Radiocesium in the Taiwan Strait and the Kuroshio east of Taiwan from 2018 to 2019

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The release of anthropogenic radiocesium to the North Pacific Ocean (NPO) has occurred in the past 60 years. Factors controlling 137Cs (half-life, 30.2 year) and 134Cs (half-life, 2.06 year) activity concentrations in the Kuroshio east of Taiwan and the Taiwan Strait (latitude 20° N–27° N, longitude 116° E–123° E) remain unclear. This study collected seawater samples throughout this region and analyzed 134Cs and 137Cs activity concentrations between 2018 and 2019. A principal component analysis (PCA) was performed to analyze the controlling factors of radiocesium. Results of all 134Cs activity concentrations were below the detection limit (0.5 Bq m⁻³). Analyses of water column 137Cs profiles revealed a primary concentration peak (2.1–2.2 Bq m⁻³) at a depth range of 200–400 m (potential density σθ: 25.3 to 26.1 kg m⁻³). The PCA result suggests that this primary peak was related to density layers in the water column. A secondary 137Cs peak (1.90 Bq m⁻³) was observed in the near-surface waters (σθ = 18.8 to 21.4 kg m⁻³) and was possibly related to upwelling and river-to-sea mixing on the shelf. In the Taiwan Strait, 137Cs activity concentrations in the near-surface waters were higher in the summer than in the winter. We suggest that upwelling facilitates the vertical transport of 137Cs at the shelf break of the western NPO.

The global fallout from the atmospheric nuclear weapons tests during the late 1950s to early 1960s is the main source of artificial radiocesium to the world ocean1,2 and can still be discerned in the surface water, the subsurface, and deep waters (up to 600–1000 m) in the western North Pacific Ocean (NPO)3. After the Fukushima Daiichi nuclear power plant (FDNPP) accident on March 11th, 2011, additional anthropogenic radiocesium with long half-life 137Cs (30.2 year) and short half-life 134Cs (2.06 year) was released into the NPO4–11. Previous studies have estimated that the total amount of 137Cs released by the FDNPP accident ranged between 15 and 27 PBq, of which a very significant portion found its way to the atmosphere and ocean5,12,13. Traceable, FDNPP-derived 137Cs (latitude 37.42° N, longitude 141.03° E) follows the pathway of the North Pacific circulation toward the eastern NPO6,9,14. Thus 60 years after the first nuclear tests in the Pacific region, the fate of long-lives radionuclides on the western NPO and adjacent marginal seas still raises social concerns about potential health risks, e.g. associated with seafood consumption and fishing industry.

Recent studies have observed that 137Cs can disperse through the Subtropical Mode Water (STMW) and Central Mode Water (CMW) in the western NPO9,15. STMW is formed right south of the Kuroshio Current and Extension (between 132° E and near the dateline) by deep vertical convection in winter16–18. CMW forms north of the Kuroshio Extension (around 36°–41° N, 160° E–165° W), hence is characterised by colder temperatures and a deeper layer (250–500 m)19. These two water masses (i.e., STMW and CMW) circulate clockwise beneath the surface of the western NPO20. When plotted against depth, the vertical profiles of 137Cs activity concentrations recorded at 20° N, 165° E in 2002 exhibited two distinct peaks: the first one at a potential density (σθ) of 25.5 kg m⁻³ (corresponding to the σθ range of STMW) and the second one at a σθ of 26.0 kg m⁻³ (corresponding to the σθ range of CMW)21. The subsequent injection of FDNPP-derived radionuclides resulted in a contamination plume spreading eastward and merging with the Oyashio Current; hence the incorporation of radiocesium in the formation of STMW and CMW and its current value as a tracer of ocean circulation4. Both water masses (i.e., STMW and CMW) extend clockwise to the western boundary of the NPO, where the Kuroshio is most intense. Branches of the Kuroshio Current intrude into the northern South China Sea.
Subtropical marginal seas of the western NPO are characterized by a monsoon cycle and seasonal coastal cur-
rents. For instance, the Taiwan Strait is affected by the northeasterly monsoon in the winter and the southwesterly
monsoon in the summer. As a result, the northern half of the Taiwan Strait is characterized by a southward-
flowing coastal current in the winter while the southern half of the Taiwan Strait is characterized by a warm,
northward-flowing current reinforced by the Kuroshio intrusion branch in the summer. Furthermore, there are
well documented upwelling regions in the study area, such as the waters off northeastern Taiwan, several
regions in the Taiwan Strait, and regions in the northern SCS. However, to the best of our knowledge,
the effects of seasonality and upwelling on the fate of radioesium in seawater in this study area remain unclear.

