General comment

This paper presents interesting data about fate of cesium in sediment from various areas impacted by the Fukushima accidental releases. Observed data are of high quality and it is no doubt that these are very useful and deserve publication. Indeed, these additional data allowed to precise the cesium inventories in the sediment compartment which is fundamental to better assess their potential as delayed cesium sources for the future. However the paper is not always easy to read and the writing has to be improved in order to help the reader in following the authors' reasoning.

Before going into details in specific remarks, it seems important to me to give a general comment that I consider particularly relevant to this paper but also to various papers dealing with cesium in sediment. Due to the fact that the finer the sediment the higher the exchange surface authors generally look for a relation between grain size and cesium concentrations. However in some cases this quest can be a vain one. Indeed, when dealing with diffuse sources such as global fallout there is no doubt that fine sediment are characterized by higher cesium contents. However this is not always true for point sources such as controlled liquid discharges or accidental releases (except of course if areas around these point sources have high silt contents which is indeed the case for example in the eastern Irish sea close to the discharge point of Sellafield). In case of point sources main factors are obviously the distance or for riverine installations also the time of the discharges (low flow vs high flow rates), the dispersion patterns of the releases, lateral transport etc...

Specific remarks

Introduction

7236 line 18-19 : TEPCO has reported some of the highest 134Cs sediment activity (2000 Bq kg-1, NRA, 2014a) *when where? This number does not seem so high! there are indeed a number of data above 2000 that have been reported by TEPCO, please check and also mention the date/location of the sampling.*

7239 line 8," If conditions are relatively stable, the 210Pbex in a given area will represent the flux to this location averaged over the last century (_ 5 half-lives)"

I think it is 210Pbxs <u>inventory</u>

Most of the section dealing with 210Pbex should be move to the methods section, in the introduction you should only mention that in order to help in evaluating the rates of both sedimentation (<u>but nothing on this point is reported in the paper …)</u> and mixing, two natural-occurring radionuclides have been studied.

2Methods

2.1 **2.1 Sample collection**

7241: *It would be useful to have the core section.*

It is mentioned that a multi-corer has been used. Did you pool several tubes for getting one sample?

7240 > 2.2 Grain size

I understand that for simplification in the presentation of the data, authors have averaged the grain size over the entire cores though measurements have been performed on each layer. Therefore why assume to have a 5% uncertainty when most of the time due to large fluctuations in layer to layer analyses the standard deviation is far greater than this. I strongly recommend to give the standard deviations in table 1.

I have to confess that I get lost with all the different kind of uncertainties the reader have to integrate when reading the paper…

2.3 Isotope measurements

Please indicate clearly the unit for the results Bq kg-1 dry? (no indication in table or figure legends as well)

7242 line 5: where layer thickness was in meters, activities were in Bq kg−1 *(I guess it is dry weight, please clarify)*

7242> 2.4 Modeling

In this part arise a confusion which appears even more clearly in the section 3.7 where, <u>if I am correct</u>, the authors consider that Db represents mixing due to bioturbation processes only when Db represent also other processes such as physical resuspension etc.. This latter process is certainly quite important in NCZ and SCZ and cannot be rule out especially since the areas (1) have been subject to a strong tsunami and we do not know if sediments have been consolidated leading then likely to easier re-suspension phenomena and (2) are regularly under influence of typhoons.

3 Results

3.1

7244 line 5 “We could not obtain … which suggest that the factors controlling local inventories may vary for the two isotopes”

Suggests and may seem inappropriate: replace suggests by confirms and delete may. There is no reason to have a good correlation between the two isotopes, their sources and entry route in the environment are completely different here.

7244 line *Please clarify sentence and redraft* :Overall… considered: greater than *what do the authors mean? High variability in both activity and inventory? greater than what? 210Pb, if it is the case there is no interest in doing so (various sources for the two isotopes etc..)*

Line 11 *Please clarify sentence* :The replicate cores… across all samples relative to the order of magnitude differences seen in cesium activities??

7244 Lines 23-25 *Last sentence of this part:*

Although particle size characteristics may not control local differences in radionuclide activities <u>where variations in size are comparatively minimal</u> *(not clear ?)*, they are important over large regional scales: *just because when looking at a larger scale there is a smoothing in the influence of factors with high variability that occur in coastal areas*

3.2

7245 lines 5 to 8 *In the coordinates change S to N*

7245 line 25The remaining cores were visually assess*? Please precise what you mean exactly .. this is far better explained in table 1 legend*

7245 line 27 These cores had the lowest average standard deviation *(see comment on 2.2 it will be useful to have theses standard deviation values for D50 and % clay, silts mentioned in table 1)*

Redundancy between lines 10-11 (…. And relatively consistent ..) and lines15-16 (D50 values remained fairly consistent..)

3.4

This part is quite difficult to read mainly because authors mentioned data not shown and I did not succeed in calculating them i.e.

