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To cite this version:

A Delaval, C Duffa, Olivier Radakovitch. A review on cesium desorption at the freshwater-seawater interface. Journal of Environmental Radioactivity, 2020, 218, pp.106255. 10.1016/j.jenvrad.2020.106255 . hal-03215088

A REVIEW ON CESIUM DESORPTION AT THE FRESHWATER-SEAWATER INTERFACE

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Acknowledgments :

The authors are indebted to the Institute for Radiological Protection and Nuclear Safety (IRSN) and to Region Sud (Provence-Alpes-Côte d'Azur) authorities for the PhD funding. We thank the CRIS (Centre de ressources en informations scientifiques et techniques) of IRSN for their help to provide various documents. This study was conducted within the Rhône Sediment Observatory (OSR) program, a multi-partner research program funded through Plan Rhône of the European Regional Development Fund (ERDF), Agence de l'eau

Rhône Méditerranée Corse, CNR, EDF and three regional councils (Region Auvergne-Rhône-Alpes, PACA and Occitanie).

Introduction

The pollution of marine ecosystems by radionuclides is a major concern for society since the beginning of the nuclear era. It may be due to different sources including: *in-situ* releases from coastal nuclear power-plants, accidents associated with nuclear vessels (vessels, missils), Naturally Occuring Radioactive Materials (NORMs) related to oil or gas production and direct or indirect global releases associated with nuclear tests or accidents on installations. A direct input is provided by atmospheric fallout and/or offshore releases (Fukushima), while an indirect input is due to the transport by rivers, collecting radionuclides from watersheds consequently to atmospheric fallout. Finally, submarine groundwater discharge may constitute a very specific direct input (Sanial et al., 2017).

Cesium radioactive isotopes (^{134}Cs and ^{137}Cs) have been extensively monitored in the environment due to their significant radioecological hazard (Garnier-Laplace et al., 2011) and their persistence (half-life of 2.4 and 30.2 years respectively). They are produced through uranium fission within nuclear reactors and thus can be found in both accident or authorized releases.

In the case of accidents and atmospheric nuclear testing, radiocesium has been spread over large spatial scales by the way of atmospheric deposition (Mattsson and Moberg, 1991), but rivers constitute an additional input to the sea (Trapeznikov et al., 1994; Yamashiki et al., 2014). These rivers bring radiocesium through the releases of reprocessing and power plants as well as by the runoff of contaminated watersheds like in Chernobyl or Fukushima (Walling and He, 1999; Garcia-Sanchez and Konoplev, 2009;

Sakaguchi et al., 2018). Cesium exists in rivers as dissolved Cs^+ with small tendency to form colloids (Onishi et al., 1981; Eyrolle and Charmasson 2004) but is mainly transported in particular form (Takahashi et al., 2017) because of its high affinity for clay minerals (Torstenfelt et al., 1982; Fan et al., 2014;)

At the river-sea interface, the important changes in physico-chemical conditions including ionic strength, solution composition and pH may induce the desorption of Cs from particles to the dissolved phase. A direct consequence is the shift of its distribution coefficient Kd (ratio between solid and liquid activities, L/Kg), decreasing from $6{,}66.10^3$ - $1{,}35.10^5$ in freshwater (Tomczak et al., 2019) to $4{,}5.10^2$ -2.10^3 in seawater (IAEA, 2004; Tagami and Uchida, 2013)

This desorption has been highlighted in laboratory experiments and *in-situ* studies, through the monitoring of dissolved activities (Matishov et al., 2006 ; Kakehi et al., 2016) or sediment inventories (Kusakabe et al., 2013). It is well recognized that clay minerals are the most important solid phase for the adsorption of radiocesium, but a fraction may also be attached to organic material, at least in contaminated watersheds near Fukushima (Naulier et al., 2017). For clays, illite is the most important and strong absorbent but kaolinite, smectite and vermiculite are also active, as well as biotite for phyllosilicate (Okumura et al., 2018)),

The negatively charged basal oxygen surfaces found on clay planar sites can form strong inner-sphere complexes with monovalent cations with low hydration energy, such as K^+,NH_4^+, Rb^+ and Cs^+ (Sposito et al., 1999). These sites have usually low affinity due to their low selectivity (Wauters et al., 1996), but they adsorb Cs^+ more efficiently than the other cations due to its lowest hydration energy (Nakao et al., 2014). This cation uptake can be effective within a few hours (Onishi et al., 1981). Other sorption occurred on edge sites, hydrated interlayer sites or frayed edge sites, corresponding to weathering fronts of micaceous minerals (Okumura et al., 2018). Finally, interlayer sites are not accessible to hydrated cations with large effective ionic radii, but are accessible to easily dehydrated cations such as Cs^+ (Zachara et al., 2002). This binding could result in a strong fixation, similar to those of native stable Cs already present in the mineral (Yin et al., 2016). However, binding on both planar and interlayer sites are partially reversible, and the increase of competitive ions in seawater (K^+, Na^+ or NH_4^+) moves the equilibrium towards a release of Cs^+, providing an additional input to the dissolved phase.

If desorption process at the freshwater-seawater interface has been already demonstrated, there is no consensus on the potential quantity of Cs that can be desorbed depending on the salinity. According to Sakaguchi et al (2018): “the desorbed value remains open for discussion”. Furthermore, the influence of the major cations on the desorption efficiency is not completely clear. According Yin et al (2016), questions remain on how each site is likely to desorb and on the various associated kinetic rates.

This paper provides a review on experimental laboratory studies conducted on Cs desorption in seawater. It aims to precise this process by identifying the values that can be expected according to the salinity, by characterizing the parameters of influence and by underlining unknowns for an eventual modelling work.

1 Material and methods

1.1 Literature search

All laboratory experiments exposing particles to a Cs (stable or radioactive) contamination in freshwater or seawater and then desorbing in seawater media were examined. The information required were the distribution of Cs between the solid and the dissolved phase.

Searches were performed to list all experimental studies carried out on stable and radioactive cesium desorption from particles samples in salt water. We included peer-reviewed and conferences papers, thesis and technical reports from 1950 to present. The databases used were Google Scholar, Scopus, and the CRIS (Centre de ressources en informations scientifiques et techniques) of IRSN.

Searches were performed using the keywords: “Caesium”, “cesium”, and “desorption”, ”remobilization”, ”leaching”, ”distribution” and “saltwater”, ”seawater”, ”sea”, ”salinity”. Studies reporting distribution of radiocesium between solid and liquid phases in the field were not considered because of the lack of information on duration of desorption, delay between adsorption and desorption as well as the salinity of the media. We focused only on experimental laboratory approach that explicitly addressed cesium desorption in saltwater (minimal salinity of 1). Three studies which did not provide the fraction of desorbed Cs from samples after the desorption experiment (or a way to obtain it such as graphs, Kd, or activities in both phases before and after experiment) were excluded. The completeness of the results obtained was considered as satisfactory when no more relevant references on the topic could be found in the selected studies. This is the “snowballing approach” proposed by (Sayers, 2007).

1.2 Database construction

A bibliographic database was created to list all publications concerning cesium desorption from particles samples in saltwater according to the enounced modalities, leading to a total of 32 publications (Table 1).

