



## Plutonium in coral archives: A good primary marker for an Anthropocene type section



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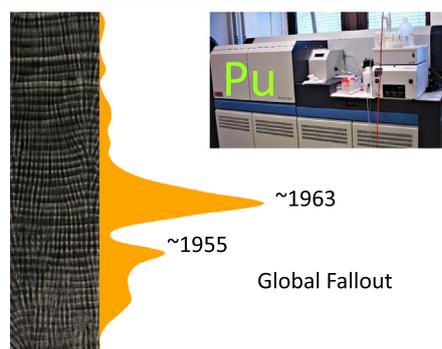
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### HIGHLIGHTS

- A new record of plutonium in a Caribbean coral is compared to seven records worldwide.
- The plutonium maxima in banded corals are asynchronous.
- The plutonium onsets in corals distant to nuclear detonation grounds are synchronous.
- Massive corals from Caribbean reefs are suitable to host an Anthropocene golden spike.

### GRAPHICAL ABSTRACT



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### ABSTRACT

While we officially live in the Holocene epoch, global warming and many other impacts of global change have led to the proposal and wide adoption of the Anthropocene to define the present geological epoch. The Anthropocene Working Group (AWG) established that it should be treated as a formal stratigraphic unit, demonstrated by a reference level commonly known as “golden spike”, still under discussion. Here we show that the onset of bomb-derived plutonium recorded in two banded massive corals from the Caribbean Sea is consistent (1955–1956 CE), so sites far from nuclear testing grounds are potentially suitable to host a type section of the Anthropocene. Coastal coral demonstration sites are feasible, could foster economic development, and may serve as focal points for scientific dissemination and environmental education.

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### 1. Introduction

The Quaternary period (2.6 Myr) is characterized by oscillations of the earth's climate between glacial and interglacial conditions. The Holocene epoch (11.7 kyr) is the present warm interglacial interval after

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the last glacial period, and officially we still live in its Meghalayan Age (last 4.2 kyr; Walker et al., 2018). During the 20<sup>th</sup> century the human species has grown rapidly, in terms of individuals and capacity of transforming nature through global economic growth, energy and resources use, and industrialization, markedly since the 1950s (great acceleration) (Steffen et al., 2007), causing profound changes in the earth system (global change; Stern et al., 1992) such as on-going climate change (Stocker et al., 2014). Humans are now the major geomorphological force (Cooper et al., 2018) and many scientists have naturally adopted the Anthropocene concept to express this severe anthropogenic transformation of the planet's surface (Crutzen and Stoermer, 2000).

The AWG (Anthropocene Working Group, International Commission on Stratigraphy) has recently voted that the Anthropocene should be treated as a formal stratigraphic unit defined by a Global Boundary Stratotype Section and Point (GSSP), i.e. a reference level that would terminate the Holocene epoch and the Meghalayan age (AWG, 2020). A synchronous base is necessary for the Anthropocene to be a formal chronostratigraphic unit of geological time. Briefly, a GSSP (commonly known as a “golden spike”, because a suite of permanent markers should indicate it) should be characterized by a well-defined event that can be identified in other parts of the world (in synchrony and globally). Other requirements include accessibility, conservation, and protection (Waters et al., 2018). The AWG also established that the base of the Anthropocene should be “one of the stratigraphic signals around the mid-twentieth century of the Common Era”, broadly coincident with the great acceleration, and has recognized that the onset of global fallout from thermonuclear tests in the 1950s may be a primary marker (Waters et al., 2015). A clear majority of the AWG members chose plutonium radionuclide signals associated with the “bomb spike” as the best primary marker for the Anthropocene, as this provides the sharpest and most globally widespread signal (Zalasiewicz et al., 2017).

