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On the barium - oxygen consumption relationship in the Mediterranean Sea: implications for mesopelagic marine snow remineralisation.

Stéphanie H.M. Jacquet[1*], Dominique Lefèvre[1], Christian Tamburini[1], Marc Garel[1], Frédéric A.C. Le Moigne[1], Nagib Bhairy[1], Marie Roumagnac[1], Sophie Guasco[1]

[1]Aix Marseille Université, CNRS/INSU, Université de Toulon, IRD, Mediterranean Institute of Oceanography (MIO), UM 110, 13288 Marseille, France

[*]Correspondence to: S. Jacquet (*stephanie.jacquet@mio.osupytheas.fr)*

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ABSTRACT

In the ocean, remineralisation rate associated with sinking particles is a crucial variable. Since the 90's, particulate biogenic barium (Ba_{xs}) has been used as an indicator of carbon remineralization by applying a transfer function relating Ba_{xs} to O_2 consumption (Dehairs's transfer function, Southern Ocean-based). Here, we tested its validity in the Mediterranean Sea (ANTARES / EMSO-LO) for the first time by investigating connections between Ba_{xs}, prokaryotic heterotrophic production (PHP) and oxygen consumption (JO_2-Opt; optodes measurement). We show that: (1) higher Ba_{xs} (409 pM; 100- 500 m) in situations where integrated PHP (PHP100/500= 0.90) is located deeper, (2) higher Ba_{xs} with increasing JO_2-Opt, and (3) similar magnitude between JO_2-Opt (3.14 mmol m^{-2} d^{-1}; 175- 450 m) and JO_2-Ba (4.59 mmol m^{-2} d^{-1}; transfer function). Overall, Ba_{xs}, PHP and JO_2 relationships follow trends observed in the Southern Ocean. We believe that such transfer function could apply in the Mediterranean Sea with no restriction.

KEY WORDS: particulate biogenic barium, mesopelagic zone, oxygen consumption, prokaryotic heterotrophic production, carbon remineralization, Mediterranean Sea

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1. INTRODUCTION

Ocean ecosystems play a critical role in the Earth's carbon (C) cycle [IPCC, 2014]. The quantification of their impacts of both present conditions and future predictions remains one of the greatest challenges in oceanography [Siegel et al., 2016]. In essence, the biological C pump is termed for the numerous processes involved in maintaining the vertical gradient in dissolved inorganic C. This includes processes such as organic matter production in surface, its export and subsequent remineralization. Most of marine snow organic C conversion (i.e. remineralization) into CO_2 by heterotrophic organisms (i.e. respiration) occurs in the mesopelagic zone (100-1000 m) [Martin et al., 1987; Buesseler and Boyd, 2009]. Globally, the flux of C exported below 1000 m depth is the key determinant of ocean carbon storage capacity [Henson et al., 2011]. However, there is no consensus on C transfer efficiency estimations from field experiments, leading to an imbalance of the water column C budget [Giering et al., 2014]. Resolving this imbalance is in the core of numerous studies in the global ocean, but also regionally, especially in the Mediterranean Sea (MedSea). Due to limited exchanges with adjacent basin and the existence of an intense overturning circulation qualitatively resembling the global one (but with shorter time scales), the MedSea is often considered as a laboratory to observe and understand the impact of transient climate variability on ecosystems and biogeochemical cycles [Malanotte-Rissoli et al., 2014]. In a context of climate changes, better constraining C fluxes and the ocean C storage capacity is of crucial importance.

Particulate barium in excess (Ba_{xs}, i.e. biogenic Ba from total particulate Ba after correction for lithogenic Ba) is a geochemical tracer of particulate organic carbon (POC) remineralization in the mesopelagic layer [Dehairs et al., 1997]. Ba_{xs} mostly occurs in the form of barite microcrystals ($BaSO_4$) at these depths. In a global ocean undersaturated with respect to barite, studies report that Ba_{xs} would precipitate inside oversaturated biogenic micro-environments during POC

