

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

3 Jelena Vidović^{a*}, Rafał Nawrot^a, Ivo Gallmetzer^a, Alexandra Haselmair^a, Adam Tomašových^b,
4 Michael Stachowitsch^c, Vlasta Čosović^d and Martin Zuschin^a

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6 ^aDepartment of Palaeontology, University of Vienna, Althanstrasse 14, 1090 Vienna, Austria

7 ^bEarth Science Institute, Slovak Academy of Sciences, Dúbravská cesta 9, 84005 Bratislava, Slovak
8 Republic

9 ^cDepartment of Limnology and Bio-Oceanography, Center of Ecology, University of Vienna,
10 Althanstrasse 14, 1090 Vienna, Austria

11 ^dDepartment of Geology, Faculty of Science, University of Zagreb, Horvatovac 102a, 10 000
12 Zagreb, Croatia

13 *Corresponding author: vidovic.jelena@gmail.com

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 ~500 years of anthropogenic pressure from mining, port and industrial
20 zones in the Gulf of Trieste, Italy. From 1600 to 1900 AD, concentrations of trace element, nutrients
21 and pollutants remain relatively constant and foraminiferal assemblages show moderate fluctuations
22 in abundance of species adapted to high sediment and nutrient supply. The only significant
23 anthropogenic activity during this period is mercury mining in the hinterlands of the gulf, releasing
24 high amounts of mercury into the bay and significantly exceeding the standards on the effects of
25 trace elements to benthic organisms. Nonetheless, the fluctuations in the concentrations of mercury
26 do not correlate with changes in the composition and diversity of foraminiferal assemblages,
27 possibly due to its nonbioavailability. A major change in the composition of foraminiferal
28 assemblages occurs at the onset of 20th century and correlates with an overall increase in organic
29 pollutants and nutrient concentrations. First, intensified agricultural and maricultural activities in

30 the first half of the 20th century are associated with an increase in nutrients and correlate with a
31 minor increase in foraminiferal diversity. Second, intensified port and industrial activities in the
32 second half of 20th century increased the normalized trace element concentrations and persistent
33 organic pollutants (PAH, PCB) in the topmost part of the core. This increase caused only minor
34 changes in the foraminiferal community probably because foraminifera in Panzano Bay were
35 exposed to elevated trace element concentrations for a long time. Our study underlines the
36 importance of using an integrated, multidisciplinary approach in reconstructing the history of
37 environmental and anthropogenic changes in marine systems. Given the prolonged human impacts
38 in coastal areas like the Gulf of Trieste, such long term baseline data are crucial for interpreting the
39 present state of marine ecosystems.

40

41 **Keywords**

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

43

44 **1. Introduction**

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

51 The impact here started nearly 500 years ago with the onset of mercury mining in the hinterland of
52 the bay (Singh and Turner 2009, Covelli et al., 2012), enhanced in the late 20th century with
53 intensifying agriculture and mariculture (Aleffi et al., 2006; Rampazzo et al., 2013; Finch et al.,
54 2014), and continued to the present times with increasing port and industrial activities

55 (thermoelectric plant) of the City of Monfalcone (Notar et al., 2001; Pozo et al., 2009).
56 Such intensive anthropogenic pressures have prompted a growing scientific effort to estimate the
57 effects of pollution on ecosystem composition here. Most attempts have addressed modifications of
58 the marine habitats that occurred in the 20th century, using only geochemical (Horvat et al., 1999,
59 Faganeli et al., 2003; Acquavita et al., 2012) or biological proxies (Solis-Weiss et al., 2007). There
60 is however, a growing tendency towards integrated assessments of its present state (Cibic et al.,
61 2007; Melis and Covelli, 2013, Franzo et al., 2015), but until today there are no multidisciplinary
62 studies assessing the long term history of the environmental changes in the northeastern Adriatic
63 and thus capturing its preindustrial, undisturbed state.
64 Such a historical record requires an integrated geochemical and paleoecological approach. Benthic
65 foraminifera, among the most abundant microorganisms in shallow and marginal marine
66 environments, are often used in paleoecological studies. This is because they are highly sensitive to
67 short term environmental changes (Schönenfeld, 2012), they have a high preservation potential and
68 thus provide an excellent temporal record of ecosystem states over the past hundreds to thousands
69 of years (Yasuhara et al., 2012).
70 The present multidisciplinary study is designed to provide a high resolution historical record of
71 environmental changes in Panzano Bay, to obtain information on the state of the ecosystem prior to
72 the onset of the most intensive impact, and to evaluate the effects of anthropogenic activities in the
73 bay. We obtained geochemical data and foraminiferal assemblages from a 1.6m long sediment core
74 containing a centennial scale record of environmental and anthropogenic changes. The core covers
75 approximately the last 500 years, as indicated by radiocarbon calibrated AAR dating of the mollusks
76 shells.
77 Taking into account the history of potential anthropogenic stressors in Panzano Bay, we assess the
78 following hypotheses: (1) agricultural and maricultural activities produce upcore increases in the
79 concentrations of organic matter, nutrients and trace elements, (2) mining activities and

80 thermoelectric plants generate a progressive enrichment of mercury and persistent organic
81 pollutants, (3) increased pollutants alter the taxonomic composition of foraminiferal assemblages
82 and cause a decline of species abundance and diversity.

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

88 **2. Study area**

89 The Gulf of Trieste is a shallow marine basin in the northernmost part of the Adriatic Sea,
90 occupying an area of about 500km², with an average water depth of 17m and a maximum of about
91 25m (Fig. 1). Seasonal variations of water temperature range between 8 to 24°C at the surface and 8
92 to 20°C in the bottom layer. The salinity of the water in the gulf is typically marine, ranging
93 between 33 and 38.5‰ (Ogorelec et al., 1991).

94 The water enters the gulf in the southeast and continues to the northwest, following the general
95 anticlockwise circulation pattern of the Adriatic Sea. However, the water circulation in the gulf is
96 mostly controlled by tides (range ~0.5m), winds (strong northeastern Bora) and seasonal variations
97 of freshwater inflow. The Isonzo/Soča and Timavo rivers are the most significant sources of
98 freshwater to the Gulf, with average inflows of about 100-130m³/s each (Ogorelec et al., 1991).

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

102 The main sediment supply comes from the Isonzo River in the north and from the weathering of the
103 Paleogene flysch deposits outcropping along the southern coast of the gulf. The sediment
104 accumulation rates are approximately 1 mm/yr in the central part of the gulf and increase to about

105 2.5mm/yr towards the mouth of the Isonzo River located in Panzano Bay (Ogorelec et al., 1991, our
106 unpublished data). Surface sediments in this area are mostly silt clays and clay silts (Zuschin and
107 Piller, 1994) occupied by a high biomass epifauna (Zuschin et al., 1999).

108 The Gulf of Trieste is affected by many sources of organic and inorganic pollutants, coming from
109 agricultural and industrial activities in the hinterland as well as from tourist and maricultural
110 activities along its coasts (Notar et al., 2001; Covelli et al., 2006). Panzano Bay is one of the highly
111 impacted areas, with organic pollution coming from mussel farms located along the eastern part of
112 the Gulf of Trieste (Melaku Canu and Solidoro, 2014) and industrial and port areas of the city of
113 Monfalcone, including a thermoelectric plant and several coal, petroleum and other cargo handling
114 piers (Fig. 1). The Monfalcone thermoelectric plant consists of four thermoelectric generator sets
115 powered by coal and fuel oil and became operative in 1965 ("The Monfalcone Thermoelectric
116 Plant"). Finally, there is substantial Hg pollution originating from the Idrija mercury mine in the
117 hinterlands and delivered to the bay through the Isonzo river flow (Horvat et al., 1999; Notar et al.,
118 2001). Idrija, situated 50km west of Ljubljana (Slovenia), was the second largest Hg mine in the
119 world, operating for nearly 500 years until its definite closure in 1995 (Faganeli et al., 2003; Covelli
120 et al., 2012). During this period, over 5 million tones of Hg ore were mined and much of the
121 residues were spread around the town and its vicinity (Miklavčič, 1999). Most of the Isonzo riverine
122 input of Hg is in particulate form (1500kg/yr), followed by dissolved Hg at 8.6kg/yr (Faganeli et al.,
123 2003). Dissolved mercury is biogeochemically reactive and tends to accumulate in certain seafood
124 from mariculture, presenting social and economic problems for the local population (Faganeli et al.,
125 2003).

