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Organochlorines and Polycyclic Aromatic Hydrocarbons as fingerprint of exposure pathways from marine sediments to biota

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Abstract

To elucidate the dynamics of a suite of organochlorine contaminants (PCBs, HCB), PAHs and Hg and verify the potential of these pollutants as reliable fingerprints of sources, an ensemble of marine sediments and organisms (finfish, shellfish species and *Mytilus galloprovincialis*) were analysed from the contaminated Augusta Bay (Southern Italy). The Hg and HCB concentration in the sediments exceeded the EQS of the Directive 2000/60/EU. Similarly, ∑PCB and selected PAHs were above the threshold limit set by regulation. The marine organisms showed Hg concentrations above CE 1881/2006. Contaminants in transplanted mussel evidenced an increased accumulation overtime and different distribution patterns between sampling sites. Analysis of the homolog composition of PCB congeners revealed comparable patterns between sediments and marine organisms and offered the opportunity to define a robust fingerprint for tracing contaminants transfer from the abiotic to the biotic compartments. These results were confirmed by the Fluoranthene/Pyrene, Hg and HCB distribution modes.

Keywords: Organochlorines, PAHs, Biota, Sediment, Source fingerprint.

1. Introduction

Understanding the transfer of contaminants from marine sediments to biota represents a critical aspect for environmental risk assessment, modelling and specific remediation actions (Gobas and Arnot, 2010). Distribution patterns of pollutants, isotope measurements, compositional analysis and investigation of variable proportions of single congeners of specific organic pollutants were shown as valuable tools for tracing sources of contamination to the trophic chains (e.g., Sahu et al., 2009; Baskaran, 2011; Rodenburg and Leidos, 2017; Habibullah-Al-Mamun et al., 2019; Bonsignore et al., 2020).

Although several actions have been taken to reduce or even eliminate input of contaminants in the environment (UNEP 2001, 2017, 2019), compounds such as Persistent Organic Pollutants (POPs) and some trace metals, including Hg, are accumulated in different environmental matrices thus representing a global environmental threat for marine organisms and human health (García-Flor et al., 2009; Bellas et al., 2011; Sanchez-Avila et al., 2012; Berrojalbiz et al., 2014; Olenycz et al., 2015; Batang et al., 2016; Jepson et al., 2016; Beiras, 2018). Due to their hydrophobic behaviour in aquatic ecosystems, most of the pollutants such as e.g. Polychlorinated biphenyls -[PCBs], Hexachlorobenzene [HCB] and Polycyclic Aromatic Hydrocarbons [PAHs], readily bind to particles (suspended in seawater and/or into the sediments) and are strongly associated with organic phase. Once in the sediments they persist for very long time (Minh et al., 2007) with high potential to be incorporated into the food web via benthic–pelagic coupling (Thomann et al., 1992). As an example of extremely stable compounds, Hexachlorobenzene (HCB) was produced for industrial and agricultural applications worldwide (Courtney, 1979; Bailey, 2001; Meijer et al., 2003), and although its production was stopped before 2000 in most countries, it is still abundant in the environment due to its long-half life in water and sediments (Barber et al., 2005). PCBs were produced in the past century, up to the 1970s-1980s, and largely used for different industrial applications (e.g. as dielectric fluid in electrical transformers and capacitors, active constituent of pesticides) (ATSDR, 2000; Crinnion, 2011). Among 209 congeners, the Stockholm Convention reports as mandatory to measure at least six indicator congeners non-dioxin-like (NDL) PCBs 28, 52, 101, 138, 153 and 180 to estimate environmental PCBs contamination (JECFA, 2018), while ICES (Webster et al., 2013) recommends the measure of 7 PCBs congeners (including also congener 118) which represent a large percentage of the total congeners measured in environmental samples and human fluids (EFSA, 2010; IARC, 2016). Likewise, Hg is a trace metal efficiently transferred and biomagnified along food web reaching high concentrations in the upper trophic levels and thus representing a risk to human health (e.g., Signa et al., 2017).

Finally, PAHs represent another group of chemicals of priority concern which include the largest known class of carcinogens and chemical mutagens (Keith and Telliard, 1979). PAHs tend to adsorb rapidly on particles (Neff, 1979; Landrum and Robbins, 1990) and since their solubility decreases with increasing molecular weight, the bioaccumulation of these chemicals from sediments to marine organisms is generally higher for those with lower molecular weight (Porte and Albaigés, 1993; Djomo et al., 1996).

Over the last decade, a number of scientific contributions explored the distribution patterns of classes of contaminants (e.g., PAHs, PCBs, PCDD/Fs, Hg and HCB) to trace transfer dynamic from the sediments to the biotic compartment, and to characterize sources of variability in contaminant bioavailability to aquatic biota (e.g. Selck et al., 2012; McLeod et al., 2015).

Fish and other key species have been used as bioindicators to investigate the presence and toxic effects of chemical pollutants (Ueno et al., 2003). It is well-known that biotransformation pathways significantly modulate absorption, distribution and excretion of organic xenobiotics in fish while filter feeders, such as bivalve, tend to accumulate these pollutants in their tissues from both water column and sediments thus providing additional information on the environmental state of pollution (Regoli et al., 1998; Wiberg et al., 2002; Olenycz et al., 2015; Farrington et al., 2016; Beyer et al., 2017).

However, PCBs congeners distribution in the biota generally remains similar to the un-weathered PCBs mixtures in the sediments (mainly when those with higher chlorine content are considered), thus allowing the identification of these sources with a satisfactory degree of confidence. Some exercises of PCBs fingerprinting in biota have been performed in the Hanford Site in Washington State (Rodenburg et al., 2015) and Portland Harbor in Oregon (Rodenburg et al., 2019) offering robust information on the transfer of contaminants from the abiotic to the biotic compartments.

In this work, we present an unprecedented dataset of Hg, HCB, PCBs and 16 priority PAHs measured on surface sediments, mussels, and several species of marine organisms from the highly contaminated marine area of Augusta Bay (southern Italy). Patterns of PCBs congeners, combined

to spatial distribution of PAHs, HCB and Hg in sediments and biota were elaborated to evaluate the fingerprint association between sediments (sources) and biota, and their opportunity for high-resolution tracing of priority contaminants sources.

2. Study area

Augusta Bay is a ~ 25 km^2 area located in the eastern Sicilian coast (Fig. 1) which hosts one of the most important harbours of the Mediterranean Sea and, since 1950s, one of the largest petrochemical complexes in Europe, characterized by oil refineries and chlor-alkali plant. Uncontrolled industrial discharges led to significant contamination of sediments by metals and organics, mainly Hg, PCBs, PAHs and HCB (ICRAM, 2008; Bellucci et al., 2012; Croudace et al., 2015), and all the area was included in the National Remediation Plan by the Italian Ministry of Environment. Several investigations have highlighted a strong contamination of the marine environment with i) abnormal levels of Hg, PCBs and HCB in sediments (ICRAM, 2005, 2008, Bellucci et al., 2012; Sprovieri et al., 2011; Orecchio and Polizzotto, 2013), ii) active fluxes of Hg from the sediment to the water column (Salvagio Manta et al., 2016; Denaro et al., 2020) and iii) Hg evasion processes to the atmosphere (Sprovieri et al., 2011; Bagnato et al., 2013). Serious impacts on the ecosystem have been documented by ecotoxicological investigations which revealed genotoxic damages and high Hg contents, exceeding the regulatory limits for food consumption in the tissues of fish and mussels (Ausili et al., 2008; ENVIRON, 2008; ICRAM, 2008; Tomasello et al., 2012; Bonsignore et al., 2013; Signa et al., 2017), thus advising for possible human health implications due to consumption of seafood from this area (Ausili et al., 2008; Bonsignore et al., 2013; Di Bella et al., 2020).

The role of polluted sediments as the main carrier of Hg to the ecosystem and local fish consumers has been further revealed throught the exploration of Hg isotopes signature in sediments and fish (Bonsignore et al., 2015).

3. Sampling activity

3.1 Sediments

The sediment sampling, carried out in October 2017 during an oceanographic cruise on board R/V "*Luigi Sanzo",* provided sediment cores from 4 stations (A3, A7, A9, A11) inside the bay, from the northern to the southern area (Fig. 1), using an oceanic box-corer. Sediment cores were immediately stored at −20 °C until the analyses.

3.2 Marine organisms

In November 2013, wild mussels (*Mytilus galloprovincialis*) with size range 3.6-6 cm, were manually collected from industrial wharf and rapidly transported to laboratory for the tissue's dissection. In the same period, a total of 62 specimens of finfish (*Sphyraena sphyraena, Mullus barbatus, Pagellus* spp.*, Sparus aurata, Serranus cabrilla, Diplodus* spp.) were obtained through local fisherman (Fig. 1).

In September 2017, mussels (*Mytilus galloprovincialis*) with size range of 4-6 cm were obtained from a commercial farm. A pool of individuals (about 2 kg) was used as control (*M. galloprovincialis CTRL*)*,* while other two pools (about 10 kg) were transplanted in the northern (*M. galloprovincialis N*) and the southern (*M. galloprovincialis S*) areas of Augusta Bay, caged in nylon net bags, secured to a rope and maintained at approximately 3 m water depth. After a period of 5 weeks, a subgroup of mussels was collected from each site (*N1* and *S1*), and an additional sampling was performed 7 months later (*N2* and *S2*).

In October 2017, a total of 96 specimens of finfish (*Sphyraena sphyraena, Trigla lucerna, Mullus barbatus, Pagellus spp., Diplodus spp.*) and shellfish (*Parapaeneus kerathurus* and *Sepia* spp.) were obtained through local fishermen (Fig. 1).

4. Analytical methods

4.1 Samples preparation

In laboratory, the sediment cores were defrosted, extruded and immediately sliced into 1 cm intervals. The samples were then dried at 35°C and homogenized by an agate mortar. For the goals of this study only the most superficial levels (between 0-10 cm) were analysed.

The total length (mm) and the weight (g) for each organism were measured and muscles and soft tissues were dissected by stainless steel scissors. Individuals of each species with comparable size were pooled, homogenized and stored at -80°C (Table 1). The tissues samples were then freeze-dried and powdered by an agate mortar prior the analyses.

4.2 Mercury determination in sediments and marine organisms

The concentration of Hg in organisms and sediments was determined by a direct mercury analyzer (milestone-DMA-80 atomic absorption spectrophotometer), according to analytical procedures reported in EPA 7473 method (2007). About 0.050 g of dried tissue and ~0.010 g of dry sediment were loaded in nickel boats and transferred into the DMA-80 system. The Certified Reference Materials-TORT-2 Lobster hepatopancreas and (PACS-2 Marine sediment, NRC-CNRC) were used to assess accuracy (estimated % error =3%) and precision (routinely better than 4%; RSD%, n = 3). About 20% of the total number of samples was duplicated to estimate reproducibility (better than 7%). Acid-cleaned laboratory materials were used in order to minimize contamination risks during sample preparation and analyses procedures. Analyses were performed at the biogeochemical laboratory of the Institute of Anthropic impacts and Sustainability in marine environment (IAS-CNR) of Capo Granitola (Trapani, Italy). Results were expressed as on wet weight basis.

