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Plutonium in Southern Hemisphere Oceans

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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.

  $\Rightarrow$Global fallout

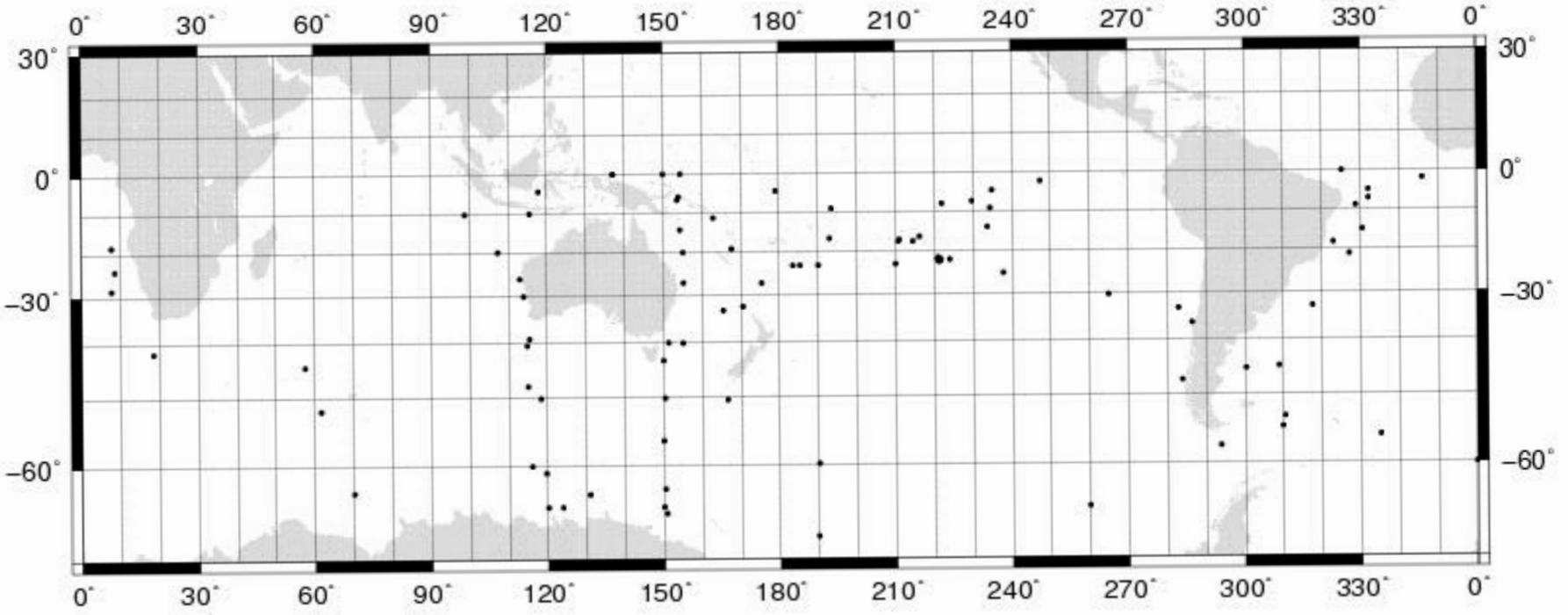
  $\Rightarrow$ Close-in fallout from the French nuclear explosions (South Pacific).

  (less contribution of close-in fallout such as Bikini explosions and radioactive discharge)

- Radioactivity measurements in the Southern Hemisphere Oceans

  $\Rightarrow$ 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 $\Rightarrow$ 48 stations (South Pacific Ocean), 20 stations (Indian Ocean), 15 stations (Atlantic Ocean)

  $\Rightarrow$ Vertical distribution(5 stations in the South Pacific Subtropical Gyre)
- Sample volume $\Rightarrow$5-60 liters of filtered seawater.
- Analytical method

