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# Radiocesium accumulation in aquatic organisms: A global synthesis from an experimentalist's perspective

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## 21 **1. Introduction**

22 While radioactive releases into the ocean have in general decreased over recent time (Ito et  
23 al., 2003), there are still many unresolved concerns in coastal areas receiving direct  
24 radioactive inputs. This is particularly true after the 2011 accident that occurred in the nuclear  
25 power plant at Fukushima where a significant amount of radiocesium was released into the  
26 marine environment (Bailly du Bois et al., 2012; Chino et al., 2011). After this accident, the  
27 activity of radiocesium increased as much as 1000 times more than the background levels  
28 observed in the coastal waters off Japan before this event (Buesseler et al., 2011, 2012;  
29 Estournel et al., 2012).

30 Radiocesium isotopes are persistent in aquatic environments ( $^{134}\text{Cs}$ :  $t_{1/2} = 2.06$  yr and  $^{137}\text{Cs}$ :  
31  $t_{1/2} = 30.17$  yr), and so can be readily bioaccumulated by aquatic organisms at the bottom of  
32 the aquatic food chain (e.g. phytoplankton and macrophytes; Fisher, 1985; Harvey and  
33 Patrick, 1967; Heldal et al., 2001; Warnau et al., 1996) and can then be transferred to higher  
34 trophic levels such as fish (Pentreath, 1963; Zhao et al., 2001; Mathews and Fisher, 2009).  
35 Furthermore, radiocesium concentrations have been measured in the field up to  $1000 \text{ Bq kg}^{-1}$   
36 ( $338 \text{ Bq kg}^{-1}$  of  $^{134}\text{Cs}$  and  $699 \text{ Bq kg}^{-1}$  of  $^{137}\text{Cs}$ ) in fish (Chen, 2013; Iwata et al., 2013; Wada  
37 et al., 2016). Such observations confirm the importance of aquatic organisms as vectors for  
38 bioaccumulation and potential biomagnification of radiocesium (Tateda et al., 2013) as has  
39 also been suggested from results of laboratory radiotracer experiments (e.g. Mathews et al.,  
40 2008; Mathews and Fisher, 2008; Pan and Wang, 2016; Zhao et al., 2001).

41 The determination of radiocesium bioaccumulation parameters in aquatic organisms under  
42 controlled laboratory conditions can be key to better understanding the significance of field  
43 measurements (Warnau and Bustamante, 2007; Wang et al., 2018). Indeed, an experimental  
44 radiotracer approach can provide relevant information about contamination pathways or  
45 uptake and depuration capacities of exposed organisms (e.g. Pouil et al., 2015; Reinardy et al.,

46 2011; Sezer et al., 2014; Wang et al., 2000). Laboratory experiments allow (1) comparing the  
47 bioaccumulation capacities of different marine organisms in fairly comparable contamination  
48 conditions, (2) obtaining information about food chain transfer, (3) delineating the major  
49 uptake pathway(s) through computation of the data, and (4) providing a clear insight into  
50 major biological mechanisms that are activated during pollution events (Metian et al., 2016).  
51 In comparison to stable isotope approaches, radiotracer techniques offer several unique  
52 advantages, such as cost effectiveness and elevated throughput of samples. Furthermore,  
53 gamma-emitting radiotracers allow radioanalysis of live organisms and thus, substantially  
54 decrease the number of sacrificed organisms and generate data with reduced biological  
55 variability (Warnau and Bustamante, 2007). Laboratory experiments also enable the selection  
56 of appropriate candidate species for carrying out biomonitoring programs (e.g. Bervoets et al.,  
57 2003; Børretzen and Salbu, 2009; Warnau et al., 1999). Thus, experimental results help to  
58 better understand and predict the dynamics of radiocesium in aquatic environments and in  
59 biota after a contamination event.

60 The present review also identifies trends and gaps in the literature, as well as offers an  
61 opportunity to outline methodologies for measuring the bioaccumulation of radiocesium in  
62 marine organisms. The experiments outlined in the database and built into this review helped  
63 scientists understand the effects of radiological depositions by controlling environmental  
64 parameters in a laboratory. Such works which were carried out under controlled conditions  
65 were then compared to field data that were collected after accidents such as Chernobyl and  
66 Fukushima for making an even deeper analysis of the consequences of a nuclear accident on  
67 the environment. Furthermore, many different models have been developed to simulate  
68 additional depositions or to outline the pathways by which contamination moves through an  
69 organism and where the contamination will accumulate in these organisms. The necessary

70 inputs for these models are also outlined in works held within this database. This review  
71 therefore aims at outlining key results of previous experiments as a toolbox for modelers.

72

## 73 **2. Material and methods**

### 74 2.1. Literature search

75 Searches were performed to list all the available experimental studies carried out on  
76 radiocesium bioaccumulation in aquatic organisms. Laboratory studies with stable cesium  
77 isotopes, a less relevant approach to studying kinetics of radiocesium bioaccumulation, were  
78 not considered in this review. Commonly used databases were searched, e.g. Elsevier, Google  
79 Scholar, Scopus and Web of Science. For each database, searches included peer-reviewed  
80 articles, conferences articles, thesis and scientific reports over the time span from 1950 to  
81 present (2018). All available citation indexes of the database core collection were included in  
82 the search. Due to differences in search functionality, coding of the searches was adapted with  
83 selected keywords: “bioaccumulation”, “cesium”, “caesium”, and “aquatic organisms”.  
84 Following searches, duplicates were deleted. Non-relevant records, studies that not explicitly  
85 addressed bioaccumulation of radiocesium by an experimental approach, and review articles  
86 were removed. Records not written in English, at least the abstract, were excluded from  
87 further analysis. The completeness of the results obtained was considered as satisfactory  
88 based on “snowballing” (i.e. checking citations on reference lists of relevant articles until no  
89 further relevant articles could be found; Sayers, 2007).

90

### 91 2.2. Database construction

92 A bibliographic database (see supplementary material) was assembled to archive all  
93 publications (book chapters, conference articles, peer-reviewed articles, reports and theses)

94 that address radiocesium bioaccumulation in aquatic organisms under controlled conditions. A  
95 total of 125 publications was finally selected.

96 The information extracted for the database fell into 6 different sections:

97 1. Paper information and objective(s) of the experiment: This section includes reference  
98 information such as title, year and authors of the publication. Objective(s) of the  
99 experiment(s) including the tested variable and the isotope of cesium used ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ )  
100 were also compiled.

101 2. Biological model information: All details are provided about the biological model such as  
102 phylogeny, diet and trophic level and habitat (e.g. benthic, demersal, pelagic). For trophic  
103 level, information was collected from databases such as FishBase (Froese and Pauly, 2018) or  
104 scientific literature on the given species. In some cases, when there was no information  
105 available, approximations have been made, taking the TL of the closest-related species (e.g.  
106 filter-feeder bivalves were considered to be at a trophic level of  $2.1 \pm 0.13$ ).

107 3. Location information: Geographical information on where the sampling and experiments  
108 took place.

109 4. Experimental conditions: This section is focused on the materials and methods information  
110 such as the uptake pathway(s) examined, if uptake and/or loss were investigated, and the level  
111 of exposure (in  $\text{Bq L}^{-1}$  or  $\text{Bq g}^{-1}$ ). Details of size and/or weights of the experimental organisms  
112 are provided. Ambient habitat conditions of the organisms including source of water used  
113 (natural or artificial), open or closed water circulation, pH, temperature and salinity are  
114 described, and acclimation and experiment duration (expressed in days) are also indicated.

115 5. Data collection methodology: Herein is indicated the type of data collected (e.g. kinetic  
116 parameters, organotropism) and the biological level considered (i.e. whole-body or specific  
117 organs and tissues). For kinetics of radiocesium accumulation, when available, information is

118 provided on modelling approaches used to describe observed trends: exponential, linear or  
119 logarithmic models.

