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**Distribution of plutonium in the Pacific Ocean and implications for
tracing of ocean circulation**

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ABSTRACT

This study examined plutonium (Pu) sources and distribution in the Pacific Ocean based on extensive field datasets over the past 50 years. The basin wide $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in surface and deep seawater ranged from 0.192 to 0.279 averaging 0.235 ± 0.019 , a value consistently higher than that of global fallout at ~ 0.180 . The distribution of $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios exhibited a decreasing trend along the North Equatorial Current-Kuroshio to their extension areas. The activity levels of $^{239+240}\text{Pu}$ in Pacific Ocean surface seawater ranged widely from 0.2 to 43.5 mBq m⁻³, and increased with latitude. We determined the Pu sourced from the Pacific Proving Grounds (PPG) and global nuclear fallout in the Pacific Ocean based on the Pu isotopic composition. Using a mixing model, we found that the PPG made the dominant Pu contribution (average = $69.6 \pm 14.4\%$) to Pacific Ocean surface seawater. The depth range of maximal $^{239+240}\text{Pu}$ activity in the Pacific Ocean was well defined, averaging 608 ± 137 m. The vertical distribution of the $^{239+240}\text{Pu}$ inventory showed most of Pu retained in the upper 3000 m, namely, the contributions of $^{239+240}\text{Pu}$ inventories at 0-1000 m and 0-3000 m depths accounted for $43.5 \pm 9.0\%$ and $75.1 \pm 12.0\%$ of the total, respectively. We identified the transport pathway of Pu-PPG in the Pacific Ocean and demonstrated that Pu isotopes hold great promise as tracers of ocean circulation. Finally, via this extensive compilation of Pu isotopic compositions in the Pacific Ocean water, we established a Pu baseline in the region.

Keywords: Plutonium, Pacific Proving Grounds, Pacific Ocean, Kuroshio, North Equatorial Current

1. Introduction

The main sources of the man-made radionuclide plutonium (Pu) in the Pacific are the deposition of global and close-in fallout due to atmospheric nuclear tests¹⁻². It has been reported that the ²³⁹⁺²⁴⁰Pu inventory is ~8.5 petabecquerels (1 PBq=10¹⁵ Bq) in the Pacific Ocean³. The environmental risk of Pu has become of considerable public concern due to its high radioactivity and danger even at small doses. Pu is also a good tracer to study ocean current trajectories⁴. Additionally, the specific Pu sources can be identified according to the characteristic value of the Pu isotopic ratio⁵.

Accurate Pu measurement is critical for the study of this element in the environment. In general, the conventional analytical method of Pu is α -spectrometry. However, there are clear disadvantages associated with this method, such as the inability to distinguish between ²³⁹Pu and ²⁴⁰Pu, requirement for labor-intensive pretreatment work, need for a large-volume sample (>100 L) and long counting time⁶. In the past 30 years, various Pu analytical methods have been developed for use with environmental samples including the rapid development of mass spectrometry (MS), e.g., multiple collector inductively coupled MS⁷, accelerator MS⁸ and ICP-MS⁹. However, the determination of Pu using the MS is expensive due to the high cost of instrumentation. Therefore, compared to routine monitoring, such as that of temperature-salinity (T-S) and nutrient data, the available field Pu datasets in the ocean are very limited. This makes it difficult to improve our understanding of the chemical behavior and distribution patterns of plutonium in the ocean. Therefore, it is necessary to synthesize a large amount of existing field Pu datasets to ascertain the Pu behavior in the ocean.

Plutonium derived from the Pacific Proving Grounds (PPG) in the equatorial Pacific islands is well known and is characterized by high atomic ratios of ²⁴⁰Pu/²³⁹Pu (0.30–0.36)¹⁰⁻¹¹. Previous studies suggested that the PPG released Pu could be transported to the northwestern Pacific via the North Equatorial Current (NEC) and Kuroshio^{1,10,12}. These studies indicated that Pu from the PPG is a promising tracer of ocean currents in the Pacific Ocean. Additionally, the input of Pu from the PPG resulted in a high Pu inventory in the North Pacific Ocean^{1,10,13-14}. However, the vertical structure of the ²³⁹⁺²⁴⁰Pu inventory remains unclear. Furthermore, it was suggested that the 2011 Fukushima Nuclear Accident (FNA) exerted no immediate Pu impact on the northwestern Pacific Ocean^{12, 15-19}. Overall,

the limited available Pu data usually cover a local area, and therefore cannot yield unbiased interpretation for the Pacific Ocean as a whole. Large Pu dataset in both surface and deep water are rarely available simultaneously for the Pacific Ocean, thus limiting our ability to fully understand the chemical behavior of Pu in the open ocean.

The present study examined the source functions and aimed to gain a better understanding of the fundamental processes that control the transport and fate of Pu by combining large field datasets obtained in the past 50 years. It provides baseline Pu data with sufficient resolution to define a basin-wide distribution. Meanwhile, it is worthwhile noting that the Japanese government plans to discharge the FNA sewage to the Pacific Ocean. Information on Pu isotopes helps in future environmental assessment and understanding of the Pu biogeochemical cycle in the Pacific Ocean, and allows proposing of future Pu studies in the ocean.

