Heavy metal uptake of near-shore benthic foraminifera during multi-metal culturing experiments

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Abstract. Heavy metal pollution originating from anthropogenic sources, e.g., mining, industry and extensive land use, is increasing in many parts of the world and influences coastal marine environments ~~for a long time~~even after the source has ceased pollution. The elevated input of heavy metals into the marine system potentially affects the biota because of their toxicity, persistence and bioaccumulation. An emerging tool for environmental applications is the heavy metal incorporation into foraminiferal ~~tests~~calcite tests, which facilitates monitoring of anthropogenic footprints on recent and past environmental systems. The aim of this study wa~~i~~s to investigate whether the incorporation of heavy metals in foraminifera is a direct function of their concentration in seawater. Culturing experiments with a mixture of dissolved chromium (Cr), manganese (Mn), nickel (Ni), copper (Cu), zinc (Zn), silver (Ag), cadmium (Cd), tin (Sn), mercury (Hg) and lead (Pb) in artificial seawater were carried out over a wide concentration range to assess the uptake of heavy metals by the near-shore foraminiferal species *Ammonia aomoriensis*, *Ammonia batava* and *Elphidium excavatum*. Seawater analyse~~i~~s ~~exhibited~~ revealed ~~the~~ increasing ~~metal~~ concentration~~ss~~ for most metals between culturing phases ~~between culturing phases~~and ~~revealed~~ high metal concentrations in the beginning of the culturing phases due to the punctual metal addition. Furthermore, a loss of metals during the culturing process was discovered, by an offset~~which lead to a deviation~~ between the ~~expected~~ added and the actual concentrations of the metals in seawater. Laser ablation ICP-MS analysis of the newly formed calcite revealed species-specific differences in the incorporation of heavy metals. The foraminiferal calcite of all three species exhibited Pb and Ag concentrations ~~reveals a~~ strongly correlated with ~~positive correlation with Pb and Ag~~ concentrations in the seawater culturing medium (partition coefficients and standard deviation for Ag: *Ammonia aomoriensis*=0.50 ±0.02, *Ammonia batava*=0.17 ±0.01, *Elphidium excavatum*=0.47 ±0.04; for Pb: *Ammonia aomoriensis*=0.39 ±0.01, *Ammonia batava*=0.52 ±0.01, *Elphidium excavatum*=0.91 ±0.01). *Ammonia aomoriensis* further showed a correlation with Mn and Cu, *A. batava* with Mn and Hg and *E. excavatum* with Cr and Ni, and partially also with Hg. However, Zn, Sn and Cd showed no clear trend for the species studied, which in case of Sn was ~~may~~ be caused by the ~~little~~ lack of variation of the seawater Sn concentration ~~se metals in seawater~~. ~~Our~~ The calibrations and the calculated partition coefficients render *A. aomoriensis*, *A. batava* and *E. excavatum* as natural archives that enable the determination of variations of some heavy metal concentrations in seawater ~~direct quantification of metals~~ in polluted and pristine environments. ~~This in turn allows monitoring of the ecosystem status of areas that are potentially under the threat of anthropogenic pollution in order to evaluate contemporary emission reduction measures.~~

1 Introduction

Particular heavy metals e.g., zinc (Zn), iron (Fe), molybdenum (Mo), cobalt (Co) and copper (Cu) serve as micronutrients (e.g., Hänsch and Mendel, 2009) for eukaryotic life and play an important role for metabolism, growth, reproduction and enzymatic activity of organisms (e.g., Martín-González et al., 2005; Gallego et al., 2007). Other metals like mercury (Hg), on the other hand, are not known to have any positive effect on the body and are therefore believed to have a higher toxic potential (Jan et al., 2015). All these metals occur naturally in the environment as geogenic traces in soils, water, rocks and, consequently, in plants and animals. However, at higher concentrations, most heavy metals become toxic and have hazardous effects on marine biota (Stankovic et al., 2014). Heavy metals are defined herein as elements with a density >7 g/cm^3 (Venugopal and Luckey, 1975) and an atomic number beyond calcium (Bjerrum, 1936; Thornton, 1995). Furthermore, they are highly persistent in the marine environment and ~~can be hardly degraded~~are not easily excreted by organisms after the uptake of these metals into their system and cells (Flora et al., 2012; Kennish, 2019). Coastal environments act as natural catchments for anthropogenic pollutants because these areas are directly affected by industry, agriculture and urban runoff (e.g., Alloway, 2013; Julian, 2015; Tansel and Rafiuddin, 2016).

In marginal seas and coastal areas, benthic foraminifera are common, and ~~they~~ the chemical composition of their calcite test can be used as proxies for changing environmental parameters like water temperature (Mg/Ca; e.g., Nürnberg et al., 1995; 1996), salinity (Na/Ca; e.g., Wit et al., 2013, Bertlich et al., 2018) and ~~, oxygen content or~~ redox conditions (Mn/Ca; Groeneveld and Filipsson, 2013b; Koho et al., 2015; 2017; Kotthoff et al., 2017; Petersen et al., 2018; Guo et al., 2019). Foraminifera take up heavy metals and incorporate them into their calcium carbonate shells during calcification (e.g., Boyle, 1981; Rosenthal et al., 1997; Dissard et al., 2009; 2010a; ~~2009;~~ 2010b; Munsel et al., 2010; Nardelli et al., 2016; Frontalini et al., 2018a; 2018b; Titelboim et al., 2018; Smith et al., 2020). Moreover, foraminifera have a short life cycle (< 1 year; e.g., Haake, 1967; Boltovskoy and Lena, 1969; Wefer, 1976; Murray, 1992) and thus, react immediately to changing environmental conditions and contamination levels of the surrounding environment. Therefore, foraminifera archive environmental signals and fossil records from sediments can be used to determine parameters of interest throughout space and time.

Species of the foraminiferal genera *Elphidium* and *Ammonia* are among the most abundant foraminiferal taxa in ~~near shore~~intertidal and shelf environments worldwide. They are found from subtidal water depths to the outer continental shelves (Murray, 1991). Furthermore, their calcite tests are often well preserved in the fossil record (Poignant et al., 2000; McGann, 2008; Xiang et al., 2008) and therefore provide the opportunity to assess past environmental conditions. The combination of all these properties make foraminifera, and especially *Elphidium* and *Ammonia* species, suitable indicators of anthropogenic pollution (e.g., Sen Gupta et al., 1996; Platon et al., 2005). As such, this group of organisms are excellent candidates for monitoring the spatial and temporal distribution of heavy metals in seawater to evaluate, for example, the effectiveness of contemporary measures of reducing emissions caused by anthropogenic inputs.

The majority of culturing studies on heavy metal incorporation into benthic foraminifera were designed to assess the influence and uptake of one particular metal, e.g., manganese (Mn) (Barras et al., 2018), copper (Cu) (De Nooijer et al., 2007), chromium (Cr) (Remmelzwaal et al., 2019), lead (Pb) (Frontalini et al., 2015), zinc (Zn) (e.g., Smith et al., 2020), mercury (Hg) (Frontalini et al., 2018a) or cadmium (Cd) (Linshy et al., 2013). This approach is adequate to detail the effects on shell chemistry, growth or physiology. Only ~~two~~one studi~~es~~y reported ~~a~~ culturing experiment~~s~~ with elevated levels of Cu, Mn and Ni ~~in the same culturing medium~~ (Munsel et al., 2010)

77 and elevated levels of Mn, Ni and Cd (Sagar et al., 2021b) in the same culturing medium. However, in reality there

78 is rarely only one metal polluting environments one but mostly instead a combination of several pollutants that

79 occur in nearly all environments affected by heavy metals pollution is usually found (e.g., Mutwakil et al., 1997;

80 Cang et al., 2004; Vlahogianni et al., 2007; Huang et al., 2011; Wokhe, 2015; Saha et al., 2017). How foraminifera

81 incorporate and react to heavy metals when they are co-exposed to more than one metal at a time is less constrained

82 to date. A mixture of different metals will lead to interactions, which may result in a more severe damage of tissue

83 than exposure to each of them individually (Tchounwou et al., 2012). For example, a co-exposure to arsenic and

84 cadmium causes a more distinct damage of human kidneys than only one of these elements (Nordberg et al., 2005).

85 Furthermore, a chronic low-dose exposure to multiple elements can cause similar synergistic effects (e.g., Wang

86 et al., 2008). It is therefore reasonable to assume that other organisms are likewise threatened more harmfully

87 harmed more when exposed to several potentially toxic elements simultaneously.

88 Here we present results from culturing studies with *Ammonia aomoriensis*, *Elphidium excavatum exvacatum* and

89 *Ammonia batava* assddressing the relationship betweenof heavy metal concentrations in seawater and

90 foraminiferal tests. The partitioning factor between the concentration of an element in the ambient seawater and

91 the calcium carbonate of the foraminifers is constrained by determining both the dissolved metal concentrations

92 in water and the metal contents of individual chambers of the foraminiferal shell that have been precipitated in the

93 culturing medium. In particular, foraminifera were grown while exposed to a combination of ten different heavy

94 metals, i.e., cadmium (Cd), copper (Cu), chromium (Cr), lead (Pb), manganese (Mn), mercury (Hg), nickel (Ni),

95 silver (Ag), tin (Sn) and zinc (Zn) over a range of concentrations that prevail in polluted near-shore environments

96 today. These metals are the most common representatives of marine heavy metal pollution (Alve, 1995; Martinez-

97 Colon et al., 2009). Once the carbonate/seawater metal partitioning coefficients are known, investigations of the

98 chemistry of benthic foraminiferal shells offer a reliable method to monitor short-term changes in the chemistry

99 and bioavailabilityof the concentration of toxic elementsheavy metals in seawater.

100

101 **2 Material and Methods**
102 **2.1 Field sampling**
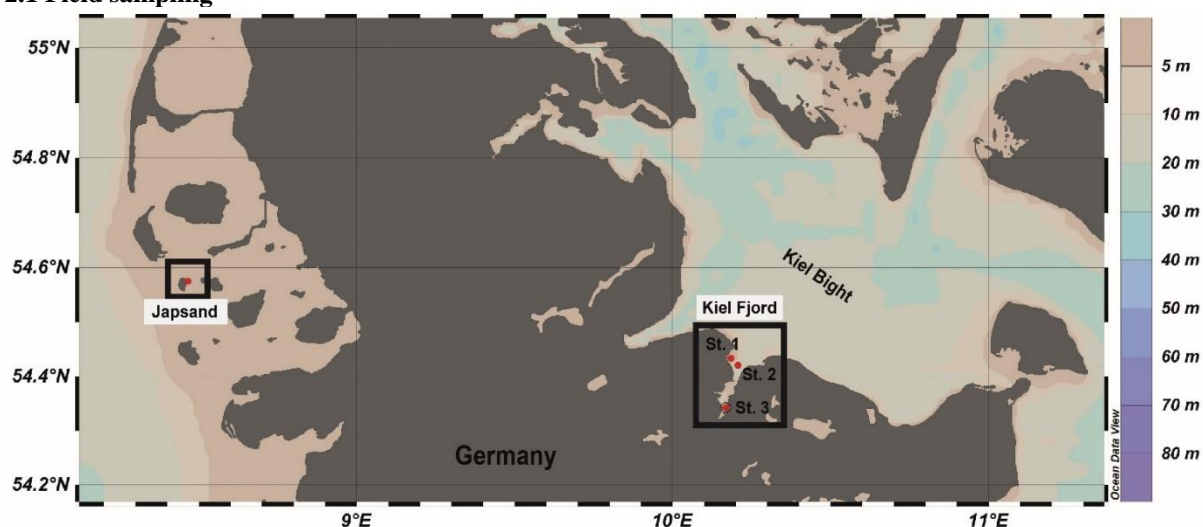
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104 **Figure 1**: Location of the sampling stations in the North Sea (Japsand area) and in the Baltic Sea (Kiel Fjord, St.1

3

105 Strander Bucht, St. 2 Laboe, St. 3 Mönkeberg). The map was drawn with Ocean Data View (Schlitzer, 2016) on

106 the basis of bathymetric data. Water d~~D~~epths in m are indicated by the colo~~u~~r scale~~code~~.

107

2.1.1 North Sea, Japsand

109 Living specimens of *A. batava* were collected at the barrier sand Japsand near Hallig Hooge in the German Wadden

110 Sea in July 2019 at two stations (St. 1: 54°34.480´N, 8°27.919´E; St. 2: 54°34.491´N, 8°27.895´E) (Fig. 1). The

111 sediment was a glacial till or Eemian clay at Station 1 and fine to medium sand at Station 2. Temperature and

112 salinity of seep waters were measured with a WTW 3210 conductivity meter in excavated holes in the vicinity.

113 The temperature at Station 1 was 21.1 ° C and at Station 2 21.6 ° C, respectively. Salinity was 34 PSU at station 1

114 and 33.6 PSU at station 2. The samples were recovered during low tide by scrapping off the uppermost centimetre

115 of the surface sediment with a spoon made out of stainless steel. Natural seawater (NSW) with a salinity of 30.3

116 PSU was collected near the sites for further processing of the samples. Once back on the nearby island Hallig

117 Hooge, the sediment was washed with NSW through stacked sieves with a mesh size of 2000 and 63 µm. The

118 2000 µm sieve was used to remove larger organisms and excess organic material (macroalgae, gastropods,

119 lugworms etc.) that could have induced anoxic conditions in the sediment during transport and storage. The residue

120 was stored in Mucasol soap-washed and acid-cleaned Emsa CLIP and CLOSE® boxes, sparged with air and some

121 algae food was provided. Back in the laboratory at GEOMAR, the residue was stored at 8 °C in a fridge until

122 culturing. These stock cultures were fed twice a week with green-coloured *Nannochloropsis* concentrate

123 (BlueBioTech) and water was partly exchanged with NSW from the sampling site once a week.

124

2.1.2 Baltic Sea, Kiel Bight

126 Living specimens of *A. aomoriensis* and *E~~.~~ excavatum* were collected from different stations in Kiel Fjord, western

127 Baltic Sea (St.1, Strander Bucht, 54°26.001´N, 10°11.1078´E; St. 2, Laboe, 54°25.254´N, 10°12.346´E; St. 3,

128 Mönkeberg, 54°20.752´N, 10°10.150´E; water depth: 12.5 m, 12.3 m and 14.3 m, respectively) in September and

129 October 2019 with F.B. Polarfuchs and F.S. Alkor, respectively (Fig. 1). A Rumohr corer (inner diameter 55 mm)

130 was used on F.B. Polarfuchs and 9 cores were taken (2 at St. 1 and 7 at St. 3). ~~One core at both stations was used~~

131 ~~for foraminiferal assemblage analysis and the first 2 cm of the~~ The sediment from ~~all other~~the cores was collected

132 in Mucasol treated and acid-cleaned plastic containers with NSW from the site.

133 ~~The sediment surface was nearly horizontal and comprised a ~ 5 mm thick fluffy layer consisting of organic~~

134 ~~detritus of a dark brownish color. Mussels, worm burrows and plant debris was found. The sediment underneath~~

135 ~~the surface layer was a very fine mud. The redox boundary was shallower than 0.5 to 1 cm as indicated by the~~

136 ~~color turning black underneath this depth, and the sediment smelling of H₂S.~~

137 On F.S. Alkor, a Reineck box corer was used (200 x 250 m~~e~~m) and 3 replicates at each station were taken (St. 1-

138 —3). The first 1 to 2 cm of the sediment surface of the box core were scrapped off with a spoon made out of

139 stainless steel and the material was stored in a Mucasol treated and acid-cleaned plastic box with NSW from the

140 location. ~~Additional samples for foraminiferal assemblage analysis were taken at each station.~~

141 Back in the laboratory at GEOMAR, the samples were treated the same way as Japsand samples from the North

142 Sea. Artificial seawater (ASW, Tropic Marin) with a salinity of 30 PSU was used for washing and storage of the
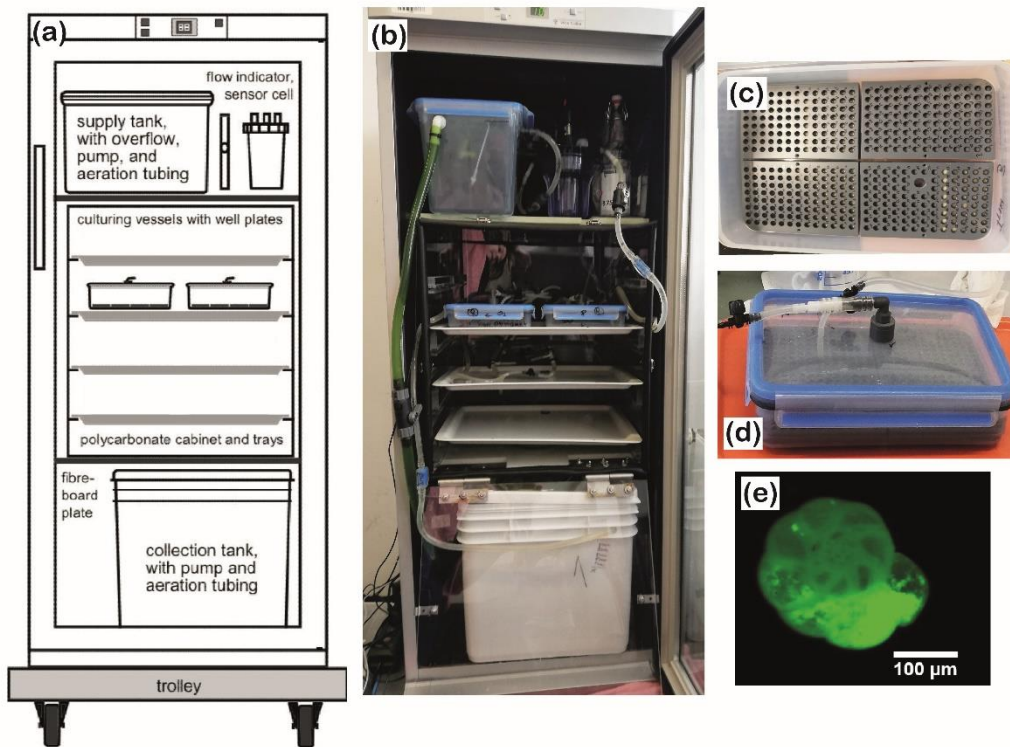
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surface samples from Kiel Fjord. The use of artificial seawater ensured that no harmful microorganism could invade the cultures.

2.2 Culturing setup
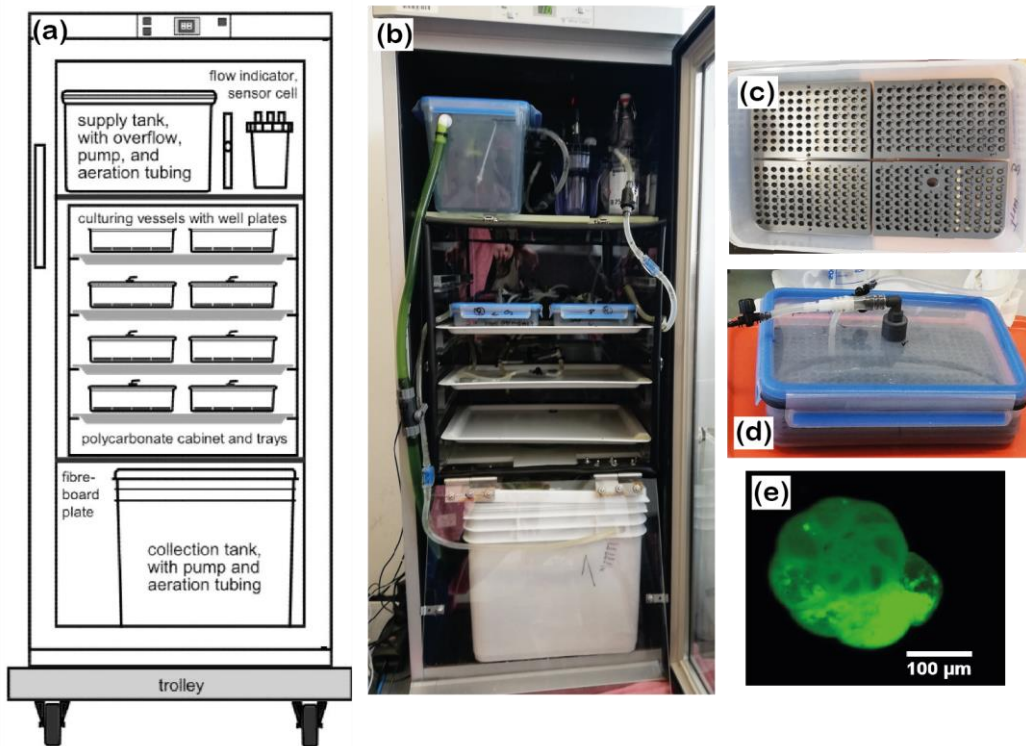
Figure 2: Culturing setup. a: conceptual draft (left, modified after Dagan et al., 2016) and b: assembly of the system (right). Tubing and hoses were omitted from the draft for clarity. c: a well plate with mounted specimens

151 and sand, d: closed culturing vessel with well plates and conduits. e: with calcein stained foraminifer under a

152 fluorescence microscope. Please note that the last 2 ½ chambers are labelledThe chambers formed in calcein are

153 and fluorescing brightly. The specimen shown in the picture was dead, cleaned and dried, which ensured that the

154 test itself and not the cytoplasm showed the fluorescence.

155

2.2.1 Picking of the samples

157 The three foraminiferal species that were used in this study have been described in detail in the literature (e.g.,

158 Lutze, 1965; Nikulina et al., 2008; Schweizer et al., 2011; Francescangeli et al., 2021; Schmidt and Schönfeld,

159 2021). For extracting the foraminiferal specimens from the sediment, about 1 cm^3 of the 63 to 2000 µm size

160 fraction was transferred to a petri dish. For maintaining optimal conditions for the foraminifera, the petri dish was

161 filled with artificial seawater (ASW) with a salinity of 30 PSU. All living specimens were picked with a paint

162 brush from this subsample and collected in a small petri dish of 55 mm diameter with ASW. The procedure was

163 repeated until the whole sample residue was screened.All plastic utensils were treated with Mucasol water and

164 rinsed with 5% HNO$_3$ prior to use. The paintbrush was cleaned with ethanol to protect the culture from harmful

165 microorganisms. Only specimens with a glossy, transparent and undamaged test were chosen. Furthermore, only

166 individuals with the cytoplasm present in more than just a couple of chambers that were connected and included

167 the innermost chambers were chosen. After picking, a drop of concentrated food (pure culture of *Nannochloropsis*,

168 green coloured algae) was added and the foraminifera were left untouched for a night.

169 Specimens that met one or more of the following criteria were considered as living and used for further procedures:

170 • The cytoplasm of the specimens was present in more than two chambers that were connected and
171   including the innermost chambers,

172 • Specimens showed a structural infill of cytoplasm with a bright green colour, indicating they took up the
173   food over night,

174 • they developed a film or strings of pseudopodia firmly sticking to sediment particles or food,

175 • they had covered themselves or gathered a cyst of sediment or food particles.

176 Specimens were identified and sorted by species, rinsed, sediment particles were removed, and the color of the

177 cytoplasm was checked before inserting them into a petri dish withstained with calcein (106 mg l^{-1}, Bernhard et

178 al., 2004) (bis[N,N-bis(carboxymethyl)aminomethyl]-fluorescein) (Sigma-Aldrich) directly before each culturing

179 phase to ensure that freshly labelled foraminifera were inserted into the culturing system for staining (Fig. 2e).

180 Staining took place immediately before the individual culturing phase 0 to 3 and lasted for 14 days. Petri dishes

181 were stored at 8 °C in a fridge, partial water exchanges and feeding of the foraminifera was performed twice a

182 week. After the staining, the foraminifera were transferred to a petri dish with ASW and left for 1 to 2 days to

183 remove excess calcein from seawater vacuoles in their cytoplasm prior to the introduction into the culturing

184 system.