120 6. Results and additional information: The data collected in the results section of the  
121 publications were compiled in this section as well as the main points discussed. Additional  
122 information about the contents of the publication was also detailed. Normalization of the data  
123 (such as unit conversion and transformation) was done for comparison purposes and is clearly  
124 indicated in the database (see supplementary material).

125

### 126 **3. Global overview of the database**

#### 127 3.1. History of radiocesium bioaccumulation in experimental studies

128 Overall analysis of the database reveals that there is no coincidence that this research began  
129 after the early developments of nuclear weapons. Weapons testing left large amounts of  
130 fission products scattered throughout the environment, and since some radioisotopes of Cs  
131 have a long half-life (e.g. 30.17 years for  $^{137}\text{Cs}$ ), the deleterious effects of this contaminant on  
132 the aquatic environment and humans can extend over several decades.

133 The importance of studies outlining the bioaccumulation of radiocesium becomes even more  
134 obvious when assessing the effects of nuclear accidents. Nevertheless, our findings indicated  
135 that the number of publications on this topic has not increased significantly following the  
136 Chernobyl nuclear accident in 1986 and later after the Fukushima Daiichi nuclear power plant  
137 accident in Japan in 2011 (Fig. 1A). It was expected that there would be a sharp increase in  
138 the number of papers written following events such as Chernobyl and Fukushima. However,  
139 since 1990, there is a consistency in the publications per decade carried out on the  
140 experimental bioaccumulation of radiocesium in aquatic organisms (Fig. 1B). While the focus  
141 of studies changes over time, the overall objective of the selected publications appears to  
142 remain constant. It seems that since the early stages of nuclear power and weapons

143 development (in the 1950-1960's), the majority of research was focused on the accumulation  
144 and retention of radiocesium in different types of marine organisms, firstly through empirical  
145 approaches (e.g. Bryan and Ward, 1962; Gutknecht, 1965; Jefferies and Hewett, 1971) and  
146 more recently using kinetic models (e.g. Belivermiş et al., 2017; Metian et al., 2016; Sezer et  
147 al., 2014).

### 148 3.2. Biological models (species and taxa)

149 There is a great taxonomy diversity in the aquatic taxa selected in the experimental studies of  
150 Cs bioaccumulation. Indeed, among the 125 publications analyzed, 110 used animals as  
151 biological models (Fig. 2A). Thus, 158 animal species were studied including mainly ray-  
152 finned fish (Actinopterygii), bivalves and malacostracans (i.e. approx. 70% of the animals  
153 studied, Fig. 2B).

154 Plants, which include both macrophytes and some microalgal species, were studied in 22  
155 publications and used 40 species from 9 different classes (Fig. 2A). Among the latter,  
156 Chlorophyceae, Florideophyceae and Ulvophyceae were the most studied (Fig. 2B).

157 Chromista, including mostly algae were rarely studied; e.g. 17 publications based on 21  
158 species. The low representation of bacteria (Fisher, 1985; Harvey, 1969a; Harvey and Patrick,  
159 1967; Vogel and Fisher, 2010) and protozoa (Williams, 1960) confirms that the research  
160 effort examining bioaccumulation of radiocesium in aquatic microorganisms is still very  
161 limited (Fig. 2A).

162

### 163 3.3. Exposure pathways

164 The experimental study of radiocesium bioaccumulation can be done through (1) an exposure  
165 via a unique pathway (so-called single-pathway approach) or (2) several pathways studied  
166 separately or together (so-called multiple-pathway approach). The latter experimental  
167 approach allows a more comprehensive understanding of the mechanisms of bioaccumulation



168 in a given species, and it is useful to estimate, through a modelling approach, the main  
169 pathways of bioaccumulation (Børretzen and Salbu, 2009; Hewett and Jefferies, 1978; Metian  
170 et al., 2016; Pentreath, 1973; Pouil et al., 2015).

171 This review indicates that the single-pathway approach was used in 79% of the publications  
172 (Fig. 3) with a main focus on the water pathway (85% of the publications concerned). Food  
173 and sediment were also studied as single pathways in 9% and 1% of the publications,  
174 respectively (Fig. 3A). In the remaining publications (5%), other pathways were considered  
175 such as radiocesium injection into the bloodstream (e.g. Peters et al., 1999) and via maternal  
176 transfer to offspring (e.g. Jeffree et al., 2015, 2018).

177 Only 21% of the studies conducted a multiple-pathway approach. Among them, the  
178 combination of water and food occurred in 69% of the publications (Fig. 3B). Unlike single-  
179 pathway studies, more information on sediment is available in multiple-pathway studies, an  
180 aspect that was dealt with in 5 publications (Amiard-Triquet, 1974, 1975; Evans, 1984; Ueda  
181 et al., 1978, 1977). Furthermore, only 3 experimental works were conducted with 3 exposure  
182 pathways: water, food and sediment (Bustamante et al., 2006; Metian et al., 2011, 2016). The  
183 multi-pathway studies allow acquiring data regarding the major pathways of radiocesium  
184 bioaccumulation under similar experimental conditions (e.g. under the same physicochemical  
185 conditions whatever the studied pathway). Since aquatic organisms are naturally exposed to  
186 radiocesium from dissolved and particulate pathways (food, sediment), a multi-pathway  
187 approach should be preferred to highlight the main source of uptake and thus better  
188 characterize the main bioaccumulation pathway of this contaminant.

189

#### 190 **4. Factors influencing radiocesium bioaccumulation**

##### 191 4.1. Temperature

192 Temperature is one of the most important environmental variables in aquatic ecosystems since  
193 it has a strong impact on the physiology of organisms. For this reason, the influence of this  
194 abiotic factor on bioaccumulation of radiocesium has been extensively studied (17  
195 publications). Interestingly, the effects of temperature are similar in different taxa. Thus, an  
196 increase in temperature leads, in most cases, to an increase of concentration factors (CFs) of  
197 dissolved radiocesium in ray-finned fish (Hiyama and Shimizu, 1964; Prihatiningsih et al.,  
198 2016a), arthropods (Bryan, 1965; Bryan and Ward, 1962), echinoderms (Hutchins et al.,  
199 1996a, b), molluscs (Qureshi et al., 2007; Wolfe and Coburn, 1970) and algae (Boisson et al.,  
200 1997; Styron et al., 1976). Elimination of radiocesium following dissolved or trophic  
201 exposure is also temperature-dependent, with usually a higher radiocesium retention (i.e.  
202 longer  $T_{b1/2}$  and lower  $k_e$ ) when temperature decreases (Cocchio et al., 1995; Hutchins et al.,  
203 1996a, b; Ugedal et al., 1992). However, there are some exceptions where the temperature had  
204 no effect on uptake (Harvey, 1969b; Lacoue-Labarthe et al., 2012) or on the elimination of  
205 radiocesium (Hutchins et al., 1998). In some cases, reverse effects have been shown with, for  
206 example, a decrease in CFs observed in the goldfish *Carassius auratus* in relation to  
207 increasing temperatures (12, 20 and 28°C, Srivastava et al., 1994). Organotropism of  
208 radiocesium can also be affected by temperature. Indeed, it has been demonstrated in the  
209 channel catfish *Ictalurus punctatus*, after radiocesium injection into the blood, that its  
210 partitioning in the peripheral tissues decreased with increased temperature (Peters et al.,  
211 1999).

212 Interestingly, a meta-analysis based on information available in the database (see  
213 Supplementary Material) was performed and revealed no general trend regarding the  
214 influence of temperature on uptake (uptake rate and CF) and retention ( $T_{b1/2}$ , absorption  
215 efficiency) of dissolved radiocesium in aquatic organisms. Overall, these results suggest that  
216 the effects of temperature, the most studied abiotic factor, on the bioaccumulation of

217 radiocesium are complex and dependent both on the species considered and other  
218 environmental factors. For these reasons modeling the effects of temperature requires special  
219 attention.

