#### 1 Hydrophobic organic contaminants are not linked to microplastic uptake in Baltic Sea herring

- 2 \*Ogonowski M.<sup>1,2</sup>, Wenman, V<sup>.1</sup>, Danielsson S<sup>.3</sup> and Gorokhova E.<sup>1</sup>
- 3
- <sup>1</sup> Department of Environmental Science and Analytical Chemistry, Stockholm University, SE-10691,
   Stockholm, Sweden
- <sup>6</sup> <sup>2</sup> AquaBiota Water Research, Löjtnantsgatan 25, SE-115 50, Stockholm, Sweden
- <sup>3</sup> Swedish Museum of Natural History, Department of Environmental Science and Monitoring, P. O. Box
   50 007, SE-104 05, Stockholm Sweden
- 9
- 10 \*corresponding author:
- 11 <u>martin.ogonowski@aces.su.se</u>
- 12 **Declarations of interest**:none
- 13

## 14 Abstract

Due to its ubiquity in the environment, microplastic debris (MP) is commonly ingested by a range 15 of aquatic organisms. A commonly held paradigm states that this can lead to lower food intake 16 17 and the bioaccumulation of organic contaminants carried by MP. However, causal links between 18 MP and contaminant levels in biota are poorly understood and *in situ* data are scarce. By examining 19 the stomach contents of herring (Clupea harengus membras) caught along the Swedish east coast 20 for the presence of plastic particles, and relating MP burden to hydrophobic organic contaminant 21 concentrations in the fish muscle, we found that MP were present in about one third of herring, although the degree of ingestion exhibited a large geographic variability. This result is in good 22 23 agreement with previous findings for Baltic Sea herring. If present, the mean number of MP 24 particles was 8.4 ind<sup>-1</sup>. MP burden normalized to gut content volume did not differ significantly between sea basins. The observed MP abundance was in agreement with values predicted by an 25 individual based model that had been parameterized for clupeids of a similar size and the ambient 26 27 MP abundances reported for surface waters in the Baltic Sea. MP abundance in the gastrointestinal 28 tract decreased with reproductive stage but increased with body size. Contaminant concentrations 29 in the muscle tissue were found to be unrelated to the number of MP present in gastrointestinal tract, suggesting a lack of correlation between the bioaccumulation of contaminants and MP 30 31 ingestion. Thus, despite their ubiquity, MP are unlikely to have a measurable impact on food intake 32 or the total body burden of environmental contaminants in Baltic herring.

33

34 **Capsule**: Ingested microplastic abundance is not correlated to hydrophobic organic contaminant 35 body burden in Baltic herring.

36

37 Keywords: Microplastic, Baltic Sea, herring, hydrophobic organic contaminants, marine monitoring

### 39 Introduction

40 Plastic debris, and especially microplastics (MP < 5 mm), can be found in a variety of aquatic

41 organisms across several trophic levels (Cole et al., 2013; Lusher et al., 2015, 2013). Due to the

42 importance of commercial fish and shellfish species for human consumption, the ingestion and

43 presence of MP in these organisms has become a matter of concern (EFSA Panel on Contaminants

44 in the Food Chain (CONTAM), 2016), and several studies have been aiming to provide a 45 quantitative assessment of MP ingestion in several fish species (Beer et al., 2017; Budimir et al.,

46 2018: Fockema et al. 2013: Lusher et al. 2013: Dummel et al. 2016)

- 46 2018; Foekema et al., 2013; Lusher et al., 2013; Rummel et al., 2016).
- 47 Several experimental studies have found a link between microplastic ingestion and decreased food
- intake (Cole et al., 2015; Ogonowski et al., 2018, 2016) and the bioaccumulation of hydrophobic
   organic chemicals (HOCs) (Besseling et al., 2013; Oliveira et al., 2012; Rochman et al., 2013b)
- 50 sorbed to the MP particles (Mato et al., 2001; Rochman et al., 2013a; Rusina et al., 2010).
- 51 However, other studies indicate that plastic polymers could have a net cleaning effect, relieving

52 the organisms of contaminants by acting as passive samplers (Gouin et al., 2011; Herzke et al.,

53 2016; Koelmans et al., 2016). Hence, the relative importance of microplastics as vectors for

54 contaminant transport remains unresolved, possibly also due to the lack of environmental data

55 linking HOC body burden to ingested MP.

56 Here, we study MP ingestion by Baltic Sea herring (Clupea harengus membras L.), a commercially relevant species. Being facultative pelagic filter-feeders (Huse and Toresen, 1996), 57 58 herring stand a high risk of MP ingestion. We used a modelling approach with reported parameters 59 on clupeid feeding, food processing and ambient MP concentrations to estimate realistic intervals 60 of MP ingestion in Baltic herring. The model was validated with *in situ* samples collected from 61 various locations along the Swedish coast, stretching about 1500km from the Bothnian Bay in the north to the Bornholm basin (Hanö Bight) in the south. Furthermore, by investigating the 62 63 relationship between consumed MP abundance and HOC concentrations in muscle tissue, we 64 assess whether MP significantly contribute to HOC body burden in Baltic Sea herring.

65

# 66 Materials and Methods

# 67 Fish collection and sample characteristics

The Baltic herring used for our analyses were collected along the Swedish east coast as a part of the Swedish National Monitoring Program for Contaminants in Marine Biota, an initiative by the Swedish Museum of Natural History (Stockholm, Sweden). In order to avoid possible bias by

71 known point sources, we randomly selected a total of 130 specimens that had been collected at

thirteen reference monitoring stations, thus providing a sufficiently large geographical area and a

representative range of HOC and MP levels for analysis (Figure 1).