185

2.2.2 Culturing system

187 We used two closed-circulation incubation systems for foraminifera (Fig. 2a, b) provided by the Institute of

188 Microbiology, Kiel University (Dagan et al., 2016, their Fig. 5; Woehle et al., 2018, their Fig. S4). The systems

189 were ~~a~~ further develop~~ment~~ed based on earlier closed-circulation systems for culturing foraminifer~~aal culturing~~

190 (Hintz et al. 2004; Haynert et al., 2011). They were slightly modified for the requirements of this study, but the

191 basic operational principle is described by ~~Dagan~~ Woehle et al., 201~~6~~8. In detail, the systems consisted of three

192 levels with different functions. They ~~are~~were built into ~~a~~ Bauknecht WLE 885 fridges for temperature control.

193 Each ~~incubation~~ system accommodated two culturing vessels, which were arranged pairwise on a tray in a

194 polycarbonate cabinet (Fig. 2a, b). The water was pumped from the collection tank at the lowest level ~~of the fridge~~

195 to the top level into the supply tank. From the supply tank, the water was directed to the culturing vessels and the

196 flow was regulated ~~by 3-way wheels, which~~ ensur~~ed~~ing that the same amount of water was provided to every

197 culturing vessel. After passing the culturing vessels, the water was redirected to the collection tank. The systems

198 were filled with 15 L of ASW with a salinity of 30.5 PSU. The water was aerated in the supply and the collection

199 tank with filtered (0.2 µm) air from outside the system~~, which passed through a 0.2 µm filter~~. Monitoring of

200 temperature and salinity were performed with a WTW 3210 conductivity meter. Uncertainty of the conductivity

201 measurements was ± 0.5% and ± 0.1 °C for temperature according to the manufacturer's test certificate. pH was

202 monitored using a pH electrode (GHL) for aquarium purposes with uncertainties of ± 0.06. All parts that were

203 introduced into the system were sterili~~z~~sed before use either by autoclaving,~~an~~ UV~~-~~-lamp exposure, or by applying

204 DanKlorix®.

205

2.2.3 Preparation for incubation

207 For the incubation of the foraminifera, well plates with cavities made from PVC were used (Fig. 2c). ~~Each well~~

208 ~~plate measured 117 x 82 mm with 104 cavities of which each one had a diameter of 6 mm and a depth of 5 mm.~~

209 All well plates had been used in previous experiments for culturing foraminifera in seawater, which ensured that

210 potentially toxic substances or additives were already released from the ~~PVC~~ material (~~Dagan et al., 2016;~~ Woehle

211 et al., 2018)~~ and should therefore not harm the foraminifera~~. Before the foraminifera were placed in the cavities,

212 each cavity was filled with sterile quartz sand up to 1.5 mm height. The cavities were subsequently filled with

213 artificial seawater and the specimens were inserted randomly. Prepared well plates were left untouched for one

214 night, to make sure that the foraminifera ~~are~~were able to spread their pseudopodial network before incubation.

215 This ensures that they ~~are~~were stabl~~e~~y anchored in the cavities and di~~d~~o not float when the culturing vessels

216 ~~are~~were filled and mounted (Haynert et al., 2011). Four well plates ~~respectively~~ were assembled in ~~an~~each airtight

217 Emsa CLIP and CLOSE® box ~~of 226 x 167 x 59 mm~~ (Fig. 2d). ~~Of these four well plates, only three were engrafted~~

218 ~~with living foraminifera, one species per plate. The fourth plates were left barren and were used to stabilize the~~

219 ~~arrangement in the culturing vessels.~~ Each culturing vessel had a lid with an inflow and an outflow conduit, for

220 which cleaned food grade Tygon® tubing w~~ere~~as used. ~~The tubing had an outside diameter of 6.3 mm, and the~~

221 ~~PVC material was considered as nontoxic because no phthalate-based emollients were used.~~ To guarantee that the

222 foraminiferal specimens ~~are~~were not flushed away by the incoming water, the inflow conduit reached almost the

223 bottom of the culturing vessel and was placed between two well plates. ~~The outflow conduit located at the lid of~~

224 ~~the vessel was attached to a custom-made PVC fitting (Fig. 2d).~~ Once all well plates were arranged in the culturing

225 vessel, the lid was equipped with an additional, elastic sealing and closed. Before the culturing vessels were placed

226 in the ~~incubators~~culturing systems, each chamber was slowly filled with ASW. Thereafter, the culturing ~~chambers~~

227 vessels were placed on the shelve in the ~~incubator~~culturing system, and were connected to the supply hoses. ~~The~~

228 ~~water supply was turned on once the system was completely filled.~~

7

229

2.2.4 Culturing experiment

The culturing experiment had four different phases. The first, phase 0 was dedicated as control phase and. ~~n~~No heavy metals were added. This phase allowed both systems to equilibrate in terms of physicochemical and biological processes and made it possible to determine the background values in terms of seawater constituents. This phase lasted 21 days. Afterwards, one system was used as the control system, where no heavy metals were added. In the other system, three phases with elevated heavy metal concentrations were performed. The phases lasted 21 days each. Tropic Marin Pro-Reef salt was mixed with deionized water for adjusting the salinity. This artificial salt contains all elements and nutrients in sufficient amounts required by marine organisms. A stock solution containing all metals of interest was mixed and ~~before each phase,~~ this solution is called the multi metal stock solution hereafter. It was added to the supply tank of the system (see Fig. 2a) (phase 1 = 1 ml, phase 2 = 10 ml, phase 3 = 150 ml) at the beginning of each phase to reach the target concentration (Table 1). Additionally, a smaller aliquot of the same multi metal stock solution (Phase 1 = 0.1 ml, Phase 2 = 1 ml, Phase 3 = 10 ml) was introduced twice a week during the three weeks of a phase. This was to counteract the loss of metals during the culturing phase through e.g., uptake of metals by foraminifera or algae or by adsorption to surfaces of the culturing system. ~~the heavy metal concentration was elevated by 1 to 1.5 order of magnitude by adding an appropriate amount of the stock solution into the supply tank of the system (see Fig. 2a) (phase 1 = 1 ml, phase 2 = 10 ml, phase 3 = 150 ml). The elevation was performed all at once and for keeping the metal concentration at the same level over the culturing phase, a smaller amount of the stock solution was fed into the system bi-weekly. Target concentrations are given in Table 1.~~ The target concentration of the elements at each phase were chosen after earlier culturing experiments with foraminifers (Mn, Cu, Ni: Munsel et al., 2010; Pb: Frontal~~l~~ini et al., 2015 & 2018~~b~~a; Zn: Nardelli et al., 2016; Cd: Linshy et al., 2013; Cu: De Nooijer et al., 2007; Le Cadre and Debenary et al., 2006; Cr: Remmelzwaal et al., 2019, Hg: Frontalini et al., 2018a) and to resemble conditions observed in threatened environments. Examples for such environments are the San Francisco Bay, California (Thomas et al., 2002), the Black Sea, Turkey (Baltas et al., 2017) or the Gulf of Chabahar, Oman Sea (Bazzi, 2014). Furthermore, the Adriatic Sea (Ag; Barriada et al., 2007), Jakarta Bay (Williams et al., 2000; Putri et al., 2012), and polluted U.S. and European rivers (Byrd and Andreae, 1982; Kannan et al., 1998; Thomas et al., 2002) were considered. Table A4 summarizes the heavy metal concentration in seawater in different areas around the world to compare to the experimental values. Additionally, the maximum metal concentration as recommended by the EPA (Environmental Protection Agency, USA) is the lower boundary of the concentration range from this study (Prothro, 1993). This was taken into account to ensure that the foraminifera were not limited in their growth and able to maintain normal physiological functions. A lower concentration than the EPA value is also covered by our study during the control phase or in the control system. ~~in polluted areas like Jakarta Bay (e.g., Williams, 2000) or at European ports (e.g., Fatoki and Mathabatha, 2001). Furthermore, recommended threshold values provided from the EPA (Environmental Protection Agency, USA) were taken into account to ensure that the foraminifera are not limited in their growth and are able to maintain normal physiological functions. However,~~ ~~T~~the heavy metal concentrations in the culturing media obtained during each phase were monitored by frequent water sampling.

Table 1: Heavy metal concentration in the multi metal stock solution, target concentration of these metals in each phase and used salt compounds. All salts used were provided in pro ~~-~~analysi~~-~~ quality and were purchased from

268 Carl Roth (CrCl$_3$ · 6 H$_2$O; SnCl$_2$ · 2 H$_2$O and PbCl$_2$), Walter CMP (CdCl$_2$) and Sigma Aldrich (MnCl$_2$ · 4 H$_2$O,

269 NiCl$_2$ · 6 H$_2$O, CuCl$_2$ · 2 H$_2$O, ZnCl$_2$, AgNO$_3$ and HgCl$_2$).

			Target conc. in µg l^{-1}		
	Salt compound	Conc. in mg l^{-1} Multi metal sStock solution	Phase 1	Phase 2	Phase 3
Chromium (Cr)	CrCl$_3$ · 6 H$_2$O	25	0.5	5	50
Manganese (Mn)	MnCl$_2$ · 4 H$_2$O	40	40	400	4000
Nickel (Ni)	NiCl$_2$ · 6 H$_2$O	5	0.1	1	10
Copper (Cu)	CuCl$_2$ *. 2 H$_2$O	2	0.05	0.5	5
Zinc (Zn)	ZnCl$_2$	50	0.8	8	80
Cadmium (Cd)	CdCl$_2$	4	0.08	0.8	8
Silver (Ag)	AgNO$_3$	3.5	0.1	1	10
Tin (Sn)	SnCl$_2$.* 2 H$_2$O	10	0.1	1	10
Mercury (Hg)	HgCl$_2$	0.04	0.01	0.1	1
Lead (Pb)	PbCl$_2$	10	0.1	1	10

270

271 Over the entire culturing period, both systems were exposed to a natural day and night cycle and the flow rate was

272 adjusted to 1.020.017 ml mins^{-1} (one drop per second) within the culturing vessels. The foraminifera were fed

273 with *Nannochloropsis* concentrate twice a week (~ 2000 µg). After 21 days (meaning after each culturing phase)

274 one culturing vessel per system was exchanged. Vessels and specimens were left in the culturing system for the

275 complete culturing phase (21 days) and no exchange took place during a culturing phase. One culturing vessel

276 containing all three species was left in the system from the beginning until the end of the experiment (from phase

277 0 to phase 3) for 84 days. Data of these specimens are not available due to time constraints caused by the outbreak

278 of the COVID-19 pandemic.

279 Temperature and salinity were kept stable at 15.0 ±0.1 °C and 30.2 ±0.3 units PSU (trace heavy metals) and at

280 14.9 ±0.2 °C and 30.4 ±0.4 units PSU (control) over the complete culturing period. As the system was mostly

281 closed, evaporation had a minor effect. Demineralized water was added when necessary to keep the salinity stable.

282 The exchanges of culturing vessels between phases inferred a partial water exchange of approximately 10 % (=

283 1.5 l) every three weeks, which ensured a repetitive renewal of water with adequate quality.

284

2.3 Water samples

2.3.1 Collection of water samples

287 Water samples for determining the heavy metal concentrations were taken frequently from the supply tanks (see

288 Fig. 2a) of both systems using acid cleaned syringes (Norm-Ject® disposable syringe, 20 ml, sterile) and sample

289 bottles (LLG narrow neck bottles, 50 ml, LDPE = Low Density Polyethylene; Hg: GL 45 Laboratory bottle 250

290 ml with blue cap and ring, boro 3.3). From the beginning of phase 1, sampling was performed once a week. Water

291 samples to be analysed for mercury concentrations had to be treated differently due to analytical constraints as

292 detailed below. The water was filtered through a 0.2 µm PES filter (CHROMAFIL Xtra disposable filters,

293 membrane material: polyether sulfone pore) for heavy metal samples and through a 0.2 µm quartz filter for Hg

294 samples (HPLC syringe filters, 30 mm glass fibre syringe filters/ nylon). Filters were rinsed with the sample water

295 before taking the samplebefore the sample was taken. Every water sample was immediately acidified with

9

296 concentrated ultrapure HCl to a pH of approximately 2 to avoid changes in the ~~trace~~ heavy metal concentrations

297 due to adsorption to the sample bottle walls or the formation of precipitates.

298

299 **2.3.2 Preparation of water samples before analysis**

300 For Mn, Zn, Ni, Pb, Cu, and Cd concentration analyses, the water samples were pre-concentrated offline ~~by~~ using

301 a SeaFAST system (ESI, USA). Twelve m~~L~~l of each sample were used to fill a ~~10 mL~~ sample loop and

302 preconcentrated by a factor of 25 using the SeaFAST column into 1.5M HNO_3. All samples were spiked with

303 indium as an internal standard for monitoring and ~~correcting for instrumental drift~~the pre-concentration procedure.

304 Both MilliQ water and bottle blanks of acidified MilliQ water (pH ~ 2) stored in the same bottles until the samples

305 were passed through the pre-concentration system. Additionally, procedural blanks which were filtered as the

306 samples were also pre-concentrated and measured. A variety of international (Open Ocean Seawater NASS-6,

307 River Water SLRS-6, Estuarine Seawater SLEW-3, all distributed by NRC-CNRC Canada) and in-house (South

308 Atlantic surface water, South Atlantic Gyre water) reference materials were pre-concentrated like the samples. All

309 samples were subsequently analysed by ICP-MS (inductively coupled plasma mass spectrometry).

310 ~~For the metals that cannot be preconcentrated by the SeaFAST system as they are not retained on the Nobias resin~~

311 Other metals (Cr, Ag and Sn) ~~samples~~ were diluted 1/25 and directly introduced into the ICP-MS as they are not

312 retained on the Nobias resin used by the SeaFAST system. The dilution was performed with indium-spiked nitric

313 acid (2%) and to match the matrix of these samples, blanks and standards with added NaCl were prepared.

314 All ~~trace~~ heavy metals except mercury were measured using an Agilent 7500ce quadrupole ICP-MS. ~~Rand r~~aw

315 intensities were calibrated with mixed standards, which were made from single element solutions~~,~~ covering a wide

316 concentration range. Additionally, a dilution series (dilution factors: 1, 1/10, 1/100 and 1/1000) of SLRS-6 of river

317 water reference material (NRC Canada; Yeghicheyan et al., 2019) was measured for quality control. Mean values

318 and relative standard deviations (RSD) derived from the reference materials are summarised in the appendix (Table

319 A2).

320 Prior to the measurements of Hg concentrations, all samples were treated with BrCl solution at least 24 hours

321 before the analysis to guarantee the oxidation and release of mercury species that ~~are~~ were possibly present in a

322 different oxidation states or phases. The BrCl was removed again ~~from the sample~~ by adding hydroxylamine

323 hydrochloride at least one hour prior to analysis before the Hg was reduced to the volatile Hg^0 species with acidic

324 $SnCl_2$ (20 % w v^{-1}) during the measuring process. All preparations of the water samples took place in a Clean Lab

325 within a ~~trace~~ metal clean atmosphere and all vials were acid cleaned prior to use. Mercury concentrations were

326 determined using a Total Mercury Manual System (Brooks Rand Model III). The reduced volatile Hg^0 was~~is~~

327 nitrogen-purged onto a gold-coated trap and released ~~from~~ again by heating before it ~~is~~ was measured via cold

328 vapour atomic fluorescence (CVAFS) under a continuous argon carrier stream. Quality control of the Hg

329 measurements was carried out by measuring mixed standards, made from single element solutions and confirmed

330 with replicate measurements throughout each analysis. The measurement uncertainty was smaller than 4.5 % RSD

331 for all analyses.

332 The calcium concentration of culture seawater was analysed using a VARIAN 720-ES ICP-OES (inductively

333 coupled plasma optical emission spectrometer). Yttrium was added as an internal spike and samples were diluted

334 1/10. IAPSO seawater standard (ORIL) was measured after every 15 samples for further quality control which

335 revealed a measurement uncertainty < 0.35 (RSD %) for the elements analysed (mean Ca concentration IAPSO

336 this study = 419.6 ± 0.15 mg l^{-1}; reference Ca concentration IAPSO Batch 161 = 423 mg l^{-1}).

337

338 **2.4 Foraminiferal samples**

339 After every culturing phase, the culturing vessels were taken out of the culturing system and foraminiferal

340 specimens where collected from their cavities within one day. The individuals were cleaned with tap water and

341 ethanol before they were mounted in cell slides to mechanically remove salt scale and organic coatings with a

342 paintbrush. Dead specimens could be identified because they lost the colour of their cytoplasm and furthermore,

343 they did not gather food and particles anymore and thus were lacking a detritus cyst by their aperture.

344 In order to check the growth of foraminifera during the culture experiment, the total number of chambers were

345 counted before and after the experiment for every specimen, whether the foraminifera had grown during the

346 experiment, the total number of chambers was counted before and after the experiment for every specimen (Table

347 2). This was performed to double check the growth in cases where calcein staining may have failed. As the

348 foraminifera were stained with calcein before the experiment, it was possible to cross-check the growth with a

349 fluorescent microscope (Zeiss Axio Imager 2) if new chambers without color fluorescence were added, and hence

350 whether the particular specimen had grown or not (Fig. 2e). Only individuals clearly showing new chambers were

351 analysed by Laser ablation ICP--MS.

352 Prior to the laser ablation analyses, the foraminifera were transferred into individual acid-leached, 500 µl micro-

353 centrifuge tubes and thoroughly cleaned, applying a procedure adapted from Martin and Lea (2002). The

354 specimens were rinsed three times with MilliQ water and introduced into the ultrasonic bath for a few seconds at

355 the lowest power setting after each rinse. Afterwards, clay and adhering particles were removed by twice rinsing

356 the sample with eEthanol twice, which was followed by three MilliQ rinses again with minimal ultrasonic

357 treatment. Oxidative cleaning was applied using 250 µl of a 0.1M NaOH and 0.3 % H_2O_2 mixture added to each

358 sample and the vials were kept for 20 min in a 90 °C water bath. Afterwards, the samples were rinsed with MilliQ

359 three times to remove the remaining chemicals. The reductive step of the cleaning procedure by Martin and Lea

360 (2002) was not applied. This step is necessary to remove metal oxides, which of course could also influence the

361 trace heavy metal concentration within the foraminiferal shell carbonate but these are usually considered to be

362 added during early deposition (e.g., Boyle, 1983) and therefore unlikely to occur during culture experiments. For

363 Laser Ablation Inductively Coupled Plasma Mass Spectroscopy (LA-ICP-MS) measurements, all cleaned

364 specimens were fixed on a double-sided adhesive tape (PLANO).

365 Micro-analytical analyses with LA-ICP-MS were performed at the Institute of Geosciences, Kiel University, using

366 a 193nm ArF excimer GeoLasPro HD system (Coherent) with a large volume ablation cell (Zurich-type

367 LDHCLAC, Fricker et al., 2011) and helium as the carrier gas with 14 mL min^{-1} H_2 added prior to passing the

368 ablation cell. For the foraminiferal samples, the pulse rate was adjusted to 4 to 5 Hz with a fluence between 2 and

369 3.5 J cm^{-2}. The spot size was set to 44 or 60 µm depending on the size of the foraminiferal chamber. All chambers

370 of a foraminifer that were built up in the culturing medium were analysed, starting from the earliest, inner chamber

371 adjacent to the calcein-stained chamber. The laser was manually stopped once it broke through the foraminiferal

372 shell. The ablated material was analysed by an tandem ICP-MS/MS instrument (8900, Agilent Scientific

Instruments) in no gas mode. The NIST SRM 612 glass (Jochum et al., 2011) was used for calibration and monitoring of instrument drift while NIST SRM 614 was measured for quality control. The glass was chosen because all elements of interest (except Hg) were reported in the literature, which was not the case for established carbonate reference materials. Glasses were ablated with a pulse rate of 10 pulses per second, an energy density of 10 J cm^{-2} and a crater size of 60 µm. Dueñas-Bohórquez et al. (2009) demonstrated that different energy densities between the foraminiferal calcite and the glass standard does not affect the analyses. Carbonate matrix reference materials coral JCp-1, giant clam JCt-1, limestone ECRM752-1 and synthetic spiked carbonate MACS-3 (Inoue et al., 2004; Jochum et al., 2019) in the form of nano-particle pellets (Garbe-Schönberg and Müller, 2014) were analysed ~~(Garbe-Schönberg and Müller, 2014)~~ for quality control. Carbonate reference material were ablated with a pulse rate of 5 pulses per second, an energy density of 5 J cm^{-2} and a crater size of 60 µm. MACS-3 was used for calibrating the mercury content in the samples as Hg is not present in the NIST SRM glasses. All results for the reference materials are ~~displayed~~ given in the appendix (Table A3). Trace element-to-calcium ratios were quantified using the following isotopes: ^{26}Mg, ^{27}Al, ^{52}Cr, ^{55}Mn, ^{60}Ni, ^{63}Cu, ^{65}Cu, ^{68}Zn, ^{107}Ag, ^{111}Cd, ^{114}Cd, ^{118}Sn, ^{201}Hg, ^{202}Hg and ^{208}Pb normalised to ^{43}Ca. If more than one isotope was measured for an element, the average concentration of these was used after data processing. Analytical u~~U~~ncertainty (in % RSD) was better than 5 % for all TE/Ca ratios. The lowest RSD % based on the NIST SRM 612 glass was 2.1 % for Mn/Ca and the highest 5.0 % for Ag/Ca. Uncertainties of all used standards and reference materials ~~are expressed~~ are summarized in Table A3. Each acquisition interval lasted for 90 seconds, started and ended with measuring 20 s of gas blank, used as the background baseline to subtract from sample intensities during the data reduction process. Furthermore, the background monitoring ensured that the system was flushed properly after a sample. In cases when foraminiferal test walls were very fragile causing the test to break very quickly and, hence, the length of the sample data acquisition interval was less than 15 seconds, these profiles were excluded from further consideration.

Transient logs of raw intensities given in counts per seconds for all isotopes measured were processed with the software Iolite (Version 4, Paton et al., 2011) producing averages of every time-resolved laser profile. The determination of element/Ca ratios ~~were~~ was performed after the method of ~~(~~Rosenthal et al.~~,~~ (1999). High values of ^{25}Mg, ^{27}Al or ^{55}Mn at the beginning of an ablation profile were related to contamination on the surface of the foraminiferal shell or remains of organic matter (e.g., Eggins et al., 2003) and these parts of the profiles were excluded from further data processing. The detection limit was defined by 3.3*SD of the gas blank in counts per seconds for every element in the raw data. Only values above this limit were used for further analyses and no data below the LOQ (limit of quantification = 10*SD) were interpreted. After processing the data with Iolite, an outlier detection of the TE/Ca ratios of the samples was performed. If trace metal values from a spot deviated more than ±2SD from the average of the samples from the corresponding culturing phase, values were defined as outliers and discarded. The number of rejected points is indicated in the supplementary material (Table S1).

All statistical tests of the TE/Ca values in the foraminiferal shell and the water were carried out using the statistical program PAST (Hammer, 2001). As the concentration of heavy metals in seawater ~~was varying~~ varied during individual phases in the metal system (Table A1 and Fig. B1 in the appendix), the mean concentration was calculated by applying an individual curve fit for every phase. The curve was either linear, exponential or a power function depending on the ~~covariance~~ trend the particular metal showed. If the type of trend was not clear, the curve type with the highest p and R^2 values were chosen. Based on these curves, water values were calculated for every day and the weighted average from all days was used for further calculations. This ensured that high

concentrations in the beginning of each phase did not influence the mean value disproportionately. The partition coefficients of the different trace metal-to-calcium ratios were calculated using the trace element (TE) and calcium ratios in calcite and seawater. The following equation was used:

$$D_{TE} = (TE/Ca)_{calcite}/(TE/Ca)_{seawater}.$$

When the correlation between the metal concentration in seawater and the metal concentration in the foraminiferal test was positive and significant ($R^2 > 0.4$, $p < 0.05$), the D_{TE}´s are derived from the mean values of all phases and represent the slope of the calculated regression line. In cases where a significant positive correlation between phases could not be identified, the D_{TE} values were calculated from the means of each phase separately and the ranges given. The regression line was forced through the origin, which is a common practice and is applied in many other studies (e.g., Lea and Spero, 1994; Munsel et al., 2010; Remmelzwaal et al., 2019; Sagar et al., 2021a). The reason for this approach is that foraminifers are expected not to incorporate any metals into their shell if the metals concentration is zero in the seawater. In cases where there was clearly a non-zero intercept (Mn of *A. batava* with phase 3 and Hg of *E. excavatum* without phase 3), obvious if the course of the regression line changed significantly or the R^2 value decreased, then the trend line was not forced through the origin.

