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Biogeosciences Discussions

Particulate biogenic barium tracer of mesopelagic carbon remineralization in the Mediterranean Sea (PEACETIME project)

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ABSTRACT

We report on the sub-basins variability of particulate organic carbon (POC) remineralization in the central and western Mediterranean Sea during a late spring period (PEACETIME cruise). POC remineralization rates (MR) were estimated using the excess non-lithogenic particulate barium (Ba_{xs}) inventories in mesopelagic waters (100-1000 m) and compared with prokaryotic heterotrophic production (PHP). MR range from 25 ± 2 to 306 ± 70 mg C m^{-2} d^{-1}. Results reveal larger MR processes in the Algerian (ALG) basin compared to the Tyrrhenian (TYR) and Ionian (ION) basins. Ba_{xs} inventories and PHP also indicates that significant remineralization occurs over the whole mesopelagic layers in the ALG basin in contrast to the ION and TYR basins where remineralization is mainly located in the upper 500 m horizon. We propose that this may be due to particle injection pumps likely driven by strong winter convection in the Western basin of the Mediterranean Sea. This implies significant differences in the remineralization length scale of POC in the central Mediterranean Sea relative to the western region.

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1. Introduction

In the ocean, remineralization rate associated with sinking particles is a crucial variable for air sea CO_2 balance (Kwon et al., 2009). Most of the sinking particulate organic carbon (POC) conversion (i.e. remineralization) into CO_2 by heterotrophic organisms (i.e. respiration) occurs within the mesopelagic zone (100-1000 m) (Martin et al., 1987; Buesseler et al., 2007; Buesseler and Boyd, 2009). A quantitative representation of this process is thus crucial to future predictions of the ocean's role in the global C cycle (IPCC, 2014). Particulate biogenic barium (Ba_{xs}) is a geochemical tracer of POC remineralization in the mesopelagic layer. Ba_{xs} occurs in the form of barite ($BaSO_4$ crystals) in the dark ocean as a byproduct of prokaryotic remineralization. In a global ocean undersaturated with respect to barite (Monnin and Cividini, 2006), Ba_{xs} precipitates inside oversaturated biogenic micro-environments during POC degradation by heterotrophic prokaryotes, through sulfate and/or barium enrichment (Dehairs et al., 1980; Stroobant et al., 1991; Bertram and Cowen, 1997; Ganeshram et al., 2003). By applying a transfer function relating Ba_{xs} to O_2 consumption (Dehairs et al., 1997) Ba_{xs} has been widely used since the 90's as an estimator of mesopelagic POC remineralization rates in various sectors of the Southern Ocean, North Pacific and North Atlantic (Cardinal et al., 2001, 2005; Dehairs et al., 2008; Jacquet et al., 2008a, 2008b, 2011, 2015; Planchon et al., 2013; Lemaitre et al., 2018). Jacquet et al. (in review) recently reported that such transfer function could be applied in the Mediterranean Sea without restriction. This last study complemented previous investigations aiming at improving the use of Ba_{xs} to estimate local processes of POC remineralization in the Mediterranean Sea (Jacquet et al., 2016; Jullion et al., 2017). In this prospect, the Mediterranean Sea represents a unique case study, mainly due to unresolved issues related to the imbalance in the regional C budget (Sternberg et al., 2008; Santinelli et al., 2010; Tanhua et al., 2013b; Malanotte-Rissoli et al., 2014).