74

- 75 The sex ratio in our sample was approximately 50:50 and uniform across sampling sites. The fish
- 76 were 3-7 years old, with an average total length of  $173 \pm 18$  mm. The reproductive phase was
- determined by gametocytic maturity and varied geographically. It ranged from immature to mature
- and also included post-spawned individuals. The entire gastrointestinal tract (GIT) was removed,
- 79 packed in aluminum foil, frozen at -20  $^{\circ}$ C, and stored until MP –analysis at the Department of
- 80 Environmental Science and Analytical Chemistry, Stockholm University, Sweden.

## 82 MP quantification in the gastrointestinal tract of fish

The GITs were individually placed in glass Petri dishes, dissected with surgical scissors, and rinsed 83 with deionized, particle-free water. Each sample was carefully examined under a stereo 84 85 microscope and any bolus items resembling plastic were extracted using stainless steel pincers and transferred to Eppendorf tubes for further analysis. To account for variable feeding rates and the 86 corresponding variability in the amount of ingested plastic, we standardized the plastic counts per 87 88 GIT using the gut fullness weighed by the mass of the fish, calculated as: gut fullness (%)  $\times$  body 89 weight (g wet weight); gut fullness was assessed using a semi-quantitative scale with five possible values: 0% (empty), 25%, 50%, 75% or 100% (full). Furthermore, stomach volume was assumed 90 91 proportional to body length (Pirhonen and Koskela, 2005).

92 Criteria for the visual identification of MP followed the recommendations of Norén (2007) and 93 Hidalgo-Ruz et al. (2012). A particle with diameter between 1-5 mm was classified as MP if all 94 the following criteria were met: (i) uniform, unnaturally bright or of an unnatural color, (ii) lack 95 of organic structures, and (iii) uniform diameter over the entire length of a fiber. Each putative 96 plastic particle was categorized according to its shape (fiber or fragment) and color. Particles with 97 diameters smaller than 1 mm were discarded while particles > 5 mm were recorded but not used 98 for further analyses. To test the accuracy of the visual identification method, a random subset of 99 20 samples containing MP (i.e., the gut contents of 20 individual fish) was analyzed using the hot

- 100 needle test (De Witte et al., 2014).
- 101
- 102 *Controls and blanks*

103 To prevent cross-contamination by airborne particles during sample examination, the dissections were performed under a Fumex local extractor; each sample being analyzed for 10 min. A Petri 104 dish filled with filtered deionized water was placed next to a test sample to serve as a blank for the 105 106 quantification and characterization of potential contamination during the analysis. A cotton lab coat and nitrile gloves were used at all times to minimize contamination by clothing. The type and 107 108 color of clothing were also recorded for each dissection event in order to back-trace potential 109 contamination. Samples that would display quantifiable amounts of background contamination 110 (plastic particles < 1 mm) were excluded.

- 111
- 112 Chemical analysis
- 113 Fish muscle samples for contaminant analysis were taken from the middle dorsal muscle layer
- and tissue sampling was performed according to standard procedures (TemaNord 1995). The
- samples were analyzed for polychlorinated biphenyls (PCB 28, 52, 101, 118, 138, 153 and 180),
- 116 organochlorine pesticides (DDE, DDD, DDT, HCB, AHCH, BHCH, and Lindane) and
- polybrominated flame retardants (BDE 28, 47, 99, 100, 153, 154 and HBCD), according to the
- 118 guidelines of the Swedish National Monitoring Program for Contaminants in Marine Biota. For
- 119 the analysis of most compounds, 10 g of muscle tissue from individual fish was used, whereas 1
- 120 g samples of muscle tissue from 10 individuals were pooled for a few analytically challenging
- 121 compounds. An overview of the analyzed contaminants and their average concentrations in
- 122 herring muscle tissue are provided in

123 Table 1, while details of the analytical procedures are given elsewhere (Bignert et al., 2016).

124

125 Data analysis and statistics

## 126 Relationships between biological factors, geography and ingested microplastic

Regional differences in microplastic abundance in the GIT (both at a station and basin level) were tested using Permanova (Anderson, 2001), while relationships between specific biological variables were tested using generalized additive models (GAM). Due to an overrepresentation of zeros in the data (overdispersion), the models were run using zero-inflated Poisson error structures. Model performance was assessed using residual plots. All analyses were performed in R 3.5.0 (R

132 Core Team, 2014).

133

134 Relationships between HOCs and ingested microplastic

Prior to statistical analyses, the measured values in samples whose chemical body burdens were below the limit of quantification (LOQ) were imputed by LOQ divided by the square root of two (Succop et al., 2004). The analyzed chemical concentrations were summed and grouped into their

respective contaminant groups (PCBs, PBDEs and organochlorine pesticides). A factor analysis

139 was performed to assess the degree of association between the chemical variables and microplastic

140 abundance in the GIT.

141

142 Modeling plastic ingestion by herring

To evaluate whether the observed number of MP in the GIT could be predicted using ambient MP abundance data and fish feeding rate, we modeled the ingestion of MP using literature-derived parameters on food uptake, egestion, and MP abundance in the study area. The two main assumptions in the model were: (i) non-selective feeding by fish on ambient MP (1 to 5 mm) and (ii) non-discriminatory gut evacuation, i.e., MP being egested at the same rate as prey remains.

