Plutonium in Southern Hemisphere Oceans


*Sophia University

E-mail: hirose45037@mail2.accsnet.ne.jp
Objective

• Review on levels and distribution of plutonium in Southern Hemisphere Ocean waters
• SHOTS results
• Roles of plutonium as an oceanic tracer (biogeochemical processes, deep water advection)
Background

• Sources of plutonium in Southern Hemisphere Oceans.
  ⇒ Global fallout
  ⇒ Close-in fallout from the French nuclear explosions (South Pacific).
    (less contribution of close-in fallout such as Bikini explosions and radioactive discharge)

• Radioactivity measurements in the Southern Hemisphere Oceans
  ⇒ Very small number of data, especially in deep waters
Plutonium in Southern Hemisphere Oceans
Sampling stations before SHOTS (HAM database)

Aoyama and Hirose, SWJ, 2004
Sampling and method

• Sampling stations ⇒ 48 stations (South Pacific Ocean), 20 stations (Indian Ocean), 15 stations (Atlantic Ocean)
⇒ Vertical distribution (5 stations in the South Pacific Subtropical Gyre)
• Sample volume ⇒ 5-60 liters of filtered seawater.
• Analytical method
  Fe-coprecipitation
  Radiochemical separation
  alpha-spectrometry (South Pacific surface water)
  ICP-MS (South Pacific vertical samples, Indian and Atlantic surface water)

Only $^{239}$Pu concentration can be determined for ICP-MS because of smaller sample volumes and low plutonium concentration.
Sampling stations including SHOTS stations
Results: Plutonium in the Southern Hemisphere Oceans

- SHOTS data: plutonium in surface waters
- Temporal change of surface plutonium
- Vertical profiles of plutonium in the South Pacific (SHOTS)
- Plutonium/$^{137}$Cs ratios as a proxy of geochemical processes (SHOTS(South Pacific), GEOSECS(South Atlantic))
- Deep plutonium
Comparison between alpha spectrometry and ICP-MS (assuming that $^{240}$Pu/$^{239}$Pu atom ratio is equal to global fallout (0.18))
$^{239}$Pu concentration in surface waters of Southern Hemisphere oceans

Hirose et al., STOTEN, 2007
Gautaud et al., Prog. Oceanogr. 2011
Temporal change of surface $^{239,240}$Pu in Southern Hemisphere oceans

**Sea area**
- Western South Pacific
- South Pacific Subtropical Gyre
- Eastern South Pacific
- Eastern Indian Ocean
- Western South Atlantic

**Half-residence time (year)**
- Western South Pacific: $34 +^{17}_{-8}$
- South Pacific Subtropical Gyre: $19 +^{2}_{-1}$
- Eastern South Pacific: $22 +^{2}_{-1}$
- Eastern Indian Ocean: $20 +^{15}_{-5}$
- Western South Atlantic: $12 +^{13}_{-6}$

**Graphs**
- **Eastern Indian Ocean**
- **Western South Atlantic**
Vertical sampling sites of BEAGLE2003

Vertical sampling sites of Pu
Cross section of $^{239}\text{Pu}$ in the South Pacific subtropical gyre

Hirose et al.,
*Prog. Oceanogr.* 2011
$^{239}\text{Pu}/^{137}\text{Cs}$ ratio is an indicator of biogeochemical processes.

1. $^{239}\text{Pu}/^{137}\text{Cs}$ ratios exponentially increased from surface to 1500 m depth.
2. The ratios were almost constant in deep water. However, lower ratios occurred in the depth range of 4000 m to 5000 m.

Hirose et al., JER, 2008
Hirose et al., Prog.Oceanogr., 2011
Vertical profiles of $^{239,240}$Pu in South Atlantic Ocean (GEOSECS; 1972)

$^{239,240}$Pu/Cs activity ratio

$^{239}$Pu/$^{137}$Cs ratios exponentially increased from surface to 1200 m depth for South Atlantic.