220

#### 221 4.2. Salinity

222 Salinity is a master variable for coastal and marine ecosystems and can play an important role  
223 in the chemical speciation of many elements and also affect the physiology of aquatic  
224 organisms. This review showed that 15 publications explicitly dealt with salinity in Animalia,  
225 Chromista and Plantae species. Most of these studies found effects of salinity on the  
226 bioaccumulation of dissolved radiocesium in several taxa. For ray-finned fish species, there  
227 are contradictory findings on the effects of salinity on bioaccumulation of dissolved  
228 radiocesium, with an increase in CFs observed at the lowest (15 psu, Zhao et al., 2001),  
229 highest (35 psu, IAEA, 1975) or intermediate (29 psu, Prihatiningsih et al., 2016a) salinity  
230 conditions. In addition, Hattink et al. (2009) have demonstrated that radiocesium uptake in  
231 European seabass is independent of salinity (approx. 1-35 psu) as well as the assimilation  
232 efficiency (AE) of ingested radiocesium. Nevertheless, in turbot *Scophthalmus maximus*,  
233 Pouil et al. (2018) found a significantly higher AE when fish are exposed to low salinity  
234 conditions (10 and 25 psu) compared to the control condition (salinity: 38 psu). These results  
235 show that effects of salinity are species-dependent, likely in relation to their salinity tolerance  
236 ranges. Among invertebrates, Topcuoğlu (2001) found that bioaccumulation of radiocesium in  
237 the isopod *Idothea primastica* was significantly enhanced in a low salinity regime (approx. 7  
238 psu). Similar findings were highlighted for the lugworm *Arenicola marina* (Amiard-Triquet,  
239 1974). Generally, the same results have been reported for bivalves (Ke et al., 2000; Qureshi et  
240 al., 2007; Wolfe and Coburn, 1970) and malacostracans (Bryan, 1961; Bryan and Ward, 1962;  
241 Bryan, 1963) for salinity values from approx. 1 to 35 psu. Not surprisingly, radiocesium CFs

242 in algae are usually higher at the lower salinity (3.5-8 psu, Carlson and Erlandsson, 1991;  
243 Styron et al., 1976). Although there are some contradictory results, especially in fish, most  
244 experimental studies have shown that salinity strongly affects the bioaccumulation of  
245 radiocesium in aquatic organisms which usually results in an increase in concentration in low  
246 salinity regimes (< 15 psu). Various mechanisms have been proposed to explain that  
247 organisms living in low salinity regimes generally contain higher radiocesium concentrations.  
248 Indeed, salinity can affect physiological conditions of the organisms (e.g. cell volume,  
249 membrane permeability, water pumping rate) and chemical element speciation. Furthermore,  
250 an increase in cation concentrations with increasing salinity affects the permeability of the  
251 membranes and increases the competition for binding sites.

252

#### 253 4.3. Water composition

254 The effect of water composition on radiocesium bioaccumulation in aquatic organisms has  
255 been studied from different angles. Since Cs is an alkali metal, it is highly soluble in the water  
256 and exists almost exclusively as the monovalent cation  $Cs^+$  in aqueous solution. This  
257 dissolved form of radiocesium is chemically similar to the potassium (K) and sodium (Na)  
258 ions. Effects of K concentrations in water on the bioaccumulation of Cs were considered in 7  
259 publications. In three species of ray-finned fish exposed via the dissolved pathway, an  
260 increase in K concentrations led to a decrease in radiocesium uptake (Cocchio et al., 1995;  
261 Srivastava et al., 1990, 1994). Similar findings have been reported for the green-lipped mussel  
262 *Perna viridis* (Ke et al., 2000) and for the microcrustacean *Daphnia magna* (Hagstroem,  
263 2002). A plausible explanation for such findings may be competitive inhibition of  
264 radiocesium uptake by the high K concentrations (Bryan, 1963). However, a larger magnitude  
265 of effects of K concentration has been observed in microorganisms (Plantae and Protozoa).  
266 Thus, Hagstroem (2002) has found a negative effect of the increase of K in water on the

267 uptake of the dissolved radiocesium in two species of Chlorophyta, *Chlamydomonas*  
268 *noctigama* and *Scenedesmus quadricauda*, while Williams (1960) showed a positive  
269 relationship between K concentrations and the uptake of radiocesium in one species of  
270 Chlorophyta, *Chlorella pyrenoidosa*, and a species of Euglenozoa, *Euglena deses*.  
271 Considering the assumption that K could change the distribution of radiocesium between the  
272 solid and the liquid phase of sediment, Bervoets et al. (2003) studied the accumulation of  
273 radiocesium from the sediment in the benthic midge larvae *Chironomus riparius* and found it  
274 was unaffected by K concentrations in water. The influence of sodium (Na) concentrations  
275 has also been examined, and some authors have demonstrated that Na has little effect on the  
276 bioaccumulation of Cs (Hagstroem, 2002), and that is also true for calcium (Cocchio et al.,  
277 1995). In addition, Fraysse et al. (2002) showed the absence of an effect of the dissolved trace  
278 metals Cd and Zn on the bioaccumulation of radiocesium in the zebra mussel *Dreissena*  
279 *polymorpha*, contrary to what they observed for two other radionuclides ( $^{57}\text{Co}$  and  $^{110\text{m}}\text{Ag}$ ).  
280 All of these results demonstrate that there is ultimately insufficient experimental works  
281 investigating the effects of water chemistry on the bioaccumulation of radiocesium, such as,  
282 for example, water hardness. Nevertheless, field investigations have shown that, in freshwater  
283 ecosystems, water hardness and conductivity play a role in the level of radiocesium in biota  
284 (Hakanson et al., 1992; Särkkä and Luukko, 1995).

285

#### 286 4.4. Cs concentration

287 The influence of environmental stable Cs concentrations on subsequent radiocesium  
288 bioaccumulation in aquatic organisms has been studied in fish and bivalves, as well as in  
289 phytoplankton and bacteria species. In the mangrove snapper *Lutjanus argentimaculatus*,  
290 Zhao et al. (2001) found that radiocesium CFs were not influenced by the stable Cs  
291 concentrations (0.006-0.6 mM), whereas the calculated uptake rate ( $k_u$ ) increased linearly

292 with increasing ambient stable Cs concentration. Similar findings have been reported for the  
293 green-lipped mussel (stable Cs concentrations of 0.006-0.6 mM; Ke et al. 2000). Such results  
294 are in agreement with Argiero et al. (1966) who did not find any effect of external  
295 radiocesium concentration on CF in another bivalve species (*Mytilus galloprovincialis*). For  
296 microorganisms, Williams (1960) found in the bacteria *Euglena deses* and in the microalga  
297 *Chlorella pyrenoidosa* that the uptake of dissolved radiocesium was directly proportional to  
298 the ambient stable Cs concentration in water (0-2.5 mM). All of these results indicate that  
299 increasing ambient Cs concentrations lead to a positive linear response of the radiocesium  
300 uptake rate in the aquatic organisms studied. In other words, this suggests that, for the limited  
301 number of aquatic organisms studied, equilibration of radiocesium uptake was not reached  
302 within the experimental Cs concentrations tested (broad range from 0 to 2.5 mM).

