

1 **Supplementary materials**

2 **Temporal and spatial variations in benthic nitrogen cycling in a temperate macro-tidal**
3 **coastal ecosystem: observation and modeling**

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23 **Table S1** Reaction network and rate formulation used in the model.

Description	Reaction formulation ^a	Rate expression ^{b,c}
Aerobic respiration	$OM + xO_2 + (2z-y)HCO_3^- \rightarrow yNH_4^+ + zHPO_4^{2-} + (x-y+2z)CO_2 + (x-y+2z)H_2O$	$R1 = k1 [OM] \cdot \frac{[O_2]}{[O_2] + kO} \cdot F_T$
Denitrification	$OM + 0.8xNO_3^- \rightarrow 0.4xN_2 + yNH_4^+ + 0.8xNO_2^- + zHPO_4^{2-} + (0.8x+y-2z)HCO_3^- + (0.2x-y+2z)CO_2 + (0.6x-y+2z)H_2O$	$R2 = k2 [OM] \cdot \frac{[NO_3^-]}{[NO_3^-] + kNO} \cdot \frac{kinO}{[O_2] + kinO} \cdot 1 - F_{DNRA} \cdot F_T$
DNRA	$OM + 0.5xNO_3^- \rightarrow (0.5x+y)NH_4^+ + (0.5xNO_2^-) + zHPO_4^{2-} + (y-2z)HCO_3^- + (x-y+2z)CO_2 + (0.5x-y+2z)H_2O$	$R3 = k2 [OM] \cdot \frac{[NO_3^-]}{[NO_3^-] + kNO} \cdot \frac{kinO}{[O_2] + kinO} \cdot F_{DNRA} \cdot F_T$
Nitrification step 1	$NH_4^+ + 1.5O_2 \rightarrow NO_2^- + H_2O$	$R4 = k_{nit1} \cdot \frac{[NH_4^+]}{[NH_4^+] + kmNH4ao} \cdot \frac{[O_2]}{[O_2] + kmO2ao} \cdot F_T$
Nitrification step 2	$NO_2^- + 0.5O_2 \rightarrow NO_3^-$	$R5 = k_{nit2} \cdot \frac{[NO_2^-]}{[NO_2^-] + khNIT} \cdot \frac{[O_2]}{[O_2] + kHO} \cdot F_T$
Anammox	$NH_4^+ + NO_2^- \rightarrow N_2 + 2H_2O$	$R6 = k_{anx} \cdot \frac{[NH_4^+]}{[NH_4^+] + khNH4} \cdot \frac{[NO_2^-]}{[NO_2^-] + khNO} \cdot \frac{kinO}{[O_2] + kinO} \cdot F_T$

24 ^a $OM = x(CH_2O)_{org} + y(NH_3)_{org} + z(H_3PO_4)_{org}$, where x, y, z represent the CNP ratios,

25 ^b F_{DNRA} = fraction of the total nitrate reduction occurring via DNRA (5%),

26 ^c F_T = temperature correction for the rates. $F_T = \frac{K_1 \cdot \exp(\gamma_1 \cdot (T - T_1))}{1 + K_1 \cdot \exp(\gamma_1 \cdot (T - T_1) - K_1)} \cdot \frac{K_2 \cdot \exp(\gamma_2 \cdot (T_2 - T))}{1 + K_2 \cdot \exp(\gamma_2 \cdot (T_2 - T) - K_2)}$, where T_1 and T_2 are the lower and upper temperature for the OM decay, respectively;

27 K_1 and K_2 are the coefficient rates for the lower and upper temperature, respectively; γ_1 and γ_2 are the temperature rate multipliers for T_1 and T_2 , respectively (see Cole and
28 Wells, 2006 for details).

29 **Table S2** Boundary conditions used in the model.

Parameters	Apr 15 th	Apr 22 nd	Jun 16 th	Jun 24 th	Aug 5 th	Sep 8 th	St. A	St. B	St. C	St. D	Unit	Sources
2015 (Nord Dumet monitoring station/St. A)							2016					
<i>Solutes</i>												
O ₂	142 (84%)	161 (95%)	142 (82%)	120 (75%)	118 (93%)	114 (101%)	198 (80%)	96 (37%)	170 (70%)	198 (79%)	μM	I
NO ₃ ⁻	17.7	13.1	1.60	1.30	0.07	0.04	0.37	6.82	0.20	0.47	μM	I
NO ₂ ⁻	0.27	0.26	0.12	0.17	0.03	0.03	0.04	0.73	0.00	0.06	μM	I
NH ₄ ⁺	1.90	3.30	1.03	1.23	0.21	0.09	0.80	4.40	1.80	1.61	μM	I
Fe ²⁺	0	0	0	0	0	0	0	0	0	0	μM	I
SO ₄ ²⁻ ^a	28	28	28	28	28	28	28	28	28	28	mM	M
<i>Solids</i>												
OM1	600	600	700	750	275	375	1,500	1,800	1,000	400	μmol C cm ⁻² yr ⁻¹	M
OM2	150	150	170	180	120	120	300	300	300	150	μmol C cm ⁻² yr ⁻¹	M
Fe(OH) ₃	75	75	75	75	75	75	75	75	75	75	μmol cm ⁻² yr ⁻¹	M

30 Solute concentrations were obtained from the measurements. Solid fluxes were constrained from the model fitting (see Methods). ^a The SO₄²⁻ concentration was constrained
31 from the measured salinity. Sources: **M**, constrained by the model fitting; **I**, independently determined from the field data.

32 **Table S3** Reaction parameter values used in the model.

