

<https://doi.org/10.5194/essd-2022-374-RC3>

### **Answer to the reviewer comments (Anonymous Referee 2)**

Thank you very much for your valuable comments. I modified the manuscript taking into account your comments.

### **Detailed specific comments**

#### **Abstract:**

- Line 10, Pg.1:

Reviewer understands that authors used mainly HAMGlobal2021 data and partly MARIS data, but authors mentioned only HAMGlobal2021 data. It is better to describe this matter correctly.

A. MARIS data were also included in the HAMGlobal2021. After published the manuscript in ESSD, researcher in the IRSN send me their data set (about 2000 records). I asked to Editor to use these additional data. These new data were also used in this analysis.

In HAMGlobal2021, the dataset produced by the IAEA Marine Radioactivity Information System (MARIS) were combined. Furthermore, we used the <sup>137</sup>Cs data reported in the IRSN database (Baily du Bois, P. et al., 2020). Finally, all these data were compiled into a single comprehensive database for this study.

The data used in this study were adopted from the HAMGlobal2021: Historical Artificial radioactivity database in the Marine environment, Global integrated version 2021 (Aoyama, 2021), which contains data from the F1NPS accident. The HAMGlobal2021 database contains information on several radionuclides (<sup>134</sup>Cs, <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>3</sup>H, <sup>239,240</sup>Pu, <sup>241</sup>Am, and <sup>14</sup>C) in the global ocean. The data were measured from 1956 to 2021. The dataset in International Atomic Energy Agency Marine Radioactivity Information System (IAEA MARIS) database were also compiled in the HAMGlobal2021. In addition to this, the data measured in the North Atlantic Ocean and its marginal seas developed by IRSN (Bois et al., 2020) were also contained in this study.

- Lines 11-17, Pg.1:

Author used data from 1956-2021 data but no statement before 1970 is found in the abstract. It is desirable to include comments before 1970.

- A. The 0.5-yr median value of  $^{137}\text{Cs}$  in each box in the Pacific Ocean, the values were gradually increased or almost constant levels in the 1950s and 1960s, and then, except in the northern North Atlantic Ocean and its marginal sea, decreased exponentially in 1970–2010, immediately before the Fukushima Nuclear Power Plant (F1NPS) accident.

### **Introduction:**

- Line 30, Lines 35-36, Line 46, Pg.2:

Authors repeatedly elaborated that the Cs137 originates from large-scale atmospheric weapons tests. Writing improvements are needed.

- A.  $^{137}\text{Cs}$  is regarded as one of the most abundant artificial radionuclides in the ocean because of its long half-life (30.17 yr) and large fission yield that originates from large-scale atmospheric weapons tests due to atmospheric nuclear weapon tests by the United States and Russian Federation. Atmospheric nuclear weapons tests occurred from 1945 to 1980. During 1945 to 1963, the large scale atmospheric nuclear weapons tests were conducted by the United States. In 1963, the Partial Nuclear Test Ban Treaty was signed and these tests in the atmosphere by the United States and Soviet Union, and Great Britain shifted to the underground. However, France continued the atmospheric test until 1974 and China until 1980. In addition,  $^{137}\text{Cs}$  has been released into the Pacific Ocean by local fallout from ground tests (e.g., UNSCEAR 2000; Aoyama et al., 2006; Aoyama, 2010; Inomata, 2010) on Bikini Atoll in the Marshall Islands between 1946 and 1958 by the United States. Because the  $^{137}\text{Cs}$  released into the atmosphere fallout onto the ocean surface, the ocean is recognized as the largest receptor of  $^{137}\text{Cs}$  on Earth. Furthermore, other sources, such as the accidental release from nuclear facilities (the Three Mile Island nuclear power plant in 1979), sea dumping of nuclear wastes from nuclear facilities carried out in 1986 in the north–central East Sea/Japan Sea by the former Soviet Union and Russian Federation, lost nuclear weapons, and the use of radioisotopes in human activities, such as industry, medicine, and science, are recognized. These contributions in the environment are minor compared to those from the dominant sources listed above (UNSCEAR, 2000; IAEA, 2005).

- Lines 54-55, Pg.2:

A. In addition,  $^{137}\text{Cs}$  has been released into the Pacific Ocean by local fallout from ground tests on Bikini Atoll in the Marshall Islands between 1946 and 1958 by the United States (e.g., UNSCEAR 2000; Aoyama et al., 2006; Aoyama, 2010; Inomata, 2010).

- Lines 80-83, Pg. 3:

-Reviewer is confused with authors statement such that the atmospheric deposition of  $^{137}\text{Cs}$  into the ocean was estimated to be 11.7-14.8 PBq (Aoyama et al., 2016b) and that the  $^{137}\text{Cs}$  inventory into the North Pacific Ocean was estimated to be 15.2-18.3 PBq (Aoyama et al., 2016b, Inomata et al., 2016; Tsubono et al., 2016). Clear description is required.

