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

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Abstract:

A better understanding of the fate of radiocesium in aquatic organisms is essential for making accurate assessments of potential impacts of radiocesium contamination on ecosystems and human health. Studies of the accumulation of ^{134}Cs and ^{137}Cs in diverse biota have been the subject of many field investigations; however, it may often be difficult to understand all the mechanisms underlying the observations reported. To complement field investigations, laboratory experiments allow better understanding the observations and predicting dynamics of Cs within aquatic ecosystems by accurately assessing bioaccumulation of Cs in living organisms. The present review summarizes selected relevant laboratory studies carried out on Cs bioaccumulation in aquatic organisms over a period of more than 60 years. To date, 125 experimental studies have been carried out on 227 species of aquatic organisms since 1957. The present review provides a synthesis of the existing literature by highlighting major findings and identifying gaps of key information that need to be further addressed in future works on this topic. Thus, influences of some environmental parameters such as water chemistry both for marine and freshwater ecosystems, and biotic factors such as the life-stages and size of the organisms on radiocesium bioaccumulation should be examined and become priority topics for future research on Cs accumulation in aquatic organisms.

Keywords: Aquatic biota; Radiocesium; Radiotracer experiment; Radioecology; Review.

1. Introduction

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

Radiocesium isotopes are persistent in aquatic environments (^{134}Cs: $t_{½}$ = 2.06 yr and ^{137}Cs: $t_{½}$ = 30.17 yr), and so can be readily bioaccumulated by aquatic organisms at the bottom of the aquatic food chain (e.g. phytoplankton and macrophytes; Fisher, 1985; Harvey and Patrick, 1967; Heldal et al., 2001; Warnau et al., 1996) and can then be transferred to higher trophic levels such as fish (Pentreath, 1963; Zhao et al., 2001; Mathews and Fisher, 2009). Furthermore, radiocesium concentrations have been measured in the field up to 1000 Bq kg^{-1} (338 Bq kg^{-1} of ^{134}Cs and 699 Bq kg^{-1} of ^{137}Cs) in fish (Chen, 2013; Iwata et al., 2013; Wada et al., 2016). Such observations confirm the importance of aquatic organisms as vectors for bioaccumulation and potential biomagnification of radiocesium (Tateda et al., 2013) as has also been suggested from results of laboratory radiotracer experiments (e.g. Mathews et al., 2008; Mathews and Fisher, 2008; Pan and Wang, 2016; Zhao et al., 2001).

The determination of radiocesium bioaccumulation parameters in aquatic organisms under controlled laboratory conditions can be key to better understanding the significance of field measurements (Warnau and Bustamante, 2007; Wang et al., 2018). Indeed, an experimental radiotracer approach can provide relevant information about contamination pathways or uptake and depuration capacities of exposed organisms (e.g. Pouil et al., 2015; Reinardy et al., 2011; Sezer et al., 2014; Wang et al., 2000). Laboratory experiments allow (1) comparing the bioaccumulation capacities of different marine organisms in fairly comparable contamination conditions, (2) obtaining information about food chain transfer, (3) delineating the major uptake pathway(s) through computation of the data, and (4) providing a clear insight into major biological mechanisms that are activated during pollution events (Metian et al., 2016). In comparison to stable isotope approaches, radiotracer techniques offer several unique advantages, such as cost effectiveness and elevated throughput of samples. Furthermore, gamma-emitting radiotracers allow radioanalysis of live organisms and thus, substantially decrease the number of sacrificed organisms and generate data with reduced biological variability (Warnau and Bustamante, 2007). Laboratory experiments also enable the selection of appropriate candidate species for carrying out biomonitoring programs (e.g. Bervoets et al., 2003; Børretzen and Salbu, 2009; Warnau et al., 1999). Thus, experimental results help to better understand and predict the dynamics of radiocesium in aquatic environments and in biota after a contamination event.

The present review also identifies trends and gaps in the literature, as well as offers an opportunity to outline methodologies for measuring the bioaccumulation of radiocesium in marine organisms. The experiments outlined in the database and built into this review helped scientists understand the effects of radiological depositions by controlling environmental parameters in a laboratory. Such works which were carried out under controlled conditions were then compared to field data that were collected after accidents such as Chernobyl and Fukushima for making an even deeper analysis of the consequences of a nuclear accident on the environment. Furthermore, many different models have been developed to simulate additional depositions or to outline the pathways by which contamination moves through an organism and where the contamination will accumulate in these organisms. The necessary inputs for these models are also outlined in works held within this database. This review therefore aims at outlining key results of previous experiments as a toolbox for modelers.

2. Material and methods

2.1. Literature search

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

2.2. Database construction

A bibliographic database (see supplementary material) was assembled to archive all publications (book chapters, conference articles, peer-reviewed articles, reports and theses) that address radiocesium bioaccumulation in aquatic organisms under controlled conditions. A total of 125 publications was finally selected.

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

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

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

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

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

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

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

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

3. Global overview of the database

3.1. History of radiocesium bioaccumulation in experimental studies

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

The importance of studies outlining the bioaccumulation of radiocesium becomes even more obvious when assessing the effects of nuclear accidents. Nevertheless, our findings indicated that the number of publications on this topic has not increased significantly following the Chernobyl nuclear accident in 1986 and later after the Fukushima Daiichi nuclear power plant accident in Japan in 2011 (Fig. 1A). It was expected that there would be a sharp increase in the number of papers written following events such as Chernobyl and Fukushima. However, since 1990, there is a consistency in the publications per decade carried out on the experimental bioaccumulation of radiocesium in aquatic organisms (Fig. 1B). While the focus of studies changes over time, the overall objective of the selected publications appears to remain constant. It seems that since the early stages of nuclear power and weapons development (in the 1950-1960's), the majority of research was focused on the accumulation and retention of radiocesium in different types of marine organisms, firstly through empirical approaches (e.g. Bryan and Ward, 1962; Gutknecht, 1965; Jefferies and Hewett, 1971) and more recently using kinetic models (e.g. Belivermiş et al., 2017; Metian et al., 2016; Sezer et al., 2014).