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degradation by heterotrophic prokaryotes in the mesopelagic zone, through sulfate and/or barium enrichment [Bertram and Cowen, 1997]. The first-ever studies on mesopelagic Ba_{xs} reported coinciding Ba_{xs} maxima with depths of dissolved O_2 minimum and pCO_2 maximum [Dehairs et al., 1987, 1997]. By using an 1D advection-diffusion model applied to highly resolved, precise O_2 profiles in the Atlantic sector of the Southern Ocean (ANTX/6 cruise; Shopova et al., 1995), Dehairs et al. [1997] established an algorithm converting mesopelagic Ba_{xs} concentration into O_2 consumption rate (JO_2) and organic C remineralized (POC remineralization rate). This transfer function has been widely used until now [Cardinal et al., 2001- Lemaitre et al., 2018]. Yet its validity has never been tested in other oceanic provinces. Recently, significant progresses were made in relating Ba_{xs}, O_2 dynamics to prokaryotic heterotrophic activity [Jacquet et al., 2015]. Nevertheless, the Dehairs transfer function has never been revised since. These advancements clearly show that Ba_{xs} is closely related with the vertical distribution of prokaryotes heterotrophic production (PHP) (the rate of change with depth), reflecting the temporal progression of POC remineralization processes. Also, in a first attempt to test the validity of the Dehairs's transfer function in other locations, Jacquet et al. [2015] confronted oxygen consumption rates (JO_2) from direct measurements (dark community respiration, DCR) to derived JO_2 from Ba_{xs} data (using the transfer function) in the Kerguelen area (Indian sector of the Southern Ocean). We revealed good convergence of JO_2 rates from these two approaches, further supporting the Dehairs's function to estimate POC remineralization rates in different biogeochemical settings of the Southern Ocean.

Here, we further investigate relationships between the mesopelagic Ba_{xs} proxy, prokaryotic activity and oxygen dynamics (Figure 1a) in the northwestern Mediterreanean Sea (MedSea), a different biogeochemical setting to those already studied (see references above). Today, observations of the various components of the MedSea biological C pump provide organic C fluxes varying by at least an order of magnitude [Santinelli et al., 2010; Ramondenc et al., 2016].

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Malanotte-Rissoli et al. [2014] reviewing unsolved issues and future directions for MedSea research highlighted the need to further investigate biogeochemical processes at intermediate (mesopelagic) and deep layers to reconcile the C budget in the Mediterranean basin. Previous particulate Ba_{xs} dataset is very scarce in the NW- MedSea, with in general very low vertical sampling resolution [Sanchez Vidal et al., 2005] or very restricted studied areas [Dehairs et al., 1987; Sternberg et al., 2008]. Here we discuss Ba_{xs}, PHP and JO_2 (from optodes measurement during incubations) at the ANTARES / EMSO-LO observatory site (Figure 1a, b). We hypothesize that the Dehairs's transfer function converting Ba_{xs} into POC remineralization also applies in a different ocean ecosystem functioning from the Southern Ocean. We suggest that the Ba_{xs} proxy can be used as routine tracer to estimate local-scale processes of mesopelagic POC remineralization in the Mediterranean basin.

2. SAMPLING AND ANALYSES

2.1 STUDY SITE

The BATMAN cruise (https://doi.org/10.17600/16011100, March 10-16 2016, *R/V* EUROPE) took place to the ANTARES / EMSO-LO observatory site (42°48'N, 6°10'E; Tamburini et al., 2013), 40 km off the coast of Toulon, southern France (Figure 1b). The hydrological and biogeochemical conditions at this site are monitored monthly in the framework of the MOOSE (Mediterranean Ocean Observing System for the Environment) program and of the EMSO (European Multidisciplinary Subsea Observatory) observation program. The hydrography displays the general three-layer MedSea system with surface, intermediate and deep waters [Hainbucher et al., 2014]. Briefly, the main water masses can be distinguished (see potential temperature – salinity diagram during the BATMAN cruise in Figure 1c): (1) Surface Water (SW); (2) Winter Intermediate Water (WIW) and Levantine Intermediate water (LIW). LIW is

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present at intermediate depths (around 400 m at ANTARES) and is characterized by temperature and a salinity maxima; (4) Mediterranean Deep Water (MDW).

2.2 ANALYSES

For particulate barium, 4 to 7 L of seawater sampled using Niskin bottles were filtered onto 47 mm polycarbonate membranes (0.4 µm porosity) under slight overpressure supplied by filtered air. Filters were rinsed with few mL of Milli-Q grade water to remove sea salt, dried (50°C) and stored in Petri dishes. Thirteen depths between surface and 2000 m were sampled by combining different casts sampled closeby in time and space (total of 28 samples). In the laboratory, we performed a total digestion of filters using a tri-acid (0.5 mL HF /1.5 mL HNO_3 / HCl 1 mL; all Optima grade) mixture in closed teflon beakers overnight at 95°C in a clean pressurized room. After evaporation close to dryness, samples were re-dissolved into 10 mL of HNO_3 2%. The solutions were analysed for Ba and other elements of interest (Na and Al) by HR-ICP-MS (High Resolution-Inductively Coupled Plasma- Mass Spectrometry; ELEMENT XR ThermoFisher). Details on sample processing and analysis are given in Cardinal et al. [2001] and Jacquet et al. [2015]. The presence of sea-salt was checked by analysing Na and the sea-salt particulate Ba contribution was found negligible. 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 [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 phytoplankton growth events.

