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A Fukushima tracer perspective on four years of North Pacific mode water evolution

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Abstract

1

2 We present the results of a multi-platform investigation that utilizes tracer information provided 3 by the 2011 release of radioisotopes from the Fukushima Dai-ichi Nuclear Power Plant to better 4 understand the pathways, mixing and transport of mode waters formed in the North Pacific Ocean. The focus is on transition region mode waters and radiocesium (¹³⁷Cs and ¹³⁴Cs) observations 5 6 obtained from the May-June 2015 GO-SHIP occupation of the 152°W line in the Northeast Pacific. 7 Samples include profiles from the surface to 1000 m and surface/subsurface pairs that provide an 8 average 1° of latitude spacing along 152°W. We find a clear Fukushima (134Cs) signal from the surface to 400 m. The core signal (134 Cs ~ 10 Bg m⁻³, 137 Cs ~ 12 Bg m⁻³) at 41°-43°N lies at 30-220 9 m where mode waters formed through deep winter mixing in 2011 outcropped in the western North 10 11 Pacific. The strongest 2015 152°W Fukushima-source radiocesium signal is associated with 12 Dense-Central Mode Waters consistent with the densest variety of these mode waters being formed 13 off the coast of Japan 4 years earlier. The radionuclide signal transited the basin along subsurface 14 mode water isopycnals mainly on the northern side of the subtropical gyre before outcropping at 15 and to the east of the 152°W line. In 2015, the densest 152°W waters with ¹³⁴Cs lie at ~435 m in 16 the bottom range of Dense-Central Mode Water at 40°N. There is a weak, but detectable, signal in 17 the boundary current off both Kodiak and Sitka. The deepest detectable ¹³⁷Cs (weapon's testing background) are found at and to the north of 45°N at 900–1000 m. With the exception of a single 18 subsurface sample near Hawaii, as of spring 2015, the southernmost ¹³⁴Cs was found above 200 m 19 20 at 30°N. A total date-corrected ¹³⁴Cs inventory of 11-16 PBq is estimated. Qualitative comparison 21 to model output suggests good consistency in terms of general location, latitudinal breadth, and 22 predicted depth of penetration, allowing discussion of the bigger picture. However, the model's 23 2015 152°W radiocesium signal is quantitatively weaker and the core is offset in latitude, 24 potentially due to the lack of consideration of atmospheric deposition.

25 Keywords

- 26 Fukushima radiocesium
- Mode water
- North Pacific
- 29 Radionuclide Tracer
- 30 Ocean Circulation

31 1. Introduction

- In the Northwest Pacific, warm tropical/subtropical waters meet cold, upwelled, nutrient rich 32 33 waters from the north in a confluence zone east of Japan. Here, a variety of mode waters are formed 34 in deep winter mixed layers, reaching 300-400 m. It is also here that the radionuclide 35 contamination from the 2011 Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident was 36 directly discharged into the ocean and where the major portion of the atmospheric deposition was 37 thought to have occurred. This study examines the spread of Northwest Pacific mode water (MW) 38 from the perspective of observations made 4 years after the accident along 152°W and in the 39 Alaska Gyre. While many different radioisotopes were released as a result of the 2011 accident, we focus on radiocesium (henceforth referred to as Cs), in particular ¹³⁷Cs (30.07-year half-life) 40 and ¹³⁴Cs (2.06-year half-life). Although the Pacific Ocean has received Cs input in the past, most 41 42 notably due to nuclear weapons testing during the 1950's and 60's, given its two-year half-life, the presence of ¹³⁴Cs is a unique diagnostic tool for indicating the presence of waters derived from the 43 FDNPP. Henceforth, water tagged with ¹³⁴Cs will be referred to as FDNPP Cs or simply Cs_F. The 44 45 Cs_F release afforded an opportunity to employ a powerful, soluble, readily detectable tracer input to the ocean at a known time and location to better understand the timing and spread of North 46 47 Pacific MW.
- Section 1 includes a review of North Pacific circulation and mode waters, and a short history of
- North Pacific Cs. Section 2 presents the Northeast Pacific observed Cs dataset and its distribution.
- Section 3 relates the observed Cs patterns with MW distributions, calculates inventories, brings
- 90 Sr estimates to bear and compares the observations to model predictions. Section 4 is a summary.
- 52 Throughout, radionuclide units are given as activity in Bq (1 Bq = 1 nucleus decay per
- second) and activity concentration Bq m^{-3} .
- 1.1 North Pacific Circulation and Mode Waters North Pacific surface circulation is composed
- of three wind driven gyres: one subtropical and two semi-distinct subpolar, the Alaska and Western
- 56 Sub-arctic Gyres (Fig. 1). The western boundary Kuroshio Current separates from the coast at
- 57 ~35°N as it turns into the open ocean. Bifurcating, the main portion flows east as a strong inertial
- 58 jet known as the Kuroshio Extension (KE), and slightly further north, influenced by Oyashio
- waters, the Kuroshio Bifurcation Front (KBF) extends eastward to become the Subarctic Current
- 60 (Oka and Qiu, 2012).

In the west, to the south of the frontal regions (KE Front, KBF and further north near the Subarctic Front) strong heat loss to the atmosphere results in vertical convective mixing in winter. These regions of deep mixing have been illustrated by February mixed layer depths (MLD) observed by historical hydrography (Talley, 1988; Suga et al., 2004), satellites (e.g. Fig. 1) and profiling floats (e.g. Whalen et al., (2012); Yoshida et al. (2015)). The resulting ventilated, low potential vorticity "mode" waters that lie slightly above or within the permanent pycnocline are classified into six classes according to their distinct but homogeneous hydrodynamical characteristics (Table 1) and are reviewed in detail by Oka and Qiu (2012).

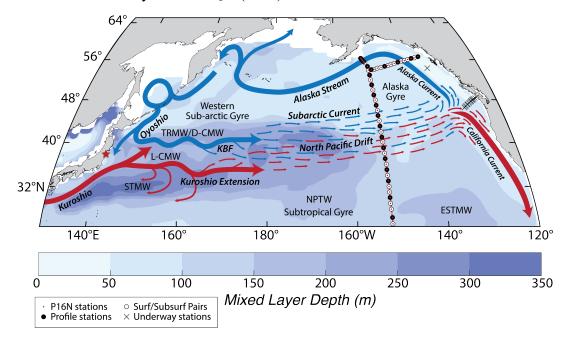


Figure 1: Schematic of North Pacific circulation overlaid on mean March mixed layer depth (blue shading, from GODAS, 2015; units: m) for the period 2011-2015. Line thickness does not represent current strength. Major currents, gyres and specific waters mass formation regions mentioned in the text are labeled: Kuroshio Bifurcation Front (KBF), Subtropical Mode Water (STMW), Light Central Mode Water (L-CMW), Dense Central Mode Water (D-CMW), Transition Region Mode Water (TRMW), Eastern Subtropical Mode Water (ESTMW) and North Pacific Tropical Water (NPTW). Hydrographic station locations for the northern end of the 2015 P16N/leg 2 occupation are shown as red dots. Black symbols indicate the location of cesium samples: open circle = subsurface/surface pair, filled circle = profile, and cross = underway surface sample. All P16N cesium stations are shown in Figure S1. The red star is FDNPP location.

North Pacific Subtropical Mode Water (STMW 24.5 < σ_0 < 25.9) is formed to the south of the KE front, and is associated with deep mixed layers (up to 400-450 m) and the anticyclonic recirculation of the Kuroshio Current system (e.g.Suga and Hanawa, 1995, Qiu et al., 2006). STMW's low potential vorticity is thought to arise largely through lateral induction (i.e. advection

across a changing MLD) supported by the strong mixed layer front found on the southern flank of the formation region, which is characterized by large surface density gradients (Suga et al. 2004).

Table 1: North Pacific Mode Water temperature, salinity and density characteristic definitions used for this study. TRMW definition comes from Saito et al. (2011). All others come from Bingham and Suga (2006) and references therein.

Mode Water	Temperature	Salinity	Density
Name	Range	Range	Anomaly (σ ₀)
	(°C)	$(g kg^{-1})$	$(kg m^{-3})$
NPTW	20.0 - 24.0	34.9 – 35.5	23.6 - 25.1
ESTMW	16.0 - 22.0	34.6 - 35.4	23.9 - 26.1
STMW	15.4 – 19.7	34.6 – 35.0	24.5 – 25.9
L-CMW	12.0 – 14.5	34.3 – 34.6	25.5 - 26.3
D-CMW	6.0 - 10.0	33.7 – 34.2	25.9 – 26.9
TRMW	4.0 - 9.0	33.0 - 34.0	26.3 - 26.6

Transition Region and Dense-Central Mode Waters (TRMW 26.3 $< \sigma_0 <$ 26.6, D-CMW, 25.9 <

 σ_0 < 26.9) are formed to the north of the KBF and to the south of the polar fronts as a zonal band of deep winter mixed layers emanating from the west coast of Japan (39-43°N) and extending beyond 160°W (Oka et al., 2011). Light Central Mode Water (L-CMW, 25.5 < σ_0 < 26.3) is formed between the KE and the KBF in regions of moderate (150-225 m) mixed layers that do not change significantly along isopycnals, where there is little diapycnal flow (Suga et al. 2004) and zonal surface density gradients are small.

Central mode water (CMW) that is formed to the west of 170°E cannot be subducted into the permanent pycnocline due to the deeper and lighter mixed layers found downstream. However, CMW formed to the east of this longitude, where downstream winter mixed layers are shoaling, can be, and therefore, can also be advected into the anticyclonic circulation of the subtropical gyre (Oka et al., 2011). This picture is complicated by dedacal-scale shifts in fronts and mesocale eddy activity linked to changes is surface wind forcing that can affect CMW formation (e.g. Oka and Qiu, 2012 and references therein; Kawakami et al., 2016). The possible effects of CMW formation variability include impacts on oxygen utilization rates (Kwon et al., 2016), carbon uptake and storage rates, and sea-surface temperatures. While interannual variations (Kawakami et al., 2016) and formation region eddy activity (Dong et al., 2017) are now being studied using Argo and satellite records, less is understood about longer timescale variations and effects outside formation regions.

- *Eastern Subtropical Mode Water* (ESTMW 23.9 $< \sigma_0 <$ 26.1) is by definition formed to the east
- of the dateline. While ESTMW forms in regions with fairly shallow MLDs (100-140 m, Katsura,
- 96 2018), which are somewhat less homogeneous along isopycnals than those of CMW,
- 97 compensating gradients in a fairly broad range of SSS and SST are responsible for minimal density
- 98 gradients, so as for CMW, low potential vorticity is achieved through to lack of diapycnal flow
- 99 (Suga et al., 2004).
- 100 North Pacific Tropical Water (NPTW 23.6 $< \sigma_0 < 25.1$), which is formed to the south of 30°N in
- the saline surface water Ekman convergence at the center of the subtropical gyre between 15°-
- 102 25°N, 160°E-120°W (Katsura et al, 2013) is also of interest because its 30°N limit may impact
- the farfield distribution of Cs_F. While warmer and saltier than North Pacifc MW, it partially
- overlaps ESTMW and STMW geographically as well as in density space.
- A number of MW characteristic definitions exist in the literature (e.g. Hautula and Reommich,
- 106 1998; Mecking and Warner, 2001; Oka and Suga, 2005; Qu et al., 2001; Oka et al., 2007, 2011,
- 2015; Rainville et al., 2014; Katsura, 2018) some of which suggest narrower or slightly offset
- ranges. We use those described in Table 1. Across the broader Pacific, these waters have been
- recognized by their natural physical characteristics obtained at the time of formation. Here, we
- have the opportunity to use an anthropogenic tracer (i.e. Cs_F) that was unintentionally introduced
- to the ocean at a time and location that coincided with water mass formation.

