Author response to reviews in *italics* for:

Radium-based estimates of cesium isotope transport and total direct ocean discharges from the Fukushima Nuclear Power Plant accident
Author(s): M.A. Charette et al.
MS No.: bg-2012-512
MS Type: Research Article
Special Issue: Impacts of the Fukushima nuclear power plant discharges on the ocean

Anonymous Referee #1

Received and published: 10 December 2012

The paper presents an interesting estimation of the offshore and vertical 134Cs fluxes from the Fukushima Nuclear Power Plant that followed the earthquake and resulting tsnunami of 11 March 2011. The radium-based methodology is appropriate to attain the objectives of the study, allowing to estimating water apparent ages as well. The manuscript is well written and structured and the covered issue is of undoubtedly relevance within the scope of BG. However, there are some general and specific comments that should be addressed before publication.

The major comment is that basic information on sampled stations and the obtained radium concentrations (not available in the cited BCO-DMO website) are missing, while there are crucial for the good comprehension of the paper and the traceability of the results. Radium isotopic activities should be explicitly given in the paper. In addition: where were the radium profiles been collected? which are the dates of sample collection? which are the surface samples processed with MnO2 acrylic fibers and those processed with cartridges?...

We have added two new data tables to the paper per the reviewer's request.

Specific comments

P16142, L10: "Radium samples" should we replaced by "Seawater samples for radium analysis"

Fixed.

P16142, L11: Figures 1a, b and c are not large enough to correctly see the stations.

We have separated Fig. 1a from b and c so that it may be seen at a larger scale.

P16143, L21: Results section. Radium concentrations for the four isotopes (at least three, if Ra-224 is below detection limit) and all the stations should definitely be included in this section in a table. The authors do not even mention 223Ra data here while they use them to estimate the

water apparent age.

We have added a new table and reference the 223Ra data in the results section per this request.

P16144, L14-15: Could the authors give a more detailed description of the detected eddy at station 29? The paper by Rypina et al., 2012 is only submitted and the eddy structure is an important feature for the interpretation of the results.

See comment below regarding L20-23 on this page.

P16144, L19-20: I would rephrase the sentence "While 228Ra/226Ra is expected to decrease with distance from shore, we found many exceptions to this rule". This is only true when diffusion dominates advection and this is clearly not the case in the present study.

We re-wrote this sentence as: "While $^{228}Ra/^{226}Ra$ is expected to decrease with distance from shore when cross-shelf mixing is dominated by diffusion, we found many exceptions to this rule suggesting that offshore transport was impacted by advection in some areas"

P16144, L20-23: Still concerning the eddy, according to the coordinates of the semipermanent eddy detected by surface drifter data (Buesseler et al., 2012), it is centered on 37°N 142.5°E and thus, closer to station 31 than station 29. If higher radium activity ratios trace larger proportions of coastal (vs open) seawater, could the authors give any explanation of why AR close to the center of the eddy (i.e. station31) are remarkably lower than in st. 29? Could it exist any other justification for the high AR in st. 29?

We added new text to the end of the results section and beginning of the discussion section to further explain the sharp gradients in Ra and Cs within the eddy. Please see the track changes version of the manuscript for details.

P16145, L9-13: The authors invoke the shoreline as the radium source but which are the processes governing the radium input to the water column? Is the dissolution of sedimented material? Are there porewaters playing a role? What about potential groundwater discharge in the region? I would recommend to better constraining their shoreline argument, what allows using the crustal average 223Ra/228Ra ratio to derive water ages.

Unfortunately we do not have Ra data from potential shoreline-based or benthic endmembers (nor is there any in the literature for the short-lived Ra isotopes to our knowledge). However, the method does not require that we know the precise source of the Ra, just its isotopic ratio. On P. 16147 line 5 we discuss the sensitivity of the method to a different assumed endmember ratio.

P16146, L20-P16147, L6: Comparison between Ra-based and numerically simulated water ages should be done more cautiously since when considering associated uncertainties, the two averaged ages are not different, with a Ra-based estimate that could be either higher or lower than the oceanographic water age.

See the above comment on the sensitivity analysis of the Ra-age model. Furthermore, in calculating the Cs isotope flux from FNPP direct discharge (eqn. 4) we use multiple methods to constrain the flux. Hence, we are taking a cautious approach in not simply applying the water age derived from a single method.

P16147, L13-P16148, L2: I found a bit surprising to estimate the vertical Cs flux due to diapycnal mixing, which is driven by intermittent patches of small scale turbulence, through an approach that only accounts for diffusion. Could the authors use a method similar to that in Li and Cai (2011) to estimate both (diffusion and advection) terms? (Li, C. and Cai W.-J., 2011. On the calculation of eddy diffusivity in the shelf water from radium isotopes: High sensitivity to advection. Journal of Marine Systems 86, 28–33.

