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Fish life-history traits are affected after chronic dietary exposure to an environmentally realistic marine mixture of PCBs and PBDEs

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Abstract :

Polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) are persistent organic pollutants that have been shown to affect fish life-history traits such as reproductive success, growth and survival. At the individual level, their toxicity and underlying mechanisms of action have been studied through experimental exposure. However, the number of experimental studies approaching marine environmental situations is scarce, i.e., in most cases, individuals are exposed to either single congeners, or single types of molecules, or high concentrations, so that results can hardly be transposed to natural populations. In the present study, we evaluated the effect of chronic dietary exposure to an environmentally realistic marine mixture of PCB and PBDE congeners on zebrafish life-history traits from larval to adult stage. Exposure was conducted through diet from the first meal and throughout the life cycle of the fish. The mixture was composed so as to approach environmentally relevant marine conditions in terms of both congener composition and concentrations. Life-history traits of exposed fish were compared to those of control individuals using several replicate populations in each treatment. We found evidence of slower body growth, but to a larger asymptotic length, and delayed spawning probability in exposed fish. In addition, offspring issued from early spawning events of exposed fish exhibited a lower larval survival under starvation condition. Given their strong dependency on life-history traits, marine fish population dynamics and associated fisheries productivity

for commercial species could be affected by such individual-level effects of PCBs and PBDEs on somatic growth, spawning probability and larval survival.

Graphical abstract

Highlights

► Effects of realistic marine PCB/PBDE mixtures on fish life-history are unknown. ► Zebrafish were chronically exposed to a realistic marine PCB/PBDE mixture via diet. ► Exposed fish grew to larger sizes but their spawning probability was delayed. ► Larval survival of offspring from early spawns was decreased under starvation. ► Environmental PCB/PBDE mixture can alter fish population dynamics via life-history.

Keywords : Contaminants, Body length, Condition, Fertilization rate, Energy allocation, Trade-off

1 Introduction

Persistent organic pollutants (POPs) gather a wide number of chemicals which are of great concern because of their persistence, bioaccumulation and toxicity. In addition, given their propensity for long-range transport, they are globally distributed in various environments worldwide including some far from source areas (Bogdal et al., 2013; Corsolini, 2009; Rigét et al., 2016). Among POPs, polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) are two families made of 209 congeners differing by the number and position of one to ten substitution by chlorine and bromine, respectively. PCBs have been used since the 1930s for various industrial purposes, such as dielectric fluids in electrical capacitors, transformers and hydraulic systems (United Nations Environment Programme, 1999), while PBDEs have been used since the 1970s as flame retardants in plastics, furniture, upholstery, electrical equipment, electronic devices, textiles and other household products (United Nations Environment Programme, 2012).

PCBs have been progressively banned in various countries since the 1970s whereas PBDEs have been banned or restricted more recently, beginning in the early 2000s. These regulations were endorsed internationally by the Stockholm Convention on POPs (United Nations Environment Programme, 2001). Today, only the commercial production of deca-PBDE is allowed, although with some restrictions in Europe. Although a decrease in their levels has been reported in biota from various locations (Byer et al., 2015; Rigét et al., 2016) and despite these restrictions, PCBs and PBDEs are still present in all environmental compartments worldwide, including aquatic ecosystems. Therefore they still represent a potential environmental concern.

PCBs and PBDEs are found in the marine environment as complex mixtures of numerous congeners. Due to their long-term persistence and elevated lipophilicity (Mizukawa et al., 2009), they are significantly bioaccumulated and biomagnified through trophic transfer, in most biotic compartments of marine ecosystems (e.g. mollusks, fish, seals; Couderc et al., 2015; Johansson et al., 2006; Letcher et al., 2009). Such bioaccumulation could be a threat for animal communities and their population dynamics through the scaling up of their individual-level effects to the population level (Vasseur and Cossu-Leguille, 2006). Notably, some scientists suspected that chemicals have contributed to the decline of some wild marine fish populations (Hamilton et al., 2015) and it has been suggested that the productivity of some marine fish stocks could be altered due to recruitment impairment caused by nursery habitat degradation in relation to pollutant accumulation (Gilliers et al., 2006; Riou et al., 2001; Rochette et al., 2010). Given the strong dependency of population dynamics on life-history (De Roos et al., 2003; Stearns, 1992), potential individual-level effects of PCBs and PBDEs on fish life-history traits could indeed affect fish population recruitment and dynamics and associated fisheries productivity for commercial species (Vasseur and Cossu-Leguille, 2006).

It is difficult, however, to ascertain the relationship between the presence of one class of chemical and its effects on biota from field observations, because of the accumulation of multiple potential stresses, including many families of chemicals, in natural environments (Baillon et al., 2016). In contrast, the experimental approach allows controlling for potential confounding effects and establishing such links without ambiguity. The effects and the underlying mechanisms of action of PCBs and PBDEs at the individual level have thus been intensively studied through experimental exposure, notably in fish. These studies have demonstrated an alteration of behavior, growth, reproductive, hepatic, and renal functions as well as of the immune and the endocrine systems in fish (Berg et al., 2011; Daouk et al., 2011; Han et al., 2011, 2013, Lyche et al., 2010, 2011; Muirhead et al., 2006; Péan et al.,

2013; Yu et al., 2015 and references therein). In particular, several studies have demonstrated that exposure of fish to either PCB or PBDE congener mixtures can affect fish life-history traits such as reproductive success, growth and survival. For example, it has been reported that long-term dietary exposure of zebrafish to a PCB mixture led to a decrease in the number of eggs per spawn and in their fertilization rate (Daouk et al., 2011). Furthermore, dietary exposure of fathead minnows to a single PBDE congener (BDE-47) was shown to reduce cumulative egg production (Muirhead et al., 2006), and McCarthy et al. (2003) showed that Atlantic croaker larvae originating from parents exposed to PCBs technical mixture (Aroclor 1254) through diet were characterized by diminished growth. In addition to their effects on reproductive success and growth, these compounds have been shown to increase fish early-life-stage mortality. Indeed, it has been reported by Foekema et al. (2014) that exposure of common sole eggs to a mixture of POPs (including PCBs and PBDEs) via the water caused acute mortality in larvae after hatching. However, although experimental studies provide valuable information on the potential effects of PCBs and PBDEs, few have focused on environmental situations (see Berg et al., 2011; Lyche et al., 2010, 2011 for experiments mimicking freshwater lake environmental situations). In most cases, the exposure conditions are indeed quite different from environmental situations because of the use of either single congeners, or single types of molecules (i.e., PCBs, or PBDEs, or PAHs), or high concentrations and results can thus hardly be transposed to natural populations. How fish life-history traits may be affected by lifelong exposure to mixtures of both PCB and PBDE congeners that are realistic for the environment therefore remains largely unknown. More precisely, questions about the effects of environmentally realistic mixtures on growth, reproduction and survival and their consequences on individual fitness and population dynamics are still pending.

In this study, we used the zebrafish model to explore the life-history effects associated with long-term dietary exposure to a mixture of PCBs and PBDEs. Due to their lipophilicity, these compounds are mostly found associated to organic matter and not in the water, so that dietary exposure is considered the major route of exposure to PCBs and PBDEs for vertebrates (Muir et al., 2003; Nyman et al., 2002). The selected congeners were chosen to approach environmentally representative conditions for marine ecosystems in terms of both concentrations and compositions. For PCBs, the mixture corresponded to the profile and concentrations found in mussels from an estuary highly-impacted by industrial and urban activities, the Seine estuary in France (Abarnou et al., 2000), which is a nursery area for many flatfish species (Riou et al., 2001). Benthic invertebrates such as mussels are indeed an important food source for many exploited fish species, notably flatfish and demersal fish. As PBDEs profiles in living organisms depend upon their metabolism (Christensen et al., 2002; Ma et al., 2013; Stapleton et al., 2004; Voorspoels et al., 2003), the mixture for this type of molecules was not defined based on specific measurements, but corresponded to a mixture at environmental concentration of the most representative congeners in marine biota and the main congener in marine sediments that were identified for priority action by OSPAR (OSPAR Commission, 2009, 2013).

These PCBs and PBDEs mixtures representative of environment situations were used for identifying life-history effects potentially transposable to wild marine fish populations with possible consequences for their population dynamics and productivity. Effects on early survival, growth, reproduction and larval survival in the progeny were specifically investigated.

2 Materials and methods

This study was conducted under the approval of the Animal Care Committee of Poitou-Charentes # 84 COMETHEA (France) under project authorization number CE2012-23.

2.1 Fish rearing

Experiments were performed with wild type strain TU zebrafish (ZFIN ID: ZDBGENO-990623-3) from a stock kept at the Fish Ecophysiology Platform (PEP - http://wwz.ifremer.fr/pep_eng) originating from the European Zebrafish Resources Center (EZRC, Karlsruhe, Germany). Fish were maintained in heterosexual groups (n = 28 ± 5 individuals per 10 L-tank) under a 14 h day/10 h night light cycle. The resulting rearing density fell within the recommended range of 1 to 5 individuals/L (Nüsslein-Volhard and Dahm, 2002; Singleman and Holtzman, 2014). Water used in the rearing system was a mixture of approximately 2/3 reverse osmosis water and 1/3 tap water, both being initially treated by sediment and charcoal filters. Water physicochemical parameters were remained constant during the experiment: temperature 27 ± 1 °C, conductivity 300 ± 50 µS cm^{-1} and pH 7.5 ± 0.5.

To produce the F0 generation, eggs were obtained by random pairwise mating of zebrafish placed together in spawning boxes the evening before collection (AquaSchwarz, Germany). Eggs from each clutch with a fertilization rate > 80% were collected the next morning in a Petri dish containing 30 mL of isotonic mixture E3 (1 L: 17.2 g NaCl, 0.76 g HCl, 2.9 g $CaCl_2$. 2 H_2O, 4.9 MgSO4. 7 H_2O) and placed at 28 °C. Twenty four hours post-fertilization (hpf), eggs from 5 clutches were mixed in a balanced way (taking the same number of eggs from each clutch) and distributed in 20 Petri dishes at a rate of 60 larvae per Petri dish. The 20 groups of 60 larvae were considered together as one replicate population. At 5 days post fertilization (dpf), the groups of 60 larvae were transferred from their Petri dishes to separate

1-L tanks. At 15 dpf, the groups of larvae were transferred to tubes inserted inside separate 10-L rearing tanks disposed on flow-through racks and were then freed into the tanks at 27 dpf (after Vignet et al., 2014). In the flow-through racks, an hourly automated addition of 150 mL of system water resulted in a daily total renewal of one third of the volume of each tank. Discarded water was collected and treated with activated charcoal before being discharged into sewers. Mesh bags filled with zeolite stones (~30 cL) were also added in each tank to guarantee water quality. Tanks were inspected daily and cleaned by siphoning if necessary. Furthermore, tanks were fully emptied and cleaned, together with zeolite bags, monthly from the first biometry at 2 or 3 months age onwards. With this rearing protocol, concentrations of ammonia, nitrites and nitrates measured were always below critical values for zebrafish (Lawrence, 2007) and even below quantification level in most cases.

