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

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Abstract

Fillet samples of marine fish collected from the East/West Greenland current (GC) and from the Baltic Sea (BS), have been investigated by gamma-ray spectrometry within the regular German monitoring program. In samples of the second half of 2011 ^{134}Cs traces have been detected, suggested to originate from the Fukushima fallout being deposited in March/April 2011 over the northern North Atlantic and accumulated by fish. The radionuclide ^{134}Cs (half-live 2 yr) was indeed detected with quite small activities at about 0.0036 Bq kg^{-1} w.w. Existing box-models describing the transport of Cs within seawater boxes of the NE Atlantic allowed estimating that ^{134}Cs contributions from other sources, i.e. from the Chernobyl fallout and from discharges by the two major European nuclear reprocessing plants, both were negligible around Greenland, while for the Chernobyl fallout a small ^{134}Cs background contribution to BS fish was estimated. Model results confirmed the level of ^{134}Cs measured in BS fish and showed its maximum to have occurred in winter 2011/2012 followed by a continuous decrease. It was also determined that ^{134}Cs activity, but not that of ^{137}Cs, showed a significant negative correlation with sampling depth (150–400 m) of GC fish; this strengthens our Fukushima fallout assumption. As a result, the Fukushima fallout in these sea areas only marginally enhanced (GC: 4 %; BS: 0.1 %) pre-Fukushima levels of individual dose rates received by human fish consumers; the addition was around 0.001 µSv following the consumption of 10 kg fish per year, which is not expected to cause concern according to present guidelines for radiation protection.

1 Introduction

On 11 March 2011, a Tsunami hit the Fukushima-Daiichi nuclear power plant (FD-NPP), which caused the loss of cooling capacity in four of its six nuclear reactors and led to the release a radionuclides into the environment. It is expected that between 6 and 47 PBq (1 PBq = 10^{15} Bq) of ^{137}Cs (half-live 30.17 yr) was directly discharged into

BGD
10, 4127–4156, 2013

the Pacific Ocean (e.g. Bailly du Bois et al., 2012) in the aftermath of the tragedy. Due to the determined $^{134}Cs:^{137}Cs$ ratio of around one, about the same amount of ^{134}Cs (half-live 2.07 yr) was discharged into the Pacific. Initially, discharge was assumed to represent the larger fraction of total Cs-releases. Therefore, many researchers and TEPCO focussed on the determination and estimation of the behaviour of Cs in Pacific waters and its behaviour in the environment, especially the uptake by biota (e.g. Buesseler et al., 2011, 2012; Honda et al., 2012; Madigan et al., 2012; Behrens et al., 2012).

The explosions of units 1 to 4 of FD-NPP also released radionuclides into the atmosphere which were detectable around the world (e.g. Hsu et al., 2012; Stohl et al., 2012; Jakobs, 2011). From these investigations the wet and dry deposition of caesium isotopes in marine areas where German monitoring was carried out after the accident at FD-NPP occurred, which are the North Sea, the Baltic Sea, and the coastal West and East Greenland currents, were estimated to be between 0.1 and 100 $Bq\,m^{-2}$.

The deposition of Fukushima fallout radionuclides, arriving during the last week in March 2011, of which an average depositions 2 $Bq\,m^{-2}$ for each of ^{134}Cs and ^{137}Cs, respectively, was measured at three locations in Denmark by Risø DTU (Nielsen, 2011) until about begin of May 2011. Furthermore, in Finland, depositions of 1.75 $Bq\,m^{-2}$ for each of ^{134}Cs and ^{137}Cs, respectively, was measured by STUK in Helsinki (Ikäheimonen, 2011) during 21 April and 14 May in 2011 using one week period samples. From Greenland, to the best of our knowledge, no such determinations are available.

Putting FD-NPP deposition estimates and the resulting concentrations measured in Greenland into perspective, they are to be compared with those of the Chernobyl accident in the Baltic Sea, which was characterized by a mean deposition of 12 000 $Bq\,^{137}Cs\,m^{-2}$ and led to an activity concentration between 50 and 5200 $Bq\,m^{-3}$ in seawater and up to 270 $Bq\,^{137}Cs\,kg^{-1}$ wet weight in fish (HELCOM, 1995). The resulting concentration factors of up to 400 imply that fish is the most reasonable test organism for monitoring of the input of caesium isotopes from FD-NPP into different

BGD
10, 4127–4156, 2013

oceans and the separation of FD-NPP from historic Cs-inputs, namely through Global Fallout and the catastrophe at Chernobyl nuclear power plant.

From this point of view, the aims of our study were (i) to determine the activity concentration of caesium isotopes in cod and redfish as economically relevant fish species in the North Atlantic Ocean and its marginal seas, (ii) separate the FD-NPP input from that from Chernobyl and Global Fallout, and (iii) validate the determinations and estimate the future behaviour of caesium isotopes in fish of the Baltic Sea using the first-order kinetics box-model previously developed within a HELCOM Working Group MORS-EG to model Chernobyl ^{137}Cs in seawater and fish of the Baltic Sea.

2 Materials and methods

2.1 Sampling and sample preparation

Biota were caught during research and monitoring cruises using the RV Walther Herwig III carried out between October 2010 and December 2011 using bottom trawls or collected from commercial catches from trawlers operating within ICES (International Council for the Exploration of the Sea) sub-divisions in the Baltic Sea (BS). Sampling positions are indicated in Fig. 1. Round fish samples selected from the catches, consisting of two to 31 individual animals (Table 1), were stored at −20 °C after sampling of 5 kg small fish or 15 kg larger fish. Larger fishes (especially cod) were already filleted on board before freezing. The dry mass of the samples was determined in the laboratory by drying at 110 °C for 2 days. Afterwards, dried samples were dry-ashed at a maximum temperature of 420 °C. The retrieved ash, which was between 50 and 70 g, was homogenized and transferred to 200 mL cylindrical PVC containers for gamma spectrometric analysis, resulting in variable filling heights and bulk densities of about 25–40 mm and 0.44–0.56 g cm^{-3}, respectively. The fish samples selected for this investigation are characterized in Table 1. Fish age was not determined.