Results relative to tissues were converted from dried to wet-weight (μg g^{-1}) applying a conversion factor previously calculated using the following formula: $C_w=C_d$ x (100-%H_2O/100) were C_d and C_w are the concentration expressed relatively to dry and wet mass respectively. %H_2O is the percentage of humidity (ranging around 80% for almost species) calculated after the freeze-drying process (Di Bella et al., 2020 and references therein).

171

172 *4.3 PCBs, HCB and PAHs determination in sediments and marine organisms*

173 The sediment and biota samples were freeze dried and homogenized, then an aliquot (about 1 and 2
174 g of tissues and sediments, respectively) was placed in an ASE 200 steel extraction cell, spiked with
175 surrogate standards (o2si smart solution ® custom deuterated PAH mix; Wellington Laboratories
176 Inc PCB mix; CDN Isotope Inc HCB standard solution) for recovery monitoring and extracted by
177 pressurized fluid extraction using a moderately polar solvent mixture. The resulting extract was
178 cleaned up according to the matrix (sediment or biota) and the analytes (organochlorines or PAHs)
179 by a combination of column chromatography on silica (PAH) and florisil (organochlorine)
180 adsorbents and a shaking of the extract with a NaOH solution (PAH in biota) or with concentrated
181 sulphuric acid (organochlorine). For the determination of organochlorine compounds in sediment
182 samples the extract was also shaken with activated copper powder for sulphur removal. The cleaned
183 up concentrated extract was spiked with injection internal standard and analysed by GC/MS/MS for
184 organochlorine and GC/MS for PAHs.

185 PAH contents in sediments were compared to values established by the Italian regulation
186 (Legislative Decree 172/2015). Benzo(a)pyrene and Fluorantene were monitored in biota,
187 specifically in molluscs as suggested by Legislative Decree 172/2015.

188 Reference materials for sediments (SRM 1941b-NIST, organics in marine sediments) and biota
189 (NIST 2974a, organics in freeze-dried mussel's tissue - *Mytilus edulis*) were analysed to estimate
190 the accuracy (recoveries for each analyte of PAHs, PCBs and HCB ranged between 94% and
191 107%). The reproducibility, estimated on tripled samples, was better than ± 10% and analytical
192 precision was routinely better than 4% (RSD%, n = 3).

193 For the PCB class of contaminants, we referred to the following notation: ∑PCBs (sum of all the
194 measured congeners), ∑PCBs[1] (sum of PCB congeners according to Italian Legislative Decree
195 172/2015 for marine sediments), ∑NDL-PCBs (PCB 28, 52, 101, 153, 138 and 180) and ∑7PCBs
196 (∑6PCBs + PCB 118).

4.4 Statistical analysis

All the statistical analyses and graphics were performed using the statistical software R 3.6.3 (R Development Core Team, 2020). Box-whisker plots of measured Hg, HCB and PCBs were used to provide synthetic and direct comparisons for these parameters among the cores. For statistical analysis, all values below the limit of detection (LOD) were set-up as ½ LOD.

A hierarchical cluster analysis (HCA) with Euclidean distance and Ward's grouping method was applied to the ratio 'individual PCB congener/ΣPCB' in order to create homogeneous clusters of samples (sediments, fishes and mussels) based on similarity of PCBs composition. The elbow method was used to determine the most statistically reliable and representative number of clusters, minimising the total intra-cluster variation.

5. Results and Discussion

5.1 Contaminants in sediments

5.1.1. Mercury

Mercury concentrations measured in the A3, A7, A9 and A11 cores ranged from 3.07 to 12.2 μg g^{-1} (Tab. 2; Tab. S1) widely exceeding (generally more than one order of magnitude) the Environmental Quality Standard (EQS), defined in accordance with Directive 2000/60/EU as criteria for the achieving of the Good Chemical and Ecological Status of water bodies, and established by the Italian regulation (Legislative Decree 172/2015; 0.3 mg kg^{-1}). The highest values (mean±1σ = 11.4±0.57 μg g^{-1}) were found in A9 core collected from the south-western sector of the bay, in front of the past chlor-alkali plant (Tab. 2; Fig 2a). The lowest concentration (mean±1σ = 3.64±0.27 μg g^{-1}) was recorded in the A3 core sampled in the northern area (Tab. 2, Fig. 2a). Finally, A7 and A11 cores, collected from the central and southern part of the bay, showed comparable levels of Hg (mean±1σ = 6.71±0.80 μg g^{-1} and 6.54±1.28 μg g^{-1}, respectively).

223

224 *5.1.2. Organochlorine compounds (HCB and PCBs) and PAHs*

225 The analysis of organochlorine compounds in sediments showed very high concentrations of HCB
226 in all the analysed cores, ranging from 1.98 to 330 ng g^{-1} (Tab. 2; Tab. S1). The mean values
227 registered from each core, again considering only the superficial 10 cm of sediments, were
228 considerably above the EQS for marine sediments (0.4 ng g^{-1}), with highest values measured in the
229 A9 core (mean±1σ = 172 ± 87.6 ng g^{-1}) (Tab. 2; Fig. 2b).

230 Also ∑PCBs and ∑7PCB registered higher values in A9 core (41.9 ± 5.60 ng g^{-1}; Tab. 2; Tab. S1;
231 Fig. 2c,d). The ∑PCBs also exceeded EQS value (8 ng g^{-1}) for sediments in all cores (Tab. 2).

232 The average concentrations of the individual PAHs were, in some cases, above the EQS indicated in
233 Tab 1/A of the Legislative Decree 172/2015 Specifically, the Anthracene concentrations (range =
234 23.4-286 ng g^{-1}) were above the EQS (24 ng g^{-1}) in all the analyzed samples; most of the A3 levels
235 showed concentrations of Benzo-a-Pyrene (mean±1σ = 64.7 ± 20.1 ng g^{-1}; EQS = 30 ng g^{-1}),
236 Benzo-b-Fluoranthene (mean±1σ = 54.6 ± 20.4 ng g^{-1}; EQS = 40 ng g^{-1}) and Benzo-k-Fluoranthene
237 (mean±1σ = 33.6 ± 24.3 ng g^{-1}; EQS = 20 ng g^{-1}) above the EQS values; furthermore, Indenopyrene
238 concentrations higher than EQS (70 ng g^{-1}) were recorded in the upper part (between 0-6 cm) of the
239 A3 core (Tab. 2; Tab. S2).

240

241 *5.2 Contaminants in marine organisms*

242 *5.2.1. Mercury*

243 The lowest Hg mean values referred to *Sepia* spp. collected in 2017 (0.21±0.08 μg g^{-1}) and *S.*
244 *aurata* sampled in 2013 (0.32±0.09 μg g^{-1}). In most cases, such as *S. sphyraena*, *T. lucerna*, *M.*
245 *barbatus*, *Diplodus* spp. and *P. kerathurus* (sampled in 2017) and *S. sphyraena, S. cabrilla* and
246 *Diplodus* spp. (sampled in 2013), the Hg levels exceed the threshold limits set by EU regulation for
247 contaminants in seafood (CE 1881/2006) (Tab. 3). Excluding the effect of age and length, the
248 contents of Hg found in organisms resulted in most cases higher than values previously measured in

249 the study area. Specifically, the concentrations measured in *Diplodus* spp. (1.49±1.77 μg g^{-1} in
250 2017) and *Pagellus* spp. (0.86±0.44 μg g^{-1} in 2017 and 0.53±0.12 μg g^{-1} in 2013) were higher than
251 those reported by Bonsignore et al. (2013), Ausili et al. (2003) and ICRAM (2005, 2008), for the
252 same species (between 0.56-0.90 μg g^{-1} and 0.36-0.41 μg g^{-1}, respectively). In red mullet (*M.*
253 *barbatus*), the concentrations relative to 2013 (0.44±0.20 μg g^{-1}) were in the same range of
254 measurements (0.46-0.82 μg g^{-1}) of the previous investigations (Bonsignore at al., 2013; ICRAM,
255 2005, 2008) while those relative to 2017 (2.11 and 1.71 μg g^{-1}) resulted significantly higher and in
256 the same range of those reported by ICRAM in 2003 (1.49-1.92 μg g^{-1}). Finally, the Hg
257 concentration in *T. lucerna* (0.66 μg g^{-1}) resulted slightly higher than that reported by ICRAM in
258 2003 (0.57 μg g^{-1}).
259 The bioavailability of Hg in the study area has been confirmed by analyses on both native and
260 caged mussels with concentrations always higher than the EQS (0.02 μg g^{-1}) (Tab.3). Wild
261 specimens collected in industrial wharf exhibited concentration of approximately about 0.2 μg g^{-1},
262 ~20-fold higher than those measured in control organisms and comparable to those observed by
263 Ausili et al. (2008) (Tab. 3). Bioaccumulation of Hg demonstrated to be very rapid in caged mussels
264 showing, after only 5 weeks, comparable values to those of native organisms; after 7 months the
265 concentrations increased by about 5-fold (Tab. 3). In particular, in the northern area, Hg in mussels
266 5 weeks after transplant (N1: 0.02±0.002 μg g^{-1}) appeared double than in the control sample (0.01
267 μg g^{-1}), while after 7 months, the Hg increase was nearly one order of magnitude higher (N2:
268 0.12±0.01 μg g^{-1}) (Tab. 3). Worthy to note, this value is reasonably in agreement with the results
269 recently reported for mussels in the Augusta Bay (0.17±0.07 μg g^{-1}; Caricato et al., 2019). In the
270 southern area, when comparing the concentration in pre-deployed mussels, the levels of Hg were
271 ~23 times higher in those collected after 5 weeks (S1: 0.23±0.02 μg g^{-1}) and up to 89 times higher
272 after 7 months (S2: 0.89 μg g^{-1}). The latter value exceeds the threshold limits set up by EU
273 regulation for contaminants in seafood (0.5 μg g^{-1}; Reg. CE 1881/2006) and appeared much higher
274 than those reported for many other Mediterranean sites (range: 0.01-0.20 μg g^{-1}; Amodio-Cocchieri

et al., 2003; Ipolyi et al., 2004; Licata et al., 2004; Cardellicchio et al., 2010; Kljaković-Gašpić et al., 2010; Spada et al., 2011, 2012, 2013; Caricato et al., 2019).

5.2.2. HCB, PCBs and PAHs

Concentrations of HCB, PCBs and PAHs measured in organisms are reported in Table 3. Values of HCB were <LOD (0.05 ng g^{-1}) in all species of finfish and shellfish sampled in 2017, except for *S. sphyraena* (1.40 ng g^{-1}) and *M. barbatus* (0.24 and 0.65 ng g^{-1}); on the other hand, in 2013 they ranged between 0.09 ±0.02 (*Pagellus* spp.) and 1.36±0.31 ng g^{-1} (*Mullus* spp.). All the concentrations are below the EQS of 10 ng g^{-1}.

