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

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1 Abstract

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
- 27 Mode water
- 28 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 $C_{s_{\rm F}}$ 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 ⁹⁰Sr estimates to bear and compares the observations to model predictions. Section 4 is a summary. 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 Sub-arctic Gyres (Fig. 1). The western boundary Kuroshio Current separates from the coast at ~35°N as it turns into the open ocean. Bifurcating, the main portion flows east as a strong inertial 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 (Oka and Qiu, 2012).

61 In the west, to the south of the frontal regions (KE Front, KBF and further north near the Subarctic 62 Front) strong heat loss to the atmosphere results in vertical convective mixing in winter. These 63 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 64 (e.g. Whalen et al., (2012); Yoshida et al. (2015)). The resulting ventilated, low potential vorticity 65 "mode" waters that lie slightly above or within the permanent pychocline are classified into six 66 classes according to their distinct but homogeneous hydrodynamical characteristics (Table 1) and 67 are reviewed in detail by Oka and Qiu (2012). 68



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.

69 *North Pacific Subtropical Mode Water* (STMW 24.5 $< \sigma_0 < 25.9$) is formed to the south of the

- 70 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).
- 72 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 < σ_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 82 83 permanent pycnocline due to the deeper and lighter mixed layers found downstream. However, 84 CMW formed to the east of this longitude, where downstream winter mixed layers are shoaling, 85 can be, and therefore, can also be advected into the anticyclonic circulation of the subtropical gyre 86 (Oka et al., 2011). This picture is complicated by dedacal-scale shifts in fronts and mesocale eddy 87 activity linked to changes is surface wind forcing that can affect CMW formation (e.g. Oka and 88 Qiu, 2012 and references therein; Kawakami et al., 2016). The possible effects of CMW formation 89 variability include impacts on oxygen utilization rates (Kwon et al., 2016), carbon uptake and 90 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 91 92 satellite records, less is understood about longer timescale variations and effects outside formation 93 regions.

94 *Eastern Subtropical Mode Water* (ESTMW 23.9 < σ_0 < 26.1) is by definition formed to the east 95 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 101 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 103 the farfield distribution of Cs_F. While warmer and saltier than North Pacifc MW, it partially 104 overlaps ESTMW and STMW geographically as well as in density space.

105 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, 107 2015; Rainville et al., 2014; Katsura, 2018) some of which suggest narrower or slightly offset 108 ranges. We use those described in Table 1. Across the broader Pacific, these waters have been 109 recognized by their natural physical characteristics obtained at the time of formation. Here, we 110 have the opportunity to use an anthropogenic tracer (i.e. Cs_F) that was unintentionally introduced 111 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 114 is fallout from atmospheric nuclear weapons testing in the late 1950s and early 1960s, which was 115 highest in mid-latitudes in the northern hemisphere (e.g. Aarkrog, 2003). Programs in the 1960s 116 and 1970s began to measure the weapon's testing Cs (Cs_{WT}) oceanic distribution along with several 117 other isotopes (e.g. tritium, ¹⁴C, ⁹⁰Sr, and Pu). Its soluble nature (e.g. <0.1% of Cs is caught on a 118 one-micron filter; Buesseler et al., 2012) allows Cs to be used as an ocean circulation tracer.

119 Additional ¹³⁷Cs inputs to the ocean from reprocessing plants in Europe peaked in the 1970s, but

120 did not impact Pacific waters. In 1986, Chernobyl became the largest accidental release of ¹³⁷Cs.

121 Although the total atmospheric release of radioisotopes from Chernobyl was about ten times that

122 of FDNPP (Steinhauser et al., 2014), only a small fraction was deposited in the North Pacific.

123 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 127 longer half-life of the ¹³⁷Cs isotope means that the ¹³⁷Cs signal that entered the surface ocean during 128 129 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 131 the concentrations of this weapons-based isotope (C_{SWT}) 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 133 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 134 135 North Pacific and the effects of this mixing on the uniformity of the Cs_{WT} background in the region 136 will be discussed in Section 3.3.

137 FDNPP Radionuclide Release: The FDNPP, which includes six reactors, is located just north of 138 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 141 atmosphere (i.e. widespread atmospheric deposition – AD) through venting, hydrogen explosions 142 and fire, and a leakage of coolant water through direct discharge into the ocean, i.e., the point-143 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±0.03 144 (Buesseler et al., 2011). While no direct fallout observations over the oceans exist and total 145 146 magnitudes are still subject to debate, transport models suggest that the bulk of the AD to the ocean 147 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 149 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 151 ocean off the FDNPP were order 68x10⁶ Bq m⁻³ (Buesseler et al., 2017 and others), representing 152 more than a 10 million-fold increase over prior activities. Both observational evidence and 153 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 155 highly energetic mixed water region in the confluence of the Kuroshio and Oyashio (Fig. 1). Cs 156 levels decreased rapidly in the months following the accident as maxima mixed vigorously and 157 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, 159 ocean Cs levels close to the FDNPP remained elevated -1000 Bq m⁻³ (outflow ≈ 10 GBq day⁻¹) 160 four years after the accident (Buesseler et al., 2011, 2017) and 20 Bq m⁻³ (outflow \approx 2 GBq day⁻¹) 161 seven years after (Kyodo News, 2018).