  Fe-coprecipitation

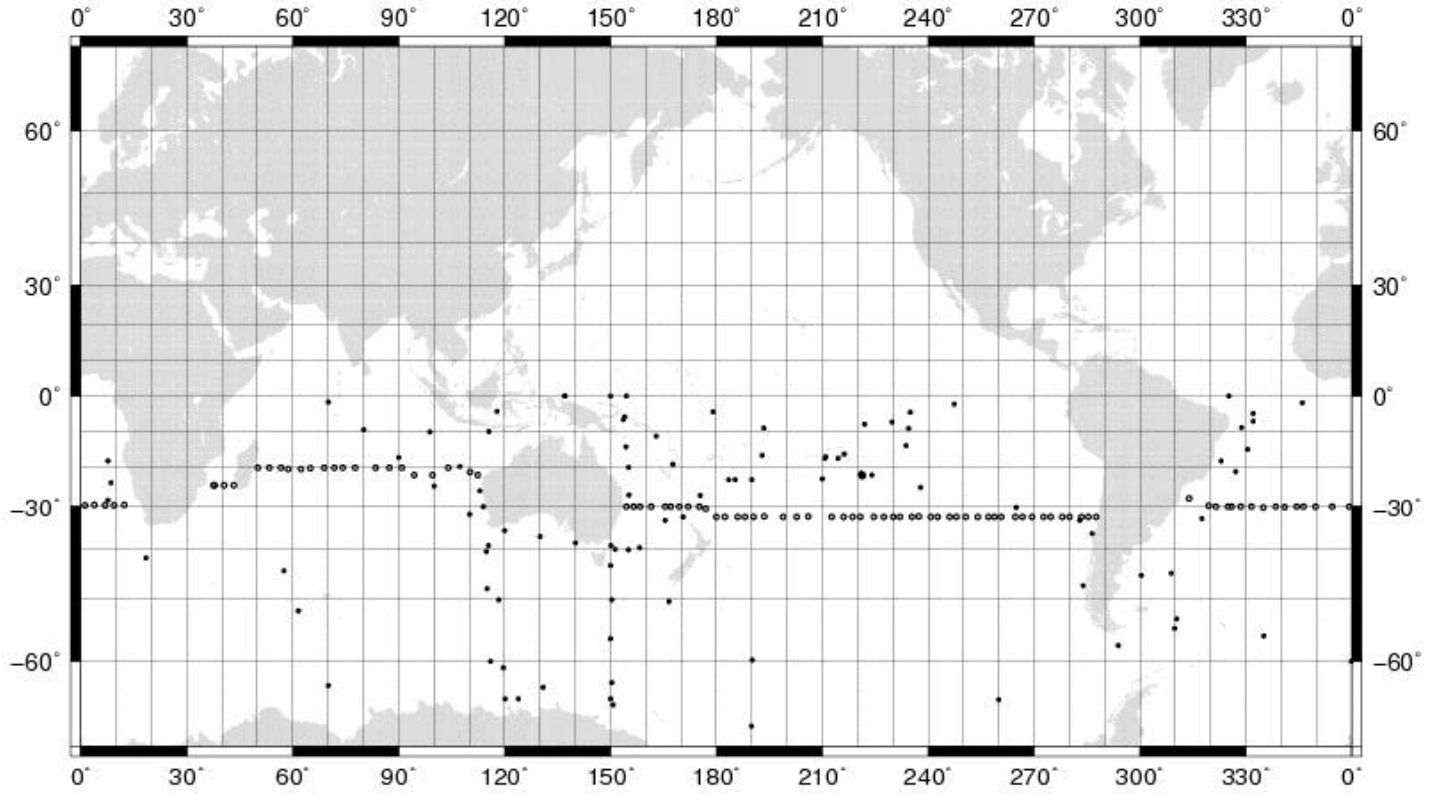
  Radiochemical separation

  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

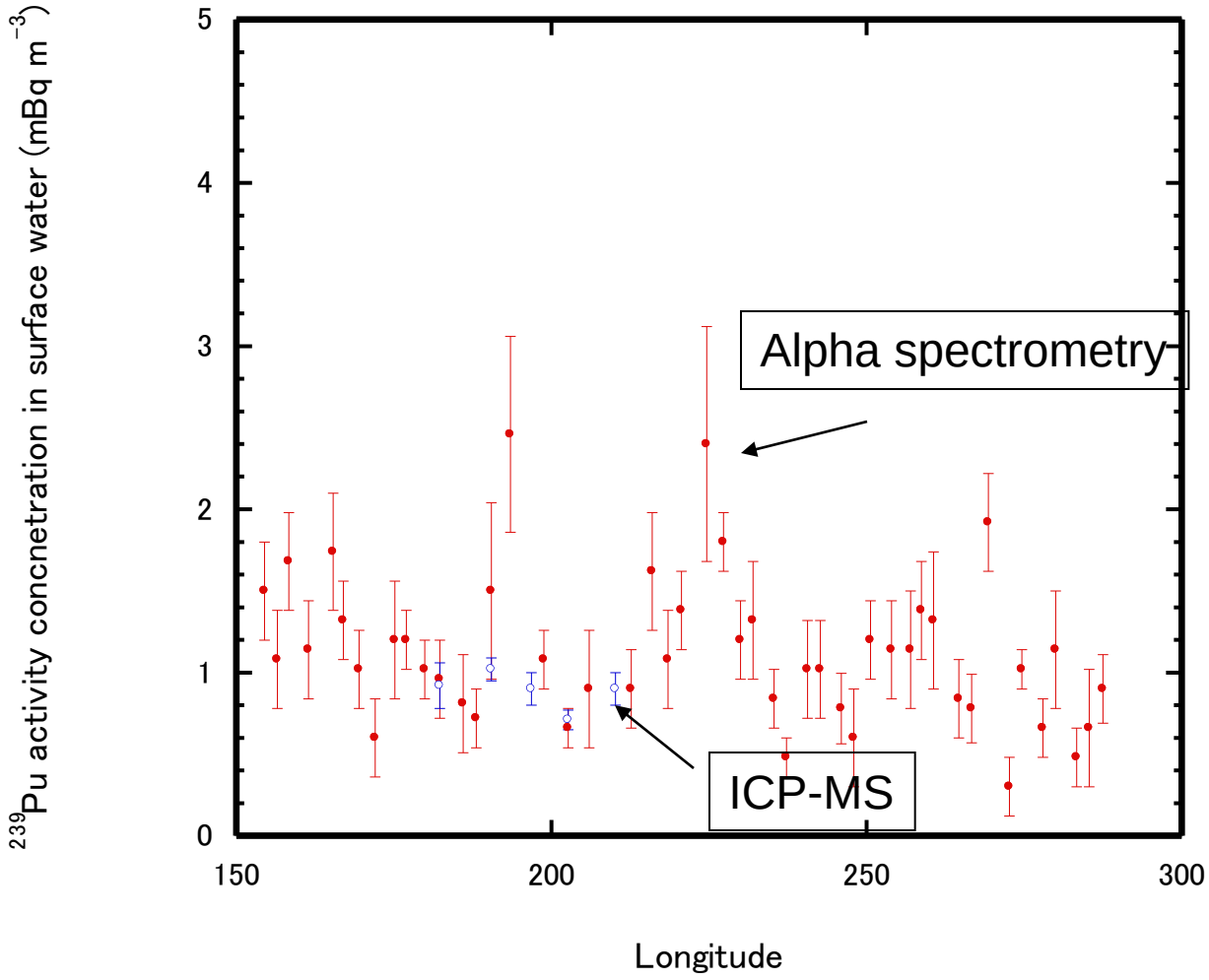


Results: Plutonium in the Southern Hemisphere Oceans

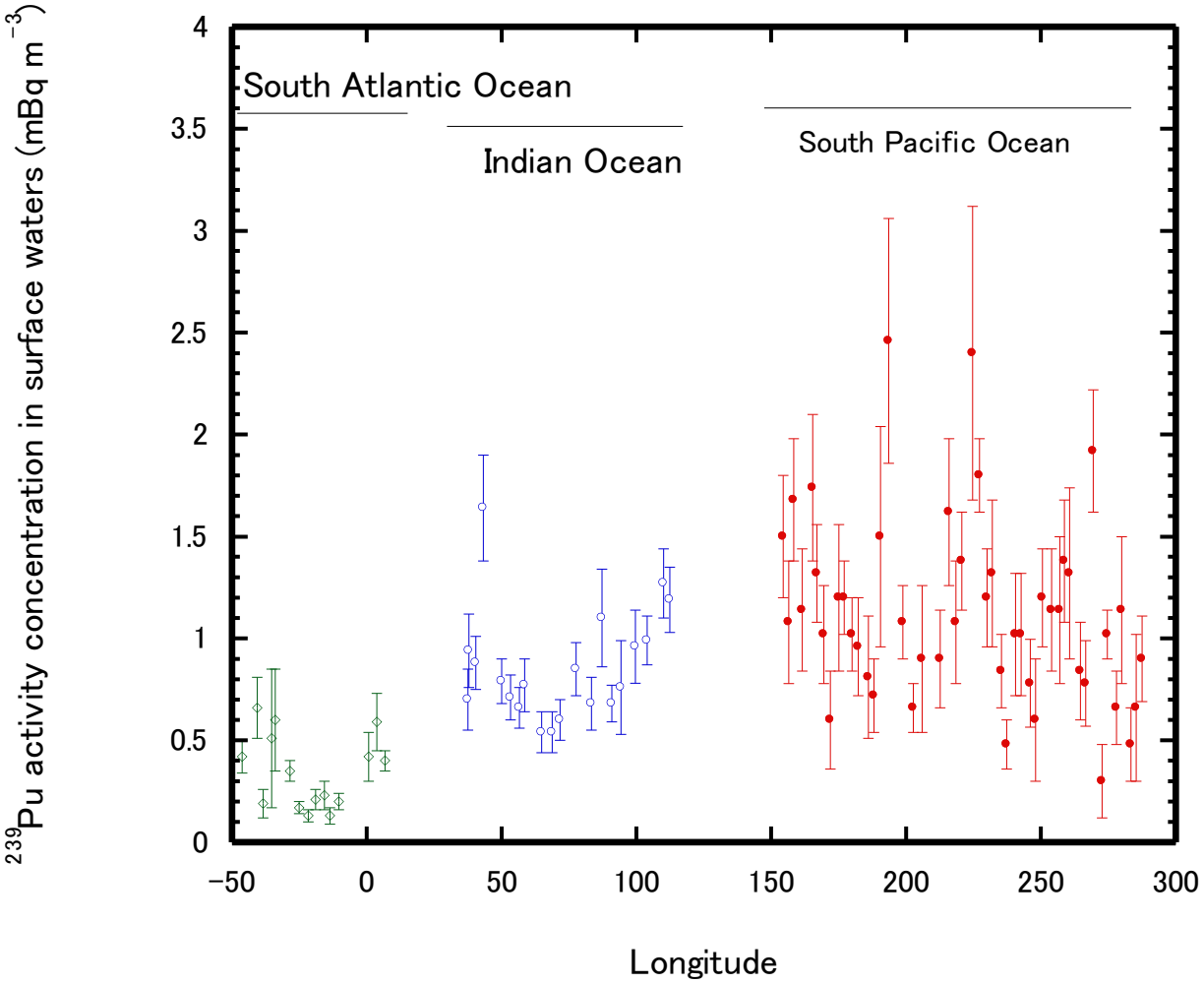
- SHOTS data: plutonium in surface waters
- Temporal change of surface plutonium
- Vertical profiles of plutonium in the South Pacific (SHOTS)