Radioactivity released to the atmosphere by thermonuclear explosions mostly in the late 1950s and early 1960s has left a signature of anthropogenic radionuclides on the entire planet. Approximately 2900 kg of <sup>239</sup>Pu were released in atmospheric explosions, corresponding to about 6.5 PBq of radioactivity. Although <sup>239</sup>Pu occurs naturally in the Earth's crust, its pre-1950 CE concentration is extremely low (~0.05 mBq/kg in typical soils; Syvitski et al., 2020). The fallout from 1945 to 1951 CE was caused by fission devices, resulting in only local contamination. Thermonuclear weapon tests that began in 1952 CE and peaked in 1961–1962 CE left a clear and global signature (Waters et al., 2016). The Limited Test-Ban Treaty of 1963 led to a rapid decline in radionuclide fallout from atmospheric tests during the late 1960s and 1970s (Waters et al., 2019). Although global fallout was spread worldwide, it was affected by i) test location, ii) wind and precipitation patterns, and iii) in the marine environment, by ocean currents and water depth. About three-quarters of global fallout occurred in the Northern Hemisphere (Livingston and Povinec, 2000), mainly in mid-latitudes (Waters et al., 2015).

Global fallout plutonium is routinely measured by mass spectrometry and alpha-spectrometry (in this case together with <sup>240</sup>Pu, half-life 6561 yr). Because of its insolubility, it is highly immobile in sediments, where it is often used to validate <sup>210</sup>Pb chronologies (Sanchez-Cabeza and Ruiz-Fernández, 2012). However, a traditional sedimentary GSSP is not optimum because the temporal resolution depends on the actual accumulation rate, and due to the small time-lapse under consideration, the depositional environment should be varved or very finely laminated. Plutonium can also be measured in hermatypic corals that are easily accessible and usually located in protected areas. Most of these corals show unambiguous annual bands that provide an accurate and precise age model. Here, we report plutonium concentrations in an annually banded coral from the National Park Puerto Morelos Reef (Caribbean Sea, Mexico), part of the Mesoamerican Barrier Reef, and synthesize published plutonium records from other sites, to

contribute towards the establishment of an Anthropocene golden spike, currently in progress by the AWG.

## 2. Materials and methods

### 2.1. Collection of published data

We sourced plutonium concentrations of seven dated coral cores from refereed and published papers (Table 1, Fig. 1), which in some cases included pre-bomb values. In one case (Lindahl et al., 2012) we digitized the plot with the free software PlotDigitizer (version 2.6.6). In another case, the coral core had been re-sampled (Froehlich et al., 2017) and we did not consider the previous data (Froehlich et al., 2016).

A “coral year” is defined between consecutive high-density bands, which usually correspond to summer conditions, and the age is assigned to the year of the younger band (Benninger and Dodge, 1986), e.g. a band spanning from summer 1962 CE to summer 1963 CE is assigned the coral year 1963 CE (and we assigned a central date of 1963.0 CE). We used the mid-year as the band age (e.g. 1963.5 CE for the natural year 1963 CE) when natural years were reported.

The most abundant plutonium isotopes are <sup>239</sup>Pu and <sup>240</sup>Pu. Plutonium analysis in the coral cores published before 1990 (Bikini, Croix, and Plantation) was performed by alpha spectrometry, which usually does not resolve both isotopes and are reported together as <sup>239,240</sup>Pu. Although spectral deconvolution could be used to determine each isotope in environmental samples (Vintró et al., 1996), this was not the case, likely because plutonium activities were low in most cases (Table 1). The shorter-lived <sup>238</sup>Pu can also be analyzed by alpha spectrometry, but it was only reported for the Bikini core where levels were high (Table 1; Noshkin et al., 1975). In this case, the <sup>240</sup>Pu/<sup>239</sup>Pu isotopic ratio was also determined by mass spectrometry. After 1990, mass spectrometry techniques (Thermal Ionization Mass Spectrometry – TIMS, Multicollector Inductively Coupled Plasma Mass Spectrometry – MC-ICP-MS and Accelerator Mass Spectrometry – AMS) were used, so both <sup>239</sup>Pu and <sup>240</sup>Pu are reported. To compare all published results, we used the <sup>239,240</sup>Pu concentration directly determined by alpha spectrometry, and in the case of mass spectrometry i) as the sum of the concentration of both isotopes, or ii) calculated from <sup>239</sup>Pu and the <sup>240</sup>Pu/<sup>239</sup>Pu isotopic ratio.

