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Short communication

Evidence of intensified biogenic silica recycling in the Black Sea after 1970

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ABSTRACT

The Black Sea has been subject to increased levels of nitrogen and phosphorus loading and a decrease in silicate input after around 1970. Changes in phytoplankton community composition from diatoms to non-diatom groups have been attributed to the decrease in silicate. However, a discrepancy between the decreasing silicate input and the increasing silicate pool in the deep sea reported elsewhere implies that another silicate source exists which challenges the current paradigm of widespread silicate limitation. In this study, we investigate changes in the dissolution state of siliceous protists over the last 140 years and show that siliceous protists became significantly more dissolved after the late 1960s indicating a reduction of the silicate pool preserved in the deep sea sediment. We hypothesize that the decline in the dissolution state is caused by increased recycling of biogenic silica in the water column due to an increased annual production driven by nitrogen enrichment.

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1. Introduction

The Black Sea has been subject to extensive human induced nutrient enrichment which has led to increased eutrophication with several effects being noted both in the coastal and open ocean ecosystems (Mee, 1992; Oguz, 2005a; Yunev et al., 2005). However, while concentrations of dissolved inorganic nitrogen (N) and phosphate (P) have increased in the entire basin, dissolved silicate (DSi) has shown a decrease after the 1970s (Cociasu et al., 1996; Konovalov and Murray, 2001) (Fig. 1A, B).

Humborg et al. (1997) reported a shift in the dominating bloom forming group after 1970 from diatoms to non-diatom species, and argued that extreme change in the DSi:N ratio (from 42 to 2.8) was the primary forcing factor. Other researchers have drawn similar conclusions on the effects of decreasing DSi concentrations; e.g. changes in the phytoplankton community composition towards non-siliceous species (Bodeanu, 1993), increasing dinoflagellate diversity vs. diatom diversity (Eker-Develi and Kideys, 2003) and changes in the diatom community composition from large to small diatoms (Mousing et al., 2013). These patterns have led to the current understanding that the productivity of silicate utilizing protists became severely DSi limited after 1970.

However, even though the decreasing DSi concentration in the upper layers of the Black Sea correlates with the reduced discharge of DSi, a discrepancy exists. Konovalov et al. (1999) and Konovalov and Murray, (2001) reported a two-fold increase in the DSi concentration in the deep anoxic waters (Fig. 1B). This increase was surprisingly high relative to the riverine discharge reported by Humborg et al. (1997) and was also surprising as a decrease in the surface DSi concentration should lead to a decreased downward flux of DSi and biogenic silica (BSi). This discrepancy raises several questions relating to the underlying mechanisms driving the increased DSi concentration in the deep layer and to the apparently missing DSi source in an environment where DSi input, in general, was decreasing.

The primary source of DSi in aquatic systems is terrestrially derived from weathering of silica-containing minerals transported to the oceans via rivers (Tréguer and De La Rocha, 2013). However, recent studies suggest that BSi recycling as a significant DSi source should also be considered (Ragueneau et al., 2010). In the Black Sea, Ragueneau et al. (2002) investigated biogeochemical transformations of the major inorganic nutrients on the north-western shelf and found evidence of intense recycling of BSi with

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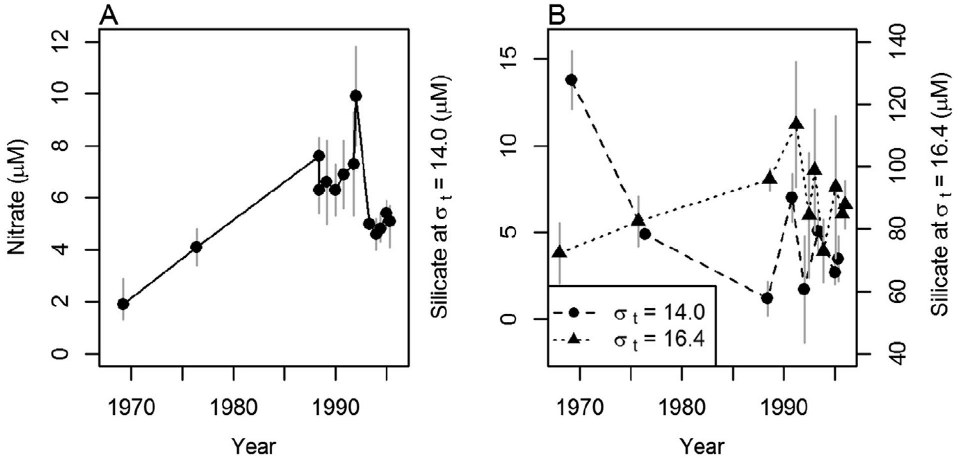

Fig. 1. Temporal patterns in the concentrations of nitrate in the upper ocean (A) and silicate at the surface (at $\sigma_t = 14.0$) and the deep sea (immediately below the permanent pycnocline at $\sigma_t = 16.4$) (B). Data are redrawn from Konovalov and Murray (2001) and represents average concentrations measured at 14 cruises in the period 1969 to 1995.

dissolution taking place both in the water column and in the sediments. Other studies have shown a similar importance of BSi recycling when other sources were limited (Adjou et al., 2011; Beucher et al., 2004).