This study reports the activity concentration of $^{134}$Cs and $^{137}$Cs over the shelf break of the western NPO and
examines the fates of these radionuclides on the shallow continental shelf. The $^{134}$Cs and $^{137}$Cs activity concentra-
tions were measured on samples collected in near-surface, subsurface or deeper waters in the Kuroshio region
east of Taiwan and the Taiwan Strait between 2018 and 2019. The origin of the $^{137}$Cs maximum in the subsurface/
deep waters and the factors controlling seasonal variations in $^{137}$Cs activity concentration in the near-surface
waters are discussed.

Methods

From 2018 to 2019, surface (<5 m), subsurface (5–200 m), and deep seawater (200–1000 m) samples were col-
lected at sites in the Kuroshio east of Taiwan and the Taiwan Strait (Fig. 1). Surface seawater samples (40 or 60 L)
were collected mostly from fishing boats by using cleaned 20-L tanks. Subsurface samples were taken by using
Niskin bottles mounted on a Conductivity–Temperature–Depth (CTD) rosette, which recorded temperature,
salinity (from conductivity), and water depth (from pressure) onboard R/Vs Ocean Researcher I, II, and III.
Sampling locations are shown in Fig. 1. Each 20-L sample was acidified using hydrochloric acid (11 M HCl,
salinity (from conductivity), and water depth (from pressure) onboard R/Vs Ocean Researcher I, II, and III.

As variables can be collectively controlled by major oceanographic mechanisms, principal component analysis
(PCA) serves as a multivariate analysis tool to catch the major features of a dataset. PCA can analyze inter-
correlations among these variables and reduces the dimension of a dataset. The major factors affecting this dataset
is thus obtained. The result is displayed as a subset of new, independent (orthogonal) variables which are referred
to as dimensions. The higher the coordinates of a dimension are, the greater the amount of co-variability among
the original variables this dimension explains. The results were presented graphically as plots with each dimension
and length represented the relationship and weight to the principal components, correspondingly. We applied
PCA using R software on our data set for which four variables had been determined: salinity, temperature, $\sigma_\theta$
and $^{137}$Cs activity concentrations from the surface layer to a depth of 400 m. The oceanographic context is then
used to interpret the meaning of each dimension.

Results

Surface water properties and the distribution of $^{134}$Cs and $^{137}$Cs. Surface water samples were obtained from depths of less than 5 m, with temperatures of 9.7–34.9 °C and salinities of 21.8–34.2 psu (Figs. 2 and 3a). Salinity, TA, and $\sigma_\theta$ displayed large variations in surface waters (Fig. 3). Salinities were comprised between 35 and 21 psu and $\sigma_\theta$ was usually lower than 24 kg m$^{-3}$ (Fig. 3a,b), indicating mixing between seawater and fresh water. The average TA in surface waters was 2243 ± 38 μmol kg$^{-1}$. NTA values associated with low $\sigma_\theta$ values, i.e., surface waters, deviated significantly from the average value determined in subsurface and deep waters (2309 μmol kg$^{-1}$) (Fig. 3d), implying that TA values in surface waters were likely to be affected by additional river TA sources.

In our samples, $^{134}$Cs activity concentrations were under the detection limit (0.5 Bq m$^{-3}$), and $^{137}$Cs activity concentra-
tions ranged from 0.5 to 2.0 Bq m$^{-3}$ (Fig. 3e,f), with an average of 1.2 ± 0.3 Bq m$^{-3}$ in the surface water (Supplementary Fig. S1). We also noticed a peak in the surface water $^{137}$Cs vertical profile (Fig. 3f), with the maximum value ranging from 1.95 to 1.96 Bq m$^{-3}$ ($\sigma_\theta$ = 18.8 to 21.4 kg m$^{-3}$).

