

Mass balance of radiocaesium derived from Fukushima accident and estimation of latest fluxes among atmosphere, land and ocean

Michio Aoyama¹, Daisuke Tsumune², Yayoi Inomata³, Yutaka Tateda²

¹ Faculty of Life and Environmental Sciences, Univ. of Tsukuba, 305-8577, Japan

² Environmental Science Research Laboratory, Central Research Institute of Electric Power Industry, 1646 Abiko, 270-1194, Japan

³ Institute of Nature and Environmental Technology, Kanazawa University, Kanazawa, 920-1192, Japan

EGU2020-12301

8 May 2020, EGU2020 online



Summary

In this presentation, we summarize mass balance of the Fukushima-derived ^{137}Cs released to the atmosphere and ocean prior to 2018 as well as the ^{137}Cs inventories on land and in the ocean, biota, and sediment. We propose that the consensus value of the total amount of ^{137}Cs released to the atmosphere was 15–21 PBq; atmospheric deposition of ^{137}Cs on land was 3–6 PBq; atmospheric deposition of ^{137}Cs on the North Pacific was 12–15 PBq; and direct discharge of ^{137}Cs to the ocean was 3–6 PBq. We also evaluated the movement of ^{137}Cs from one domain to another for several years after the accident. We calculated that the amount of ^{137}Cs transported by rivers might be 40 TBq. The annual deposition of ^{137}Cs due to resuspension at Okuma during the period 2014–2018 was 4–10 TBq year⁻¹. The ^{137}Cs discharged to the ocean was 0.73–1.0 TBq year⁻¹ in 2016–2018. The integrated amount of FNPP1-derived ^{137}Cs that entered the Sea of Japan from the Pacific Ocean from 2011 until 2017 was 0.27 ± 0.02 PBq, 6.4 % of the estimated amount of FNPP1-derived ^{137}Cs in Subtropical Mode Water in the North Pacific. The integrated amount of FNPP1-derived ^{137}Cs that returned to the North Pacific Ocean through the Tsugaru Strait from the Sea of Japan was 0.11 ± 0.01 PBq. Decontamination efforts removed 0.134 PBq of ^{137}Cs from surface soil prior to February 2019, an amount that corresponded to 4 % of the ^{137}Cs deposited on land in Japan.

Presentation outline

- Why is mass balance important?
- Mass balance in the environment of radioactivity originating from the Fukushima accident
- Amount and initial distribution of artificial radioactive materials released by the Fukushima accident
- Behavior in the environment: Estimation of the amount of movement after being released to land and ocean (land to land, land to sea, one sea area to another sea area, etc.)

Why is mass balance important?

- The conservation of mass is the basic law.
- Somewhere in the reports and discussions that are out of balance.
- If you don't see the whole picture, you won't know the truth.
- There is an example of not integrating the integrated region in order to obtain the quantity when reporting the quantity, or integrating it by covering only part of it with actual constraints. The same was true at the time of the Fukushima accident.

Mass balance : Fukushima accident

$$\sum R_i = \sum I_j$$

the law of conservation of mass

Where R_i are released amount to each domain and I_j are inventory in each domain.

i : 1=atmosphere, 2=direct discharge

j : 1=atmosphere (zero), 2= land, 3=ocean, 4=sediment, 5= biota

$$R_1 + R_2 = I_1 + I_2 + I_3 + I_4 + I_5 = \text{total in the core}$$

$$R_1 - I_2 = I_3 - R_2$$

Need to look at whole structure



It is a story of a group of blind men (or **men in the dark**) who touch an elephant to learn what it is like. Each one feels a different part, but only one part, such as the side or the tusk.

They then compare notes and learn that they are in complete disagreement

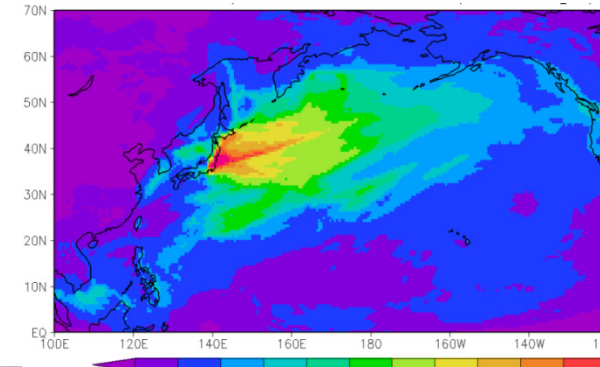
^{137}Cs (^{134}Cs) mass balance at the beginning

