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Long term behavior of tritium activity concentration in coastal regions of Fukushima and the North Pacific Ocean since 1970s

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Current status of Fukushima Daiichi decommissioning project, treated water, and future collaboration among RP experts

22 June 2021 online JHPS and KARP

Objectives of this talk

- Present the long-term history of 137Cs and 3H in the marine environment to understand their behavior.
- Present the Fukushima accident impact in the marine environment based on observations and model simulations.
- Estimate the impacts of planned tritium release based on simple proportional calculation and published results of model simulation.

Contents of this talk

- Major long-lived artificial/natural radionuclides in the marine environment
- Long term behavior of 137Cs and tritium in the marine environment at the coastal region of Japan, the Sea of Japan, and the Pacific Ocean
- A few model simulations of 137Cs and tritium
- Simple proportional calculation of the possible impact of 3H release based on 137Cs observations in case of FNPP1 accident
- summary

Table 1 List of major long lived artificial/natural radionuclides in marine environment

Isotope	Fission yield (%)a	$T_{1/2}$ (yr)	Radiation	Sources (ranked by importance)
^{3}H		12.32	β	Natural, Nuclear fuel reprocessing plant discharges, Atmospheric weapons tests
^{14}C		5.70×10^{3}	β	Natural, Atmospheric weapons tests
^{99}Tc		2.11×10^{5}	β	coastal discharges, atmospheric weapons tests
^{137}Cs	5.57	30.2	γ, β	Atmospheric weapons tests, Nuclear fuel reprocessing plant discharges, Fukushima, Chernobyl
^{134}Cs		2.06	γ, β	Fukushima, Chernobyl
^{90}Sr	3.50	28.8	β	Atmospheric weapons tests, Nuclear fuel reprocessing plant discharges
^{129}I		1.57×10^{7}	γ, βb	Nuclear fuel reprocessing plant discharges, Natural
^{239}Pu		2.41×10^{4}	α	Atmospheric weapons tests
^{240}Pu		6.56×10^{3}	α	Atmospheric weapons tests

Aoyama, 2019, Artificial radionuclides, Encyclopedia of Ocean Sciences

Source of radionuclides in our environment unit: PBq

source terms	year	137Cs	134Cs	90Sr	3H	106Ru	133Xe	131I
Atmospheric weapons tests (Northern Hemisphere only)	1970	765+-79*		310#	240000##			
Atmospheric weapons tests (North Pacific Ocean only)	1970	290+-30*						
Nuclear facility accidents								
Windscale**	1957	0.044	0.0011	0.00022			14	0.59
Three Mile Island***	1979						74-370	0.00056
Chernobyl****	1986	85	47	10			6500	1760
Fukushima (release to atmosphere)	2011	15-20	15-20	0.01-0.2###			11000####	160
Fukushima (direct discharge to ocean)	2011	3.5+-0.7	3.5+-0.7	around 1$	0.05$$			20$$
Liquid effluents from nucler fuel reprocessiong plants at UK and France $$$$								
	1970–1979	29.78		3.83	11.7	7.7		
	1980–1984	9.15		1.63	13.6	4.1		
	1985–1989	0.46		0.47	23.6	2.1		
	1990–1994	0.13		0.26	35.4	0.3		
	1995–1999	0.07		0.26	69.4	0.1		
	2000–2004	0.05		0.10	73.1	0.1		
	2005–2008	0.06		0.03	48.9	0.0		

*:Aoyama et al., 2006 **:SCOP50 ***:THE KEMENY REPORT ON THE ACCIDENT AT THREE MILE ISLAND (October, 1979)https://tmi2kml.inl.gov/Documents/2a-Kemeny/Presidents%20Commission,%20Technical%20Assessment%20Task%20Force,%20Vol.%20I%20(1979-10-30).pdf ****:UNSCERA2008
#:An activity ratio between 137Cs and 90Sr at Tokyo during the period from 1963 to 1965 times 137Cs amount in Aoyama et al., 2006
##:Galeriu and Melintescu, 2011, global inventory ###: assumed that 137Cs vs. 90 Sr activity ratio is 100
####:Report of the Japanese Government to the IAEA Ministerial Conference on Nuclear Safety - The Accident at TEPCO's Fukushima Nuclear Power Stations -, June 2011
$:Povinec et al., 2012 $$:Takahata et al., 2018 $$$:Tsumune et a;., 2012. 131I/137Cs activity ratio is 5.7.
$$$$Reproduced from Table 40 in United Nations, "ANNEX C", in Sources and Effects of Ionizing Radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, 2000 Report to the General Assembly, with scientific Annexes, I, United Nations publication, Austria, 2000, 158–291 (2000) for the period from 1970 to 1994. And, annual data was summarized from three EU reports European Commission Radiation Protection 127, 143 and 164 for the period from 1995 to 2008 (Van der Stricht and Janssens, 2001, 2005, 2010)

Comparison of environmental characteristics

^{3}H

- HTO
- Evaporation/Precipitation
- Not easy to conduct to simulation after injected into marine environment because of evaporation, precipitation. Need to consider fresh water flux from rivers/underground water
- Concentration Factor is ca. 1 for fish filet in marine environment (rapid equilibrium between tissue-free water tritium, TFWT, and the environment)

^{137}Cs

- Cs^{+} cation: NaCl, KCl, CsCl in ocean
- Less mobility on land
- No evaporation, resuspension on land
- Wet and dry deposition
- Easy to conduct to simulation after injected into marine environment because move as seawater
- Concentration Factor is ca. 100 for fish filet in marine environment