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The present work is part of the PEACETIME project (ProcEss studies at the Air-sEa Interface after dust deposition in the MEditerranean sea) (http://peacetime-project.org/). PEACETIME aimed at studying the chain of processes occurring in the Mediterranean Sea after atmospheric deposition (Saharan dust), and to put them in perspective on-going environmental changes (Guieu et al., in review, this special issue). Dust deposition is a major source of micro- and macro-nutrients that can stimulate the biological carbon pump through organic matter production (i.e. primary production) and its subsequent export and remineralization in the water column (Pabortsava et al., 2017). Overall, the aims of the present contribution to the PEACTIME project were: (1) to document particulate biogenic Ba_{xs} in different ecoregions of the western and central parts of the Mediterranean Sea. Previous Ba_{xs} data in the Mediterranean Sea are relatively scarce with limited vertical sampling resolution (Sanchez-Vidal et al., 2005) or restricted locations (Dehairs et al., 1987; Sternberg et al., 2008); (2) to estimate Ba_{xs}-based POC remineralization rates at mesopelagic depths (MR) according to the Dehairs' transfer function (Dehairs et al., 1997; Jacquet et al., in review), and (3) to determine the relationship between Ba_{xs} and environmental variability, including dust deposition, and (4) assess potential differences in remineralization length scale of POC in the various ecoregions of the Mediterranean Sea.

2. Material and methods

2.1 Study area

The PEACETIME cruise (https://doi.org/10.17600/17000300) was conducted during late spring conditions from May 10 to June 11, 2017 (French R/V Pourquoi pas?) in the western and central Mediterranean (Figure 1a). Three main ecoregions (Reygondeau et al., 2014; Ayata et al., 2018) were crossed during the cruise: the Algero-Provençal basin (later referred to as ALG), the Tyrrhenian basin (TYR) and the Ionian basin (ION). The hydrography

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displays the general three-layer Mediterranean system with surface, intermediate and deep waters (Tamburini et al., 2013; Tanhua et al., 2013b; Hainbucher et al., 2014, Malanotte-Rizzoli et al., 2014). Briefly, the main water masses can be distinguished (see potential temperature – salinity diagram during the PEACETIME cruise in Figure 1b): (1) from west to east surface Atlantic Water (SW) is gradually replaced by Ionian surface Water (ISW) and Levantine Surface water (LSW); (2) Winter Intermediate Water (WIW) and Levantine Intermediate water (LIW). LIW is present at intermediate depths (from 200 to 800 m) and is characterized by a local maximum of salinity and a local minimum of dissolved oxygen concentration; (4) Mediterranean Deep Water (MDW).

2.2 Barium sampling and sample processing

Thirteen stations were sampled for particulate barium from surface to 2000 m (thirty depths in total) in the ALG (stations #1, #2, #3, #10, #Fast, #9 and #4), TYR (stations #5, Tyrr and #6) and ION (stations #8, #7 and #Ion) basins. Three of these stations were sampled twice on different days (long stations #Fast, #Tyrr and #Ion), but due to technical problem no particulate barium data are available for the second visit at station #Ion.

We followed the same Ba sample processing and analysis as in Jacquet et al. (2015). Briefly, 4 to 6 L of seawater sampled using Niskin bottles were filtered onto 47 mm polycarbonate membranes (0.4 µm porosity). Filters were rinsed with MQ water grade, dried (50°C) and stored for later analysis. In the laboratory, we performed a total digestion of filters. Solutions were analysed for Ba and other elements of interest (i.e. Al, Sr and Ca) by HR-ICP-MS (High Resolution-Inductively Coupled Plasma- Mass Spectrometry; ELEMENT XR, Thermo).

Particulate biogenic barium in excess (hereafter referred to as Ba_{xs}) was calculated as the difference between total Ba and lithogenic Ba using Al as the lithogenic reference element (Dymond et al., 1992; Taylor and Mc.Lennan, 1985). The standard uncertainty (Ellison et al.,

2000) on Ba_{xs} concentration ranges between 5.0 and 5.5%. The term "in excess" is used to indicate that concentrations are larger than the Ba_{xs} background. The background (or residual value) is considered as "preformed" Ba_{xs} at zero oxygen consumption left over after transfer and partial dissolution of Ba_{xs} produced during degradation of previous particles export events. This background Ba_{xs} value likely depends on the saturation state of the water with respect to barite ($BaSO_4$, the main phase of particulate biogenic barium). Saturation indexes were reported in Jacquet et al. (2016) over a high resolution and quasi-zonal Mediterranean transect (M84/3 cruise; Tanhua et al., 2013a). They revealed that the water column throughout the study area is largely undersaturated, with saturation state ranging between 0.2 and 0.6. A background Ba_{xs} value of 130 pM was recently reported in Jacquet et al. (in review). It is close to the average Ba_{xs} contents observed at greater depth (>1000 m) in the present study (see below).