3 Results

3.1 Survival Rates/ Growth rates / Reproductions

Table 2: Number of inserted and recovered foraminifera from the different systems (C = control system, M = metal system) and phases (0–3). Numbers of living individuals after the experiment and individuals that formed chambers during their individual culturing phase are given in %. Note that the percentage of living foraminifera is based on the number of foraminifera that could be recovered alive and not on the number of inserted individuals. The number of laser spots is indicated as well.

	C0	C1	C2	C3	M0	M1	M2	M3	Total
No. of inserted individuals									
Ammonia aomoriensis	50	24	20	20	19	70	70	72	345
Ammonia batava	22	20	20	20	16	43	72	72	285
Elphidium excavatum	45	24	20	20	19	70	69	70	337
Total	117	68	60	60	54	183	211	214	967
No. of recovered individuals									
Ammonia aomoriensis	43	20	10	19	11	57	58	56	274
Ammonia batava	11	15	16	14	7	29	65	56	213
Elphidium excavatum	36	20	20	14	7	62	58	53	270
Total	90	55	46	47	25	148	181	165	757
Living individuals (end of experiment) in %									
Ammonia aomoriensis	86	100	80	100	90.9	100	81	98.2	92.0
Ammonia batava	81.8	100	100	92.9	100	100	100	100	96.8
Elphidium excavatum	91.7	100	95	92.9	100	88.7	91.4	94.3	94.3

Total	86.5	100	91.7	95.3	97.0	96.2	90.8	97.5	94.4
Ind. that formed chambers (end of the experiment) in %									
Ammonia aomoriensis	62.8	84.2	100	93.8	81.8	100	92.3	90	88.1
Ammonia batava	45.5	85.7	100	100	71.4	100	100	100	87.8
Elphidium excavatum	69.4	65	56.3	38.5	57.1	67.7	75	62.3	61.4
Total	59.2	78.3	85.4	77.4	70.1	89.2	89.1	84.1	79.1
No. of laser spots									
Ammonia aomoriensis	22	18	17	20	9	39	40	36	201
Ammonia batava	14	20	19	19	6	17	52	57	204
Elphidium excavatum	14	13	13	12	1	36	24	31	144
Total	50	51	49	51	16	92	116	124	549

On average 74.5 % of the specimens inserted into the experiment could be recovered after their individual culturing phase of 21 days and 94.4 % of these recovered specimens survived. Approximately 79.1 % of the surviving specimens also formed at least one new chamber. Fewer specimens of *E. excavatum* formed new chambers (61.4 %) than *A. batava* (87.8%) or *A. aomoriensis* (88.1 %) (Table 2). On average, *E. excavatum* formed only one or rarely two new chambers, whereas both *Ammonia* species formed usually more than four new chambers. Reproduction happened very sporadically ~~concerning~~ occurring in between 2 and 6 specimens per phase, on average 5 %, for the two *Ammonia* species but not for *E. excavatum*. No malformed chambers were observed in specimens that were recovered from the ~~trace~~ heavy-metal contaminated system.

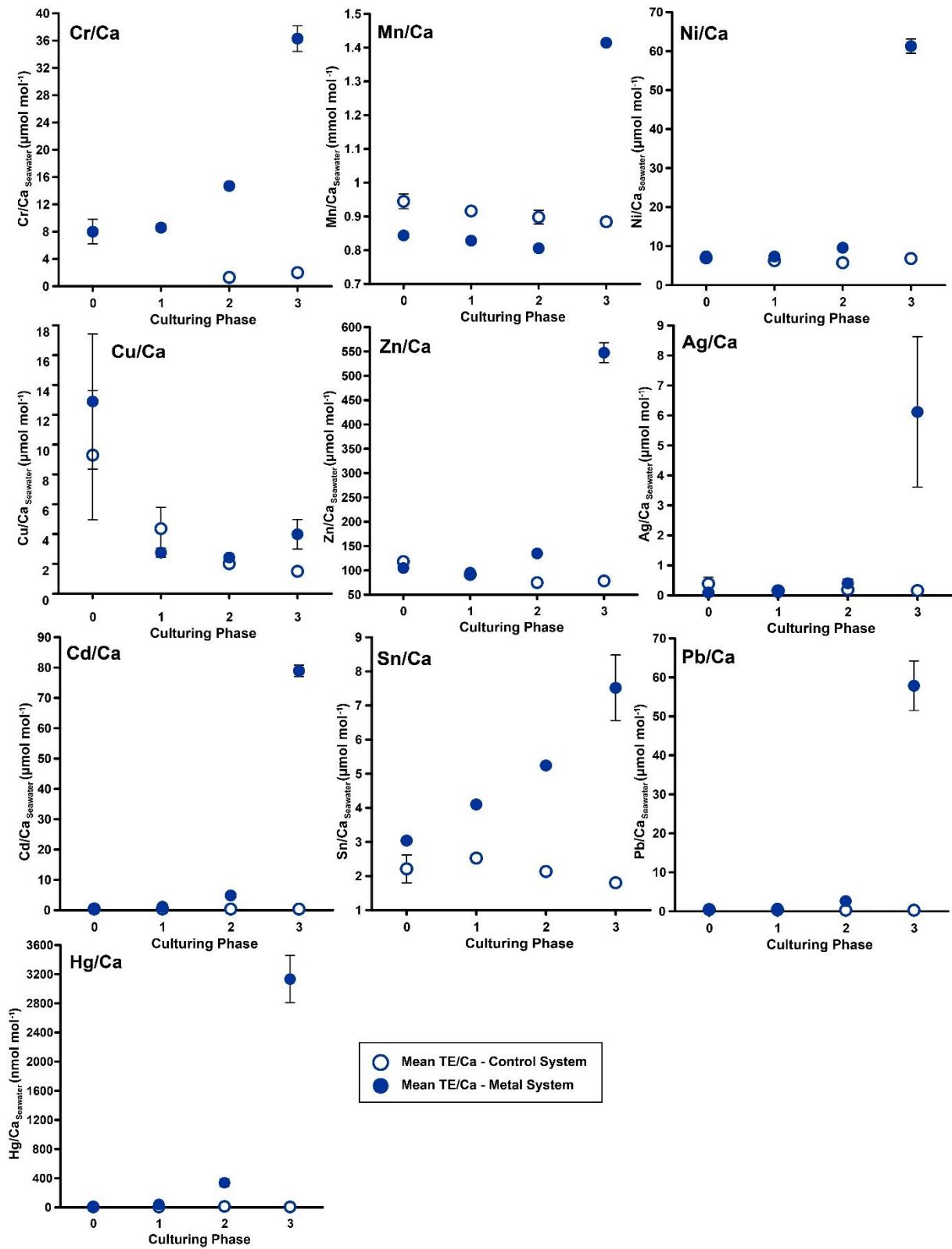**3.2 Culturing media**
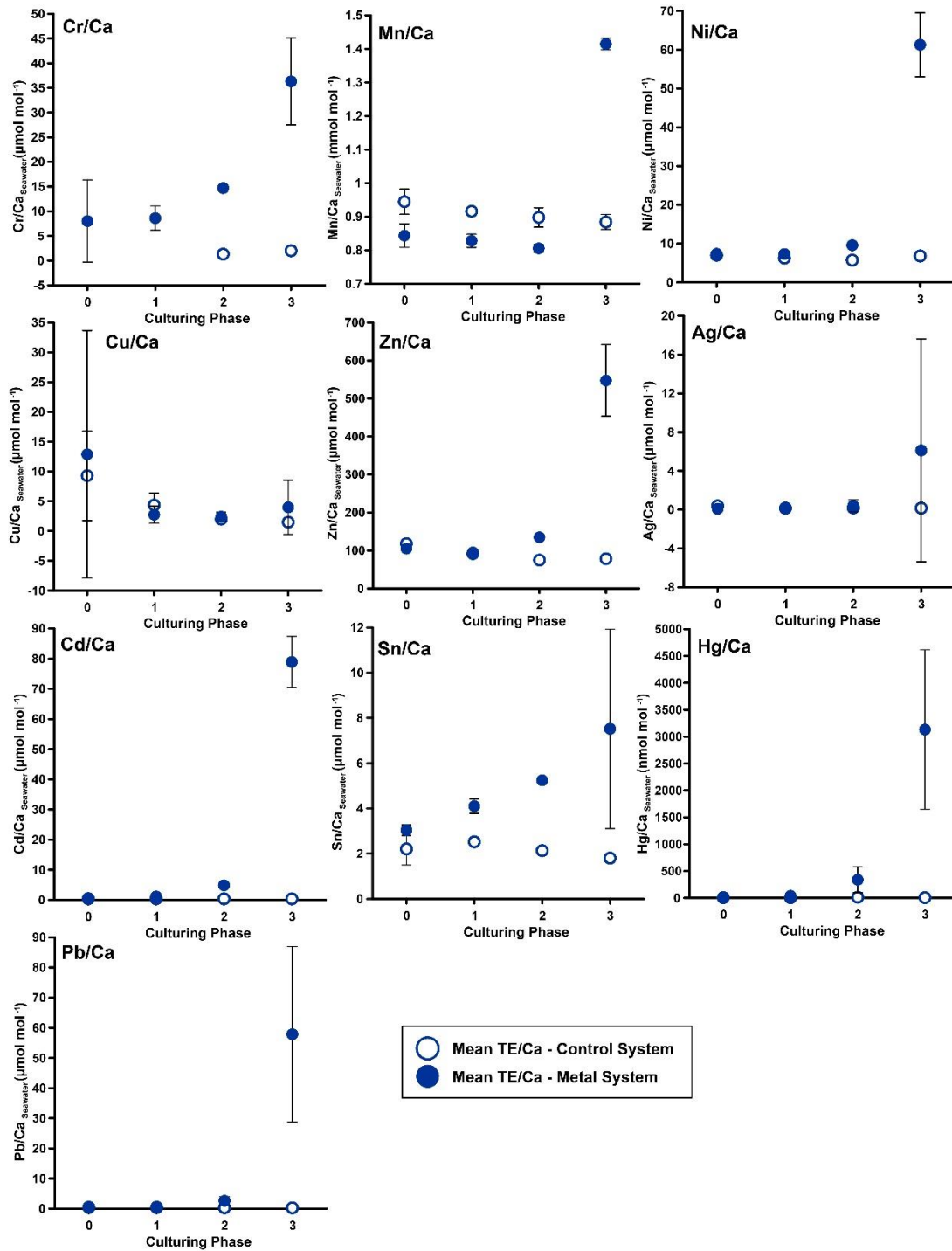
Table 3: Weighted mean TE/Ca values in the culturing medium of the control and the metal system ± the standard error of the mean (standard deviation $\sigma/\sqrt{n}$). Furthermore, the factors between the target concentrations (Table 1) and the measured concentrations as well as the factors between individual phases are given. Values given without a standard error originate from only one measurement. Averaged TE/Ca values of a phase were calculated based on single values measured on samples from different days during the culturing phase. These single values can be found in the Appendix ~~All values the calculations are based on can be found in the appendix~~ (Table A1). BDL = below detection limit.

	Cr/Ca	Mn/Ca	Ni/Ca	Cu/Ca	Zn/Ca	Ag/Ca	Cd/Ca	Sn/Ca	Hg/Ca	Pb/Ca
Control System	µmol mol^{-1}	mmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	nmol mol^{-1}	µmol mol^{-1}
Phase 0	BDL	0.94 ± 0.02	7.0 ± 0.1	9.3 ± 4.3	118.3 ± 4.5	0.43 ± 0.214	0.41 ± 0.001	2.2 ± 0.4	5.8 ± 0.6	0.44 ± 0.06
Phase 1	BDL	0.92 ± 0.00	6.3 ± 0.1	4.4 ± 1.4	91.6 ± 1.1	0.19 ± 0.013	0.41 ± 0.002	2.5 ± 0.1	4.5 ± 1.0	0.39 ± 0.02
Phase 2	1.3 ± 0.3	0.90 ± 0.02	5.7 ± 0.1	2.1 ± 0.2	74.8 ± 2.0	0.19 ± 0.003	0.38 ± 0.006	2.1 ± 0.1	13.2 ± 5.8	0.31 ± 0.02
Phase 3	2.0 ± 0.4	0.89 ± 0.01	6.8 ± 0.3	1.5 ± 0.1	78.3 ± 0.8	0.16 ± 0.009	0.37 ± 0.006	1.8 ± 0.1	5.8 ± 1.8	0.28 ± 0.01
Metal System	µmol mol^{-1}	mmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	nmol mol^{-1}	µmol mol^{-1}
Phase 0	8.0 ± 1.8	0.84 ± 0.01	7.4 ± 0.1	12.9 ± 4.5	104.8 ± 1.4	0.09 ± 0.02	0.43 ± 0.002	3.0 ± 0.1	5.28	0.50 ± 0.04

Phase 1	8.6 ± 0.5	0.83 ± 0.004	7.3 ± 0.1	2.8 ± 0.3	95.2 ± 0.3	0.10 ± 0.02	1.12 ± 0.01	4.1 ± 0.1	39.7 ± 2.7	0.69 ± 0.03
Phase 2	14.7 ± 0.1	0.81 ± 0.003	9.6 ± 0.1	2.4 ± 0.2	134.8 ± 0.5	0.40 ± 0.14	4.86 ± 0.03	5.2 ± 0.03	337.6 ± 52.1	2.63 ± 0.3
Phase 3	36.3 ± 1.9	1.41 ± 0.004	61.3 ± 1.8	4.0 ± 1.0	547.5 ± 20.5	6.1 ± 2.5	78.92 ± 1.9	7.5 ± 1.0	3132.4 ± 323.7	57.84 ± 6.4
Factor between target conc. and measured conc.										
Phase 1	17.2	20.8	73.0	56.0	119.0	1.0	14.0	41.0	4.0	6.9
Phase 2	2.9	2.0	9.6	4.8	16.9	0.4	6.1	5.2	3.4	2.6
Phase 3	0.7	0.4	6.1	0.8	6.8	0.6	9.9	0.8	3.1	5.8
Factor between Phases										
Phase 0-1	1.1	1.0	1.0	0.2	0.9	1.1	2.6	1.4	7.5	1.4
Phase 1-2	1.7	1.0	1.3	0.9	1.4	4.0	4.3	1.3	8.5	3.8
Phase 2-3	2.5	1.7	6.4	1.7	4.1	15.3	16.2	1.4	9.3	22.0

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Figure 3: Weighted mean TE/Ca values in the culturing medium in μmol mol^{-1}. Error bars display the standard error of the mean (standard deviation $\sigma/\sqrt{n}$). Open symbols represent the control system, where no extra metals were added during the complete culturing period (phase 0 to 3) and closed symbols represent the metal system. In this system, phase 0 is the control phase without any extra added metals and for phase 1 to 3, the heavy metal concentration in the culturing medium was elevated. Note that the standard error is comparably high in phase 3 because the heavy metal concentration in this phase varied more strongly, which is shown in the appendix (Table A1, Fig. B1). Therefore, this error is derived from the real values in the seawater and not from analytical uncertainties. Note that the Cr/Ca values from the control system in phase 0 and 1 are not given as these values were below the detection limit.

17

465

In phases 1 and 0 the concentration in both systems were nearly equal for most elements. Only Cr and Sn had slightly elevated concentrations in the metal system~~.~~ Furthermore,~~,~~ ~~whereas~~ Cu ~~and Mn~~ concentration ~~were~~ was higher in the ~~control~~ metal system in phase 0 and phase 3 (Fig. 3). ~~This also holds true for Mn in~~ In phase 2, ~~when~~ all ~~other~~ metals but Mn and Cu showed higher concentrations in the metal system than in the control system. Mn concentrations were higher in the control system during phase 0 to phase 2. In phase 3, the concentration of all heavy metals were elevated in the metal system ~~as~~ compared to the control system. The variation of the metal concentration was ~~in both systems~~ highest in phase 3, in both systems, for all elements but Cu, which showed ~~the~~ highest variation in phase 0 (Fig. 3). The control system generally displayed a smaller degree of variation than the metal system.

Even though, the aim was to maintain the target concentrations shown in Table 1 during the 21 days of each culturing period by the bi-weekly addition of an aliquot of the multi metal stock solution, t~~T~~he target concentration of the metals was not ~~accomplished~~ obtained for most metals in phase 1 and 2, the only exception wa~~s~~is Ag in phase 1 (Table 3). The difference factors between the target and measured concentration was highest (> 50) for Ni, Cu and Zn in phase 1 and ~~gets~~decreased ~~smaller~~ in phase 2 and 3. ~~Generally, all elements but Mn were concentrated higher in phases 1 and 2 than expected.~~ In phase 3, metals Cr, Mn, Cu, Ag and Sn reached concentrations closer (factor 0.4~~-~~—0.8) to the target concentration and Ni, Zn, Cd, Hg and Pb concentrations were ~~concentrated~~ higher (factor 3.1~~-~~—9.9) than expected. Furthermore, the ~~factor between individual phases (Table 3)~~change in metal concentration was small for the transition from phase 0 to 1 (factor <-1.4) for all elements but Cd (factor 2.6) and Hg (factor 7.5). ~~Same patterns can be seen between phase 1 and 2, while the difference between phase 2 and 3 was more distinct (factor > 4) for Ni, Zn, Ag, Cd, Pb and Hg. Mn, Cu and Sn showed little variation between phase 2 and 3 (factor < 1.7). Generally, the factor between each phase should have been approximately 10, which was not achieved in most cases. Exceptions were Ag, Cd and Pb, which had factors >15 between phase 2 and 3. Furthermore, Hg showed concentrations that were higher by a factor around 10 between all phases (phase 0-1 = 7.5, phase 1-2 = 8.5, phase 2-3 = 9.3).~~

490

3.3.1 Incorporation of ~~trace~~ heavy metals into the foraminiferal shell

Table 4: Mean ~~trace element~~heavy metal–to–calcium values of *A. aomoriensis*, *A. batava* and *E. excavatum* in the control and the metal system. Errors are standard errors of the mean (standard deviation $\sigma/\sqrt{n}$). Values marked with an asterisk were derived from only one laser spot and thus are not considered for further discussion. Furthermore, the calculated D_{TE} values, the slope of the linear regression line (OLS—–Ordinary Least Squares) of all means, Pearson's correlation coefficient (R^2) and its significance (p) are given for the calculation with all phases and when removing phase 3 from the calculations. Cases where the regression lines were forced through the origin are indicated. ~~It´s also indicated whether the regression line is forced through the origin or not~~. In cases when a regression did not show significant correlation, the D_{TE} range ~~separately~~calculated separately from the individual phases is given. In cases when the regression was significant, the D_{TE} values represent the slope of the regression line. Ph = Phase, SD = Standard deviation. Values in Table S1 are the basis of all calculations.

	Phase	Cr/Ca	Mn/Ca	Ni/Ca	Cu/Ca	Zn/Ca
Control System		μmol mol^{-1}	mmol mol^{-1}	μmol mol^{-1}	μmol mol^{-1}	μmol mol^{-1}

18

A. aomoriensis	0	18.6 ± 2.5	0.11 ± 0.02	1.3 ± 0.2	5.6 ± 0.9	53.2 ± 8.8
	1	12.6 ± 0.6	0.53 ± 0.12	5.9 ± 0.8	8.6 ± 1.0	34.2 ± 4.7
	2	13.6 ± 0.5	0.27 ± 0.07	2.1 ± 0.2	3.6 ± 0.2	18.6 ± 1.9
	3	10.2 ± 0.6	0.43 ± 0.08	4.3 ± 0.7	8.1 ± 2.0	29.5 ± 6.1
A. batava	0	11.6 ± 0.7	0.04 ± 0.01	1.4 ± 0.2	7.2 ± 1.1	23.9 ± 4.5
	1	10.9 ± 0.5	0.03 ± 0.00	2.6 ± 0.3	5.9 ± 0.6	17.8 ± 1.3
	2	9.0 ± 0.3	0.03 ± 0.00	0.9 ± 0.1	5.0 ± 1.0	12.9 ± 1.4
	3	9.1 ± 0.4	0.03 ± 0.01	1.9 ± 0.2	6.5 ± 1.3	14.9 ± 2.2
E. excavatum	0	22.9 ± 2.9	0.43 ± 0.13	9.4 ± 2.5	22.3 ± 7.9	28.1 ± 4.5
	1	88.9 ± 34.1	2.29 ± 0.56	7.8 ± 1.9	20.3 ± 8.0	48.9 ± 12.1
	2	16.2 ± 1.7	1.55 ± 0.26	5.9 ± 1.0	6.7 ± 1.4	21.9 ± 2.9
	3	26.7 ± 3.3	1.88 ± 0.55	4.4 ± 0.6	4.7 ± 0.7	16.8 ± 2.0

Metal System

A. aomoriensis	0	16.0 ± 0.5	0.08 ± 0.02	5.5 ± 0.9	15.2 ± 2.6	29.8 ± 5.1
	1	14.0 ± 0.7	0.39 ± 0.08	3.1 ± 0.3	6.7 ± 0.7	30.0 ± 4.0
	2	11.1 ± 0.3	0.20 ± 0.05	5.3 ± 0.5	5.8 ± 0.5	28.3 ± 2.3
	3	14.1 ± 1.0	0.71 ± 0.12	3.8 ± 0.3	6.3 ± 1.5	42.2 ± 6.1
A. batava	0	16.5 ± 0.7	0.07 ± 0.01	1.1 ± 0.1	7.7 ± 1.6	68.0 ± 9.6
	1	15.2 ± 1.2	0.04 ± 0.01	1.8 ± 0.3	2.5 ± 0.6	20.7 ± 2.7
	2	9.7 ± 0.2	0.02 ± 0.00	1.8 ± 0.1	8.3 ± 1.8	12.9 ± 1.2
	3	12.2 ± 0.3	0.17 ± 0.04	2.9 ± 0.2	8.3 ± 1.2	49.8 ± 3.5
E. excavatum	0	17.30*	0.29*	4.30*	12.20*	26.70*
	1	32.9 ± 3.4	0.70 ± 0.12	8.2 ± 1.1	12.8 ± 1.8	18.5 ± 0.9
	2	41.8 ± 5.2	0.77 ± 0.15	8.6 ± 1.1	11.5 ± 1.5	29.8 ± 3.6
	3	54.1 ± 8.2	0.88 ± 0.15	17.0 ± 2.2	22.6 ± 3.6	43.1 ± 3.3

Calculations with Phase 3

A. aomoriensis

Slope of regression line ±SD		0.38 ±0.30		1.18 ±0.25	
Correlation coefficient (R^2)		0.83		0.80	
Significance (p)		0.05		0.05	
D_{TE} ±SD	0.4-10.3	0.38 ±0.30	0.06-0.94	1.18 ±0.25	0.08-0.45
Forced through origin	Single points	Yes	Single points	Yes	Single points

A. batava

Slope of regression line ±SD		0.23 ±0.04			
Correlation coefficient (R^2)		0.84			
Significance (p)		0.001			
D_{TE} ±SD	0.4-6.8	0.23 ±0.04	0.05-0.41	0.60-4.35	0.09-0.65
Forced through origin	Single points	No	Single points	Single points	Single points

E. excavatum

Slope of regression line ±SD	2.1 ±0.28		0.19 ±0.04		
Correlation coefficient (R^2)	0.82		0.79		
Significance (p)	0.01		0.003		
D_{TE} ±SD	2.1 ±0.28	0.34-2.50	0.19 ±0.04	0.95-5.67	0.08-0.53
Forced through origin	Yes	Single points	~~Yes~~No	Single points	Single points

Calculations without Phase 3

A. aomoriensis

Slope of regression line ±SD					
Correlation coefficient (R^2)					
Significance (p)					
D_{TE} ±SD	0.74-10.3	0.09-0.53	0.19-0.94	0.61-5.42	0.21-0.45
Forced through origin	Single points	Single points	Single points	Single points	Single points

A. batava

Slope of regression line ±SD					
Correlation coefficient (R^2)					
Significance (p)					
D_{TE} ±SD	0.65-6.8	0.02-0.08	0.15-0.41	0.60-4.35	0.10-0.65
Forced through origin	Single points	Single points	Single points	Single points	Single points

E. excavatum

Slope of regression line ±SD					
Correlation coefficient (R^2)					
Significance (p)					
D_{TE} ±SD	2.5-13.4	0.34-2.50	0.64-1.35	0.95-4.73	0.22-0.53

Forced through origin		Single points	Single points	Single points	Single points	Single points

503   **Table 4** continued.