Different kinds of information were extracted from this database and reported into sections described hereunder. Some are presented in Table 1:

- Paper information and objectives of the experiment: This section includes reference information such as title, year and authors of the publication.
- Sampling location information: Geographical information on the sampling sites as well as available information on main physico-chemical parameters: grain-size, cation exchange capacity (CEC), organic content and clay content. It is important to note that samples can be different and include suspended particles in the water column, sediments or soils. The results obtained on samples that were modified are not taken into account (sieving of grain-size or removal of specific mineral phases; Ouaniche, 1984; El Assy et al., 1991).
- Experimental design: This section focused on the materials and methods information such as the conditions of cesium uptake and the isotope used. Four main experimental designs were found and differentiated for our work (see the scheme of their classification on Figure 2).
    - ✓ 1) desorption of Stable Cesium **(SCs)** corresponding to ^{133}Cs naturally incorporated into the mineral material of the particles (not spiked). A good example of the experiment is provided by Takata et al. (2015).
    - ✓ 2) desorption of RadioCesium **(RCs)** from particles which have been exposed to ^{134}Cs or ^{137}Cs in the field (Yamasaki et al., 2016).

- ✓ 3) desorption of RadioCesium from particles spiked (^{134}Cs or ^{137}Cs) in Laboratory and in Freshwater media (**RCs.Lab.Fw**; ex. Onodera et al., 2017).
- ✓ 4) desorption of RadioCesium from particles spiked in Laboratory in Saltwater media at salinity>1 (**RCs.Lab.Sw**; ex. Oughton et al., 1997).

- Experimental conditions: this section reports experimental parameters such as salinity, ratio between sample mass and volume of seawater, use of natural or artificial seawater, duration of the sorption and desorption phases.
- Results collection methodology: All the results concerning the distribution of cesium between the solid and the dissolved phases after exposition to saltwater were collected. When a study reported multiple values corresponding to change of the experimental conditions or of the sample used, all results were also collected. Results were homogenized into a percentage of Cs activity (or concentration for stable Cs) desorbed in seawater. The section reports the methodology used to obtain this percentage which are values: (i) directly provided by the authors, (ii) calculated from graphical extraction with plot digitizer, (iii) calculated from activities of (iv) the sample or from the Kd provided by the authors.

2 Global overview of the database

2.1 Worldwide and historic concerns on radiocesium desorption at river-sea interfaces

The 32 studies selected correspond to 68 samples and a total of 502 experimental results. 87% of these samples were sediments, 10% suspended particles and 3% soils. The sediments were collected mostly in river (56 %), sea or bays (30%), or estuaries (14%). The majority of studies (14) were interested in the fate of radiocesium sorbed onto soils after atmospheric depositions due to global fallout or nuclear accidents. Also, most of the sediment or suspended particle samples were collected at the output of contaminated watersheds as shown by the locations of the sites (figure 1), mostly related with the Fukushima accident or with nuclear power-plant releases. Only one study reported data directly in relation with the Chernobyl accident (Davison et al., 1993). Four specific studies were also conducted in the Artic Sea in relation with the dumping of radioactive material (17000 containers and 19 vessels) and the potential release from high activity sediments.

The aim of the other works (5) was related to the understanding of stable elements transfer at the river-sea interface, using of their radioactive analogs.

2.2 Influence of the experimental design

The values of Cs desorption percentage are reported in Figure 2. They relate the initial activity onto the particles, i.e. 100% indicates that all radiocesium was released into dissolved phase. They are directly issued from the document or calculated according different methodologies (Table 1). When this value in % was not given in the study, it was calculated as following:

$$\%des = 1 - \frac{A_{solid\ final}}{A_{solid\ initial}} \times 100 \tag{1}$$

Or

$$\%des = 1 - \frac{A_{liquid\ final} * V_{liq\ final}}{A_{solid\ initial} * M_{solid}} \times 100 \tag{2}$$

Where $A_{solid\ initial}$ and $A_{solid\ final}$ are the massic activities (Bq/kg) of the sample before and after desorption, $A_{liquid\ final}$ the dissolved activity after the desorption experiment (Bq/L), M_{solid} the mass of sample used (kg) and $V_{liq\ final}$ the mass of water in the media (L).

The design of the experiment may have an influence on the results. Indeed, while the desorption protocol was similar between experiments, the Cs sorption (in the lab or in the environment) was very different. This could lead to different desorption dynamics depending on the solid to liquid ratio used for the contamination (Onodera et al., 2017), or the ionic composition of the spiked medium as shown by Benes et al. (1992) in freshwater. Consequently, results were separated into 4 different experimental designs defined in section 1.2, and the distribution of the percentage of desorption obtained are presented in Figure 2.

The range of desorption rate for experiments SCs and RCs varied greatly since they are limited between 0 and respectively 10 and 64%. They present however the same modal class at 2-6%. On the other hand, experiments RCs.Lab.Fw and RCs.Lab.Sw show wider and multimodal distributions, extending from 6 to 86 % and 7 to 88%.

Our aim is to evaluate the state of knowledge and major uncertainties on the fate of radiocesium at the river-sea interface. We will focus on the RCs and RCs.Lab.Fw experiments because SCs concerns stable Cs and RCs.Lab.Sw sorption phase occurs in seawater. These two experiments answer to different questions described below. They were designed to give an order of magnitude of the Cs released after a sorption in freshwater, and they did not look at the influence of various physico-chemical parameters.

The experimental design RCs includes (but not only) riverine particles or sediments issued from contaminated watersheds (Sakaguchi et al., 2018) or contaminated directly in the river (Standring et al., 2002). These particles are likely to be transported from river to sea especially during a flood event (Takata et al., 2015).

The experimental design RCs.Lab.Fw aims to reproduce the fate of suspended particles or bottom sediments contaminated after a radioactive release directly into freshwater (river), but desorption is still considered into seawater (Ciffroy et al., 2003). In general, such experiment is more difficult to carry out because it requires an additional sorption phase using radioactive sources. This could explain the lower number of available results.

3 Overview on experiments

Results of the RCs experiments follow a log-normal distribution (Fig. 2 and 3). The percentages of desorption range between 0 and 65%, but the modal class 2-6% gathers more than 47% of the experimental results (93/197). Furthermore, it should be noted that values above 30% are issued from three studies only, and two of them used sediments from Bombay Harbour (Desai et al., 1994; Patel, 1978). These authors reported little information on their samples. The last and maximal value of 65 % comes from the Ravenglass estuary (Stanners and Aston, 1982).

In contrary, results of RCs.Lab.Fw do not follow any classical distribution (Fig 2 and 4) and present many modes. Low desorption below 10% concerned only particles with solid to liquid ratios at or above 10000 mg/L. This concentration is already above most of those observed in rivers, even in floods (Meybeck and Moatar, 2014; Sadaoui et al., 2016). The highest desorption value (86%) was found for particles from the Tamar estuary (Bale, 1987).

3.1 Influence of salinity

There is no significant statistical relation between the salinity and the desorbed fraction or Kd, when the results of the 32 studies related to RCs and RCs.Lab.Fw experiments are plotted together (Fig 5). However, most of these studies tested just one salinity value, and only two of them reported results over a range of salinity. In RCs experiments, Sakaguchi et al. (2018) tested two salinities (16.5 and 33) using successive desorption experiments, and each time the desorption at the 33 salinity medium increases by a factor 1.5-2 compared to those at 16.5. Patel (1978) found a linear relation between salinity and the amount of desorbed Cs for two different samples of the Bombay Harbour for salinities ranging between 9.5 and 38.