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used to interpret the meaning of each dimension.
We arbitrarily divided the study area into geographic sectors and listed the average $^{137}\text{Cs}$ values for each sector in Table 1. The average values of $^{137}\text{Cs}$ activity concentrations in each sector were similar, with $^{137}\text{Cs} = 1.2 \pm 0.3 \text{ Bq m}^{-3}$ in the Taiwan Strait and $1.2 \pm 0.2 \text{ Bq m}^{-3}$ in the Kuroshio and its adjacent waters (Table 1).
In the surface waters, the $^{137}$Cs values binned in one-degree latitude bands were higher between 25 to 26° N than the ones between 21 to 22° N in the Kuroshio and its adjacent waters (approximately to the east of 121° E in our study area) (Fig. 4a). Binned $^{137}$Cs activity concentrations were close to each other to the west of 121° E (Fig. 4b).

The sea surface temperature (SST) distribution in 2018 displayed a general pattern, where SST values of less than 25 °C were found in the southern ECS and the northern Taiwan Strait while SST values over 27 °C were common for the Kuroshio and the Luzon Strait (Fig. 5). The SST in the northern Taiwan Strait and southern ECS showed strong seasonal variations, displaying SST over 25 °C during the summer-like months (i.e., July–September) and less than 25 °C during winter-like months (i.e., January–March) (Supplementary Fig. S2). The SST in shelf waters of the northern Taiwan Strait displayed stronger seasonal variations than those in the pelagic Kuroshio waters east of Taiwan.

In the shelf waters, including the Taiwan Strait and the waters off northern Taiwan, the average $^{137}$Cs activity concentration was statistically higher in August than in February (two-tailed $t$-test, $p < 0.05$) (Fig. 6a). A statistically significant relationship was observed between monthly variations of $^{137}$Cs and temperature (Fig. 6b). Monthly values of $^{137}$Cs (Fig. 6a) increased from winter to summer and again started to decrease in fall, implying a seasonal cycle of $^{137}$Cs in surface shelf waters.

Subsurface and deep water properties and distributions of $^{134}$Cs and $^{137}$Cs. Subsurface/deep water samples were taken from depths of 5–1000 m, with temperatures of 4.7–28.2 °C, and salinities over 21.0 to 34.0 psu (Fig. 2). The T–S characteristics of the subsurface and deep waters (Fig. 2b) of the Kuroshio-influenced region of the NPO covered the signals of STMW ($\sigma_\theta$: ~ 25.6 kg m$^{-3}$), Kuroshio Tropical Water (KTW, $T = 17.0$ °C, $S = 34.6$ psu), and CMW ($\sigma_\theta$ ~ 26.1 kg m$^{-3}$). TA values in the subsurface and deep waters varied in a narrow range (Fig. 3c). The average and standard deviation of NTA value for waters between 5 to 400 m was 2309 ± 5 μmol kg$^{-1}$ (Fig. 3d).

At station NTU2 in the Kuroshio region, $^{134}$Cs activities were below the detection limit (0.5 Bq m$^{-3}$), and $^{137}$Cs activities were lower than 2.5 Bq m$^{-3}$ from the surface to a depth of 1000 m (Fig. 3e). Moreover, two layers of elevated $^{137}$Cs activities were observed: $^{137}$Cs activities were mostly higher than 1 Bq m$^{-3}$ from 0 to 400 m and also higher than 2 Bq m$^{-3}$ from 200 to 400 m. By contrast, they were lower than 1 Bq m$^{-3}$ from 600 to 1000 m (Fig. 3f). A synthesis of these results showed that low $^{137}$Cs activity concentrations (1.5 to 0.6 Bq m$^{-3}$) corresponded to the high $\sigma_\theta$ (> 25 kg m$^{-3}$) waters between 400 and 1000 m (Fig. 3e,f).

Discussion

The first three dimensions of the PCA results explained 91% of all variations, including sample temperature, salinity, and $\sigma_\theta$ and $^{137}$Cs activity concentration above the depth of 400 m. Dimension 1 explained 54% of the variations (Fig. 7a,b) and was dominated by $\sigma_\theta$, temperature, and salinity (Table 2). $\sigma_\theta$, salinity, and $^{137}$Cs were positively correlated with Dimension 1 while the temperature was negatively correlated (Fig. 7a). We suggest that Dimension 1 represents density-induced water layer distributions and explains the primary peak of $^{137}$Cs in the deep waters. This suggestion is consistent with the fact that $\sigma_\theta$ is positively correlated with the variables that have a high loading on Dimension 1 (Fig. 7c) and is a marker for the two layers of elevated $^{137}$Cs in the top 400 m of the water column.