112 1.2 Radiocesium in the Pacific

- 113 History of Ocean Cs prior to FDNPP Release: The largest input of cesium isotopes to the ocean
- is fallout from atmospheric nuclear weapons testing in the late 1950s and early 1960s, which was
- highest in mid-latitudes in the northern hemisphere (e.g. Aarkrog, 2003). Programs in the 1960s
- and 1970s began to measure the weapon's testing Cs (Cs_{WT}) oceanic distribution along with several
- other isotopes (e.g. tritium, ¹⁴C, ⁹⁰Sr, and Pu). Its soluble nature (e.g. <0.1% of Cs is caught on a
- one-micron filter; Buesseler et al., 2012) allows Cs to be used as an ocean circulation tracer.
- Additional ¹³⁷Cs inputs to the ocean from reprocessing plants in Europe peaked in the 1970s, but
- did not impact Pacific waters. In 1986, Chernobyl became the largest accidental release of ¹³⁷Cs.
- Although the total atmospheric release of radioisotopes from Chernobyl was about ten times that
- of FDNPP (Steinhauser et al., 2014), only a small fraction was deposited in the North Pacific.
- Therefore, the 15-20 PBq (e.g. Inomata et al., 2018; Aoyama et al., 2019) of input via atmosphere

- and ocean from the 2011 FDNPP accident represents the largest accidental source of radioisotopes
- to this basin. It increased the existing 2011 North Pacific Cs inventory of 69 PBq (Aoyama et al.,
- 126 2012) by 20-27% (Inomata et al., 2016; Aoyama et al 2016b, 2019).
- With its short half-life, ¹³⁴Cs uniquely "tags" water as being of Fukushima origin (i.e. Cs_F). The
- longer half-life of the ¹³⁷Cs isotope means that the ¹³⁷Cs signal that entered the surface ocean during
- nuclear weapons testing prior to the treaty signed in 1963 is still measurable today (Bowen et al.,
- 130 1980; United Nations, 2000). More than half a century of ocean mixing and dispersion has lowered
- the concentrations of this weapons-based isotope (Cs_{WT}) to background levels of 1.0–2.0 Bq m⁻³
- 132 (Aoyama and Hirose, 2008; Inomata et al., 2009; Aoyama et al., 2012). Due to the lack of deep
- water formation fallout delivery to the surface Pacific Ocean has tended to remain in the upper
- water column. How deeply the ¹³⁷Cs has been mixed down over the last half century in the eastern
- North Pacific and the effects of this mixing on the uniformity of the Cs_{WT} background in the region
- will be discussed in Section 3.3.
- 137 FDNPP Radionuclide Release: The FDNPP, which includes six reactors, is located just north of
- the Kuroshio separation point on the northeast coast of Japan (37°25'N, 142°2'E, Fig. 1). In March
- 139 2011, the offshore earthquake, and resulting tsunami and aftershocks caused a loss of power at the
- 140 FDNPP. Soon thereafter, destabilization led to both a release of radioactive materials to the
- atmosphere (i.e. widespread atmospheric deposition AD) through venting, hydrogen explosions
- and fire, and a leakage of coolant water through direct discharge into the ocean, i.e., the point-
- source direct ocean discharge DOD just off the FDNPP (Chino et al., 2011; Butler, 2011;
- NSCJ, 2011, Buesseler et al., 2017). Both AD and DOD had a ¹³⁴Cs/¹³⁷Cs ratio of 0.99±0.03
- 145 (Buesseler et al., 2011). While no direct fallout observations over the oceans exist and total
- magnitudes are still subject to debate, transport models suggest that the bulk of the AD to the ocean
- occurred to the northeast of the plant (e.g. Stohl et al., 2012; Tsubono et al., 2016, Aoyama et al.,
- 148 2016b) and that about three-quarters of the contamination that entered the atmosphere landed in
- the Northwest Pacific in near-shore waters (Buesseler et al., 2017; Inomata et al., 2018).
- 150 Injected from the shoreline, at its peak on 6 April 2011, the highest Cs activities measured in the
- ocean off the FDNPP were order 68×10^6 Bq m⁻³ (Buesseler et al., 2017 and others), representing
- more than a 10 million-fold increase over prior activities. Both observational evidence and
- numerical model simulations indicate relatively rapid (within a few months) advection of

- 154 contaminants eastward (Buesseler et al., 2012; Tsumune et al., 2012; Rypina et al., 2013) into the
- highly energetic mixed water region in the confluence of the Kuroshio and Oyashio (Fig. 1). Cs
- levels decreased rapidly in the months following the accident as maxima mixed vigorously and
- moved offshore carried by the strong and highly turbulent Kuroshio Current and KE (Rossi et al.
- 158 2013). Although, 90% of the DOD occurred by 12 April, 2011, attesting to continued leakage,
- ocean Cs levels close to the FDNPP remained elevated 1000 Bq m⁻³ (outflow ≈ 10 GBq day⁻¹)
- four years after the accident (Buesseler et al., 2011, 2017) and 20 Bq m⁻³ (outflow \approx 2 GBq day⁻¹)
- seven years after (Kyodo News, 2018).

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1.3 Ocean Focused Investigations of FDNPP Radionuclides

- In 2011, the Japanese government (MEXT, 2011b) and the Tokyo Electric Power Company
- 164 (TEPCO, 2011) monitored direct ocean discharge within 30 km of the coast. Near shore
- monitoring (to within 1 km of the power plant) continues to today (TEPCO, 2017; Kyodo News,
- 166 2018). Repeat surveys have been undertaken in the western basin near Japan (e.g. Men et al., 2015,
- Kumamoto et al., 2014, 2017), but observations of the radionuclide activity in the open-ocean have
- been sporadic as opportunities have arisen. These observations include samples scattered across
- the basin (e.g. Aoyama et al., 2011, 2012, 2013, 2016a,b, 2019; Kameník et al., 2013), and more
- 170 recent samples obtained as ancillary measurements on transects (e.g. Yoshida et al., 2015;
- Kaeriyama et al., 2016; Kumamoto et al., 2016b) and in the eastern basin (Smith et al., 2015,
- 172 2017), as well as samples provided by both members of the science community and the public
- along the U.S./Canadian shoreline (ORO, 2013). Surface drifters deployed near the FDNPP in
- early summer 2011 showed trajectories crossing the basin generally following the large-scale
- ocean circulation after one year (Rypina et al 2013). From June 2011 observations it was
- determined that long-lived isolated eddies and western boundary current dynamics were the
- dominant factors defining Cs_F dispersion in the far western North Pacific on a 3-month time-scale
- 178 (Rypina et al., 2013). As the contaminated water spread horizontally across the North Pacific basin,
- the modeling studies of Behrens et al. (2012) and Rossi et al. (2013) were among the first to report
- substantial vertical intrusion of Cs into the ocean interior, especially when the surface plume met
- late-winter/spring deep pycnostads in well-known MW formation regions.
- 182 Four meridional sections at 147°E (30 Oct. 2012 5 Nov. 2012), 149°E (20 Dec. 2011 9 Feb.
- 183 2012), 155°E (25 Oct. 2012 29 Oct. 2012) and 165°E (10 Jun. 16 Jul. 2012) provided early

- observational evidence of entrainment, subduction and advection of ¹³⁴Cs in MW to the north and
- south of the Kuroshio. About 9-months after the accident, observations at 149°E suggested that
- the bulk of the ¹³⁴Cs passed through the transition region (35°-40°N) between the surface and 200
- 187 m (σ_{θ} =26.4-26.8, maximum ~16 Bq m⁻³) (Kumamoto et al., 2014). By the fall of 2012, at 147°E
- the signal remaining in the transition region was concentrated at $38^{\circ}-40^{\circ}N$ ($\sigma_{\theta}=26.1-26.4$,
- maximum ~14 Bq m⁻³) at 150 m (Kaeriyama et al., 2016, values decay corrected to Apr 6, 2011).
- At 155°E, the transition region signal had a similar maximum (~15 Bq m⁻³ at 38.5°N, σ_{θ} =26.6),
- but also a recognizable 100-300 core across from 34N° to 40°N. At 36°-38°N there were secondary
- maxima (~14 Bq m⁻³) at 400 m, σ_{θ} =26.4 that extended down to 500 m (Kaeriyama et al., 2016).
- These meridional lines also saw the subduction, capping and spreading of STMW to the south of
- the KE. In January-February 2012, a subsurface ¹³⁴Cs maximum (~18 Bq m⁻³ at 400 m) at 149°E,
- 195 34°N was observed and this 300-400 m core, although attenuating southward, was detectable as
- far south as 20°N (Kumamoto et al., 2014). By the fall of 2012, the ¹³⁴Cs core was observed at
- 197 200-400 m at 147°E, throughout the 30°-35°N region with maxima 10-14 Bq m⁻³ in the 24.6 to
- 198 26.1 σ_{θ} range (Kaeriyama et al., 2016). These observations are consistent with those made a few
- months earlier at 165°E, which presented double ¹³⁴Cs cores to the south and north of the KE (at
- $\sim 30^{\circ}$ N /150 m and $\sim 40^{\circ}$ N/300 m, respectively) in a pattern of detectable signal that spread from
- 201 ~23-46°N (Aoyama, 2016a).
- 202 Yoshida et al. (2015) investigated the spread of Cs_F on a 2-year timescale utilizing the samples
- obtained from the variety of cruises including the 2013 P02 transect that crossed the basin at 30°N.
- They found that at this latitude the leading edge of the surface Cs_F signal had reached 174°W by
- 205 the spring of 2012. A year later, with an average advection speed of 5 cm s⁻¹ it was observed at
- 206 160.6°W. This rate of spread at 30°N agreed well with drifter and model estimates (Aoyama et al.
- 207 2016a). Although the 30°N subsurface data set was sparse, it was found that two years after the
- 208 accident the maximum Cs_F signal west of 173.3°E lay at ~300 m, but shoaled sharply to 200 m
- 209 near the dateline and remained subsurface until the signal disappeared at 160.6°W.
- 210 At 40°N, Cs_F had reached the dateline by July 2012 suggesting an average advection speed of 8.5
- 211 cm s⁻¹, which slowed to 5~ cm s⁻¹ by the time the signal reached 160° to 170°W (Yoshida et al.,
- 212 2015; Inomata et al., 2018). In the northeast, Cs levels were being monitored along Line-P (Fig.
- S1). Here, Smith et al., (2015) first detected ¹³⁷Cs greater than the background (which they took to

- be ~1 Bq m⁻³) ~1500 km west of the Canadian coast in June 2012. A year later, order 1 Bq m⁻³
- values were observed on the Canadian Shelf and by early 2014, ¹³⁷Cs values as high as 2 Bq m⁻³
- were observed throughout Line-P (Smith et al., 2017). These results were corroborated by 2014
- observations obtained between Dutch Harbor, AK and Eureka, WA, where a clear FDNPP surface
- signal with ¹³⁴Cs values of 2-4 Bq m⁻³ was found in the Alaska Gyre along with one higher value
- 219 (134 Cs = 5.4 Bq m⁻³) off the coast WA (Yoshida et al., 2015).
- Compiled surface ¹³⁷Cs measurements obtained between July 2011 and December 2012 across a
- broad expanse of the North Pacific Ocean (Aoyama et al., 2016a) suggest that in the Northeast
- Pacific, after the first few months, the effects of Fukushima sourced AD could not be distinguished
- from the weapons testing background. This result is supported by the findings of Buesseler et al.,
- 224 (2017) in their compilation of surface data ¹³⁷Cs that extends to April 2016 (see their figure 2) and
- Kumamoto et al. (2016b) who concluded that the main bulk of the AD occurred in the Bering Sea
- and between 25°N and 50°N in the northwestern basin. So, while there was certainly some
- 227 atmospheric delivery of Cs_F to the Northeast Pacific, the early lack of substantially elevated ¹³⁴Cs
- in the surface ocean to the east of the dateline suggests that Cs_F found at 152°W in the spring of
- 229 2015, whether AD or DOD, was input to the ocean in the west and advected eastward. As the
- 230 ¹³⁴Cs/¹³⁷Cs ratio in the AD and DOD were the same, the two are indistinguishable in our Cs data
- set. Therefore, we presume that the ¹³⁴Cs signal observed in 2015 at 152°W is derived from a
- combination of the FDNPP DOD and AD that entered the ocean in the western basin. Likewise,
- we assume that patterns of ¹³⁷Cs that mimic those of ¹³⁴Cs are from an FDNPP source (we will
- come back to this in Section 3.3).
- Here, we present the 2015 distribution of Cs_F in the Northeast Pacific along 152°W. Using these
- observations, we seek to elucidate the history of mode waters that have traversed the basin over
- the previous 4 years.