Fish were fed three times per day, twice with pellets in the morning and the evening, and once with freshly hatched crustaceans (*Artemia salina*) at noon. Food pellet size was adapted to fish's mouth size using their age as a proxy. From 5 to 60 dpf, fish were fed sequentially with 100, 200 and 300 µm SDS (Special Diet Service; Dietex international, United Kingdom) with 5 to 10 days of overlap; from 60 to 70 dpf, fish were fed with a mixture of 300 µm SDS and Inicio^{+} 500 µm (Biomar, France), and from 70 dpf onwards, they were fed with Inicio^{+} 500 µm only.

2.2 Fish exposure

Fish were exposed to contaminants through food pellets spiked with a mixture of PCB and PBDE congeners following the food-pellet size depending on age sequence presented above. The spiking procedure was similar for all pellet sizes except for vessel and solution volumes that were of course adapted to the quantity of food to be spiked. PCB and PBDE congeners

188 used in contaminated diet and their concentrations were chosen in order to represent
189 environmental conditions.

190 For PCBs, the choice was based on the contamination levels and profiles reported in mussels
191 from the Seine estuary, one of the most contaminated site along the French coastlines
192 (Abarnou et al., 2000). This choice was justified by the fact that many exploited fish species,
193 especially flatfish (e.g. sole, plaice, turbot) and demersal fish (e.g. cod, haddock, seabass),
194 feed largely on benthic invertebrates and/or have nursery grounds in industrialized estuaries.
195 More precisely, contaminated food was spiked with a mixture of 22 PCB congeners, i.e.,
196 congeners CB-8, CB-18, CB-28, CB-31, CB-44, CB-49, CB-52, CB-77, CB-101, CB-105,
197 CB-110, CB-118, CB-128, CB-132, CB-138, CB-149, CB-153, CB-156, CB-170, CB-180,
198 CB-187 and CB-194 at targeted concentrations between 28 and 280 ng g^{-1} ww (wet weight)
199 per congener. The 22 congeners used covered a wide range of chlorinated substitutions (2-8)
200 and a range of hydrophobicity (log K_{ow} - octanol/water partition coefficient) from 5.07 to 7.80
201 (Hawker and Connell, 1988).

202 For PBDEs, targeted contamination levels and profiles were based on the 6 most
203 representative congeners in marine biota and the main congener in marine sediments that were
204 identified for priority action by OSPAR (OSPAR Commission, 2009, 2013). The rationale for
205 this choice is that PBDE congeners are metabolized more easily and quickly than PCB
206 congeners in low trophic level marine organisms (Grimm et al., 2015; Zhang et al., 2016). As
207 a result, the PBDE congener profiles found in an organism's tissue will depend on the species
208 considered as PBDE metabolism varies between species. It follows that we did not want to
209 define the target profile of PBDE congeners based on measurements on a particular species.
210 More specifically, contaminated food was also spiked with a mixture of 7 PBDE congeners,
211 i.e., congeners BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-183 for marine biota
212 and congener BDE-209 for marine sediments at targeted concentrations of between 10 and

213 200 ng g^{-1} ww per congener. The reason why BDE-209 was included is that, despite the fact it
214 is not listed among the main congeners in marine biota in general as it disappears at high
215 trophic levels (Burd et al., 2014) and in pelagic biota (Desforges et al., 2014), it is the main
216 PBDE congener found in marine sediments and thus one of the main in benthic invertebrates
217 (together with BDE-47, BDE-99 and BDE-100; Burd et al., 2014; Dinn et al., 2012). As
218 explained previously, many exploited fish are benthic or demersal species feeding largely on
219 benthic invertebrates, so that it was sensible to include BDE-209 in the mixture tested. The 7
220 congeners used covered a wide range of brominated substitutions (3 - 10) and a range of
221 hydrophobicity (log K_{ow}) from 6.7 to 12.1 (Kelly et al., 2008).

222 The precise targeted and measured concentrations for each PCB and PBDE congener are
223 available in Table S1. The contaminated diet was prepared after dilution of a stock PCB and
224 PBDE solution in isooctane solvent for incorporation as described in Daouk et al. (2011). The
225 control diet was prepared in the same manner, i.e., with isooctane solvent but without addition
226 of POPs. Control and contaminated diets were prepared with food of all sizes.

227 Fish were fed from their first meal (5 dpf) with either a control diet or a contaminated diet
228 using the feeding schedule described earlier. A total of 5 replicate populations, each
229 composed of 9 ± 1 tanks per dietary treatment (46 tanks per treatment in total, see Table S3
230 for details), were used in this study to increase the significance and confidence level of the
231 experimental results. Hereafter, we will refer to fish exposed to the control and the
232 contaminated diet as SOLV (as solvent) and MIX (as mixture) fish, respectively.

233 **2.3 PCB and PBDE analyses**

234 PCBs and PBDEs were analyzed in all batches of MIX (n = 12) and SOLV diets (n = 16), and
235 in MIX (PCBs: $n_{females} = n_{males} = 3$; PBDEs: $n_{females} = n_{males} = 3$) and SOLV fish (PCBs: $n_{females}$
236 $= n_{males} = 1$; PBDEs: $n_{females} = n_{males} = 3$) at 180 dpf using the methods described in Daouk et

al. (2011) and Munschy et al. (2011), respectively. Briefly, PCBs were extracted using a hot Soxhlet apparatus (Soxtec), purified sequentially with concentrated sulfuric acid and by adsorption chromatography on Florisil column, and analyzed using gas chromatography equipped with an electron capture detector (GC-µECD). PBDEs were analyzed using Accelerated Solvent Extraction (ASE, Dionex Corp., USA) followed by gel permeation chromatography, silica and alumina column, and concentrated sulfuric acid treatment prior to quantification by gas chromatography coupled to mass spectrometry (GC-MS) in electron capture negative ionisation mode (ECNI). Analyses on fish were done on entire individuals.

2.4 Quality assurance/quality control

2.4.1 PCB analysis

The calibration of the system was performed within a large range using a multi-point (6) calibration curve to define the linearity range of our detector (ECD) for all contaminants. The relative precision of the method was checked for this type of samples by the analysis of 6 aliquots of a homogeneous preparation of fish (laboratory control card). The results showed coefficients of variation of < 10 % for all congeners, which indicates a satisfactory reproducibility of the method. Detailed information is provided in Supplementary Text S2.

2.4.2 PBDE analysis

Quality Assurance / quality Control procedures were implemented for each batch of 8 to 10 samples, including procedural blanks, use of recovery surrogates in all samples, analysis of certified reference material and participation to inter-comparison exercises. Detailed information can be found in Munschy et al. (2011) and an update is provided in Supplementary Text S2.

2.5 Trait monitoring

The following description covers trait monitoring of all replicates at once while some details (number of tanks, precise date of biometries, etc.) may vary between replicates. Such details are given in supplementary Table S3 and corresponding variation is indicated as standard deviation or minimum and maximum values in the main text below.

2.5.1 Survival

Early survival of individuals was followed by counting daily the number of live fish in each tank of each replicate from 5 dpf to the juvenile stage (30 ± 6 dpf, mean ± SD; see date for each replicate in Table S3) for all replicates.

Juvenile/adult survival was monitored monthly from 30 ± 6 dpf to 183±4 dpf for all replicates on the occasion of growth monitoring (see below for growth monitoring and Table S3 for precise dates of biometries). As individuals were sampled from various tanks within each replicate during this period for additional analyses but without keeping the information of the tanks sampled, data had to be aggregated at the replicate level.

2.5.2 Growth

Growth of all fish was monitored monthly from 65 ± 10 dpf to between 181 and 362 dpf depending on the replicate (see dates for each replicate in Table S3). On each occasion, individual standard length (mm) and body weight (g) were measured after anesthesia with benzocaine (Vignet et al., 2014). Sex was determined whenever possible based on morphological clues, generally from around 3 months old onwards. All replicates were used for this monitoring.

2.5.3 Reproduction test

After 72 ± 11 dpf, reproduction was monitored for 17 ± 7 days by placing two spawning boxes into each rearing tank (see dates for each replicate in Table S3). Eggs were collected the next morning and sorted to count the total number of eggs as well as the number of fertilized ones. The fertilization rate was then calculated for each spawning event. The total number of spawning events obtained relative to solicitation number (equal to the number of inspected rearing tanks per treatment) was calculated for each treatment and replicate. For each tank, sex-ratio s, mean individual food consumption c, number of individuals N, length of females l_f, and length of males l_m were measured and the age a of fish was recorded. In this test, the length of both males and females from each tank was measured on day 30 ± 18 (depending on the replicate) only to avoid repetitive stress during the test. Reproduction monitoring was very time-consuming and due to manpower limitations only a selection of replicates (1, 2 and 4, 28 tanks in total per treatment) was used for this assessment.

2.5.4 Larval survival in the progeny

A survival experiment was performed on unfed F1 larvae of replicate 4. The purpose of this experiment was to determine whether the exposure of parents (F0) to PCB and PBDE congeners could affect the survival of their offspring larvae (F1).

Survival of unfed larvae was monitored on two occasions corresponding to the 1st and 10th spawning events of F0 fish. On each occasion, survival was studied on 3 different clutches from 10 rearing tanks per dietary treatment. Thirty eggs were collected from each clutch and kept in a Petri dish in 30 mL of E3 medium at 28°C until the end of the survival experiment.

In total, the survival of 900 larvae per treatment and spawning event (1^{st} and 10^{th}) was monitored (n = 30 eggs × 3 clutches × 10 tanks × 2 treatments × 2 dates = 3600 larvae).

2.6 Statistical analyses

2.6.1 Modeling approach

Statistical analyses were performed in R version 3.2.2 (R Development Core Team, 2015). All traits were analyzed using mixed-effects models. Random effects were used to account for variability due to rearing tanks and/or replication depending on the trait considered: two nested random effects, namely replicates and tanks nested within replicates (replicate/tank), were included in models describing F0 generation traits, except for juvenile/adult survival for which only replicates were included due to data aggregation, and clutches nested within tanks were included in the model describing unfed F1 larvae survival. Random effects affected either the model intercept only or both model intercept and slopes. For each trait, the fixed part of the model included a dietary treatment effect (T, MIX versus SOLV diet) to assess the effect of fish exposure to the mixture of PCBs and PBDEs plus relevant covariates likely to biologically affect the considered trait (see description below for each trait as well as Table S4 for details on fixed and random effects).

For each trait, the full model was reduced by backward selection in two steps: the random part of the model was reduced first and the fixed part was selected afterwards (Pinheiro and Bates, 2000; Zuur et al., 2009). Selection was based on significance of the effects at a 5% alpha risk threshold determined by likelihood ratio tests (LRT) between nested models while respecting the marginality of the effects. Such LRTs are supposed to follow a χ^2 distribution under the null hypothesis (type II tests; Fox and Weisberg, 2011). Diagnostics based on residuals were used to assess the adequacy of the reduced model and compliance to the underlying

assumptions. Variables were transformed whenever necessary to ensure that the residuals followed the assumed error distribution (Table S4). Finally, the effects of treatment T and other active covariates were estimated from the reduced models and their significance was tested by LRT. When the reduced models did not include the treatment effect T, the test provided for this effect corresponds to the LRT test during the selection procedure. Only the fixed part of the selected models will be presented in the Results section. Random effects are mere technical parameters included to account for variation due to tanks and replication and thus to obtain robust estimates and significance tests of fixed effects against such variation. Therefore, detailed information on the random part is given in Table S5 in Supplementary Material.