The highest coordinate of Dimension 2 corresponds to the secondary peak of $^{137}$Cs in the surface waters (Fig. 7d), implying that Dimension 2 represents a factor that led to the secondary $^{137}$Cs peak. The plot of Dimension 2 coordinates against $\sigma_\theta$ (Fig. 7d) can be divided into a high $\sigma_\theta$ (> 22 kg m$^{-3}$) arm and a low $\sigma_\theta$ (< 22 kg m$^{-3}$) arm.
Figure 3. Vertical distributions of salinity, TA, NTA, and $^{137}$Cs concentration. In the subsurface and deep waters, where (a) salinities varied from 33.7 to 34.9 long the vertical profile, (b) TA values varied from 2284 to 2360 μmol kg$^{-1}$, (c) $^{137}$Cs activity concentrations displayed the primary peak at a depth range of 200–400 m, and where (d) $\sigma_\theta$ varied from 25.2 to 26.1 kg m$^{-3}$, (e) NTA was 2309 ± 5 μmol kg$^{-1}$, (f) the primary peak of $^{137}$Cs was observed. (d) NTA with low $\sigma_\theta$ values deviated from 2309 in the near-surface waters. (f) The secondary peak of $^{137}$Cs activity concentration displayed $\sigma_\theta$ ranging from 19 to 22 kg m$^{-3}$ in the near-surface waters. All panels share the same legend. Data from NTU2 were noted with triangle markers in corresponding solid circles.
arm. As both temperature and salinity were positively correlated with Dimension 2 (Fig. 7a, Table 2), we hypothesize that the high $\sigma_\theta$ arm was caused by upwelling which transports high salinity water toward the surface and reduces the thickness of the lens of warm surface water in this study area. Upwelling near the coast of Taiwan and in the Taiwan Strait is known to transport subsurface waters (as deep as 80 m to 100 m) to the near-surface layer. The vertical velocity of upwelling off northeastern Taiwan (Fig. 1)\textsuperscript{42–45} has been estimated to be 15 m day\textsuperscript{-1} on the shelf and over 40 m day\textsuperscript{-1} at the shelf edge\textsuperscript{19,46}. The $\sigma_\theta$ of water with salinity = 34.3 psu, temperature = 18.3 °C, and $\sigma_\theta$ = 25.0 kg m\textsuperscript{-3} at a depth of 200 m, can decrease to less than 22 kg m\textsuperscript{-3} if the water temperature increases to 28 °C at 1 m during the upwelling process (Fig. 2a). This annual upwelling to the northeastern Taiwan may lead to the higher binned $^{137}$Cs between 25 and 26° N than the others to the east of 121°E in the near-surface waters (Fig. 4a). Another driving force of annual upwelling is internal tide, which can induce upwelling in the waters off northeastern Taiwan\textsuperscript{47} and off southern Taiwan\textsuperscript{48–50}. Moreover, this transition $\sigma_\theta$ of 22 kg m\textsuperscript{-3} in Fig. 7d was also consistent with the transition range of $\sigma_\theta$ where NTA positively deviated from 2309 ± 5 μmol kg\textsuperscript{-1} in the near-surface water (Fig. 3d). This deviation of NTA in low $\sigma_\theta$ waters indicates freshwater inputs from terrestrial runoff. To sum up, we suggest that mixing between riverine freshwater and seawater is responsible for the lower $\sigma_\theta$ arm while upwelling drives the higher $\sigma_\theta$ arm in Fig. 7d.

Dimension 3 accounted for no more of the variance than $^{137}$Cs itself (Table 2, Fig. 7b), suggesting that it was controlled by the chemical characteristics of $^{137}$Cs. MacKenzie et al.\textsuperscript{51} have argued that high freshwater discharge from land can remobilize $^{137}$Cs from surface sediments (< 10 cm). Future work should be conducted towards a dynamic representation of radionuclide transfer among freshwater, seawater, sediment, and the biological compartments\textsuperscript{11}.