2. Materials and methods

2.1. Study area

The Pacific Ocean is the largest ocean worldwide, and is characterized by a unique ocean circulation pattern, which makes the region of particular interest for oceanographic studies. The Pacific Ocean's surface circulation includes the North Pacific surface circulation and South Pacific surface circulation (Figure 1)²⁰⁻²⁴. The North Pacific surface circulation is mainly consisted of the eastward subarctic current and the westward NEC²⁵. The NEC is divided into the Kuroshio (northern branch) and the Mindanao Currents (southern branch)²⁶. The western boundary current - Kuroshio plays an important role in heat and mass transport between the equatorial and northwestern Pacific²⁷. The eastward-flowing Kuroshio extension splits into a southward branch and an eastward flow in the nearby of the Shatsky Rise. The former serves to create Kuroshio Countercurrent and the latter becomes the North Pacific Current²¹. The eastern boundary current in the North Pacific's subtropical gyre is the California Current. In contrast, the South Pacific surface circulation mainly comprises the South Pacific Current (Figure 1)²⁸ and South Equatorial Current²¹. The western and eastern boundary currents in the South Pacific are the East Australian current and the Peru current, respectively²¹.

2.2. Data sources

We reviewed over 400 datasets of $^{239+240}\text{Pu}$ in Pacific Ocean encompassing the past 50 years, and including surface and deep water measurement. These Pu data were mainly extracted from published databases including ScienceDirect, the Web of Science and Scopus, Google Scholar, using the keywords “plutonium and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio” as well as the primary keyword “Pacific Ocean seawater”. Values reported throughout this study represent the mean $\pm$ standard deviation (SD).

3. Results and discussion

3.1. Horizontal distribution of Pu isotopes in the Pacific Ocean

3.1.1. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios

Over 50 datasets of $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in Pacific Ocean surface seawater were summarized from previously published studies^{4,12,29-33} and their horizontal distribution was plotted in Figure 2. The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios over the period 1973-2015 varied from 0.195 to 0.279, averaging 0.242 ± 0.021 (n=37). Spatially, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in the Kuroshio mainstream zone and its extension of the northwestern Pacific Ocean varied from 0.243 to 0.263 (average= 0.253 ± 0.007 , n=7) and from 0.228 to 0.232 (average= 0.230 ± 0.003 , n=2), respectively¹². The distribution of the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio exhibited a gradual decrease in the direction of the subarctic gyre, declining along the Kuroshio from the eastern Philippines to southern Japan (Figure 2a). In contrast, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios off the Kuroshio zone varied from 0.199 to 0.244, averaging 0.222 ± 0.019 (n=8), values which were lower than those in the mainstream zone of Kuroshio¹². It is noteworthy that higher $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios were observed in the converging zone between the Kuroshio and the Oyashio, ranging from 0.233 to 0.279 (average= 0.257 ± 0.015 , n=11)^{29,32}. In the subarctic zone, $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios were in the range of 0.237-0.254, averaging 0.247 ± 0.007 (n=6)²⁹, i.e., were lower than those in the mainstream zone of Kuroshio. In the South Pacific Ocean, $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios varied from 0.195 to 0.215 (average= 0.205 ± 0.014 , n=2)³¹, and were thus slightly lower than those in the North Pacific.

Overall, the $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in Pacific Ocean surface seawater are

substantially higher (by 34.4%) than the values from global fallout, indicating that this area has received non-global Pu fallout. Potential non-global fallout sources of Pu include low-latitude above-ground nuclear weapons test sites located within the basin (i.e., the PPG), Fukushima derived from nuclear contamination, the Chernobyl nuclear accident and Asian dust derived from aboveground nuclear tests conducted by the People's Republic of China (PRC) at the Lop Nor and/or the Semipalatinsk sites. During the period 1952-1958, extensive US nuclear bomb tests were carried on the Pacific Marshall Islands, which yielded fallout accounting for > 50% of total fallout at the time¹⁰. It is well known that these events resulted in higher $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios (0.30–0.36)¹¹. The contribution of Chernobyl-derived Pu was confirmed to be negligible in the North Pacific Ocean³⁴. The atomic ratios of $^{241}\text{Pu}/^{239}\text{Pu}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ in seawater and marine sediments suggested that the Pu signal from the immediate impact and atmospheric deposition from the FNA was minor in the Pacific Ocean^{12,16,18,35-37}. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratio of ~0.180 in Chinese soil profiles is similar to the global fallout, suggesting that the Lop Nor and Semipalatinsk nuclear tests did not appear to contribute Pu to Chinese soil and the China Seas^{4,12,38-40}. Considering the large distance of the Pacific Ocean from the Lop Nor and Semipalatinsk nuclear test sites relative to the China Seas, the Pu contribution from these sites appears to be negligible. Plutonium from the PPG was confirmed to be the only non-global source in the Pacific Ocean. Therefore, we constrained the Pu sources in the Pacific Ocean to include the PPG and global fallout, the former being continuously supplied to the Pacific Ocean via ocean circulation after the banning of nuclear tests (e.g., NEC and Kuroshio, North Pacific Current) and resulting in the differential distribution of Pu activity between North Pacific and South Pacific¹.

3.1.2 $^{239+240}\text{Pu}$ activity

The $^{239+240}\text{Pu}$ activities in Pacific Ocean surface seawater over the period 1973–2015 derived from previous studies are summarized in Figure 2. They show a wide range from 0.2 to 43.5 mBq m⁻³, averaging 5.4 ± 7.5 mBq m⁻³ (n=58)^{4,12-14, 29-33, 41-42}. In the North Pacific Ocean, the $^{239+240}\text{Pu}$ activity of surface seawater varied from 0.6 to 43.5 mBq m⁻³, averaging 5.9 ± 7.7 mBq m⁻³ (n=50). In the South Pacific Ocean, they varied from 0.2 to 17.0 mBq m⁻³, with a mean value of 2.7 ± 5.8 mBq m⁻³ (n=8). In the present study, we highlight the analysis and discussion of $^{239+240}\text{Pu}$ activity in the North Pacific in light of the additional

field data included.