In the following sections, we describe how the fixed part of the full model was defined for each trait.

2.6.2 Survival

Time to death at early and juvenile/adult stages was modeled using survival analysis (Therneau and Grambsch, 2000) as the data analyzed here are right-censored data because some individuals were still alive at intermediate sampling dates (individuals sampled for additional analyses) and at the end of the study. More precisely, two mixed-effects Cox proportional hazards models (COXME) were used to estimate how the hazard rate, i.e., the death rate, was affected by the contamination treatment T as a fixed effect (Table S4) during early and juvenile/adult stages. Cox models allow the death rate to be modeled as being affected proportionally (i.e., multiplicatively) by the explanatory variables. In practice, a logarithmic link function is used to allow for a linear predictor of the death rate. The COXME models were implemented using the *coxme* package in R (Therneau, 2015).

2.6.3 Growth

Fish growth was modeled according to the Von Bertalanffy growth (VBG) function using a non-linear mixed-effects model (NLME; Pinheiro and Bates, 2000). The VBG equation describes fish length $l(a)$ (mm) according to individual age a (days) as

$$l(a) = l_\infty - (l_\infty - l_0)e^{-k(a-a_0)}$$

where l_∞ is the asymptotic standard length (mm), l_0 is the initial standard length (mm) at age a_0 (days), a_0 is the age at the first biometric measurement and k is the growth rate coefficient (day^{-1}). To ensure well-behaved residuals of the fitted model, the VBG function to the power of 3 was fitted to length data to the power of 3. This is because the VBG function was initially developed to describe growth in mass and that individual mass is roughly proportional to individual length to the power of 3 (Von Bertalanffy, 1938).

Nested within the VBG function, the three parameters of this model, l_0, l_∞, and k were themselves modeled as depending linearly on fixed effects, namely treatment T only for l_0, and treatment T, individual's sex S and their interaction for l_∞ and k, as well as on random effects (replicate/tank; Table S4). l_0 was not modeled as dependent on sex as no sexual length dimorphism was observed at age a_0. The NLME model was implemented using the *nlme* package in R (Pinheiro et al., 2016).

2.6.4 Condition

Fish condition, defined as length-specific weight, was modeled via the length-weight allometry $W = al^b$ linearized by log-transformation (Froese, 2006):

368 $$\log W = \log a + b \log l$$

369 where W is wet weight (g), l is standard length (mm), $\log a$ is the regression intercept and b
370 is the regression slope. A linear mixed-effects model (LME) was used to analyze how
371 treatment T, individual's sex S and their interaction taken as fixed effects modified this
372 relationship (Table S4). The LME model was fitted using the *nlme* package in R (Pinheiro et
373 al., 2016).

374 ***2.6.5 Spawning probability***

375 A generalized linear mixed-effects model (GLMM) with binomial error distribution and logit
376 link function was used to analyze the effect of PCBs and PBDEs exposure on spawning
377 probability based on the number of spawning events relative to solicitation number in
378 reproduction tests (i.e., the number of trials to obtain a spawn). Fixed effects included
379 treatment T, but also age a to assess how spawning probability increased with age, plus their
380 interaction, as well as the mean length of females $\bar{l}_f$, the mean length of males $\bar{l}_m$, and the
381 mean individual food consumption c in the rearing tank as covariates (Table S4). In this
382 analysis, all continuous explanatory variables were standardized, i.e., centered and scaled to
383 unit variance. The GLMM model was fitted using the *lme4* package in R (Bates et al., 2015).

384 ***2.6.6 Number of eggs***

385 To test whether PCB and PBDE congener mixtures affect reproductive output, the number of
386 eggs produced by each rearing tank during reproduction tests was modeled using an LME
387 model. The fixed effects included in the full-model were treatment T, age a (as clutch size
388 often increases with age in fish), their interaction, plus the mean individual food consumption
389 c, the number of individuals N, and the sex-ratio s in the rearing tank as covariates (Table
390 S4). A Box-Cox transformation was applied to the response variable (eggs number) to obtain

a normal distribution of the residuals and all continuous explanatory variables were standardized. The LME model was fitted using the *nlme* package in R (Pinheiro et al., 2016).

2.6.7 Fertilization rate

Fertilization rate, obtained as the number of fertilized eggs relative to the total number of eggs in each rearing tank during reproduction tests was analyzed in the same way as spawning probability (see description above).

2.6.8 Larval survival in the progeny

As for early survival of the F0 generation, survival of unfed F1 larvae was analyzed using a COXME model. In this case, data were uncensored data as all individuals were dead at the end of the experiment. The fixed effects included treatment T, occasion O (1^{st} and 10^{th} F0 spawning event) and their interaction.

3 Results

3.1 PCB and PBDE concentrations in diet and fish

PCB and PBDE concentrations in MIX and SOLV diets are presented in Table S1. In MIX diet (n = 12), the total concentrations, i.e., summed across congeners, were 1932.3 ± 90.4 ng g^{-1} ww for PCBs and 479.8 ± 50.8 ng g^{-1} ww for PBDEs. In SOLV diet (n = 16), the total concentrations were circa 245 and 522 times lower, i.e., 7.9 ± 3.5 ng g^{-1} ww for PCBs and 0.92 ± 0.36 ng g^{-1} ww for PBDEs.

In MIX fish, the total concentrations of PCBs (n = 3) and PBDEs (n = 3) at 180 dpf in entire individuals were respectively 2188.3 ± 132.26 and 110.9 ± 1.14 ng g^{-1} ww in females, and 2140 ± 73.95 and 96.4 ± 8.65 ng g^{-1} ww in males. In SOLV fish, the total concentrations of

412 PCBs (n = 1) and PBDEs (n = 3) at 180 dpf were respectively 26.2 and 1.1 ± 0.31 ng g^{-1} ww
413 in females, and 26.7 and 2.7 ± 0.54 ng g^{-1} ww in males.

414 **3.2 Survival**

415 Early survival probabilities at the end of the survival experiment (30 ± 6 dpf) ranged from
416 47% to 89% and from 51% to 87% in MIX and SOLV fish, respectively (Kaplan-Meier
417 curve; Fig. 1). Juvenile/adult survival probabilities at 183 ± 4 dpf ranged from 44% to 73%
418 and from 38% to 70% in MIX and SOLV fish, respectively (Kaplan-Meier curve; Fig. 2). No
419 significant effect of the treatment on both early and juvenile/adult death rate was detected (*T*
420 effect Table 1; Fig. 1; Fig. 2).
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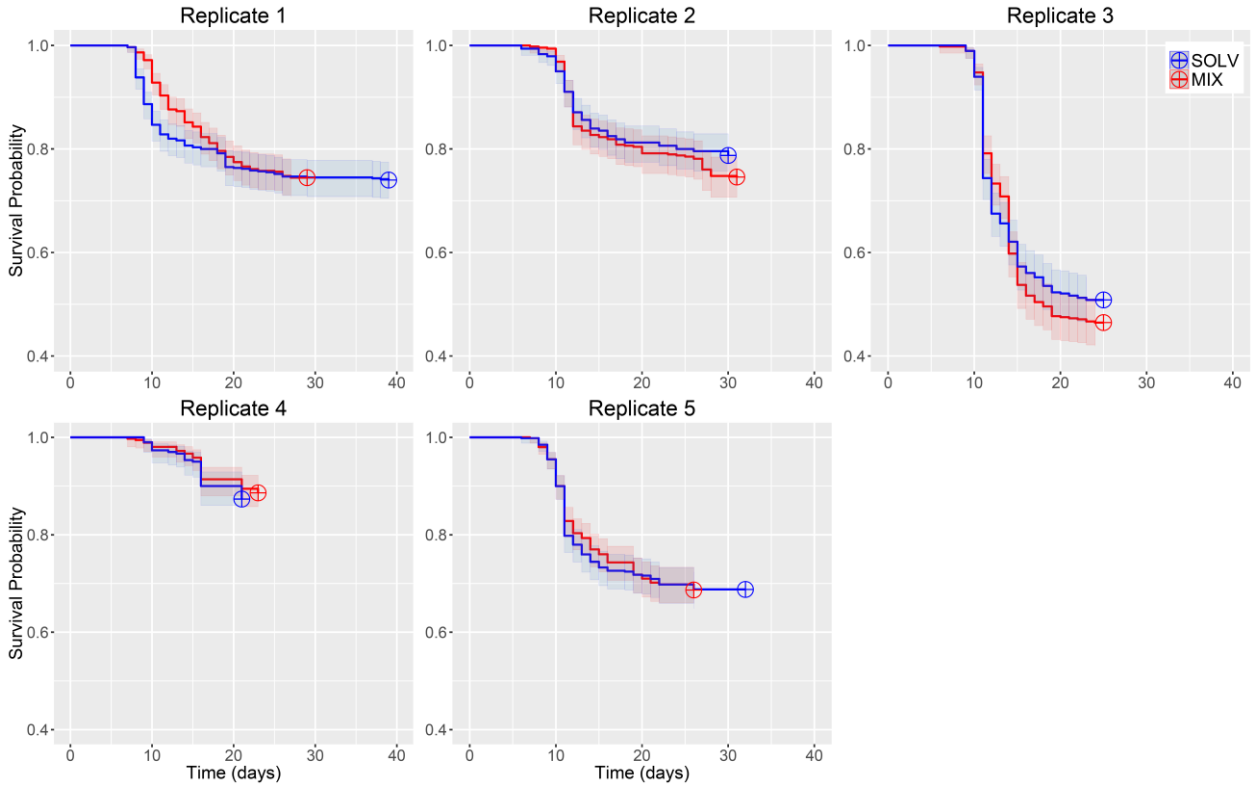
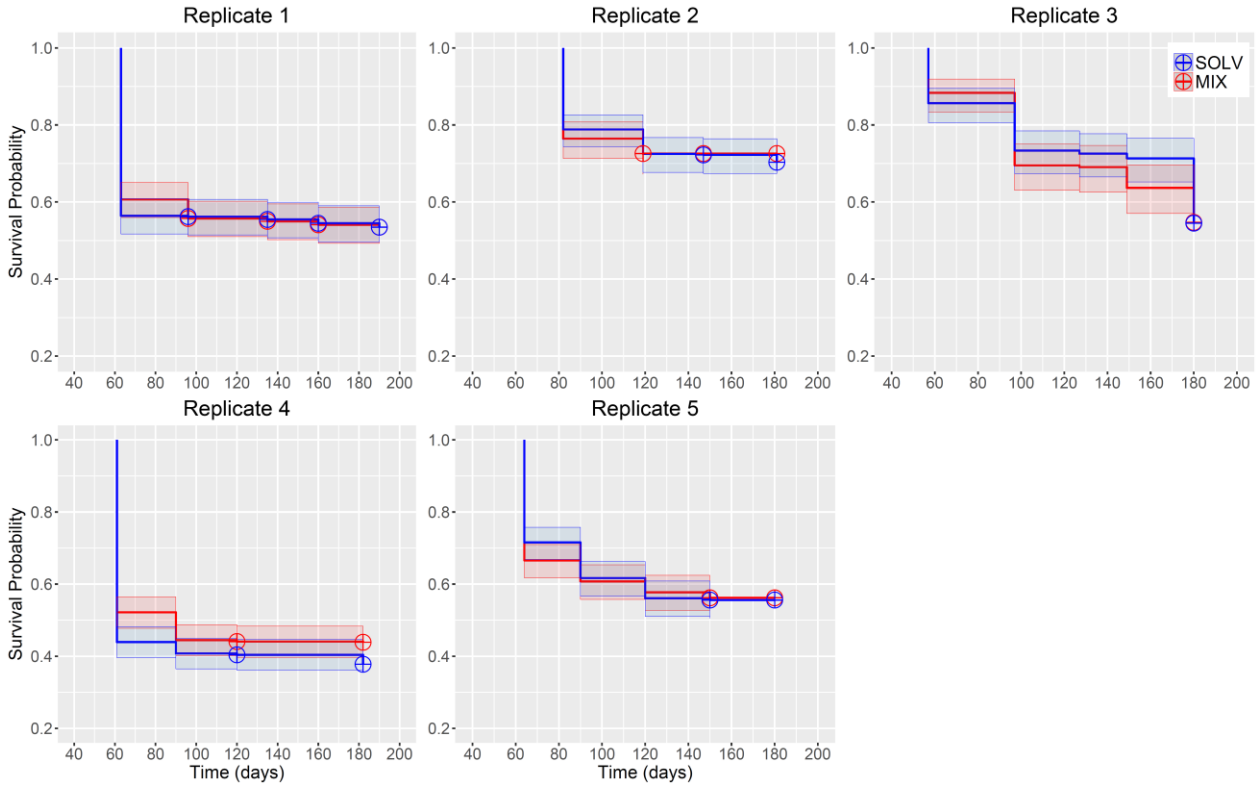


