Does the Fukushima NPP disaster affect the Caesium activity of North Atlantic Ocean fish?

bg-2012-649

(07 June 2013)

Final Response to comments given by reviewer Dr. Pröhl on 03 May 2013

We assume that Dr. Pröhl is the one who commented earlier as "reviewer #2" on 14 Apr 2013.

We thank Dr. Gerhard Pröhl very much for careful reading and appreciate very much the helpful and important comments und many suggestions how to improve our paper.

Many of the comments given by Dr. Pröhl had already been considered for the minor revision in February 2013 (see below, following 'Previous changes'), as noted by Dr. Nies, while a few points have been re-considered here by us as described in the following.

The question of how many layers of boxes (and their thickness) we have used within boxmodel calculations has been clarified for the minor revision. However, For the Baltic Sea case, we have explicitly specified now in section 2.3, Page 7, L 13 ff, that we have used the surface-layer as well as the bottom-layer of the three boxes, in total 6 box layers, for averaging the modeled Cs concentrations seawater. The layer thicknesses have been specified there. The modified text reads now:

Therefore, each of the three water boxes included a surface and a bottom layer; within the model, the surface layer thickness is about one third of the total depth. For calculating the activities in fish, the modeled activity concentrations in seawater were averaged over the selected six box layers by weighting with their associated box-layer volumes.

Our "significance factor" SF used for gamma-ray spectrometry purposes has been clarified in section 3.1, Page 8 L 8. This helps preventing from being mixed up with an otherwise common abbreviation "SF" for "Sellafield".

In the last sentence of section of the first paragraph in section 3.3 the word "decay" was replaced by the word "decrease".

The ¹³⁴Cs contribution will decrease from 2012 onwards (Fig. 3).

The last sentence of section 2.3 has been modified again as follows:

This is caused by the compartment model's specific property to instantaneously perfectly mix the analyte within the overall box volume. This means in the case of the Greenland boxes that complete mixing is assumed over a large depth of 330 m in the surface layer, while the surface boxes are shallower in the Baltic Sea. In reality one would expect a decreasing vertical activity concentration profile after fresh deposition on the seawater surface.

The proposal by Dr. Pröhl to include the dose contribution by intake of the naturallyoccurring ²¹⁰Po in the discussion of dose in section 3.3 had been taken into account already for the previous minor revision (see below, after 'Previous changes'). On pages 5 and 6 we changed or extended small parts of sentences (underlined in the following), just for clarification:

... feeding on smaller fish (food fish).

A direct uptake of Cs from seawater has been included according to Brown et al. (2006).

... linked to the physical half-live of the radionuclide and the biological half-lives <u>of food and fish</u>, respectively;

Missing explanation added: $k_{\rm F}$ denotes the predating fish's rate of ingesting food (g⁻¹ g⁻¹ d⁻¹).

Previous changes

Minor changes, 04 Mar 2013:

page 3, line 17: a comma missing after "respectively" page 6, line 15: Please replace "HELCOM, 2012." By "HELCOM (2012).". page 7, line 4: Please replace " ... has been included Brown et al. (2006)." By "... has been included according to Brown et al. (2006)." page 11, line 12: Please replace "un-realistic" by "unrealistic" page 19, line 6: Please remove the blank line page 19, line 20: Please replace "HELCOM" by " Helsinki Commission (HELCOM)" Caption to Figure 1: Please replace "red fish" by "redfish"

First stage of review: response to reviewers, 09 Feb 2013

We thank the reviewers very much for their careful reading and appreciate very much their helpful and important comments und many suggestions how to improve our paper. We therefore have added some more text now and re-defined the sub-model for Cs uptake by fish and parameters used therein.

"massic activity":

This term, designating "activity per unit mass", was taken from the metrology field. For example, NIST and IAEA are using it since long within reports on results of intercomparison exercises. However, the German metrology institute (PTB) recommended us now to use "activity concentration", even if this related to mass. So, we replaced this term as recommended by PTB throughout the whole paper.

Response to Reviewer 1:

The **counting durations** of the samples measured were partly about 1 week including the 2010 samples and about 2 weeks for the other part of samples, including the 2011 Greenland samples. This was indicated in Table 1.

Although this might be considered as a rather limited data set for concluding that Fukushimaderived fallout is the source, we also rely on model calculations of this fallout deposition, especially Jakobs et al. (2011), which demonstrated that it can be expected just the over the southern part of Greenland.

We therefore have extended the first and the last sentence of the Conclusions section.

Concentration factor model – in time-dependent formulation

We are aware of the fact that Cs is ingested by fish mainly from food and to a much lesser extent directly from seawater; we relied on a series of papers by Rowan & Rasmussen.

Instead, we have used a simple concentration factor model as it is widely used within radioecology. By including a Cs elimination rate constant for the fish, which corresponds to what we have called "biological half-live", we enabled it to cover also the time-dependent case (dynamic in time), which may be required for the "short-time" event of Fukushima fallout deposited to the North Atlantic. The concentration factor is a simple concept which, in the steady state case, integrates the food chain and uptake effects into one single number, by dividing the activity concentration of the fish by the activity concentration in water. While the Cs concentration factor, as published by the report cited as IAEA (2004), is known sufficiently well for fish, the elimination rates (corresponding to the biological half-lives) are less well known for individual species.

According to your presumptions we felt that we should extend the section 2.3 by taking also a 2-compartment model into account, which considers predating fish feeding on food fish. It considers also specific exponential growth of the predator and takes Cs intake directly from seawater into account. We also commented now shortly on the effects of such parameters.