For RCs.Lab.Fw, Reynolds and Gloyna (1964) found a positive linear relation between released Cs and salinity, whereas Ciffroy et al. (2003) and Bale (1987) found that the same fraction of Cs is desorbed for low (respectively 13 and 10) and high salinity (34), similarly to a threshold over which salinity had no more influence on the fraction of desorbed Cs. It must be noted however that in both studies, the desorbed fraction increased significantly in the range 0 to 10 of salinity.

It is impossible from these various results to parametrize a relation between the salinity and desorption of Cs from the particles. This might be due to the various solutions chemistry used for these experiments, ranging from natural seawater to monoionic solutions of Na^+. Indeed, the use of monoionic solutions (Na^+, K^+) as desorption medium leads to different effects. Mukai et al. (2018), Onodera et al. (2017), and Desai et al. (1994) observed that for the use of K^+ solutions always reached a plateau in the Cs desorption, which was similar whatever the range of K+ concentrations, respectively 10^{-5} to 1 M, 0.01 to 1 M and 0.01 to 1.6 M. On the other hand, desorption increased linearly with Na^+, using respective ranges of 10^{-5} to 1M, 0.47 to 1M, and 0.01 to 1.6 M. By comparison, the mean concentrations of K^+ and Na^+ in seawater at 35 salinity are 0.01 and 0.47M, and their molar ratio is 47.

These authors explained their results by the fact that Na^+ has a higher hydration energy and the capacity to enhance the interfoliar layer of clays, releasing thus the Cs stored on these sites. On the other hand, K^+ has a lower hydration energy and cannot release this Cs. It is only involved in the cation exchange reaction occurring on surficial sites.

In addition, Mukai et al., (2018), and Onodera et al., (2017) both found that an increase of K^+ concentration from 0.001 to 0.01 M (seawater) combined with Na^+ (at 1M or 0.47 M) decreased the desorption by a factor 2 (60% to 30% for Onodera) or even 4 (80% to 20% for Mukai et al), which is also the amount of desorption observed with natural seawater in their experiment.

These authors and Desai et al., (1994), argue that the “shrinking” or “collapsing” effect of K^+ on the interfoliar gap of clays (Mela, 1962) inhibits Cs release from these interfoliar sites. Increasing salinity undoubtedly influences the desorption due to Na^+ and K^+ and their mechanisms discussed above. However, while desorption increases linearly with Na^+ alone, addition of K^+, around 0.001 to 0.01M range (corresponding to concentrations in seawater of salinity 3.5-35) leads to a desorption plateau, due to its shrinking properties on clay. These results suggest that the desorption no longer significantly increases once a minimal salinity has been reached (>3.5). This is in agreement with Figure 5 showing that Kd are similar in seawater regardless of salinity. This could also explain the salinity threshold found by Ciffroy et al. (2003) and Bale (1987).

Care must thus be taken on desorption results obtained only with Na^+ solution at high concentrations which would lead to an overestimation of the amount of desorbed Cs due to the absence of the inhibitive effect of K^+.

3.2 Influence of the duration of the desorption phase

Studies performing experiments in the form of successive desorption (Sakaguchi 2018; Berthail, 2015; Ouaniche, 1984; Yamasaki et al 2016) were not taken into account as each time that a new desorption media is added, equilibrium is moved, allowing additional Cs to be released.

Durations of desorption phase for RCs ranged between half an hour and 8 months. The desorption was not more important by extending the experiment over one week (Takata et al 2015), one month (Otosaka and Kobayashi, 2013) or two months (Standring et al. (2002). Only Stanners & Aston (1982) found that the amount of released Cs is still increasing after few days in their experiments.

The desorption phase for RCs.Lab.Fw ranged between ten minutes and ten days. The five studies of RCs.Lab.Fw experiments looking at desorption over time showed a fast increase of release within the first hours and then a plateau. The timescales of these five studies differed and we integrated rate laws in order to compare the desorption rates (Jannasch et al., 1988; Simonsen and Saetra, 2019). This approach assumes that trace element uptake and release, including Cs, are the sum of a complex series of first order reversible reactions (Okumura et al, 2018; Jannasch et al., 1988) .

M_d and M_s being the metal in dissolved and solid phases, each reaction can be written as :

$$M_d \overset{k_i}{\underset{k_{-i}}{\rightleftarrows}} M_s$$

Each first-order reaction should verify the linearized equation :

$$-\ln\left(\frac{[M_d]-[M_d]_e}{[M_t]-[M_d]_e}\right)=(k_i+k_{-i}).t \qquad (3)$$

where $[M_d]_e$ is the metal in dissolved phase at equilibrium and M_t the total metal in the solid-liquid system. Demonstration of the equation is available in Simonsen and Saetra, (2019) and Jannasch et al. (1988).

Plotting the left member of equation (3) in function of time provides the slope.

Because the number of experimental data points (n≈5) for each desorption studies was too small, multiple linearization was not possible and the equation was derived only one time for each experiments (16) with the available points. Observations ranged between 0.17 and 168 hours. One analytical curve by experiment is then plotted using exponential transformation of equation (3).The 16 curves are presented in Figure 6.

The integration allows to define 3 cases (Figure 6):

- The equation was verified but a significant intercept (different from zero) exists (7/16 experiments; black curves)
- The equation was not verified (slope not significant at 0.05 error) and a significant intercept exists (7/16; dotted blue curves)
- The equation was not verified (2/16) and there is no significant intercept at 0.05 error (dotted red curves)

The significant intercept observed on 14 studies proves the existence of one or more desorption processes at a timescale inferior to one hour (Jannasch et al., 1988). The fraction of Cs released by these fast processes on the total released is 86 % ± 13 (mean of the intercepts). The non-significant slopes (9/16; case 2 and 3) can be due to the fact that an increasing number of data within the plateau relatively to those in the rising limb minimizes the trend.

These observations lead to the conclusion that Cs desorption in this kind of experimental design is governed by one or more processes on a timescale inferior to one hour releasing in average 86% of the available Cs, and less frequently by an additional (or more) reaction on timescales inferior to 7 days. This justify the use of at least two reactions for an accurate representation of desorption process for timescales over one hour.

In modeling, these reactions are usually taken into account through two successive desorption kinetics involving surficial and interlayer sites of clays, they have led to a two box model (or more) used for both saltwater and freshwater (e.g. Comans and Hockley, 1992).

The kinetic of the first reaction is given by the slope coefficient between time 0 (origin) and the first point of measure. These overall rates (k_1+k_{-1}) obtained for the 16 studies range between 1.10^{-4} and 1.10^{-3} s^{-1} (half-reaction time, $t_{1/2}$, of 0.2 to 2 hours) similar to those obtained in freshwater (Ciffroy et al., 2003; Benes et al., 1992; Comans and Hockley, 1992). They are theoretically related to surficial sites. The second reaction, theoretically related to interlayer sites, presents overall rates ranging between 5.10^{-6} and $1.10^{-5}s^{-1}$ (half-reaction time of two days to one week) in seawater, half an order of magnitude above those in freshwater (1.10^{-6} to 5.10^{-6}); (Benes et al., 1992; Comans and Hockley, 1992; Ciffroy et al., 2003).

