

## Responses to the Referee 1 comments:

1)

„Many statistical analyses make the manuscript complex and confusing. Indeed, the result of NMDS is not fully discussed. What foraminiferal species are related to NMDS 1 and 2, and what can we learn from the temporal changes of NMDS 1 and 2? Figure 7 is not fully used in discussion. So, I think there is no need to show figure 7 in this manuscript”.

We think that unconstrained ordination - NMDS (Figure 7) - is important because it directly depicts the differences between foraminiferal assemblages from different stratigraphic intervals/time periods, independently of environmental data. We note that the stratigraphic order is rather lost in both RDA and clustering - they just show two distinct groups, with no trends and no continuous compositional gradient. We have rather decided to move Figure 8 with constrained ordination to the Supplement (see also below). We have expanded the description of NMDS in results, related it to the information on what taxa are associated with positive and negative scores, and have now provided some further explanation in the chapters Results and Discussion:

Chapter Results, page 14:

*“In the NMDS space, samples from the lower and middle part of the core (150-35cm) are associated with positive scores along the first axis and are tightly grouped, indicating relatively homogeneous faunal composition... In contrast, the samples from the upper 35cm of the core are associated with negative scores along the first axis and are widely distributed in the ordination space. The separation of the uppermost part of the core from the rest of the core in the ordination space suggests a continuous, but a relatively strong shift in the assemblage composition at the onset of the 20<sup>th</sup> century (Fig. 7).”*

Chapter Discussion, page 19:

*„However, the overall assemblage composition in the 20<sup>th</sup> century changed markedly relatively to the pre-20<sup>th</sup> century assemblage composition. The uppermost parts of the core show a very strong and directional change in composition lasting up to the present (Fig. 7), whereas the lower and middle parts of the core were characterized by a relatively constant taxonomic composition, with a much smaller multivariate dispersion in NMDS (Fig. 7).”*

„I think that figure 6 (b) and (c) are enough for the conclusion of the manuscript. It is also same to figure 8. What is PC1 in figure 8? I think PC1 is the result of PCA of geochemical data (Fig. 4). However, the values are not equal”.

As explained above, the results of NMDS analysis directly show stratigraphic order of foraminiferal assemblages and thus we would like to keep it as a part of the manuscript. The component PC1 in the Figure 8 (RDA plot) is based on relative abundances of foraminifera, while the geochemical data are reduced to total nitrogen concentration along RDA1. Because there is only one environmental variable selected by the forward model selection in the redundancy analysis (TN), there is only one RDA axis. We have clarified the caption to Figure 8 (in the revised version Figure S1) as follows:

*“Given that only one environmental variable is used, the second axis corresponds to the first principal component that visualizes the residual compositional variation unconstrained by geochemical data. “*

We moved the Figure 8 to supplementary data and added the Table S4 to the main part of the manuscript (The results of the forward model selection in the redundancy analysis; in the revised version named Table 1), because it provides detailed information about elements that explain the highest proportion of variation in assemblage composition. The results shown in this table are important for the conclusions of the manuscript, as previously explained on the page 21, lines 517-519, page 23, lines 577-578).

2)

„The authors concluded that the foraminiferal community has adapted to naturally elevated trace element concentrations, but such adaptation cannot be evaluated from this study because anthropogenic impact is found even in the bottom of the core”.

To avoid confusion, we have omitted “naturally”.

3)

„There are no explanations about dash line, solid line, and grey circle in figure 2. The explanations are needed in caption. I think dash line means the range between maximum and minimum ages of shells. If so, the range of ages between 90cm and 120cm are very wide (i.e. it shows “modern” to “old” ages).“

We have added our clarification to the caption of the Figure 2 as follows:

*"The dashed line represent the total range of ages of *Varicorbula gibba*, i.e., bounded by the minimum and maximum age, the solid line represents the inter-quartile range of ages, i.e., it is bounded by the 25th and 75th quantiles, and the grey circle refers to the median age. We note that age range and inter-quartile age range increase downcore and death assemblages in the basal core increments are time-averaged to few centuries, most likely due to bioturbational mixing. Therefore, although median age of *V. gibba* in the lowermost (145-150 cm) increment is 1616 AD, interquartile range of ages of *V. gibba* in this increment includes shells that died in the 16th century."*

Additionally, as the lowermost sample includes shells that died in the 16th century, the core effectively captures the past 500 years of environmental history in the Bay of Panzano. We corrected this within the title and the manuscript and added an explanation about this in chapter Chronological framework, page 12:

*“Median age of *V. gibba* in the lowermost (145-150 cm) increment is 1616 AD, but interquartile range of ages of *V. gibba* in this increment includes shells that died in the 16th century. Therefore, we used median ages to set the chronology of events, but the core effectively captures the past 500 years of environmental history in Panzano Bay (Fig. 2).”*

Moreover, calculated sedimentation rate between 120cm and 140cm is very high (ca. 2cm/yr). The concentration of Al decreases abruptly during this period. This may indicate the change of depositional environment. Evaluations about these points are needed”.

If we use median age in each increment, the difference of ~100 years between 120 and 140cm would imply sedimentation rate ~0.2cm per year, not 2cm per year. Although some elements (like Al) show some fluctuations that are in phase with grain size variations, absolute changes in sediment grain size are very minor (as already described in manuscript at page 12, lines 295-299) and we thus think that it is difficult to infer changes in environment based on it. We additionally discuss this in chapter Discussion, page 16:

*"In the Panzano Bay sediments, trace (Cr, Cu, Ni, Cd, Mn) and conservative elements (Fe, Al) have relatively higher concentrations in the lower part of the core and a pronounced decrease in the upper 35cm. The only discrepancy of this trend occurred in the latest 17<sup>th</sup> century, when the concentrations of all these elements declined, with simultaneous change in grain size (Fig. 3). Although this elemental fluctuation is in phase with grain size variations, absolute changes in sediment grain size are very minor and it is thus difficult to infer changes in environment based on it."*

„The authors argue that short term decline of Ammonia sp. in the latest 17th century may have caused by the increases of pollutants, because Ammonia sp. is sensitive to pollutants (p. 17 line 410 to 413). However, this short term drop is only one sample, and the drop of Al during this period may indicate the change of depositional environment as mentioned above. It may be the taphonomic effect. Indeed, it seems that the change in grain size distribution occurs simultaneously with Al drop. So, we cannot discuss the decrease of Ammonia in relation to anthropogenic impacts. Moreover, the authors describe that the increase of Ammonia sp. during the late 20th century is correlated with the increase of persistent organic pollutants (p. 21 line 511 to 516). These two interpretations of Ammonia species are inconsistent (Ammonia sp. is sensitive or tolerant to pollution? Ammonia sp. increases after 1950 when some pollutants increase rapidly.)”.

To avoid ambiguity and avoid difficulty with disentangling sampling issues and taphonomic effects, we have omitted the inference about the decline in *Ammonia* sp. in the lower part of the core from the Discussion (pages 17 and 18, lines 421-425, 428-431). We keep the information that most *Ammonia* species are tolerant to pollution.

4)

„Major foraminiferal change during 1700s to 1800s is fluctuation of Valvulineria species (Fig. 5). The authors argue that the distinct peak of Valvulineria in the early 19th century coincides with the coldest and most humid phases of the LIA by citing previous study. However, the same distinct peak of Valvulineria also occurs in the early 18th century. The authors also

describe that Valvulineria is adapted to large seasonal variability of organic matter, periodic hypoxic conditions, increased fluvial runoff and increased turbidity. However, nutrient concentrations and grain size are relatively stable rather than variable. The authors do not discuss this point”.

We have downplayed the inference about seasonality, mainly because of the likely effect of time averaging and mixing. The constancy of nutrient concentrations can be further explained by less dense spacing of samples where nutrients were measured. In the Discussion (page 18), we have added:

*“In contrast to fluctuations in abundance of these foraminifer taxa, vertical changes in the concentration of nutrients in the lower and middle part of the core are mild. However, the spacing of increments that were analyzed for nutrients is larger than dense spacing of increments analyzed for the composition of foraminiferal assemblages. In addition, concentrations of nutrients, grain size distribution and vertical changes in foraminiferal assemblages are likely further affected by vertical homogenization by bioturbation, as evidenced by decadal to centennial time averaging of *Varicorbula gibba* (Fig. 2), thus making difficult to detect the effects of environmental fluctuations occurring at higher (seasonal or yearly) temporal resolution“*

5)

„The authors describe that high abundances of Non keeled Elphidium, Valvulineria, and Ammonia during the 17th to 19th century suggest strong seasonal variations of river runoff and organic matter input based on the result of RDA (p. 17 line 425 to 429). However, I think that RDA results are strongly influenced by top 20cm (20th century) data of the core, because N. tot is relatively stable below 20cm. Gradual decrease of N. tot may indicate decomposition process. Indeed, if my understanding is right, high positive correlations occur within top 20cm (Figs. 4 and 8)“.

Yes, this is correct; we have removed our reference to RDA in this part of the Discussion (page 19, lines 455, 461). Our inference here is primarily based on known and described ecological requirements of these taxa.

Regarding nitrogen concentrations, we have added this statement into Discussion (page 20):

*„Although upward increasing concentrations of TOC and TN can be partly related to their recycling dynamic, the corresponding increase in pollutants (PAH and PCB) and other observations of major increase in pollutants and organic enrichments in the Gulf of Trieste (Heath et al., 2006) imply that the nutrient increase also reflects intensifying agricultural and maricultural activities in the Gulf of Trieste during the 20<sup>th</sup> century. “*

6)

„The authors propose the three hypotheses in introduction section, so inspection result of the hypotheses should be written in conclusion section. Especially, hypothesis three (relationship between foram diversity and pollutants) was not incompatible with the result. Many previous

studies have already suggested that early phase of eutrophication cases increase in foram diversity“.

We added the inspection of the hypotheses as follows:

In the Discussion, page 20:

*„The increase in diversity observed in the uppermost parts of the core not only correlates with nutrient enrichment (in accord with observations that early stages of eutrophication can increase species richness, Martinez-Colón et al. 2009) but also with higher concentrations of pollutants, thus rather contrasting with the hypothesis that pollution inevitably decreases species richness.“*

In the conclusions (pages 24 and 25):

*"This is reflected in increasing concentrations of trace elements and persistent organic pollutants (PAH, PCB), as well as in progressive nutrient enrichment, as it was presumed within the first two hypotheses. However, mining activity did not produce a progressive enrichment of mercury as anticipated in the second hypothesis, due to the improvement of the methods for the metal recovery. Increased pollutants did not cause a decline of species abundance and diversity as suggested by the third hypothesis, as foraminiferal response to such anthropogenic impacts in Panzano Bay are shaped by their long history of adaptation to elevated trace element concentrations, but also as initial stages of eutrophication can positively affect species richness."*

7)

„p. 18 line 453 to p. 19 line 473: The authors associate the increases in the abundances of Miliolinella, Triloculina, and Haynesina with enhanced microalgal biomass (mainly diatoms) as a consequence of nutrient enrichment. However, certain Elphidium species feed diatoms and prefer organic rich sediments. Ammonia tepida decreases during this period, but A. tepida is herbivorous and tolerant to all kinds of stress conditions, including organic enrichment as the authors describe in the manuscript. So, this faunal change cannot be explained only by enhanced microalgal biomass and nutrient enrichment“.

It is true that certain *Elphidium* species feed on diatoms, but those are only keeled species. Non-keeled *Elphidium* species prefer an infaunal mode of life and can be associated with food enrichment of the sediments. In Panzano Bay, only non-keeled *Elphidium* species dominate in the lower part of the core and we relate their abundances with the LIA, increased river run-off and organic matter input.

In the 20<sup>th</sup> century, taxonomic change of foraminiferal communities happened, including the decrease of non-keeled elphidiids and increase of herbivorous taxa. Such a shift in the trophic mode of foraminiferal species we interpreted as an indication of enhanced phytoplankton, reflecting higher nutrient levels. Therefore, non-keeled elphidiids decline here probably because of the shift in the available food. This inference is explained in the manuscript (page 18, lines 432-436, pages 20, 21, lines 502-508, page 21, lines 509-514).

Yes, *A. tepida* slightly decreases during this period and does not fit this trend. However, *Ammonia* sp. and *A. inflata* increase here. We added a comment about it on page 20:

*„Additionally, in the second part of the 20<sup>th</sup> century herbivorous genus Ammonia also slightly increases its abundances, with only A. tepida not following this trend.“*

9)

„p. 20 line 504 to p. 21 line 506: The authors describe that the presence of PAH is probably related to industrial activities, and it started to increase from the middle of the 20th century. However, it seems that PAH concentration start to increase from the latest 18th century, although it increases rapidly from the middle of the 20th century. So, industrial activities was advanced from the latest 18th century“.