Fig. 1. Effect of dietary exposure to a PCB and PBDE congeners mixture on the early death rate of zebrafish. Curves represent the estimated decrease in individuals' survival probability with time by Kaplan-Meier estimator for MIX (red) and SOLV (blue) treatment, respectively, and shaded areas are the corresponding confidence intervals.



436

437 **Fig. 2.** Effect of dietary exposure to a PCB and PBDE congeners mixture on the juvenile and adult death rate of
438 zebrafish. Curves represent the estimated decrease in individuals' survival probability with time by Kaplan-
439 Meier estimator for MIX (red) and SOLV (blue) treatment, respectively, and shaded areas are the corresponding
440 confidence intervals.
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Table 1. Results of mixed-effects models testing for the effects of dietary treatment (MIX vs. SOLV) and other explanatory variables on the life-history traits of zebrafish. For categorical variables, effects are estimated for one category (indicated between parentheses) relative to the reference one taken as the intercept, namely MIX relative to SOLV for treatment T, and males M relative to females F for sex S.

Function	*Trait*	*Effect*	*Estimate*	*s.e.*	*df*	*χ2*	*p-value*
Survival	Early death rate	baseline hazard (SOLV)	n.a.	n.a.	n.a.	n.a.	n.a.
		T (MIX)	0.034	0.072	1	0.23	0.631
Survival	Juvenile/adult death rate	baseline hazard (SOLV)	n.a.	n.a.	n.a.	n.a.	n.a.
		T (MIX)	- 0.087	0.048	1	3.29	0.070
Growth	l_0	intercept (SOLV)	2.946	0.024	n.a.	n.a.	n.a.
		T (MIX)	0.019	0.016	1	1.97	0.160
Growth	k	intercept (F/SOLV)	- 4.317	0.119	n.a.	n.a.	n.a.
		S (M)	0.112	0.027	1	19.56	**<0.001**
		T (MIX)	- 0.141	0.047	1	6.40	**0.011**
Growth	l_∞	intercept (F/SOLV)	3.527	0.005	n.a.	n.a.	n.a.
		S (M)	- 0.067	0.005	1	171.52	**<0.001**
		T (MIX)	0.024	0.006	1	11.35	**<0.001**
Condition	W	Intercept (F/SOLV)	- 11.573	0.040	n.a.	n.a.	n.a.
		$\log(l)$	3.247	0.008	1	29057.02	**<0.001**
		S (M)	- 0.165	0.003	1	2219.74	**<0.001**
		T (MIX)	- 0.010	0.008	1	1.84	0.175
Reproduction	Spawning probability	intercept (SOLV)	0.829	0.238	n.a.	n.a.	n.a.
		a	2.033	0.248	1	127.21	**<0.001**
		$\bar{l}_f$	- 0.746	0.228	1	10.70	**0.001**
		$\bar{l}_m$	0.600	0.211	1	8.06	**0.004**
		T (MIX)	- 0.464	0.287	1	1.30	0.253
		$T \times a$	- 0.924	0.275	1	11.30	**<0.001**
Reproduction	Number of eggs	intercept (SOLV)	10.022	0.294	n.a.	n.a.	n.a.
		a	1.409	0.166	1	72.21	**<0.001**
		N	- 0.646	0.193	1	11.29	**<0.001**
		T (MIX)	- 0.486	0.403	1	1.46	0.227
Reproduction	Fertilization rate	intercept (SOLV)	- 0.175	1.246	n.a.	n.a.	n.a.
		a	0.978	0.478	1	4.19	**<0.041**
		T (MIX)	0.041	0.346	1	0.014	0.906

p-values < 0.05 are in bold.

3.3 Growth

Growth data revealed sexual length dimorphism, females being larger than males, and suggested that MIX fish grew to larger lengths than SOLV fish (Fig. 3). These observations were confirmed by the VBG model. Males grew at a faster rate k than females but to smaller asymptotic length l_∞ (S effect on both parameters, Table 1). More importantly, the VBG model showed that MIX fish grew at slower rate k (- 13.00 % for MIX individuals relative to

SOLV ones; T effect on parameter k, Table 1) but to larger asymptotic length l_∞ than SOLV fish (+ 2.40 % for MIX individuals relative to SOLV ones; T effect on l_∞, Table 1) and that these effects were independent of sex (no significant $T \times S$ interaction was found and it was therefore not kept in the model). In contrast, no significant effect of the treatment was observed on initial length l_0 (T effect, Table 1), which is the length at first biometric measurement.

3.4 Condition

The length-weight relationship differed significantly across sexes (main S effect, Table 1; Fig. 4), males being lighter than females at equivalent length (- 15.23 % of weight difference at 16, 24 and 32 mm for males). In contrast, there was no significant difference between MIX and SOLV fish, indicating that exposure had no effect on fish condition (main T effect, Table 1; Fig. 4). Moreover, the absence of interactions between $\log(l)$ and both S and T in the reduced model indicated that the shape of the allometric relationship between length and weight, determined by the exponent $b = 3.247$ ($\log(l)$ effect, Table 1), was unaffected by sex or treatment (Fig. 4).

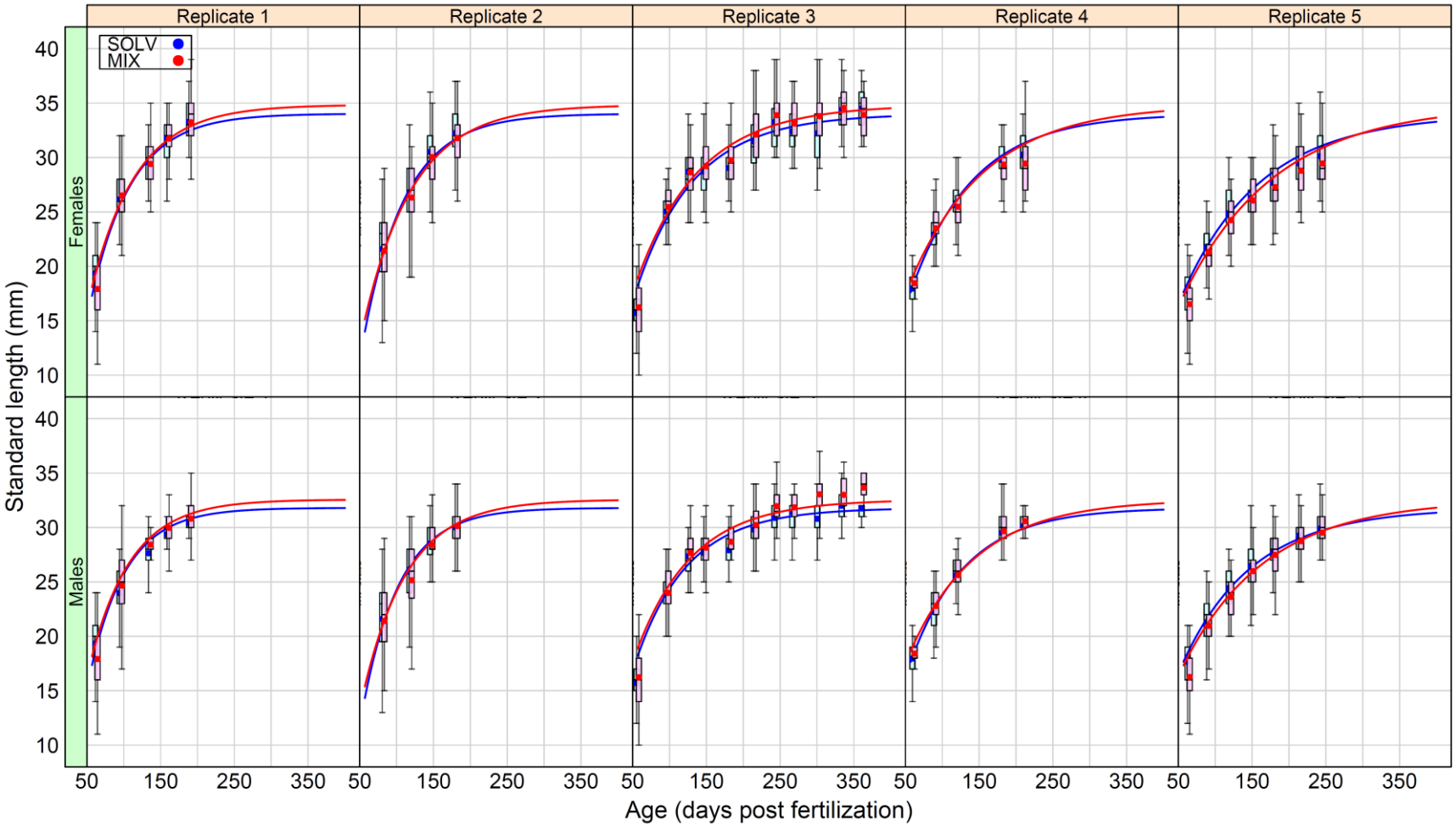


Fig. 3. Effect of dietary exposure to a PCB and PBDE congeners mixture on growth (in terms of standard length) in zebrafish. Rows correspond to the sex of fish and columns to the replicates. Boxplots represent observations per treatment (MIX in red and SOLV in blue) and curves represent the fitted model per treatment. For boxplots, the bottom and top of the box are the first and the third quartiles of the data distribution, the horizontal segment and the dot inside the box are the median and the mean, respectively, whiskers represent the most extreme data point within the 1.5 interquartile range.