HCB together with PCBs are ubiquitous contaminants in Mediterranean marine coastal areas (Solé et al., 2000). HCB can be stored in sediments and accumulated in benthic organisms. Being a lipophilic compound, it has a greater affinity for tissues with high lipid content, thus explaining the low concentrations measured in almost all the muscle samples of fish analysed in this study. Other authors have reported data related to HCB in fishes, but a direct comparison appears problematic due to the high variability of investigated organs/tissues (skin, gills or mussels' tissues) and the different used reference system (wet, dry or lipid weight) (Domingo and Bocio, 2007). Nonetheless, when comparable, our results are within the range of concentration reported from other studies for the Mediterranean Sea (Rodríguez-Hernández et al., 2016; Junquè et al., 2018).

PCBs were detected in all the analysed species. The mean values of ∑PCBs ranged from 28.7 ng g^{-1} (*Diplodus* spp.) to 241.7 ng g^{-1} (*S. sphyraena*). The ∑NDL-PCBs showed variable concentrations among the analysed species, with a mean value above the limit of 75 ng g^{-1} (European Commission, 2011) measured in *S. sphyraena* (168 ng g^{-1}) and *M. barbatus* (88.2 ng g^{-1}) collected in 2017 and *Mullus spp*. sampled in 2013 (96.2 ng g^{-1}). All the analysed species sampled in 2013 and 2017, exceeded ∑7PCBs threshold limit (10 ng g^{-1}) with the lowest and highest value measured in *T. lucerna* (19.0 ng g^{-1}) and *S. sphyraena* (180.9 ng g^{-1}), respectively (Tab. 3); this value represents the

300 limit above which effects on marine organisms might be expected according to the Ecotoxicological
301 Assessment Criteria (EACs) of the OSPAR Convention (Campillo et al., 2017). A direct
302 comparison of the ∑7PCBs with a previous study (Ferrante et al., 2007) on various edible species
303 sampled in the Gulf of Naples (Southern Tyrrhenian Sea) showed systematically higher
304 concentrations in samples from Augusta Bay.
305 The highly-chlorinated congeners CB153 and CB138 (hexa-chlorinated) and CB180 (epta-
306 chlorinated) exhibited the higher abundance in biota (Tab. 3) and dominated the PCB group in all
307 the analysed samples, thus mirroring results from other studies (Miao et al., 2000; Solé et al., 2000;
308 Green and Knutzen, 2003; Castro-Jimenez et al., 2008; Storelli et al., 2009; Scarpato et al., 2010;
309 Xia et al., 2012; Suarez et al., 2013; Herceg-Romanic et al., 2014; Mohebbi- Nozar et al., 2014;
310 Kampire et al., 2015; Batang et al., 2016). The major occurrence of these congeners is related to the
311 high degree of chlorination on the aromatic rings which results in a lower degradation rate by
312 organisms and a potential bioaccumulation (Jönssonn et al., 2003; Storelli et al., 2009). Specific
313 differences in PCBs content could reflect the dissimilar behaviour and feeding patterns, trophic
314 levels, physiological status of organisms or metabolic detoxification capacity (Ashley et al., 2003)
315 typical of the studied species. In particular, *S. sphyraena*, which is an active predatory fish,
316 occupying a high level in the trophic chain (Premolatha and Manojkumar, 1990), showed the
317 highest concentration of PCBs with respect to other species. Also, the *Mullus* species exhibited
318 higher values of ∑PCBs most likely related to their ecological features (bottom-feeder) and
319 consequently greater probability of exposure to PCBs from sediments.
320 PAHs were measured only in organisms sampled in 2017, revealing differences among the analysed
321 species both as total content and individual PAHs (Tab. 3). Finfish and shellfish showed a major
322 content of low molecular weight (LMW) PAHs, while high molecular weight (HMW) PAHs were
323 all <LOD (0.8 ng g^{-1}). The ∑PAHs ranged between 9.22 ng g^{-1} (*S. sphyraena*) and 22.0 ng g^{-1}
324 (*Sepia* spp.). The Benzo-a-pyrene (BaP), used as reference of PAHs presence in seafood (Reg. EC

325 1881/2006 as amended), was <LOD in all finfish and shellfish except in mussels. The latter exceed
326 threshold limit of Benzo-a-pyrene established for molluscs (5 ng g^{-1}; Legislative Decree 172/2015).
327 The OCs pollutants in transplanted mussels showed higher values than those measured in the
328 control samples, suggesting significant bioaccumulation (Tab. 3). HCB detected in transplanted
329 mussel samples revealed differences between the two sites, with always higher HCB values in
330 samples from the southern sites over both the sampling periods (S1 and S2), with levels up to 7 time
331 higher (1.91 and 3.15 ng g^{-1}, respectively) than those measured in the northern site (0.44 and 0.41
332 ng g^{-1}) (Tab. 3). Recorded values, compared to those from other Mediterranean areas (Ferrante et
333 al., 2007), resulted higher and suggested that mussels from Augusta Bay were exposed to high-
334 levels of this pollutant.
335 The ∑PCBs in S1 mussels (35.2 ng g^{-1}) was >3 times than values measured in N1 organisms
336 (∑PCBs= 11.66 ng g^{-1}). A higher content of ∑PCBs, compared to N2 (27.1 ng g^{-1}), was confirmed
337 in S2 mussels (39.3 ng g^{-1}). Furthermore, ∑7PCBs resulted above the OSPAR limit in all samples
338 except in N1 mussels. The analysis of PCB congeners showed that the penta (101, 118), hexa (138-
339 153) and epta-chlorinated PCBs (180) were generally more abundant than the less chlorinated forms
340 (28 and 52) (Tab. 3) reflecting previous data reported by Ferrante et al. (2007), Beiras et al. (2012)
341 and Campillo et al. (2017).
342 Also, PAHs concentrations were higher in transplanted mussels (∑PAHs ranged between 34.2 and
343 76.4 ng g^{-1}) than in the control sample (∑PAHs= 25.0 ng g^{-1}) with B(a)P showed values above the
344 EQS (5 ng g^{-1}) (Tab. 3). In particular, S1 and S2 organisms exhibited a marked increase of B(a)P
345 (11.3 and 11.02 ng g^{-1}, respectively) in relation to the initial value of control samples (4.3 ng g^{-1}).
346 All samples showed values above the limit of 30 ng g^{-1} for Fluoranthene (Tab. 3).
347

348 **Discussion and conclusive remarks**

349 Recent studies revealed a systematic correlation between PCBs in biota and in nearly un-weathered

350 PCB mixtures in sediments (Rodenburg et al., 2015, 2019). Several investigations (e.g., Wiegel
351 and Wu, 2000; Bedard et al., 2005; Zanaroli et al., 2015; Praveckova et al., 2016) evidenced that
352 dehalogenation processes affect PCBs under strictly anaerobic conditions and low redox potential
353 and thus changing the congener patterns from highly to low chlorinated congeners.
354 The PCBs composition in surface sediments appears dominated by the higher chlorinated penta,
355 hexa and hepta CBs, accounting for >60% of ∑PCBs in all samples (Fig. 3). As previously
356 mentioned, differences in ∑PCBs content measured in the various species, primarily depend on a
357 number of different ecological features (Ashley et al., 2003; Arnot and Gobas, 2006; Martinez-Silva
358 et al., 2018) and might reflect a non-homogeneous proportion of the various congeners.
359 Nonetheless, the compositional patterns of the PCB congeners in the studied fishes appear
360 reasonably comparable (Fig. 3), and their distribution mode in finfish and shellfish maintains a
361 systematic order of concentration: hexa->penta->hepta->tetra->tri-CBs, contributing for 58%, 24%,
362 21%, 4.5% and 0.3% (Fig. 3). Particularly, the major contribution of penta and hexa-CBs is in
363 excellent agreement with data reported for biota by other authors (Naso et al., 2005; Ferrante et al.,
364 2007; Howell et al., 2008; Pan et al., 2016; Habibullah-Al-Mamun et al., 2019). The mussels
365 showed a similar profile although with a minor percentage of epta-CBs accounting for 5.1-8.5% of
366 ∑PCBs (Fig. 3).

367 The composition pattern of the 7 PCB congeners (28, 52, 101, 118, 138, 153 and 180) in all samples
368 was explored by cluster analysis (Fig. 4), using the elbow method to identify the optimal number of
369 groups primarily driving the high variability in the studied data population. Except for *Diplodus*
370 spp., all the finfish and shellfish samples appear strictly grouped in the cluster together with
371 sediment samples from the A9 core, while all the mussel samples and the other sediment samples
372 (core A3, A7 and A11) grouped in two separate clusters (Fig. 4).

373 The statistically significant relationship between sediments from the A9 core and finfish and
374 shellfish suggests a first-order, although robust, insight on the intimate link between the abiotic and

375 biotic compartments and reasonably supports in the study area, the use of the PCB congeners as
376 potential source fingerprint for these contaminants. Indeed, sediments from the A9 core showed
377 highly comparable percentages of high-chlorinated congeners (CB 153, 138 and 180) whereas
378 mussels (native and transplanted) and sediments from the A3, A7 and A11 cores evidence different
379 PCB patterns. Although the period of permanence of the organisms in the area of the A9 core could
380 not be precisely assessed, our results suggest that this zone would represent a primary
381 contamination 'hot spot' area for Augusta Bay. Differently, the other sediment cores, characterized
382 by different and relatively lower PCBs concentration, might represent secondary sources of these
383 contaminants to the biotic compartment.

384 Analogue pieces of evidence, supporting transfer of contaminants from sediment to biota, again
385 emerge from the analysis of the distribution patterns of specific PAHs, which in finfish and
386 shellfish are commonly dominated by LMW hydrocarbons, in particular Fluoranthene and Pyrene.
387 Indeed, these two molecules have a large range of stability and are good indicators of
388 thermodynamic vs. kinetic processes (Soclo et al., 2000). The Fluoranthene/Pyrene has been
389 frequently applied to identify and characterize sources of PAHs in the marine environment (Magi et
390 al., 2002). On the other side, the distribution pattern of PAHs in mussels has been reported to reflect
391 accumulation of bioavailable fraction from the water column (Baumard et al., 1998) with
392 preferential accumulation of LMW PAHs (Varanasi and Gmur, 1981; Broman et al., 1990). The
393 dotchart in figure 5 evidence a significant correlation between biota and sediments, with
394 comparable Fluoranthene/Pyrene ratios in finfish, shellfish and A9 sediments. Worthy to note, also
395 Fluoranthene/Pyrene measured in N2, S1 and S2 mussels which seem to mirror a potential impact
396 from the A9 sediments. This could primarily suggest an effect of that 'hot-spot' area in terms of
397 resuspension and re-distribution of highly contaminated particles on a relatively wider distance
398 within the study area (see Denaro et al., 2020 for a complete modelling discussion on the dynamic
399 within Augusta Bay). Thus, if PCBs seem to offer a primary fingerprint to follow sediment-to-biota

transfer of contaminants, the combined use of specific PAHs patterns convincingly supports the hypotheses of dynamic transfer of those organics in the various environmental compartments. Particularly, a clear 'hot-spot' effect of the A9 sediments on the investigated biotic compartment can be also documented taking into account the Hg and HCB concentration patterns mirroring coherent higher concentrations in the analysed benthic fish. Conclusively, the ensemble of achieved results suggests that combined information from a wide spectrum of contaminants providesa consistent fingerprint to trace their transfer from sediments to biotic compartments and thus supports specific remediation decisions and sediment mitigation strategies. Despite the statistical robustness, the obtained results could primarily reflect specific dynamics of local biogeochemistry and a critical role of local effects on their distribution patterns, mainly at the abiotic and biological marine interfaces. However, these promising results encourage to the use of PCB congeners and pattern of pollutants as fingerprint of exposure pathways from marine sediments to biota in other marine environmental contests.