Parameters	Apr 15 th	Apr 22 nd	Jun 16 th	Jun 24 th	Aug 5 th	Sep 8 th	St. A	St. B	St. C	St. D	Unit	Source	Ref.	Description
2015 (Nord Dumet monitoring station/St. A)							2016							
<i>Fixed</i>														
C:N OM1	10	10	10	10	10	10	10	10	10	10		M		C:N ratio for OM1
C:N OM2	15	15	15	15	15	15	15	15	15	15		M		C:N ratio for OM2
k _O	5	5	5	5	5	5	5	5	5	5	μM	L	2, 4, 5	Limitation of O ₂ for aerobic oxidation
k _{NO}	5	5	5	5	5	5	5	5	5	5	μM	L	2, 4, 5	Limitation of NO ₃ ⁻ for denitrification
k _{mNH4ao}	5	5	5	5	5	5	5	5	5	5	μM	L	1	Limitation of NH ₄ ⁺ for nitrification step 1
k _{mO2ao}	5	5	5	5	5	5	5	5	5	5	μM	L	1	Limitation of O ₂ for nitrification step 1
k _{hNT}	1	1	1	1	1	1	1	1	1	1	μM	L	1	Limitation of NO ₂ ⁻ for nitrification step 2
k _{HO}	5	5	5	5	5	5	5	5	5	5	μM	L	1	Limitation of O ₂ for nitrification step 2
k _{hno}	1	1	1	1	1	1	1	1	1	1	μM	L	3	Limitation of NO ₂ ⁻ for anammox
k _{hnh}	1	1	1	1	1	1	1	1	1	1	μM	L	3	Limitation of NH ₄ ⁺ for anammox
k _{inO}	6	6	6	6	6	6	6	6	6	6	μM	L	6	Inhibition of O ₂ for anoxic processes
k ₁ OM1	16	16	16	16	16	16	16	16	16	16	yr ⁻¹	M		Rate constant for aerobic oxidation of OM1
k ₁ OM2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	yr ⁻¹	M		Rate constant for aerobic oxidation of OM2
k ₂ OM1	5	5	5	5	5	5	5	5	5	5	yr ⁻¹	M		Rate constant for denitrification and DNRA of OM1
k ₂ OM2	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1	yr ⁻¹	M		Rate constant for denitrification and DNRA of OM2
<i>Adjusted</i>														
k _{nit} 1	90	80	180	180	100	190	20	5	50	60	μmol cm ⁻³ yr ⁻¹	M		Maximum rate for nitrification step 1
k _{nit} 2	200	195	550	550	400	540	30	50	190	150	μmol cm ⁻³ yr ⁻¹	M		Maximum rate for nitrification step 2
k _{anx}	0.03	0.05	0.02	0.02	0.04	0.02	0.02	0.02	0.02	0.02	μmol cm ⁻³ yr ⁻¹	M		Maximum rate for anammox
<i>Measured</i>														
∅	0.83	0.84	0.88	0.85	0.85	0.82	0.87	0.92	0.59	0.40	cm ³ cm ⁻³	I		Sediment porosity
ρ	2.89	2.49	3.26	2.74	3.08	3.06	2.71	4.45	2.50	1.93	g cm ⁻³	I		Sediment density
T°	11.1	11.9	14.4	14.9	17.9	17.8	16.5	13.8	17.3	15.8	°C	I		Bottom water temperature

33 The sources of the parameter values are indicated by the following codes: **M**, constrained by the model fitting; **I**, independently determined from the field data, **L**, literature
34 value, with references given as follows: 1. Ward (1986); 2. Wang and VanCappellen (1996); 3. Dalsgaard and Thamdrup (2002); 4. Canavan et al. (2006); 5. Dale et al. (2016);
35 6. Akbarzadeh et al. (2018).

36 **Table S4** Model parameter values for the transport processes that were invariable within the
 37 season and sampling station.

Parameters	Value	Description	Unit	Sources
Db	3	Bioturbation coefficient at $x < 2$ cm	$\text{cm}^2 \text{yr}^{-1}$	M
	1.5	Bioturbation coefficient at $2 \text{ cm} \leq x \leq 4$ cm	$\text{cm}^2 \text{yr}^{-1}$	M
	0.3	Bioturbation coefficient at $4 \text{ cm} \leq x \leq 7$ cm	$\text{cm}^2 \text{yr}^{-1}$	M
	$0.1 * (1 - \exp(x - 20))$	Bioturbation coefficient at $x > 7$ cm	$\text{cm}^2 \text{yr}^{-1}$	M
α	3	Bioirrigation coefficient at $x < 2$ cm	yr^{-1}	M
	1.5	Bioirrigation coefficient at $2 \text{ cm} \leq x \leq 4$ cm	yr^{-1}	M
	0.3	Bioirrigation coefficient at $4 \text{ cm} \leq x \leq 7$ cm	yr^{-1}	M
	$0.1 * (1 - \exp(x - 20))$	Bioirrigation coefficient at $x > 7$ cm	yr^{-1}	M
ω	0.5	Burial velocity	cm yr^{-1}	M

38 See Table S2 for the source indications
 39

40 **Characteristic time-scales for the sediment processes**

41 *1. Diffusion time-scale*

42 Diffusion time-scales calculated from the modified Einstein-Smoluchowski equation (Jørgensen
43 and Revsbech, 1985):

44 $t_{DIFF} = z^2/2D_s$ eq. 1

45 where t_{DIFF} is the diffusion time-scales (d), z is the characteristic length (cm), and D_s is the
46 diffusion coefficient ($\text{cm}^2 \text{s}^{-1}$), corrected with the bottom water temperature and sediment
47 porosity for each measurement. The diffusion time-scale was calculated for the upper 2 cm
48 layer, where the exchange between the sediment and overlying water occurs most actively.

49 **Table S5** Diffusion time-scales for NH_4^+ , NO_3^- and DON over the first 2 cm sediment layer.

Date	$D_s \text{ NH}_4^+$	$D_s \text{ NO}_3^-$	$D_s \text{ DON}^*$	$t_{DIFF} \text{ NH}_4^+$	$t_{DIFF} \text{ NO}_3^-$	$t_{DIFF} \text{ DON}$
	(x $10^{-6} \text{ cm}^2 \text{ s}^{-1}$)			(d)		
April 15 th .	1.4	1.4	0.17	1.6	1.7	13.2
April 22 nd	1.4	1.4	0.18	1.6	1.6	13.2
June 16 th	1.5	1.5	0.19	1.5	1.5	12.4
June 24 th	1.6	1.5	0.19	1.5	1.5	12.5
August 5 th	1.7	1.6	0.21	1.4	1.4	11.3
September 8 th	1.7	1.6	0.21	1.4	1.4	11.1

50 t_{DIFF} = the diffusion time-scale; D_s = the diffusion coefficient. *The D_s of DON was estimated using the empirical
51 relationship between the free solution diffusion coefficient (D_o) and the molecular weight (MW) for the various
52 organic compounds at 25°C in distilled water reported by Burdige et al. (1992), assuming a fixed average MW of
53 2500 Daltons. The obtained values were then corrected for in situ temperature using the Stoke-Einstein equation
54 and translated to D_s after correction for sediment porosity (Boudreau, 1997).