These two reactions are considered to occur consecutively (Comans and Hockley, 1992; Oughton et al., 1997; Ciffroy et al., 2001) or in parallel (Benes et al., 1992; Yamamoto et al., 2015). Since the fast desorption rate of surficial Cs is usually two orders of magnitude higher, most of the slowly released pool of Cs within interlayers must remain available even if the reactions are in parallel. In any case, this difference is not so much important for timescales inferior to one week and does not change the goodness of fit of the models (Benes et al., 1992; Borretzen and Salbu, 2002).

3.3 Influence of the cesium distribution into particles prior to desorption

The existence of different sites with their own affinity for Cs implies that preliminar distribution of cesium on these sites influences the quantities and the dynamics of desorbed Cs. When plotting the values of Kd obtained after desorption in seawater from experiment RCs.Lab.Fw in function of their Kd after adsorption in freshwater (8 studies; Fig 7) we observe a positive and significant correlation (R^2=0.43, p-value =2.10^{-14}) and a decrease by a factor 6 in seawater. This leads to the hypothesis that particles presenting high affinity with cesium in freshwater (high Kd) partially keep this affinity in seawater. Consequently, we investigated factors influencing cesium repartition on material and consequences for desorption.

3.3.1 Influence of material:

Authors generally refer to three components as main absorbents for Cs^+ with different properties:

-strongly sorbing non-expansible 2:1 phyllosilicates such as micaceous mineral (biotite) or illite with relatively low cation-exchange capacity (CEC) with high affinity sites (Nakao et al., 2014).

-weakly sorbing expansible 2:1 phyllosilicate such as smectite with relatively high CEC (Mckinley et al., 2001).

- organic matter with usually high CEC (Valcke and Cremers, 1994).

Since particles composition is a heterogeneous mixing of mineral and organic phases in different proportions, we can expect a high spatio-temporal variability in the proportion of available sites with their own capacity to sorb Cs. Such heterogeneity may explains the discrepancies in distribution coefficients in freshwater between sites (Boyer et al., 2018; Takahashi et al., 2017). In the case where riverine particles directly reach the coastal zone (small rivers, sea with no tides), the characteristics of the particles regarding Cs fixation can be conserved in seawater, explaining the good relation of Fig 7.

The important role of the nature of particulate material is highlighted in RCs.Lab.Fw, by the fact that two samples from the same river presents a similar desorption, even if they were not collected at the same time. Ferrer (1983) and Ouaniche (1984) obtained 40% of desorption in the Rhône river; Clanton (1968) and Reynolds and Gloyna (1964) also obtained 40% in Guadalupe river. Ciffroy et al (2001) used two samples from the Loire river collected over 6 months, and they found 60-80% of desorption for both.

3.3.2 Influence of the contamination process

Sites with high affinity (FES: Frayed Edge Site) contributes predominantly to Cs sorption at low concentrations under 10^{-8}M (Sawhney, 1970; Bradbury and Baeyens, 2000). However, they also present the lowest capacity (Nakao et al., 2014) and constitutes only 0.25% of the CEC of the illite (Bradbury and Baeyens, 2000). Once these sites are saturated, excess Cs goes to surficial sites with lower affinity.

Usually the particles spiked for the RCs.Lab.Fw experiment present higher activities than those for RCs. This potentially results in a higher fraction of Cs in surficial sites, more subject to a rapid remobilization.

Beyond that, an increase of solid-to-liquid ratio during the contamination phase leads to a lower desorption. Onodera et al. (2017) found that the desorbed fraction of radiocesium followed a negative power law with a ratio ranging from 0.17 g/L to 10 g/L during the spiking phase. According to the authors, a larger number of strong adsorption sites were available when the solid-to-liquid ratio was higher, leading to an increase of the fraction of strongly adsorbed Cs onto particles and, as a result, desorption became more difficult. Finally, it is possible that a longer contact time with the Cs solution decreases the amount of extractable Cs from all sites (Okumura et al, 2018).

Comans and Hockley (1992) argued that radiocesium is able to migrate from basal surface sites to interlayer sites and can be incorporated into the mineral as stable Cs. This migration called " aging effect" may impact its mobility; (Madruga and Cremers, 1997; Roig et al., 2007). This effect has been studied in a 1M Na^+ solution only by Mukai et al. (2018) on weathered biotites from Fukushima. They found that a sorption time longer than one week decreased desorbed fraction from 90% to less than 10%, compared to a one day or one weak sorption. Biotite sampled at the same site, containing radiocesium post-Fukushima (RCs like experiment) desorbed the same amount of radiocesium. However, it should be noted that this experiment was conducted at pH 4.5, clearly different from those of seawater.

The aging effect has been more studied in experiments using freshwater for both the adsorption and desorption phase. Ciffroy et al (2001) found that the fraction of Cs released after 24 h decreases by a factor 2 when the duration of the contact time of the sorption phase increases from 30 min to 100 hr. Benes et al (1992) also found that a sorption phase of 5 and 21 days instead of 2 hours decreased the fraction of released Cs by a factor 1.3 and 2 respectively.

From these experiments it seems that a longer sorption phase allows to fix Cs more irreversibly, at least in freshwater media. These results could partly explain why the desorption in RCs experiment is usually lower than for RCs.Lab.Fw and closer to SCs experiment. Indeed the contact time for the samples used in RCs is longer because it corresponds to the delay between the *in-situ* sample contamination and its desorption once the particles are exposed to seawater. This longer delay facilitates Cs migration into the mineral lattice with strong affinity.

4 Discussion

The objective of this review is to summarize data from literature in order to precise the efficiency of Cs desorption processes into seawater medium. We show that results issued from two main experimental designs may be used for that purpose. In the first design, RCs, particles have been exposed to ^{134}Cs or ^{137}Cs in the field, whereas the second one, RCS.Lab.Fw, concerns the desorption of radiocesium from particles spiked (^{134}Cs or ^{137}Cs) in laboratory and in freshwater media.

In the case of field exposition, most of the values of desorption fractions are around 3% (figure 2), and this is valid for both radiocesium (RCs) and stable Cs (SCs). Furthermore, any sample of particles collected in the environment consequently to a contamination by ^{137}Cs or ^{134}Cs (RCs experiment) released in 95% of cases one third or less of its radiocesium (Figure 3). These results have to be compared with the value proposed by IAEA for the proportion of desorbable Cs in coastal sediments: 20% (IAEA, 2004). Above 20% the figure 3 shows that a plateau is effectively reached, but this value is clearly above the mode at 3% determined here.

Important points are that the efficiency of desorption (i.e. the fraction of Cs released) does not depend on the duration of desorption, and that an increase of salinity does not influence so much the desorbed fraction.

In RCs.Lab.Fw samples were highly contaminated and thus different mineral sites with different sorption isotherms may have stored cesium, including those with low affinity. Overall amount of released Cs is higher than for RCs experiment and could reach sometime 87%. Usually, two kinetic rates are observed and 80% of equilibrium is reached in one hour. A short duration of contamination reduces the “aging” effect resulting in a consequent pool of easily released cesium.

Here, the percentages of desorbed Cs do not follow any regular distribution.

