**Report to:** Natural Resources Wales

**From:** Keith Barnham, (Correspondence author: [redacted]
Emeritus Professor of Physics, Imperial College London
Denis Henshaw,
Emeritus Professor of Human Radiation Effects, University of Bristol

**Date:** 25th August 2021

# Further evidence for Plutonium Microparticles in the Hinkley Point sediment in EDF's dredging application, Part 2 TR534

## Introduction

For EDF's dredging application that preceded the first Hinkley Point C (HPC) dredge and dump operation in 2018, CEFAS did not directly test the sediment for the alpha emitters sensitive to plutonium contamination. They relied on interpreting measurements of the gamma emission from the americium isotope Am241 to infer the contamination of Pu(241). CEFAS assumed that the plutonium has similar isotopic composition to that discharged from the Sellafield reprocessing plant, to determine an upper limit for plutonium contamination.

The Marine Monitoring Organisation (MMO) has recently approved a new dredging application for which CEFAS have studied 27 core samples for alpha emissions. The CEFAS report [1] concludes that concentrations of the plutonium radionuclides were lower than those estimated using gamma emissions. This report demonstrates that this conclusion is based on an unscientific and inappropriate analysis of the measurements. Instead, the data in Ref.1 confirms the existence of significant amounts of plutonium in the core samples. Some of the evidence suggests the plutonium is in particulate form.

This report should be read in conjunction with Ref. 2, submitted to National Resources Wales (NRW) through the Petitions Committee on 9-3-21. This contained evidence from the report NRPB-M173 for plutonium in the historic waste discharges from Hinkley Point A (HPA) in the form of plutonium microparticles (PMPs).

## 1) CEFAS unscientific interpretation of the alpha measurements

CEFAS conclusion [1] that the alpha spectroscopy data from the core samples indicates plutonium contamination lower than the estimates determined from an analysis of the gamma results, was based on the summary Table 2. Averages of specific activities of Pu(239+240) and AM(241) are presented there. One can only assume that the MMO assessors approved the application without reference to Table 4 in the Appendix where the measurements of all 27 cores are presented. Table 4 clearly shows that these averages were made of data sets that varied by factors of more than 1000. Such an error would be unacceptable in an undergraduate science report that averaged data varying by a factor of 10. CEFAS should have been aware it was inappropriate to average such data sets.

Even if the MMO assessors had only studied Table 2 they should have been aware that the Pu(239+240) and Am(241) averages are inconsistent with the gamma data. The Pu and Am concentration averages were approximately equal (0.12 Bq/kg and 013 Bq/kg respectively) for the alpha data. However, the gamma results in Table 2 differ by a factor of

1.75 (Pu(239+240) = 0.95 Bq/kg and Am = 1.66 Bq/kg). Either the alpha or gamma results in Table 2 are wrong, or both. This should have prompted a close inspection of Table 4. Both results are incorrect as explained in the Sections 2) and 3)

When comparing radionuclides concentrations extracted from gamma and alpha measurements on the same core samples, it is necessary to make a large correction because gamma radiation penetrates far more easily through the mud than alpha particles. Alpha radiation only travels a very short distance in sediment. Hence, the deeper the alpha emitter in the sample the more attenuated the alpha signal when it reaches the detector compared to a gamma emitter at the same depth. It does not appear that such corrections were made to the alpha measurements.

The CEFAS gamma estimate for the plutonium concentration assumed the plutonium had isotopic compositions similar to Sellafield discharges. They should therefore have been aware that the plutonium in the Sellafield discharges is in the form of PMPs which need to be extracted from the sediment by well researched separation techniques **before** the alpha spectrometry measurements are performed. This is another major error which, as discussed in Section 3, severely underestimates the plutonium contamination and its threat to public health.

## 2) Evidence for plutonium in 24 of the core samples in Table 4

In Fig.1 the results of Table 4 for the specific activity of Pu(239+20) are plotted against the Am241 values. The scales are logarithmic to cover the large range of values of the respective signals as discussed in Section 1. The data closely follows a power law dependence over two decades with exponent close to unity. Though the plutonium results in the 3rd and 4th decades are more spread, as expected because the measurements are reaching the limits of detectability, it is impressive that, even below 0.01 Bq/kg, the results are consistent with the trend in the regions of higher detection efficiency.

