

Dear Dr Truede,

Thanks for your comments. We have carefully considered your comments and reviewers comments and replied to every comment individually. This should make it quite easy to follow our replies and associated changes in the manuscript.

Best regards,

Carlos

Associate Editor Decision: Reconsider after major revisions (19 May 2015) by Tina Treude

Comments to the Author:

Dear Dr. Sanz-Lazaro and colleagues,

thank you very much for submitting your revised manuscript to BG. I apologize for the long time it took to get the second round of reviews back to you, but I had to contact almost 40 scientists to find 3 available reviewers.

As you can see from the reports, the reviews are diverse (from conditional accept to major revisions). For your second revision I would like you to concentrate on Report #1 and #2. The revised manuscript will be sent back to 1-2 reviewers for the final round of review. After this point, I will accept only minor revisions.

Please do provide concentration vs. time data in the supplementary. These data were requested by one of your first reviewers and I agree that they are useful. They should be easy for you to provide.

REPLY: We are unsure about this request. We show the fluxes based on concentration changes vs. time in Fig. 2 and this is usually the level of detail presented in this type of studies. As an example we have included all the raw data needed to calculate the nutrient fluxes shown in Fig. 2 for the treatment at 16°C – OM (see supplementary material in the file: bg-2014-572-supplement-Nutrient integration 16-OM.zip). Same procedure was done for all the other treatments. If you find it relevant we can provide the data for the rest of the treatments, but we find the information to be so detailed and too much for a potential reader of the paper.

Because one of the reviewers had trouble accessing the San-Lazaro 2011 paper (which could be the case for other readers as well), I highly recommend that in your present study non-standard methods are introduced in sufficient detail so that readers are able to understand the setup without studying the 2011 paper.

REPLY: All methods we been described with enough detail so readers could be able to repeat the experiment. We have added the analytical procedure of TCO₂ analysis, since we have added this data in a Fig. 1 as suggested by a referee.

REPLY TO REFEREE 1:

The manuscript describes an experiment on metabolic enhancement in sediments and nutrient efflux to the water column by temperature increase and organic enrichment. The experiment involved a blank set of cores (no addition) and a set of enriched cores, that were incubated at 3 different temperatures. As can be expected, mineralization rates and nutrient efflux were enhanced by temperature and organic addition.

The manuscript is written in correct English. I did not like the large amount of numbers in the text. Generally, data should be in figures or tables, or in the text, but not in both. Of course it is the best to have the data in figures. The text should then guide the reader through the data, by pointing to trends and remarkable data.

REPLY: In the Material & Methods section, we have deleted all the data that is shown in figures and tables from the text. We have also removed redundant numbers from the text in the results section. We have followed the referee guidelines, aiming to guide the reader and highlighting the most important data in the manuscript.

The Figures could use improvement. Figure 1 is not needed, the data in the text indicate clearly that Fe(III) pools are variable and so large (ca 30 mmol/L sediment) that they cannot be influenced by the treatments. Instead, I would have liked to see the mineralization rates, which are now hidden in a line in the method section (171-173).

REPLY: We have deleted Fig. 1 and now this figure is the mineralization rates. In the M&M, we have explained how the TCO₂ fluxes were calculated and we have taken out the text related with the trends of the new figure to avoid repetition.

Figures 2 should be larger.

REPLY: Done

In Figure 3 we see two trends: a linear increase of the P efflux with T, N efflux seems to increase faster than linear. The exponential trajectory in Fig. 3b is an overinterpretation. Just connect the points and state in the text that it increases non-linearly.

REPLY: The regression corresponding to the trajectory in Fig. 3b has been selected with a standard criteria that it is used to fit trajectories, this is relying on AICc. We think that the figure is better explained by showing the regression. Otherwise it would not be consistent to show the regression on Fig. 3a and not in Fig. 3b because the best fit of the trajectory is non-linear. It is a general procedure in scientific papers to plot the regression in the graph as long as it is significant. Additionally, we think that the interpretation of the figure (such as it increases linearly or non-linearly) should be written in the text, not in the legend. So we have done this in the text. Anyway, we understand the point of the referee that it is better to say "non-linear" than "exponential". We have changed it throughout the text, only leaving it when it referred to the best fit found according to AICc in the result section.

The fate of the worms is not mentioned, yet is important. Did they survive the whole incubation? Did you see them pump? Frequency? The experiments where fishfood was added will have had much more active and well fed worms. Addition of organics enhances the activity of worms, hence the aeration of the sediments.

REPLY: We have added in the M&M: "Unfortunately irrigation rates and visual observations showed that the added worms died in three +OM cores at 26 degrees, and results from these cores were omitted from further analysis. For the rest of the cores no polychaetes were found dead during the experiment, and active *N. diversicolor* were observed in all the rest of the cores. The bromide incubations indicated that *N. diversicolor* ventilated their burrows with the same intensity regardless of OM-enrichment level".

The introduction is excellent, but it does not lead to a hypothesis, nor an explicit statement on why these measurements needed to be done. Yet this is implicit in there. It is a very good overview of the complexity of the matter. A very complicated set of processes is influencing nutrient release, and the outcome cannot be easily predicted, so must be measured. A text along these lines would be enough to explain why this study needed to be done.

REPLY: We have added before the aim: "Thus, to improve our forecasting capacity related to climate change, estimates of temperature driven changes in sediment nutrient release are needed."

The complexity of the sediment processes is not reflected in the experimental set up. As both phosphate and inorganic N are divided over several pools, and DIN is further subjected to various process steps, it is hard to link production rates to efflux. Phosphate is partially bound to different fractions of iron-oxides and organics and for a small part dissolved. Phosphate release is stimulated by Fe(III) reduction and inhibited by Fe oxidation and both take place. In the enrichment experiment, Fe reduction will be stimulated due to higher e-donor supply, and Fe oxidation will be stimulated due to higher worm activity, which process wins is not a priori clear. Therefore good that this is measured, from the data it appears that Fe reduction and P release is stimulated but that most P remains stuck. It should be mentioned that the P binding capacity is not easily exhausted, as there is tens of mmoles of Fe(III) per liter of sediment. It is thus not surprising that only a fraction of the phosphate is released. Mineralization produces ammonium, which is also strongly bound to sediments, and is converted to NO_x and finally N₂. Also nitrification will be stimulated by worm activity, which is again stimulated by feeding.

REPLY: We agree with the referee in all their comments, only, bioirrigation rates were similar despite the organic enrichment level (see Sanz-Lázaro et al 2011). We have added in the 3rd paragraph of the discussion: "In this experiment, the PO₄³⁻ binding capacity was not easily exhausted due to the large pool of Fe (III) (tens of mmoles) in the first cm of the sediment.". Sanz-Lazaro, C., Valdemarsen T., Marin A., and Holmer M.: Effect of temperature on biogeochemistry of marine organic-enriched systems: implications in a global warming scenario, Ecol. Appl., 21, 2664-2677, 2011.

The efflux of the measured nutrients is therefore determined by too many processes (both enhancing and inhibiting effluxes) to make a mechanistic interpretation. Also, the efflux rates cannot be directly, so linearly, linked to mineralization rates. Efflux of DIN in whatever form is ecologically relevant, therefore, beside ammonium also NO_x should have been measured.

REPLY: We agree with the referee, but we found that the data presented here has enough relevance. We previously wrote this in the discussion related with this issue: "Despite so, NH₄⁺ is mostly the dominating form of dissolved inorganic N effluxing from organic enriched sediments (Christensen et al. 2000; Holmer et al. 2003), while coupled nitrification-denitrification rarely exceeds 1-2 mmol m⁻² d⁻¹ in marine sediments (Middelburg et al. 1996). We are therefore confident that the NH₄⁺ release rates can act as a proxy for total inorganic N release to the water column."

The manuscript can thus be much shorter. Most of the numbers should be removed from the text and left in tables and figures. Also the discussion should be focused. Higher T and sediment enrichment significantly increase mineralization rates and much less stimulate nutrient efflux. The processes stimulating and inhibiting the effluxes of the different nutrients can shortly be reviewed. Much more cannot be said.