^{137}Cs activity concentration in surface water

Red: 56N at FNPP1, Fukushima coast
Black: Pacific Ocean

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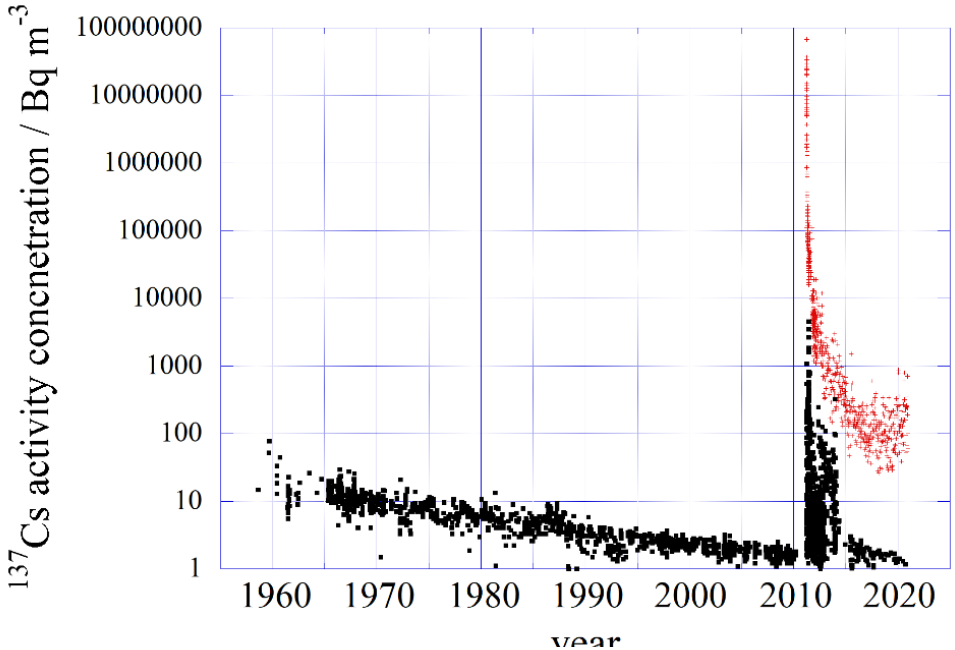
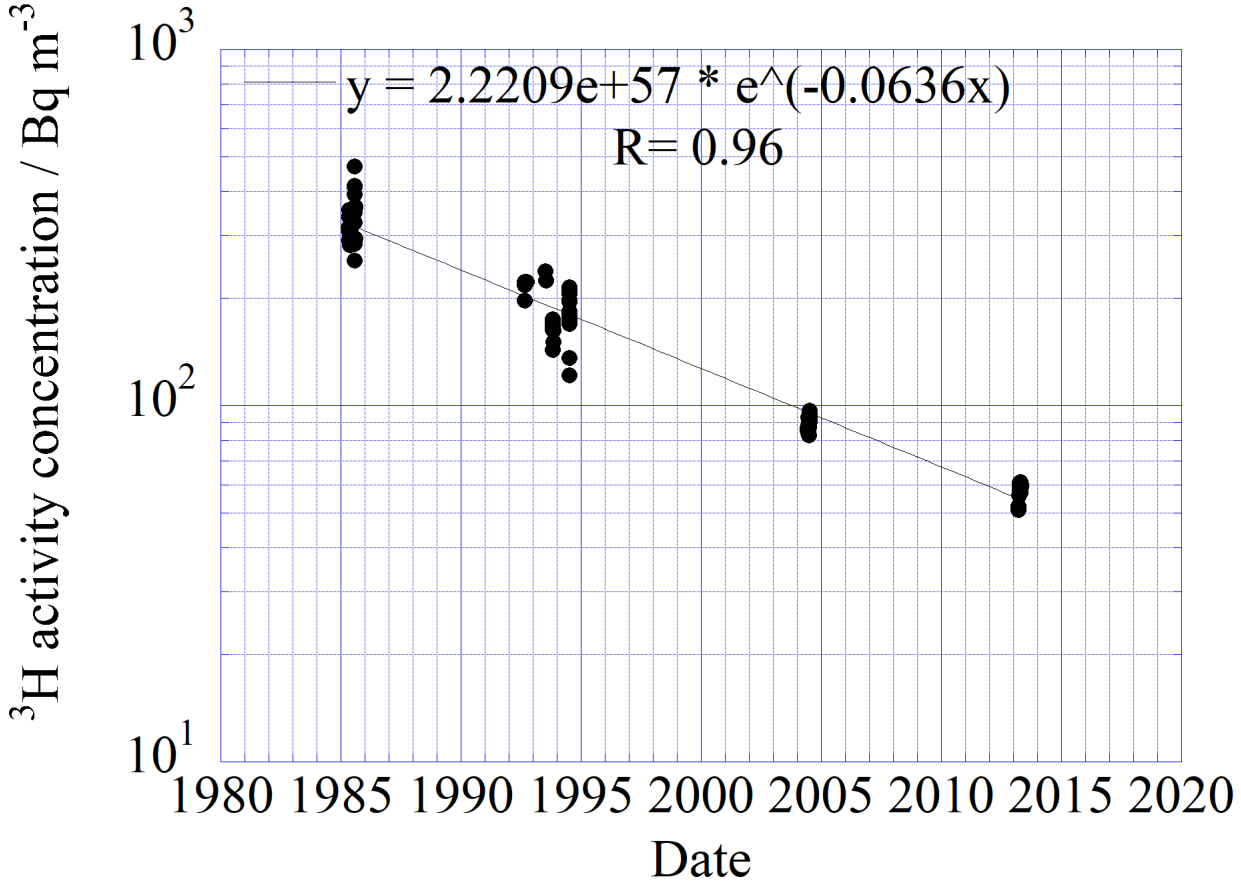


Apparent half residence time of 137Cs in the Pacific Ocean before 2011 :
ca. 16 years

Left ; Aoyama, 2019, Artificial radionuclides, Encyclopedia of Ocean Sciences
right : 青山、岩波科学2021年4月号

^{3}H activity concentration in the mid latitude of western Pacific Ocean



Apparent half residence time:
ca. 10 years. Less advection/diffusion compared with 137Cs which means source exist for 3H

Aoyama, M., Charmasson, S., Hamajima, Y., Duffa, C., Tsumune, D., and Tateda, Y.: Tritium activity concentration and behaviour in coastal regions of Fukushima in 2014, Biogeosciences Discuss. [preprint], https://doi.org/10.5194/bg-2021-10, in review, 2021.