3.2. Biological models (species and taxa)

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

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

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

3.3. Exposure pathways

The experimental study of radiocesium bioaccumulation can be done through (1) an exposure via a unique pathway (so-called single-pathway approach) or (2) several pathways studied separately or together (so-called multiple-pathway approach). The latter experimental approach allows a more comprehensive understanding of the mechanisms of bioaccumulation in a given species, and it is useful to estimate, through a modelling approach, the main pathways of bioaccumulation (Børretzen and Salbu, 2009; Hewett and Jefferies, 1978; Metian et al., 2016; Pentreath, 1973; Pouil et al., 2015).

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

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

4. Factors influencing radiocesium bioaccumulation

4.1. Temperature

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

Interestingly, a meta-analysis based on information available in the database (see Supplementary Material) was performed and revealed no general trend regarding the influence of temperature on uptake (uptake rate and CF) and retention ($T_{b1/2}$, absorption efficiency) of dissolved radiocesium in aquatic organisms. Overall, these results suggest that the effects of temperature, the most studied abiotic factor, on the bioaccumulation of radiocesium are complex and dependent both on the species considered and other environmental factors. For these reasons modeling the effects of temperature requires special attention.

4.2. Salinity

Salinity is a master variable for coastal and marine ecosystems and can play an important role in the chemical speciation of many elements and also affect the physiology of aquatic organisms. This review showed that 15 publications explicitly dealt with salinity in Animalia, Chromista and Plantae species. Most of these studies found effects of salinity on the bioaccumulation of dissolved radiocesium in several taxa. For ray-finned fish species, there are contradictory findings on the effects of salinity on bioaccumulation of dissolved radiocesium, with an increase in CFs observed at the lowest (15 psu, Zhao et al., 2001), highest (35 psu, IAEA, 1975) or intermediate (29 psu, Prihatiningsih et al., 2016a) salinity conditions. In addition, Hattink et al. (2009) have demonstrated that radiocesium uptake in European seabass is independent of salinity (approx. 1-35 psu) as well as the assimilation efficiency (AE) of ingested radiocesium. Nevertheless, in turbot *Scophthalmus maximus*, Pouil et al. (2018) found a significantly higher AE when fish are exposed to low salinity conditions (10 and 25 psu) compared to the control condition (salinity: 38 psu). These results show that effects of salinity are species-dependent, likely in relation to their salinity tolerance ranges. Among invertebrates, Topcuoğlu (2001) found that bioaccumulation of radiocesium in the isopod *Idothea primastica* was significantly enhanced in a low salinity regime (approx. 7 psu). Similar findings were highlighted for the lugworm *Arenicola marina* (Amiard-Triquet, 1974). Generally, the same results have been reported for bivalves (Ke et al., 2000; Qureshi et al., 2007; Wolfe and Coburn, 1970) and malacostracans (Bryan, 1961; Bryan and Ward, 1962; Bryan, 1963) for salinity values from approx. 1 to 35 psu. Not surprisingly, radiocesium CFs in algae are usually higher at the lower salinity (3.5-8 psu, Carlson and Erlandsson, 1991; Styron et al., 1976). Although there are some contradictory results, especially in fish, most experimental studies have shown that salinity strongly affects the bioaccumulation of radiocesium in aquatic organisms which usually results in an increase in concentration in low salinity regimes (< 15 psu). Various mechanisms have been proposed to explain that organisms living in low salinity regimes generally contain higher radiocesium concentrations. Indeed, salinity can affect physiological conditions of the organisms (e.g. cell volume, membrane permeability, water pumping rate) and chemical element speciation. Furthermore, an increase in cation concentrations with increasing salinity affects the permeability of the membranes and increases the competition for binding sites.

4.3. Water composition

The effect of water composition on radiocesium bioaccumulation in aquatic organisms has been studied from different angles. Since Cs is an alkali metal, it is highly soluble in the water and exists almost exclusively as the monovalent cation Cs^+ in aqueous solution. This dissolved form of radiocesium is chemically similar to the potassium (K) and sodium (Na) ions. Effects of K concentrations in water on the bioaccumulation of Cs were considered in 7 publications. In three species of ray-finned fish exposed via the dissolved pathway, an increase in K concentrations led to a decrease in radiocesium uptake (Cocchio et al., 1995; Srivastava et al., 1990, 1994). Similar findings have been reported for the green-lipped mussel *Perna viridis* (Ke et al., 2000) and for the microcrustacean *Daphnia magna* (Hagstroem, 2002). A plausible explanation for such findings may be competitive inhibition of radiocesium uptake by the high K concentrations (Bryan, 1963). However, a larger magnitude of effects of K concentration has been observed in microorganisms (Plantae and Protozoa). Thus, Hagstroem (2002) has found a negative effect of the increase of K in water on the uptake of the dissolved radiocesium in two species of Chlorophyta, *Chlamydomonas noctigama* and *Scenedesmus quadricauda*, while Williams (1960) showed a positive relationship between K concentrations and the uptake of radiocesium in one species of Chlorophyta, *Chlorella pyrenoidosa*, and a species of Euglenozoa, *Euglena deses*. Considering the assumption that K could change the distribution of radiocesium between the solid and the liquid phase of sediment, Bervoets et al. (2003) studied the accumulation of radiocesium from the sediment in the benthic midge larvae *Chironomus riparius* and found it was unaffected by K concentrations in water. The influence of sodium (Na) concentrations has also been examined, and some authors have demonstrated that Na has little effect on the bioaccumulation of Cs (Hagstroem, 2002), and that is also true for calcium (Cocchio et al., 1995). In addition, Fraysse et al. (2002) showed the absence of an effect of the dissolved trace metals Cd and Zn on the bioaccumulation of radiocesium in the zebra mussel *Dreissena polymorpha*, contrary to what they observed for two other radionuclides (^{57}Co and ^{110m}Ag). All of these results demonstrate that there is ultimately insufficient experimental works investigating the effects of water chemistry on the bioaccumulation of radiocesium, such as, for example, water hardness. Nevertheless, field investigations have shown that, in freshwater ecosystems, water hardness and conductivity play a role in the level of radiocesium in biota (Hakanson et al., 1992; Särkkä and Luukko, 1995).