126 **3. Methods**

127 **3.1. Sampling**

128 Three sediment cores, two for sedimentological and one for foraminiferal analyses, 1.6m long with
129 a diameter of 9cm, were acquired using an UWITEC piston corer with hammer action (Gallmetzer

130 et al., 2006) from a research vessel in summer 2013. The drilling station is located in the central
131 part of Panzano Bay (45°44,122' N; 13°36,029' E) at a water depth of 12.5m. The uppermost 20cm
132 of each core were sliced into 2cm thick intervals in order to attain high resolution data. The rest of
133 the core was sliced into 5cm thick samples. For analytical purposes and in order to improve
134 compatibility with the lower part of the core, the uppermost 2cm thick samples were merged into
135 4cm thick intervals (reducing the number of samples from 36 to 31). Sediment samples were used
136 to determine grain size, the content of major, minor and trace elements, nutrients and persistent
137 organic pollutants. Core chronology is based on molluscan shells dated by ^{14}C calibrated amino acid
138 racemisation.

139 **3.2. Sediment parameters**

140 The grain size of 36 samples was analyzed using a sedigraph (SediGraph III 5120 Particle Size
141 Analyzer) for the small fractions ($<63\mu\text{m}$) and dry sieving for fractions from $<63\mu\text{m}$ to $>1\text{mm}$. The
142 sediments were classified according to the Shepard's classification (1954).

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

149 To analyze elemental concentrations, each sediment sample was gently squeezed to break down
150 aggregates and screened through a PE sieve to remove particles bigger than 1mm. A part of the
151 screened sediment was dried in an oven at 105°C until reaching a constant weight (to measure water
152 content). The dried sediment was ground to powder using an agate mortar and pestle before further
153 analyzing the contents of heavy metals and As. The sample (about 0.4g d.w.) was digested with 8ml
154 HNO_3 in a microwave oven (Multiwave 3000, Anton Paar, Austria). The digested material was left

155 to cool at room temperature and then filtered through a 0.45 μ m nitrocellulose membrane filter. The
156 filtered digestates were diluted with distilled deionized water to 40ml in a volumetric flask
157 (USEPA, 1994A). The concentrations of the elements (Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Zn)
158 were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES) (Optima
159 2100DV, Perkin Elmer, USA) (USEPA, 1994B). Mercury analyses were carried out using atomic
160 absorption spectrophotometry with cold vapor (Analyst 100, Perkin Elmer, USA) (USEPA, 1976).
161 The quality acceptance protocols required that one blank sample or one certified reference material
162 (BCR-277r estuarine sediment, Community Bureau of Reference) were digested and analyzed with
163 each batch of fifteen samples. The blank results indicated that the analytical procedure was free
164 from contamination because the concentrations of all metals were below the respective method
165 detection limits. Mean recovery from the certified material ranged between 84% (Zn) and 103%
166 (Hg), except for Al (40%) because the extraction method was not strong enough to break crystalline
167 aluminosilicates. The analytical precision, determined using five replicates of homogenized
168 samples, was estimated to be better than 10% for all elements. Calibration for ICP-AES and AAS
169 analysis was achieved with prepared external standards via the standard curve approach. Full
170 calibration was performed after every set of 48 samples. The method detection limit for element
171 analysis was defined as 3 times the standard deviation of 10 blank measurements.
172 Carbon and nitrogen determination was performed following the method of Hedges and Stern
173 (1984), using an elemental analyzer (CHN 2400, Perkin Elmer, USA). The total concentration (TC)
174 and TN) was determined on an aliquot of the sample as is (about 10mg of dry sediment); the
175 organic fraction of carbon (TOC) was determined after treatment of another aliquot of the sample
176 with acid vapors. The inorganic fraction (C) was calculated by the difference between the total and
177 organic concentrations. For the instrument calibration, before each daily series of analyzes, three
178 replicates of Acetanilide standard were performed. For the quality acceptance protocols one blank
179 sample every five samples was analyzed.

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

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

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

205 Raw concentrations of Hg, Cr, Pb, As, Cd and PCB were compared to Italian sediment quality
206 guidelines (SQG), following the directive D. L.vo n. 172 of 13/10/2015, whereas PAH and Ni
207 threshold concentrations were taken from directive DM 367/2003. Additionally, raw concentrations
208 were compared to two sediment quality criteria used around the world: effects range low (ERL),
209 representing the threshold level below which effects to benthic organisms rarely occur, and effects
210 range medium (ERM), above which effects are likely to occur (Burton, 2002). Finally, trace
211 elements were **normalized** to a reference element (Al) in order to compensate for grain size and
212 mineralogical effects on the metal variability in samples (Covelli et al., 2006).

213 **3.3. Foraminiferal analyses**

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

230 (Barras et al., 2014; Alve et al., 2016; Dimiza et al., 2016) and Paleobiology Database
231 (Behrensmeyer and Turner, 2013). A full list of used sources is given in the Supplement: Table S1.

232 **3.4. Statistical analyses**

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

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

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

255 environmental variables and NMDS axis 1 scores for the subset of samples with available values of
256 elemental concentrations.

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

266 To identify the timing of the major shifts in assemblage composition, we performed chronological
267 clustering, a type of constrained cluster analysis that takes into account the temporal sequence of
268 samples (Birks, 2012), by using the CONISS algorithm (constrained incremental sum of squares
269 agglomerative clustering) implemented in “chclust” function from the “rioja” package (Juggins,
270 2015). The number of significantly distinct temporal bins was determined by comparing the amount
271 of variance accounted for by a given number of clusters to a random expectation based on the
272 broken stick model (Bennett, 1996). Clustering was performed on the Bray-Curtis distance matrix
273 based on relative abundance data. All statistical analyses were performed in R 3.2.1 (R Core Team,
274 2014) using “vegan” (Oksanen et al., 2015) and “rioja” (Juggins, 2015) packages.

275 **3.5. Chronological framework**

276 Core chronology is based on the radiocarbon calibrated amino acid racemization dating of the
277 bivalve species *Varicorbula gibba*. First, 13 shells of *V. gibba* were selected for ^{14}C dating and
278 analyzed at the Poznan Radiocarbon Laboratory. Radiocarbon ages were converted to calendar
279 years using Calib7.1 (Stuiver and Reimer, 1993), the Marine13 data (Reimer et al., 2013), and a

280 regional marine reservoir correction (R) in the northeastern Adriatic equal to = -61 years (standard
281 deviation = 50 years) (Siani et al., 2000). The extent of amino acid racemization (AAR) in 329
282 shells was analyzed at Northern Arizona University using reverse phase high pressure liquid
283 chromatography (RP-HPLC) and the procedures of Kaufman and Manley (1998). Thirty specimens
284 of *V. gibba* were randomly selected from eleven, more or less evenly spaced, 4cm or 5cm thick
285 intervals covering the whole core thickness. The rate of AAR was calibrated based on the13 shells
286 dated with ^{14}C and three live collected individuals with the Bayesian model fitting according to
287 Allen et al. (2013). The time dependent reaction kinetic model with the initial D/L value estimated
288 from data and lognormal uncertainty showed the best calibration between D/L values of aspartic
289 acid and calendar ages. AAR data in 18 shells did not pass screening criteria, and ages of 311
290 specimens in total were used for core chronology. Median age of *V. gibba* in the lowermost (145-
291 150 cm) increment is 1616 AD, but interquartile range of ages of *V. gibba* in this increment
292 includes shells that died in the 16th century. Therefore, we used median ages to set the chronology
293 of events, but the core effectively captures the past 500 years of environmental history in Panzano
294 Bay (Fig. 2).

295 **4. Results**

296 **4.1. Sediment parameters and geochemistry**

297 The grain size distribution is rather homogeneous throughout the Panzano Bay core, with only a
298 slight increase in the contribution of the >1mm fraction in the uppermost part (up to 9%). The
299 sediment in the lowermost part of the core is composed of silty clay (50-55% clay). Starting from
300 135cm toward the upper section of the core, the amount of clay decreases to 44-50% and the
301 sediment changes into clayey silt (Fig. 3).