162 **1.3 Ocean Focused Investigations of FDNPP Radionuclides**

163 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 165 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, 167 Kumamoto et al., 2014, 2017), but observations of the radionuclide activity in the open-ocean have 168 been sporadic as opportunities have arisen. These observations include samples scattered across 169 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; 171 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 173 along the U.S./Canadian shoreline (ORO, 2013). Surface drifters deployed near the FDNPP in 174 early summer 2011 showed trajectories crossing the basin generally following the large-scale 175 ocean circulation after one year (Rypina et al 2013). From June 2011 observations it was 176 determined that long-lived isolated eddies and western boundary current dynamics were the 177 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, 179 the modeling studies of Behrens et al. (2012) and Rossi et al. (2013) were among the first to report 180 substantial vertical intrusion of Cs into the ocean interior, especially when the surface plume met 181 late-winter/spring deep pycnostads in well-known MW formation regions.

Four meridional sections at 147°E (30 Oct. 2012 - 5 Nov. 2012), 149°E (20 Dec. 2011 - 9 Feb.
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 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°-40°N (σ_{θ} =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),

192 maxima (~14 Bq m⁻³) at 400 m, σ_{θ} =26.4 that extended down to 500 m (Kaeriyama et al., 2016).

191

but also a recognizable 100-300 core across from 34N° to 40°N. At 36°-38°N there were secondary

- 193 These meridional lines also saw the subduction, capping and spreading of STMW to the south of 194 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 196 200-400 m at 147°E, throughout the 30°-35°N region with maxima 10-14 Bq m⁻³ in the 24.6 to 197 $26.1\sigma_{\theta}$ range (Kaeriyama et al., 2016). These observations are consistent with those made a few 198 199 months earlier at 165°E, which presented double ¹³⁴Cs cores to the south and north of the KE (at 200 \sim 30°N /150 m and \sim 40°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 C_{SF} on a 2-year timescale utilizing the samples 203 obtained from the variety of cruises including the 2013 P02 transect that crossed the basin at 30°N. 204 They found that at this latitude the leading edge of the surface Cs_F signal had reached 174°W by the spring of 2012. A year later, with an average advection speed of 5 cm s⁻¹ it was observed at 205 206 160.6°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.
- At 40°N, Cs_F had reached the dateline by July 2012 suggesting an average advection speed of 8.5 cm s⁻¹, which slowed to 5~ cm s⁻¹ by the time the signal reached 160° to 170°W (Yoshida et al., 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 (¹³⁴Cs = 5.4 Bq m⁻³) off the coast WA (Yoshida et al., 2015).

- 220 Compiled surface ¹³⁷Cs measurements obtained between July 2011 and December 2012 across a 221 broad expanse of the North Pacific Ocean (Aoyama et al., 2016a) suggest that in the Northeast 222 Pacific, after the first few months, the effects of Fukushima sourced AD could not be distinguished 223 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 225 Kumamoto et al. (2016b) who concluded that the main bulk of the AD occurred in the Bering Sea 226 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 228 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 231 set. Therefore, we presume that the ¹³⁴Cs signal observed in 2015 at 152°W is derived from a 232 combination of the FDNPP DOD and AD that entered the ocean in the western basin. Likewise, 233 we assume that patterns of ¹³⁷Cs that mimic those of ¹³⁴Cs are from an FDNPP source (we will 234 come back to this in Section 3.3).
- Here, we present the 2015 distribution of C_{sF} 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.

238 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 radionuclideonly 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 onregular hydrographic casts produced another 155 samples.

245 These included surface samples obtained from the underway seawater system as the ship steamed 246 from the outer boundary of the U.S. Exclusive Economic Zone (EEZ) into Seattle, a few short 247 profiles in shallower waters coming up the slope toward Kodiak and Sitka, and about 50 248 surface/subsurface pairs. This sampling scheme provided $\sim 1^{\circ}$ spacing along 152°W and a more 249 broadly spaced survey across the Alaska Gyre (Fig. 1, S1). The 'cross-gyre' transect began at 250 54.1°N, 151.1°W, 100 km east of the meridional line and with an average latitude of 55.5°N 251 followed a portion of the 1993 WOCE P17N survey track. Of the 395 samples taken, 292 were on 252 the 152°W line, 86 were on the cross-gyre transect, 15 were underway samples in the U.S. EEZ, 253 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 |

254 2.2 Chemical Analysis

Here we present results from three radioisotopes: ¹³⁴Cs, ¹³⁷Cs, and briefly ⁹⁰Sr (half-life = 28.90 yeas). Approximately 20 L seawater samples were collected into cubitainers via CTD rosette for ¹³⁷Cs, ¹³⁴Cs and ⁹⁰Sr analyses. The samples were analyzed onshore at the Buesseler Laboratory at Woods Hole Oceanographic Institution using an ion-exchange resin-based method– potassiumnickel 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 $2.0\pm0.0\%$) and detector count error (varies by sample). The average recovery was $98.5\%\pm1.7\%$ 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.