REPLY: We have reduced the text accordingly. See the track changes.

Finally, the title should be changed. The authors did not study climate change. Climate change involves storm frequencies, precipitation patterns, sealevel rise, a bit of temperature (much less than the current yearly variation). They only studied the effect of temperature and nutrients, so suggested: Effects of temperature and organic pollution on nutrient cycling in marine sediments

REPLY: Done. Additionally, We have added "climate change" as a keyword.

REPLY TO REFEREE 2:

Review of:

Effects of global climate change and organic pollution on nutrient cycling in marine sediments

By Sanz-Lazaro, Valdemarsen and Holmer

I have read both the original and revised manuscripts as well as author responses to previous reviewer comments. The paper provides new evidence on the potential effects of seafloor warming on benthic N and P fluxes. The results do come out as expected from previous studies and theoretical considerations, but these findings are still valuable. While the most recent version of the paper is an improvement over the original one, I think the manuscript can benefit further revisions along the lines outlined below:

A general concern of mine is to what extent are the authors confident that the measured TCO₂ originated from added labile OM? Two other mechanisms can be induced or accelerated by the imposed experimental conditions and can contribute to TCO₂ release:

REPLY: TCO₂ measurements are a broadly used surrogate of oxidation/mineralization rates in sediment biogeochemistry experiments.

1- Calcite dissolution induced by decreasing pH in the sediment column. Were there any pH measurements?

REPLY: We do not have pH measurements, but do not think significant calcite dissolution was likely to occur since the sediment used for the experiment was carbonate poor and overlying water was periodically changed. We have been more precise in the M&M to avoid confusion and we have added “every week” in the paragraph before the “Nutrient fluxes” subsection, and so the sentence now is: “During the experiment the water in each tank was kept aerated and changed every week.”.

2- TCO₂ originating from the oxidation of old, less labile or refractory carbon pools. There are a number of studies showing that addition of labile OM could also effect the mobilization of refractory organic carbon. Generally known as priming effect, this pathway could affect the TCO₂ release from the experimental substrates.

With the data at hand these processes are difficult to constrain but some discussion of these processes are needed and it may improve the interpretation of the data.

(If these processes were discussed in the companion paper – Lazaro et al 2011b – I cannot double check this since I do not have access to this paper.)

REPLY: We agree with the reviewer that the priming effect is relevant to consider in ecological studies (van Nugteren et al. 2009). However, we do not think having a priming effect is of great importance for the net-outcome of our research, where the main goal was to quantify the net effect of temperature change on total benthic metabolism. Having a priming effect would add some uncertainty to our budget considerations regarding the amount of added organic matter and nutrients being mineralized in +OM treatments, but would not impact the temperature trajectories of total mineralization.

van Nugteren, P., Moodley, L., Brummer, G. J., Heip, C. H., Herman, P. M., & Middelburg, J. J. (2009). Seafloor ecosystem functioning: the importance of organic matter priming. *Marine biology*, 156(11), 2277-2287.

A few other points:

L18: First sentence of abstract: difficult to read, please consider to re-organize.

REPLY: **Done**

L48: This is optional but I would prefer to read “oxidation” instead of mineralization.

REPLY: **We have used through the whole document mineralization, and we think there can be used any of both but we prefer to use “mineralization” and use the same Word throughout the Ms so to keep in consistent and avoid confusion.**

L50: No need for ‘mainly’. In the coastal sediments, all OM is microbially oxidized.

REPLY: **When Eukaryotic organisms feed they also oxidize organic matter, which is normally around 10-20 % of the total oxidation of the OM.**

L53: This sentence is confusing. Do the authors imply that iron is reduced during P adsorption (the use of ferric and then ferrous)? Or do they mean that adsorption to Fe minerals is one P sequestration pathway and the formation of Fe(II)-P minerals is another one?

REPLY: **We have rewritten the sentence to clarify these points: “PO₄³⁻ can be sequestered in the sediment by adsorption to ferric (oxy)hydroxides and by forming solid ferrous phosphates (Jorgensen 1983). Sulfides resulting from sulfate reduction may mobilize PO₄³⁻ by reducing ferric oxyhydroxides (Roden and Edmonds 1997) and by dissolving Fe minerals containing PO₄³⁻ (Gachter and Muller 2003)”.**

L59: I suggest the replacement of ‘reduced’ with ‘reducing’

REPLY: **Done**

L63: This sentence is problematic as NO₃, NO₂ and N₂ does not exit the biological cycle.

REPLY: **We have changed the sentence to: “In contrast to NH₄⁺, NO₃⁻ and NO₂⁻, N₂ can not be directly used by primary producers.”.**

L99: I think in this section a brief recap of the methods should still be given.

REPLY: **We have tried to do so.**

L324: I would replace “pools” with “forms”

REPLY: **Done**

REPLY TO REFEREE 3:

The above manuscript presents interesting and partly novel information on the likely effect of multiple (two) stressors, namely rising temperatures and OM enrichment, on benthic fluxes across the sediment/ water interface in shallow subtidal soft sediments. I share the concerns raised by previous reviewers, namely about the lack of nitrite and nitrate data, or nitrification rate measurements, and the lack of information on fluxes during light/ dark cycles, which limit the ability to unravel the ecological/ biogeochemical processes underlying the recorded trends. However, I also feel that the authors have done their best in addressing the reviewer's comments and acknowledging the limitations of their study in the manuscript.

REPLY: We have tried to do our best by adding the previous comments in the 1st revision, by adding the paragraph before the conclusion, and some sentences two paragraphs before.

I don't think further revision would significantly improve the manuscript and recommend to accept it for publication.

1 | Effects of temperature change and organic pollution on nutrient cycling in marine sediments

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4 | Carlos Sanz-Lázaro^{1,2*}, Thomas Valdemarsen¹ & Marianne Holmer¹

Con formato: Francés
(Francia)

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6 | ¹Department of Biology, University of Southern Denmark, Campusvej 55, 5230 Odense,
7 | Denmark

8 | ²Present address: Departamento de Ciencias del Mar y Biología Aplicada, Universidad de
9 | Alicante, P.O. Box 99, E-03080 Alicante, Spain.

10

11 | *Corresponding author:

12 | E-mail: carsanz@ua.es, carsanzla@gmail.com

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(Francia)

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Con formato: Francés
(Francia)

14 | Running title: Climate warming effect on nutrient cycling

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(Francia)

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17 Abstract

18 Increasing ocean temperature due to climate change is an important anthropogenic driver of
19 ecological change in coastal systems. In these systems, sediments play a major role in nutrient
20 cycling. Our ability to predict ecological consequences of climate change is enhanced by
21 simulating real scenarios. Based on predicted climate change scenarios, we tested the effect of
22 temperature and organic pollution on nutrient release from coastal sediments to the water column
23 in a mesocosm experiment. PO_4^{3-} release rates from sediments followed the same trends as
24 organic matter mineralization rates, and increased linearly with temperature and were
25 significantly higher under organic pollution than under non-polluted conditions. NH_4^+ release
26 only increased significantly when the temperature rise was above 6°C , and was significantly
27 higher in organic polluted compared to non-polluted sediments. Nutrient release to the water
28 column was only a fraction from the mineralized organic matter, suggesting PO_4^{3-} retention and
29 NH_4^+ oxidation in the sediment. Bioturbation and bioirrigation appeared to be key processes
30 responsible for this behaviour. Considering that the primary production of most marine basins is
31 N-limited, the excess release of NH_4^+ at a temperature rise > 6 degrees could enhance water
32 column primary productivity, which may lead to the deterioration of the environmental quality.
33 Climate change effects are expected to be accelerated in areas affected by organic pollution.