55 *2. Residence time of the solutes*

56 The residence time of the solutes at steady-state can be calculated by dividing the stock of the
57 solutes by the flux of these solutes.

58 $t_{RES} = Stock/DifFlux$ eq. 2

59 where t_r is the residence time of the solutes (d), *Stock* is the average stock of the solutes over the
60 first 2 cm sediment layer ($\mu\text{mol m}^{-2}$), and *DifFlux* is the diffusive fluxes of the solutes
61 ($\mu\text{mol m}^{-2} \text{ d}^{-1}$).

62 **Table S6** Residence time of NH_4^+ , NO_3^- and DON over the first 2 cm sediment layer.

Date	Stock			DifFlux			t_{RES}		
	NH_4^+	NO_3^-	DON	NH_4^+	NO_3^-	DON	NH_4^+	NO_3^-	DON
	($\mu\text{mol m}^{-2}$)			($\mu\text{mol m}^{-2} \text{ d}^{-1}$)			(d)		
April 15 th .	3,138	376	7,473	427	-247	148	7.4	1.5	50
April 22 nd	3,699	294	8,156	541	-99	157	6.8	3.0	52
June 16 th	3,487	130	6,471	518	17	175	6.7	7.5	37
June 24 th	2,858	198	9,469	439	44	225	6.5	4.5	42
August 5 th	3,384	84	6,322	589	21	193	5.7	4.1	33
September 8 th	3,795	113	6,916	567	22	212	6.7	5.2	33

63 * Diffusive fluxes calculated from the concentration gradient at the SWI using Fick's first law of diffusion
64 (Boudreau, 1997; Schulz, 2006)

65 **Sensitivity analysis**

66 The sensitivity analysis was carried out by imposing changes in the model boundary
 67 condition (Table S3) and parameter values (Table S2 & S4) for each sampling date. The
 68 contribution of NO_2^- to the DIN fluxes was considered negligible (< 5% of the NH_4^+ and NO_3^-
 69 fluxes) and therefore not included in the sensitivity analysis. A stepwise approach was applied
 70 by manually changing the parameter values individually (Table S7) and observing the model
 71 response on the NH_4^+ and NO_3^- fluxes. The model was run by imposing each change in these
 72 values one by one for each sampling date. A combination effect between the parameters was not
 73 tested in the sensitivity analysis in the present study. The response was calculated as a
 74 percentage of change in the NH_4^+ and NO_3^- fluxes with regard to best fits (baseline model).

75 **Table S7** List of tested parameters for the sensitivity analysis.

Parameters	Description
Anoxic	Bottom water O_2 concentrations are equal to zero
BW $\text{NO}_3^- = 10\text{x}$ (100x)	Increase in the bottom water NO_3^- concentrations by 10 fold (or 100 fold for August and September)
OM1 2x (3x)	Increase in the deposition of OM1 by 2 fold (or 3x for August)
OM1 <=> OM2	Inversing the proportion of OM1 and OM2
k_1 OM1 = 2x	Increase in the rate constant for the aerobic oxidation of OM1 by 2 fold
k_1 OM1 = 1/2 x	Decrease in the rate constant for the aerobic oxidation of OM1 by one-half
C/N OM1 = 106/16	Imposed change in the C/N ratio compared to that in living phytoplankton (i.e., Redfield ratio)
Db = 10x	Increase in the bioturbation coefficient by 10 fold
$\alpha = 10\text{x}$	Increase in the bioirrigation coefficient by 10 fold

76 **Table S8** Density of macrofauna in the incubated sediment cores of the temporal study carried
 77 out at the ND monitoring station in 2015.

Species names	Apr 15 th	Apr 22 nd	Jun 16 th	Jun 24 th	Aug 5 th	Sep 8 th	Total species/m ²
Crustacea							
<i>Asthenognathus atlanticus</i>	1	2					78
<i>Philocheras bispinosus bispinosus</i>			1				26
Echinoderms							
<i>Acrocnida brachiata</i>	1				1		52
<i>Amphiura filiformis</i>	6	6	4	22		6	1,146
<i>Labidoplax digitata</i>					2		52
<i>Leptopentacta elongata</i>			1	2	2		130
<i>Synaptidae</i>							
Molluscs							
<i>Abra nitida</i>			1		2	1	104
<i>Corbula gibba</i>		1					26
<i>Kurtiella bidentata</i>	5		1	3		4	339
<i>Nassarius pygmaeus</i>					1	1	52
<i>Nucula nitidosa</i>	27	27	15	24	3	5	2,631
<i>Philina aperta</i>				1	2		78
<i>Spisula solida</i>	1						26
<i>Turritella communis</i>				1	1		52
Annelids							
<i>Aphelochaeta</i>						1	26
<i>Chaetozone</i>						1	26
<i>Glycera unicornis</i>	1					1	52
<i>Heteromastus filiformis</i>		2				6	208
<i>Labioleanira yhleni</i>	1	4	2	2	1		260
<i>Magelona</i>			1		4		130
Maldanidae		1	1	4	1		182
<i>Malmgrenia lilianae</i>	3	1		3			182
<i>Nephtys</i>	1	1					52
<i>Pholoe baltica</i>			1	1			52
<i>Sternaspis scutata</i>	5	3	5	1	8		573
Cnidaria							
<i>Edwardsidae</i>					1		26
<i>Halcapa</i>							
<i>Virgularia</i>					1	4	130
Others							
<i>Nemertea</i>		2	1			1	104
<i>Phoronidien</i>		1				1	52