273 Samples were analyzed for ⁹⁰Sr in the Buesseler Laboratory via beta counting of its daughter product 90 Y (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 Bq·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 283 Hydrography Program (CCHDO, 2019) and the IAEA MARIS database (MARIS, 2019).

284 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.

294 *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 ¹³⁴Cs 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 ¹³⁴Cs activity (10.3±0.8 Bq m⁻³) is found at 50-100 dbar at 43°N (Fig. 2c). At 30°N all ¹³⁴Cs values are >= 1 Bq m⁻³. The southernmost significant ¹³⁴Cs measurement, which is also subsurface (140 dbar), appears as an



304 outlier $(0.4\pm0.1 \text{ Bq m}^{-3})$ above the detection limit at 20°N as no other is found south of 30°N at 305 any depth (Fig. 2). Not so for ¹³⁷Cs. Detectable ¹³⁷Cs (Fig. S2) is found throughout the section 306 from the southernmost profile at 3°N to the Alaska Shelf. The only break is between 12° and 22°N 307 where there are no samples in the shallowest bin. The southern boundary of the ¹³⁴Cs signal tends 308 northward with depth, a pattern made clearer by a latitude/pressure section plot (Fig. 3).

North of $\sim 36^{\circ}$ N, both the maximum value in each profile and the deepest detectable signals shoal to the north. The Cs_F follows the shoaling of the isopycnals northward. The abrupt boundary at 30°N is clear in the ¹³⁴Cs sections.

The ¹³⁷Cs signal confirms the ¹³⁴Cs findings (Fig. 3b, d) with maximum activities in similar geographic, depth and density spaces. Note, that although the signal is weak, the southern 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 south. Whether or not this signal is following isopycnals will be discussed in the analysis of the circulation and local Cs_{WT} background (Section 3.3).

- 318 A 134 Cs section (Fig. 3a, c) highlights the subsurface and compact nature of Cs_F at 152°W. The
- 319 two largest activities, those greater than 10 Bq m⁻³, are both found at 43°N. The 10 largest values
- 320 are confined between 25.2 and $26.5\sigma_0$ all MW densities. Even the outlier at 20°N and the deepest
- detectable signal (at 645 dbar) still lie with MW density bounds as defined in Table 1.

Figure 2: Geographic distribution of detectable P16N 134 Cs 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 137 Cs.



322

Figure 3: a) ¹³⁴Cs 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 ¹³⁷Cs. 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 ¹³⁷Cs. Note, ¹³⁷Cs gray scale below 3 Bq m⁻³. This value has been chosen to facilitate comparison to the ¹³⁴Cs figure and to improve the representation of weaker signals. Although it is representative of the maximum remaining Cs_{WT} in the region, it should not be taken as a hard and fast estimate as Cs_{WT} has a latitudinal dependence (see Section 3.3).

- 323
- 324 *Radiocesium distribution on the zonal transect crossing the Alaska Gyre*: As noted earlier, some
- 325 low, but significant, ¹³⁴Cs values were observed across the gyre (Fig. 2). At first glance the mainly
- 326 surface signal seems best described as spots of activity; in the east, interior to the slope approaching
- 327 Sitka, in the west near 152°W, and in the center of the gyre. The latter two and especially the
- 328 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 ± 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 ¹³⁷Cs cross-gyre maximum (3.8 ± 0.2 Bq 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.



349

Figure 4: a) Significant ¹³⁴Cs 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 ¹³⁷Cs. 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 ¹³⁷Cs signals; c) Same as (a) in longitude- σ_0 space with gray curves representing 100, 200 and 300 dbar; d) Same as (c) for ¹³⁷Cs. Note, that depths, densities and color bar are not the same as those in Fig. 3. Equivalent for ¹³⁷Cs down to 1000 dbar is provided in the supplementary material (Fig. S3).

350 **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 351 352 observations indicate a stronger radionuclide signal associated with denser waters. The eastern 353 boundary along the North American west coast is well known as an upwelling system and here we 354 see that as surface waters become denser on the coastal approach, CsF traces the shoaling 355 isopycnals where they outcrop. The ¹³⁷Cs observations (Fig. S4) support this result, though not 356 quite as clearly as they are clouded by variable and upwelling Cs_{WT}. Just to the north (48.5°-50°N), 357 Line-P (Fig. S1) density sections based on observations show shoaling isopycnals – supporting the 358 idea of inshore upwelling within 500 km of the coast in this region and timeframe (see August 359 2015 panels of Smith et al. (2017) figure 2).



Figure 5: Surface ¹³⁴Cs 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 ¹³⁷Cs is provided as supplementary Fig. S4. Equivalent figures in pressure space are not shown as these all surface measurements.