In the northwest Pacific, $^{239+240}\text{Pu}$ activities in Kuroshio water were in the range of 1.15-1.93 mBq m^{-3} , averaging $1.60 \pm 0.25 \text{ mBq m}^{-3}$ ¹². Outside the Kuroshio zone, they ranged from 1.18 to 2.16 (average = $1.62 \pm 0.30 \text{ mBq m}^{-3}$), and were comparable to those of the Kuroshio mainstream¹². In the Oyashio and the Kuroshio converging zone, $^{239+240}\text{Pu}$ activities ranged from 1.46 to 43.50 mBq m^{-3} (average = $8.52 \pm 12.11 \text{ mBq m}^{-3}$, $n=11$)^{29,32}, and were thus higher than those of the Kuroshio mainstream. In turn, in the subarctic zone, $^{239+240}\text{Pu}$ activities varied from 2.30 to 20.20 mBq m^{-3} , averaging $11.97 \pm 7.40 \text{ mBq m}^{-3}$ ($n=6$)²⁹. To determine whether $^{239+240}\text{Pu}$ activities differed within and outside the Kuroshio, an f-test and t-test were conducted using these combined data⁴³. Results showed that $^{239+240}\text{Pu}$ activity in the Kuroshio water was significantly lower than those outside the Kuroshio zone (Student t-test, $p < 0.05$). This suggests that the former is readily scavenged leading to a decline in the supply of Pu.

The bulk of Pu in Kuroshio water should originate mainly from the PPG. The physicochemical characteristics of particles that packaged Pu are closely related to those occurring under nuclear testing conditions¹⁰. In general, close-in fallout from the PPG was enclosed by calcium compounds such as calcium oxides and calcium hydroxides⁴⁴. The size of PPG particles that package Pu is typically over 1 micron⁴⁵. It is well known that Pu from the PPG adhering to relatively larger particles is more efficiently scavenged during downward transport and deposition. In contrast, the size of global fallout particles that package Pu is typically 0.1–1 micron⁴⁴. Plutonium from global fallout adhering to relatively smaller particles is less efficiently scavenged in the open ocean. The retention time of Pu from the PPG in water is thus shorter than that of global fallout^{4,12}. Additionally, it is reported that elevated diazotrophic concentration in Kuroshio water usually stimulates the growth of diatoms and increases the export flux of particles⁴⁶⁻⁴⁷. Actually, the *Trichodesmium* (nitrogen fixer) is abundant in Kuroshio water, and is positively correlated with the concentration of large diatom cells ($>10 \mu\text{m}$) ($p < 0.05$)⁴⁸. The vertical distribution of diatom abundance in Kuroshio water is consistent with that of *Trichodesmium*, in that the maximum abundance of both appeared at the surface⁴⁸⁻⁴⁹. In contrast, *Trichodesmium* in the zone outside the Kuroshio exhibited a low concentration and its vertical distribution showed no surface maxima⁵⁰. This indicated that the zone outside the Kuroshio potentially

has a lower scavenging efficiency than Kuroshio seawater, although particle abundances does not differ significantly in these two areas⁵¹. The lower $^{239+240}\text{Pu}$ activities in Kuroshio seawater should thus be attributable to its higher scavenging efficiency due to the abundance of *Trichodesmium*.

At present, the bulk of $^{239+240}\text{Pu}$ in Kuroshio water should originate mainly from the seawater and sediment discharge from PPG contaminated lagoons. $^{239+240}\text{Pu}$ activities in seawater and sediments of PPG lagoons were determined to be $\sim 455 \text{ mBq m}^{-3}$ and $\sim 99 \text{ Bq kg}^{-1}$, respectively⁵². They are 2-3 orders of magnitude higher than the background value in the earth's environment. It is estimated that the export flux of $^{239+240}\text{Pu}$ from the PPG is ca. 0.5 Terabecquerel/year in the North Pacific ($1 \text{ TBq} = 10^{12} \text{ Bq}$)^{4,12}. Nevertheless, this annual export flux is several orders of magnitude smaller than the delivery rate via the deposition of PPG close-in fallout over the period 1952-1958^{12,52}. Thus, the lower $^{239+240}\text{Pu}$ activities in Kuroshio water should be closely related to the decline in the PPG supply with time.

Overall, the lower $^{239+240}\text{Pu}$ activity in Kuroshio water is linked primarily to the higher scavenging efficiency of Kuroshio water, coinciding with a decline in the PPG supply over time. The fact that the radioactivity levels of $^{239+240}\text{Pu}$ in Kuroshio seawater were substantially lower than those outside the Kuroshio zone argues for high Pu accumulation outside the Kuroshio zone and is consistent with the rapid scavenging of Pu-PPG in Kuroshio water.

The horizontal distribution of $^{239+240}\text{Pu}$ activities shown in Figure 2b is characterized by three key features: 1) the $^{239+240}\text{Pu}$ activity in the North Pacific Ocean (average $\sim 5.9 \text{ mBq m}^{-3}$) was higher than in the South Pacific Ocean (average $\sim 2.7 \text{ mBq m}^{-3}$); 2) the $^{239+240}\text{Pu}$ activity in the North Pacific Ocean at first increased with latitude and subsequently decreased with latitude (i.e., the maximum $^{239+240}\text{Pu}$ activity appeared at mid-latitudes); 3) the $^{239+240}\text{Pu}$ activity in the South Pacific Ocean gradually decreased with increasing the latitude. This distribution pattern of $^{239+240}\text{Pu}$ activity is influenced by the source, transport and scavenging of Pu. Large-scale atmospheric testing of nuclear weapons was carried out at mid-high latitudes of the northern hemisphere between the 1950s to early 1960s⁵³⁻⁵⁴, such that fallout debris was largely injected into the stratosphere where it remained for about a year and was then deposited globally (so called global fallout)⁵⁴. Therefore, Pu in the Pacific Ocean mainly originates from the deposition of global

fallout^{1,10,53} and is estimated to result in a delivery of 2.59 PBq to the North Pacific Ocean¹. The latitudinal distribution of Pu is consistent with the deposition pattern of global fallout¹. In the 1950s, close-in fallout from the PPG was deposited in the Pacific Ocean, and Bowen et al. (1980)¹ estimated that ca. 6.44 PBq of ²³⁹⁺²⁴⁰Pu-PPG was delivered to the North Pacific Ocean. Following the moratorium of weapons tests, the NEC could bring PPG contaminants westward, entering the Kuroshio and southward, entering the Equatorial Countercurrent (see later discussion).