	Phase	Ag/Ca	Cd/Ca	Sn/Ca	Hg/Ca	Pb/Ca
Control System		μmol mol^{-1}	μmol mol^{-1}	μmol mol^{-1}	nmol mol^{-1}	μmol mol^{-1}
A. aomoriensis	0	0.27 ± 0.08	7.6 ± 1.0	0.33 ± 0.07	1.54 ± 0.46	1.23 ± 0.22
	1	0.28 ± 0.05	3.8 ± 0.3	1.60 ± 0.30	3.11 ± 0.68	1.14 ± 0.16
	2	0.16 ± 0.04	3.6 ± 0.2	0.21 ± 0.03	1.13 ± 0.31	0.81 ± 0.10
	3	0.31 ± 0.11	2.9 ± 0.2	0.19 ± 0.03	8.02 ± 1.72	1.45 ± 0.42
A. batava	0	0.09 ± 0.03	4.7 ± 0.5	0.27 ± 0.05	1.3 ± 0.4	0.67 ± 0.10
	1	0.07 ± 0.01	2.5 ± 0.2	0.65 ± 0.09	1.2 ± 0.3	0.29 ± 0.03
	2	0.05 ± 0.00	2.7 ± 0.1	0.08 ± 0.02	1.5 ± 0.4	0.39 ± 0.03
	3	0.06 ± 0.01	1.9 ± 0.1	0.10 ± 0.02	4.4 ± 0.6	0.36 ± 0.05
E. excavatum	0	0.22 ± 0.09	3.6 ± 1.1	0.99 ± 0.40	15.0 ± 4.4	1.83 ± 0.59
	1	0.07 ± 0.01	20.1 ± 9.2	8.21 ± 2.63	83.0 ± 33.4	2.22 ± 0.54
	2	0.10 ± 0.03	1.2 ± 0.2	0.45 ± 0.08	16.9 ± 3.8	0.94 ± 0.10
	3	0.04 ± 0.01	2.3 ± 0.4	0.27 ± 0.03	35.8 ± 6.3	0.55 ± 0.11
Metal System						
A. aomoriensis	0	0.08 ± 0.03	4.9 ± 0.3	0.62 ± 0.09	2.6 ± 0.6	1.17 ± 0.24
	1	0.25 ± 0.04	4.0 ± 0.4	0.84 ± 0.10	1.8 ± 0.2	0.90 ± 0.13
	2	0.52 ± 0.08	5.5 ± 0.4	1.70 ± 0.17	9.1 ± 1.7	3.85 ± 0.45
	3	3.03 ± 0.39	5.4 ± 0.4	0.55 ± 0.10	10.3 ± 1.3	22.14 ± 2.37
A. batava	0	0.06 ± 0.03	6.2 ± 0.2	0.19 ± 0.04	1.0 ± 0.2	1.27 ± 0.08
	1	0.04 ± 0.01	3.1 ± 0.3	0.59 ± 0.12	0.2 ± 0.0	0.42 ± 0.07
	2	0.18 ± 0.04	3.1 ± 0.2	0.46 ± 0.06	4.5 ± 1.1	0.52 ± 0.05
	3	1.05 ± 0.17	6.5 ± 0.3	0.21 ± 0.02	7.7 ± 1.0	29.82 ± 3.70
E. excavatum	0	0.40^{*}	5.60^{*}	0.18^{*}	6.80^{*}	1.59^{*}
	1	0.03 ± 0.01	3.0 ± 0.3	2.63 ± 0.32	85.7 ± 19.7	1.36 ± 0.15
	2	0.69 ± 0.18	3.9 ± 0.5	2.89 ± 0.47	120.4 ± 44.7	4.61 ± 0.86
	3	2.84 ± 0.64	4.7 ± 0.5	2.74 ± 0.42	94.9 ± 16.2	52.51 ± 6.17
Calculations with Phase 3						
A. aomoriensis						
Slope of regression line $\pm$SD		0.50 ± 0.02				0.39 ± 0.01
Correlation coefficient (R^2)		0.97				0.97
Significance (p)		< 0.0001				< 0.0001
$D_{TE} \pm$SD		0.50 ± 0.02	0.07-18.49	0.07-0.63	0.003-1.39	0.39 ± 0.01
Forced through origin		Yes	Single points	Single points	Single points	Yes
A. batava						
Slope of regression line $\pm$SD		0.17 ± 0.01			0.003 ± 0.001	0.52 ± 0.01
Correlation coefficient (R^2)		0.98			0.63	1
Significance (p)		< 0.0001			0.01	< 0.0001
$D_{TE} \pm$SD		0.17 ± 0.01	0.08-14.42	0.03-0.26	0.003 ± 0.001	0.52 ± 0.01
Forced through origin		Yes	Single points	Single points	Yes	Yes
E. excavatum						
Slope of regression line $\pm$SD		0.47 ± 0.04				0.91 ± 0.01
Correlation coefficient (R^2)		0.96				1
Significance (p)		< 0.0001				< 0.0001
$D_{TE} \pm$SD		0.47 ± 0.04	0.06-49.45	0.06-3.25	0.03-18.51	0.91 ± 0.01
Forced through origin		Yes	Single points	Single points	Single points	Yes
Calculations without Phase 3						
A. aomoriensis						
Slope of regression line $\pm$SD						1.6 ± 0.17
Correlation coefficient (R^2)						0.91
Significance (p)						< 0.001
$D_{TE} \pm$SD		0.70-2.57	1.14-18.49	0.10-0.63	0.003-1.39	1.60 ± 0.17
Forced through origin		Single points	Single points	Single points	Single points	Yes
A. batava						
Slope of regression line $\pm$SD		0.35 ± 0.09				
Correlation coefficient (R^2)		0.91				

Significance (p)	0.03				
D_{TE} ±SD	0.35 ±0.09	0.63-14.42	0.04-0.26	0.005-0.76	0.20-5.52
Forced through origin	Yes	Single points	Single points	Single points	Single points
E. excavatum					
Slope of regression line ±SD				0.26 ±0.11	2 ±0.28
Correlation coefficient (R^2)				0.53	0.90
Significance (p)				0.05	0.003
D_{TE} ±SD	0.23-4.25	0.80-49.45	0.06-3.25	0.26 ±0.11	2.0 ±0.28
Forced through origin	Single points	Single points	Single points	No	Yes

504

505 Measurable incorporation into the foraminiferal calcite was found for all the ~~trace~~ heavy metals analysed but the
506 degree of incorporation varied profoundly within and between species (Fig. 4 and Table 4). In both systems, the
507 ~~trace~~ heavy metal concentration in _E. excavatum_ was higher than in the other species (_A. aomoriensis_ and _A._
508 _batava_) for Cr, Mn, Ni, ~~Cu,~~ Hg and Sn. This trend is also visible but less pronounced in the Cu values of the
509 control system.

510 Cr, Ni, Cu, Zn, Cd, Pb and Ag values of _A. aomoriensis_ displayed the highest standard error of the mean paired
511 with highest concentrations in the water in the metal system. Sn, Mn and Hg did not show any clear pattern. In the
512 control system, all ~~trace~~ heavy metal concentrations had higher standard errors of the mean when the concentration
513 of these metals in the culturing medium was higher. The trend was also shown in _A. batava_ and _E. excavatum_ for
514 all heavy metals of the control and the metal system. Note that even though no extra metals were added to the
515 culturing medium of the control system, differences in the heavy metal concentration occurred (Fig. 3 and Table
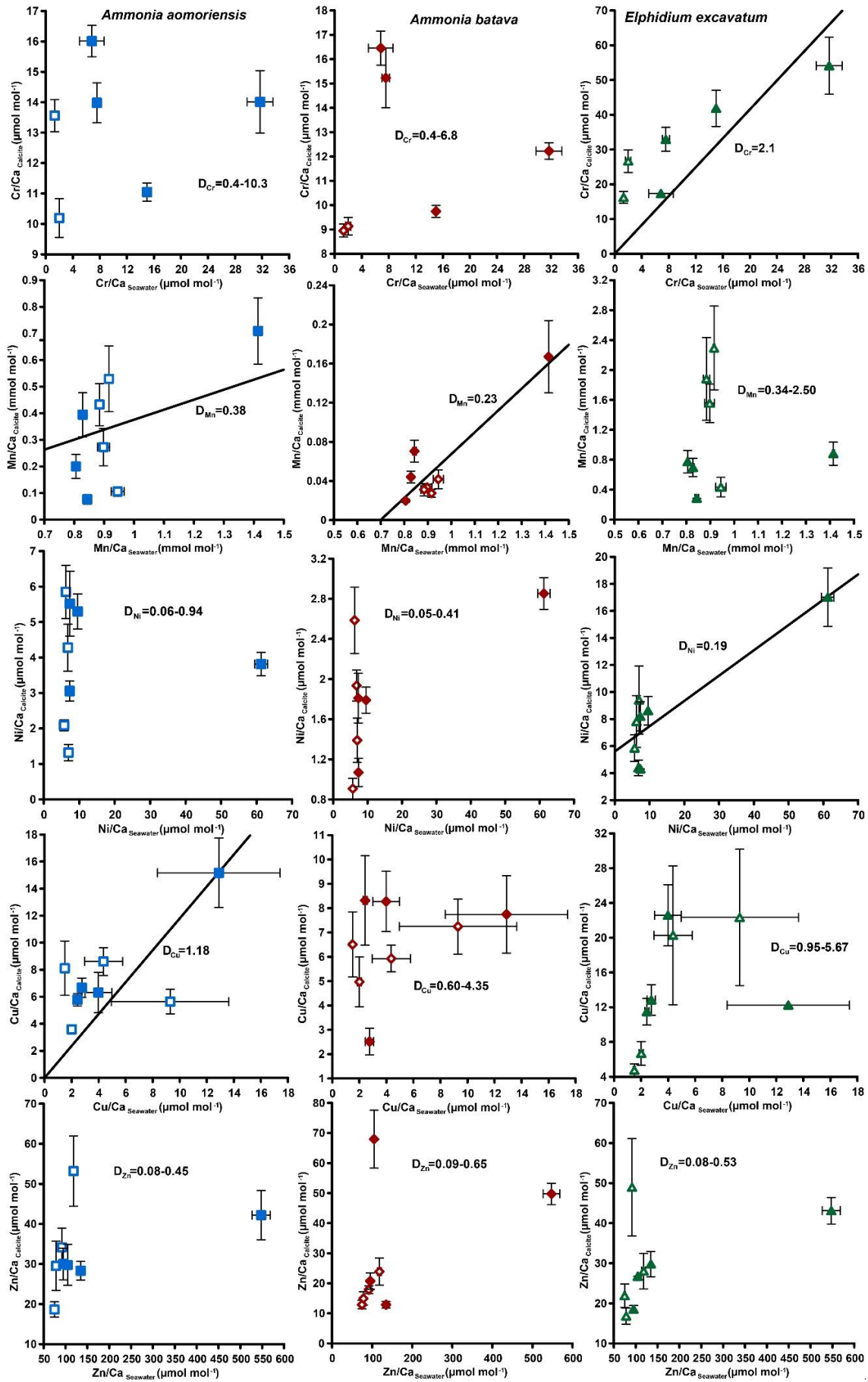516 3).

517 Calculations were performed with and without phase 3 of the metal system (Fig. 4, Fig. B2 and Table 4) to address
518 a possible overload effect when it comes to higher metal concentrations in the seawater.

519 When phase 3 was included, a strong positive correlation ($R^2 > 0.9$, $p \leq 0.05$) ~~of~~ between Ag and Pb concentrations
520 in the foraminiferal shell and ~~in~~ the culturing medium was ~~recognised~~ found for~~in~~ all three species. Furthermore,
521 _A. batava_ also displayed~~revealed~~ a positive correlation for Hg ($R^2 = 0.63$, $p < 0.01$), _A. aomoriensis_ for Cu ($R^2 =$
522 0.80, $p < 0.05$) and _E. excavatum_ for Cr ($R^2 = 0.82$, $p < 0.01$) and Ni ($R^2 = 0.79$, $p < 0.003$). Weaker but still
523 significant positive correlations were recorded for Mn ($R^2 > 0.84$, $p \leq 0.05$) for both _Ammonia_ species. An
524 indistinct correlation of the concentration in the seawater and in the foraminiferal test was recognised for Zn in all
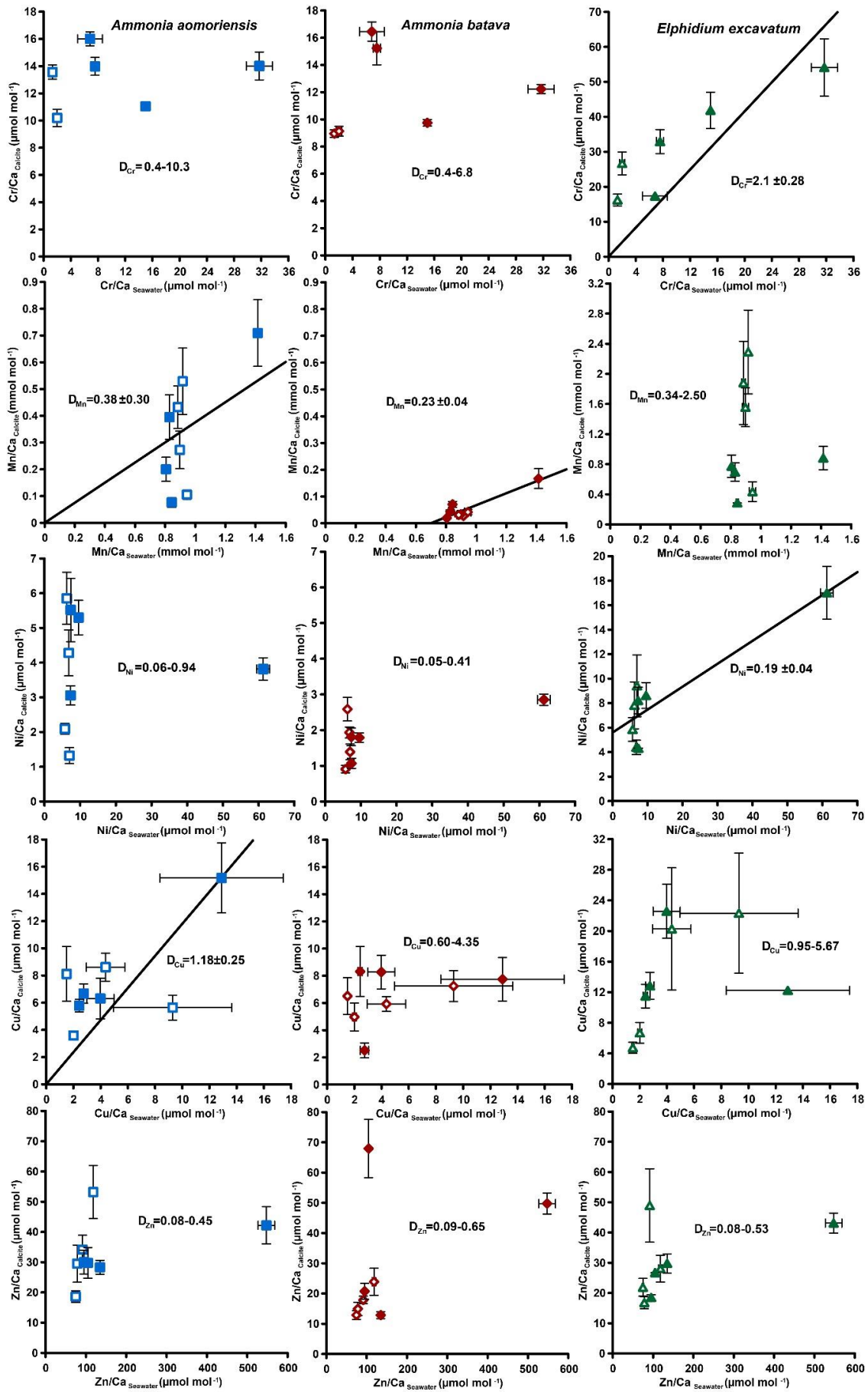525 three species, whereas Cd and Sn showed no covariance (Fig. 4 and Table 4).

526 When phase 3 was excluded from the calculations, _A. aomoriensis_ and _E. excavatum_ showed a positive correlation
527 for Pb ($R^2 > 0.9$, $p \leq 0.003$), _A. batava_ for Ag ($R^2 = 0.91$, $p = 0.03$) and in _E. excavatum_ Hg correlated weaker
528 positively ($R^2 > 0.53$, $p \leq 0.05$). All other elements show no significant correlation (Fig. 4 and Table 4).
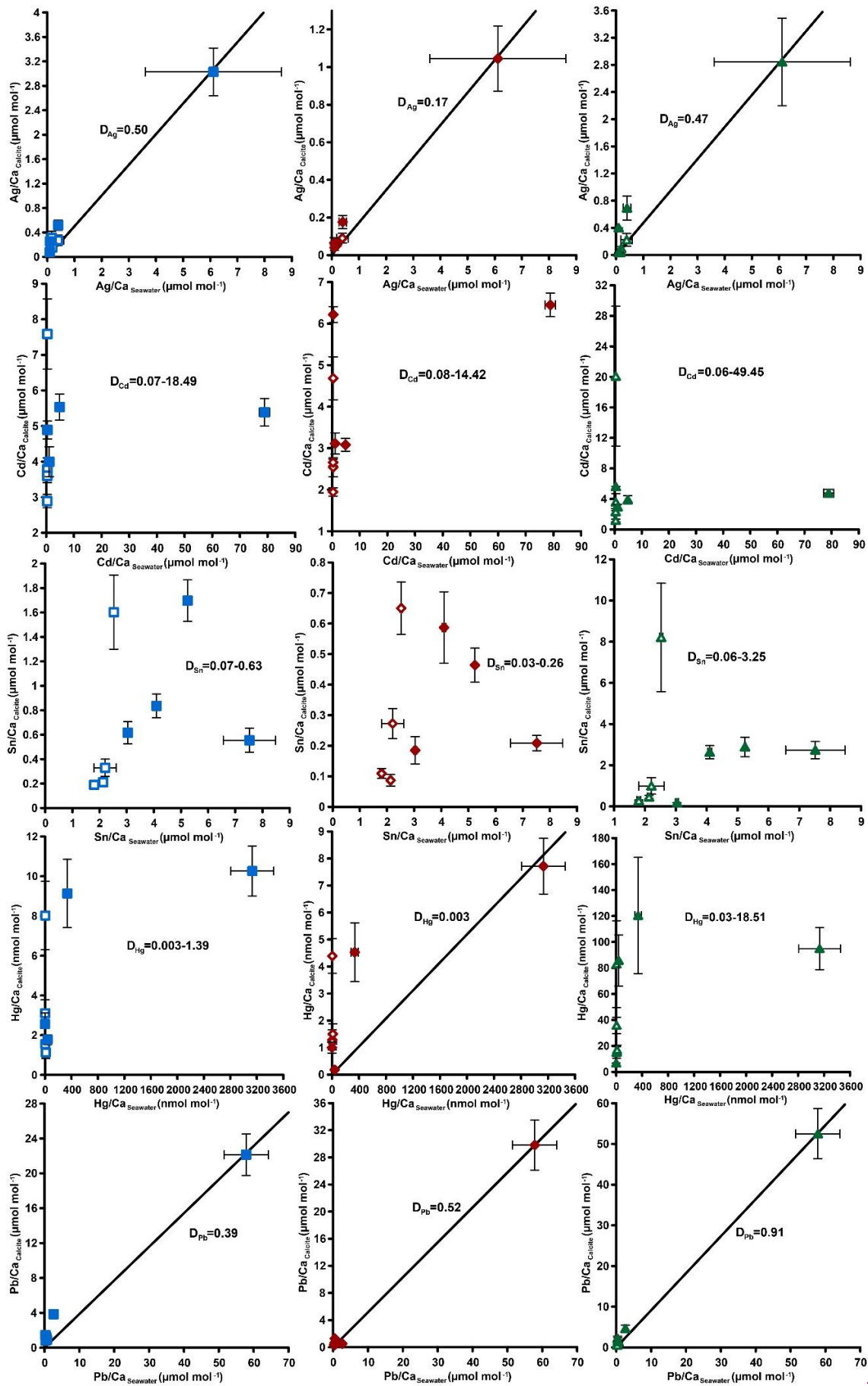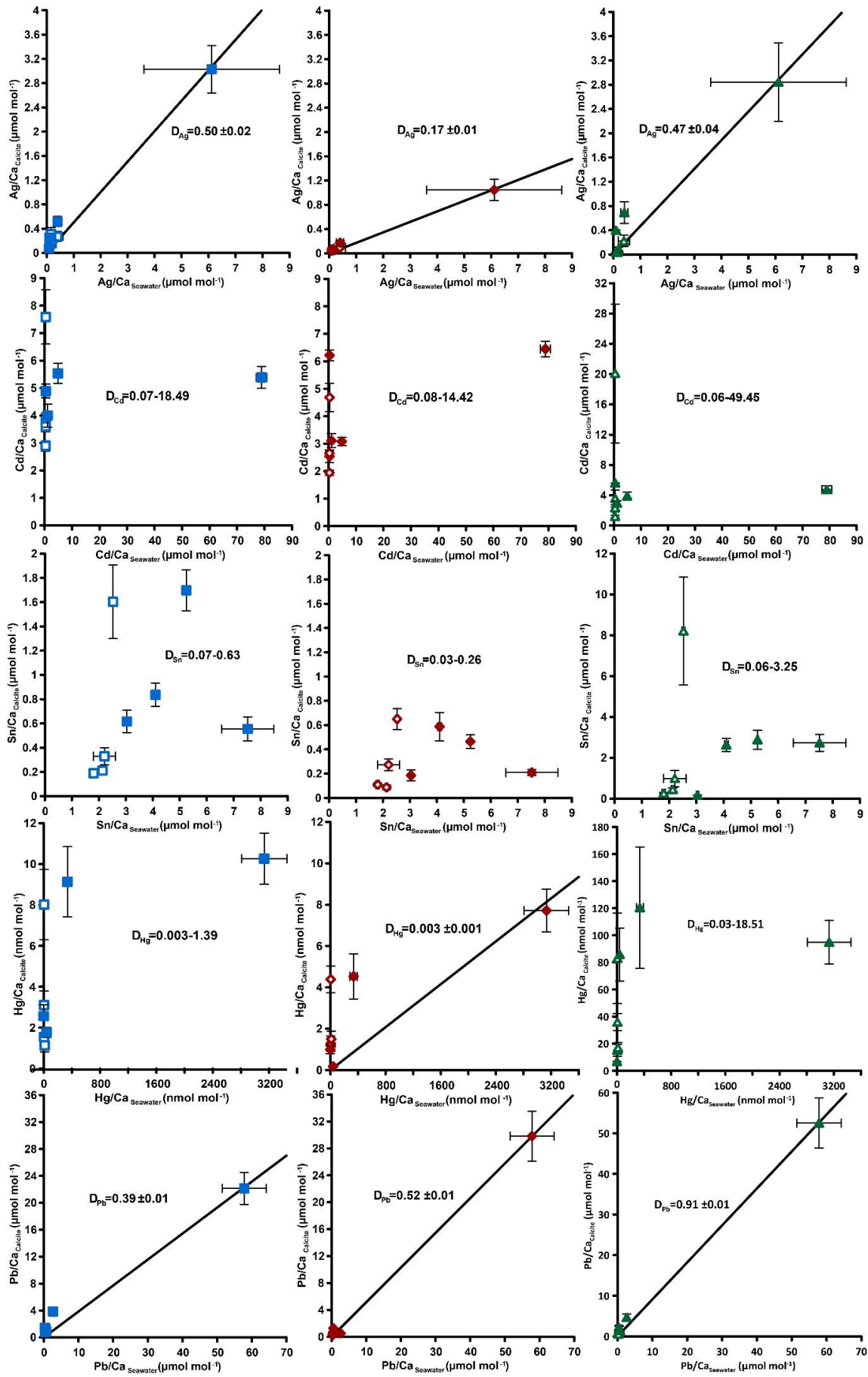
529

530   **3.3.2 Partition coefficient (D_{TE})**

Ammonia aomoriensis

D_{Cr}=0.4-10.3

D_{Mn}=0.38

D_{Ni}=0.06-0.94

D_{Cu}=1.18

D_{Zn}=0.08-0.45

Ammonia batava

D_{Cr}=0.4-6.8

D_{Mn}=0.23

D_{Ni}=0.05-0.41

D_{Cu}=0.60-4.35

D_{Zn}=0.09-0.65

Elphidium excavatum

D_{Cr}=2.1

D_{Mn}=0.34-2.50

D_{Ni}=0.19

D_{Cu}=0.95-5.67

D_{Zn}=0.08-0.53

531

23

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25

535

536 **Figure 4**: Mean TE/Ca values in the foraminiferal calcite versus the mean TE/Ca values in the corresponding

537 culturing medium based on phase 0 to 3. Each data point represents the mean value of all laser ablation ICP—-

538 MS measurements on single foraminiferal chambers built up during the individual culturing phase plotted against

539 the mean metal concentrations in the seawater averaged over the culturing phase (Table 3). Because calculating p-

540 and R^2 values of the regression lines and the D_{TE}´s with the mean per phase resulted in comparable values to when

541 calculating with the overall dataset, we considered this approach adequate. Error bars symbolize the standard error

542 of the mean. The linear regression line (± standard deviation) is displayed when elements showed a significant

543 correlation between seawater and calcite. -D_{TE}'s of *E. excavatum* where considered without values for Phase 0 of

544 the Metal System as only data from one newly formed chamber are available. All values can be found in Table 4.