Experimental design RCs.Lab.Fw looks like an “ideal” case based on the fact that there is “no aging”, no change in equilibrium between the fixation of Cs onto particles and their arrival into brackish waters. However, it may correspond to the case were suspended particles are contaminated in freshwater and rapidly reach the sea because of short transit times in rivers. In this case, desorption could be as high as 80 %, and it is not possible to predict it regarding the irregular distribution observed here. In case of an accident, an evolution of the Cs desorbed may be thus expected with time, related to the duration of transit of the particles between their contamination and the sea.

This desorption is also dependent on salinity until a certain threshold. Studies showed that for salinity above 2, concentrations of K^+ (5.10^{-5} M) and Na^+ (0.027 M) are in theory sufficient to desorb superficial Cs, but there was no change in desorption from 10 to 35 of salinity.

The repartition of Cs onto different sites of the mineral may thus partly explains the differences in desorbed Cs found between both experiments. The various proportions of adsorption sites between samples plus a possible aging effect are an additional source of variability. For each area of interest, we thus recommend to use a sample representative of the site for the sorption/desorption experiments, in order to properly discuss the dynamics of Cs transfer.

5 Conclusion

Desorption of radiocesium from contaminated particles was observed at any salinity above 3, and the fraction desorbed ranges widely from 0 to 86%. For particles containing radiocesium due to an environmental contamination, this range extends from 0 to 64 % with a peak at 3%. Particles containing radiocesium issued from a contamination in laboratory show higher values of Cs released and a reaction implying two first-order kinetics. The first one is really short with a half-life reaction time about one hour, whereas it is of a few days for the second one. These two parallel or successive reactions must be related to sites with different affinity for Cs, and thus radiocesium distribution on mineral prior to desorption is a key factor to take into account.

As a result, a two kinetic box-model calibrated for each site of interest is suitable to predict more accurately desorbed cesium at the interface. However, these conceptual models should be supported by recent developments on clay mineral structures and avoid over parametrization.

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Figures List

Figure 1 : Locations of the sampling sites relative to the documents reporting results on cesium desorption. The symbols refer to the sources of contamination of the particles

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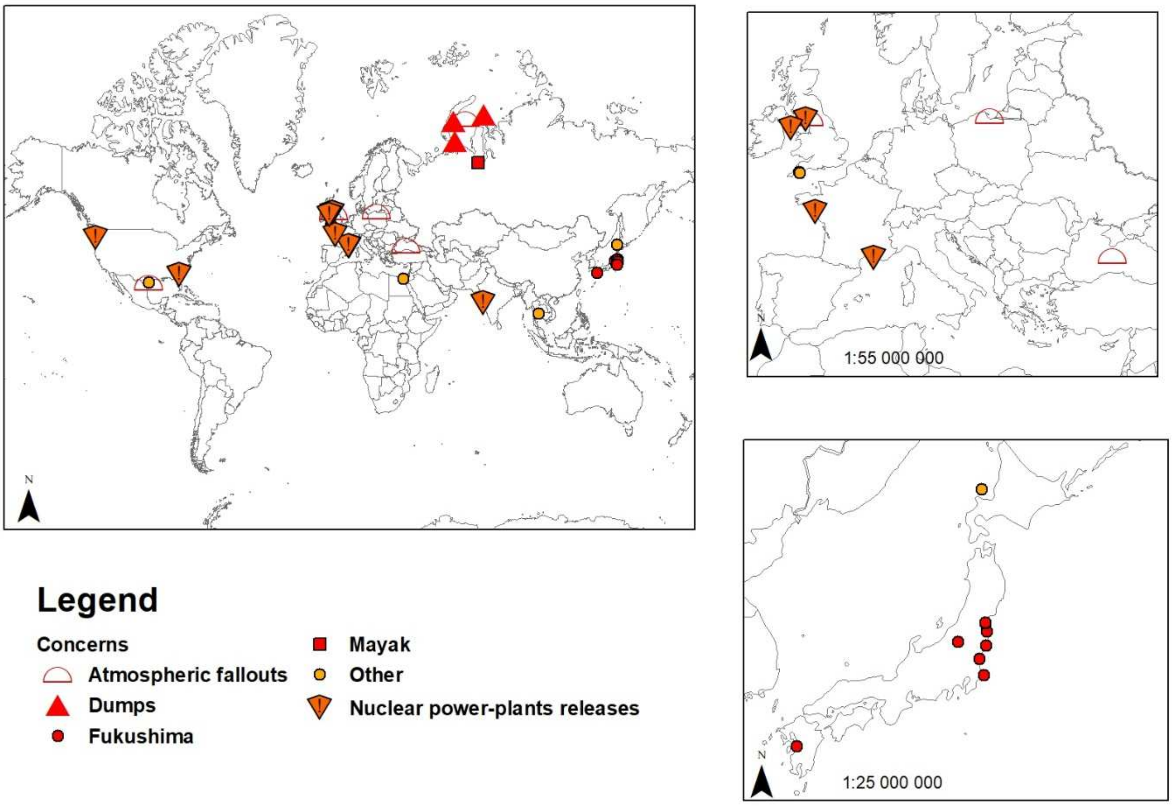
Table List

Table 1：main information of the studies compiled in this article



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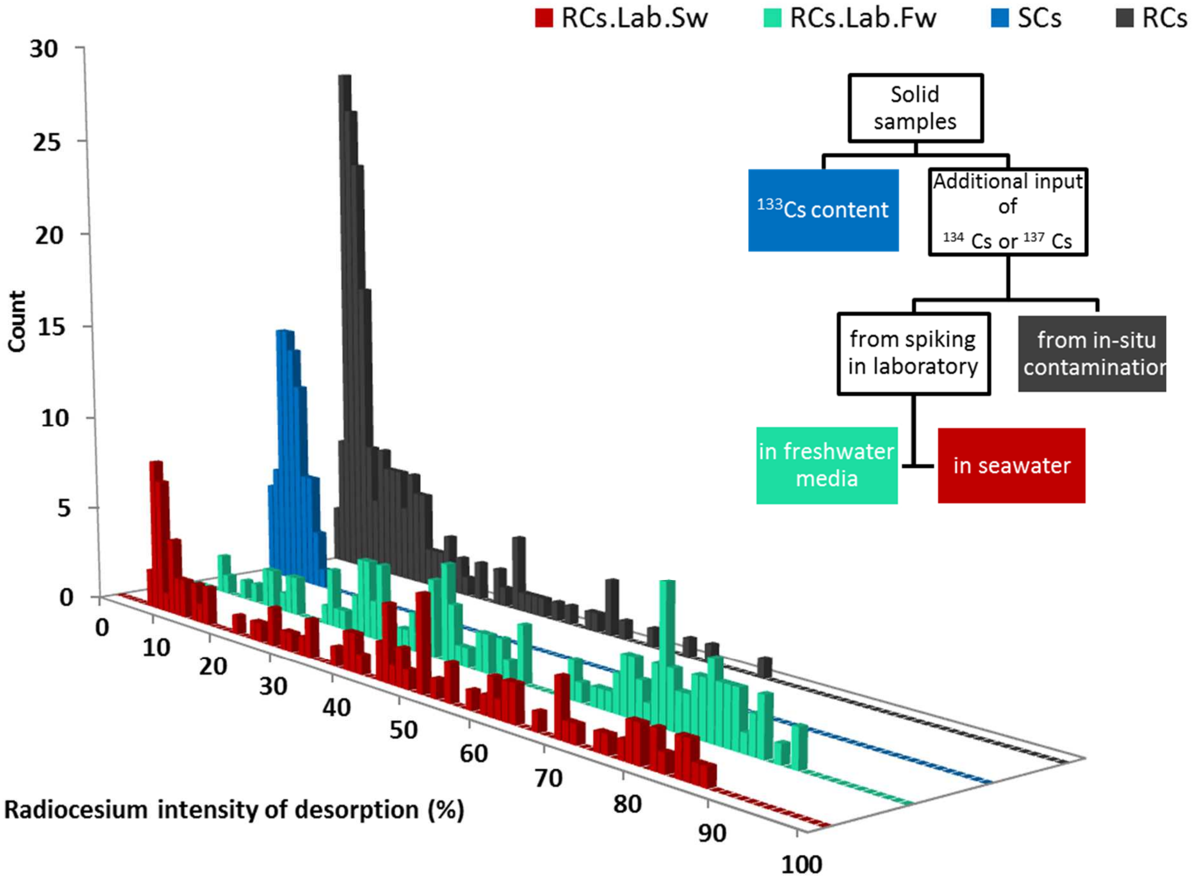