2. The 2015 Radionuclide Dataset, Chemical Analysis and Distribution

239 2.1 Sampling

- In the spring of 2015 (Table 2) 395 radionuclide samples were obtained from the GO-SHIP
- occupation of the 152°W line P16N (Figs. 1, S1) beginning with twenty 12-sample radionuclide-
- only casts each going from ~10 m to ~1000 m. Use of the underway seawater system on station

and efficient use of water remaining in the bottles after all other sampling had been completed on regular hydrographic casts produced another 155 samples.

These included surface samples obtained from the underway seawater system as the ship steamed from the outer boundary of the U.S. Exclusive Economic Zone (EEZ) into Seattle, a few short profiles in shallower waters coming up the slope toward Kodiak and Sitka, and about 50 surface/subsurface pairs. This sampling scheme provided ~1° spacing along 152°W and a more broadly spaced survey across the Alaska Gyre (Fig. 1, S1). The 'cross-gyre' transect began at 54.1°N, 151.1°W, 100 km east of the meridional line and with an average latitude of 55.5°N followed a portion of the 1993 WOCE P17N survey track. Of the 395 samples taken, 292 were on the 152°W line, 86 were on the cross-gyre transect, 15 were underway samples in the U.S. EEZ, and 2 were lost due to technical difficulties reducing the final number to 393.

Table 2: Start and end times and locations of the 2015 P16N radiocesium sampling.

Component	Start	End	Month/Day	Month/Day
	Location	Location	Start	End
Leg 1	3.1°N, 152.0°W	21.5°N, 152.0°W	4/29/2015	5/11/2015
Leg 2	22.5°N, 152.0°W	56.4°N, 152.0°W	5/27/2015	6/18/2015
Cross-Gyre	54.1°N, 151.1°W	56.8°N, 136.0°W	6/19/2015	6/23/2015
Underway	54.3°N, 134.9°W	48.5°N, 124.8°W	6/24/2015	6/27/2016

2.2 Chemical Analysis

Here we present results from three radioisotopes: 134 Cs, 137 Cs, and briefly 90 Sr (half-life = 28.90 yeas). Approximately 20 L seawater samples were collected into cubitainers via CTD rosette for 137 Cs, 134 Cs and 90 Sr analyses. The samples were analyzed onshore at the Buesseler Laboratory at Woods Hole Oceanographic Institution using an ion-exchange resin-based method– potassium-nickel hexacyanoferrate (II) (KNiFC-Pan) (Šebasta 1997; Breier et al., 2016) for γ-counting on high-purity germanium well-type detectors for an average counting time of 50 hours.

Measurement errors include propagated recovery error (average 2.0±0.0%) and detector count error (varies by sample). The average recovery was 98.5%±1.7% with a range of 81%-100% and a median 99.0%. Decay correction ¹³⁴Cs (¹³⁷Cs) measurement error ranged from 0.02 to 1.30 (0.01 to 0.60) Bq m⁻³. Determined at the time of measurement, the average detection limits for both ¹³⁴Cs and ¹³⁷Cs are 0.1 Bq m⁻³ for Buesseler Laboratory measurements (Pike et al., 2012) and 0.03 Bq

- m⁻³ for the few low count samples that were sent to the HADES Laboratory (HADES, 2020). For example, the deep 650 dbar ¹³⁴Cs at 35°N (0.04±0.02 Bq m⁻³) was measured underground at HADES. Of samples sent to the underground lab, WHOI (HADES) found detectable ¹³⁷Cs in 43 (71) and detectable ¹³⁴Cs in 1 (25). No samples south of 30°N were measured at HADES. Note, no samples south of 30°N were measured at HADES. Henceforth, the term *significant* implies that a measurement is both greater than the detection limit and the estimated decay-corrected
- measurement error.

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273 Samples were analyzed for 90Sr in the Buesseler Laboratory via beta counting of its daughter product ⁹⁰Y (half-life = 64.1 hours) by a method adapted from Tazoe et al. (2017). Fe hydroxide 274 275 co-precipitated samples were filtered through a 25 mm QMA filter, dried, and fixed onto beta 276 mounts that were counted on Risø beta detectors (Risø National Lab; Roskilde, Denmark). The detection limit ranged from $0.2 - 0.4 \text{ Bg} \cdot \text{m}^{-3}$ for an approximately 90 hour counting time. Aliquots 277 were taken before and after adsorption for stable ¹³⁷Cs, ¹³⁴Cs, ⁹⁰Sr, and ⁹⁰Y to determine extraction 278 279 recovery via inductively coupled plasma mass spectrometer (Thermo iCAP Qc). All results 280 reported here, unless otherwise noted, are based on concentration decay corrected to the major 281 liquid discharge event date of 6 April 2011 (Buesseler et al., 2011). All sampling locations and the 282 associated Cs activities have been submitted to the US Global Ocean Carbon and Repeat

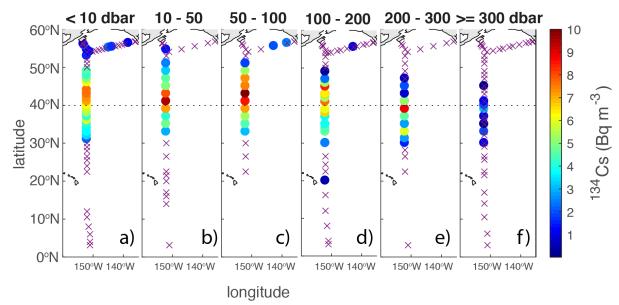
Hydrography Program (CCHDO, 2019) and the IAEA MARIS database (MARIS, 2019).

2.3 The 2015 Northeast Pacific Radionuclide Distribution

- 285 On P16N, Cs_F is evident from 30°N to the coast of Alaska (Fig. 2). Of the 393 Cs samples, 123 286 (31%) have significant ¹³⁴Cs activities (i.e. above the detection limit and greater than their 287 measurement error). As expected, due to the existence of the CswT background, a larger number 372 (95%) had significant ¹³⁷Cs. Of the 82 surface ¹³⁴Cs samples (Fig. 2a), 37 (45%) have 288 289 significant activities. There are marked southern and northern boundaries to the surface signal at 290 30°N and 49°N. Significant subsurface activities are apparent on the approach up the northern 291 slope (within the Alaska Stream) and scattered across the Alaska Gyre including in the path of the 292 Alaska Current. We return to the cross-gyre leg in the next subsection and here focus on the 292 293 152°W samples.
 - Radiocesium distribution along the 152°W Meridian:

Along 152°W with 106 (36%) of the ¹³⁴Cs samples having significant activities. 278 (95%) of the 152°W samples had significant ¹³⁷Cs signals (Fig. S2). Of the 186 samples with ¹³⁴Cs below the detection limit, 172 had detectable ¹³⁷Cs.

The minimum significant 152°W surface 134 Cs is 0.04 ± 0.02 Bq m⁻³. The five largest surface values (> 7.0 Bq m⁻³) lie along 152°W between 41°N and 44°N. The next highest (6.1 ± 0.6 Bq m⁻³) is longitudinally contiguous at 40°N. Deeper in the water column (Fig. 2b-f) the greatest activities are slightly more spread between about 39°N and 45°N. The highest 134 Cs activity (10.3±0.8 Bq m⁻³) is found at 50-100 dbar at 43°N (Fig. 2c). At 30°N all 134 Cs values are >= 1 Bq m⁻³. The southernmost significant 134 Cs measurement, which is also subsurface (140 dbar), appears as an



outlier (0.4±0.1 Bq m⁻³) above the detection limit at 20°N as no other is found south of 30°N at any depth (Fig. 2). Not so for ¹³⁷Cs. Detectable ¹³⁷Cs (Fig. S2) is found throughout the section from the southernmost profile at 3°N to the Alaska Shelf. The only break is between 12° and 22°N where there are no samples in the shallowest bin. The southern boundary of the ¹³⁴Cs signal tends northward with depth, a pattern made clearer by a latitude/pressure section plot (Fig. 3).

309 North of ~36°N, both the maximum value in each profile and the deepest detectable signals shoal 310 to the north. The Cs_F follows the shoaling of the isopycnals northward. The abrupt boundary at 311 30°N is clear in the ¹³⁴Cs sections. The ¹³⁷Cs signal confirms the ¹³⁴Cs findings (Fig. 3b, d) with maximum activities in similar 312 313 geographic, depth and density spaces. Note, that although the signal is weak, the southern 314 boundary is less abrupt giving the impression that the measurement at 20°N (that appeared as an outlier in the ¹³⁴Cs distribution) is the southernmost extension of the deepest signal shoaling to the 315 316 south. Whether or not this signal is following isopycnals will be discussed in the analysis of the 317 circulation and local Cs_{WT} background (Section 3.3). 318 A ¹³⁴Cs section (Fig. 3a, c) highlights the subsurface and compact nature of Cs_F at 152°W. The two largest activities, those greater than 10 Bg m⁻³, are both found at 43°N. The 10 largest values 319 320 are confined between 25.2 and 26.5σ₀, all MW densities. Even the outlier at 20°N and the deepest 321 detectable signal (at 645 dbar) still lie with MW density bounds as defined in Table 1.

Figure 2: Geographic distribution of detectable P16N ¹³⁴Cs in pressure bins, shallow/left to deep/right: a) 0-10 dbar; b) 10-50 dbar; c) 50-100 dbar; d) 100-200 dbar; e) 200-300 dbar; and f) below 300 dbar. Upper surface pressure boundaries are inclusive. Colors indicate magnitude of activity decay corrected to April 6, 2011. Color bar is saturated at 10 Bq m⁻³. Higher activities are plotted over lower activities, so be aware that deeper samples may overlay shallower samples. Values below the detection limit are shown by 'x' (see text). Fig. S2 is the equivalent for ¹³⁷Cs.