The importance of this calculation was is not so much the magnitude of the vertical flux, but rather the relative importance of the vertical flux relative to the horizontal flux. A more sophisticated mixing model (using data with limited vertical spatial resolution) would not change our conclusion that the horizontal flux was orders of magnitude more important for FNPP Cs dilution in the coastal ocean.

P16148, L14: Are the authors referring to "228Ra data" or "228Ra/226Ra data"? Actually, the reader cannot reproduce the estimation of the Kz (0.7 m2d-1) with the data supplied in the manuscript.

Fixed. The addition of the data table should allow future readers to reproduce these mixing rates.

P16149, L27: The term "It" should be replaced by "Ii" according to the definitions given in the text. Again, data on collection time is required for the reproduction of the calculations made.

Fixed.

Anonymous Referee #2

Received and published: 31 December 2012

Review of the manuscript of Biogeosciences MS No.: bg-2012-512 Title: Radium- based estimates of cesium isotope transport and total direct ocean discharges from the Fukushima Nuclear Power Plant accident Author(s): M.A. Charette, C.F. Breier, P.B. Henderson, S.M. Pike, I.I. Rypina, S.R. Jayne, and K.O. Buesseler

Major comments: In this manuscript the authors used radium isotopes to help to derive vertical and horizontal transports of Cs from NPP. The results may be used to estimate transports of other radionuclides released from NPP and evaluate the impacts of NPP. From this perspective this

manuscript fits the special issue. However, a few terms have to be clarified before the results are accepted for publication.

There is a mixed use of 'water age', 'mixing rate', and 'replacement time' in this manuscript. 'Water age' was defined as the time the water parcel left NPP, so it is based on Lagrange observation, while 'replacement time' and 'mixing rate' are terms based on Euler observation.

We have checked the manuscript for inconsistencies in the terminology used; there were two occurrences where we made changes (noted in the annotated version of the paper).

Water age is reflective of replacement time or mixing rate. But they may not be equivalent in values to each other. In terms of the water age, the correlation between the numerical model age and the Ra water age was not good enough to state they are consistent with each other. On Page 11 Line 17, the authors stated that the oceanographic numerical model also suggests a good correlation (Fig. 6) between the Ra-based water age and Cs age. But Fig. 6 shows that at 9 out of 26 stations there are big differences (greater than 10 days, about 1/3 of the average ages) between the two ages. Therefore, uncertainties for the ages and fluxes should be provided to help evaluate the consistency of the two ages.

The uncertainty on the individual age estimates (Ra-based) and the range of ages from the oceanographic model are reported in Figure 4. We have included these uncertainties in our new data table as requested by Reviewer #1.

Minor comments: On Page 2 Line 22, "FNPP" is not consistent with what was defined on the same page Line 5. On Page 3 Line 17, Why was 'NPPs' used here, not 'NPP'?

Fixed.

On Page 4 Line 15, 'pump from 0.5 m' should be 'pump from a water depth of 0.5 m'.

Fixed.

On Page 6 Line 4, 'FNPP' should be 'NPP';

Fixed.

Line 6, '. . .was roughly the same. . .' should be changed to '...were roughly the same...';

Fixed.

Line 9, 'The outer box stations were significantly lower ranging from <2-333 Bq m-3' is suggested to change to 'The outer box stations had significantly lower Cs activities ranging <2-333 Bq m-3'.

Fixed.

On Page 10 Line 9, 'station' should be capitalized;

Fixed.

Line 20, the first ‘flux’ should be removed.

Fixed.

On Page 12 Line 25, ‘of’ should be added before ‘the local. . .’.

Fixed.

On Page 13 Line 10, ‘, which have proven useful. . .’ should be changed to ‘, which have been proven useful. . .’.

Fixed.

On Page 17, texts in Fig. 1A are too small to read, especially the station IDs.

As noted above, this figure has been enlarged.

On Pages 20 & 22, there is no station with an ID of non-integer as shown in Fig. 1A, but on the x-axis there are some non-integer Station IDs.

These were too large to fit within the station circles. We have annotated the figure caption to indicate the location of the “Half”stations (they follow immediately the integer stations.

Please correct it or better explain it. On Page 21, the peaks in 228/226Ra ratio and 134Cs (or the lower values near the surface) were not explained in the text.

We added the following text to the discussion: “Slighly higher ^{134}Cs and $^{228}Ra/^{226}Ra$ just below the mixed layer at 35 m may be the remnants of a slightly older, coastal-seawater rich water mass.”