Trace Levels of Fukushima Disaster Radionuclides in East Pacific Albacore


**DOI**: 10.1021/es500129b

**Publisher**: American Chemical Society

**Version**: Version of Record

**Terms of Use**: http://cdss.library.oregonstate.edu/sa-termsofuse
Trace Levels of Fukushima Disaster Radionuclides in East Pacific Albacore

Delvan R. Neville,*† A. Jason Phillips,‡ Richard D. Brodeur,§ and Kathryn A. Higley†

†Department of Nuclear Engineering & Radiation Health Physics, and ‡College of Earth, Ocean, and Atmospheric Sciences (CEOAS), Oregon State University, Corvallis, Oregon 97331, United States
§Northwest Fisheries Science Center, National Oceanic and Atmospheric Administration (NOAA) Fisheries, Newport, Oregon 97365, United States

ABSTRACT: The Fukushima Daiichi power station released several radionuclides into the Pacific following the March 2011 earthquake and tsunami. A total of 26 Pacific albacore (Thunnus alalunga) caught off the Pacific Northwest U.S. coast between 2008 and 2012 were analyzed for $^{137}$Cs and Fukushima-attributed $^{134}$Cs. Both 2011 (2 of 2) and several 2012 (10 of 17) edible tissue samples exhibited increased activity concentrations of $^{137}$Cs (234–824 mBq/kg of wet weight) and $^{134}$Cs (18.2–356 mBq/kg of wet weight). The remaining 2012 samples and all pre-Fukushima (2008–2009) samples possessed lower $^{137}$Cs activity concentrations (103–272 mBq/kg of wet weight) with no detectable $^{134}$Cs activity. Age, as indicated by fork length, was a strong predictor for both the presence and concentration of $^{134}$Cs ($p < 0.001$). Notably, many migration-aged fish did not exhibit any $^{134}$Cs, suggesting that they had not recently migrated near Japan. None of the tested samples would represent a significant change in annual radiation dose if consumed by humans.

INTRODUCTION

In the aftermath of the 2011 Tōhoku magnitude 9.0 earthquake, the hydrogen explosions from three reactor buildings at the Fukushima nuclear plant in Japan in March 2011 released a substantial amount of radioactive particles into the atmosphere and ocean. Fission products, including $^{134}$Cs and $^{137}$Cs, were released from the Fukushima Daiichi nuclear power station and are still making their way into the food chain. Surveys in the region off Japan have recorded elevated levels of radionuclides in the water and biota in the month following the accident, up to 600 km offshore of the release site.

The ocean ecological dynamics of a nuclear accident are not yet well-defined from previous accidental releases from reactors. The Sellafield fuel reprocessing plant in the United Kingdom released far more $^{137}$Cs from a year of normal operations in the 1970s than during its 1954 “Windscale” graphite fire. The nuclear accident at Three Mile Island in New York released only noble gases and iodines with undetectable oceanic contribution. The Chernobyl, Ukraine oceanic contributions were minor compared to the terrestrial inputs and arose mainly via global atmospheric fallout. $^{134}$Cs is produced primarily in nuclear fuel from neutron activation of stable $^{133}$Cs. Very little is produced in nuclear weapons testing, because all other fission products with 134 nucleons undergo $\beta$-decay to stable $^{133}$Xe or $^{133}$Ba rather than to $^{134}$Cs. $^{134}$Cs has a short half-life (2 years), and U.S. and Canadian reactors do not release radioactive waste into the ocean. Thus, no other known $^{134}$Cs source is available on the West Coast other than Fukushima.

