North Pacific distribution and budget of radiocesium released by the 2011 Fukushima nuclear

accident

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Objectives

- To reveal temporal and spatial distribution of radiocaesium released from Fukushima in the North Pacific Ocean
- To estimate total amount of radiocaesium released from Fukushima NPP accident including atmospheric release and direct discharge

我々の研究の特徴

北太平洋全域での観測結果から出発し、変動の大き い事故サイト周辺を除く北太平洋で積分した人工放射能 総量を推定し使用した。また大気および海洋の輸送モデ ルも最先端のものを使用している。

外国の研究も含めて先行研究の弱点:複数のモデル や気象条件の解析から判っていることは、放出された人 工放射能の2/3以上は北太平洋に降下したことである。 しかし、大気への放出量を推定しているすべての研究は 陸上(日本および諸外国)での観測値のみに依っている。 従ってそれらの推定の不確かさが必然的に大きくなる。

得られた成果の自己評価:現時点で最も真実に近い 数字を得た。 Temporal and spatial distribution of radiocaesium released from Fukushima in the North Pacific Ocean Sampling summary and other data source

•NYK ships 7 cruises more than 150 samples, 79 samples done, 2 liters surface only until end of May 2011.

•NYK ships 10 cruieses from Aug. 2011-Now

•Tansei-maru, Hakuho-maru and Keifu-maru surface and vertical samples

放射性セシウムの高精度分析と不確かさ

- 収率99%程度で安定した抽出(Aoyama and Hirose 2008)事実上の世界標準法
- 金沢大学尾小屋地下測定室での測定:総量で1mBq以下まで測定可能
- ¹³⁷Csは表面水すべての値を決定:核実験起 源分と整合
- ¹³⁴Csは0.4mBq L⁻¹以下を下限値以下: 福島 事故由来分を正しく評価できた

Table S2. Radiocaesium activity in the North Pacific Ocean in April/May 2011

| Station | Depth | Latitud | e Longitud | e Date | ¹³⁴ Cs | ¹³⁷ Cs | _ | |
|-------------------|---------------|----------------|-----------------|----------------|--------------------------|--------------------------|-----------------|--------------------------------------|
| | dbar | 240503 | 1 10 0 00 | | Bq m ⁻³ | Bq m ⁻³ | ⊢xam | nnle of resul |
| 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 | 14/.60°E | 20110401 | $1000 \pm /1$ | 1080 ± 60 | | |
| NYK11-044 | 0 | 35.0/°N | 146.44°E | 20110401 | 34 ± 2.6 | 36.8 ± 2.1 | | |
| NY <u>K11-00/</u> | 0 | <u>38.18°N</u> | <u>154.9/°E</u> | | 1/./ <u>+</u> 1.6 | 21.5 ± 1.3 | 101 | 105 |
| | ituc | le L | ongiti | ude | Date | - | 134 Cs | 13^{\prime} Cs |
| NY] | | | U | | | D | $\alpha = 3$ | $\mathbf{D} \propto \mathbf{m}^{-3}$ |
| NY] | | | | | | D | pq m ° | <u>БЧШ°</u> |
| NY 36 6 | NoV. | J 1/ | 7 600 | \mathbf{F} 2 | 01104 | N1 1 | 000 471 | 1080 ± 60 |
| NY | | · · · · · · · | | | | | | 1000 ± 00 |
| NYI 26 C |) 5 01 | T 1' | 70 00 | о г (| 0 | 06 1 | $1 \cdot 0 \in$ | 2 4 0 4 |
| NYI 30.3 | 0.5Γ | N I | /0.99 | E 4 | 201104 | 00 1 | 11 ± 0.0 | 2.4 ± 0.4 |
| $\frac{NY}{NY}$ | 7 0N | J 1 | 67 75 | oW 🤇 | 01104 | 07 | ND | 18 ± 0.2 |
| NYL VZI | | | | | | U / | | 1.0 ± 0.2 |
| 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 | Fuksuhima | Fukushima+ bomb |
| 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 | 1340 | s 137Cs activity ratio |
| 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 | of Fu | kushima radioactivitv |
| NYK11-029 | 0 | 38.18°N | 134.97°W | 20110411 | ND | 1.4 ± 0.2 | - | -9 |
| 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 | <u>n aa .</u> | +- 0 02 |
| 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 | Bues | seller et al., 2011 |
| 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 | | |

NYK11 ¹³⁷Cs April 2011



NYK11 ¹³⁷Cs May 2011



NYK11 ¹³⁴Cs April 2011



NYK11 ¹³⁴Cs May 2011



¹³⁴Cs (left) and ¹³⁷Cs(right) in April-May 2011



GMD 2012 Jan 06 01 35 12 NYK_KT_MR_results_Cx137

¹³⁷Cs distribution before Fukushima



Fig. S1. 3-D ¹³⁷Cs in the Pacific Ocean before Fukushima accident in 2011



¹³⁷Cs trend in the subtropical gyre in the western North Pacific Ocean.(Aoyama, M., Hirose, K., *TheScientificWorldJOURNAL*, 4, 200-215, 2004 and update)