The issue of the biological half-live ("150 days") is now also discussed in section 2.3.

effective half-live of 18.6 years

Your comment is correct; we extend this sentence as follows:

After decay correction with an effective half-live of 18.6 years, by which the 137Cs concentration in the North Atlantic seawater decreases (IAEA, 2005; the box 28 given therein),

Similarly, we will extend the sentence a few lines beneath:

Extrapolating a ¹³⁷Cs seawater time trend for the corresponding water box number 28 taken from IAEA (2005) and using ...

Dose estimation section 3.3

According to your quite valuable remarks, but also those of Reviewer 2, we extended section 3.3 and clarified that we are discussing dose to humans. We added another paragraph in this section about a comparison with doses to humans obtained by ingesting 210Po with consumed fish. We also stated now the origin of the dose conversion factors we used. The 40K data we have included in Table 2 for completeness will not be discussed further.

Response to Reviewer 2:

First point:

Our investigation, also in the Greenland Current area, was part of our monitoring program. Although we did not expect direct Fukushima fallout deposited to the North Atlantic as a matter of radiological concern, so did however the public in Germany; at least, we got a lot of questions by the public how Fukushima could affect the North Atlantic and its fish. By doing this routine investigation we clearly did that also in a scientific way. We hope that we put this now clear enough in the first part of the abstract.

Second point: Scientific value – ¹³⁴Cs, an ideal tracer:

We see your point on this generally and discussed this internally.

Due to the small ¹³⁴Cs-actitivity concentrations determined in fish through fallout from FD-NPP, the decay of ¹³⁴Cs and its small decreasing activity concentration caused by mixing inside the water column, it questionable whether 134Cs is really an ideal tracer for monitoring of dispersion processes. However in our case it was possible to determined he vertical mixing of 134Cs in water through the determination of 134Cs in fish. Therefore we added a sentence to the conclusions section addressing this topic.

Specific remarks:

Throughout the paper, the term "massic activity" is used. This sounds unusual. It is proposed to use "activity concentration" or "activity per unit mass" instead.

→ Has been replaced.

Page 1, L 5/6/7

This sentence is not clear. Why was only in late 2011 the Fukushima fallout from March/ April relevant for the Cs-134 traces in fish?

→ The first sentences of the abstract have been re-written, thereby also removing this point.

Page 5, L 5:

The term "ICES" should be defined.

 \rightarrow This has been explained now in the small section 2.1.

Page 5, L 14/20:

Is it possible to provide a reference for the assumed half-life of 100 days? How sensitive are the results to the half-life?

- → The first question has been discussed now in section 2.3, in the second paragraph following the new equation (3): different values between Greenland and Baltic Sea areas.
- → The second is discussed in the third (new) paragraph of section 3.2. We hope this is sufficient.

Page 5 L 20 ff

How many layers are assumed? What is the thickness of these layers? Does it depend on the individual boxes?

- → These first two points have been covered in new paragraphs: section 2.3, third paragraph after Eq. (3) (BS) and in section 3.1, the paragraph beginning with "Dahlgaard (1995)".
- → In the Greenland case there are only small differences between the two layers of the "Irminger Sea" box; in the BS case, the differences of modeled activity concentration are

larger between surface and associated boxes, and also between the surface boxes. This is expressed by the standard deviation curves shown Figures 2 and 3.

Page 6, L 15:

The term "SF" should be defined when it appears first. It is mentioned in the legend of Table 2, however in could also be defined in a footnote on page 6.

→ This has been explained now directly in the first part of the first paragraph of section 3.1. It was already explained within the title text of Table 2.

Page 6, L3-5:

The first part of the last sentence of section 2.3 should be rewritten.

→ This has been re-written at the end of section 2.3.

Page 7, L 15-20: What is behind the "water box from IAEA (2005)"? This should be explained in more detail.

→ We outlined this in a bit more explicit way and hope that it fits.

Page 8, L 17:

"... was **safely** detected ... should be replaced by " was **definitely** detected"

➔ Has been done.

Page 8, L 20/22:

This sentence should be rewritten. How is the "box volume weighted average massic activity calculated? Does it include the activity in different layers? (see above)

→ We have deleted that part of the sentence in section 3.2, second paragraph. We have outlined this section 2.3, in the third paragraph following Eq. (3). Different activity concentrations in different layers are taken into account in theses averages.

Page 8, L 25:

"... caused by " should be replaced by "... caused by the still large contribution of Cs-137 deposited during the Chernobyl accident".

 \rightarrow We corrected that in section 3.2, in the middle of the second paragraph.

Page 9, L23/25

Clarification is needed. Why is this upper limit? Cs-134 will not only decay from 2012. This should be more clearly written.

→ We deleted this sentence concerning the "upper limit" and replaced the word "decay" by "decrease".

Page 9/10: Conclusions

The possible exposures estimated due to intake of fish are extremely low. These exposures should be put in context by providing a comparison with the average exposures from natural sources in general and with the possible exposure due to the intake of the natural radionuclide Po-210 with the ingestion of fish in particular.

It could be discussed whether the Cs-134 can be used as a tracer for estimating the vertical exchange of water.

The dependence of the Cs-134 activity concentration in fish on the depth of the catch as indicated in Figure 3 should be highlighted in the conclusions.

→ We have included such a sentence in the Conclusions and a further sentence on the 134Cs feature as a good tracer for vertical mixing.

Table 3:

The values are given with 3, in some cases even with 4 significant digits. This looks overly accurate.

→ We have changed the number precision to 2 significant decimals.