- Plutonium/^{137}Cs ratios as a proxy of geochemical processes (SHOTS(South Pacific), GEOSECS(South Atlantic))
- Deep plutonium

Comparison between alpha spectrometry and ICP-MS

(assuming that $^{240}Pu/^{239}Pu$ atom ratio is equal to global fallout (0.18))



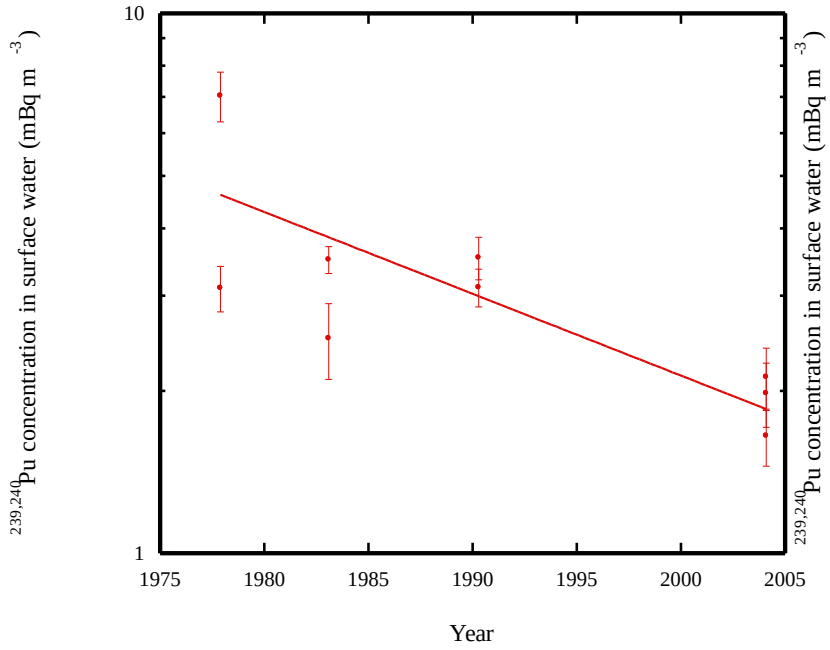
^{239}Pu concentration in surface waters of Southern Hemisphere oceans



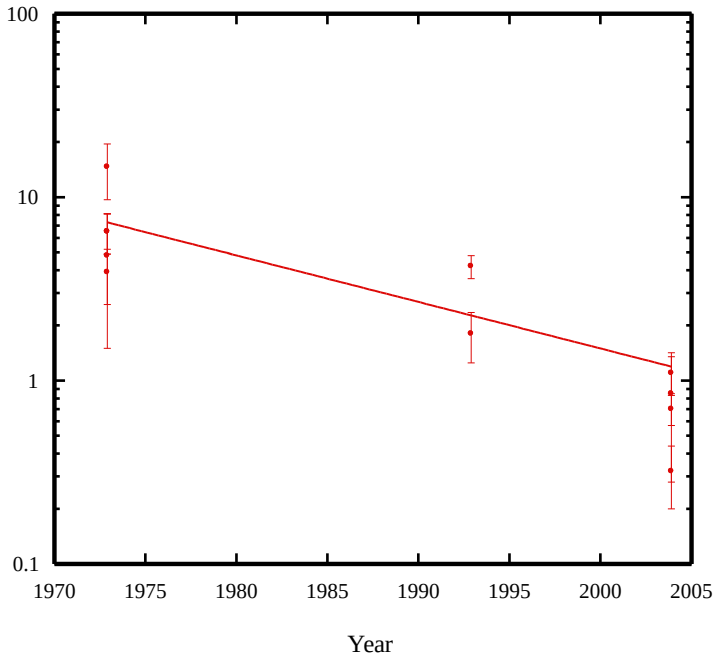
Hirose et al., STOTEN, 2007
Gautaud et al., Prog. Oceanogr. 2011