### 2.2. Analysis of the Puerto Morelos coral

The Puerto Morelos reef is part of the Mesoamerican Coral Reef, in the Western Caribbean Sea and northeastern Yucatan Peninsula of Mexico, and since 1998 it is a protected national park. The reef is close to the Chicxulub bolide impact location that marks the Cretaceous-Paleogene boundary, although its stratotype is found at El Kef (Tunisia).

Using an underwater drill, we collected a ~1 m skeleton core of an *Orbicella faveolata* colony from the Puerto Morelos reef lagoon, Mexico, at ~7 m depth during summer 2016 (Table 1). The growth of *O. faveolata* is controlled by light and temperature, and shows clear annual bands that were dated by sclerochronology with calibrated digital X-ray images, following the methodology described by Rico-Esenaro et al. (2019). The core spanned ~100 yr and plutonium was determined in samples from 1931 to 1977 CE (Supplementary material).

To obtain enough mass for analysis, the outer core slab was used, sclerochronology was transported from the adjacent slab, and yearly aliquots of circa 5 g were sampled. We determined plutonium isotopes by mass spectrometry, following the method described by Röllin et al. (2009). Briefly, samples were dissolved by fusion and purification was performed with TEVA (Triskem International) resins. We measured plutonium isotopes with a Neptune Plus high-resolution double-focusing multicollector ICP-MS (Thermo Scientific Inc.) equipped with nine Faraday collectors and five ion counters. Aqueous solutions were introduced into the ICP-MS using a CETAC Aridus II desolvator (Elemental Scientific Inc.). We assured quality by analyzing the certified reference

**Table 1**

Summary of plutonium records in coral cores. Ages correspond to the mid-point of the analyzed band, except in Enewetak, where bands were sampled sub-annually and they correspond to the reported age. When no pre-bomb values were available, the  $^{239,240}\text{Pu}$  onset year was estimated (-) as the age of the oldest band.

Code	Location Country	Species	Timespan (CE)	$^{239,240}\text{Pu}$ range (mBq kg $^{-1}$ ) year of maximum value (CE)	$^{240}\text{Pu}/^{239}\text{Pu}$ range year of maximum value (CE)	$^{239,240}\text{Pu}$ onset year (CE)
<i>Pacific Ocean</i>						
Bikini	Bikini Atoll	<i>Favites virens</i>	1954–1972	3 700–1 441 890	0.15–0.24	~1954.5
	Marshall Islands			~1954.5	1956	
Enewetak	Enewetak Atoll	<i>Porites lutea</i>	1952–1965	1 100–140 000	0.07–0.58	~1952.8
	Marshall Islands			1958.9	1952.8	
Guam	Guam Island	<i>Porites lobata</i>	1943–1999	4–4 543	0.05–0.46	1951
	USA territory			1954	1953	
Ishigaki	Ishigaki Island	<i>Porites</i> sp.	1937–1997	5–1 073	0.07–0.47	1951
	Japan			1953	1952	
Hawaii	French Frigate Shoals	<i>Porites</i> sp.	1955–1964	150–367	0.19–0.24	~1955
	Hawaii, USA			~1955	1956	
<i>Atlantic Ocean</i>						
Croix	St. Croix	<i>Orbicella annularis</i>	1951–1980	0.2–17	NA	1955
	Virgin Islands, USA			1964		
Plantation	Plantation Key	<i>Orbicella annularis</i>	1957–1967	8–61	NA	~1957
	Florida, USA			1964		
Morelos	Puerto Morelos	<i>Orbicella faveolata</i>	1931–1977	1–98	0.08–0.25	1956
	Mexico			1966	1952	

References: Bikini (Noshkin et al., 1975), Enewetak (Froehlich et al., 2017), Guam (Lindahl et al., 2011), Ishigaki (Lindahl et al., 2012), Hawaii (Buesseler, 1997), Croix (Benninger and Dodge, 1986), Plantation (Purdy and Druffel, 1989), Morelos (this work). NA: not available.

materials IAEA-327, IAEA-384, and IAEA-447. All measurements were close to the certified value.