3.2. Vertical distribution of Pu isotopes in the Pacific Ocean

The vertical profile of Pu isotopes in the water column at 25 sites in the Pacific Ocean was summarized^{1,12,14,29,31,33,41,55} and plotted in Figure 3.

3.2.1. ²⁴⁰Pu/²³⁹Pu atom ratios

Vertical depth profile of ²⁴⁰Pu/²³⁹Pu atom ratios in the Pacific Ocean are shown in Figure 3a. Those in the water column varied from 0.192 to 0.255, averaging 0.229 ± 0.015 (n=46). Spatially, the ²⁴⁰Pu/²³⁹Pu atomic ratios in the North Pacific Ocean varied from 0.215 to 0.255, averaging 0.231 ± 0.011 (n=37), and the corresponding values in the South Pacific Ocean ranged from 0.192 to 0.251, averaging 0.220 ± 0.024 (n=9). They were consistently higher than those of global fallout, suggesting that the Pu signature from non-global fallout occurs throughout the water column (0-6000 m). High ²⁴⁰Pu/²³⁹Pu atom ratios are potentially attributed to the PPG-Pu signature, via deposition in the 1950s and subsequent transport in ocean circulation. The vertical patterns of ²⁴⁰Pu/²³⁹Pu atom ratios showed a slight increase with depth in the Pacific Ocean (Figure 3a). For example, the atom ratio at the AQ7 station of the North Pacific increased from ~0.215 at the surface to ~0.230 in the mid-water column. Similarly, the atom ratio at the AQ13 station of the South Pacific increased from ~0.195 at the surface to ~0.215 in the intermediate water. Large particles packaged Pu-PPG was preferentially scavenged during downward transport and deposition¹⁰. At mid-depth, an increase in the ²⁴⁰Pu/²³⁹Pu atom ratio was caused by Pu recycling during downward transport, where PPG-Pu goes back to the water column because of particle remineralization^{6,12}.

3.2.2. ²³⁹⁺²⁴⁰Pu activity

Vertical patterns of ²³⁹⁺²⁴⁰Pu activity in the Pacific Ocean are shown in Figure 3b-3c. Activities in the water column of the Pacific Ocean varied from 0.2 to 76.7 mBq m⁻³,

averaging 13.4 ± 14.4 mBq m⁻³ (n=315). Those in the North Pacific Ocean varied from 0.6 to 76.7 mBq m⁻³, averaging 16.5 ± 15.7 mBq m⁻³ (n=212), while they ranged from 0.2 to 50.0 mBq m⁻³ in the South Pacific Ocean, with a mean value of 7.0 ± 7.9 mBq m⁻³ (n=103). Thus, overall, ²³⁹⁺²⁴⁰Pu activities in the North Pacific are higher than those in the South Pacific. The vertical ²³⁹⁺²⁴⁰Pu activity profile showed an initial tendency to increase with increasing water depth, featuring a sub-surface maximum, followed by a slower decrease with depth. This distribution agrees with the typical Pu pattern found in oceanic regimes¹.

The maximal ²³⁹⁺²⁴⁰Pu activity in the water column of the Pacific Ocean displayed a wide range of 300-900 m, averaging 608 ± 137 m (n=25) (Figure 4a). In the North Pacific Ocean, the maximum ²³⁹⁺²⁴⁰Pu activity ranged from 405 to 900 m, averaging 625 ± 117 m (n=17). In contrast, it ranged from 300 to 800 m in the South Pacific Ocean, averaging 572 ± 174 m (n=8). The depth of maximum ²³⁹⁺²⁴⁰Pu activity in the North Pacific is thus greater than that in the South Pacific. The depth of maximum Pu activity is also greater than the deep chlorophyll maximum (DCM < 100 m) in the Pacific⁵⁶⁻⁵⁷. As shown in Figure 4b, the depth of maximum ²³⁹⁺²⁴⁰Pu activity in the Pacific Ocean increases with increasing latitude, i.e., the depth in the high latitude zone is greater than that at low latitudes. The relationship between the depth of maximal Pu activity and longitude is also plotted in Figure 4c, showing that this depth in the eastern hemisphere is more variable than in the western hemisphere. Indeed, it was determined that particle concentration was higher in the upper 100 m (DCM layer), which would enhance Pu scavenging in this layer. Beyond 100 m, particle concentrations remained low, and regeneration due to decomposition of particles by microorganisms during downward transport is expected to be dominant^{4,58-59}.