Trend of tritium activity concentration in the mid latitude of the Nor Atlantic Ocean

360 Bq m-3

240 Bq m-3

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120 Bq m-3

1 TU = 118 Bq m-3

Apparent half residence time:
ca. 10 years same as that in the NP

Pierre-Emmanuel Oms, Pascal Bailly du Bois, Franck Dumas, Pascal Lazure, Mehdi Morillon, Claire Voiseux, Cedric Le Corre, Catherine Cossonnet, Luc Solier, Pascal Morin, Inventory and distribution of tritium in the oceans in 2016, Science of The Total Environment, Volume 656, 2019,

^{137}Cs activity concentration in surface water in 1970-1979(left) and 2011(right)

Higher in the NH, lower in the SH

Higher close to the sources

Two figures were deleted from here

Aoyama, M., 2018. Long-range transport of radiocaesium derived from global fallout and the Fukushima accident in the Pacific Ocean since 1953 through 2017-Part I: Source term and surface transport, J Radioanal Nucl Chem, 318, 1519-1542. doi: 10.1007/s10967-018-6244-z

1TU = 118 Bq m-3

Again, Higher in the NH, lower in the SH and higher at close to the sources regions same as the feature of 137Cs

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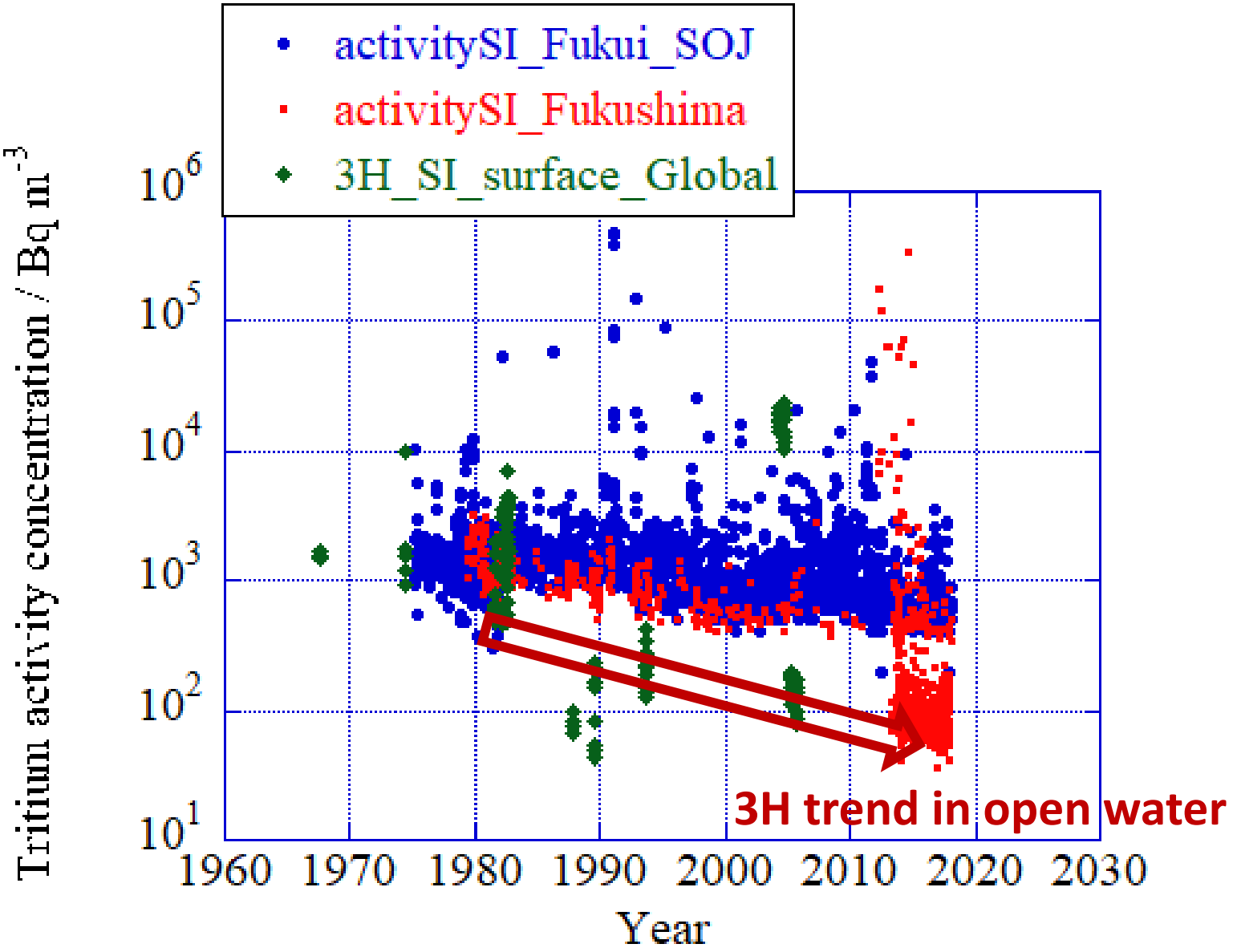
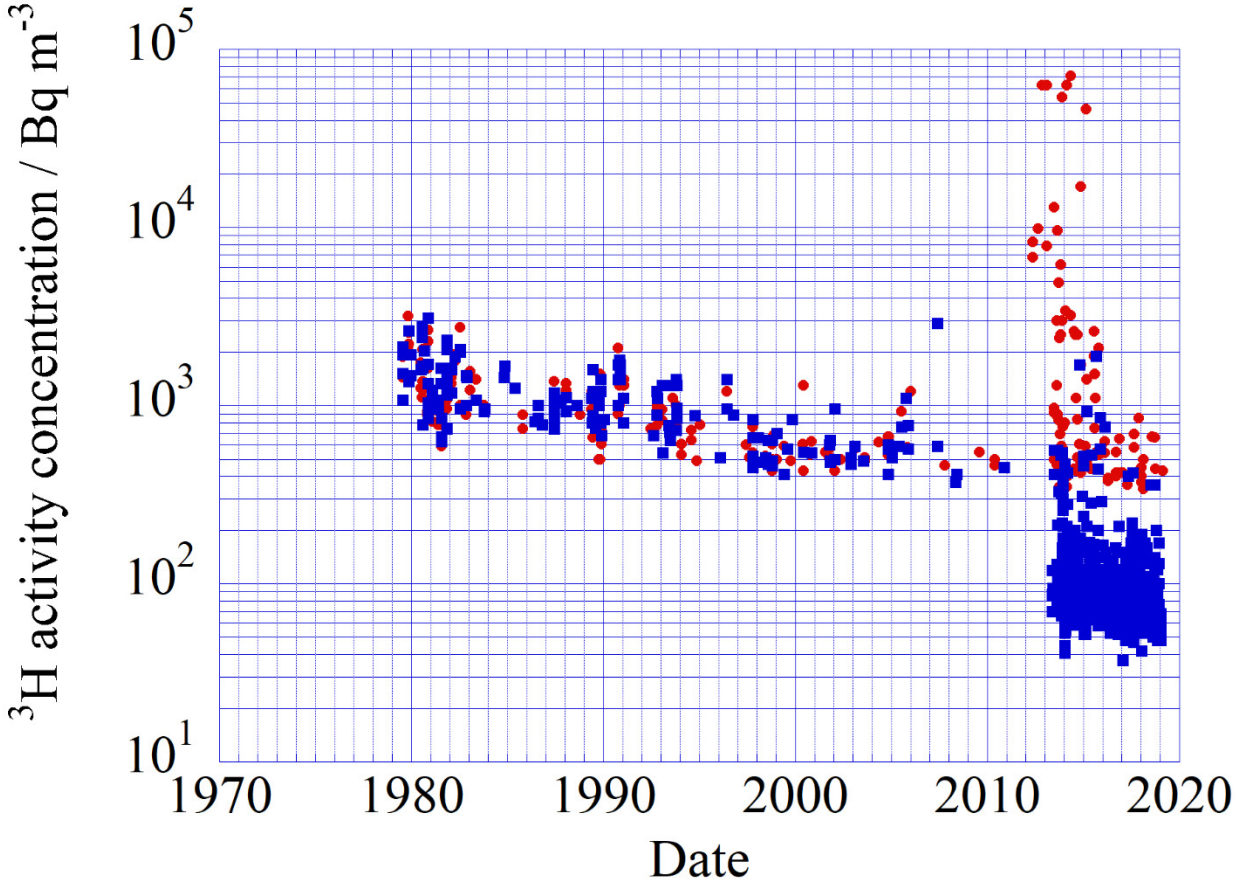
Fig. 4. **Mean decay-corrected tritium concentrations** in the oceans between 0 and 500 m depth (TU16), between years 1967 and 2016. Boxes boundaries and decay-corrected and non- decay–corrected tritium concentrations are listed in Table 2. If several measurements are available at one location over time, the average concentration is plotted. New tritium concentrations from this article are represented with squares, while data compiled from the literature are shown with circles. Nuclear power plants are indicated with red circles (data obtained from http://GlobalEnergyObservatory.org).