BGD
10, 4127–4156, 2013

2.2 Gammaspectrometric analysis

2.2.1 Spectrometry basics

For the analysis of gamma emitting radionuclides three high purity germanium-detectors (2 × *n*-type; 1 × *p*-type) with relative efficiencies between 36 and 55 % and energy resolutions of around 1.9 keV, both at 1.33 MeV. The detectors are protected against environmental radiation by standard low-level lead shieldings of 10 cm lead and inner layers of copper, cadmium and plexi glass. Two Multichannnel analyzer (MCA) modules were used for recording the spectra with 4096 channels. The MCA spectra were transferred into an external spectrum database used by the GamW software for spectrum evaluating which is an in-house development. Some details of the spectrum analysis techniques used in that program are given in Appendix A, in which also some measures for improving the detection of ^{134}Cs are described. Radionuclide activities always refer to wet weight.

The full-energy peak efficiency curves were re-calibrated in January 2011 using a liquid multi-radionuclide standard solution 7503ML (Eckert & Ziegler, Braunschweig, Germany; 11 radionuclides including ^{210}Pb and ^{241}Am) covering the energy range of 46 to 1836 keV; relative standard uncertainties of the activity concentrations were about 1 %. Two-dimensional efficiency curves, depending on gamma-energy and filling height in the container were fitted to the measured efficiency data for four heights between 12–56 mm). As the calculations of true coincidence summing corrections (TCS), which require the knowledge of an efficiency curve as pre-requisite, they were included as part of the extended fitting function used during curve fitting; see Appendix A for more information about TCS corrections.

The standard counting durations for screening on caesium isotopes was 160 h. When there was some evidence about the presence of ^{134}Cs, the measurements of those samples were prolonged to 320–410 h for improving the counting statistics and lowering the detection limit. The ISO 11929 decision threshold was used as a criterion for the detection of ^{134}Cs (see Appendix A for details).

BGD
10, 4127–4156, 2013

2.2.2 Quality control

The stability of the detection efficiencies is controlled by counting every few weeks a ^{152}Eu point source on the detectors. The detector background is measured two times a year by collecting two or three one-week spectra, the peak net counting rates are evaluated statistically, also by comparing the results with those of the sum spectra obtained by summing up these 2–3 spectra per detector. For external quality control the lab participates in national (Federal Office for Radiation Protection (BfS), Berlin, Germany, water samples) as well as IAEA-EL intercomparison exercises with marine origin samples. The latest IAEA intercomparison test sample measured with the new calibration from January 2011 was a seaweed sample (code IAEA-446). Our results for mean activity concentrations (i.e. activities per unit mass) of ^{40}K and ^{137}Cs differed only 1.1 % from the preliminary IAEA reference values (under evaluation). More information about the intercomparison exercises our lab participated in, but also about GamW built-in utilities for routinely testing evaluations of complex spectra, can be found in HELCOM (2012).

2.3 Modeling Cs in fish

For validation of the activity concentrations for ^{134}Cs and ^{137}Cs measured in Baltic Sea fish and giving an estimation of the future behavior of the Cs-isotopes in fish, the HELCOM box-model for the Baltic Sea case has been used (HELCOM, 1995; Kanisch et al., 2000). In this compartment model, the Baltic Sea and the North Sea are separated into several water boxes in which the transport of radionuclides between water boxes, sorption and remobilization of radionuclides to/from the sediment, the physical decay of radionuclides, and the input from anthropogenic sources is described by first-order differential equations. Modeling results have recently been presented in a HELCOM Thematic Assessment Report (HELCOM, 2009); see also Nielsen et al. (2010). The model takes time-dependent uptake by biota into account. For the special case of the fallout deposited to the Baltic Sea, it has been extended to a two-compartment model

BGD
10, 4127–4156, 2013

by considering piscivorous fish (or predating fish in the following) feeding on smaller fish. We refer to Rowan and Rasmussen (1996, 1997), but also to Smith (2006), Fievet and Plet (2003), and Wang et al. (2000), respectively, for the underlying bioenergetics based assumptions. A direct uptake of Cs from seawater has been included according to Brown et al. (2006). The link between the model's seawater activity concentrations, $c_w(t)$ in $\mathrm{Bq\,L^{-1}}$, and the activity concentrations in food fish and the predating fish, $c_{fd}(t)$ and $c_F(t)$ in $\mathrm{Bq\,kg^{-1}}$ w.w., respectively, is given by a system of two coupled first-order differential equations solved by a Runge-Kutta method (Press et al., 1992). Including specific growth of the predating fish the equations are:

$$\frac{dc_{fd}(t)}{dt} = K_{fd}(\lambda_{bio,fd} + \lambda_p)\,c_w(t) + k_w c_w(t) - (\lambda_{bio,fd} + \lambda_p)\,c_{fd}(t), \quad (1)$$

$$\frac{dc_F(t)}{dt} = k_F\,\alpha\,c_{fd}(t) + k_w c_w(t) - (\lambda_{bio,F} + \lambda_p + \lambda_G)\,c_F(t), \quad (2)$$

with rate constants λ_p, $\lambda_{bio,fd}$ and $\lambda_{bio,F}$ ($\mathrm{d^{-1}}$) linked to the physical half-live of the radionuclide and the biological half-lives, respectively; λ_G is the specific growth rate ($\mathrm{g\,g^{-1}\,d^{-1}}$ (or $\%\,\mathrm{d^{-1}}$) of the predator fish; K_{fd} and K_F are the concentration factors of food fish and predator, respectively, in $\mathrm{L\,kg^{-1}}$; α is the food assimilation efficiency of the predator; for k_w denoting the uptake rate of Cs from water a value of $0.04\,\mathrm{d^{-1}}$ was used. The general relation between T_{bio} and λ_{bio} is $\lambda_{bio} = \ln 2/T_{bio}$. An average value of $0.0015\,\mathrm{g\,g^{-1}\,d^{-1}}$ was estimated as λ_G for BS cod, with a typical (total) length of about 50 cm, from the weight-based von Bertalanffy growth parameters, which are available in the Fishbase database (Froese and Pauly, 2011).The quantities k_F and α can be eliminated by expressing by analogy the first term of Eq. (2) like that of Eq. (1) and taking the relation $c_{fd}(t) = K_{fd}c_w(t)$ into account:

$$k_F \alpha K_{fd} = K_F(\lambda_{bio,F} + \lambda_p + \lambda_G) \quad (3)$$

By implementing this in the model, assuming a single-time input to the Baltic sea water box surfaces, activity concentrations of fish were modeled by using a caesium

BGD
10, 4127–4156, 2013

concentration factor K_F of 164 L kg^{-1} (HELCOM, 2009; due to lower salinity in the Baltic Sea it is larger than the value of 100 L kg^{-1} used otherwise for marine fish; IAEA, 2004a) and an assumed value of 150 days for the biological half-live $T_{bio,F}$ (corresponding to the rate constant of the Cs elimination from fish). For food fish (smaller fish), assumed values of 50 L kg^{-1} and 80 d were used for K_{fd} and $T_{bio,fd}$, respectively.