3.3. ²³⁹⁺²⁴⁰Pu inventory in the Pacific Ocean

The ²³⁹⁺²⁴⁰Pu inventory provides a useful indicator to assess its accumulation in the ocean. The Pacific Ocean water column ²³⁹⁺²⁴⁰Pu inventories are summarized and plotted in Figure 5 to determine their latitudinal spatial distribution. The inventory in the Pacific Ocean showed a wide range of 7.3-148.0 Bq m⁻², with an average of 67.7 ± 40.7 Bq m⁻² (n=49). In the North Pacific it varied from 28.2 to 148.0 Bq m⁻², averaging 81.2 ± 35.8 Bq m⁻² (n=38), whereas it ranged from 7.3 to 39.7 Bq m⁻² in the South Pacific, with a mean value of 20.8 ± 9.8 Bq m⁻² (n=11). The ²³⁹⁺²⁴⁰Pu inventory in the North Pacific is markedly

higher than that in the South Pacific (Figure 5a), which is potentially related to the large-scale nuclear bombing conducted in the northern hemisphere⁶⁰. Notably, a high $^{239+240}\text{Pu}$ inventory was observed in the northwest Pacific (Figure 5a), presumably due to the high deposition flux of global fallout and the Pu input of PPG via the NEC and Kuroshio current. The latitudinal distribution of $^{239+240}\text{Pu}$ inventory is plotted in Figure 5b, showing that values were higher than the deposition flux of global fallout in the corresponding latitudinal zone. This further suggests that the Pacific Ocean has been receiving Pu from the PPG. The water column $^{239+240}\text{Pu}$ inventories increase with latitude in the North Pacific, whereas the inverse occurs in the South Pacific, i.e., they decrease with increasing latitude.

The $^{239+240}\text{Pu}$ inventory at depth > 3000 m (except for HY15) in the Pacific Ocean and percent of the total at three different depth ranges (0-1000 m, 0-2000 m and 0-3000 m) are listed in Table 1. The vertical distribution of the $^{239+240}\text{Pu}$ inventory in the Pacific Ocean showed considerable heterogeneity with water depth (Figure 6a), such that most of $^{239+240}\text{Pu}$ inventory occurs in the upper 1000 m. In the South Pacific, the $^{239+240}\text{Pu}$ inventories at depths of 0-1000 m, 1000-2000 m and 2000-3000 m at the stations sampled ranged from 4.1-21.1 Bq m⁻² (average=9.0±5.7 Bq m⁻²), 1.6-10.4 Bq m⁻² (average=4.4±3.2 Bq m⁻²), 1.0-11.5 Bq m⁻² (average=4.5±4.1 Bq m⁻²), respectively. Therefore, the $^{239+240}\text{Pu}$ inventories at 0-1000 m are ca. twice as high as those at 1000-2000 m and 2000-3000 m. Correspondingly, the $^{239+240}\text{Pu}$ inventory contribution at 0-1000 m, 1000-2000 m and 2000-3000 m to the total Pu inventory varied from 32.7-56.0% (average=44.0±8.3%), 12.9-28.7% (average=20.0±4.6%), 7.7-33.8% (average=18.0±8.3%), respectively (Figure 6b). The percent $^{239+240}\text{Pu}$ out of the total at 0-3000 m depth varied from 67.2-100.0% (average=82.0±11.5%). The $^{239+240}\text{Pu}$ inventories in the North Pacific Ocean at 0-1000 m, 1000-2000 m and 2000-3000 m ranged from 9.6-45.2 Bq m⁻² (average=25.7±10.6 Bq m⁻²), 3.3-20.5 Bq m⁻² (average=9.6±5.9 Bq m⁻²), 1.7-16.3 Bq m⁻² (average=6.8±4.5 Bq m⁻²), respectively. The $^{239+240}\text{Pu}$ inventories at 0-1000 m are higher than those at 1000-3000 m. In turn, the percent contribution of $^{239+240}\text{Pu}$ inventories to the total in the North Pacific Ocean at 0-1000 m, 1000-2000 m and 2000-3000 m varied from 29.7-65.5% (average=43.3±9.5%), 6.7-26.9% (average=15.7±5.6%), 2.4-33.3% (average=12.1±8.0%), respectively. This contribution in the North Pacific Ocean at 0-3000 m ranged from 53.9 to 85.0% (average=71.1±10.7%). Overall, the contribution of $^{239+240}\text{Pu}$ inventories in the

Pacific Ocean at 0-1000m and 0-3000 m accounted on average for $43.5 \pm 9.0\%$ and $75.1 \pm 12.0\%$ of the total, respectively. This vertical structure of the $^{239+240}\text{Pu}$ inventory is helpful to estimate the Pu inventory in the ocean and better characterize Pu accumulation in general.

The $^{239+240}\text{Pu}$ inventory in the water column is calculated by integrating the activity measured at each depth⁶¹. Here, we made an attempt to quantify the stoichiometry between the $^{239+240}\text{Pu}$ inventory and depth via analysis of a large number of field data in the Pacific Ocean. The plot of $^{239+240}\text{Pu}$ inventory (y) vs. depth (x) using this extensive dataset is shown in Figure 7 and shows that they are positively correlated. Linear equations describing these data are also shown in Figure 7 for the South Pacific Ocean ($y=0.0049x$, $R^2=0.6675$) and the North Pacific Ocean ($y=0.0149x$, $R^2=0.8548$). Thus, overall, the slopes of the linear equations indicate that the modeled $^{239+240}\text{Pu}$ inventory in the North Pacific Ocean is ca. three times that in the South Pacific Ocean at the same depth. Additionally, we inferred the boundary equations of $y=0.0027x$ ($R^2=0.9919$) in the South Pacific Ocean and $y=0.0227x$ ($R^2=0.9960$) in the North Pacific Ocean, respectively. We can therefore roughly calculate the $^{239+240}\text{Pu}$ inventory at a given depth from these relationships. Using a simple first order calculation, the total amount of $^{239+240}\text{Pu}$ is estimated to be ~ 9.8 PBq ($3.5\text{-}10.7$ PBq) in terms of the average depth ($x=3957$ m) and area ($S=1.81 \times 10^{14}$ m²) of the Pacific Ocean, a value comparable to previous estimates ($8.5\text{-}9.06$ PBq)^{1,3,12}. The above well defined relationship between the Pu inventory and depth allows quickly calculation of the Pu inventory in the ocean.