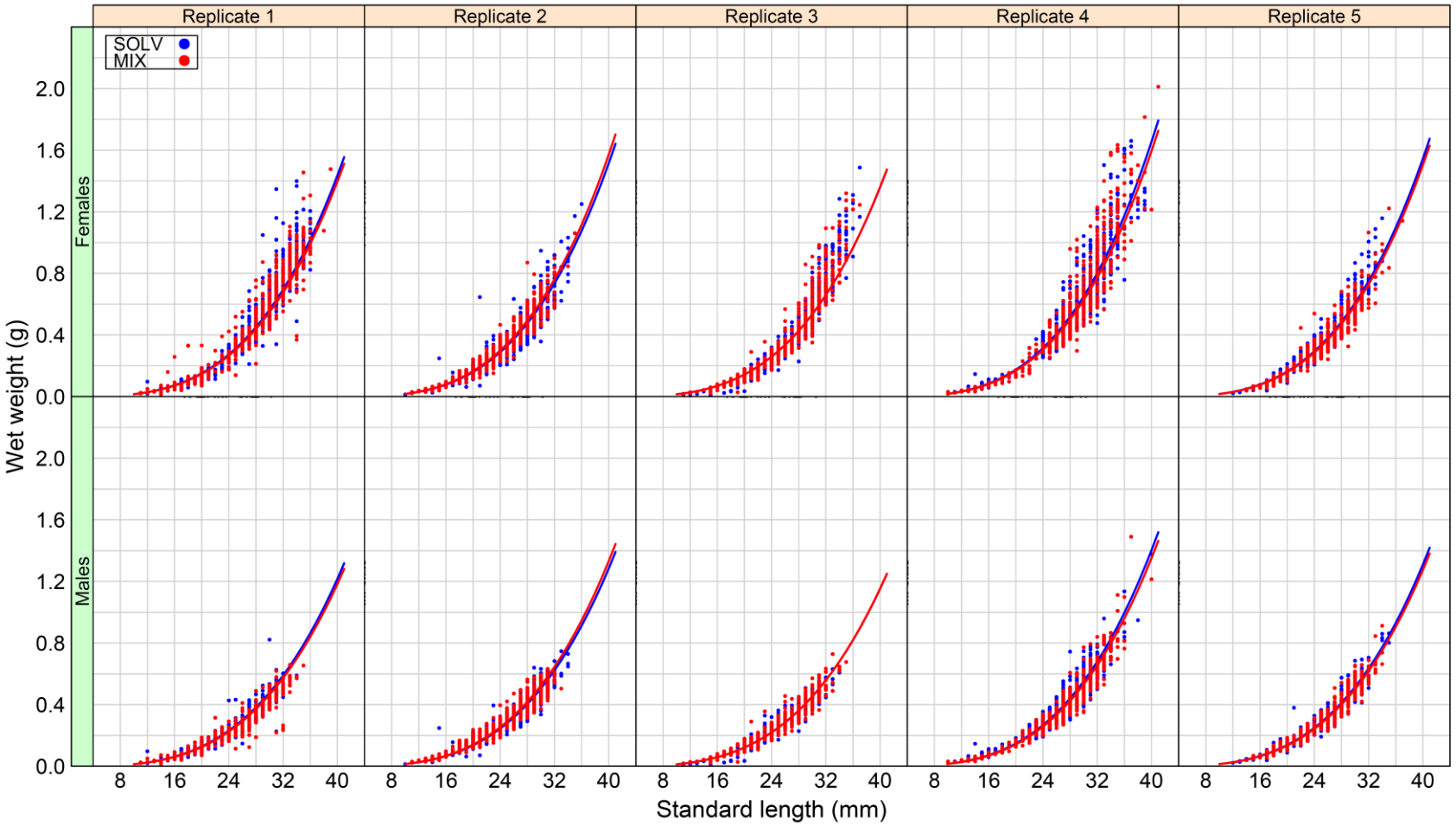


Fig. 4. Effect of dietary exposure to a PCB and PBDE congeners mixture on zebrafish body condition. Rows correspond to the sex of fish and columns to the replicates. Dots represent observations per treatment (MIX in red and SOLV in blue) and curves represent the fitted model per treatment.

3.5 Spawning probability

Spawning probability increased significantly with age and average length of male and decreased with average length of female (a, $\bar{l}_m$, $\bar{l}_f$, effects, Table 1; Fig. 5). As mean individual food consumption c had no effect on spawning probability, this covariate was not kept in the reduced model. The overall spawning probability was not affected by exposure to contaminant (no significant T effect, Table 1) but there was a significant interaction effect between treatment and age ($T \times a$ effect, Table 1) showing that spawning probability in MIX fish increased with age at a slower rate than in SOLV fish (Fig 5). Consequently, over the duration of the reproduction test, SOLV fish had a significantly higher likelihood to spawn compared to MIX fish.

3.6 Number of eggs

The comparison of the cumulative number of eggs produced by all successful spawning events throughout the reproduction test indicated a similar number of eggs produced by MIX fish (110 ± 82 eggs per female) than by SOLV fish (122 ± 82 eggs per female) (T effect Table 1; Fig. 5). In contrast, the number of eggs produced increased significantly with age and decreased significantly with the number of individuals in tanks (a and N effects, Table 1). Although the number of eggs produced by MIX fish seemed to increase with age faster than for SOLV fish (Fig. 5), the interaction between treatment and age was not significant ($T \times a$ effect that was not kept in the model).

3.7 Fertilization rate

Fertilization rate increased significantly with age (a effect, Table 1; Fig. 5) but there was no significant difference between MIX and SOLV fish in terms of both the overall level of

fertilization rate and its increase with age (T and $T \times a$ effects, Table 1; the latter was not kept in the model; Fig. 5).

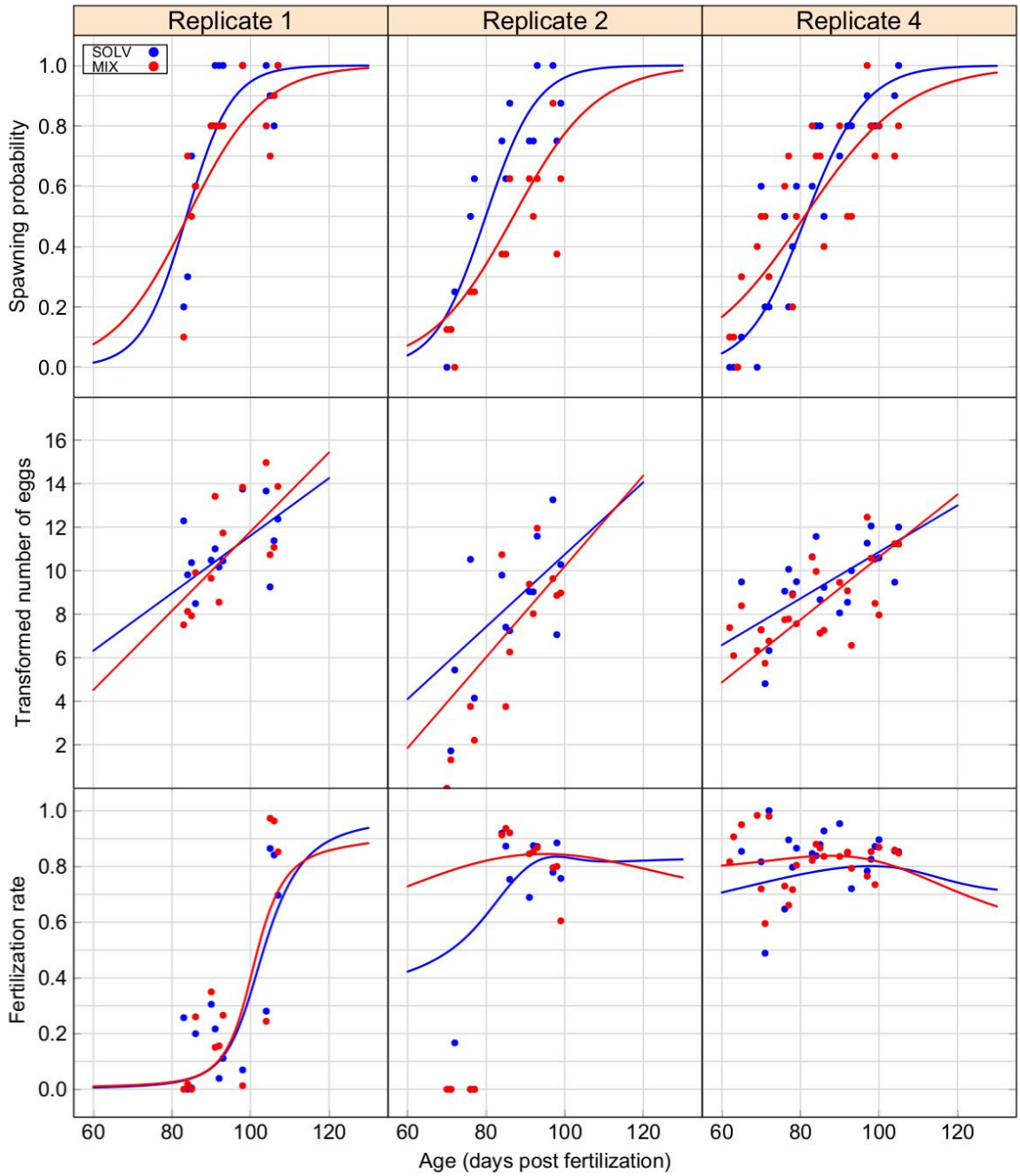


Fig. 5. Effect of dietary exposure to a PCB and PBDE congeners mixture on reproduction traits in zebrafish. Rows correspond to the spawning probability, number of eggs (Box-Cox transformed) and fertilization rate, respectively. Traits are represented as functions of age (dpf) in the three replicates (in columns) used for the reproduction test. Dots are for observed reproduction traits in rearing tanks for MIX (in red) and SOLV (in blue) fish and curves represent the fitted model per treatment.