7246 line 26 Average surface activities (top 3 cm) in the 20 cores ranged from 2.1±0.1 to 640±40 Bq kg−1 for 137Cs and from 0 to 550±30 Bq kg−1 for 134Cs (Supplement S6).*We have no mean to check these numbers which I guess were calculated taking into account activities and weight of each layer over the first 3cm). In addition it would help the reader to mention in which core the min and max are found. In the table S6 (which is very useful) give the layer thickness in cm rather than in m! and precise for the Bq kg-1 and m-2 if it is wet or dry weight*

In addition when calculating average ± should be standard deviation?

7246 line 28 *sentence not clear*: The OZ sediment activities ranged from 0 to 13±1 Bq kg−1 for 134Cs, while the MCZ activities ranged from 2.7±0.5 to 57±3 Bq kg−1 for 134Cs. > *It was impossible to find what exactly these number refer to…Are they still averaged activities over the top 3cm ?*

7247 line 5 : within 3 km of the FDNPP, contained an average of 550±30 Bq kg−1 5 in the top 3 cm *perhaps useful to precise with the highest content in the first centimeter.*

7247 Lines 6-8 With the exception of this single core in the NCZ, the cores in the SCZ had the highest AVERAGE (?) activities of all zones, with 134Cs values ranging from 167±7 to 230±10 Bq kg−1.

If I understand correctly the first $ of 3.4 refer all to the average activities in the top 3cm, please be clear in the writing.

7247 Lines 12-13 The MCZ cores generally showed similar exponentially decreasing *CESIUM* activities with depth with the exception of core 11, which showed a pronounced ~~cesium~~ peak between 1 and 4 cm.

7247 The penetration depth of 134Cs was deeper here, on average, than in the AZ and OZ core. *In order not to mix the various terms average I suggest rewriting as follows Generally, the penetration depth of 134Cs was deeper here, ~~on average,~~ than in the AZ and OZ cores.*

7247 line 17 Penetration depths for 134Cs were at least 16cm in the SCZ. *This sentence is not very informative, it would be more useful to say that the entire thickness of the sampled layer was labelled with 134Cs in both SCZ and NCZ. Perhaps useful also to comment on the vertical profile in core 14 compared to cores 12 and 13 due to its higher contents in sand (higher mean D50 and lower % silt and clay though these latter data are averaged so authors have to check if it is really the case)*

3.5 Cesium inventories

7247 Line 25 : Total 134Cs and 137Cs inventories ranged from 0 to 74000±2000 Bqm−2 (COR XX) and 21±1 to 73000±2000 Bqm−2 (CORE YY), respectively (Table 1). *In order to help the reader please indicate the core number where min and max are found.*

7248 Line 13-16: We observed inventories consistent with weapons testing fallout in the AZ and OZ cores (134Cs/137Cs of 0 to 0.86). Larger inventories and 134Cs/137Cs ratios of 15 _ 1 in most of the MCZ, NCZ, and SCZ cores suggested negligible contributions from weapons testing 137Cs.

I suggest to modify as follows :
We observed inventories consistent with weapons testing fallout in the AZ and OZ (134Cs/137Cs = 0). Larger inventories and 134Cs/137Cs ratios of ~1 in most of the MCZ, NCZ, and SCZ cores suggested negligible contributions from weapons testing 137Cs.
Indeed there is no more 134Cs in global fallout, cores 5 and 6 are closer to the MCZ and represent a kind of transition inventories between the two cases i.e. only weapons testing and mainly Fukushima influence.

7248 Line 22 /The percentages for <u>those</u> (?) cores in the MCZ ranged from 0 to 33% for 134Cs and from 10 to 36% for 137Cs

7248 Line 23: The average inventory below 3 cm in the MCZ cores attributed to Fukushima (134Cs) was 15±16 %, which agreed closely with the Otosaka and Kato 25 (2014) 134Cs average from this zone of 19%. When we combined the two datasets for this zone (*n* = 15) the average inventory below 3 cm was 18±16 %.
I suggest authors underlined the very high variability in inventories in MCZ

7249 lines 3-6: Figure 3a…. despite that the **<u>remaining core inventories</u>** …. I am getting lost, *what do the authors mean?*

7249 *from line 10 to 20 I have the feeling that one important factor is missing i.e. lateral transport, this is an important factor in coastal zones especially with a point source; and indeed the authors mention this in the section 3.6 when dealing with 210Pb activities (lines 8-10 p7250)*

7255 line 10-20 *I am not really convinced by the main influence of grain size. Don't you think that this reflects a mixing of various factors such as grain size, dispersion of the releases in coastal areas, lateral transport…*

3.6 210Pbex and 234Thex activities and inventories
7249 line 21 : 210Pbex <u>surface</u> activities ranged from 12±3 <u>(core XX)</u> to 2000±100 Bq kg−1 <u>(core YY),</u> *indicate the cores concerned to help the reader, in addition clarify the word surface, is it 0-1, 0-3cm? are they averages?*

Does the authors have an explanation for 210Pbxs profile in 4-0Z which is a deep location (3259m)?