3.4. Quantitative estimate of the Pu-PPG contribution to the Pacific Ocean

This study confirms that the PPG continuously provides Pu source to the Pacific Ocean, and excludes other possible sources, such as the Chernobyl and FNA^{4,12,34}. The combined field atom ratios of Pu in the Pacific Ocean were also found to lie between the global fallout and the PPG (Figure 8), suggesting that Pu in the Pacific Ocean originates from both global fallout and the PPG. The Pu-PPG contribution in the Pacific Ocean can be calculated using the following equation⁶²:

$$(Pu)_K = (Pu)_P + (Pu)_G \quad (1)$$

where (Pu) represents the $^{239+240}\text{Pu}$ activity; the subscripts P , G and K indicate the

361 PPG, global fallout and the Pacific sample, respectively. The partitioning between $(Pu)_P$
 362 and $(Pu)_G$ can be expressed as:

$$363 \quad Y = \frac{(Pu)_P}{(Pu)_G} \quad (2)$$

364 Based on mass balance considerations, equation (3) can be deduced from equations (1) and
 365 (2) as:

$$366 \quad Y = \frac{(Pu)_P}{(Pu)_G} = \frac{(R_G - R_K)(1 + 3.674R_P)}{(R_K - R_P)(1 + 3.674R_G)} \quad (3)$$

367 where R represents the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio, and the coefficient 3.674 is a conversion
 368 factor between the activity ratio and atom ratio of $^{240}\text{Pu}/^{239}\text{Pu}$.

369 while the Pu-PPG contribution to the Pacific sample can be expressed as:

$$370 \quad \frac{(Pu)_P}{(Pu)_K} = \frac{Y}{1 + Y} \quad (4)$$

371 In the calculation, R_K is the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio in the Pacific sample, R_G
 372 averages 0.180 ± 0.014 ⁶³ and R_P averages 0.330 ± 0.030 ¹¹. Plutonium from the PPG was
 373 estimated to average $70.7 \pm 12.9\%$ and $29.0 \pm 4.2\%$ of surface seawater in the North Pacific
 374 and South Pacific, respectively. Overall, the Pu-PPG contributed $69.6 \pm 14.4\%$ to the
 375 Pacific Ocean surface seawater, values is slightly higher than the results in the Pacific's
 376 marginal seas, e.g., the East China Sea ($\sim 36\%$)⁶⁴, South China Sea ($\sim 42\%$)^{4,30} and Japan
 377 Sea ($\sim 33\%$)⁶⁵. The spatial distribution of the PPG contribution to surface seawater in the
 378 Pacific Ocean is plotted in Figure 9, showing a high PPG contribution in the transport
 379 pathway, e.g., the Kuroshio. The Pu-PPG contribution to the North Pacific was significantly
 380 higher (by a factor of 3) than that to the South Pacific. A higher Pu-PPG fraction was
 381 determined in the intersection zone between the Kuroshio and the Oyashio⁶⁶, a finding
 382 potentially related to the continuous PPG input via the NEC and Kuroshio^{4,12}.

383 **3.5. Pu transport pathway in the Pacific Ocean and implication for ocean circulation**

384 The $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios (0.192–0.279) of surface and deep water in the Pacific
 385 Ocean were higher than those of global fallout (average= 0.180 ± 0.014)⁶³, suggesting that
 386 this ocean region must have received Pu from the Marshall Islands PPG characterized by a

high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio (range=0.30-0.36)¹¹. The Pu input from the PPG to the Pacific Ocean can be divided into two periods. During the period 1952-1958, large-scale US nuclear tests were conducted at the Marshall Islands PPG including the Enewetak and Bikini Atolls with high-energy thermonuclear detonations¹⁰. The Mike test at the Enewetak Atoll in 1952 and the Bravo test at the Bikini Atoll in 1954 yielded a fission of 15 Mt and 15 Mt, respectively⁶⁰. They were also the only ones characterized significantly high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios⁶⁰. Depending upon local wind conditions and atmospheric deposition patterns⁶⁷, 50% of two high-energy thermonuclear detonations was deposited preferentially within the vicinity of the test sites (close-in tropospheric fallout)^{7,33,60}. It is estimated that ca. 60% of the $^{239+240}\text{Pu}$ inventory in the North Pacific originated from the PPG¹. Even in the subarctic zone far away from the PPG, high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios (0.247 ± 0.007 , $n=7$) were observed in 1988^{29,33}.

Plutonium deposited in the Pacific Ocean is further transported via oceanic current circulation. For example, Pu from the PPG circulates in the subarctic zone via the Alaska stream, and Oyashio and Alaska currents⁶⁸. Thus, after banning of US nuclear weapons testing in the Marshall Islands, Pu from the PPG was mainly dispersed throughout the Pacific Ocean by circulation currents (e.g., NEC and Kuroshio), resulting in the ubiquitous Pu-PPG signature in this region. The NEC, originating approximately from the PPG, transported the high activity of $^{239+240}\text{Pu}$ westward and subsequently fed into its northward and southward bifurcations off the Philippines, namely, the Kuroshio Current and North Equatorial Countercurrent (NECC). In the North Pacific, Pu from the PPG is transported northwestwardly by the Kuroshio. This high $^{240}\text{Pu}/^{239}\text{Pu}$ “stream” along the NEC-Kuroshio was evident when compared with $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios outside this pathway in the western Pacific Ocean^{12,30} and its adjacent marginal seas^{4,30,65,69-72}. Persistently high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios ($0.224\text{--}0.279$, average= 0.251 ± 0.018 , $n=14$) were observed in the inter-frontal zone ($30^{\circ}\text{--}40^{\circ}$ N, $140^{\circ}\text{--}170^{\circ}$ E) between the Kuroshio and the Oyashio during the period 1988-2014^{4,12,29,32-33,66}; this is potentially related to the continuous Pu-PPG supply via the NEC-Kuroshio. Correspondingly, high $^{239+240}\text{Pu}$ activities and inventories were also observed in this region. Indeed, this region is also the formation zone of Subtropical Mode Water (STMW)^{20-21,73}, which penetrates at ca. 300–500 m depth²². The formation and circulation of the STMW and Central Mode Water (CMW) has been in detail