A. Atmospheric deposition of  $^{137}\text{Cs}$  into the ocean mean that  $^{137}\text{Cs}$  deposition amount from the atmosphere in the ocean surface. The  $^{137}\text{Cs}$  inventory means that  $^{137}\text{Cs}$  existed amount into the surface mixed layer. In order to clear these mean, the author modified as follows.

The atmospheric deposited amount of  $^{137}\text{Cs}$  into the ocean from the atmosphere was estimated to be 11.7-14.8 PBq (Aoyama et al., 2016b). Directly discharged liquid  $^{137}\text{Cs}$  from the FINPS was estimated to be  $3.6 \pm 0.7$  PBq by using the observation data around the FINPS and model simulation (Tsumune et al., 2012, 2013). The  $^{137}\text{Cs}$  inventory into the North Pacific Ocean in the surface mixed layer was estimated to be 15.2-18.3 PBq (Aoyama et al., 2016b), which are consistent with the estimated values by optical statistical analysis (Inomata et al., 2016) and model simulation (Tsubono et al., 2016).

- Regarding the statement “directly discharged liquid  $^{137}\text{Cs}$  from the FINPS was estimated to be  $3.6 \pm 0.7$  PBq (Tsumune et al., 2012, 2013)”, comments that there are various estimates in literature need to be added.

A. The released  $^{137}\text{Cs}$  by the FINPS accident were investigated by numerous researches and summarized in Busseler et al. (2017). However, we used the values by considering the mass balances among atmosphere, land, and ocean. The atmospheric deposited amount of  $^{137}\text{Cs}$  into the ocean from the atmosphere was estimated to be 11.7-14.8 PBq (Aoyama et al., 2016b). Directly discharged liquid  $^{137}\text{Cs}$  from the FINPS was estimated to be  $3.6 \pm 0.7$  PBq by using the observation data around the FINPS and model simulation (Tsumune et al., 2012, 2013). The  $^{137}\text{Cs}$  inventory into the North Pacific Ocean in the surface mixed layer was estimated to be 15.2-18.3 PBq

- Lines 148-151, Pg. 5:

-It is not clear what the statement “latitudinal and longitudinal distributions, and the locations of global fallout, reprocessing plants, and nuclear power plants” represents. “latitudinal and longitudinal distributions of global fallout, and the locations of reprocessing plants, and nuclear power plants”??

A. Actually, distribution of  $^{137}\text{Cs}$  are controlled by the ocean current and location of point source such as the reprocessing plants. Because  $^{137}\text{Cs}$  exists as water soluble, spatial distribution and temporal variation of  $^{137}\text{Cs}$  are related with these horizontally and vertically, and sources. This part was modified as, “The boxes were determined based on the ocean current and location of sources.” In addition, author added the location and feature of each box related with ocean current in more detail.

Measured  $^{137}\text{Cs}$  data, however, was very limited and it is impossible to cover the distribution of  $^{137}\text{Cs}$  in the global ocean. In this study, the global ocean was divided into 37 boxes to investigate the temporal variations in  $^{137}\text{Cs}$  activity concentrations in surface seawaters by using the available almost all data (Inomata and Aoyama, 2022a) (Figure 1). These boxes were divided by showing the latitudinal and longitudinal distributions based on the known ocean currents (IAEA, 2005; Open University, 2004), the latitudinal distributions of global fallout, location of reprocessing plants and F1NPS under the assumptions that  $^{137}\text{Cs}$  activity concentrations in the box is almost same (Hirose et al., 2003; Inomata et al., 2009; IAEA, 2005). Subarctic North Pacific Ocean (Box1, north  $40^\circ\text{N}$ ) is the highest atmospheric deposition of  $^{137}\text{Cs}$  occurred in the 1960s in the Pacific Ocean, western North Pacific Ocean and eastern North Pacific Ocean (Box2, Box 3), which locate in  $25\text{-}40^\circ\text{N}$ , are upstream and downstream of Kuroshio extension. These three regions are influenced the  $^{137}\text{Cs}$  contamination derived from the F1NPS accident. Subtropical western and eastern North Pacific Ocean (Box 4, Box 5) are downstream and upstream of the north Equatorial Current associated with the subtropical Gyre. Subtropical western and eastern North Pacific Ocean includes the California Gyre ( $5\text{-}25^\circ\text{N}$ ). These boxes include the contamination of local fallout such as the Bikini Atoll. Western and eastern equatorial Pacific Ocean (Box 6, 7) are downstream and upstream of the South Equatorial Current. And upwelling of seawater occurs in the eastern Southern Pacific Ocean. Western and eastern subtropical North Pacific Ocean (Box 8, 9) are down stream and upstream of the weak South Equatorial Current. Eastern subtropical South Pacific Ocean includes the French nuclear weapons test sites. The western Southern Pacific Ocean ( $25\text{-}40^\circ\text{S}$ ) is Tasmania Sea (Box 10). Eastern South Pacific Ocean ( $25\text{-}40^\circ\text{S}$ ) is mid-latitude region of the South Pacific Ocean and includes South Pacific Current (Box 11). Eastern Southern Ocean ( $40\text{-}60^\circ\text{S}$ ) is affected by the Antarctic Circumpolar Current (Box 12). Antarctic Ocean (below  $60^\circ\text{S}$ ) are divided into three; Antarctic sector for Pacific (Box13), Indian (Box 36), and Atlantic (Box 37) and locate the polar front and continental water boundary. In the Middle Southern Ocean (Box 19), the Leeuwin Current, which flow to southward along the continental