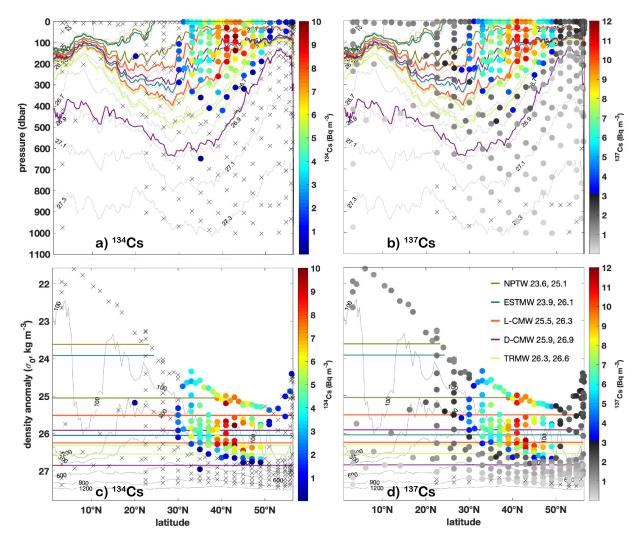


Figure 3: a) 134 Cs along 152°W in latitude-pressure space. Colors indicate magnitude of activity decay corrected to April 6, 2011. Color bar is saturated at 10 Bq m⁻³. Gray x symbols are values below detection limit; gray lines are potential density anomaly relative to 0 dbar; colored lines are boundaries for each Table 1 water mass according to the legend in (d); b) Same as (a) for 137 Cs. Color bar saturated at 12 Bq m⁻³; c) same as (a) in latitude- σ_0 space with gray curves representing pressure contours; d) Same as (c) for 137 Cs. Note, 137 Cs gray scale below 3 Bq m⁻³. This value has been chosen to facilitate comparison to the 134 Cs figure and to improve the representation of weaker signals. Although it is representative of the maximum remaining CswT in the region, it should not be taken as a hard and fast estimate as CswT has a latitudinal dependence (see Section 3.3).

Radiocesium distribution on the zonal transect crossing the Alaska Gyre: As noted earlier, some low, but significant, ¹³⁴Cs values were observed across the gyre (Fig. 2). At first glance the mainly surface signal seems best described as spots of activity; in the east, interior to the slope approaching Sitka, in the west near 152°W, and in the center of the gyre. The latter two and especially the central gyre signal reached deeper into the water column, but still only to ~140 dbar (Fig. 2c-d).

As on the meridional line, the maximum 134 Cs activity (2.5 \pm 0.4 Bq m⁻³) lies subsurface (80 dbar 329 330 at 138.3°W, 56.4°N). The section plot (Fig. 4a) provides some further insight as it suggests that 331 the ¹³⁴Cs signal is mainly located above the pycnocline. The exception is the profile taken in the 332 steeply sloping isopycnals at 149°W where ¹³⁴Cs has been subducted locally in the front or has 333 arrived at this location in the denser waters (e.g. perhaps TRMWs/D-CMWs outlined by light 334 green/purple contours) of the boundary current. Whichever the case, Cs_F is being advected around 335 the gyre in the boundary current which, according to both the density field in Fig. 4 and the P16N 336 Lowered Acoustic Doppler Current Profiler (LADCP) direct velocity measurements is lying 337 offshore of the easternmost stations. 338 The 137 Cs cross-gyre maximum (3.8 \pm 0.2 Bg m⁻³ found at the surface at 144.8°W, 55.2°N) was 339 not co-located with the ¹³⁴Cs maximum (which had an associated ¹³⁷Cs of 3.1 Bq m⁻³). Nevertheless, the pattern of ¹³⁷Cs (Fig. 4b) is similar and fills in some of the gaps left by the ¹³⁴Cs 340 341 measurements in the near surface. All ¹³⁷Cs with activity greater than the canonical background of 1-2 Bq m⁻³ were found shallower than the upper boundary of L-CMW (25.5 σ_0 , orange curve). 342 343 Note also that Fig. 4 only shows the samples going down to 300 dbar. The deepest detectable cross-344 gyre ¹³⁷Cs was found at 1000 dbar at the longitude of the deepest ¹³⁴Cs value (142.5°W, Fig. S3). However, ¹³⁷Cs was not detected everywhere along the gyre section below 500 dbar. To the west, 345 toward 152°W and away from the central portion of the cross-gyre transect, the ¹³⁷Cs signal shoals. 346 At the westernmost cross-gyre profile (151.1°W) the deepest ¹³⁷Cs signals lies at ~450 dbar, well 347 348 above the deepest cross-gyre signals at 1000 dbar.

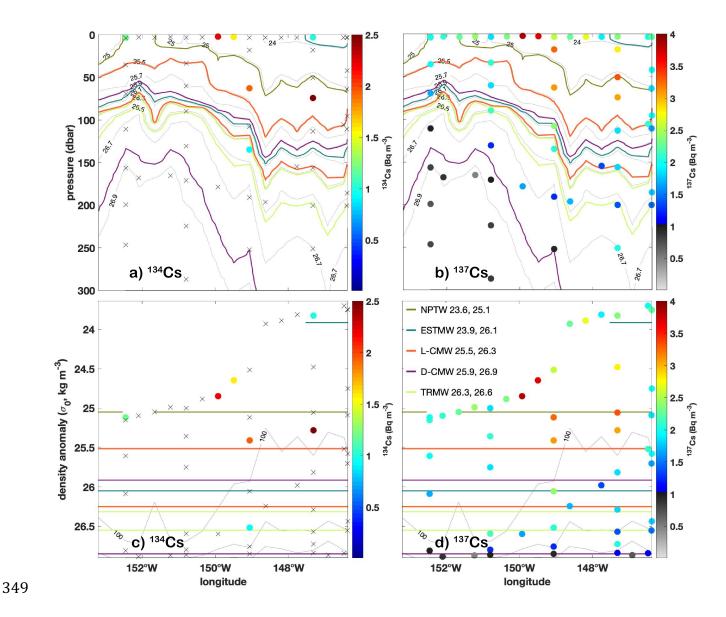


Figure 4: a) Significant 134 Cs along the cross-gyre transect in longitude-pressure space. Colors indicate magnitude of activity decay corrected to April 6, 2011. Color bar is saturated at 2.5 Bq m⁻³. Gray x symbols indicate values below detection limit; gray lines are potential density anomaly relative to 0 dbar; water mass density boundaries are indicated in color as in Figure 4; b) Same as (a) for 137 Cs. Color bar saturated at 4.0 Bq m⁻³ and a change to gray scale occurs at 1 Bq m⁻³. As for Fig. 3, gray scale does not represent C_{WT}, but is provided to improve the representation of weaker, but detectable 137 Cs signals; c) Same as (a) in longitude-σ₀ space with gray curves representing 100, 200 and 300 dbar; d) Same as (c) for 137 Cs. Note, that depths, densities and color bar are not the same as those in Fig. 3. Equivalent for 137 Cs down to 1000 dbar is provided in the supplementary material (Fig. S3).

Radiocesium distribution on underway leg into Seattle: As in the cross-gyre samples, the ¹³⁴Cs signal in the underway samples is not strong (< 2.5 Bq m⁻³), but is significant (Fig. 5). The observations indicate a stronger radionuclide signal associated with denser waters. The eastern boundary along the North American west coast is well known as an upwelling system and here we see that as surface waters become denser on the coastal approach, Cs_F traces the shoaling isopycnals where they outcrop. The ¹³⁷Cs observations (Fig. S4) support this result, though not quite as clearly as they are clouded by variable and upwelling Csw_T. Just to the north (48.5°-50°N), Line-P (Fig. S1) density sections based on observations show shoaling isopycnals – supporting the idea of inshore upwelling within 500 km of the coast in this region and timeframe (see August 2015 panels of Smith et al. (2017) figure 2).

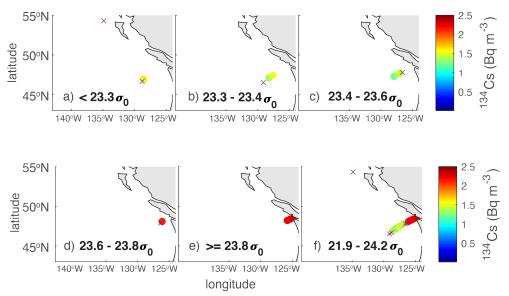


Figure 5: Surface 134 Cs from underway samples in density anomaly bins. Densities are calculated from the ship's underway seawater system temperature and salinity sensor data (see explanation in the text). a) Density less than 23.3 σ_0 ; b) 23.3-23.4 σ_0 ; c) 23.4-23.6 σ_0 ; d) 23.6-23.8 σ_0 ; e) greater than 23.8 σ_0 ; and f) the full range 21.9-24.2 σ_0 . Upper surface density boundaries are inclusive. Colors indicate magnitude of activity decay corrected to April 6, 2011. Color bar is saturated at 2.5 Bq m⁻³. Magenta x symbols indicate values less than the detection limit of 0.1 Bq m⁻³. Equivalent figure for 137 Cs is provided as supplementary Fig. S4. Equivalent figures in pressure space are not shown as these all surface measurements.

In summary, the P16N Cs sampling provides a May-June 2015 picture of Cs_F in the northeast Pacific with the deepest significant ^{134}Cs measurement at ~ 650 dbar; the ten highest values between 39°N-45°N from just below the surface to 260 dbar; a maximum signal at 43°N (70 and 145 dbar); and an abrupt cutoff at 30°N. These observations in the east are consistent with earlier findings in the west (e.g. Kumamoto et al., 2014; Kaeriyama et al., 2016) with a stronger signal

subsurface in the MW densities. In the eastern basin there is a shoaling of Cs_F northward along 152°W with the MW, an indication of the transport of a weak surface signal around the Alaska Gyre in the boundary current, and a stronger subsurface signal away from the coast at ~55°N. The interior gyre presents a 1000 dbar deep bowl shaped ¹³⁷Cs signal with the deepest values shoaling toward the coast to the east and to ~450 m to the west at 152°W. Lastly, a pattern of upwelling Cs_F was found on the coastal approach into Seattle.

3. Discussion

The timing of maximum FDNPP AD and DOD introduced to the western North Pacific in late winter/early spring and the proximate North Pacific MW formation regions made it reasonable to assume from the beginning that Cs_F would find its way into MW. As discussed earlier, many studies both observation- and model-based have borne this assumption out (e.g. Rossi et al., 2013; Kumamoto et al., 2014, 2017; Yoshida et al., 2015; Kaeriyama et al., 2016, Aoyama et al., 2016a; Kumamoto et al., 2017; Smith et al., 2017). Updating Kaeriyama et al. (2014), Kaeriyama et al. (2016) estimate that by the fall of 2012, 4.2 \pm 1.1 PBq Cs_F had penetrated STMWs, leading Inomata et al. (2018) to conclude that 2.5 \pm 0.9 PBq Cs_F had been injected into CMWs. While only 22% of the 152°W water column can be characterized as one or more of the MW according to all three Table 1 characteristics (Θ , S, and σ_0) nearly 75% have potential densities in MW ranges, so we take the opportunity to determine the relative distribution and their water column Cs_F inventory.

3.1 The Source of Mode Waters found at 152°W

The L-CMW formation region lies directly in the path of the DOD between the fronts to the north and south (Fig. 1). However, in Θ/S space (Fig. 6), while one of ten largest observed ¹³⁴Cs activities (largest dots circled in black) is associated with L-CMW densities, nine of these ten as well as the deepest signals are associated with D-CMW, and in particular a regime where D-CMW characteristics overlap those of TRMW. Kumamoto et al. (2014) and Kaeriyama et al. (2016) reported that between 147°-155°E most of signal found in the 2012 transition region (35°-40°N) was in "CMW", but more precisely it was found in waters with TRMW/D-CMW densities.

MW in 2015 at 152°W and 2011-2015 MW Formation: To explain why we see Cs_F so clearly in D-CMW and TRMW at 152°W, but far less so in L-CMW and STMW, we follow the work of Bingham and Suga (2006, henceforth BS06) to determine where and when these waters formed over the 2011-2015 timeframe and how they have been distributed since. BS06 compared the

extent of MW formation regions in 2006 based on Argo float data to the climatological average formation regions suggested by the World Ocean Atlas 2001 historical hydrography (Conkright et al., 2002). They reported that the overall distribution of suggested STMW formation sites in that single year were similar to the climatological distribution. They also found that the denser CMW formation regions were quite patchy and limited in 2006, and concluded that they varied more spatially and temporally than those of STMW. BS06 did not look at TRMW because it was not defined as a distinct MW until the following year (Saito et al., 2007, 2011).