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Oxygen concentrations were measured using optical oxygen sensor (Aanderaa 4330-Optodes) at 4 depths in the mesopelagic layer (175, 250, 450 and 1000 m). In total each of the 8 optodes (two per depths) were placed into a sealed 1L borosilicate glass bottles incubated at a fixed temperature of 13°C in thermo-regulated baths for 24 to 48 hours. Oxygen consumption rates (later referred to as JO_2-Opt) were calculated from oxygen concentration evolution with time applying linear model calculations.

Prokaryotic heterotrophic production (PHP) estimation was measured over time course experiments at *in situ* temperature (13°C) following the protocol described in Tamburini et al. [2002]. ^{3}H-leucine labelled tracer [Kirchman, 1993] was used. To calculate prokaryotic heterotrophic production, we used the empirical conversion factor of 1.55 ng C per pmol of incorporated leucine according to Simon and Azam [1989], assuming that isotope dilution was negligible under these saturating concentrations.

3. RESULTS AND DISCUSSION

3.1 Barium vertical distribution

Particulate biogenic Ba_{xs}, particulate Al (pAl) and biogenic Ba fraction profiles in the upper 1000 m at ANTARES are reported in Figure 2a. Ba_{xs} concentrations range from 12 to 719 pM. The biogenic Ba fraction range from 51 to 91 % of the total particulate Ba signal. Particulate Al concentrations (pAl) are low and range from 8 to 170 nM. Ba_{xs} concentrations are low in surface water (<100 pM) where the lithogenic fraction reaches 43 to 49 % in the upper 70 m. From previous studies we know that Ba_{xs} in surface waters is distributed over different, mainly non-barite biogenic phases, and incorporated into or adsorbed onto phytoplankton material. As such these do not reflect POC remineralization processes, in contrast to mesopelagic waters where Ba_{xs} is mainly composed of barite formed during prokaryotic degradation of organic matter. At

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ANTARES the Ba_{xs} profile displays a mesopelagic Ba_{xs} maximum between 100 and 500 m, reaching up to 719 pM at 175 m. Ba is mostly biogenic at these depths (> 80 %). Ba_{xs} concentrations then decrease below 500 m to reach a background value of around 130 pM (see BKG in Figure 2). Note that the MedSea is largely undersaturated with respect to barite, with saturation state ranging between 0.2 and 0.6 over the basin [Jacquet et al., 2016; Jullion et al., 2017]. For comparison, the Ba background value in the Southern Ocean reaches 180 to 200 pM below 1000 m [Dehairs et al., 1997; Jacquet et al. 2015]. Previously, Sternberg et al. [2008] reported the seasonal evolution of Ba_{xs} profiles at the DYFAMED station (43°25'N-7°52'E; BARMED project) northeast from ANTARES (Figure 1c) in the NW-MedSea. The present Ba_{xs} profile at ANTARES (March 2016) is very similar to the Ba_{xs} profile measured in March 2003 at DYFAMED (Figure 2a). The slight difference between Ba_{xs} profiles in the upper 75 m suggests more Ba bounded and/or adsorbed onto phytoplankton material during BARMED. Both profiles present a Ba_{xs} maximum in the upper mesopelagic zone between 150 and 200 m. Below this maximum, Ba_{xs} concentrations gradually decrease to reach around 130 pM between 500 and 1000 m (this study). A similar value was reached between 500 and 600 m at the DYFAMED station over the whole studied period (between February and June 2003; Sternberg et al., 2008).