15–20 PBq to the atmosphere

Aoyama et al., 2016

3-6 PBq to the land

Aoyama et al., 2016



12 - 15 PBq to the ocean

Aoyama et al., 2016

Total in the Ocean: 15-18 PBq

Aoyama et al., 2016, Tsubono et al., 2016, Inomata et al., 2016

3.6 ± 0.7 PBq to the ocean

(Tsumune et al., 2013)

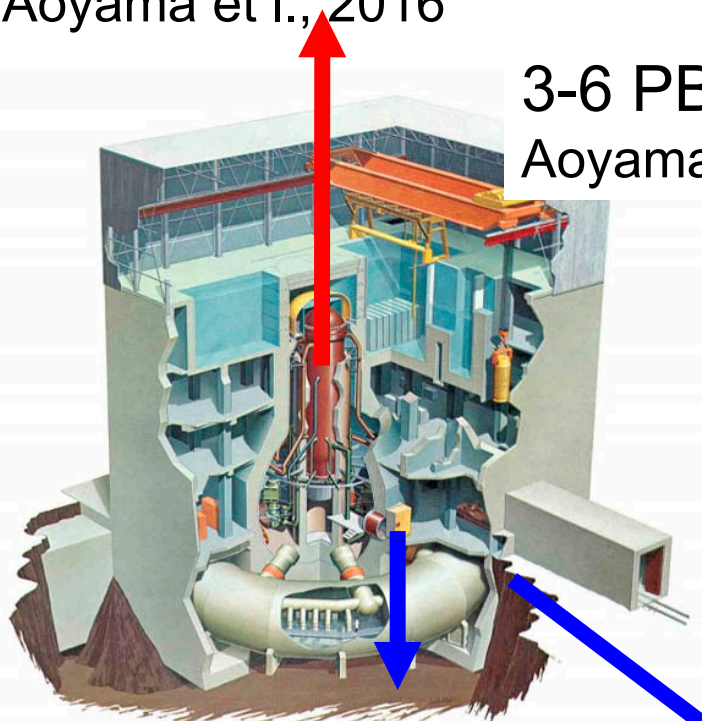
230 PBq recovered

TEPCO unpublished data

140 PBq in stagnant water

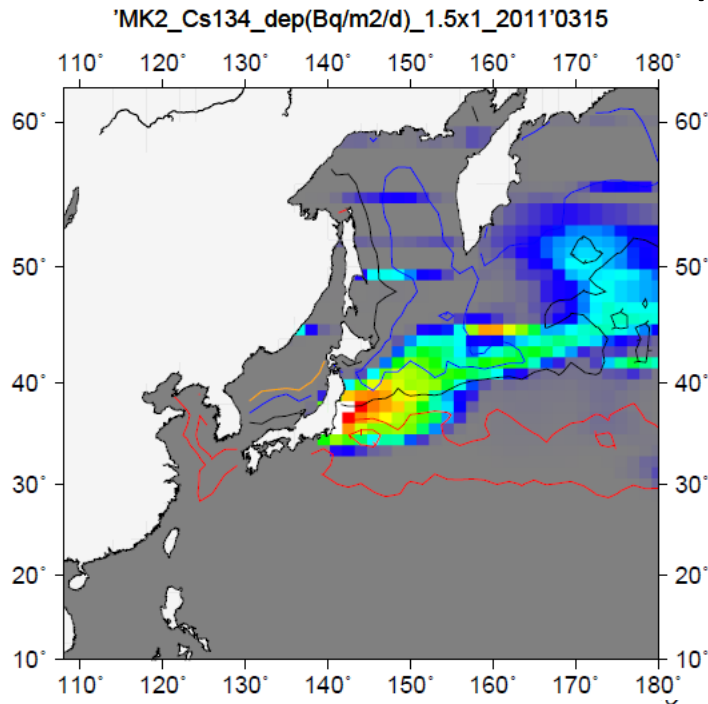
700 PBq was in the three core

(Nishihara et al., 2011)