Brown et al. (2006) used in their simulation study a range from 69 to 385 days for marine fish and a "representative" value of 116 d. We used a value of 100 d for modeling the behavior of Sellafield-derived ^{134}Cs in Greenland current fish with a single-fish-compartment version; this case can be considered as very close to a steady-state situation; thus a value more appropriate than 100 d is not considered necessary. We did not try to model the Fukushima-derived ^{134}Cs in Greenland fish, because we are lacking of an appropriate sub-model describing how the ^{134}Cs, deposited to the surface, would be mixed within the vertical water column. However, in the Baltic Sea, with its significantly lower salinity compared to the Atlantic Ocean, the biological half-lives may be significantly larger for fresh water fish than for pure marine fish (Rowan and Rasmussen, 1996, 1997). Therefore, a value of 150 days used for $T_{bio,F}$ in the Baltic is only a rough estimate.

The fish samples originating from the Baltic Sea were collected from the water boxes 75, 81 and 83, which represent the Belt Sea and the Baltic Proper (Fig. 1). The water depths and the salinities are quite variable, with gradients from west to east: between about 30 m and 160 m (depth), between 15 and 7.5 psu (surface salinity) and between 20 and 8 psu (bottom salinity) (HELCOM, 2010). Therefore, each water box 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 six selected boxes by weighting with their associated volumes. A value of 2 Bq m^{-2} (see introduction) for each of the caesium isotopes was used for the Fukushima fallout deposited to the Baltic Sea surface boxes.

BGD
10, 4127–4156, 2013

For the Greenland case, another box-model was used which extends the HELCOM box model with respect to the seas in and around the Arctic (Nielsen et al., 1995); as regards the Baltic Sea the results from this extended model agreed very well with results of the other model. The extended box-model was used for clarifying possible ^{134}Cs background sources in the Greenland case. However, it was not used to model the short-term behavior of deposited Fukushima ^{134}Cs in seawater and in fish around Greenland. If one would do this anyway, it would lead to unrealistically small estimated concentrations in seawater and fish. This is caused by the compartment model specific property to instantaneously perfectly mix the analyte within the overall box volume, which, due to the much deeper and larger box volumes, means much larger dilution as compared to that in the Baltic Sea.

3 Results and discussion

3.1 Greenland seas – Cs-activity concentrations and source identification for fish

The results of the Cs measurements of Greenland fish are given in Table 2. The ^{137}Cs activities showed a quite similar distribution pattern of 4 samples each between 2010 and 2011. It is found that ^{134}Cs was clearly detected in the two 2011 fish samples (17704 and 17705) collected at the end of October 2011 in the West Greenland current with ISO 11929 related significance factors $SF > 2.3 \gg 1$; SF is the ratio (^{134}Cs activity)/(^{134}Cs decision threshold), the latter being calculated according to ISO 11929. The activity concentrations found, around 0.005 Bq kg^{-1}, are extremely low. In contrast, in the two fish samples from the East Greenland current (17702 and 17703) ^{134}Cs was about two times smaller and thereby hardly detectable, $SF < 1$. The activity ratio ^{134}Cs/^{137}Cs was about 0.013 for sample 17703 while it showed the identical value of 0.022 for the two West Greenland current samples. As the ^{134}Cs/^{137}Cs ratio of FD-NPP-depositions was described to be close to

BGD
10, 4127–4156, 2013

one, the FD-NPP derived ^{137}Cs must also be expected in fish at the same size as ^{134}Cs. Therefore, for the two West Greenland current samples of 2011, the ratio $^{137}Cs^{Fuku}/^{137}Cs^{pre\text{-}Fuku} = {}^{134}Cs^{Fuku}/(^{137}Cs^{measured} - {}^{134}Cs^{Fuku})$ is about 0.023, which means that the Fukushima deposition added at maximum about 2 % to the existing pre-Fukushima ^{137}Cs in the West Greenland current, while it was about or less than 1 % in the East Greenland current samples. Whether the surface seawater activity concentrations in the two regions were significantly different cannot be deduced because due to other sampling strategy reasons the fish was sampled in stratified layers; average depths were around 200 m and about 400 m in the West Greenland and East Greenland site, respectively (see Table 1). For the rather fresh fallout situation the Cs isotopes with FD-NPP origin are not fully mixed over depths down to nearly 400 m; therefore, the difference of ^{134}Cs values in fish between the two sites may simply be a consequence of different sampling/water depths when we assume that the fish stay for a longer time in these layers. The ^{134}Cs and ^{137}Cs values of fish were plotted versus sampling depth (Fig. 4). This reveals that the depth dependence of ^{137}Cs is rather weak, which means that slight ^{137}Cs differences found between the two sampling sites probably do not depend on sampling depth. However, the sampling depth dependency is quite strong for ^{134}Cs. Thus, this observed sampling depth dependency, showing an incomplete vertical mixing of ^{134}Cs in the water column, supports our assumption that ^{134}Cs originates from the deposition of FD-NPP fallout.

For the coastal East Greenland current, Dahlgaard et al. (2004) found surface ^{137}Cs concentrations of about 3–4 Bq m^{-3} in 2001; they measured concentrations decreasing from 5–2.4 Bq m^{-3} in this current, from north to south in the eastern branch and in the coastal West Greenland current up to the north again. They also determined ^{137}Cs in two fish samples collected 1999 in the East Greenland current with values of 0.35 and 0.47 Bq kg^{-1} w.w After decay correction with an effective half-live of 18.6 yr, by which the ^{137}Cs concentration in the North Atlantic seawater and in fish decreases (IAEA, 2005; according to data for the box 28 given therein), their average then amounts to 0.27 Bq kg^{-1} in 2010. This is rather close to our 2010 and 2011-averages of about

BGD
10, 4127–4156, 2013

0.20 Bq kg^{-1} (from Table 2) for the two sites in the East Greenland current. Extrapolating a ^{137}Cs seawater time trend for the corresponding water box number 28 taken from IAEA (2005) and using a concentration factor of 100 L kg^{-1} leads to a value of around 0.2 Bq kg^{-1} in fish. Our samples from the West Greenland current show only slightly larger values of about 0.25 Bq kg^{-1}.