Temporal change of surface ^{239,240}Pu in Southern Hemisphere oceans

Sea area	*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	$12\ ^{+13}_{-6}$

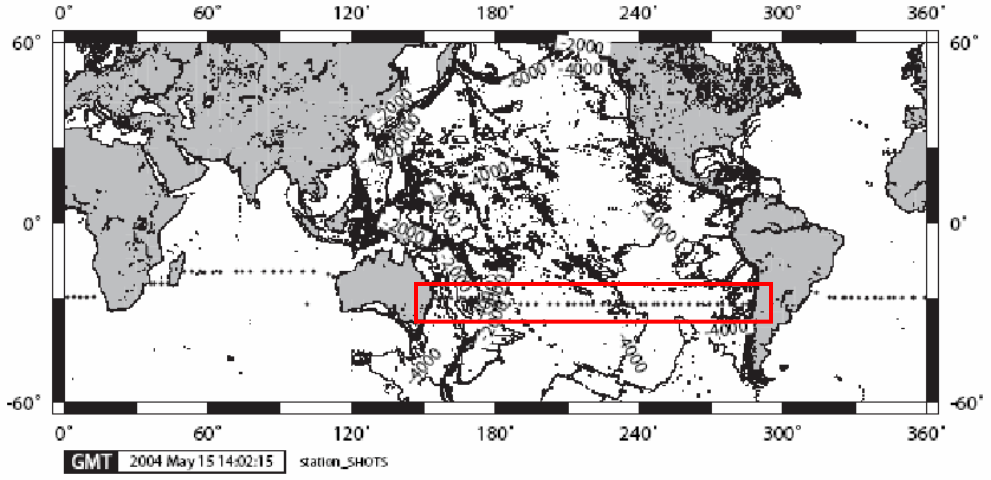


Eastern Indian Ocean

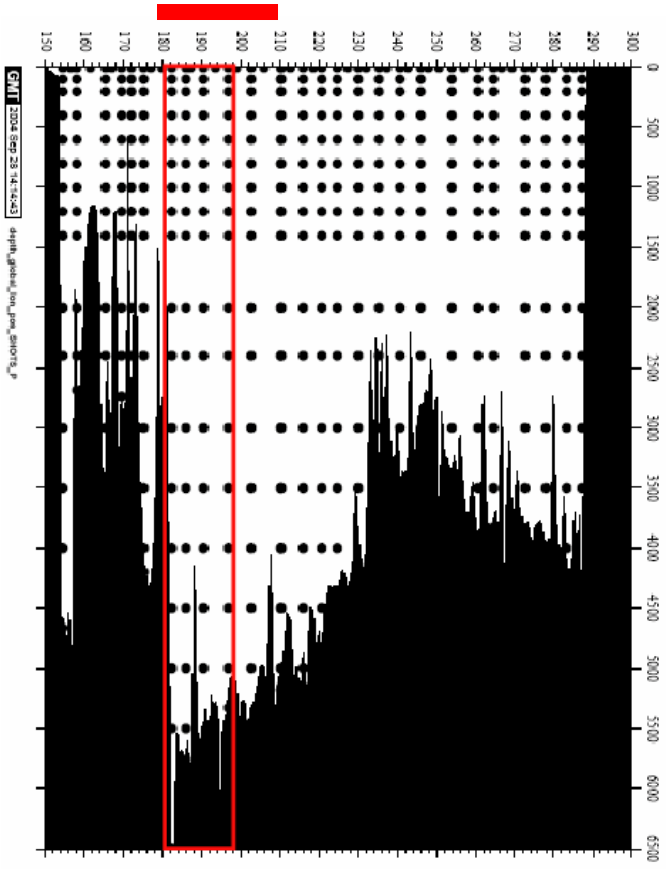


Western South Atlantic

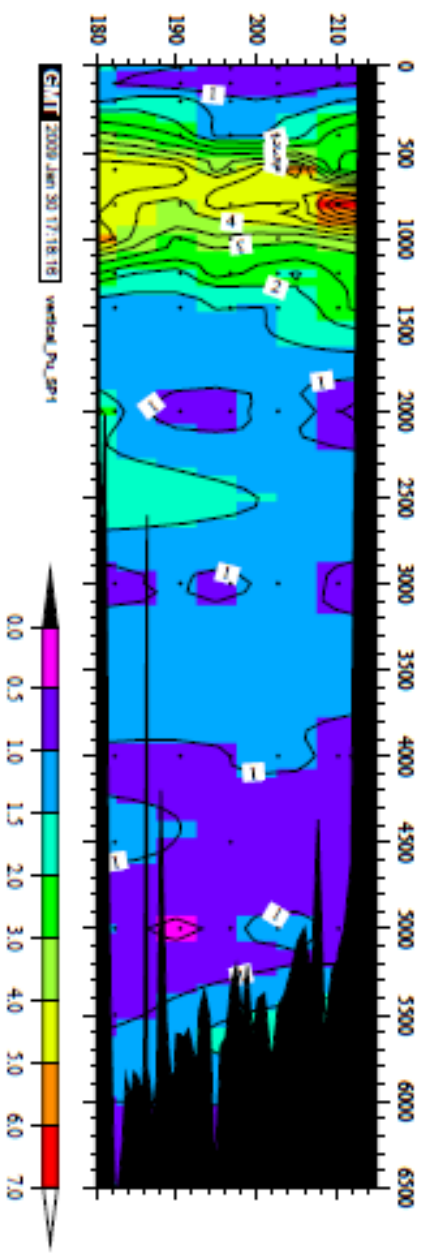
Vertical sampling sites of BEAGLE2003

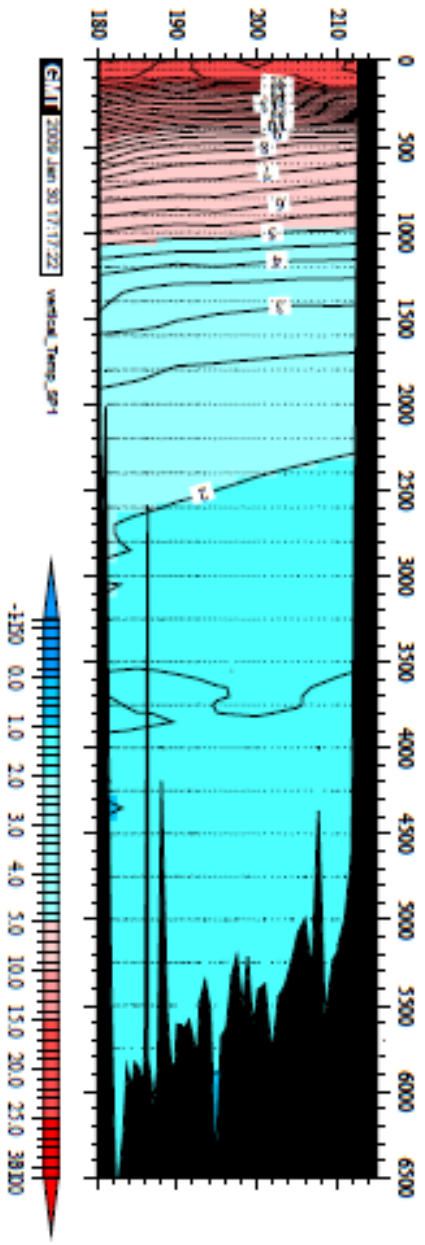


Vertical sampling sites of Pu

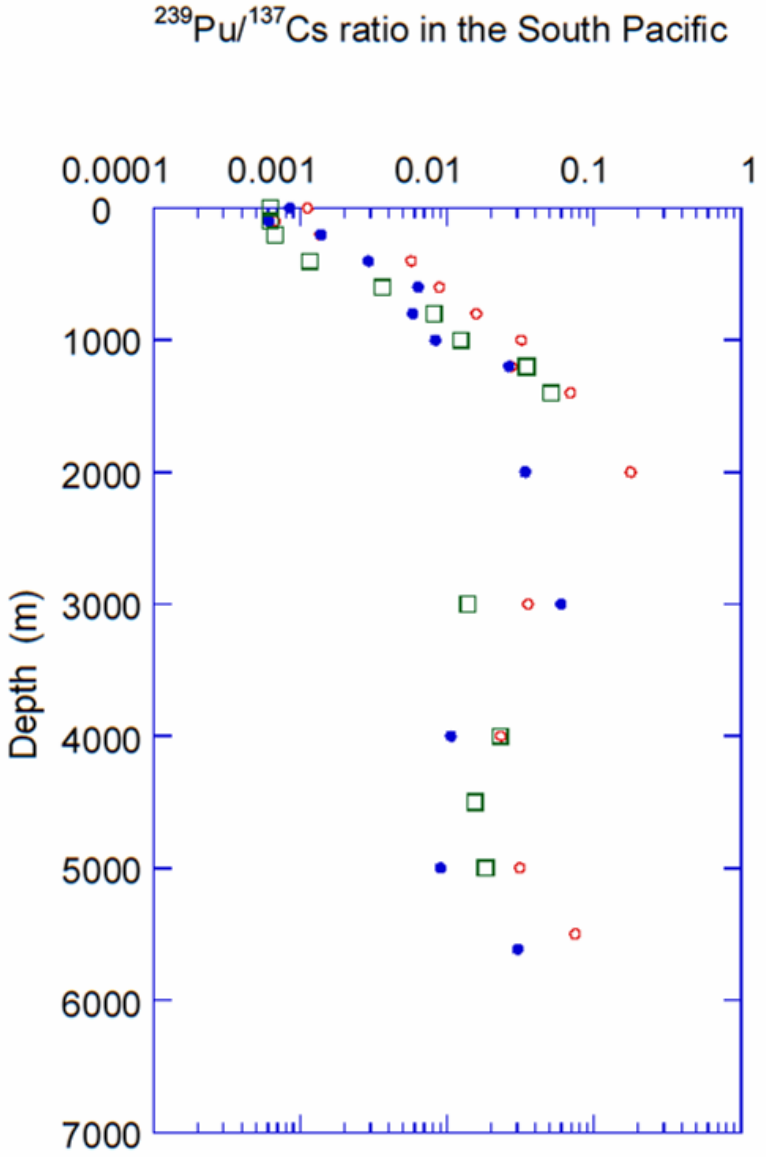