### 3. Results and discussion

#### 3.1. Plutonium activities and ratios

$^{239}\text{Pu}$  activities and  $^{240}\text{Pu}/^{239}\text{Pu}$  ratios in coral skeletons depend on many variables, such as species, environment, distance to nuclear test sites, and transport processes. Maximum  $^{239,240}\text{Pu}$  activities in coral cores show a striking range of five orders of magnitude, from 17 mBq kg $^{-1}$  (St. Croix, Table 1; Benninger and Dodge, 1986) to 1.4 10 $^6$  mBq kg $^{-1}$  (Bikini Atoll, Noshkin et al., 1975). Many Pacific Ocean cores were collected within or close to nuclear weapons testing grounds (Fig. 1), notably the Marshall Islands (Pacific Proving Grounds - PPG: Bikini and Enewetak atolls), and were directly exposed to close-in bomb fallout.

The first of a series of atmospheric tests of large-yield thermonuclear weapons was performed in Enewetak Atoll (Ivy Mike test) in 1952. A *Porites lutea* core (Froehlich et al., 2017) sub-annually sampled showed plutonium maxima in 1952, 1957, and 1958 CE, reflecting the coral exposure to different tests. In Bikini Atoll, large-yield tests occurred between 1954 and 1958, and a *Favites virens* core showed a maximum in 1954 CE (Noshkin et al., 1975). In other Pacific sites, maxima were also attributed to fallout and transport by large oceanographic currents: a *Porites lobata* core from Guam peaked in 1954 CE (Lindahl et al., 2011), a *Porites* core from Ishigaki island peaked in 1953 CE (Lindahl et al., 2012), and a *Porites* core from French Frigate Shoals Island, Hawaii, likely peaked in 1956 CE (Buesseler, 1997).

During the early 1960s, most atmospheric tests were performed by the former USSR, over 50% of them in Novaya Zemlya (Arctic Ocean), including the largest thermonuclear detonation in 1961, with a yield of 50 MT (UNSCEAR, 2000), leading to the 1963 global fallout maximum. Atlantic coral cores, remote from the main nuclear test sites, showed low plutonium activities (Table 1). Maximum activities in St. Croix and the Florida Keys (Purdy and Druffel, 1989) were observed in 1964 CE. Some delay was expected because plutonium reaches the coral environment by direct atmospheric fallout and continental runoff. The longer delay in Puerto Morelos (1966 CE) was likely caused by a slower transport from the reef lagoon catchment and the absence of rivers in the Yucatan Peninsula. In summary, plutonium concentrations in the Pacific cores showed peaks of different amplitude in the time interval