34

35 Keywords: climate change, eutrophication, global warming, nitrogen, organic matter enrichment,
36 phosphorus.

Eliminado: , where

Eliminado: especially when the interactions among drivers may not be just additive

Eliminado: of

Eliminado: , sea water temperature

37 1. Introduction

38 Ocean temperature rise due to climate change is considered among the most severe

39 anthropogenic drivers of ecological change in marine systems, especially in coastal areas

40 (Halpern et al. 2008). Sea surface temperature (SST) in marine systems is expected to rise 0.2°C

41 per decade (IPCC 2007), but in relatively enclosed and shallow coastal areas, such as the Baltic

42 Sea, temperature has risen much more rapidly (1°C in last decade; (Mackenzie and Schiedek

43 2007; Belkin 2009) and similar rises are expected for the next decades (Doscher and Meier 2004).

44 In these areas, heterotrophic processes in sediments prevail due to the generally low availability
45 of light in the seabed due to eutrophication and high input of labile organic matter (Conley et al.
46 2009).

47 Coastal areas are climate-sensitive systems (IPCC 2007) that play an important role in the

48 mineralization of organic matter (OM) (Middelburg et al. 1997). During mineralization, organic

49 phosphorous (P) and nitrogen (N) are transformed into inorganic forms (PO_4^{3-} and NH_4^+ ,

50 respectively). OM is mineralized mainly by microbial processes, which are strongly influenced

51 by temperature (Robador et al. 2009). Consequently, climate change may affect nutrient

52 regeneration rates in coastal ecosystems.

53 PO_4^{3-} can be sequestered in the sediment by adsorption to ferric (oxy)hydroxides and by
54 forming solid ferrous phosphates (Jorgensen 1983). Sulfides resulting from sulfate reduction may

55 mobilize PO_4^{3-} by reducing ferric oxyhydroxides (Roden and Edmonds 1997) and by dissolving

56 Fe minerals containing PO_4^{3-} (Gachter and Muller 2003). Despite the complex interactions

57 among the Fe, S and P pools, PO_4^{3-} release to the water column is mainly dependent on the redox

58 conditions in the sediment surface, where oxidized Fe keep PO_4^{3-} immobilized. Thus seasonal

59 increases in sediment metabolic rates due to temperature rise may lead to reducing conditions in

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60 the sediment surface, resulting in PO_4^{3-} release during summer (Jensen et al. 1995; Cowan and
61 Boynton 1996). Mineralized N is mainly released to the water column as NH_4^+ (Jorgensen 1983).
62 NH_4^+ can be further transformed into NO_3^- and NO_2^- , through nitrification, and end up as N_2
63 through denitrification or anammox (Thamdrup 2012). In contrast to NH_4^+ , NO_3^- and NO_2^- , N_2
64 can not be directly used by primary producers. Consequently, denitrification results in N removal
65 from the ecosystem preventing eutrophication (Thamdrup and Dalsgaard 2008). Marine coastal
66 sediments play a major role in supporting primary productivity of the water column by supplying
67 a large part of the nutrient demand for phytoplankton (Nixon 1981; Kelly et al. 1985). Increases
68 in sea water temperature and input of OM to the sediment, are both expected to stimulate the
69 release of inorganic nutrients from the sediment to the water column, which may in turn
70 stimulate primary productivity. Nutrient increase above a certain threshold may lead to algal
71 blooms and subsequent hypoxic/anoxic events, resulting in the deterioration of ecological status
72 (Gray et al. 2002).

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73 Coastal areas gather the greatest human population densities in the world, resulting in
74 high anthropogenic pressure on coastal ecosystems. Thus, many coastal areas are subject to
75 multiple stressors (Halpern et al. 2008) such as different kinds of pollution and global warming.
76 Among the different types of pollution, organic pollution, also know as organic enrichment, is
77 one of the most common in densely populated coastal areas (Islam and Tanaka 2004). As for
78 temperature, organic enrichment enhances sediment metabolism, mainly by stimulating
79 anaerobic pathways and especially sulphate reduction (Valdemarsen et al. 2009). This effect is
80 magnified when organic pollution and temperature rise co-occur (Sanz-Lazaro et al. 2011b).

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81 Our ability to predict the ecological consequences of climate change is enhanced by
82 simulating realistic future scenarios. Hence, it is important to conduct experiments to elucidate

83 not only the effects of individual drivers of ecological change but also the interactions among
84 them, especially when the interactions among these drivers are not just additive (Falkenberg et
85 al. 2013). Despite the current concern of global change, scarce research effort has been taken on
86 understanding how temperature rise affect the cycling of N and P in coastal sediments
87 (Alsterberg et al. 2012). In fact, the accuracy of predictive biogeochemical models related to the
88 effect of climate change on the coastal eutrophication is restrained due to limited knowledge
89 (Meier et al. 2011). Thus, to improve our forecasting capacity related to climate change,

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90 estimates of temperature driven changes in sediment nutrient release are needed.

91 The aim of this work is to examine the effects of temperature rise and organic enrichment
92 on sediment nutrient release derived from heterotrophic processes. We used a mesocosm
93 approach and hypothesized that temperature rise due to climate change increases PO_4^{3-} and NH_4^+
94 release from the sediments as a consequence of increased metabolic rates. We expected an
95 increase of the release of nutrients with temperature, with a more marked effect in organic
96 enriched than in non-organic enriched sediments, due to stimulation of anaerobic processes
97 resulting in PO_4^{3-} and NH_4^+ release.

98

99

100 2. Materials and methods

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102 This experiment is the second part of a study. The results on carbon and sulphur cycles are
103 presented in Sanz-Lázaro et al. (2011b), where detailed information on the methods it is given.

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105 2.1. Collection of sediment and polychaetes

106 The sediment used for experiments was well sorted, organic poor, Fe-rich sand [0.4 %DW
107 particulate organic carbon (POC), 125 $\mu\text{mol cm}^{-3}$ reactive Fe and 220 μm average grain size]
108 collected [at 1 m depth](#) in Fænø Sund, Denmark (for further details see [Valdemarsen et al. 2009](#)).

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109 The macrofaunal organism used for experiments was the polychaete *Nereis diversicolor*, which
110 was chosen based on its ability to bioirrigate surface sediment and influence microbial reaction
111 rates ([Kristensen 2000](#); [Mermillod-Blondin et al. 2004](#)). *N. diversicolor* was collected from
112 Fællesstrand in the outer part of Odense Fjord, Denmark.

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114 2.2. Experimental setup

115 Sediment was split into either control (-OM) or organically enriched sediment (+OM).

116 Enrichment was performed by adding 92 g labile OM [finely ground fish feed, Ecolife, Dansk
117 Ørredfoder with 49.4% DW POC, 8.1% DW total nitrogen organic N (TN), 0.9% DW total P
118 (TP)] to 20 L of sediment. The enrichment corresponded to 26 mol POC m^{-2} , which is
119 comparable to the annual OM deposition at fish or mussel farms ([Callier et al. 2006](#); [Holmer et al.](#)
120 [2007](#); [Sanz-Lazaro et al. 2011a](#)).

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121 Sediment was packed into 36 core liners (8 cm internal diameter; i.d., 35 cm sediment
122 depth) which were distributed in three tanks maintained at 16, 22 or 26°C containing 65 L GF/F-
123 filtered seawater from Fænø Sund with a salinity of 17 psu. Each tank contained 6 cores of each
124 -OM and +OM, sediment. The 16°C treatment was chosen as the reference level of present mean
125 SST for summer months in temperate areas of the Baltic Sea ([Kristensen 1993](#); [Holmer and](#)
126 [Kristensen 1996](#)). The 22°C and 26°C treatments were chosen as SST climate change scenarios
127 in 60 and 100 years based on SST temperature rise observed in the last decades ([Mackenzie and](#)

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128 Schiedek 2007;Belkin 2009) and expected raises (Doscher and Meier 2004) (1°C per decade in
129 both cases).

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130 The sediment in the cores was left 3 days to compact and acclimitize. Then three *N.*

131 *diversicolor* were added to each core to simulate the natural density (Delefosse et al. 2012). The
132 time of polychaete addition was assumed the beginning of the experiment (t = 0). *N. diversicolor*

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133 bioirrigation rates were previously reported in Sanz-Lázaro et al. (2011b). They were calculated

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134 based on Br⁻-addition experiments and were on average 13-24 L m⁻² d⁻¹ in the different

135 treatments. There were no significant (p < 0.05) effects of temperature or of OM enrichment on

136 bioirrigation rates and all cores were bioirrigated to 6–8 cm depth (Sanz-Lazaro et al. 2011b).

