

$^{134}\text{Cs}$  and  $^{137}\text{Cs}$  in the North Pacific Ocean derived  
from the TEPCO Fukushima Dai-ichi Nuclear Power  
Plant accident, Japan in March 2011:  
Transport processes and estimation of  $^{134}\text{Cs}$  and  
 $^{137}\text{Cs}$  inventories

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Radioactivity in the North Pacific

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# Research team

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# Objectives

- To describe long term behavior of TEPCO FNPP1 released radiocaesium in the coastal region and the North Pacific Ocean based on the observations and model simulations during the period from just after the accident to July 2015.
- To estimate total amount of radiocaesium and other radionuclides released from TEPCO Fukushima NPP1 accident including atmospheric release and direct discharge to the ocean based on observations on land and ocean and model simulations. The obtained numbers should be verified by the facts.

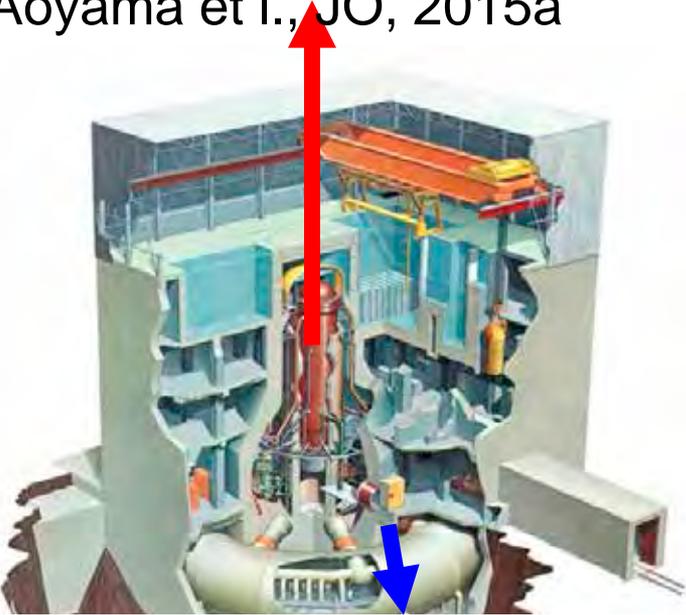
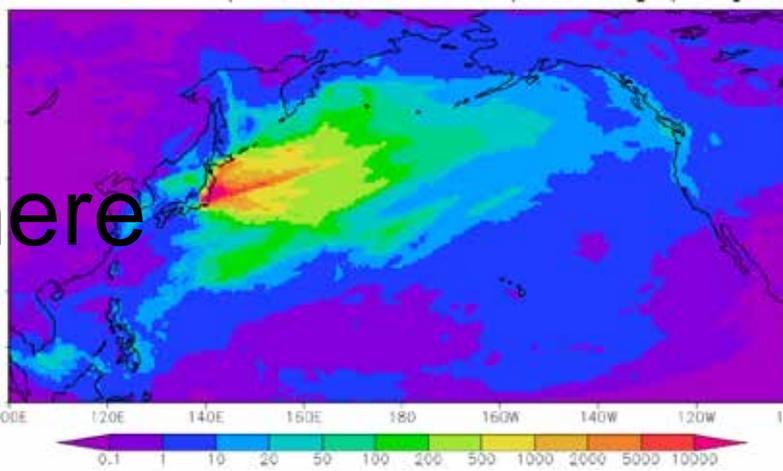
# Talk outlines

- Ultra low background measurement
- Fallout history since 1945 and 3-D distribution of  $^{137}\text{Cs}$  before Fukushima accident in the Pacific Ocean
- Distribution of Fukushima radiocaesium by observations and model simulations
- Estimation of total amount of released radiocaesium to the atmosphere and the ocean
- Conclusions

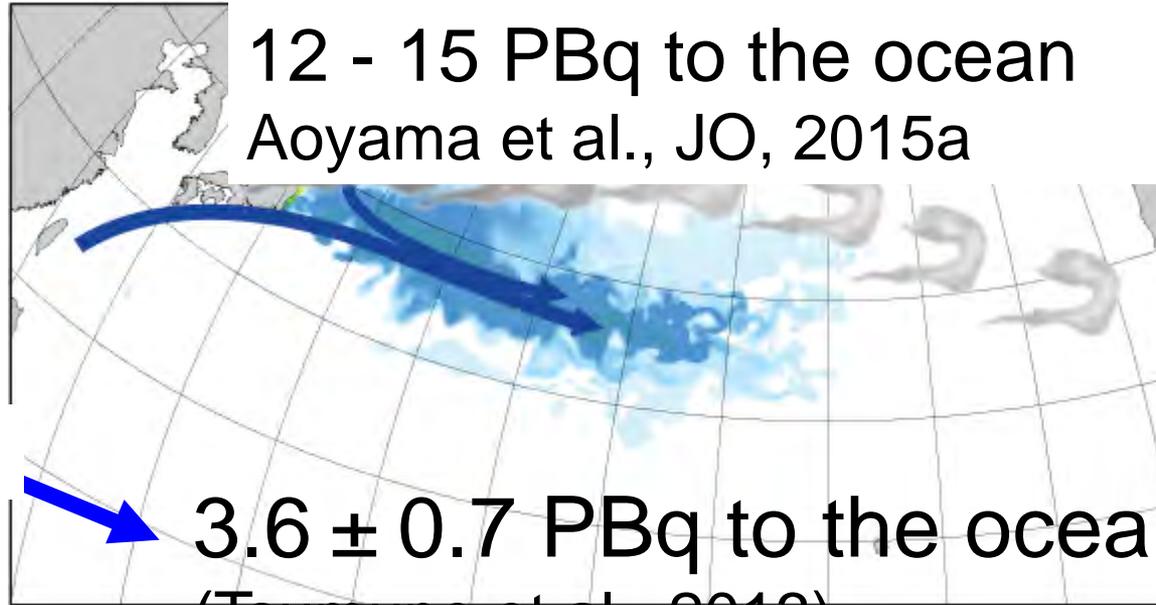
# $^{137}\text{Cs}$ mass balance

14–17 PBq to the atmosphere

Aoyama et al., JO, 2015a



12 - 15 PBq to the ocean  
Aoyama et al., JO, 2015a



140 PBq in stagnant water

Boiling Water Reactor Systems "Nuclear Reactor Concepts" Workshop Manual, U.S. NRC

$3.6 \pm 0.7$  PBq to the ocean  
(Tsumune et al., 2013)

700 PBq was in the three core  
(Nishihara et al., 2011)

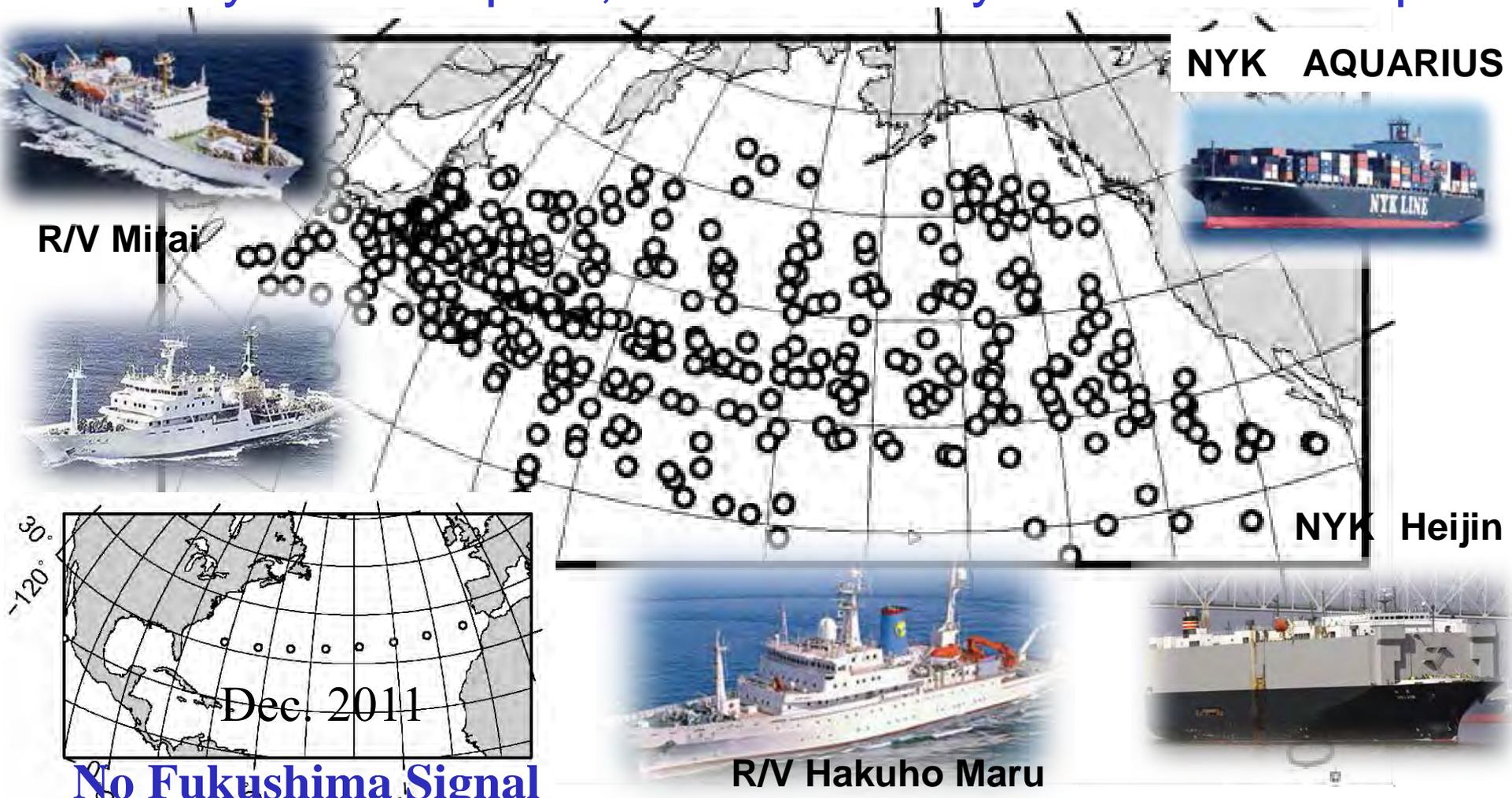
230 PBq recovered

TEPCO unpublished data

440 Sampling locations during the period from March 2011 to Dec 2012 for surface water. 2 liter samples => (1 mBq in the sample)  
low level techniques necessary because

Area  $13 \times 10^6 \text{ km}^2$ , depth 100 m, volume  $1.3 \times 10^{15} \text{ m}^3$

If activity is  $0.5 \text{ Bq m}^{-3}$ , total inventory will be 0.7 PBq



# Two underground laboratories No. 1 in Japan

Ogoya underground laboratory  
(Kanazawa University)