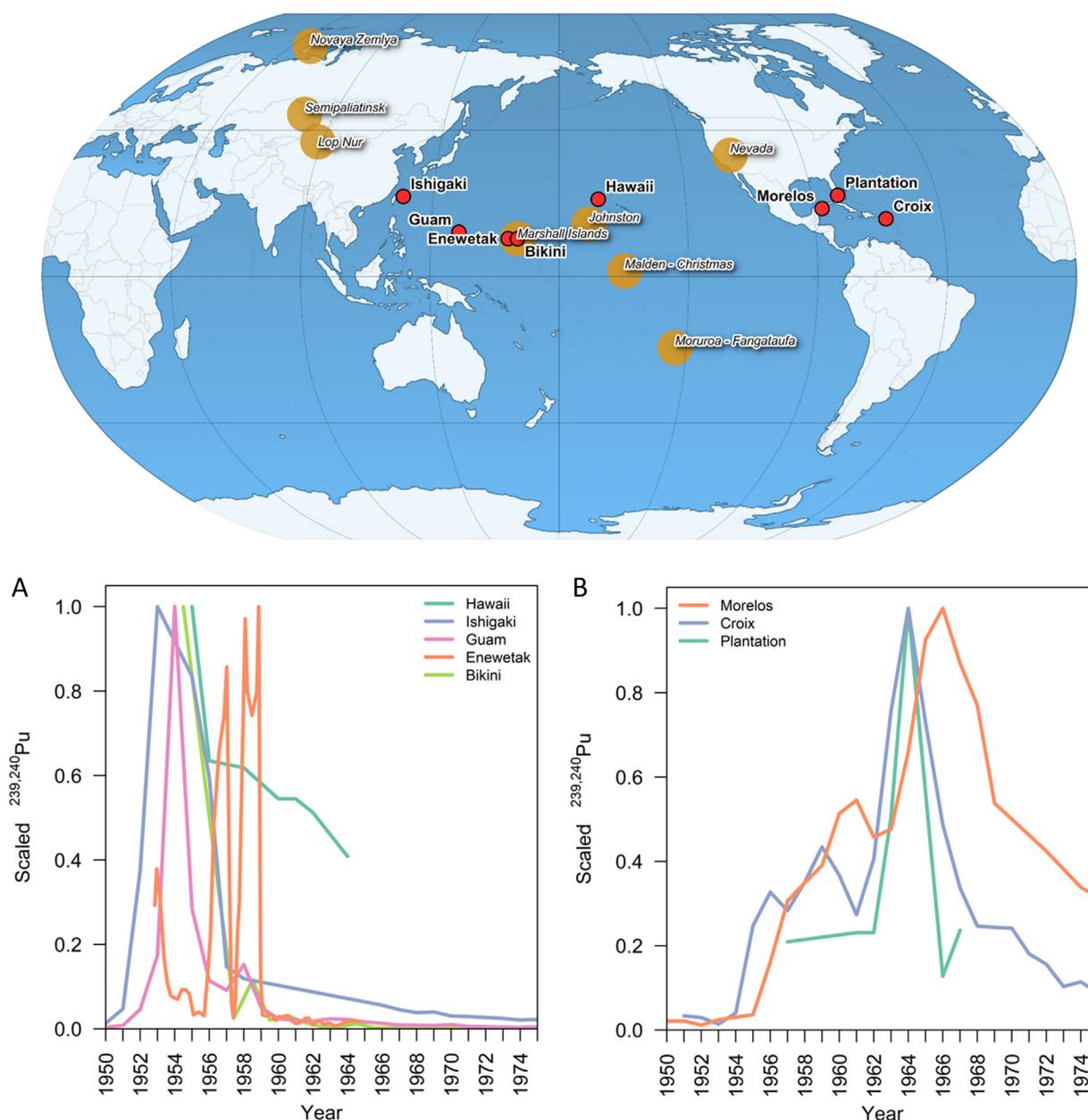
1952–1958 CE, whereas cores from the Atlantic peaked in the interval 1964–1966 CE, closer to the 1963 CE global fallout atmospheric maximum.

The spatio-temporal variability of plutonium isotopic ratios is also high. The highest plutonium ratios were reported in Enewetak (0.58, 1952 CE), Ishigaki (0.47, 1952 CE), and Guam (0.46, 1953 CE). In Bikini, the largest ratio (0.24) was observed in 1958 CE. The plutonium ratio in Pacific corals since the end of large tests in PPG (1958 CE) was similar to the Bikini ratio (Guam: 0.23; Ishigaki: 0.21; Hawaii: 0.20) because the yield at Bikini was about three times larger than at Enewetak. In the early 1960s, the large tests in Novaya Zemlya led to a global fallout plutonium ratio of  $0.176 \pm 0.014$  (Krey et al., 1976; confirmed by Kelley et al., 1999). In Puerto Morelos, the plutonium ratio in 1956 CE was 0.28, close to the Bikini Atoll, and 0.18 during 1959–1969 CE. Therefore, the variability in the timing of plutonium maxima and ratios preclude considering them as good markers of the Anthropocene lower boundary.