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137 Bioirrigation rates in the present experiment were similar to the bioirrigation measured

138 previously for other polychaetes (Quintana et al. 2013) and within the range of the bioirrigation

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139 performed by natural macrofaunal communities (Valdemarsen et al. 2010). Unfortunately

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140 irrigation rates and visual observations showed that the added worms died in three +OM cores at

141 26 degrees, and results from these cores were omitted from further analysis. For the rest of the

142 cores no polychaetes were found dead during the experiment, and active *N. diversicolor* were

143 observed in all the rest of the cores. The bromide incubations indicated that *N. diversicolor*

144 ventilated their burrows with the same intensity regardless of OM-enrichment level.

145 Additionally, six cores with 5 cm i.d. were also filled with –OM and +OM sediment to

146 determine initial element pools in the sediment. Cores were closed at the bottom with rubber

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147 stoppers and ~20 cm sediment was added, leaving a 10-12 cm headspace above the sediment.

148 During the experiment the water in each tank was kept aerated and changed every week. The

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149 cores were kept submerged and in darkness throughout the experiment (25-39 days). More

150 specific details can be found in Sanz-Lázaro et al. (2011b).

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152 2.3. Nutrient fluxes

153 NH_4^+ and PO_4^{3-} fluxes between sediment and water were measured every 2-4 days during the
154 first 2 weeks and every week during the rest of the experiment. During flux measurements, the
155 water column of each sediment core was sampled and cores were closed with rubber stoppers.
156 Incubations were ended after 3-5 h (-OM) or 1-2 h (+OM), where the rubber stoppers were
157 removed and the water column was sampled again. All samples were GF/F-filtered, transferred
158 to 20 mL plastic vials and frozen (-20°C).

159

160 2.4. Sectioning of cores

161 The three 5 cm i.d. sediment cores of each sediment type (-OM and +OM) were initially
162 sectioned ($t = 0$; henceforth referred to as initial cores) and the remaining cores from every
163 temperature treatment were sectioned at the end of the experiment (henceforth, final cores).

164 Cores were sectioned into 1 cm slices to 2 cm depth and into 2 cm slices to 16 cm depth. Every
165 sediment slice was homogenized and subsampled for different analysis. One subsample from
166 each sediment slice was used to measure reactive Fe and phosphate bound to reactive Fe. A
167 subsample of sediment was used for TP measurement. The remaining sediment from each slice
168 was used to determine TN, sediment density and water content. All analytical methods are
169 described below. Since temperature has a strong stimulatory effect on microbial reaction rates,
170 the duration of the temperature treatments varied to prevent porewater sulfate depletion and a
171 shift in sediment metabolism towards methanogenesis. Thus, the sectioning of the sediments at
172 16, 22 and 26°C was performed after 39, 32 and 25 days, respectively.

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174 2.5. Sediment metabolic rates

175 C mineralization rates were estimated as time-integrated total CO₂ (TCO₂) sediment-water
176 column flux divided by the experiment duration as described in Sanz-Lázaro *et al.* (2011b) (Fig.
177 1). TCO₂ fluxes were measured following same procedures as for nutrient fluxes and TCO₂
178 concentration was analyzed by flow injection analysis (Hall and Aller 1992).

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180 2.6. Analyses

181 NH₄⁺ and PO₄³⁻ were analyzed spectrophotometrically on a Lachat QuikChem 8500
182 autoanalyzer. Reactive Fe was extracted on ~0.2 g of sediment subsamples with 5 ml of 0.5 M
183 HCl shaken for 30 min. Then samples were centrifuged for 5 min (3000 rpm, ca. 1000 × g) and
184 GF/F filtered. The supernatant was analyzed by the ferrozine method (Stookey 1970). Fe (II) was
185 determined on the untreated extract while total Fe [Fe(II)+Fe(III)] was determined after
186 reduction with hydroxylamine (Lovley and Phillips 1987). Fe (III) was determined as the
187 difference between total Fe and Fe(II). PO₄³⁻ in HCl extracts was also analyzed
188 spectrophotometrically after addition of Molybdate reagent (Koroleff 1983) to get a measure of
189 Fe-bound P. Total P was determined on combusted sediment subsamples (520°C, 2 h), which
190 were boiled in 1 M HCl for 1 h. The supernatant was analyzed for PO₄³⁻ as described above. TN
191 was determined on a Carlo Erba CHN EA1108 elemental analyzer according to Kristensen and
192 Andersen (1987). Sediment density was determined gravimetrically by weighing a known
193 volume of sediment using cut off syringes. Water content was measured as weight loss after
194 drying (105°C, >12 h).

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196 2.7. Data analysis

197 Significant differences in the content of solid phase nutrients were tested by pair-wise t-tests.
198 Comparisons were done between initial -OM and +OM cores, and between initial and each final
199 treatment within each sediment type (-OM and +OM).

200 Average nutrient efflux rates were estimated as time-integrated nutrient effluxes divided
201 by the experiment duration. To compare the trends of the time-integrated effluxes between -OM
202 and +OM treatments along temperature we did regression models considering temperature the
203 continuous covariate and OM enrichment as a fixed factor. According to the data on the trends of
204 the time-integrated effluxes with increasing temperature we hypothesized different regression
205 models. We used the second order Akaike Information Criterion (AICc) to choose the best
206 model, since it is a good compromise between fitting and complexity of the model and it is
207 recommended when the sample size is relatively low (Burnham and Anderson 2002). In all
208 regressions temperature was centered at 26°C, so intercepts showed mean values of effluxes at
209 this temperature. The significance of the regression coefficients was tested by ANOVA.
210 Homocedasticity was checked using Levene's test and normality with p-p plots. Analyses were
211 run in R (v. 2.15.0) and linear regressions were implemented using the *lm* function (R
212 Development Core Team 2012). All data were reported as mean ± standard error (SE) and
213 statistical tests were conducted with a significance level of $\alpha = 0.05$.

214 We calculated the molar ratio of C:N:P in the added OM based on its composition and the
215 molar C:N:P ratio in the sediment based on POC, TN and TP content in unenriched and enriched
216 sediment. We estimated C:N:P ratios of organic matter being mineralized based on TCO₂, NH₄⁺
217 and PO₄³⁻ effluxes. C and TCO₂ effluxes were reported in Sanz-Lázaro *et al.* (2011b).

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220 3. RESULTS

221

222 3.1. P in sediment

223 TP concentration in initial -OM and +OM sediment showed similar values (Table 1). The initial
224 enrichment with fish feed should have resulted in 0.4 mol m^{-2} higher TP in +OM cores than -OM
225 cores (4.7 % of the TP in the sediment), but this was not evident from our measurements
226 probably due to high variability between subsamples. Hence, there were no detectable
227 differences in the TP content between initial -OM and +OM cores ($p>0.05$). Additionally, there
228 were no significant differences on TP content between the initial and each final treatment within
229 each sediment type (-OM and +OM). Fe-bound P at the beginning of the experiment also showed
230 similar values between -OM and +OM cores (Table 2).

Eliminado: was 50.8 ± 3.2 and $51.2 \pm 0.3 \mu\text{mol cm}^{-3}$, respectively. Depth-integrated (0-16 cm) TP thus amounted to 8.2 ± 0.5 and $8.2 \pm 0.1 \text{ mol m}^{-2}$ in initial -OM and +OM cores, respectively

Eliminado: Depth-integrated (0-16 cm)

Eliminado: corresponded to 2.5 ± 0.1 and $2.1 \pm 0.2 \text{ mol m}^{-2}$ in initial

Eliminado: sediment, respectively. By the end, Fe-bound P ranged between 2.1 ± 0.2 and $2.6 \pm 0.3 \text{ mmol m}^{-2}$ in -OM cores, and between 2.0 ± 0.2 and $2.6 \pm 0.3 \text{ mmol m}^{-2}$ in +OM cores

232 3.2. N in sediment

233 PON content in initial -OM cores was significantly lower ($p<0.05$) than +OM cores.
234 Additionally, there were no significant differences on PON content between the initial and each
235 final treatment within each sediment type (-OM and +OM). The initial enrichment with fish feed
236 should have resulted in extra 3.6 mol m^{-2} (46.8 % of the PON concentration in the sediment),
237 which was close to the measured enrichment (28.6 % of the PON in the sediment) (Table 1).