545 An enlarged graph based on the calculations without phase 3 is provided in the appendix (Fig. B2).

546

547 Partition coefficients for the different trace elements were deduced from molar foraminiferal test TE/Ca and the

548 values from the corresponding culturing medium. Note that the D_{TE} values represent the slope of the regression

549 line when there was a positive correlation detected and were furthermore separately calculated for every individual

550 phase. The range of the calculations based on every individual phase are given when no positive correlation could

551 be detected. Furthermore, calculations were performed with and without phase 3 (Fig. 4 , Fig. B2 and Table 4).

552 Generally, the values varied between 0.003 and 49.45. The majority of D_{TE} were lower than 1 in *A. aomoriensis*

553 (with phase 3 = 61 %, without phase 3 = 57%) and *A. batava* (with phase 3 = 75%, without phase 3 = 73%), i.e.,

554 uptake but no enrichment took place. D_{TE} values derived from *E. excavatum* on the other hand showed a smaller

555 proportion < 1 (with phase 3 = 47%, without phase 3 = 42%). Note that D_{TE} of *E. excavatum* were considered

556 without values for Phase 0 as there were data from only one newly formed chamber available. For most elements

557 (Cr, Mn, Ni, Cu, Cd, Sn, Pb and Hg) D_{TE} derived from *E. excavatum* wereare higher than D_{TE} from the two

558 *Ammonia* species (Table 4, Fig. 4), which showed comparable D_{TE} values for most elements. One exception is Zn,

559 where all D_{Zn} built the exception because all values -wereare within a similar range (D_{Zn} ~ 0.08-0.65) independent

560 of the species. For *A. aomoriensis* D_{Cu} was > 1 and D_{Cd} as well as D_{Pb} were also > 1 when phase 3 was excluded

561 from the calculations. *A. batava* showed D_{TE} values > 1 for most Cr and Cd values (excluding phase 3 and 2) when

562 considering calculations from the individual phases. *Elphidium.* excavatum displayed D_{TE} values > 1 for Cr and

563 Cu for the calculations with phase 3 and also for Pb without phase 3. Furthermore, D_{Cd} and D_{Hg} of *E. excavatum*

564 were also mostly > 1 when looking at calculations based on individual phases (Table 4, Fig. 4). The highest

565 variation between minimum and maximum D_{TE} for all species was found for Cd and Hg. In addition, some

566 elements showed partition coefficients > 1 in a couple of phases and/ or in one of the two culturing systems (e.g.,

567 *E. excavatum*: Mn = Control System Phase 1 to 3; *A. batava*: Cu = all but Control System Phase 0 and Metal

568 System Phase 1; Pb = Control System and Metal System Phase 1; *A. aomoriensis*: Cr = Control System Phase 2

569 and 3, Metal System Phase 1; Ag = Control System Phase 1 and 3, Metal System Phase 2 and 3, Hg: Control

570 System Phase 3, see Table 4, Fig. 4).

571

4. Discussion

4.1 Experimental Uncertainties

Calcein was used for staining the foraminiferal test before they were placed into the culturing system. It can be assumed, that a period of 1 or 2 days for removing excess calcein was sufficient because the youngest chambers were not stained. Calcein binds to Ca and is incorporated into the mineralised calcium carbonate (Bernhard et al., 2004). It is conceivable that the heavy metal incorporation could also be affected by calcein. However, no evidence for such effects has been found so far in a variety of studies (e.g., Hintz et al., 2006; De Nooijer et al., 2007; Dissard et al., 2009). Furthermore, calcein was only used prior to the experiment to mark the last chamber that was grown outside the culturing system. Therefore, the incorporation of the metals measured in subsequent chambers was not affected by the calcein application.

The element concentrations within the culturing medium of each culturing phase were comparably stable for most elements in the control system. In the metal system, the variations were higher, which is due to the punctual input of the multi metal stock solution for reaching the next phase concentration (Table A1, Fig. B1). This sudden ~~adding~~ addition of metals resulted in a high peak concentration in the beginning of the new phase, which equilibrated after a while. This trend was~~is~~ most pronounced in phase 3 as the added amount of the multi metal stock solution was highest for this phase, which was~~is~~ also why the standard error of this phase was~~is~~ comparably high. Furthermore, the variations of the metal concentrations were in a comparable range than those presented in other culturing studies (e.g., Marechal-Abram et al., 2004; De Nooijer et al., 2007; Munsel et al., 2010; Remmelzwaal et al., 2019). Generally, many other studies (e.g., Remmelzwaal et al., 2019; Sagar et al., 2021a; Titelboim et al., 2021) measured the heavy metal concentration in the seawater less frequently than done in this study. Therefore, the stability of metal concentrations during the culturing phases of those studies are often inferred. Furthermore, pollution events in nature are in most cases not persistent and stable but transient as was mirrored by the concentration changes in our experiments.

~~When taking into account that the amount of the stock solution added to the culturing medium of the metal system at the beginning of each culturing phase was elevated 1 to 1.5 order of magnitudes between phases,~~ T~~t~~he measured metal concentrations in the culturing seawater were~~are~~ smaller than expected (Table 3) ~~for phases 0, 1 and 2~~. This in combination with the varying metal concentration within one phase suggested~~s~~ that several processes were~~are~~ affecting the concentration in such a complex culturing system. One possible mechanism was~~is~~ sorption of the metals onto surfaces (e.g., tubing, culturing vessels, plates, organic matter or the foraminiferal test itself), which could have lowered the metal concentration in the culturing medium. Therefore, sorption could have contributed to the overall budget of the metals. On the other hand, Cu appeared~~s~~ to have been released from components of the culturing system even though the system was cleaned before use and was operated with seawater for 14 days before the experiments begun. For instance, the concentration of Cu was high in phase 0, where no metals were added suggesting release from system parts. In phase 1, the Cu concentration decreased meaning the contamination derived from the system was removed by a process similar to that observed for the other metals after additions were made. Similar effects have been reported by De Nooijer et al. (2007) for Cu and Havach et al. (2001) for Cd. Other processes like the uptake of the metals by the foraminifera itself and the growth of algae could further have an influence on the metal concentration in the culturing medium. Germs of algae were introduced accidentally together with the living foraminifera and grew during the experiment. Such processes are difficult to predict and

even more challenging to avoid but probably mirror real environments more realistically better than sterile petri dish experiments do (e.g., Havach et al., 2001; Hintz et al., 2004; Munsel et al., 2010).

Neither the survival rate nor the formation of new chambers was influenced by the elevated metal concentrations during the culturing period. These features were rather constant between the four different phases. Furthermore, no test morphology malformations of the test morphology were recognised. Elevated heavy metal concentrations are thought to induce a higher rate of malformations in benthic foraminifera (e.g., Sharifi et al., 1991; Yanko et al., 1998), whereas recent studies constrained them as a reaction to stressful environments, not necessarily created by high heavy metal concentrations (Frontalini and Coccioni, 2008; Polovodova and Schönfeld, 2008). The lack of malformations in our experiments suggesteds that the foraminifera were neither poisoned by elevated trace heavy metal concentrations nor stressed too much by strongly varying environmental parameters, maintaining a normal metabolism and growth. Reproduction was generally very rare, which may indicate that the conditions were not ideal. In field studies foraminiferal reproduction has been linked to short periods of elevated food supply (e.g., Lee et al., 1969; Gooday, 1988; Schönfeld and Numberger, 2007). The regular feeding of foraminifera in our experiment twice a week at constant rates therefore probably did not provide supply levels that trigger reproduction. Nevertheless, it can be assumed that a sufficient amount of food was provided because after the experiments, leftovers covering the sediment surfaces in the cavities were evident. This would have likely been consumed by the foraminifera if they would have needed more. Furthermore, the foraminifera calcified, which would not be the case if any malnourishment occurred (e.g., Lee et al., 1991; Kurtarkar et al., 2019). Therefore, the nutritional status is unlikely to have influenced the metal uptake by the foraminifera.

The calibrations between the heavy metal concentration in seawater and the foraminiferal shell rely on the TE/Ca values from phase 3 because the difference in seawater concentration was highest compared to other phases. Nevertheless, data points from other phases do play a role and forcing through the origin adds a further fixed point. High variability for D_{TE} values like observed here for Cd or Cu is difficult to explain. Such variability suggests there are factors affecting these metals we do not understand and therefore it is also important to show the data for these elements. Furthermore, the experimental design, especially the mixture of metals, was chosen to best simulate metal conditions in real environments, which could naturally enhance the variability of D_{TE}. This knowledge is indispensable for the application of heavy metal concentrations in foraminifera as a proxy for the heavy metal concentration in seawater.

Calcein was used for staining the foraminiferal test before they were placed into the culturing system. Calcein binds to Ca and is incorporated into the mineralised calcium carbonate (Bernhard et al., 2004). It is conceivable that the trace metal incorporation could also be affected by calcein. However, no evidence has been found by a variety of studies (e.g., Hintz et al., 2006; De Nooijer et al., 2007; Dissard et al., 2009). Furthermore, calcein was only used prior to the experiment to mark the last chamber that was grown outside the culturing system. Therefore, the incorporation of the metals measured in subsequent chambers was not affected by the calcein application.

4.2 Incorporation of heavy metals in the foraminiferal test

Many heavy metals have been demonstrated to be incorporated into the foraminiferal shell (e.g., Cr: Remmelzwaal et al., 2019; Mn: Koho et al, 2015; 2017; Barras et al., 2018; Cu: De Nooijer et al., 2007; Ni: Munsel et al., 2010; Hg: Frontalini et al., 2018a; Cd: Havach et al., 2001; Pb: Frontalini et al., 2018b; Titelboim et al., 2018; Sagar et

650 al., 2021a; 2021b; Zn: Marchitto et al., 2000; Van Dijk et al., 2017), and the incorporation of all of these metals

651 has been ~~was~~ measured here. Additionally, to the best of our knowledge, Sn and Ag were investigated here for the

652 first time. The levels observed were well above control values indicating an elevated incorporation of Ag and Sn

653 into the foraminiferal test calcite with increasing metal concentrations in seawater.

654 Different factors can influence the incorporation of these metals into the foraminiferal test. First of all, the uptake

655 depends on metabolic~~al~~ pathways during the calcification process. Fundamental biomineralization processes of

656 foraminifera are the subject of an ongoing discussion and several (partly) competing models have been proposed

657 ~~so far~~ (e.g., Elderfield and Erez, 1996; Erez, 2003; De Nooijer et al., 2009b, 2014; Nehrke et al., 2013). One model

658 proposes that the foraminifera take up ions directly from the surrounding seawater by endocytosis or by building

659 seawater vacuoles, which are transported to the site of calcification (SOC) (Elderfield and Erez, 1996; Erez 2003;

660 De Nooijer et al., 2009b; 2009a; Khalifa et al., 2016). The SOC is located outside the foraminiferal cell and the

661 formation of new calcite takes place in this zone (see ~~e.g.,~~ De Nooijer et al., 2014 for a summary and illustration).

662 There is~~are~~ evidence that this ~~part~~ SOC is separated from the surrounding seawater (e.g., Spindler, 1978; Bé et al.,

663 1979; De Nooijer et al., 2009b; 2014; Glas et al., 2012; Nehrke et al., 2013). The other competing model suggests

664 that the uptake of ions and the transport to the SOC is performed directly from the seawater across the cell

665 membrane by active trans-membrane-transports (TMT) and/ or passive transport via gaps in the pseudopodial

666 network of the foraminifera (Nehrke et al., 2013; De Nooijer et al., 2014). The dependenc~~ey~~ of heavy metal

667 concentrations in the foraminiferal test on their seawater concentration relies on the prevailing mechanism. ~~A~~

668 ~~B~~biomineralization based on endocytosis ~~does not control the amount of ions that is introduced into the~~

669 ~~foraminiferal cell and is transported to the SOC. Consequently, processes like Rayleigh fractionation are most~~

670 ~~important (e.g., Elderfield and Erez, 1996). This~~ would ~~also mean~~ infer that the metal concentration in the seawater

671 is directly mirrored by their concentration in the foraminiferal shell, which ~~cannot be~~ is not generally supported by

672 the results of our study except for Ag and Pb. ~~Indeed, several~~Several ~~elements~~ metals showed partition coefficients

673 > 1 or <1 when the D_{TE}´s ~~were~~are calculated separately for each culturing phase ~~(see Results section in this~~

674 ~~manuscript)~~. Only Pb and Cr in *E. excavatum* and Cu and Pb in *A. aomoriensis* consistently displayed mean D_{TE}´s

675 > 1 paired with a positive correlation of the concentration in seawater and in the foraminiferal shell, which could

676 indicate a non-selective uptake of these metals~~.~~ meaning uptake not only driven by the chemical properties of the

677 ion such as the size of the metal ion itself. If this would have been the case, D_{TE} values > than 1 would be expected

678 especially for metals ions that are smaller than Ca (Rimstidt et al., 1998). On the other hand, the D_{TE} values of

679 many elements (Ni, Zn, Cd, Hg, Pb) dramatically decreased with increasing concentration in the seawater in the

680 highest metal treatment in all species (Fig. 4). This kind of overload effect ~~was~~ has also been noted ~~also noted~~ by

681 Nardelli et al. (2016) for Zn, by Barras et al. (2018) for Mn, by Mewes et al., (2015) for Mg and~~or~~ by Munsel et

682 al. (2010) for Ni. Nardelli et al. (2016) suggested~~,~~ that some biological mechanism expulse or block these metals

683 if the concentration is ~~getting~~ too high and ~~an~~ imminent intoxication is probable, which may be managed by

684 controlling the ion uptake via TMT. Therefore, it may well be possible that the highest concentration of the metals

685 in our study was close to the tipping point of the biological mechanism taking over and protecting the organism.

686 Besides biologically controlled factors, ~~also~~ physicochemical properties also play an important role when it comes

687 to the uptake of ions. One chemical factor is the aqueous speciation and solubility of the metals. Metals with a free

688 ion form with a charge of 2+ are more similar to Ca^{2+}, which makes incorporation more likely (Railsback, 1999).

689 Nearly all metals in this study were added as dissolved chlorides and therefore had a charge of 2+. The only

690   exceptions were Ag, which was added as $AgNO_3$ with a charge of 1+ and Cr, which was added as $CrCl_3*6\ H_20$.

691   The charge of the cation as such does not seem to make a major difference as Ag was incorporated into all three

692   species and Cr into *E. excavatum* with a significant positive correlation to concentrations in the culturing medium.

693   Furthermore, it is possible that the oxidative state of the ~~elements is changing~~ elements changed due to their pH

694   dependency, which will be discussed for every element separately. Furthermore, other ions with a charge of 1+

695   are also known to be incorporated in calcite. Examples are Li^+ (e.g., Delaney et al., 1985; Hall et al., 2004) and

696   Na^+ (e.g., Wit et al., 2013; Bertlich et al., 2018), which are believed to occupy interstitial positions in calcite where

697   the calcite lattice has defects (Ishikawa and Ichikuni, 1984; Okumura and Kitano, 1986). In addition, rare earth

698   elements with a charge of 3+ are also detected in the foraminiferal calcite (e.g., Haley et al., 2005; Roberts et al.,

699   2012).

700   The aqueous speciation of many metals is strongly influenced by the pH (e.g., Förstner, 1993; Pagnanelli et al.,

701   2003; Spurgeon et al., 2006; Powell et al., 2015; Huang et al., 2017). As the pH during the experiment was stable

702   around 8.0 ± 0.1 (measured twice a week), speciation changes between phases due to varying pH values can be

703   excluded. However, it is possible that some metals were not available in a form that could be readily incorporated

704   in the calcite such as the free ion or carbonate species. Cr was ~~is~~ not available in an optimal speciation to substitute

705   Ca as a pH of 8 would favour Cr^{3+} or Cr^{4+} as well as oxides and hydroxides (Elderfield, 1970; Geisler and Schmidt,

706   1991). Furthermore, the used Cr-salt may not have dissolved completely, even though the multi metal stock

707   solution was heated and stirred during the process. Both in combination may lead to the small variation ~~between~~

708   ~~the different phases~~ in the seawater concentrations between the different phases. Interferences that could possibl~~y~~e

709   have influence~~d~~ the Cr measurements in the water samples are chlorine oxides or hydroxides (e.g., Tan and

710   Horlick, 1986; McLaren et al., 1987, Reed et al., 1994; Laborda et al., 1994). ~~As NaCl blanks were measured,~~

711   ~~these interferences are most likely monitored and can be excluded as a biasing factor. Furthermore,~~

712   ~~M~~measurements of reference materials revealed slightly elevated ~~accurate~~ Cr concentrations compared to those

713   presented in the literature (Table A2), which ~~also corroborates~~indicates that ~~the assumption that these~~ interferences

714   ~~can be neglected~~could be responsible for some of the observed variability for Cr. Similar pH dependant processes

715   could also have a~~e~~ffected Cu ~~as a pH around 8, like in this experiments, favours copper carbonates over free Cu^{2+}~~

716   ~~ions (e.g., Escudero et al., 2008, Millero et al., 2009), which means that the best available speciation was not~~

717   ~~prevailing during the experiments.~~ .Nevertheless, Cu and Cr were taken up by all species and therefore, this factor

718   cannot be decisive when it comes to incorporation of these metals into the foraminiferal shell.