- 360 In summary, the P16N Cs sampling provides a May-June 2015 picture of Cs_F in the northeast
- 361 Pacific with the deepest significant 134 Cs measurement at ~ 650 dbar; the ten highest values
- 362 between 39°N-45°N from just below the surface to 260 dbar; a maximum signal at 43°N (70 and
- 363 145 dbar); and an abrupt cutoff at 30°N. These observations in the east are consistent with earlier
- 505 115 dour, and an abrupt cutoff at 50 10. These observations in the cast are consistent with curren
- 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.

371 3. Discussion

The timing of maximum FDNPP AD and DOD introduced to the western North Pacific in late 372 373 winter/early spring and the proximate North Pacific MW formation regions made it reasonable to 374 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; 375 376 Kumamoto et al., 2014, 2017; Yoshida et al., 2015; Kaeriyama et al., 2016, Aoyama et al., 2016a; 377 Kumamoto et al., 2017; Smith et al., 2017). Updating Kaeriyama et al. (2014), Kaeriyama et al. 378 (2016) estimate that by the fall of 2012, 4.2 ± 1.1 PBq Cs_F had penetrated STMWs, leading Inomata 379 et al. (2018) to conclude that 2.5±0.9 PBq Cs_F had been injected into CMWs. While only 22% of 380 the 152°W water column can be characterized as one or more of the MW according to all three 381 Table 1 characteristics (Θ , S, and σ_0) nearly 75% have potential densities in MW ranges, so we 382 take the opportunity to determine the relative distribution and their water column Cs_F inventory.

383 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

- surface (Fig. 7, light blue dots) as capped MW formed at some recent point in time, not necessarily
 2011-2015, and locations where those properties exist at both 10 dbar and 100 dbar (Fig. 7, pink
- 409 dots) as 2011-2015 MW formation.



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.

- 410 The distribution of the Argo database MLDs (Fig. S6, based on Holte et al., (2017)) generally
- 411 agrees with the combined distribution from all our Fig. 7 panels. However, it is not possible to
- 412 create a one to one comparison. For example, due to the deeper STMW, CMW and TRMW
- 413 formation region MLDs, NPTW formation is washed out in the Fig. S6 representation, as is the
- 414 westward extension of ESTMW formation (see Katsura (2018), his figure 7).
- 415 Focusing on the areas with MW properties in the upper water column during January-March
- 416 timeframe between 2011-2015 (Fig. 7 pink dots), it can be seen that while the north/south extent
- 417 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, 442 the STMW region is blocked by the Kuroshio and KE fronts (e.g. Rypina et al., 2013). 443 Nevertheless, results from the western basin suggest that Cs_F contaminated waters have been 444 subducted across the front (e.g. Kumamoto et al., 2014; Garraffo et al., 2014; Yoshida et al., 2015; 445 Aoyama et al., 2019). These waters have entered the recirculation where STMW forms (e.g. 446 Aoyama et al., 2019). Such southward crossing is expected to occur more easily further out in the 447 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).

449 Looking deeper in the water column (Fig. 8) provides insight into where the various MWs exist 450 inside and outside their formation regions. All five of the water masses discussed overlap with the 451 152°W line at some location and depth in the subtropics or to the north (Figs. 7-8). From the 452 surface to 100 dbar, NPTW exists well south of 30°N, and it recedes southward with depth (Fig. 453 8e): a pattern that suggests that what little Cs_F found to the south of 30°N may be associated with 454 NPTW. It is, however, also true that ESTMW (Figs. 7b, 8b) water properties exist in the same 455 region and with a much broader fingerprint at depth. Therefore, we are not in a position to say 456 whether the few Cs_F observations found to the south of 30°N are the result of local mixing of AD 457 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).

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

462 circulation. However, while the former does so predominantly on the southern (in)side of gyre, the
463 latter takes the northern (out)side route. This suggests that L-CMW tagged with Cs_F may have

464 turned southward and eastward before arriving at 152°W. This pattern is consistent with the

465 observations of ¹³⁷Cs_{WT} in L-CMW in the subtropical gyre further east at 165°E (e.g. Aoyama et

466 al., 2008, their figure 3). Note, that at depth, subducted L-CMW may reach the 152°W (Fig. 8c),

467 but only to the south of 30°N where no Cs_F was found in 2015, i.e. timescales longer than 4-years

468 may be necessary. Subsurface waters with D-CMW and TRMW properties extend to and beyond

469 152°W (Fig. 8a, d). At 400 dbar, (Fig. 8, light blue dots) the pressure of our deepest non-outlying

470 Cs_F signals (Fig. 3a), both TRMW and D-CMW cross 152°W, TRMW at and just south of 40°N,

471 and D-CMW in a broad region from \sim 43°N to 18°N.