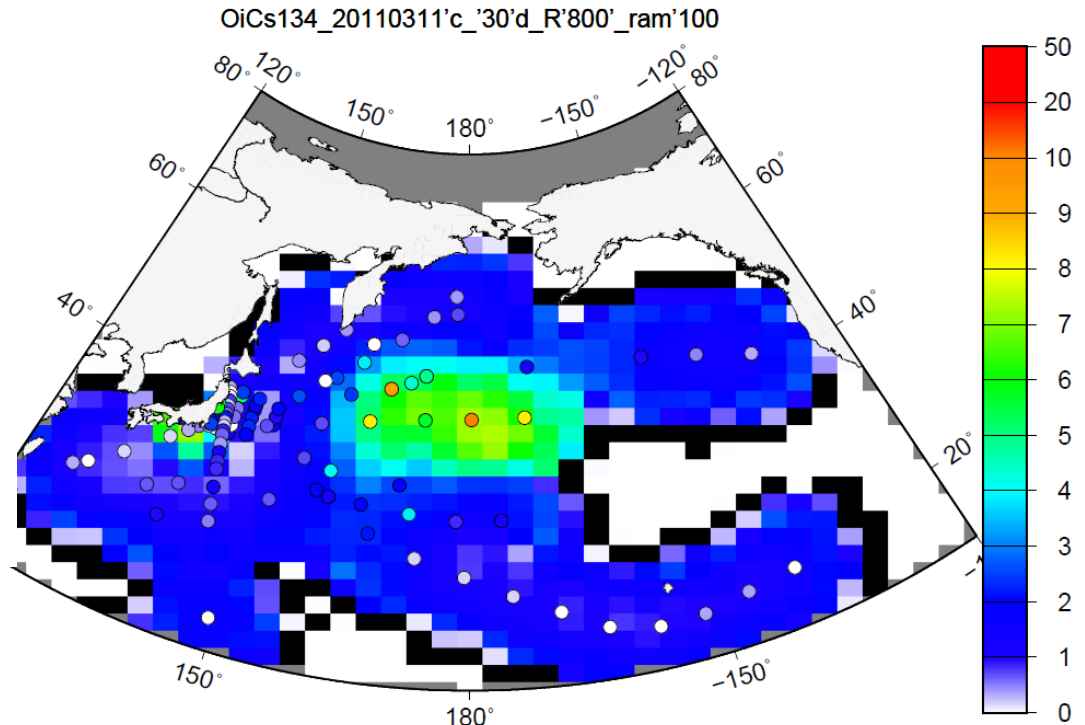


Mass balance in the ocean

Inomata et al., 2018



Estimate of ^{134}Cs deposition in STMW, LCMW, DCMW, TRMW in the western North Pacific Ocean.



Distribution of ^{134}Cs in Oct.-Dec. 2012.
Decay corrected to 11 Mar. 2011.

Estimated inventory in surface seawater
OICs134; 7.9 ± 1.4 PBq
OICs137; 13 ± 0.93 PBq

Mass balance calculations in the ocean interior

$$\Sigma R_i = \Sigma I_j \quad (1)$$

$$R_1 + R_2 = I_1 + I_2 + I_3 + I_4 + I_5 \quad (2)$$

R_1 : Atmospheric deposition

(11.7-14.8 PBq; Aoyama et al., 2016b)

R_2 : Direct discharge

(3.5 ± 0.7 PBq; Tsumune et al., 2013)

$R_1 + R_2$: 15.2 to 18.3 PBq

15.3 ± 2.6 PBq (Inomata et al., 2016) by OI

$$I_3 = (R_1 + R_2) - I_1 - I_2 \quad (3)$$

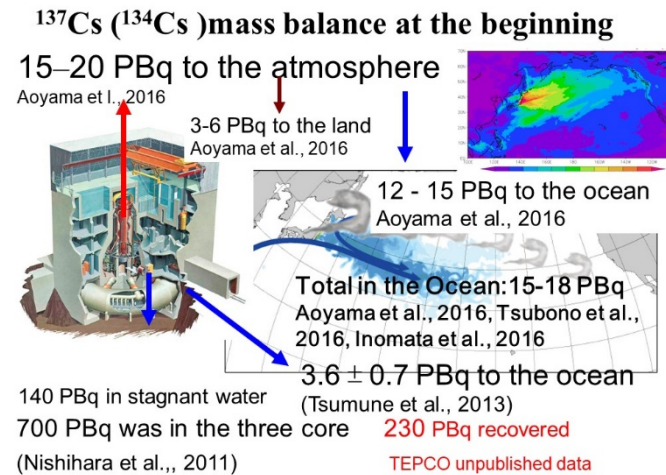
I_1 Surface water inventory 7.9 ± 1.4 PBq

I_2 STMW 4.2 ± 1.1 PBq (Kaeriyama et al., 2016)

I_3 CMW 2.5 ± 0.9 PBq (Inomata et al., 2018)

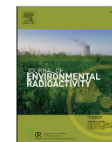
I_4 Biota negligible 200 GBq (Aoyama et al., 2019)

I_5 Sediment negligible 130 ± 60 TBq (Kusakabe et al., 2014)



Issues we should know during 7 years after the accident

- 1) Relatively higher deposition, due to flux from forest?
- 2) Continuous release from the site and other sources, eg. river water and resuspension.
- 3) Transport from STMW in the North pacific to Sea of Japan
- 4) Transport in the surface layer and into ocean interior
- 5) Results of de-contamination work



Mass balance and latest fluxes of radiocesium derived from the Fukushima accident in the western North Pacific Ocean and coastal regions of Japan