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730 **Figure Captions**
731
732 **Figure 1:** Map and details of the sampling in Augusta Bay.
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734 **Figure 2:** Box plot of the concentrations of Hg (a), HCB (b), ∑PCBs (c) and ∑PCBs NDL (d) in
735 sediment cores.
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737 **Figure 3:** Relative contribution of PCB homologs (% composition) in sediments, seafood and
738 mussel samples.
739
740 **Figure 4:** Heatmap generated from hierarchical clustering analysis. The dendrograms of sample
741 clustering (on the left) and of congener clustering (on the top) were added. The colour bars inside
742 the graph indicate the different proportion of congeners (X axis) for each sample (Y axis). The rows
743 were splitted based on number of identified clusters.
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745 **Figure 5:** Dotchart of Fluo/Py in sediments and marine organisms
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Tables

	Specie	Category	Habitat	Total lenght range (med ±st.dv) cm	n. total individuals	n. pool
2013	*Mytilus galloprovincialis*	Molluscs	benthic	3.6-6 (4.60±0.58)	200	20
	Sphyraena sphyraena	Fish	pelagic	46-44 (45±1.41)	2	2
	Mullus barbatus	Fish	benthic	17.5-21 (18.7±1.14)	30	6
	Pagellus spp.	Fish	demersal	15-26 (20±3.25)	15	5
	Sparus aurata	Fish	demersal	21-24 (21.6±1.21)	9	3
	Serranus cabrilla	Fish	demersal	20	1	1
	Diplodus spp.	Fish	demersal	14.1-16.3 (15.1±1.1)	3	2
2017	*Sphyraena sphyraena*	Fish	pelagic	37.4	1	1
	Trigla lucerna	Fish	benthic	22.3-22.5	2	1
	Mullus barbatus	Fish	benthic	17.7-20.5 (18.8±1.1)	6	2
	Pagellus spp.	Fish	demersal	15.3-25.4 (18.9±2.7)	37	8
	Diplodus spp.	Fish	demersal	13.5-20 (16.8 ±3.2)	4	3
	Penaeus kerathurus	Crustacean	benthic	4.5-6.5 (5.2±0.3)	39	4
	Sepia spp.	Molluscs	bento-nectonic	9.9-15.5 (12.5±2.0)	7	6
	Mytilus galloprovincialis N	Molluscs	benthic	4.4-5.9 (5.20±1.06)	392	13
	Mytilus galloprovincialis S	Molluscs	benthic	5.0-5.4 (5.2±0.3)	284	10

Table 1. Characteristics, number of specimens and pool of marine organisms caught in the study area.

CORE	Unit	A3 mean ±st.dv	A7 mean ±st.dv	A9 mean ±st.dv	A 11 mean ±st.dv
Hg	µg g $^{-1}$	**3.64±0.27**	**6.71±0.80**	**11.4 ±0.57**	**6.54±1.28**
HCB	ng g $^{-1}$	**8.53 ± 2.06**	**25.3± 8.03**	**172.6 ± 87.6**	**22.8 ± 14.45**
PCB 28	ng g $^{-1}$	10.36 ± 6.66	2.26 ± 0.82	0.40 ± 0.12	9.65 ± 12.6
PCB 52	ng g $^{-1}$	0.42 ± 0.53	<LOD	1.35 ± 0.30	0.56 ± 0.94
PCB 81	ng g $^{-1}$	<LOD	<LOD	<LOD	<LOD
PCB 77	ng g $^{-1}$	<LOD	<LOD	0.72±0.13	<LOD
PCB 101	ng g $^{-1}$	<LOD	<LOD	5.22 ± 1.40	<LOD
PCB-118	ng g $^{-1}$	6.90 ± 4.66	0.97 ± 0.77	3.79 ± 0.96	8.74 ± 16.9
PCB 114	ng g $^{-1}$	<LOD	<LOD	<LOD	<LOD
PCB 123	ng g $^{-1}$	<LOD	<LOD	11.2 ± 34.6	<LOD
PCB 153	ng g $^{-1}$	10.4 ± 5.77	3.43±1.95	10.4 ± 1.07	16.2 ± 13.3
PCB 105	ng g $^{-1}$	1.18 ± 1.88	<LOD	1.26 ± 0.80	1.24 ± 2.84
PCB 138	ng g $^{-1}$	7.59 ± 7.11	2.83 ± 2.56	9.72 ± 1.53	17.3 ± 20.0
PCB 126	ng g $^{-1}$	<LOD	<LOD	1.06 ± 0.24	<LOD
PCB 128+157	ng g $^{-1}$	<LOD	0.16±0.20	0.62 ± 0.22	1.10 ± 1.21
PCB 156	ng g $^{-1}$	<LOD	0.19 ± 0.29	0.58 ± 0.34	0.80 ± 1.65
PCB 167	ng g $^{-1}$	<LOD	<LOD	1.04 ± 0.08	0.84 ± 1.75
PCB 180	ng g $^{-1}$	0.81 ± 1.17	1.53 ± 1.58	7.89 ±1.45	4.30 ± 3.26
PCB 169	ng g $^{-1}$	<LOD	<LOD	<LOD	<LOD
PCB 170	ng g $^{-1}$	<LOD	0.34 ± 0.54	3.85 ± 0.36	2.35±2.03
PCB 189	ng g $^{-1}$	<LOD	<LOD	0.25 ± 0.09	<LOD
∑ PCBs	ng g $^{-1}$	38.7 ± 26.2	12.6 ± 6.7	59.5 ± 35.6	38.2 ± 19.9
∑ PCBs[1]	ng g $^{-1}$	**29.6 ± 26.6**	**11.8 ±6.50**	**41.9 ± 5.60**	**34.3 ± 16.2**
∑ 7PCB	ng g $^{-1}$	36.6 ± 24.4	11.2 ± 6.39	38.7 ± 5.20	32.2 ± 13.6
Phenanthrene	ng g $^{-1}$	37.6 ± 8.52	26.4 ± 6.56	67.8 ± 23.3	27.7 ± 11.1
Anthracene	ng g $^{-1}$	**180 ± 68.9**	**186.8 ± 29.6**	**56.3 ± 14.6**	**91.7 ± 27.6**
Fluoranthene	ng g $^{-1}$	61.8 ± 32.2	35.3 ± 18.1	45.2 ± 24.7	27.5 ± 12.8
Pyrene	ng g $^{-1}$	33.1 ± 13.7	24.5 ± 8.11	61.2 ± 22.3	25.4 ± 10.2
Benzo-a-Anthracene	ng g $^{-1}$	34.2 ± 11.1	17.0 ± 6.18	26.4 ± 10.6	13.5 ± 3.94
Chrysene	ng g $^{-1}$	38.5 ± 10.2	20.6 ± 6.72	48.2 ± 13.1	17.8 ± 4.99
Benzo-b-Fluoranthene	ng g $^{-1}$	**54.6 ± 20.4**	32.5 ± 7.45	30.5 ± 8.51	14.4 ± 4.24
Benzo-k-Fluoranthene	ng g $^{-1}$	**33.6 ± 24.3**	9.26 ± 2.54	10.3 ± 4.59	7.09 ± 2.03

Benzo-a-Pyrene	ng g^{-1}	**64.6 ± 20.1**	29.3 ± 8.53	23.5 ± 6.94	20.5 ± 6.42
Indeno-123-cd-Pyrene	ng g^{-1}	**70.1 ± 25.5**	19.5 ± 6.19	9.88 ± 5.95	13.1 ± 2.30
Dibenzo-ah-Anthracene	ng g^{-1}	12.9 ± 6.30	1.87 ± 1.36	7.90 ± 4.29	2.91 ± 1.94
Benzo-ghi-Perylene	ng g^{-1}	50.6 ± 21.7	17.6 ± 7.72	11.15 ± 5.30	15.7 ± 4.06
∑ PAHs	ng g^{-1}	671.87 ± 125.97	420 ± 72.73	398 ± 103	277 ± 64.8

Values exceeding environmental quality standards (Dlgs. 172/2015) are indicated in bold
∑ PCBs[1]: sum of PCBs congeners indicated by Dlgs.172/2015
∑ 7PCB: sum of CB 28+52+101+118+138+153+180