Dahlgaard (1995) estimated a transport time of 7–10 yr of such discharges to the East Greenland current. For testing possible contributions of the Sellafield-derived ^{134}Cs background to our measurements in fish, the box-model extended with respect to the Arctic Sea (see Sect. 2.3) was used. ^{137}Cs values around 0.1 Bq kg^{-1} were obtained from this model using the surface layer (thickness 330 m) of the relative large box "Irminger Sea" and a fish concentration factor of 100 L kg^{-1}. This is only 2.5 times lower than our measured values, i.e. not unrealistic. However, the corresponding values modeled for ^{134}Cs in fish, around 4×10^{-6} Bq kg^{-1}, were 1000 times below our measured ^{134}Cs values. This finding assures that the ^{134}Cs measured in fish originates from the FD-NPP fallout deposition. Due to reasons similar to those given at the end of Sect. 2.3, but also because the Chernobyl Cs deposition (Bq m^{-2}) to the Greenland current is assumed to be much smaller than in the Baltic Sea, it can be assumed that Chernobyl-derived ^{134}Cs in fish around Greenland would be much smaller than that estimated by modeling in the Baltic Sea and therefore would not contribute to the measured ^{134}Cs values.

3.2 Baltic Sea – Cs-activity concentrations and source identification for fish

Compared to the seas around Greenland with larger water flows, the rate of Baltic Sea water exchange with the adjacent North Sea is rather small. In the Baltic the salinities therefore are lower due to freshwater inflows. The Chernobyl ^{137}Cs seawater inventory in the southern part of the Baltic Sea (Baltic Proper) is decreasing with an effective half-live of around 15 yr (HELCOM, 2009).

BGD
10, 4127–4156, 2013

Results of measurements are included in Table 2; the samples from the additional retrospective screening of many other spectra for the existence of ^{134}Cs are included. In the Baltic Sea, due to the impact by the Chernobyl fallout, the ^{137}Cs activity concentrations in fish are around 7 Bq kg^{-1} being about 35 times larger than in the Greenland currents. ^{134}Cs was found in only three of the six samples with larger counting durations. The ISO 11929 related significance factors SF of the ^{134}Cs values where closer to one, with about 1.36 at maximum, but for the other samples at around 1 or even below 1. Thus, when including the four values from screening values, ^{134}Cs was definitely detected in only 50 % of the samples. Therefore, methods for left-censored data (Helsel and Cohn, 1988; Helsel, 2005) were applied for obtaining descriptive statistics; mean and median values were about 0.0036 Bq kg^{-1}. The Baltic Sea model was used for clarifying the origin of ^{134}Cs. Figures 2 and 3 shows the curves modeled in fish and measured values, of ^{137}Cs and ^{134}Cs, respectively. The lower and upper boundary (dashed) curves (±2 s) were calculated from the standard deviation of the 6 individual box activity concentrations.

It had been tested how setting up the fish kinetic sub-model affects the shape of the modeled curve of ^{134}Cs. Within the curves (Fig. 3), mainly their increasing part and the height of the maximum were found to be more variable, while the decrease following the maximum is determined by the decrease of modeled ^{134}Cs in seawater (near to equilibrium between water and fish). Lowering the biological half-live as well as adding specific fish growth to the single compartment sub-model resulted in a steeper increase of the ^{134}Cs activity concentration before reaching the maximum. Introducing the food fish compartment has the effect of some delay in this increase. Figure 3 shows the results of two model variants: (i) single-compartment model neglecting specific growth and Cs intake from water (upper graph) and (ii) two-compartment model taking both of these effects into account (lower graph). Therefore, the result obtained with a simple single-compartment sub-model is considered not bad taken as a first guess.

The model curve for ^{137}Cs in fish, which is mainly caused by the still large contribution of ^{137}Cs deposited during the Chernobyl accident, is unaffected by the FD-NPP

BGD
10, 4127–4156, 2013

deposition, the increase is as small as the line thickness in Fig. 2a. For ^{134}Cs however, the effect of FD-NPP-fallout is clearly visible in the model, but with very low values of the order of 0.004 Bq kg^{-1} (w.w.) being about 2000 times smaller than those of ^{137}Cs. Note, that the ^{134}Cs modeled for the pre-FD-NPP time is still the continuously decaying activity from the Chernobyl accident (effective half-live about 15 yr; HELCOM, 2009). Additionally, it has been tested that possible ^{134}Cs contributions originating from small annual liquid discharges of the Baltic Sea area nuclear power plants (see HELCOM, 2009, chapter 2) would hardly be identifiable in the graph for ^{134}Cs.

The good agreement between modeled and measured ^{137}Cs values suggests that the modeled ^{134}Cs values are as reliable as for ^{137}Cs. As the two Cs isotopes have a different origin one might argue that the very recently introduced ^{134}Cs behaves differently from ^{137}Cs with respect to mixing in the seawater. However, apart from one measured value from 19 April 2011, the measured ^{134}Cs values from the post-Fukushima time are well within the lower and upper model boundaries (Fig. 2); this also holds for ^{137}Cs. When continuing the upper ^{134}Cs boundary curve from the beginning of 2011, i.e. when assuming no Fukushima input, this curve (pure Chernobyl-^{134}Cs) during 2011 would also safely lie beneath the measured values. Therefore, we conclude, that the major contribution to ^{134}Cs values measured in fish, i.e. on average about 0.0026 Bq kg^{-1}, is related to the FD-NPP fallout deposition. The addition of ^{137}Cs from FD-NPP to existing ^{137}Cs in Baltic Sea fish corresponds to an increase of only 0.04 %.

3.3 Dose considerations about the Fukushima contribution

The contribution of the FD-NPP fallout deposition to effective dose rates received by the human individual through consumption of fish is estimated in relation to the corresponding pre-FD-NPP dose rates. Assuming an annual human consumption of 10 kg fish fillets and estimating the FD-NPP contribution from averages of measured ^{134}Cs values in fish with using the activity ratio ^{134}Cs/^{137}Cs of about 1 in March 2011 (neglecting the ^{134}Cs decay), dose rate contributions to human individuals are obtained

BGD
10, 4127–4156, 2013

which are given in Table 3; the dose conversion coefficients (ICRP, 1996) are given in Table 3. The extremely low effective dose rates obtained for the Fukushima contribution were 0.0011 µSv yr^{-1} and 0.00083 µSv yr^{-1} for the Greenland and Baltic Sea areas, respectively. A main result is that the relative increase of the effective dose rate, caused by the deposition of Fukushima fallout, is about 4 % for the Greenland case, but only about 0.1 % in the Baltic Sea case. The ^{134}Cs contribution will decrease from 2012 onwards (Fig. 3).

