Effect of sediment characteristics on sorption of $^{137}$Cs at the sediment-water interface

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Abstract: Sediments play an important role on diffusion fate of radionuclide in aquatic systems. In the present study investigated the sorption behavior of $^{137}$Cs in four different characteristic of marine sediments (clay content, CEC, organic matter) that collected from the upper gulf of Thailand by using batch experiment. The sorption process is depend on characteristic of marine sediment samples which found distribution coefficient ($K_d$) increase with increase CEC and % clay content that indicated the degree of sorption of $^{137}$Cs to the sediment were strongly dependent on particle size. On the other hand the $K_d$ to decrease with increase organic matter and particle size. Thus, marine sediment samples that high clay content effect on efficiency sorption of $^{137}$Cs increasing (St.1, St. Angsila, St.21 and St. Bangsaen, respectively) but organic matter is contrast effect on $^{137}$Cs sorption that there is a little influence on the sorption.

Keywords - $^{137}$Cs, Sorption, Distribution coefficient, Marine sediment, radionuclide.

I. Introduction

Radiocesium have been introduced into the environment by various sources, the most well documented being fallout from atmospheric nuclear weapons testing in the 1950s and 1960s and later the accident at the Chernobyl nuclear power plant in 1986 and last at Fukushima nuclear power plant in march 2011. In present, radiocesium still remain to ocean water, marine sediment and marine life that reported indicates the concentration in ocean water pose no direct threat to humans or marine biota but accumulation in marine sediment could be of concern for decades [4]. Marine sediment was important factor on diffusion and accumulation of radiocesium in environment. Radiocesium can be transfer from water to sediment by direct adsorption that efficiency of adsorption depend on characteristic of marine sediment such as type of clay mineral, clay content, soil texture, cation exchange capacity (CEC), particle size and organic matter. $^{137}$Cs is strongly adsorbed onto clay mineral that adsorption sites have been reported at interlayer sheet or fried edge site (FES) [3].

CEC levels of sediment depend on amount of clay content (soil texture) and soil organic matter that clay particles and organic matter in sediment tends to negatively active charged. Organic matter also plays an important role in adsorbing $^{137}$Cs, even though affinity is lower than of clay mineral [9]. Several studies have shown that organic components in soil are closely related with $^{137}$Cs concentration [2] [10] [11]. Organic matter in sediment to reduced $^{137}$Cs adsorption by clay mineral, maybe by impeding the collapse of interlayers following adsorption [13]. One of the more important factors influencing the transport of particle- reactive chemicals is the particle size distribution of sediment. Furthermore, the particle size distribution of sediment that important factor influencing the transport of $^{137}$Cs in sediment [1] [6] [14]. The concentrations of $^{137}$Cs in the fine fraction higher than the coarser fraction that indicated the $^{137}$Cs concentration decrease with an increase in particle size [8].

The aim of the present work was to investigate the sorption behavior of $^{137}$Cs in four different characteristic of marine sediment by using the batch method. Correct determination of $^{137}$Cs adsorption mechanics on sediment is important for understanding the fate of this pollutant in environment.

II. Material and method

2.1 Sediment and analysis

Four surface marine sediment samples near coastal area from the upper gulf of Thailand were chosen to give a range of physical properties, e.g., clay content affecting $^{137}$Cs sorption. The sediments were freeze-dried, passed through a 2 mm sieve and stored before analysis.

Particle size analyses were made using the hydrometer method with a 2 hour reading for clay content. Organic matter was determined from loss on ignition determine by the difference of sediment weight after dry at 105°C and then at 550°C. Cation exchange capacity was measured according to the ammonium acetate method.
The marine sediment samples were treated in triplicate and the mean reported. Samae-sarn sea water (salinity, 32 psi and pH 8.3) used in the experiments was filtered through 0.22 µm Millipore filters.

2.2 Sorption experiments

The marine sediment sorption capacity was investigated in this study by batch method. Sorption experiments were performed in 20 ml screw cap centrifuge tubes containing 0.2 g of sediments. Use one concentration activity, 1090.7 Bq/ml of $^{137}$Cs. The ratio of mass of sediment to volume of solution was 1:50. After shaking for 168 h the solution was immediately separated from suspensions by centrifuging. The solutions from the above treatment were analyzed for $^{137}$Cs using a Gamma ray spectrometry (NaI). In all cases, standards and blank were always treated in the same way as the samples to minimize matrix interferences during the analysis.