3.2 Prokaryotic heterotrophic production

The particulate excess Ba (>BKG) is centred in the upper mesopelagic zone between 100 and 500 m and reflects that POC remineralization mainly occurred at these depths (Figure 2a). Depth-weighted average (DWA) Ba_{xs} content (409 pM) was calculated over this entire depth interval. Figure 2b shows column-integrated PHP at 100 m over column-integrated PHP at 500 m (PHP100/500= 0.90), according to the relationship obtained during KEOPS1 (summer) and KEOPS2 (spring; out plateau stations) cruises in the Southern Ocean [Jacquet et al., 2008; 2015] and #DY032 cruise (2015, *R/V* DISCOVERY) at the PAP (Porcupine Abyssal Plain) observatory in the northeast Atlantic (49°N, 16.5 °W) (personal data). Results at the ANTARES / EMSO-LO site follow the trend previously reported in the Southern Ocean, indicating higher DWA Ba_{xs} in situations where a significant part of column-integrated PHP is located deeper in the water column (high Int. PHPx1/IntPHPx2 ratio; Figure 2b). These previous studies revealed that the shape of the column-integrated PHP profile (i.e. the attenuation gradient) is important in setting the Ba_{xs} signal in the mesopelagic zone (Dehairs et al., 2008; Jacquet et al., 2008, 2015]. Indeed, mesopelagic Ba_{xs} appears reduced when most of the column-integrated PHP is limited to the upper layer (indicating an efficient remineralization in surface), compared to situations where a significant part of integrated PHP is located deeper in the water column (reflecting significant deep PHP activity, POC export and subsequent remineralization) (Figure 2b). Our MedSea results are located along the trend defined in the Southern Ocean during KEOPS1 cruise. It is generally considered that Ba_{xs} (barite) forms inside sulfate and/or barium oversaturated biogenic micro-environments during POC degradation by heterotrophic prokaryotes. However, it is unclear whether barite formation at mesopelagic depths is (directly or indirectly) bacterially induced or bacterially influenced. Overall, our results strengthen the close link between the water column Ba_{xs} distribution and respiration (organic matter degradation).

3.3 Oxygen- barium relationship

The relationship we obtained at ANTARES between Ba_{xs} concentrations and oxygen consumption rates from optodes measurements (JO_2-Opt) is reported in Figure 3a. JO_2-Opt range from 0.11 to 5.85 µmol L^{-1} d^{-1}. The relationship indicates higher Ba_{xs} concentrations with increasing JO_2-Opt. An interesting feature is the intercept at zero JO_2-Opt (around 128 pM)

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which further supports the Ba BKG value at ANTARES (130 pM) determined from measured Ba_{xs} profiles (Figure 3a).

We applied a similar approach as reported in Jacquet et al. (2015) where we show the correlation between JO_2 obtained from dark community respiration DCR (winkler titration; JO_2-DCR) data integration in the water column and JO_2 based on Ba_{xs} content (Dehairs's transfer function; later referred to as JO_2-Ba). Similarly, to estimate JO_2-Ba in the present study we used the following equation [Dehairs et al., 1997]:

$$JO_2\text{-Ba} = (Ba_{xs} - Ba\ BKG)/17450 \quad (1)$$

A Ba BKG value of 130 pM was used (see above). JO_2-Ba is confronted to JO_2-Opt integrated over the same layer depth (between 175 and 450 m; Figure 3b). JO_2 rates are of the same order of magnitude (JO_2-Ba= 4.59 mmol m^{-2} d^{-1} and JO_2-opt= 3.14 mmol m^{-2} d^{-1}). The slight difference could be explained by the integration time of both methods: few hours to days for the incubations vs. few days to weeks for Ba_{xs} (seasonal build-up; Jacquet et al., 2007). JO_2 rates calculated in the present work are 3 times higher than those reported in the Southern Ocean during KEOPS1 [Jacquet et al., 2015] but they are in good agreement with the Ba_{xs} vs JO_2 trend (Figure 3b). Overall, our results indicate similar Ba_{xs} - JO_2 relationship in the Southern Ocean and the Mediterranean Sea. This further supports the universal validity of the Dehairs's transfer function in the present study.

3.4 Estimated particles remineralisation rates and implications

In order to provide a Ba_{xs}-derived estimate of POC remineralization rate (MR) at the ANTARES / EMSO-LO observatory during BATMAN cruise, we converted JO_2-Ba into C respired using the Redfield (RR) C/O_2 molar ratio (127/175; Broecker et al., 1985) multiplied by the depth layer considered (Z) [Dehairs et al., 1997]:

$$MR = Z \times JO_2\text{-}Ba \times RR \quad (2)$$

We obtain a POC remineralization rate of 11 mmol C m^{-2} d^{-1} (10% RSD). This is within the range of previously published remineralization fluxes in the Mediterranean Sea from sediment trap [Sanchez-Vidal et al., 2005] and from thorium-derived data [Speicher et al., 2006]. It is also in good agreement with recent POC flux attenuation combining drifting sediment traps and underwater vision profilers [Ramondenc et al., 2016].