described in elsewhere²². STMW mainly comprises Oyashio and Kuroshio water, accounting for ~45% and ~55%, respectively^{66,74}. Thus, high $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios of surface seawater could be transported to outside of the Kuroshio zone by spreading and subduction of STMW. And then, the STMW spreads near the subtropical front via advection over the Kuroshio recirculation region⁷⁵⁻⁷⁷. Then, the Pu signature from the PPG is continuously transported into the CMW formation area by the Kuroshio extension and North Pacific Current⁷⁸. In general, the CMW firstly spreads eastward along the North Pacific Current, turns southward and then westward²². Plutonium from the PPG is further transported to the tropical zone by CMW spreading and subduction. In the South Pacific Ocean, Pu from the PPG is transported by the NECC and South Equatorial Current. Therefore, Pu from the PPG is also a useful potential tracer for oceanic circulation in the Pacific Ocean.

4. Conclusions

$^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in both deep and surface water were determined to be 0.192–0.279 during the period 1973–2015, indicating the input of non-global Pu source to the Pacific Ocean. The horizontal distribution of $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios showed a gradual decrease in the direction of the subarctic gyre, declining along the Kuroshio from the eastern Philippines to southern Japan. The $^{239+240}\text{Pu}$ activities of surface seawater in the Pacific Ocean were determined to be 0.2–43.5 mBq m⁻³, showing an increase with increasing latitude. The present study confirms the non-global Pu from the PPG by comparing Pu isotopic compositions between the zones within and outside the circulation current region. Plutonium sources are constrained by the PPG and global fallout in the Pacific Ocean. The present study further revealed that the PPG contributed on average $70.7 \pm 12.9\%$ and $29.0 \pm 4.2\%$ of Pu to the North Pacific and the South Pacific, respectively.

The depth range of maximal $^{239+240}\text{Pu}$ activity in the Pacific Ocean was well defined at 300–900 m, averaging 608 ± 137 m. The $^{239+240}\text{Pu}$ inventory in the Pacific Ocean has a varied vertical structure, namely, the percent $^{239+240}\text{Pu}$ inventory at 0–1000m and 0–3000 m accounted for $43.5 \pm 9.0\%$ and $75.1 \pm 12.0\%$ of the total, respectively. Additionally, the Pacific Ocean is mainly impacted by PPG Pu through early atmospheric deposition and subsequent ocean circulation currents. We established the transport pathway of Pu from the

PPG in the Pacific Ocean and demonstrated that Pu was an excellent tracer of oceanic circulation. Finally, via this new compilation of Pu isotopic compositions in the Pacific Ocean, we have established a background for the coming Pu studies associated with the nuclear sewage from the FNA discharged into the Pacific Ocean.

Additional work includes the understanding Pu biogeochemistry and the fate of this element in the Pacific Ocean. We conclude that Pu from the PPG serves as a promising tracer that could be widely used to study ocean circulation currents in the Pacific Ocean. Certainly, Pu isotopic composition in the circulation currents, e.g., the NEC and CMW, needs to be further investigated. Plutonium isotopes need to be continuously monitored in lagoonal seawater discharges, sediment and marine organisms in the PPG to accurately evaluate Pu's ecological impact. Finally, future developments aimed at increasing the sensitivity of different synchrotron radiation (SR)-X-ray microbeams and mass spectrometric techniques will allow improved characterization of the distribution and speciation of Pu in the Pacific Ocean.

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Author Contributions

Junwen Wu: Conceptualization, Funding acquisition, Writing-original draft & editing. **Jisheng Chen:** Methodology-collecting the data. **Cui Wang:** Writing-review &

editing. **Zhaoyong Zheng**: Review & editing. **Liang Wang**: Review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Figure 1. Schematic view of current circulation in the Pacific Ocean. Green solid and dashed ellipses represent the formation and spreading areas, respectively, of Subtropical Mode Water (STMW), respectively. Purple solid and dotted ellipses represent the formation and spreading areas, respectively, of Central Mode Water (CMW). Red and blue dotted lines indicate surface circulation currents of the subtropical and subarctic gyres, respectively. Black dotted lines indicate surface circulation in the South Pacific Ocean. Red triangle marks the Pacific Proving Grounds (PPG). This figure is based on previous publications^{20,22-24}. This figure was prepared with the free software Ocean Data View (ODV 5.1.2) (Schlitzer, R., Ocean Data View, <https://odv.awi.de>, 2018)

Figure 2. Lateral distribution of the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio (a) and $^{239+240}\text{Pu}$ activity (mBq m^{-3}) (b) in the Pacific Ocean. The Pu atom ratios and activities are derived from previous publications^{4,12,14,29-33,41}. This figure was prepared with the free software Ocean Data View (ODV 5.1.2) (Schlitzer, R., Ocean Data View, <https://odv.awi.de>, 2018)

Figure 3. Sampling sites for analyzing Pu in water column from the Pacific Ocean (a) and vertical depth profile of the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio (b) and $^{239+240}\text{Pu}$ activity (mBq m^{-3}) at various sampling stations in the North Pacific Ocean (c) and South Pacific Ocean (d). PPG= Pacific Proving Grounds. The Pu data in the water column are cited from previous publications^{1,12,14,29,31,33,41,55}. This figure was prepared with the free software Ocean Data View (ODV 5.1.2) (Schlitzer, R., Ocean Data View, <https://odv.awi.de>, 2018) (a) and Sigma-Plot professional 10.0 software (b–d).