shelf off the western Australia (22°S to 35°S) in the Southern Ocean (Box 17), transport eastward. The Southern Ocean is characterised as subtropical gyre, and connected with the Antarctic Circumpolar Current from the Atlantic Ocean side in the upstream and the Pacific Ocean side in the downstream. The Antarctic Circumpolar Current is banded structure with several current and formed as narrow jet with sharp front. Indian Ocean (Box 16) is connected to Indonesian Archipelago (Box 35) by Indonesian through flow from the Pacific Ocean. The Arabian Ocean (Box 15, 10°S-30°N) is affected by the equatorial current system and the circulation in the Northern Indian Ocean associated with monsoon system. In the marginal seas of the North Pacific Ocean, South China Sea (Box 33), Eastern China Sea (Box 32), Japan Sea (Box 14), and Okhotsk Sea (Box 31) are classified. The Eastern China Sea influence of the bifurcation of Kuroshio Current and downstream of western North Pacific Ocean and connected to the Japan Sea via Tsushima Warm Current. The northward transported seawater in the Japan Sea is connected to the Sea of Okhotsk. The Bering Sea (Box 34) is downstream of the subarctic North Pacific Ocean and upstream of the Arctic Ocean. The Atlantic Ocean was divided into three, South Atlantic Ocean (Box 28, 60°S-30°S), Central Atlantic Ocean (Box 29, 30°S-15°N), and North Atlantic Ocean (Box 30, 15°N-45°N). The South Atlantic Ocean is connected with the Southern Ocean via the Agulhas Current. The Irish Sea (Box 23) and English Channel (Box 24) are considered as the <sup>137</sup>Cs direct discharged region. The North Sea (Box 22), Barents Sea and Coast of Norway (Box 20), Baltic Sea (Box 21), Arctic Ocean (Box 18) are downstream of the northern North Atlantic Ocean and affected the inflow the <sup>137</sup>Cs derived from the Irish Sea and English Channel. The northern North Atlantic Ocean (Box 25) received <sup>137</sup>Cs global fallout by the large scale weapons tests in the 1950s and 1960s. The Baltic Sea (Box 21), the Mediterranean Sea (Box 27), the Black Sea (Box 26) received the fallout of <sup>137</sup>Cs from the Chernobyl accident.

- -Spatial distribution of Cs137 concentrations is uniform? In each box?
  - A. Measured <sup>137</sup>Cs data, however, was very limited and it is impossible to cover the distribution of <sup>137</sup>Cs in the global ocean. In this study, the global ocean was divided into 37 boxes to investigate the temporal variations in <sup>137</sup>Cs activity concentrations in surface seawaters by using the available almost all data (Inomata and Aoyama, 2022a) (Figure 1). These boxes were divided by showing the latitudinal and longitudinal distributions based on the known ocean currents (IAEA, 2005; Open University, 2004), the latitudinal distributions of global fallout, location of reprocessing plants and FINPS under the assumptions that <sup>137</sup>Cs activity concentrations in the box is almost same (Hirose et al., 2003; Inomata et al., 2009; IAEA, 2005).
- -Horizontal and vertical transport of ocean water is almost same? In each box? It is needed to elaborate in detail the basis of this assumption somewhere in this manuscript.

There are several questions on the assumption. What is the assumption in shallow marginal seas? Are the horizontal and vertical transports of ocean water time-invariant or time-varying? If each box is further divided into two, the assumption such that horizontal and vertical transports of ocean water are almost same is still valid?

A.