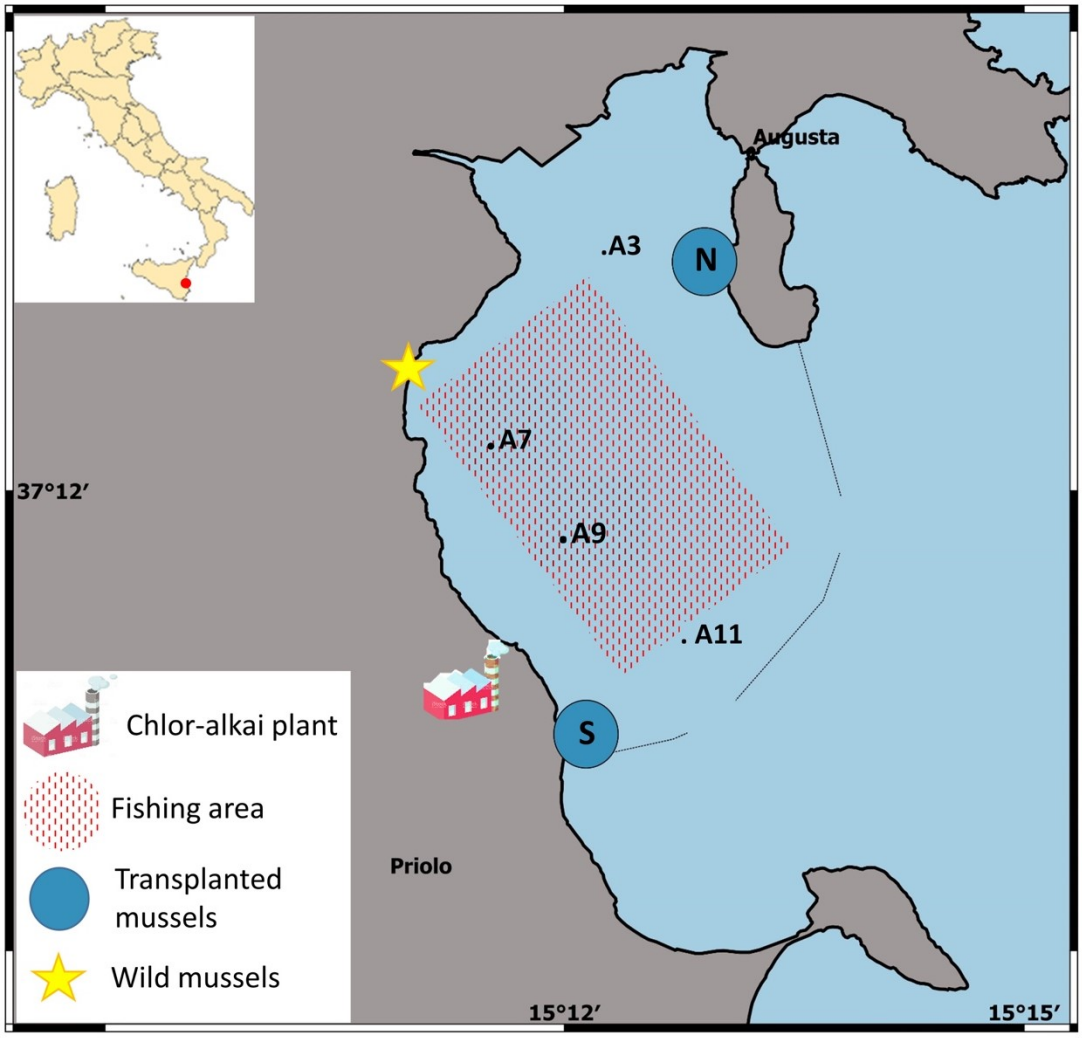
Table 2. Hg, HCB, PCBs and PAHs concentrations (d.w.) in sediment collected in the sampling site.

Parameters	Unit	Sampling 2013							Sampling 2017											
		S. sphyraena	*M. barbatus*	*Pagellus spp.*	*S.aurata*	*S. cabrilla*	*Diplodus spp.*	*M.galloprovincialis wild*	*S. sphyraena*	*T. lucerna*	*M. barbatus*	*Pagellus spp.*	*Diplodus spp.*	*P. kerathurus*	*Sepia spp.*	*M. gallorprovincialis*				
																CTRL	*N1*	*S1*	*N2*	*S2*
		med ±st.dv							med ±st.dv											
Hg	µg g^{-1}	**0.52 ±0.04** [a]	0.44 ± 0.20	0.53 ± 0.12	0.32 ± 0.09	**0.77** [a]	**0.503±0.137** [a]	**0.2 ± 0.04** [a]	**0.78** [a]	**0.66** [a]	**2.11;1.71** [a]	0.86±0.44	**1.49±1.77** [a]	**0.59±0.02** [a]	0.21 ± 0.08	0.01	**0.02 ± 0.002** [b]	**0.23 ± 0.02** [b]	**0.12 ± 0.01** [b]	**0.89** [a,b]
HCB	ng g^{-1}	n.d	1.36 ± 0.31	0.09 ± 0.02	0.60 ± 0.43	n.d	n.d	0.13 ± 0.00	1.40	<LOD	0.24;0.65	<LOD	<LOD	<LOD	<LOD	0.35	0.44	1.91	0.41	3.15
PCB 28	ng g^{-1}	n.d	0.36 ± 0.06	0.06 ± 0.01	0.16±0.09	n.d	n.d	0.15±0.00	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PCB 52	ng g^{-1}	n.d	6.31 ± 2.27	2.77 ± 0.63	4.16±2.32	n.d	n.d	4.95±1.54	2.03	<LOD	1.29;2.42	0.48 ± 0.50	<LOD	<LOD	<LOD	<LOD	<LOD	0.72	<LOD	<LOD
PCB 81	ng g^{-1}	n.d	0.59 ± 0.10	0.18 ± 0.08	0.21±0.11	n.d	n.d	0.26±0.01	<LOD	<LOD	1.69;<LOD	<LOD	<LOD	0.41 ± 0.34	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PCB 77	ng g^{-1}	n.d	<LOD	<LOD	<LOD	n.d	n.d	0.05±0.01	2.01	<LOD	1.0;1.49	0.69 ± 0.61	0.40	0.64 ± 0.45	0.65 ± 0.55	<LOD	<LOD	<LOD	<LOD	<LOD
PCB 101	ng g^{-1}	n.d	8.29 ± 4.97	3.89±1.46	4.66±2.39	n.d	n.d	5.82±1.22	12.7	1.17	3.45;5.46	3.17 ± 2.93	0.42	1.54 ± 0.48	0.93 ± 1.05	0.39	1.36	3.91	2.26	2.70
PCB-118	ng g^{-1}	n.d	9.91 ± 1.95	3.29 ± 1.30	5.03±2.41	n.d	n.d	2.87±0.44	12.7	<LOD	6.72;7.26	1.96 ±2.42	2.37	1.70 ± 1.79	<LOD	0.47	1.09	2.84	3.39	3.20
PCB 114	ng g^{-1}	n.d	0.08 ± 0.06	<LOD	0.05±0.03	n.d	n.d	<LOD	<LOD	<LOD	7.56;<LOD	2.46 ±2.86	2.59	2.91 ± 1.35	0.38 ± 0.55	<LOD	0.59	2.50	2.88	2.76
PCB 123	ng g^{-1}	n.d	<LOD	<LOD	<LOD	n.d	n.d	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PCB 153	ng g^{-1}	n.d	36.68 ± 2.93	13.5±4.40	18.5±8.28	n.d	n.d	10.6±1.37	62.8	7.81	40.30	16.6 ± 12.3	0.74	10.3 ± 0.87	10.58 ± 8.81	0.93	2.58	9.64	8.44	13.66
PCB 105	ng g^{-1}	n.d	1.78 ± 0.66	0.67±0.28	1.09±0.67	n.d	n.d	0.83±0.01	2.61	<LOD	1.01;1.32	0.52 ± 0.31	0.66	0.68 ± 0.38	<LOD	<LOD	0.41	0.56	0.66	0.49
PCB 138	ng g^{-1}	n.d	27.29 ± 2.51	10.6±3.64	13.9±6.06	n.d	n.d	8.46±0.97	52.4	5.80	35.34;3.19	15.7 ± 9.8	9.22	10.5 ± 0.99	1.81 ± 1.93	0.78	2.87	9.53	5.51	11.26
PCB 126	ng g^{-1}	n.d	0.36 ± 0.04	0.13±0.07	0.16±0.09	n.d	n.d	0.14±0.01	39.6	6.68	38.84;22.20	8.70 ± 9.35	<LOD	5.02 ± 5.87	4.53 ± 6.83	<LOD	<LOD	0.42	<LOD	0.58
PCB 128+157+167	ng g^{-1}	n.d	4.32	1.68	2.28	n.d	n.d	1.75	1.70	0.69	2.29	1.79	0.49	0.77	0.77	0.32	0.45	1.35	1.11	0.70
PCB 156	ng g^{-1}	n.d	1.42 ± 0.14	0.67±0.26	0.73±0.32	n.d	n.d	0.44±0.02	1.64	0.72	4.02;3.01	1.51 ± 1.35	1.00	1.12 ± 0.37	<LOD	<LOD	<LOD	0.35	0.32	0.44
PCB 180	ng g^{-1}	n.d	17.31 ± 2.69	8.39±1.92	6.84±3.12	n.d	n.d	1.45±0.06	38.1	3.76	25.24;19;49	10.9 ± 7.17	8.24	5.22 ± 0.62	8.42 ± 6.61	<LOD	0.63	1.98	1.05	1.63
PCB 169	ng g^{-1}	n.d	<LOD	<LOD	<LOD	n.d	n.d	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
PCB 170	ng g^{-1}	n.d	6.77 ± 0.52	3.55±0.82	3.31±1.47	n.d	n.d	0.42±0.03	12.44	1.83	0.43;9.32	3.07 ± 2.28	1.46	1.82±1.12	0.44 ± 0.36	<LOD	0.16	0.52	0.31	0.73
PCB 189	ng g^{-1}	n.d	0.32 ± 0.04	0.14±0.04	0.15±0.08	n.d	n.d	0.07±0.01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
∑ PCB$_S$	ng g^{-1}	n.d	121.8 ± 1.47	49.5±13.9	1.3±22.2	n.d	n.d	38.4±5.27	241.7	30.1	175.1;113.8	68.5 ± 37.8	28.70	43.6 ± 6.93	30.1± 24.0	4.98	11.7	35.2	27.1	39.3
∑ NDL-PCB	ng g^{-1}	n.d	96.2 ± 9.81	39.2±10.6	48.3±16.2	n.d	n.d	31.5±5.0	**168.2** [a]	18.9	**110.5;**66.3 [a]	47.0 ± 31.2	18.90	27.9 ± 2.32	22.1 ± 17.5	2.59	7.87	25.94	17.58	29.57
∑ 7PCB	ng g^{-1}	n.d	**106 ± 11.4**	**42.5 ±11.9**	**53.3±18.5**	n.d	n.d	34.4±5.5	**180.90**	**19.0**	**117.2; 73.6**	**48.9 ± 32.7**	**21.30**	**29.7 ± 2.87**	**22.4 ± 17.7**	3.06	8.96	**28.8**	**20.9**	**32.8**
Phenanthrene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	1.52	3.97	7.50;8.56	2.64 ± 1.07	1.33	3.61 ± 1.54	2.15±1.69	4.60	3.74	2.91	2.77	1.63
Anthracene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	6.57	11.83
Fluoranthene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	1.54	2.85;1.40	2.53 ± 2.90	3.16	0.81 ± 0.48	4.32± 3.26	3.02	13.73	4.07	7.62	8.96
Pyrene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	3.70	5.34	7.49;5.18	9.03 ± 11.36	12.73	3.60 ± 1.05	11.97 ± 12.30	6.80	7.42	5.85	10.09	26.99
Benzo-a-Anthracene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.68	0.65	3.61	1.09
Chrysene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	3.08	1.19	5.09	2.74
Benzo-b-Fluoranthene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	2.11	1.24	5.21	2.93
Benzo-k-Fluoranthene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.94	0.69
Benzo-a-Pyrene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	4.31	**8.50** [a,b]	**11.3** [a,b]	**6.57** [a,b]	**11.0** [a,b]
Indeno-123-cd-Pyrene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	3.42	15.72	5.35	8.49	7.16
Dibenzo-ah-Anthracene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD
Benzo-ghi-Perylene	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.20	0.97
∑ PAHs	ng g^{-1}	n.d	n.d	n.d	n.d	n.d	n.d	n.d.	9.22	14.4	21.4;18.7	18.0 ± 13.8	21.70	11.6 ± 3.00	22.0 ± 16.9	25.0	57.6	34.2	59.6	76.4

Values exceeding reference limits are expressed in bold; [(a)] Reg CE 1881/2006 as amended; [(b)] Dlgs.172/2015; ∑ 7PCB sum of CB 28+52+101+118+138+153+180 (Webster et al., 2013).