302 Principal component analysis (PCA) based on raw elemental concentrations illustrates the
303 correlation between elements, with the first two axes explaining 74.8% of the variance of the data
304 (Fig. 4). This approach distinguishes two major groups of elements with different vertical

305 distribution trends (Fig. 3, Table S2), and three elements (Hg, As, C) that do not fall into this
306 grouping and have distinct position in the ordination space. The first group comprises trace (Cr, Cu,
307 Ni, Cd, Mn) and conservative elements (Fe, Al), characterized by positive mutual correlations
308 (Table S3), relatively higher concentrations in the lower part of the core and a pronounced decrease
309 in the upper 35cm. The second group includes organic and inorganic pollutants and nutrients whose
310 raw concentrations are stable (Pb, Zn, PCB) or increase only slightly (PCB, TN, TOC, P) in the
311 lower part of the core, but sharply increase in the uppermost 35cm. Concentrations of PAH increase
312 markedly in the upper 35 cm, although they also show high values at 75cm. Normalization to Al
313 reveals two pronounced peaks in the concentrations of the elements from the first group: at 125-
314 130cm core depth and in the uppermost 10cm. The latter peak is also visible in normalized Pb and
315 Zn values (Fig. 3). The concentration of Hg sharply increases from 12.7mg/kg at 145-150cm of the
316 core depth, to 44.7mg/kg at 100-130cm. The Hg values then decrease upcore to a minimum in the
317 surface sediment (8.2mg/kg). Concentrations of As vary in the lower core (2.1-9mg/kg) but
318 gradually decrease to 4.3mg/kg in the surface sediment (0-20cm). Normalization to Al reveals one
319 concentration peak of As in the upper 10cm; it coincides with the peak of all other trace elements.
320 Total carbon remains constant throughout the core (8-9.35%), except for the lowermost part (3.7%
321 C).

322 **4.2. Trends in foraminiferal assemblages**

323 A total of 69 benthic foraminiferal species were identified in the sediments from Panzano Bay, with
324 raw species richness varying between 29 and 41 species in individual samples (26-36 species after
325 rarefaction to 240 individuals; Table S1). The highest percentage of individuals belongs to the
326 suborder Rotaliina (63-89%), followed by Miliolina (8-29%) and Textulariina (1.5-11%). Relative
327 abundances of suborders are generally constant throughout the core and vary notably in the
328 uppermost 20cm only (Fig. 5). Diversity is high throughout the core and increases only in the
329 second half of the 20th century. Values of Fisher α index vary from 7.5 in the lower core to 12 in the

330 uppermost sample; the exponential of Shannon index ranges from 14 to 23 and shows the same
331 vertical trend (Fig. 6A).

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

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

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

355 Within each of the two major groups of samples, further clusters are recognizable, defined by the
356 breaks at 85 and 20cm (Fig. 6C). The lowermost part of the core (150-85cm) corresponds to the
357 period from ~1600 to ~1800 AD and has variable foraminiferal distribution trends. The middle part
358 of the core (85-35 cm, ~1800 to ~1860 AD) is characterized by more stable foraminiferal
359 abundances and a pronounced decline of the genus *Valvulineria*. At 35-20cm (~1860 to 1950 AD)
360 the diversity of the foraminiferal assemblages slightly increases, as do the abundances of epiphytic
361 species. The uppermost sediment (20-0cm, 1950 until today) is characterized by a further increase
362 in biodiversity and in the abundance of textulariids (Figs. 5 and 6A).

363 **4.3. Relationship between foraminiferal assemblages and geochemical proxies**

364 NMDS axis 1 scores are positively correlated with concentrations of Cu, Ni, Cd, Mn, Fe, Al and
365 negatively with total nitrogen and PCB (Table S3). The amount of clay does not correlate with axis
366 1 scores (Table S3). Total nitrogen content explains the highest proportion of variation in
367 assemblage composition (42.4%) and is the only explanatory variable included in the RDA analysis
368 following the forward model selection procedure (Table 1). However, other elements closely
369 (positively or negatively) correlate with total nitrogen content and explain a significant amount of
370 variation in single RDA analyses (Table 1), including TOC, organic pollutants (PAH and PCB), and
371 trace elements (Mn, Fe, Ni, Cu, Cd, Zn). The assemblages from the topmost sediment layers (20th
372 century) are clearly separated from the middle core assemblages and from assemblages at the base
373 along RDA axis 1 (Fig. S1). Therefore, this separation reflects the stratigraphic increase in the
374 content of nitrogen, organic carbon and pollutants and the stratigraphic decline in several trace
375 elements (Fig. 3).

376 **5. Discussion**

377 **5.1. The effects of agricultural and maricultural activities**

378 The agricultural use of pesticides and of organic or inorganic fertilizers releases considerable
379 amounts of pollutants into the environment (Campos, 2003; He et al., 2005; Finch et al., 2014).

380 Pesticides contain pollutant elements such as As, Hg, Cr and Pb (Campos, 2003), while fertilizer
381 contamination includes the discharge of macronutrients (N, P, K) and trace elements, including Co,
382 Cu, Fe, Mn and Zn (Finch et al., 2014). Maricultural activities, in turn, disperse organic matter
383 (waste feed and feces) and nutrients (Mantzavrikos et al., 2007), leading to elevated concentrations
384 of P, TN and TOC in the sediment (Holby and Hall, 1991; Hall et al, 1990, 1992; Mook et al.,
385 2012).

386 In the Panzano Bay sediments, trace (Cr, Cu, Ni, Cd, Mn) and conservative elements (Fe, Al) have
387 relatively higher concentrations in the lower part of the core and a pronounced decrease in the upper
388 35cm. The only discrepancy of this trend occurred in the latest 17th century, when the
389 concentrations of all these elements declined, with simultaneous change in grain size (Fig. 3).
390 Although these elements show some fluctuations that are in phase with grain size variations,
391 absolute changes in sediment grain size are very minor and it is thus difficult to infer changes in
392 environment based on it. Few elements (Cd, Cr and Pb) sporadically and slightly exceed the limits
393 imposed by the Italian SQG (Fig. 3, Table S2). Only Ni has elevated values throughout the core,
394 even when compared to the standards evaluating the effects of trace elements to benthic organisms
395 (ERL and ERM). Ni, Cd and Cr have a high positive correlation with the major constituents of clay
396 minerals (Al and Fe), the main scavengers of trace elements (Romano et al., 2013). This points to a
397 possible grain size and mineralogical effect on the accumulation of these elements throughout the
398 core because the sediment in Panzano Bay is composed of silt and clay 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 elements/Al
401 ratios) are usually applied (Covelli et al., 2006; Romano et al., 2013). Normalized concentrations of
402 Cr, Cu, Ni, Cd, Zn and Mn in Panzano Bay are low before the 1950s and, together with As and Pb,
403 increase only in the last 30 years (Fig. 3). Such an increase can reflect the rapid development of
404 technology and the intensification of agricultural activities during the 20th 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 **500** years.

418 **5.2 Changes in foraminiferal assemblages before the 20th century**

419 During the period 1600-1900 AD, foraminiferal assemblages in Panzano Bay are characterized by
420 stable diversity indices and by a relatively high abundance of stress tolerant genera and species,
421 including *Valvulineria* sp., *Ammonia* sp., *A. tepida* and non keeled elphidiids (Figs. 5 and 6F).
422 The genus *Ammonia* (and especially *A. tepida*) is usually described as being tolerant to all kinds of
423 stress conditions, including organic and heavy metal pollution (Jorissen, 1988; Coccioni et al.,
424 1997; Armynot du Châtelet et al., 2004; Ferraro et al., 2006; Frontalini and Coccioni, 2008). Non
425 keeled *Elphidium* species prefer an infaunal mode of life (Murray, 2006) and can be associated with
426 food enrichment of the sediments (Donnici and Serandrei Barbero, 2002; Vidović et al., 2009,
427 2014). These requirements are similar to those of the infaunal genus *Valvulineria*, which is adapted
428 to large seasonal variability of organic matter and periodic hypoxic conditions (Jorissen, 1987;
429 Donnici and Serandrei Barbero, 2002; Piva et al., 2008). *Valvulineria* is considered to be

430 representative of environmental conditions prevailing during the “Little Ice Age” (LIA), that
431 include enhanced rainfall, increased fluvial runoff and increased turbidity (Piva et al., 2008).
432 Interestingly, the distinct peak of *Valvulineria* in Panzano Bay in the early 19th century (Fig. 5)
433 coincides with the maximum abundances of this genus in sediments from the central and south
434 Adriatic. This peak is attributed to one of the coldest and most humid phases of the LIA,
435 characterized by substantially increased river discharge (Piva et al. 2008). **The second peak of**
436 *Valvulineria* in Panzano Bay occurred in the 18th century (Fig. 5), thus pointing that humid
437 conditions prevailed in the bay also during this period. To conclude, high abundances of *Ammonia*,
438 non keeled *Elphidium* and *Valvulineria* genera during the 17th to 19th century suggest high river
439 runoff and high organic matter input in Panzano Bay. In contrast to fluctuations in abundance of
440 these foraminiferal taxa, vertical changes in the concentration of nutrients in the lower and middle
441 part of the core are mild. However, the spacing of increments that were analyzed for nutrients is
442 larger than dense spacing of increments analyzed for the composition of foraminiferal assemblages.
443 In addition, concentrations of nutrients, grain size distribution and vertical changes in foraminiferal
444 assemblages are likely further affected by vertical homogenization by bioturbation, as evidenced by
445 decadal to centennial time averaging of *Varicorbula gibba* (Fig. 2), thus making difficult to detect
446 the effects of environmental fluctuations occurring at higher (seasonal or yearly) temporal
447 resolution.