Measured  $^{137}\text{Cs}$  data, however, was very limited and it is impossible to cover the distribution of  $^{137}\text{Cs}$  in the global ocean unlike the remote sensing data and/or weather data. In this study, the global ocean was divided into 37 boxes to investigate the temporal variations in  $^{137}\text{Cs}$  activity concentrations in surface seawaters (Inomata and Aoyama, 2022a) (Figure 1). These boxes were divided based on the known ocean currents (IAEA, 2005; Open University, 2004), location of reprocessing plants and F1NPS under the assumptions that  $^{137}\text{Cs}$  activity concentrations in the box is almost same and sources of  $^{137}\text{Cs}$  are established (Hirose et al., 2003; Inomata et al., 2009; IAEA, 2005). Marginal Seas such as Japan Sea are based on the definition of IHO (1953). The temporal variation of  $^{137}\text{Cs}$  activity concentrations in the surface seawater in each box were investigated by using the available almost all data. The box divided by showing the latitudinal and longitudinal distributions, oceanographic parameters, and the latitudinal distributions of global fallout. Subarctic ocean (north  $40^\circ\text{N}$ ) is the highest atmospheric deposition of  $^{137}\text{Cs}$  occurred in the 1960s in the Pacific Ocean, western North Pacific Ocean and eastern North Pacific Ocean ( $25\text{-}40^\circ\text{N}$ ) are upstream and downstream of Kuroshio extension. These three regions are influenced the  $^{137}\text{Cs}$  contamination derived from the F1NPS accident. Subtropical western and eastern North Pacific Ocean ( $5\text{-}25^\circ\text{N}$ ) are downstream and upstream of the north Equatorial Current associated with the subtropical Gyre. Subtropical western and eastern North Pacific Ocean includes the California Gyre. These boxes include the contamination of local fallout such as the Bikini Atoll. Western and eastern equatorial Pacific Ocean are downstream and upstream of the South Equatorial Current. And upwelling of sweater occurs in the eastern Southern Pacific Ocean. Subtropical western and eastern North Pacific Ocean are down stream and upstream of the weak South Equatorial Current. Eastern subtropical South Pacific Ocean includes the French nuclear weapons test sites. The eastern Southern Pacific Ocean ( $25\text{-}40^\circ\text{S}$ ) is Tasmania Sea. Eastern South Pacific Ocean ( $25\text{-}40^\circ\text{S}$ ) is mid-latitude region of the South Pacific Ocean and includes South Pacific Current. Eastern Southern Ocean ( $40\text{-}60^\circ\text{S}$ ) is affected by the Antarctic Circumpolar Current. Antarctic Ocean (below  $60^\circ\text{S}$ ) are divided into three; Antarctic sector for Pacific, Indian, and Atlantic and locate the polar front and continental water boundary. Middle Southern Ocean is connected to the Indian sector of the Southern Ocean. Indian Ocean is connected to Indonesian Archipelago by Indonesian through

flow and also connected to the Arabian Sea. In the marginal seas of the North Pacific Ocean, South China Sea, Eastern China Sea, Japan Sea, and Okhotsk Sea are classified. The Eastern China Sea influence of the bifurcation of Kuroshio Current and downstream of western North Pacific Ocean and connected to the Japan Sea via Tsushima Warm Current. The northward transported seawater in the Japan Sea is connected to the Sea of Okhotsk. The Bering Sea is downstream of the subarctic North Pacific Ocean and upstream of the Arctic Ocean. The Atlantic Ocean was divided into three, South Atlantic Ocean (60°S-30°S), Central Atlantic Ocean (30°S-15°N), and North Atlantic Ocean (15°N-45°N). The South Atlantic Ocean is connected with the Southern Ocean. The Irish Sea and English Channel are considered as the  $^{137}\text{Cs}$  direct discharged region. The North Sea, Barents Sea and Coast of Norway, Baltic Sea, Arctic Ocean are down stream of the northern North Atlantic Ocean and affected the inflow the  $^{137}\text{Cs}$  derived from the Irish Sea and English Channel. The northern North Atlantic Ocean received  $^{137}\text{Cs}$  global fallout by the large scale weapons tests in the 1950s and 1960s. The Baltic Sea, the Mediterranean Sea, the Black Sea received the fallout of  $^{137}\text{Cs}$  from the Chernobyl accident.

- Lines 148-160, Pg. 5:

Elaborate why the box configuration is changed.

- A. Taking into account the ocean current, these boxes were further divided. The location of Box and its ocean current, and seawater flow pattern are explained in the manuscript related with the above question. In Box2, 23, and 25, we set several sub-boxes, because the significantly larger values cause the larger  $^{137}\text{Cs}$  values in the box.

The boxes corresponding to the source region, such as the Irish Sea (Box 23; Boxes 23.1-23.5) for the Sellafield plant and the northern North Atlantic Ocean (Box 25; Boxes 25.1 and 25.2) and western North Pacific Ocean (Box 2; Boxes 2.0-2.6) for the F1NPS accident, were divided into several sub regions, because significantly large values around the discharged region cause to larger values to estimate the  $^{137}\text{Cs}$  inventory.

## 2. Data and methods:

- Lines 193-194, Pg. 10:

Authors state that the currents and major source of  $\text{Cs}137$  in the surface water has a 0.5-year time interval. It is not clear what “the currents” and the major source? Describe the detailed

information. One more thing, are the 5-year interval data instantaneous values or some mean values?

A. When we calculate the values in t-yr, the data within  $t \pm 0.5$  yr were used to calculate median value. The t year interval data is instantaneous values at the target year.