Michio Aoyama^{a,*}, Daisuke Tsumune^b, Yayoi Inomata^c, Yutaka Tateda^b

^a Center for Research in Isotopes and Environmental Dynamics, Univ. of Tsukuba, Tsukuba, Japan

^b Environmental Science Research Laboratory, Central Research Institute of Electric Power Industry, Chiba, Japan

^c Institute of Nature and Environmental Technology, Kanazawa University, Ishikawa, Japan

ARTICLE INFO

Keywords:

Fukushima accident
Mass balance
Radiocesium
¹³⁷Cs
Inventory
Flux

ABSTRACT

This article summarizes and discusses mass balance calculations of the activities of Fukushima-derived ¹³⁷Cs released to the atmosphere and ocean prior to 2018 as well as the ¹³⁷Cs inventories on land and in the ocean, biota, and sediment. We propose that the consensus value of the total amount of ¹³⁷Cs released to the atmosphere was 15–21 PBq; atmospheric deposition of ¹³⁷Cs on land was 3–6 PBq; atmospheric deposition of ¹³⁷Cs on the North Pacific was 12–15 PBq; and direct discharge of ¹³⁷Cs to the ocean was 3–6 PBq. We also evaluated the movement of ¹³⁷Cs from one domain to another for several years after the accident. We calculated that the amount of ¹³⁷Cs transported by rivers might be 40 TBq. The annual deposition of ¹³⁷Cs due to resuspension at Okuma during the period 2014–2018 was 4–10 TBq year⁻¹. The ¹³⁷Cs discharged to the ocean was 0.73–1.0 TBq year⁻¹ in 2016–2018. The integrated amount of FNPP1-derived ¹³⁷Cs that entered the Sea of Japan from the Pacific Ocean from 2011 until 2017 was 270 ± 20 TBq, 6.4% of the estimated amount of FNPP1-derived ¹³⁷Cs in Subtropical Mode Water in the North Pacific. The integrated amount of FNPP1-derived ¹³⁷Cs that returned to the North Pacific Ocean through the Tsugaru Strait from the Sea of Japan was 110 ± 10 TBq. Decontamination efforts removed 134 TBq of ¹³⁷Cs from surface soil prior to February 2019, an amount that corresponded to 4% of the ¹³⁷Cs deposited on land in Japan.

1. Introduction

The total amount of radionuclides released to the environment from the Fukushima Dai-ichi Nuclear Power Plant, hereafter FNPP1, as a result of the accident in March 2011 as well as the impact of those radionuclides on biota, and especially humans, have been among the major concerns related to the FNPP1 accident. The radionuclide of principal concern with respect to human health has been radiocesium, and it is thus particularly important to know how much radiocesium was released to the environment. Many articles and several review articles have already been published concerning this issue (Buessler et al., 2017; IAEA, 2015; Mathieu et al., 2018; Smith, 2014), but there has been no discussion based on mass balances between the atmosphere, land, and ocean. It is important to consider mass balances in discussions of the total amount of radionuclides released to the environment because the law of conservation of mass is a basic principle, and mass balance is one of the strongest constraints on estimates of the total amount of radionuclides released to the environment and to inventories

in the air, on the land, and in the North Pacific Ocean.

In this paper, we have summarized the results of studies of the amounts of Fukushima-derived ¹³⁷Cs that were released to the atmosphere and ocean as well as estimates of the ¹³⁷Cs inventories on land, in the ocean, in biota, and in sediments. We propose consensus values of these inventories based on mass balance considerations. Finally, we discuss the fluxes of ¹³⁷Cs between domains for several years after the FNPP1 accident. We consider in particular fluxes from the land to the ocean via rivers, releases from the accident site to the ocean, and delayed effects of the accident associated with resuspension from the land to the atmosphere, deposition from the atmosphere onto the land and ocean, and transport of FNPP1-derived ¹³⁷Cs from the North Pacific Ocean to the Sea of Japan. Finally, we consider the total amount of ¹³⁷Cs in surface soil removed by human activity as a part of decontamination work.

* Corresponding author.

E-mail address: michio.aoyama@ied.tsukuba.ac.jp (M. Aoyama).