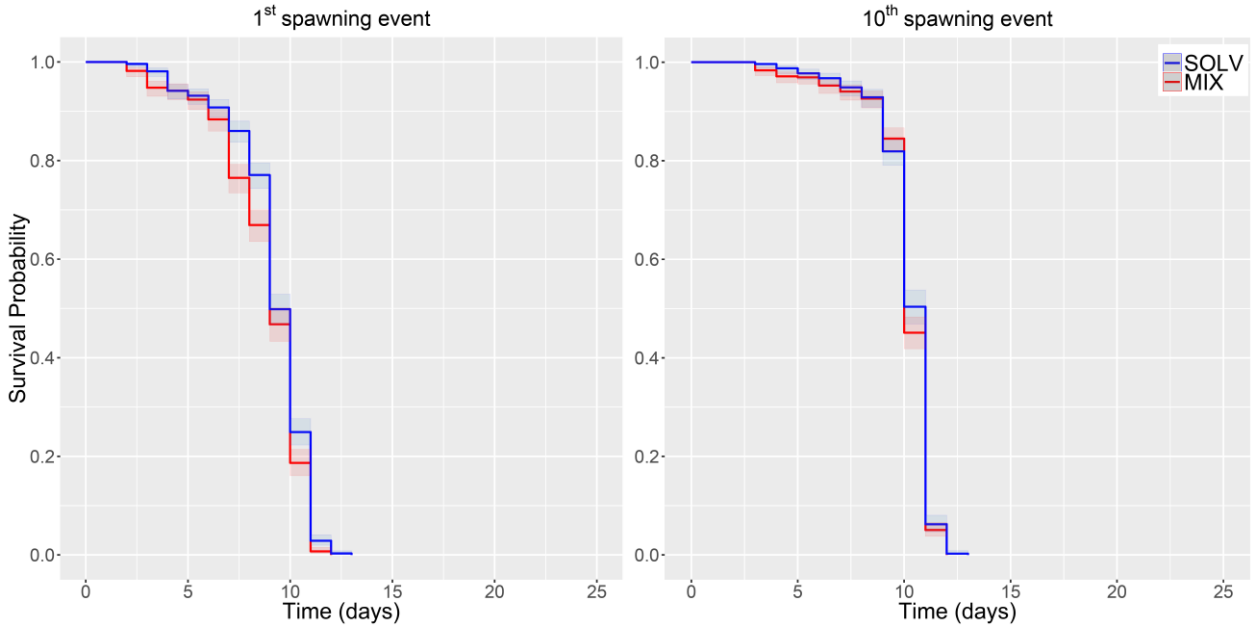
3.8 Larval survival in the progeny

The overall level of survival of unfed larvae was unaffected by exposure to contaminants (T effect, Table 2; Fig. 6), but survival differed significantly according to the spawning event considered (O effect, Table 2) and this difference was dependent on treatment ($T \times O$ interaction effect, Table 2). More precisely, survival probability was higher at the 10th F0 spawning event than at the 1st F0 spawning event and this difference was more pronounced for larvae originating from MIX progenitors than from SOLV ones (Table 2). This analysis was followed by a post-hoc multiple comparison test (Hothorn et al., 2008) to assess which groups differed from the rest. The test revealed that, on the 1st F0 spawning event, the survival probability of larvae produced by SOLV F0 fish was higher than that of larvae produced by MIX F0 fish (post-hoc test MIX/1st spawn vs. SOLV/1st spawn: Z = - 2.55, p-value = 0.021, Fig. 6, 1st spawning event) whereas no difference was detected on the 10th spawning event (post-hoc test MIX/10th spawn vs. SOLV/10th spawn: Z = 0.08, p-value = 1, Fig. 6, 10th spawning event).

Table 2. Analysis of the effects of dietary treatment (MIX vs. SOLV) on zebrafish F1 larval survival in non-feeding conditions by a mixed-effects Cox proportional hazards model. For categorical variables, effects are estimated for one category (indicated between parentheses) relative to the reference one taken as the intercept, namely MIX relative to SOLV for treatment T, and second observation date (10th spawning event) relative to the first one (1st spawning event) for spawning event O.

Trait	*Effect*	*Estimate*	*s.e.*	*df*	*χ2*	*p-value*
Larval survival in progeny	baseline hazard (SOLV/1st spawn)	n.a.	n.a.	n.a.	n.a.	n.a.
	O (10th spawn)	- 0.405	0.283	1	30.73	**<0.001**
	T (MIX)	0.986	0.318	1	1.65	0.198
	$T \times O$	- 1.053	0.440	1	8.92	**0.003**

p-values < 0.05 are in bold.



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555 **Fig. 6.** Effect of parental (F0) dietary exposure to a PCB and PBDE congeners mixture on the survival of unfed
556 offspring (F1) larvae in zebrafish. Curves represent the estimated decrease in individuals' survival probability
557 with time by Kaplan-Meier estimator for MIX (red) and SOLV (blue) treatment, respectively, and shaded areas
558 are the corresponding confidence intervals.
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4 Discussion

4.1 PCB and PBDE levels in MIX and SOLV fish

The very low concentration of PCB and PBDE congeners measured in SOLV diet showed that no external or cross-contamination between treatments occurred during diet preparation and exposure experiment. In contrast, MIX diet was actually contaminated with spiking efficiencies ranging from 86 to 130% of targeted concentrations (Table S1). As a result, concentrations of PCB and PBDE congeners measured in both females and males were clearly higher in MIX fish than in SOLV ones (80-85 times and 35-100 times, respectively). PBDE levels in fish at the end of the experiment were in the range of those reported in fish from European industrialized estuaries (Law et al., 2006). PCB levels at the end of the experiment were also in the range of those encountered in the environment (Robinson et al., 2017). These results show the ability of these contaminants to accumulate in fish tissues through trophic contamination and allow us to assign the observed differences between MIX and SOLV fish life-history traits to the effects of the trophic transfer of the PCB/PBDE mixture.

4.2 Effects on survival of exposed fish

No difference in early and juvenile/adult survival was observed between SOLV and MIX fish. Our result on early survival is in accordance with a study describing the effects of exposure to a single congener of PBDE (BDE-47) in zebrafish on roughly the same part of the life-cycle (Chou et al., 2010). PCBs and PBDEs have been shown to accumulate over time in fish tissues in experimental conditions (Daouk et al., 2011; Nyholm et al., 2009). However, as food was spiked at environmental doses, the contamination levels in tissues was relatively low during the entire life cycle, which may explain the absence of acute mortality in MIX fish at any stage. The second point that might explain the absence of a difference in survival between

MIX and SOLV fish, specifically at early stage, is that the exposure began at the larval stage from mouth opening (at 5 dpf) and not at the embryonic stage, which has been described as the most critical life-stage (Belanger et al., 2010). Indeed, previous studies in sole showed that exposure to POPs (among which PCBs and PBDEs) at the embryonic stage, i.e. eggs, via the water can lead to a delayed effect on survival during larval stage from mouth opening until metamorphosis (Foekema et al., 2012). Furthermore, Lema et al. (2007) also observed a decrease in zebrafish larval survival after exposure to high concentrations of BDE-47 from embryonic to larval stages. These studies therefore suggest that exposure to POPs at early embryonic stages, as for example through maternal transfer of accumulated pollutants into eggs, may affect larval survival. In the present study, such potential effects were not tested through the analysis of exposed fish early survival, but through the analysis of the larval survival of their progeny (see subsection "Larval survival of exposed fish progeny" below).

4.3 Effects on growth of exposed fish

In both sexes, MIX fish grew to larger lengths (asymptotic length l_∞) than SOLV fish but at a slower pace (growth rate coefficient k). This result contrasts with other studies in which long term exposure to POP mixtures were performed. Dietary exposure of zebrafish to mixtures of polycyclic aromatic hydrocarbons (PAHs) produced a decrease in both weight and length (Vignet et al., 2014), the amplitude of which depended on PAHs concentrations and fish's sex. Such reduction in length growth as well as reduction in body condition was also observed after a short exposure via the water of sole juveniles to a PAH mixture at high concentration mimicking an oil spill (Gilliers et al., 2012). In another experiment, the dietary exposure of zebrafish to environmentally realistic freshwater mixtures of POPs (including PCBs and PBDEs as part of the major contaminants identified) produced a significant increase in weight without changes in fish length, i.e. a significant increase in fish condition (Nourizadeh-

Lillabadi et al., 2009). Analyses of gene expression indicated disruption of endogenous hormone function, including genes involved in growth regulation (Berg et al., 2011; Lyche et al., 2011, 2010; Nourizadeh-Lillabadi et al., 2009). Taken together, these reports suggest that POP mixtures can increase or decrease growth in length and/or in weight depending on their chemical composition, which is indicative of the triggering of different mechanisms. However, in the above mentioned studies, the ratio of weight to length is either unchanged as in the present study (Vignet et al., 2014), increased (Nourizadeh-Lillabadi et al., 2009) or decreased (Gilliers et al., 2012). Regarding fish condition, our results showed no effect of an environmentally realistic marine mixture of PCBs and PBDEs on fish condition or length-weight relationship, suggesting that weight was affected by exposure only according to its allometric relationship with length. In zebrafish, early growth rate is very important in juveniles and slows down at the time of sexual maturation (Gómez-Requeni et al., 2010). The higher growth rate in MIX fish may therefore be related to changes in the sexual maturation process, which may be delayed in MIX fish.

4.4 Effects on reproduction of exposed fish

Three different aspects of reproduction were evaluated, namely the spawning probability, the number of eggs and the fertilization rate. Spawning probability increased with age faster in SOLV fish than in MIX fish, thus revealing a delay in reproduction in the latter. This result is contradictory with the earlier onset of puberty observed after exposure of zebrafish to a freshwater POP mixture (Nourizadeh-Lillabadi et al., 2009). However, in this earlier study and as previously stated, exposure produced an increase in female condition, which is one important positive driver of maturation and thus spawning probability in fish (Grift et al., 2007; Mollet et al., 2007; Uusi-Heikkilä et al., 2011; Wright, 2007). Numerous studies have demonstrated that POPs may act as endocrine disrupters by disrupting hormone pathways that regulate reproductive functions, leading to a decrease in reproductive success components

641 such as egg production and fertilization rate in fish (reviewed in Mills and Chichester, 2005;
642 Yu et al., 2015). For instance, Muirhead et al. (2006) showed that exposure to BDE-47 caused
643 a significant reduction in mature sperm in fathead minnows, suggesting that PBDEs can affect
644 male reproductive function and reduce male fertility. In the present study, the delayed
645 increase in reproductive output observed in MIX fish could also be explained by such
646 disruption caused by PCBs and PBDEs producing a delay in follicle maturation as this has
647 been shown after exposure to PCBs (Daouk et al., 2011) or PAHs (Vignet et al., 2016).

648 In the present study, MIX fish produced on average similar eggs per female than SOLV fish.
649 Previous works have shown that PCBs and PBDEs reduce clutch size in zebrafish (Kuiper et
650 al., 2008; Muirhead et al., 2006; Örn et al., 1998). Note however that contaminants were
651 administered at very high doses in these studies unlike in the present study during which the
652 mixture of PCBs and PBDEs was administered at doses representative of the marine
653 environment. Hence, PCBs and PBDEs might have not reached a sufficient concentration in
654 fish tissue to alter clutch size in the present study. For example, Kuiper et al. (2008) observed
655 no effect of BDE-71 (commercial pentabromodiphenylether mixture) on zebrafish egg
656 production at environmentally relevant exposure, but they suspected a decrease in egg
657 production when fish were exposed to higher levels of BDE-71. However, this decrease in
658 egg production was not statistically significant, likewise in the present study. In addition to
659 the number of eggs, several other reproduction traits can be used in order to give more
660 information on realized fecundity or reproductive output. Indeed, realized fecundity can be
661 seen as the combination of spawning probability and clutch size. In the present study, the
662 combination of delayed spawning probability and an unaltered increase in clutch size with age
663 can thus be interpreted as a diminution of realized fecundity at young ages for MIX treatment.
664 Besides fecundity, fertilization rate did not differ between MIX and SOLV fish. This contrasts
665 with previous studies in zebrafish where exposure to environmentally relevant concentrations

of PCB congeners via diet (Daouk et al., 2011) or of BDE-71 via water (Han et al., 2013) could reduce fertilization success. However, the effect of a mixture of PCBs and PBDEs may differ from those assessed for a single type of POPs (Daouk et al., 2011) or a single congener (Han et al., 2013) because of potentially differing mechanisms of action between PCBs and PBDEs as well as synergistic and antagonistic effects. In addition, fertilization can be highly variable within one treatment, which makes mean fertilization rate a poor predictor (Vignet et al., 2016).