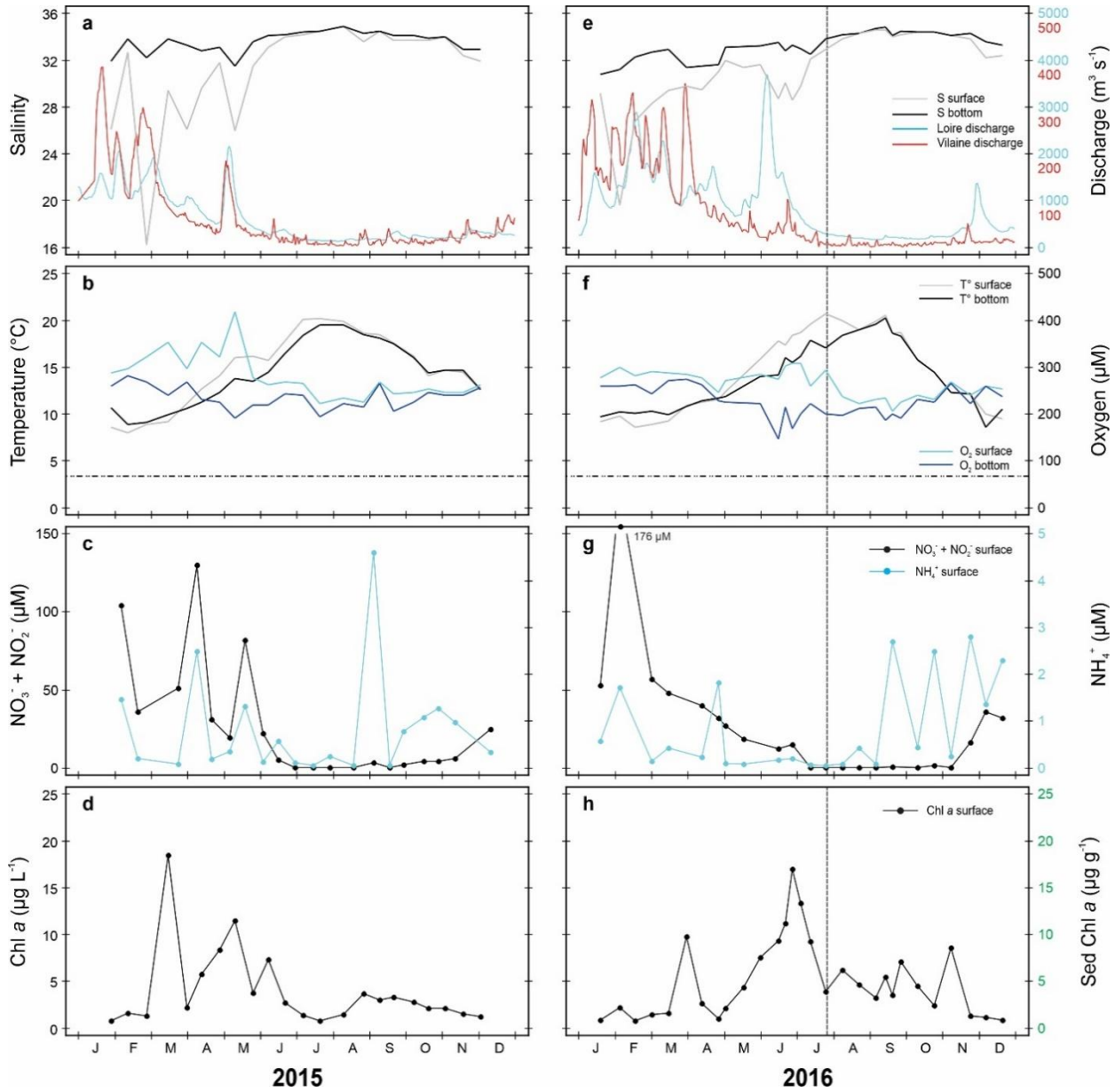
78 * Sediment core surface area = 64 cm²

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80 References

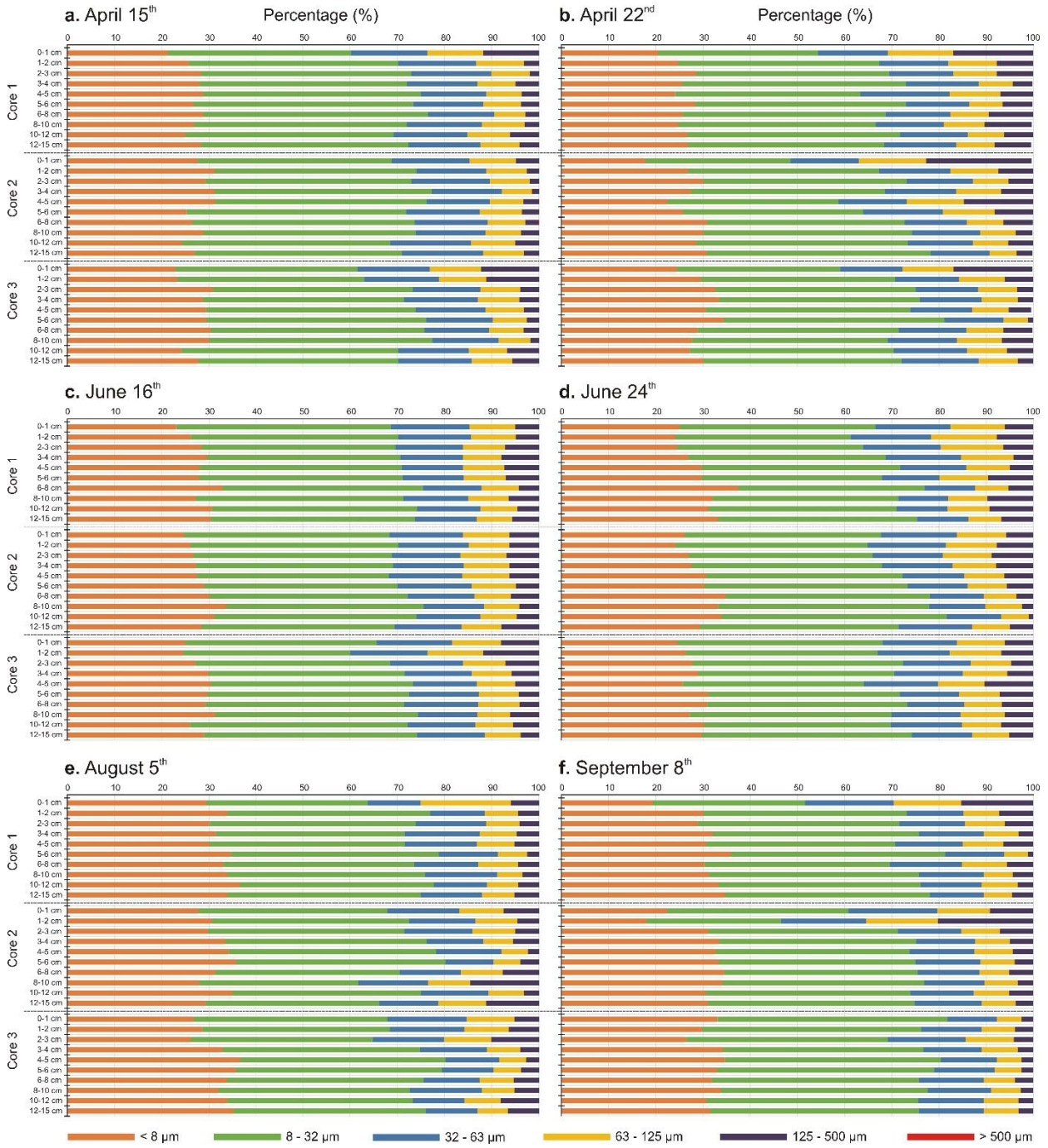
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115