303

#### 304 4.5. pH

305 That the physiological processes of aquatic organisms are affected by changes of pH is  
306 especially important in the context of ocean acidification. For this reason, effects of pH are  
307 increasingly being considered in ecotoxicology studies. Nevertheless, reports in the literature  
308 on the influence of pH on radiocesium bioaccumulation remain rare. Indeed, only 4  
309 publications dealing with pH have been identified. In ray-finned fish species, the Atlantic  
310 salmon *Salmo salar* and the brown trout *Salmo trutta trutta*, which were maintained in  
311 freshwater at two pH values (5.00 and 7.40), the Cs uptake rate was significantly reduced at  
312 low pH, but efflux rates were little affected (Morgan et al., 1993). More recently, pH  
313 experiments have been carried out on marine invertebrates; e.g. in cuttlefish eggs of *Sepia*  
314 *officinalis* (Lacoue-Labarthe et al., 2012) and in the Manila clam *Ruditapes philippinarum*  
315 (Sezer et al., 2018). In a comparative study, Lacoue-Labarthe et al. (2018) showed no  
316 influence of pH on the bioaccumulation of radiocesium in the variegated scallop

317 *Mimachlamys varia* and the kuruma shrimp *Penaeus japonicus*. These four studies did not  
318 show any significant difference in the bioaccumulation of dissolved Cs in either the molluscs  
319 (bivalves and cephalopods) or the arthropod exposed to low pH (minimum values of 7.60). At  
320 the organ and tissue level, Lacoue-Labarthe et al. (2012) found that the fraction of  
321 radiocesium associated with the perivitelline fluid of the cuttlefish eggs was higher at lower  
322 pH levels than at normal pH, whereas radiocesium in the eggshell was lower at pH 7.60 than  
323 at pH 8.10. The same authors attributed this result to an increase in the concentration of H<sup>+</sup>  
324 that may reduce the radionuclide adsorption on the eggshell or epithelia through increasing  
325 competition between cations for the binding sites. Thus current knowledge, based on very few  
326 publications, suggests that the influence of pH on the bioaccumulation of radiocesium in  
327 aquatic organisms is limited.

328

#### 329 4.6. Species

330 Interspecific difference is one of the most studied factors influencing the bioaccumulation of  
331 radiocesium in aquatic organisms (49 publications). Thus, bioaccumulation of radiocesium in  
332 190 species of Animalia, Bacteria, Chromista, Plantae has been compared in the literature.  
333 Examples showing differences in bioaccumulation in phylogenetically-close species are  
334 numerous. Differences between these species have been found after exposures from food (e.g.  
335 Hewett and Jefferies, 1978; Pan and Wang, 2016; Warnau et al., 2002), sediment (e.g.  
336 Amiard-Triquet, 1975; Marc Metian et al., 2016; Ueda et al., 1978) and water (e.g. Baptist  
337 and Price, 1962; Bryan and Ward, 1962; Harvey and Patrick, 1967; Haldal et al., 2001).  
338 Linking taxonomy, phylogeny and radiocesium bioaccumulation can be very complex (Brown  
339 et al., 2019). Nevertheless, a simple meta-analysis of data from the database revealed  
340 differences in the CFs and AEs observed among the different kingdoms (Animalia, Bacteria,  
341 Chromista and Plantae). Thus, CFs of dissolved radiocesium in these different taxonomic

342 groups were ranked in the following decreasing order Bacteria  $\geq$  Chromista > Animalia  $\geq$   
343 Plantae ( $p < 0.05$ , Fig. 4A). AEs of ingested radiocesium in the different classes were ranked  
344 in the following decreasing order Asteroidae  $\geq$  Elasmobranchii > Actinopterygii  $\geq$  Gastropoda  
345  $\geq$  Malacostraca  $\geq$  Polychaeta  $\geq$  Bivalvia ( $p < 0.05$ , Fig. 4B). These meta-analyses allow  
346 highlighting global trends, but their interpretation must take into account the large disparities  
347 in the study of the different taxonomic groups (see Section 3.2) that may affect results.

348

#### 349 4.7. Size and life-stages

350 In aquatic organisms, such as invertebrates and fish, age and size are correlated. The database  
351 analysis revealed a relative abundance of information on the influence of these variables on  
352 the bioaccumulation of radiocesium in various species of ray-finned fish (Actinopterygii) and  
353 molluscs (bivalves and cephalopods). Thus, in ray-finned fish, some publications have  
354 demonstrated higher CFs of dissolved radiocesium in small or medium size individuals  
355 compared to larger ones (Malek, 1998, 1999; Suzuki et al., 1992). Similarly, Morgan et al.  
356 (1993) stated that juvenile brown trout are more susceptible to bioaccumulate dissolved  
357 radiocesium than adults. Furthermore, Ugedal et al. (1992) reported a higher retention of  
358 radiocesium in small individuals of brown trout.

359 For molluscs, a higher ability to bioaccumulate dissolved radiocesium has been shown in  
360 smaller (= younger) individuals of bivalve mussels (*M. galloprovincialis* and *P. viridis*)  
361 through the measurements of  $k_u$  or CF (Argiero et al., 1966; Ke et al., 2000). Nevertheless,  
362 GÜngör et al. (2001) and Nolan and Dahlgaard (1991) have shown more contrasting results  
363 with no significant difference of CF or  $T_{b1/2}$  between mussels (*M. edulis* and *M.*  
364 *galloprovincialis*) of different sizes. Such differences observed for the same species (or a  
365 phylogenetically closely related species) can be explained, at least partially, by the size ranges  
366 used which vary greatly in these publications. Indeed, while Ke et al. (2000) used individuals



367 of 3-4 cm shell length, Güngör et al. (2001) and Nolan and Dahlgaard (1991) have made their  
368 observations on a larger size range (approx. 2.8-6.5 cm shell length. In the cephalopod *S.*  
369 *officinalis*, Bustamante et al. (2006) demonstrated a higher assimilation efficiency (i.e. AE)  
370 and retention (i.e.  $T_{b1/2}$ ) of ingested radiocesium in juveniles compared to adults indicating  
371 that the greater ability of smaller (= younger) individuals to bioaccumulate radiocesium can  
372 be also true when radiocesium is taken up from food. The authors stated that these  
373 differences could be related to the decrease of digestive metabolism with age in cephalopods,  
374 with the consequence of a higher efficiency of digestion process in smaller (= younger)  
375 individuals.

376 Interestingly, even though this was not the main purpose of their study, Warnau et al. (1996)  
377 showed in plants (the Neptune grass *Posidonia oceanica*) and the killer algae (*Caulerpa*  
378 *taxifolia*) that adult leaves have a higher radiocesium retention time (i.e. slower depuration)  
379 than that in younger leaves. This is one of the few publications available on the influence of  
380 size or stage of life on the bioaccumulation of radiocesium in plants.

381

#### 382 4.8. Food quality and starvation

383 Food quality (type of natural prey and compounded food) is well-known to affect the  
384 assimilation of trace elements in aquatic organisms. For radiocesium, effects of food quality  
385 have been investigated in ray-finned fish and in several invertebrate species (arthropods and  
386 molluscs). Zhao et al. (2001) found in the mangrove snapper (*L. argentimaculatus*) that there  
387 was no significant difference in radiocesium AE when fed with different prey. Similar results  
388 were found in three species of ray-finned fish with contrasting feeding habits (Pan and Wang,  
389 2016). In bivalves, no significant effect of food has been shown in the Manila clam *Ruditapes*  
390 *philippinarum* although AE varied slightly between the experimental treatments (Belivermiş  
391 et al., 2017). Thus, interestingly, the type of food seems to have very limited effect on the

392 assimilation of radiocesium in aquatic organisms. Furthermore, uptake of dissolved  
393 radiocesium was not affected by starvation as has been shown for several species of  
394 Malacostraca (Bryan, 1961; Bryan and Ward, 1962).