The 0.5-yr median values of the surface  $^{137}\text{Cs}$  concentrations in each box were produced by the grid value producing command of block median programs (Wessel et al., 2013). The block median reads the arbitrary data (x, y, z) and calculates the median value in a grid defined in the setting range. In the case of t-year, the data within  $t \pm 0.5$  years were used to calculate the median values.

● Lines 217, Pg. 11:

is used without definition and without physical meaning (Eq. (3) should appear in advance)

A. The apparent half time ( $T_{ap}$ ) of  $^{137}\text{Cs}$  was calculated using the following equations:

$$^{137}\text{Cs} = ^{137}\text{Cs}_0 \exp(-\lambda_{cs, \text{apparent}} t) \quad (1)$$

$$\lambda_{cs, \text{apparent}} = \lambda_{cs, \text{ocean}} + \lambda_{cs, \text{decay}} \quad (2)$$

$$T_{ap} = 0.693 / (\lambda_{cs, \text{apparent}}) \quad (3)$$

$$T_{po} = 0.693 / (\lambda_{cs, \text{ocean}}) \quad (4)$$

where  $\lambda_{cs, \text{apparent}}$ ,  $\lambda_{cs, \text{ocean}}$ , and  $\lambda_{cs, \text{decay}}$  are the decay constants for apparent decay, physical oceanographic decay, and radioactive decay, respectively.  $\lambda_{cs, \text{apparent}}$  is estimated by using the regression line of the 0.5-yr average value of  $^{137}\text{Cs}$  as shown in (1).  $T_{po}$  is the apparent half residence time by causing the oceanic physical processes and  $\lambda_{cs, \text{ocean}}$  was estimated  $\lambda_{cs, \text{apparent}}$  and  $\lambda_{cs, \text{decay}}$  in equation (2). The longer  $T_{ap}$  and negative values of  $T_{po}$  mean that  $^{137}\text{Cs}$  is flowed into the downstream region to the upstream region. However, the exponentially decreasing trend was disturbed by several unexpected accidents, such as the Chernobyl accident in 1986 and the F1NPS accident in 2011, as well as direct discharge from nuclear reprocessing power plants.  $T_{ap}$ , therefore, was estimated for several periods, taking into account the source contribution as follows.  $T_{ap1}$  is before 1970 (periods with nuclear weapon tests at a global scale),  $T_{ap2}$  is the period from 1970 to 1986-1990 (until



the Chernobyl accident), Tap3 is from 1990 to 2010 (after the Chernobyl accident), and Tap4 is after 2011 (after F1NPS accident).

- Lines 221-224, Pg. 10:

-What is the definition of  $\tau$  ?

-What is the definition of  $T_{po}$  and its physical meaning?

A.  $T_{po}$  is physical oceanographic apparent half residence time. In equation (1)-(4), decreased  $^{137}\text{Cs}$  activity concentrations are controlled by radioactive decay and oceanographic physical processes.

$$^{137}\text{Cs} = ^{137}\text{Cs}_0 \exp(-\lambda_{cs, \text{apparent}} t) \quad (1)$$

$$\lambda_{cs, \text{apparent}} = \lambda_{cs, \text{ocean}} + \lambda_{cs, \text{decay}} \quad (2)$$

$$T_{ap} = 0.693 / (\lambda_{cs, \text{apparent}}) \quad (3)$$

$$T_{po} = 0.693 / (\lambda_{cs, \text{ocean}}) \quad (4)$$

The longer  $T_{ap}$  and negative values of  $T_{po}$  mean that  $^{137}\text{Cs}$  is flowed into the downstream region to the upstream region.

Line 281-282. Pages 12

In contrast, a longer  $T_{ap}$  as well as a negative  $T_{po}$  value means that  $^{137}\text{Cs}$  is preserved in the region for a longer time and/or there is an influx of water mass with higher  $^{137}\text{Cs}$  in the region compared to the  $^{137}\text{Cs}$  outflow from the region.

- Lines 279-280, Pg. 13:

Authors state that the maximum monthly mixed layer depth was used because Cs137 is easily transported to the subsurface under deeper mixed layer. Hard to understand why easier subsurface transport is necessary. How about using mean monthly mixed layer depth?

A. The mixed layer depth was the monthly time interval with seasonal variation that is deeper in winter and shallower in summer. It is recognised that sea water subducted from the ocean surface in the mode water formation region associated with the winter convective mixing because of the lower buoyancy from the ocean surface (Hanawa and Tally, 2001). The flow through the winter mixed layer ventilate the sea water into the ocean interior. The maximum monthly mixed layer depth in each box, mainly winter month, was used to calculate the  $^{137}\text{Cs}$  inventory in the mixed layer.

- Lines 293, Pg. 13:

Authors described “horizontal” transport as “outflow to the downstream box” transport. Reviewer thinks it is incorrect because there can be inflow-related transport. “horizontal (net outflow to the downstream box) transport” needs to be used.