Y. Hahajima

**11 of 18 Ge detectors were used**



Located inside a tunnel – a former Cu-mine  
Covered by 135 m rock

Shields from old lead from  
Kanazawa castle

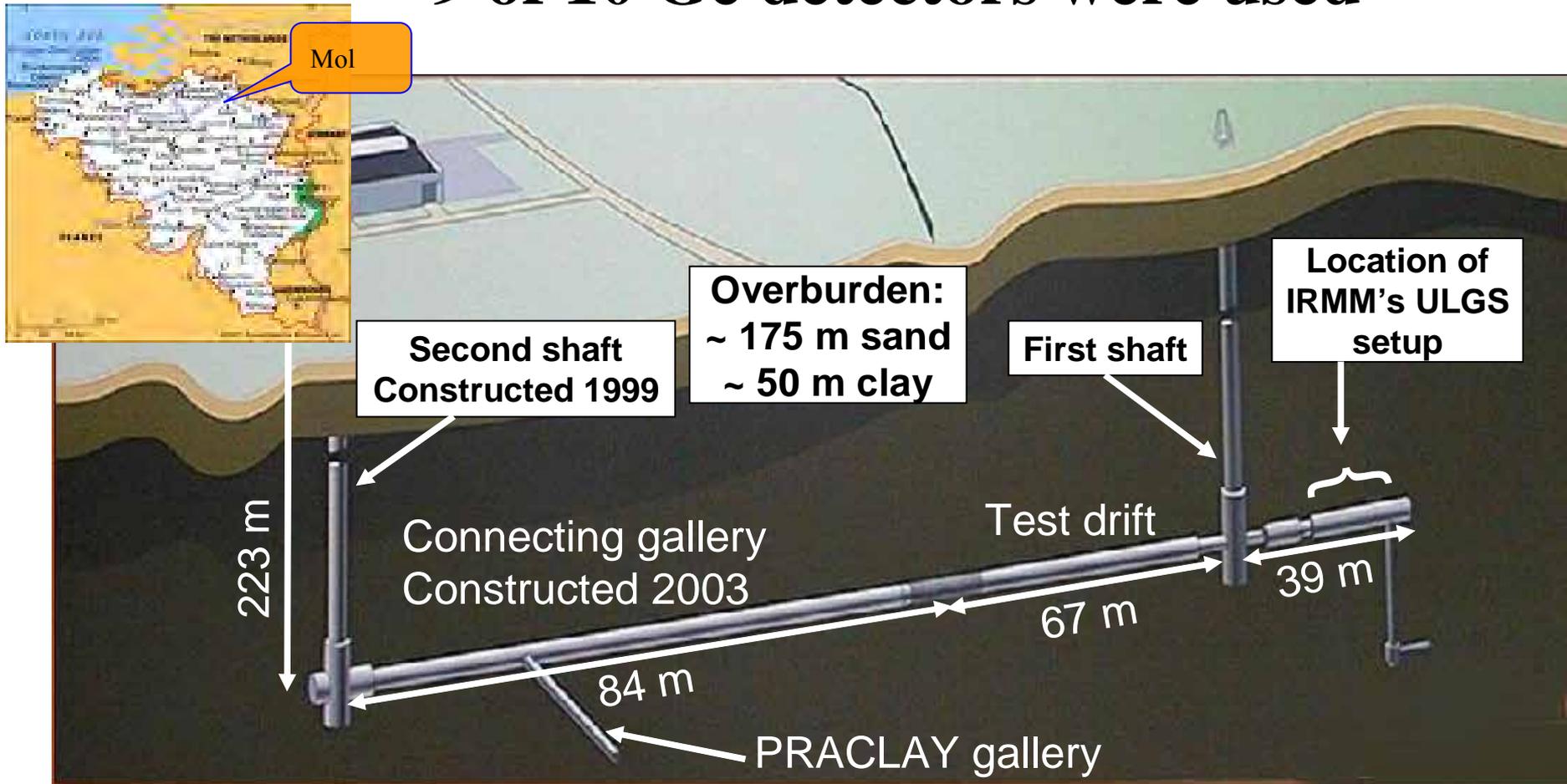
# Two underground laboratories No. 2 in the EU

HADES underground laboratory

(EC-JRC-IRMM, located at the premises of SCK in Mol, Belgium)

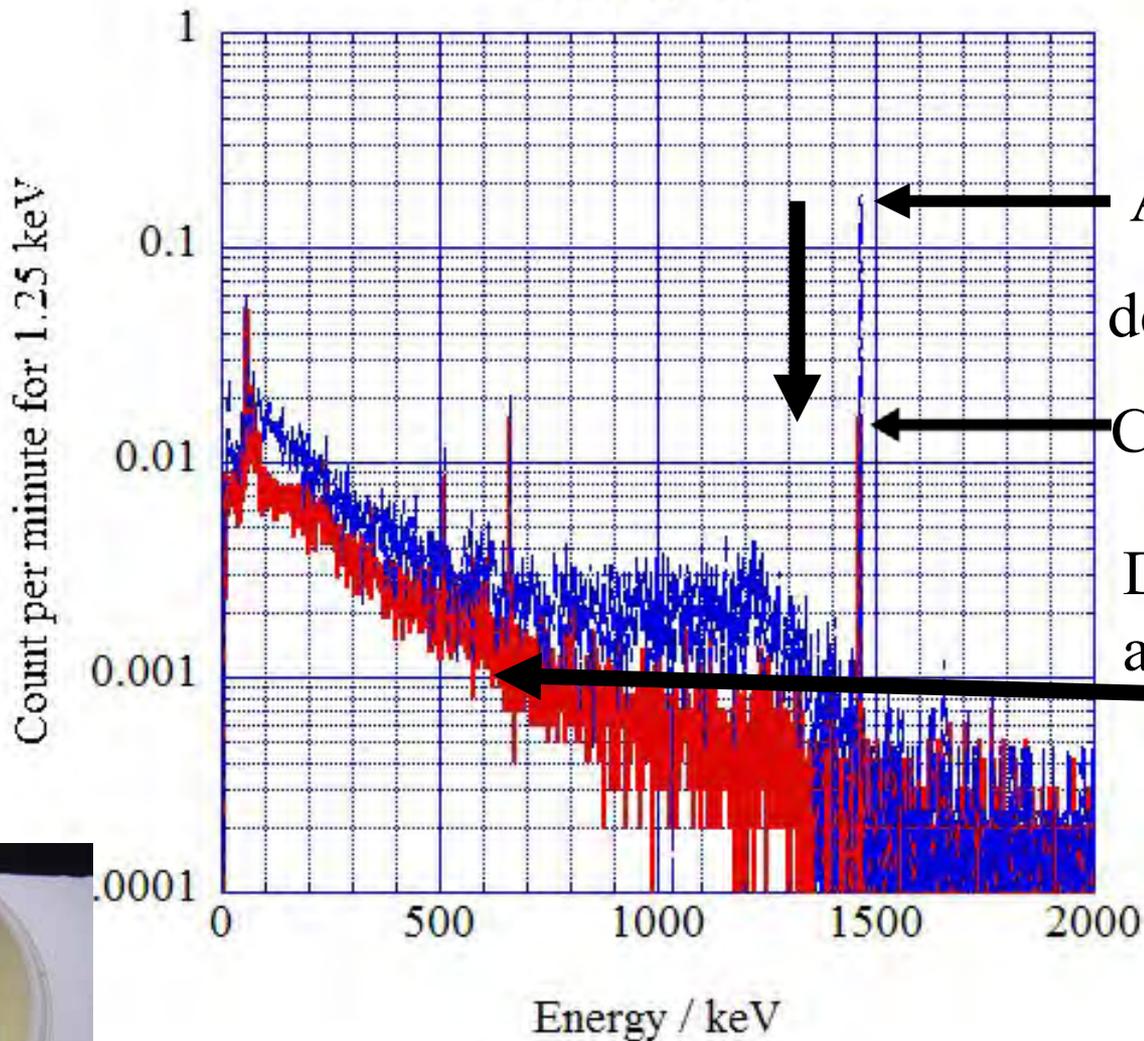
M. Hult

## 9 of 10 Ge detectors were used



# Double treatment by Aoyama and Hirose 2008 to reduce $^{40}\text{K}$

KH1108-273



AMP/Cs compound  
decrease  $^{40}\text{K}$  content

$\text{Cs}_2\text{Pt}(\text{Cl})_4$  precipitate

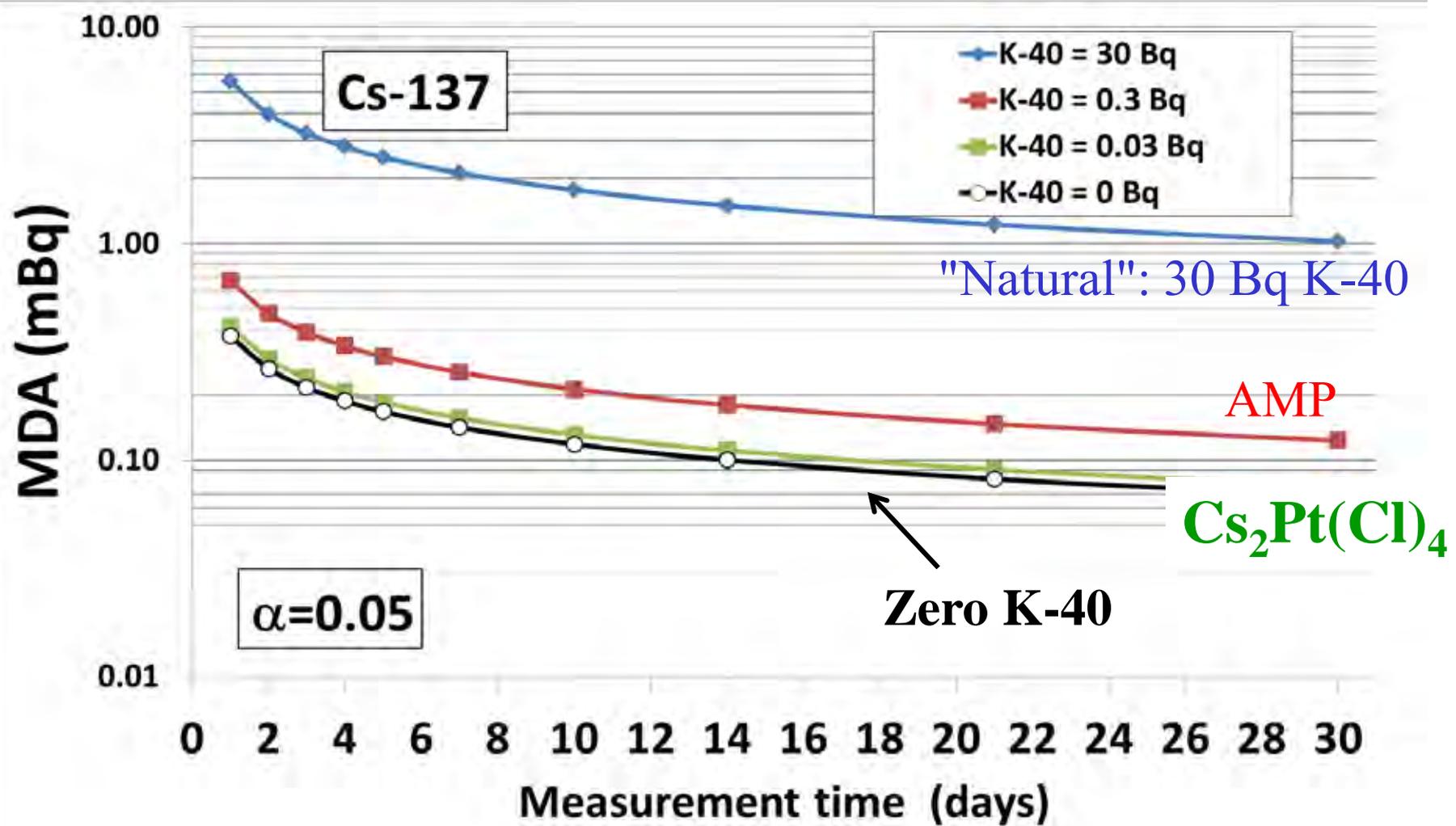
Decrease compton as expected



<- AMP/Cs compound

$\text{Cs}_2\text{Pt}(\text{Cl})_4$  precipitate ->





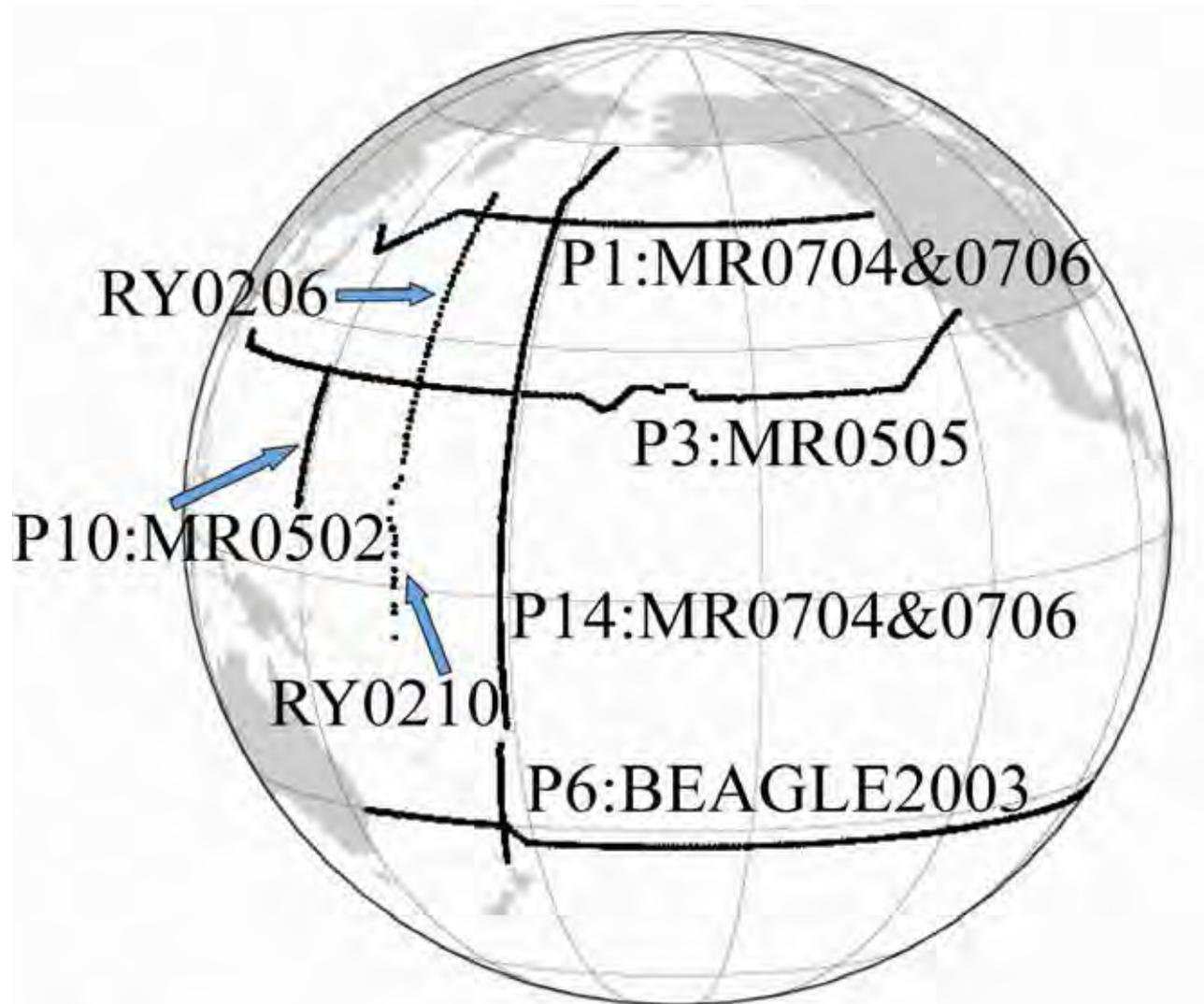
Detection limits (MDA) for  $^{137}\text{Cs}$  in a 4-g sea water precipitate sample, measured in a 2-kg well-detector (Ge-12 in HADES), as a function of measurement time for different activities of  $^{40}\text{K}$  in the sample.