The seasonal variation of the $^{137}$Cs activity concentration in the Taiwan Strait (Fig. 6a) reflects the seasonal intrusions of waters from Kuroshio intrusion, ECS, and SCS. Wu et al.\textsuperscript{19} corrected their $^{137}$Cs activity concentrations to the same date as this study, leading to a mean of 0.71 ± 0.27 Bq m\textsuperscript{-3} in the surface ECS and an average of 0.92 ± 0.28 Bq m\textsuperscript{-3} in the surface SCS. These results are consistent with our observation that southward-flowing cold waters with low $^{137}$Cs values intruded into the northern half of the shallow Taiwan Strait during the winter while the reverse took place during the summer (Fig. 6a)\textsuperscript{22,24,25}. In addition to the Kuroshio offshoot transporting warm waters with higher $^{137}$Cs values in the summer, the Pearl river plume can also intrude into the southern Taiwan Strait during the summer\textsuperscript{26}. It follows that the slightly lower $^{137}$Cs activity concentration during July

| Sub-division        | $^{137}$Cs Mean | STD\textsuperscript{a} |
|---------------------|-----------------|------------------------|
| Taiwan strait       | 1.2             | 0.3                    |
| KC and adjacent water| 1.2             | 0.2                    |

Table 1. Average $^{137}$Cs values for the Taiwan Strait and the Kuroshio Current (KC) east of Taiwan (2018–2019) in Bq m\textsuperscript{-3}. \textsuperscript{a}STD represents the standard deviation of the corresponding mean values.
indicates that the effect of the intrusion of Kuroshio is more than offset by the amount of warm water with lower
\(^{137}\)Cs activity originating from the SCS.

The maximum in \(^{137}\)Cs activity concentration was observed in a specific range of \(\sigma_\theta\) in the subsurface and deep
waters (Fig. 3e,f), implying a lateral transport along the 125–400 m depth horizon in addition to local atmos-
pheric fallout. Local and modern atmospheric \(^{137}\)Cs fallout can only affect the average \(^{137}\)Cs values in the surface
waters (Table 1). \(^{137}\)Cs activity concentrations in the subsurface water of the study area displayed characteristics
(\(\sigma_\theta = 25.2\) and 26.1 kg m\(^{-3}\)) which were similar to those of STMW and CMW (\(\sigma_\theta = 25.3\) to 26.3 kg m\(^{-3}\)) in NPO
(Fig. 3). Some surface NTA values and also the average NTA value in subsurface waters (2309 ± 5 μmol kg\(^{-1}\))
of this study were consistent with NTA values previously reported in the surface western NPO (2301 ± 9 to
2299 ± 5.4 μmol kg\(^{-1}\))\(^5\). The maximum \(^{137}\)Cs activity concentration at a depth of 300 to 400 m at 165° E before
the FDNPP event should be 1.65 Bq m\(^{-3}\) (corrected to January 1st, 2020)\(^3\). After the FDNPP event in June/July
2012, \(^{137}\)Cs activity concentrations at depths of between 0 and 600 m in the western NPO (25 to 45° N, 165°
E) were between 2.1 and higher than 8.4 Bq m\(^{-3}\) (corrected to January 1st, 2020)\(^4\). As the half-life of \(^{134}\)Cs is
shorter than that of \(^{137}\)Cs, \(^{134}\)Cs/\(^{137}\)Cs is assumed to be 1.000 at 165° E; it became 0.095 after 7.5 year (July 2012
December 2020). Since \(^{137}\)Cs activity concentration is already low (<2.1 Bq m\(^{-3}\)) in this study area, \(^{134}\)Cs is
expected to be <0.2 Bq m\(^{-3}\) which is lower than the detection limit (0.5 Bq m\(^{-3}\)) in this study.