Cross section of ^{239}Pu in the South Pacific subtropical gyre





Hirose et al.,
Prog. Oceanogr. 2011

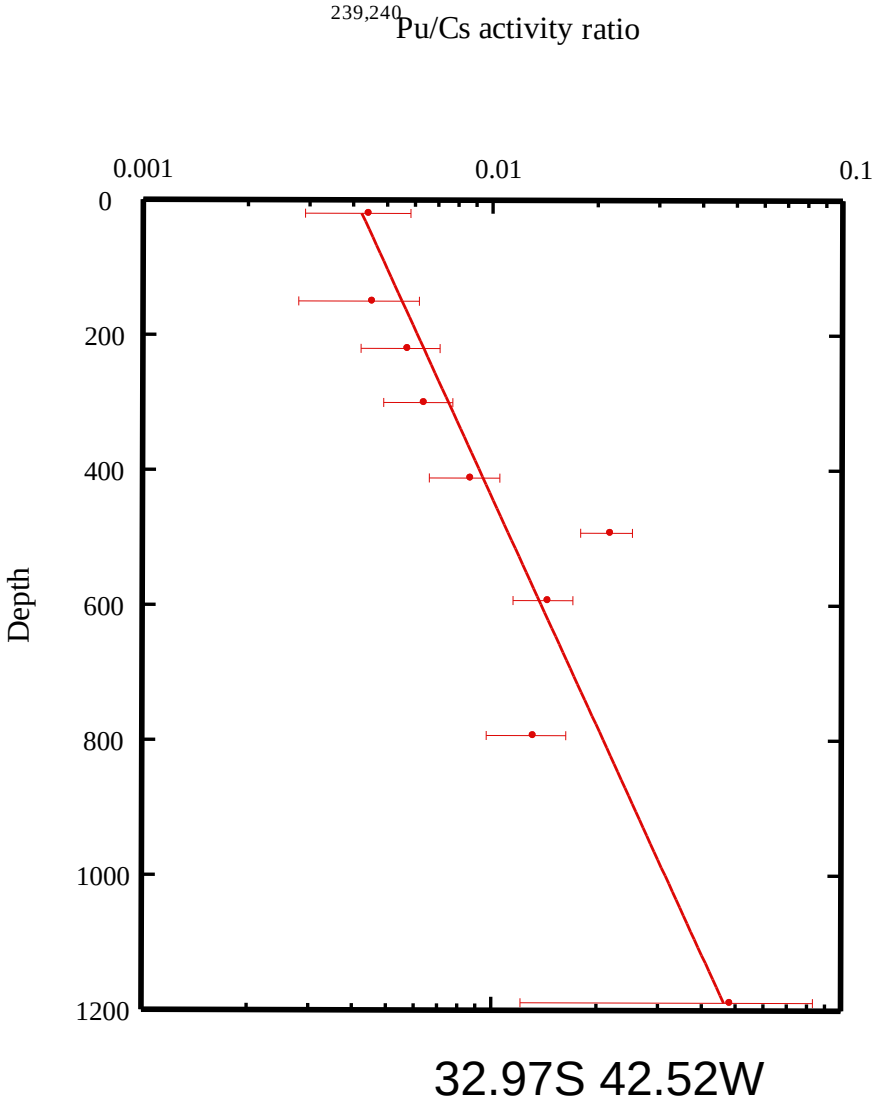


^{239}Pu/^{137}Cs ratio is an indicator of Biogeochemical processes.

1. ^{239}Pu/^{137}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}Pu/^{137}Cs ratios exponentially increased from surface to 1200 m depth for South Atlantic.

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 x 10^{-3}) comparing with that in global fallout (0.009)