Pierre-Emmanuel Oms, Pascal Bailly du Bois, Franck Dumas, Pascal Lazure, Mehdi Morillon, Claire Voiseux, Cedric Le Corre, Catherine Cossonnet, Luc Solier, Pascal Morin, Inventory and distribution of tritium in the oceans in 2016,
Science of The Total Environment, Volume 656, 2019

The features close to Japan: the SOJ (@Fukui) and Fukushima coast vs. open water



Details of ^{3}H trend at stations close to Fukushima NPP since end of 1970s



Red: just in front of FNPP1

Blue coastal stations close to FNPP1

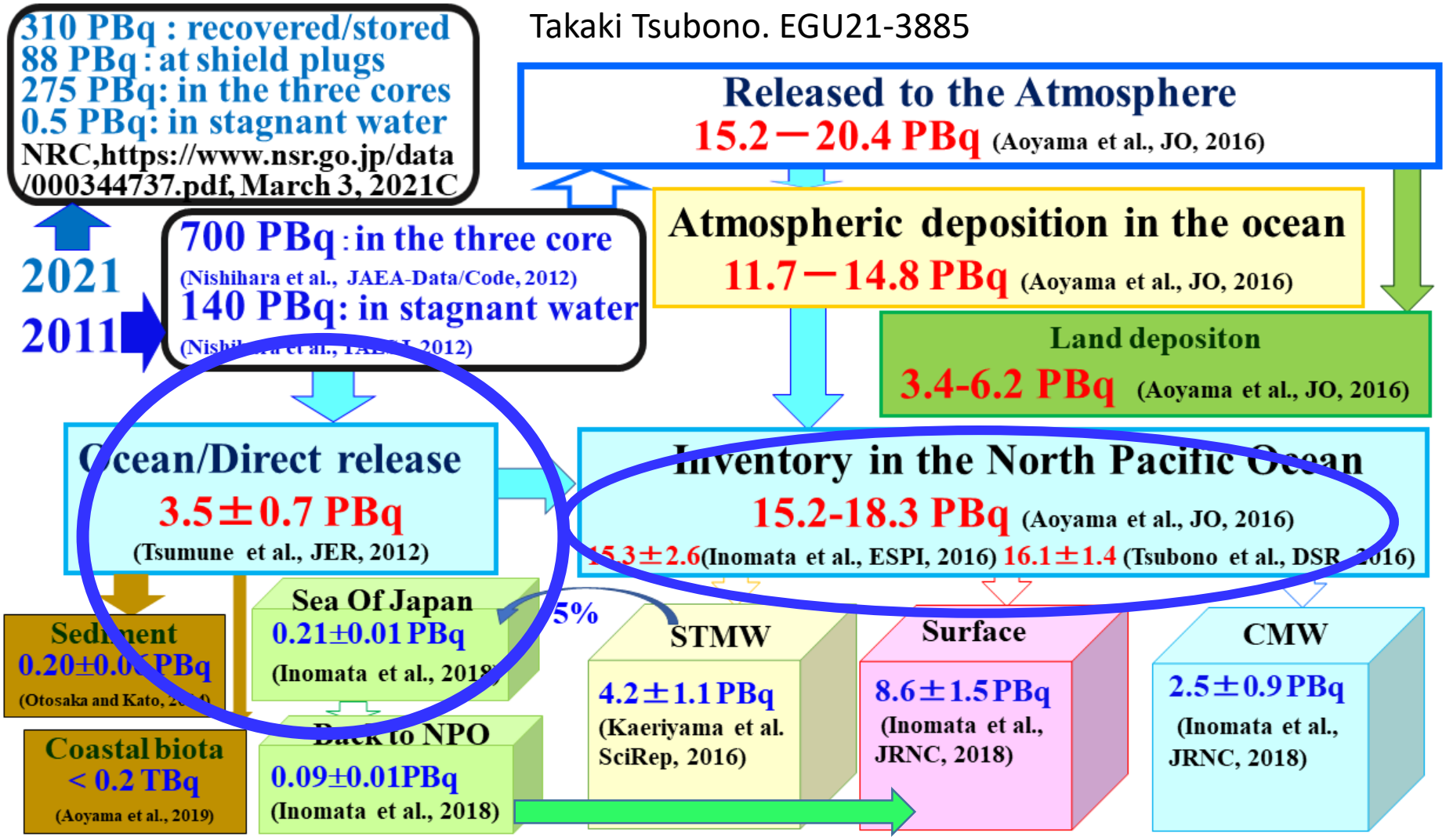
Before 2011, planed 3H release made 3H activity concentration as well mixed, but after the accident 3H activity concentration in front of FNPP1 increase sporadically due to un-controled release while lowest activity was close to that in open water.