Then, the amount of MP in the GIT at any given time, t, can be written as the mass balance between uptakeand loss rates (Eq. 1):

150

152

151 
$$MP_t = MP(t - dt) + (IR - Eg) dt,$$
 (1)

where *IR* and *Eg* are the ingestion and egestion rates (number of MP  $h^{-1}$ ), respectively. They can be written as:

$$156 IR = CMP \times FE \times CR (2)$$

157 158 and

159

 $\begin{array}{ll} 160 & Eg = GER \times MP_t \ , \\ 161 \end{array} \tag{3}$ 

- 162 where *CMP* is the ambient MP concentration (number of MP  $L^{-1}$ ), *FE* the effective feeding period
- 163 (unitless; proportion of time spent feeding per day), CR the clearance rate (L h<sup>-1</sup>; the volume of water
- 164 swept clear of particles per individual and hour, and *GER* the gut evacuation rate  $(h^{-1})$ .
- 165

## 166 Model parameterization

167 In the absence of published rates for adult Baltic herring, we used the gut evacuation and clearance rates

- 168 of European pilchard (Sardina pilchardus) (Table 2), which is a clupeid with a feeding ecology similar to
- Baltic herring (c.f. Costalago and Palomera, 2014; Möllmann et al., 2004). Since neither herring nor
- pilchard are particular night time feeders (Arrhenius and Hansson, 1994; Costalago and Palomera, 2014),
  the effective feeding period was set to the average number of daylight hours in Sundsvall, Sweden
- 1/1 the effective recomp period was set to the average number of daying nours in Sundsvall, Sweden 1/2 (62°23′28″ N, 17°18′22″ E) during August; as this represents both the latitudinal and temporal midpoints
- 172 (02 25 26 10, 17 16 22 E) during August, as this repres
- 174
- 175 To determine a plausible range of MP burden under environmentally relevant conditions, we
- modeled two extreme scenarios. In the first and the more conservative scenario, we used the lowest  $\frac{1}{2}$
- reported ambient MP abundance for the northern Baltic proper (0.19 MP m<sup>-3</sup>, Gewert *et al.* 2017)
- established from surface manta trawls (335  $\mu$ m mesh, MP size range = 0.335-5 mm) as well as the
- 179 lowest reported experimental clearance rates when using *Brachionus plicatilis* (~190  $\mu$ m) and 180 *Artemia salina* nauplii ( $\leq$  724  $\mu$ m) as a prey (Garrido et al., 2007). In the second scenario, we used
- 180 Artemia salina nauplii ( $\leq$  724 µm) as a prey (Garrido et al., 2007). In the second scenario, we used 181 the highest known ambient MP abundance of 1.13 MP m<sup>-3</sup> (Gewert et al., 2017) and the highest
- 182 clearance rates observed when feeding on low densities of fish eggs (Garrido et al., 2007).
- 183

# 184 **Results**

# 185 Observed microplastic concentrations in the GIT

186 Microplastic particles identified by visual inspection were found in 37 out of the 130 analyzed herring 187 (33.8%; range: 0 to 51 pieces of plastic fiber and/or fragments per individual). In those 37 individuals, the 188 mean abundance was 8.4 particles ind<sup>-1</sup>. The dominant type of MP were fibers of various colors (87.6%), 189 while fragments were less frequent (12.4%). When excluding black particles that had been identified as

- 190 non-plastic by the hot needle-test, the proportion of fibers decreased slightly (86.2%).
- 191 All procedural blanks contained plastic (mainly single fibers) of unknown origin. However, these particles 192 all were < 1 mm and did thus not measurably contribute to the MP counts used for statistical analysis. The
- 193 variation in total MP burden between stations and basins was high (Table 3), and we did not find any
- significant differences between the normalized abundances of MP from the different basins (*station* nested
- 195 within *basin* as a random factor, pseudo  $F_{4,117} = 1.06$ , p = 0.40).
- 196
- 197 Predicted microplastic concentrations in the GIT
- 198 The model scenario using the lowest reported ambient MP concentrations (Gewert et al., 2017) and
- 199 clearance rates (Garrido et al., 2007) yielded an average number of MP in the gastrointestinal tract of 0.01
- 200 particles ind<sup>-1</sup> for clupeid fish of sizes similar to those analyzed here. The highest MP abundance and CR
- 201 modelling scenario resulted in an average number of MP in the GIT of 0.88 particles ind<sup>-1</sup>.

### 203 Linkage between ingested plastic and HOCs

We found no relationship between the number of consumed MP and the concentration of any of the HOCs (Figure 2). Microplastic occurrence in GIT displayed a significant negative loading on the first axis and positive loading on the second. In contrast, the organochlorine pesticides and PBDEs loaded strongly only on the first axis, while the PCBs displayed some degree of positive loading on both axes. Hence, no contaminant group loaded in the same direction as MP abundance.

209

## 210 Biological factors related to MP occurrence in fish GIT

The amount of plastic found in the GIT was positively related to fish body weight for individuals > 50 g (GAM,  $\chi^2 = 306.7$ , p < 0.0001, Figure 3 A), but not in the smaller fish. In contrast, an inverse

relationship was found for individuals in their *reproductive phase*, where MP occurrence in the GIT was significantly lower in fish that had reached sexual maturity and recently spawned ( $\chi^2 =$ 

215 78.5, p < 0.0001, Figure 3 B). Both *Gut fullness* (Figure 3 C) and *Age* (Figure 3 D) displayed weak

216 bell-shaped relationships with MP occurrence. However, albeit significant (GAM,  $\chi^2 = 17.7$ , p <

217 0.0002,  $\chi^2 = 32.4$ , p < 0.001, *Gut fullness* and *Age*, respectively), these relationships were not