395

#### 396 4.9. Trophic ecology

397 Meta-analyses were conducted to characterize the influence of trophic ecology on the ability  
398 of aquatic organisms to accumulate radiocesium. Thus, data available on major kinetic  
399 parameters determined respectively from dissolved (CF) and trophic (AE) exposures were  
400 represented as a function of the trophic level of each study (Fig. 5). Regarding CFs, no clear  
401 relationship could be established. However, the results indicate that organisms belonging to  
402 the lowest trophic level (i.e. 1, primary producers) are likely to reach very high CFs values ( $>$   
403 1000, Fig. 5A) in contrast to consumers (trophic levels  $>$  1, Fig. 5A). These results suggest  
404 that, although considerable variabilities exist within each trophic level group, there is no  
405 general trend for the radiocesium CFs to increase with increasing trophic level as suggested in  
406 some previous studies (Fisher et al., 1999; Wang et al., 2000; Zhao et al., 2001). In fact, the  
407 results in Figure 5A even suggest a tendency to decrease with increasing trophic level.  
408 Regarding AE of ingested radiocesium, the meta-analysis showed a trend towards a linear  
409 increase in AE as a function of trophic level (Fig. 5B), a finding that can partly explain why  
410 Cs is one of the few trace elements which show a biomagnification potential at the top level of  
411 food chain (Mathews et al., 2008; Mathews and Fisher, 2008; Zhao et al., 2001).

412

### 413 **5. Organotropism of Cs in aquatic organisms**

414 Measurements of the distribution of radiocesium in organs and tissues are important to  
415 understand the site-specificity of radiocesium binding, to provide additional mechanistic  
416 information potentially helpful in the interpretation of results from whole-body kinetic

417 measurements, and to furnish additional information for modelling. This literature review  
418 reveals that radiocesium organotropism is relatively poorly studied in laboratory experiments.  
419 Indeed, specific data on the distribution of radiocesium in organs and tissues, expressed as  
420 percentages, have been reported in only 35 publications. Results concerning Cs organotropism  
421 are always difficult to compare between studies since it is rarely the same body compartments  
422 that are considered, and because there is an internal redistribution of the bioaccumulated Cs,  
423 i.e. the time of sampling can greatly affect the results of organotropism (e.g. Onat and  
424 Topcuoğlu, 1999; Wang et al., 2000). Nevertheless, some results are notably similar between  
425 studies. Indeed, in ray-finned fish many have demonstrated a high proportion of Cs in muscles  
426 (>50%) after exposure by the dissolved route (Guimarães, 1992; Jeffree et al., 2006b; Malek,  
427 1999, 1998; Malek et al., 2004; Twining et al., 1996) or by injection into the blood (Peters et  
428 al., 1999). In bivalves, results are contrasted with absorption of radiocesium in the shell  
429 surface that can be species-dependent (Ke et al., 2000; Metian et al., 2016; Onat and  
430 Topcuoğlu, 1999; Pouil et al., 2015) and pathway-dependent (Metian et al., 2016; Metian et  
431 al., 2011; Pouil et al., 2015). Nevertheless, care needs to be taken in interpreting these results.  
432 Indeed, rinsing methods of organisms for removal of adsorbed radiocesium before carrying  
433 out radiocesium measurements are sometimes not adequately reported and can therefore lead  
434 to an overestimation of radiocesium on the external surfaces (Cresswell et al., 2017).

435

## 436 **6. Gaps and perspectives**

437 Much has been done over the last 60 years in radioecological research to better assess  
438 radiocesium dynamics, with a main focus on fish and a few abiotic parameters. Figure 6  
439 highlights the research efforts on bioaccumulation and on a series of factors influencing the  
440 bioaccumulation of radiocesium in aquatic organisms. It also brings gaps of knowledge to the  
441 fore, identified by the limited number of studies and/or unclearly explained effects. It is

442 especially true for (1) some abiotic environmental factors such as water chemistry (e.g.  
443 chemical composition and pH) and (2) biotic factors (the life-stages and size of the  
444 organisms). All the listed factors should be looked at and become priority topics for further  
445 investigations on radiocesium accumulation in aquatic organisms. Therefore, future research  
446 on this topic should include the effect of abiotic factors (single or multiple factors) and  
447 examine some species that have not been investigated to date. For instance, there is a need to  
448 focus future work on small organisms that constitute food for fish, and to investigate some  
449 abiotic factors that have not been examined to date such as seawater deoxygenation. In  
450 addition, it would be important to better assess the main uptake pathway in a wider range of  
451 taxa, not considering only water and food but also sediment.

452 In future experimental research on radiocesium in aquatic organism, a special effort should  
453 be made to examine food transfer. Indeed, radiocesium enters aquatic food chains primarily  
454 from the aqueous phase into plankton (phyto- and zoo-) which is then consumed and highly  
455 assimilated by a variety of organisms including fish (Thomas et al., 2018). This gap is  
456 confirmed by our meta-analysis (Fig. 5). In fact, one recent modeling approach has indicated  
457 that 99% of the total body burden of radiocesium in fish is diet-driven in both marine and  
458 freshwater environments (Thomas et al., 2018).

459

## 460 **7. Conclusion**

461 As summarized in this review, laboratory-based investigations and subsequent meta-analyses  
462 are proven useful to identify general trends regarding the factors influencing the  
463 bioaccumulation of radiocesium isotopes, and thus better understand their transfer in aquatic  
464 environments after accidental contaminations. In addition, our database available as  
465 supplementary material, provides an exhaustive source of experimental data useful for  
466 modeling purposes.

467

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833

834 **Captions to figures**

835

836 Figure 1. Number of publications dealing with experimental studies of radiocesium  
837 bioaccumulation in aquatic organisms (A) per year in relation with the Chernobyl and  
838 Fukushima accidents, and (B) per decade.

839

840 Figure 2. Taxa used as biological models to study bioaccumulation of radiocesium expressed  
841 (A) by kingdom, and (B) by class.

842

843 Figure 3. Proportion and pathways considered in experimental studies conducted by (A)  
844 single-pathway approach, and (B) multiple-pathway approach.

845

846 Figure 4. Influence of phylogeny on (A) Concentration Factor (CF) values determined from  
847 dissolved exposure in the different kingdoms, and (B) Assimilation Efficiency (AE) values  
848 calculated after trophic exposure in different classes of aquatic animals. Whiskers represent  
849 both the max and min values, and the black line represents the median values. Small case  
850 letters (a and b) denote statistical differences ( $p_{\text{Kruskal-Wallis}} < 0.05$ ).