Figure 5. Sea surface distributions of seawater temperature. SST is an important factor while understanding
the oceanic condition. High SST (MODIS 2018) was detected in the Kuroshio east of Taiwan and its intrusion
into the southeastern Taiwan Strait. Low SST was identified along the coastline from the East China Sea (ECS)
to the western side of the Taiwan Strait. The white dashed line indicates the contour line of \(18 \, ^\circ\)C obtained from
January. Monthly SST data was collected from AQUA-MODIS with a spatial resolution of \(1^\circ \times 1^\circ\) (https://neo.sci.
gsfc.nasa.gov/view.php?datasetId=MYD28M). This data was accessed on July 3, 2021. This image was created by
Ocean Data View (Version 4.7.5) (Schlitzer, R., Ocean Data View, https://odv.awi.de, 2016).

Figure 6. Monthly variations in sea-surface \(^{137}\)Cs activity concentration in the Taiwan Strait and southern ECS.
(a) \(^{137}\)Cs activity concentration and corresponding seawater temperature were measured at the study sites on
both sides of the Taiwan Strait and the waters off northern Taiwan (refer to Fig. 1b). Both parameters displayed
seasonal variations: low in winter and high in summer. (b) The monthly \(^{137}\)Cs activity concentration was
statistically correlated to its corresponding seawater temperature.
The increase between the estimated $^{137}\text{Cs}$ activity concentration before FDNPP ($1.65$ Bq m$^{-3}$) and the maximum measured $^{137}\text{Cs}$ value after FDNPP ($2.1$ Bq m$^{-3}$) in this study (both corrected to January 1st, 2020) implies an additional $^{137}\text{Cs}$ activity concentration of $2.1 - 1.65 = 0.45$ Bq m$^{-3}$ during this time period. This increase is the result of complex physical transportation from NPO to the study area. Kamidaira et al.\(^{55}\) reported that approximately 43% of FDNPP-derived $^{137}\text{Cs}$ could be delivered to below the mixed layer through eddy processes.

**Figure 7.** Results of principal component analysis (PCA) and sample coordinates. (a,b) $\sigma_\theta$ dominated variations in Dimension 1; increasing temperature and salinity dominated variations in Dimension 2; and $^{137}\text{Cs}$ dominated variations in Dimension 3. (c) The coordinates of individual results in Dimension 1 were consistent with the primary peak of $^{137}\text{Cs}$ defined along the stratified water column. (d) The peak coordinate of Dimension 2 was consistent with the secondary $^{137}\text{Cs}$ peak along the $\sigma_\theta$ gradient.

**Table 2.** Principal components.

|          | Dim. 1 | Dim. 2 | Dim. 3 |
|----------|--------|--------|--------|
| Temperature | − 0.659 | 0.696  | 0.284  |
| Salinity  | 0.728  | 0.682  | − 0.061 |
| $\sigma_\theta$ | 0.974  | 0.055  | − 0.213 |
| $^{137}\text{Cs}$ activity | 0.530  | − 0.174 | 0.830  |
addition, $^{137}$Cs in STMW and CMW appears to be transported clockwise toward the western boundary of the NPO\cite{36,37,38}. This subsurface or deep-water layer corresponding to $\sigma_\theta=26.7 \text{ kg m}^{-3}$ can further rise westward and reach the shelf break surrounding Taiwan\cite{36,37,38}. East of Taiwan, the Kuroshio is a swift and powerful current that can reach as deep as 400–600 m\cite{39}. While the primary $^{137}$Cs maximum centered around $\sigma_\theta$ of 25.3 to 26.1 kg m$^{-3}$ is suggested to be from lateral transportation from the NPO, there is still a research gap between pelagic studies in the NPO and shelf break data in this study area. For example, impacts of interleaving\cite{40} and meso-scale eddies\cite{41} on the cross-Kuroshio transport mechanism of $^{137}$Cs are still unclear. Direct evidence to constrain the origin and evolution of the $^{137}$Cs maxima along 165°E to this study area is needed in the future. We suggest integrating multiple chemical tracers to study the complex circulation across the pelagic NPO to its western shelf boundary in the future.

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Author contributions
W.-J.H. analyzed the data and wrote the main manuscript and C.T.A.C. edited and supervised this project. M.T.L. coordinates and supervises this project and the sample analysis. K.C.H. prepared all figures and organized data. K.J.K. was involved in the discussion and collected samples. M.A.L. contribute satellite data. M.A.L,
Y.J.Y., and S.J. edited this manuscript and collected samples. All authors designed the experiments and reviewed this manuscript.

**Competing interests**
The authors declare no competing interests.

**Additional information**

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