4.4. Cs concentration

The influence of environmental stable Cs concentrations on subsequent radiocesium bioaccumulation in aquatic organisms has been studied in fish and bivalves, as well as in phytoplankton and bacteria species. In the mangrove snapper *Lutjanus argentimaculatus*, Zhao et al. (2001) found that radiocesium CFs were not influenced by the stable Cs concentrations (0.006-0.6 mM), whereas the calculated uptake rate (k_u) increased linearly with increasing ambient stable Cs concentration. Similar findings have been reported for the green-lipped mussel (stable Cs concentrations of 0.006-0.6 mM; Ke et al. 2000). Such results are in agreement with Argiero et al. (1966) who did not find any effect of external radiocesium concentration on CF in another bivalve species (*Mytilus galloprovincialis*). For microorganisms, Williams (1960) found in the bacteria *Euglena deses* and in the microalga *Chlorella pyrenoidosa* that the uptake of dissolved radiocesium was directly proportional to the ambient stable Cs concentration in water (0-2.5 mM). All of these results indicate that increasing ambient Cs concentrations lead to a positive linear response of the radiocesium uptake rate in the aquatic organisms studied. In other words, this suggests that, for the limited number of aquatic organisms studied, equilibration of radiocesium uptake was not reached within the experimental Cs concentrations tested (broad range from 0 to 2.5 mM).

4.5. pH

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

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

4.6. Species

Interspecific difference is one of the most studied factors influencing the bioaccumulation of radiocesium in aquatic organisms (49 publications). Thus, bioaccumulation of radiocesium in 190 species of Animalia, Bacteria, Chromista, Plantae has been compared in the literature. Examples showing differences in bioaccumulation in phylogenetically-close species are numerous. Differences between these species have been found after exposures from food (e.g. Hewett and Jefferies, 1978; Pan and Wang, 2016; Warnau et al., 2002), sediment (e.g. Amiard-Triquet, 1975; Marc Metian et al., 2016; Ueda et al., 1978) and water (e.g. Baptist and Price, 1962; Bryan and Ward, 1962; Harvey and Patrick, 1967; Heldal et al., 2001). Linking taxonomy, phylogeny and radiocesium bioaccumulation can be very complex (Brown et al., 2019). Nevertheless, a simple meta-analysis of data from the database revealed differences in the CFs and AEs observed among the different kingdoms (Animalia, Bacteria, Chromista and Plantae). Thus, CFs of dissolved radiocesium in these different taxonomic groups were ranked in the following decreasing order Bacteria ≥ Chromista > Animalia ≥ Plantae ($p < 0.05$, Fig. 4A). AEs of ingested radiocesium in the different classes were ranked in the following decreasing order Asteroidae ≥ Elasmobranchii > Actinopterygii ≥ Gastropoda ≥ Malacostraca ≥ Polychaeta ≥ Bivalvia ($p < 0.05$, Fig. 4B). These meta-analyses allow highlighting global trends, but their interpretation must take into account the large disparities in the study of the different taxonomic groups (see Section 3.2) that may affect results.

4.7. Size and life-stages

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

For molluscs, a higher ability to bioaccumulate dissolved radiocesium has been shown in smaller (= younger) individuals of bivalve mussels (*M. galloprovincialis* and *P. viridis*) through the measurements of k_u or CF (Argiero et al., 1966; Ke et al., 2000). Nevertheless, Güngör et al. (2001) and Nolan and Dahlgaard (1991) have shown more contrasting results with no significant difference of CF or $T_{b1/2}$ between mussels (*M. edulis* and *M. galloprovincialis*) of different sizes. Such differences observed for the same species (or a phylogenetically closely related species) can be explained, at least partially, by the size ranges used which vary greatly in these publications. Indeed, while Ke et al. (2000) used individuals of 3-4 cm shell length, Güngor et al. (2001) and Nolan and Dahlgaard (1991) have made their observations on a larger size range (approx. 2.8-6.5 cm shell length. In the cephalopod *S. officinalis*, Bustamante et al. (2006) demonstrated a higher assimilation efficiency (i.e. AE) and retention (i.e. $T_{b1/2}$) of ingested radiocesium in juveniles compared to adults indicating that the greater ability of smaller (= younger) individuals to bioaccumulate radiocesium can be also true when radiocesium is taken up from food. The authors stated that these differences could be related to the decrease of digestive metabolism with age in cephalopods, with the consequence of a higher efficiency of digestion process in smaller (= younger) individuals.