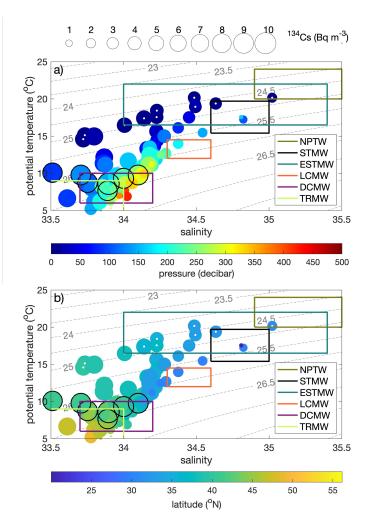


Figure 6: Potential temperature/salinity distribution of waters associated with 134 Cs samples on the 152°W line. Size indicates magnitude of activity (scale provided at top of the figure). Surface (~5 m) samples have white dots in their centers. Ten largest values are outlined in black. Colored rectangles and contours outline the Θ/S and σ_0 space for each water mass as defined in Table 1, respectively. Colors match those used in Fig. 4. Circle colors indicate a) pressure and b) latitude associated with each sample. Equivalent figure for 137 Cs is provided as supplementary Fig. S5.

Here, we employ some 113 thousand 2011-2015 Argo float profiles (Argo, 2019) to identify both the formations sites and subsurface extent of North Pacific MW since the FDNPP accident. Following BS06, we used the winter (January-March) MW properties at 10 dbar to identify possible formation regions between 2011 and 2015 (Fig. 7, dark blue dots), noting that these can also represent outcrops. We interpret locations with MW properties at 100 dbar but not at the

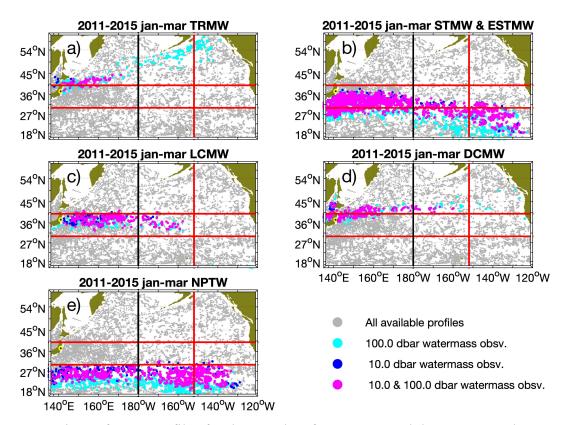


Figure 7: Locations of Argo profiles for the months of January-March between March 2011 and June 2015 (gray dots). Profiles with the temperature, salinity and density characteristics of a) TRMW, b) STMW west of the dateline and ESTMW east of the dateline, c) L-CMW, d) D-CMW and e) NPTW at 10 dbar (dark blue dots, i.e. possible formation sites), at 100 dbar (light blue dots, i.e. capped subsurface sites), and at both 10 dbar and 100 dbar (pink dots, i.e. formation sites). Water mass definitions are provided in Table 1. For reference: black line at 180° longitude, red lines at 30°N, 40°N and 152°W. Yellow circle with black dot on Japan coast indicates FDNPP.

The distribution of the Argo database MLDs (Fig. S6, based on Holte et al., (2017)) generally agrees with the combined distribution from all our Fig. 7 panels. However, it is not possible to create a one to one comparison. For example, due to the deeper STMW, CMW and TRMW formation region MLDs, NPTW formation is washed out in the Fig. S6 representation, as is the westward extension of ESTMW formation (see Katsura (2018), his figure 7).

Focusing on the areas with MW properties in the upper water column during January-March timeframe between 2011-2015 (Fig. 7 pink dots), it can be seen that while the north/south extent of ESTMW (Fig. 7b) at the dateline was similar (27°-34°N), in 2011-2015 STMW formed in a

418 narrower band (27°-37°N) than in either 2006 or the climatological past (20°-37°N) (See BS06 419 their figure 3). This difference highlights the large inter-annual variability in geographical 420 delimitation and spatial extent exhibited by these MW formation regions. While such variability 421 might be studied by treating the Argo database year-by-year, observational gaps hinder such 422 analysis. In addition, our main period of interest (2011-2015) is temporally and spatially well 423 represented by Argo, which give us confidence in our results. We therefore leave investigation of 424 inter-annual variability to future research. 425 The combination of multiple years of data, averages out inter-annual variability, returning less 426 patchy CMW formation sites (Fig. 7c-d) than the 2006 data alone. The patterns suggest the Cs_F 427 could well have been mixed/remixed into different MWs in different years as it was advected 428 through the various mid-basin formation regions. L-CMW has a geographical extent similar to the 429 climatology while D-CMW extends further south in the far west (~37°-44°N for 2011-2015 430 compared to 40°-44°N for the climatology), suggesting D-CMW formation directly off the coast 431 from the FDNPP during the 4 years of interest. The model-based results of Cedarholm et al. (2019) 432 support this suggestion, reporting that the densest varieties of MW (as dense as $26.8\sigma_{\Theta}$) were 433 formed in 2011 in the transition region. TRMW formation sites (Fig. 7a) during the more recent 434 time frame are limited to a relatively thin line (40°-43°N) west of 170°E. To the south, possible 435 NPTW formation sites span most of the basin, expanding in latitude toward the east, but remaining 436 to the south of 30°N. This is consistent with the Nie et al. (2016) analysis of ECCO model output 437 that determined that NPTW subducts within the 20°-30°N band along 150°E-130°W. Of all these 438 suggested formation regions, only ESTMW and NPTW overlap with the 152°W line. They both 439 do so to the south of 30°N, indicating MW found to the north would have to have been advected 440 into the region. 441 While all MW formation regions in the western basin lie within close proximity of the FDNPP,

the STMW region is blocked by the Kuroshio and KE fronts (e.g. Rypina et al., 2013).

Nevertheless, results from the western basin suggest that Cs_F contaminated waters have been

subducted across the front (e.g. Kumamoto et al., 2014; Garraffo et al., 2014; Yoshida et al., 2015;

Aoyama et al., 2019). These waters have entered the recirculation where STMW forms (e.g.

Aoyama et al., 2019). Such southward crossing is expected to occur more easily further out in the

basin where the current becomes more diffusive (Rypina et al. 2014), has indeed occurred (e.g.

448 Yoshida et al., 2015; Cedarholm et al. 2019).

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Looking deeper in the water column (Fig. 8) provides insight into where the various MWs exist inside and outside their formation regions. All five of the water masses discussed overlap with the 152°W line at some location and depth in the subtropics or to the north (Figs. 7-8). From the surface to 100 dbar, NPTW exists well south of 30°N, and it recedes southward with depth (Fig. 8e): a pattern that suggests that what little Cs_F found to the south of 30°N may be associated with NPTW. It is, however, also true that ESTMW (Figs. 7b, 8b) water properties exist in the same region and with a much broader fingerprint at depth. Therefore, we are not in a position to say whether the few Cs_F observations found to the south of 30°N are the result of local mixing of AD or trans-basin advection.

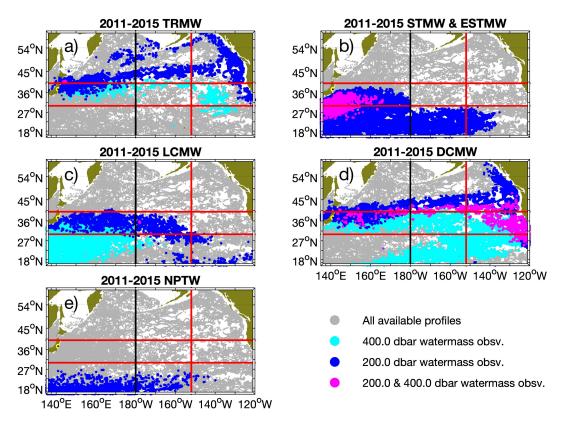


Figure 8: Same as Fig. 8 except showing all available Argo profiles between March 2011 and June 2015 (gray dots) with the specified water mass characteristics at 200 dbar (dark blue dots), 400 dbar (light blue dots), and both 200 dbar and 400 dbar (pink dots).

MW in 2015 at 152°W and Circulation: The previous discussion explains why we might expect to see the strongest 152°W Cs_F signal in D-CMW/TRMW, but does not necessarily explain the weakness of the L-CMW signal (Fig. 6a). Upon exiting their respective formation regions, L-CMW and D-CMW/TRMW are both advected eastward in the anticyclonic subtropical gyre

circulation. However, while the former does so predominantly on the southern (in)side of gyre, the latter takes the northern (out)side route. This suggests that L-CMW tagged with Cs_F may have turned southward and eastward before arriving at 152°W. This pattern is consistent with the observations of ¹³⁷Cs_{WT} in L-CMW in the subtropical gyre further east at 165°E (e.g. Aoyama et al., 2008, their figure 3). Note, that at depth, subducted L-CMW may reach the 152°W (Fig. 8c), but only to the south of 30°N where no Cs_F was found in 2015, i.e. timescales longer than 4-years may be necessary. Subsurface waters with D-CMW and TRMW properties extend to and beyond 152°W (Fig. 8a, d). At 400 dbar, (Fig. 8, light blue dots) the pressure of our deepest non-outlying Cs_F signals (Fig. 3a), both TRMW and D-CMW cross 152°W, TRMW at and just south of 40°N, and D-CMW in a broad region from ~43°N to 18°N.

At these pressures, TRMW and D-CMW properties are found in a sweeping curve around the subtropical gyre – a signal not seen in the 2015 Cs_F observations, which are limited by the 4-year timescale. A similar timescale (~ 3-5 years) is necessary for the core of the radionuclide plume to be simulated at about 500 m near 150°W/30°N (Rossi et al. 2013, see their figure 4 and Section 3.5). Water at both 200 dbar and 400 dbar (pink dots) at 152°W have D-CMW properties between about 39°-43°N and so spans of our maximum Cs_F signal. At 200 dbar, D-CMW/TRMW properties overlap at ~45°N and extend around the boundary of the gyre in the Alaska Current, but only TRMW properties are seen further west in the Alaska Stream. They are also seen at 100 dbar in this boundary current (Fig. 7a light blue dots). Both patterns suggest that near Sitka, the measured Cs_F could be associated with upwelled versions of either of these water masses, but it is TRMW that has brought Cs_F fully around the Alaska Gyre to where it was measured off the Alaska Shelf near Kodiak. This process is consistent with Line-P findings (Smith et al., 2017) and the Rossi et al. (2013) modeling study.

3.2 152°W Radiocesium Inventories

To obtain inventory estimates, water properties along 152°W were interpolated (but not extrapolated) onto a regular vertical (2 dbar) and meridional grid (0.05° latitude) using a natural nearest neighbor interpolation. Given the relative lack of deep observations, all inventory calculations were performed between 0 and 700 m (a depth chosen to include our deepest ¹³⁴Cs signal). Integrating vertically (Fig. 9a) the maximum water column inventories of both ¹³⁷Cs and ¹³⁴Cs are found at ~40°N (2.9x10³ Bq m⁻² and 1.9x10³ Bq m⁻² per meter of latitude, respectively).