Absorbed $^{137}$Cs activity concentration in the solid phase was determined from the difference between the initial $^{137}$Cs activity concentration and the equilibrium concentration. The equilibrium distribution coefficient ($K_d$) was used to characterize $^{137}$Cs sorption: $K_d = C_s / C_l$

Where $C_s$ is the $^{137}$Cs activity concentration sorbed on solid matrix (Bq/kg) and $C_l$ is the equilibrium contaminant concentration in solution (Bq/l)

<table>
<thead>
<tr>
<th>Parameter</th>
<th>St. 1</th>
<th>St. 21</th>
<th>St. Angsila</th>
<th>St. Bangsaen</th>
</tr>
</thead>
<tbody>
<tr>
<td>Clay%</td>
<td>81</td>
<td>61</td>
<td>65.2</td>
<td>3</td>
</tr>
<tr>
<td>Silt %</td>
<td>7</td>
<td>20</td>
<td>18.8</td>
<td>11</td>
</tr>
<tr>
<td>Sand%</td>
<td>11</td>
<td>19</td>
<td>23</td>
<td>86</td>
</tr>
<tr>
<td>Texture</td>
<td>Clay</td>
<td>Clay</td>
<td>Clay</td>
<td>Loamy sand</td>
</tr>
<tr>
<td>Organic matter (%)</td>
<td>6.43</td>
<td>10.08</td>
<td>7.61</td>
<td>0.68</td>
</tr>
<tr>
<td>CEC (cmol/kg)</td>
<td>29.17</td>
<td>21.95</td>
<td>19.76</td>
<td>0.63</td>
</tr>
</tbody>
</table>

Table 1 Physicochemical properties of the selected marine sediment (n=3).

III. Result and Discussion

Selected physicochemical properties of the studies marine sediment are present in Table1. The marine sediments are clay texture but St. Bangsaen is loamy sand and contained various amounts of organic matter that range between 0.68 - 10.08%. The cation exchange capacity (CEC) was highly correlated to clay content ($r$ =0.97 ***, p<0.05; Fig.1).

The effect of marine sediment characteristic on $^{137}$Cs sorption was highly significant that St.1 and St. Angsila higher $K_d$ value than St.21 and St.Bangsaen respectively (325.59±23.52, 279.36±22.25, 211.95±11.84 and 8.92±0.99) (Fig. 2). The $K_d$ values indicate that the clay sediment (St.1, St.21 and St. Angsila) exhibit the higher sorption of $^{137}$Cs than the loamy sand sediment (St. Bangsaen). The minimum sorption of $^{137}$Cs was displayed by loamy sand sediment, with the lowest CEC. In the order to evaluate the relationships between the $K_d$ and sediment parameters, correlation analysis was employed.

Figure 1 Correlation between clay content and CEC of the studied marine sediments.
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Figure 2 The $K_d$ values of $^{137}$Cs as a function of four studied marine sediments.

The result of correlation between % clay content and $K_d$ value of $^{137}$Cs is showed in Fig. 3 that $K_d$ value of $^{137}$Cs had significant positive correlation with % clay content ($r=0.99$, $P<0.05$). The particle size (%Clay content) distribution plays an important role on sorption of $^{137}$Cs and the distribution coefficient exhibited a certain correlation with clay content and CEC (Fig. 3). This supports the hypothesis that the clay fraction of soil, with consists by sheet silicate minerals, has a high affinity for sorption of $^{137}$Cs [7]. Texture of sediment determined most size particle of sediment sample that refer specific area of sediment of sediment. The specific surface area is increase as a particle size become small that important for sorption capacity and ion exchange capacity [14].

Figure 3 Correlation between $K_d$ value of $^{137}$Cs and % clay content.

Figure 4 Correlation between $K_d$ value of $^{137}$Cs and % organic matter.

The relation between the $K_d$ value of $^{137}$Cs and the % organic matter that showed not significant correlation ($r=0.76$, $p>0.05$) (Fig 4). The result indicated that organic matter not effect on $^{137}$Cs sorption.
However, several authors have found that organic matter have effect on Cs sorption which organic matter dose decrease the adsorption of Cs on clay mineral [5][12]. On the other hand, considered sediment sample from St.1, St.21 and St.Angsila which same texture is clay type found that increase organic matter effect on $^{137}$Cs sorption trend to efficiency decrease. Organic matter were anionic or negative charges, the reason for decrease Cs adsorption clearly cannot be competition for the FES and reducing the selectivity of minerals for cations, probably by organic macromolecules impeding the collapse of FES, which organic matter catch couple, adsorption decreasing with increasing organic matter added [13]. Another reason, organic matter that coating of the clay mineral surface is likely to be limited to positive charged edge site caused by broken Al-OH group that decrease positively charged edge site or adsorption site for fix Cs.

IV. Conclusions

We studied various characteristic of marine sediments near the upper Gulf of Thailand that has effect on $^{137}$Cs adsorption. $^{137}$Cs adsorption is increased with the increasing of % clay content and CEC. In the case of organic matter, it is decrease with an increasing in organic matter.

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References