Aoyama, M., Charmasson, S., Hamajima, Y., Duffa, C., Tsumune, D., and Tateda, Y.: Tritium activity concentration and behaviour in coastal regions of Fukushima in 2014, Biogeosciences Discuss. [preprint], https://doi.org/10.5194/bg-2021-10, in review, 2021.

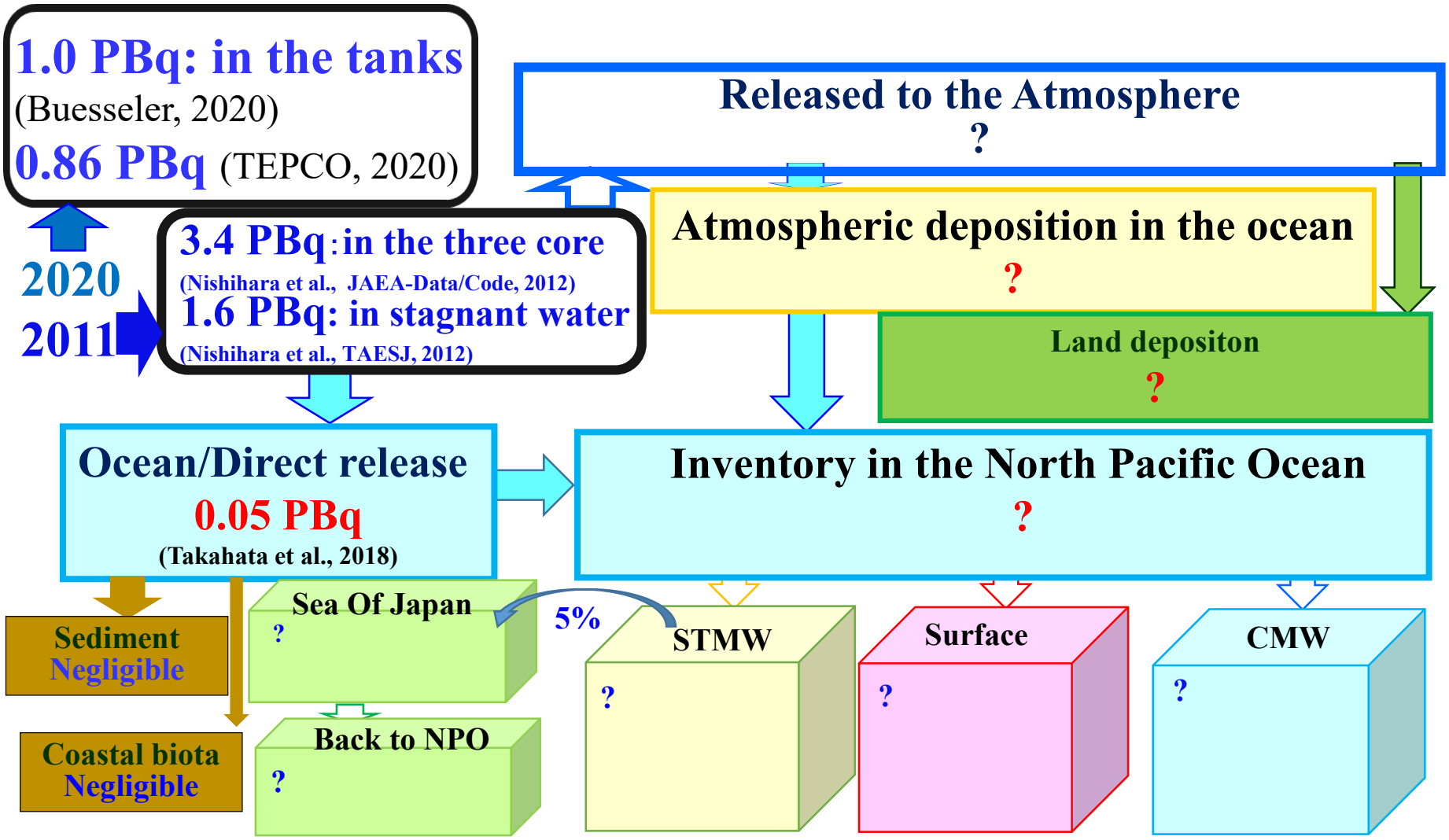
Final mass balance of radiocaesium released from FNPP1 on March 2011
Radioactivity presented here are for ^{137}Cs (^{134}Cs) as of 11 March 2011

We know many!!