- 218 particularly strong and likely of low biological importance.
- 219

### 220 Discussion

## 221 Microplastics are only a small dietary component in planktivorous herring

Microplastics were found in about one third of the gastrointestinal tracts, with a high proportion 222 223 of fibers (88 %). While these values are in good agreement with those reported by Lusher et al. (2013) for the English Channel (36.5% of individuals containing MP, with 68% being fibers) and 224 Beer et al. (2017) for the central Baltic Sea (20% containing MP, with 93% fibers), other studies 225 found considerably lower MP prevalence and fiber contributions. Both Foekema et al. (2013) and 226 Rummel et al. (2016) found plastics in only 2% of herring samples from the North Sea and the 227 Southern Baltic Sea, with fibers accounting for less than 10% of MP. Having excluded fibers from 228 229 their analyses, Budimir et al. (2018) reported a frequency of occurrence as low as 1.8% in herring from the northern Baltic Sea. These discrepancies between different studies could be related to 230 differences in fish size and gut fullness. For example, Foekema et al. (2013) used fish that were 231 232 considerably larger (>200 mm total length) which most likely already had switched from filter 233 feeding to raptorial feeding on larger prey (Huse and Toresen, 1996). This change in feeding behavior would result in a lower ingestion rate of zooplankton-sized plastic particles and thus 234 235 result in lower overall MP burden. In the study by Rummel et al. (2016), many fish stomachs were empty, which probably was related to arrested feeding related to spawning, and possibly, stress-236 induced gut evacuation caused by the fish sampling (Vinson and Angradi, 2011; Wilkins, 1967). 237 238 This lends further support to our findings that MP burden increases with fish size and decreases 239 with reproductive phase. In addition, one would expect the amount of ingested MP to scale with gut bolus size or gut fullness. However, our findings do not support this assumption. This could 240 241 be due to one of our underlying model assumptions (see methods), namely that the rate of plastic egestion is equal to the egestion of natural food. If the egestion of plastic in herring occurs at a 242 slower rate, as observed with plastic fibers in amphipods (Au et al., 2015), one would observe a 243 higher rate of accumulation of MP in the GIT. While normalizing the ingested MP content by gut 244 fullness seems to have little impact on the absolute amount of ingested particles (as indicated by 245 the associated weak bell-shaped response curve), fish size appears be a better candidate for 246

standardizing internalized MP concentrations. Nevertheless, there are several other factors that
must be considered. They include ontogenetic changes in feeding modes, behavior, maturity levels,
fishing methods, and time of capture (diurnal differences in feeding activity).

250

# 251 No correlation between MP presence and HOCs

The so-called "Trojan horse" effect (Cole et al., 2011) denotes a transfer of hydrophobic 252 contaminants from ingested plastics to biota. While this effect has been demonstrated under 253 254 laboratory conditions (Batel et al., 2016; Besseling et al., 2013; Browne et al., 2013; Rochman et al., 2013b), recent modelling studies seem to indicate that compared to MP, natural sources play a 255 much more important role in explaining HOC bioaccumulation patterns in aquatic organisms 256 257 (Koelmans, 2015; Koelmans et al., 2016). Since we did not find any correlation between HOC concentrations in herring muscle tissue and MP abundance in the GIT, it could be argued that 258 259 omitting MP with diameters < 1 mm from our analyses, could have biased the results by ignoring 260 the potential influence a higher total surface area and thus higher HOC desorption rates (Hartmann et al., 2017; Hendriks et al., 2001). However, this is rather unlikely since filter feeding herring 261 have a relatively low capacity to retain smaller particles due to their rather wide gill raker spacing 262 (Gibson, 1988). In fact, this line of reasoning has been confirmed by several other studies who 263 found a predominant retention of MP with diameters > 1 mm (Beer et al., 2017; Lenz et al., 2016). 264 Given the short residence time (Grigorakis et al., 2017) of internalized plastics and the slow 265 desorption kinetics of many HOCs, we would moreover only expect a rather weak correlation 266 267 between the two, or no correlation at all.

268

Although causality is difficult to prove using environmental samples where many different 269 parameters contribute to the contaminant body burden of an organism (Hartmann et al., 2017), our 270 271 findings strongly suggest that there is no tenable relationship between the amount of ingested 272 microplastic particles and tissue contaminant concentrations (Fig. 2). This findings contrasts the 273 currently held paradigm according to which microplastics are an important source of HOCs in 274 aquatic organisms (Mato et al., 2001; Rochman et al., 2013b). Similarly, no correlation has been 275 found between the amount of ingested plastic and HOC concentrations in northern fulmars (Fulmarus glacialis) from the Norwegian coast (Herzke et al., 2016), although the birds had 276 277 ingested much larger amounts of plastic and their gut passage times for plastic particles are several orders of magnitude longer than for herring (Ryan, 2015). In light of present and past findings 278 279 regarding the role of MP as vectors for contaminant transport, our study further strengthens the 280 view that MP only play a minor or even negligible role in explaining contaminant bioaccumulation 281 in aquatic biota.

- 282
- 283 Conclusions

Our findings suggest that microplastic ingestion by filter-feeding fish is relatively low, even in a semi-enclosed sea like the Baltic, where the MP loading is expectedly high. A rather large discrepancy exists in the literature with regard to the interpretation of MP ingestion demonstrating the urgent need to find suitable standards for MP quantification in gut content analyses. Several studies also demonstrated that biological factors such as fish size and reproductive state may affect both feeding in general and selectivity towards MP, and hence MP burden. We found no correlation whatsoever between the amount of ingested plastic and muscle tissue HOC concentrations. These results provide an additional challenge to the commonly held paradigm that
 MP are important vectors for the transfer of contaminants to biota.

293

#### 294 Acknowledgements

295 This work was supported through the Joint Programming Initiative Healthy and Productive Seas and Oceans (JPI-Oceans) WEATHER-MIC project by the Swedish Research Council for 296 Environment, Agricultural Sciences and Spatial Planning (FORMAS) [grant number 942-2015-297 298 1866], FORMAS project; ir-PLAST [grant number 2015-932] and through the joint Baltic Sea 299 research and development programme (BONUS) MICROPOLL project by the Swedish Innovation Agency VINNOVA [grant number 2017-00979]. The study was designed, analyzed 300 301 and compiled solely at the responsibility of the authors without the involvement of the funding 302 agencies. We would also like to thank XpertScientific, Barcelona, Spain for proof reading the 303 manuscript.