Beyond exposure to PCBs and PBDEs, spawning probability and the number of eggs produced could also depend on other factors. Previous studies have shown that the probability of spawning in zebrafish increases with length of female (Paull et al., 2008; Spence and Smith, 2006; Uusi-Heikkilä et al., 2010) and male (Pyron, 2003). The latter is in agreement with our results that showed an increase in spawning probability with average male length that could be explained by female mating preferences for larger males (Pyron, 2003). In contrast, the observed decrease in spawning probability with average female length has never been reported before and seems rather counter-intuitive. However, another aspect that should be taken into account is the fact that fish used for reproduction tests were young adults. In zebrafish, female reach sexual maturity earlier than males (Gonzales, 2012), which is associated to larger weight and condition (Cousin et al., 2012; Gómez-Requeni et al., 2010). Therefore mature females are larger than males of the same age, which was confirmed by our results on growth and condition. In turn, if the preference of females for larger males implies that males should be larger than females themselves, then such choosiness could favor reproduction of the smaller females as they could find larger males more easily, especially in experimental populations where all individuals have the same age.

4.5 Larval survival of exposed fish progeny

Maternal transfer of contaminants in general − i.e., heavy metals, organochlorine pesticides, PAHs, PCBs − to offspring has been well documented in many species, including birds (Ackerman et al., 2016; Bargar et al., 2001), amphibians (Metts et al., 2013), and reptiles (Rauschenberger et al., 2007). In fish, several studies have focused more specifically on the maternal transfer of POPs to eggs, particularly PCBs and PBDEs, because of their high concentrations and widespread occurrence in aquatic environments (e.g. Daouk et al., 2011; Miller, 1993; Niimi, 1983; Nyholm et al., 2008; Yu et al., 2011; Zhang et al., 2010). These compounds have lipophilic properties that provide a route for transfer from a female's stored lipids to offspring through eggs. In other words, PCBs and PBDEs present in a female's body fat are transported to its oocytes via egg yolk lipoproteins derived from vitellogenin (Nyholm et al., 2008; Russell et al., 1999; Ungerer and Thomas, 1996; Zhang et al., 2010). Given such maternal transfer and the documented detrimental effect on larval performances of exposure to POPs at embryonic stage (Foekema et al., 2012, 2014; Grimes et al., 2008; Yu et al., 2011), the elevated mortality observed in unfed larvae descending from exposed fish in the present study was expected.

However, survival probability in unfed larvae descending from MIX fish increased from the 1st to the 10th spawning event so as to become similar to that of larvae originating from SOLV fish. This increase may be related to a gain in egg quality as females age and thus between early and later spawns, which is a common phenomenon in teleost fish (Brooks et al., 1997; Brunel, 2010; Nasiadka and Clark, 2012; Trippel et al., 1997). More precisely, one may hypothesizes that there was a gain in egg quality between the 1st and the 10th spawning event that compensated for the detrimental effect of PCBs and PBDEs transferred from mothers to their eggs. These results suggest that the negative effect of a realistic marine mixture of PCBs and PBDEs on larval survival may be mostly expressed in young females' progeny.

4.6 Energy-allocation interpretation and life-history implications

In terms of bioenergetics, life-history traits are considered as resulting from the allocation of limited energy resources acquired through feeding to three main compartments: maintenance, growth and reproduction (Partridge et al., 1991; Perrin and Sibly, 1993; Stearns, 1992). These compartments are supposed to be linked by energy-based trade-offs, so that any increase in resource allocation to one trait should be correlated with a decrease in allocation to the others. This is for example suggested by the slowing-down of growth at the beginning of sexual maturation (Gómez-Requeni et al., 2010). In our study, the increase in individual growth and the decrease in the reproductive output observed in exposed fish can be interpreted in terms of the energetic trade-off between growth and reproduction. In other words, it suggests that the mixture of PCBs and PBDEs altered the allocation of energy between these two compartments, with fish diverting more energy towards somatic growth at the expense of the reproductive function, and hence, MIX fish grew larger than SOLV fish but reproduced later. These results are consistent with the fact that PCBs and PBDEs are known endocrine disruptors favoring obesity (Berg et al., 2011; Lyche et al., 2011, 2010; Nourizadeh-Lillabadi et al., 2009) and impairing reproduction (Mills and Chichester, 2005; Yu et al., 2015), which can be seen as favoring energy investment towards soma against of gonads. These direct bioenergetic effects linked to the suspected endocrine activity of the PCB/PBDE mixture are additional to and independent from the indirect bioenergetics effect on basal metabolism and energy dedicated to maintenance due to detoxification that is often observed in contaminated individuals (Jørgensen et al., 2016; Kumaraguru and Beamish, 1983; Newman and Clements, 2008). The latter is supposed to be relatively weak in our case given that the PCB/PBDE mixture was administered at low environmental doses and that lethal effects were only observed at larval stage in the progeny.

In contrast to their suspected effect on the energetic trade-off between somatic growth and reproduction, a purely obesogenic effect of the PCB and PBDE mixture seems unlikely. Despite the fact that a variety of environmental endocrine disrupting chemicals such as POPs can cause changes in fat mass and subsequent obesity as it has been observed in fish (Berg et al., 2011; Lyche et al., 2010, 2011; Nourizadeh-Lillabadi et al., 2009), humans (Dirinck et al., 2011; Grün and Blumberg, 2009) and rats (Grün and Blumberg, 2009), the present study shows indeed no difference in condition between MIX and SOLV fish. Still, the present results do not allow rejecting the possibility that these compounds could cause simultaneous but independent changes in body growth and the reproductive function.

Another question is the net effect of these changes in growth and reproduction on individuals' reproductive output throughout the whole life-cycle, and thus their fitness. Given that fecundity is known to increase with length in teleost fish species (Kamler, 2012), it remains to be investigated whether the potential gain in fecundity due to the increase in body growth could (over-)compensate for delayed reproduction and potentially smaller clutch size at a given age. A study based on bioenergetics modeling may help to answer this question and to confirm the alteration of the energetic trade-off between growth and reproduction. Bioenergetic models at the individual level are often used to describe the effects of chemical stressors and their physiological modes of action (e.g. feeding, maintenance, reproduction, growth) (Álvarez et al., 2006; Augustine et al., 2012; Martin et al., 2013). They are also especially well adapted for extrapolating individual effects of stressors to populations (Beaudouin et al., 2015; Jager and Klok, 2010; Kooijman and Metz, 1984; Martin et al., 2013). One of the best-tested and most extensive bioenergetic approaches is the Dynamic Energy Budget theory (DEB) (Kooijman, 2010; Sousa et al., 2010), which describes the rates at which an individual organism acquires energy and utilizes it for three energetic compartments, namely maintenance, growth and reproduction or maturity. Developing such a

model to investigate the effects of an environmentally realistic marine mixture of PCBs and PBDEs would allow assessing (i) whether changes in growth and reproduction can be independent or not by testing theoretically different physiological mode of actions and (ii) whether they could compensate each other in terms of consequences on lifetime reproductive output and fitness.

4.7 Potential consequences of life-history effects of environmental mixtures of PCBs and PBDEs on fish population dynamics

Contaminant exposure may have serious implications for population dynamics and, consequently, on the structure of ecosystems as shown in many species (Barnthouse et al., 1990; Klok and de Roos, 1996; Munns et al., 1997). However, the effects of POPs on the population dynamics of fish, especially on exploited marine fish, are still poorly investigated. This study shows that chronic dietary exposure to an environmentally realistic marine mixture of POPs can delay reproduction and decrease progeny larval survival under starvation conditions while increasing body growth in zebrafish. We acknowledge that the results reported here using a freshwater species may not fully apply to marine fish, especially when considering the additional metabolic costs due to osmotic pressure maintenance in an hyperosmotic environment. There are however a large number of articles reporting similar toxicity pathways in marine and freshwater fish species for a number of biological functions such as growth (Bodiguel et al., 2009; Daouk et al., 2011), behavior (Gravato and Guilhermino, 2009; Oliveira et al., 2012; Vignet et al., 2017) and reproduction (Sun et al., 2015; Vignet et al., 2016). These studies therefore support the hypothesis that the present results on a freshwater fish species are indicative of what could happen in marine fish. Under the assumption that these life-history effects can be transposed to marine teleost fish in the wild, they may have detrimental consequences on their population dynamics.

Marine fish population recruitment is known to depend strongly on food availability, especially during the critical period corresponding to the transition from endogenous to exogenous feeding (Hjort, 1914), and on the match or mismatch between larvae emergence timing and that of their prey (Cushing, 1990), which can partly depend on hydrodynamic factors (Cury and Roy, 1989; Lasker, 1978). Starvation episodes during larval stages are common and generally lead to lower recruitment and smaller year classes (Cushing, 1990; Leaf and Friedland, 2014; Lusseau et al., 2014; Pritt et al., 2014). The results of this study on progeny larval survival suggest that exposure to PCBs and PBDEs in the marine environment could amplify the detrimental consequences of starvation on larval survival and the resulting decrease in recruitment level due to maternal transfer. This is in line with the hypothesis that the recruitment of some exploited marine fish species could be impaired due to nursery degradation partly related to the accumulation of PCBs and PBDEs (Gilliers et al., 2006; Riou et al., 2001; Rochette et al., 2010, 2012). Moreover, lower reproductive output because of delayed reproduction may also decrease recruitment although this effect may be partly compensated for by the observed increase in body growth and the associated increase in fecundity due to its dependency on length.

Finally, these life-history effects may also render commercial fish populations more vulnerable to exploitation. Increased growth could increase fishing mortality at young ages as fisheries select most often larger individuals (Garcia et al., 2012) while delayed reproduction could diminish spawning stock biomass (Enberg et al., 2010; Fiorentino et al., 2008) and lower larval survival could decrease its reproductive output. Moreover, the typical diminution in the average age of the spawning stock due to fishing (Berkeley et al., 2004; Brunel, 2010; Ottersen et al., 2006) could favor the expression of the decrease in larval survival due to the exposure to PCBs and PBDEs documented in the present study, and thus amplify the potential negative effect on total reproductive output. As a result, population growth rates of exploited

fish could diminish under exposure to POPs so that they may sustain lower exploitation levels and produce lower fishing yields.

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Supplementary material

Table S1. Diet composition. List of PCB and PBDE congeners used in this work along with the chlorine and bromine numbers. Targeted and measured congeners concentrations (ng g-1 ww) in MIX (n = 12) diet are indicated (mean ± SD), as well as the spiking efficiency. In the SOLV diet (n=16), the concentration of each congener is also indicated. n.a.: not available. LOD: limit of detection.