**Ca. 0.1 mBq of  $^{137}\text{Cs}$  can be measured**

# samples and range of $^{137}\text{Cs}$ activities

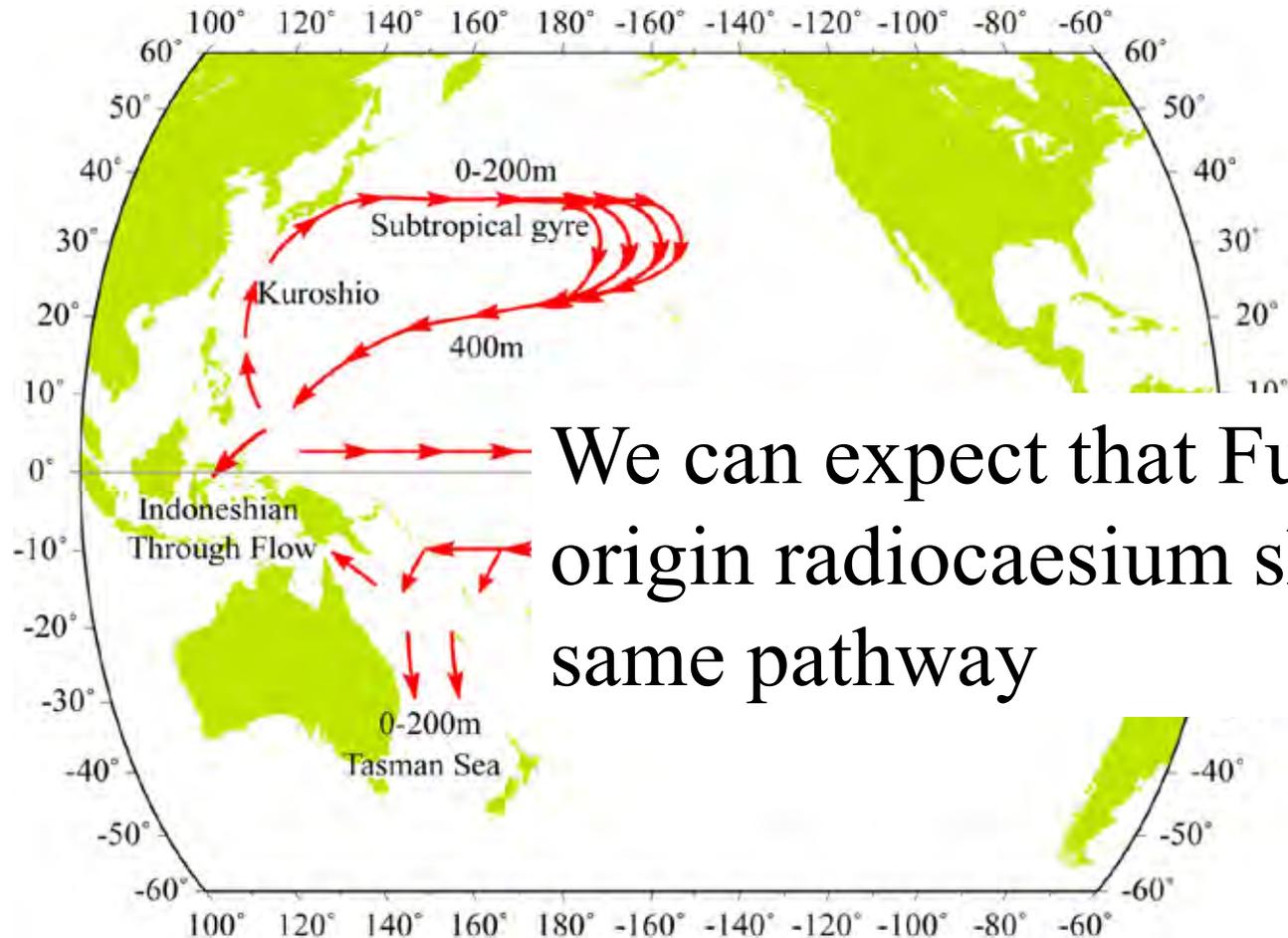
- Surface seawater 2 liters 1 mBq 1 Bq
- Subsurface seawater 10 liters 1 mBq 0.1 Bq
- Deep seawater 100-200 liters 1 mBq 5 mBq
- Particles 10 mg 100 mg 0.1 mBq 5 mBq
- Contents in the stomach of fish 0.1 mBq 1 mBq

# Observations in the Pacific Ocean from 2002 to 2007





# A pathway of weapons tests derived $^{137}\text{Cs}$ in the North Pacific Ocean, tracer of sea water movement

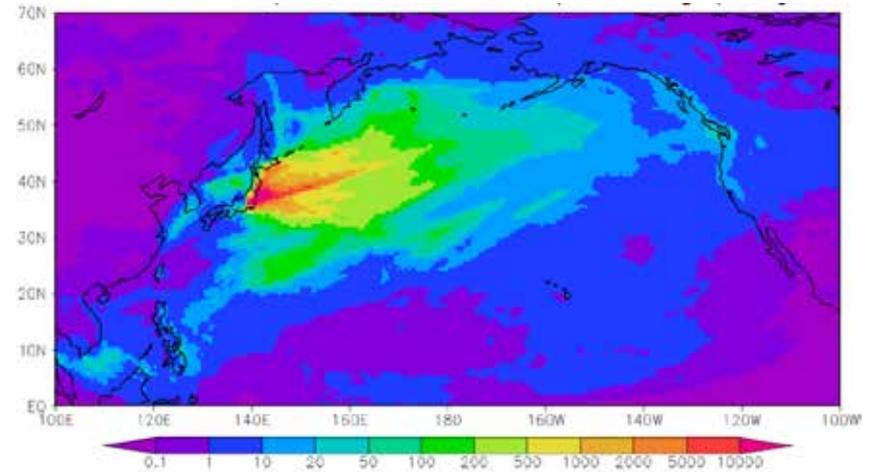


We can expect that Fukushima origin radiocaesium should go same pathway

Fig. S2 Possible pathway of  $^{137}\text{Cs}$  in the Pacific Ocean based on 3-D observations of  $^{137}\text{Cs}$  in 2000s (Aoyama et al., 2013)

# Possible pathways to the Ocean

- Atmospheric deposition
- Direct release
- Groundwater discharge
- Freshwater runoff from the 1F NPP site
- River runoff (initial stage)
- Planned release of low-level waste water

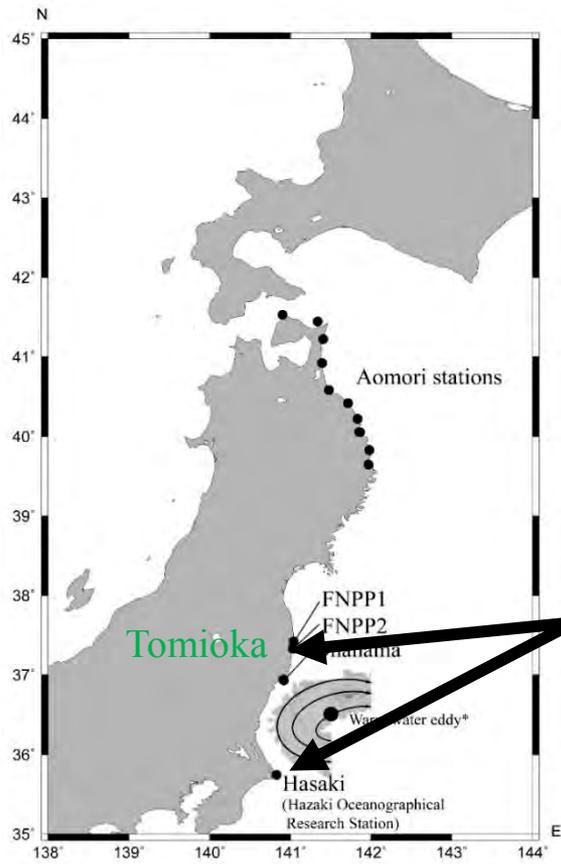
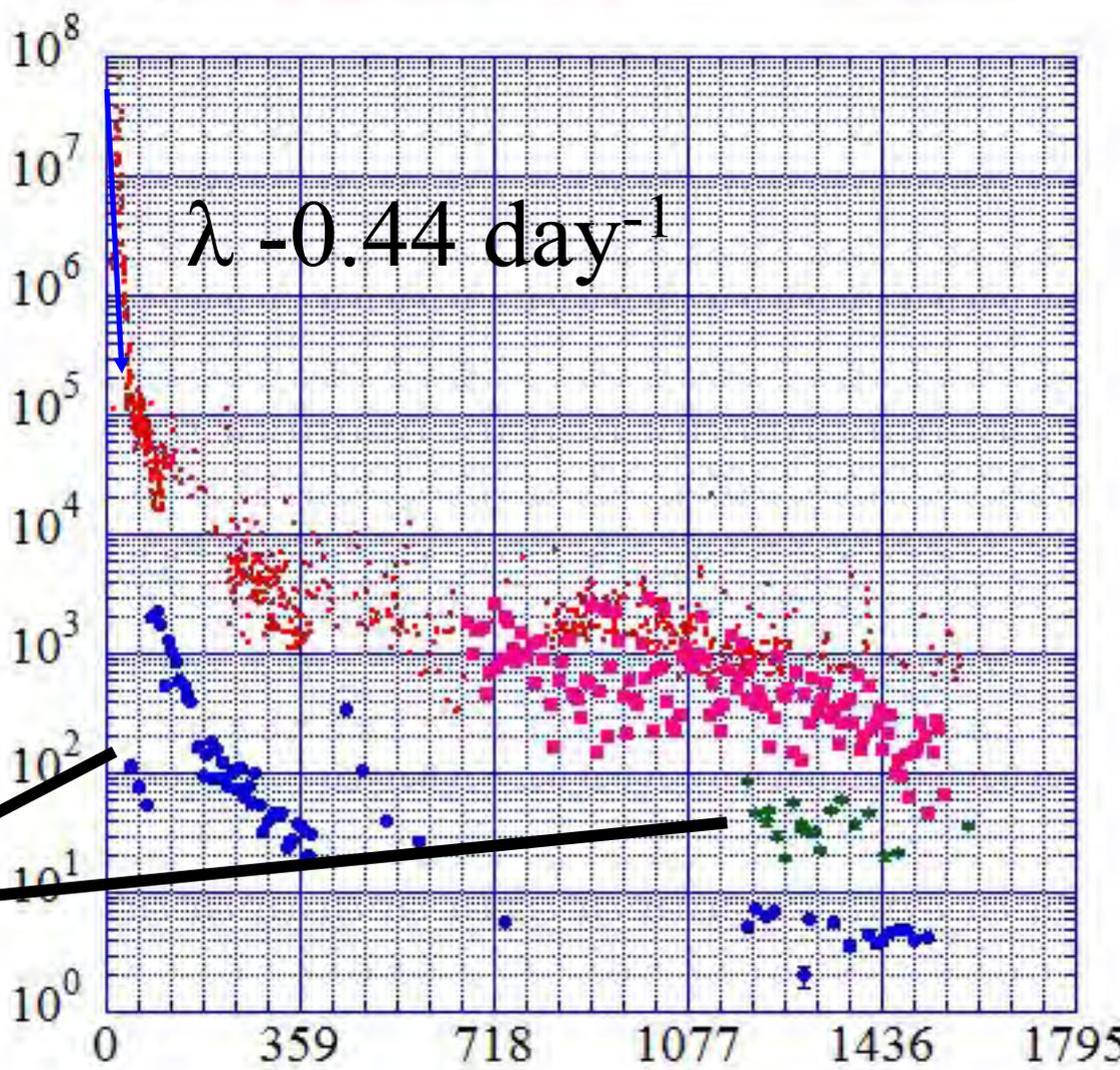
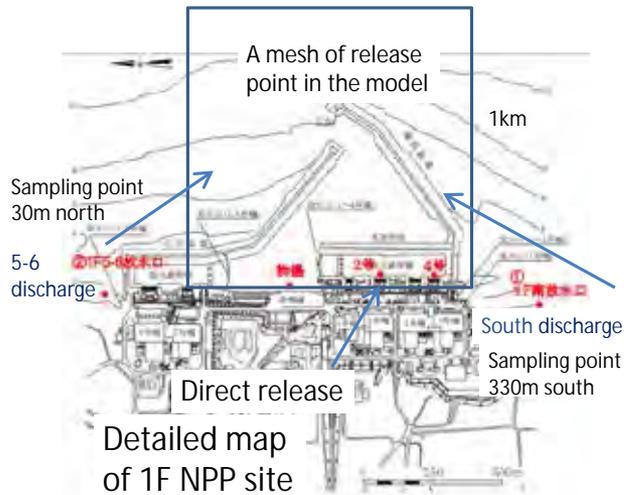