448 The foraminiferal community from Panzano Bay is also highly correlated with several trace
449 elements (Table S3) that accumulated in fine grained sediments during this period (as discussed
450 above). Although the assemblages show no effects of elevated trace element concentrations in terms
451 of decline of species abundance or diversity, they remain dominated by taxa tolerant to both
452 chemical and organic pollution (Fig. 6F), as observed also in other foraminiferal assemblage in the
453 northern Adriatic (Jorissen, 1987; Donnici and Serandrei Barbero, 2002). These results imply that
454 the community in Panzano Bay has a long history of adaptation to elevated trace element

455 concentrations.

456 **5.3 Changes in foraminiferal assemblages during the 20th century**

457 With the onset of the 20th century, the diversity of foraminiferal assemblages slightly increased
458 (mainly with the increase of infaunal taxa, as reported in Naeher et al., 2012), but this trend
459 becomes pronounced only from 1950 AD onwards (Figs. 6A and 6E). However, the overall
460 assemblage composition in the 20th century changed markedly, relatively to the pre-20th century
461 assemblage composition. The uppermost parts of the core show a very strong and directional
462 change in composition lasting up to the present (Fig. 7), whereas the lower and middle parts of the
463 core were characterized by a relatively constant taxonomic composition, with a much smaller
464 multivariate dispersion in NMDS (Fig. 7).

465 Nutrient concentrations (TN, TOC and P) increase markedly in the uppermost parts of the core (Fig.
466 3). Although upward increasing concentrations of TOC and TN can be partly related to their
467 recycling dynamic, the corresponding increase in pollutants (PAH and PCB) and other observations
468 of major increase in pollutants and organic enrichments in the Gulf of Trieste (Heath et al., 2006)
469 imply that the nutrient increase also reflects intensifying agricultural and maricultural activities in
470 the Gulf of Trieste during the 20th century. The increase in diversity observed in the uppermost parts
471 of the core not only correlates with nutrient enrichment (in accord with observations that early
472 stages of eutrophication can increase species richness, Martinez-Colón et al., 2009) but also with
473 higher concentrations of pollutants, thus rather contrasting with the hypothesis that pollution
474 inevitably decreases species richness.

475 Moreover, total N and P in Panzano Bay sediments are similar to the values measured in sediments
476 beneath adjacent mussel farms (Rampazzo et al., 2013; Franzo et al., 2014). Mussel farming here
477 became an important activity by the middle of the 20th century, reaching peak production in 1990
478 (Melaku Canu and Solidoro, 2014). Intense mussel biodeposition enriches surface sediments
479 underneath the farms in organic matter, causing anoxic conditions (Rampazzo et al., 2013).

480 Nonetheless, the impact of this farming does not significantly alter the overall coastal marine
481 system (Danovaro et al., 2004; Vidović et al., 2009, 2014). Rather, strong winds disperse and
482 resuspend surface organic rich sediments over the broader area of the gulf (Franzo et al., 2014).
483 Besides the increase in diversity, the 20th century is marked by a taxonomic change in foraminiferal
484 assemblages: the abundances of *Valvulineria* sp. and non keeled elphidiids decrease, whereas
485 *Haynesina* sp. and epiphytic genera (*Miliolinella* sp., *Triloculina* sp.) become more abundant (Figs.
486 5 and 6D). Additionally, in the second part of the 20th century herbivorous genus *Ammonia* also
487 slightly increases its abundances, with only *A. tepida* not following this trend. Relatively higher
488 abundances of these epiphytic, herbivorous genera during this period suggest the presence of
489 seagrasses or macroalgal meadows near the sampling station (e.g., Langer, 1993; Mateu-Vicens et
490 al., 2010). Furthermore, a slight shift in the trophic mode of foraminiferal species in the 20th century
491 (increase of herbivorous taxa) indicates enhanced phytoplankton productivity, probably reflecting
492 higher nutrient levels. The distribution of the genera *Miliolinella* and *Triloculina* in the Gulf of
493 Trieste has already been related to their feeding preference for diatoms in addition to organic
494 detritus and bacteria (Hohenegger et al., 1993).
495 *Haynesina*, another genus commonly found in the sediments in the upper part of the core, is also
496 herbivorous, known to be tolerant to high concentrations of organic matter (Debenay et al., 2001;
497 Armynot du Châtelet et al., 2004; Murray, 2006; Romano et al., 2008). Higher abundances of
498 *Haynesina*, together with the increase in overall foraminiferal diversity, may be related to the 20th
499 century nutrient enrichment (Fig. 3) because the representatives of this genus indirectly benefit by
500 feeding on enhanced microalgal biomass (Ward et al., 2003). The faunal shift in dominance of
501 *Valvulineria* to *Haynesina*, together with higher abundance of epiphytic species, suggests milder
502 seasonal variations of river discharge and enhanced microalgal biomass as a consequence of
503 nutrient enrichment. These conclusions are supported by the RDA analysis, pointing to organic
504 enrichment as a key factor controlling the composition of foraminiferal communities in Panzano

505 Bay (Fig. S1, Table 1).

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

510 **5.4. Idrija mercury mine**

511 The activity of the Idria mercury mine is well recorded in Panzano Bay sediments. The Hg
512 concentrations during the last 500 years are high and significantly exceed the limits imposed by the
513 Italian SQG, but also ERL and ERM standards (Fig. 3, Table S2). Interestingly, there are some
514 distinct trends: the concentrations are considerably higher during the 18th century and decrease in
515 the 19th and 20th century (Fig. 3), corresponding to the history of the mine: the onset of its
516 significant impact on Panzano Bay occurred in the 18th century, when mining activity sharply
517 increased (Covelli et al., 2012). In the early 19th century, metal recovery from the mine improved,
518 thus releasing less Hg into the river (Covelli et al., 2006).

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

526 **5.5. The port of Monfalcone**

527 Panzano Bay is also affected by the industrial and port activities of the City of Monfalcone.
528 Although the first port features were established in the early 19th century, the port as it is known
529 today was designed and built in the 1930s (“CPM”). In 1965, a thermoelectric plant powered by

530 coal and fuel oil was opened in the industrial area ("The Monfalcone Thermoelectric Plant"). One of
531 the main byproducts of coal and oil combustion are persistent organic pollutants: polycyclic
532 aromatic hydrocarbons (PAH) and polychlorinated biphenyl (PCB), contaminants that potentially
533 form highly carcinogenic and mutagenic derivatives (Notar et al., 2001; Pozo et al., 2009).
534 Moreover, the use of antifouling paints in ports produces trace elements as residues, namely Cu and
535 Zn, but also Cd, Cr, Ni and Pb (Singh and Turner, 2009).

536 The presence of PAH and PCB in Panzano Bay sediments is probably related to industrial activities
537 in the port. Their concentrations are low throughout the core and start to increase in the 19th century
538 and especially since the middle of the 20th century (Fig. 3), corresponding to the opening of the
539 thermoelectric plant. However not even the highest measured concentrations exceed Italian SQG, or
540 ERL and ERM values. In contrast, the concentration peaks of As, Cr, Cu, Ni, Cd, Zn and Pb in the
541 late 20th century reflect not only agricultural sources (see above) but also intensifying port
542 (antifouling paints) and industrial activities (coal and oil burning).

543 Certain changes in foraminiferal taxonomic composition correlate with the concentrations of
544 persistent organic pollutants (as detected by RDA in Table 1). These include the decrease of the
545 genus *Valvulineria* and the increase in the abundance of taxa tolerant to chemical pollution (Fig.
546 6F), primarily the genus *Ammonia* (Fig. 5). Nevertheless, as the genus *Ammonia* is also known to
547 tolerate organic enrichment, a synergistic interaction of both processes (chemical and organic
548 pollution) may have caused such community change.

549 **5.4. The chronology of environmental changes in Panzano Bay over the last 500 years**

550 Integration of foraminiferal and geochemical proxies, combined with a robust chronological
551 framework based on extensive radiometric dating of mollusk shells, reveals four major phases in the
552 recent history of Panzano Bay.

553 During the 17th and 18th century, the effects of port activities, as well as of agriculture in the
554 surrounding area on the composition of foraminiferal assemblages are negligible. In the early 18th

555 century, the release of high amounts of mercury into the environment is related to increasing
556 activity at the Idrija mercury mine (Faganeli et al., 2003). These high inputs, however, did not affect
557 foraminiferal communities because the dominant particulate form of Hg is probably not
558 bioavailable. Environmental conditions in Panzano Bay during this period were probably
559 characterized by fluctuations in the discharge of the Isonzo River and thus in the amount of organic
560 matter input. The foraminiferal community was therefore composed predominantly of stress tolerant
561 species adapted to such unstable conditions.