- **The $^{239}Pu/^{137}Cs$ ratios exponentially increased from surface to 1500 m depth.**

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

Stn.	$R_{Pu/Cs,o}$ x 10^3	λ value	HRD(m)	correlation factor
175	0.95	0.0032	220	0.967
156	0.74	0.0028	250	0.959
145	0.43	0.0034	200	0.986
136	0.52	0.0031	220	0.991
127	0.53	0.0037	190	0.957

HRD: half-regeneration depth

Vertical changes of $^{239,240}Pu/^{137}Cs$ in shallow layer (South Atlantic)

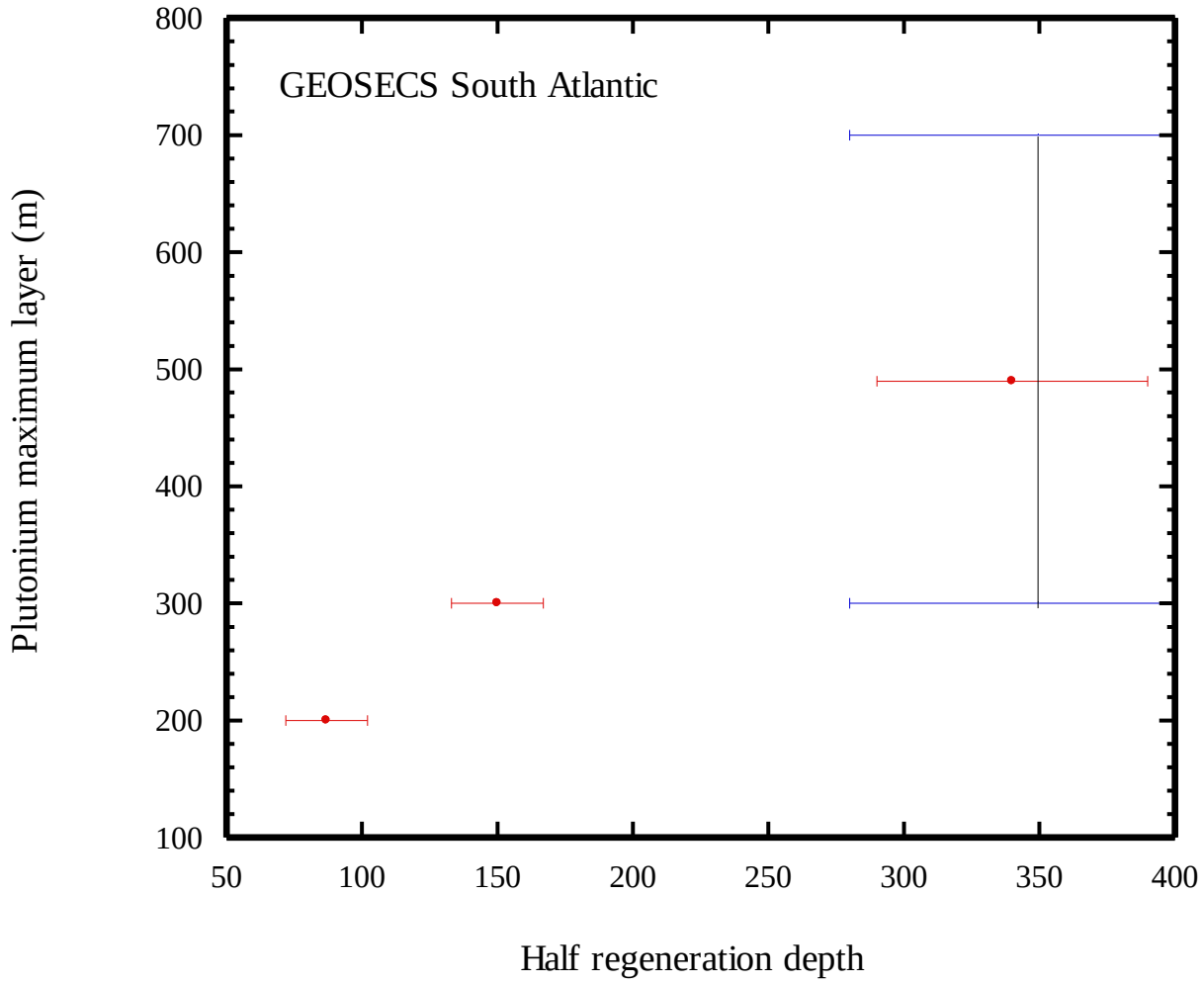
- **The $^{239,240}Pu/^{137}Cs$ ratios in surface layers**