851

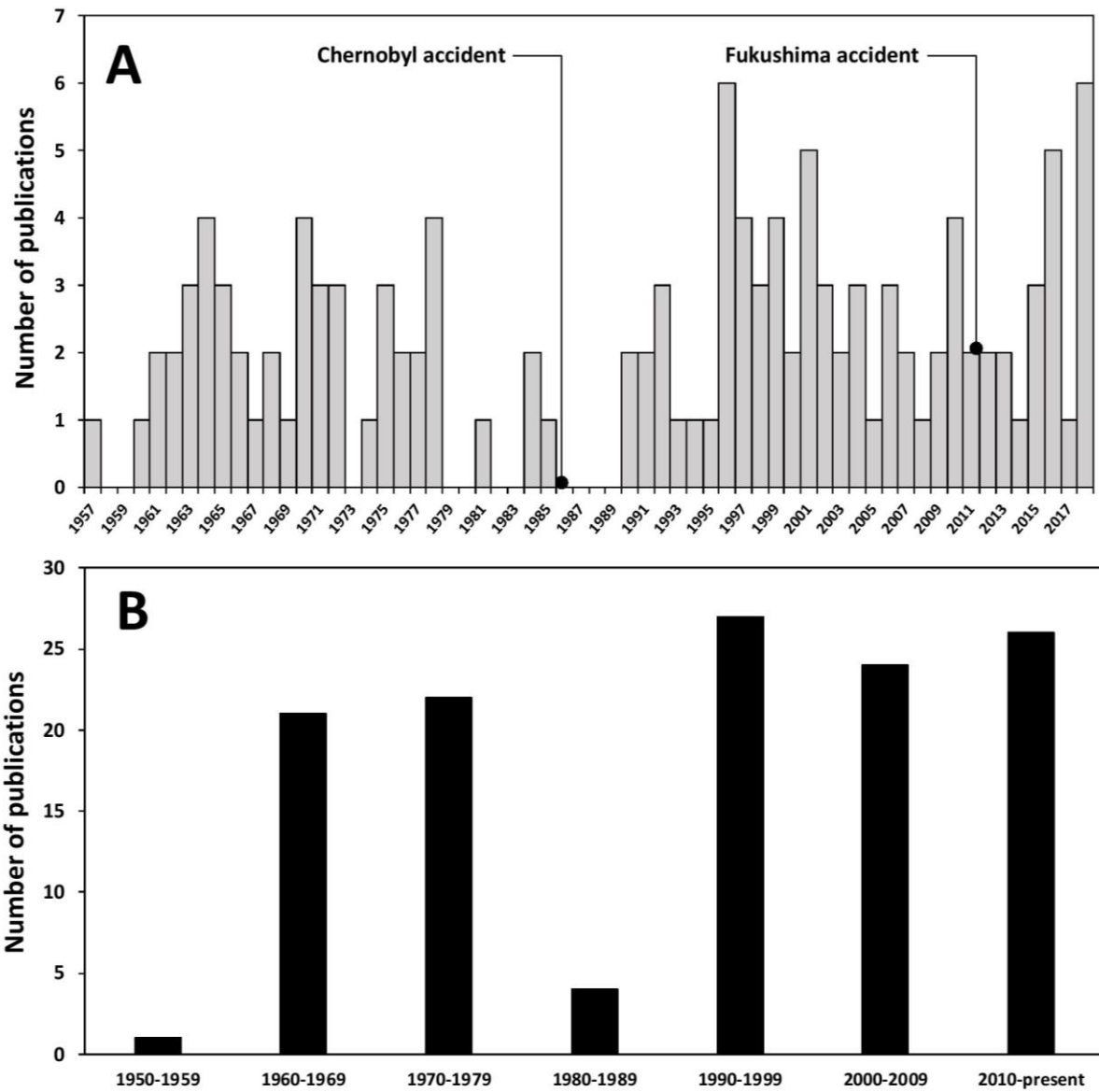
852 Figure 5. Influence of trophic level (mean values, ranking from 1 for autotroph producers to 5  
853 for higher heterotroph consumers) on (A) Concentration Factor (CF) values determined from  
854 dissolved exposure, and (B) Assimilation Efficiency (AE) calculated after trophic exposure to  
855 aquatic organisms.

856

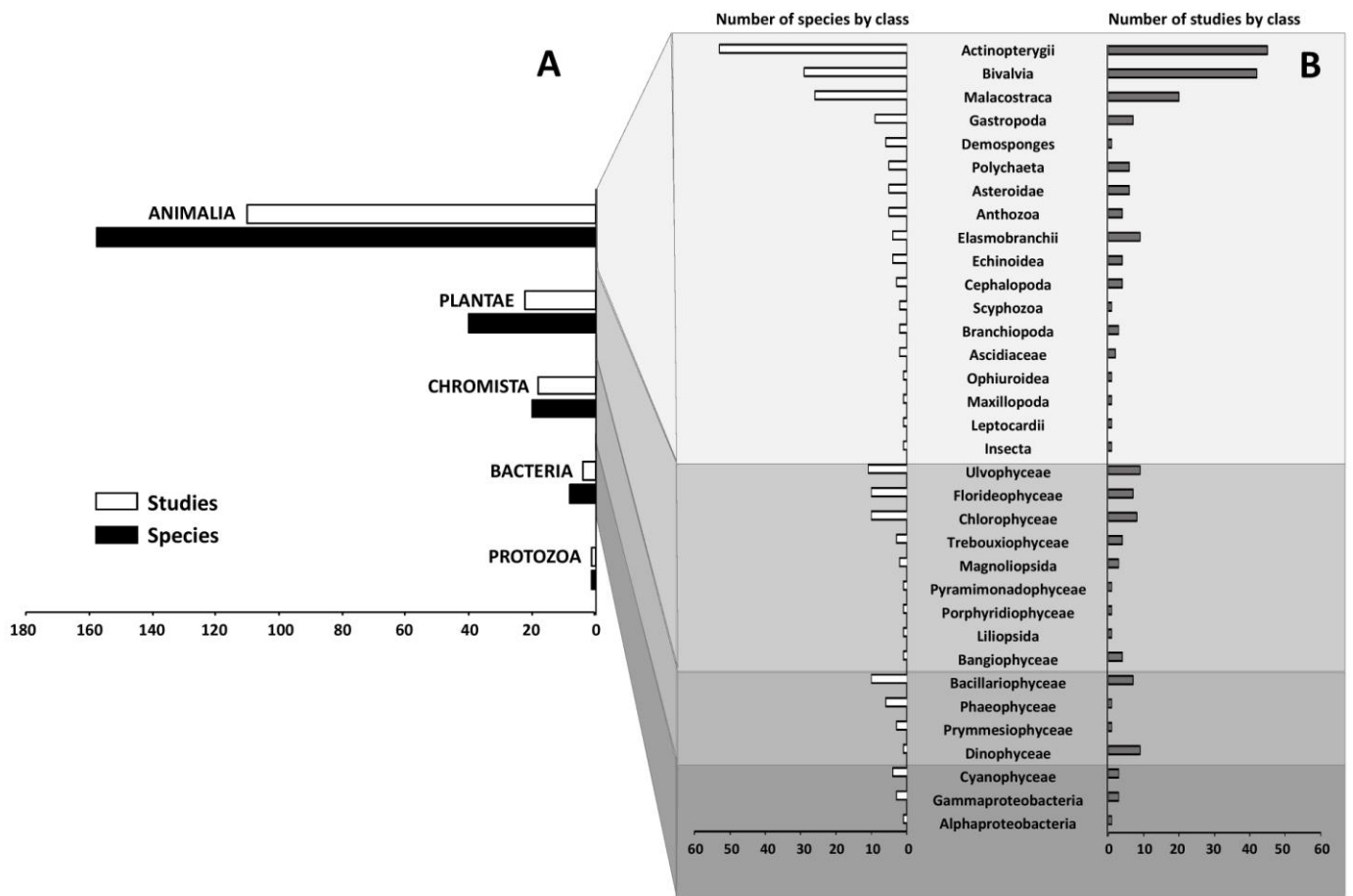
857 Figure 6. Synthesis of the different factors influencing the bioaccumulation of radiocesium in  
858 aquatic organisms which have been studied, and their relative occurrence in the literature.

859 \* Effects of antibiotics, cell density, food preparation and sex.