To put these extremely low dose rates into context they may be compared with those dose rates received by humans from the alpha-ray emitting radionuclide ^{210}Po, a naturally-occurring radionuclide which is also accumulated by fish. From average ^{210}Po activities per unit mass of about 0.94 Bq kg^{-1} w.w. in Baltic Sea fish (HELCOM, 2009) the human fish consumer receives a dose of 11 µSv per year after consumption of 10 kg fish per year; a ^{210}Po dose conversion coefficient of 1.2×10^{-6} Sv Bq^{-1} has been considered (ICRP, 1996). In comparison, this dose rate is larger than that from the Fukushima contribution by more than a factor of 10^4. The data in Table 3 also imply that even the dose rate received from Chernobyl-derived ^{137}Cs in Baltic Sea cod (0.88 µSv yr^{-1}) is nearly a hundred-fold larger than that of the Fukushima contribution. Within a dose assessment for consumption of sea fish (Aarkrog et al., 1997), a ^{210}Po value in fish of 2.4 Bq kg^{-1} was used which, taking the meanwhile larger ICRP dose conversion coefficient into account, would result in a dose rate of 29 µSv yr^{-1} for consuming 10 kg yr^{-1}.

4 Conclusions

During our study, we determined caesium isotopes in fish samples from the Greenland and the Baltic Sea originating from FD-NPP fallout deposited over the North Atlantic as suggested from other publications on modeling its airborne deposition. ^{134}Cs became detectable by using large amounts of fish fillets and an extended counting duration as compared to our standard method. In the Greenland Sea, being much

BGD
10, 4127–4156, 2013

deeper compared to the Baltic Sea, we observed a significant negative correlation ($p = 0.0018$) of ^{134}Cs activities in fish with sampling depths (150–400 m) while this was not true for ^{137}Cs. This indicates that ^{134}Cs was not yet mixed in the water column which substantiates the Fukushima fallout as its origin, while a major part of ^{137}Cs originates from global fallout and is well mixed over this depth. It also indicates the general feature of ^{134}Cs as an ideal tracer that it is well suited for studying processes of vertical mixing of fallout in an ocean when the ^{134}Cs levels prior to the fallout are extremely low.

The additional contribution through FD-NPP fallout to the effective dose received by members of the public by consumption of fish was calculated to be extremely low, e.g. four orders of magnitude lower than the effective dose rate received at the same time by the naturally occurring ^{210}Po, which is persistent in fish. Due to the short duration of deposition at sea it is expected that levels of caesium isotopes from FD-NPP will continue to decrease in seawater by vertical and horizontal mixing and by physical decay. Therefore, our results indicate that, although based on a limited set of data, the improved monitoring of ^{134}Cs in fish, also in other northern areas such as the Barents Sea, in combination with the NE Atlantic waters box model, are essential tools to maintain consumer safety.

Appendix A

Details of Gamma-ray spectrometric analysis with GamW

A numerical procedure for geometry dependent self-attenuation corrections, accounting also for different elemental compositions of fish ash and the calibration solution, is included in GamW. Calculated self-attenuation corrections at 605 keV were between 0.91 and 0.94.

The method used for the TCS calculations including X-ray contributions was that of Novcović et al. (2007); see also Kanisch et al., 2009. The extension from point to

BGD
10, 4127–4156, 2013

volume sources was achieved by the "LS-curve" method (Vidmar and Korun, 2006; Vidmar and Kanisch, 2010; Lépy et al., 2012). Approximate total efficiency curves required by such calculations were obtained with the gamma-ray spectrometry simulation tool GESPECOR (Sima et al., 2001). The linear energy calibration is re-calculated for each sample spectrum according to well-known radionuclide energies in the spectrum. The spectrum evaluation then starts with a peak search followed by nuclide library based complementation of the list of peaks; peak multipletts are fitted by non-linear weighted least squares (Press et al., 1992; Levenberg-Marquardt subroutine MRQMIN). The peak shape function used (Koskelo et al., 1981) consists of a Gaussian with low-energy tailing, extended by adding a step-function to the 1–3 parameter background-continuum polynomial. The method described by Uher et al. (2010), in fact being a penalized weighted least squares method, was implemented into the MRQMIN-package for stabilizing the fits especially in the case of small peaks, which is achieved by adding a penalty term to the Chi-square to be minimized for the non-linear parameters (width, location, relative step height and the tailing parameter of a peak). Thus, such parameters are allowed to vary thereby accounting for slight deviations from calibration values as they may occur during long-term measurements, and, on the other hand, they are prevented from leaving safe value domains which guaranties convergence of the fit.

Net background peak counting rates are subtracted from the corresponding gross peak counting rates. Then, decay corrected massic radionuclide activities are calculated by solving a linear matrix equation system by the linear least squares method according to the standard ISO 11929 (2010; Annex C.5 therein) taking peak efficiency correlations into account (Glavic-Cindro et al., 2004). Major relative standard uncertainties were about 1.5 % (peak efficiency around 662 keV), 2.9 % (preparing the fish ash as counting source including the sample wet/dry weight ratio uncertainty), about 0.5 % (gamma emission probabilities), 1 % (self-attenuation corrections) and around 2 % (TCS).

Decision thresholds and detection limits were calculated for each sample measurement by applying the ISO 11929 concept to the matrix-based least-squares procedure,

BGD
10, 4127–4156, 2013

i.e., not only for the easier-to-solve single-peak case of ^{137}Cs, but also for ^{134}Cs for which one or two lines (at 604.8 and 796.0 keV) have been used; in the latter case the detection limit value is lower than for a single peak. Treating this by using a weighted mean rather than by least-squares has been considered by Vivier et al. (2012). Values of $k_{1-\alpha} = 3$ and $k_{1-\beta} = 1.645$ were used for the quantiles of the normal distribution, the larger $k_{1-\alpha}$ value was used for preventing from finding too many non-significant peaks (De Geer, 2004).