We have modified our statement as follows (chapter Discussion, page 22):

*“The presence of PAH and PCB in Panzano Bay sediments is probably related to industrial activities in the port. Their concentrations are low throughout the core and start to increase in the 19<sup>th</sup> century and especially since the middle of the 20<sup>th</sup> century (Fig. 3)..., “*

10)

„Foraminiferal discussions in subsection 5.1 and 5.3 are same although different pollutants are described in each section. So, subsection 5.1 and 5.3 should be combined to avoid confusion.“

Subsections 5.1. and 5.3. concern each source of pollution separately, namely maricultural and agricultural in the chapter 5.1. and port activities in the chapter 5.3.

We estimate the effects of maricultural and agricultural activities based on trace element concentrations and nutrients. Consequently, we provide the information on the responses of foraminiferal assemblages to elevated trace element concentrations and nutrients.

In the chapter 5.3. we evaluate the effects of port activities by using the concentrations of persistent organic pollutants: polycyclic aromatic hydrocarbons (PAH) and polychlorinated biphenyl (PCB) and we discuss possible effects of these pollutants on foraminiferal assemblages. Therefore, we would like to retain these subsections as they are.

Additionally, we further split the Discussion into pre-20th and 20<sup>th</sup> century changes in foraminiferal assemblages.

„Figure 4: In the text, the authors describe “the first two axes explaining 75.8% of the variance”, but the value of PC2 is 14.8% in the figure 4 (sum of axes is 74.8%). Which is correct? Names of each arrow are piled and indistinct. Please redraw in the clearest way possible. Figure 5 : “sp.” is not italic. „References: Di Leonardo et al. (2006) and Solis-Weiss et al. (2001) are not cited in the text. There are discrepancies of publish year between the text and the references. P. 18 Line 440: Naehrer et al. (2014) P. 11 Line 270: R Core Team (2015) P. 4 Line 104: Xuschin and Piller (1994)“.

Figure 4: The correct number is 74,8% of variance. We redrew the Figure 4 as suggested and corrected all listed spelling mistakes.

## **Responses to the Referee 2 comments:**

it may be is useful to show Idrija in the map of Fig. 1

We agree, but Idria mercury mine is located outside the coverage of the map, far away in the central Slovenia. Therefore, we added the direction of the mercury input on the map of Fig. 1.

diversity indices: There is no need to measure diversity in three ways when one is enough

We use three different indices, as they depict the diversity in a different way. Also, we use them to prove that the results are consistent and not dependent on the used metric.

Additionally, these indices are frequently used in ecological studies (and not only in the ones dealing with foraminifera) to define the ecological status of the research area. Therefore, by keeping all three of them in this manuscript, we would thus allow easy comparison of results between different studies.

Line 209: normalised

We corrected this spelling mistake.

Line 307: 12.74mg/kg do not correspond to the reported in Table S2

Actually, it does correspond to the table, but it was not written clearly. The submitted version was: “*The concentration of Hg sharply increases at 100-130cm, from 12.74mg/kg to 44.7mg/kg.*”

And the revised version is: “*The concentration of Hg sharply increases from 12.74mg/kg at 145-150cm of the core depth, to 44.7mg/kg at 100-130cm.*”

Line 367: Campos et al., 2003 missing in References

Campos et al., 2003 is spelling mistake. It is Campos, 2003 and we corrected it accordingly.

Line 489: Aquavita should be Acquavita

We corrected this spelling mistake.

Line 1047: 1993 should be 1994

We corrected this spelling mistake.

1 **Anthropogenically induced environmental changes in the northeastern Adriatic Sea in the last**  
2 **400-500 years (Panzano Bay, Gulf of Trieste)**

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14

15 **Abstract**

16 Shallow and sheltered marine embayments in urbanized areas are prone to the accumulation of  
17 pollutants, but little is known about the historical baselines of such marine ecosystems. Here we  
18 study foraminiferal assemblages, geochemical proxies and sedimentological data from 1.6m long  
19 sediment cores to uncover ~~~400-500~~ years of anthropogenic pressure from mining, port and  
20 industrial zones in the Gulf of Trieste, Italy.

21 From 1600 to 1900 AD, normalized element concentrations and foraminiferal assemblages point to  
22 negligible effects of agricultural activities. The only significant anthropogenic activity during this  
23 period is mercury mining in the hinterlands of the gulf, releasing high amounts of mercury into the  
24 bay and significantly exceeding ~~today's Italian sediment quality guidelines (SQG) and~~ the standards  
25 on the effects of trace elements to benthic organisms ~~(ERL and ERM)~~. Nonetheless, the fluctuations  
26 in the concentrations of mercury do not correlate with changes in the composition and diversity of  
27 foraminiferal assemblages due to its nonbioavailability. Intensified agricultural and maricultural  
28 activities in the first half of the 20<sup>th</sup> century caused slight nutrient enrichment and a minor increase  
29 in foraminiferal diversity. Intensified port and industrial activities in the second half of 20<sup>th</sup> century



30 | increased the normalizzed trace element concentrations and persistent organic pollutants (PAH,  
31 | PCB) in the topmost part of the core, ~~with solely Ni exceeding Italian SQG, ERL and ERM~~. This  
32 | increase caused only minor changes in the foraminiferal community because foraminifera in  
33 | Panzano Bay have a long history of adaptation to ~~naturally~~-elevated trace element concentrations.  
34 | Our study underlines the importance of using an integrated, multidisciplinary approach in  
35 | reconstructing the history of environmental and anthropogenic changes in marine systems. Given  
36 | the prolonged human impacts in coastal areas like the Gulf of Trieste, such long term baseline data  
37 | are crucial for interpreting the present state of marine ecosystems.

38

### 39 **Keywords**

40 Marine pollution, Trace elements, Nutrients, Persistent organic pollutants, Benthic foraminifera

41

### 42 **1. Introduction**

43 | The northern Adriatic Sea is densely urbanized and polluted (Lotze et al., 2006; Cozzi and Giani,  
44 | 2011), and the areas around the Po River, the Venice Lagoon and in the Gulf of Trieste bear the  
45 | highest pressure (Solis-Weiss et al., 2007; Raccanelli et al., 2009). Panzano Bay, located in the  
46 | northeastern part of the Gulf of Trieste, is a shallow and sheltered embayment prone to the  
47 | accumulation of pollutants, with recent anthropogenic pressure coming from agricultural,  
48 | maricultural, mining and industrial activities (Horvat et al., 1999).

49 | The impact here started nearly 500 years ago with the onset of mercury mining in the hinterland of  
50 | the bay (Singh and Turner 2009, Covelli et al., 2012), enhanced in the late 20<sup>th</sup> century with  
51 | intensifying agriculture and mariculture (Aleffi et al., 2006; Rampazzo et al., 2013; Finch et al.,  
52 | 2014), and continued to the present times with increasing port and industrial activities  
53 | (thermoelectric plant) of the City of Monfalcone (Notar et al., 2001; Pozo et al., 2009).

54 | Such intensive anthropogenic pressures have prompted a growing scientific effort to estimate the

55 effects of pollution on ecosystem composition here. Most attempts have addressed modifications of  
56 the marine habitats that occurred in the 20<sup>th</sup> century, using only geochemical (Horvat et al., 1999,  
57 Faganeli et al., 2003; Acquavita et al., 2012) or biological proxies (Solis-Weiss et al., 2007). There  
58 is however, a growing tendency towards integrated assessments of its present state (Cibic et al.,  
59 2007; Melis and Covelli, 2013, Franzo et al., 2015), but until today there are no multidisciplinary  
60 studies assessing the long term history of the environmental changes in the northeastern Adriatic  
61 and thus capturing its preindustrial, undisturbed state.

62 Such a historical record requires an integrated geochemical and paleoecological approach. Benthic  
63 foraminifera, among the most abundant microorganisms in shallow and marginal marine  
64 environments, are often used in paleoecological studies. This is because they are highly sensitive to  
65 short term environmental changes (Schönfeld, 2012), they have a high preservation potential and  
66 thus provide an excellent temporal record of ecosystem states over the past hundreds to thousands  
67 of years (Yasuhara et al., 2012).

68 The present multidisciplinary study is designed to provide a high resolution historical record of  
69 environmental changes in Panzano Bay, to obtain information on the state of the ecosystem prior to  
70 the onset of the most intensive impact, and to evaluate the effects of anthropogenic activities in the  
71 bay. We obtained geochemical data and foraminiferal assemblages from a 1.6m long sediment core  
72 containing a centennial scale record of environmental and anthropogenic changes. The core covers  
73 approximately the last ~~400~~500 years, as indicated by radiocarbon calibrated AAR dating of the  
74 molluskes shells (Tomasovych et al., submitted).

75 Taking into account the history of potential anthropogenic stressors in Panzano Bay, we assess the  
76 following hypotheses: (1) agricultural and maricultural activities produce upcore increases in the  
77 concentrations of organic matter, nutrients and trace elements, (2) mining activities and  
78 thermoelectric plants generate a progressive enrichment of mercury and persistent organic  
79 pollutants, (3) increased pollutants alter the taxonomic composition of foraminiferal assemblages

80 and cause a decline of species abundance and diversity.

81 To test these hypotheses, we evaluate the pollution in the bay using geochemical proxies (major,  
82 minor and trace elements, nutrients, persistent organic pollutants) and quantify the composition and  
83 diversity of foraminiferal assemblages. Finally, we reconstruct the chronology of environmental  
84 changes in Panzano Bay over the last ~~400~~500 years and underline the applicability of our results to  
85 disturbed shallow coastal ecosystems elsewhere.

## 86 **2. Study area**

87 The Gulf of Trieste is a shallow marine basin in the northernmost part of the Adriatic Sea,  
88 occupying an area of about 500 km<sup>2</sup>, with an average water depth of 17m and a maximum of about  
89 25m (Fig. 1). Seasonal variations of water temperature range between 8 to 24°C at the surface and 8  
90 to 20°C in the bottom layer. The salinity of the water in the gulf is typically marine, ranging  
91 between 33 and 38.5‰ (Ogorelec et al., 1991).

92 The water enters the gulf in the southeast and continues to the northwest, following the general  
93 anticlockwise circulation pattern of the Adriatic Sea. However, the water circulation in the gulf is  
94 mostly controlled by tides (range ~0.5m), winds (strong northeastern Bora) and seasonal variations  
95 of freshwater inflow. The Isonzo/Soča and Timavo rivers are the most significant sources of  
96 freshwater to the Gulf, with average inflows of about 100-130m<sup>3</sup>/s each (Ogorelec et al., 1991).

97 The Gulf of Trieste generally shows mesotrophic to oligotrophic conditions, with episodic  
98 eutrophication events, accompanied by summer thermal stratification of the water column  
99 (Ogorelec et al., 1991; Horvat et al., 1999; Turk et al., 2007).

100 The main sediment supply comes from the Isonzo River in the north and from the weathering of the  
101 Paleogene flysch deposits outcropping along the southern coast of the gulf. The sediment  
102 accumulation rates are approximately 1 mm/yr in the central part of the gulf and increase to about  
103 2.5mm/yr towards the mouth of the Isonzo River located in Panzano Bay (Ogorelec et al., 1991, our  
104 unpublished data). Surface sediments in this area are mostly silt clays and clay silts (Zuschin and

105 Piller, 1994) occupied by a high biomass epifauna (Zuschin et al., 1999).  
106 The Gulf of Trieste is affected by many sources of organic and inorganic pollutants, coming from  
107 agricultural and industrial activities in the hinterland as well as from tourist and maricultural  
108 activities along its coasts (Notar et al., 2001; Covelli et al., 2006). Panzano Bay is one of the highly  
109 impacted areas, with organic pollution coming from mussel farms located along the eastern part of  
110 the Gulf of Trieste (Melaku Canu and Solidoro, 2014) and industrial and port areas of the city of  
111 Monfalcone, including a thermoelectric plant and several coal, petroleum and other cargo handling  
112 piers (Fig. 1). The Monfalcone thermoelectric plant consists of four thermoelectric generator sets  
113 powered by coal and fuel oil and became operative in 1965 ("The Monfalcone Thermoelectric  
114 Plant"). Finally, there is substantial Hg pollution originating from the Idrija mercury mine in the  
115 hinterlands and delivered to the bay through the Isonzo river flow (Horvat et al., 1999; Notar et al.,  
116 2001). Idrija, situated 50km west of Ljubljana (Slovenia), was the second largest Hg mine in the  
117 world, operating for nearly 500 years until its definite closure in 1995 (Faganeli et al., 2003;  
118 Covelli et al., 2012). During this period, over 5 million tonnes of Hg ore were mined and much of  
119 the residues were spread around the town and its vicinity (Miklavčič, 1999). Most of the Isonzo  
120 riverine input of Hg is in particulate form (1500kg/yr), followed by dissolved Hg at 8.6kg/yr  
121 (Faganeli et al., 2003). Dissolved mercury is biogeochemically reactive and tends to accumulate in  
122 certain seafood from mariculture, presenting social and economic problems for the local population  
123 (Faganeli et al., 2003).