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

4.8. Food quality and starvation

Food quality (type of natural prey and compounded food) is well-known to affect the assimilation of trace elements in aquatic organisms. For radiocesium, effects of food quality have been investigated in ray-finned fish and in several invertebrate species (arthropods and molluscs). Zhao et al. (2001) found in the mangrove snapper (*L. argentimaculatus*) that there was no significant difference in radiocesium AE when fed with different prey. Similar results were found in three species of ray-finned fish with contrasting feeding habits (Pan and Wang, 2016). In bivalves, no significant effect of food has been shown in the Manila clam *Ruditapes philippinarum* although AE varied slightly between the experimental treatments (Belivermiş et al., 2017). Thus, interestingly, the type of food seems to have very limited effect on the assimilation of radiocesium in aquatic organisms. Furthermore, uptake of dissolved radiocesium was not affected by starvation as has been shown for several species of Malacostraca (Bryan, 1961; Bryan and Ward, 1962).

4.9. Trophic ecology

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

5. Organotropism of Cs in aquatic organisms

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

6. Gaps and perspectives

Much has been done over the last 60 years in radioecological research to better assess radiocesium dynamics, with a main focus on fish and a few abiotic parameters. Figure 6 highlights the research efforts on bioaccumulation and on a series of factors influencing the bioaccumulation of radiocesium in aquatic organisms. It also brings gaps of knowledge to the fore, identified by the limited number of studies and/or unclearly explained effects. It is especially true for (1) some abiotic environmental factors such as water chemistry (e.g. chemical composition and pH) and (2) biotic factors (the life-stages and size of the organisms). All the listed factors should be looked at and become priority topics for further investigations on radiocesium accumulation in aquatic organisms. Therefore, future research on this topic should include the effect of abiotic factors (single or multiple factors) and examine some species that have not been investigated to date. For instance, there is a need to focus future work on small organisms that constitute food for fish, and to investigate some abiotic factors that have not been examined to date such as seawater deoxygenation. In addition, it would be important to better assess the main uptake pathway in a wider range of taxa, not considering only water and food but also sediment.

