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

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Summary

In this presentation, we summarize the 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 $0.27 \pm 0.02$ 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 $0.11 \pm 0.01$ 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

\[ \Sigma R_i = \Sigma I_j \]

The law of conservation of mass

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

\( i: 1=\text{atmosphere}, 2=\text{direct discharge} \)

\( J: 1=\text{atmosphere (zero)}, 2=\text{land}, 3=\text{ocean}, 4=\text{sediment}, 5=\text{biota} \)

\( R_1+R_2=I_1+I_2+I_3+I_4+I_5=\text{<total in the core}> \)

\( R_1-I_2=I_3-R_2 \)
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)

140 PBq in stagnant water

700 PBq was in the three core

(Nishihara et al., 2011)

230 PBq recovered

TEPCO unpublished data
Estimate of $^{134}\text{Cs}$ deposition in STMW, LCMW, DCMW, TRMW in the western North Pacific Ocean.


Estimated inventory in surface seawater OICs$^{134}$; $7.9 \pm 1.4$ PBq
OICs$^{137}$; $13 \pm 0.93$ PBq
Mass balance calculations in the ocean interior

$$\Sigma R_i = \Sigma I_j$$  \hspace{1cm} (1)

$$R_1 + R_2 = I_1 + I_2 + I_3 + I_4 + I_5$$  \hspace{1cm} (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

$$I_3 = (R_1 + R_2) - I_1 - I_2$$  \hspace{1cm} (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 radionuclides derived from the fukushima accident in the western North Pacific Ocean and coastal regions of Japan

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ABSTRACT

This article summarizes and discusses mass balance calculations of the activities of Fukushima-derived 137Cs released to the atmosphere and oceans prior to 2018 as well as the 137Cs inventories on land and in the ocean, biota, and sediments. We estimated that the maximum 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 60 TBr. The annual deposition of 137Cs due to resuspension at Okuma during the period 2014–2018 was 4.1 TBr y−1. The 137Cs discharged to the ocean was 0.2 TBr y−1 in 2015–2016; 54 TBr y−1 in 2017; and 270 TBr y−1 in 2018–2019. The integrated amount of FNPP1-derived 137Cs that entered the Sea of Japan from the Pacific Ocean from 2011 until 2017 was 270 ± 20 TBr, 6.9% 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 TBr. Decontamination efforts removed 154 TBr 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 Daiichi 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 radionuclides of principal concern with respect to human health has been radium-226, and it is thus particularly important to know how much radionuclide was released to the environment. Many articles and several review articles have already been published concerning this issue (Iwadare et al., 2017; IAEA, 2015; Mathieu et al., 2018; Smith, 2014), but there has been no discussion based on mass balances between the atmosphere, the ocean, 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 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.

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

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 $^{137}\text{Cs}$ 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)

**North Pacific Ocean** $^{134}$Cs
15.2-18.3 PBq
(Aoyama et al., JO, 2016)

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)

**Ocean/Direct release**
3.5 ± 0.7 PBq
(Tsumune et al., 2012)

SOJ
0.27±0.02 PBq
(42%)

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

Surface
7.9 ± 1.4 PBq

CMW
2.5 ± 0.9 PBq

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

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

**Inomata et al., JRNC, 2018b, ENVIRA2019**
Schematic diagram of transport/inventory of radiocaesium

- 7.9 ± 1.4 PBq
- 4.2 ± 1.1 PBq
- 2.5 ± 0.9 PBq
- 0.27 ± 0.02 PBq
- 0.11 ± 0.01 PBq
Conclusions 1

We propose that the consensus amount of total atmospheric release of 137Cs was 15–21 PBq. The fallout of 137Cs from the atmosphere was 3–6 PBq onto the land and 12–15 PBq onto the North Pacific. The direct discharge of 137Cs to the ocean was 3–6 PBq. The total inventory of 137Cs in the North Pacific was 15–18 PBq. We also estimated the inventory of 137Cs in the surface layer and CMW to be $7.9 \pm 1.4$ and $2.5 \pm 0.9$ 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 137Cs activity at the 56N canal in 2016–2018 corresponded to 0.73–1.0 TBq year−1 of 137Cs discharged to open water from the FNPP1 site. The integrated amount of FNPP1-derived 137Cs that entered the SOJ from the North Pacific Ocean until 2017 was estimated to be $0.270 \pm 0.002$ PBq, whereas $0.11 \pm 0.01$ PBq returned to the North Pacific Ocean through the Tsugaru Strait. Decontamination efforts were estimated to have removed 0.134 PBq of 137Cs from surface soil, an amount that corresponds to 4.5 % of 137Cs deposited on land in Japan prior to February 2019.
Thank you for your attention!!