<https://doi.org/10.1016/j.jenvrad.2020.106206>

Received 10 October 2019; Received in revised form 6 February 2020; Accepted 12 February 2020

Available online 21 February 2020

0265-931X/© 2020 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

Movement during 7 years after the accident

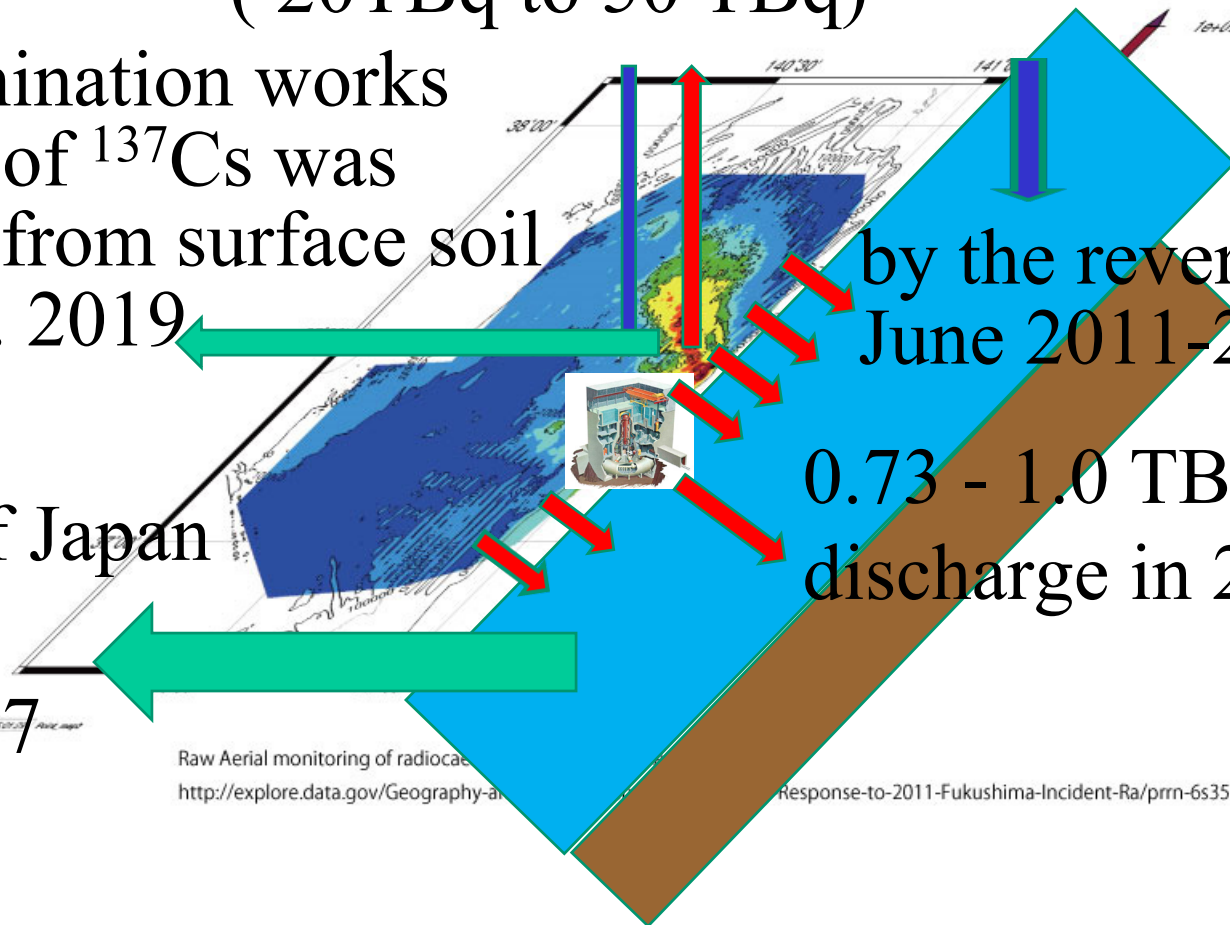
4 TBq year⁻¹ to 10 TBq year⁻¹ flux
as resuspension and fallout 2014-2018
(20TBq to 50 TBq)

decontamination works
134 TBq of ^{137}Cs was
removed from surface soil
until Feb. 2019