Michio Aoyama, Yayoi Inomata , Daisuke Tsumune, and Takaki Tsubono. EGU21-3885

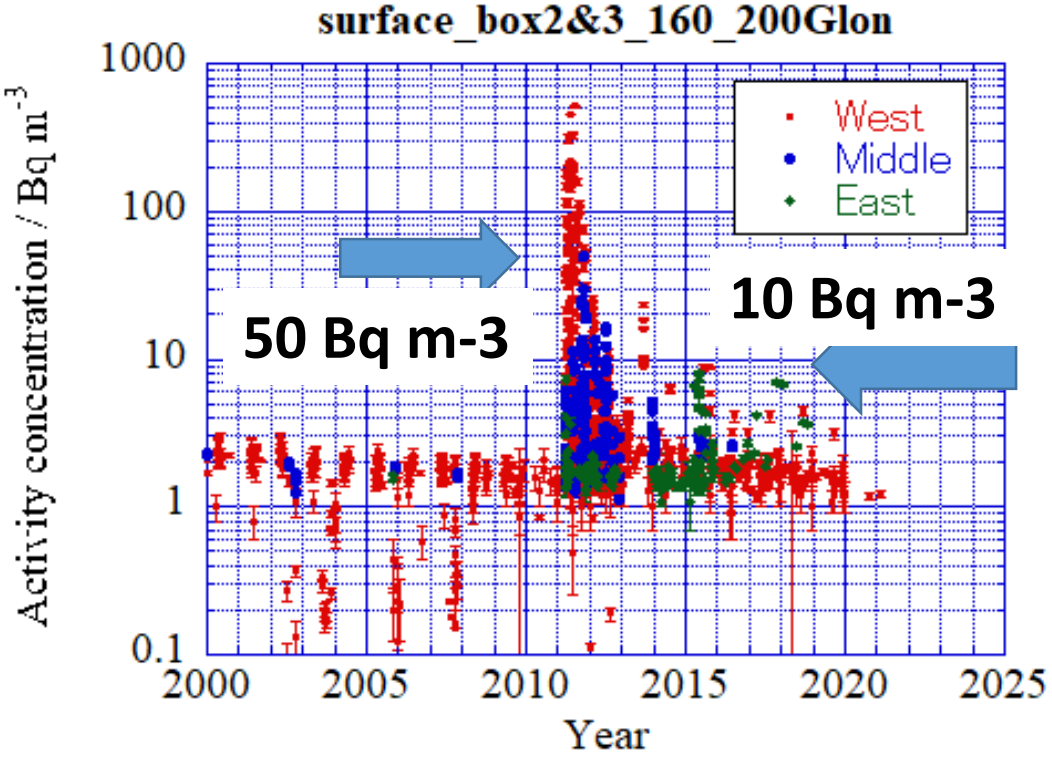


Mass balance of tritium: we know a few



North Pacific model simulation of ^{137}Cs (lower panel) and observed activity at mid latitude of the North Pacific Ocean (right)

50 Bq m^{-3} at the middle of boxes2&3 and 10 Bq m^{-3} at US coast were observed.



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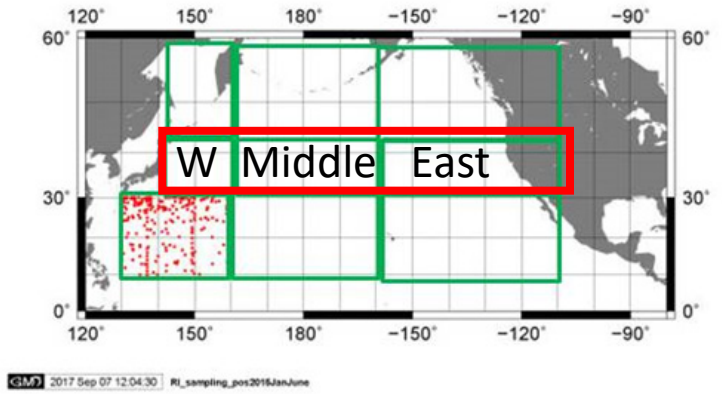

Tsubono et al., 2016 source: direct discharge and atmospheric deposition

Main surface transport pathways were Kuroshio and Kuroshio extension, subarctic gyre and subtropical gyre. A part of released radiocaesium was in the STMW and was transported to the SOJ.

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青山道夫、「10年間福島事故で海洋に放出された放射性物質を追いかけて見えたこと」Isotope News No.773、2021

Signal of Fukushima accident in SOJ: 1-2 Bq m^{-3} increase from 1.5 Bq m^{-3}

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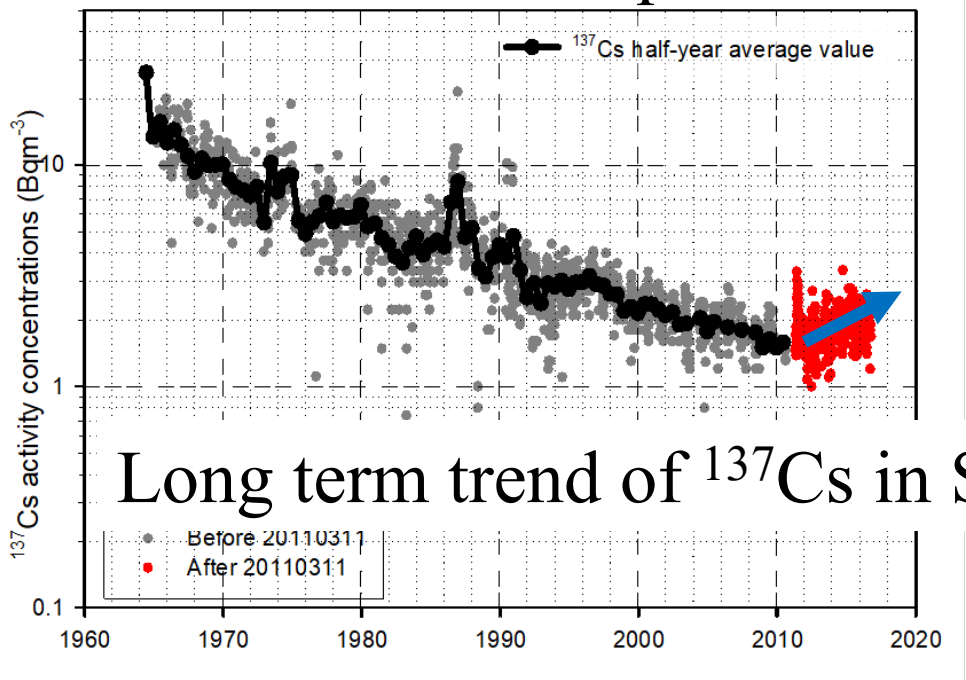
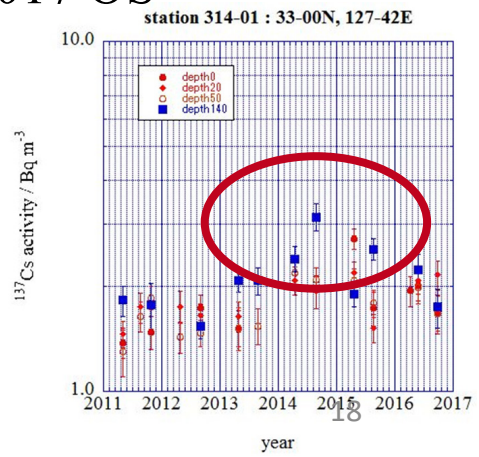