Simulated atmospheric deposition of  $^{137}\text{Cs}$  ( $\text{Bq m}^{-2}$ ) (by Masingar II of MRI) (Aoyama et al., JO, 2015a)

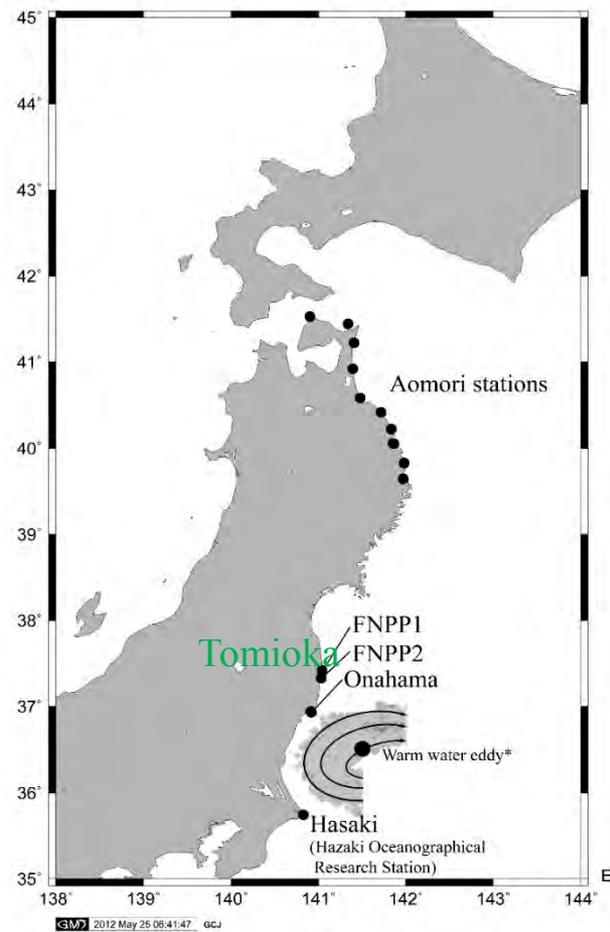
$^{137}\text{Cs}$  :  $1.8\text{E}+12 \text{ Bq/m}^3$

TEPCO Press release

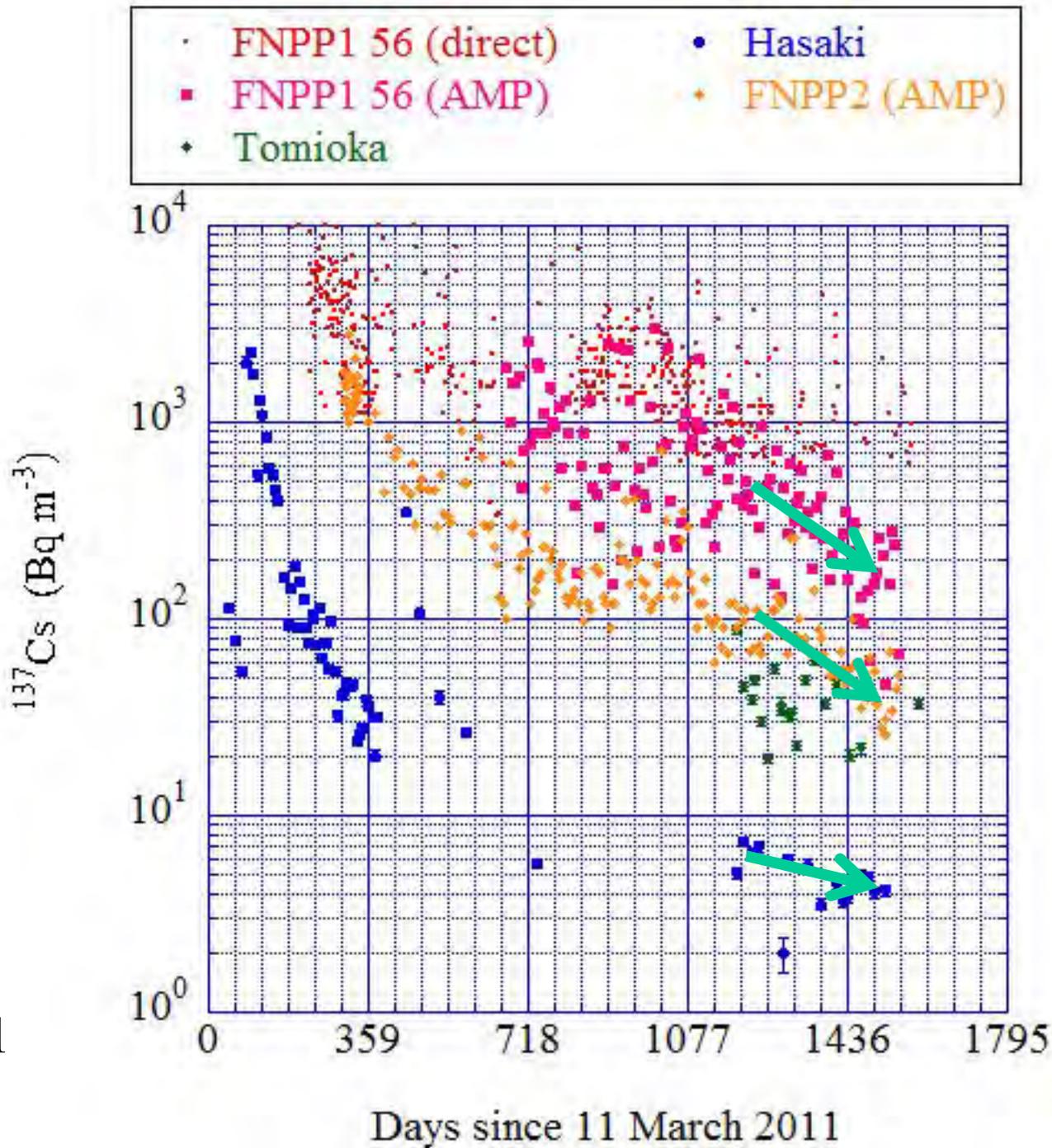




Days since 11 March 2011



Flux from FNPP1  
 2013 30 GBq day<sup>-1</sup>  
 2014 10 GBq day<sup>-1</sup>  
 2015 a few GBq day<sup>-1</sup>



Days since 11 March 2011

# Radiocaesium in surface water in April-May 2011

Station	Depth dbar	Latitude	Longitude	Date	<sup>134</sup> Cs Bq m <sup>-3</sup>	<sup>137</sup> Cs Bq m <sup>-3</sup>
NYK11-043	0	34.95°N	143.86°E	20110331	135 ± 10	150 ± 8
NYK11-001	0	35.68°N	143.77°E	20110401	507 ± 33	546 ± 28
NYK11-003	0	36.60°N	147.60°E	20110401	1000 ± 71	1080 ± 60
NYK11-044	0	35.07°N	146.44°E	20110401	34 ± 2.6	36.8 ± 2.1
NYK11-007	0	38.18°N	154.97°E	20110402	17.7 ± 1.6	21.5 ± 1.3

examples

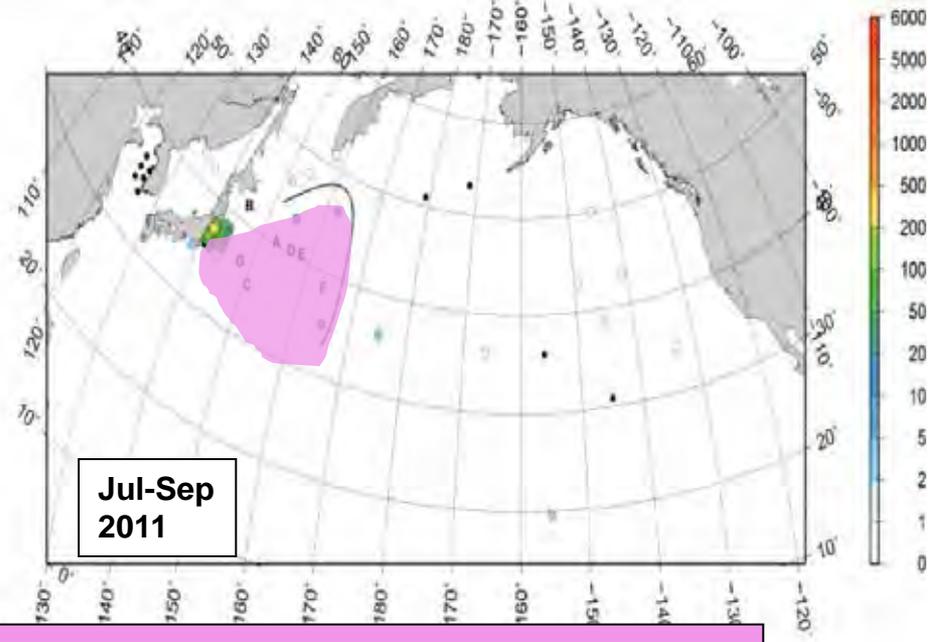
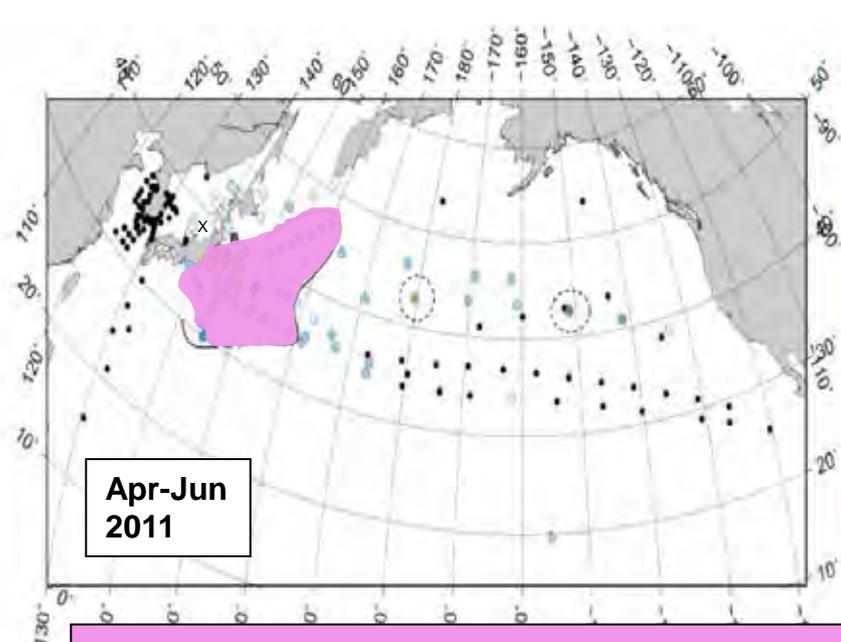
Latitude	Longitude	Date	<sup>134</sup> Cs Bq m <sup>-3</sup>	<sup>137</sup> Cs Bq m <sup>-3</sup>
36.60°N	147.60°E	20110401	1000 ± 71	1080 ± 60
36.35°N	178.99°E	20110406	1.1 ± 0.6	2.4 ± 0.4
41.12°N	167.75°W	20110407	ND	1.8 ± 0.2