Konovalov and Murray (2001) found an increase in the sulfide concentration in the deep Black Sea after around 1970 indicating increased primary productivity and downward flux of particulate organic matter (POM). The concomitant increase in DSi the deep sea suggested increased sinking of BSi in the form of diatom-generated POM. This would, however, imply an activation of the silicate utilizing protists rather than limitation after 1970. Although part of DSi could originate from coastal shelf sediments (Konovalov and Murray, 2001; Friedl et al., 1998), a decreasing DSi concentration in the open euphotic part of the Black Sea would also lead to increased dissolution and potential recycling of BSi in the water column. It is thus possible that some of the DSi in the deep sea could originate from the water column because a larger percentage of BSi did not reach the sea floor.

Thus, in this study we hypothesize that the increase in DSi concentration in the deep layers of the Black Sea originates from increased dissolution of sinking BSi, and to investigate this we examine the dissolution state of siliceous protists in two sediment cores from the southern open Black Sea.

2. Materials and methods

Two sediment cores (22-MUC-1 and 25-MUC-1) were collected with a multiple corer in the southern Black Sea on board the RV Meteor (cruise M72-3b) in March 2007 (Fig. 1 in Mousing et al., 2013). The uppermost 10 cm of the cores were removed, cut into samples of 0.5 cm width (depth), and freeze dried for 96 h (Heto FD5-66). The core was dated using ^{210}Pb-dating and CRS-modeling (Constant Rate of Supply) (Appleby, 2001).

Permanent slides preserving the siliceous protists (microfossils) were prepared following the method of Renberg (1990) and enumerated using phase contrast at 1000x on an Olympus BH-2 light microscope. Changes in the species abundance and composition as well as a detailed description of the methodology are described in Mousing et al. (2013).

While enumerating, microfossils were categorized as being either ‘pristine’ (showing no signs of dissolution; Fig. 2A, C) or ‘dissolute’ (showing signs of dissolution; Fig. 2B, D). At least 200 microfossils were evaluated in each sample resulting in more than 2400 individual assessment of dissolution state. From these assessments, the dissolution state of each sample was estimated by calculating the Dissolution Index (F_i) (Flower and Likhoshway, 1993; Ryves et al., 2001) following Equation (1):

$$F_i = \sum_j^m x_{ij} \bigg/ \sum_j^m X_{ij}, \tag{1}$$

where x_{ij} is the count of pristine microfossils of species j (of m) in sample i and X_{ij} is the total count of microfossils of species j (of m) in sample i. The equation produces a number between 1 and 0 where 1 describes a perfectly preserved sample without any signs of dissolution and 0 describes a very badly preserved sample where all observed microfossils show signs of dissolution.

Ryves et al. (2001) found that F_i was strongly correlated to the percentage of dissolved biogenic silicate (DBSi%) in relation to the original content (i.e. before the beginning of the dissolution experiment). Thus, F_i can provide insight into BSi recycling. In order to quantify the temporal changes in DBSi% we first modeled F_i as a function of time (year) using a Generalized Additive Model (GAM: Wood, 2006). Using this model, we calculated yearly values of F_i on which we applied the empirical relationship suggested by Ryves et al. (2001) following Equation (2):

$$DBSi\% = (0.97 - F_i)*100\%; (F_i < 0.97) \tag{2}$$

3. Results and discussion

The dissolution index (F_i) shows the same temporal pattern in both cores (Fig. 3A): From the 1860s to around 1950 there is a gradual increase in F_i from ca. 0 to 0.5. After the late 1960s, however, the pattern is reversed showing a decrease in F_i until around 2000. Together, the development in F_i in both cores clearly reflects a shift in the dissolution state of the siliceous protists from general improvement through time before the 1960 to degradation after the 1960s. By calculating the temporal changes in percentage dissolved BSi (DBSi%) (Fig. 3B) we estimate the relative change in the BSi pool preserved in the sediment. The pattern shows a decrease in the DBSi% from ca. 95 %–50 % in the period prior to around 1950 indicating a rapid increase BSi preserved in the sediment. After the late 1960s, however, the DBSi% increases indicating a decrease in the BSi inventory in the sediment. This decrease in the BSi

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