### 124 **3. Methods**

#### 125 **3.1. Sampling**

126 Three sediment cores, two for sedimentological and one for foraminiferal analyses, 1.6m long with  
127 a diameter of 9cm, were acquired using an UWITEC piston corer with hammer action (Gallmetzer  
128 et al., ~~in press~~2016) from a research vessel in summer 2013. The drilling station is located in the  
129 central part of Panzano Bay (45°44,122' N; 13°36,029' E) at a water depth of 12.5m. The

130 uppermost 20cm of each core were sliced into 2cm thick intervals in order to attain high resolution  
131 data. The rest of the core was sliced into 5cm thick samples. For analytical purposes and in order to  
132 improve compatibility with the lower part of the core, the uppermost 2cm thick samples were  
133 merged into 4cm thick intervals (reducing the number of samples from 36 to 31). Sediment samples  
134 were used to determine grain size, the content of major, minor and trace elements, nutrients and  
135 persistent organic pollutants. Core chronology is based on molluscan shells dated by <sup>14</sup>C calibrated  
136 amino acid racemisation.

### 137 3.2. Sediment parameters

138 The grain size of 36 samples was analysed using a sedigraph (SediGraph III 5120 Particle Size  
139 Analyzer) for the small fractions (<63µm) and dry sieving for fractions from <63µm to >1mm. The  
140 sediments were classified according to the Shepard's classification (1954).

141 The concentrations of elements, nutrients and pollutants were determined at specific core sections:  
142 1cm, 5cm, 9cm, 24cm, 46cm, 69cm, 85cm, 105cm, 126cm and 151cm core depth. Geochemical  
143 analyses included the content of major (Fe, Al), minor (Mn, P) and trace elements (As, Cd, Cr, Cu,  
144 Hg, Ni, Pb, Zn), nutrients (total organic carbon – TOC and total nitrogen – ~~TN-tot~~), persistent  
145 organic pollutants (polycyclic aromatic hydrocarbons – PAH, polychlorinated biphenyls – PCB)  
146 and total inorganic carbon (~~C-tot~~).

147 To analyse elemental concentrations, each sediment sample was gently squeezed to break down  
148 aggregates and screened through a PE sieve to remove particles bigger than 1mm. A part of the  
149 screened sediment was dried in an oven at 105°C until reaching a constant weight (to measure water  
150 content). The dried sediment was ground to powder using an agate mortar and pestle before further  
151 analysing the contents of heavy metals and As. The sample (about 0.4g d.w.) was digested with  
152 8ml HNO<sub>3</sub> in a microwave oven (Multiwave 3000, Anton Paar, Austria). The digested material was  
153 left to cool at room temperature and then filtered through a 0.45µm nitrocellulose membrane filter.

154 The filtered digestates were diluted with distilled deionised water to 40ml in a volumetric flask

155 (USEPA, 1994A). The concentrations of the elements (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn)  
156 were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) (Optima  
157 2100DV, Perkin Elmer, USA) (USEPA, 1994B). Mercury analyses were carried out using atomic  
158 absorption spectrophotometry with cold vapor (Analyst 100, Perkin Elmer, USA) (USEPA, 1976).  
159 The quality acceptance protocols required that one blank sample or one certified reference material  
160 (BCR-277r estuarine sediment, Community Bureau of Reference) were digested and analyzed with  
161 each batch of fifteen samples. The blank results indicated that the analytical procedure was free  
162 from contamination because the concentrations of all metals were below the respective method  
163 detection limits. Mean recovery from the certified material ranged between 84% (Zn) and 103%  
164 (Hg), except for Al (40%) because the extraction method was not strong enough to break crystalline  
165 aluminosilicates. The analytical precision, determined using five replicates of homogenized samples,  
166 was estimated to be better than 10% for all elements. Calibration for ICP-AES and AAS analysis  
167 was achieved with prepared external standards via the standard curve approach. Full calibration was  
168 performed after every set of 48 samples. The method detection limit for element analysis was  
169 defined as 3 times the standard deviation of 10 blank measurements.

170 Carbon and nitrogen determination was performed following the method of Hedges and Stern  
171 (1984), using an elemental analyzer (CHN 2400, Perkin Elmer, USA). The total concentration- (TC  
172 and TN) was determined on an aliquot of the sample as is (about 10mg of dry sediment); the  
173 organic fraction of carbon (TOC) was determined after treatment of another aliquot of the sample  
174 with acid vapors. The inorganic fraction (C~~-tot~~) was calculated by the difference between the total  
175 and organic concentrations. For the instrument calibration, before each daily series of analyzes,  
176 three replicates of Acetanilide standard were performed. For the quality acceptance protocols one  
177 blank sample every five samples was analyzed.

178 To analyze the concentrations of persistent organic pollutants, sediment samples were thoroughly  
179 mixed, sieved through a 1mm mesh to remove any debris, and subsequently air dried in the dark at

180 room temperature for 48h on hexane rinsed aluminum foil. The dry samples were finely ground in  
181 an agate mortar. The extraction was performed using a Microwave Sample Preparation System  
182 (Multiwave 3000, Anton Paar Graz, Austria), in accordance with the EPA recommendation (method  
183 3546). Two grams of dried sediments were weighed into lined microwave extraction vessels. Then,  
184 a 25ml 1:1 acetone/hexane solvent mixture was added. The vessels were then assembled as  
185 instructed by the manufacturer and the extraction was conducted during 15 min at 110°C and 6-10  
186 bars. At the end of the oven program, vessels were cooled to room temperature and the extracts  
187 were filtered and rinsed with the same solvent mixture.

188 The samples were concentrated in a rotating evaporator (Rotavapor-R Buchi, CH), and the sulphur  
189 compounds were removed by soaking the extracts with activated copper powder. Purification and  
190 fractionation were performed by eluting extracts through chromatography glass columns packed  
191 with Silica gel/Alumina/Florisil (4+4+1gr). The first fraction, containing PCBs, was eluted with  
192 25ml of n-hexane, whereas the second fraction, containing the PAHs, was eluted with 30ml of 8:2  
193 n-hexane/methylene chloride solvent mixture (Fossato et al., 1996, 1998). After concentration with  
194 a rotary evaporator, the samples were ready for the instrumental analysis.

195 The identification of PAHs and PCBs was based on matching retention time, and the quantification  
196 was obtained from calibration curves established for each compound by analyzing four external  
197 standards. Average determination coefficients  $R^2$  of the calibration curves exceeded 0.99 for both  
198 PAH and PCB, and the relative standard deviations of the calibration factors were always less than  
199 20% (average 10%). The detection limits were 0.05-0.1ng/g and 0.05ng/g for PAHs and PCBs,  
200 respectively. Blanks were run for the entire procedure. Recovery and accuracy were validated with  
201 IAEA-417 and IAEA-159 sediment sample certified reference materials. Laboratory methods were  
202 also validated by intercalibration activities (IAEA, 2001, 2007, 2012).

203 Raw concentrations of Hg, Cr, Pb, As, Cd and PCB were compared to Italian sediment quality  
204 guidelines (SQG), following the directive D. L.vo n. 172 of 13/10/2015, whereas PAH and Ni

205 threshold concentrations were taken from directive DM 367/2003. Additionally, raw concentrations  
206 were compared to two sediment quality criteria used around the world: effects range low (ERL),  
207 representing the threshold level below which effects to benthic organisms rarely occur, and effects  
208 range medium (ERM), above which effects are likely to occur (Burton, 2002). Finally, trace  
209 elements were normalized to a reference element (Al) in order to compensate for grain size and  
210 mineralogical effects on the metal variability in samples (Covelli et al., 2006).

### 211 3.3. Foraminiferal analyses

212 A total of 36 sediment samples were washed with water through a set of sieves with 63, 125, 250  
213 and 500µm meshes. Each obtained subsample was split with a microsplitter in order to yield around  
214 300 specimens. Individuals were identified under a binocular microscope following the  
215 classification of Loeblich and Tappan (1987) and Cimerman and Langer (1991). Foraminiferal  
216 species were categorized into different ecological categories: according to their substrate relation  
217 (infaunal, epifaunal, epiphytic, epifaunal/infaunal and an epiphytic/infaunal group for ambiguous  
218 literature data) and according to their tolerance to different types of pollution, namely organic or  
219 chemical (referring to trace elements and persistent organic pollutants). (hereafter referred to as  
220 stress tolerant taxa). This classification was based on a total of 84 sources, including studies carried  
221 out in the Adriatic (Jorissen, 1987, 1988; Van der Zwaan & Jorissen, 1991; Jorissen et al., 1992;  
222 Barmawidjaja et al., 1992, 1995; Hohenegger et al., 1993; Vaniček et al., 2000; Donnici and  
223 Serandrei Barbero, 2002; Albani et al., 2007,; Frontalini and Coccioni, 2008, 2011; Di Bella et al.,  
224 2008, 2013; Vidović et al., 2009, 2014; Coccioni et al., 2009; Popadić et al., 2013; Melis and  
225 Covelli, 2013; Langlet et al., 2013, 2014). Additionally we used important primary and secondary  
226 literature about foraminiferal ecology (Murray, 1991, 2006; Langer, 1993), the most recent studies  
227 about improved tools and methods when using benthic foraminifera in environmental monitoring  
228 (Barras et al., 2014; Alve et al., 2016; Dimiza et al., 2016) and Paleobiology Database  
229 (Behrensmeyer and Turner, 2013). A full list of used sources is given in the Supplement: Table S1.



### 230 3.4. Statistical analyses

231 Before further statistical treatment, 18 environmental variables (grain size and raw concentrations  
232 of nutrients and organic and inorganic pollutants) were checked for normality, log transformed  
233 when non normal distribution was detected, and z standardized to account for different units and  
234 scales. Pearson correlations among environmental variables and principal component analysis (PCA)  
235 based on these 18 variables were performed to assess their collinearity and stratigraphic distribution.  
236 Only clay content was used in the multivariate analyses because other grain size fractions correlate  
237 with the percentage of clay.

238 The total foraminiferal assemblages were used in all analyses by pooling all mesh size fractions for  
239 each sample. Species diversity was measured using species richness, the exponential of Shannon  
240 entropy, and Fisher's  $\alpha$ . The exponent of the Shannon index (H) corresponds to the number of  
241 equally abundant species that would produce the given value of H (Hill, 1973; Jost, 2006). As all  
242 three diversity measures strongly depend on the number of sampled individuals, we rarefied our  
243 abundance data down to the size of the smallest sample (240 specimens). This procedure was  
244 repeated 1000 times and the mean values of species richness,  $\exp(H)$  and  $\alpha$  with corresponding  
245 95% confidence intervals were computed across all iterations.

246 Species relative abundance data were square root transformed before multivariate analyses. Non  
247 metric multidimensional scaling (NMDS) based on Bray-Curtis distances was used to visualize  
248 gradients in community composition. Rescaling the NMDS space according to the underlying  
249 dissimilarity matrix and rotating it with the principal component analysis maximized the  
250 compositional variation among samples along the first ordination axis (Oksanen et al., 2015).  
251 NMDS axis 1 scores thus correspond to the relative position of samples along the main gradient in  
252 species composition. The Pearson correlation was used to measure the association between the  
253 environmental variables and NMDS axis 1 scores for the subset of samples with available values of  
254 elemental concentrations.

255 Redundancy analysis (RDA) combined with the forward model selection approach was employed to  
256 quantify variation in the multivariate composition of foraminiferal assemblages explained by  
257 environmental variables. The effects of environmental variables were first tested in single  
258 regressions. Most environmental variables, however, show some degree of collinearity, and the  
259 forward model selection approach was thus employed to find a subset of factors that maximizes the  
260 explanatory power of environmental variables. At each step of the model building algorithm, an  
261 environmental variable with the highest partial  $R^2$  was added while considering the effects of the  
262 already selected variables, and the significance of the additional contribution was evaluated through  
263 a permutation test (10 000 permutations) (Blanchet et al., 2008).

264 To identify the timing of the major shifts in assemblage composition, we performed chronological  
265 clustering, a type of constrained cluster analysis that takes into account the temporal sequence of  
266 samples (Birks, 2012), by using the CONISS algorithm (constrained incremental sum of squares  
267 agglomerative clustering) implemented in “chclust” function from the “rioja” package (Juggins,  
268 2015). The number of significantly distinct temporal bins was determined by comparing the amount  
269 of variance accounted for by a given number of clusters to a random expectation based on the  
270 broken stick model (Bennett, 1996). Clustering was performed on the Bray-Curtis distance matrix  
271 based on relative abundance data. All statistical analyses were performed in R 3.2.1 (R Core Team,  
272 2014~~5~~) using “vegan” (Oksanen et al., 2015) and “rioja” (Juggins, 2015) packages.