116 **Fig. S1** Variations in the salinity and river discharge (Loire and Vilaine) (a, e), temperature and
 117 dissolved O₂ (b, f), NO₃⁻ + NO₂⁻ and NH₄⁺ (c, g), Chl *a* in the water column and sediment
 118 surface (d, h), at the Ouest Loscolo monitoring station in 2015 (left) and 2016 (right) from
 119 January to December. Dashed-dotted horizontal line (panels a & e): hypoxia threshold (63 μM;
 120 Middelburg and Levin, 2009; Zhang et al., 2010). Vertical lines: dates of the sediment
 121 investigations.



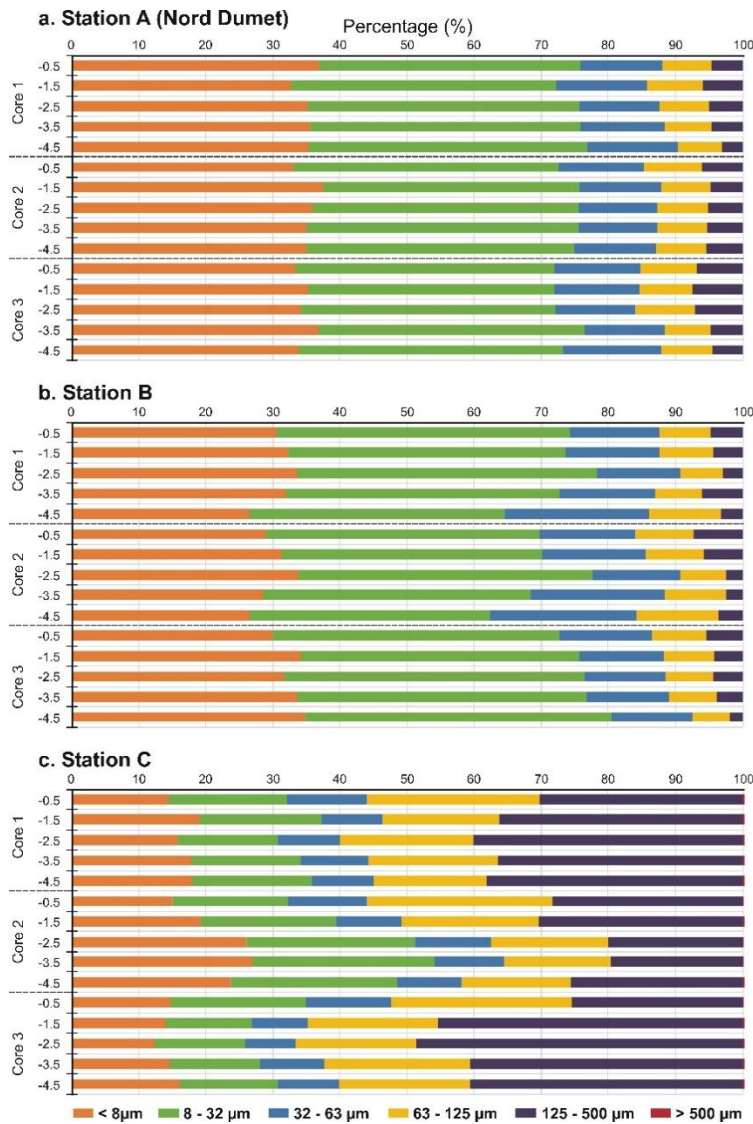
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124 **Fig. S2** Depth profile of the grain size distribution at the Nord Dumet monitoring station (St. A)
 125 in triplicate sediment cores for the temporal study from April to September 2015. The y-axis
 126 represents each section of the sediment layers in the triplicate cores.