#### 305 **References**

- Anderson, M.J., 2001. A new method for non-parametric multivariate analysis of variance. Austral Ecol.
   26, 32–46. https://doi.org/10.1111/j.1442-9993.2001.01070.pp.x
- Arrhenius, F., Hansson, S., 1994. In situ food consumption by young-of-the-year Baltic Sea herring
   Clupea harengus: a test of predictions from a bioenergetics model. Mar. Ecol. Prog. Ser. 110,
   145–149.
- Au, S.Y., Bruce, T.F., Bridges, W.C., Klaine, S.J., 2015. Responses of Hyalella azteca to acute and
   chronic microplastic exposures: Effects of Microplastic Exposure on Hyalella azteca. Environ.
   Toxicol. Chem. 34, 2564–2572. https://doi.org/10.1002/etc.3093
- Batel, A., Linti, F., Scherer, M., Erdinger, L., Braunbeck, T., 2016. Transfer of benzo[a]pyrene from
   microplastics to Artemia nauplii and further to zebrafish via a trophic food web experiment:
   CYP1A induction and visual tracking of persistent organic pollutants. Environ. Toxicol. Chem.
   35, 1656–1666. https://doi.org/10.1002/etc.3361
- Beer, S., Garm, A., Huwer, B., Dierking, J., Nielsen, T.G., 2017. No increase in marine microplastic
   concentration over the last three decades A case study from the Baltic Sea. Sci. Total Environ.
   https://doi.org/10.1016/j.scitotenv.2017.10.101
- Besseling, E., Wegner, A., Foekema, E.M., van den Heuvel-Greve, M.J., Koelmans, A.A., 2013. Effects
   of Microplastic on Fitness and PCB Bioaccumulation by the Lugworm Arenicola marina (L.).
   Environ. Sci. Technol. 47, 593–600. https://doi.org/10.1021/es302763x
- Bignert, A., Danielsson, S., Faxneld, S., Nyberg, E., 2016. Comments Concerning the National Swedish
   Contaminant Monitoring Programme in Marine Biota (No. 5:2016). Stockholm, Sweden.
- Browne, M.A., Niven, S.J., Galloway, T.S., Rowland, S.J., Thompson, R.C., 2013. Microplastic Moves
   Pollutants and Additives to Worms, Reducing Functions Linked to Health and Biodiversity. Curr.
   Biol. 23, 2388–2392. https://doi.org/10.1016/j.cub.2013.10.012
- Budimir, S., Setälä, O., Lehtiniemi, M., 2018. Effective and easy to use extraction method shows low
   numbers of microplastics in offshore planktivorous fish from the northern Baltic Sea. Mar. Pollut.
   Bull. 127, 586–592. https://doi.org/10.1016/j.marpolbul.2017.12.054
- Cole, M., Lindeque, P., Fileman, E., Halsband, C., Galloway, T.S., 2015. The Impact of Polystyrene
   Microplastics on Feeding, Function and Fecundity in the Marine Copepod Calanus helgolandicus.
   Environ. Sci. Technol. 49, 1130–1137. https://doi.org/10.1021/es504525u
- Cole, M., Lindeque, P., Fileman, E., Halsband, C., Goodhead, R., Moger, J., Galloway, T.S., 2013.
   Microplastic Ingestion by Zooplankton. Environ. Sci. Technol. 47, 6646–6655.
   https://doi.org/10.1021/es400663f
- Cole, M., Lindeque, P., Halsband, C., Galloway, T.S., 2011. Microplastics as contaminants in the marine
   environment: A review. Mar. Pollut. Bull. 62, 2588–2597.
   https://doi.org/10.1016/j.marpolbul.2011.09.025
- Costalago, D., Palomera, I., 2014. Feeding of European pilchard (Sardina pilchardus) in the northwestern
   Mediterranean: from late larvae to adults. Sci. Mar. 78, 41–54.
   https://doi.org/10.3989/scimar.03898.06D
- De Witte, B., Devriese, L., Bekaert, K., Hoffman, S., Vandermeersch, G., Cooreman, K., Robbens, J.,
   2014. Quality assessment of the blue mussel (Mytilus edulis): Comparison between commercial and wild types. Mar. Pollut. Bull. 85, 146–155. https://doi.org/10.1016/j.marpolbul.2014.06.006