### 273 **3.5. Chronological framework**

274 Core chronology is based on the radiocarbon calibrated amino acid racemization dating of the  
275 bivalve species *Varicorbula gibba* (Tomasovych et al., submitted). First, 13 shells of *V. gibba* were  
276 selected for  $^{14}\text{C}$  dating and analyzed at the Poznan Radiocarbon Laboratory. Radiocarbon ages  
277 were converted to calendar years using Calib7.1 (Stuiver and Reimer, 1993), the Marine13 data  
278 (Reimer et al., 2013), and a regional marine reservoir correction ( $\Delta R$ ) in the northeastern Adriatic  
279 equal to = -61 years (standard deviation = 50 years) (Siani et al., 2000). The extent of amino acid

280 racemization (AAR) in 329 shells was analyzed at Northern Arizona University using reverse  
281 phase high pressure liquid chromatography (RP-HPLC) and the procedures of Kaufman and Manley  
282 (1998). Thirty specimens of *V. gibba* were randomly selected from eleven, more or less evenly  
283 spaced, 4cm or 5cm thick intervals covering the whole core thickness. The rate of AAR was  
284 calibrated based on the 13 shells dated with  $^{14}\text{C}$  and three live collected individuals with the  
285 Bayesian model fitting according to Allen et al. (2013). The time dependent reaction kinetic model  
286 with the initial D/L value estimated from data and lognormal uncertainty showed the best  
287 calibration between D/L values of aspartic acid and calendar ages. AAR data in 18 shells did not  
288 pass screening criteria, and ages of 311 specimens in total were used for core chronology (Fig. 2).  
289 Median age of *V. gibba* in the lowermost (145-150cm) increment is 1616 AD, but interquartile  
290 range of ages of *V. gibba* in this increment includes shells that died in the 16<sup>th</sup> century. Therefore,  
291 we used median ages to set the chronology of events, but the core effectively captures the past 500  
292 years of environmental history in Panzano Bay (Fig. 2).

## 293 4. Results

### 294 4.1. Sediment parameters and geochemistry

295 The grain size distribution is rather homogeneous throughout the Panzano Bay core, with only a  
296 slight increase in the contribution of the >1mm fraction in the uppermost part (up to 8.9%). The  
297 sediment in the lowermost part of the core is composed of silty clay (50.4-54.5% clay). Starting  
298 from 135cm toward the upper section of the core, the amount of clay decreases to 43.5-50% and the  
299 sediment changes into clayey silt (Fig. 3).

300 Principal component analysis (PCA) based on raw elemental concentrations illustrates the  
301 correlation between elements, with the first two axes explaining 75.8% of the variance of the data  
302 (Fig. 4). This approach distinguishes two major groups of elements with different vertical  
303 distribution trends (Fig. 3, Table S2), and three elements (Hg, As, C~~tot~~) that do not fall into this  
304 grouping and have distinct position in the ordination space. The first group comprises trace (Cr, Cu,

305 Ni, Cd, Mn) and conservative elements (Fe, Al), characterized by positive mutual correlations  
306 (Table S3) and a pronounced decrease in the upper 35cm. The second group includes organic and  
307 inorganic pollutants and nutrients whose raw concentrations are stable (Pb, Zn, PCB) or increase  
308 only slightly (PAH, ~~TN-tot~~, TOC, P) in the lower part of the core, but sharply increase in the  
309 uppermost 35cm. Concentrations of PAH increase markedly in the upper 35 cm, although it also  
310 shows high values at 75cm. Normalization to Al reveals two pronounced peaks in the  
311 concentrations of the elements from the first group: at 125-130cm core depth and in the uppermost  
312 10cm. The latter peak is also visible in normalized Pb and Zn values (Fig. 3). The concentration of  
313 Hg sharply increases from 12.74mg/kg at 145-150cm of the core depth, 100-130cm, from  
314 12.74mg/kg to 44.7mg/kg at 100-130cm. The Hg values then decrease upcore to a minimum in the  
315 surface sediment (8.22mg/kg). Concentrations of As vary in the lower core (2.14-9mg/kg) but  
316 gradually decrease to 4.3mg/kg in the surface sediment (0-20cm). Normalization to Al reveals one  
317 concentration peak of As in the upper 10cm; it coincides with the peak of all other trace elements.  
318 Total carbon remains constant throughout the core (8-9.35%), except for the lowermost part (3.7%  
319 ~~C-tot~~).

#### 320 **4.2. Trends in foraminiferal assemblages**

321 A total of 69 benthic foraminiferal species were identified in the sediments from Panzano Bay, with  
322 raw species richness varying between 29 and 41 species in individual samples (~~26.2-36.4~~ species  
323 after rarefaction to 240 individuals; Table S1). The highest percentage of individuals belongs to the  
324 suborder Rotaliina (63-89%), followed by Miliolina (8-29%) and Textulariina (1.5-11%). Relative  
325 abundances of suborders are generally stable throughout the core and vary notably only in the  
326 uppermost 20cm (Fig. 5). Diversity is high throughout the core and increases only in the second half  
327 of the 20<sup>th</sup> century. Values of Fisher  $\alpha$  index vary from 7.5 in the lower core to 12 in the uppermost  
328 sample; the exponential of Shannon index ranges from ~~14.2~~ to 23 and shows the same vertical trend  
329 (Fig. 6A).

330 Epifaunal/infaunal and infaunal taxa dominate the assemblages, having variable abundances in the  
331 lower core (late 17<sup>th</sup> and 18<sup>th</sup> century) and more stable abundances in its upper part (Fig. 6D). In  
332 contrast, the number of infaunal species increases distinctly during the 20<sup>th</sup> century (Fig. 6E).

333 Foraminiferal species tolerant to both chemical and organic pollution dominate the assemblages  
334 (40-60%), with maximal abundances in the 18<sup>th</sup>, 19<sup>th</sup> and the second half of the 20<sup>th</sup> century.  
335 Species known to tolerate only organic pollution make up 18.5 to ~~42~~41.7% of the assemblage and  
336 have opposite temporal trends than the organic/chemical group, with decreasing trends in the above  
337 mentioned time intervals (Fig. 6F, Table S1).

338 NMDS ordination and chronological cluster analysis of the assemblages reveal two main groups of  
339 samples, with the major shift in relative species abundances starting around 35cm (Figs. 6B, 6C and  
340 7). This depth approximately corresponds to the late 19<sup>th</sup> century, ~1860 AD (Fig. 2). In the NMDS  
341 space, samples from the lower and middle part of the core (150-35cm) are associated with positive  
342 scores along the first axis and are tightly grouped, indicating relatively homogeneous faunal  
343 composition. These assemblages are characterized by dominance of *Valvulineria* sp., *Nonionella*  
344 sp., non keeled elphidiids, *Ammonia* sp., *A. tepida* and *Haynesina depressula* (Fig. 5). The 130-  
345 135cm sample (latest 17<sup>th</sup> century) represents an outlier with unusually low abundance of *Ammonia*  
346 sp. and an increased share of epiphytic species. In contrast, the samples from the upper 35cm of the  
347 core are associated with negative scores along the first axis and are widely distributed in the  
348 ordination space, ~~with gradually decreasing axis 1 scores.~~ The separation of the uppermost part of  
349 the core from the rest of the core in the ordination space ~~This~~ suggests a continuous, but relatively  
350 strong shift in the assemblage composition at the onset of the 20<sup>th</sup> century (Fig. 7) ~~in the uppermost~~  
351 ~~core~~. Here, the major drop in the abundance of *Valvulineria* sp. and non keeled elphidiids is  
352 accompanied by a growing share of *Miliolinella* sp., *Triloculina* sp., *Haynesina* sp. and *Nonion* sp.  
353 (Fig. 5).

354 Within each of the two major groups of samples, further clusters are recognizable, defined by the

355 breaks at 85 and 20cm (Fig. 6C). The lowermost part of the core (150-85cm) corresponds to the  
356 period from ~1600 to ~1800 AD and has variable foraminiferal distribution trends. The middle part  
357 of the core (85-35 cm, ~1800 to ~1860 AD) is character~~ized~~ed by more stable foraminiferal  
358 abundances and a pronounced decline of the genus *Valvulinaria*. At 35-20cm (~1860 to 1950 AD)  
359 the diversity of the foraminiferal assemblages starts to increase, as do the abundances of epiphytic  
360 species. The uppermost sediment (20-0cm, 1950 until today) is character~~ized~~ed by a further increase  
361 in biodiversity and in the abundance of textulariids (Figs. 5 and 6A).

### 362 4.3. Relationship between foraminiferal assemblages and geochemical proxies

363 NMDS axis 1 scores are positively correlated with concentrations of Cu, Ni, Cd, Mn, Fe, Al and  
364 negatively with ~~total-nitrogen~~TN and PCB (Table S3). The amount of clay does not correlate with  
365 axis 1 scores (Table S3).

366 Total nitrogen content explains the highest proportion of variation in assemblage composition  
367 (42.4%) and is the only explanatory variable included in the RDA analysis following the forward  
368 model selection procedure (Table S41). Nonetheless, other elements that closely (positively or  
369 negatively) correlate with total nitrogen content explain a significant amount of variation in single  
370 RDA analyses (Table S41), including TOC, organic pollutants (PAH and PCB), and trace elements  
371 (Mn, Fe, Ni, Cu, Cd, Zn). The assemblages from the topmost sediment layers (20<sup>th</sup> century) are  
372 clearly separated from the middle core assemblages and from assemblages at the base along RDA  
373 axis 1 (Fig. S1). This separation reflects the stratigraphic increase in the content of nitrogen, organic  
374 carbon and pollutants and the stratigraphic decline in several trace elements (Fig. 83).

## 375 5. Discussion

### 376 5.1. The effects of agricultural and maricultural activities

377 The agricultural use of pesticides and of organic or inorganic fertilizers releases considerable  
378 amounts of pollutants into the environment (Campos-~~et al.~~, 2003; He et al., 2005; Finch et al., 2014).  
379 Pesticides contain pollutant elements such as As, Hg, Cr and Pb (Campos, 2003), while fertilizer

380 contamination includes the discharge of macronutrients (N, P, K) and trace elements, including Co,  
381 Cu, Fe, Mn and Zn (Finch et al., 2014). Maricultural activities, in turn, disperse organic matter  
382 (waste feed and faeces) and nutrients (Mantzavarakos et al., 2007), leading to elevated  
383 concentrations of P, N and TOC in the sediment (Holby and Hall, 1991; Hall et al, 1990, 1992;  
384 Mook et al., 2012).

385 In the Panzano Bay sediments, trace (Cr, Cu, Ni, Cd, Mn) and conservative elements (Fe, Al) have  
386 relatively higher concentrations in the lower part of the core and a pronounced decrease in the upper  
387 35cm. The only discrepancy of this trend occurred in the latest 17<sup>th</sup> century, when the  
388 concentrations of all these elements declined, with simultaneous change in grain size (Fig. 3).  
389 Although this elemental fluctuation is in phase with grain size variations, absolute changes in  
390 sediment grain size are very minor and it is thus difficult to infer changes in environment based on  
391 it. In the Panzano Bay sediments, only few elements (Cd, Cr and Pb) sporadically and slightly  
392 exceed the limits imposed by the Italian SQG (Fig. 3, Table S2). Only Ni has elevated values  
393 throughout the core, even when compared to the standards evaluating the effects of trace elements  
394 to benthic organisms (ERL and ERM). Ni, Cd and Cr have a high positive correlation with the  
395 major constituents of clay minerals (Al and Fe), the main scavengers of trace elements (Romano et  
396 al., 2013). This points to a possible grain size and mineralogical effect on the accumulation of these  
397 elements throughout the core because the sediment in Panzano Bay is composed of silt and clay  
398 fractions (Fig. 3).

399 In order to account for such natural processes, to identify background levels and to determine  
400 excess trace elements related to anthropogenic contamination, normalized values (trace  
401 elements/Al ratios) are usually applied (Covelli et al., 2006; Romano et al., 2013). Normalized  
402 concentrations of Cr, Cu, Ni, Cd, Zn and Mn in Panzano Bay are low before the 1950s and, together  
403 with As and Pb, increase only in the last 30 years (Fig. 3). Such an increase can reflect the rapid  
404 development of technology and the intensification of agricultural activities during the 20<sup>th</sup> century.

405 Similar vertical trends have been recorded in the Marano Lagoon, located 20km west of Panzano  
406 Bay (Covelli et al., 2013). The Ni concentrations are almost the same in the two areas, while Pb  
407 values are slightly higher in Panzano Bay (starting from 1980 until today). The additional source of  
408 Pb here could come from industrial or port activities (see below).

409 The responses of foraminiferal assemblages to elevated trace element concentrations generally  
410 include declining species abundance and diversity as well as altered taxonomic composition  
411 because more sensitive species die off and more tolerant taxa prevail (Debenay et al., 2000;  
412 Coccioni et al., 2009). Foraminifera can assimilate potentially toxic elements by ingesting  
413 contaminated detritus or algae, but also by incorporating these elements during test crystallization,  
414 leading to test abnormalities (Le Cadre and Debenay, 2006; Frontalini et al., 2009; Martinez-Colón  
415 et al., 2009). In foraminiferal assemblages from Panzano Bay, however, no test abnormalities  
416 occurred, indicating that the threshold of elemental concentrations for such an impact was never  
417 reached during the last 5400 years.