2.3 Prokaryotic heterotrophic production

Prokaryotic heterotrophic production (PHP) estimation was measured by incorporating L-[4,5-3H]-Leucine (3H-Leu, 109 [TC1] Ci mmol−1 of specific activity, PerkinElmer®) to get a final saturation concentration of 20 nM in surface until 800 m depth and 10 nM [TC2] below 800m depthn, and incubated between 4 hours for sample from 0-200 m depth, 6 hours for sample between 200-800 m depth and 10 hours for sample below 800m depth. Deep prokaryotic heterotrophic production (PHP) was measured over time course experiments at *in situ* temperature in the dark following the protocol described in Tamburini et al. (2002). At the end of incubation, triplicate 40 mL formaldehyde-killed blanks and triplicate 40 mL were incubated at at *in situ* temperature (13°C). At the end of incubation, samples were fixed with 2% final concentration formaldehyde and stored at 4°C until filtration. The detailed protocol is also detailed in Kirchman (1993). Epipelagic layers (0- 250 m) were incubated using the microcentrifuge technique at 20 nM final concentations (see Van

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Wambeke et al., this issue, submitted). To calculate prokaryotic heterotrophic production, we used the empirical conversion factor of 1.5 ng C per pmol of incorporated leucine according to Simon and Azam (1989). Indeed, isotope dilution was negligible under these saturating concentrations as checked occasionally with concentration kinetics.

2.4 POC remineralization rates

We recently reported on the validity of the Dehairs's transfer function (Dehairs et al., 1997) in the Mediterranean basin to estimate mesopelagic POC remineralization (Jacquet et al., in review). We applied the similar approach to estimate remineralization rates (MR):

MR = [(Ba_{xs} –Ba BKG)/17450] x Z x RR (Eq.1)

With Z the depth layer considered and RR the Redfield C/O_2 molar ratio (127/175; Broecker et al., 1985). As reported above, a Ba background (BKG) value of 130 pM was used.

3. Results

Particulate biogenic Ba_{xs}, biogenic Ba fraction (%) and particulate Al, Sr and Ca concentrations are reported in Figure 2 in the upper 2000 m along a zonal transect crossing the three main sub-basins. PHP rates are also reported in Figure 2 in the upper 500 m along the same transect.

At stations #9, #Tyrr, #8 and #Ion, Ba_{xs} concentrations in the upper 100 m are quite high (>5000 pM), with values reaching up to 11700 pM (80 m at station #Tyrr) (Figure 2a). The very high Ba_{xs} contents in the surface are quite unusual, though similar values were occasionally observed in earlier Southern Ocean studies (Dehairs et al., 1991, 1997; Jacquet et al., 2007b, 2008a,b). The high Ba_{xs} values at stations #Tyrr and #Ion are associated with higher Sr and Ca concentrations (Figure 2d, e).

The upper mesopelagic layer (100-500 m) shows the characteristic large Ba excess (maximum), as illustrated in Figure 3a. The lithogenic impact on the Ba_{xs} signal is relatively low except at stations #4, #5 and #Tyrr where it reached >20 % at some depths in the water column (Figure 2b). The Ba_{xs} maximum at stations in the ALG basin and at station #7 in the ION basin presents a deeper extent compared to stations in the TYR basin (Figure 2a). At station #Ion Ba_{xs} maximum coincides with higher Ca concentrations between 300 and 400 m (Figure 2e). However, the Ba_{xs} maximum is not restricted to the upper mesopelagic layer and also extents deeper. This is especially salient at stations in the ALG basin. At the other stations Ba_{xs} concentrations below 500 m decrease to reach the background value of 130 pM. Among stations sampled twice for barium during the cruise, station #Fast presents similar Ba_{xs} profiles except between 400 and 1000 m with lower concentrations measured during the second visit (3 days later; Figure 3a). In contrast, at station #Tyrr differences between Ba_{xs} profiles mainly occurred in the surface layer and upper mesopelagic layer, with relatively higher Ba_{xs} peaks during the second visit (2 days later).