Surface ocean currents are not expected to introduce the liquid plume with radionuclides derived from the Fukushima Daiichi nuclear power station to U.S. waters until 2014–2016. However, fish species, such as Pacific albacore (Thunnus alalunga), are known to make trans-Pacific migrations that can bring them near Japan, and they could serve as transport vectors for these radionuclides. Madigan et al. determined that the closely related species Pacific bluefin tuna (Thunnus orientalis) caught near California did in fact transport radionuclides across the Pacific. On the basis of the high trophic level and rapid trans-Pacific migration of albacore, the hypothesis was put forth that $^{134}$Cs tracer activity and elevated $^{137}$Cs activity would be found in west coast U.S. caught albacore as early as summer 2011, just a few months after the primary release at Fukushima. In this study, we examined levels of radioactive Cs in specimens of albacore collected prior to and after the Fukushima release to better understand the recent migration patterns in albacore caught along the U.S. Pacific Northwest coast.

MATERIALS AND METHODS

Collections at sea were made as part of research cruises conducted by the National Marine Fisheries Service during the...
at half-maximum (fwhm) \( = 1.33 \text{ MeV} \), and 1.0 keV resolution (fwhm) at 122 keV.

Methods were certified using IAEA-414 freeze-dried fish tissue standards. The chemical yield for cesium through the drying and ashing process was unity, within the range of counting uncertainty (\( \sigma = 4.074\% \)). To account for differences in detection geometry arising from differing volumes of ash, samples of known activity were counted at various fill volumes and a weighted least-squares fit for the absolute efficiency based on the fill volume was produced. Uncertainties in count rates, mass, geometry-altered efficiency, and chemical yield (on the basis of the yield using the IAEA-414 standards) were propagated. In total, 7 albacore from 2008, 2 from 2011, and 17 from 2012 were analyzed for \(^{137}\text{Cs}\) and \(^{134}\text{Cs}\) in either the edible portion (loins) or the carcass.

### RESULTS AND DISCUSSION

The complete set of results is presented in Table 1, with samples ordered by year of collection. Because many of the available samples were carcasses rather than whole fish, the ratio of activity concentration between the loins and the carcasses was estimated from five albacore that had both loins and carcass available [mean = 0.97, and standard error (SE) = 0.14]. The estimated loin concentrations were calculated from this ratio to allow for cross-comparison between all albacore sampled. A similar approach was applied to the 2011 sample that already had the loins and carcass mixed together, to estimate the activity concentration in the edible portion to that in the whole body that was ashed (mean = 0.95, and SE = 0.08). The ratio of \(^{137}\text{Cs}/^{134}\text{Cs}\) for Fukushima-derived tracers is believed to be 1:1 when back-calculated to March 11, 2011. This back calculation allowed for quantifying how much of \(^{137}\text{Cs}\) present in a sample was derived from Fukushima based on what \(^{134}\text{Cs}\) was measured in the animal. Because \(^{134}\text{Cs}\) is relatively short-lived (2 year half-life), there are no other realistic sources for \(^{134}\text{Cs}\) in the northern California Current to alter this ratio prior to Fukushima. Figure 2 presents the (a) total \(^{137}\text{Cs}\) concentration in each of the loins, (b) \(^{134}\text{Cs}\) concentrations in the loins, and (c) extrapolated non-Fukushima \(^{137}\text{Cs}\) derived from a 1:1 March 11, 2011 ratio.

Because the Fukushima release was the sole source of environmental \(^{134}\text{Cs}\) available, one would expect the pre-existing \(^{137}\text{Cs}\) concentrations in albacore in 2008 to agree well with the \(^{137}\text{Cs}\) that could not be attributed to Fukushima via \(^{134}\text{Cs}\). As shown in Figure 2c, there is good agreement between \(^{137}\text{Cs}\) in pre-Fukushima samples (2008 series) and the non-Fukushima attributed \(^{137}\text{Cs}\) in post-Fukushima samples (2011 and 2012 series). The statistics support the visual conclusion that 1:1 is a good estimate of the Fukushima-derived \(^{134}\text{Cs}/^{137}\text{Cs}\) activity ratio. There is no significant difference between \(^{137}\text{Cs}\) in 2008 albacore and the non-Fukushima \(^{137}\text{Cs}\) in 2011 and 2012 fish (one-way analysis of variance (ANOVA); \( p = 0.1881 \)), and there is no significant difference between total \(^{137}\text{Cs}\) in those 2012 fish with no detectable \(^{134}\text{Cs}\) and non-Fukushima \(^{137}\text{Cs}\) in 2012 fish with detectable \(^{134}\text{Cs}\) (one-way ANOVA; \( p = 0.5864 \)). Total radiocesium was on average 198% higher in \(^{134}\text{Cs}\)-contaminated samples than in uncontaminated samples from any year.