NYK11-091	0	35.15°N	158.95°E	20110405	3.4 ± 0.7	7.5 ± 0.8
NYK11-092	0	35.32°N	161.70°E	20110405	3.4 ± 0.6	5.3 ± 0.4
NYK11-095	0	36.35°N	178.99°E	20110406	1.1 ± 0.6	2.4 ± 0.4
NYK11-021	0	41.12°N	167.75°W	20110407	ND	1.8 ± 0.2
NYK11-023	0	42.33°N	159.88°W	20110408	0.6 ± 0.3	1.9 ± 0.2
NYK11-025	0	43.00°N	151.95°W	20110409	ND	1.8 ± 0.2
NYK11-055	0	33.46°N	154.15°W	20110409	ND	1.4 ± 0.2
NYK11-098	0	36.84°N	163.23°W	20110409	ND	1.9 ± 0.2
NYK11-027	0	43.62°N	143.57°W	20110410	0.7 ± 0.3	2.3 ± 0.2
NYK11-056	0	32.52°N	146.59°W	20110410	ND	1.8 ± 0.2
NYK11-100	0	35.88°N	151.92°W	20110410	ND	1.8 ± 0.2
NYK11-125	0	33.29°N	142.20°E	20110410	3.1 ± 0.6	3.5 ± 0.4
NYK11-029	0	38.18°N	134.97°W	20110411	ND	1.4 ± 0.2
NYK11-057	0	31.32°N	140.20°W	20110411	ND	1.9 ± 0.2
NYK11-101	0	34.97°N	146.43°W	20110411	ND	2 ± 0.2
NYK11-127	0	35.36°N	147.57°E	20110411	2.2 ± 0.5	3.3 ± 0.4
NYK11-102	0	33.92°N	141.12°W	20110412	ND	2 ± 0.2
NYK11-103	0	32.50°N	135.86°W	20110412	ND	1.8 ± 0.2
NYK11-129	0	39.01°N	152.70°E	20110412	1.9 ± 0.4	3.6 ± 0.3
NYK11-058	0	28.94°N	130.73°W	20110413	ND	1.6 ± 0.2

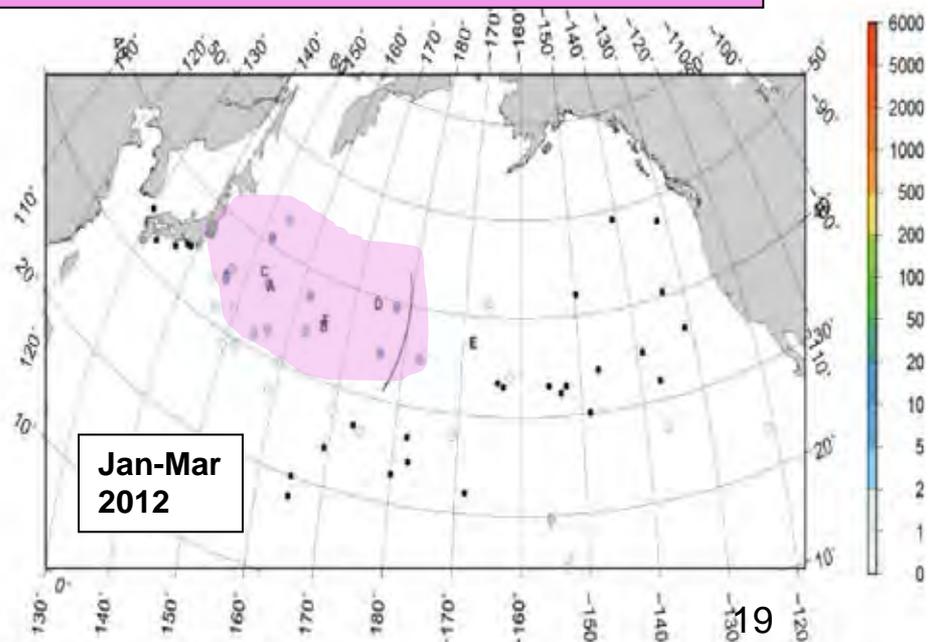
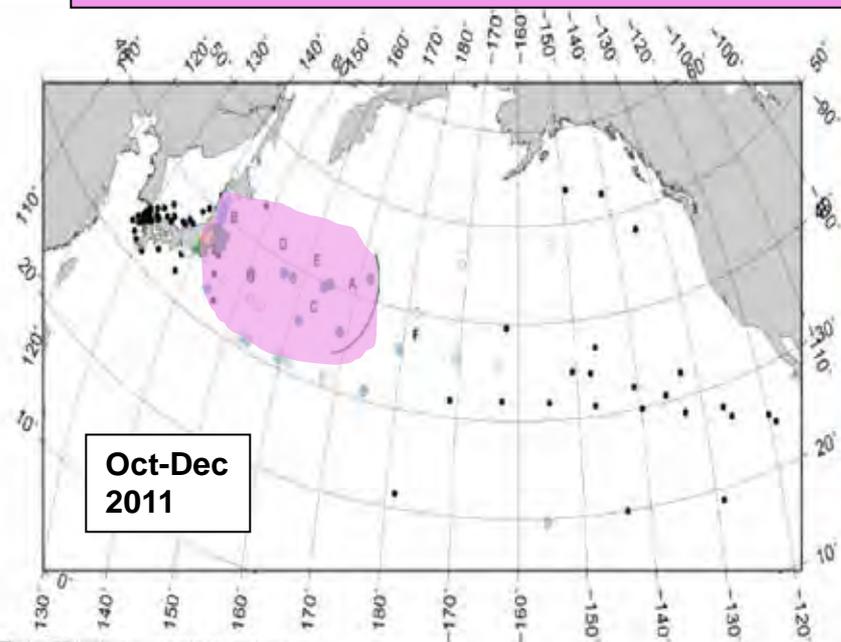
Fukushima

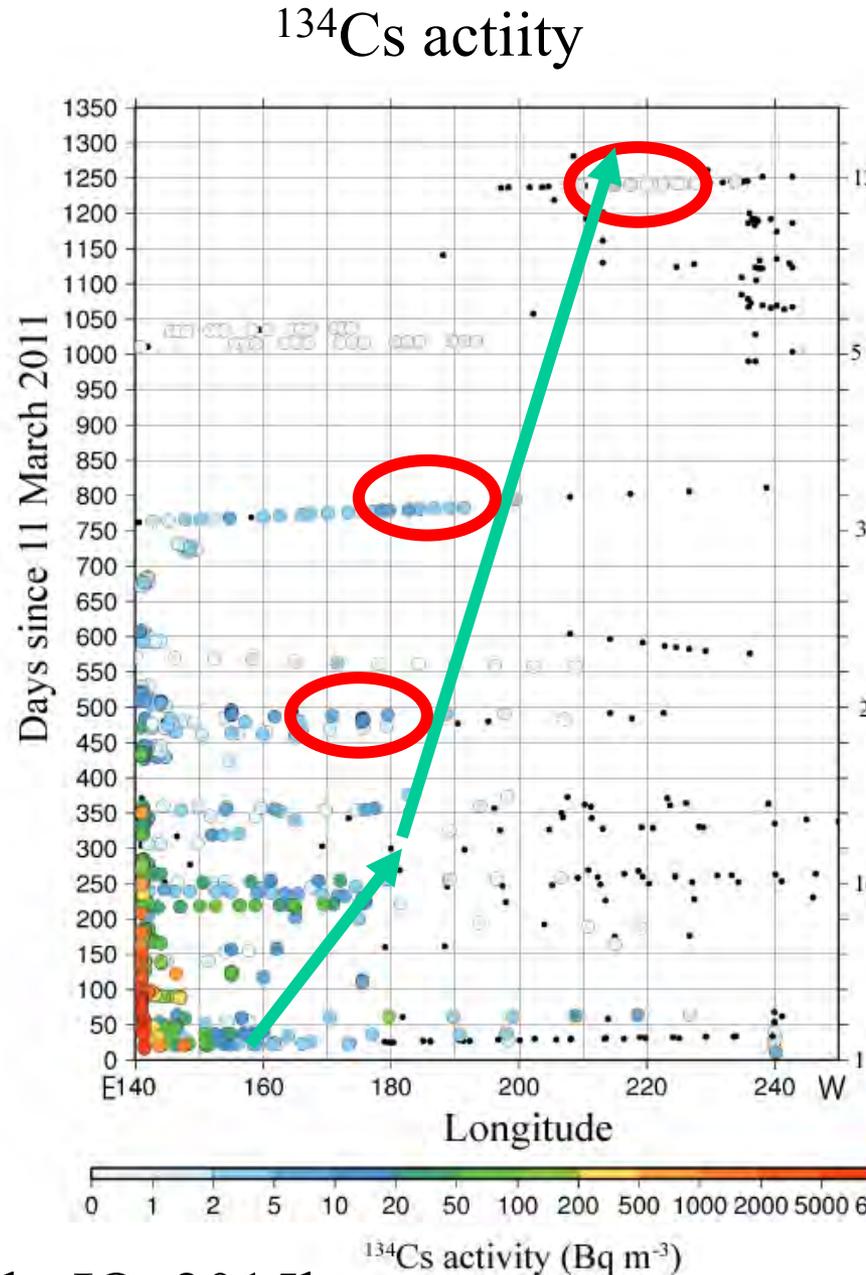
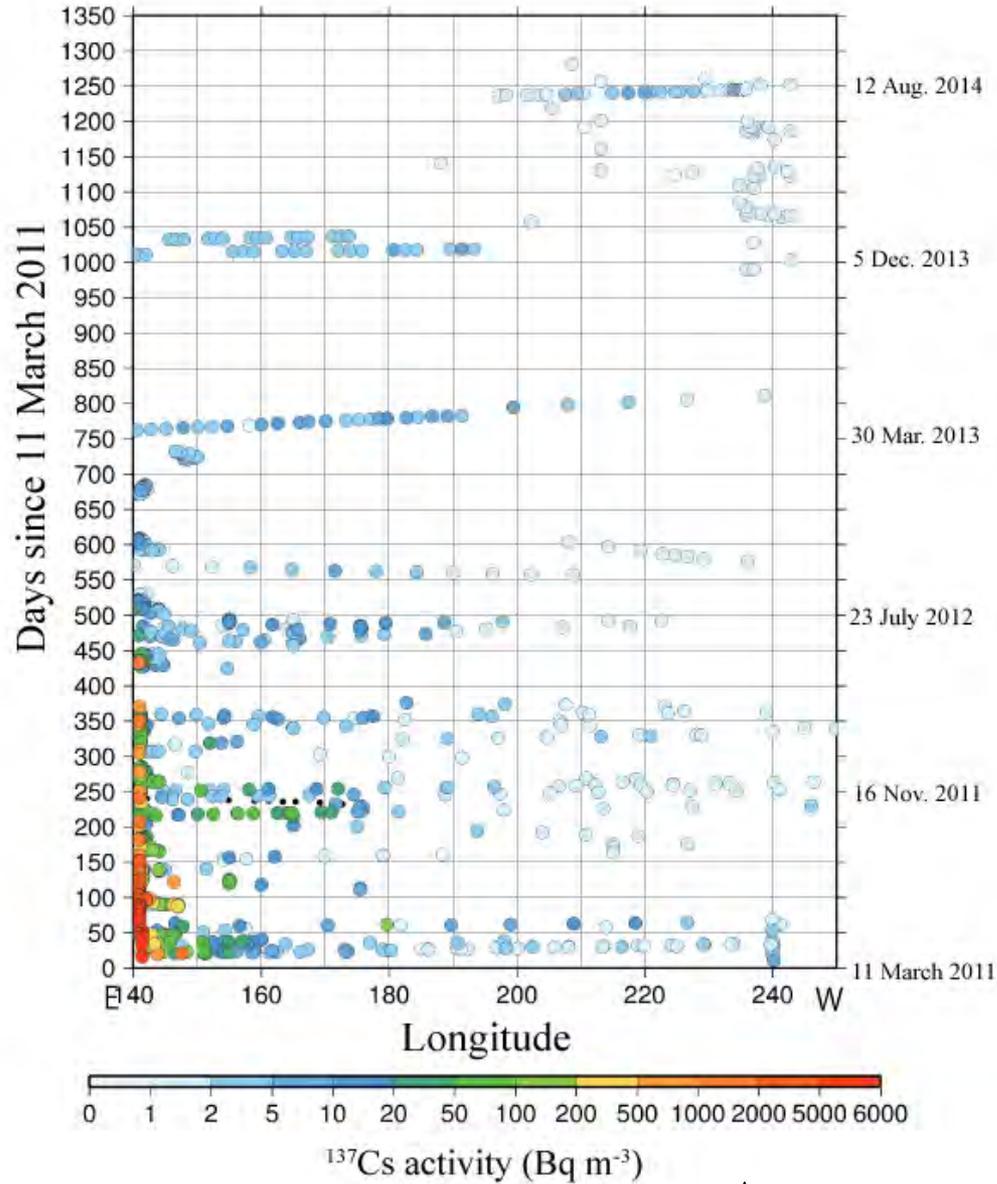
Fukushima+ bomb