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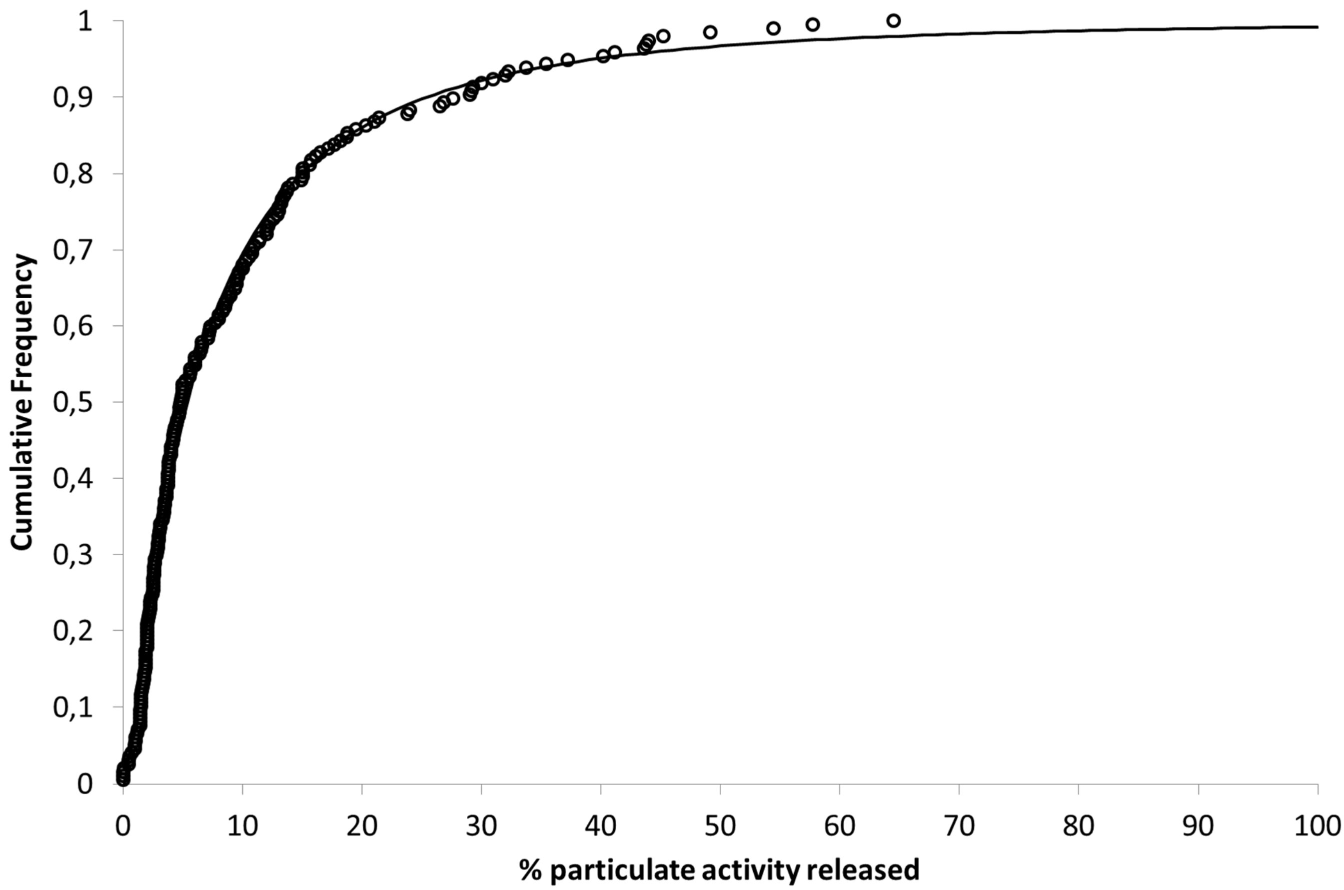

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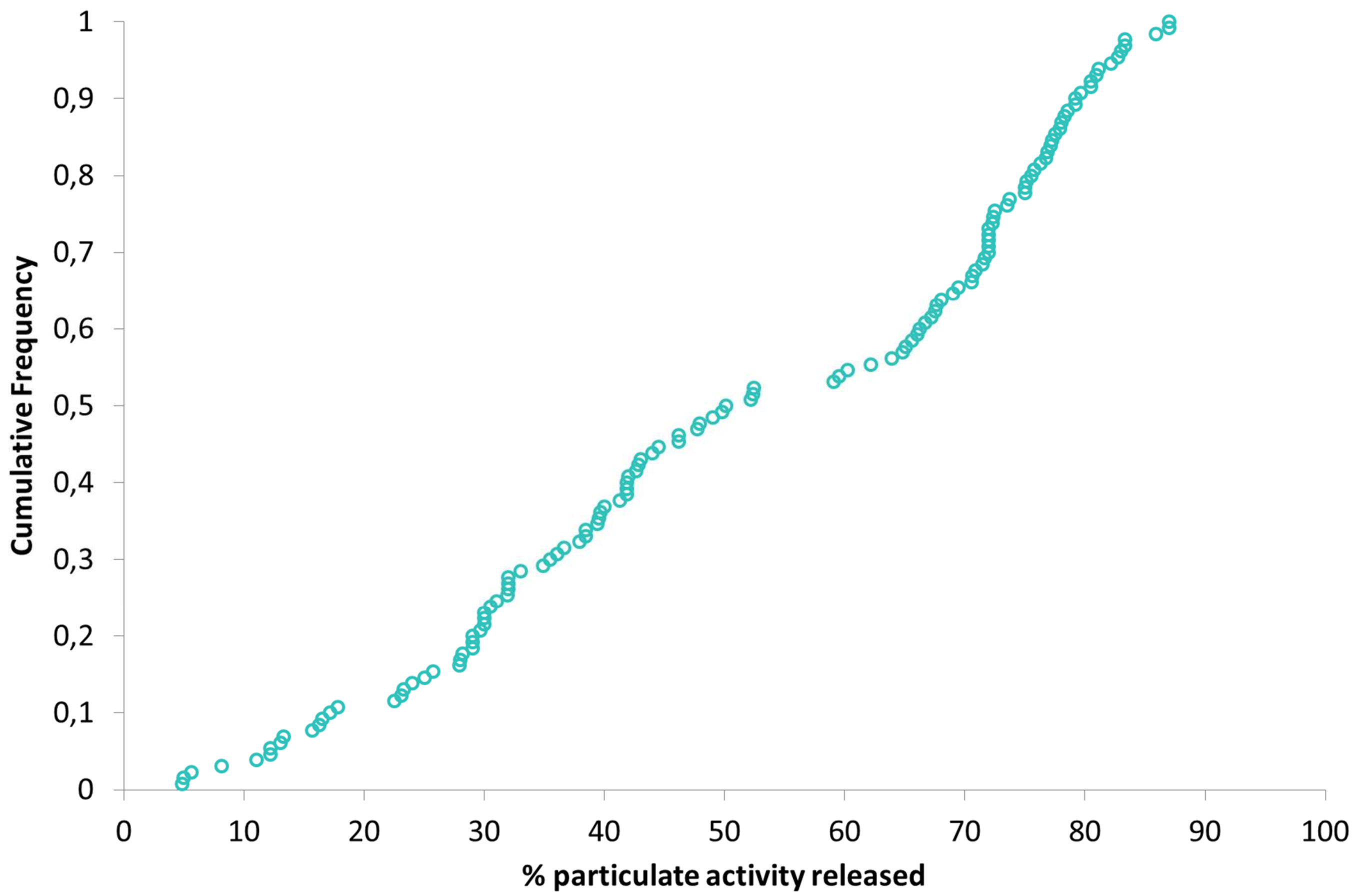