## 418 5.2 Changes in foraminiferal assemblages before the 20<sup>th</sup> century

419 During the period 1600-1900 AD, foraminiferal assemblages in Panzano Bay are characterized by  
420 stable diversity indices and a high, variable abundance of stress tolerant genera and species,  
421 including *Valvulineria* sp., *Ammonia* sp., *A. tepida* and non keeled elphidiids (Figs. 5 and 6F). ~~The~~  
422 ~~only major discrepancy of this trend occurred in the latest 17<sup>th</sup> century, when species richness and~~  
423 ~~Fisher's  $\alpha$  decrease, following an abrupt, short term decline of *Ammonia* sp. (Figs. 5 and 6A). This~~  
424 ~~event approximately coincides with a peak of Cr, Cu, Ni and Cd recorded just above this interval~~  
425 ~~(Fig. 3).~~ The genus *Ammonia* (and especially *A. tepida*) is usually described as being tolerant to all  
426 kinds of stress conditions, including organic and heavy metal pollution (Jorissen, 1988; Coccioni et  
427 al., 1997; Armynot du Châtelet et al., 2004; Ferraro et al., 2006; Frontalini and Coccioni, 2008).  
428 ~~However, some species of this genus (namely *Ammonia parkinsoniana*) are known for their poor~~  
429 ~~tolerance to high levels of trace elements (Jorissen, 1988; Frontalini and Coccioni, 2008; Coccioni~~



430 ~~et al., 2009). Such a sensitivity to increased trace elemental concentrations of certain *Ammonia*~~  
431 ~~species may have caused the decline of *Ammonia* sp. in this interval.~~

432 Non keeled *Elphidium* species prefer an infaunal mode of life (Murray, 2006) and can be associated  
433 with food enrichment of the sediments (Donnici and Serandrei Barbero, 2002; Vidović et al., 2009,  
434 2014). ~~The~~ these requirements are similar to the infaunal genus *Valvulineria*, which is adapted to  
435 large seasonal variability of organic matter and periodic hypoxic conditions (Jorissen, 1987;  
436 Donnici and Serandrei Barbero, 2002; Piva et al., 2008). Moreover, *Valvulineria* is considered to be  
437 representative of environmental conditions prevailing during the “Little Ice Age” (LIA), that  
438 include enhanced rainfall, increased fluvial runoff and increased turbidity (Piva et al., 2008).  
439 Interestingly, the distinct peak of *Valvulineria* in Panzano Bay in the early 19<sup>th</sup> century (Fig. 5)  
440 coincides with the maximal abundances of this genus in sediments from the central and south  
441 Adriatic. This peak is attributed to one of the coldest and most humid phases of the LIA,  
442 characterized by substantially increased river discharge (Piva et al. 2008). The second peak of  
443 *Valvulineria* in Panzano Bay occurred in the 18<sup>th</sup> century (Fig. 5), thus pointing that humid  
444 conditions prevailed in the bay also during this period. To conclude,

445 hHigh abundances of *Ammonia*, non keeled *Elphidium* and *Valvulineria* ~~the above three genera~~  
446 during the 17<sup>th</sup> to 19<sup>th</sup> century suggest fluctuations ~~strong seasonal variations~~ of the river runoff and  
447 organic matter input in Panzano Bay. In contrast to fluctuations in abundance of these foraminiferal  
448 taxa, vertical changes in the concentration of nutrients in the lower and middle part of the core are  
449 mild. However, the spacing of increments that were analyzed for nutrients is larger than dense  
450 spacing of increments analyzed for the composition of foraminiferal assemblages. In addition,  
451 concentrations of nutrients, grain size distribution and vertical changes in foraminiferal assemblages  
452 are likely further affected by vertical homogenization by bioturbation, as evidenced by decadal to  
453 centennial time averaging of *Varicorbula gibba* (Fig. 2), thus making difficult to detect the effects  
454 of environmental fluctuations occurring at higher (seasonal or yearly) temporal resolution.

455 ~~Accordingly, food availability was the primary controlling factor for foraminiferal community~~  
456 ~~composition during this time. This conclusion is supported by the RDA results, which show high~~  
457 ~~correlation between organic enrichment (N<sub>tot</sub>) and assemblage composition (Fig. 8, Table S4).~~  
458 ~~Similar correlations between community composition/diversity and the type of substrate/food~~  
459 ~~availability (higher abundance of infauna in organic rich muddy sediments) have been reported for~~  
460 ~~modern foraminiferal assemblages in the northern Adriatic (Jorissen, 1987; Donnici and Serandrei~~  
461 ~~Barbero, 2002).~~ The foraminiferal community from Panzano Bay is also highly correlated with  
462 several trace elements (Table S3) that ~~naturally~~ accumulated in fine grained sediments during this  
463 period (as discussed above). Although the assemblages show no effects of elevated trace element  
464 concentrations in terms of decline of species abundance or diversity, they remain dominated by taxa  
465 tolerant to both chemical and organic pollution (Fig. 6F), as observed also in other foraminiferal  
466 assemblage in the northern Adriatic (Jorissen, 1987; Donnici and Serandrei Barbero, 2002). These  
467 results imply that the community in Panzano Bay has a long history of adaptation to elevated trace  
468 element concentrations.

### 469 5.3 Changes in foraminiferal assemblages during the 20<sup>th</sup> century

470 With the onset of the 20<sup>th</sup> century, the diversity of foraminiferal assemblages starts to increase  
471 (mainly with the increase of infaunal taxa, as reported in Naeher et al., ~~2014~~2012), but this trend  
472 becomes pronounced only from 1950 AD onwards (Figs. 6A and 6E). However, the overall  
473 assemblage composition in the 20<sup>th</sup> century changed markedly, relatively to the pre-20<sup>th</sup> century  
474 assemblage composition. The uppermost parts of the core show a very strong and directional  
475 change in composition lasting up to the present (Fig. 7), whereas the lower and middle parts of the  
476 core were characterized by a relatively constant taxonomic composition, with a much smaller  
477 multivariate dispersion in NMDS (Fig. 7). Nutrient concentrations (~~TN<sub>tot</sub>~~, TOC and P) display the  
478 same dynamics of increase during this period (Fig. 3), ~~and we interpret these to have caused the~~  
479 ~~observed increase in foraminiferal diversity. The nutrient increase can be attributed both to~~

480 ~~intensifying agricultural and maricultural activities.~~ Although upward increasing concentrations of  
481 TOC and TN can be partly related to their recycling dynamic, the corresponding increase in  
482 pollutants (PAH and PCB) and other observations of major increase in pollutants and organic  
483 enrichments in the Gulf of Trieste (Heath et al., 2006) imply that the nutrient increase also reflects  
484 intensifying agricultural and maricultural activities in the Gulf of Trieste during the 20<sup>th</sup> century.  
485 The increase in diversity observed in the uppermost parts of the core not only correlates with  
486 nutrient enrichment (in accord with observations that early stages of eutrophication can increase  
487 species richness, Martinez-Colón et al., 2009) but also with higher concentrations of pollutants, thus  
488 rather contrasting with the hypothesis that pollution inevitably decreases species richness.

489 Moreover, ~~total~~-TN and P in Panzano Bay sediments are similar to the values measured in  
490 sediments beneath adjacent mussel farms (Rampazzo et al., 2013; Franzo et al., 2014). Mussel  
491 farming here became an important activity by the middle of the 20<sup>th</sup> century, reaching peak  
492 production in 1990 (Melaku Canu and Solidoro, 2014). Intense mussel biodeposition enriches  
493 surface sediments underneath the farms in organic matter, causing anoxic conditions (Rampazzo et  
494 al., 2013). Nonetheless, the impact of this farming does not significantly alter the overall coastal  
495 marine system (Danovaro et al., 2004; Vidović et al., 2009, 2014). Rather, strong winds disperse  
496 and resuspend surface organic rich sediments over the broader area of the gulf (Franzo et al., 2014).  
497 Besides the increase in diversity, the 20<sup>th</sup> century is marked by a taxonomic change in foraminiferal  
498 assemblages: the abundances of *Valvulineria* sp., *Ammonia* sp. and non keeled elphidiids decrease,  
499 whereas *Haynesina* sp. and epiphytic genera (*Miliolinella* sp., *Triloculina* sp.) become more  
500 abundant (Figs. 5 and 6D). Additionally, in the second part of the 20<sup>th</sup> century herbivorous genus  
501 *Ammonia* also slightly increases its abundances, with only *A. tepida* not following this trend.  
502 Relatively higher abundances of these epiphytic, herbivorous genera during this period suggest the  
503 presence of seagrasses or macroalgae meadows near the sampling station (e.g., Langer, 1993;  
504 Mateu-Vicens et al., 2010). Furthermore, a slight shift in the trophic mode of foraminiferal species

505 in the 20<sup>th</sup> century (increase of herbivorous taxa) indicates enhanced phytoplankton, probably  
506 reflecting higher nutrient levels. The distribution of the genera *Miliolinella* and *Triloculina* in the  
507 Gulf of Trieste has already been related to their feeding preference for diatoms in addition to  
508 organic detritus and bacteria (Hohenegger et al., 1993).

509 *Haynesina*, another genus commonly found in the studied sediments, is also herbivorous, known to  
510 be tolerant to high concentrations of organic matter (Debenay et al., 2001; Armynot du Châtelet et  
511 al., 2004; Murray, 2006; Romano et al., 2008). Higher abundances of *Haynesina*, together with the  
512 increase in overall foraminiferal diversity, may be related to the 20<sup>th</sup> century nutrient enrichment  
513 (Fig. 3) because the representatives of this genus indirectly benefit by feeding on enhanced  
514 microalgal biomass (Ward et al., 2003). The faunal shift in dominance of *Valvulineria* to  
515 *Haynesina*, together with higher abundance of epiphytic species, suggests milder seasonal  
516 variations of river discharge and enhanced microalgal biomass as a consequence of nutrient  
517 enrichment. These conclusions are supported by the RDA analysis, pointing to organic enrichment  
518 as a key factor controlling the composition of foraminiferal communities in Panzano Bay (Fig. 8,  
519 Table S41).

520 Finally, the increase in abundance of the suborder Textulariina in this uppermost part of the core  
521 may be the result of taphonomic processes: agglutinated taxa are susceptible to postdepositional  
522 degradation, and the destruction of their tests explains the downcore reduction of their relative  
523 abundances (Diz and Francés, 2009).

#### 524 **5.42. Idrija mercury mine**

525 The activity of the Idria mercury mine is well recorded in Panzano Bay sediments. The Hg  
526 concentrations during the last 5400 years are high and significantly exceed the limits imposed by  
527 the Italian SQG, but also ERL and ERM standards (Fig. 3, Table S2). Interestingly, there are some  
528 distinct trends: the concentrations are considerably higher during the 18<sup>th</sup> century and decrease in  
529 the 19<sup>th</sup> and 20<sup>th</sup> century (Fig. 3), corresponding to the history of the mine: the onset of its

530 significant impact on Panzano Bay occurred in the 18<sup>th</sup> century, when mining activity sharply  
531 increased (Covelli et al., 2012). In the early 19<sup>th</sup> century, metal recovery from the mine improved,  
532 thus releasing less Hg into the river (Covelli et al., 2006).

533 Foraminiferal assemblages in Panzano Bay remained mostly unaffected by these elevated Hg values  
534 throughout the observed period. This implies that speciation of mercury and the bioavailability of  
535 its species are more relevant than its total concentration (Martinez-Colón et al., 2009; Acquavita et  
536 al., 2012). Most of the Hg enters Panzano Bay in particulate (unreactive) form, with only a small  
537 fraction of dissolved Hg (Faganeli et al., 2003). This suggests that the mercury species found here  
538 are not accessible to foraminiferal assemblages or, if they are bioavailable, their concentrations do  
539 not reach values sufficient to produce toxic effects.

### 540 **5.53. The port of Monfalcone**

541 Panzano Bay is also affected by the industrial and port activities of the City of Monfalcone.  
542 Although the first port features were established in the early 19<sup>th</sup> century, the port as it is known  
543 today was designed and built in the 1930s (“CPM”). In 1965, a thermoelectric plant powered by  
544 coal and fuel oil was opened in the industrial area (“The Monfalcone Thermoelectric Plant”). One of  
545 the main byproducts of coal and oil combustion are persistent organic pollutants: polycyclic  
546 aromatic hydrocarbons (PAH) and polychlorinated biphenyl (PCB), contaminants that potentially  
547 form highly carcinogenic and mutagenic derivatives (Notar et al., 2001; Pozo et al., 2009).  
548 Moreover, the use of antifouling paints in ports produces trace elements as residues, namely Cu and  
549 Zn, but also Cd, Cr, Ni and Pb (Singh and Turner, 2009).

550 The presence of PAH and PCB in Panzano Bay sediments is probably related to industrial activities  
551 in the port. Their concentrations are low throughout the core and start to increase in the 19<sup>th</sup> century  
552 and especially since from the middle of the 20<sup>th</sup> century ~~on~~ (Fig. 3), corresponding to the opening of  
553 the thermoelectric plant. However not even the highest measured concentrations exceed Italian  
554 SQG, or ERL and ERM values. In contrast, the concentration peaks of As, Cr, Cu, Ni, Cd, Zn and

555 Pb in the late 20<sup>th</sup> century reflect not only agricultural sources (see above) but also intensifying port  
556 (antifouling paints) and industrial activities (coal and oil burning).