Aoyama et al., 2013 BG

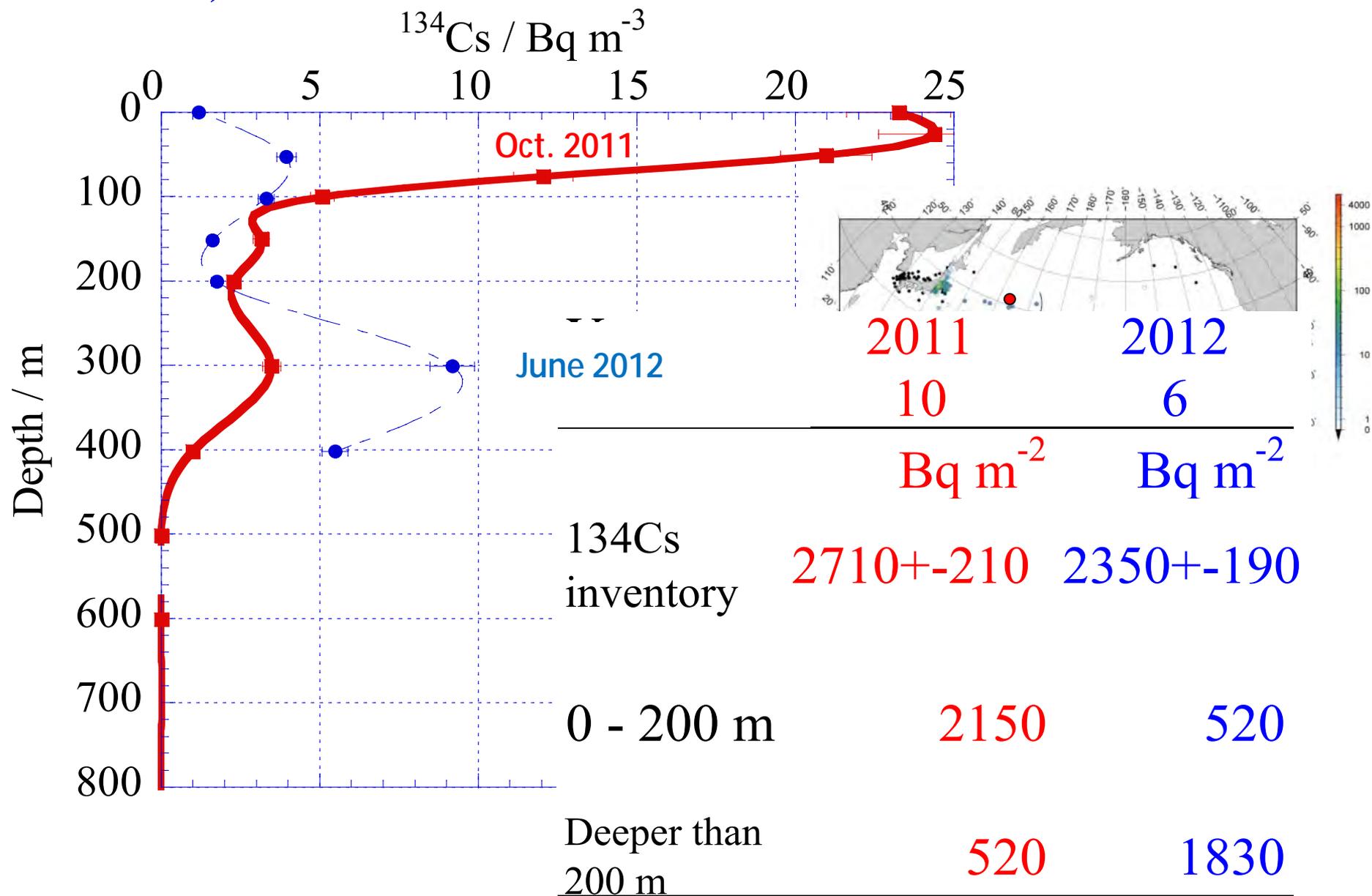


**Surface speed of contaminated water ca. 7 km day<sup>-1</sup>**



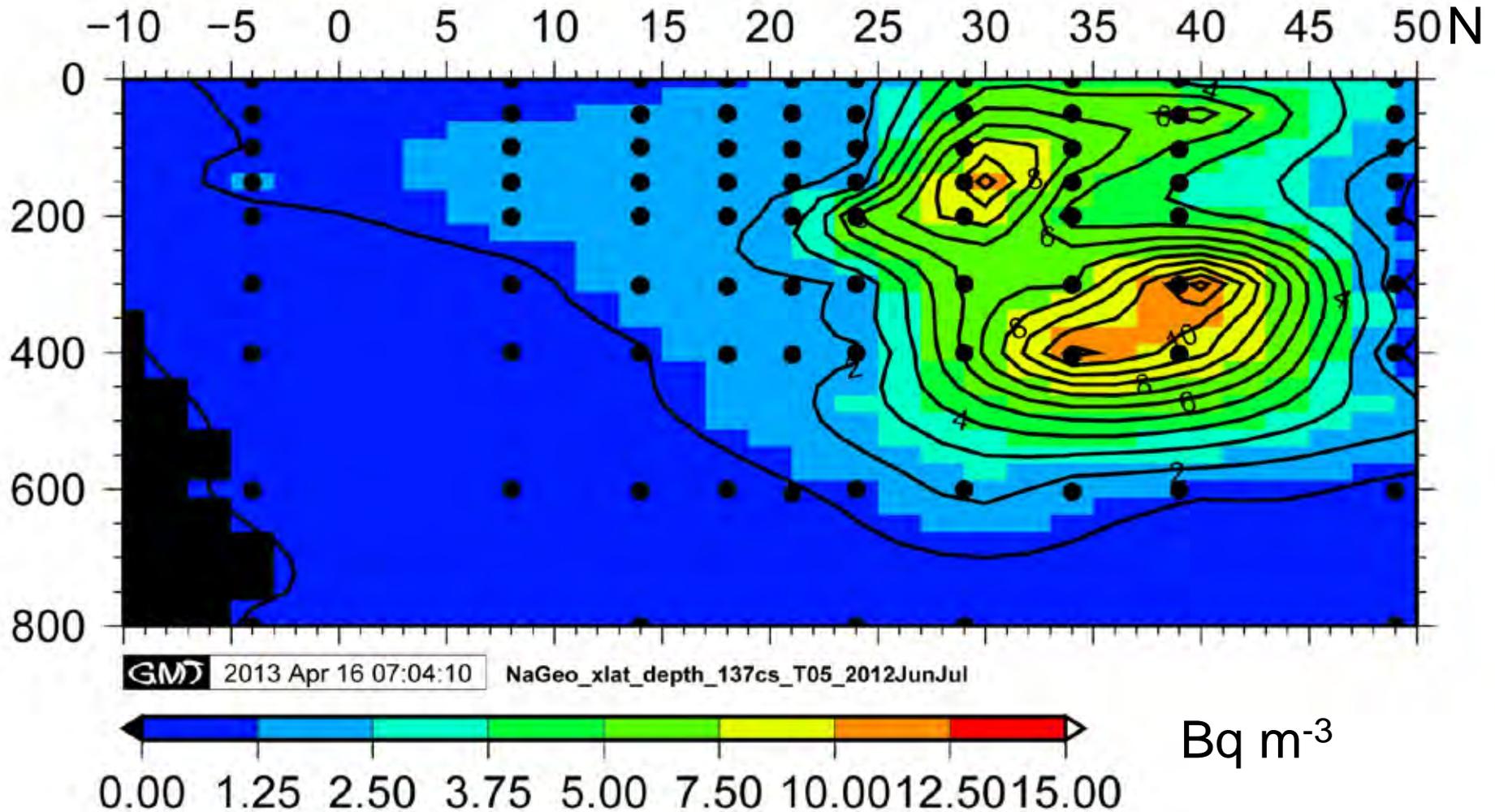


# Over time, Cs moves east and subducted in the ocean interior



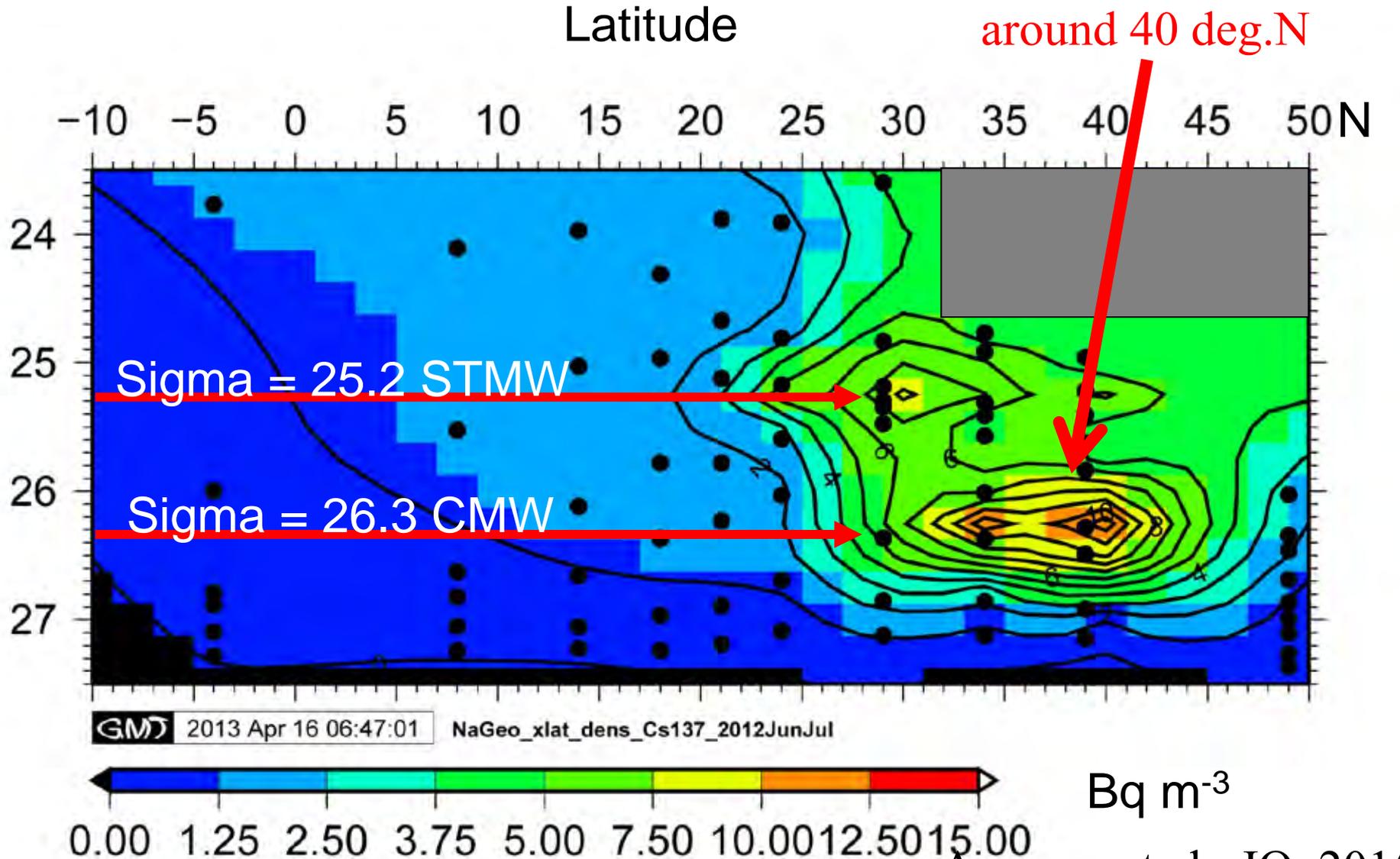
# $^{137}\text{Cs}$ cross section along 165 °E in June 2012