#### 3.2. Plutonium onset in corals

An alternative time marker is the onset of plutonium concentrations in corals. In the Pacific Ocean, the onset reflects the first large detonations of thermonuclear weapons (1952 in Enewetak Atoll and 1954 in Bikini Atoll). The Hawaii core does not allow a precise determination of the onset, although 1955 CE might be a reasonable estimation.

The immediate incorporation of plutonium to the coral skeleton (Enewetak core, Froehlich et al., 2017) and the high resemblance of the records of  $^{90}\text{Sr}$  (which should reflect the coral calcification process) and plutonium (Purdy and Druffel, 1989), indicate that plutonium uptake is biologically mediated. Laboratory experiments of coprecipitation of several radionuclides (including plutonium) with calcium carbonate, also indicate that uranium (an analog of oxidized plutonium) uptake is regulated by biological processes (Meece and Benninger, 1993). However, non-biotic uptake of plutonium along the skeleton surface might also occur, such as sorption and coprecipitation with calcium carbonate (Meece and Benninger, 1993), and trapping of suspended fine particles, which would explain the detection of plutonium in older sections. When reported (four coral records) we used pre-bomb plutonium concentrations to establish background means and variabilities. For each core, we calculated the mean and standard deviation of the three oldest pre-bomb values, and we established the plutonium onset when the younger value was larger than the mean



**Fig. 1.** Plutonium concentrations in coral cores. Map: sampling locations of eight coral cores (closed red points, Table 1; see also Supplementary material) and main sites of atmospheric nuclear tests (open orange circles). Plots show plutonium concentrations (scaled by the maximum concentration of each core, unitless) for Pacific (A) and Atlantic (B) corals. Concentrations and uncertainties can be consulted in the dataset provided as Supplementary material. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

plus two standard deviations (95% confidence). The number of pre-bomb values was increased by one until the condition was reached. In the distant sites, the onset was 1955 CE (St. Croix) and 1956 CE (Puerto Morelos). In both Guam and Ishigaki the onset occurred in 1951 CE, likely reflecting the impact of the Ivy Mike explosion in 1952, assuming a one-year uncertainty of the sclerochronology.

The plutonium onset in coral colonies in sites distant from nuclear testing grounds is a good primary marker to select the Anthropocene GSSP location. Although plutonium activities in these corals are low, there is enough technical capability to identify the plutonium onset in banded corals with annual resolution. The plutonium signal is clear in all the species analyzed, and we suggest that this should be the case for all banding hermatypic corals.

A clear correlation of the plutonium onset is also found in ice cores (Gabrieli and Barbante, 2014; Arienzo et al., 2016), where the visual age control is also very good and the absence of mixing can be easily

assessed, but their worldwide distribution is limited. On the other hand, both coastal sediment cores (Corcho-Alvarado et al., 2014; Xu et al., 2018) and lake sediment cores (Thomas and Briner, 2009; Hancock et al., 2011) can show a similar onset, although the age control is limited by the sampling resolution, the sediment accumulation rate, and the dating uncertainties. Other secondary markers, such as  $^{90}\text{Sr}$  and  $^{14}\text{C}$ , might be useful to confirm the onset.

### 3.3. The Anthropocene GSSP location

Some questions arise from the stratigraphic conditions established for a “perfect” GSSP. The corals reviewed here are tropical and from the Northern Hemisphere (NH). Although no plutonium records have been published in gorgonian and scleractinian cold-water corals, they record other Anthropocene tracers such as metals and other radionuclides. As these corals are usually not banded, the use of other

radiometric dating methods (such as  $^{210}\text{Pb}$ ,  $^{226}\text{Ra}$ , and U/Th dating) might be needed.