by the revers 40 TBq
June 2011-2016

To Sea of Japan
270 TBq
Until 2017

0.73 - 1.0 TBq year⁻¹
discharge in 2016-2018

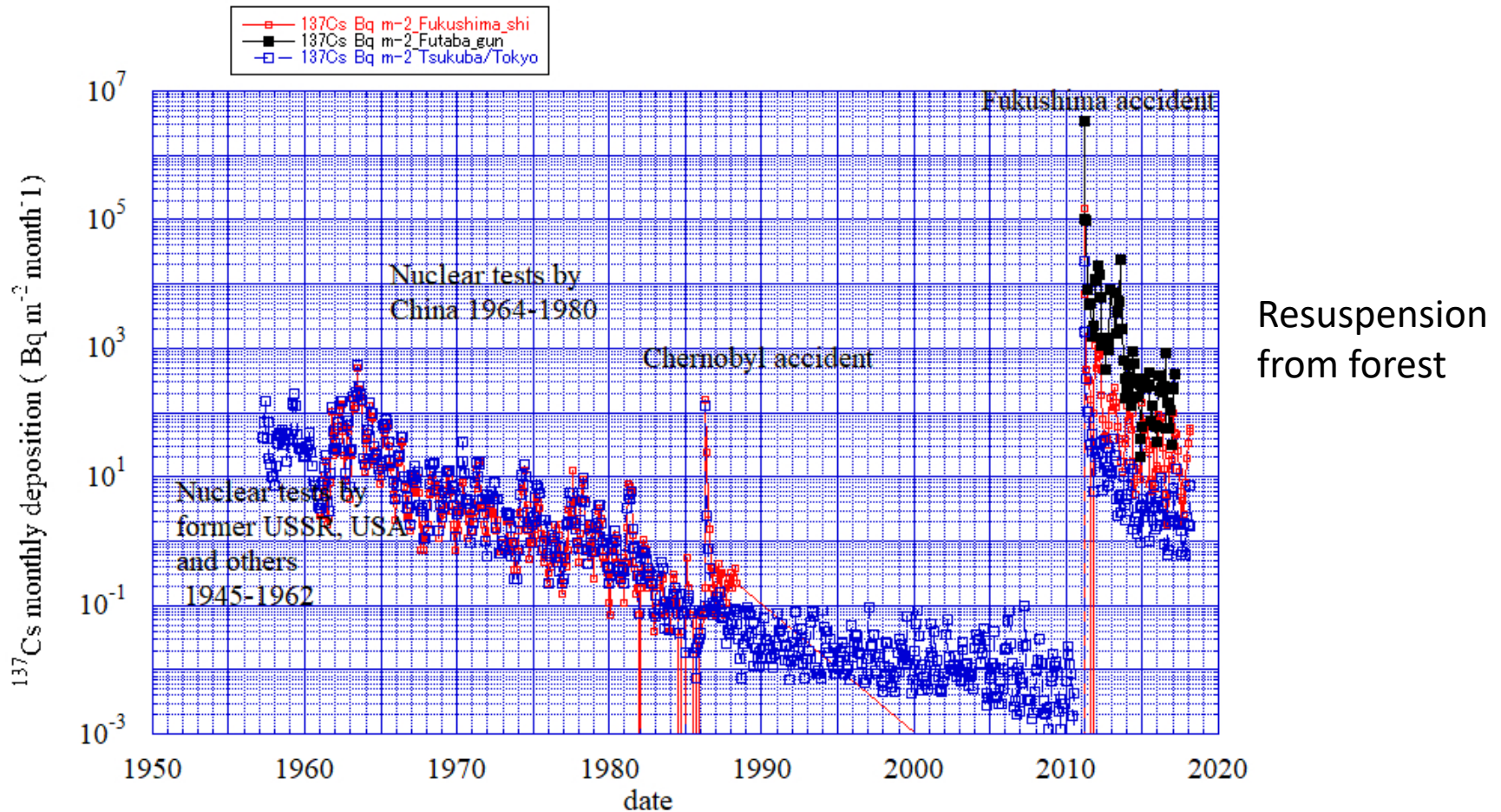


Details are in Aoyama et al., 2020, JER, 10.1016/j.jenvrad.2020.106206

resuspension from forest

Observed monthly deposition of ^{137}Cs at

Tokyo/Tsukuba, Fukushima-Shi and Futaba-Gun

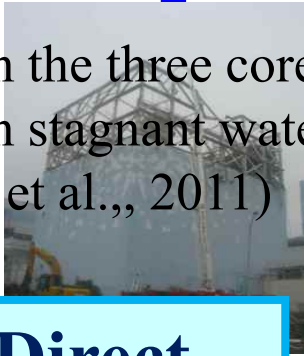


Aoyama, 2019, Kagaku July 2019, Iwanami, in Japanese

Mass balance of FNPP1 derived radiocaesium

Atmospheric input 15.2 – 20.4 PBq

700 PBq in the three core
140 PBq in stagnant water
(Nishihara et al., 2011)



Atmospheric deposition

Total 11.7 – 14.8 PBq (Aoyama et al., JO, 2016)

Land/3.4-6.2 PBq
(Aoyama et al., JO, 2016)

Ocean/Direct release

3.5 ± 0.7 PBq
(Tsumune et al., 2012)

North Pacific Ocean ^{134}Cs

15.2-18.3 PBq (Aoyama et al., JO, 2016): Total
 15.3 ± 2.6 (Inomata et al., 2016) **16.1 ± 1.4** (Tsubono et al., 2016)
(STMW 2.2-4.9 PBq, CMW 7.5-9.3 PBq by the models)

SOJ

6.4%

0.27 ± 0.02 PBq

(42%)