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

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

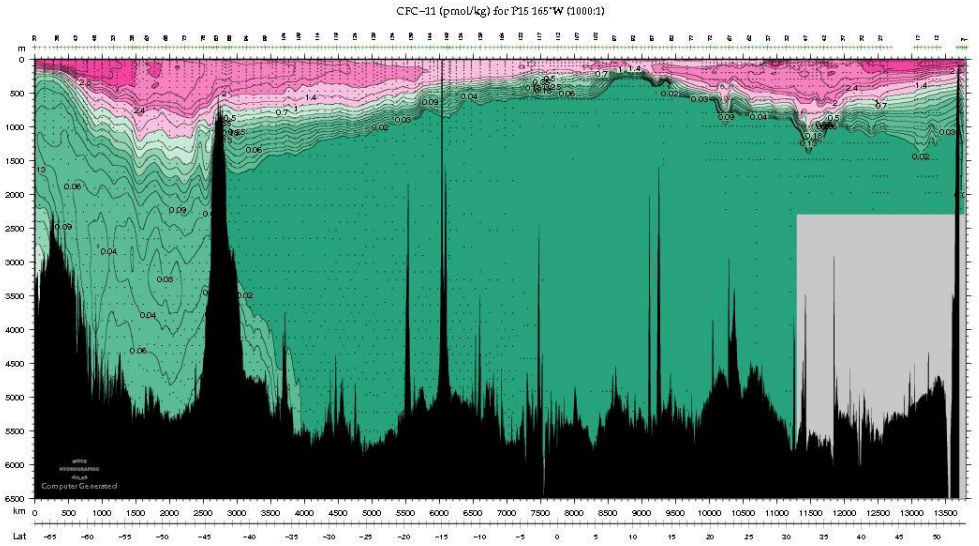
Latitude	$R_{Pu/Cs,o} \times 10^3$	λ value	HRD(m)	correlation factor
15S	0.9	0.0080	87	0.960
21S	1.3	0.0046	150	0.997
33S	4.1	0.0021	340	0.942
45S	4.0	0.0020	350	0.919

Relationship between plutonium maximum layer depth and half-regeneration depth

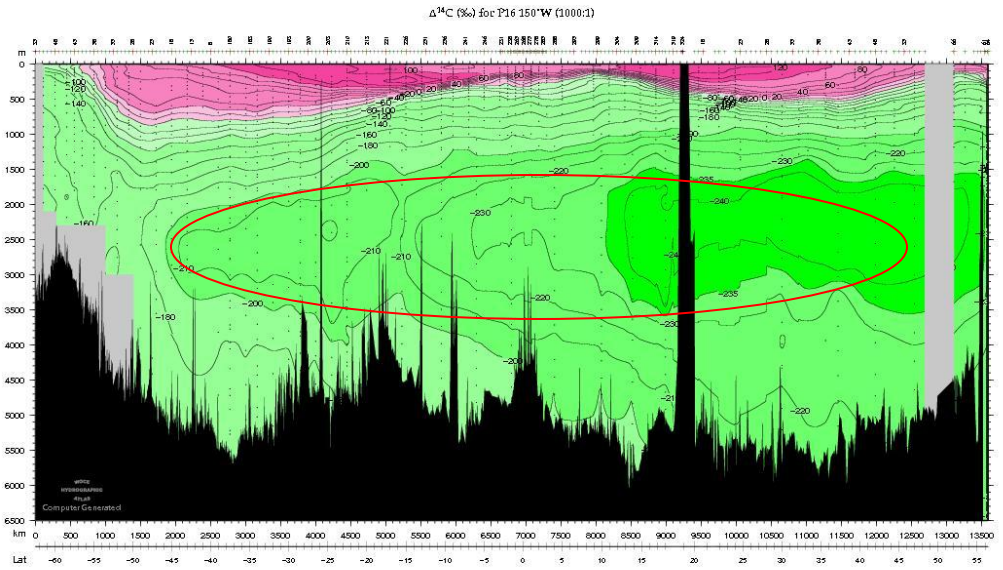


Chemical tracers in deep waters of the Pacific Ocean

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



North Pacific Deep Water (2000 - 3000 m depth)