Figure 4. Depth variation of maximum $^{239+240}\text{Pu}$ activity at various stations in the South and North Pacific Ocean (a) and distribution pattern of maxima with latitude (b) and longitude (c). These figures were prepared with Sigma-Plot professional 10.0 software.

Figure 5. Spatial (a) and latitudinal (b) distribution of the $^{239+240}\text{Pu}$ inventory (Bq m⁻²) in Pacific Ocean seawater. Blue and red bars in (b) represent the $^{239+240}\text{Pu}$ inventory observed in the Pacific Ocean and deposition flux of global fallout in a different latitudinal zone, respectively. $^{239+240}\text{Pu}$ inventories originate from previous publications^{1,14,29,31,33,41,55,79}. This figure was prepared with the free software Ocean Data View (ODV 5.1.2) (Schlitzer, R., Ocean Data View, <https://odv.awi.de>, 2018) (a) and Sigma-Plot professional 10.0 software (b).

Figure 6. Vertical distribution of the $^{239+240}\text{Pu}$ inventory (a) and percent of the total (b) at three different depth ranges and sampling stations. These figures were prepared with Sigma-Plot professional 10.0 software.

Figure 7. Linear fitted relationship between the $^{239+240}\text{Pu}$ inventory (y) and water depth (x) in the Pacific Ocean. Values indicate the mean $\pm$ standard deviation (SD); R^2 =coefficient of determination. This figure was prepared with Sigma-Plot professional 10.0 software.

Figure 8. Relationship between the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio and the reciprocal of $^{239+240}\text{Pu}$ activity for surface seawater and profile seawater in the Pacific Ocean. The blue circles represent the surface seawater data and the red squares indicate the profile seawater data. Horizontal black dashed lines represent the average $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio (0.180 ± 0.014) of global fallout⁶³ and cyan dotted lines represent the range of the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio (0.30–0.36) characteristic of the Pacific Proving Grounds (PPG)¹⁰⁻¹¹. This figure was prepared with Sigma-Plot professional 10.0 software.

Figure 9. Spatial distribution of the contribution of Pu from the PPG to surface seawater in the Pacific Ocean. This map was drawn using the free software Ocean Data View (ODV 5.1.2) (Schlitzer, R., Ocean Data View, <https://odv.awi.de>, 2018).

Table 1. $^{239+240}\text{Pu}$ inventory in the Pacific Ocean and percent of the total at three different depth ranges

station	water depth (m)	$^{239+240}\text{Pu}$ inventory (Bq m^{-2})				Pu inventory percent		
		0-1000m	0-2000m	0-3000m	0-bottom	0- 1000m	0- 2000m	0- 3000m
HY17	4037	5.13±0.49	7.24±0.70	8.48±0.84	9.94±1.02	51.6%	72.8%	85.3%
HY18	4411	5.54±0.38	7.49±0.59	8.88±0.74	11.65±1.03	47.6%	64.3%	76.2%
HY15	2867	4.08±0.38	6.17±0.83	7.28±1.09	7.28±1.09	56.0%	84.8%	100.0%
HY12	4114	5.79±0.46	7.39±0.71	8.34±0.86	12.42±0.99	46.6%	59.5%	67.2%
HY11	4680	8.38±0.59	13.83±0.98	18.14±1.38	25.66±1.97	32.7%	53.9%	70.7%
HY9	3882	13.46±1.26	21.18±1.96	29.65±2.46	39.74±2.95	33.9%	53.3%	74.6%
AQ13	3200	8.17±0.25	11.87±0.90	21.17±1.80	21.10±0.60	38.7%	56.3%	90.1%
HY6	3219	21.13±1.47	31.52±2.46	43.06±3.11	46.87±3.39	45.1%	67.3%	91.9%
AQ7	5500	9.60±0.41	14.58±1.67	19.29±2.12	28.20±1.20	34.0%	51.7%	68.4%
HY3	3635	22.30±1.68	31.09±2.47	41.35±3.32	50.03±3.92	44.6%	62.1%	82.7%
NP4	5933	38.95±2.71	59.46±4.40	74.34±6.80	131.0±4.0	29.7%	45.4%	56.8%
NP3	5728	45.21±3.59	52.51±4.77	54.16±5.65	69.0±4.0	65.5%	76.1%	78.5%
HY2	4208	26.40±2.07	33.80±3.62	39.72±4.22	48.77±5.08	54.1%	69.3%	81.4%
HY1	5309	17.37±1.53	21.37±2.08	23.72±2.38	36.13±3.07	48.1%	59.2%	65.7%
NP6	9754	29.02±2.64	34.41±3.28	36.80±3.73	66.0±3.0	44.0%	52.1%	55.8%
NP2	5843	41.47±2.56	61.22±4.17	66.35±4.86	107.0±3.0	38.8%	57.2%	62.0%
NP1	4085	22.04±2.29	25.32±4.40	41.65±6.25	49.0±4.0	45.0%	51.7%	85.0%
NP5	4004	30.73±3.55	40.13±4.69	47.08±5.77	60.0±3.0	51.2%	66.9%	78.5%
DR10	5600	15.57±2.69	21.60±3.43	25.65±4.15	47.56±6.73	32.7%	45.4%	53.9%
DR13	3930	13.42±1.21	21.82±2.20	26.32±2.86	36.58±3.97	36.7%	59.7%	72.0%
NP8	4090	25.74±2.92	45.14±5.67	54.24±6.77	72.0±4.0	35.8%	62.7%	75.3%
NP7	3920	21.89±3.66	31.64±4.71	38.07±5.61	48.0±3.0	45.6%	65.9%	79.3%