562 During the 19th century, metal recovery at the Idrija mine improved (Covelli et al., 2006) and less
563 mercury was released into the bay. The onset of maricultural activities here area dates back to this
564 period, when bivalve farming was established along the eastern coast of the Gulf of Trieste (Melaku
565 Canu and Solidoro, 2014). This also marks the construction of the port of Monfalcone. The effects
566 of bivalve farming as well as of agricultural and port activities remain negligible during this period.
567 The first half of the 20th century, however, is marked by rapid technological development. In
568 Panzano Bay, agricultural, maricultural and port activities intensified. The associated slight increase
569 of nutrients caused an increase in foraminiferal diversity and a shift in the trophic mode of the
570 species.

571 In the second part of the 20th century, the Monfalcone thermoelectric plant, powered by coal and
572 fuel oil, became operative. This slightly increased the concentrations of persistent organic pollutants
573 caused a minor change in the foraminiferal community composition. The nutrient increase that
574 started in the early 20th century extended to this period. As a consequence, the trend of increasing
575 foraminiferal diversity continues until today.

576 **6. Conclusions**

577 The chronology of changes in the geochemical composition of sediments and foraminiferal
578 assemblages in shallow and sheltered marine embayments of the northern Adriatic reflects
579 agricultural and industrial development, coastal eutrophication and natural variations. Mercury is a

580 major pollutant in the area, whose concentrations during the last 500 years have significantly
581 exceeded Italian sediment quality guidelines, ERL and ERM. Surprisingly, these high
582 concentrations have not affected the ecosystem because the mercury species are not bioavailable to
583 foraminifera.

584 The impact of agricultural, maricultural and industrial activities intensified during the second half of
585 the 20th century and is ongoing. This is reflected in increasing concentrations of trace elements and
586 persistent organic pollutants (PAH, PCB), as well as in progressive nutrient enrichment, as it was
587 presumed within the first two hypotheses. However, mining activity did not produce a progressive
588 enrichment of mercury as anticipated in the second hypothesis, due to the improvement of the
589 methods for the metal recovery. Increased pollutants did not cause a decline of species abundance
590 and diversity as suggested by the third hypothesis, as foraminiferal response to such anthropogenic
591 impacts in Panzano Bay are shaped by their long history of adaptation to elevated trace element
592 concentrations, but also as initial stages of eutrophication can positively affect species richness.
593 Consequently, the shift in community composition during the 20th century reflects a combination of
594 factors, including the recorded increase of pollutants, varying natural conditions, but also a natural,
595 preindustrial predisposition of foraminifera here to tolerate trace elemental pollution.

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

600 **Acknowledgements**

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606 Zonta from the Institute of Marine Sciences in Venice, Italy, carried out the geochemical analyses.
607 Finally, our gratitude goes to Dr. Fabrizio Frontalini and the two anonymous reviewers, whose
608 suggestions helped us improve the manuscript.

609

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

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

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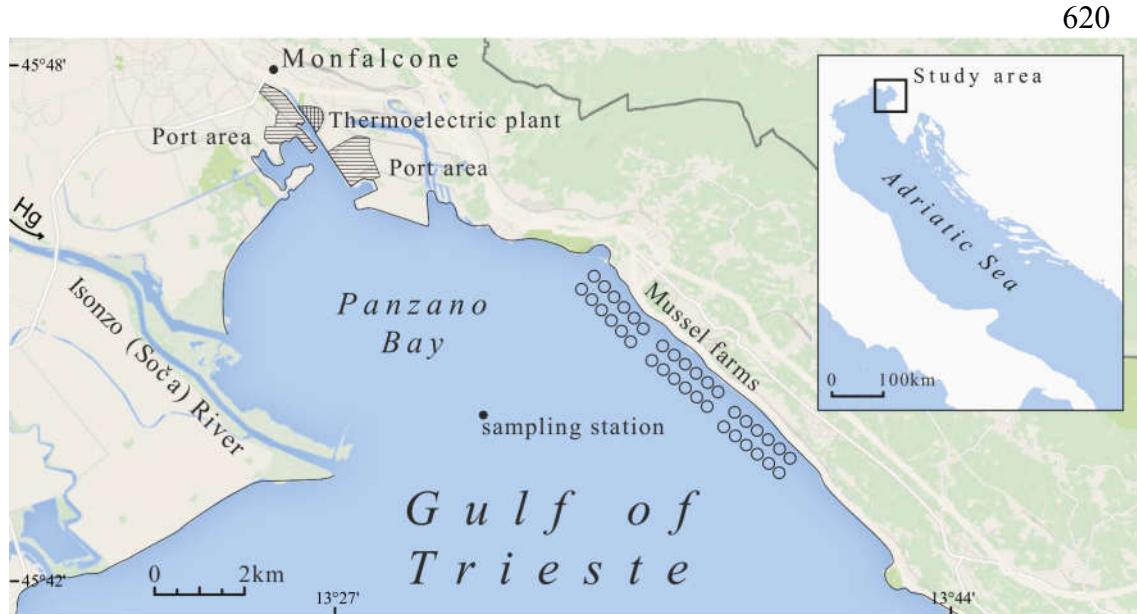
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619 **Figure 1.** Study area and location of sampling site in Panzano Bay.