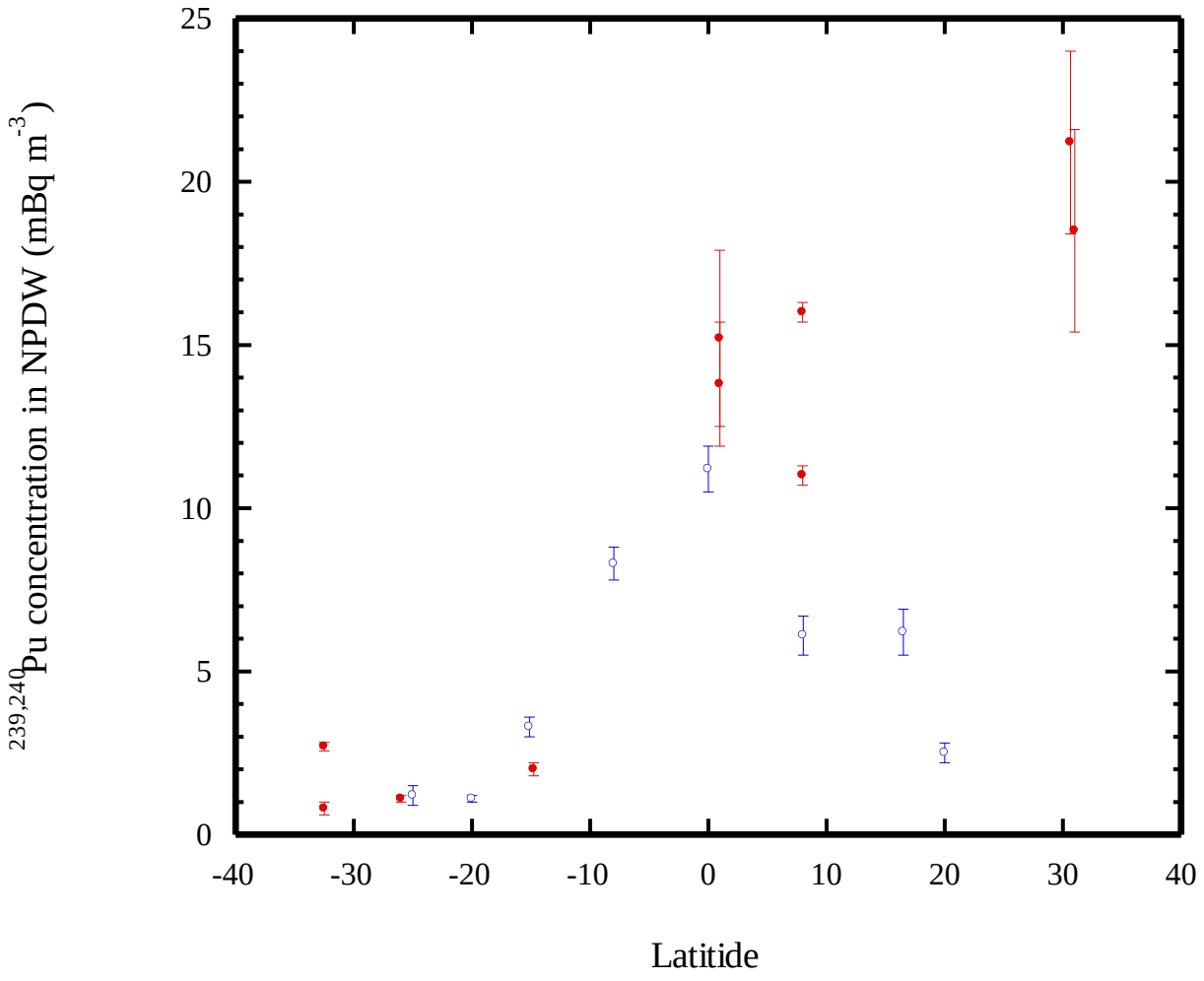
Plutonium in deep water

- Weak biological activities $\Rightarrow$less important biogeochemical processes
- The $^{239}Pu/^{137}Cs$ ratios in deep water showed no increase with increasing depth.

  The relatively low values occurred the depth range from 4000 to 5000 m depth.

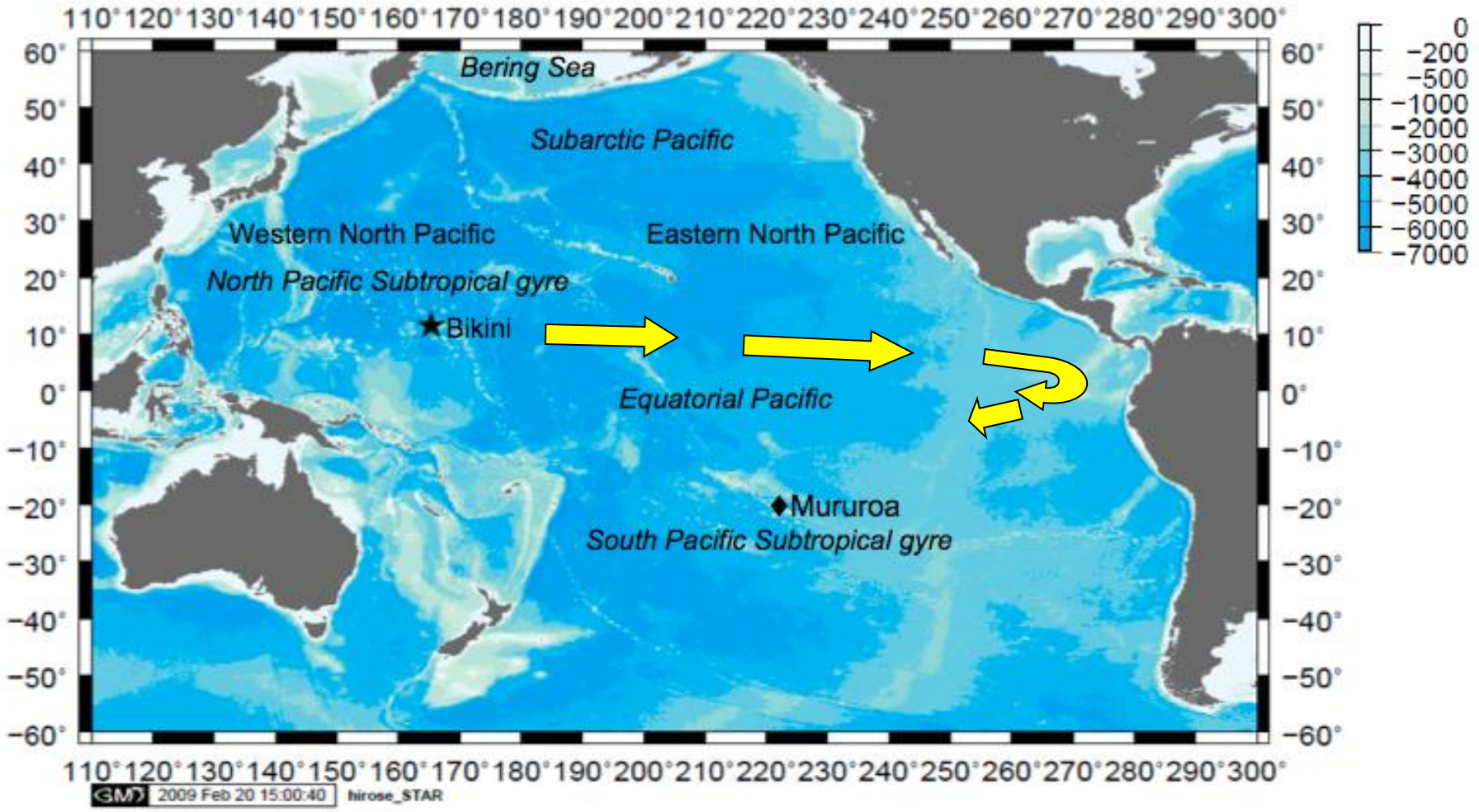
  $\Rightarrow$0.01- 0.03 (0.009: global fallout)
- Plutonium in the South Pacific deep water is supplied by advection rather than biogeochemical processes.

^{239,240}Pu concentration in the North Pacific Deep Water (2000 - 3000 m) (sampling period: 1999 - 2003)



Closed circle: Central Pacific
Open circle: Eastern Pacific , *Kinoshita et al., Sci. Total Environ., 2011*

Possible pathway of the North Pacific Deep Water (2000-3000 m)



Bikini-derived Pu with higher $^{240}Pu/^{239}Pu$ 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!