A. Line 342-344, Pages 14;

In the marine environment,  $^{137}\text{Cs}$  activity concentrations after 1970 were dominantly controlled by radioactive decay and physical ocean processes, such as horizontal, which mean net outflow to the downstream box for sea water current  $U$  and  $V$  component, and downwards transport below the surface mixed layer for seawater current  $W$  component.

- Lines 296-302, Pg. 13:

-Subscripts in  $C_{i, \text{box}}$  and  $C_{0, \text{box}}$  in (8) are a little bit confusing. Better to use notations with two subscripts (including box number  $I$  and time).

A.  $C_{\text{box}, t1}$ ,  $C_{\text{box}, t0}$  were used in the equation.

Line 347-354, Page 14

$[^{137}\text{C inventory}_{\text{box}, t1}] = [^{137}\text{Cs inventory}_{\text{box}, t0}] - [^{137}\text{Cs inventory}_{\text{box}, t0} \times \exp(-0.693/T_{1/2} \times \Delta t)] - [\text{net outflow to the downstream box of } C_{I_{\text{box}, t0}}] - [\text{downwards transport of } ^{137}\text{Cs inventory}_{\text{box}, t0} \text{ below the mixed layer}]$  (8)

where  $^{137}\text{Cs inventory}_{\text{box}, t0}$  is the  $^{137}\text{Cs}$  inventory by using 0.5-yr  $^{137}\text{Cs}$  average value and mixed layer depth in each box in the initial year and  $^{137}\text{Cs inventory}_{\text{box}, t1}$  is the  $^{137}\text{Cs}$  inventory by using 0.5-yr  $^{137}\text{Cs}$  average value in each box after the  $\Delta t$  year. This mass balance was estimated to every 5 years from 1975 to 2015. In the case of 1970, the value of the initial year in each box was  $^{137}\text{Cs}$  deposition

amount until 1970. In fact, distinguishing between net outflow to the downstream box and downwards-transported  $^{137}\text{Cs}$  amounts was very difficult in this study.

- Describe how the “initial year” is defined.

This mass balance was estimated to every 5 years from 1975 to 2015. In the case of 1970, the value of the initial year in each box was  $^{137}\text{Cs}$  deposition amount until 1970. In fact, distinguishing between net outflow to the downstream box and downwards-transported  $^{137}\text{Cs}$  amounts was very difficult in this study.

- To reviewer’s knowledge, the transport is composed of advective and diffusive fluxes. Explain which data were used for the fluxes. If only advective flux was used, elaborate which current (u,v and w) data were used. Furthermore, authors mentioned that distinguishing the horizontal and downward transports were difficult. Explain why. No w velocity?

A. Actually, seawater transport was controlled by ocean current. And seawater speed and direction consists with u,v, and w. However, in this study, I did not analysis of ocean current, because of the data is discrete. I mean that “the net outflow to the downstream box“ correspond to u and v component, and “downward transport below the surface sea water” correspond to w component. “The net outflow to the downstream box” is controlled by advective and diffusive fluxes.

In the marine environment,  $^{137}\text{Cs}$  activity concentrations after 1970 were dominantly controlled by radioactive decay and physical ocean processes, such as horizontal, which mean net outflow to the downstream box for sea water current U and V component, and downwards transport below the surface mixed layer for seawater current W component, except for the contribution from accidental release (the Chernobyl accident in 1986 and the Fukushima accident in 2011) and direct discharge from nuclear reprocessing plants.

### 3. Results

- Line 308, Pg. 13:

Authors state that correlation coefficient is between 0.51 and 1.0. It appears that the range is very large. Elaborate why. And clarify the correlation coefficient between what?

A. Additional data against to the previous analysis were caused to the difference of the average data. However, this is not an essential issue in this manuscript. I delete this description.

● Lines 561-562, Pg. 28:

Describe why the model results are considerably different from the estimate by Aoyama et al. (2006) and this study.

A. However, these estimations are almost 1.4 times larger than those in the estimation by using a model simulation (UNSCEAR, 1993), with an estimated value of 545 PBq (Aoyama, 2019). The large difference in the meridional distribution in the mid-latitude. These corresponds to have larger  $^{137}\text{Cs}$  fallout region, where Kuroshio Current and its extension areas (latitude 20-40°N) in the Pacific Ocean and Gulf stream transport area (latitude 30-50°N) in the Atlantic Ocean. It was also reported that the  $^{137}\text{Cs}$  water column inventory in the North Pacific Ocean was 2-3 times larger than those in the cumulative  $^{137}\text{Cs}$  fallout amount in the same latitude in the modelling results in UNSCEAR (1993) (Aoyama, 2019). Because reconstructed  $^{137}\text{Cs}$  deposition in Aoyama et al. (2006) was based on the historical observed data, uncertainty of model would cause the underestimation of  $^{137}\text{Cs}$  deposition amount.