719   If the incorporation of metals would be straightforward and would only depend on the speciation of the metal and

720   other physicochemical factors, the behaviour of the metals would mostly be influenced by the ionic radius in

721   combination with the charge of the metal ions as described for carbonate minerals by Rimstidt et al. (1998). The

722   endocytotic pathway of seawater ~~components~~ into the foraminifer ~~provokes~~ should produce a behaviour of ion

723   incorporation comparable to inorganic calcite precipitation. It was found that cations are incorporated into

724   inorganic calcite by substitution of Ca^{2+} (e.g., Reeder et al., 1999), especially when the effective ionic radius of

725   these ions is comparable to the one of calcium (= 1.0 Å).
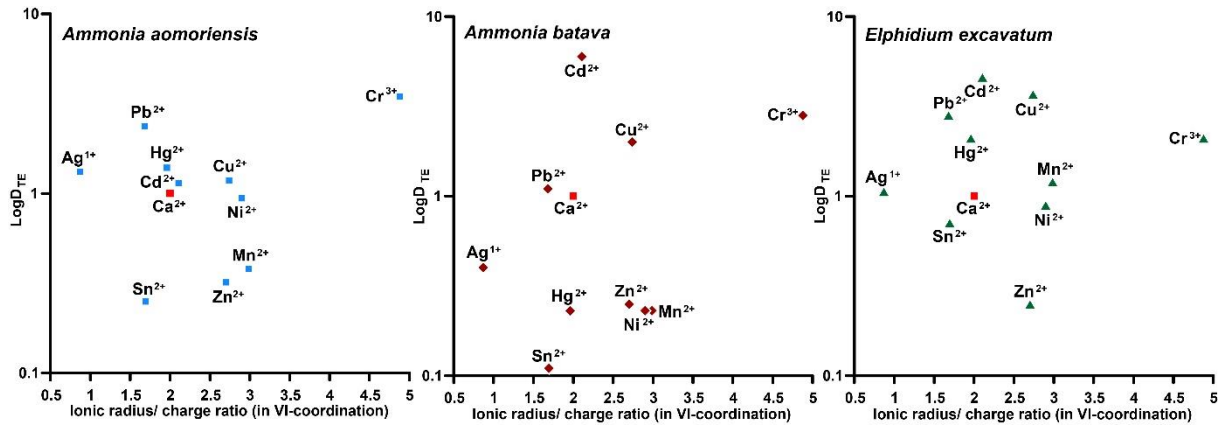
Figure 5: Partition coefficients (log D_{TE}) in dependency of the ionic radius-to-charge ratio (values from Shannon and Prewitt, 1969) in 6-fold coordination for the trace elements analysed in this study. Displayed mean D_{TE}'s are derived from single values calculated for each culturing phase individually (Metal and Control System Phase 0 to 3) and not from the regression lines. Calcium is marked in red. Blue squares represent values from *A. aomoriensis* (left), red diamonds represent values from *A. batava* (middle) and green triangles represent values from *E. excavatum* (right).
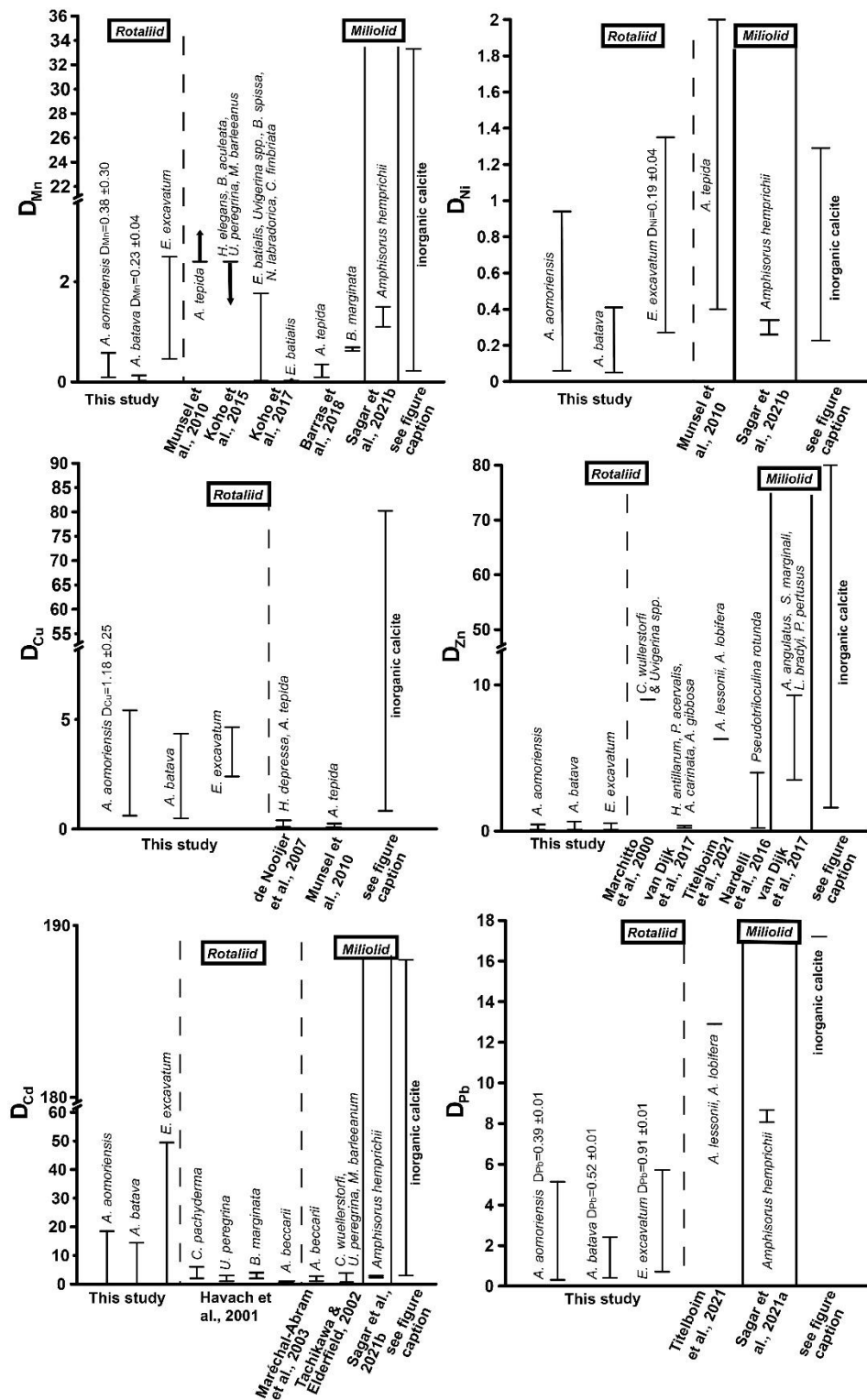
Figure 5: Comparison of D_{TE} values of this study with D_{TE} values from literature of different rotaliid and miliolid foraminiferal species. The range of D_{TE} based on the different culturing phases is given and if a correlation between the heavy metal concentration in seawater and the foraminiferal shell was detected, the mean D_{TE} value ±SD (=slope of the regression line) is also indicated. Note that the x-axis was clipped for some elements. (Literature for inorganic calcite D_{TE} values: Ni = Rimstidt et al., 1998; Alvarez et al., 2021; Mn = Lorens, 1981; Dromgoole and Walter, 1990; Wang et al., 2021; Cu =Kitano et al., 1973; 1980; Wang et al, 2021; Zn = Kitano et al., 1973;

740 1980; Rimstidt et al., 1998; Wang et al., 2021; Cd = Rimstidt et al., 1998; Day and Henderson, 2013; Pb = Rimstidt
741 et al., 1998.)

742

743 Some metals like Mn, Zn and Cu are known to be fundamentally necessary as micro-nutrients to maintain
744 biological and physiological function of a cell (e.g., Mertz, 1981; Tchounwou et al., 2012; Martinez-Colon et al.,
745 2009; Maret, 2016). Therefore, these elements should preferentially be taken up into the foraminiferal cell, where
746 they ~~were~~ are used for further processes. This in turn could lead to the consumption of these metals before they
747 can be incorporated into the foraminiferal tests. The artificial sea salt used in this study ensured that these elements
748 were present in a sufficient amount of micronutrients. All of these ions have a similar ionic radius (Cu = 0.73 Å,
749 Mn = 0.67 Å, Zn = 0.74 Å) in six-fold coordination (Rimstidt et al., 1998), which would also suggest, that their
750 behaviour is comparable. The ionic radii are much smaller than that of Ca, but are rather similar to Mg (0.72 Å,
751 Rimstidt et al., 1998) ~~(Fig. 5).~~.

752 Mn showed~~s~~ a positive correlation between its concentration in seawater and the foraminiferal test in the two
753 *Ammonia* species when the calculations include~~d~~ phase 3. This indicates that this element serves as a well-behaved
754 proxy influenced mainly by its concentration in seawater. However, *E. excavatum* di~~do~~d not show this positive
755 correlation. D_{Mn} values of this study were comparable with rotaliid and miliolid species and partly with D_{Mn} values
756 from inorganic precipitation (Fig. 5). ~~Furthermore, D_{Mn} values of this study calculated with phase 3 (A.~~
757 ~~aomoriensis D_{Mn} = 0.38, A. batava D_{Mn} = 0.23, E. excavatum D_{Mn} = 0.34-2.50) are comparable in range of those~~
758 ~~presented in Koho et al. (2015, 2017: D_{Mn} = 0.34-2.50) and Barras et al. (2018: D_{Mn} = 0.09-0.35), but lower than~~
759 ~~Munsel et al. (2010) reported (D_{Mn} > 2.4) except for E. excavatum.~~ Species-specific ~~differences~~ partition
760 coefficients of elements like Mg or Na are already reported in the literature~~in partition coefficients not only for~~
761 ~~Mn but also for other elements like Mg or Na are common phenomena~~ (e.g., Toyofuku et al., 2011; Barras et al.,
762 2018; Wit et al., 2013) and could also explain the ~~offset~~ different ~~of the~~ D_{TE} values ~~from~~ of *E. excavatum* in this
763 study (see below). Furthermore, it is known that the presence of toxic metals such as Cd, Ni or Hg can inhibit the
764 uptake of essential metals like Mn into the cell if these metals are present in low concentrations (e.g., Sunda and
765 Huntsman, 1998a, 1998b). It is possible that this mechanism is more pronounced in *E. excavatum* than in the
766 *Ammonia* species. Zn was clearly incorporated above control level~~s~~ into all three species, but it´s behaviour wa~~s~~s
767 influenced by more factors than the concentration of Zn in the culturing medium. ~~This can be inferred by the fact~~
768 ~~that there was no significant correlation recognised between Zn concentration in calcite and seawater~~ (Fig. 4, Table
769 4). D_{Zn} values of this study are in good agreement with those calculated by Van Dijk et al. (2017) for four hyaline
770 species ~~(D_{Zn} = 0.2-0.36)~~ and Nardelli et al. (2016) for the miliolid *Pseudotriloculina rotunda* (Fig. 5) ~~(D_{Zn} = 0.2-~~
771 ~~4.0), especially when phase 3 is excluded from the calculation (this study, without phase 3: A. aomoriensis D_{Zn} =~~
772 ~~0.21-0.45, A. batava D_{Zn} = 0.10-0.65, E. excavatum D_{Zn} = 0.22-0.53).~~ Other studies reported higher values.
773 ~~between 3.5 and 9, though for rotaliid taxa like Cibicidoides wullerstorfi and Uvigerina spp. (Marchitto et al.,~~
774 ~~2000; Van Dijk et al., 2017).~~ It is again possible that the mixture of metals ~~is inhibiting~~ inhibited the uptake of
775 essential metals like Zn similar to Mn. Cu showed~~s~~ a simple well-behaved proxy behaviour with a significant
776 positive correlation in *A. aomoriensis* but not in the other two species. The D_{Cu} presented ~~by Munsel et al. (2010)~~in
777 the literature for rotaliid species ~~(D_{Cu} = 0.08-0.25) and De Nooijer et al. (2007) (D_{Cu} = 0.1-0.4) are~~is lower than
778 D_{Cu} from this study. Inorganic values were mostly higher (Fig. 5). ~~(A. aomoriensis D_{Cu} = 1.18, A. batava D_{Cu} =~~
779 ~~0.60-4.35, E. excavatum D_{Cu} = 0.95-5.67). Only the lowest values of this study are in the same order of magnitude.~~

These differences could arise from the lower concentration of Cu in this study or from the mixture of metals. It is also reported, that the exposure to more than one metal can cause an increased uptake of another metal into the cell (Archibald and Duong, 1984; Martinez-Finley et al., 2012; Bruins et al., 2000; Shafiq et al., 1991). If more Cu is taken up into the cell~~, well may be that~~ after the usage of Cu as micro-nutrient~~,~~ more Cu is left over and ~~is actively~~could possibly be deposited into the calcite. It is therefore conceivable that one particular metal in our study was effecting a co-uptake of Cu, which lead to an elevated incorporation into the calcite as compared to other studies.

The non-~~ ~~essential elements Hg, Cd and Pb are not used in physiological processes and are therefore believed to have a higher toxic potential (Barbier et al., 2005; Raikwar et al, 2008; Ali and Khan, 2019). This ~~in turn~~could first of all make the foraminifera prevent the uptake of these metals into their cell. ~~lead to the assumption, that these metals are incorporated into the foraminiferal cell to a smaller amount. This could also~~But if the uptake of heavy metals into the cell cannot be prevented, the foraminifera may remove the metals to their shell instead of keeping them in their cell. ~~result in an enhanced removal of these metals~~This is a common mechanism for avoiding intoxication~~, because they are potentially harmful for marine life and could trigger mechanisms that prevent the foraminifera from intoxication~~ as reported for various organisms (benthic foraminifera: Bresler and Yanko, 1995; Yeast: Adle et al., 2007; Bacteria: Shaw and Dussan, 2015; Microalgae: Duque et al., 2019). Furthermore, this would mean that~~it could increase~~ the ~~input~~incorporation of these metals into the foraminiferal calcite ~~as a further removing mechanism~~increases. The ionic radii of Pb^{2+} in calcite-coordination is 1.19 Å, which is remarkably higher than those of Hg^{2+} (1.02 Å) and Cd^{2+} (0.95 Å), which are comparable to Ca.~~ (Fig. 5).~~ This similarity should also favour the incorporation of Cd and Hg into calcite, which holds only partly true, as Cd showe~~d~~s no trends with complex behaviour, but Hg ~~correlates~~was linearly incorporated in *A. batava* and in *E. excavatum* if the high concentrations of phase 3 are excluded~~is not integrated into the calculations~~. ~~This indicated that the incorporation of Cd is not straight-forward and is indeed depending on more complex factors than seawater concentration of Cd. Nevertheless, Cd is incorporated well above control level in all three species. Because the ionic radius of Pb is bigger than that of Ca a smaller degree of Ca^{2+}-substitution following the ionic radius to charge ratio theory after Rimstidt et al. (1998) is expected. This is not the case as~~ Pb emerged as a well-behaved proxy under these experimental conditions with a.~~ A~~ll three species incorporat~~ing~~ed Pb ~~with a significant positive trend indicating that the main controlling factor is the seawater concentration of Pb~~linearly (Fig. 4, Table 4). ~~Calculated D_{Pb} values including phase 3 of this study are around 1 for *E. excavatum* (D_{Pb} = 0.91), but lower for *A. aomorienis* (D_{Pb} = 0.39) and *A. batava* (D_{Pb} = 0.52). When excluding phase 3, D_{Pb} values of *E. excavatum* (D_{Pb} = 2.0) and *A. aomoriensis* (D_{Pb} = 1.6) are getting even higher, which is may connected to the overload effect.~~ When c~~C~~omparing the D_{Pb} values ~~around 8.4 determined by Sagar et al. (2021), who cultured the large benthic foraminifer *Amphisorus hemprichii*, a symbiont bearing, miliolid species, with varying Pb concentrations (0.5-80 µm l⁻¹) that are comparable to our concentration range (~ 0.11-30 µm l⁻¹), the D_{Pb} of this study are a little lower.~~ in the literature, our D_{Pb} are slightly lower (Fig. 5). ~~Nevertheless, D_{Pb} values from the present study are partly higher in individual phases (*A. aomorienis* D_{Pb}: individual phases without phase 3 = 1.3-5.14; *A. batava* D_{Pb}: Control System Phase 3 = 2.52; *E. excavatum* D_{Pb}: Control System Phase 0-2 and Metal System Phase 1 D_{Pb} ≥ 3) and thereby match the values from Sagar et al. (2021) better.~~ For Hg, no partition coefficients ~~were~~are published so far. D_{Cd} values from different studies ~~ranged between 0.7 and 4~~ (Havach et al., 2001; Tachikawa and Elderfield, 2002; Maréchal-Abram et al., 2004, Sagar at al., 2021b)~~ and are overall lower than D_{Cd} values from the present study (*A. aomoriensis* D_{Cd} = 0.07-18.49, *A. batava* D_{Cd} = 0.08-14.42, *E. excavatum* D_{Cd} = 0.06-49.45), but mean D_{Cd} values are of the same~~

821 ~~order of magnitude.~~ have overall a smaller range of D_{Cd} values than found here (Fig. 5). The greater variability in
822 D_{Cd} of our study makes a comparison difficult. ~~Cd is known to mimic other metals that are essential to diverse~~
823 ~~biological functions (Martelli et al., 2006; Urani et al., 2015; Losada Ros et al., 2020). Consequently, one may~~
824 ~~argue that cadmium follows a Trojan horse strategy to get assimilated (Martelli et al., 2006), which could also be~~
825 ~~the case for Hg. Smith et al. (2020) found a strong correlation between the Cd concentration in the seawater and~~
826 ~~the foraminiferal shell in the species *Haynesina germanica, Ammonia tepida, Quinqueloculina sabulosa* and~~
827 ~~*Triloculina oblonga*. This trend was not present in our study, as no species showed a covariance (Fig. 4, Table 4)~~
828 ~~indicating the influence of more complex mechanisms than simple incorporation of the seawater ion concentration.~~
829 ~~Investigations on deep-water benthic foraminifera like *Cibicidoides wuellerstorfi, Uvigerina peregrine* and~~
830 ~~*Melonis barleeanum* revealed a smaller range in the D_{Cd} values (e.g., McCorkle et al., 1995; Tachikawa and~~
831 ~~Elderfield, 2002), which most likely originated from the smaller variability of the Cd concentration in their~~
832 ~~seawater as compared to this study.~~

833 The importance of other metals like Sn, Cr, Ag and Ni is not fully understood yet but some of them are believed
834 to have certain biological functions in the cells of animals or plants (Horovitz, 1988; Mertz, 1993; Lukaski, 1999;
835 Pilon-Smits et al., 2009; Hänsch & Mendel, 2009; Chen et al., 2009). For example, Ni is important for plants and
836 bacteria (Poonkothai and Vijayavathi, 2012; Maret, 2016). The ionic radii of these metals in calcite-coordination
837 is rather different (Sn = 1.18 Å; Ag = 1.15 Å; Cr = 0.62 Å; Ni = 0.69 Å) and deviate from the ionic radius of
838 ~~calcite~~Ca^{2+}, too~~.~~. ~~(Fig. 5).~~

839 Ni was incorporated with a positive trend in *E. excavatum,* but with no clear trend in the *Ammonia* species (Fig. 4,
840 Table 4). ~~Munsel et al. (2010) found a similar trend in *A. tepida* and their calculated~~ D_{Ni} values ~~(D_{Ni} = 0.4-2.0)~~
841 from rotaliid and miliolid foraminifera and from inorganic calcite ~~-~~ are in good agreement with our results (Fig. 5).
842 ~~when the highest metal phase of this study is not taken into account (*A. aomoriensis* D_{Ni} = 0.19-0.94, *A. batava*~~
843 ~~D_{Ni} = 0.15-0.41, *E. excavatum* D_{Ni} = 0.64-1.35).~~ Ag exhibited a strong positive correlation between seawater and
844 foraminiferal shell in all three foraminiferal species. Partition coefficients for Ag (*A. aomoriensis* D_{Ag} = 0.5~~0 ±0.02~~
845 ~~6~~, *A. batava* D_{Ag} = 0.17 ±0.01, *E. excavatum* D_{Ag} = 0.47 ±0.04) cannot be compared to other studies as no literature
846 data are~~is~~ available~~, but the general trend, the ionic radius and the D_{TE} values are comparable to other elements in~~
847 ~~this study e.g., to Pb. Ag and Ni both display a well behaved proxy for the estimation of seawater concentration~~
848 ~~of these ions (Fig. 4, Table 4).~~.

849 Cr and Sn, on the other hand, were not incorporated in a higher amount when the concentration of these metals in
850 the culturing medium was raised, except for Cr in *E. excavatum*, which showed a positive correlation. The D_{Cr}
851 values presented in Remmelzwaal et al. (2019) (D_{Cr} > 107), based on culturing experiments with the tropical,
852 symbiont bearing foraminifera *Amphistegina spp.*, are at least one order of magnitude higher than D_{Cr} values in
853 this study (*A. aomoriensis* D_{Cr} = 0.74-10.3, *A. batava* D_{Cr} = 0.4-6.8, *E. excavatum* D_{Cr} = 2.1 ±0.28). ~~For Sn, no~~
854 ~~comparative studies are available.~~ One possible reason for dynamics of Cr are the comparable low concentrations
855 in the culturing medium and furthermore, the differences between the phases ~~are~~ were also very low (Fig. 3, Fig.
856 B1 and Table 3). It may be that the concentration of Cr needs to be further elevated and the concentration range
857 needs to be extended before the foraminifera are able to incorporate Cr with significant differences between
858 concentrations. For Sn, no comparative studies are available so w~~W~~e may speculate that the same could apply for
859 Sn. ~~Besides, Remmelzwaal et al. (2019) suggested, that Cr in foraminiferal shells is mainly a result of post-~~
860 ~~depositional overprinting. Diagenetic processes are very unlikely to play a role in our experiments~~Nevertheless,

861 we ~~, which would explain, why we do not~~ recognised a correlation between the concentration of Cr in the culturing

862 medium and in the foraminiferal calcite of *E. excavatum*, but not for both *Ammonia* species.

863

4.3 Interspecies variability

865 The three different species cultured in this study clearly incorporated the same metal in different ways, which is

866 most visible in the overall higher TE/Ca values of *E. excavatum* compared to species from the genus *Ammonia*

867 (Fig. 4 and 5, Table 4). Koho et al. (2017) suggested that these differences in the incorporation result from different

868 microhabitats used by different foraminiferal species. This might be true in nature. In our experiments, however,

869 the sediment in the cavities was only a few mm thick and no redox horizon was recognised when recovering the

870 foraminifera after the experiment. Therefore, all foraminifera were living in the same microhabitat. Leftover food

871 may have created a microhabitat but this effect would have been the same in all cavities and therefore cannot

872 account for the differences between the species.

873 ~~Another possible reason for the difference between *E. excavatum* and *Ammonia* species is their nutrition strategy.~~

874 ~~As discussed above, D$_{TE}$ values were markedly higher in symbiont-bearing species. *Ammonia* species do not~~

875 ~~harbour endosymbionts (Jauffrais et al., 2016), whereas at least five intertidal *Elphidium* species were husbanding~~

876 ~~diatom chloroplasts, including *E. excavatum* (e.g., Pillet et al., 2011; Cesborn et al., 2017). However, an earlier~~

877 ~~study could not corroborate the assumption that *Elphidium* species living at greater water depth in the Baltic Sea~~

878 ~~may contain endosymbiontic zooxantellae (Schönfeld and Numberger, 2007).~~ In our experiment, dead

879 *Nannochloropsis* were fed, which is certainly not the preferred food source for *E. excavatum* (Pillet et al., 2011).

880 This could lead to a slower growth and *E. excavatum* built on average only 1 chamber during the individual

881 culturing period of 21 days while *Ammonia* species built more than four chambers. Furthermore, *E. excavatum* did

882 not reproduce, even though the culturing period is close to the generation time of this species (Haake, 1962). When

883 growth is slower, it could be possible that a higher amount of a metal is incorporated into the shell, which would

884 lead to higher TE/Ca values in this species. It is possible that a more preferred food source would have stimulated

885 enhanced growth and influenced the incorporation of heavy metal into the shells of *E. excavatum*. For instance,

886 the closely related species *E. clavatum* prefers bacillariophycean diatoms (Schönfeld and Numberger, 2007). It

887 may also be possible that *E. excavatum* is simply a slower growing species than *Ammonia*, which seems not to be

888 necessarily connected to a specific food source (e.g., Haynert et al., 2020). One could assume that a slower growth

889 would provide more time to remove potentially toxic metals from the cell to the foraminiferal shell, which could

890 explain why *E. excavatum* incorporated a higher metal concentration than *A. aomoriensis* and *A. batava*.

891 Another possibility for the higher metal concentration found in *E. excavatum* is the timing of chamber formation.

892 As *E. excavatum* formed on average one new chamber, it is possible that this chamber was formed during the high

893 peak in the metal concentration during the beginning of the culturing phases (Fig. B1, Table A1). This could in

894 turn lead to a higher uptake of the metals and apparently higher D$_{TE}$ values. Both *Ammonia* species on the other

895 hand, formed more chambers, which makes it most likely that ~~not only~~ the first high concentrations did not

896 particularly influences ~~their~~ their overall D$_{TE}$ value. Unfortunately, it is not possible to constrain exactly when the

897 specimens formed their new chambers. It was checked whether the evolution of the metal concentration in seawater

898 of phase 3 was reflected in the intra-test (chamber to chamber) data for the two *Ammonia* species. Particularly, the

899 initial high concentration of certain heavy metals was found in the first chambers of very few individuals after the

staining (i.e. the first chamber built in culture). This is most likely due to the individual timing of calcification. Furthermore, it could also be possible that the foraminifera did not calcify during the first high peak due to an initial intoxication. Therefore, a mean value over the whole culturing phase was considered as most representative.

Comparing *Ammonia* and *Elphidium* species showed that the D_{TE} of the *Ammonia* species of this study are partly comparable to literature data (Fig. 5).