472 At these pressures, TRMW and D-CMW properties are found in a sweeping curve around the 473 subtropical gyre – a signal not seen in the 2015 Cs_F observations, which are limited by the 4-year 474 timescale. A similar timescale (~ 3-5 years) is necessary for the core of the radionuclide plume to 475 be simulated at about 500 m near 150°W/30°N (Rossi et al. 2013, see their figure 4 and Section 476 3.5). Water at both 200 dbar and 400 dbar (pink dots) at 152°W have D-CMW properties between 477 about 39°-43°N and so spans of our maximum CsF signal. At 200 dbar, D-CMW/TRMW 478 properties overlap at ~45°N and extend around the boundary of the gyre in the Alaska Current, but 479 only TRMW properties are seen further west in the Alaska Stream. They are also seen at 100 dbar 480 in this boundary current (Fig. 7a light blue dots). Both patterns suggest that near Sitka, the 481 measured CsF could be associated with upwelled versions of either of these water masses, but it is 482 TRMW that has brought Cs_F fully around the Alaska Gyre to where it was measured off the Alaska 483 Shelf near Kodiak. This process is consistent with Line-P findings (Smith et al., 2017) and the 484 Rossi et al. (2013) modeling study.

485 **3.2 152°W Radiocesium Inventories**

To obtain inventory estimates, water properties along $152^{\circ}W$ 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 ¹³⁴Cs 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 520 significant portion of the water column with the latitude- σ_0 criteria (difference between the solid red and black dotted curves) as not all water masses present are accounted for in the definitionsused.



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.

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 Cs_{WT} 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 554 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 $\sim 70\%$ 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.

564 **3.3 Background** ¹³⁷Cs

565 By focusing on the 134 Cs results, we have thus far avoided making a distinction between the 566 weapons testing 137 Cs (137 Cs_{WT}) background and the more recent FDNPP contamination. Here, we 567 turn our attention to 137 Cs_{WT}. Note that in the following discussion, if a specific reference year is 568 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 569 570 30-35°N and 140°E-120°W (i.e. crossing 152°W and henceforth referred to as GZ for GEOSECS 571 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 572 573 near surface values in 1973 of ~8.5 Bq m⁻³, and a subsurface maximum at 50-150 m of ~10 Bq m⁻ 574 ³ that decreased to 5 Bg m⁻³ by 500 m and to less than 0.3 Bg m⁻³ by \sim 600 m. On GM there was a 575 fairly linear decrease with latitude at the surface from the 8.5 Bq m⁻³ at 30-35°N to somewhere in the range of 1.7-3.4 Bq m⁻³ at the equator. The vertical gradient was steeper further south (\sim 5 Bq 576 m^{-3} at 200 m and less than 0.3 Bg m^{-3} at 300-400 m between 0°-15°N). 577

- 578 Ignoring mixing and advection for now, by 2011 the surface values on GZ at 152°W would have
- 579 decayed to 3.5 Bq m⁻³ and the subsurface maximum to 4.3 Bq m⁻³ (Fig. 10a, green dashed curve).
- 580 Consistent with the P16N observations (Fig. 3b), the decay corrected Bowen et al. (1980) vertical
- 581 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
- 583 be 2.3 Bq m⁻³ in 2011, or about a two-thirds of the decay corrected Bowen (1980) value.
- 584 Subtracting this fractional difference between the two surface estimates from the Bowen curve

(Fig. 10a, green dashed curve) as a multiplicative offset gives a profile estimate for 2000 (Fig10a,
blue dashed curve) that assumes the shape of the profile did not change over time.



Figure 10: Estimates of $152^{\circ}W^{137}Cs$ background calculated as $^{137}Cs_{WT} =$ observed ^{137}Cs minus predicted ^{137}Cs (calculated from ^{134}Cs observations and a decay corrected $^{137}Cs/^{134}Cs$ 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 ^{134}Cs to $^{137}Cs_{WT}$ magnitude, same symbols as in (a); c) geographical location of the symbols in (a); d) latitude-pressure section of significant $^{137}Cs_{WT}$ (i.e. red asterisks in (a)) plus to the south 30°N (see separating black line), observed ^{137}Cs (circled in black) at locations where no ^{134}Cs was detectd ^{137}Cs . Color contours represent water mass boundaries as in Fig. 3; e) Same as (d) in potential density anomaly-latitude space. (Units: Bq m⁻³)

587 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 588 589 spring 2015 and applying it to our ¹³⁴Cs observations a predicted FDNPP-only ¹³⁷Cs estimate can be obtained. The background ¹³⁷Cs_{WT} estimate is the difference between the observed and 590 591 predicted ¹³⁷Cs (Fig. 10a, colored symbols and black solid curve). We estimate the mean (crosssection area-averaged) 2011 northeast Pacific ${}^{137}Cs_{WT}$ to be 1.2±0.1 Bg m⁻³ (1.3±0.1 Bg m⁻³ along 592 593 152° 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 594 underway section off the coast of Seattle).

Comparing the patterns ¹³⁷Cs_{WT} (Fig. 10) suggest first that: a) the subsurface maximum vertical 595 596 gradient reported by Bowen et al. (1980) still exists and in 2015 was on average 2 Bq m⁻³ at ~100 597 m; b) the surface latitudinal gradient they suggested may also still exist beginning at \sim 45°N; c) while south of 35°N the surface values are generally less than 2 Bq m⁻³, the decay corrected 598 599 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 600 ¹³⁴Cs signal (Fig. 10b), the spread in ¹³⁷Cs_{WT} decreases with decreasing ¹³⁴Cs, i.e. increasing depth. 601 602 This last, speaks both to the latitudinal limits on the ¹³⁴Cs signal at depth and to the relationship 603 between the 4- and 40-year mixing timescales.