● Lines 630-631, Pg. 30:

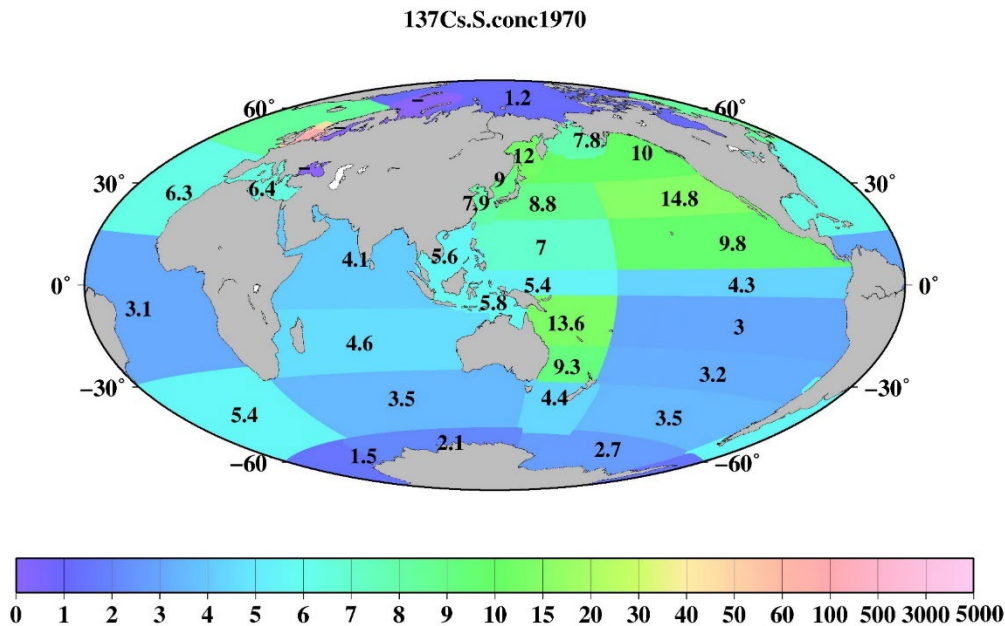
Better to use 0.0 PBq instead of 0 PBq.

A. The  $^{137}\text{Cs}$  deposition amount is the lowest in the Pacific sector (0.05 PBq), Atlantic sector (0.0 PBq), and Indian sector (0.0 PBq) of the Antarctic Ocean.

● Lines 770, 785, Pg. 37:

Hard to see the max. value of scale bars.

These Figures modified as follows:



- Lines 923-924, Pg. 48:

The mixed layer depth can have strong seasonal variability in some marginal seas where vertically well mixed in winter while stratified in summer. Elaborate how authors deal with.

A. The monthly mixed layer depth shows seasonal variation that is deeper in winter and shallower in summer. It is recognized that sea water subducted from the ocean surface in the mode water formation region associated with the winter convective mixing because of the lower buoyancy from the ocean surface (Hanawa and Tally, 2001). The flow through the winter mixed layer ventilate the sea water into the ocean interior. The maximum monthly mixed layer depth in each box was used to calculate the <sup>137</sup>Cs inventory in the mixed layer. In fact, <sup>137</sup>Cs activity concentrations were increased after 1-2 years at the FINPS accident in the Eastern China Sea and the Japan Sea. It appeared that <sup>137</sup>Cs released into the FINPS accident deposited in the subtropical mode water formation region in the North Pacific Ocean, and then subducted into the subsurface layer, following then transported westward, and reached to the bottom of the Eastern China Sea. The FINPS derived <sup>137</sup>Cs were upward transport into the surface seawater and then transported into the Japan Sea (Inomata et al.,

2018a,b). Therefore, maximum mixed layer depth associated with the mode water formation was used in this study.

The mixed layer depth was the monthly time interval with seasonal variation that is deeper in winter and shallower in summer. It is recognised that sea water subducted from the ocean surface in the mode water formation region associated with the winter convective mixing because of the lower buoyancy from the ocean surface (Hanawa and Tally, 2001). The flow through the winter mixed layer ventilate the sea water into the ocean interior. The maximum monthly mixed layer depth in each box, mainly winter month, was used to calculate the  $^{137}\text{Cs}$  inventory in the mixed layer.

- Lines 984-986, Pg. 51:

Authors state that the  $^{137}\text{Cs}$  discharged into the Irish Sea.1 was transported into the Irish Sea.2, followed by transport to the northern North Atlantic Ocean.1, North Sea, and Barents Sea and coast of Norway. Reviewer is interested in its transport direction in Irish Sea. To reviewer's knowledge, simulation by Prandle (1983) showed that Cs137 moved to the north channel of Irish Sea. How about in this study? Discuss this matter.

- A. Actually Irish Sea.2 area include the north channel of Irish Sea. Also this  $^{137}\text{Cs}$  transport pattern analyzed in this study is consistent with the general circulation of seawater transport by Bois et al. (2020). Unfortunately, I did not find the paper, Prandle (1983).