- EFSA Panel on Contaminants in the Food Chain (CONTAM), 2016. Presence of microplastics and
   nanoplastics in food, with particular focus on seafood. EFSA J. 14, n/a-n/a.
   https://doi.org/10.2903/j.efsa.2016.4501
- Foekema, E.M., De Gruijter, C., Mergia, M.T., van Franeker, J.A., Murk, A.J., Koelmans, A.A., 2013.
  Plastic in North Sea Fish. Environ. Sci. Technol. 47, 8818–8824.
  https://doi.org/10.1021/es400931b
- Garrido, S., Marçalo, A., Zwolinski, J., Lingen, C.D. van der, 2007. Laboratory investigations on the
   effect of prey size and concentration on the feeding behaviour of Sardina pilchardus. Mar. Ecol.
   Prog. Ser. 330, 189–199. https://doi.org/10.3354/meps330189
- Gewert, B., Ogonowski, M., Barth, A., MacLeod, M., 2017. Abundance and composition of near surface
   microplastics and plastic debris in the Stockholm Archipelago, Baltic Sea. Mar. Pollut. Bull. 120,
   292–302. https://doi.org/10.1016/j.marpolbul.2017.04.062
- Gibson, R.N., 1988. Development, morphometry and particle retention capability of the gill rakers in the
   herring, Clupea harengus L. J. Fish Biol. 32, 949–962. https://doi.org/10.1111/j.1095 8649.1988.tb05438.x
- Gouin, T., Roche, N., Lohmann, R., Hodges, G., 2011. A Thermodynamic Approach for Assessing the
   Environmental Exposure of Chemicals Absorbed to Microplastic. Environ. Sci. Technol. 45,
   1466–1472. https://doi.org/10.1021/es1032025
- Grigorakis, S., Mason, S.A., Drouillard, K.G., 2017. Determination of the gut retention of plastic
   microbeads and microfibers in goldfish (Carassius auratus). Chemosphere 169, 233–238.
   https://doi.org/10.1016/j.chemosphere.2016.11.055
- Hartmann, N.B., Rist, S., Bodin, J., Jensen, L.H., Schmidt, S.N., Mayer, P., Meibom, A., Baun, A., 2017.
   Microplastics as vectors for environmental contaminants: Exploring sorption, desorption, and
   transfer to biota. Integr. Environ. Assess. Manag. 13, 488–493. https://doi.org/10.1002/ieam.1904
- Hendriks, A.J., van der Linde, A., Cornelissen, G., Sijm, D.T.H.M., 2001. The power of size. 1. Rate
  constants and equilibrium ratios for accumulation of organic substances related to octanol-water
  partition ratio and species weight. Environ. Toxicol. Chem. 20, 1399–1420.
  https://doi.org/10.1002/etc.5620200703
- Herzke, D., Anker-Nilssen, T., Nøst, T.H., Götsch, A., Christensen-Dalsgaard, S., Langset, M., Fangel,
   K., Koelmans, A.A., 2016. Negligible Impact of Ingested Microplastics on Tissue Concentrations
   of Persistent Organic Pollutants in Northern Fulmars off Coastal Norway. Environ. Sci. Technol.
   50, 1924–1933. https://doi.org/10.1021/acs.est.5b04663
- Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the Marine Environment:
   A Review of the Methods Used for Identification and Quantification. Environ. Sci. Technol. 46,
   3060–3075. https://doi.org/10.1021/es2031505
- Huse, G., Toresen, R., 1996. A comparative study of the feeding habits of herring (clupea harengus,
   clupeidae, 1.) and capelin (mallotus villosus, osmeridae, müller) in the barents sea. Sarsia 81,
   143–153. https://doi.org/10.1080/00364827.1996.10413618
- Koelmans, A.A., 2015. Modeling the Role of Microplastics in Bioaccumulation of Organic Chemicals to
   Marine Aquatic Organisms. A Critical Review, in: Bergmann, M., Gutow, L., Klages, M. (Eds.),
   Marine Anthropogenic Litter. Springer International Publishing, Cham, pp. 309–324.
- Koelmans, A.A., Bakir, A., Burton, G.A., Janssen, C.R., 2016. Microplastic as a Vector for Chemicals in
   the Aquatic Environment: Critical Review and Model-Supported Reinterpretation of Empirical
   Studies. Environ. Sci. Technol. 3315–3326. https://doi.org/10.1021/acs.est.5b06069

- Lenz, R., Enders, K., Beer, S., Kirk Sørensen, T., Stedmon, C.A., 2016. Analysis of microplastic in the
   stomachs of herring and cod from the North Sea and Baltic Sea. DTU Aqua National Institute of
   Aquatic Resources.
- Lusher, A.L., Hernandez-Milian, G., O'Brien, J., Berrow, S., O'Connor, I., Officer, R., 2015.
   Microplastic and macroplastic ingestion by a deep diving, oceanic cetacean: The True's beaked
   whale Mesoplodon mirus. Environ. Pollut. 199, 185–191.
   https://doi.org/10.1016/j.envpol.2015.01.023
- Lusher, A.L., McHugh, M., Thompson, R.C., 2013. Occurrence of microplastics in the gastrointestinal
   tract of pelagic and demersal fish from the English Channel. Mar. Pollut. Bull. 67, 94–99.
   https://doi.org/10.1016/j.marpolbul.2012.11.028
- Mato, Y., Isobe, T., Takada, H., Kanehiro, H., Ohtake, C., Kaminuma, T., 2001. Plastic Resin Pellets as a
   Transport Medium for Toxic Chemicals in the Marine Environment. Environ. Sci. Technol. 35,
   318–324. https://doi.org/10.1021/es0010498
- Möllmann, C., Kornilovs, G., Fetter, M., Koster, F.W., 2004. Feeding ecology of central Baltic Sea
   herring and sprat. J. Fish Biol. 65, 1563–1581.
- 406 Norén, F., 2007. Small plastic particles in Coastal Swedish waters. KIMO Swed.
- 407 Ogonowski, M., Gerdes, Z., Gorokhova, E., 2018. What we know and what we think we know about
   408 microplastic effects A critical perspective. Curr. Opin. Environ. Sci. Health 1, 41–46.
   409 https://doi.org/10.1016/j.coesh.2017.09.001
- 410 Ogonowski, M., Schür, C., Jarsén, Å., Gorokhova, E., 2016. The Effects of Natural and Anthropogenic
   411 Microparticles on Individual Fitness in Daphnia magna. PLOS ONE 11, e0155063.
   412 https://doi.org/10.1371/journal.pone.0155063
- Oliveira, M., Ribeiro, A., Guilhermino, L., 2012. Effects of short-term exposure to microplastics and
  pyrene on Pomatoschistus microps (Teleostei, Gobiidae). Comp. Biochem. Physiol. A. Mol.
  Integr. Physiol. 163, Supplement, S20. https://doi.org/10.1016/j.cbpa.2012.05.063
- 416 Pirhonen, J., Koskela, J., 2005. Indirect estimation of stomach volume of rainbow trout Oncorhynchus
   417 mykiss (Walbaum). Aquac. Res. 36, 851–856. https://doi.org/10.1111/j.1365-2109.2005.01293.x
- R Core Team, 2014. R: A language and environment for statistical computing. R Foundation for
   Statistical Computing, Vienna, Austria.
- Rochman, C.M., Hoh, E., Hentschel, B.T., Kaye, S., 2013a. Long-Term Field Measurement of Sorption
   of Organic Contaminants to Five Types of Plastic Pellets: Implications for Plastic Marine Debris.
   Environ. Sci. Technol. 47, 1646–1654. https://doi.org/10.1021/es303700s
- Rochman, C.M., Hoh, E., Kurobe, T., Teh, S.J., 2013b. Ingested plastic transfers hazardous chemicals to
   fish and induces hepatic stress. Sci. Rep. 3. https://doi.org/10.1038/srep03263
- Rummel, C.D., Löder, M.G.J., Fricke, N.F., Lang, T., Griebeler, E.-M., Janke, M., Gerdts, G., 2016.
  Plastic ingestion by pelagic and demersal fish from the North Sea and Baltic Sea. Mar. Pollut.
  Bull. 102, 134–141. https://doi.org/10.1016/j.marpolbul.2015.11.043
- Rusina, T.P., Smedes, F., Klanova, J., 2010. Diffusion coefficients of polychlorinated biphenyls and
   polycyclic aromatic hydrocarbons in polydimethylsiloxane and low-density polyethylene
   polymers. J. Appl. Polym. Sci. 116, 1803–1810. https://doi.org/10.1002/app.31704
- 431 Ryan, P.G., 2015. How quickly do albatrosses and petrels digest plastic particles? Environ. Pollut. 207,
  432 438–440. https://doi.org/10.1016/j.envpol.2015.08.005