Length was an excellent predictor of the occurrence of \(^{134}\text{Cs}\) (one-way ANOVA; \( p < 0.004 \)) in the 2012 samples. A least-squares linear regression for the model \( A = \beta_0 + L/\beta_1 \), explained a reasonable proportion of the variance in \(^{134}\text{Cs}\) (\( R^2 = 0.5688 \)), where \( A \) is the activity concentration in Bq/kg, \( L \) is the fork length in millimeters, \( \beta_0 = -1.074 \) Bq/kg...
Limiting the regression to only samples with detectable \(137\text{Cs}\) reduced the significance but did not substantially change either coefficient: \(\beta_4 = -1.042 \text{ Bq/kg} \ (p = 0.097)\), and \(\beta_1 = 1.652 \text{ mBq mm}^{-1} \text{ kg}^{-1} \ (p = 0.063)\). The relationship between length and \(137\text{Cs}\) is presented in Figure 3, where dashed bars represent the minimum detectable activity (MDA) for those samples that did not appear to contain any \(134\text{Cs}\). Some small age-based accumulation of non-Fukushima \(137\text{Cs}\) was evident in both modern and pre-Fukushima albacore as well (Figure 4), but the relationship was not statistically significant \((p = 0.137)\) for all albacore as a whole and only marginally significant \((p = 0.088)\) for albacore that contained no \(134\text{Cs}\). Figures 3 and 4 are both demarcated to show estimated age classes.\textsuperscript{14} Note that, because the uncertainty in non-Fukushima \(137\text{Cs}\) depends upon both the total \(137\text{Cs}\) detected and the total \(134\text{Cs}\) detected, most of the age 4 fish have much higher uncertainties in Figure 4, even though the quantity of non-Fukushima \(137\text{Cs}\) is the same. They have more total \(137\text{Cs}\) and \(134\text{Cs}\) than the age 3 fish, and the majority of age 3 fish have no detected \(134\text{Cs}\) at all.

Our findings have substantial relevance to food safety concerns. The derived intervention level for total radiocesium in food \((134\text{Cs} + 137\text{Cs})\) is 1200 Bq/kg in the United States.\textsuperscript{15} The highest total radiocesium in any sample was approximately 1180 mBq/kg (Table 1), 0.1% of the U.S. Food and Drug Administration (FDA) level of concern for radiocesium and about 1% of the typical activity concentration of naturally occurring \(40\text{K}\). The committed effective dose equivalent (CEDE) is the radiation safety dose term used to account for the total dose received over a lifetime as a result of ingesting radionuclides. This accounts for the rate at which the radionuclide decays and the rate at which it is eliminated from the body biologically. On the basis of the CEDE per unit activity for \(40\text{K}\), our findings have substantial relevance to food safety concerns. On the basis of the CEDE per unit activity for \(40\text{K}\), the committed effective dose equivalent (CEDE) is the radiation safety dose term used to account for the total dose received over a lifetime as a result of ingesting radionuclides. This accounts for the rate at which the radionuclide decays and the rate at which it is eliminated from the body biologically. On the basis of the CEDE per unit activity for \(40\text{K}\), our findings have substantial relevance to food safety concerns. On the basis of the CEDE per unit activity for \(40\text{K}\), the committed effective dose equivalent (CEDE) is the radiation safety dose term used to account for the total dose received over a lifetime as a result of ingesting radionuclides. This accounts for the rate at which the radionuclide decays and the rate at which it is eliminated from the body biologically. On the basis of the CEDE per unit activity for \(40\text{K}\), our findings have substantial relevance to food safety concerns.
134Cs and 137Cs,\textsuperscript{16} consuming 1 kg of the loins of the highest activity sample corresponds with a CEDE of 18 nSv or 0.0006% of the annual dose of radiation from natural sources for the average American.\textsuperscript{17} Although the trace levels that we detected will be useful in estimating migration history for albacore, they do not appear to be significant to food safety.