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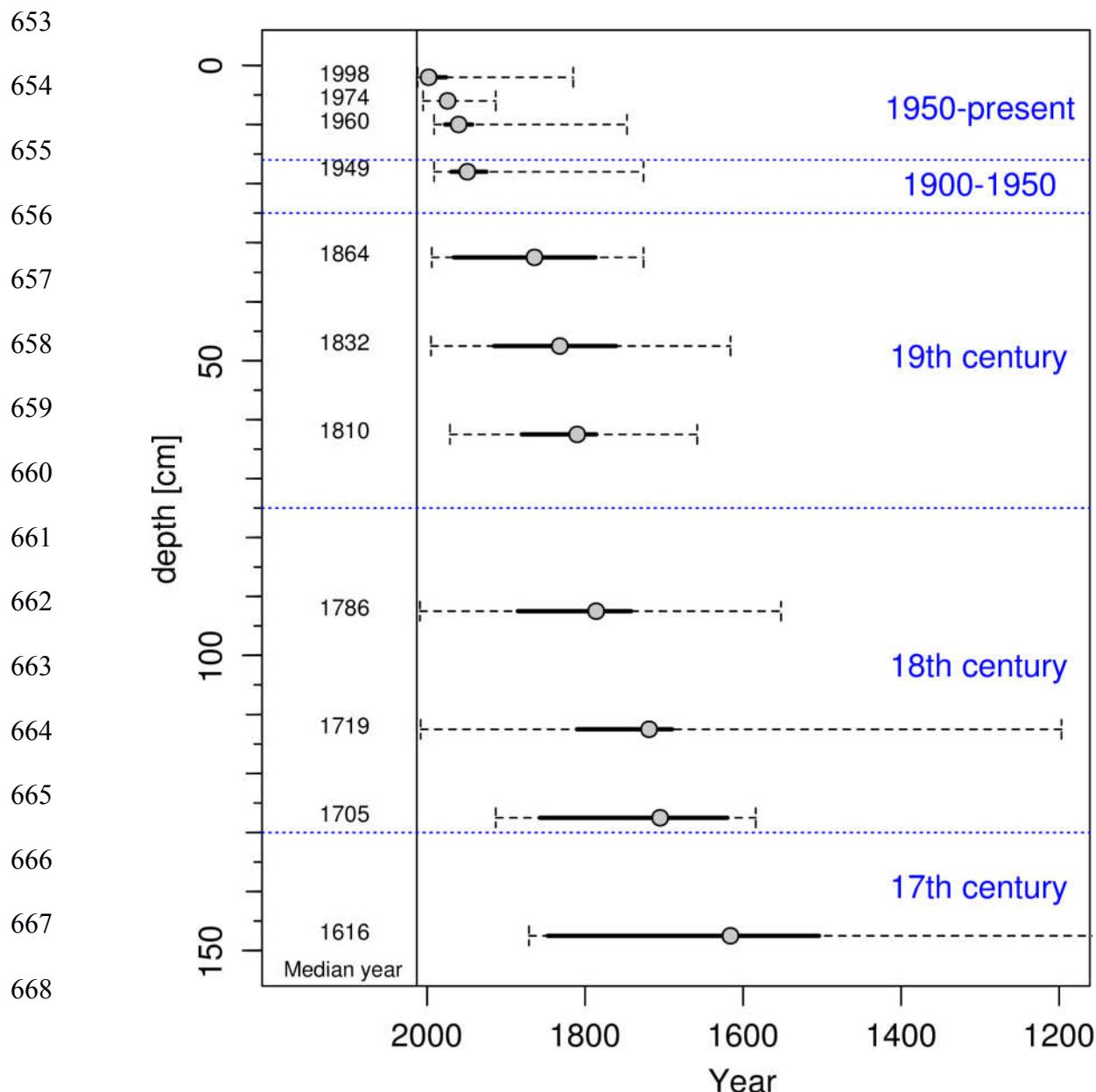
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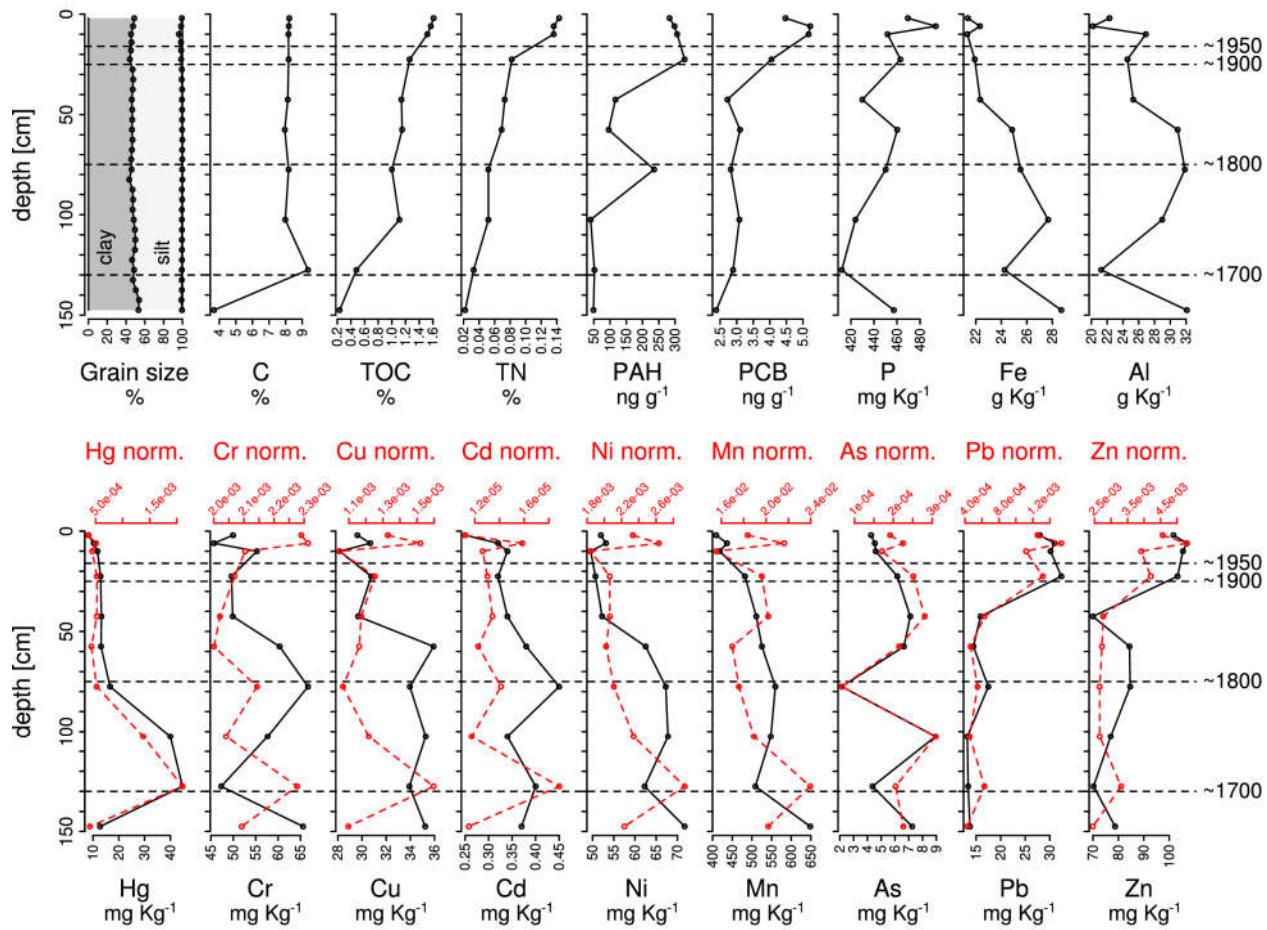
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644 **Figure 2.** Radiocarbon calibrated AAR dating of shells of the bivalve species *Varicorbula gibba*.
 645 The dashed line represent the total range of ages of *V. gibba*, i.e., bounded by the minimum and
 646 maximum age, the solid line represents the inter-quartile range of ages, i.e., it is bounded by the
 647 25th and 75th quantiles, and the grey circle refers to the median age. We note that age range and
 648 inter-quartile age range increase downcore and death assemblages in the basal core increments are
 649 time-averaged to few centuries, most likely due to bioturbational mixing. Therefore, although
 650 median age of *V. gibba* in the lowermost (145-150 cm) increment is 1616 AD, interquartile range of
 651 ages of *V. gibba* in this increment includes shells that died in the 16th century.