PHP rates decrease from west to east in surface waters (Figure 2f). At station #Fast, PHP rates decreased from 49.4 ng C l^{-1} h^{-1} in surface to values between 7.5 and 11.1 ng C l^{-1} h^{-1} at 100 m depth and below 6.05 ng C l^{-1} h^{-1} below 200 m depth (Figure 3d). Same trend can be found in #Tyrr and #Ion stations with a rate in surface waters of around 36 and below 27 ng C l^{-1} h^{-1} respectively (Figure 3e and 3f).

Depth-weighted average (DWAv) concentrations of Ba_{xs} are reported in Table 1 for the upper (100-500 m) and entire (100-1000 m) mesopelagic layer. Since the base of the mixed layer was generally shallower than 100 m, this depth is taken as the upper boundary of the mesopelagic domain. DWAv range from 221 to 979 pM. On average stations located in the ALG basin present higher DWAv than in the TYR and ION basins. DWAv Ba_{xs} values remained rather stable over the 3–day period at station #Fast and even displayed a slight decrease from 527 to 381 pM (100-1000 m), corroborating above-mentioned profiles variation. In contrast, at station #Tyrr DWAv Ba_{xs} values for the 100-500 m layer increased from 329 to 810 pM over the 2–day period. Also, for all stations DWAv Ba_{xs} values globally increase when considering the whole mesopelagic layer, reflecting that significant Ba_{xs} is still extending up to 1000 m depth.

4. Discussion

4.1 Ba_{xs} distributions across the sub-basins

The very high Ba_{xs} concentrations reported in the surface layer at stations #9, #Tyrr, #8 and #Ion are associated with Sr and Ca maxima, likely linked with potentially ballasted phytoplankton-derived material. Similar observations were previously reported in the Southern Ocean, revealing that in the surface water particulate Ba_{xs} is incorporated into or adsorbed onto biogenic material, with barite being a minor component (Dehairs et al., 1991, 1997; Jacquet et al., 2007a, 2008a, b). Below the surface layer, Ba_{xs} presents the characteristic maximum reflecting remineralization processes in the mesopelagic layer. The maximum extents up to 1000 m in the ALG and ION basins, while it is mostly located in the upper 500 m in the TYR basin. The lithogenic impact on the Ba_{xs} signal is relatively low, except at stations #4, #5 and #Tyrr where it is moderated. A dust deposition event occurred over a large area including the southern Tyrrhenian Sea during PEACETIME, starting on May 10 (Guieu et al., this issue) at the beginning of the campaign. Particulate Al concentrations and estimated lithogenic Ba fraction were sampled at theses stations 5 to 12 days after the event and would potentially reflect a slight impact of this dust event in depth.

4.2 Mesopelagic Ba_{xs} and prokaryotic heterotrophic production

Previous studies highlighted the relationship between the mesopelagic Ba_{xs} and the vertical distribution of prokaryotic heterotrophic production (PHP), reflecting the temporal progression of POC remineralization processes. Figure 3d-f shows PHP profiles in the upper 2000 m at long station #Fast, #Tyrr and #Ion. The whole dataset is discussed in Van Wambeke et al. (submitted, this issue). Figure 4 shows column-integrated PHP at 100 m over