This pattern is also consistent with general pattern of seawater transport in this region (Bois et al., 2020).

#### 4. Discussion

- Line 1259, Pg. 67:

Hard to understand the expression “outflowed  $^{137}\text{Cs}$  is larger than the flow of  $^{137}\text{Cs}$ ”. Improvement in writing is required.

Line 933-949

Fig. 15 shows the spatiotemporal variations in the  $^{137}\text{Cs}$  density in the surface mixed layer in the North Pacific Ocean, subtropical North Pacific Ocean, equatorial Pacific Ocean, and subtropical western South Pacific Ocean. In the western North Pacific Ocean, except for the highest  $^{137}\text{Cs}$  density in 1960,  $^{137}\text{Cs}$  density increased and reached to 1964, and then decreases exponentially. However, in

the eastern North Pacific Ocean, the  $^{137}\text{Cs}$  density increased until 1966 and then decreased exponentially. The 2 years timelag that reached the maximum value was caused by horizontal transport from the western North Pacific Ocean and accumulated in the eastern North Pacific Ocean (Inomata et al., 2012). In the subtropical western and eastern North Pacific Ocean, and eastern equatorial Pacific Ocean, the  $^{137}\text{Cs}$  density was almost constant in the 1970s and the 1980s. After the 1990s, the  $^{137}\text{Cs}$  density decreased gradually. In the western equatorial Pacific Ocean and western subtropical South Pacific Ocean, the  $^{137}\text{Cs}$  density increased gradually until the 1980s and then decreased after the 1990s. As shown in Table 3, Tap2 in the eastern North Pacific Ocean, which is estimated to be 8.8 years, is shorter than that in the western North Pacific Ocean (16.9 years). This suggests that the outflowed  $^{137}\text{Cs}$  amount in the eastern North Pacific Ocean was larger than the inflowed  $^{137}\text{Cs}$  amount from the western North Pacific Ocean. The Tap2 in the western subtropical North Pacific Ocean is estimated to be 34.1 years and Tpo is estimated to be -260.7 years. This mean that the  $^{137}\text{Cs}$  was accumulated in this region: The seawater with higher  $^{137}\text{Cs}$  activity concentrations moves southwards with subsidence associated with the North Pacific subtropical gyre, followed by westwards transport and subduction in the central and eastern subtropical North Pacific Ocean (Inomata et al.,2012). The increased  $^{137}\text{Cs}$  density in the western equatorial Pacific Ocean and the western subtropical South Pacific Ocean, as shown in Fig. 15, would result in a supply of seawater with higher  $^{137}\text{Cs}$  activity concentrations.

- Lines 1356-1357, Pg. 67:

Regarding the statement “although the contribution of directly discharged  $^{137}\text{Cs}$  from the F1NPS might be included in the values in the western North Pacific Ocean”, comment when it happened. 2011? 2012? That is, how long does it take the directly released  $^{137}\text{Cs}$  moves from Fukushima NPP to the southern sea region of Kuroshio extension?

A. Direct discharge of  $^{137}\text{Cs}$  continued. However, most of discharged  $^{137}\text{Cs}$  occurred from 26March to 6 April, 2011, and then decreased (Tsumune, 2013). Because the atmospheric deposited  $^{137}\text{Cs}$  and in flowed  $^{137}\text{Cs}$  did not distinguished in this analysis, this part was deleted.

Line 1042-1043

These were caused by the atmospheric deposition of  $^{137}\text{Cs}$  derived from the F1NPS accident.

- Lines 1399-1357, Pg. 75:

The expression “Cs137 is transported via advection” is not correct. “via advection” needs to be changed to “via advection and diffusion”. Authors appear to neglect diffusion, right?

A. I added the tem “diffusion”.

The  $^{137}\text{Cs}$  deposited into the ocean surface is transported via advection and diffusion in the surface seawater

● Lines 1406-1408, Pg. 75:

Authors mentioned inflow/outflow, upstream box and downstream box. Reviewer hardly understand how such information can be obtained. Up to now authors had never mentioned current data. Clarify this point.

Positive values (red) indicate that inflowed  $^{137}\text{Cs}$ , whereas negative values (blue) indicate that outflowed  $^{137}\text{Cs}$  in each area.

● Line 1428, Pg. 75:

Authors mention “considering the current system”. What is the source of the current system?

Then, these were transported southwards with subsidence associated with California Current and westwards in the equatorial Pacific Ocean.  $^{137}\text{Cs}$  moved southwards due to subduction in the eastern subtropical North Pacific Ocean and upwelled in the western/eastern equatorial Pacific Ocean.

● Lines 1011-1714, Pg. 94:

What is represented by “1~2 years” in Fig. 22 shown in North Pacific?

A.  $^{137}\text{Cs}$  deposited by the large-scale nuclear weapons tests in the western North Pacific Ocean was transported eastwards within 1-2 years.