D_{TE} values are known to be generally higher in tropical high-Mg calcite taxa like *Amphistegina* (e.g., Titelboim et al., 2021) and also high-Mg miliolid taxa like *Amphisorus* (e.g., Sagar et al., 2021a) incorporate a higher amount of metals compared to rotaliid low-Mg taxa like *Ammonia* or *Elphidium*. Comparing our data with high-Mg species, it is visible that this trend can be partly confirmed (Fig. 5). For Mn, both *Ammonia* species of this study show lower values than miliolid species but D_{Mn} of *E. excavatum* is comparable. D_{Ni} values of *A. hemprichii* determined by Sagar et al. (2021b) display the same range as the values for low-Mg species here and furthermore D_{Zn} values of the miliolid *P. rotunda* (Nardelli et al., 2017) overlap with our findings. On the other hand, D_{Zn} values from miliolids in van Dijk et al., (2017) and high-Mg rotaliids from Titelboim et al. (2021) are much higher. The same trend is observed for D_{Pb} (Titelboim et al., 2021; Sagar et al., 2021a). When comparing the Zn/Ca concentration in the foraminiferal shell directly to values from Titelboim et al. (2018), who analysed the Cu, Zn and Pb concentration in rotaliid and miliolid species from a field site, our values show similarities with both groups. Zn/Ca in the foraminiferal calcite of our study was a maximal ~ 68 µmol/mol, which is slightly lower than reported in Titelboim et al. (2018) for the low-Mg species *Pararotalia calcariformata* (195 µmol/mol), but much lower than Zn/Ca reported for the high-Mg species *Lachlanella* (2540 µmol/mol). Differences between the low-Mg species may be due to different concentrations in the seawater the foraminifera grew in. As the seawater metal concentration is not given in Titelboim et al. (2018) this cannot be evaluated. It may also be possible hat high-Mg species have more defects in their tests, which would result in more interstitial space, leading to more space for ions other than Ca. Maximum Cu/Ca values of our study are ~ 23 µmol/mol in *E. excavatum*, which fits the findings of Titelboim et al. (2018) for rotaliid species (*P. calcariformata* ~21 µmol/mol) and is lower than in high-Mg species (*Lachlanella* ~ 186 µmol/mol). Pb/Ca of ~ 12 µmol/mol in *P. calcariformata* described by Titelboim et al. (2018) are lower than found here (max. Pb/Ca in *E. excavatum* of this study ~ 53 µmol/mol), whereas our findings are more comparable to *Lachlanella* (Pb/Ca ~ 125 µmol/mol).

4.4 Application of TE/Ca values in the foraminiferal shell

Table 5: Comparison of the heavy metal concentrations in different regions of the world to values used for the culturing experiments in this study. It is indicated whether the values of this study are comparable to environmental values or if values from this study are higher or lower. (EPA = Environmental Protection Agency, USA)

Element	Study area	Concentration in µg l^{-1}	Comparable?	Reference
		0.06-4.61		This study
	EPA Recommended Values (acute)	1.9	yes	Prothro, 1993
Ag	Restronguet Creek, U.K. + Adriatic Sea	0.0025-0.03	yes	Barriada et al., 2007
	Ibaraki coast + Watarase river	0.014-0.03	yes	Shijo et al., 1989
Cd		0.14-30.61		This study

	Location	Value	Comparable	Reference
	EPA Recommended Values (chronic)	7.9	yes	Prothro, 1993
	Suva, Fiji	150-250	no, low	Arikibe and Prasad, 2020
	Black Sea in Rize, Turkey	1-3	yes	Baltas et al., 2017
	Gulf of Chabahar, Oman Sea	0.15-0.19	yes	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	200-1580	no, low	Chakraborty et al., 2014
	East London + Port Elizabeth harbours, U.K.	200-72600	no, low	Fatoki and Mathabatha, 2001
	Yalujiang Estuary, China	0.83-1.33	yes	Li et al., 2017
	Gulf San Jorge, Argentina	0.01-0.09	yes	Muse et al., 1999
	Alang-Sosiya ship scrapping yard, Gulf of Cambay, India	34-560	yes	Reddy et al., 2005
	Kamal estuary, Jakarta	0.01-0.02	no, high	Putri et al., 2012
	Jakarta Bay	0.04-0.104	yes	Williams et al., 2000
	Kepez harbor of Canakkale, Turkey	19-73800	yes	Yılmaz and Sadikoglu, 2011
Cr		0.1-14.0		**This study**
	EPA Recommended Values (chronic)	50	no, low	Prothro, 1993
	Gulf of Chabahar, Oman Sea	20.16-21.46	yes	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	260-3010	no, low	Chakraborty et al., 2014
	Yalujiang Estuary, China	0.113-0.14	yes	Li et al., 2017
	Gulf San Jorge, Argentina	0.04-0.5	yes	Muse et al., 1998
	Jakarta Bay	0.511-5.25	yes	Williams et al., 2000
	Alang-Sosiya ship scrapping yard, Gulf of Cambay, India	35-765	no, low	Reddy et al., 2005
Cu		0.6-6.2		**This study**
	EPA Recommended Values (chronic)	3.1	yes	Prothro, 1993
	Suva, Fiji	880-10290	no, low	Arikibe and Prasad, 2020
	Black Sea in Rize, Turkey	30-242	no, low	Baltas et al., 2017
	Gulf of Chabahar, Oman Sea	3.37-5.74	yes	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	1350-1850	no, low	Chakraborty et al., 2014
	East London + Port Elizabeth harbours, U.K.	500-42600	no, low	Fatoki and Mathabatha, 2001
	Yalujiang Estuary, China	1.8-4.7	yes	Li et al., 2017
	Gulf San Jorge, Argentina	0.02-0.65	yes	Muse et al., 1998
	Jakarta Bay	0.405-4.04	yes	Williams et al., 2000
	Alang-Sosiya ship scrapping yard, Gulf of Cambay, India	32-3939	yes	Reddy et al., 2005
Hg		0.00035-0.273		**This study**
	EPA Recommended Values (chronic)	0.94	yes	Prothro, 1993
	South Florida Estuaries	0.0034-0.0074	yes	Kannan et al., 1998
	Guadalupe River and San Francisco Bay, California	0.0017-0.135	yes	Thomas et al., 2002
	Vembanad, India	0.0024-0.206	yes	Ramasamy et al., 2017
	Kamal estuary, Jakarta	0.1-0.2	yes	Putri et al., 2011
	Yalujiang Estuary, China	0.006-0.049	yes	Li et al., 2017
Mn		320-549		**This study**
	Black Sea in Rize, Turkey	3-14	yes	Baltas et al., 2017
	Gulf of Chabahar, Oman Sea	15.43-24.76	no, high	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	13000-18000	no, low	Chakraborty et al., 2014

	East London + Port Elizabeth harbours, U.K.	300-23900	yes	Fatoki and Mathabatha, 2001
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	31-4920	yes	Reddy et al., 2005
Ni		2.3-24.3		**This study**
	EPA Recommended Values (chronic)	8.2	yes	Prothro, 1993
	Suva, Fiji	230-800	no, low	Arikibe and Prasad, 2020
	Black Sea in Rize, Turkey	0.006-0.036	yes	Baltas et al., 2017
	Gulf of Chabahar, Oman Sea	16.42-17.14	yes	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	190-330	no, low	Chakraborty et al., 2014
	Jakarta Bay	0.058-5.25	yes	Williams et al., 2000
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	32-944	yes	Reddy et al., 2005
Pb		0.11-28.35		**This study**
	EPA Recommended Values (chronic)	5.6	yes	Prothro, 1993
	Suva, Fiji	880-1770	no, low	Arikibe and Prasad, 2020
	Black Sea in Rize, Turkey	6-130	yes	Baltas et al., 2017
	Gulf of Chabahar, Oman Sea	4.24-4.25	yes	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	20-120	yes	Chakraborty et al., 2014
	East London + Port Elizabeth harbours, U.K.	600-16300	no, low	Fatoki and Mathabatha, 2001
	Yalujiang Estuary, China	0.4-1.8	yes	Li et al., 2017
	Gulf San Jorge, Argentina	0.1-0.5	yes	Muse et al., 1998
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	30-2036	yes	Reddy et al., 2005
	Kamal estuary, Jakarta	1.3-4	yes	Putri et al., 2011
	Jakarta Bay	0.485-3.62	yes	Williams et al., 2000
	Kepez harbor of Canakkale, Turkey	49-9390	yes	Yılmaz and Sadikoglu, 2011
Sn		0.86-3.95		**This study**
	estuarine seawater, Galicia Coast, Spain	0.53-1.23	yes	Bermejo-Barrera et al., 1999
	U.S. and European rivers	0.0001-0.1	yes	Byrd and Andreae, 1982
Zn		30.0-226.9		**This study**
	EPA Recommended Values (chronic)	81	yes	Prothro, 1993
	Suva, Fiji	80-1450	yes	Arikibe and Prasad, 2020
	Black Sea in Rize, Turkey	38-178	yes	Baltas et al., 2017
	Gulf of Chabahar, Oman Sea	18.01-22.62	yes	Bazzi, 2014
	Gulf of Kutch, Arabian Sea	11000-31000	no, low	Chakraborty et al., 2014
	East London + Port Elizabeth harbours, U.K.	500-27600	yes	Fatoki and Mathabatha, 2001
	Yalujiang Estuary, China	9.2-19.6	yes	Li et al., 2017
	Gulf San Jorge, Argentina	0.01-0.55	no, high	Muse et al., 1998
	Jakarta Bay	2-30.1	yes	Williams et al., 2000
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	33-5832	yes	Reddy et al., 2005

932

933 During the past years, many studies were performed to assess the pollution level of seawater. The range of heavy

934 metal concentrations in the culturing medium of this study are compared to the metal concentrations in polluted

935 environments measured over the past 40 years in different regions all over the world (Table 5). The data

936 ~~demonstrate that the concentration range from our study is in good agreement with threatened environments in~~
937 ~~San Francisco Bay, California (Thomas et al., 2002), the Black Sea, Turkey (Baltas et al., 2017), the Gulf of~~
938 ~~Chabahar, Oman Sea (Bazzi, 2014), the Restronguet Creek, U.K., the Adriatic Sea (Ag; Barriada et al., 2007), the~~
939 ~~Yalujiang Estuary, China (Li et al., 2017), the Gulf of San Jorge, Argentina (Muse et al., 1999), Vembanad and~~
940 ~~the Gulf of Cambay, India (Ramasamy et al., 2017; Reddy et al., 2005), Kepez harbor of Canakkale, Turkey~~
941 ~~(Yılmaz and Sadikoglu, 2011), Jakarta (Williams et al., 2000; Putri et al., 2012) and with polluted U.S. and~~
942 ~~European rivers (Byrd and Andreae, 1982; Kannan et al., 1998; Thomas et al., 2002). Furthermore, the maximum~~
943 ~~metal concentration as recommended by the EPA is the lower boundary of the concentration range from this study~~
944 ~~(Prothro, 1993). A lower concentration than the EPA value is also covered by our study during the control phase~~
945 ~~or in the control system. This enables us to assess metal levels at the very beginning of the harmful pollution phase~~
946 ~~in environments, which could be used as an early-warning system for the ecological status of an area (Sagar et al.,~~
947 ~~2021). Furthermore, it allows to assess the effectiveness of contamination reducing measures. This advantage will~~
948 ~~be important in the future for the possibility to intervene or to apply more promising measures within an adequate~~
949 ~~time frame. In some regions of the world, seawater heavy metal concentrations are higher than in this study.~~
950 ~~Examples are Suva, Fiji (Arikibe and Prasad, 2020), the Gulf of Kutch, Arabian Sea (Chakraborty et al., 2014) or~~
951 ~~the East London and Port Elizabeth harbours, U.K. (Fatoki and Mathabatha, 2001) (Table 5). These areas seem to~~
952 ~~be extremely polluted, which would make it necessary to apply a higher metal concentration to the cultured~~
953 ~~foraminifera if a reconstruction covering these values should be made. However, this study clearly indicates a~~
954 ~~reduced uptake of metals of interest, when the concentration of these metals in the seawater is exceeding a certain~~
955 ~~threshold value (here between phase 2 and 3). This will make it generally difficult to model extreme high pollution~~
956 ~~levels. Indeed, it is possible to distinguish between a heavy and a moderate pollution level. Overall, the~~
957 ~~concentration of heavy metals in seawater should be decreasing all over the world due to a rigorous legislation for~~
958 ~~reduction of the heavy metal input into the environment, and due to various emission reducing measures that are~~
959 ~~applied already. This means that the concentration range of metals covered by this study is adequate for future~~
960 ~~research and monitoring of polluted systems.~~

961

962 **5 Conclusion**

963 ~~The aim of this study was to assess the incorporation of heavy metals into the foraminiferal calcite as a function~~
964 ~~of their concentration in the seawater the foraminifera calcified in. C~~Culturing experiments with different
965 foraminiferal species (*A. aomoriensis*, *A. batava* and *E. excavatum*) that were exposed to a mixture of ten different
966 metals (Cr, Mn, Ni, Cu, Zn, Ag, Cd, Sn, Hg and Pb) at varying concentrations (Table 3, Fig. 3, Fig. B1) were
967 carried out ~~to gain further insights into the uptake of heavy metals.~~and ~~l~~Laser ablation ICP-MS analysis of the
968 newly formed calcite revealed the following:

969   1.   ~~-~~All metals used in this study were incorporated into the foraminiferal calcite of all three species (Fig. 4,
970   Table 4).

971   2.   ~~S~~species-specific differences in the incorporation of heavy metals occurred. ~~Nevertheless, all metals used~~
972   ~~in this study were incorporated into the foraminiferal calcite of all three species (Fig. 4, Table 4).~~

3. The following metals showed a positive correlation between the metal concentration in seawater and the foraminiferal calcite inferring that the uptake of these metals mainly depends on its concentration in seawater:

   a. *Ammonia aomoriensis*: D_{Mn} = 0.38 ±0.3, D_{Cu} = 1.18 ±0.25, D_{Ag} = 0.50 ±0.02, D_{Pb} = 0.39 ±0.01

   b. *Ammonia batava*: D_{Mn} = 0.23 ±0.04, D_{Ag} = 0.17 ±0.01, D_{Hg} = 0.003 ±0.001; D_{Pb} = 0.52 ±0.01

   c. *Elphidium excavatum*: D_{Cr} = 2.1 ±0.28, D_{Ni} = 0.19 ±0.04, D_{Ag} = 0.47 ±0.04, D_{Pb} = 0.91 ±0.01

4. Other metals like Zn, Sn and Cd showed no clear correlation between seawater and calcite, which may be linked to the mixture of metals leading to synergetic effects.

5. D_{TE} values of Ni, Zn, Cd, Hg and Pb decreased with increasing heavy metal concentration in the seawater, which may be evidence for an early protective mechanism, prior to damage, reduced growth or death of the organism.

Some elements showed a behaviour inferring that the uptake of these metals mainly depends on its concentration in seawater, which was indicated by strong positive correlations between the metal concentration in seawater and in the foraminiferal calcite. All three species showed a strong positive correlation between Pb and Ag in the water and their calcite. *A. aomoriensis* further revealed a slightly weaker correlation for Mn and Cu. *A. batava* holds a strong positive correlation for Mn and Hg. *E. excavatum* depicts a strong positive correlation for Cr and Ni. Other elements like Cd and Zn showed a more complicated behaviour indicating that factors other than seawater concentration are effecting the uptake of these metals, which is demonstrated by no clear correlation between seawater and calcite metal values. The reasons for this different behaviour are yet unclear. D_{TE} values of Ni, Zn, Cd, Hg and Pb decrease with increasing metal concentration in the seawater, which is most prominent in the highest metal treatment in all species. This could be due to an overload effect arising when the concentration of the metals is exceeding a certain threshold and could potentially be harmful or even lethal for the organism. This in turn could lead to a removal or a prevention from uptake of the metal (Nardelli et al., 2016). The results of this study facilitate the determination of variations in a reconstruction of the heavy metal concentration in seawater for those elements showing a correlation between TE/Ca ratios in calcite and seawater (*A. aomoriensis* = Mn, Cu, Ag, Pb; *A. batava* = Mn, Ag, Hg, Pb; *E. excavatum* = Cr, Ni, Ag, Pb). Such estimates can be based on foraminiferal samples from the fossil sediment cores record and recent surface sediments, and. This facilitates a monitoring of anthropogenic footprints on the environment today and in the past. Foraminifera offer the opportunity of long- and short-term monitoring of the heavy metal concentration because they are storing environmental signals over a period of time and not only at one point in time. The presented D_{TE}'s allow a direct quantification of metal concentrations in polluted and pristine areas. The foraminiferal species considered prevail in nearly all coastal environments worldwide, except polar latitudes. This in combination provides a powerful tool for monitoring the ecosystem status in various areas of interest.

Appendix

Appendix A: Additional Tables

1010 **Table A1**: TE/Ca$_{Seawater}$ values from single weeks during the culturing period of the metal system. Measurements

1011 were carried out with ICP-MS. ~~This values~~These values are the basis for the calculations of the mean TE/Ca values

1012 in Table 3 and for figure B1.

Metal System			Sampling date	Cr/Ca	Mn/Ca	Ni/Ca	Cu/Ca	Zn/Ca	Ag/Ca	Cd/Ca	Sn/Ca	Hg/Ca	Pb/Ca
	Phase	Day		µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	nmol mol^{-1}	µmol mol^{-1}
FR0 W2	0	10	10.2.20	12.80	818.54	7.60	27.75	100.19	0.16	0.44	3.20		0.63
FR0 W3	0	17	19.2.20	3.16	858.94	7.23	3.74	107.69	0.05	0.43	2.94	5.28	0.43
FR1 W1	1	2	27.2.20	13.59	862.52	7.08	6.25	97.45	0.37	1.00	4.98	43.07	1.03
FR1 W2	1	9	5.3.20	5.86	796.65	6.69	2.23	93.09	0.04	1.06	3.87	19.13	0.69
FR1 W3	1	13	9.3.20	7.03	819.38	6.86	2.14	95.50	0.06	1.08	4.23	27.17	0.62
FR1 W4	1	20	16.3.20	7.75	844.23	7.94	2.77	95.75	0.11	1.19	4.11	60.20	0.68
FR2 W1	2	2	19.3.20	13.68	825.59	10.02	4.15	129.09	1.88	5.20	5.37	933.50	5.70
FR2 W2	2	8	26.3.20	16.49	820.63	9.75	2.78	134.85	0.41	4.96	5.46	494.26	3.07
FR2 W3	2	15	2.4.20	13.31	811.64	9.44	2.23	132.12	0.31	4.89	5.10	287.70	2.50
FR2 W4	2	19	6.4.20	15.47	789.96	9.77	2.23	135.50	0.33	4.75	5.19	210.66	2.20
FR3 W1	3	2	9.4.20	52.74	1558.73	74.72	15.89	772.38	31.53	87.65	18.31	6123.75	125.25
FR3 W2	3	7	14.4.20	39.90	1281.58	46.73	3.67	455.31	7.95	61.37	11.84		70.27
FR3 W3	3	16	23.4.20	26.97	1469.59	66.07	3.55	579.52	4.13	84.82	5.87	2858.26	53.51
FR3 W4	3	20	27.4.20	25.59	1397.18	65.00	3.01	550.78	4.31	84.23	5.02	1640.01	45.72

1013

1014 **Table A2**: Average concentration, RSD (1σ in %), literature values, accuracy in comparison to literature values

1015 and number of measurements of the reference materials SLRS-6, SLEW-3, in-house reference materials (South

1016 Atlantic surface water and South Atlantic Gyre water) and NASS-6 measured with ICP-MS. Average

1017 concentration, RSD and accuracy values displayed here are averaged from single measuring days. Cr values are

1018 analysed after dilution of the samples and all other elements were analyses after preconcentration with a SeaFAST

1019 system. NRCC – National Research Council Canada. *Values originated from 1:10 dilution of SLRS-6.

Reference Materials	Cr	Mn	Ni	Cu	Zn	Cd	Pb
SLRS-6	nmol kg^{-1}	nmol kg^{-1}	nmol kg^{-1}	nmol kg^{-1}	nmol kg^{-1}	nmol kg^{-1}	nmol kg^{-1}
Average conc.	4732	52956	9811	338014*	31391*	62	786
RSD%	3.5	3.9	6.0	1.7*	7.2*	12.8	0.8
Yeghicheyan et al., 2019	4509	38616	10496	376378*	26920*	56	820
Accuracy	0.96	0.74	1.08	1.11*	0.86*	0.90	1.04
Number	4	11	11	13*	13*	7	7
SLEW-3							
Average conc.		40007	17508	22907	4442	343	
RSD%		4.3	3.5	4.2	9.1	4.8	
Leonhard et al., 2002		29326	20958	24409	3074	427	
Accuracy		0.74	1.21	1.07	0.78	1.28	
Number		12	12	12	12	12	
South Atlantic Gyre water							
Average conc.		1615	2189	2649	5614		
RSD%		6.2	3.7	5.3	13.2		

Number		10	10	10	10	
South Atlantic surface water						
Average conc.		1959	2417	2646	39718	
RSD%		6.8	2.8	5.8	2.2	
Number		6	6	6	6	
NASS-6						
Average conc.	6747	11162	3557	5206	5158	169
RSD%	15.9	5.2	3.2	3.0	25.3	7.0
NRCC	2293	9654	5129	3528	3931	165
Accuracy	0.34	0.87	0.76	0.35	0.81	0.98
Number	9	11	11	11	11	2

1020

1021 **Table A3**: Average concentration, RSD (1 σ in %), literature values, accuracy in comparison to literature values
1022 and number of measurements of the reference materials NIST SRM 610, NIST SRM 614, JCt-1, JCp-1, MACS-3
1023 and ECRM752-1 measured with LA-ICP-MS. Please note that for the ECRM752-1 no reported values for the
1024 elements of interest are available, which is also the case for some elements in other reference materials. It is
1025 important to note that the Hg/Ca values in the NIST glasses are not reliable as Hg is volatile and most likely
1026 volatilized during the glass formation. Average concentration, RSD and accuracy values displayed here are
1027 averaged from single measuring days.

Reference materials	Cr/Ca	Mn/Ca	Ni/Ca	Cu/Ca	Zn/Ca	Ag/Ca	Cd/Ca	Sn/Ca	Hg/Ca	Pb/Ca
NIST SRM 612	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	µmol mol^{-1}	mmol mol^{-1}
Mean value	0.33	0.34	0.33	0.29	0.37	0.08	0.12	0.15	2.26	0.10
RSD%	3.49	2.09	2.30	4.05	2.97	5.01	3.90	2.08	26.32	2.52
Jochum et al., 2011	0.33	0.33	0.31	0.28	0.28	0.10	0.12	0.15		0.09
Number of spots	69	75	75	75	73	73	75	75	41	75
NIST SRM 614	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	nmol mol^{-1}	µmol mol^{-1}
Mean value	19.28	10.31	8.43	15.86	67.58	2.13	15.53	5.97	20.93	5.23
RSD%	10.57	4.47	4.66	3.03	2.44	4.92	5.69	2.98	20.69	1.98
Jochum et al., 2011	10.78	12.18	8.83	10.16	20.11	1.83	2.35	6.67		5.28
Accuracy	0.57	1.19	1.06	0.64	0.30	0.86	0.23	1.12		1.01
Number of spots	35	38	37	39	38	38	38	39	19	39
MACS-3	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	mmol mol^{-1}	µmol mol^{-1}	mmol mol^{-1}
Mean value	0.21	0.97	0.093	0.17	0.13	0.065	0.041	0.042	5.11	0.026
RSD%	1.60	1.36	1.90	1.92	2.19	6.37	2.83	2.68	9.23	2.18
Jochum et al., 2019	0.23	0.99	0.10	0.19	0.20	0.054	0.051	0.049	5.41	0.031
Accuracy	1.13	1.02	1.09	1.11	1.50	0.84	1.24	1.15	1.07	1.16
Number of spots	45	45	44	46	46	42	46	46	44	46
JCt-1NP	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	µmol mol^{-1}	nmol mol^{-1}	µmol mol^{-1}
Mean value	6.16	0.91	0.37	1.14	1.46	0.01	1.60	2.30	8.93	0.063
RSD%	14.25	15.59	9.56	7.44	10.37	6.57	11.75	5.06	23.95	5.86
Jochum et al., 2019	0.93	1.01	1.03	1.48						0.064
Accuracy	0.15	1.19	2.71	1.31						1.04

Number of spots	44	38	45	47	45	11	46	13	26	48
JCp-1NP	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	nmol mol⁻¹	μmol mol⁻¹
Mean value	9.61	2.11	0.50	0.84	1.81	0.02	0.98	0.06	8.25	0.13
RSD%	7.91	4.62	6.89	6.36	6.53	11.34	11.08	10.68	20.96	6.15
Jochum et al., 2019	1.27	2.16	1.05	1.29	3.53					0.15
Accuracy	0.15	1.06	2.10	1.25	1.96					1.19
Number of spots	37	41	41	40	41	21	36	30	21	47
ECRM752-1	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	μmol mol⁻¹	nmol mol⁻¹	μmol mol⁻¹
Mean value	14.75	144.44	3.87	2.34	8.40	0.01	1.54	0.04	19.14	0.86
RSD%	7.78	2.54	4.97	6.21	2.37	87.11	7.76	9.22	18.03	3.82
Number of spots	27	31	26	28	27	15	29	24	19	31

1028

1029 **Table A4**: Comparison of the heavy metal concentrations in seawater of different regions of the world to values
1030 used for the culturing experiments in this study. It is indicated whether the values of this study are comparable to
1031 environmental values or if values from this study are higher or lower. EPA = Environmental Protection Agency,
1032 USA, FI = Field Injection, SF-ICP-MS = Sector Field Inductively Coupled Plasma Mass Spectrometry, GF =
1033 Graphite Atomic, (F)AAS = (Flame) Graphite Atomic Absorption Spectrometry, APDC-MIBK = Ammonium
1034 Pyrrolidine Dithiocarbamat-Methyl Isobutyle Ketone, ASV = Anodic Stripping Voltammetry, AES = Atomic
1035 Emission Spectrometry, CVAFS = Cold Vapor Atomic Fluorescence Spectrometry, FPD = Flame Photometric
1036 Detector.