557 Certain changes in foraminiferal taxonomic composition correlate with the concentrations of  
558 persistent organic pollutants (as detected by RDA in Table S4). These include the decrease of the  
559 genus *Valvulineria* and the increase in the abundance of taxa tolerant to chemical pollution (Fig. 6F),  
560 primarily the genus *Ammonia* (Fig. 5). Nevertheless, as the genus *Ammonia* is also known to  
561 tolerate organic enrichment, a synergistic interaction of both processes (chemical and organic  
562 pollution) may have caused such community change.

#### 563 | **5.46. The chronology of environmental changes in Panzano Bay over the last 400 years**

564 Integration of foraminiferal and geochemical proxies, combined with a robust chronological  
565 framework based on extensive radiometric dating of mollus~~ke~~ shells, reveals four major phases in  
566 the recent history of Panzano Bay.

567 During the 17<sup>th</sup> and 18<sup>th</sup> century, the effects of port activities, as well as of agriculture in the  
568 surrounding area on the composition of foraminiferal assemblages are negligible. In the early 18<sup>th</sup>  
569 century, the release of high amounts of mercury into the environment is related to increasing  
570 activity at the Idrija mercury mine (Faganeli et al., 2003). These high inputs, however, did not  
571 affect foraminiferal communities because the dominant particulate form of Hg is not bioavailable.

572 Environmental conditions in Panzano Bay during this period ~~are~~ were probably characterized by  
573 ~~strong seasonal variations~~ fluctuations in the discharge of the Isonzo River ~~runoff~~ and thus in the  
574 amount of variable organic matter input. ~~The~~ foraminiferal community was therefore composed  
575 predominantly of stress tolerant species adapted to such unstable conditions.

576 During the 19<sup>th</sup> century, metal recovery at the Idrija mine improved (Covelli et al., 2006) and less  
577 mercury was released into the bay. The onset of maricultural activities here area dates back to this  
578 period, when bivalve farming was established along the eastern coast of the Gulf of Trieste (Melaku  
579 Canu and Solidoro, 2014). This also marks the construction of the port of Monfalcone. The effects

580 of bivalve farming as well as of agricultural and port activities remain negligible during this period.  
581 The first half of the 20<sup>th</sup> century, however, is marked by rapid technological development. In  
582 Panzano Bay, agricultural, maricultural and port activities intensified. The associated slight increase  
583 of nutrients caused an increase in foraminiferal diversity and a shift in the trophic mode of the  
584 species.

585 In the second part of the 20<sup>th</sup> century, the Monfalcone thermoelectric plant, powered by coal and  
586 fuel oil, became operative. This slightly increased the concentrations of persistent organic pollutants  
587 caused a minor change in the foraminiferal community composition. The nutrient increase that  
588 started in the early 20<sup>th</sup> century extended to this period. As a consequence, the trend of increasing  
589 foraminiferal diversity continues until today.

## 590 **6. Conclusions**

591 The chronology of changes in the geochemical composition of sediments and foraminiferal  
592 assemblages in shallow and sheltered marine embayments of the northern Adriatic reflects  
593 agricultural and industrial development, coastal eutrophication and natural variations. Mercury is a  
594 major pollutant in the area, whose concentrations during the last 5400 years have significantly  
595 exceeded Italian sediment quality guidelines, ERL and ERM. Surprisingly, these high  
596 concentrations have not affected the ecosystem because the mercury species are not bioavailable to  
597 foraminifera.

598 The impact of agricultural, maricultural and industrial activities intensified during the second half of  
599 the 20<sup>th</sup> century and is ongoing. This is reflected in increasing concentrations of trace elements and  
600 persistent organic pollutants (PAH, PCB), as well as in progressive nutrient enrichment, as it was  
601 presumed within the first two hypotheses. However, mining activity did not produce a progressive  
602 enrichment of mercury as anticipated in the second hypothesis, due to the improvement of the  
603 methods for the metal recovery. Increased pollutants did not cause a decline of species abundance  
604 and diversity as suggested by the third hypothesis, as-

605 ~~The~~ foraminiferal response to such anthropogenic impacts in Panzano Bay are shaped by their long  
606 history of adaptation to ~~naturally~~ elevated trace element concentrations, but also as initial stages of  
607 eutrophication can positively affect species richness. Consequently, the shift in community  
608 composition during the 20<sup>th</sup> century reflects a combination of factors, including the recorded  
609 increase of pollutants, varying natural conditions, but also a ~~natural~~, preindustrial predisposition of  
610 foraminifera here to tolerate trace elemental pollution.

611 This combination of factors - and therefore our results - are clearly applicable to many other  
612 shallow coastal areas impacted by human activities, which are largely synchronized ~~on~~ a global  
613 scale. Finally, our approach points to the importance of using long term baseline data for evaluating  
614 the environmental and ecological status of present day marine ecosystems.

#### 615 **Acknowledgements**

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623 suggestions helped us improve the manuscript.

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630 **Table 1.** Results of the forward model selection in the redundancy analysis (10000 permutations  
 631 were used). Proportion of variance explained in the community data ( $R^2$ ), F-statistic and P-values  
 632 from permutation tests are reported for (a) models with a single explanatory variable and (b) for the  
 633 effects of a second variable added to the model already including total nitrogen.

<b>a)</b>				<b>b)</b>			
<b>Variable</b>	<b>Variance explained</b>	<b>F</b>	<b>Pr(&gt;F)</b>	<b>Variable</b>	<b>Variance explained</b>	<b>F</b>	<b>Pr(&gt;F)</b>
<b>TN</b>	<u>0.424</u>	<u>5.885</u>	<u>0.001</u>	<b>clay</b>	<u>0.179</u>	<u>1.522</u>	<u>0.103</u>
<b>PCB</b>	<u>0.410</u>	<u>5.553</u>	<u>0.001</u>	<b>Ni</b>	<u>0.177</u>	<u>1.507</u>	<u>0.104</u>
<b>Zn</b>	<u>0.339</u>	<u>4.105</u>	<u>0.007</u>	<b>Fe</b>	<u>0.177</u>	<u>1.502</u>	<u>0.095</u>
<b>Mn</b>	<u>0.339</u>	<u>4.101</u>	<u>0.002</u>	<b>Cd</b>	<u>0.169</u>	<u>1.421</u>	<u>0.139</u>
<b>Pb</b>	<u>0.328</u>	<u>3.899</u>	<u>0.011</u>	<b>Pb</b>	<u>0.155</u>	<u>1.286</u>	<u>0.217</u>
<b>Cd</b>	<u>0.324</u>	<u>3.835</u>	<u>0.003</u>	<b>PAH</b>	<u>0.155</u>	<u>1.280</u>	<u>0.235</u>
<b>TOC</b>	<u>0.323</u>	<u>3.825</u>	<u>0.008</u>	<b>Cu</b>	<u>0.132</u>	<u>1.062</u>	<u>0.406</u>
<b>Cu</b>	<u>0.266</u>	<u>2.903</u>	<u>0.036</u>	<b>TOC</b>	<u>0.125</u>	<u>0.998</u>	<u>0.472</u>
<b>Ni</b>	<u>0.248</u>	<u>2.644</u>	<u>0.047</u>	<b>Zn</b>	<u>0.124</u>	<u>0.991</u>	<u>0.473</u>
<b>Fe</b>	<u>0.225</u>	<u>2.322</u>	<u>0.057</u>	<b>PCB</b>	<u>0.120</u>	<u>0.951</u>	<u>0.543</u>
<b>PAH</b>	<u>0.225</u>	<u>2.320</u>	<u>0.063</u>	<b>Hg</b>	<u>0.114</u>	<u>0.897</u>	<u>0.587</u>
<b>P</b>	<u>0.185</u>	<u>1.817</u>	<u>0.121</u>	<b>C</b>	<u>0.113</u>	<u>0.891</u>	<u>0.538</u>
<b>Hg</b>	<u>0.168</u>	<u>1.612</u>	<u>0.128</u>	<b>As</b>	<u>0.103</u>	<u>0.807</u>	<u>0.667</u>
<b>Al</b>	<u>0.157</u>	<u>1.491</u>	<u>0.169</u>	<b>Mn</b>	<u>0.084</u>	<u>0.638</u>	<u>0.823</u>
<b>Cr</b>	<u>0.128</u>	<u>1.176</u>	<u>0.269</u>	<b>P</b>	<u>0.072</u>	<u>0.543</u>	<u>0.924</u>
<b>C</b>	<u>0.082</u>	<u>0.712</u>	<u>0.654</u>	<b>Al</b>	<u>0.055</u>	<u>0.411</u>	<u>0.977</u>
<b>clay</b>	<u>0.081</u>	<u>0.702</u>	<u>0.649</u>	<b>Cr</b>	<u>0.052</u>	<u>0.381</u>	<u>0.981</u>
<b>As</b>	<u>0.077</u>	<u>0.665</u>	<u>0.689</u>				

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644 **Figure 1.** Study area and location of sampling site in Panzano Bay.