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

7. Conclusion

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

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Captions to figures

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

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

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

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

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

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

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

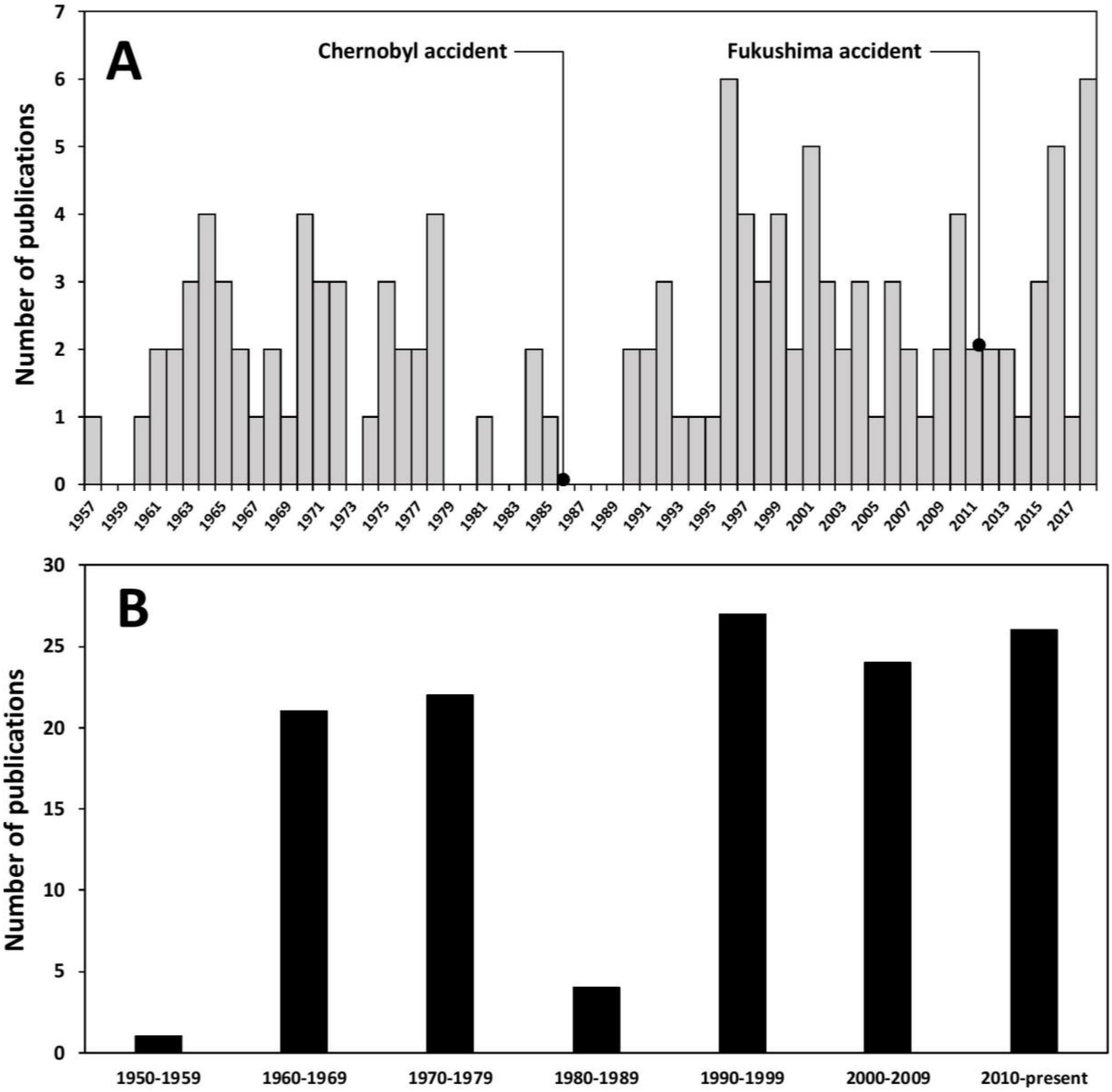
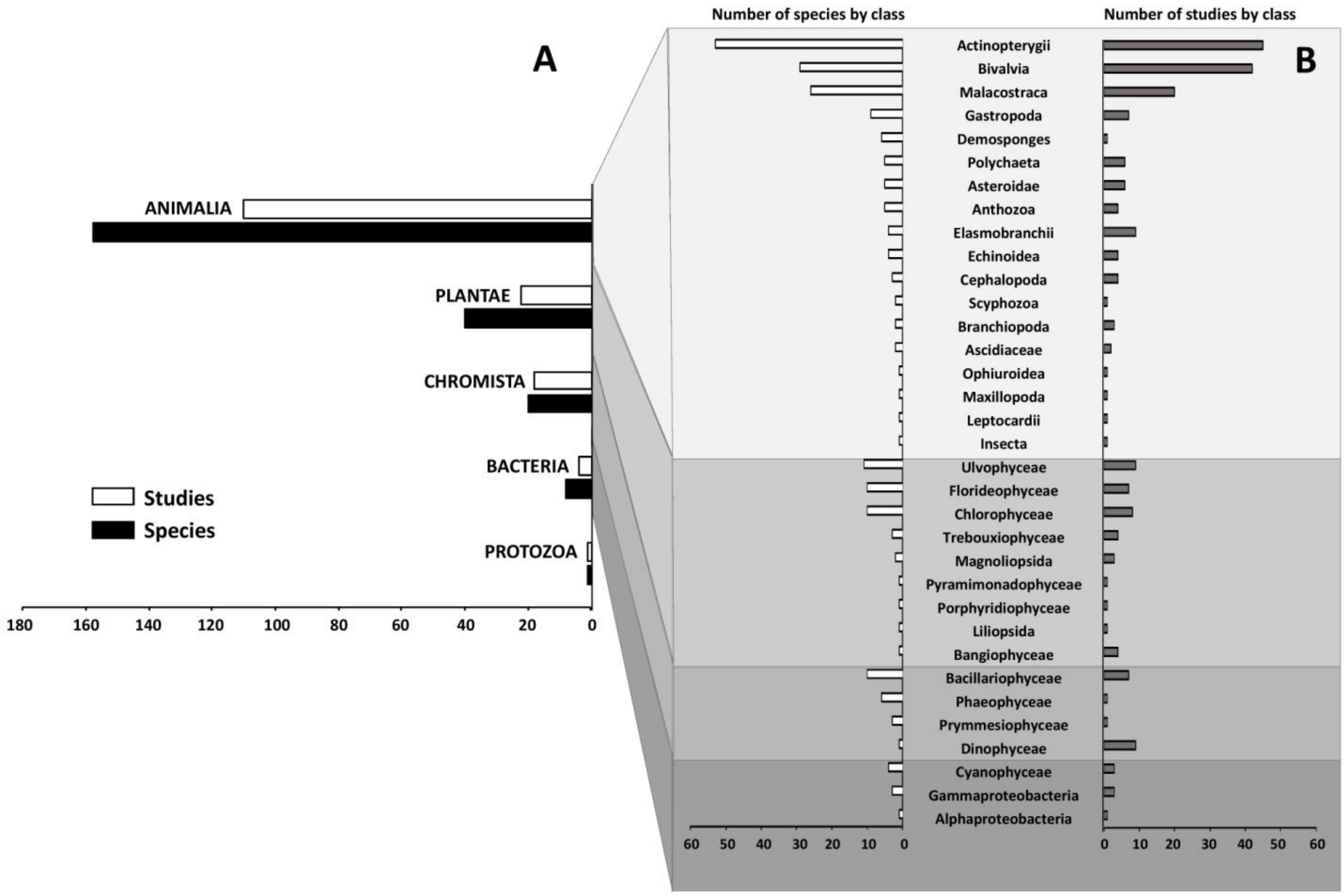
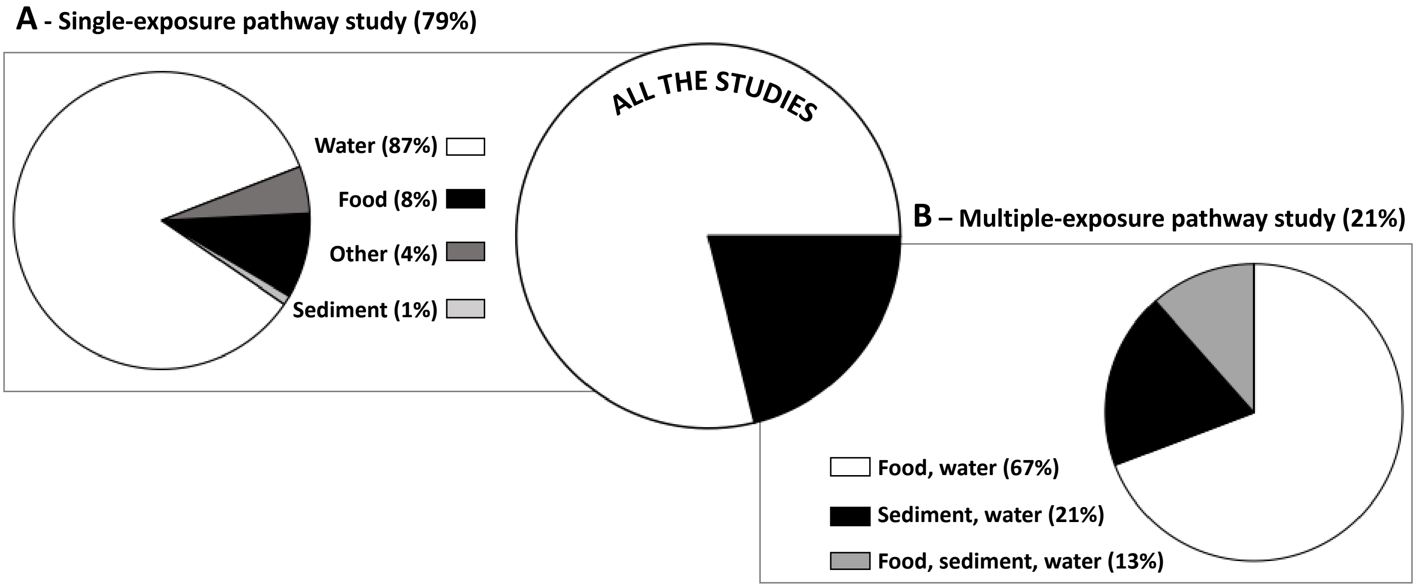