604 The difference between the mean profiles suggested by data from the 1970's, the 1990's and 2015 605 is a result not only of the uncertainties in the estimates themselves but also mixing and stirring 606 over time. Over the last 43 years the rate of non-decay related change in ¹³⁷Cs_{WT} (Fig. 10a, gray dot-dash curve = green curve minus black curve, henceforth $\Delta^{137}Cs_{WT}$) has developed a shape. 607 608 which like the pattern of Cs_F, is at least partially related to the mixing and advection of western 609 basin waters. While above \sim 50 m, lighter waters such as ESTMW dominate (Fig. 10d, above upper 610 orange contour) and from 50-200 m, L-CMW contributes (below upper orange contour), the 611 difference (the bulge in the gray dot-dash curve centered at about 100 m) is not significant. If one 612 assumes that south of 30°N (black line Fig. 10d,e) where ¹³⁴Cs is not detectable, the distribution of ¹³⁷Cs (values circled in black Fig. 10d,e) represents ¹³⁷Cs_{WT}, the same conclusion holds –waters 613 614 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±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 Bg 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 m¹³⁷Cs_{WT} is well-mixed), it suggests that western basin mixing in the transition region into which CsF 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} Cs_{WT} 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 624 625 in ¹³⁷Cs_{WT} at 165°N in STMW and L-CMW.

626 **3.4 What ⁹⁰Sr can and cannot tell us**

627 Strontium-90 (28.79-year half-life) was also measured at some of the same locations as Cs on the 152°W line (Fig. 11, S8a-e). Like ¹³⁷Cs, there are two sources of ⁹⁰Sr, namely FDNPP (⁹⁰Sr_F) and 628 weapons testing fallout (90 Sr_{WT}). The latter has a 137 Cs/ 90 Sr ratio of 1.46±0.16 (Bowen et al., 1980) 629 and an average surface ocean ⁹⁰Sr activity of ~1 Bq m⁻³ (Povinec et al, 2012). The 2011 630 $(^{137}Cs/^{90}Sr)_F$ ratio was much higher (~1000, Castrillejo et al., 2015) for AD due to the low volatility 631 632 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 633 distinguish AD and DOD fractions (Fig. S8f-g). 634

The highest 2015 152°W ¹³⁷Cs activities are order 10 Bq m⁻³. Assuming this 10 Bq m⁻³ of ¹³⁷Cs_F has been added to the existing system, then it follows that with $(^{137}Cs/^{90}Sr)_F$ ratios of 1000 and 40, an additional 0.01 and 0.25 Bq m⁻³ of ⁹⁰Sr from AD and DOD would be observed, respectively. Given the ¹³⁷Cs and ⁹⁰Sr detection limits (see Section 2.2, and note no Cs samples sent to HADES Lab Cs measured ⁹⁰Sr) and the 152°W surface background levels of 1-2 Bq m⁻³ for ¹³⁷Cs and ~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) 90 Sr (method explained in Section 3.4). Solid triangles indicate predicted 137 Cs_{WT} estimates and open triangles indicate observed 137 Cs_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 643 644 the constant ${}^{137}Cs/{}^{90}Sr$ ratio (Fig. S8g) in that source to calculate the ${}^{137}Cs_{WT}$ and derive the ${}^{137}Cs_{F}$ component by differencing the observed ¹³⁷Cs and estimated ¹³⁷Cs_{WT}. This technique is similar to 645 646 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 647 activities are low (Fig. 11a). The ⁹⁰Sr technique suggests similar latitudinal and density gradients 648 in ¹³⁷Cs_{WT} (Fig. 11b). In the near future, when ¹³⁴Cs has decayed beyond measuring capabilities, 649 90 Sr may be the only way to satisfactorily separate 137 Cs_{WT} from 137 Cs_F. 650

651 **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

654 along isopycnal surfaces within the ocean interior would produce subsurface Cs_F maxima in our

655 eastern basin observations. The depth of these maxima would depend on the ventilation intensity, 656 i.e. the depths of winter mixed layers that occurred following the discharge. A more complete 657 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 658 659 surface intensified Cs_F maxima in the eastern basin. Later (2014-2021), Cs-enriched MWs formed 660 in the western basin during the previous winters would reach the subsurface Northeast Pacific 661 through complex 3-dimensional pathways (Rossi et al. 2013, 2014; henceforth RET1314). The 662 RET1314 simulations suggested that tracer concentrations would be primarily determined by the 663 mixed layer concentrations at the time and location of MW formation, while the presence or 664 absence of observed tracer along the sampled section would be mainly determined by the small-665 scale 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 666 subsurface maxima of order of 1 Bq m⁻³ were suggested south of 40°N over 2014-2021 period 667 668 (RET1314).