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



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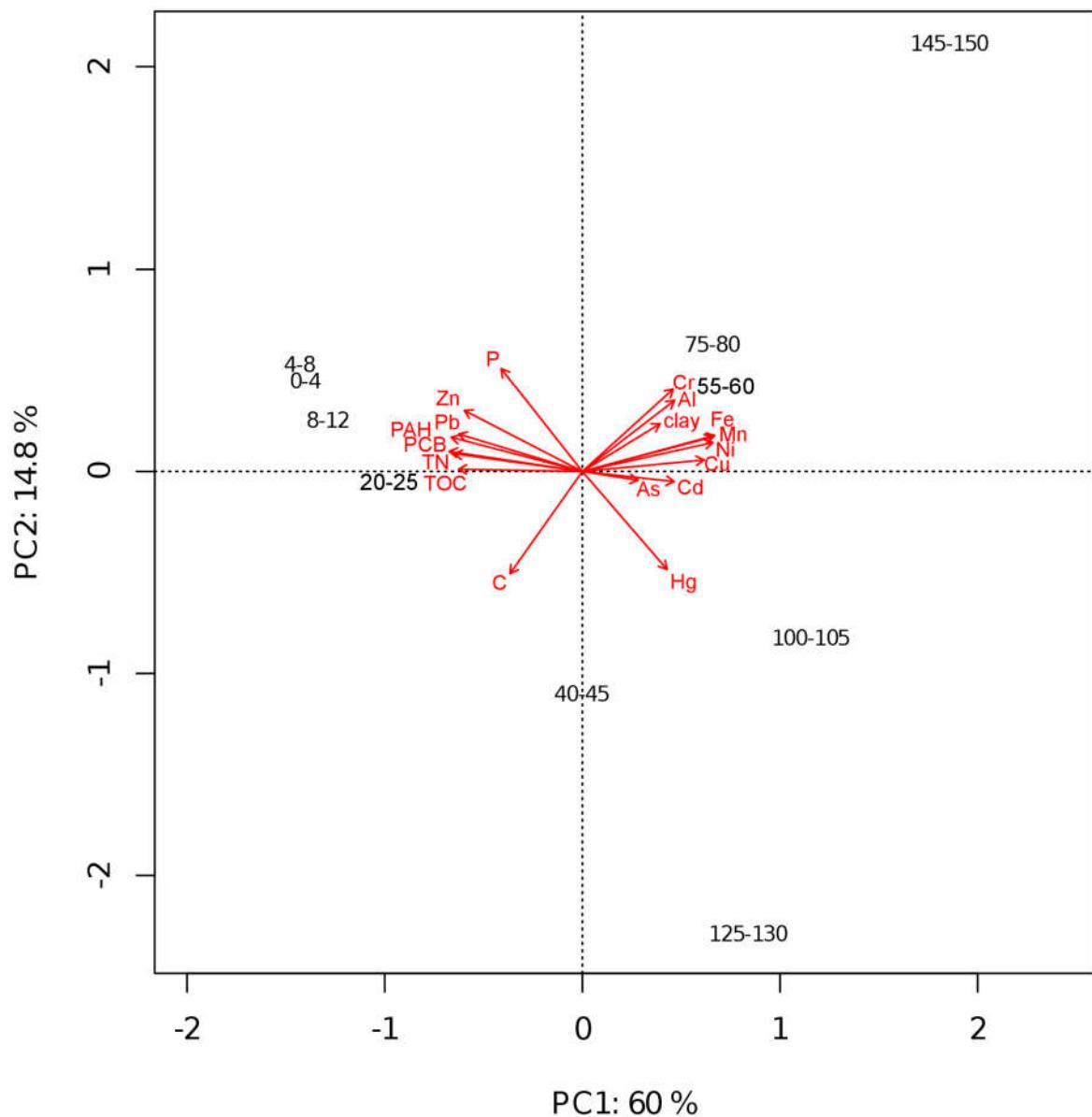
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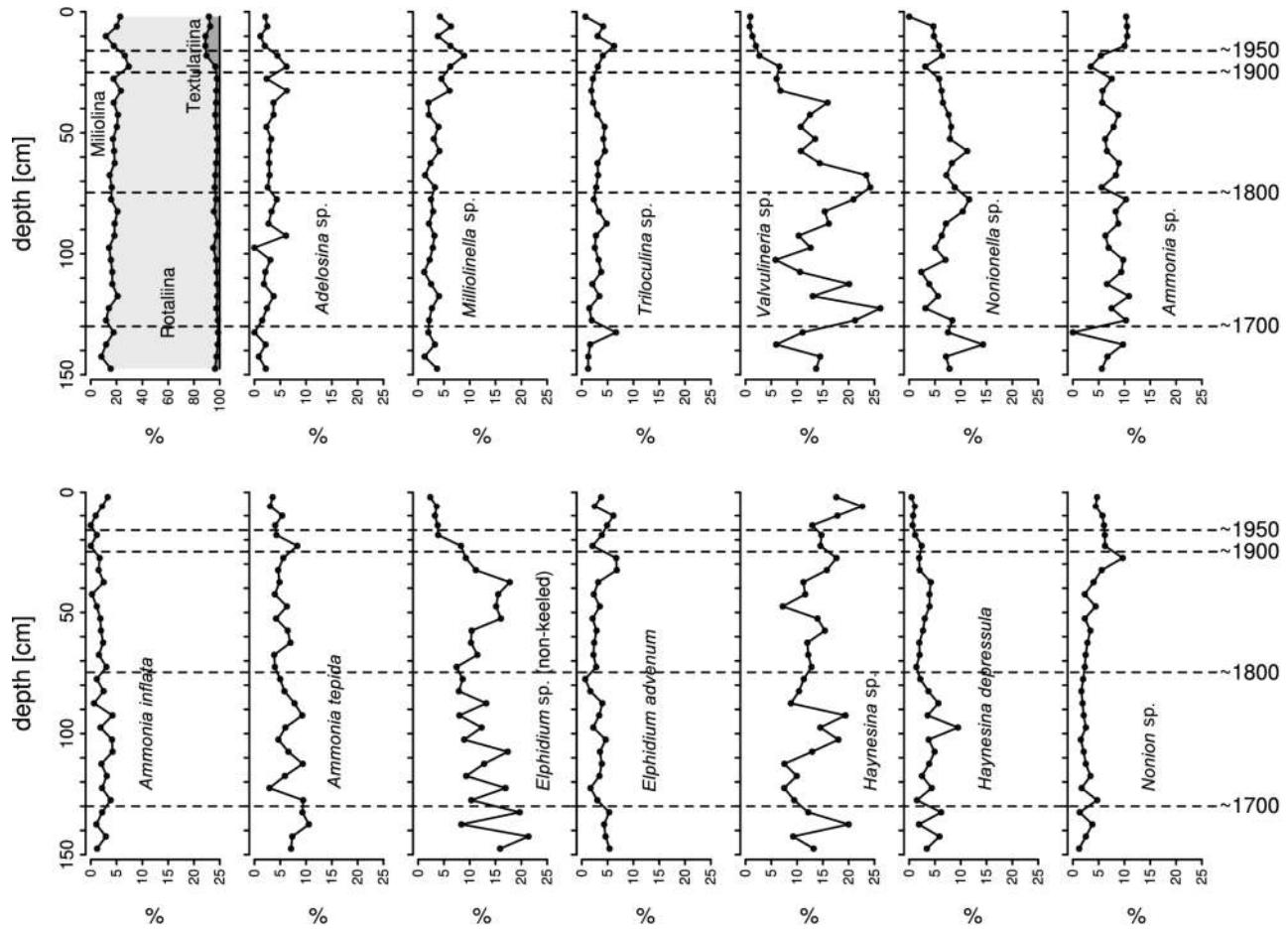
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679 **Figure 4.** Principal component analysis (PCA) based on concentrations of 17 geochemical variables
680 observed in nine increments shows marked differences between the lower and upper parts of the
681 core.



