	**C:P Flux Ratio**		
		Twilight Zone (100 - 1000 m)	Deep (> 1000 m)	All
Tropical Atlantic	-	334 (12)	-	334 (12)
Tropical Pacific	124	318 (1)	267 (11)	274 (12)
Subtropical North Atlantic	200	279 (9)	191 (4)	274 (13)
Subtropical North Pacific	154	254 (11)	243 (3)	248 (14)
Subpolar North Pacific	94	142 (3)		142 (3)
Global	146	294 (36)	243 (18)	273 (54)
Sediment Trap Types				
Moored	N/A	274 (7)	243 (18)	265 (25)
Surface tethered		316 (26)	-	316 (26)
Free-drifting		142 (3)	-	142 (3)

* Median C:P of suspended POM from Martiny et al. (2014)

Table S3. Summary of analysis of variance (ANOVA). Log-transformed C:P flux ratio was analyzed on a two-factorial ANOVA, using region and depth (twilight zone or deep) as independent factors and C:P flux ratios as the dependent variable. Asterisk (*) signifies statistical significance ($p < 0.05$).

Factor	MS	df	F	p
Region	0.4505	4	7.633	8.37e-5*
Depth	0.3683	1	6.249	0.0161*
Region x Depth	0.1289	2	2.184	0.1241
Residuals	0.0590	46		

Table S4. Summary of BATS timeseries analysis.

Martin curve parameter	**n**	**Median**	**t-test**
b_P (PP)	106	1.28	p = 0.0015 (p < 0.05)
b_C (POC)	111	0.98	

Table S5. Summary of BATS timeseries analysis. The key biogeochemical parameters, including the Martin *b* for POP, were objectively calibrated specifically for the ECCO transport matrix field to match observed PO_4, O_2, and NO_3 (Kriest et al., 2020; Figure S1). Phytoplankton power law parameter values for P:C ($\boldsymbol{s_{PO_4}^{P:C}}$ and $\boldsymbol{s_T^{P:C}}$) are weighted-mean values across all phytoplankton types based on meta-analysis of Tanioka & Matsumoto (2020) (their Table 2).

Parameter	Description	Unit	Value
Calibrated Parameters from Kriest et al. 2020 for MOPS + ECCO TMs			
b (= b_P)	Martin *b* for POP	-	1.46
$R_{-O_2:P}$	$-O_2:P$ of remineralization	mol: mol	151.1
μ_{NFix}	Maximum nitrogen fixation rate	d^{-1}	2.29
DIN_{min}	Threshold nitrate concentration for denitrification	mmol m^{-3}	16.0
K_{DIN}	Half-saturation constant for nitrate	mmol m^{-3}	23.1
K_{O_2}	Half-saturation constant for aerobic remineralization	mmol m^{-3}	1.07
Parameters for new organic carbon module			
b_C	Martin *b* for POC	-	Variable (Control = 1.46)
$[P:C]_{PHY,ref}$	Reference P:C molar ratio of phytoplankton	mol: mol	1/117
$[P:C]_{zoo,ref}$	Reference P:C molar ratio of zooplankton	mol: mol	1/117
$[PO_4]_0$	Reference PO_4	mmol m^{-3}	0.57
T_0	Reference T	◦K	291
$s_{PO_4}^{P:C}$	Sensitivity of phytoplankton P:C to PO_4	-	0.21
$s_T^{P:C}$	Sensitivity of phytoplankton P:C to T	-	-3.6
H	Homeostatic parameter of zooplankton	-	0.08

Table S6. Summary of sensitivity model run results with varying Martin *b* for POC (b_C). Martin *b* for POP (b_P) is constant at 1.46 in all runs. All sensitivity experiments are conducted for 1000 years following 3000 years of the spin-up run.

Run ID	Gas exchange	C:P	b_c	b_p	$r_{C:P}$ at 100 m (mol C:mol P)	$r_{C:P}$ (mol C:mol P)	OCS_{soft} (PgC)	$C_{buffered}$ (PgC)	Revelle Factor, R	$V_{OC} \cdot \overline{DIC_{eq}}$ (PgC)	pCO_{2a} (ppm)	POC export at 100 m (PgC yr^{-1})
201203a (Control)	Regular	Variable	1.46	1.46	151	159	2701	3251	12.7	33811	277.8	9.5
201204r	Fast	Variable	0.75	1.46	151	309	5255	3439	10.0	31580	130.2	15.7
201204h	Fast		1	1.46	151	219	3717	3344	11.3	32958	205.2	13.2
201204q	Fast		1.25	1.46	151	168	2858	3263	12.5	33685	267.2	11.3
201204d (Default)	Fast		1.46	1.46	151	142	2412	3220	13.2	34046	307.2	10.1
201204p	Fast		1.75	1.46	151	119	2019	3186	14.0	34352	347.2	8.8
201204j	Fast	Fixed (Redfield)	1.46	1.46	117	117	1990	3184	14.1	34374	351.2	7.1