Special measures for improving the detection of ^{134}Cs. For reducing the uncertainty of the fitted areas of the background continuum polynomial, the multiplet regions-of-interest (ROI) around 605 and 796 keV, respectively, were extended to the range of about 560 keV up to > 630 keV, which often included also the ^{137}Cs peak at 661.7 keV, and from 782 keV to 825 keV, respectively. Peaks from naturally-occurring radionuclides, also found in the separately measured background spectra, appeared at 609.3 keV (^{214}Bi) and at 794.9 keV (^{228}Ac), the latter lying quite close to the ^{134}Cs peak at 795.9 keV. Especially the spectra measured with the larger detector 4 showed more clearly an extra gamma-ray peak at around 596 keV as well as an additional broad background continuum contribution with a slow exponential decay towards energies above about 620 keV. This is due to neutrons caused by cosmic ray interactions in the shielding materials, which then by interactions within the detector produce special gamma-ray peaks (e.g. at 596 keV) and, amongst others, this broad "neutron-peak" above 596 keV due to inelastic scattering with Ge (Heusser, 1995; Jovančević et al., 2010); it is much easier recognized in background spectra in which the ^{40}K related Compton-background is much smaller than in fish ash samples. The functional representation of this "neutron-induced peak" given by Siiskonen and Toivonen (2005) has been implemented here in a simplified way as a further background component in the total fitting function. Figure A1 shows an example for the fitting region containing the 605 and 662 keV peaks in the spectrum of sample 17705 (Greenland site).

The upper limit of relative standard uncertainties of single peaks entering the matrix equation mentioned above was raised to 50 %; however, according to ISO 11929

BGD
10, 4127–4156, 2013

final activity values were considered as being detected only if they were above the decision threshold. TCS corrections for ^{134}Cs being comparable between its two major lines were estimated at between about 14 % and 24 % for the three detectors. For the fish measurements uncertainty budgets of activity concentrations (IAEA, 2004b) were dominated by contributions of sample preparation, peak efficiency and self-attenuation correction for ^{40}K and ^{137}Cs, while the counting rates contributed most dominantly in the case of ^{134}Cs.

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BGD
10, 4127–4156, 2013

Table 1. Characterization of fish samples (cod: *Gadus morhua*; redfish: *Sebastes marinus*; whiting: *Merlangius merlangius*).

Sample ID	Sampling date	species	Depth of catch [M]	Average fish weight [kg]	Average fish length [cm]	# of specimen	Wet sample mass (fillets) [kg]	mass ratio wet/dry	ash mass [g]	counting duration [h]
Greenland:										
17605	15.10.2010	redfish	394	6.04	67	2	3.03	3.26	34.58	160
17606	27.10.2010	cod	291	5.54	86	2	2.70	5.11	40.35	160
17607	25.10.2010	cod	417	3.32	84	2	3.25	4.75	45.67	160
17608	25.10.2010	redfish	156	5.59	56	2	1.64	3.93	20.08	160
17702	21.10.2011	redfish	375	1.38	50	6	3.99	3.75	50.66	410
17703	21.10.2011	cod	339	6.0	82	4	4.52	5.02	67.91	390
17704	29.10.2011	cod	144	7.3	92	4	4.14	4.86	57.11	390
17705	30.10.2011	redfish	243	1.78	50	6	4.09	3.98	53.66	390
Baltic Sea:										
17649	05.05.2011	cod	78	0.95	44	17	3.07	5.31	39.13	320
17650	05.05.2011	cod	78	1.77	55	13	4.31	5.28	56.60	320
17707	04.12.2011	cod	22	3.22	66	7	3.69	4.37	49.98	320
17708	05.12.2011	whiting	33	0.75	42	31	5.33	4.16	66.29	368
17716	12.12.2011	cod	50	0.87	46	25	4.36	5.48	54.21	394
17717	13.12.2011	cod	92	0.94	47	26	4.11	5.41	50.29	394
Baltic Sea, from screening										
17647	19.04.2011	cod	70	0.73	40	22	3.04	5.39	38.60	160
17670	18.09.2011	cod	4	1.11	48	15	3.69	5.40	48.57	160
17679	04.09.2011	cod	45	0.81	43	27	5.00	5.23	66.18	160
17720	14.12.2011	cod	42	1.01	46	26	5.56	4.93	76.29	160

BGD
10, 4127–4156, 2013

Table 2. Decay corrected activity concentrations A of ^{134}Cs, ^{137}Cs and ^{40}K and associated relative uncertainties (u_{rel}) in fish fillet samples from the Greenland and Baltic Sea sites; the decision threshold DT (not shown) is well approximated by the value DL/1.548; SF is a significance factor, i.e. the ratio A/DT; "non-detects" are documented as < DL.

sample ID	species	^{134}Cs					^{137}Cs		^{40}K	
		DetLim) (DL)	A	γ lines	u_{rel}	SF	A	u_{rel}	A	u_{rel}
		Bq kg^{-1} w.w.	Bq kg^{-1} w.w.		%		Bq kg^{-1} w.w.	%	Bq kg^{-1} w.w.	%
Greenland:										
17605	redfish	0.0072	< 0.0072	0			0.183	4.0	106	3.6
17606	cod	0.0093	< 0.0093	0			0.209	4.1	136	3.6
17607	cod	0.0062	< 0.0062	0			0.279	3.7	138	3.6
17608	redfish	0.0100	< 0.0100	0			0.269	4.0	132	3.6
17702	redfish	0.0032	< 0.0032	0			0.201	3.7	114	3.6
17703	cod	0.0046	0.0027	1	35.6	0.91	0.216	3.8	127	3.6
17704	cod	0.0036	0.0058	2	13.4	2.54	0.260	3.8	134	3.6
17705	redfish	0.0028	0.0042	2	15.1	2.31	0.188	3.7	116	3.6
Baltic Sea:										
17649	cod	0.0046	0.0039	1	24.2	1.35	6.72	3.6	130	3.6
17650	cod	0.0063	< 0.0063	0			6.81	3.7	132	3.6
17707	cod	0.0043	0.0037	2	23.7	1.37	6.51	3.8	136	3.6
17708	whiting	0.0035	0.0026	1	27.8	1.16	7.30	3.6	117	3.6
17716	cod	0.0051	< 0.0051	0			7.30	3.6	123	3.6
17717	cod	0.0044	< 0.0044	0			7.00	3.7	124	3.6
Baltic Sea, from screening										
17647	cod	0.0083	0.0050	1	34.5	0.96	8.18	3.7	130	3.6
17670	cod	0.0058	0.0042	1	28.6	1.13	4.29	3.6	131	3.6
17679	cod	0.0055	0.0034	1	33.1	0.98	4.18	3.6	127	3.6
17720	cod	0.0045	0.0035	2	26.0	1.24	4.81	3.6	135	3.6

BGD
10, 4127–4156, 2013

Table 3. Average activity concentrations ($Bq\,kg^{-1}$) and effective dose rate ($\mu Sv\,yr^{-1}$) contributions to human individuals from consumption ($10\,kg\,yr^{-1}$) of fish from 2011; dose conversion factors ($Sv\,Bq^{-1}$) used were 1.9×10^{-8} and 1.3×10^{-8} for ^{134}Cs and ^{137}Cs, respectively; note that $^{134}Cs^{pre\text{-}Fukush}$, not occurring in the table, can be neglected.