Plutonium has also been reported in Southern Hemisphere (SH) corals near detonation sites (Moruroa and Fangataufa; Ellis, 1986; Hrnccek et al., 2005). Although plutonium global fallout has been lower in SH, it has also been detected in diverse environmental matrices and locations, for example Southern Ocean surface waters (Hirose et al., 2007), sediments from coastal lagoons, near-shore zones, and lakes in Australia and New Zealand (Hancock et al., 2011), soils from Chile (Chamizo et al., 2011), and even ice and biota from Antarctica (Cutter et al., 1979; Szufa et al., 2018). Furthermore, the onset of plutonium isotopes should also occur in the mid-1950s (Hancock et al., 2011), synchronous with the NH onset. In any case, the plutonium signal should be highest in the NH, where most atmospheric tests of thermonuclear weapons occurred.

#### 4. Conclusions and perspectives

Plutonium concentrations in banded corals are valuable records of global fallout. While maxima are mostly asynchronous and timing is largely affected by nearby detonations, the plutonium onsets in distant sites from nuclear testing grounds are synchronous, including the Southern Hemisphere. This allows for global correlation, either in corals (likely including cold-water corals) or auxiliary stratotypes such as lake and coastal sediment cores, and ice cores. Secondary markers related to both nuclear weapon testing (such as  $^{90}\text{Sr}$  and  $^{14}\text{C}$ ) and global change (such as heavy metals,  $\delta^{13}\text{C}$  and nitrates; Waters et al., 2018) are also widely available. Finally, as coral skeletons are calcium carbonate rocks, a physical golden spike of the Anthropocene in corals is feasible.

Some limitations of the plutonium onset in corals as a primary marker for an Anthropocene type section must be recognized, but “all boundaries are a compromise, since the Earth System is so complex... no boundary is perfect” (Zalasiewicz et al., 2019). Although corals can be sampled with an annual (or even better) resolution, the uncertainty of the best-known GSSP (the Holocene) is ~100 yr (Walker et al., 2009). Therefore, small (a few years) differences of the plutonium onset recorded in corals, caused by proximity to nuclear weapon test sites, plutonium transport with ocean currents (including vertical mixing and scavenging with particles) or from terrestrial runoff, should not be a concern, provided the primary marker (plutonium) is well defined and identified.

Because massive banded corals are mainly found in the tropics, a protected and accessible monument/demonstration site (even submarine) is possible. For example, the National Park Puerto Morelos Reef, protected by Mexican laws, is not far from a major tourist destination (Cancun, Mexico), with large travel facilities and numerous tourist service providers, including diving. Such a site could include many important aspects of the Anthropocene, as hermatypic coral cores are also valuable records of other global change indicators, notably rising temperature, ocean acidification, and contamination. Such a demonstration site could be replicated in other locations, serving as unique sites for local sustainable development, promotion of environmental policies, scientific dissemination, and much-needed environmental education.

#### CRedit authorship contribution statement

JASC, JPCG, PM and ACRF conceptualized the project, acquired funds, provided resources and supervised project development; JASC and ACRF administered the project; JASC, SER and JACA performed analysis, data curation and formal analysis; JASC provided the original draft, and all authors contributed to writing, review and 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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#### Data statement

All data used in this work is provided as Supplementary material.

#### Appendix A. Supplementary data

It includes an MS-Excel file with information on all plutonium records in coral cores used in this work. To compare with previously published data, we report  $^{239,240}\text{Pu}$  concentrations and  $^{240}\text{Pu}/^{239}\text{Pu}$  atom ratios. From these, other variables (such as  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  activities, and  $^{240}\text{Pu}/^{239}\text{Pu}$  activity ratios) can be derived. The data fields are: Site code (see Table 1), Location 1, Location 2, Country, Latitude N (deg), Longitude E (deg), Species, Age: mean age of the analyzed band or coral section, Pu-239,240 (mBq/kg), u(Pu-239,240): uncertainty, Pu-240/Pu-239: atom ratio. u(Pu-240/Pu-239): uncertainty, 1st author, Publication year. Supplementary data to this article can be found online at doi:<https://doi.org/10.1016/j.scitotenv.2021.145077>.

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