## Start of southward travel into ocean interior



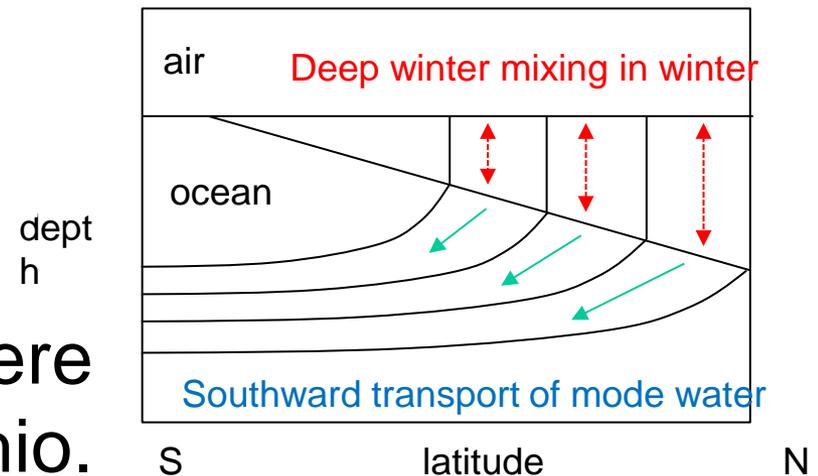
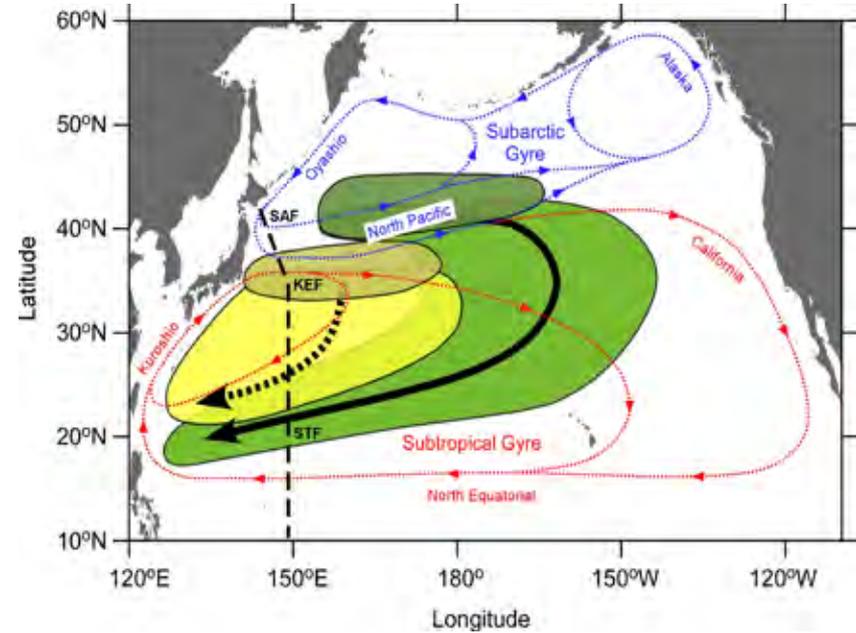
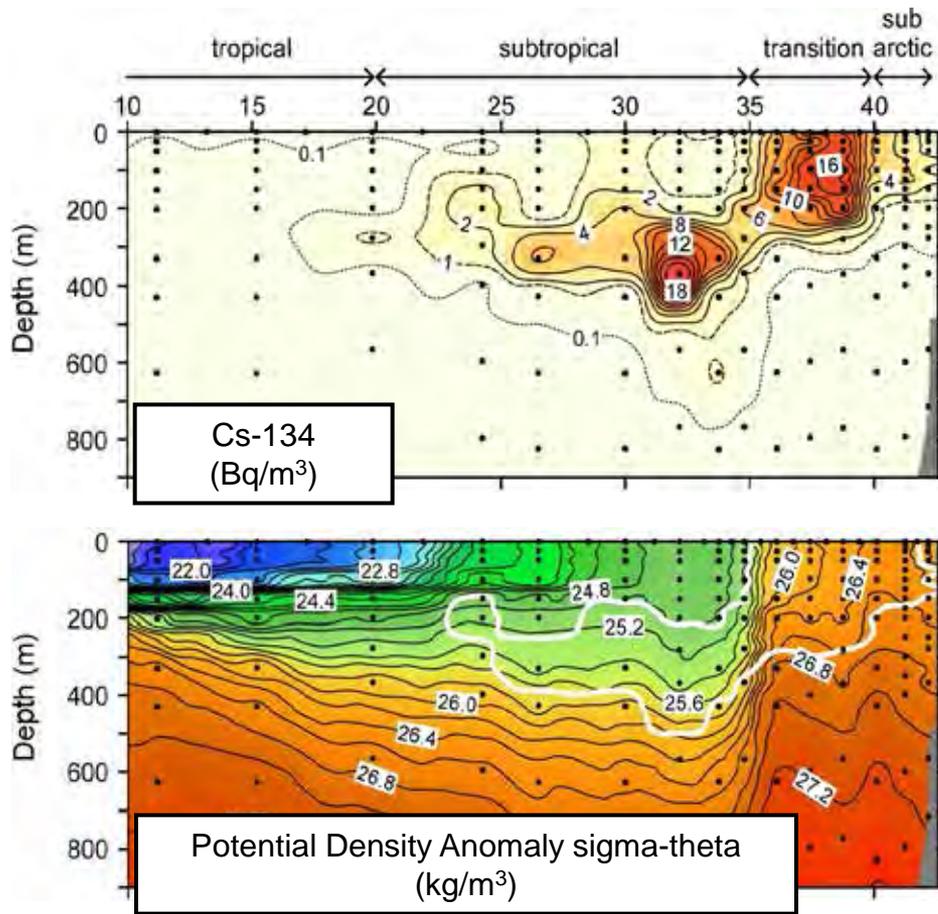
# $^{137}\text{Cs}$ cross section along 165 °E in June 2012

## Latitude vs. density



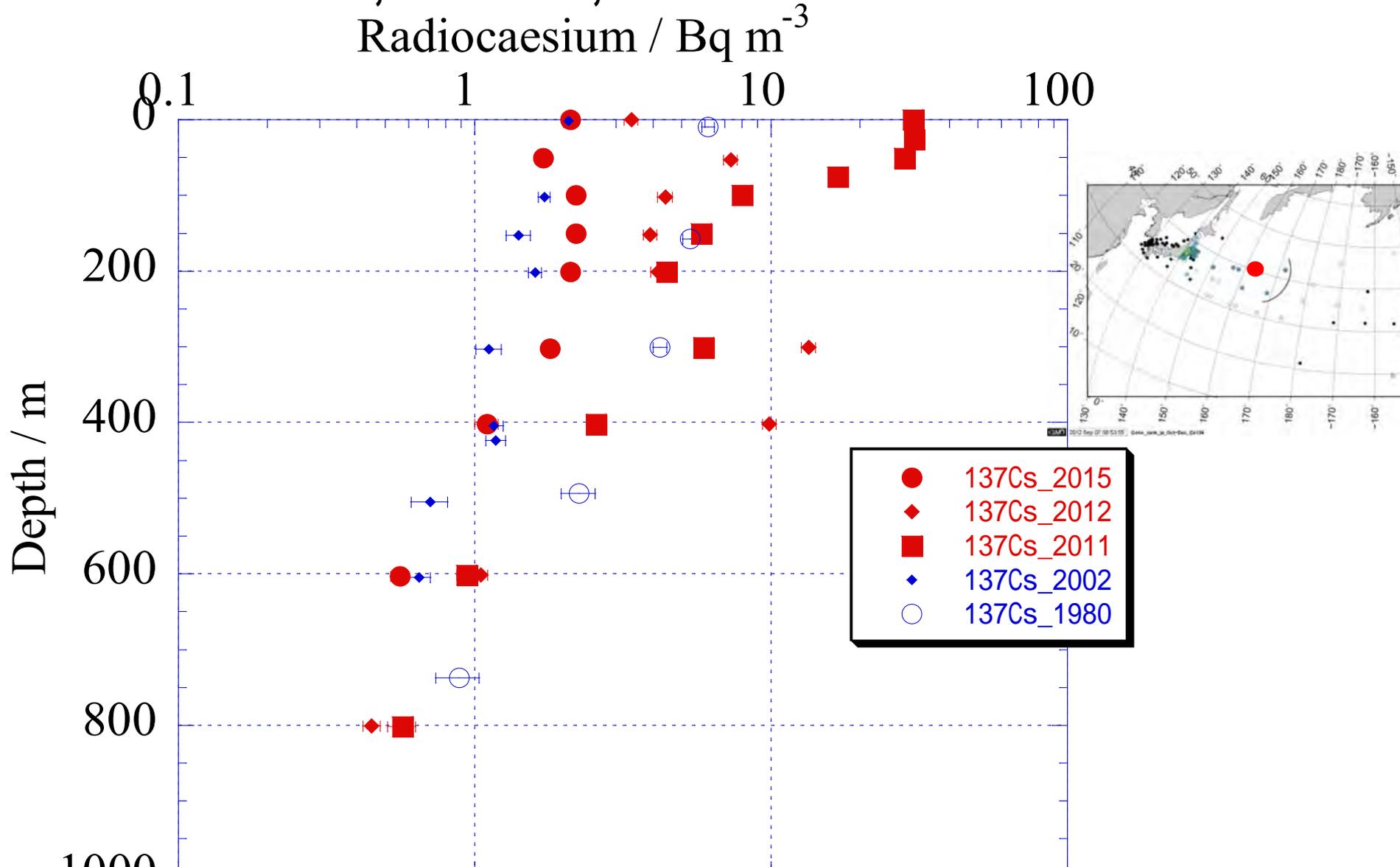
# A $^{137}\text{Cs}$ section along 149 deg. E on Nov.-Dec. 2012

Kumamoto et al. Sci. Rep. (2014)

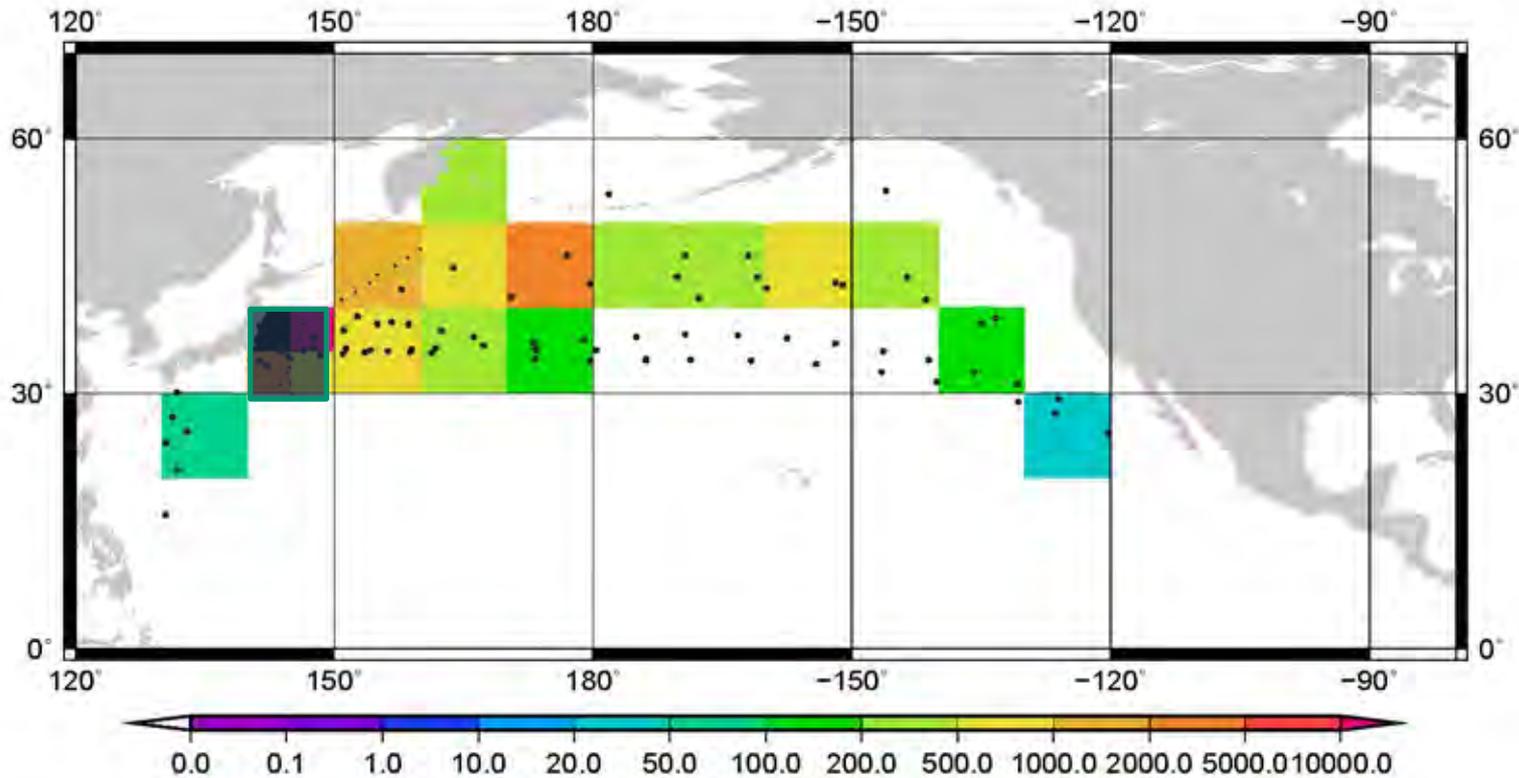


About 6 PBq of  $^{137}\text{Cs}$  were transported south of Kuroshio. (40% of total amount in the sea)