Eliminado: concentration

Eliminado: and

Eliminado: sediment was 47.5 ± 2.4 and $61.4 \pm 1.6 \mu\text{mol cm}^{-3}$, respectively. Thus depth-integrated (0-16 cm) PON was 7.7 ± 0.4 and $9.8 \pm 0.3 \text{ mol m}^{-2}$ in initial -OM and +OM cores, respectively (Table 1). The enrichment resulted in significantly higher PON in +OM cores ($p<0.05$).

239 3.3. Fe in sediment

240 Pools of reactive Fe (III) were very similar between initial -OM and +OM cores, as well as
241 between treatments at the end of the experiment (Table 2). Fe (III) concentration was high in the
242 first cm of the sediment being 32.9 ± 8.0 , 35.5 ± 4.3 and $25.1 \pm 5.6 \mu\text{mol cm}^{-3}$ in -OM cores at

Eliminado: Depth-integrated (0-16 cm) p

Eliminado: 2.5 ± 0.1 and $2.3 \pm 0.2 \text{ mol}$

Eliminado: m^{-2} in

Eliminado: sediment, respectively. After the experiment, the pools of reactive Fe (III) were 2.0 ± 0.3 , 1.8 ± 0.1 and $2.3 \pm 0.5 \text{ mmol m}^{-2}$ in -OM cores at 16, 22, and 26°C, respectively, and 1.9 ± 0.2 , 2.5 ± 0.3 and $2.3 \pm 0.5 \text{ mmol m}^{-2}$ in +OM cores at 16, 22, and 26°C

243 16, 22, and 26°C, respectively, and 12.7 ± 3.6 , 24.0 ± 6.9 and 28.5 ± 6.2 $\mu\text{mol cm}^{-3}$ in +OM cores
244 at 16, 22, and 26°C.

Eliminado: (Fig. 1)

246 3.4. Nutrient release

247 PO_4^{3-} efflux showed a similar temporal pattern in the different treatments: an increase in the first
248 days, a peak at intermediate stages and then decreasing effluxes towards the end (Fig. 2). The
249 peaks of PO_4^{3-} efflux were stimulated by OM enrichment and temperature. In -OM cores PO_4^{3-}
250 efflux ranged from -0.3 to 0.3, -0.4 to 0.8 and -0.2 to 0.7 $\text{mmol m}^{-2} \text{d}^{-1}$ at 16, 22 and 26°C,
251 respectively. In + OM cores PO_4^{3-} efflux ranged from -0.2 to 1.0, -0.2 to 1.9 and 0.0 to 1.4 mmol
252 $\text{m}^{-2} \text{d}^{-1}$ at 16, 22 and 26°C, respectively.

253 Average PO_4^{3-} efflux over the whole experiment was 1.7 ± 1.2 , 3.5 ± 1.7 and 4.3 ± 1.1
254 mmol m^{-2} in -OM cores at 16, 22, and 26°C, respectively. In +OM cores total PO_4^{3-} effluxes
255 were 6.6 ± 1.2 , 11.4 ± 2.0 and 13.2 ± 1.8 mmol m^{-2} in +OM cores at 16, 22, and 26°C,
256 respectively, corresponding to 4.2, 7.6 and 9.3 % of the total P added in fish feed (Table 1).
257 Average PO_4^{3-} efflux showed a linear increasing trend along temperature in both -OM and +OM
258 cores (Fig 3a). The AICc showed that PO_4^{3-} effluxes were best fitted with a linear regression
259 (Table A1). The regression analysis showed that average PO_4^{3-} efflux increased linearly with
260 temperature ($p < 0.05$) in -OM and +OM cores. There were significant differences between -OM
261 and +OM treatments at 26°C ($p < 0.05$) and the significant interaction term of the regression
262 ($p < 0.05$) indicated a steeper temperature response in +OM compared to -OM treatments (Table
263 3).

264 The overall trend of NH_4^+ -efflux was similar to PO_4^{3-} -efflux. NH_4^+ effluxes increased at
265 the beginning, peaked and then decreased towards the end (Fig. 2). The range of the NH_4^+

266 effluxes was stimulated by both organic enrichment and temperature. In -OM cores NH_4^+ efflux
267 ranged from -12.5 to 27.8, -20.7 to 35.8 and -16.2 to 54.6 $\text{mmol m}^{-2} \text{d}^{-1}$ at 16, 22 and 26°C,
268 respectively. In +OM cores NH_4^+ efflux ranged from 0.0 to 58.8, 0.0 to 60.0 and 3.1 to 55.6
269 $\text{mmol m}^{-2} \text{d}^{-1}$ at 16, 22 and 26°C, respectively.

270 Average NH_4^+ efflux over the whole experiment was 217 ± 48 , 112 ± 58 and 339 ± 106
271 mmol m^{-2} in -OM cores at 16, 22, and 26°C, respectively. Total NH_4^+ effluxes in +OM cores
272 were 559 ± 113 , 525 ± 102 and 577 ± 132 mmol m^{-2} at 16, 22, and 26°C, corresponding to 17.9,
273 17.4 and 20.1 % of N added in fish feed (Table 1). The trend of average NH_4^+ -effluxes over the
274 whole experiment was not as clear as for PO_4^{3-} effluxes (Fig. 3b). NH_4^+ effluxes were clearly
275 stimulated at 26°C in both sediment types, however. According to the AICc, the trend of NH_4^+
276 efflux over temperature was best fitted with an exponential regression (Table A1). The
277 regression analysis showed significant increasing NH_4^+ effluxes with temperature following a
278 non-linear trajectory ($p < 0.05$) in both -OM and +OM sediment. There were significant
279 differences between -OM and +OM treatments at 26°C ($p < 0.05$). The interaction term of the
280 regression did not show significant differences. Thus, NH_4^+ efflux in -OM and +OM cores
281 followed a similar trend with temperature although at different ranges (Table 3).

Eliminado: n

Eliminado: exponential

282 283 3.5. C:N:P ratios

284 The C:N, C:P and N:P ratios of the added OM were 5.2, 21.3 and 4.1, respectively. C:N, C:P and
285 N:P ratios were similar in the initial -OM and +OM cores. The ratios among C, N and P at the
286 end of the experiment were calculated based on CO_2 , NH_4^+ and PO_4^{3-} effluxes. C:N, C:P and N:P
287 ratios at the end of the experiment were generally greater in -OM than in +OM cores, although
288 there was not a clear trend along temperature treatments (Table 4).

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Eliminado: sediment

Eliminado: were 12.7 and 11.6, respectively.

Eliminado: I

Eliminado: C:N, C:P and N:P ranged 13.3 - 25.3, 656 - 988 and 25.9 - 65.3, respectively, while in +OM cores, C:N, C:P and N:P ranged 21.6 - 26.8, 1157 - 1866 and 43.7 - 86.5, respectively

289

290

291 4. Discussion

292 Our results show that temperature rise resulted in different trends of sediment
 293 nutrient release of PO_4^{3-} and NH_4^+ under both organic and non-organic enrichment conditions.
 294 While sediment PO_4^{3-} release followed a linear trend with increasing temperature, the NH_4^+
 295 release from sediment show ~~non-linear~~ trends, notably increasing when temperature increments
 296 were above 6°C.