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column-integrated PHP at 500 m vs. DWAv Ba_{xs} calculated over the entire 100-500 m depth interval. Results are confronted to the relationships obtained in the Southern Ocean (Jacquet et al., 2008a, 20015) and recently in the northeast Atlantic and northwestern Mediterranean Sea (PAP and ANTARES/EMSO-LO observatory sites, respectively) (Jacquet et al., in review). Results during PEACETIME follow the same trend, indicating higher DWAv Ba_{xs} in situation where a significant part of column-integrated PHP is located deeper in the water column (high Int.PHPx1/IntPHPx2 ratio, Figure 4). During KEOPS2, the lowest DWAv were reported for stations located in a meander and reflected the occurrence of different (earlier) stages of bloom advancement compared to the other stations (sea "season advancement" in Figure 4). Similarly, station #5 and #Tyrr2 would reflect the temporal evolution of the establishment of mesopelagic remineralization processes in the TYR basin compared to the other basins. Measurements performed during the second visit at station #Tyrr4 two days later corroborate this hypothesis and would potentially reflect an increase of remineralization processes in the upper mesopelagic depths.

4.3 Mesopelagic C remineralization

We translated mesopelagic Ba_{xs} DWAv into POC remineralization rates (MR) using Equ. (1). Figure 5 reports MR rates for the upper and entire mesopelagic layer. MR range from 25 ± 2 to 306 ± 70 mg C m^{-2} d^{-1}. We observe a large difference in MR rates in the ALG basin depending on the integration depths. This is especially salient at station #9. These results reveal significant remineralization occurred between 500 and 1000 m. In contrast, in the ION and TYR basins MR rates reflect that remineralization is mainly located in the upper 500 m horizon. As mentioned above, stations in the TYR region experienced a dust event and our particulate Al concentrations and estimated lithogenic Ba fraction presented a slight impact of this event. Despite the small increase in MR rates at station #Tyrr between the two visits, we can wonder whether the overall impact of the dust event could result in lower and upper

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mesopelagic layer-restricted MR processes. This would suggest that by providing ballast material (dust), and thereby decreasing of the exposition time of particles to prokaryotic remineralization (Pabortsava et al., 2017), the dust event reduced MR at station #Tyrr. Another dust event occurred on June 5 a few hours after the first sampling at station #Fast in the ALG basin. Again, it is very interesting to observe a decrease in MR rates between the two visits in the 500-1000 m depth layer at station #Fast. However, station #Fast does not present any evidence of an impact on particulate Al concentrations and estimated lithogenic Ba fraction. Furthermore, it is obvious that, in contrast to variables change in the surface layer, observable response in mesopelagic Ba_{xs} and remineralization processes to a single dust event would require continuous observations over few days to week, from an initial state just before the event to a complete process of POC production, subsequent export and remineralization.

Overall, the present study indicates that the ALG basin is the place where higher mesopelagic remineralization processes occur. These new results confirm previous work of Jullion et al. (2017) reporting dissolved Ba and Parametric Optimum Multiparameter (POMP) calculation-derived POC remineralization fluxes (from 156 to 348 mg C m^{-2} d^{-1}). These authors showed significant differences in the mesopelagic MR between the west and the east of the Mediterreanean basin, suggesting an additional export pathway and subsequent remineralization driven by the winter deep convection in the western basin. By providing a detailed Ba_{xs} dataset in the ALG, TYR and ION sub-basin, the present work would confirm the hypothesis of particle injection pump (Boyd et al., 2019) at western stations in the Mediterranean Sea.

5. Conclusion

The present paper presents an expended dataset on the Ba_{xs} distribution in the western and

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central Mediterranean Sea in late spring 2017. Our results reveal the Ba_{xs} distribution globally extends deeper in the ALG basin. Derived MR fluxes are also higher in this basin. The relationship between DWAv Ba_{xs} and PHP in the mesopelagic layer corroborates the trend of higher Ba_{xs} in situation of significant deeper PHP activity and subsequent remineralization.