# Vertical profiles of $^{137}\text{Cs}$ in 1980, 2002, 2011, 2012 and 2015

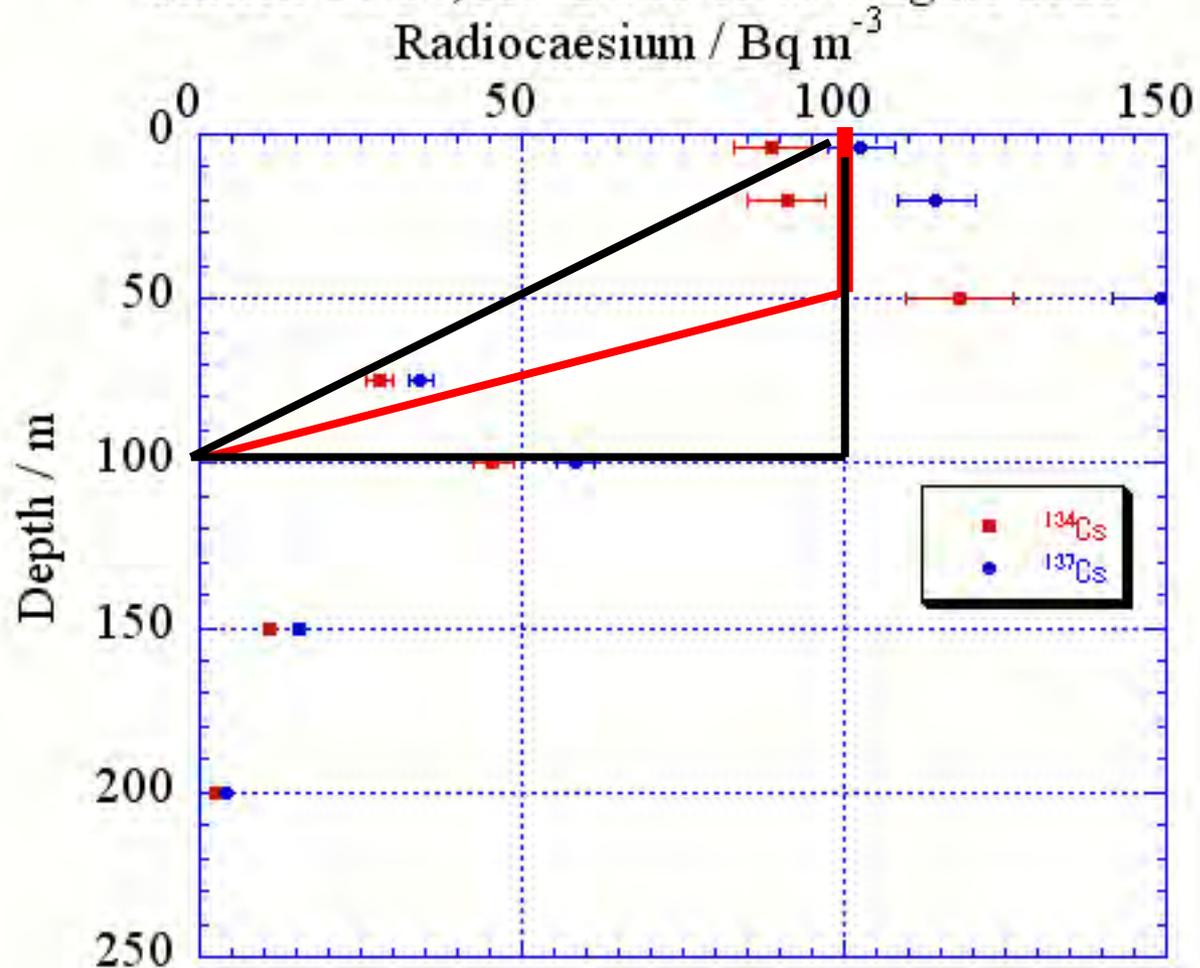


# A simple method by Aoyama et al., JO, 2015a calculate inventory in the ocean



**Fig S10.** 10 deg mesh for integration of radiocaesium in  $\text{Bq m}^{-2}$   
Black dots show observation location.

**Vertical profile of radiocaesium at 38-25N, 142-50E,  
station GJT4, KT-11-21 on 29 August 2011**



Our assumption to estimate inventory

Table S7. Estimation of total amount in the North Pacific Ocean

Longitude	Latitude	Area km <sup>2</sup>	<sup>134</sup> Cs surface Bq m <sup>-3</sup>	Inventory Bq m <sup>-2</sup>	Total amount in a grid PBq
<b>170</b>	<b>40</b>	<b>875218</b>	<b>57</b>	<b>4275</b>	<b>3.7</b>
<b>150</b>	<b>40</b>	<b>758595</b>	<b>25.5</b>	<b>1913</b>	<b>1.5</b>
<b>150</b>	<b>30</b>	<b>1011562</b>	<b>7.4</b>	<b>555</b>	<b>0.6</b>
<b>200</b>	<b>40</b>	<b>875223</b>	<b>7.05</b>	<b>529</b>	<b>0.5</b>
<b>160</b>	<b>40</b>	<b>875211</b>	<b>6.8</b>	<b>510</b>	<b>0.4</b>
<b>210</b>	<b>40</b>	<b>875223</b>	<b>4.4</b>	<b>330</b>	<b>0.3</b>
<b>160</b>	<b>30</b>	<b>1011562</b>	<b>3.4</b>	<b>255</b>	<b>0.3</b>
<b>180</b>	<b>40</b>	<b>875223</b>	<b>3.3</b>	<b>248</b>	<b>0.2</b>
<b>160</b>	<b>50</b>	<b>611243</b>	<b>3</b>	<b>225</b>	<b>0.1</b>
<b>190</b>	<b>40</b>	<b>875223</b>	<b>3</b>	<b>225</b>	<b>0.2</b>
<b>170</b>	<b>30</b>	<b>1011562</b>	<b>1.7</b>	<b>128</b>	<b>0.1</b>
<b>220</b>	<b>30</b>	<b>1011562</b>	<b>1.5</b>	<b>113</b>	<b>0.1</b>
<b>130</b>	<b>20</b>	<b>1116711</b>	<b>0.7</b>	<b>53</b>	<b>0.06</b>
<b>230</b>	<b>20</b>	<b>1116920</b>	<b>0.6</b>	<b>45</b>	<b>0.05</b>
<b>Total</b>					<b>8.1 +-2.8</b>

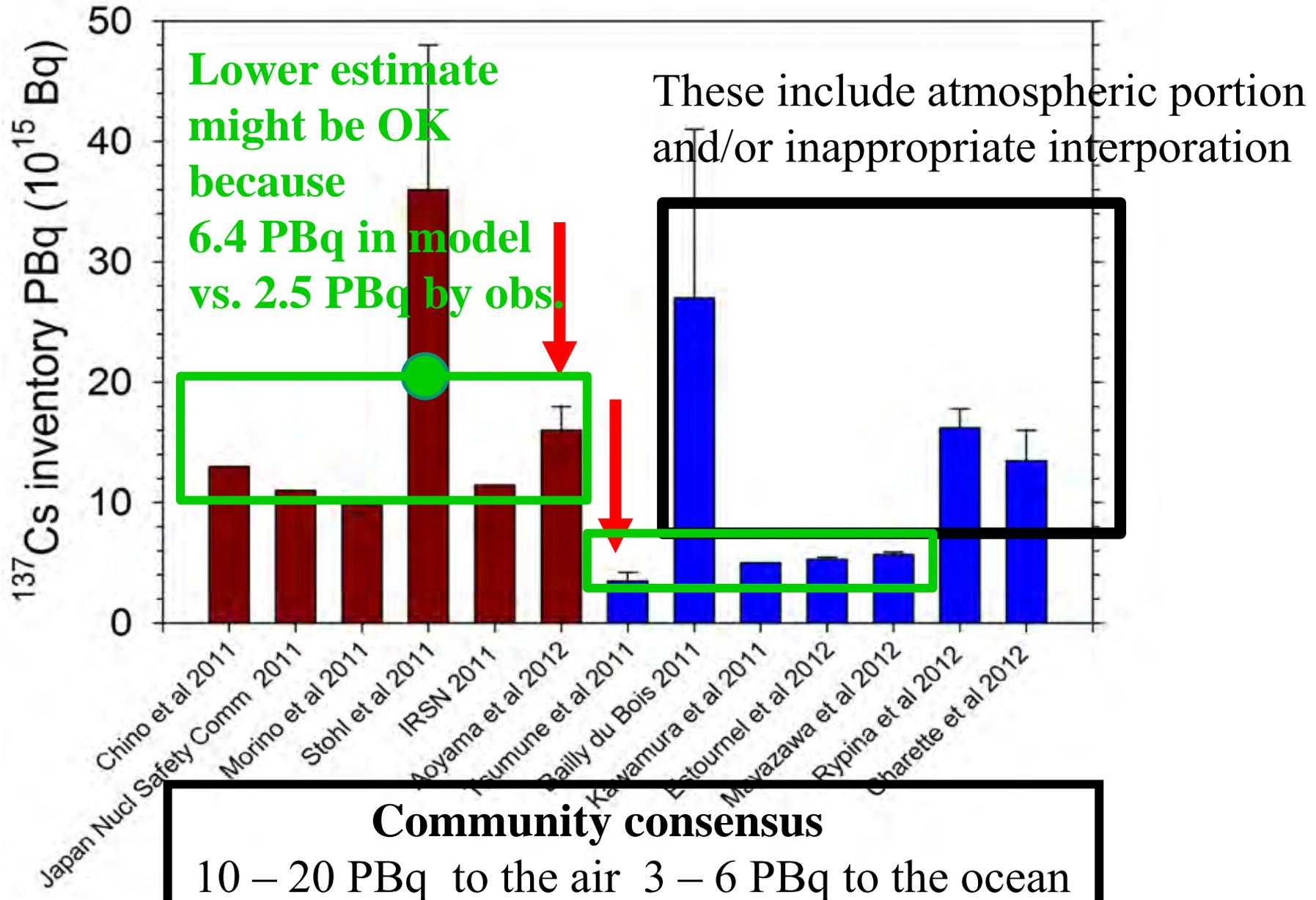
**Table 1. Summary of inventories in the ocean, source terms and land deposit**

Parameters	Symbol	MASIN PBq	MASIN PBq	PM/r PBq	uncerta PBq	remarks
<b>Open Ocean in the North Pacific Ocean</b>						
<b>Observed inventory</b>	<b>a</b>	<b>8.1</b>	<b>8.1</b>	<b>8.1</b>	<b>2.8</b>	<b>This study</b>
<b>Modeled inventory</b>	<b>b</b>	<b>4.7</b>	<b>3.5</b>	<b>3.8</b>		<b>This study</b>
<b>Factor by source term correction</b>	<b>f=a/b</b>	<b>1.7</b>	<b>2.3</b>	<b>2.1</b>		<b>This study</b>
<b>Source term correction</b>						
<b>First guess source term</b>	<b>s</b>	<b>8.8</b>	<b>8.8</b>	<b>8.8</b>		<b>This study</b>
<b>Corrected source term</b>	<b>s*f</b>	<b>15.2</b>	<b>20.4</b>	<b>18.8</b>		<b>This study</b>
<b>North Pacific Ocean</b>						
<b>Total inventory in the model</b>	<b>g</b>	<b>10.3</b>	<b>9.9</b>	<b>9.4</b>		<b>This study</b>
<b>Direct discharge</b>	<b>d</b>	<b>3.5</b>	<b>3.5</b>	<b>3.5</b>	<b>0.7</b>	<b>Tsumune et al. 2011</b>
<b>Atmospheric portion in the model</b>	<b>g-d</b>	<b>6.8</b>	<b>6.4</b>	<b>5.9</b>		<b>This study</b>
<b>Corrected atmospheric portion</b>	<b>(g-d)*f</b>	<b>11.7</b>	<b>14.8</b>	<b>12.6</b>		<b>This study</b>

# Estimates of total releases from Fukushima

To the air

Direct discharge to the Ocean



# Aoyama et al., JO 2015a vs. UNSCEAR 2013

	Aoyama	UNSCEAR
Fukushima direct discharge to the North Pacific Ocean	<b>3.5 +- 0.7</b> PBq 3 – 6 PBq	3 - 6 PBq
Fukushima deposition in the North Pacific Ocean	<b>12 - 15</b> PBq	5 – 8 PBq
Fukushima deposition on land	<b>2.5 - 3</b> PBq	no estimation ( 1 – 12 PBq??)
Total release to the air	<b>14-17</b> PBq 10 – 20 PBq	6 – 20 PBq

Huge inconsistency in UNSCEAR estimation

# Conclusions

- After July 2012, the activities of  $^{137}\text{Cs}$  in surface water at near FNPP1 site were still kept around  $1000 \text{ Bq m}^{-3}$  which corresponds flux of about  $10 \text{ GBq day}^{-1}$ . A zonal speed of FNPP1 derived radiocaesium in surface water at mid latitude in the North Pacific Ocean was  $7 \text{ km day}^{-1}$ ,  $8 \text{ cm s}^{-1}$  until March 2012, however it after March 2012 till August 2014 was ca.  $3 \text{ km day}^{-1}$ ,  $3.5 \text{ cm s}^{-1}$ . Then FNPP1 derived radiocaesium arrive at west coast of US continent as expected.

# Conclusions-2

- Clear increase of  $^{134}\text{Cs}$  due to STMW formation was observed as well as increase of  $^{134}\text{Cs}$  due to CMW formation. These observations indicate that mode waters formation were most effective pathway to introduce Fukushima derived radiocaesium into ocean interior about one year time scale. More than 80 % of released radiocaesium might be in mode waters ( sub surface layers ) and NOT in surface region as expected, too.