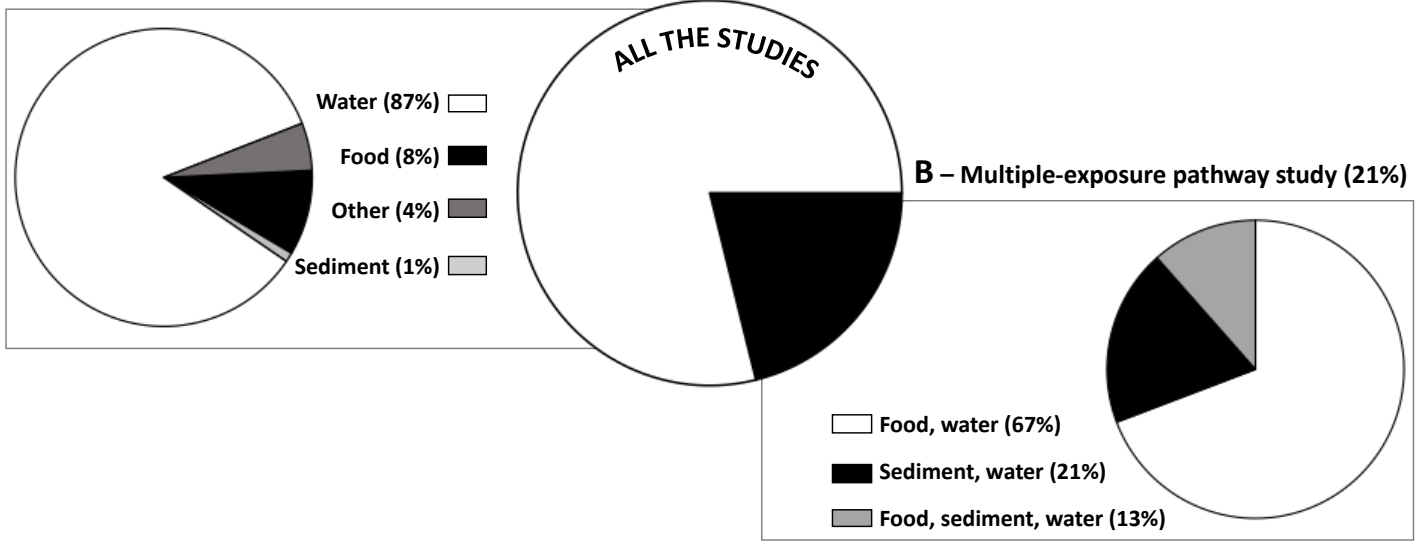


860 Figure 1

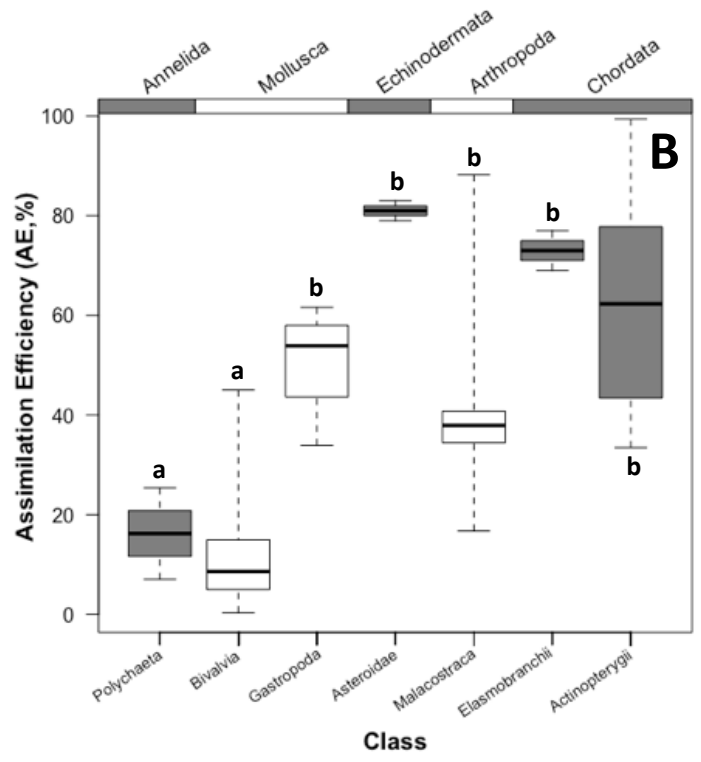
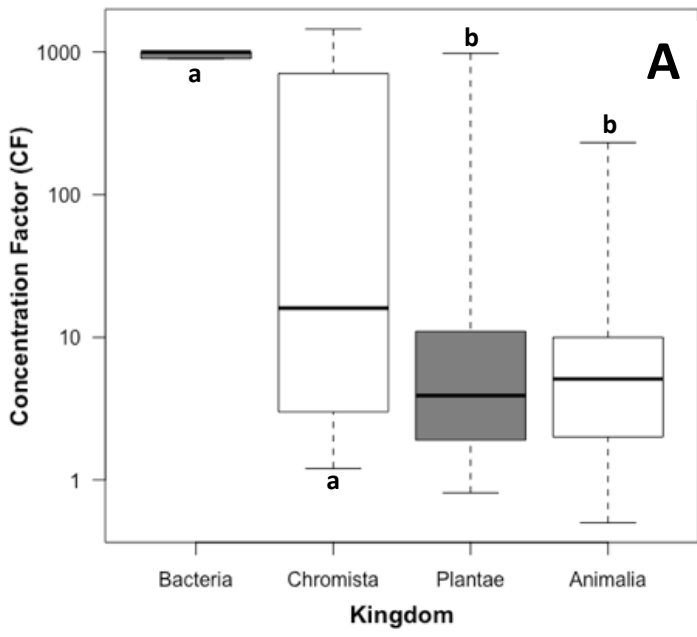


861 Figure 2

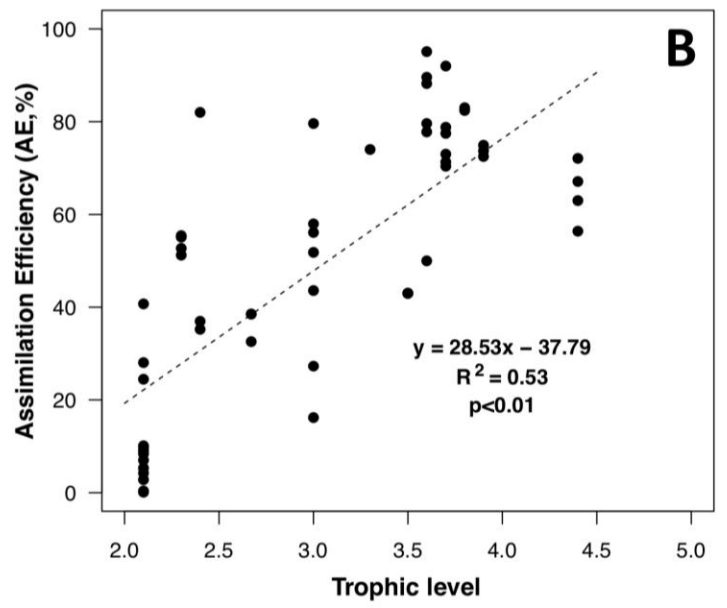
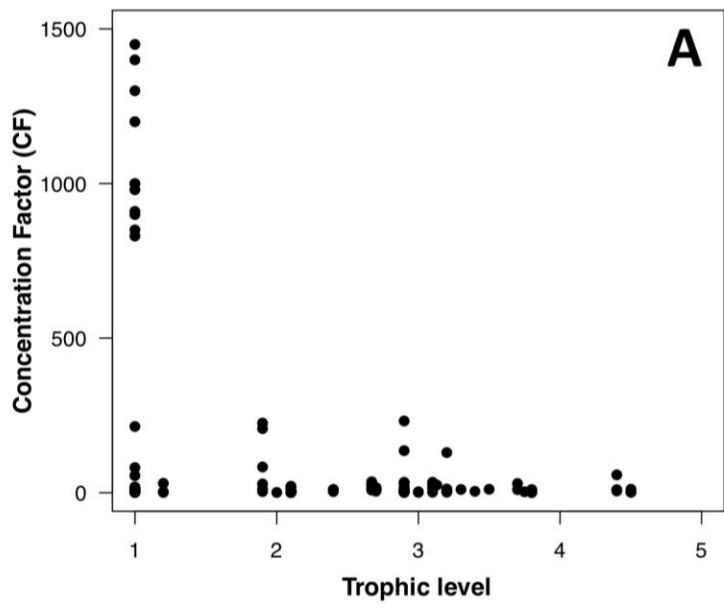
**A - Single-exposure pathway study (79%)**