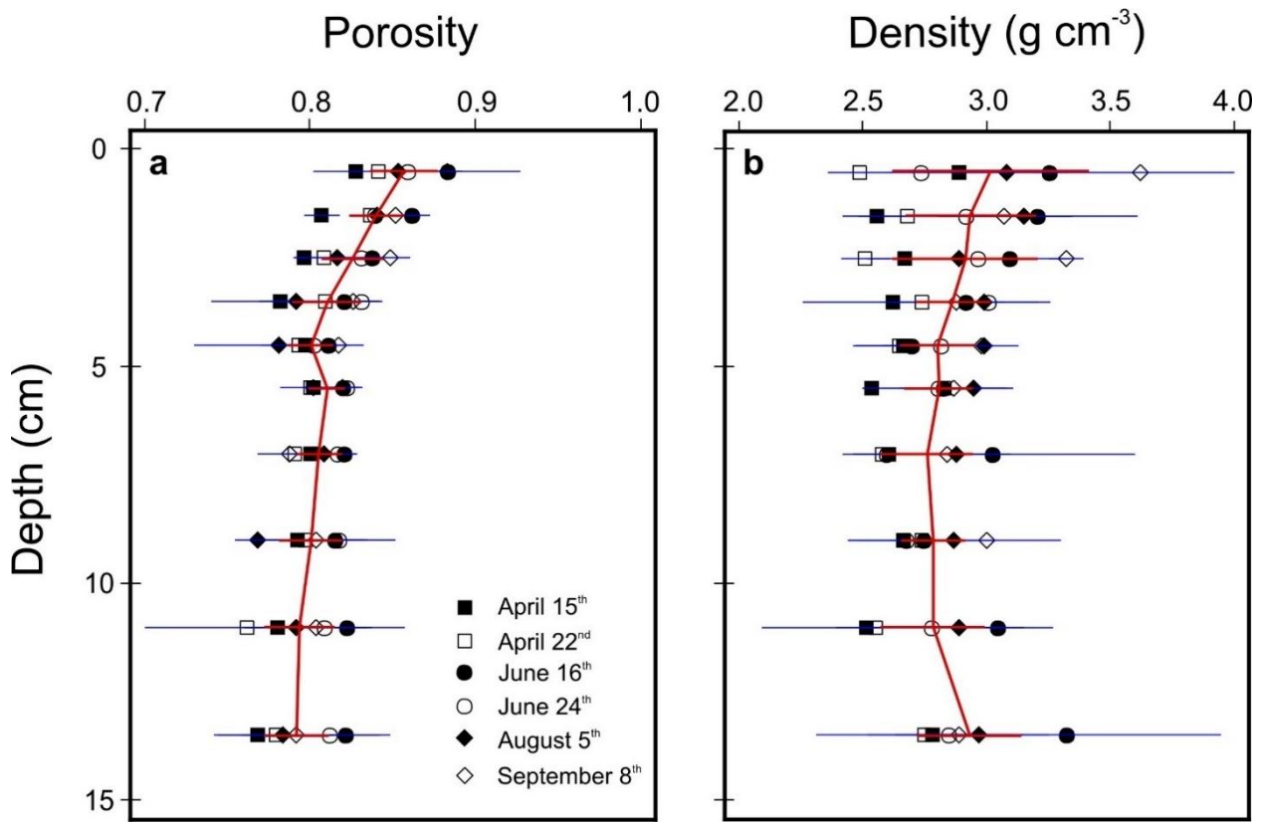
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129 **Fig. S3** Depth profile of the grain size distribution in the triplicate sediment cores at Station A, B
 130 & C for the spatial study carried out in July 2016. The y-axis represents each section of the
 131 sediment layers in the triplicate cores. Right panel: image of a sediment core sampled at Station
 132 D. There was no analysis of the grain size distribution for Station D.



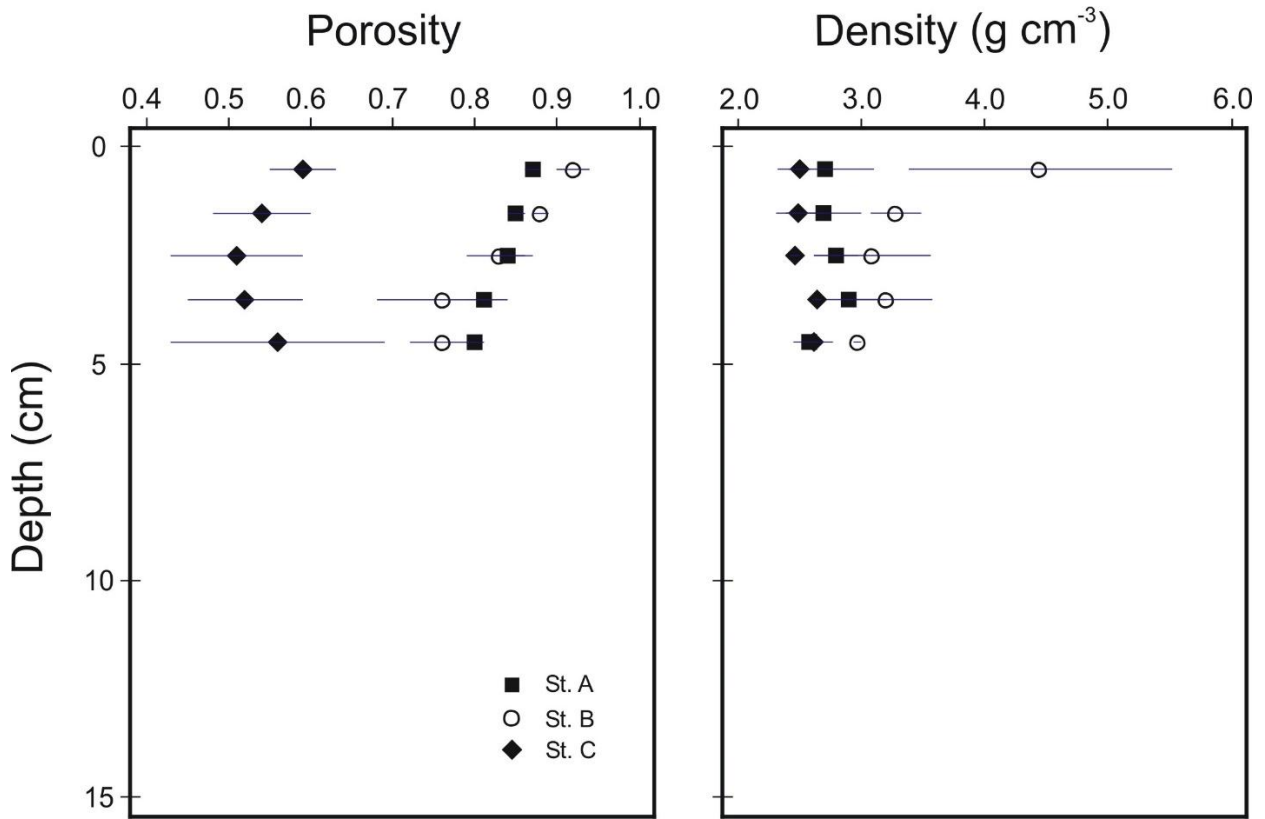
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135 **Fig. S4** Depth profiles of the sediment porosity (a) and density (b) at the Nord Dumet
136 monitoring station (St. A) for the temporal study carried out from April to September 2015. The
137 solid horizontal lines are the standard error of the triplicate sediment cores. The red curves are
138 the average of all of the values with the standard error ($n = 60$).