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



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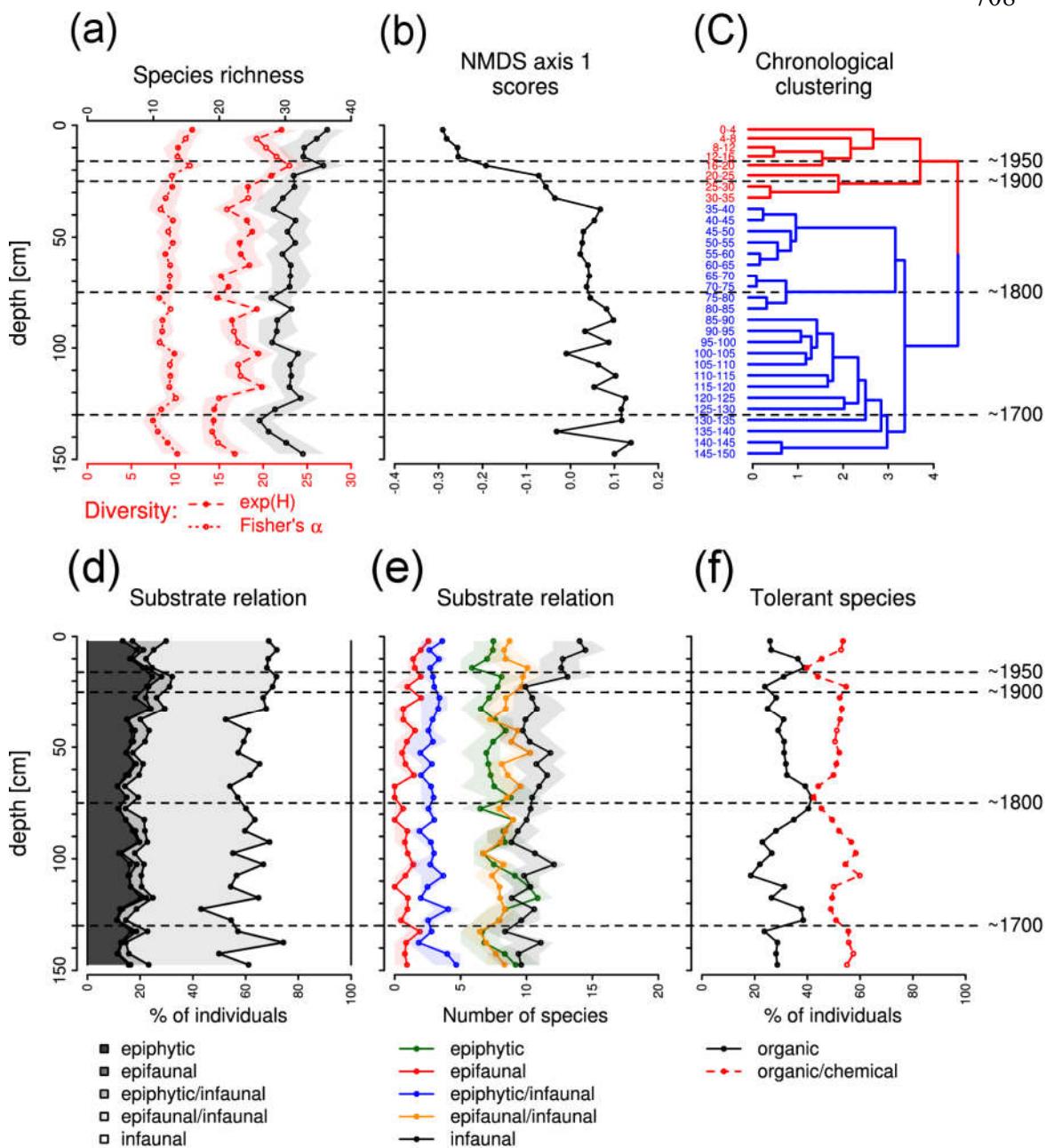
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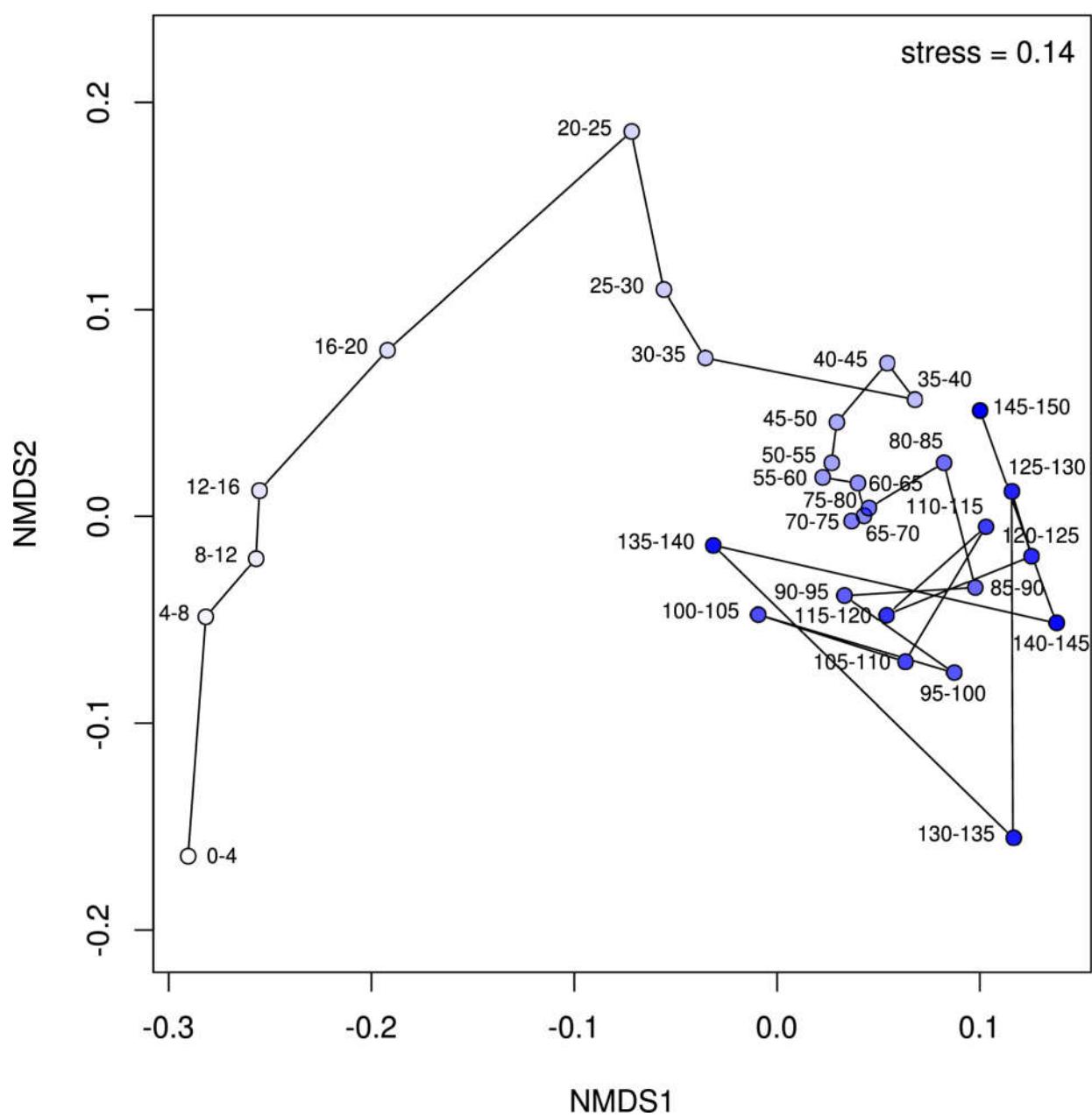
698 **Figure 6.** (a) Temporal trends in species richness, Fisher's α index, the exponent of the Shannon
 699 index. Shown are mean values with 95% CI after rarefying to 240 individuals per sample. (b)
 700 Temporal trends in species composition summarized by NMDS axis 1 scores, (c) Chronological
 701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

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



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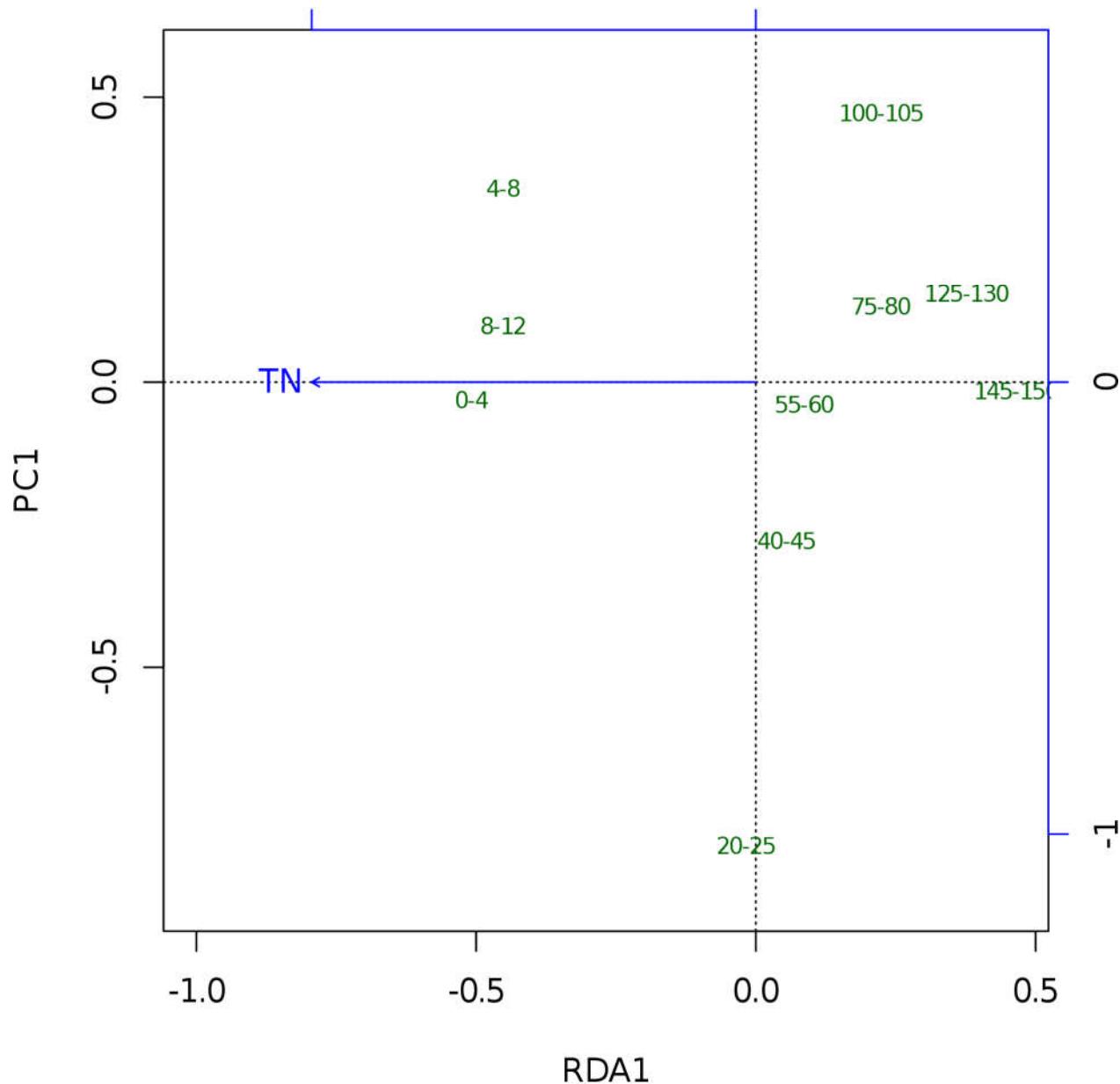
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733 **Figure S1.** Redundancy analysis triplot of the foraminiferal and geochemical data based on the
734 results of the forward model selection. Nitrogen concentrations were the only selected variable that
735 explains 42.4 % of the total variance in the foraminiferal community composition. Given that only
736 one environmental variable is used, the second axis corresponds to the first principal component
737 that visualizes the residual compositional variation unconstrained by geochemical data.



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741 **Titles for supplementary tables**

742 **Table S1.** (a) Absolute abundances of foraminiferal species with their ecological characteristics and
743 tolerances to organic and chemical pollutants, (b) full list of all used sources; (c) diversity of
744 foraminiferal community including rarefied species richness, Shannon index, the exponential of
745 Shannon index and Fisher's α .

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

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

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