Age has long been held as a determinant in Pacific albacore migration patterns.\textsuperscript{8} Starting at age 2, albacore are thought to migrate between Japan and the U.S. until age 5. After age 5, they then remain in the Japanese fishery waters or move south to subtropical waters of the west Pacific. Our data also show a strong relationship between age and inferred migration near Japan. 134Cs concentrations were strongly correlated with fork length, and fork length has been used to estimate age of Pacific albacore.\textsuperscript{13} Both 3- and 4-year-old fish caught in summer 2012 would have had two opportunities to have migrated to Japan and back, but it is only the 4-year-old fish that always had high 134Cs concentrations. It may be that the 2-year-old fish are less likely than the 3-year-old fish to make the trans-Pacific migration to Japan and back to the U.S. the following year. Childers et al.\textsuperscript{7} demonstrated that the migration patterns of juvenile albacore departing the U.S. coast are more complex than a simple annual back-and-forth, observing five different migration patterns, only one of which resulted in an albacore reaching Japanese waters. Large net distances traveled from release and capture (>500 km) were limited to only 660–780 mm fork length juveniles (5 of 20 albacore recovered), comprising the largest of the age 3 juveniles and all of the age 4 juveniles, with a single 4-year-old albacore actually reaching Japan. This suggests that our 3-year-old fish had lower concentrations of 134Cs than the 4-year-old fish because 2012 was likely the first time these age 3 albacore made the trans-Pacific trip. The 4-year-old albacore, however, have now most likely crossed it twice. Monitoring the radiocesium concentrations of the stock in 2013 and beyond across a wider sampling region may address whether this difference is indeed a difference in migration patterns or whether it is simply age-based accumulation of a heavy metal contaminant.

Future work will include sampling in both northern and southern U.S. albacore fisheries, analyzing samples for other Fukushima-related radionuclides, and potentially acquiring albacore from the western Pacific. We hope to help address a long-standing question first raised by Brock,\textsuperscript{18} much studied by Laurs,\textsuperscript{19} and recently addressed by Barr;\textsuperscript{20} whether Pacific albacore on the U.S. west coast are composed of one stock or two subsstocks. The theory for the latter suggests that juveniles (age 2–5) in the northeastern Pacific are two stocks separated

---

**Figure 3.** Mean 134Cs concentrations versus fork length with different estimated age classes enclosed in ovals. Error bars are SE of the mean.

**Figure 4.** Mean non-Fukushima-attributed 137Cs in all samples tested with different estimated age classes enclosed in ovals. Error bars are SE of the mean.
spatially north/south of 40° N, with only the northern substock migrating to Japan. None of the samples in the data reported here came from the southern region (they spanned from 44.4° N to 47.47° N; Figure 1). Nonetheless, we calculated and found no statistically significant relationship between latitude and the presence of 134Cs (one-way ANOVA; p = 0.4875) nor between the quantity of 134Cs and latitude within those that did have detectable 134Cs (p = 0.3512).

**AUTHOR INFORMATION**

**Corresponding Author**
*Telephone: 541-602-8005. E-mail: dnevill@gmail.com.*

**Notes**
The authors declare no competing financial interest.

**ACKNOWLEDGMENTS**
The authors thank the Office of Science Director of the National Marine Fisheries Service for helping to fund this work. The authors thank Keith Bosley for providing pre-Fukushima albacore samples. The authors thank the crew and scientists on the RV Shimada that assisted with the capture and processing of albacore at sea and Mario Gomez-Fernandez and Junwei Jia for assistance in processing samples. Finally, the authors thank Drs. Lorenzo Ciannelli, Bill Pearcy, and Walt Dickhoff for comments on earlier versions of the manuscript and three anonymous journal reviewers.

**REFERENCES**