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Figures

Figure 1: (a) Map of the study area showing the three sub-basins (ALG, TYRR and ION) with stations' locations; (b) Potential temperature – salinity diagram with isopynals (kg m^{-3}) for PEACETIME profiles. Graph constructed using Ocean Data View (Schlitzer, 2002).

Figure 2: (a-e) Sections of particulate biogenic Ba (Ba_{xs}, pM), Al (pM), Sr (pM) and Ca (nM) concentrations, and % Ba_{xs} (biogenic fraction) in the upper 2000 m water column. (f) Section of PHP (ngC L^{-1} h^{-1}) is reported in the upper 500 m. Graph constructed using Ocean Data View (Schlitzer, 2002).

Figure 3: Ba_{xs} (a-c; pM) and PHP (d-f; ngC L^{-1} h^{-1}) profiles in the upper 2000 m at long stations #Fast, #Tyrr and #Ion.

Figure 4: Integrated PHP in the upper 100 m over integrated PHP in the upper 500 m versus depth-weighted average (DWA) mesopelagic Ba_{xs} (pM) over the 150-500 depth interval during PEACETIME. The same relations are reported for KEOPS1 and KEOPS2 cruises (Southern Ocean; Jacquet et al., 2015) and at the PAP (NE-Atlantic) and ANTARES/EMSO-LO (NW-Mediterranean Sea) observatory sites (Jacquet et al., in review, biogeosciences).

Figure 5: POC remineralization fluxes (mg C m^{-2} $d^{-}1$) in the upper (100-500 m) and entire (100-1000 m) mesopelagic layer in the ALG, TYR and ION basins.

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Tables

Table 1: Depth-weighted average (DWAv) concentrations of Ba_{xs} (pM) and remineralization rates (MR; mg C m^{-2} d^{-1}) for the upper (100-500 m) and entire (100-1000 m) mesopelagic layer.

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Figure 1

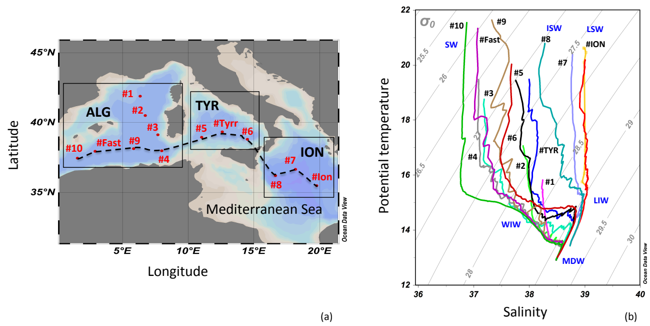

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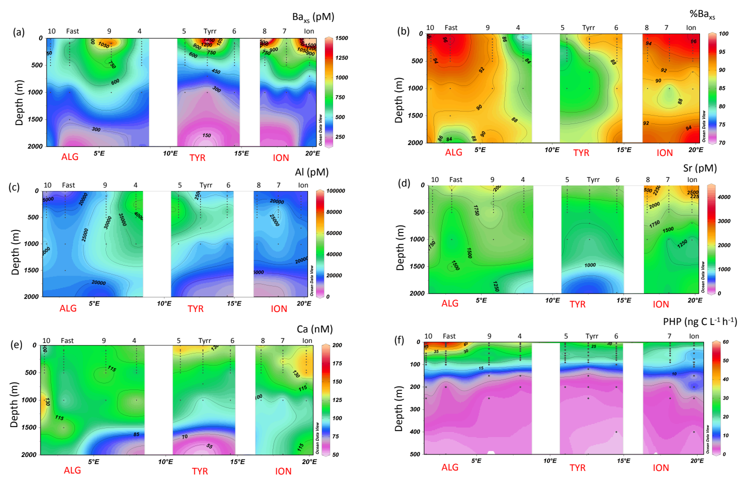
Figure 2