A
Chernobyl accident
Fukushima accident
Number of publications
0
1
2
3
4
5
6
7
1957
1959
1961
1963
1965
1967
1969
1971
1973
1975
1977
1979
1981
1983
1985
1987
1989
1991
1993
1995
1997
1999
2001
2003
2005
2007
2009
2011
2013
2015
2017
B
Number of publications
0
5
10
15
20
25
30
1950-1959
1960-1969
1970-1979
1980-1989
1990-1999
2000-2009
2010-present

Figure 1


A
B
ANIMALIA
PLANTAE
CHROMISTA
BACTERIA
PROTOZOA
Studies
Species
180 160 140 120 100 80 60 40 20 0
Number of species by class
Number of studies by class
Actinopterygii
Bivalvia
Malacostraca
Gastropoda
Demosponges
Polychaeta
Asteroidae
Anthozoa
Elasmobranchii
Echinoidea
Cephalopoda
Scyphozoa
Branchiopoda
Ascidiaceae
Ophiuroidea
Maxillopoda
Leptocardii
Insecta
Ulvophyceae
Florideophyceae
Chlorophyceae
Trebouxiophyceae
Magnoliopsida
Pyramimonadophyceae
Porphyridiophyceae
Liliopsida
Bangiophyceae
Bacillariophyceae
Phaeophyceae
Prymmesiophyceae
Dinophyceae
Cyanophyceae
Gammaproteobacteria
Alphaproteobacteria
60 50 40 30 20 10 0
0 10 20 30 40 50 60

861 Figure 2


A - Single-exposure pathway study (79%)
Water (87%)
Food (8%)
Other (4%)
Sediment (1%)
ALL THE STUDIES
B – Multiple-exposure pathway study (21%)
Food, water (67%)
Sediment, water (21%)
Food, sediment, water (13%)