Long term trend of ^{137}Cs in SOJ

left : Aoyama et al., 2017, JARI Observed results in 2015

right : Inomata and Aoyama, 2017 OS

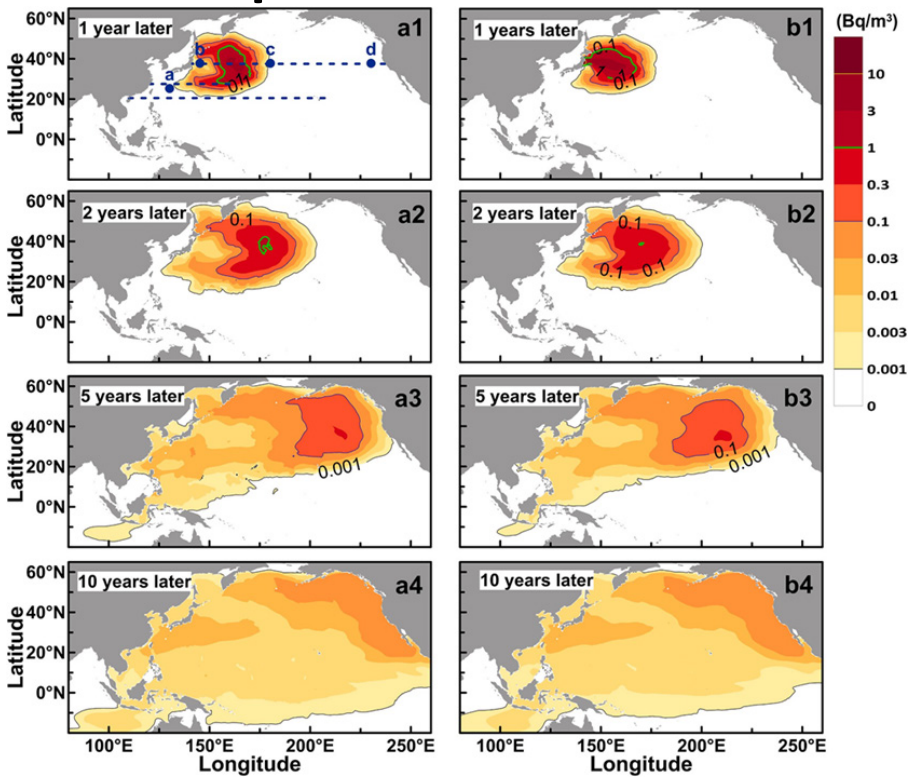


right : Data source: KINS/ER-092(2016)and previous reports

A comparison of simulations

^{3}H

50-70 Bq m^{-3} BG not included

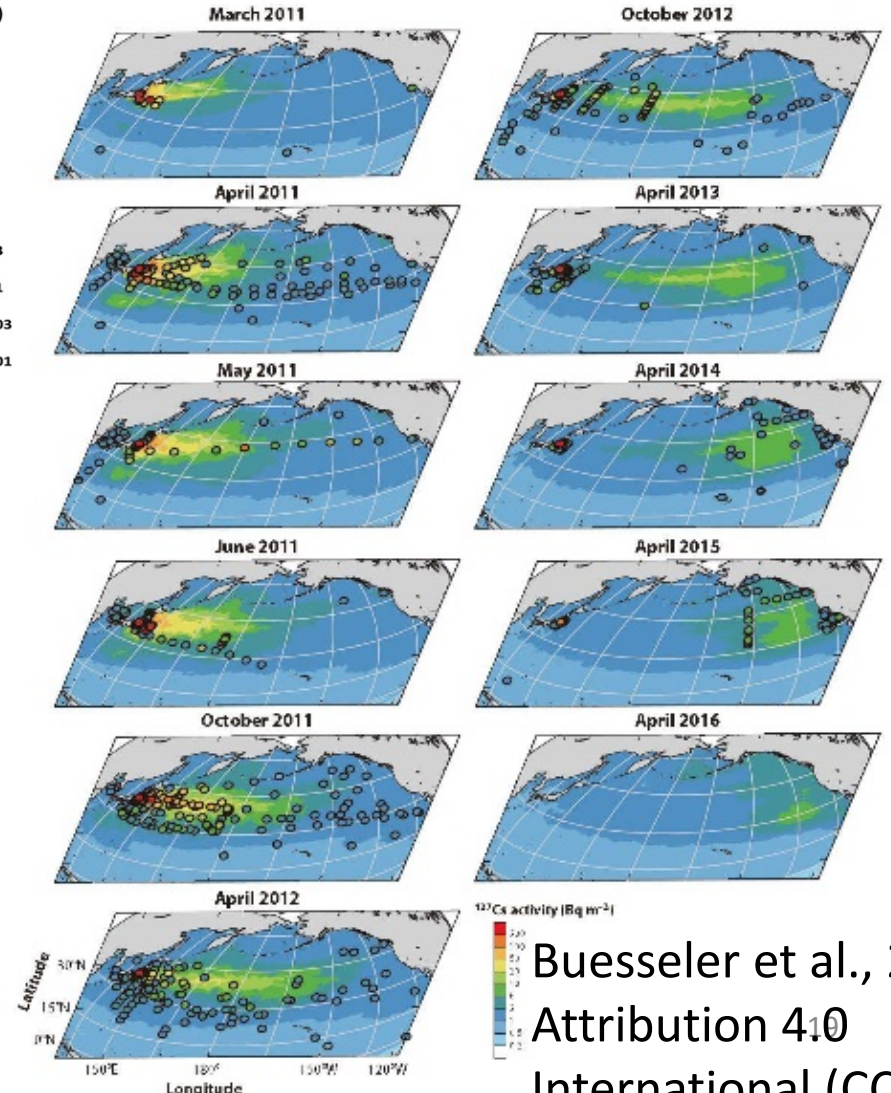


Left: 1.2 PBq 1 month release

Right: 1.2 PBq 1 year release

^{137}Cs

1.5 Bq m^{-3} BG included



Result of a proportional calculation : ^{3}H vs. ^{137}Cs in the middle/east in the North Pacific and the SOJ

	Source		Pacific Ocean	SOJ
			Bq m-3	Bq m-3
Pre-Fukushima 137Cs	weapons tests	observation	1.5	1.5
Pre-Fukushima 3H	natural, weapons tests and nuclear power plants	observation	50-70	500-10000
15-18 PBq release	direct release and atmopsheric deposition for about one month	observation	10-50	1-2
			one year	3-5 years
1.5 PBq release	**controled release for one month**	**proportional calculation by MA**	**1-5**	**0.1-0.2**
1.2 PBq release	controled release for one month	Zhao et al.	around 2	0.01
0.1 PBq release	controled release for one year planed by TEPCO		max. 0.33	max. 0.01

Evaluation at the coastal region TEPCO released simulation results.