669 The RET1314 particle trajectory simulations based on the Ocean model For the Earth Simulator 670 (OFES) eddy-resolving velocity fields, the most realistic in the region, provides an excellent 671 qualitative of comparison between the 152°W observations and a model prediction (Fig. 12). Note, 672 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 673 674 point source (larger than our DOD+AD inventory estimate, Section 3.2) within 30 km of the 675 FDNPP, assumed no background C_{SWT} and neglected AD. Despite these acknowledged 676 limitations, there are many qualitative similarities between the observed and simulated 152°W Cs_F 677 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 678 activity is the same order of magnitude (~10 Bq m⁻³ compared to observed ${}^{137}Cs_F + {}^{137}Cs_{WT}$ of ~12 679 Bq m⁻³); and maximum signal lies subsurface in the upper water column but includes a surface 680 681 signature. That said, on closer inspection a number of both qualitative and quantitative differences 682 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 $^{134}Cs_F$ 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 706 Fig. 12c), but the modeled signal is also more diffuse than the observed, particularly to the south 707 where there is no indication of the observed ${}^{134}C_8$ 30°N cutoff (see southward extension of the 708 outer black contour (Fig. 12c). To the north, we note that the Alaska Current signal is missing from 709 the model (Fig. 12b, see shaded circles north of 50°N). The specific dynamics that would go into 710 explaining the mismatches are not straightforward because as emphasized by Rossi et al. (2013), 711 the processes that produce the distribution depend both on remote effects (e.g. three-dimensional 712 MW pathways and tracer concentration, primarily acquired at the time and location of MW 713 formation) and on local effects (e.g. upwelling strength, mean depth of upwelling sources, local 714 lateral advection and mixing (see also Smith et al. (2017)).

715 Modeled vs. actual velocity fields: The latitudinal locations of the Kuroshio, the KE and KBF and 716 the diffusive nature of the model are all likely reasons for the differences between the observed 717 and modeled distributions. The modeled results indeed consist of an ensemble of simulations 718 (averaging out the pathways of multiple years of currents (1980-2008), i.e. multiple latitudinal 719 locations of the core axes) and multiple years of western basin water mass formation that do not 720 include those from the actual 2011-2015 period. It is the "ensemble approach" that produces an 721 overall more diffuse pattern and contributes to the pronounced southward extension of the modeled 722 cores along 152°W as it "aggregates" different cores together, allowing for more opportunities for 723 crossing the subtropical and subpolar gyre boundaries before arrival in the eastern basin.

724*Modeled vs. actual water mass formation:* A possible contributor to the more southerly model725 C_{SF} core is that between 1980 and 2008 more modeled L-CMW formed in the transition region726(Fig. 1) than between 2011 and 2012 (Kumamoto et al., 2014; Kaeriyama et al., 2016; Cedarholm727et al., 2019), which would result in a more southerly eastward flow of Cs_F (Oka and Qiu, 2012).728This highlights the non-negligible role of the prominent inter-annual variability of MW formation729and its effect on our understanding of the long-term fate of oceanic tracers and properties.

730 Modeled vs. actual tracer source: Another difference between modeled and observed Cs 731 distributions is that the model did not include an AD source as it was poorly understood at the time 732 the model runs were performed. The fact that the southern core in the model is overestimated 733 suggests that modeled L-CMW has carried a substantial portion of the DOD signal across the 734 basin. The region of maximum AD was to the northwest of the FDNPP (e.g. Stohl et al., 2012), 735 that is, in the region of denser D-CMW/TRMW formation. Therefore, by neglecting such surface 736 inputs, the model underestimates the northern core as compared to the strong D-CMW signal seen 737 in the observations. Following this train of thought, it suggests that AD may be the dominant 738 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.

755 4. Summary and Conclusions

In 2015, nearly 400 individual samples for measurement of radionuclides (¹³⁴Cs, ¹³⁷Cs, ⁹⁰Sr) were 756 757 collected along 152°W, across the Alaska Gyre and on a 200 nautical mile transect in the EEZ off 758 Seattle. The samples presented maximum magnitudes, decay corrected to April 2011, of order of 10 Bq m⁻³ (¹³⁴Cs), 12 Bq m⁻³ (¹³⁷Cs) and 2.5 Bq m⁻³ (⁹⁰Sr). These radionuclide tracers carry a 759 760 signature of MW formed in the northwest 3-4 years earlier after they had traversed some 7500 km 761 beneath the surface to arrive at 152°W. Here, these waters penetrate to about 300 m at 30°N, 400 762 m at 40°N, and shoal with the density surfaces that rise toward the North American coast. Covering 763 the distance between the FDNPP and 35°N, 152°W (location of deepest ¹³⁴Cs) in 4 years suggests 764 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 765 al. (2019) for ¹³⁷Cs export from the western basin in 2011. Interestingly, it is also similar to the 4.6 766 cm s⁻¹ reported by Taguchi et al. (2014) as the average 0-400 m eastward propagation speed of 767 768 ocean heat content anomalies crossing the basin between 40°N and 48°N (see their figure 1). This 769 comparison brings the discussion full circle – back to one motivation for this study mentioned in 770 the introduction (Section 1.1): information about MW circulation gleaned from Cs_F could be useful 771 to our understanding of MW impacts on other surface-related variables such as oxygen utilization, 772 carbon uptake and storage rates, as well as sea-surface temperature variability.