Element	Study area	Concentration in μg l⁻¹	Comparable?	Reference	Pretreatment + measurement technique
Ag		0.06-4.61		**This study**	Dilution + ICP-MS
	EPA Recommended Values (acute)	1.9	yes	Prothro, 1993	
	Restronguet Creek, U.K. + Adriatic Sea	0.0025-0.03	yes	Barriada et al., 2007	FI preconc.+ SF-ICP-MS
	Ibaraki coast + Watarase river	0.014-0.03	yes	Shijo et al., 1989	Solvent extraction, Microscale backextraction + GFAAS
Cd		0.14-30.61		**This study**	SeaFAST preconc. + ICP-MS
	EPA Recommended Values (chronic)	7.9	yes	Prothro, 1993	-
	Suva, Fiji	150-250	no, low	Arikibe and Prasad, 2020	FAAS
	Black Sea in Rize, Turkey	1-3	yes	Baltas et al., 2017	ICP-MS
	Gulf of Chabahar, Oman Sea	0.15-0.19	yes	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	200-1580	no, low	Chakraborty et al., 2014	AAS
	East London + Port Elizabeth harbours, U.K.	200-72600	no, low	Fatoki and Mathabatha, 2001	APDC-MIBK procedure + AAS
	Yalujiang Estuary, China	0.83-1.33	yes	Li et al., 2017	ICP-MS
	Gulf San Jorge, Argentina	0.01-0.09	yes	Muse et al., 1999	APDC-MIBK procedure + AAS
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	34-560	yes	Reddy et al., 2005	APDC-MIBK procedure + FAAS
	Kamal estuary, Jakarta	0.01-0.02	no, high	Putri et al., 2012	AAS

	Location	Value	Exceeds	Reference	Method
	Jakarta Bay	0.04-0.104	yes	Williams et al., 2000	ASV
	Kepez harbor of Canakkale, Turkey	19-73800	yes	Yılmaz and Sadikoglu, 2011	Sample mineralization + ICP-AES
Cr		0.1-14.0		**This study**	Dilution + ICP-MS
	EPA Recommended Values (chronic)	50	no, low	Prothro, 1993	-
	Gulf of Chabahar, Oman Sea	20.16-21.46	yes	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	260-3010	no, low	Chakraborty et al., 2014	AAS
	Yalujiang Estuary, China	0.113-0.14	yes	Li et al., 2017	ICP-MS
	Gulf San Jorge, Argentina	0.04-0.5	yes	Muse et al., 1999	APDC-MIBK procedure + AAS
	Jakarta Bay	0.511-5.25	yes	Williams et al., 2000	ASV
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	35-765	no, low	Reddy et al., 2005	APDC-MIBK procedure + FAAS
Cu		0.6-6.2		**This study**	SeaFAST preconc. + ICP-MS
	EPA Recommended Values (chronic)	3.1	yes	Prothro, 1993	-
	Suva, Fiji	880-10290	no, low	Arikibe and Prasad, 2020	FAAS
	Black Sea in Rize, Turkey	30-242	no, low	Baltas et al., 2017	ICP-MS
	Gulf of Chabahar, Oman Sea	3.37-5.74	yes	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	1350-1850	no, low	Chakraborty et al., 2014	AAS
	East London + Port Elizabeth harbours, U.K.	500-42600	no, low	Fatoki and Mathabatha, 2001	APDC-MIBK procedure + AAS
	Yalujiang Estuary, China	1.8-4.7	yes	Li et al., 2017	ICP-MS
	Gulf San Jorge, Argentina	0.02-0.65	yes	Muse et al., 1999	APDC-MIBK procedure + AAS
	Jakarta Bay	0.405-4.04	yes	Williams et al., 2000	ASV
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	32-3939	yes	Reddy et al., 2005	APDC-MIBK procedure + FAAS
Hg		0.00035-0.273		**This study**	amalgamation + CVAFS
	EPA Recommended Values (chronic)	0.94	yes	Prothro, 1993	-
	South Florida Estuaries	0.0034-0.0074	yes	Kannan et al., 1998	amalgamation + CVAFS
	Guadalupe River and San Francisco Bay, California	0.0017-0.135	yes	Thomas et al., 2002	amalgamation + CVAFS
	Vembanad, India	0.0024-0.206	yes	Ramasamy et al., 2017	amalgamation + CVAFS
	Kamal estuary, Jakarta	0.1-0.2	yes	Putri et al., 2011	GFAAS
	Yalujiang Estuary, China	0.006-0.049	yes	Li et al., 2017	AFS
Mn		320-549		**This study**	SeaFAST preconc. + ICP-MS
	Black Sea in Rize, Turkey	3-14	yes	Baltas et al., 2017	ICP-MS
	Gulf of Chabahar, Oman Sea	15.43-24.76	no, high	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	13000-18000	no, low	Chakraborty et al., 2014	AAS
	East London + Port Elizabeth harbours, U.K.	300-23900	yes	Fatoki and Mathabatha, 2001	APDC-MIBK procedure + AAS

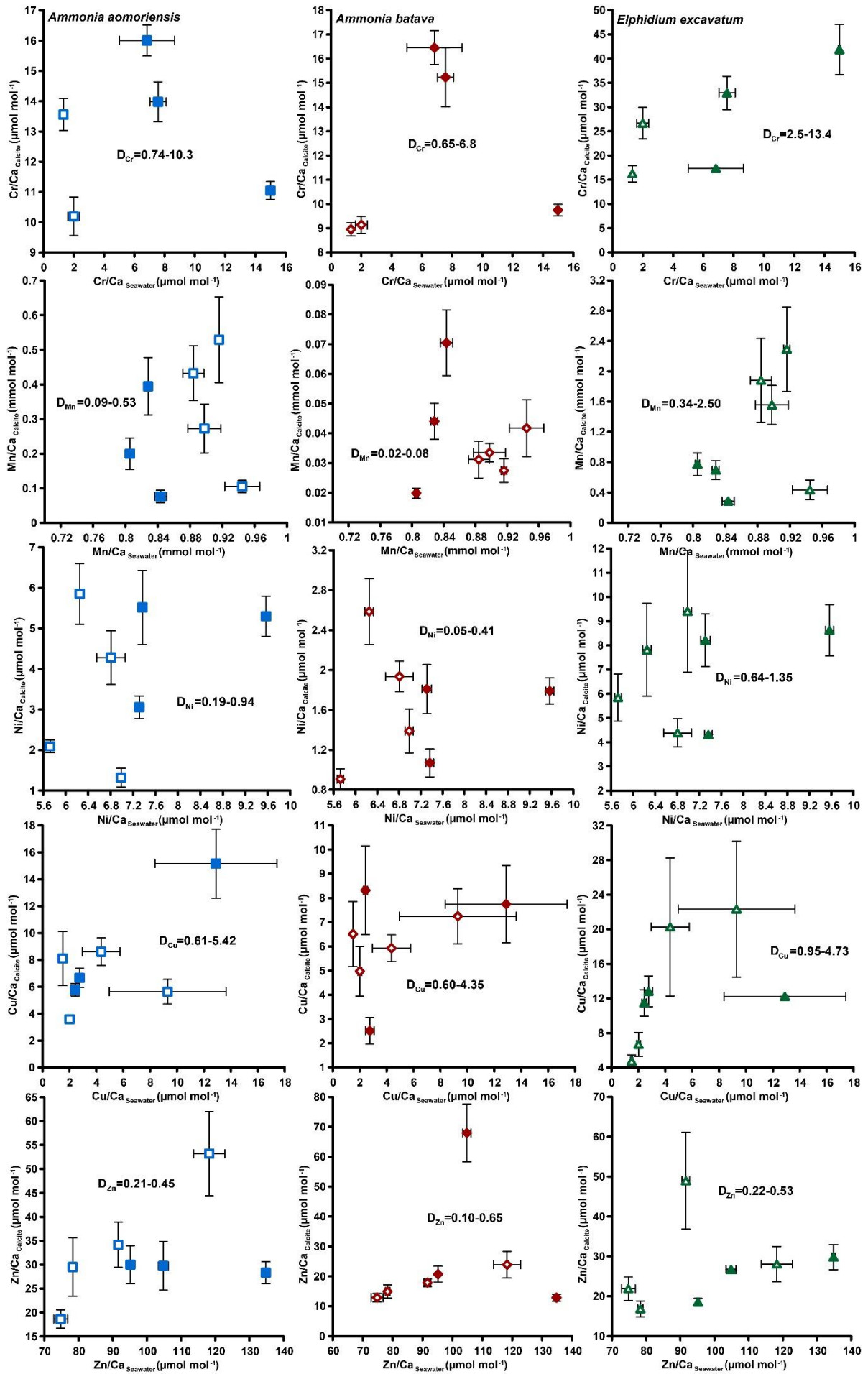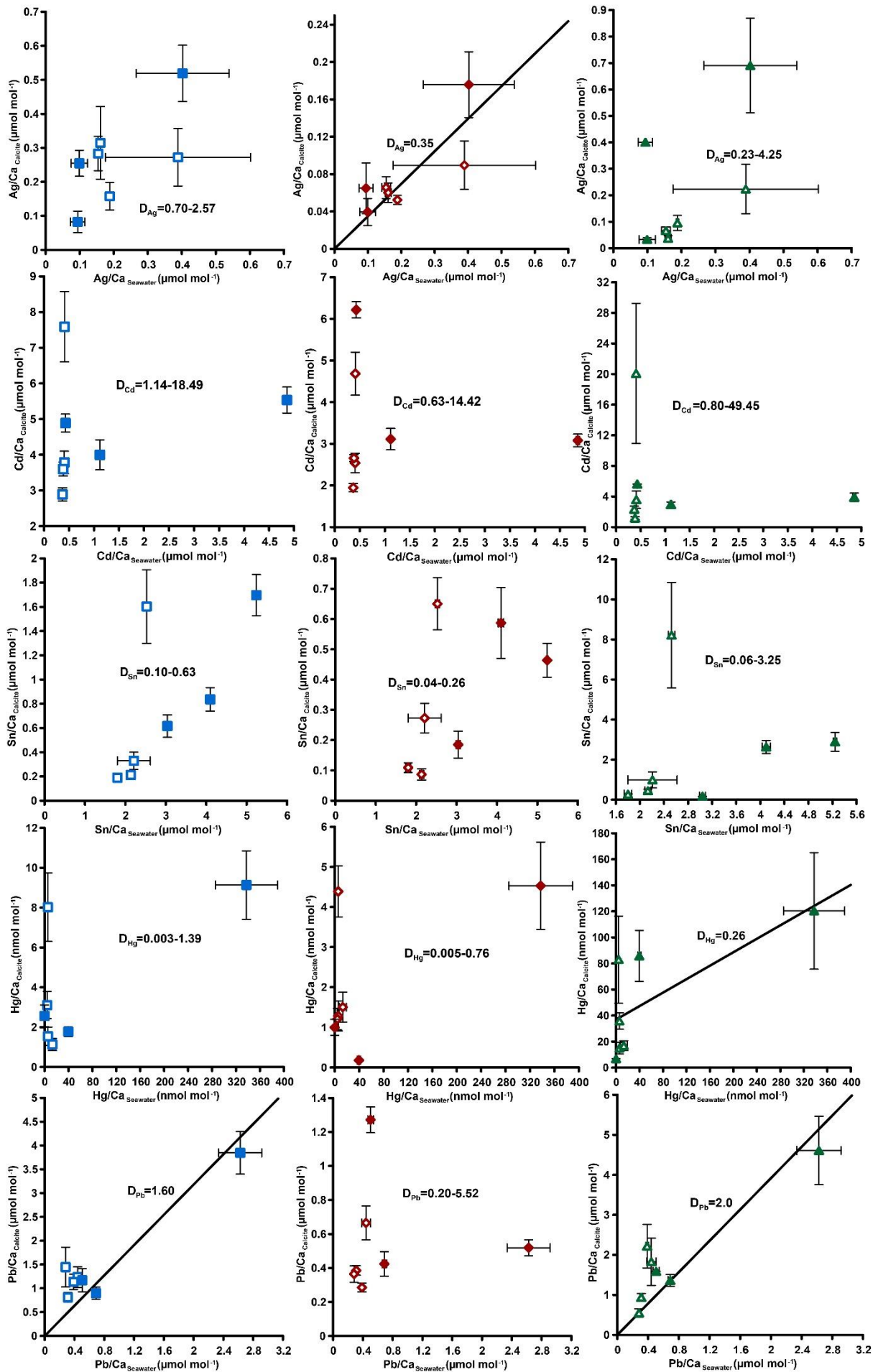|				
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	31-4920	yes	Reddy et al., 2005	APDC-MIBK procedure + FAAS
Ni		2.3-24.3		**This study**	SeaFAST preconc. + ICP-MS
	EPA Recommended Values (chronic)	8.2	yes	Prothro, 1993	-
	Suva, Fiji	230-800	no, low	Arikibe and Prasad, 2020	FAAS
	Black Sea in Rize, Turkey	0.006-0.036	yes	Baltas et al., 2017	ICP-MS
	Gulf of Chabahar, Oman Sea	16.42-17.14	yes	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	190-330	no, low	Chakraborty et al., 2014	AAS
	Jakarta Bay	0.058-5.25	yes	Williams et al., 2000	ASV
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	32-944	yes	Reddy et al., 2005	APDC-MIBK procedure + FAAS
Pb		0.11-28.35		**This study**	SeaFAST preconc. + ICP-MS
	EPA Recommended Values (chronic)	5.6	yes	Prothro, 1993	-
	Suva, Fiji	880-1770	no, low	Arikibe and Prasad, 2020	FAAS
	Black Sea in Rize, Turkey	6-130	yes	Baltas et al., 2017	ICP-MS
	Gulf of Chabahar, Oman Sea	4.24-4.25	yes	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	20-120	yes	Chakraborty et al., 2014	AAS
	East London + Port Elizabeth harbours, U.K.	600-16300	no, low	Fatoki and Mathabatha, 2001	APDC-MIBK procedure + AAS
	Yalujiang Estuary, China	0.4-1.8	yes	Li et al., 2017	ICP-MS
	Gulf San Jorge, Argentina	0.1-0.5	yes	Muse et al., 1999	APDC-MIBK procedure + AAS
	Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	30-2036	yes	Reddy et al., 2005	APDC-MIBK procedure + FAAS
	Kamal estuary, Jakarta	1.3-4	yes	Putri et al., 2011	AAS
	Jakarta Bay	0.485-3.62	yes	Williams et al., 2000	ASV
	Kepez harbor of Canakkale, Turkey	49-9390	yes	Yılmaz and Sadikoglu, 2011	sample mineralization + ICP-AES
Sn		0.86-3.95		**This study**	Dilution + ICP-MS
	estuarine seawater, Galicia Coast, Spain	0.53-1.23	yes	Bermejo-Barrera et al., 1999	hydride generation + AAS
	U.S. and European rivers	0.0001-0.1	yes	Byrd and Andreae, 1982	hybride generation + FPD
Zn		30.0-226.9		**This study**	SeaFAST preconc. + ICP-MS
	EPA Recommended Values (chronic)	81	yes	Prothro, 1993	-
	Suva, Fiji	80-1450	yes	Arikibe and Prasad, 2020	FAAS
	Black Sea in Rize, Turkey	38-178	yes	Baltas et al., 2017	ICP-MS
	Gulf of Chabahar, Oman Sea	18.01-22.62	yes	Bazzi, 2014	APDC-MIBK procedure + FAAS
	Gulf of Kutch, Arabian Sea	11000-31000	no, low	Chakraborty et al., 2014	AAS
	East London + Port Elizabeth harbours, U.K.	500-27600	yes	Fatoki and Mathabatha, 2001	APDC-MIBK procedure + AAS
	Yalujiang Estuary, China	9.2-19.6	yes	Li et al., 2017	ICP-MS
	Gulf San Jorge, Argentina	0.01-0.55	no, high	Muse et al., 1999	APDC-MIBK procedure + AAS

Jakarta Bay	2-30.1	yes	Williams et al., 2000	ASV
Alang–Sosiya ship scrapping yard, Gulf of Cambay, India	33-5832	yes	Reddy et al., 2005	APDC-MIBK procedure + FAAS

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1039 **Appendix B: Additional Figures**

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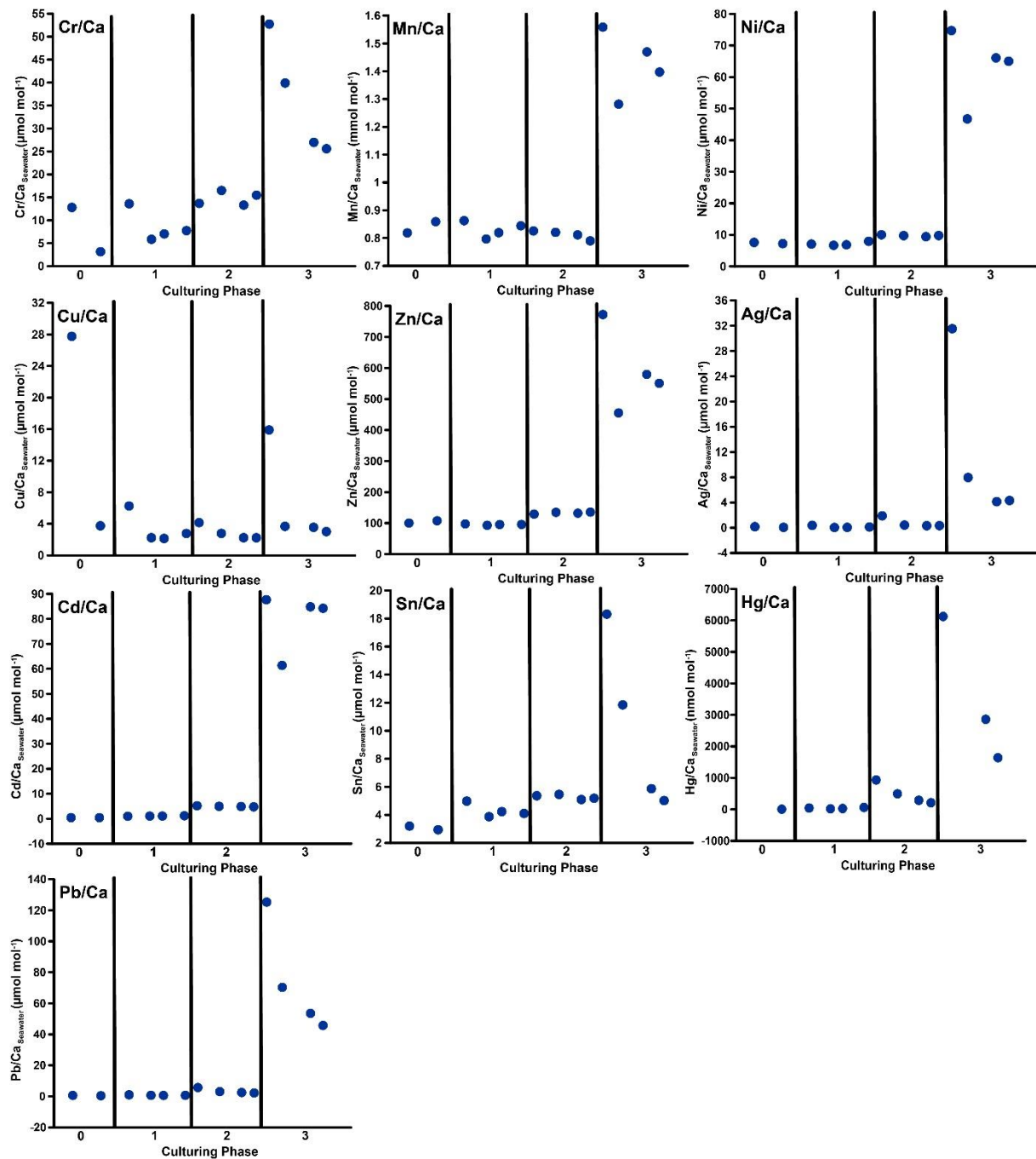
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1042 **Figure B1**: TE/Ca values in the culturing medium of the metal system in µmol mol^{-1} or nmol mol^{-1} divided by

1043 individual culturing phases. In this system, phase 0 is the control phase without any extra added metals and for

1044 phase 1 to 3, the heavy metal concentration in the culturing medium was elevated. The data the figure is based on

1045 can be found in Table A1.

48

	Mean TE/Ca Control System - *A. aomoriensis*
	Mean TE/Ca Metal System - *A. aomoriensis*
	Mean TE/Ca Control System - *A. batava*
	Mean TE/Ca Metal System - *A. batava*
	Mean TE/Ca Control System - *E. excavatum*
	Mean TE/Ca Metal System - *E. excavatum*
	Regression line

Figure B2: Mean TE/Ca values in the foraminiferal calcite versus the mean TE/Ca values in the corresponding culturing medium without phase 3. Each data point represents the mean value of all laser ablation ICP – MS measurements on single foraminiferal chambers built up during the individual culturing phase plotted against the mean metal concentrations in the seawater averaged over the culturing phase (Table 3). Error bars symbolize the standard error of the mean. The linear regression line is based on the calculations excluding phase 3 and is only displayed when elements showed a significant correlation between seawater and calcite. D_{TE}'s of *E. excavatum* where considered without values for Phase 0 as only data from one newly formed chamber are available. All values can be found in Table 4.

Supplements

Table S1–S3: TE/Ca$_{Calcite}$ values from *Ammonia aomoriensis* (Table S1)*, Ammonia batava* (Table S2) and *Elphidium excavatum* (Table S3). Values represent single laser ablation spots on foraminiferal chambers that were formed during the individual culturing period in the control and the metal system. Only values above the detection limits of the individual element are presented. Furthermore, outliers are also excluded. These values are the basis for the calculation of the mean TE/Ca values in Table 4 and Fig. 4. The sample ID indicates the species (AA = *A. aomoriensis*, AB = *A. batava*, E = *E. excavatum*), the culturing phase, the system (R = metal system, L = control system), the individual and the chamber that was ablated, starting from the innermost chamber going to the youngest one.

Data availability

All data generated or analysed during this study are included in this published article and its supplementary information files.

Author contribution

This study was initiated by JS and EH. SS collected the samples, cultured the foraminifera, processed the samples in the laboratory and acquired, analysed and interpreted the water and foraminiferal data. JS helped with the sampling logistics, design and implementation of the culturing experiments. EH advised and helped with the processing and analysis of the water samples and EH and DGS advised and helped with the measurements of the foraminiferal samples. SS wrote the manuscript with all the authors contributing to the discussion and data interpretation, and editing of the work.

Competing interests

The authors declare that they have no conflict of interest.

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