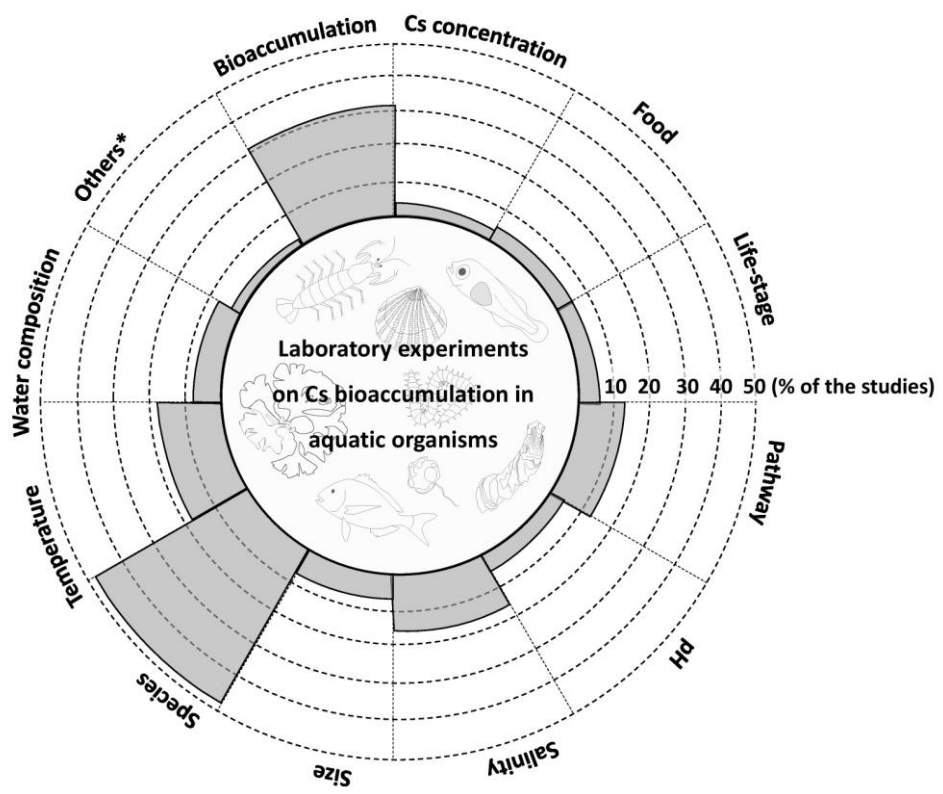
862 Figure 3



863 Figure 4



864 Figure 5



865 Figure 6

866 Table 1. Summary of the abiotic factors whose effects have been studied in relation to the  
 867 bioaccumulation of radiocesium in aquatic organisms

Variable	Pathway	Kingdom	Number of publications
Cs concentration	Food Water	Animalia Plantae Protozoa	4 <sup>a</sup>
Pathway	Food Water Sediment	Animalia	13 <sup>b</sup>
pH	Water	Animalia	4 <sup>c</sup>
Salinity	Food Water Sediment	Animalia Chromista Plantae	15 <sup>d</sup>
Temperature	Food Injection in the blood Oral administration Water	Animalia Bacteria Chromista Plantae	17 <sup>e</sup>
Water composition	Food Sediment Water	Animalia Plantae Protozoa	8 <sup>f</sup>
Others*	Water	Protozoa Plantae	3 <sup>g</sup>

868 <sup>a</sup> (Argiero et al., 1966; Ke et al., 2000; Williams, 1960; Zhao et al., 2001)

869 <sup>b</sup> (Børretzen and Salbu, 2009; Bustamante et al., 2006; Hansman et al., 2018 ; Metian et al., 2011; Pouil et al., 2015; Prihatiningsih et al.,  
 870 2016b; Reinardy et al., 2011; Sezer et al., 2014; Suzuki et al., 1992; Topcuoğlu and Van Downen, 1997; Ueda et al., 1977; Warnau et al.,  
 871 1996; Zhao et al., 2001)

872 <sup>c</sup> (Lacoue-Labarthe et al., 2012, 2018; Morgan et al., 1993; Sezer et al., 2018)

873 <sup>d</sup> (Amiard-Triquet, 1974; Bryan, 1963, 1961; Bryan and Ward, 1962; Carlson and Erlandsson, 1991; Hattink et al., 2009; IAEA, 1975; Ke et  
 874 al., 2000; Pouil et al., 2018a; Prihatiningsih et al., 2016a; Qureshi et al., 2007; Styron et al., 1976; Topcuoğlu, 2001; Wolfe and Coburn,  
 875 1970; Zhao et al., 2001)

876 <sup>e</sup> (Boisson et al., 1997; Bryan, 1965; Bryan and Ward, 1962; Cocchio et al., 1995; Harvey, 1969b; Hiyama and Shimizu, 1964; Hutchins et  
 877 al., 1996a, 1996b, 1998; Lacoue-Labarthe et al., 2012; Peters et al., 1999; Prihatiningsih et al., 2016a; Qureshi et al., 2007; Srivastava et al.,  
 878 1994; Styron et al., 1976; Ugedal et al., 1992; Wolfe and Coburn, 1970)

879 <sup>f</sup> (Bervoets et al., 2003; Cocchio et al., 1995; Fraysse et al., 2002; Hagstroem, 2002; Ke et al., 2000; Srivastava et al., 1990, 1994; Williams,  
 880 1960)

881 <sup>g</sup> (Jeffree et al., 2018; Malek et al., 2004a; Williams, 1960)

882 \* Effects of antibiotics and food preparation

883 Table 2. Summary of the biotic factors whose effects have been studied in relation to the  
 884 bioaccumulation of radiocesium in aquatic organisms

Variable	Pathway	Kingdom	Number of publications
Bioaccumulation capacity	Food Sediment Water Maternal	Animalia Bacteria Plantae	31 <sup>a</sup>
Food	Food	Animalia	5 <sup>b</sup>
Life-stage	Food Water Sediment	Animalia	5 <sup>c</sup>
Size	Oral administration Water	Animalia	6 <sup>d</sup>
Species	Food Oral administration Sediment Water	Animalia Bacteria Chromista Plantae	49 <sup>e</sup>
Sex	Water	Animalia Protozoa Plantae	2 <sup>f</sup>

885 <sup>a</sup> (Adam et al., 2001; Ancellin et al., 1965 ; Cranmore and Harrison, 1975; Evans, 1984; Fisher, 1985; Fowler et al., 1971; Fowler and  
 886 Teyssié, 1997; Garnier-Laplace et al., 1997; Gil Corisco and Carreiro, 1990; Guimarães, 1992; Güngör et al., 2001; Harrison, 1972; Harvey,  
 887 1969b; Hewett and Jefferies, 1976; Ivanov, 1972; Jeffrey et al., 2018, 2015, 2013, 2007, 2006a; Kalaycı et al., 2013; Kimura, 1984; Lacoue-  
 888 Labarthe et al., 2010; Malek et al., 2004; Milcent et al., 1996; Norfaizal, 2010; Onat and Topcuoğlu, 1999; Twining et al., 1996; Varinlioglu  
 889 et al., 2015; Warnau et al., 1999; Woodhead, 1970)

890 <sup>b</sup> (Belivermiş et al., 2017; Bryan, 1961; Bryan and Ward, 1962; Pan and Wang, 2016; Zhao et al., 2001)

891 <sup>c</sup> (Argiero et al., 1966; Bustamante et al., 2006; Kimura and Honda, 1977; Morgan et al., 1993; Suzuki et al., 1992)

892 <sup>d</sup> (Güngör et al., 2001; Ke et al., 2000; Malek, 1999, 1998; Nolan and Dahlgaard, 1991; Ugedal et al., 1992)

893 <sup>e</sup> (Adam and Garnier-Laplace, 2003 ; Amiard-Triquet, 1975; Ancellin and Vilquin, 1968; Avarguès et al., 1972, 1968; Baptist and Price,  
 894 1962; Bonotto et al., 1981, 1978; Boroughs et al., 1957; Bryan, 1961; Bryan et al., 1966; Bryan, 1963; Bryan and Ward, 1962; Corcoran,  
 895 1963; Forseth et al., 1998; Fowler et al., 2004; Fraizier and Vilquin, 1971; Genta-Jouve et al., 2012; Gutknecht, 1965; Harvey, 1969b;  
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900 <sup>f</sup> (Bryan, 1965; Williams, 1960)

901 \*Effects of cell density and sex