Radionuclide/Source	Greenland: [$Bq\,kg^{-1}$]	[$\mu Sv\,yr^{-1}$]	Baltic Sea: [$Bq\,kg^{-1}$]	[$\mu Sv\,yr^{-1}$]
Fukushima:				
$^{134}Cs^{Fukush}$	0.0035	6.7E-04	0.0026	4.9E-04
$^{137}Cs^{Fukush} \approx {}^{134}Cs^{Fukush}$	0.0035	4.6E-04	0.0026	3.4E-04
total F.		1.1E-03		8.3E-04
$^{137}Cs^{measured}$: as measured	0.21	0.027	6.8	0.88
$^{137}Cs^{pre\text{-}Fukush} = (^{137}Cs^{measured} - {}^{137}Cs^{Fukush})$	0.21	0.027	6.8	0.88
dose rate ratio: total F. / $^{137}Cs^{pre\text{-}Fukush}$ = relative increase of the dose rate by Fukush.		0.042		0.00094

BGD
10, 4127–4156, 2013

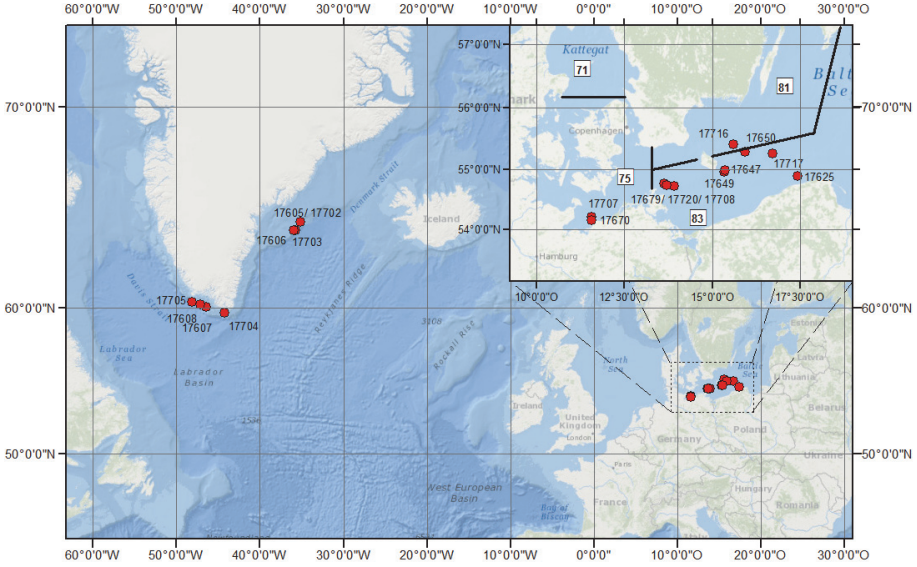

Fig. 1. Sites for sampling of cod, redfish and whiting (see Table 1 for sample details) for analysis of radioactivity in the Labrador Basin and the Baltic Sea (Scale of the large map is 1 : 42 000 000, Scale of the small map is 1 : 10 000 000); The small map from the Baltic Sea already contains the borders and nomenclature of the water boxes (numbers in boxes; 71 = Kattegat, surface 75 = Belt Sea, surface; 81 = Baltic Sea West, surface; 83 = Baltic Sea East, surface) for estimation of the Cs-behaviour using the modified HELCOM Box-model (see HELCOM, 1995, and Kanisch et al., 2000, for details)

BGD
10, 4127–4156, 2013

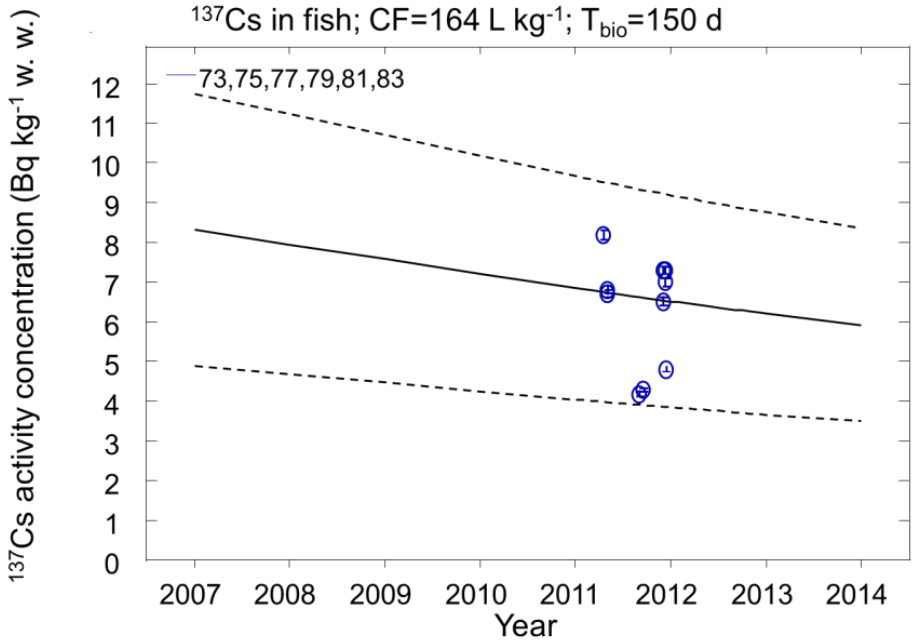

Fig. 2. Curves of ^{137}Cs activity per unit mass in fish using the two-compartment fish model, volume-averaged over the surface boxes 75, 81 and 83 and their associated bottom-layer boxes, and measured values (circles) of ^{137}Cs in Baltic Sea fish. Individual measured values are given with ±2 s-bars; the dashed lines represent the ±2 s model standard deviation curves (see text).