Eliminado: exponential

297 4.1. PO_4^{3-} release

298 The measured PO_4^{3-} effluxes at 16°C were within the range of effluxes measured in coastal
 299 sediments in the Baltic Sea (Sundby et al. 1992; Jensen et al. 1995). PO_4^{3-} effluxes increased
 300 linearly with temperature in both -OM and +OM treatments, but the slope of the regression was
 301 significantly steeper in +OM cores compared to -OM cores. This suggests that the temperature
 302 dependent increase in PO_4^{3-} effluxes is enhanced by OM enrichment. The temperature effect on
 303 PO_4^{3-} release was comparable to the temperature effect on total C-metabolism in both organic
 304 enriched and non-organic enriched conditions (Sanz-Lazaro et al. 2011b). However, the release
 305 of PO_4^{3-} was much lower than that of TCO_2 , as indicated by high C:P ratios, suggesting that
 306 organic P was either less labile compared to organic C or that inorganic PO_4^{3-} was retained in the
 307 sediment to a larger extent than C. Taking into account that the added OM (fish feed) is highly
 308 labile, our data suggest PO_4^{3-} retention in the sediment, which could be associated with
 309 adsorption to oxidized forms of Fe (Jensen et al. 1995).

Eliminado: natural

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Eliminado: Thus, a 10°C rise lead to 4.3 and 3.4 times stimulated PO_4^{3-} -efflux of in -OM and +OM cores, respectively, while this temperature rise stimulated TCO_2 -efflux 3.0 and 2.2 times in -OM and +OM cores, respectively. This indicated that PO_4^{3-} release occurred in proportion to total sediment metabolism.

Eliminado: above 600 in all treatments

Eliminado: This was far below the C:P ratio in the sediment (11.6 and 13.5 for -OM and +OM treatments, respectively) and the C:P ratio in the added OM (21.3),

Eliminado: For each temperature, we estimated the mineralized OM out of the OM added OM by subtracting the TCO_2 production in the +OM treatments to the TCO_2 production in the -OM treatments. We found that 55-77% of the added OM was mineralized, whereas P efflux only corresponded to 4.2-9.3 % of the added P (Table 2).

Eliminado: OM

Eliminado: ,

Eliminado: ,

Código de campo cambiado

Eliminado: pools

Código de campo cambiado

310 P-retention in marine sediments is controlled by the forms of P, Fe and S in the sediment,
 311 interactions between these pools and the sediment redox-conditions (Roden and Edmonds

1997;Gachter and Muller 2003). Generally, in sediments with an oxic surface layer, oxidized Fe acts as a lid for PO_4^{3-} , which is adsorbed to ferric Fe preventing its release to the water column (Rozan et al. 2002). In this experiment, the PO_4^{3-} binding capacity was not easily exhausted due to the large pool of Fe (III) (tens of mmoles) in the first cm of the sediment. This hypothesis was also supported by the fact that the levels of PO_4^{3-} bound to Fe (III) showed low variation between initial and final cores. The oxic conditions of the sediment surface were maintained in all treatments despite metabolism enhancement and subsequent increase in sulphide production, which was especially notable in the +OM treatments. The continuous oxygenation of surface sediment due to *N. diversicolor* bioturbation and bioirrigation (Sanz-Lazaro et al. 2011b) was probably critical for maintaining an oxic sediment surface with high Fe(III) concentrations, since dramatically stimulated sediment metabolism are expected to notably diminish the Fe(III) pool of the sediment in the absence of macrofauna (Valdemarsen et al. 2009). All these results suggest that the capacity of the sediment to retain PO_4^{3-} was maintained despite high metabolic rates due to organic enrichment and elevated temperatures.

Eliminado: mobilization

Código de campo cambiado

Eliminado: All the treatments maintained a considerable pool of oxidized Fe(III) (1.8 to 2.5 mol m⁻²), in the same range as measured at the beginning of the experiment (2.3 to 2.5 mol m⁻²) indicating that the PO_4^{3-} retention capacity of the sediment was maintained despite elevated temperature and organic enrichment. Similarly,

Código de campo cambiado

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The PO_4^{3-} retention may be diminished in sediments with a low pool of Fe(III) (Rozan et al. 2002) and/or with no bioturbating macrofauna (Bartoli et al. 2009). Hence under such conditions the PO_4^{3-} efflux could be much higher than measured in this experiment. A longer experiment could also have resulted in decreased PO_4^{3-} retention, because of exhaustion of the binding sites of PO_4^{3-} with Fe (III) (Jorgensen 1996). Nevertheless, the capacity of the sediment to retain PO_4^{3-} is notable considering that the amount of OM added in this experiment corresponded to the total OM deposited on the seabed below fish farms during a year, and that the OM added has a high reactivity, which implies that most of the OM is mineralized initially. Thus, marine sediment can act as a relevant sink of P, even under severe scenarios of

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335 temperature rise and organic enrichment, as long as it has a large enough pool of Fe and the
336 macrofauna keeps the surface of the sediment under oxic conditions.

337

338 4.2. NH_4^+ release

339 Rates of NH_4^+ -effluxes at 16°C were within the range reported from previous laboratory
340 experiments at similar conditions with non-enriched or enriched sediment (Valdemarsen et al.

Código de campo cambiado

341 2009;Valdemarsen et al. 2010). The 1-3 times temperature stimulation of NH_4^+ -efflux were in
342 the same range observed in sediments from coastal areas (i.e. temperature rise ca. 10°C during

Código de campo cambiado

343 summer resulted in one- to four-fold increases in NH_4^+ -efflux) (Kristensen 1993). As with PO_4^{3-} ,
344 the release rates of NH_4^+ were always higher in +OM cores than in -OM treatments for a given

345 temperature, highlighting that OM enrichment had a stimulatory effect on NH_4^+ release. This was
346 expected since high NH_4^+ -release is often observed in sediments enriched with labile organic

Código de campo cambiado

347 matter (Christensen et al. 2000;Valdemarsen et al. 2012). NH_4^+ release was not stimulated by

Eliminado: exponentially

348 temperature between 16-22 °C, but above 22°C NH_4^+ release increased non-linearly in both -OM
349 and +OM treatments. The notable increase of NH_4^+ release at a temperature increment of 10°C in

350 both -OM and +OM treatments, could led us to speculate that the efficiency of NH_4^+ oxidizing
351 pathways is lowered at high temperatures, nevertheless the data of C:N ratios pointed to an

352 opposite hypothesis.

353 The C:N ratios of C and N release during the experiment were always higher than the
354 C:N ratios of the sediment OM, in particular at high temperature rise. This suggests that part of

Eliminado: The C:N ratio of the added OM was 5.2, whereas the C:N ratios of the sediment OM at the beginning of the experiment was close to 12 in both -OM and +OM cores.

355 the mineralized N is not released to the water column as NH_4^+ , but transformed into other N

Eliminado: (C:N ratios of C and N release were 25.3 and 25.0 for a temperature increment of 6°C, 15.1 and 26.8 for a temperature increment of 10°C, in -OM and +OM cores, respectively)

356 compounds. NH_4^+ could be nitrified in the oxidized surface sediment or in *N. divericolor*

Eliminado: the

357 burrows, and subsequently reduced to N_2 via anammox or denitrification (Thamdrup 2012).

Código de campo cambiado

358 Previous studies show that both denitrification and anammox are stimulated by increasing
 359 temperatures (Nowicki 1994; Alsterberg et al. 2012; Canion et al. 2013) and by availability of OM
 360 (Nowicki 1994; Thamdrup and Dalsgaard 2002; Engstrom et al. 2005) which supports the data
 361 from this experiment. Nevertheless, evidence based on NO_3^- and NO_2^- data should be needed to
 362 confirm this hypothesis. Despite so, NH_4^+ is the dominating form of dissolved inorganic N
 363 effluxing from organic enriched sediments (Christensen et al. 2000; Holmer et al. 2003), while
 364 coupled nitrification-denitrification rarely exceeds $1\text{-}2\text{ mmol m}^{-2}\text{ d}^{-1}$ in marine sediments
 365 (Middelburg et al. 1996). We are therefore confident that the NH_4^+ release rates can act as a
 366 proxy for total inorganic N release to the water column.