Table 3. Concentrations (w.w.) of Hg, HCB, PCBs and PAHs in mare organisms from the Augusta Bay.

Figures



Core	Bottom depth (m)	Latitude N	Longitude E	Core length (cm)
A3	19.8	37°13'3.70"	15°12'27.56"	16
A7	18.6	37°12'17.24"	15°11'40.94"	21
A9	23	37°11'36.18"	15°12'08.98"	18
A11	22.5	37°11'0.37"	15°12'48.69"	11

Figure 1: Map and details of the sampling in Augusta Bay.

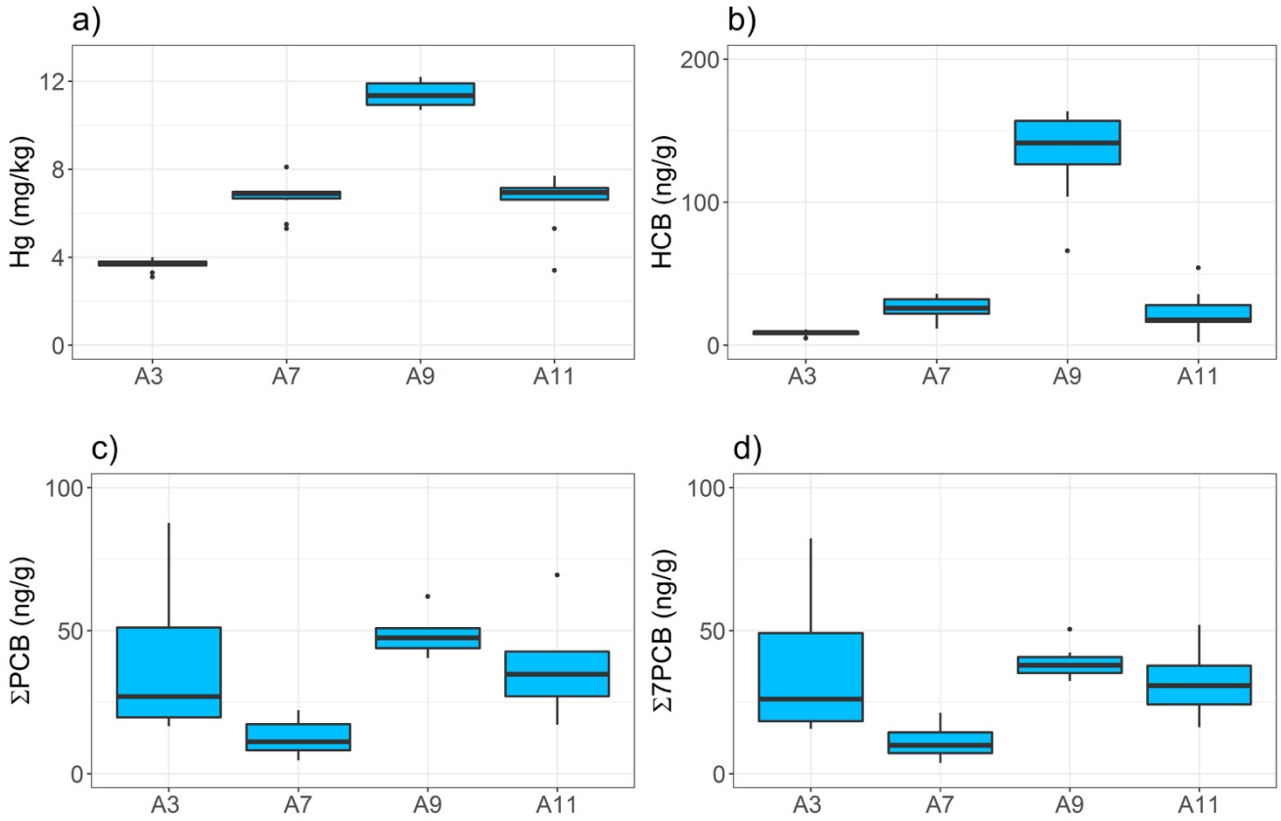


Figure 2: Box plot of the concentrations of Hg (a), HCB (b), ∑PCBs (c) and ∑PCBs NDL (d) in sediment cores.

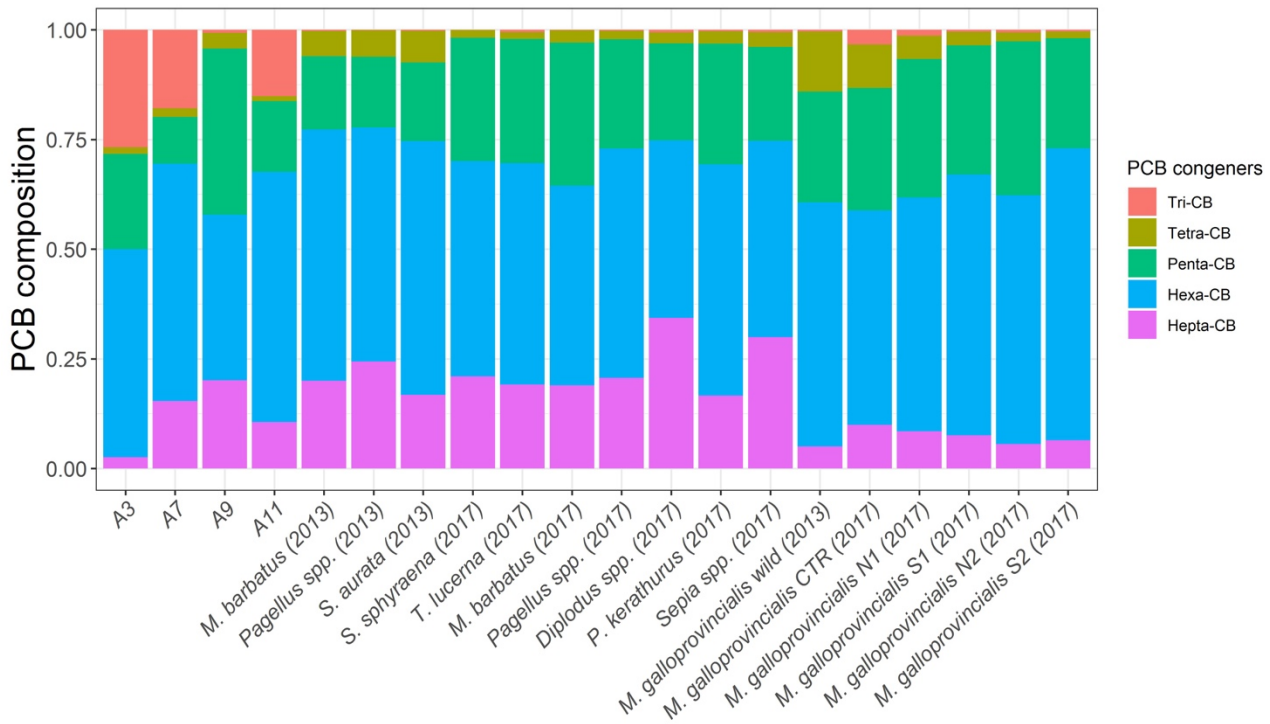


Figure 3: Relative contribution of PCB homologs (% composition) in sediments, seafood and mussel samples.

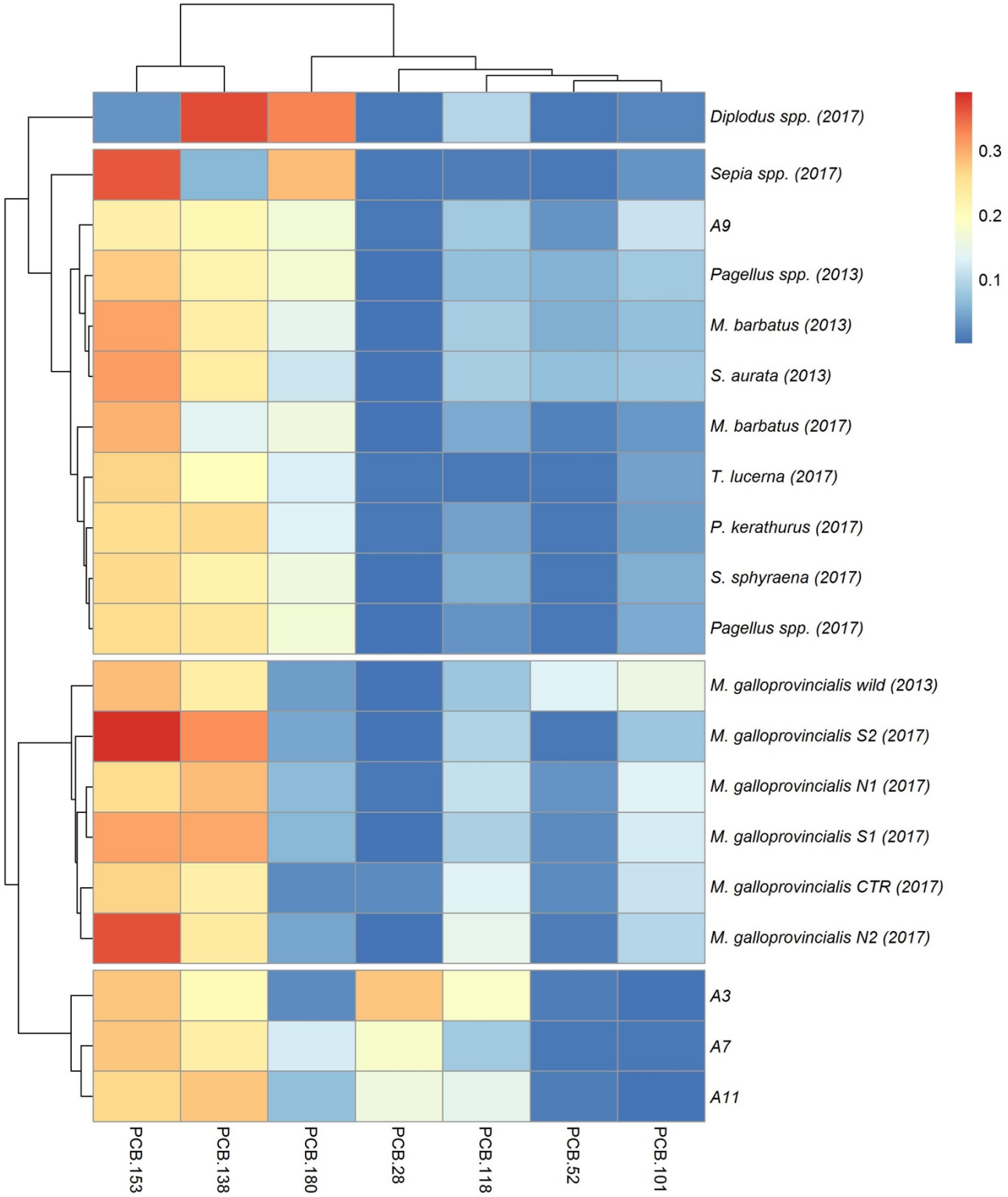


Figure 4: Heatmap generated from hierarchical clustering analysis. The dendrograms of sample clustering (on the left) and of congener clustering (on the top) were added. The colour bars inside the graph indicate the different proportion of congeners (X axis) for each sample (Y axis). The rows were splitted based on number of identified clusters.