BGD
10, 4127–4156, 2013

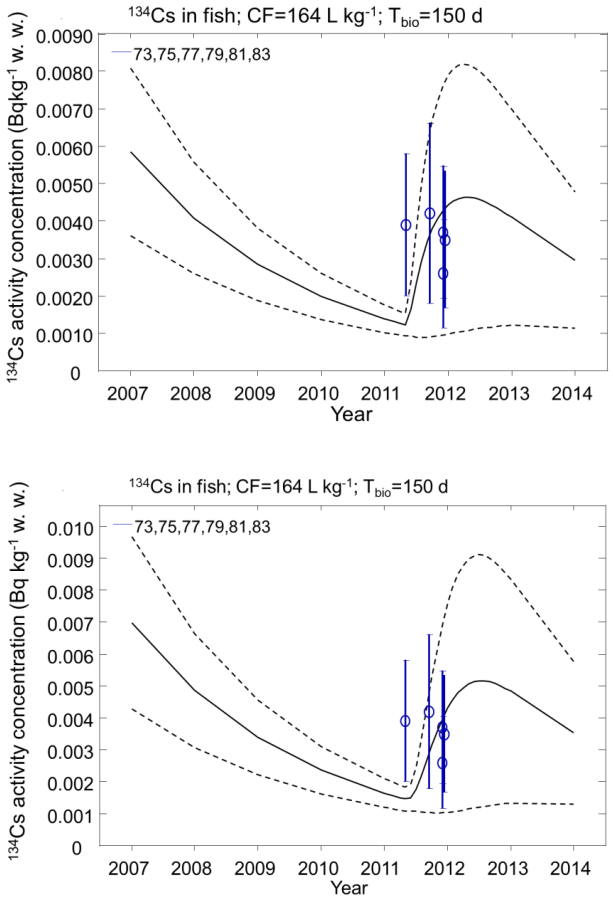

Fig. 3. Curves of ^{134}Cs activity per unit mass in fish using two models and measured values of ^{134}Cs in Baltic Sea fish. Upper part: single compartment model curves with ignoring growth G and Cs uptake from water k_w; lower part: two-compartment model used with G and k_w values given in the text; otherwise the same as in Fig. 2.

BGD
10, 4127–4156, 2013

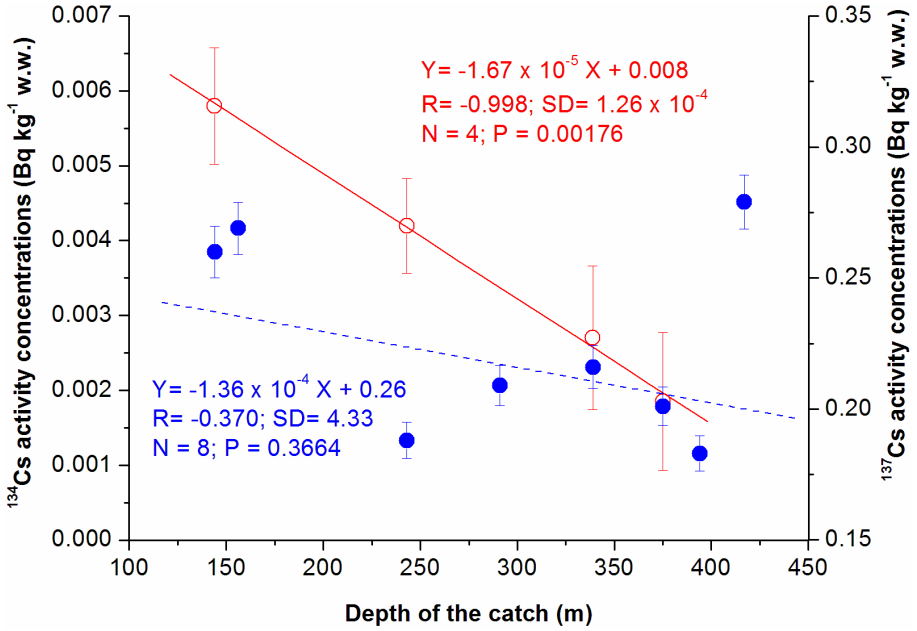

Fig. 4. Activity concentrations of ^{134}Cs (open circles, left scale, 2011) and of ^{137}Cs (closed circles, right scale, 2010 + 2011) in fish fillets plotted versus sampling depth; measurement uncertainties given are ±1 s; Note: the smallest of the ^{134}Cs values represents a "non-detect" with DL of 0.0032, for which a replacement value was plotted;

BGD
10, 4127–4156, 2013

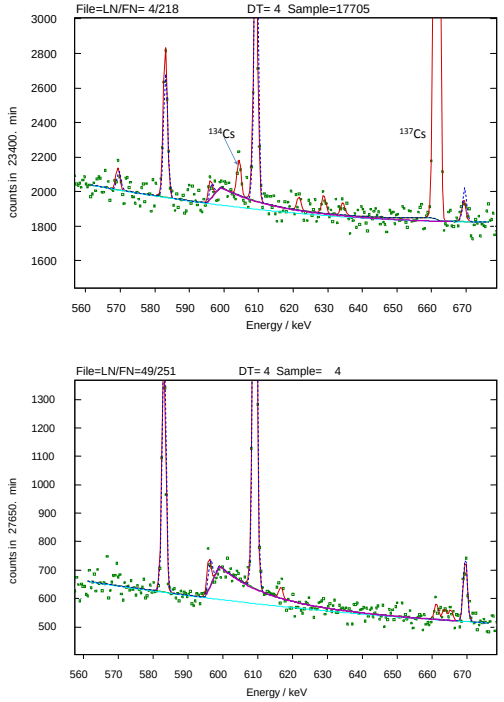

Fig. A1. Upper part: ^{134}Cs (at 605 keV) and ^{137}Cs (at 662 keV) peaks fitted in the region 560-665 keV ($\chi^2_R = 1.21$); the red colored curves show the peak fitting functions residing above a 3-parameter polynomial background continuum (light cyan); dashed peak curves indicate the peaks sizes obtained from separate background measurements. Note the broad neutron-induced background peak beneath the peaks of 605 and 609 keV; after accounting for different counting durations its fitted height parameter agrees to within 8 % with that fitted in the separate background spectrum. The three small peaks between 609 and 662 keV were found to be not significant. Lower part: spectrum region of the corresponding background spectrum showing more clearly the neutron-induced background peak beneath and no ^{134}Cs, respectively.

BGD
10, 4127–4156, 2013