32.97S 42.52W
Vertical changes of $^{239}$Pu/$^{137}$Cs in shallow layer (South Pacific)

- The $^{239}$Pu/$^{137}$Cs ratios in surface layers
  
  Low values ($0.56 - 1.1 \times 10^{-3}$) comparing with that in global fallout (0.009)

- The $^{239}$Pu/$^{137}$Cs ratios exponentially increased from surface to 1500 m depth.

$$R_{\text{Pu/Cs}(z)} = R_{\text{Pu/Cs},o} \exp(\lambda z) \text{ HRD} = 0.693/\lambda$$

<table>
<thead>
<tr>
<th>Stn.</th>
<th>$R_{\text{Pu/Cs},o} \times 10^3$</th>
<th>$\lambda$ value</th>
<th>HRD(m)</th>
<th>correlation factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>175</td>
<td>0.95</td>
<td>0.0032</td>
<td>220</td>
<td>0.967</td>
</tr>
<tr>
<td>156</td>
<td>0.74</td>
<td>0.0028</td>
<td>250</td>
<td>0.959</td>
</tr>
<tr>
<td>145</td>
<td>0.43</td>
<td>0.0034</td>
<td>200</td>
<td>0.986</td>
</tr>
<tr>
<td>136</td>
<td>0.52</td>
<td>0.0031</td>
<td>220</td>
<td>0.991</td>
</tr>
<tr>
<td>127</td>
<td>0.53</td>
<td>0.0037</td>
<td>190</td>
<td>0.957</td>
</tr>
</tbody>
</table>

HRD: half-regeneration depth
Vertical changes of $^{239,240}\text{Pu}/^{137}\text{Cs}$ in shallow layer (South Atlantic)

- The $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios in surface layers
  Low values ($2.0 - 4.1 \times 10^{-3}$) comparing with that in global fallout (0.008) (GEOSECS, 1972)
- The $^{239,240}\text{Pu}/^{137}\text{Cs}$ ratios exponentially increased from surface to about 1000 m depth.

\[
R_{\text{Pu/Cs}}(z) = R_{\text{Pu/Cs},o} \exp(\lambda z) \quad \text{HRD} = 0.693/\lambda
\]

<table>
<thead>
<tr>
<th>Latitude</th>
<th>$R_{\text{Pu/Cs},o} \times 10^3$</th>
<th>$\lambda$ value</th>
<th>HRD(m)</th>
<th>correlation factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>15\degree S</td>
<td>0.9</td>
<td>0.0080</td>
<td>87</td>
<td>0.960</td>
</tr>
<tr>
<td>21\degree S</td>
<td>1.3</td>
<td>0.0046</td>
<td>150</td>
<td>0.997</td>
</tr>
<tr>
<td>33\degree S</td>
<td>4.1</td>
<td>0.0021</td>
<td>340</td>
<td>0.942</td>
</tr>
<tr>
<td>45\degree S</td>
<td>4.0</td>
<td>0.0020</td>
<td>350</td>
<td>0.919</td>
</tr>
</tbody>
</table>
Relationship between plutonium maximum layer depth and half-regeneration depth

GEOSECS South Atlantic
Chemical tracers in deep waters of the Pacific Ocean

Latitudinal distributions of CFC-11 (P-15) and C-14(P-14) (WOCE Atlas)

North Pacific Deep Water (2000 - 3000 m depth)
Plutonium in deep water

- Weak biological activities $\Rightarrow$ less important biogeochemical processes
- The $^{239}\text{Pu}/^{137}\text{Cs}$ ratios in deep water showed no increase with increasing depth. The relatively low values occurred the depth range from 4000 to 5000 m depth. $\Rightarrow 0.01 - 0.03$ (0.009: global fallout)
- Plutonium in the South Pacific deep water is supplied by advection rather than biogeochemical processes.

Closed circle: Central Pacific
Possible pathway of the North Pacific Deep Water (2000-3000 m)

Bikini-derived Pu with higher $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios is tracing decadal flow of the North Pacific Deep Water.
Conclusion

- A level of $^{239}$Pu activity concentration in the South Pacific surface waters is similar to that in the Indian Ocean, and higher than that in the South Atlantic.
- The $^{239}$Pu/$^{137}$Cs ratio in the South Pacific and South Atlantic, a proxy of biogeochemical processes, exponentially increased in shallow layer (0 – ca.1500 m).
- Plutonium in deep waters (2000-3000 m) of the Pacific showed latitudinal distribution with high in the North Pacific and low in the South Pacific. Plutonium is a transient tracer of the North Pacific Deep Water.
Plutonium is the most powerful tracer to solve ocean processes.

Thank you for your attention!