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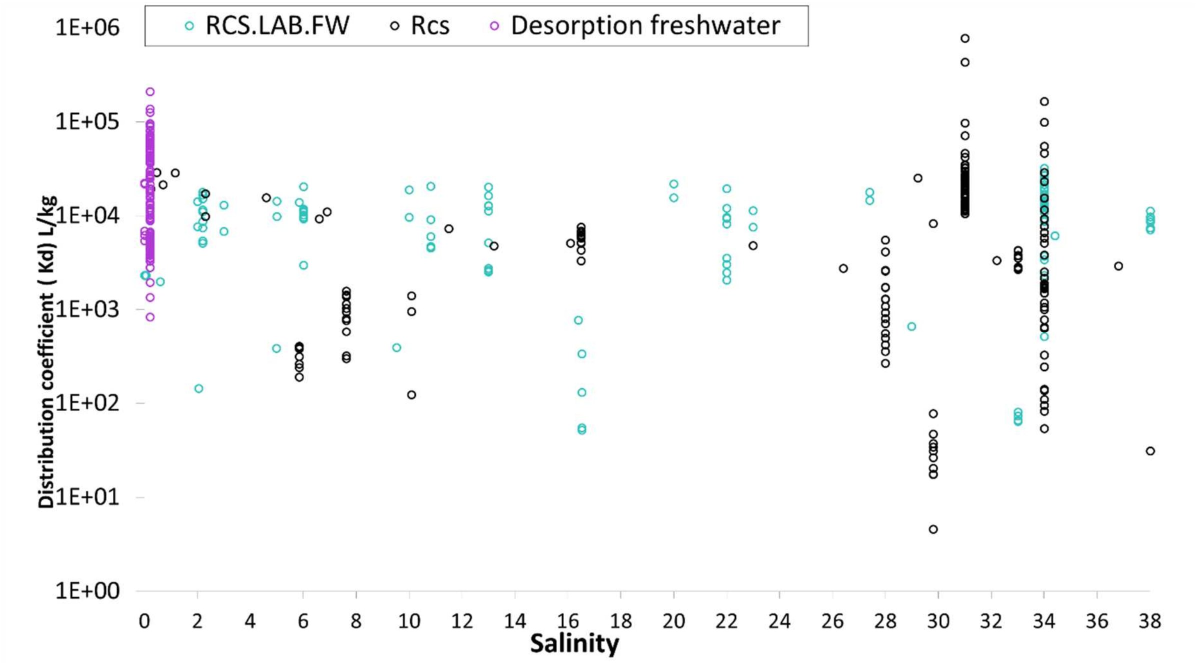

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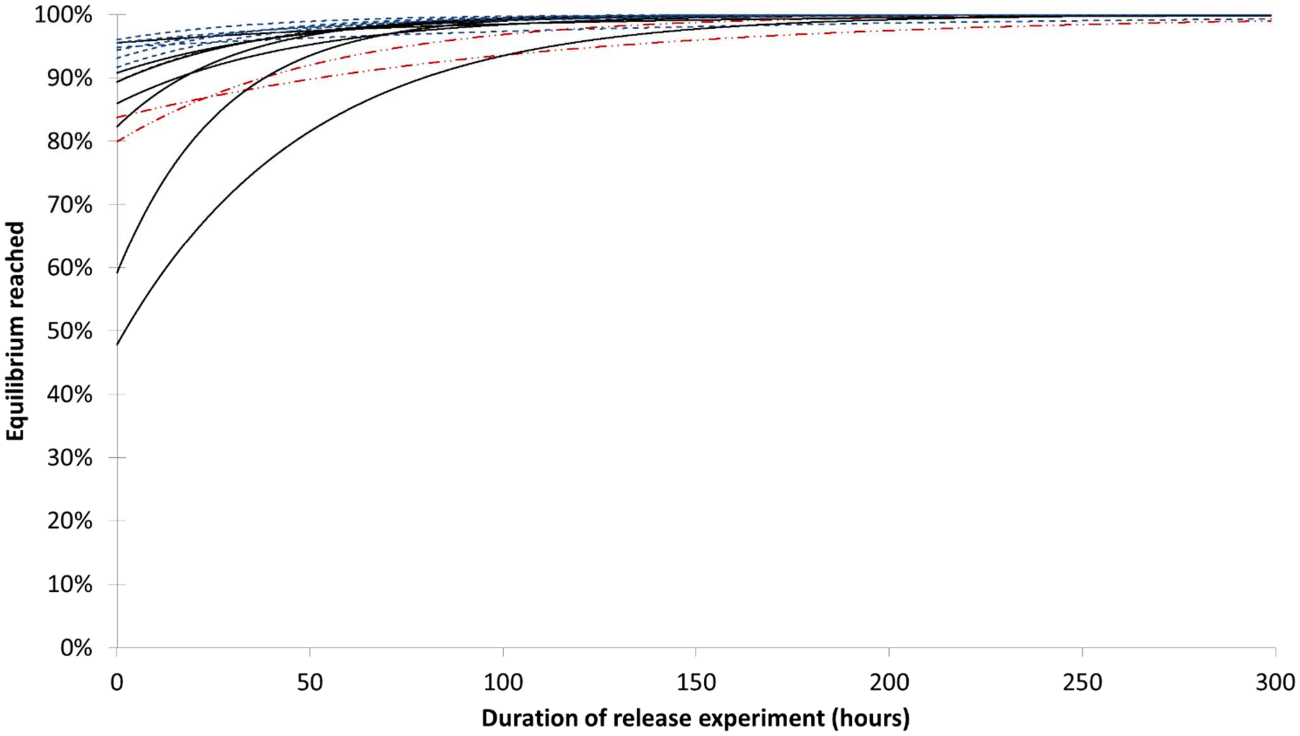