Re-NPO

0.11 ± 0.01 PBq **2.7%**

STMW

**4.2 ± 1.1
PBq**

(Kaeriyama et al. 2016)

Surface

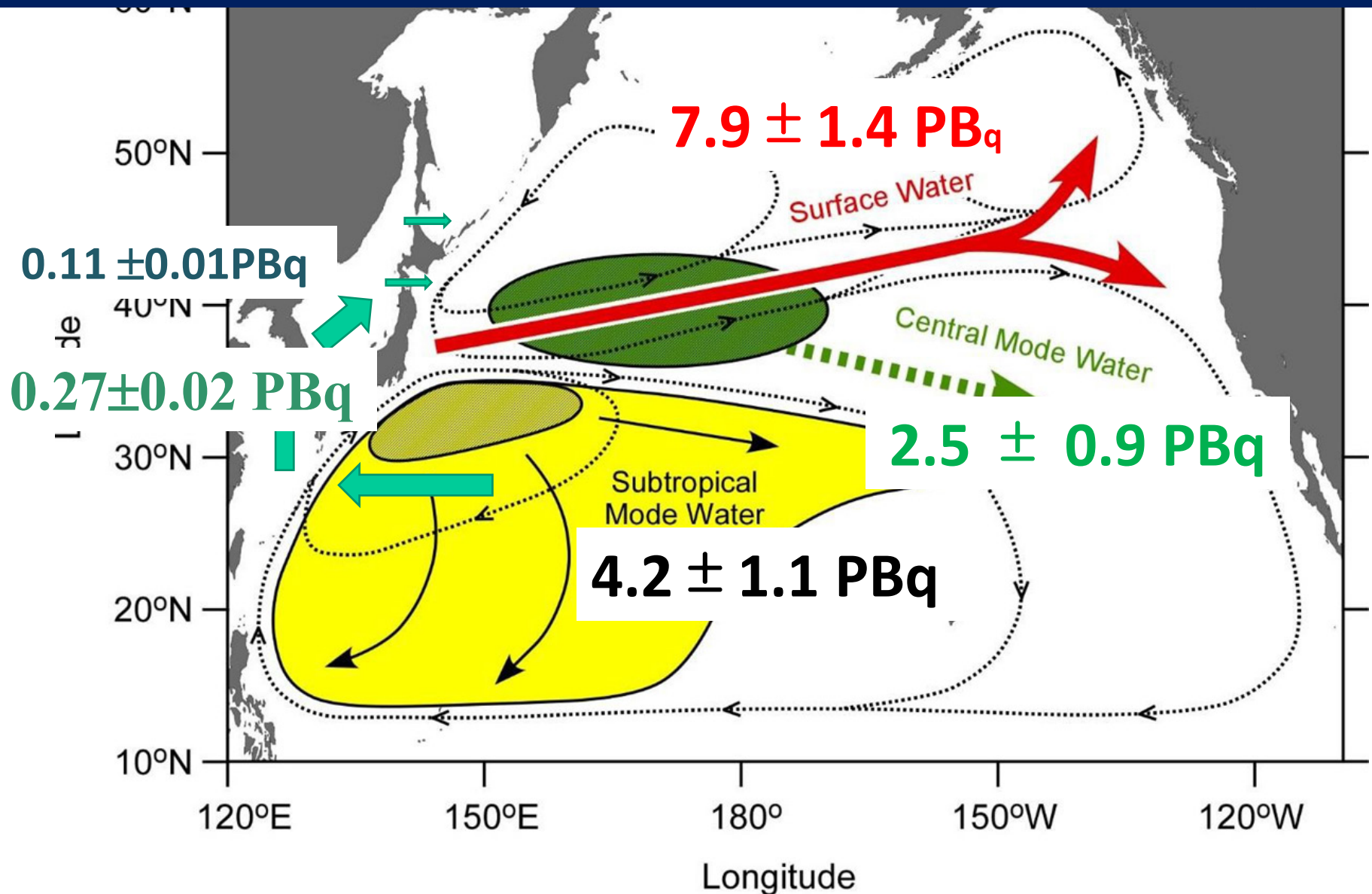
**7.9 ± 1.4
PBq**

Inomata et al., JRNC, 2018b, ENVIRA2019

CMW

**2.5 ± 0.9
PBq**

Schematic diagram of transport/inventory of radiocaesium



Conclusions 1

We propose that the consensus amount of total atmospheric release of ^{137}Cs was 15–21 PBq. The fallout of ^{137}Cs from the atmosphere was 3–6 PBq onto the land and 12–15 PBq onto the North Pacific. The direct discharge of ^{137}Cs to the ocean was 3–6 PBq. The total inventory of ^{137}Cs in the North Pacific was 15–18 PBq. We also estimated the inventory of ^{137}Cs in the surface layer and CMW to be 7.9 ± 1.4 and 2.5 ± 0.9 PBq, respectively.