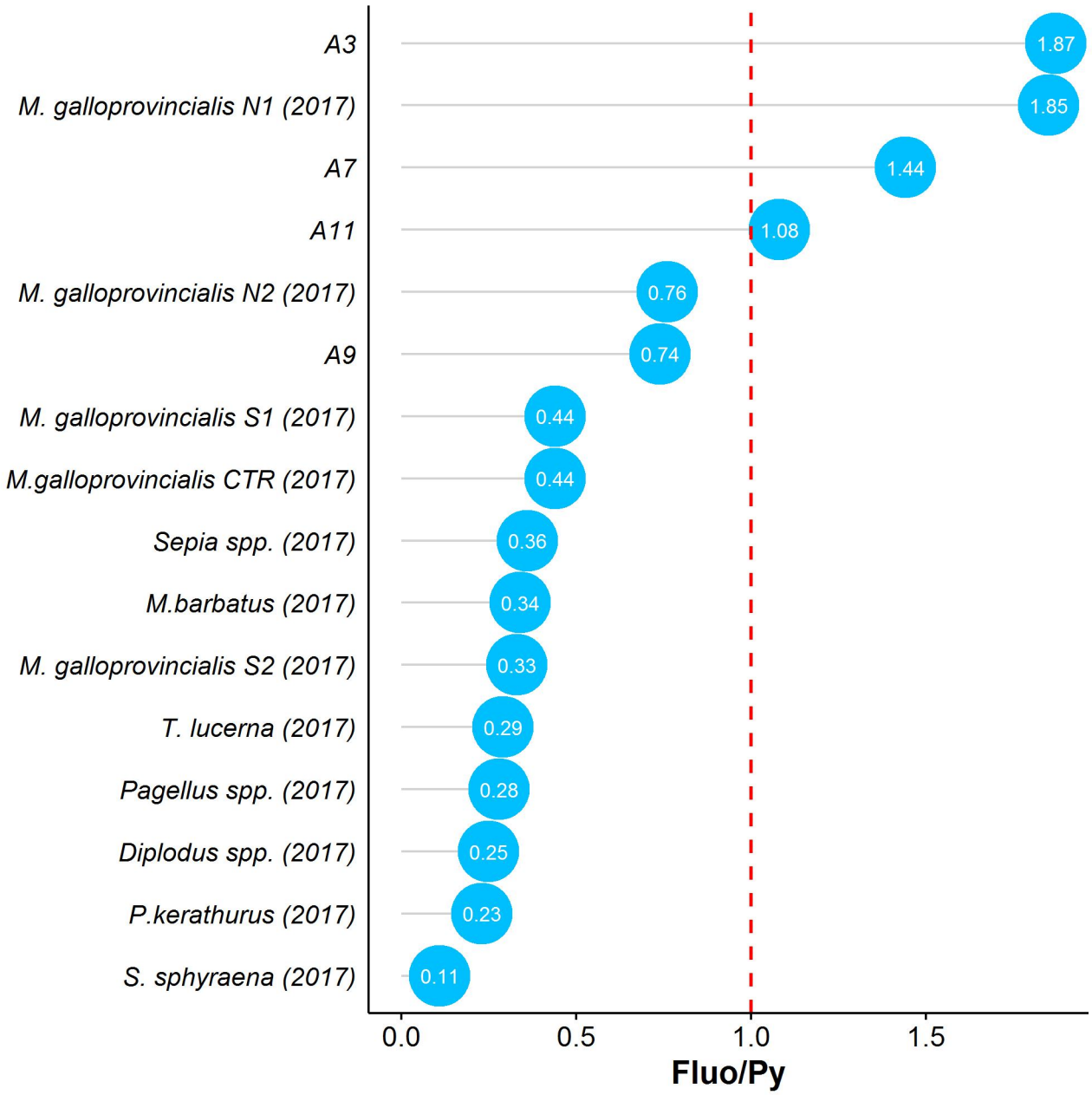


Figure 5: Dotchart of Fluo/Py in sediments and marine organisms

Supplementary materials

Table S1 Hg, HCB and PCBs concentrations (d.w.) in 0-10 cm levels of sediment collected in the sampling site.

Table S1 Hg, HCB and PCBs concentrations (d.w.) in 0-10 cm levels of sediment collected in the sampling site.

Core	level	Hg	HCB	PCB 28	PCB 52	PCB 81	PCB 77	PCB 101	PCB 118	PCB 114	PCB 123	PCB 153	PCB 105	PCB 138	PCB 126	PCB 128+157	PCB 156	PCB 167	PCB 180	PCB 169	PCB 170	PCB 189	∑ PCBs	∑ PCBs[1]	∑ 7PCB
	cm	µg g^{-1}	ng g^{-1}																						
A 3	0-1	3.73	10.6	5.85	0.31	<LOD	<LOD	<LOD	3.77	<LOD	<LOD	6.34	<LOD	4.42	<LOD	<LOD	<LOD	<LOD	1.66	<LOD	<LOD	<LOD	23.3	22.9	22.4
	1-2	3.07	11.2	4.98	<LOD	<LOD	<LOD	<LOD	3.36	<LOD	<LOD	5.91	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	1.21	<LOD	<LOD	<LOD	16.6	16.1	15.7
	2-3	3.73	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	3-4	3.81	6.29	14.7	<LOD	<LOD	<LOD	<LOD	8.23	<LOD	<LOD	14.7	<LOD	7.09	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	45.9	45.4	45.0
	4-5	3.71	9.06	4.55	<LOD	<LOD	<LOD	<LOD	2.74	<LOD	<LOD	5.90	<LOD	3.90	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	18.2	17.8	17.3
	5-6	3.96	8.42	16.0	0.41	<LOD	<LOD	<LOD	12.2	<LOD	<LOD	15.41	4.2	14.4	<LOD	<LOD	<LOD	<LOD	3.25	<LOD	<LOD	<LOD	66.8	62.2	61.8
	6-7	3.56	8.98	4.92	0.78	<LOD	<LOD	<LOD	3.59	<LOD	<LOD	5.37	0.65	3.92	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	20.2	19.2	18.7
	7-8	3.77	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	8-9	3.81	8.72	9.41	<LOD	<LOD	<LOD	<LOD	6.05	<LOD	<LOD	9.17	<LOD	4.92	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	30.7	30.2	29.8
	9-10	3.31	4.95	22.4	1.58	<LOD	<LOD	0.24	15.3	<LOD	<LOD	20.7	4.21	21.96	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	87.7	82.8	82.3
mean		**3.64**	**8.53**	10.36	0.42	<LOD	<LOD	<LOD	6.90	<LOD	<LOD	<LOD	1.18	7.59	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	38.7	**29.6**	**36.6**
st.dv		**0.27** [a]	**2.06** [a]	6.66	0.53				4.66			5.77	1.88	7.11					1.17				26.2	**26.6** [a]	**24.4** [b]
A 7	0-1	6.89	23.0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	1-2	6.98	ND	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	2-3	6.99	16.0	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	3-4	6.93	11.6	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.

	4-5	6.65	32.1	1.62	<LOD	<LOD	<LOD	<LOD	1.02	<LOD	<LOD	0.83	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	4.67	4.22	3.77
	5-6	6.88	28.8	1.74	<LOD	<LOD	<LOD	<LOD	1.24	<LOD	<LOD	4.46	<LOD	5.00	<LOD	<LOD	0.78	0.43	2.66	<LOD	1.43	<LOD	18.6	16.4	15.3
	6-7	6.92	32.8	2.01	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	4.15	<LOD	3.56	<LOD	0.56	<LOD	<LOD	2.04	<LOD	<LOD	<LOD	13.4	12.9	12.0
	7-8	5.26	22.1	3.79	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	2.47	<LOD	0.47	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	7.92	7.47	7.02
	8-9	5.47	26.0	1.82	0.21	<LOD	<LOD	<LOD	1.43	<LOD	<LOD	2.32	<LOD	1.52	<LOD	<LOD	<LOD	<LOD	0.44	<LOD	0.29	<LOD	8.93	8.27	7.82
	9-10	8.08	35.8	<LOD	<LOD	<LOD	<LOD	<LOD	2.00	<LOD	<LOD	6.35	<LOD	6.38	<LOD	<LOD	<LOD	<LOD	3.90	<LOD	<LOD	<LOD	22.2	21.8	21.3
mean		**6.71**	**25.3**	2.26	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	0.19	<LOD	1.53	<LOD	<LOD	<LOD	<LOD	**11.8**	**11.2**
st.dv		**0.80[a]**	**8.03 [a]**	0.82					0.77			1.95					0.29		1.58					**6.50 [a]**	**6.39 [b]**