- Succop, P.A., Clark, S., Chen, M., Galke, W., 2004. Imputation of data values that are less than a
  detection limit. J. Occup. Environ. Hyg. 1, 436–441. https://doi.org/10.1080/15459620490462797
- Vinson, M.R., Angradi, T.R., 2011. Stomach Emptiness in Fishes: Sources of Variation and Study Design
   Implications. Rev. Fish. Sci. 19, 63–73. https://doi.org/10.1080/10641262.2010.536856
- Wilkins, N.P., 1967. Starvation of the herring, Clupea harengus L.: survival and some gross biochemical
   changes. Comp. Biochem. Physiol. 23, 503–518.

#### 440 Figures and tables

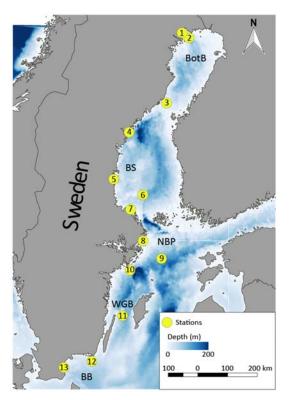
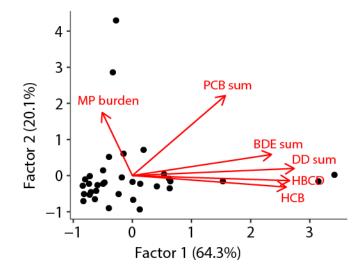




Figure 1. Sampling sites within the Swedish National Monitoring Program for Contaminants in Marine
Biota included in this study, BotB Bothnian Bay, BS Bothnian Sea, NBP Northern Baltic Proper, WGB
Western Gotland Basin and BB Bornholm Basin. 1 Rånefjärden, 2 Harufjärden, 3 Holmöarna, 4
Gaviksfjärden, 5 Långvindsfjärden, 6 Bothnian Sea offshore site, 7 Ängsskärsklubb, 8 Lagnö, 9 Baltic
proper offshore site, 10 Landsort, 11 Byxelkrok, 12 Utlängan and 13 Western Hanö bight.

448



449

450 **Figure 2.** Factor scores (axes) and loadings (arrows) of contaminants (HBCD, HCB and the sum

451 of PCBs, BDEs and DDs) and average plastic content (*MP burden*).

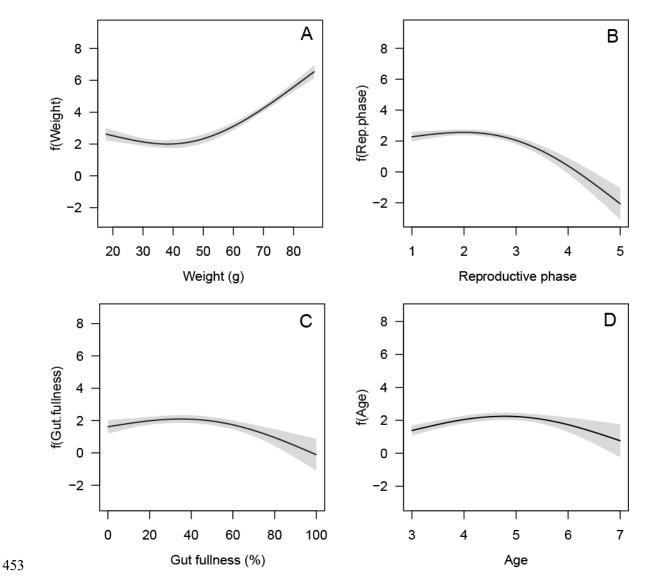


Figure 3. Generalized additive model partial response curves for the measured biological variables: *body weight* (A), *reproductive phase* (B), *gut fullness* (C) and *age* (D). The classes for *reproductive phase* correspond to: 1 = immature, 2 = developing gonads, 3 = mature gonads, 4 = post-spawned and 5 = deformed gonads. The vertical axes, indicate the relative influence of each explanatory variable on the prediction on the base of partial residuals. Grey bands indicate 95% confidence intervals for each curve.