492 Integrating along 152°W and assuming a 1 m width, we find local along-line (3.0°-56.45°N) water column inventories of 8.3x10⁹ Bg and 3.2x10⁹ Bg for ¹³⁷Cs and ¹³⁴Cs, respectively. The difference 493 494 between these two inventories (Fig. 9a, dotted line) is our first rough estimate of the shape of the 495 Cs background at 152°W. To determine the fraction of the Cs_F inventory in each of the water 496 masses present in the region, we define three additional waters masses so as to cover most of the 497 water column above 700 m: (SSURF) surface water to the south of 30°N, defined as warmer, 498 fresher and lighter than any in Table 1; (GASURF) Gulf of Alaska surface water defined to the 499 north of 40°N and includes waters colder, fresher and lighter than those defined in Table 1; and 500 (GAIW) Gulf of Alaska Intermediate Water, which spans a range of temperatures and salinities 501 but in particular, is denser than 26.9 σ_0 (You, 2000; Kouketsu, 2007). 502 Ouantifying what can already be generally seen in the pattern of isopycnal contours in Fig. 3, 44% 503 of the 152° W water column can be associated with waters whose potential temperature (θ), salinity 504 (S) and potential density anomaly (σ_0) (rectangles in Fig. 6) lie in the range of one or more of the 505 six water masses presented in Table 1 (plus SSURF, GASURF, and GAIW). If only density is 506 used, 96% is captured, but there is so much overlap in definitions that comparison among 507 contributions is not possible. Neither of these results is surprising given the number of Cs 508 observations that do not overlap with the water mass rectangles outlined in Fig. 6 and the extreme 509 overlap in density definitions found in Table 1. Therefore, we use σ_0 along with the broad water 510 mass latitudinal limits that were identified using the Argo profile data (Section 3.1), i.e. 30°N for 511 subtropical and tropical waters and 32°N for L-CMW. With these criteria 74% of the 152°W water 512 column is captured. The ¹³⁴Cs inventory to the south of 30°N (Fig. 9b) is unreliable as there are few Cs samples and 513 only one detected 134 Cs value. Further, while θ /S are available south of 30°N, the overlap in density 514 515 limits means there is little one can say about which water mass is responsible for bringing the one 516 small Cs signal to the line other than the general conclusion that ¹³⁴Cs is present in lighter 517 subtropical waters. Between 30°N and 32°N, the effect of both CMWs becomes evident as L-518 CMW and D-CMW together account for all the Cs_F present. The prescribed 32°N cutoff in L-519 CMW (Fig. 9b pink curve) is clearly non-physical, and between 32°N and 40°N we are missing a

significant portion of the water column with the latitude- σ_0 criteria (difference between the solid

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Figure 9: a) Blue solid, red solid and black dotted curves indicate the water column inventory of ¹³⁷Cs, ¹³⁴Cs, ¹³⁷Cs-¹³⁴Cs in each square meter along the meridional 152°W line. Crosses indicate location where half the total inventory has been accumulated integrating from south to north. For ¹³⁴Cs this halfway point (40.05°N) almost coincides with maximum inventory (40.75°N). For ¹³⁷Cs the halfway point (36.35°N) is offset from the maximum (40.15°N). Red asterisk indicates FDNPP latitude; b) ¹³⁴Cs - red curve is the same as in (a), other colored curves represent the water column inventory component for each of the water masses discussed in the text. The dotted blue curve represents the sum of all water mass components. Note, due to overlap in water mass definitions the sum can be greater than the total. Likewise, because not all water is represented by these water masses the sum can be less than the total. All values are decay corrected to April 6, 2011.

30°N

latitude

40°N

50°N

60°N

0°N

10°N

20°N

524 If the formation region θ , S, and σ_0 are used to identify the L-CMW contribution, the cutoff is still 525 at 32°N, but using σ_0 alone, the sum of the D- and L-CMW contributions is one and a half times 526 the total. Therefore, for the purpose of this inventory calculation we define the missing portion as 527 modified CMW, which is either spicier (i.e. warmer and saltier) than newly formed D-CMW 528 (expanding the purple Fig. 6 rectangle up and to the right) and/or less spicy (i.e. cooler and fresher) 529 than newly formed L-CMW (expanding the orange Fig. 6 rectangle down and to the left). With 530 this modified CMW definition, of the ¹³⁴Cs signal at 152°W: 55% is associated with D-CMW, 5% 531 is L-CMW defined to exist south of 32°N, 18% is modified CMW found between 32° and 40°N 532 and another 18% is TRMW most of which is carried by waters to the north of 40°N. STMW and 533 GASURF contribute 3% and 7%, respectively and as of 2015 at 152°W less 1% of the ¹³⁴Cs had 534 made it into NPTW, SSURF and GAIW. 535 To compare to previous estimates, one needs an estimate of the total North Pacific inventory. We 536 used the spring 2015 longitude limit of the Cs_F signal suggested by the Rossi et al. (2013) ensemble 537 of simulations (see Section 3.5) to obtain a maximum western extent of 180°. To the east, we used 538 the pattern of surface ¹³⁷Cs produced by Smith et al. (2017, their figure 4, June-August panel) 539 based P16N data combined with observations from the summer 2015 Line-P and CCGS S. W. 540 Laurier cruises. Along with a vertical distribution based on the 152°W observations and an 541 assumed average Csw_T of 1.5 Bq m⁻³ this produces a total North Pacific inventory (date corrected 542 to April 6, 2011) of 13.3 PBq (7.1 PBq to the west of 152°W and 6.2 PBq to the east). 543 There are numerous uncertainties in this calculation. If the reach of the signal to the west were 544 10% more/less (to 176°E/176°W instead 180°) the total inventory estimate would 545 increase/decrease by ~1 PBq. If the magnitude of the signal to the west were 10% more/less, the 546 western component would be ~0.7 PBq larger/smaller. Distance matters in the broad Pacific. If the 547 signal core to the west lies closer to 40°N than 45°N, as does the core of the eastward flow, the 548 inventory estimate could be ~0.6 PBq larger. A 10% larger core in the unobserved region to the 549 southeast of 47°N, 152°W, could further increase the estimate by ~0.5 PBq. Therefore, a conservative range for the total ¹³⁴Cs inventory would be 11-16 PBq. Therefore, a conservative 550 551 range for the total ¹³⁴Cs inventory would be 11-16 PBq. This range is consistent with the previous 552 estimates discussed, e.g. the DOD + AD estimates of 15-16 PBq (Inomata et al., 2016; Tsubono et 553 al., 2016). How much AD and DOD individually contribute to this estimate depends on how much

of each signal reached the 152°W line as sampled here.

555 In conclusion, based on the 152°W observations, the North Pacific Cs_F inventory is dominated by 556 D-CMW and TRMW which together account for ~70% of the total. To rigorously combine the 557 2015 meridional dataset with observations taken at different times, geographic locations and 558 depths to compute a total inventory is nontrivial. The physical processes involved (3-dimensional 559 transport and dispersion) are associated with strong variability at monthly, seasonal and annual 560 time-scales making disentangling and/or correcting multiple datasets to construct an "aggregated 561 dataset" is a complex task. That said, a related project is presently underway to combine the many 562 ocean Cs observations now available (MARIS, 2019) to investigate the inventory distribution 563 throughout the full basin.

3.3 Background ¹³⁷Cs

- By focusing on the ¹³⁴Cs results, we have thus far avoided making a distinction between the
- weapons testing ¹³⁷Cs (¹³⁷Cs_{WT}) background and the more recent FDNPP contamination. Here, we
- turn our attention to ¹³⁷Cs_{WT}. Note that in the following discussion, if a specific reference year is
- not provided (e.g. "in year value was"), values are decay corrected to April 2011.
- In 1980, Bowen et al. published estimates of ¹³⁷Cs_{WT} based on a zonal GEOSECS section between
- 30-35°N and 140°E-120°W (i.e. crossing 152°W and henceforth referred to as GZ for GEOSECS
- Zonal) and a meridional section at 120-128°W, 15S°-35°N (henceforth GM), east of our section
- but covering some of the same latitudes (Fig. S7). On GZ at 152°W, ¹³⁷Cs observations suggested
- near surface values in 1973 of ~8.5 Bq m⁻³, and a subsurface maximum at 50-150 m of ~10 Bq m⁻³
- 574 3 that decreased to 5 Bq m⁻³ by 500 m and to less than 0.3 Bq m⁻³ by ~600 m. On GM there was a
- fairly linear decrease with latitude at the surface from the 8.5 Bq m⁻³ at 30-35°N to somewhere in
- 576 the range of 1.7-3.4 Bq m⁻³ at the equator. The vertical gradient was steeper further south (~5 Bq
- 577 m^{-3} at 200 m and less than 0.3 Bq m^{-3} at 300-400 m between 0°-15°N).
- Ignoring mixing and advection for now, by 2011 the surface values on GZ at 152°W would have
- decayed to 3.5 Bq m⁻³ and the subsurface maximum to 4.3 Bq m⁻³ (Fig. 10a, green dashed curve).
- Consistent with the P16N observations (Fig. 3b), the decay corrected Bowen et al. (1980) vertical
- gradient implies 2011 values of less than 0.1 Bq m⁻³ at ~600 m. Livingston and Povinec (2000)
- reported a northeast Pacific surface ¹³⁷Cs value of 3 Bq m⁻³ in 2000, which decay corrected would
- be 2.3 Bq m⁻³ in 2011, or about a two-thirds of the decay corrected Bowen (1980) value.
- Subtracting this fractional difference between the two surface estimates from the Bowen curve

blue dashed curve) that assumes the shape of the profile did not change over time.

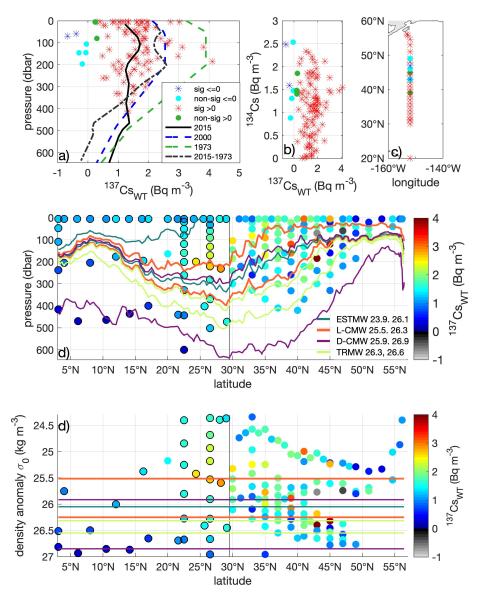


Figure 10: Estimates of 152°W ¹³⁷Cs background calculated as ¹³⁷Cs_{WT} = observed ¹³⁷Cs minus predicted ¹³⁷Cs (calculated from ¹³⁴Cs observations and a decay corrected ¹³⁷Cs/¹³⁴Cs ratio). a) Asterisks (circles) are values that are (not) significantly different from zero, where significant implies a 95% confidence level. Red/green (blue/cyan) symbols are greater (less) than 0. Black solid line is the weighted mean of the red asterisks. Historical estimates shown as green and blue dashed curves are explained in the text. Grey dot-dash curve is the green minus the black curves; b) Comparison of ¹³⁴Cs to ¹³⁷Cs_{WT} magnitude, same symbols as in (a); c) geographical location of the symbols in (a); d) latitude-pressure section of significant ¹³⁷Cs_{WT} (i.e. red asterisks in (a)) plus to the south 30°N (see separating black line), observed ¹³⁷Cs (circled in black) at locations where no ¹³⁴Cs was detectd¹³⁷Cs. Color contours represent water mass boundaries as in Fig. 3; e) Same as (d) in potential density anomaly-latitude space. (Units: Bg m⁻³)

Here we have the opportunity to estimate the shape of the background field from the 2015 observations. By decay correcting 2011 FDNPP ¹³⁴Cs/¹³⁷Cs ratio (1.0, Buesseler et al., 2011) to spring 2015 and applying it to our ¹³⁴Cs observations a predicted FDNPP-only ¹³⁷Cs estimate can be obtained. The background 137CswT estimate is the difference between the observed and predicted ¹³⁷Cs (Fig. 10a, colored symbols and black solid curve). We estimate the mean (crosssection area-averaged) 2011 northeast Pacific ¹³⁷Csw_T to be 1.2±0.1 Bq m⁻³ (1.3±0.1 Bq m⁻³ along 152°W, 1.1±0.1 Bq m⁻³ across the gyre, and 0.34±0.1 Bq m⁻³ in the surface waters along the short underway section off the coast of Seattle).