Past and present

TEPCO: Tokyo Electric Power Company, TEPCO News Press Releases 2011

http://www.tepco.co.jp/en/press/corp-com/release/

30km offshore: Ministry of Education, Culture, Sports, Science and Technology- Japan (MEXT). Readings of Sea Area Monitoring at Post Out of Fukushima Dai-ichi NPP <u>http://www.mext.go.jp/english/radioactivity_level/detail/13041</u> <u>92.htm</u>

Historical: Aoyama, M., Hirose, K., *TheScientificWorldJOURNAL*, **4**, 200-215, 2004 and update

Fukushima: http://www.pref.fukushima.jp/j/kaisui1115.pdf

Nuclear tests by Nuclear tests by China

1971

1976

1981

former USSR, USA, etc

MEXT:http://radioactivity.mext.go.jp/ja/monitoring_around_Fuku

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shimaNPP_sea
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NYK: This study
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10¹

100

10-1

1955

1986

Chernobyl Accident

1991

1996

2007

2002

2012

3/20

4/29

6/8

7/18

2011

8/27

10/6

11/15



Fig. S3 Region of interest A, B, C and D



Fig S4. ¹³⁷Cs and ¹³⁴Cs activity in region A: eastern North Pacific Ocean







Fig S7. ¹³⁷Cs and ¹³⁴Cs activity in region D: close to the FNPP1 site

Summary of temporal and spatial distribution of

Fukushima radiocaesium

- Fukushima radiocaesium deposited on wide area in the North Pacific Ocean, especially in western part of subarctic gyre and subtropical gyre.
- In the eastern North Pacific Ocean, maximum ¹³⁴Cs activity was ca. 10 Bq m⁻³ in April/May 2011.
- Detected ¹³⁴Cs was Fukushima origin and ¹³⁷Cs was Fukushima+ pre-existing bomb

8 steps to estimate total amount of released radiocaesium from Fukushima NPP accident

We already have direct discharge estimation. $(3.5 \pm 0.7 PBq, Tsumune et al., 2011, presentation 14:45 today)$

Step 1: prepare first guess of source term scenario of atmospheric release. (8.8 PBq)

Step 2: Three atmospheric transport models to simulate daily deposition

Step 3: Pass daily deposition to an ocean model in the North Pacific Ocean.

Step 4: simulate radiocaesium activity in the sea

Step 5: Compare simulated activities and observed activities to evaluate model simulations.

Step 6: Calculate inventory based on the ocean observations $(8.1 \pm 2.8 \text{ PBq})$ and in the models.

Step 7: Compare observed inventory and model simulated inventories to have correction factors of the first guess of the released scenario.(factors were 1.7-2.3)

Stop Q. Evaluate land partiana and accord



Figure 3 Model results vs. observation results comparis

Step 6 calculate inventory in the ocean



Fig S10. 10 deg mesh for integration of radiocaesium in Bq m⁻²

Black dots show observation location.

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

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

Total

8.1 +-2.8



Our assumption to estimate inventory

Summary of budget of 137**Cs**

- Global fallout as of 1970 $290 \pm$ 30PBq^a
- Observed inventory as of 2002 85 PBq b
- Estimated inventory as of 2011 69 PBq b
- Fukushima direct discharge to the North **Pacific Ocean** 3.5 ± 0.7 PBq ^c
- Fukushima atmospheric release 15 - 20 PBq^d
- Fukushima deposition on land 3.8 - 6.6PBq^d

a: Aoyama, M., Hirose, K., Igarashi, Y., J. ENVIRON. MONITOR., **8**,431-438, 2006 b: Aoyama unpublished data estimeted 3-D costribution of ¹⁵⁷Cs **19** – **24**

- c: **Tsum** e^d et al., 2011,
- d: this study

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Estimates of total releases from Fukushima



Conclusions

1. Temporal and spatial distributions of radiocaesium from Fukushima were clearly observed in the North Pacific Ocean and coupled atmospheric/ocean models simulate the activity of radiocaesium well.

2. Total amount of the atmospheric deposition in the North Pacific Ocean from Fukushima was 12 - 15 PBq for both ¹³⁴Cs and ¹³⁷Cs. Newly added 12 - 15 PBq by atmospheric deposition and 3.5 ± 0.7 PBq of direct discharge made the 22-27 % increase of total ¹³⁷Cs inventory in the North Pacific Ocean.

3. Our study also gives us that total amount of released radiocaesium from the Fukushima NPP1 accident to the air was about 15 - 20 PBq for both ¹³⁴Cs and ¹³⁷Cs. Total deposition on land might be around 3.4 - 6.2 PBq which corresponds to 23 -33% of total amount to the air.

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possible pathway of radiocaesium



Fig. S2 Possible pathway of ¹³⁷Cs in the Pacific Ocean