Chemical group	Chamical anaziaa	Abbreviation	Mean concentration in fish muscle	S.D	Median	Min	Max
Chemical group Polychlorinated biphenyls (PCBs)	Chemical species 2,4,4'-PCB	PCB 28	<u>(µg/g)</u> 0.0039	0.0019	0.0033	0.0018	0.0106
Folychionnated biphenyls (FCBS)	2,4,4 -FCB 2,2',5,5'-PCB	PCB 20 PCB 52	0.0039	0.0019	0.0057	0.0018	0.0100
	2,2',4,5,5'-PCB	PCB 32 PCB 101	0.0007	0.0044	0.0037	0.0023	0.0799
		PCB 101 PCB 118					
	2,3',4,4',5'-PCB		0.0204	0.0135	0.0155	0.0062	0.0694
	2,2',3,4,4',5'-PCB	PCB 138	0.0591	0.0408	0.0444	0.0152	0.1896
	2,2',4,4',5,5'-PCB	PCB 153	0.0417	0.0280	0.0345	0.0120	
	2,2',3,4,4',5,5'-PCB	PCB 180	0.0181	0.0114	0.0149	0.0029	0.0551
Organochlorine pesticides	4,4'-DDT	DDT	0.0205	0.0218	0.0134	0.0037	0.0966
	4,4'-DDE	DDE	0.1055	0.0960	0.0815	0.0160	0.4130
	4,4'-DDD	DDD	0.0284	0.0333	0.0163	0.0016	0.1391
	α-1,2,3,4,5,6-Hexachlorocyclohexane	AHCH	0.0029	0.0005	0.0030	0.0018	0.0039
	β-1,2,3,4,5,6-Hexachlorocyclohexane	BHCH	0.0056	0.0027	0.0057	0.0018	0.0099
Brominated flame retardants	γ-1,2,3,4,5,6-Hexachlorocyclohexane	Lindane	0.0029	0.0005	0.0030	0.0018	0.0039
(BDEs)	2,4,4'-TriBDE	BDE 28	0.0002	0.0001	0.0002	0.0001	0.0005
	2,2',4,4'-TetraBDE	BDE 47	0.0051	0.0032	0.0041	0.0016	0.0160
	2,2',4,4',5-PentaBDE	BDE 99	0.0012	0.0009	0.0009	0.0005	0.0044
	2,2',4,4',6-PentaBDE	BDE 100	0.0012	0.0007	0.0011	0.0004	0.0035
	2,2',4,4',5,5'-HexaBDE	BDE 153	0.0002	0.0002	0.0002	0.0001	0.0009
	2,2',4,4',5,6'-HexaBDE 1,2,5,6,9,10-	BDE 154	0.0006	0.0004	0.0005	0.0002	0.0018
	Hexabromocyclododecane	HBCD	0.0114	0.0096	0.0088	0.0025	0.0470
Other	Hexachlorobenzene	HCB	0.0289	0.0209	0.0246	0.0114	0.1013

# 459 Table 1. Overview of the chemicals analyzed in herring muscle tissue and their average concentrations (μg g<sup>-1</sup> fish muscle).

Parameter	Unit	Average/estimate	Min	Max	Species	Meaning	Reference
CMP	MP L <sup>-1</sup>		0.00019	0.00113		MP concentration in water column.	Gewert et al. 2017
CR	L ind. <sup>-1</sup> h <sup>-</sup>		21.6	304.2	Sardina pilchardus	Clearance rate.	Garrido et al. 2007
FE	unitless	0.625			Clupea harengus	Feeding efficiency.	Batty et al. 1986
GER	h <sup>-1</sup>	0.259	-0.215ª	0.733	Sardina pilchardus	Gut evacuation rate.	Costalago & Palomera 2014
IR	MP h <sup>-1</sup>					Ingestion, amount MP ingested at time <i>t</i> .	
MP	MP					Number of MP in fish stomach at time <i>t</i> .	
——Eg	MP h <sup>-1</sup>					Amount MP egested at time <i>t</i> .	

## 461 **Table 2.** Parameters used to model microplastic ingestion in Baltic Sea herring.

<sup>a</sup> The minimum estimate for GER is the reported lower 95% confidence limit of the mean. A negative gut evacuation rate is not biologically meaningful, therfore the mean value was used as the minimum rate in the model.

**Table 3.** Descriptive statistics for the microplastics recovered from the gastrointestinal tract of Baltic Sea herring. The data are presented as particle shape and color as well as total MP burden (mean of presences) and range (min-max) Values are given per basin and ordered north to south.

	Fiber			Fragment				Black fibers incl.					Black fibers excl.					
Basin	Black	Red	Brown	Green	Clear	Black	Red	Green	Prevalence (%)	Median MP content	Mean MP content	Mean MP content of presences	Range (min- max)	Prevalence (%)	Median MP content	Mean MP content	Mean MP content of presences	Range (min- max)
Bothnian Bay	0-7	0-38	0-4	0-0	0-3	0-0	0-3	0-12	31.8	0	4.1	9.5	0-38	35.1	0	3.5	8.2	0-38
Bothnian Sea	0-8	0-18	0-0	0-0	0-5	0-7	0-0	0-2	27.3	0	1.3	5.2	0-18	24.3	0	1.0	4.3	0-18
Northern Baltic Proper	0-1	0-0	0-0	0-0	0-51	0-0	0-2	0-2	13.6	0	6.7	32.8	0-51	10.8	0	6.6	32.7	0-51
Western Gotland Basin	0-1	0-3	0-0	0-1	0-1	0-0	0-7	0-0	18.2	0	1.0	2.7	0-7	18.9	0	0.9	2.6	0-7
Bornholm Basin	0-1	0-13	0-0	0-0	0-2	0-0	0-0	0-0	9.1	0	0.9	4.5	0-13	10.8	0	0.9	4.3	0-13
Total	0-8	0-38	0-4	0-1	0-51	0-7	0-7	0-12	33.8	0	2.7	9.1	0-51	28.5	0	2.4	8.4	0-51