480 Figure 3

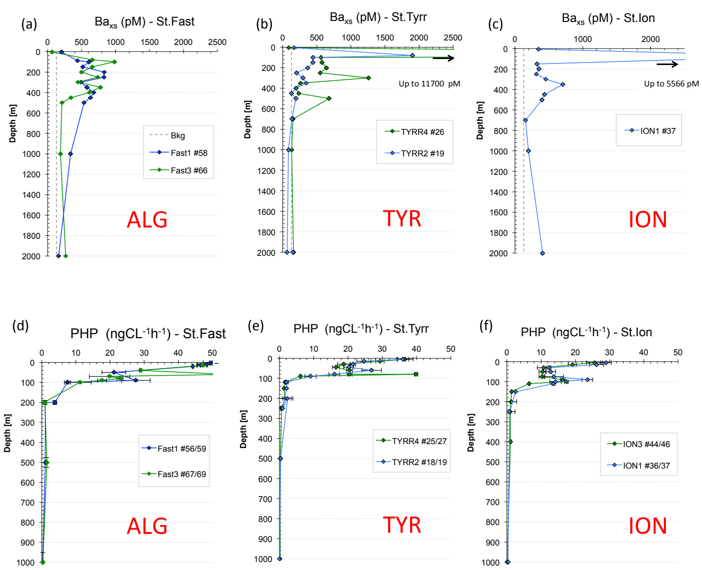


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Figure 4

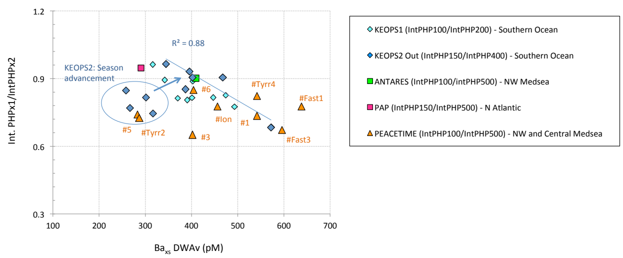

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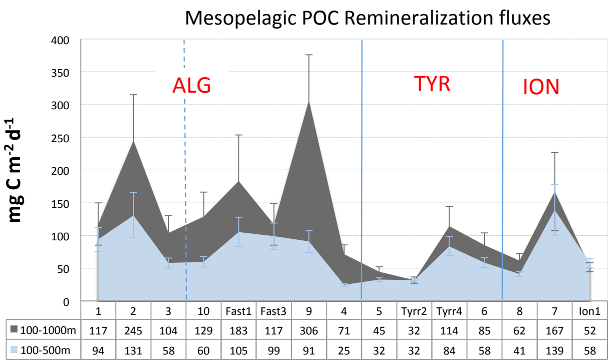
Figure 5



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489 Table 1

Basin	Station #	Mesopelagic layer	DWAv Ba_{xs} [pM]	MR [mg C m^{-2} d^{-1}]	Stnd error (%)
Algero-Provençal	1	upper	542	94	20
	1	entire	374	117	27
	2	upper	717	131	26
	2	entire	645	245	28
	3	upper	402	58	13
	3	entire	353	104	25
	4	upper	243	25	8
	4	entire	281	71	20
	9	upper	981	91	19
	9	entire	979	306	23
	Fast1	upper	638	105	21
	Fast1	entire	527	183	38
	Fast3	upper	596	99	20
	Fast3	entire	381	117	27
	10	upper	418	60	13
	10	entire	410	129	29
Tyrrhenian	5	upper	283	32	9
	5	entire	226	45	17
	Tyrr2	upper	284	32	9
	Tyrr2	entire	200	32	15
	Tyrr4	upper	542	84	17
	Tyrr4	entire	380	114	26
	6	upper	404	58	13
	6	entire	313	85	22
Ionian	7	upper	769	139	28
	7	entire	485	167	36
	ION	upper	456	58	13
	ION	entire	315	52	13
	8	upper	363	41	10
	8	entire	273	62	18