Comparing the patterns ¹³⁷Cs_{WT} (Fig. 10) suggest first that: a) the subsurface maximum vertical gradient reported by Bowen et al. (1980) still exists and in 2015 was on average 2 Bq m⁻³ at ~100 m; b) the surface latitudinal gradient they suggested may also still exist beginning at ~45°N; c) while south of 35°N the surface values are generally less than 2 Bq m⁻³, the decay corrected Livingston and Povinec (2000) value and maximum surface values are slightly higher than this estimate; and d) while the magnitude of ¹³⁷Cs_{WT} is not dependent on the magnitude of the measured ¹³⁴Cs signal (Fig. 10b), the spread in ¹³⁷Cs_{WT} decreases with decreasing ¹³⁴Cs, i.e. increasing depth. This last, speaks both to the latitudinal limits on the ¹³⁴Cs signal at depth and to the relationship between the 4- and 40-year mixing timescales.

The difference between the mean profiles suggested by data from the 1970's, the 1990's and 2015 is a result not only of the uncertainties in the estimates themselves but also mixing and stirring over time. Over the last 43 years the rate of non-decay related change in $^{137}\text{Csw}\text{T}$ (Fig. 10a, gray dot-dash curve = green curve minus black curve, henceforth $\Delta^{137}\text{Csw}\text{T}$) has developed a shape, which like the pattern of Cs_F , is at least partially related to the mixing and advection of western basin waters. While above ~50 m, lighter waters such as ESTMW dominate (Fig. 10d, above upper orange contour) and from 50-200 m, L-CMW contributes (below upper orange contour), the difference (the bulge in the gray dot-dash curve centered at about 100 m) is not significant. If one assumes that south of 30°N (black line Fig. 10d,e) where ^{134}Cs is not detectable, the distribution of ^{137}Cs (values circled in black Fig. 10d,e) represents $^{137}\text{Csw}_T$, the same conclusion holds –waters just at and above L-CMW dominate even as the isopycnals descend southward.

Between 1973 and 2015 the mean $\Delta^{137}Cs_{WT}$ above 200 m has been 0.55 \pm 0.02 Bq m⁻³ decade⁻¹.

From 200-450 m (the deepest at which multiple ¹³⁴Cs observations are available) the mean

 Δ^{137} CswT is 0.35 ± 0.15 Bq m⁻³ decade⁻¹ and is associated with a steady decrease between the two 617 depths of 14±7 Bg m⁻³ decade⁻¹ km⁻¹. As this slope is solely dependent on the Bowen et al. (1980) 618 619 1973 values (the 2015 observations suggest the 200-500 m ¹³⁷Cs_{WT} is well-mixed), it suggests that western basin mixing in the transition region into which Cs_F was discharged has been 620 621 deeper/denser than that seen on average over the last 40 years. The lack of significant gradient with depth in Δ^{137} CswT above 200 m (mid-range of western basin transition region winter MLD, 622 623 Fig. S7), suggests that in the mean for these waters mixing on the 40-year and 4-year time scales are more similar. This result is consistent with Aoyama et al. (2008) who found subsurface maxima 624

3.4 What 90Sr can and cannot tell us

in ¹³⁷Cs_{WT} at 165°N in STMW and L-CMW.

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- 627 Strontium-90 (28.79-year half-life) was also measured at some of the same locations as Cs on the
- 628 152° W line (Fig. 11, S8a-e). Like 137 Cs, there are two sources of 90 Sr, namely FDNPP (90 Sr_F) and
- 629 weapons testing fallout (90 Sr_{WT}). The latter has a 137 Cs/ 90 Sr ratio of 1.46±0.16 (Bowen et al., 1980)
- and an average surface ocean ⁹⁰Sr activity of ~1 Bq m⁻³ (Povinec et al, 2012). The 2011
- 631 (137Cs/90Sr)_F ratio was much higher (~1000, Castrillejo et al., 2015) for AD due to the low volatility
- of Sr relative to Cs and 39±1 ratio for DOD (Casacuberta et al., 2013; Buesseler et al., 2017).
- Although the AD and DOD ratios are different, observed levels of ¹³⁷Cs were not large enough to
- distinguish AD and DOD fractions (Fig. S8f-g).
- The highest 2015 152°W 137 Cs activities are order 10 Bq m⁻³. Assuming this 10 Bq m⁻³ of 137 Cs_F
- has been added to the existing system, then it follows that with (137Cs/90Sr)_F ratios of 1000 and 40,
- an additional 0.01 and 0.25 Bq m^{-3} of 90 Sr from AD and DOD would be observed, respectively.
- 638 Given the ¹³⁷Cs and ⁹⁰Sr detection limits (see Section 2.2, and note no Cs samples sent to HADES
- Lab Cs measured 90 Sr) and the 152°W surface background levels of 1-2 Bq m⁻³ for 137 Cs and \sim 1.3
- Bq m⁻³ for ⁹⁰Sr, we can conclude that additional ⁹⁰Sr_F would not be seen in the measured ¹³⁷Cs/⁹⁰Sr

Figure 11: ¹³⁷Cs predicted from: a) observed ¹³⁴Cs (method explained in Section 3.3) and b) ⁹⁰Sr (method explained in Section 3.4). Solid triangles indicate predicted ¹³⁷Cs_{WT} estimates and open triangles indicate observed ¹³⁷Cs_F. Colors in (b) represent predicted values at different latitudes along the 152°W line. Note, here values are decay corrected to the day of collection. These profiles are shown individually in Fig. S8.

ratio. Consistent with this conclusion, only one sample had ⁹⁰Sr elevated over background by more than the associated measurement error (Fig S8f).

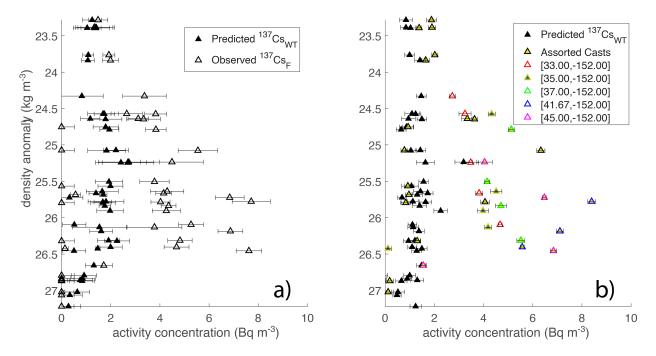


Figure 11: ¹³⁷Cs predicted from: a) observed ¹³⁴Cs (method explained in Section 3.3) and b) ⁹⁰Sr (method explained in Section 3.4). Solid triangles indicate predicted ¹³⁷Cs_{WT} estimates and open triangles indicate observed ¹³⁷Cs_F. Colors in (b) represent predicted values at different latitudes along the 152°W line. Note, here values are decay corrected to the day of collection. These profiles are shown individually in Fig. S8.

If instead we assume that all measured ⁹⁰Sr came from weapons testing, we can take advantage of the constant ¹³⁷Cs/⁹⁰Sr ratio (Fig. S8g) in that source to calculate the ¹³⁷Cs_{WT} and derive the ¹³⁷Cs_F component by differencing the observed ¹³⁷Cs and estimated ¹³⁷Cs_{WT}. This technique is similar to that used in Section 3.3. In fact, the ⁹⁰Sr method has a smaller associated error and can therefore be more useful than the ¹³⁷Cs/¹³⁴Cs method deeper in the water column where radionuclide activities are low (Fig. 11a). The ⁹⁰Sr technique suggests similar latitudinal and density gradients in ¹³⁷Cs_{WT} (Fig. 11b). In the near future, when ¹³⁴Cs has decayed beyond measuring capabilities, ⁹⁰Sr may be the only way to satisfactorily separate ¹³⁷Cs_{WT} from ¹³⁷Cs_F.

3.5 Comparison to Model Predictions

Based on observed density surfaces and the modeling work of Behrens et al. (2012), it was hypothesized that deep winter mixing in the western basin and subsequent eastward advection along isopycnal surfaces within the ocean interior would produce subsurface Cs_F maxima in our

eastern basin observations. The depth of these maxima would depend on the ventilation intensity, i.e. the depths of winter mixed layers that occurred following the discharge. A more complete scenario would be that the predominant surface advection associated with intense mixing through a rich eddy field would be the main dynamics responsible for the first (2012-2016) invasion of surface intensified Cs_F maxima in the eastern basin. Later (2014-2021), Cs-enriched MWs formed in the western basin during the previous winters would reach the subsurface Northeast Pacific through complex 3-dimensional pathways (Rossi et al. 2013, 2014; henceforth RET1314). The RET1314 simulations suggested that tracer concentrations would be primarily determined by the mixed layer concentrations at the time and location of MW formation, while the presence or absence of observed tracer along the sampled section would be mainly determined by the smallscale variability of interior pathways. In the Northeast Pacific, the model predicted surface ¹³⁷Cs_F maxima of the order of 10 Bq m⁻³ to the north of 40°N throughout 2012-2016 period, while subsurface maxima of order of 1 Bq m⁻³ were suggested south of 40°N over 2014-2021 period (RET1314). The RET1314 particle trajectory simulations based on the Ocean model For the Earth Simulator (OFES) eddy-resolving velocity fields, the most realistic in the region, provides an excellent qualitative of comparison between the 152°W observations and a model prediction (Fig. 12). Note, although we compare to model results extending to 2015, the model was not rerun for our comparison. The RET1314 model tracer is FDNPP DOD ¹³⁷Cs_F. The simulations used a 22 PBq point source (larger than our DOD+AD inventory estimate, Section 3.2) within 30 km of the FDNPP, assumed no background CswT and neglected AD. Despite these acknowledged limitations, there are many qualitative similarities between the observed and simulated 152°W Cs_F signals: the latitudinal distributions as of May 2015 (Fig. 12a) are centered in generally similar location (~25-50°N, Fig. 12b,c); they extend to a similar depth (~600 m, Fig. 12b,c); the maximum activity is the same order of magnitude (\sim 10 Bq m⁻³ compared to observed $^{137}\text{Cs}_F + ^{137}\text{Cs}_{WT}$ of \sim 12 Bq m⁻³); and maximum signal lies subsurface in the upper water column but includes a surface signature. That said, on closer inspection a number of both qualitative and quantitative differences

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become apparent.

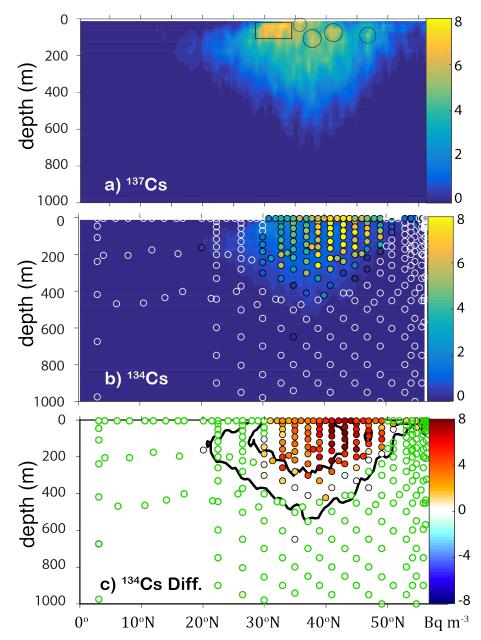


Figure 12: a) Model simulation estimate of May 2015 distribution of ¹³⁷Cs along the meridional 152°W line, black rectangle and circles highlight higher activity cores mentioned in the text; b) the same as (a) but for ¹³⁴Cs calculated using ¹³⁴Cs/¹³⁷Cs ratio decay corrected to 2015, shaded black /empty white circles indicate observed values and non-detectable samples; c) shaded black circles indicate the observed minus model ¹³⁴Cs differences, empty green circles indicate non-detectable samples, black contours are isolines of ¹³⁴Cs, where the outer line (0.25 Bq m³) broadly represents the edge of model simulated plume and the inner line (1.0 Bq m³) highlights the core of the simulated plume. Note, -8 to 8 Bq m³ color axis highlights the positive nature of these differences.