367 Despite that bioturbation and bioirrigation by macrofauna promotes OM mineralization,
 368 in the present experiment, both PO_4^{3-} and NH_4^+ effluxes were below the expected levels of the
 369 mineralized organic matter. In the case of PO_4^{3-} , this can be explained by the PO_4^{3-} retention
 370 capacity of the sediment that is promoted by macrofauna activity (Bartoli et al. 2009). In the case
 371 of NH_4^+ , these results could be explained because macrofauna promotes NH_4^+ -oxidizing
 372 processes and the coupling between nitrification and denitrification (Gilbert et al. 1995). Thus,
 373 sediment reworking processes of macrofauna promote nutrient recycling while preventing
 374 eutrophication.

375 Additionally it should be considered that the experiment was performed in darkness, as
 376 the experiment was designed to simulate the generally low availability of light at the sediment
 377 surface in relatively enclosed and shallow coastal areas exposed to eutrophication, such as in the
 378 Danish coastal areas. Nevertheless, in other areas with more light availability autotrophic
 379 processes are also important. This could lead to different results with regards to nutrient release
 380 rates from the sediment.

Eliminado: ;Nowicki 1994

Código de campo cambiado

Eliminado: Engstrom et al. 2005;

Código de campo cambiado

Eliminado: mostly

Código de campo cambiado

Código de campo cambiado

Eliminado: produced

Código de campo cambiado

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Eliminado: prevent

Eliminado: While there is, to some extent, experimental manipulative experiments that focus on the effects of carbon mineralization rates of marine sediments on warming scenarios (Finke and Jorgensen 2008; Robador et al. 2009; Sanz-Lazaro et al. 2011b), less information is available on the mineralization rates of N and P. Marine sediments act as an important source of nutrients for water column primary production (Nixon 1981). Global warming may lead to eutrophication of the water column by increasing the nutrient release from sediments. For both nutrients, the effluxes increased with temperature, but followed significantly different trends. PO_4^{3-} effluxes increased linearly, highlighting that P mineralization is temperature dependent. On the other hand, NH_4^+ effluxes showed an exponentially increasing trend suggesting that N mineralization is expected to be independent from temperature rises of up to 6°C. Since in most marine basins, primary production in the water column is limited by N (Zehr and Kudela 2011), the predicted increases in NH_4^+ effluxes may have important consequences for temperature rises above 6°C. As an example, in the Baltic Sea, N demand of phytoplankton is $5.2\text{ mmol m}^{-2}\text{ d}^{-1}$ (Koop et al. 1990). Thus, in this case, phytoplankton demand of N would be surpassed above temperature increments of 6°C or with no increments in temperature, under non- and organic enrichment conditions, respectively. Under these scenarios, the excess of NH_4^+ efflux could stimulate primary production, which may lead to excessive proliferation of algae. ¶

381 In conclusion, in future scenarios of climate change, sediment PO_4^{3-} release is expected to
382 increase following the trends observed for OM mineralization rates in the sediment. The
383 magnitude of the release of PO_4^{3-} is influenced not only by local OM deposition rates but also by
384 sediment characteristics, particularly, the amount of reactive Fe in sediment. NH_4^+ release from
385 sediment is not expected to increase substantially with temperature increments of up to 6°C , in
386 organic polluted as well as in non-polluted sediments. However, more severe temperature rises
387 may be associated with a non-linear increase in the release of NH_4^+ from sediments. As most
388 marine basins are N-limited, the excess release of NH_4^+ may set in motion a cascade of negative
389 effects leading to deteriorating environmental quality. These effects are expected to be more
390 detrimental in organic polluted areas as well as in coastal basins with no or restricted exchange
391 with ocean waters.

Eliminado: n

Eliminado: exponential

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549 Tables

550

551 Table 1: Depth-integrated (0–16 cm) pools of P and N (mean ± SE) in the sediment used in the
552 experiment (n=3, initial concentration) and the estimates on the amount of nutrient mineralized
553 during the experiment based on nutrient sediment-water column fluxes (n=6). Values inside
554 square brackets indicate the % of the nutrient mineralized out of the total added.

555

	Initial concentration (mmol m ⁻²)	Mineralized (mmol m ⁻²), by temperature		
		16°C	22°C	26°C
-OM				
P	8178 ± 548	2.65 ± 1.12	4.35 ± 1.71	5.64 ± 1.42
N	7662 ± 401	252 ± 57.5	141 ± 59.3	441 ± 138
+OM				
P	8205 ± 72.1	7.54 ± 1.33 [4.2]	13.54 ± 2.4 [7.6]	16.49 ± 2.24 [9.3]
N	9854 ± 292	641 ± 130 [17.9]	623 ± 121 [17.4]	721 ± 165 [20.1]

556 Notes: The 16°C treatment corresponds to the present mean sea surface temperature (SST) for summer
557 months in the Baltic Sea. The 22 and 26°C treatments were chosen as SST climate change scenarios in
558 60 and 100 years based on SST temperature rise observed in the last decades and expected raises (1°C
559 per decade in both cases). –OM and +OM correspond to non- and organic polluted treatments,
560 respectively. The organic matter addition to +OM cores corresponded to 26 mol POC m⁻², which is
561 comparable to the annual organic matter deposition in areas under the influence of mussel or fish farms.
562 The final cores were maintained at 16, 22, or 26°C in separate tanks containing filtered seawater.

563 Table 2: Depth-integrated (0–16 cm) pools of reactive Fe (III) and P bound to reactive Fe (mean
 564 \pm SE) for initial cores (n = 3) and final cores (n = 6). See notes at Table 1 for explanation about
 565 treatments.

566

	Initial concentration (mol m ⁻²)	Final concentration (mmol m ⁻²), by temperature		
		16°C	22°C	26°C
-OM				
Reactive Fe (III)	2.5 \pm 0.1	2.0 \pm 0.3	1.8 \pm 0.1	2.3 \pm 0.5
P bound to Fe	2.5 \pm 0.1	2.1 \pm 0.2	2.5 \pm 0.2	2.6 \pm 0.3
+OM				
Reactive Fe (III)	2.3 \pm 0.2	1.9 \pm 0.2	2.5 \pm 0.3	2.3 \pm 0.5
P bound to Fe	2.1 \pm 0.2	2.4 \pm 0.2	2.6 \pm 0.3	2.0 \pm 0.2

567

568

569 Table 3: Coefficients (means at 26°C) of the regression model for PO_4^{3-} and NH_4^+ sediment-
 570 water column flux (i.e. efflux) rates along temperature for non-organic polluted (-OM) and
 571 organic polluted (+OM) treatments. Regression model for PO_4^{3-} corresponds to a first order
 572 polynomial regression, while for NH_4^+ correspond to an exponential regression (Table A1).
 573 Significant effects ($p < 0.05$) are indicated in bold.
 574

	PO_4^{3-} efflux rate	NH_4^+ efflux rate
	Coefficient (SE)	Coefficient (SE)
Intercept	0.207 (0.047)	13.58 (3.26)
OM	0.420 (0.076)	12.45 (5.27)
Temperature	0.016 (0.008)	0.946 (0.524)
OM x Temperature	0.028 (0.012)	0.108 (0.803)

575

576

577 Table 4: Initial C:N:P ratio of the organic matter in the sediment estimated from particulate
578 organic carbon, total nitrogen and total phosphorus and overall C:N:P ratio estimated from
579 nutrient and total CO₂ flux over the experimental period. C and total CO₂ flux data was reported
580 in Sanz-Lázaro *et al.* (2011b). See notes at table 1 for explanation of treatments.

Código de campo cambiado

	Initial ratio in the sediment	Overall ratio of the effluxes, by temperature		
		16°C	22°C	26°C
-OM				
C:N	12.7	13.3	25.3	15.1
C:P	11.6	849	656	988
N:P	0.9	63.8	25.9	65.3
+OM				
C:N	11.6	21.6	25.0	26.8
C:P	13.5	1866	1157	1174
N:P	1.2	86.5	46.3	43.7

581

582

583 Figure legends

584

585 Figure 1: Mineralization rates, shown as flux rates of total CO₂ (mean ± SE, n=6) versus
586 temperature under non- (-OM) and organic polluted (+OM) conditions modified from Sanz-
587 Lázaro et al. (2011b). Lines are shown to visualize the trajectory but do not represent a
588 regression.