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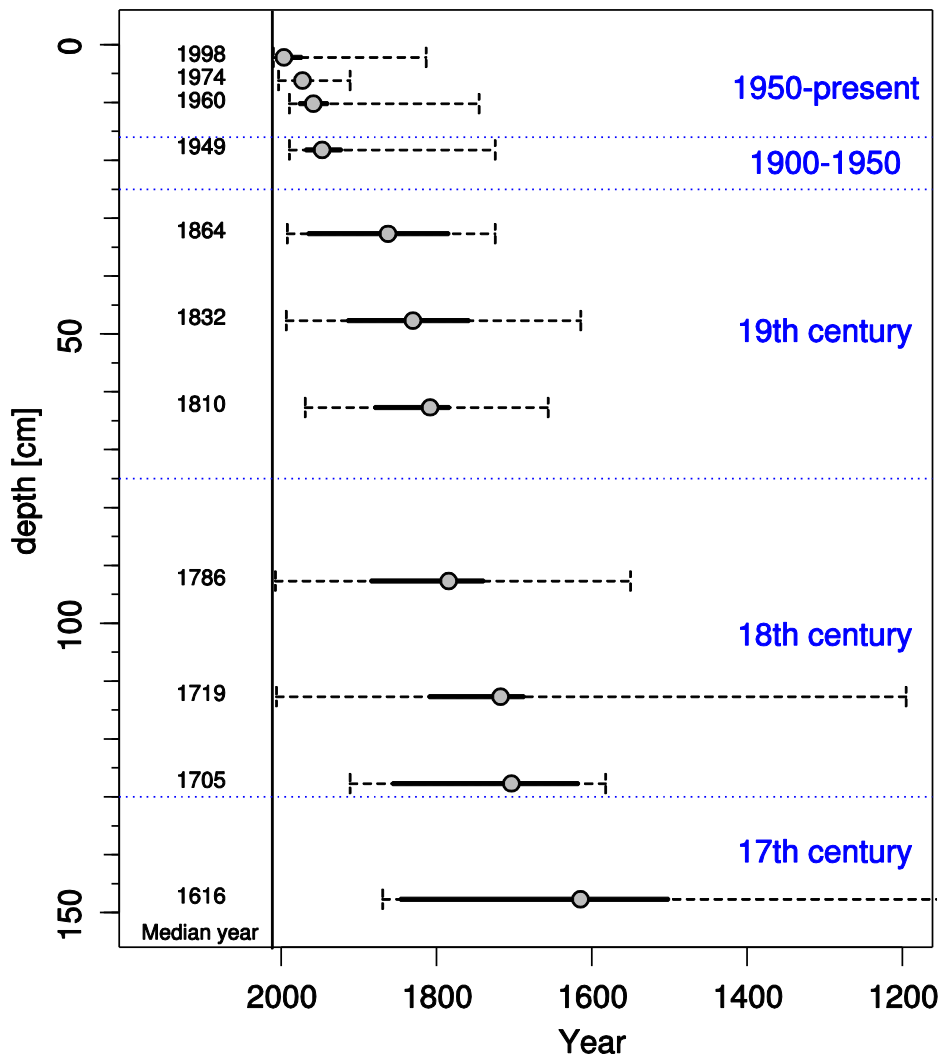
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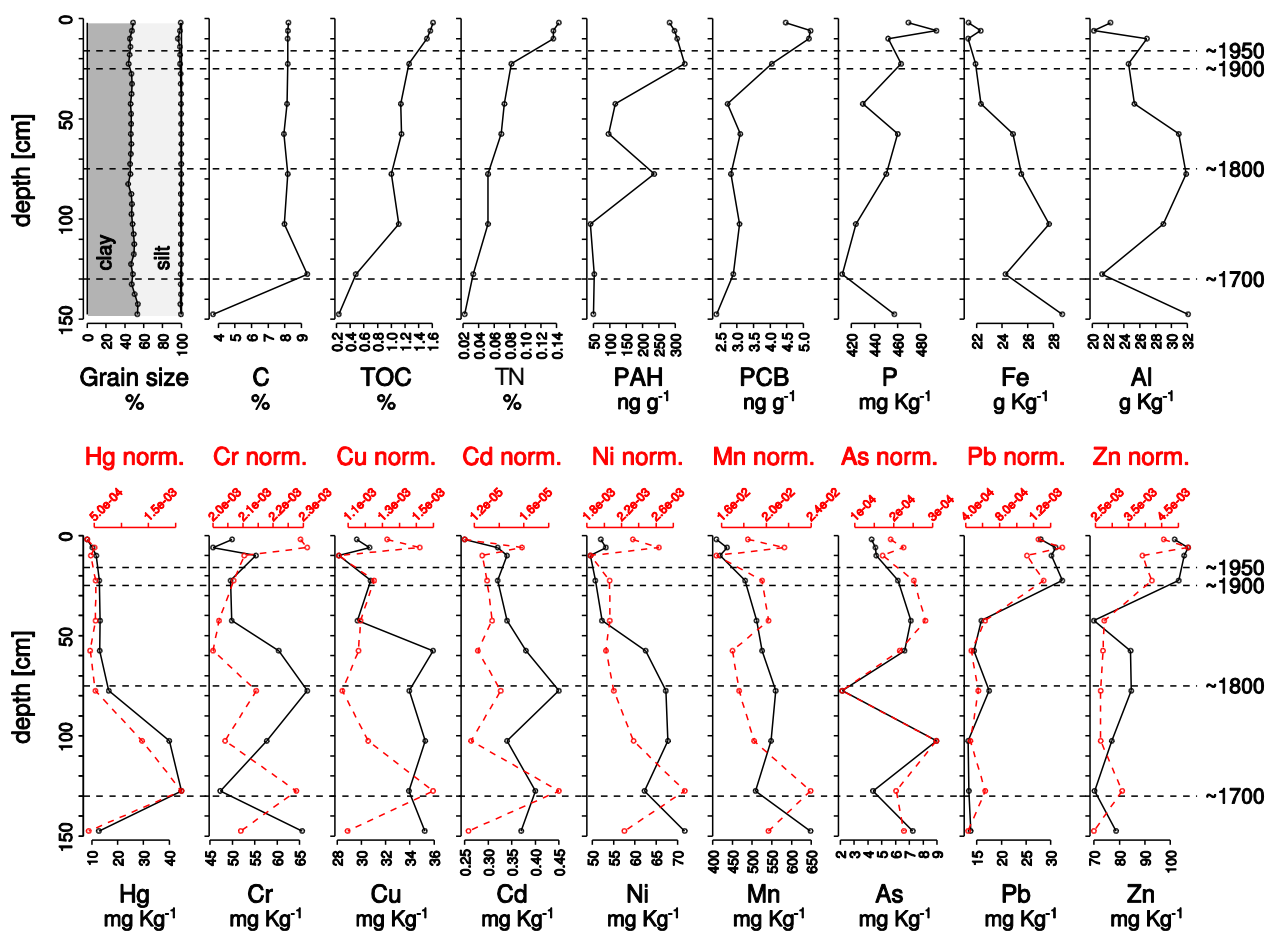
662 **Figure 2.** Radiocarbon calibrated AAR dating of shells of the bivalve species *Varicorbula gibba*.  
 663 The dashed line represent the total range of ages of *V. gibba*, i.e., bounded by the minimum and  
 664 maximum age, the solid line represents the inter-quartile range of ages, i.e., it is bounded by the  
 665 25th and 75th quantiles, and the grey circle refers to the median age. We note that age range and  
 666 inter-quartile age range increase downcore and death assemblages in the basal core increments are  
 667 time-averaged to few centuries, most likely due to bioturbational mixing. Therefore, although  
 668 median age of *V. gibba* in the lowermost (145-150cm) increment is 1616 AD, interquartile range of  
 669 ages of *V. gibba* in this increment includes shells that died in the 16<sup>th</sup> century.



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672 **Figure 3.** Vertical changes in grain size, major, minor and trace elements, nutrients and persistent  
 673 organic pollutants.



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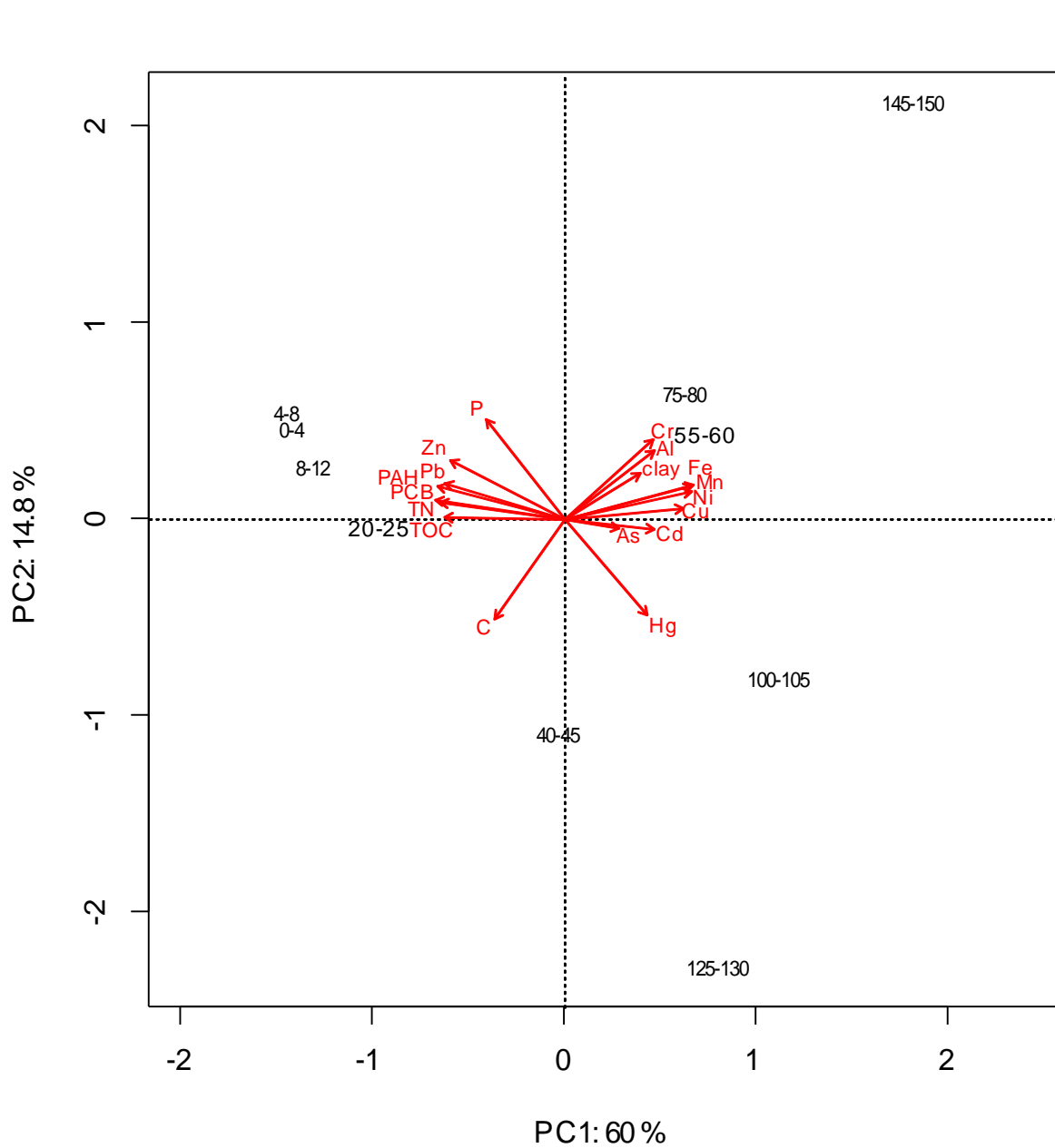
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683 **Figure 4.** Principal component analysis (PCA) plot of geochemical data.



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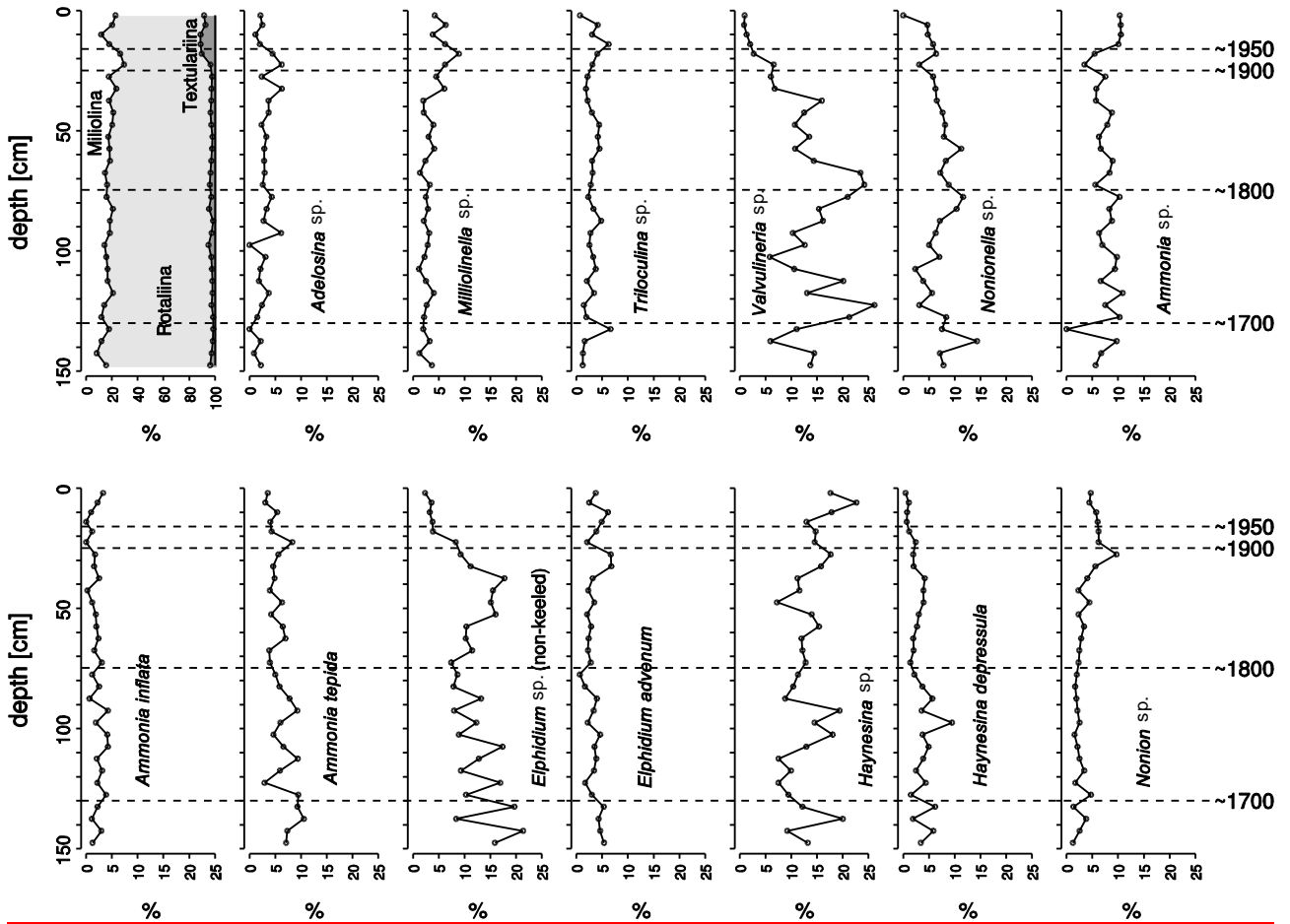
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691 **Figure 5.** Temporal trends in the relative abundance of foraminiferal suborders (following  
 692 suprageneric classification of Loeblich and Tappan, 1987) and dominant genera and species  
 693 (represented by >2 % of individuals in the pooled data).