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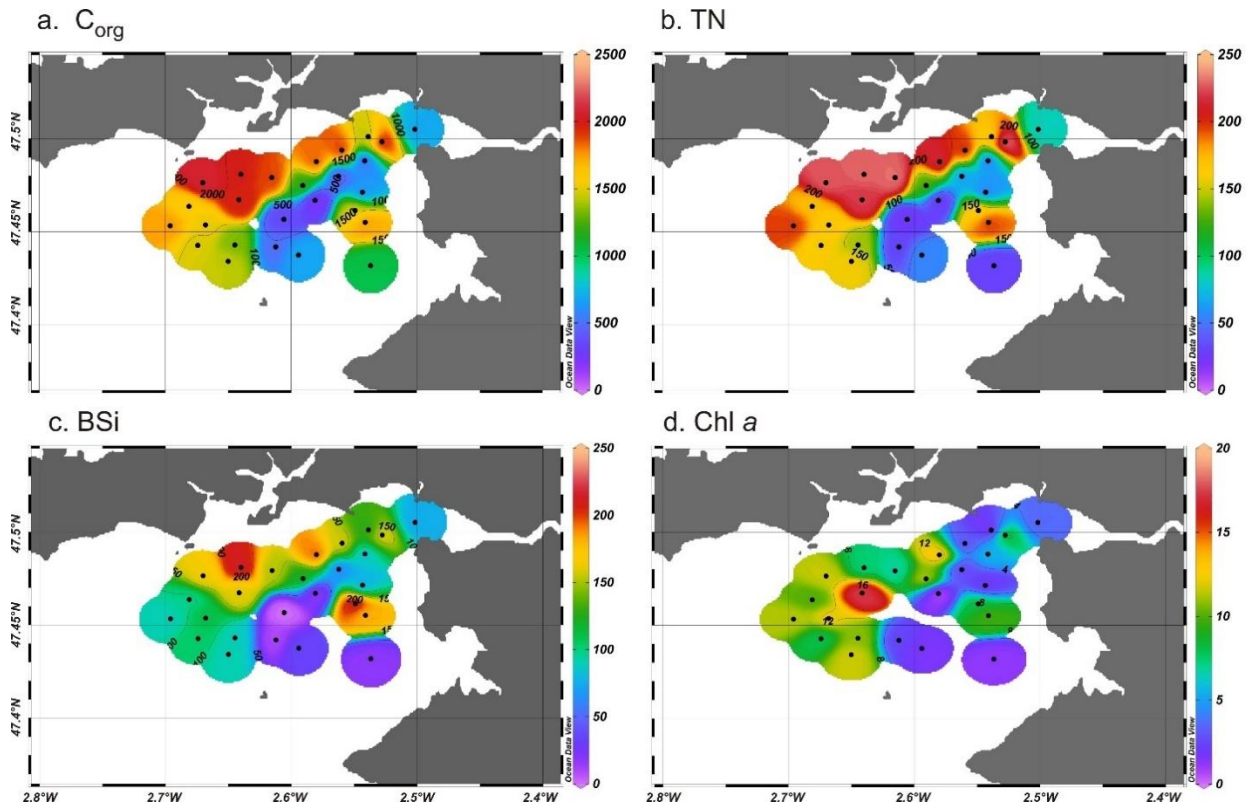
141 **Fig. S5** Depth profiles of the sediment porosity (a) and density (b) at Station A, B & C for the
142 spatial study carried out in July 2016. The solid horizontal lines are the standard error of the
143 triplicate sediment cores. Note: there was no measurement for Station D (gravel).



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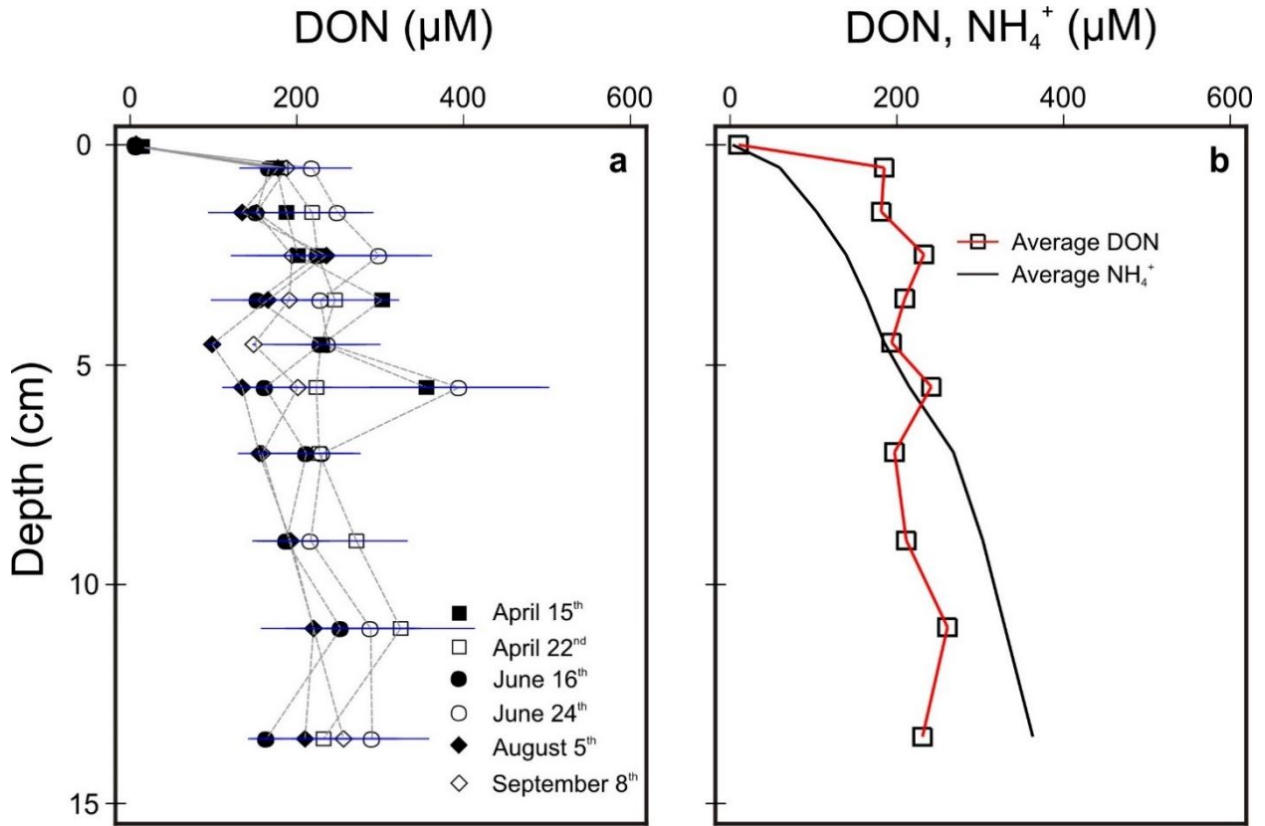
146 **Fig. S6** Concentrations of C_{org} , total N, BSi (in $\mu\text{mol g}^{-1}$) and Chl a (in $\mu\text{g g}^{-1}$) at the sediment
147 surface (5 cm and 1 cm for Chl a) in April 2016. The data was interpolated using an automatic
148 weighted-average gridding with the Ocean Data View software.