7250 line 3 : 210Pbex inventories, ranging from 2700±200 (<u>core XX</u>) to 28000±1000 Bqm−2 (<u>core YY)</u>
Add underlined information

7250 Line 3-5: 210Pbex inventories, ranging from 2700±200 to 28000±1000 Bqm−2, reflect changes in grain size, water depth and local processes, and give support to the similar trends observed for cesium inventories (Table 1, Figs. 1 and 3b)
There is no reason that the two isotopes (Cs and Pb) have the same trends since their sources are completely different.

Lines 13-14: An exponential regression of 210Pbex inventories vs. percent clay indicated a strong relationship ($R^2 > 0.9$) between grain size and inventories. *Right and this confirms the previous remark.*

7250 line 24-25: 234Thex activities, ranging from 20±10 (core XX) to 1300±100 Bq kg−1 (core YY) in the top 0 to 0.5 cm (Fig. 2)
Surface (0 to 3 cm) 234Thex inventories peaked at 2400±300 Bqm−2 in the SCZ (core ZZ).
Add underlined information

Lines 1-2: "intra-zonal variability was high and more often than not inventories did not vary negatively with increasing depth". *I suggest to replace vary negatively by decrease?*

$3.7 Bioturbation estimates
See remark on § 2.4, this is especially true for coastal zones such as NCZ and SCZ where profiles show strong mixing processes that can be related to both bioturbation and physical reworking by currents in areas often affected by typhoons and storms. And indeed the authors mentioned this factor lines 1-5 p7252 but p7251 lines 11 they wrote that "Mixing rates in the SCZ and NCZ reflected intense bioturbation, with full core 210Pbex-derived estimates starting at 11 cm2 yr−1 and the majority of rates being unquantifiable due to the vertical 210Pbex Profiles ».

Lines 16-17 reworking by physical factors also decrease with water depth….

Clarify this section and others in this respect.
In addition I am not sure that mixing rates derived for SCZ are correct due to the shape in 210Pbxs vertical profiles…and therefore calculations in 3.8 may not be correct

7252 3.8 Cesium modelling
Line 20 add input or release after Fukushima maximum

7253 3.9 Inventories
comment: it is always very difficult to assess inventories especially with data with such high variability but this allows to give orders of magnitude.

7253 *Line 9 add* ***about*** *after contained*

7253 line 6-7 "Because of the inventory variability and grain size influence in the NCZ and SCZ" *I do not see the idea behind…*
I get lost in this section line 17 it is said "we used cores recovered in February 2012 by MEXT (Kusakabe et al., 2013)" when I thought that cores reported by Kusakabe were in group (4) and cores recovered in February only were in group(3), please clarify in order to help the reader. *It would be helpful to give the column number, I guess that here we are in the second set of columns of table 3b with data from (1),(2) and(3). May be also useful to give in*

the table the total number of data used (if I am correct n=18 for the first set, n=50 for the second and n=199 for the third
Clarify also with fig 5 in this respect MEXT (3)=? What about (4)? May be there is no need to mention (1) (2) and (3) in fig 5 if they do not mean the same than in table 3 and only mention "This study, OTKA and MEXT"…
Check also the end of section 3.9

7253 Lines 26-27 "The addition of 50 locations slightly increased the sediment inventory estimate to 100±40 TBq", *There is no increase since we had already 100 TBq line 5! May be the reason is that the authors mentioned rounded numbers for the total inventory since if we sum the numbers mentioned in the table the first estimate is 96 and the second one is 104… but the uncertainties are very high and therefore they are not statistically different….*

7254 line 2: *replace Fig 4 by Fig 5*

7254 line 16-17 "The MCZ contained between 15 and 18% of the total 137Cs in each case and made up 30% of the total area". *I suggest rewriting as follows The MCZ represents 30% of the total surface area and contained 15 to 18% of the total 137Cs inventory.*

7254 line 28 "…and consistently showed the greatest variability in inventories".
I do not understand… in table 3b standard deviations (I guess it is 1 sigma standard deviation? Please precise in the legend) to the mean are not higher for SCZ and NCZ compared to the other zones in % of the inventories, for example in the first column for OZ 160 (the standard deviation) represents 94% of the mean when for SCZ 13 000 represents 81% of the mean….

Fig. 5 and 6, *why the 199 data mentioned for calculating the third set of inventories (table 3b) were not taken into account here ? can you explain.*

7255 line 9-10 "… which we suggest reflects the importance of grain size distribution in these zones (Fig. 3a). *see previous comment.*

The compiled inventories from 150 to 1500m showed a stronger relationship to water depth (R^2 =0.30) *Is this number statistically significant?*

7256 line 2*: <u>robust</u> is perhaps too strong due to the very high variability encountered in each zone.*

7257 line 3: *mixing rates*

7257 lines 5-10 If riverine inputs appear to be important at least locally what do you think about the decay correction of the data to 6th April?