684 While the maxima are similar, the source estimates for the model (22 PBq) and observations (11-685 16 PBq) are different. Scaling the model maximum by the ratio of the estimated source sizes results 686 in a model prediction (~5-7 Bq m⁻³) that is half what is observed. Also, the 152°W distribution details are different. While the model distribution of ¹³⁷Cs_F contains pockets of higher subsurface 687 688 values at about 37°N, 41°N, and possibly, 47°N at 50-125 m (Fig. 12a, northern circles), the major 689 core lies to the south at 30-35°N, 0-100 m (Fig. 12a rectangle) and the maximum lies very close 690 to the surface (upper right corner of rectangle and southernmost circle). The observed maximum 691 core lies at 41-44°N 0-200 m with a maximum at 100 m (Figs. 3b, 12b). Note, however, that due 692 to the differences in magnitude there are few locations where simulated values are greater than 693 observed values (Fig. 12c). 694 The above ¹³⁷Cs_F comparison provides insights into the "bigger picture" and continuity with the 695 earlier literature - RET1314 and others who discuss these works, but comparing observed and modeled ¹³⁴Cs_F is more rigorous because there is no measurable ¹³⁴Cs_{WT}. Applying the same 696 ¹³⁴Cs/¹³⁷Cs ratio used in Section 3.3 to the RET1314 estimates highlights the differences between 697 698 the modeled particle trajectory results and the observations (Fig 12b). The observed ¹³⁴Cs_F 699 (maximum of ~ 10 Bq m⁻³) is significantly greater than that modeled (~ 3.5 Bq m⁻³, 1.8 - 2.5 scaling 700 by estimated source size ratio) resulting in observed minus modeled differences that are positive 701 everywhere in the region of the cores (Fig. 12c). 702 The differences in the center of the stronger southern modeled core (30°-35°N) are actually 703 somewhat less than those found to the north because the mid-range observed signal has about the 704 same magnitude as the most intense modeled values. There is not only a north/south offset in the 705 location of the core (see the offset of the inner black contour from the darkest shaded circles in

somewhat less than those found to the north because the mid-range observed signal has about the same magnitude as the most intense modeled values. There is not only a north/south offset in the location of the core (see the offset of the inner black contour from the darkest shaded circles in Fig. 12c), but the modeled signal is also more diffuse than the observed, particularly to the south where there is no indication of the observed ¹³⁴Cs 30°N cutoff (see southward extension of the outer black contour (Fig. 12c). To the north, we note that the Alaska Current signal is missing from the model (Fig. 12b, see shaded circles north of 50°N). The specific dynamics that would go into explaining the mismatches are not straightforward because as emphasized by Rossi et al. (2013), the processes that produce the distribution depend both on remote effects (e.g. three-dimensional MW pathways and tracer concentration, primarily acquired at the time and location of MW formation) and on local effects (e.g. upwelling strength, mean depth of upwelling sources, local lateral advection and mixing (see also Smith et al. (2017)).

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Modeled vs. actual velocity fields: The latitudinal locations of the Kuroshio, the KE and KBF and the diffusive nature of the model are all likely reasons for the differences between the observed and modeled distributions. The modeled results indeed consist of an ensemble of simulations (averaging out the pathways of multiple years of currents (1980-2008), i.e. multiple latitudinal locations of the core axes) and multiple years of western basin water mass formation that do not include those from the actual 2011-2015 period. It is the "ensemble approach" that produces an overall more diffuse pattern and contributes to the pronounced southward extension of the modeled cores along 152°W as it "aggregates" different cores together, allowing for more opportunities for crossing the subtropical and subpolar gyre boundaries before arrival in the eastern basin.

Modeled vs. actual water mass formation: A possible contributor to the more southerly model
Cs_F core is that between 1980 and 2008 more modeled L-CMW formed in the transition region
(Fig. 1) than between 2011 and 2012 (Kumamoto et al., 2014; Kaeriyama et al., 2016; Cedarholm
et al., 2019), which would result in a more southerly eastward flow of Cs_F (Oka and Qiu, 2012).
This highlights the non-negligible role of the prominent inter-annual variability of MW formation
and its effect on our understanding of the long-term fate of oceanic tracers and properties.

Modeled vs. actual tracer source: Another difference between modeled and observed Cs distributions is that the model did not include an AD source as it was poorly understood at the time the model runs were performed. The fact that the southern core in the model is overestimated suggests that modeled L-CMW has carried a substantial portion of the DOD signal across the basin. The region of maximum AD was to the northwest of the FDNPP (e.g. Stohl et al., 2012), that is, in the region of denser D-CMW/TRMW formation. Therefore, by neglecting such surface inputs, the model underestimates the northern core as compared to the strong D-CMW signal seen in the observations. Following this train of thought, it suggests that AD may be the dominant source of the 152°W signal.

RET1314 projected that the portion of ¹³⁷Cs_F that had been subducted into the interior ocean would return to the surface via coastal upwelling along the west coast of North America, some years after the initial surface report in 2012-2013 that was solely due to horizontal currents. They suggested that this process would take about 6-9 years after DOD, that is 2017-2021 (later than 2015). If, as discussed above, the bulk of modeled signature was advected in L-CMW on the inside edge of the subtropical gyre, rather than D-CMW/TRMW on the outside edge, its arrival in the north would

- have be slowed. It is also interesting to consider the lone ¹³⁴Cs observation obtained at 20°N. The model, like the observations, finds the tracer signal to be subsurface at this latitude, and as the model does not include AD, its signal cannot be AD mixed down into ESTMW in 2011, but must instead have been DOD advected in lighter waters.
- It is worth noting that other factors contributing to the differences are the patchiness of tracer distributions in the real ocean associated with the discrete and non-synoptic sampling that could bias the "observational" perspective. Indeed, if a southern core exists, it is possible that the sampling missed it due to submesoscale variability in the real ocean. From this perspective, our model-data comparison highlights how the model prediction and observations complement each other to reconstruct the "true" tracer fate.

4. Summary and Conclusions

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- In 2015, nearly 400 individual samples for measurement of radionuclides (134Cs, 137Cs, 90Sr) were collected along 152°W, across the Alaska Gyre and on a 200 nautical mile transect in the EEZ off Seattle. The samples presented maximum magnitudes, decay corrected to April 2011, of order of 10 Bg m⁻³ (¹³⁴Cs), 12 Bg m⁻³ (¹³⁷Cs) and 2.5 Bg m⁻³ (⁹⁰Sr). These radionuclide tracers carry a signature of MW formed in the northwest 3-4 years earlier after they had traversed some 7500 km beneath the surface to arrive at 152°W. Here, these waters penetrate to about 300 m at 30°N, 400 m at 40°N, and shoal with the density surfaces that rise toward the North American coast. Covering the distance between the FDNPP and 35°N, 152°W (location of deepest ¹³⁴Cs) in 4 years suggests a cross-basin advection speed that averages about 4.8 cm s⁻¹. This is faster than the mean western basin 3 cm s⁻¹ reported by Oka et al. (2011), but similar to the estimates reported by Kumamoto et al. (2019) for ¹³⁷Cs export from the western basin in 2011. Interestingly, it is also similar to the 4.6 cm s⁻¹ reported by Taguchi et al. (2014) as the average 0-400 m eastward propagation speed of ocean heat content anomalies crossing the basin between 40°N and 48°N (see their figure 1). This comparison brings the discussion full circle – back to one motivation for this study mentioned in the introduction (Section 1.1): information about MW circulation gleaned from Cs_F could be useful to our understanding of MW impacts on other surface-related variables such as oxygen utilization, carbon uptake and storage rates, as well as sea-surface temperature variability.
- 773 The observations suggest that more than half the Cs_F was brought to 152°W in D-CMW with 774 significant, but smaller, contributions supplied by TRMW, L-CMW, and a modified version of

either or both varieties of CMW. There was a clear cutoff in the Cs_F signature at 30°N, the estimated northern limit of subtropical and tropical waters, indicating that CMW's stayed within the North Pacific Current over this time frame and did not diffuse southward west of this longitude. This result is consistent with analysis of Argo float profiles, but inconsistent with the model-based particle trajectory experiments of Rossi et al. (2013, 2014). The core of the strongest Cs_F signal (39°-45°N) extended from just below the surface to ~260 m. Cs_F observations also indicated transport of a weak surface signal around the Alaska Gyre in the boundary current and an upwelling of Cs_F near the Seattle coast, which is consistent with Line-P findings (e.g. Smith et al., 2015, 2017). The 152°W observations suggest a basin-wide 2011 Cs inventory of 11-16 PBq.

The ¹³⁷Cs observations support the results found using FDNPP fingerprint of ¹³⁴Cs and also provide some information about the weapons testing background. Using all P16N ¹³⁴Cs measurements we find a mean 2011 Northeast Pacific ¹³⁷Cs_{WT} activity of 1.2±0.1 Bq m⁻³ (at 152°W the mean ¹³⁷Cs_{WT} is 1.3±0.1 Bq m⁻³). Over the last half century, this weapons signal has penetrated down to 900-1000 m in the Alaska Gyre (the deepest detection between 37°N and 50°N) and to ~400 m throughout the subtropical gyre and into the tropics. Earlier studies reported a latitudinal dependence for ¹³⁷Cs_{WT} in the Northeast Pacific and the 2015 Cs and Sr both suggest that this dependence, although weaker than a few decades ago, appears to have been retained.

The analysis of 152°W 2015 Cs suggests that it was the denser variety of mode waters (D-CMW/TRMW) that brought most of the signal eastward. Analyses of ARGO floats show that this is because there was formation of denser than usual waters in the transition region in the 2011-2012 timeframe. As observed by others (e.g. Inomata et al., 2018; Aoyama et al., 2016a), these waters were advected into the North Pacific Current. The comparison to the DOD based simulations of RET1314 suggests that this D-CMW/TRMW signal may also be attributable to western basin AD, rather than DOD. Although some evidence of L-CMW (commonly formed in the transition region) is seen at 152°W, with the clear cutoff in the Cs_F at 30°N (the boundary with both E-STMW and subtropical gyre waters) it is likely that the bulk of Cs_F in L-CMW was subducted and advected eastward on southern side of the KE, and drawn into the western basin recirculation. Unlike Cs_F (4-year timescale), weapons testing background (50-year timescale) does appear to be associated with L-CMW. This result is consistent with analysis of observations at 165°E (Aoyama et al. 2008).

The release of Cs_F has afforded the opportunity to employ a powerful, soluble, readily detectable tracer discharged into the ocean at a known time and location to better understand North Pacific dynamics. Moving beyond the question of the extent and speed with which FDNPP radioactivity has spread, we have begun to examine what this unique tracer can tell us about the underlying physical processes and evolution of the 2011 vintage of MWs. Future efforts will look to combine these 2015 observations with other data collected across the basin as well as model output and non-radionuclide water property observations. Together, these data will allow investigation into the role of North Pacific MW in transporting, mixing and storing the characteristics they gain from the atmosphere at the time of formation and will improve our understanding of the MW role in the cycles associated with ocean heat content, biological production, nutrient cycling as well as CO₂ uptake and ocean acidification.

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