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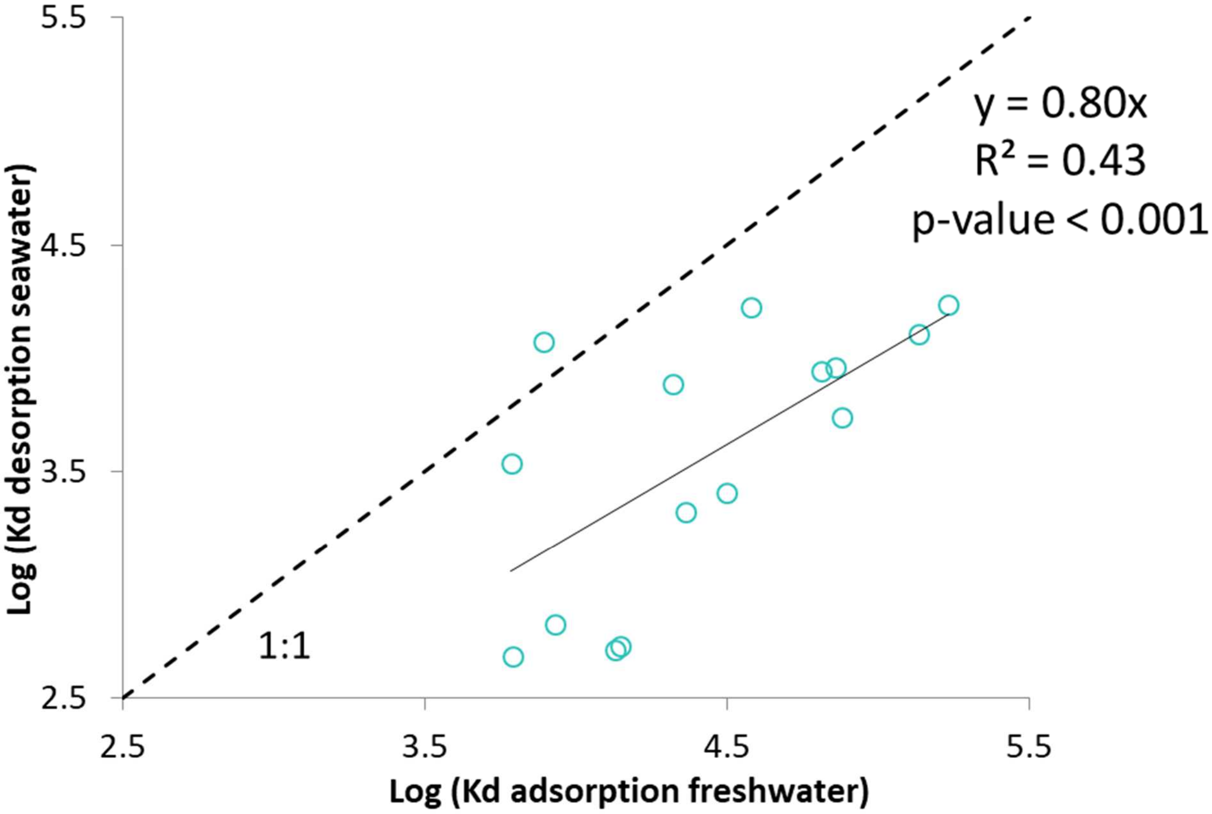

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References	Sample type	Number of samples used in the study	Desorption medium	Experimental design				Studies used for linearization	Studies used for Freshwater Seawater Kd comparison	Origin of the data used in the study
				SCs	RCs	RCs.Lab.Fw	RCs.Lab.Sw			
(Reynolds and Gloyna, 1964)	Guadalupe river sediments	1	San Antonio bay water			X	X		X	Graphical extraction
(Clanton, 1968)	Guadalupe river sediments	1	San Antonio bay water			X		X	X	Graphical extraction
(Cheng, 1968)	Terrigenous sediments Hokkaido	1	Japanese seawater			X				Graphical extraction
(Ganguly et al., 1968)	Bombay harbour sediments	1	Bombay harbour water				X			Provided
(Robertson et al., 1972)	Columbia river sediments	1	Pacific ocean water		X					Graphical extraction
(Brisbin et al., 1974)	Savannah river sediments	1	NaCl solution		X					Reported by Onishi et al., (1981)
(Patel, 1978)	Bombay harbour sediments	2	Bombay harbour water		X					Graphical extraction
(Stanners and Aston, 1982)	Ravenglass estuary sediments	1	Irish seawater		X					Calculation from activities
(Evans et al., 1983)	Savannah river pond sediments	2	NaCl solution		X					Provided
(Ferrer, 1983)	Suspended particles Rhône river	3	Artificial seawater			X		X	X	Graphical extraction
(Ouaniche, 1984)	Rhône sediments	2	Rade de Toulon water		X	X			X	Provided
(Bale, 1987)	Tamar estuary sediments	1	Manche seawater			X			X	Graphical extraction
(El-Assy et al., 1991)	Suez canal sediments	1	Suez gulf water			X		X		Graphical extraction
(Davison et al., 1993)	Esthwaite Water lake sediments	1	Artificial seawater		X					Provided
(Desai et al., 1994)	Bombay harbour sediments	1	Bombay harbour water		X					Provided
(Kleinot, 1994)	Tamar estuary sediments	1	Artificial seawater				X			Graphical extraction
(Knapinska-Skiba et al., 1994)	Baltic sea sediments	2	Gdansk Bay water		X		X			Graphical extraction
(Oughton et al., 1997)	Kara sea sediments	4	Oslo Fjord water		X	X	X			Graphical extraction and calculations from Kd
(Fuhrmann et al., 2001)	Kara sea sediments	1	Atlantic ocean water				X			Graphical extraction
(Borretzen and Salbu, 2002)	Stepovogo Fjord sediments	1	Kara seawater				X			Graphical extraction
(Standring et al., 2002)	Mayak ponds sediments	4	Artificial seawater		X					Calculation from activities
(Topcuo□lu	Black sea	4	Blacksea				X			Graphical extraction

et al., 2002)	sediments		water							
(Ciffroy et al., 2003)	Suspended particles Loire river	2	Atlantic ocean water			X		X	X	Graphical extraction and calculations from Kd
(Saengkul et al., 2013)	Bang Pakong estuary sediments	2	Thaïland gulf water				X			Graphical extraction
(Tagami and Uchida, 2013)	Fukushima soil	1	Pacific ocean water			X				Provided
(Otosaka and Kobayashi, 2013)	Marine sediments Fukushima	2	Pacific ocean water		X					Graphical extraction
(Fan et al., 2014)	Abukama and Kuchibuto river sediments	4	Artificial seawater			X			X	Graphical extraction and calculations from Kd
(Berthail, 2015)	Nitta and Otta river sediments and Sellafield estuary sediments	3	Seawater (origin unspecified)		X					Provided
(Takata et al., 2015)	Abukama, Tone, Kuji and Naka river sediments	12	Pacific ocean water	X	X					Graphical extraction
(Yamasaki et al., 2016)	Kuma river sediments	1	Artificial seawater		X					Graphical extraction
(Onodera et al., 2017)	Suspended particles Abukama and Natsui rivers.	2	Artificial seawater, pacific ocean water and NaCl solution			X		X	X	Graphical extraction
(Sakaguchi et al., 2018)	Fukushima soil	1	Seawater nearby Kikajima island	X	X					Calculation from activities

Table 1 : main information of the studies compiled in this article