A 9	0-1	10.7	65.9	0.32	1.01	0.09	0.55	3.88	2.81	<LOD	0.15	8.93	0.59	8.96	0.83	0.49	0.56	0.89	6.44	<LOD	3.46	0.26	40.4	35.0	32.4
	1-2	11.6	134	0.38	2.01	<LOD	0.89	8.40	5.90	0.03	0.50	12.7	1.45	13.5	0.94	0.96	1.31	0.92	7.54	0.13	4.00	0.32	62.0	54.8	50.5
	2-3	10.7	331	0.55	1.37	<LOD	0.78	5.74	2.95	<LOD	110	11.0	1.19	10.3	1.25	0.66	0.76	1.04	7.32	0.17	4.16	0.39	160	42.9	39.2
	3-4	12.2	140	0.29	1.17	<LOD	0.52	4.29	3.08	<LOD	0.44	9.63	0.76	9.03	0.93	0.50	0.76	1.09	6.69	0.22	3.44	0.26	43.3	37.2	34.2
	4-5	11.3	328	0.34	1.12	<LOD	0.58	4.42	3.33	0.01	0.26	9.96	0.62	9.34	0.84	0.54	0.44	1.10	7.99	<LOD	4.24	0.25	45.5	39.1	36.5
	5-6	11.3	142	0.48	1.25	<LOD	0.82	4.72	3.65	<LOD	0.15	10.13	0.53	9.22	1.24	0.48	0.73	1.08	11.62	<LOD	4.44	0.13	50.9	44.5	41.1
	6-7	12.0	155	0.34	1.25	<LOD	0.82	4.21	3.47	0.02	<LOD	9.55	1.05	8.11	0.99	0.43	0.48	1.02	7.84	<LOD	3.76	0.32	43.9	37.7	34.8
	7-8	11.4	104	0.36	1.27	<LOD	0.79	5.23	4.19	<LOD	<LOD	10.8	2.97	9.68	1.18	0.44	0.40	1.09	8.42	<LOD	3.70	0.12	50.8	42.9	39.9
	8-9	12.1	162	0.31	1.72	<LOD	0.61	6.74	4.86	<LOD	<LOD	11.0	1.18	10.62	0.83	0.67	0.29	0.99	6.99	<LOD	3.90	0.14	51.2	44.8	42.3
	9-10	10.8	164	0.64	1.30	<LOD	0.82	4.52	3.70	0.03	<LOD	9.96	2.28	8.45	1.55	1.07	0.08	1.14	7.99	<LOD	3.40	0.32	47.5	40.2	36.6
mean		**11.4**	**172.6**	0.40	1.35	<LOD	<LOD	5.22	3.79	<LOD	11.2	10.4	1.26	9.72	1.06	0.62	0.58	1.04	7.89	<LOD	<LOD	0.25	59.5	**41.9**	**38.7**
st.dv		**0.57 [a]**	**87.6 [a]**	0.12	0.30		0.13	1.40	0.96		34.6	1.07	0.80	1.53	0.24	0.22	0.34	0.08	1.45		0.36	0.09	35.6	**5.60 [a]**	**5.20 [b]**

A 11	0-1	6.94	17.4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	1-2	7.18	27.9	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	2-3	7.04	35.5	5.05	<LOD	<LOD	<LOD	<LOD	5.08	<LOD	<LOD	14.3	<LOD	19.1	<LOD	3.0	4.2	4.4	8.3	<LOD	5.4	<LOD	69.5	59.4	52.0
	3-4	7.74	54.1	4.30	<LOD	<LOD	<LOD	<LOD	2.09	<LOD	<LOD	6.44	<LOD	2.1	<LOD	<LOD	<LOD	<LOD	1.1	<LOD	<LOD	<LOD	17.1	16.7	16.2
	4-5	6.97	28.2	3.10	<LOD	<LOD	<LOD	<LOD	2.01	<LOD	<LOD	9.07	<LOD	4.3	<LOD	0.4	0.3	0.4	5.7	<LOD	1.2	<LOD	27.1	25.2	24.2
	5-6	6.63	16.0	5.18	0.64	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	12.3	<LOD	8.7	<LOD	1.0	<LOD	<LOD	3.8	<LOD	2.3	<LOD	34.8	32.1	30.8

	6-7	6.69	17.4	5.03	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	<LOD	12.2	<LOD	13.4	<LOD	<LOD	<LOD	<LOD	6.9	<LOD	4.1	<LOD	42.7	38.2	37.8
	7-8	7.45	18.0	35.3	2.42	<LOD	<LOD	<LOD	43.11	<LOD	<LOD	42.8	7.04	56.1	<LOD	2.2	<LOD	<LOD	<LOD	<LOD	1.1	<LOD	n.d.	n.d.	n.d.
	8-9	5.33	11.6	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
	9-10	3.40	1.98	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.
mean		**6.54**	**22.8**	9.65	0.56	<LOD	<LOD	<LOD	8.74	<LOD	<LOD	16.2	1.24	17.3	<LOD	1.10	0.80	0.84	4.30	<LOD	2.35	<LOD	38.2	**34.3**	**32.2**
st.dv		**1.28 [a]**	**14.45[a]**	12.6	0.94				16.9			13.3	2.84	20.0		1.21	1.65	1.75	3.26		2.03		19.9	**16.2[a]**	**13.6 [b]**

Table S2: PAHs concentrations (d.w.) in 0-10 cm levels of sediment collected in the sampling site.

Core	Layer	PHENANTHRENE	ANTHRACENE	FLUORANTHENE	PYRENE	BENZO[a]ANTHRACENE	CHRYSENE	BENZO[b]FLUORANTHENE	BENZO[K]FLUORANTHENE	BENZO[a]PYRENE	INDENO[123-cd]PYRENE	DIBENZO[ah]ANTHRACENE	BENZO[ghi]PERYLENE	∑PAHs
	cm	*ng g $^{-1}$*												
A3	0-1	53.5	136	15.9	15.0	20.9	30.9	63.0	91.9	77.1	112.2	29.6	96.6	742
	1-2	28.9	87.0	50.7	37.8	26.7	28.0	52.5	20.9	53.8	76.5	10.3	56.9	530
	2-3	49.9	118	128.9	65.8	61.8	64.1	84.6	37.8	111.4	104.0	13.7	72.4	913
	3-4	33.6	158	49.5	27.6	27.9	32.2	60.0	24.6	55.2	68.1	8.3	37.2	583
	4-5	39.3	230	63.4	29.8	31.5	35.6	48.9	23.3	54.2	74.0	14.6	49.5	694
	5-6	35.4	264	52.3	26.4	32.1	37.1	87.2	24.7	68.9	83.1	14.5	55.5	782
	6-7	32.9	286	44.6	36.0	34.5	40.7	30.0	13.5	37.4	49.6	9.4	21.7	636
	7-8	26.8	111	54.9	24.7	29.9	33.1	55.5	59.3	52.2	44.3	9.6	32.1	533
	8-9	35.6	184	52.7	26.9	35.4	38.5	29.2	16.6	62.6	32.0	8.9	36.1	558
	9-10	40.4	227	105.5	41.2	41.5	44.4	34.9	23.1	73.7	56.8	10.5	48.5	747
mean		**37.6**	**180**	**61.8**	**33.1**	**34.2**	**38.5**	**54.6**	**33.6**	**64.6**	**70.1**	**12.9**	**50.6**	**671.87**
st.dv		**8.52**	**68.9**	**32.2**	**13.7**	**11.1**	**10.2**	**20.4**	**24.3**	**20.12**	**25.5**	**6.30**	**21.7**	**125.97**
A7	0-1	41.8	186	81.1	34.3	32.7	37.8	49.8	14.8	50.2	34.4	4.77	30.9	599
	1-2	28.7	112	45.1	38.3	18.9	22.5	34.0	10.8	32.2	23.0	3.62	24.4	394
	2-3	28.0	184	37.5	25.8	18.6	23.1	33.0	7.2	29.1	20.4	1.85	25.5	434
	3-4	25.6	184	33.9	22.8	15.3	17.8	33.0	7.7	27.8	19.0	1.62	15.9	404

	4-5	23.9	201	29.3	21.5	15.8	18.9	35.4	9.0	27.4	21.6	1.41	16.7	422
	5-6	23.0	218	28.4	20.9	13.4	16.5	30.0	9.9	24.8	16.0	1.01	11.2	413
	6-7	28.2	211	32.2	25.7	17.7	19.9	32.8	8.7	29.6	18.1	1.72	20.1	446
	7-8	18.3	177	16.5	13.6	10.5	14.4	23.1	6.6	18.2	13.1	0.00	8.1	319
	8-9	19.2	209	18.7	12.7	11.6	14.7	23.0	6.7	21.9	14.1	1.54	7.2	360
	9-10	27.8	186	30.7	28.8	15.6	20.4	31.4	11.1	31.3	14.7	1.17	16.1	415
mean		26.4	**186.8**	35.3	24.5	17.0	20.6	32.5	9.26	29.3	19.5	1.87	17.6	420
st.dv		6.56	**29.6**	18.1	8.11	6.18	6.72	7.45	2.54	8.53	6.19	1.36	7.72	72.73

A 9	0-1	54.6	70.4	31.6	37.1	18.9	37.7	24.3	9.28	19.1	6.20	6.92	19.8	336
	1-2	46.1	76.2	32.6	40.7	18.8	37.0	25.2	6.61	18.0	6.27	7.53	13.7	329
	2-3	59.3	47.5	37.2	61.1	25.2	58.2	31.3	7.20	24.3	9.05	6.91	10.2	378
	3-4	60.0	65.7	34.2	50.6	20.3	40.1	24.6	7.74	20.0	9.54	6.22	9.12	348
	4-5	68.6	53.0	57.9	71.8	36.7	59.5	36.2	12.00	30.9	16.67	13.37	20.7	477
	5-6	130	23.4	112	118	53.0	78.0	51.9	22.43	39.7	23.91	3.75	9.90	666
	6-7	64.4	61.0	35.0	55.7	23.2	41.2	26.6	9.59	24.6	4.39	2.94	5.93	355
	7-8	62.0	56.3	34.3	60.2	22.6	41.7	27.4	9.59	19.1	7.03	16.34	6.91	364
	8-9	76.0	59.1	41.9	61.4	23.5	46.9	32.2	10.68	21.4	8.89	10.46	6.50	399
	9-10	57.4	50.3	34.9	55.7	21.7	41.6	25.0	7.61	18.1	6.89	4.57	8.64	332
mean		67.8	**56.3**	45.2	61.2	26.4	48.2	30.5	10.3	23.5	9.88	7.90	11.15	398
st.dv		67.8 ± 23.3	**14.6**	24.7	22.3	10.6	13.1	8.51	4.59	6.94	5.95	4.29	5.30	103

A 11	0-1	25.2	78.2	19.6	22.2	12.8	17.1	18.0	6.10	21.7	13.3	3.55	22.2	260
	1-2	28.3	93.7	18.3	24.7	11.6	15.9	14.4	6.56	20.5	14.7	4.20	19.3	272
	2-3	52.6	84.0	41.8	32.9	15.5	20.6	16.9	9.14	25.1	14.7	5.79	17.6	337
	3-4	28.9	65.6	29.4	41.2	15.6	19.5	14.5	7.80	21.8	13.9	3.71	17.7	280
	4-5	32.3	112.9	25.2	21.9	13.7	17.7	11.9	5.27	17.4	14.0	4.89	14.7	292
	5-6	21.2	96.3	27.2	21.3	14.5	18.8	15.0	10.3	22.5	14.2	3.06	16.4	281
	6-7	33.8	85.8	54.8	40.4	20.8	27.5	21.6	9.26	32.2	14.9	0.00	15.1	356
	7-8	27.4	158.1	30.2	25.5	14.7	19.9	15.5	7.10	21.9	13.6	1.89	15.5	351

	8-9	12.3	61.9	16.4	13.8	9.0	11.7	7.53	5.54	13.5	8.57	0.00	7.71	168
	9-10	15.8	80.2	12.1	10.1	6.34	9.24	8.54	3.82	8.51	9.15	2.01	11.3	177
mean		27.7	**91.7**	27.5	25.4	13.5	17.8	14.4	7.09	20.5	13.1	2.91	15.7	277
st.dv		11.1	**27.6**	12.8	10.2	3.94	4.99	4.24	2.03	206.42	2.30	1.94	4.06	64.8