Anonymous Referee #1

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1 Summary

Tsumune et al. combine measurements of 137-Cs off Fukushima, Japan with circulation models to derive estimates of 131-I, 134-Cs and 137-Cs discharged accidentally to the ocean from the Daiichi nuclear power plant during the period March 2011 to March 2012. Their main result is that the direct release from the plant to the sea (as opposed to that contamination that entered the ocean via rivers, groundwater inflow or atmospheric deposition) amounts to 11.1 2.2 pBq 131I, 3.5 ± 0.7 pBq 134Cs, and 3.6 ± 0:7 pBq 137Cs.

2 Evaluation

The paper is, in its present form, not suitable for publication in Biogeoscience. My concern is that the method used to derive the discharge estimates is flawed or, alternatively, that the errors associated to the respective estimates do not reflect the real uncertainty.

We are grateful to referee #1 for valuable comments. We answered them as follows.

3 Comments/concerns

The authors assume that there is a linear relationship between the evolution of observed 137-Cs activities at the surface ocean close to the nuclear power plant and the temporal evolution of the discharge of 137-Cs. Based on this assumption they construct an emission scenario of 137-Cs (Fig. 2), apparently by fitting a rather arbitrary curve (ln. 14 - 17 states the already scaled functions) through the temporal evolution of observed activities. In a second step the amplitude of the emission scenario is scaled such that the simulated activities are similar to the observations (Fig. 9). The emissions of 134-Cs and 131-I are derived by multiplying the 137-Cs estimate with conversion factors based on measured 134-Cs/137-Cs activities ratios in the seawater after the discharge and based on the 131-I/137-Cs activity ratio measured in a puddle of water, respectively. In the latter case the conversion factor decreases with time because 131-I decays much quicker than 137-Cs.

We used ^{134}Cs/^{137}Cs and ^{131}I/^{137}Cs activity ratio at 26 March 2011 in a puddle of water. After that we considered the decay of ^{134}Cs and ^{131}I for this estimation as we shown in

Fig. 3 (Fig. 2 in the original version).

I find the approach for a number of reasons problematic:
The authors implicitly assume a linear relationship between observed 137-activities at the "5-6 and south discharge canals near the 1F NPP site" throughout the emission period March 2011 to March 2012. This, in turn, implies that the flushing rate (or water renewal) at the site is constant with time. Given the complicated nature of near-shore (tidal and wind driven) dynamics in general this seems unlikely to hold.

We did not assume that the flushing rate at the site is constant with time. Because we determined the release rate to be fitted to temporally average observed ^{137}Cs activity, we assume that the "average" flushing rate at the site is constant with time; the assumption is supported by Kanda (2013). As we mentioned in 3.5.1 (3.5.1(1) in the original version), "there were two high-activity peaks on 26 March and 7 April 2011, during which time release rates of ^{137}Cs were assumed to be constant in the simulation. The changes of ^{137}Cs activity were caused by changes in the coastal current system." Our simulated ^{137}Cs activities changed due to the variability of current because we consider the nature of near-shore wind driven dynamics (tidal is negligible in front of Fukushima for this simulation).

The scaling of the emission scenario (i.e., the multiplication of the qualitative evolution derived from Fig.2 with a factor such that the simulated concentrations appear similar to the observations shown in Fig. 9) implicitly assumes that the flushing rate or water renewal at the "5-6 and south discharge canals near the 1F NPP site" is realistically simulated by the model. If the simulated circulation were too sluggish (vivid), consequently, their discharge estimates would be biased low (high). Hence an evaluation of the model circulation at the "5-6 and south discharge canals near the 1F NPP site" is key. However, such an analysis is included.

It is impossible to quantitatively validate the simulated mass-flux divergence near 1FNPP at the timing of the accident because it requires comprehensive field measurements. We added the description in 3.5, "We obtained the direct release rate of ^{137}Cs based on the simulation and measured data near 1F NPP. We simulated ^{137}Cs activities in a regional ocean with considering direct release, atmospheric deposition and inflow. Estimated direct release rate was constrained by the measured ^{137}Cs activity near 1F (5,6 and south discharge canal). Here we compared simulated ^{137}Cs

activities with other measured data mainly to validate our estimation of direct release for 1 year in the wider domain than Tsumune et al. (2012).” We acknowledge that our estimate has uncertainty; such model bias can be reduced by model intercomparison study. This is out of scope of this study; we are planning to address this issue in future study.

There is a 2-3 order of magnitude scatter in the early on observed activities which are used to constrain the discharge (Fig. 2 and Fig. 9). How could that transfer to an error of only 20% in the 3.6 ± 0.7 pBq 137Cs discharge estimate?

We described the method in previous paper (Tsumune et al., 2012). The errors were obtained from daily averaged data in Figure 9 (S-Fig. 6 in the original version). Magnitude of scatter in the early period is about 1 order, not 2-3 order. Daily averaged simulated results represented 1 order of magnitude scatter as shown in Figure 9 (S-Fig. 6 in the original version).

The 131-I estimate was derived by scaling the 137-Cs estimate assuming that on 26 March 2011 the 131-I/137-Cs ratio was 5.7 because this ratio was measured "in a puddle of water in the basement of the 1F NPP reactor 2 turbine building". I am not an expert. But I expect that there is considerable uncertainty as concerns the question how this ratio can be applied to derive the 131-I discharge from the 137-Cs estimate

We added description in 3.2
“The $^{131}I/^{137}Cs$ activity ratio should not change during direct release and the oceanic transport because ^{131}I and ^{137}Cs are dissolved form in the ocean water and they have weak interaction with biogenic particles.”