The observations suggest that more than half the Cs_F was brought to $152^{\circ}W$ in D-CMW with significant, but smaller, contributions supplied by TRMW, L-CMW, and a modified version of 775 either or both varieties of CMW. There was a clear cutoff in the Cs_F signature at 30°N, the 776 estimated northern limit of subtropical and tropical waters, indicating that CMW's stayed within 777 the North Pacific Current over this time frame and did not diffuse southward west of this longitude. 778 This result is consistent with analysis of Argo float profiles, but inconsistent with the model-based 779 particle trajectory experiments of Rossi et al. (2013, 2014). The core of the strongest Cs_F signal 780 (39°-45°N) extended from just below the surface to ~260 m. CsF observations also indicated 781 transport of a weak surface signal around the Alaska Gyre in the boundary current and an upwelling 782 of C_{s_F} near the Seattle coast, which is consistent with Line-P findings (e.g. Smith et al., 2015, 783 2017). The 152°W observations suggest a basin-wide 2011 Cs inventory of 11-16 PBq.

784 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 785 786 measurements we find a mean 2011 Northeast Pacific ¹³⁷Cs_{WT} activity of 1.2±0.1 Bq m⁻³ (at 152°W 787 the mean ${}^{137}Cs_{WT}$ is 1.3±0.1 Bq m⁻³). Over the last half century, this weapons signal has penetrated 788 down to 900-1000 m in the Alaska Gyre (the deepest detection between 37°N and 50°N) and to 789 \sim 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 790 791 dependence, although weaker than a few decades ago, appears to have been retained.

792 The analysis of 152°W 2015 Cs suggests that it was the denser variety of mode waters (D-793 CMW/TRMW) that brought most of the signal eastward. Analyses of ARGO floats show that this 794 is because there was formation of denser than usual waters in the transition region in the 2011-795 2012 timeframe. As observed by others (e.g. Inomata et al., 2018; Aoyama et al., 2016a), these 796 waters were advected into the North Pacific Current. The comparison to the DOD based 797 simulations of RET1314 suggests that this D-CMW/TRMW signal may also be attributable to 798 western basin AD, rather than DOD. Although some evidence of L-CMW (commonly formed in 799 the transition region) is seen at 152°W, with the clear cutoff in the Cs_F at 30°N (the boundary with 800 both E-STMW and subtropical gyre waters) it is likely that the bulk of Cs_F in L-CMW was 801 subducted and advected eastward on southern side of the KE, and drawn into the western basin 802 recirculation. Unlike Cs_F (4-year timescale), weapons testing background (50-year timescale) does 803 appear to be associated with L-CMW. This result is consistent with analysis of observations at 804 165°E (Aoyama et al. 2008).

805 The release of C_{SF} has afforded the opportunity to employ a powerful, soluble, readily detectable 806 tracer discharged into the ocean at a known time and location to better understand North Pacific 807 dynamics. Moving beyond the question of the extent and speed with which FDNPP radioactivity 808 has spread, we have begun to examine what this unique tracer can tell us about the underlying 809 physical processes and evolution of the 2011 vintage of MWs. Future efforts will look to combine 810 these 2015 observations with other data collected across the basin as well as model output and 811 non-radionuclide water property observations. Together, these data will allow investigation into 812 the role of North Pacific MW in transporting, mixing and storing the characteristics they gain from 813 the atmosphere at the time of formation and will improve our understanding of the MW role in the 814 cycles associated with ocean heat content, biological production, nutrient cycling as well as CO_2 815 uptake and ocean acidification.

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Data Sources and Availability: The P16N dataset, including the Cs measurements, was collected through the U.S. GO-SHIP program (<u>https://usgoship.ucsd.edu/</u>) and made freely available through CCHDO (2019). The Cs measurements have also been contributed to MARIS (2019) through which they are also freely available. The Argo float profiles were collected and are made freely available by the International Argo Program and the national programs that contribute to it. (<u>http://www.argo.ucsd.edu</u>, <u>http://argo.jcommops.org</u>). The Argo Program is part of the Global Ocean Observing System.

849 Contributions: A. Macdonald organized the collection of the P16N Cs data set, performed the 850 physical analysis of the Cs data, wrote the manuscript and created all figures except those 851 mentioned below. S. Yoshida, I. Rypina contributed in the physical analysis of the Cs 852 observations. S. Pike collected and shipped the P16N samples, measured many of the samples, 853 produced the original Cs data set and wrote the section on the chemical analysis. K. O. Buesseler 854 led the chemical analysis and contributed to the chemistry described herein. S. Jayne contributed 855 expertise on the mode water formation region and Northwest Pacific circulation. V. Rossi provided 856 the model-based particle tracking results and figures, and contributed to the text on the same. J. Kenyon provided the figures and contributed the text pertaining to ⁹⁰Sr. All co-authors contributed 857 858 through comments, suggestions and the editing of the full manuscript.

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