862 Figure 3

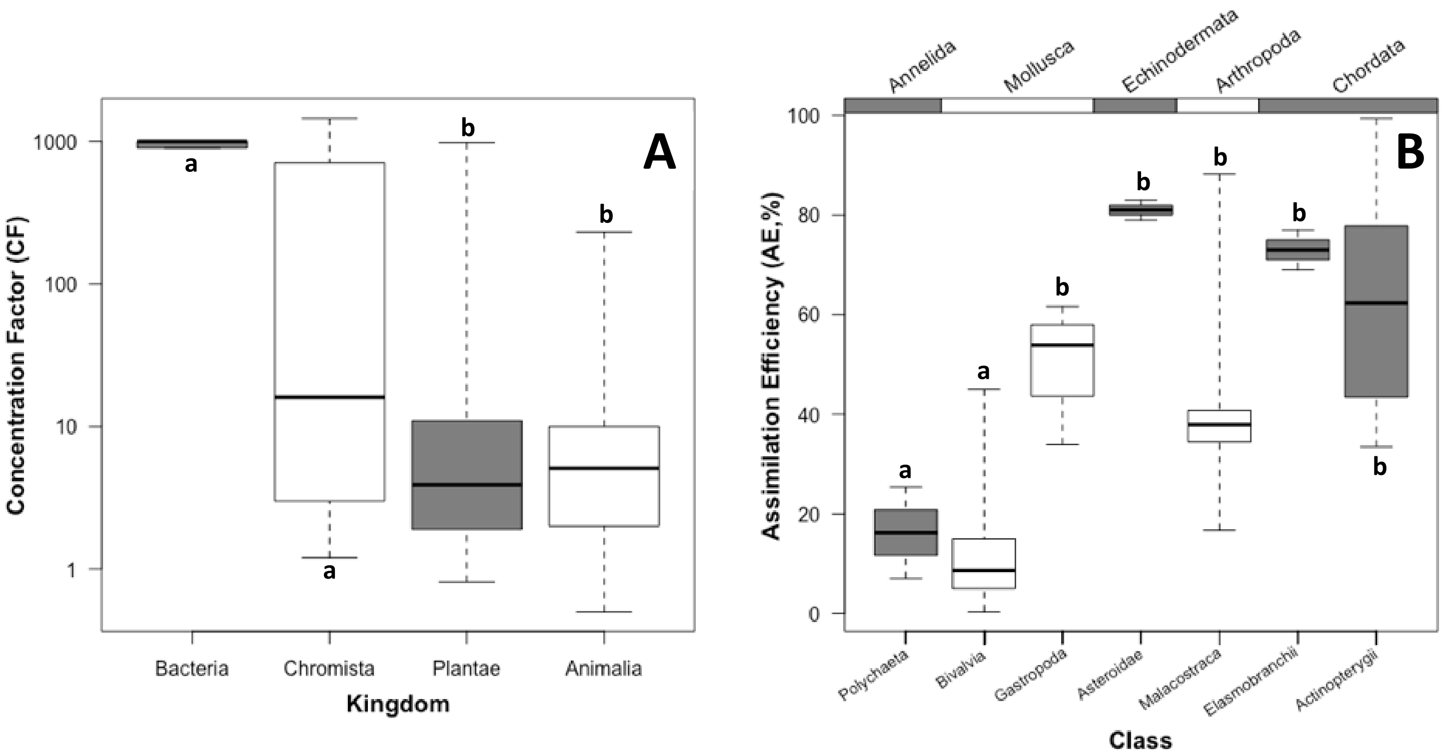

A
Concentration Factor (CF)
1000
100
10
1
a
a
b
b
Bacteria
Chromista
Plantae
Animalia
Kingdom
B
Annelida
Mollusca
Echinodermata
Arthropoda
Chordata
Assimilation Efficiency (AE,%)
100
80
60
40
20
0
a
a
b
b
b
b
b
Polychaeta
Bivalvia
Gastropoda
Asteroidae
Malacostraca
Elasmobranchii
Actinopterygii
Class

Figure 4

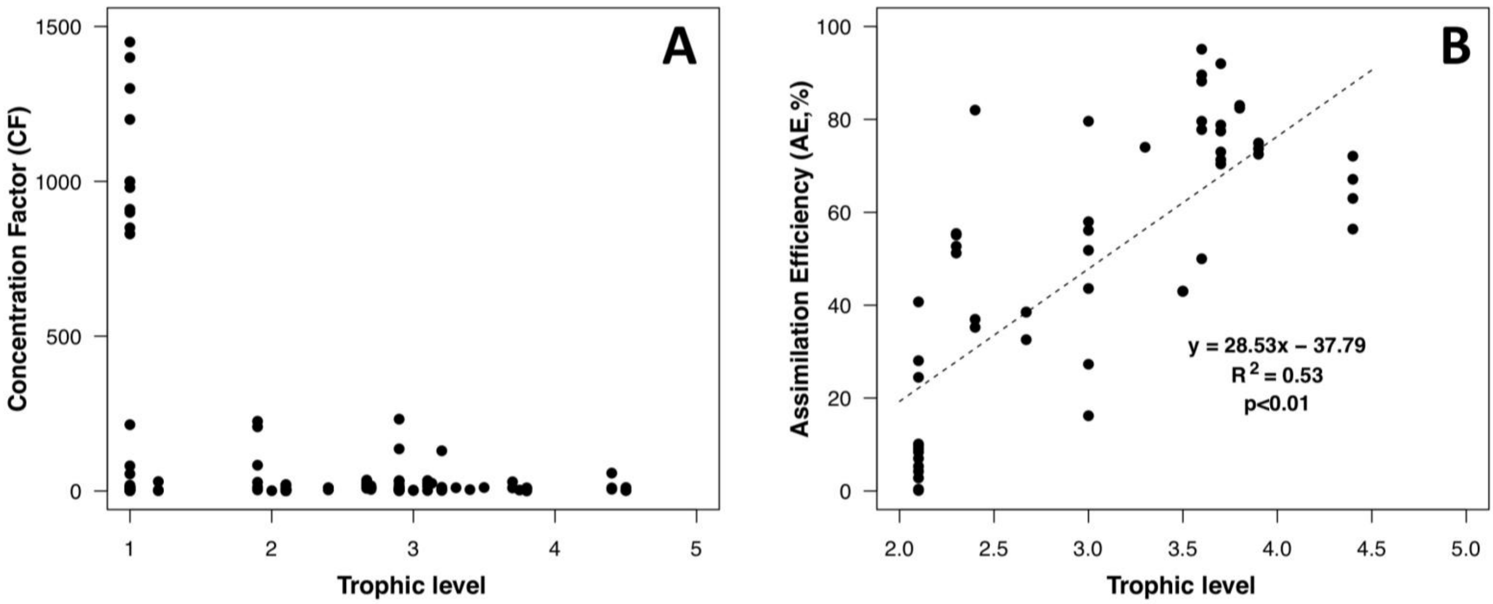

A
Concentration Factor (CF)
1500
1000
500
0
1
2
3
4
5
Trophic level
B
Assimilation Efficiency (AE,%)
100
80
60
40
20
0
2.0
2.5
3.0
3.5
4.0
4.5
5.0
Trophic level
y = 28.53x − 37.79
R 2 = 0.53
p<0.01