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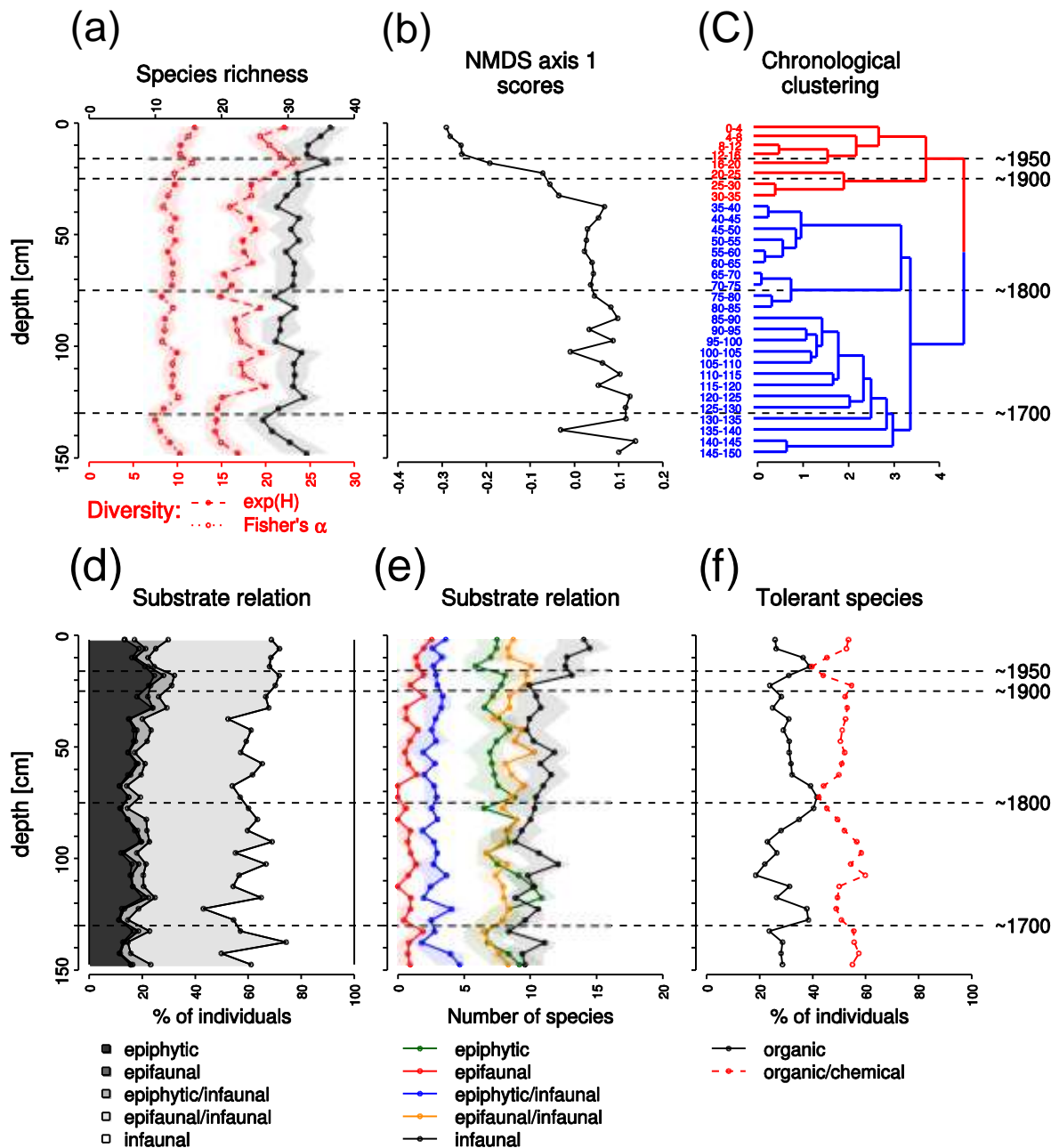
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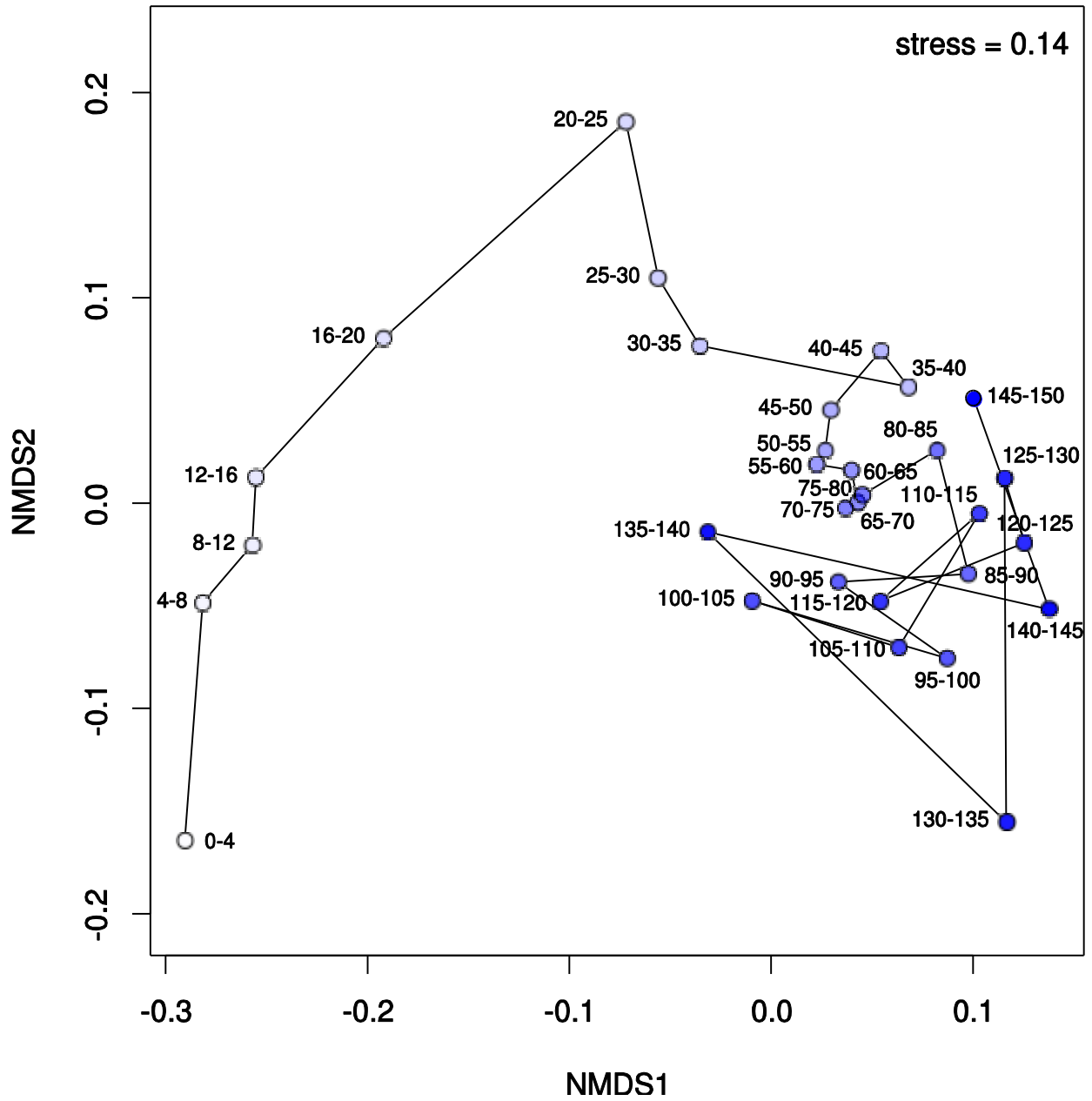
704 **Figure 6.** (a) Temporal trends in species richness, Fisher's  $\alpha$  index, the exponent of the Shannon  
 705 index. Shown are mean values with 95% CI after rarefying to 240 individuals per sample. (b)  
 706 Temporal trends in species composition summarized by NMDS axis 1 scores, (c) Chronological  
 707 clustering of foraminiferal assemblages (different colours group samples separated by a major  
 708 compositional shift, with the number of temporal bins determined by the broken stick model), (d)  
 709 Temporal trends in the relative abundances of substrate relation groups, (e) Temporal trends in the  
 710 rarefied species richness of each substrate relation group, (f) Temporal trends in the relative  
 711 abundances of two foraminiferal groups according to their tolerance to different types of pollution  
 712 (organic and organic/chemical). Figures D-F are plotted based on data in Table S1.



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715 **Figure 7.** Non metric multidimensional scaling ordination of the foraminiferal assemblages.



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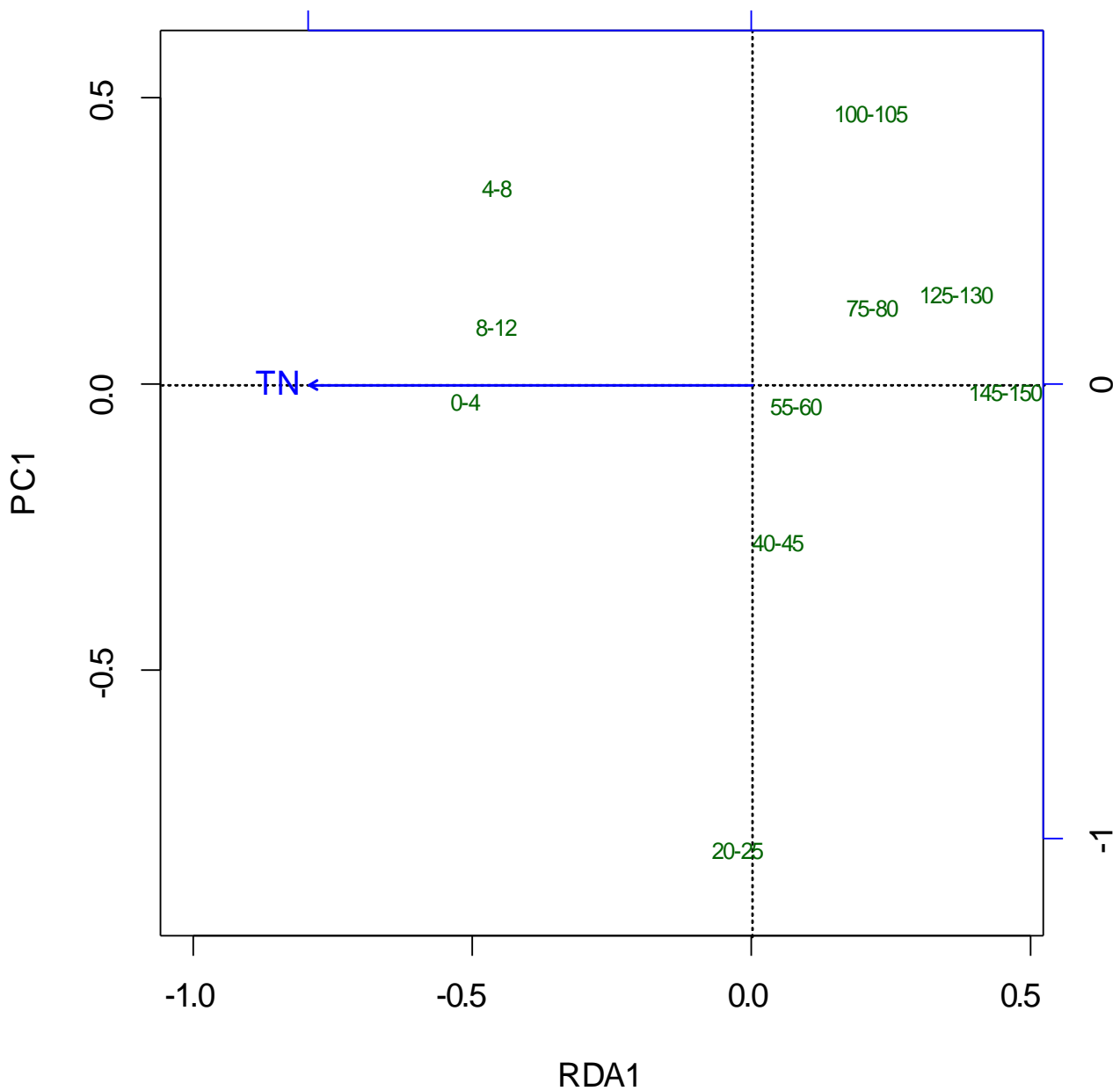
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724 **Figure 8S1.** Redundancy analysis triplot of the foraminiferal and geochemical data based on the  
725 results of the forward model selection. Nitrogen concentrations were the only selected variable. The  
726 first canonical axis explains 42.4 % of the total variance in the data. Nitrogen concentrations were  
727 the only selected variable that explains 42.4 % of the total variance in the foraminiferal community  
728 composition. Given that only one environmental variable is used, the second axis corresponds to the  
729 first principal component that visualizes the residual compositional variation unconstrained by  
730 geochemical data.



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733 **Titles for supplementary tables**

734 **Table S1.** (a) Absolute abundances of foraminiferal species with their ecological characteristics and  
735 tolerances to organic and chemical pollutants, (b) full list of all used sources; (c) diversity of  
736 foraminiferal community including rarefied species richness, Shannon index, the exponential of  
737 Shannon index and Fisher's  $\alpha$ .

738 **Table S2.** Concentrations of major, minor and trace elements, Italian national sediment quality  
739 guidelines (SQG), ERL and ERM standards.

740 **Table S3.** Pearson's correlation coefficients between clay content, geochemical data, NMDS axis 1  
741 scores and rarefied species richness in the subset of samples with measured elemental  
742 concentrations.

743 ~~**Table S4.** Results of the forward model selection in the redundancy analysis. Proportion of variance~~  
744 ~~explained in the community data ( $R^2$ ), F-statistic and P-values from permutation tests are reported~~  
745 ~~for (a) models with a single explanatory variable and (b) for the effects of a second variable added~~  
746 ~~to the model already including total nitrogen.~~

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