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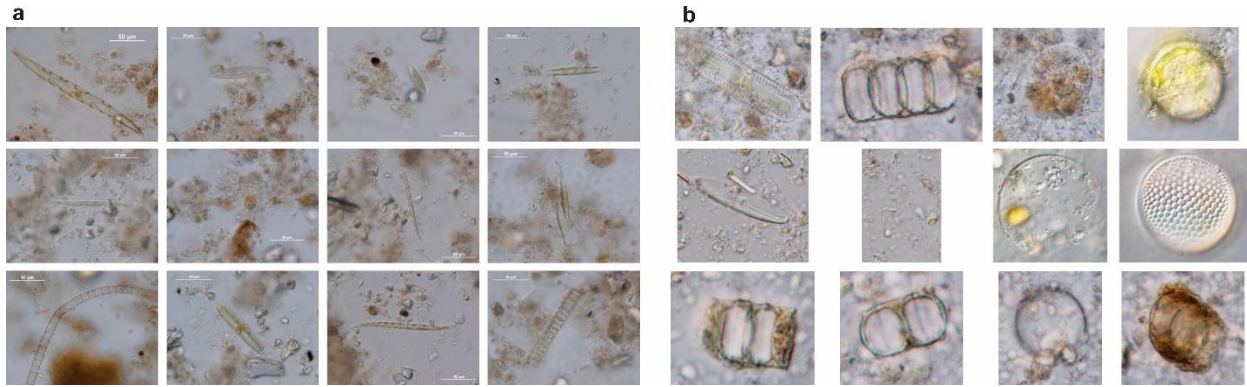
151 **Fig. S7** DON depth profiles at the Nord Dumet monitoring station (St. A) for the temporal study
 152 carried out from April to September 2015 (a) and global average of the measured DON and
 153 NH_4^+ values (b). The solid horizontal lines are the standard error of the triplicate sediment cores.



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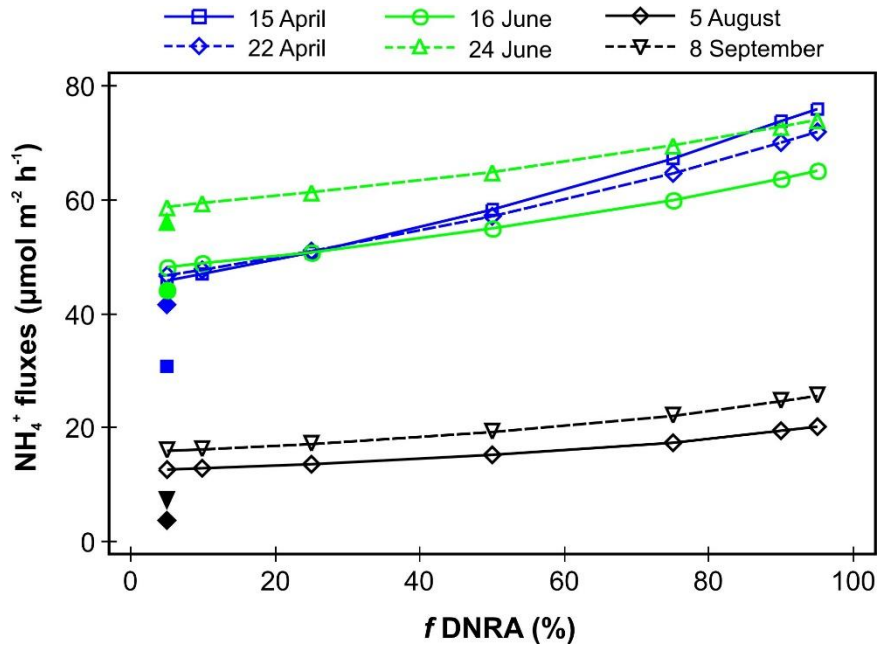
156 **Fig. S8** Microscopic observation of microalgae cells on the surface sediment at the Nord Dumet
157 monitoring station (St. A) during the period of study from April to June (a) and from August to
158 September (b) 2015.



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162 **Fig. S9** Modeled NH_4^+ fluxes in 2015 across the SWI as a function of the various $f\text{DNRA}$
163 values. The various colored lines with the colored symbols represent the sampling dates. The
164 solid symbols are the NH_4^+ fluxes measured at the Nord Dumet station.



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