Congener	Br/Cl	MIX diet					SOLV diet			
		Targeted concentration (ng g^{-1})	Measured concentration (ng g^{-1})			Spiking efficiency (%)	Measured concentration (ng g^{-1})			Occurrence (n)
CB-8	2	28	27.20	±	1.21	97.1	0.20	±	0.05	3
CB-18	3	38	32.70	±	1.37	86.1	0.60	±	0.28	4
CB-28	3	75	69.00	±	3.35	92	0.30	±	0.07	5
CB-31	3	42	47.40	±	1.74	112.9	0.30	±	0.05	4
CB-44	4	65	66.50	±	1.83	102.3	0.40	±	0.23	4
CB-49	4	54	53.20	±	4.67	98.5	n.a.		n.a.	n.a.
CB-52	4	65	64.10	±	2.76	98.6	0.20	±	0.04	6
CB-77	4	30	31.80	±	1.54	106	n.a.		n.a.	n.a.
CB-101	5	150	151.90	±	6.47	101.3	0.90	±	0.48	8
CB-105	5	76	78.90	±	5.43	103.8	0.40	±	0.18	8
CB-110	5	166	170.60	±	7.03	102.8	0.60	±	0.19	8
CB-118	5	110	107.30	±	3.05	97.5	0.60	±	0.16	8
CB-128	6	37	32.60	±	1.30	88.1	0.20	±	0.07	6
CB-132	6	71	68.70	±	6.46	96.8	0.20	±	0.11	5
CB-138	6	207	199.60	±	4.94	96.4	0.60	±	0.30	8
CB-149	6	158	161.70	±	2.47	102.3	0.80	±	0.24	8
CB-153	6	280	270.40	±	14.24	96.6	0.60	±	0.42	8
CB-156	6	38	34.10	±	3.71	89.7	0.10	±	0.01	7
CB-170	7	71	69.10	±	4.76	97.3	<LOD		-	8
CB-180	7	130	129.30	±	5.55	99.4	0.30	±	0.12	8
CB-187	7	35	32.60	±	2.91	93.1	0.60	±	0.45	4
CB-194	8	35	33.60	±	3.56	96	<LOD		-	4
Sum PCBs		1991	1932.30	±	90.35	97.05	7.90		3.45	
BDE-28	3	9.95	10.78	±	0.60	108.3	0.03	±	0.01	5
BDE-47	4	49.97	49.93	±	2.91	99.9	0.35	±	0.13	16
BDE-100	5	14.94	15.95	±	1.19	106.8	0.09	±	0.04	16
BDE-99	5	30.25	30.25	±	1.63	100.0	0.11	±	0.07	16
BDE-153	6	9.74	10.18	±	0.56	104.5	0.03	±	0.004	6
BDE-183	7	100.48	130.20	±	12.65	129.6	0.14		-	1
BDE-209	10	195.73	232.53	±	31.27	118.8	0.17	±	0.11	16
Sum PBDEs		411.1	479.82	±	50.81	116.7	0.92	±	0.36	

S2 text

Quality assurance/quality control

PCB analysis

The calibration of the system was performed within quite a range using a multi-point (6) calibration curve to define the linearity range of our detector (ECD) for all contaminants, for example from 4 to 2000 $pg.\mu l^{-1}$ for CB-153. The relative precision of the method was checked for this type of samples by the analysis of 6 aliquots of a homogeneous preparation of fish (laboratory control card). The results showed coefficients of variation of less than 10 % for all congeners, which indicates a satisfactory reproducibility of the method. During the analysis of the real samples, analytical blanks were systematically measured every 10 samples. The blank were about 0.2 $pg.\mu l^{-1}$, 20 times less than the concentrations of the lowest standard of CB-153 (i.e., <0.1 $ng.g^{-1}$), and much less for other determinants. Surrogate recoveries were 86 ± 6 % for CB-30, 97 ± 8 % for CB-198 and 102 ± 9 % for CB-209 (mean ± SD calculated on n = 93 analyses) and no surrogate correction was applied for the reported concentrations.

Finally, 6 replicates of a Standard Reference Material, SRM2977 (mussel tissue) were analysed in order to determine the accuracy and precision of the method. PCB recoveries varied between 77 and 115 %. The concentrations of the real samples were not corrected for recoveries. Moreover, the RSD values ranged from 3.8 to 19.6 %, with a mean of 9.7 % for all PCBs. All these results were in agreement with certified reference.

The limit of quantification (LOQ) was estimated taking into account a signal to noise ratio of 3, the injection volume (1µl), the volume of the concentrated extract before injection (250 µl) and the extracted sample mass. Average value varied between 2 to 15 pg depending to congener.

PBDE analysis

Quality Assurance / quality Control procedures were implemented for each batch of 8 to 10 samples, including procedural blanks, use of recovery surrogates in all samples, analysis of certified reference material and participation to inter-comparison exercises. Detailed information can be found in Munschy et al. (2011) and an update is given below.

Procedural blank samples were analysed in every batch of 8 to 10 samples using glass powder, extracted and processed in the same manner as the fish samples. Blanks were contamination-free, with the exception of BDE-209, which was found at 0.5 ± 0.3 pg injected (average value calculated on 14 blanks). BDE-209 concentrations were corrected from blank values in every sample. The values obtained in each blank were used to correct the values in samples analysed in the same batch.

The limit of quantification (LOQ) was calculated for each sample taking into account a signal to noise ratio of 3, the injection volume, the volume of the concentrated extract before injection and the extracted sample mass. Average value was 0.09 ± 0.05 ng g^{-1} ww (n = 93).

Surrogate recoveries were 88 ± 8 % for BDE-139 and 77 ± 17 % for ^{13}C BDE-209 (mean ± SD calculated on n = 93 analyses) and no surrogate correction was applied for the reported concentrations.

Certified reference material (WMF-01, supplied by Wellington laboratories Inc., Ontario, Canada) was routinely included in each sequence of samples and the results were within ± 2 times the standard deviations of the means of the certified values. WMF-01 consists of a freeze-dried fish sample for which certified or indicative values are given for 7 congeners (namely, BDE-28, BDE-47, BDE-99, BDE-100, BDE-153, BDE-154, BDE-183) at concentrations of between 0.53 ± 0.40 ng g^{-1} dry weight (dw) for BDE-183 and 123.2 ± 24.8 ng g^{-1} dw for BDE-47.

The laboratory regularly takes part in Quality Assurance of Information for Marine Environmental Monitoring in Europe (QUASIMEME) inter-comparison exercises for PBDEs in

biota and our Z-scores are satisfactory, i.e., between - 2 and + 2 (for example, they were between - 0.28 and - 0.77 in 2015 and between - 0.83 and + 0.32 in 2016).

Table S3. Trait monitoring protocol for each replicate.

Replicates	Birth date	Number of tanks		Survival monitoring				Growth monitoring		Reproduction monitoring		
				Early		Juvenile/adult		Number of biometries	Ages (dpf) at biometry	Starting date (age in dpf)	End date	Duration (days)
		SOLV	MIX	Starting date (5 dpf)	End date (age in dpf)	Starting date (age in dpf)	End date (age in dpf)					
R1	19/02/2014	10	10	24/02/2014	30/03/2014 (39)	30/03/2014 (39)	28/08/2014 (190)	5	63, 96, 135, 160, 190	13/05/2014 (83)	06/06/2014	13
R2	12/03/2014	8	8	17/03/2014	12/04/2014 (31)	12/04/2014 (31)	09/09/2014 (181)	4	82, 119, 147, 181	21/05/2014 (70)	19/06/2014	14
R3	01/10/2014	9	8	06/10/2014	26/10/2014 (25)	26/10/2014 (25)	30/03/2015 (180)	11	57, 97, 127, 149, 180, 210, 240, 268, 302, 335, 362	-	-	-
R4	26/11/2014	10	10	01/12/2014	19/12/2014 (23)	19/12/2014 (23)	27/05/2015 (182)	5	61, 90, 120, 182, 211	27/01/2015 (62)	11/03/2015	25
R5	01/04/2015	10	10	06/04/2015	03/05/2015 (32)	03/05/2015 (32)	28/09/2015 (180)	7	64, 90, 120, 150, 180, 215, 244	-	-	-

Table S4. Statistical analyses and models used in this study. COXME: mixed-effects Cox proportional hazards model; NLME: non-linear mixed-effects model; LME: linear mixed-effects model; GLMM: generalized linear mixed-effects model. The following symbols are used in formulae for fixed effects : T dietary treatment, S individual's sex, l individual's length, a individual's age, $\bar{l}_f$ average female length in the tank, $\bar{l}_m$ average male length in the tank, c mean individual food consumption in the tank, N number of individuals in the tank, s sex-ratio in the tank, O number of the spawning event.

Generation	Function	Trait	Replicate	Model	Distribution	Link function	Variable transformation	Formula (fixed-effects)	Random effects	Random effects sources
F0	Survival	Early death rate	All	COXME	Semi-parametric	Log	Identity	T	Intercept	replicate/tank
F0	Survival	Juvenile/adult death rate	All	COXME	Semi-parametric	Log	Identity	T	Intercept	replicate
F0	Growth	Initial length l_0	All	NLME	Gaussian	Identity	Identity	T	Intercept	replicate/tank
F0	Growth	Growth rate k	All	NLME	Gaussian	Identity	Identity	$S+T+S\times T$	Intercept	replicate/tank
F0	Growth	Asymptotic length l_∞	All	NLME	Gaussian	Identity	Identity	$S+T+S\times T$	Intercept	replicate/tank
F0	Condition	Weight	All	LME	Gaussian	Identity	Log	$S+T+S\times T+\log(l)+\log(l)\times T+\log(l)\times S$	Intercept	replicate/tank
F0	Reproduction	Spawning probability	R1, R2, R4	GLMM	Binomial	Logit	Identity	$a+\bar{l}_f+\bar{l}_m+c+T+T\times a$	Intercept and slope ($a+T$)	replicate/tank
F0	Reproduction	Number of eggs	R1, R2, R4	LME	Gaussian	Identity	Box-Cox (λ=0.3)	$a+c+N+s+T+T\times a$	Intercept and slope ($a+T$)	replicate/tank
F0	Reproduction	Fertilization rate	R1, R2, R4	GLMM	Binomial	Logit	Identity	$a+\bar{l}_f+\bar{l}_m+c+T+T\times a$	Intercept and slope ($a+T$)	replicate/tank
F1	Larval survival	Larval death rate	R4	COXME	Semi-parametric	Log	Identity	$O+T+T\times O$	Intercept	tank/clutch

Table S5. Random effects testing using Likelihood Ratio Test (LRT).

Generation	*Function*	*Trait*	**Random effects**	**Random effects sources**	*df*	*χ2*	*p-value*
F0	Survival	Early death rate	Intercept	Replicate	1	82.53	**<0.001**
				Replicate/tank	1	21.11	**<0.001**
	Survival	Juvenile/adult death rate	Intercept	Replicate	1	259.79	**<0.001**
F0	Growth	l_0	Intercept	Replicate	1	37.57	**<0.001**
				Replicate/tank	1	107.68	**<0.001**
	Growth	k	Intercept	Replicate	1	99.02	**<0.001**
				Replicate/tank	1	8.76	**0.003**
	Growth	l_∞	Intercept	Replicate	1	4.54	**0.033**
				Replicate/tank	1	60.28	**<0.001**
F0	Condition	W	Intercept	Replicate	1	100.24	**<0.001**
				Replicate/tank	1	243.18	**<0.001**
F0	Reproduction	Spawning probability	Intercept and slope ($a + T$)	Replicate	6	1.06	0.983
				Replicate/tank	6	32.65	**<0.001**
F0	Reproduction	Number of eggs	Intercept and slope ($a + T$)	Replicate	6	3.51	0.742
				Replicate/tank	6	16.27	**0.012**
F0	Reproduction	Fertilization rate	Intercept and slope ($a + T$)	Replicate	6	66.39	**<0.001**
				Replicate/tank	6	6150.70	**<0.001**
F1	Larval survival	Larval death rate	Intercept	Tank	1	334.48	**<0.001**
				Tank/clutch	1	1007.20	**<0.001**

p-values < 0.05 are in bold.