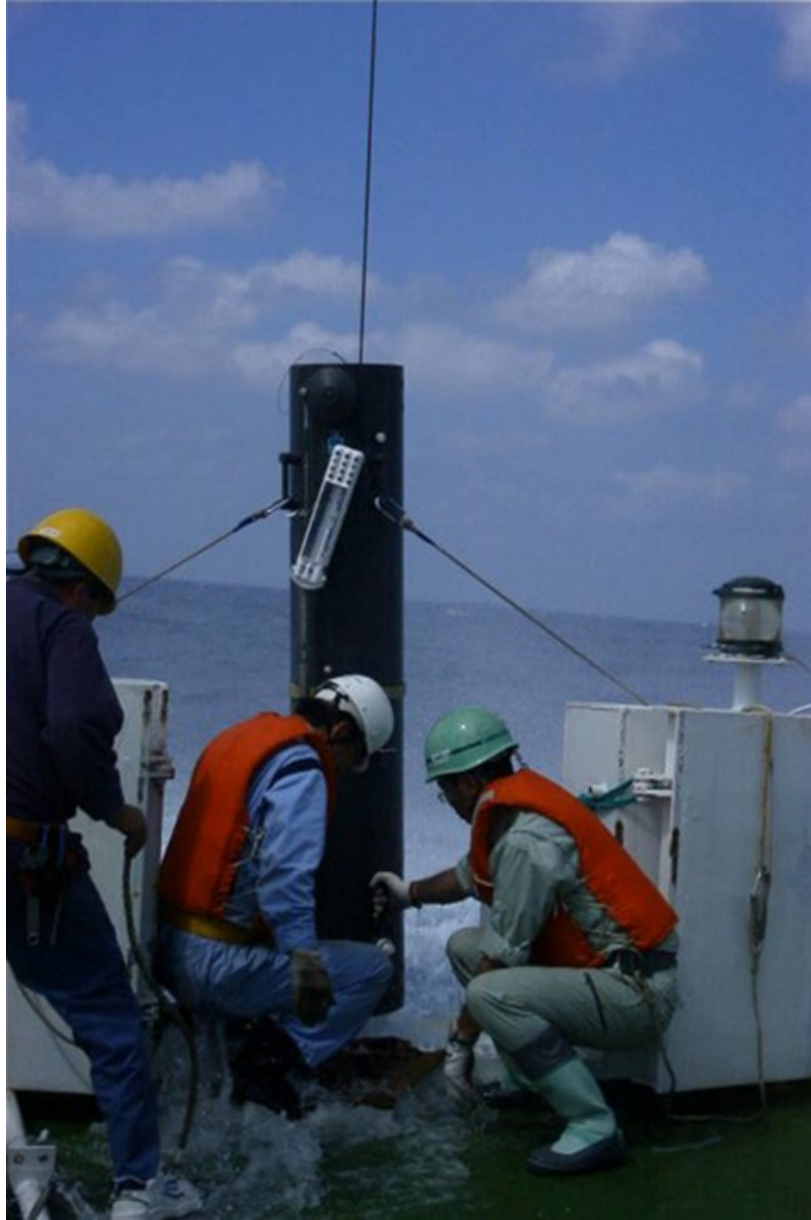
Conclusions 2

The amount of ^{137}Cs transported by rivers from land to the ocean for several years after the FNPP1 accident might be 0.04 PBq, which corresponds to 1.3 % of the ^{137}Cs deposited in the Fukushima region of Japan. The annual deposition of ^{137}Cs at Okuma during the period 2014–2018 means that 4–10 TBq year⁻¹ was resuspended from the land to the atmosphere, an amount that corresponds to about 0.1–0.3 % of the total amount of ^{137}Cs deposited on land in Japan.

Conclusions 3

The ^{137}Cs activity at the 56N canal in 2016–2018 corresponded to $0.73\text{--}1.0\text{ TBq year}^{-1}$ of ^{137}Cs discharged to open water from the FNPP1 site. The integrated amount of FNPP1-derived ^{137}Cs that entered the SOJ from the North Pacific Ocean until 2017 was estimated to be $0.270 \pm 0.002\text{ PBq}$, whereas $0.11 \pm 0.01\text{ PBq}$ returned to the North Pacific Ocean through the Tsugaru Strait.

Decontamination efforts were estimated to have removed 0.134 PBq of ^{137}Cs from surface soil, an amount that corresponds to 4.5% of ^{137}Cs deposited on land in Japan prior to February 2019.



Thank you for your attention!!