Eliminado: Pools

Con formato: Subíndice

Eliminado: (Sanz-Lazaro et al. 2011b)

Eliminado: of reactive Fe(III) versus depth for initial cores (n=3) and final cores (n=6) in the different temperature treatments.

Código de campo cambiado

589

590 Figure 2: PO₄³⁻ and NH₄⁺ efflux rates (n=6, mean ± SE) during the experiment at the three
591 temperature scenarios under non- (-OM) and organic enrichment (+OM) conditions. Dotted lines
592 are the line of reference corresponding to 0.

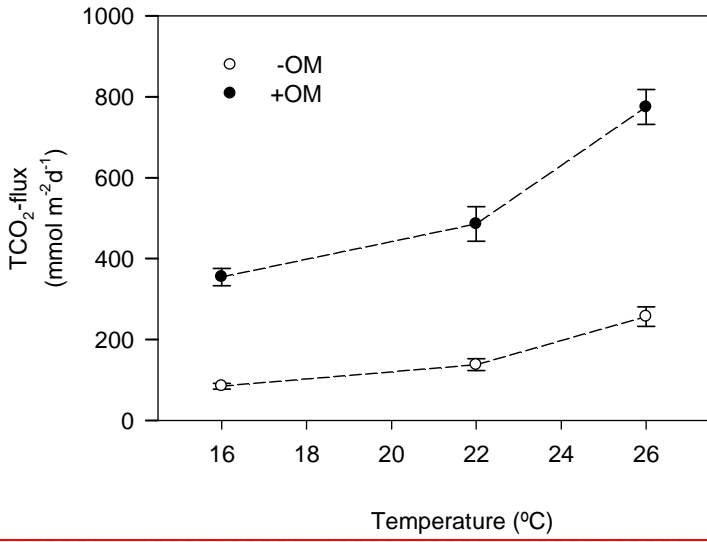
593

594 Figure 3: Nutrient sediment-water column flux rates (n=6, mean ± SE) versus temperature under
595 non- (-OM) and organic polluted (+OM) conditions. Lines indicate significant (p<0.05)
596 regressions for -OM and +OM treatments. Type of regression was chosen according to the
597 AICc (corrected Akaike Information Criterion) (Table A1). R² refers to the whole regression
598 model for each nutrient which includes the factor OM.

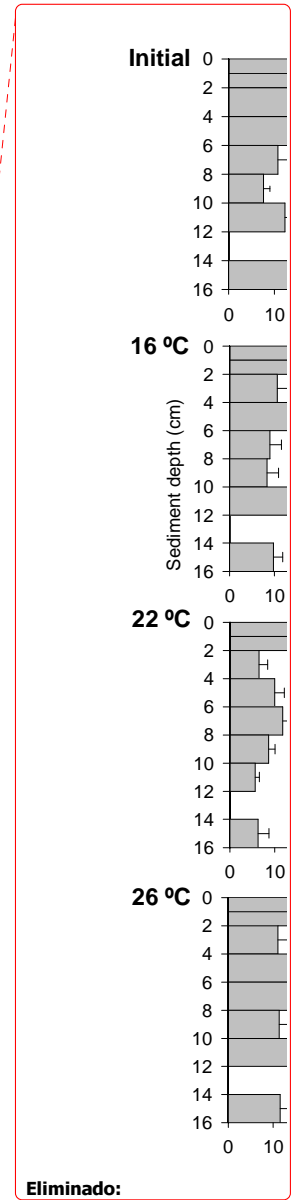
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600 Figure 1

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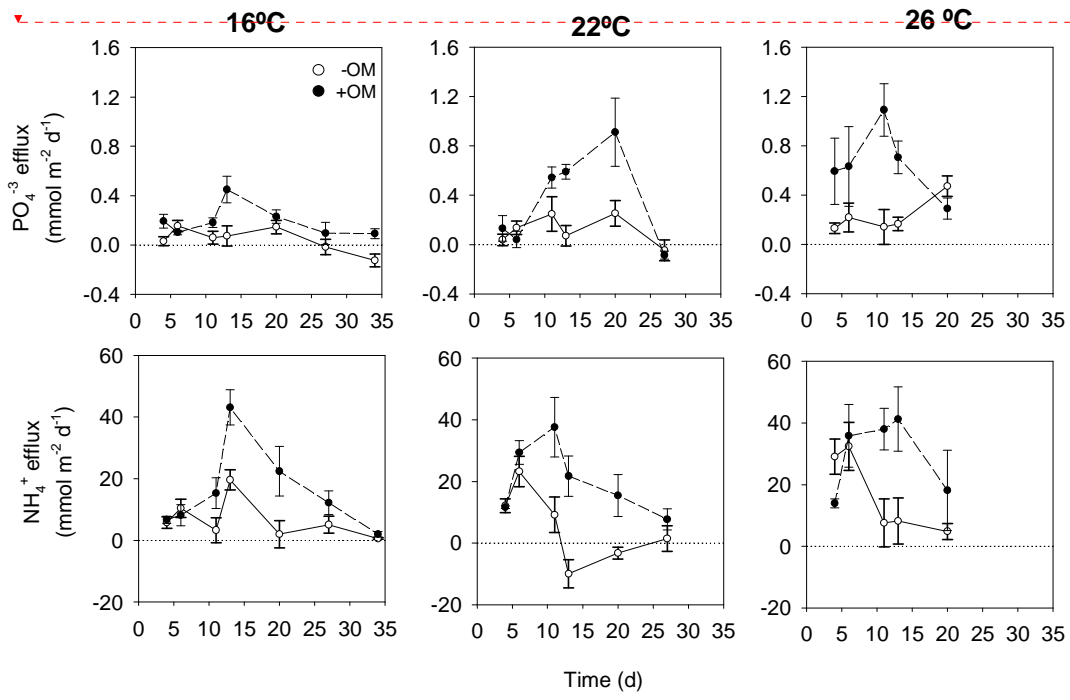


603 Figure 2

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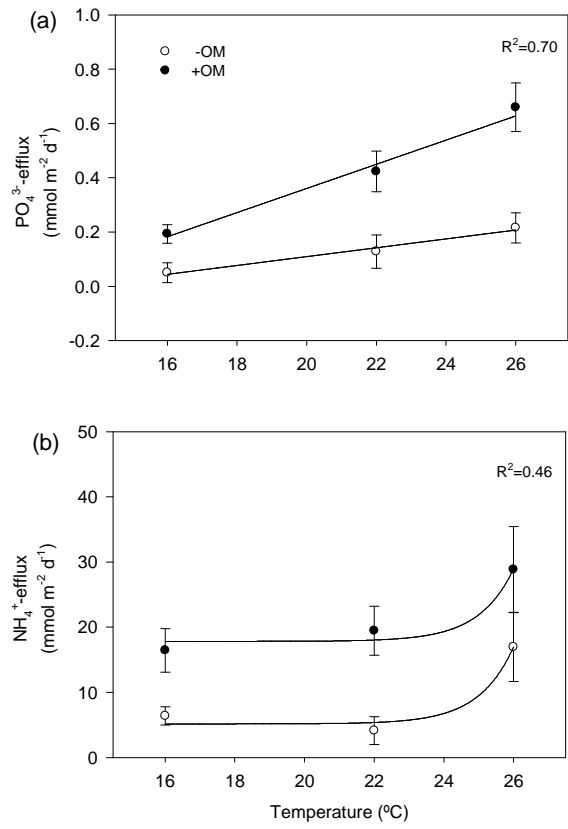
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607 Figure 3

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611 Appendix

612

613 Table A1: AICc (corrected Akaike Information Criterion) of the different regression models for

614 PO_4^{3-} and NH_4^+ efflux rates.

615

	AICc	
	PO_4^{3-} efflux rates	NH_4^+ efflux rates
$y_i = \beta_0 + \beta_1 x_i$	-31.940	247.60
$y_i = \beta_0 + \beta_1 e^{x_i}$	-23.032	243.04
$y_i = \beta_0 + \beta_1 x_i + \beta_2 x_i^2$	-26.321	248.63
$y_i = \beta_0 + \beta_1 \frac{1}{x_i}$	-30.336	248.95

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