The present paper brings a first insight into the connections of Ba_{xs}, PHP and JO_2 at the ANTARES/EMSO-LO observatory site in the northwestern Mediterranean Sea during the BATMAN (2016) cruise. Our results reveal a strong relationship between Ba_{xs} contents and measured JO_2 rates. Also, DWA Ba_{xs} vs. column integrated PHP, as well as measured vs. Ba_{xs}-based JO_2 relationships follow trends previously reported in the Southern Ocean where the Dehairs's function was first established to estimate POC remineralisation rate. Results from the present study would indicate that this function can also be applied in the Mediterranean basin provided that adequate Ba_{xs} background values are estimated. From a global climate perspective, the Ba_{xs} tool will help to better balance the MedSea water column C budget. It will contribute to gain focus on the emerging picture of the C transfer efficiency (strength of the biological pump).

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Figure captions

Figure 1: (a) Schematic representation of the convergence of the different estimators of oxygen consumption and C remineralization rates from the "oxygen dynamics", "barium proxy" and "prokaryotic activity" tools; (b) Location of the BATMAN cruise at the ANTARES observatory site in the NW-Mediterranean Sea (42°48'N, 6°10'E); (b) Potential temperature - salinity - depth plots and isopycals for BATMAN profiles. SW : Surface Water, WIW : Winter Intermediate Water, LIW : Levantine Intermediate Water, DMW : Deep Mediterranean Water. Graph constructed using Ocean Data View (Schlitzer, 2002; Ocean Data View; http://www.awi-bremerhaven.de/GEO/ODV)

Figure 2: (a) Particulate biogenic Ba_{xs} (pM) and particulate Al (nM) profiles next to the biogenic Ba fraction (%) in the upper 1000 m at ANTARES. The grey area represents a biogenic Ba fraction larger than 80 %. BKG: Ba_{xs} background. Ba_{xs} profile (pM) at DYFAMED : data from Sternberg et al. (2008); (b) ANTARES ratio plot (green square) of 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. Regression of the same ratio is reported for KEOPS1 (out plateau stations) and KEOPS2 (Southern Ocean; Jacquet et al., 2015) and #DY032 (PAP station, NE-Atlantic; pers. data) cruises.

Figure 3: (a) Relationship between Ba_{xs} concentrations (pM) and oxygen consumption rates (µmol L^{-1} d^{-1}) from optodes measurements (JO_2-Opt) at ANTARES; (b) Confrontation of oxygen consumption rates (mmol m^{-2} d^{-1}) obtained from different methods: optodes measurements (this work), dark community respiration DCR (winkler titration; JO_2-DCR; Jacquet et al., 2015;

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KEOPS1), and (Dehairs's transfer function calculation based on Ba_{xs} content (Dehairs et al., 1997).

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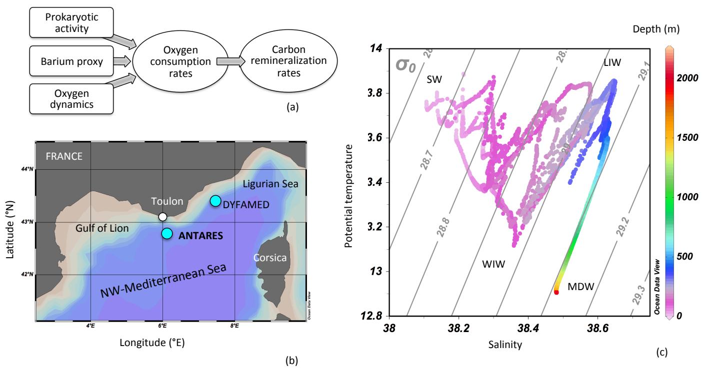
Figure 1



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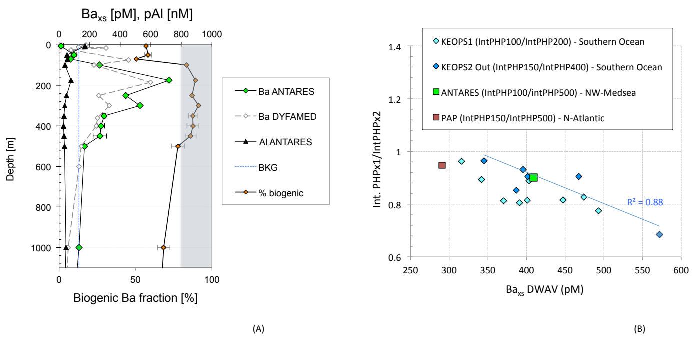

Figure 2

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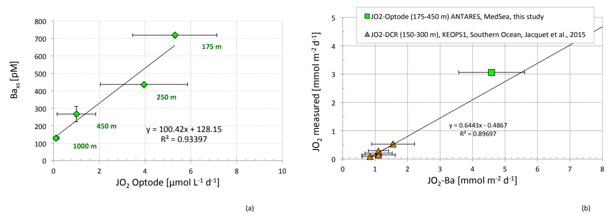

Figure 3