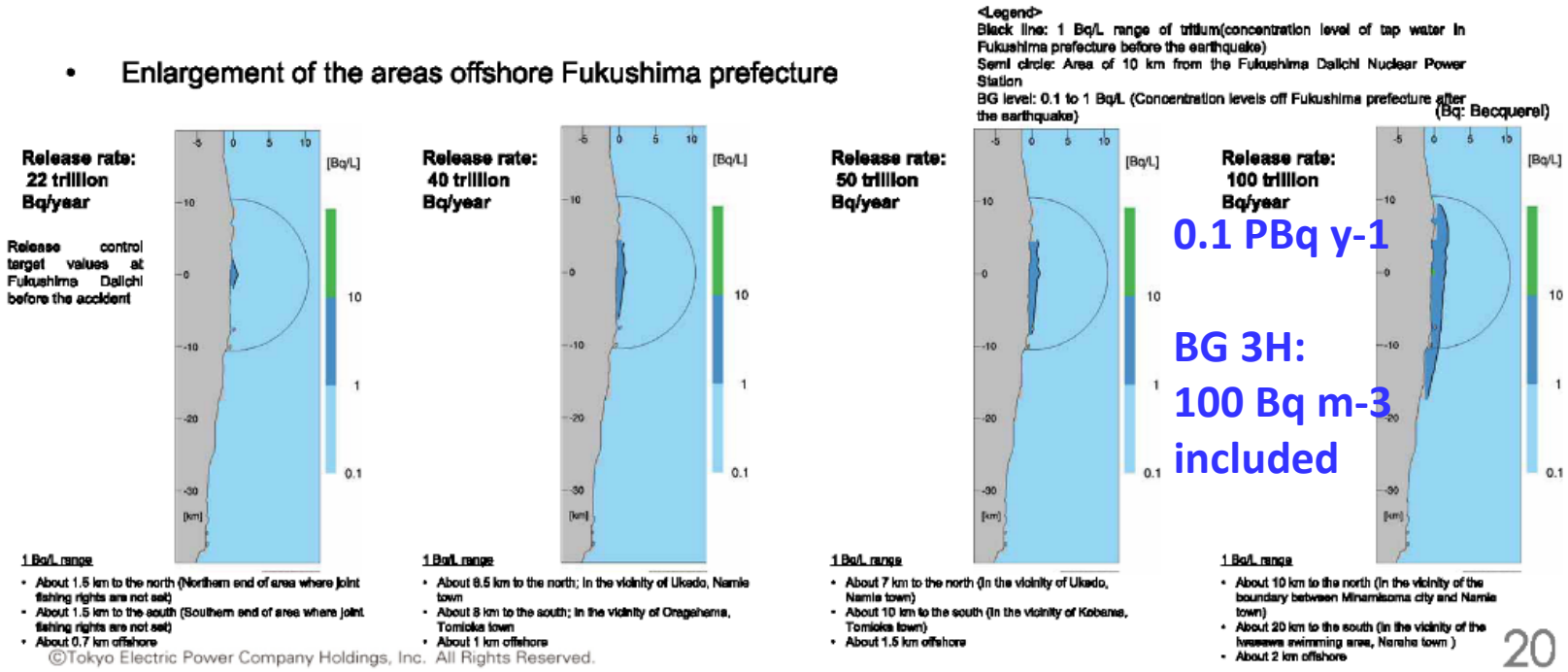
b. Discharge into the Sea: Simulation of Diffusion [1/2]

TEPCO

- Simulation conditions (Model verified with measured data for cesium-137)
  - Target sea area: About 500 km north-south and about 600 km offshore focusing on Fukushima Prefecture
  - Resolution: 1 km mesh in horizontal direction, 30 layers in vertical direction with respect to water depth (up to depth of 1 km)
  - Weather conditions, etc.: Uses wind speed, air pressure, temperature, humidity, and precipitation from January to December 2014

(Including flow conditions off Fukushima Prefecture (Kuroshio current / Mid-range eddies))

- Enlargement of the areas offshore Fukushima prefecture



https://www.tepco.co.jp/en/decommission/progress/watertreatment/images/200324.pdf

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Evaluation at the coastal region by a proportional calculation

	Source		56N at FNPP1
			Bq m-3
Pre-Fukushima 137Cs	weapons tests	observation	1.5
Pre-Fukushima 3H	natural and weapons tests	observation (Aoyama et al., 2021)	**67 +- 20**
15-18 PBq release	direct release and atmopsheric deposition for about one month	observation	**6.7×10^{7}**
			within a day
1.5 PBq release	**controled release for one month**	**scaling factor (Tsumune et al., 2012)**	**2.5×10^{6}**
1.2 PBq release	controled release for one month	Zhao et al.	not shown
0.1 PBq release	controled release for one year planed by TEPCO	**scaling factor (Tsumune et al., 2012)**	**13×10^{3}**
0.1 PBq release	controled release for one year planed by TEPCO	**TEPCO simulation BG as 100 Bq m-3**	**Activity>1000 area 30km x 2km**

Other radionuclides in waters in the tanks

- 3H is not an issue regarding with release of waters in the tanks as shown in the previous slides except very close to the release site.
- On the contrary, other radionuclides impacts such as C-14, I-129, Cs-137, Sr-90, Ru-106, similar to previous releases from nuclear fuel reprocessing plant in UK and France until the 1990s (UNSCAEAR 2000), should be evaluated before release. The impact of Co-60 and Sb-125 should be also evaluated.

summary

- We know what happened in the ^{137}Cs case after the TEPCO Fukushima accident based on observations and model simulations as shown in my talk.
- Based on the observed facts, we can evaluate the impact of tritium releases, which is planed by TEPCO, by only doing a simple proportional calculation without computer model simulations.
- It is safe to say for me that nobody can detect the tritium increase in open water, the North Pacific, and the SOJ except areas very close to the release site.
- Close to the site, impact to community living there or doing fisheries should be evaluated in terms of social science, not natural science.


御清聴ありがとうございました
Thank you for your attention!