Figure 5

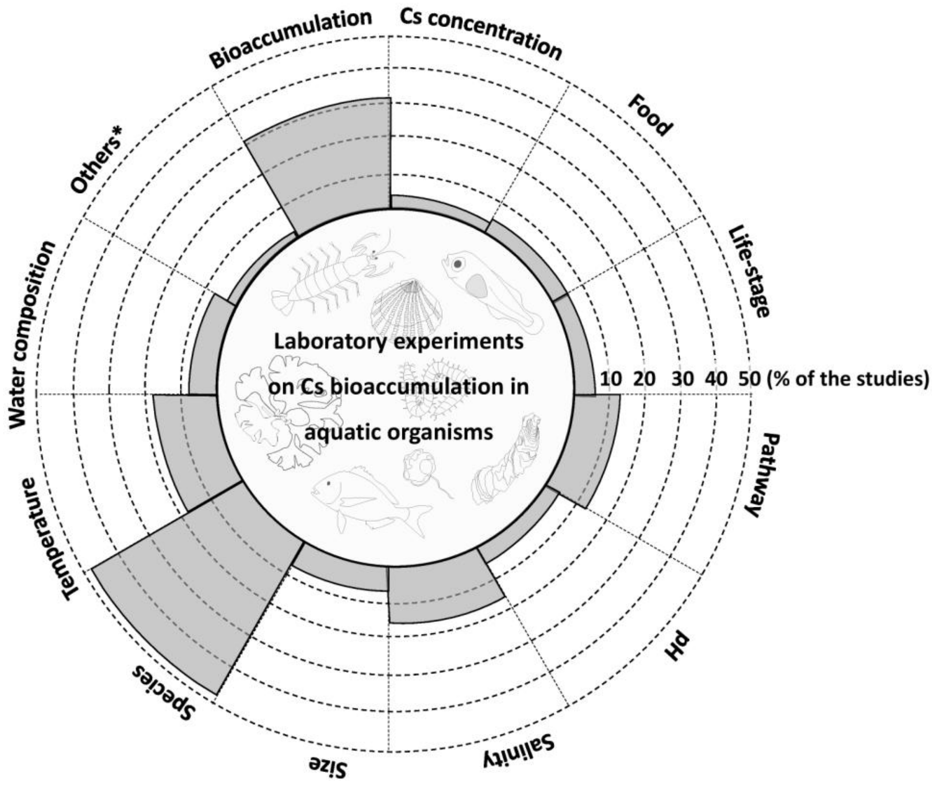

Bioaccumulation
Cs concentration
Food
Life-stage
Pathway
pH
Salinity
Size
Species
Temperature
Water composition
Others*
10 20 30 40 50 (% of the studies)
Laboratory experiments
on Cs bioaccumulation in
aquatic organisms

Figure 6

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

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

[a] (Argiero et al., 1966; Ke et al., 2000; Williams, 1960; Zhao et al., 2001)

[b] (Børretzen and Salbu, 2009; Bustamante et al., 2006; Hansman et al., 2018 ; Metian et al., 2011; Pouil et al., 2015; Prihatiningsih et al., 2016b; Reinardy et al., 2011; Sezer et al., 2014; Suzuki et al., 1992; Topcuoğlu and Van Dowen, 1997; Ueda et al., 1977; Warnau et al., 1996; Zhao et al., 2001)

[c] (Lacoue-Labarthe et al., 2012, 2018; Morgan et al., 1993; Sezer et al., 2018)

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

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

[f] (Bervoets et al., 2003; Cocchio et al., 1995; Fraysse et al., 2002; Hagstroem, 2002; Ke et al., 2000; Srivastava et al., 1990, 1994; Williams, 1960)

[g] (Jeffree et al., 2018; Malek et al., 2004a; Williams, 1960)

[*] Effects of antibiotics and food preparation

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

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

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

[b] (Belivermiş et al., 2017; Bryan, 1961; Bryan and Ward, 1962; Pan and Wang, 2016; Zhao et al., 2001)

[c] (Argiero et al., 1966; Bustamante et al., 2006; Kimura and Honda, 1977; Morgan et al., 1993; Suzuki et al., 1992)

[d] (Güngör et al., 2001; Ke et al., 2000; Malek, 1999, 1998; Nolan and Dahlgaard, 1991; Ugedal et al., 1992)

[e] (Adam and Garnier-Laplace, 2003 ; Amiard-Triquet, 1975; Ancellin and Vilquin, 1968; Avarguès et al., 1972, 1968; Baptist and Price, 1962; Bonotto et al., 1981, 1978; Boroughs et al., 1957; Bryan, 1961; Bryan et al., 1966; Bryan, 1963; Bryan and Ward, 1962; Corcoran, 1963; Forseth et al., 1998; Fowler et al., 2004; Fraizier and Vilquin, 1971; Genta-Jouve et al., 2012; Gutknecht, 1965; Harvey, 1969b; Harvey and Patrick, 1967; Heldal et al., 2001; Hewett and Jefferies, 1978; Hiyama and Shimizu, 1964; IAEA, 1975; Jefferies and Hewett, 1971; Jeffree et al., 2010, 2006b; King, 1964; Lacoue-Labarthe et al., 2018 ; Lemée et al., 1970; Mathews et al., 2008; Metian et al., 2016, 2005; Morgan, 1964; Pan and Wang, 2016; Polikarpov, 1964, 1961; Pouil et al., 2018b ; Styron et al., 1976; Suzuki et al., 1992, 1978; Topcuoğlu, 2001; Ueda et al., 1978; Vogel and Fisher, 2010; Wang et al., 2016, 2000, Warnau et al., 2002, 1996)

[f] (Bryan, 1965; Williams, 1960)

*Effects of cell density and sex