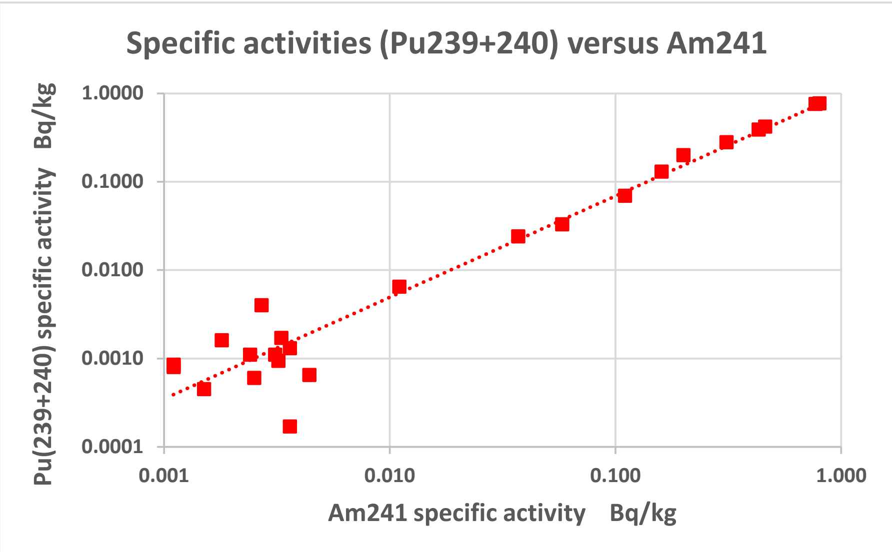


**Fig. 1.** The specific activity (i.e. concentration) of Pu(239+240) is plotted against that of Am(241) on logarithmic scales. The data in the lower decades has a wider spread, as expected as the limits of detection approach. However, the it is still consistent with the trend in the highest decades.

Am241 originates from the Pu241 isotope that was generated at the time the fuel was removed from the reactor core. The consistency of the trend in Fig. 1 over 24 of the 27 core samples tested suggests that all the Am241 was generated from this plutonium in one or more fuel elements that were removed from the HPA reactor around the same time after the same length of exposure. This is the case with the fuel elements containing weapons grade plutonium that were discharged from HPA in 1968 [3,4]. Serious accidents damaged the cladding of the spent fuel elements in the cooling ponds in 1969 [2].

**The self-consistency of the data in Fig 1. confirms the presence of plutonium in 24 of the 27 core samples in Table 4 of Ref.1.**

This report will now concentrate on what can be learnt from the core samples with highest Pu(239+240) and Am(241) specific activity in Table 4. Fig. 2 shows the specific activity of the 10 core samples with the highest Pu(239+240) concentration, plotted against the ratio of this specific activity to the Am(241). The dotted blue line (at 0.572) is the value of the ratio Pu(239+240)/Am(241) predicted by CEFAS on the basis of their gamma measurements assuming the PU(241) content of the plutonium is similar to that found in historic discharges from Sellafield.

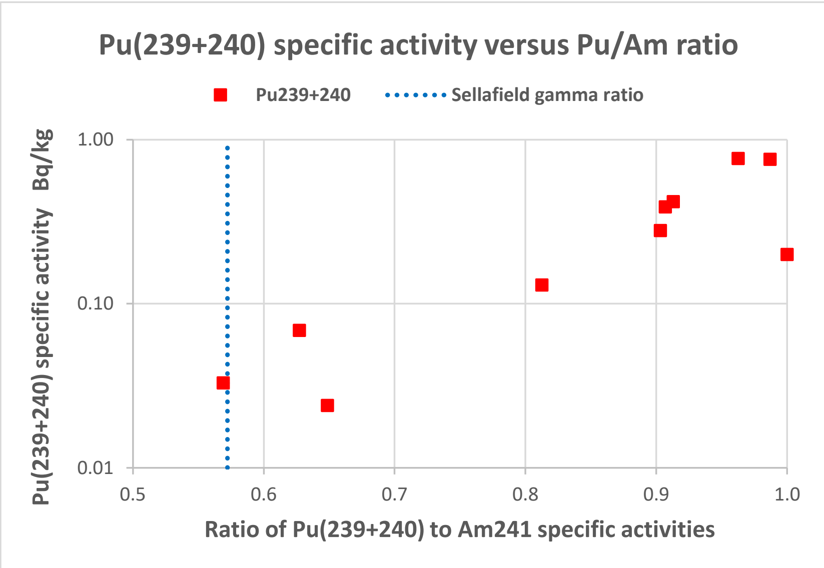


**Fig. 2.** Specific activities of the 10 core samples with the highest Pu(239+240) concentrations plotted against the ratio of Pu(239+240) to Am(241).

CEFAS concluded that, because the plutonium concentration calculated from their (erroneous) averaging of the Pu(239+240) results in Table 4 is **lower** than they derive from their analysis of the gamma data, the alpha measurements can be ignored. However, Fig. 1 shows that 9 of the 10 core samples with the highest plutonium concentration all have a plutonium to americium concentration ratio **higher** than that in the CEFAS gamma analysis.

**Fig. 2 shows that the CEFAS gamma analysis requires correction and is not a "conservative" estimate of the plutonium concentration as they claim.**

The remaining 17 core samples not included in Fig.2 have concentration ratios Pu(239+240)/Am(241) lower than 0.5, but none have ratios greater than 1. This suggests the fall in this ratio results from different sediment penetrations at the different alpha radiation energies from Pu and Am(241). This further confirms the importance of correcting the alpha signal for penetration depth that CEFAS ignored.

In addition to the alpha measurements of Pu(239+240) and Am(241), Table 4 contains the results of measurement of the alpha signal for the plutonium isotope Pu(238). Fig. 3 plots the Pu(239+241) and the Pu(238) concentrations against the Am(241) specific activity for the 10 core samples with the highest plutonium concentration.

The consistency of these isotopic ratio with the trend lines in Fig. 3 is further evidence for **the presence of plutonium radionuclides in 10 of the core samples**.

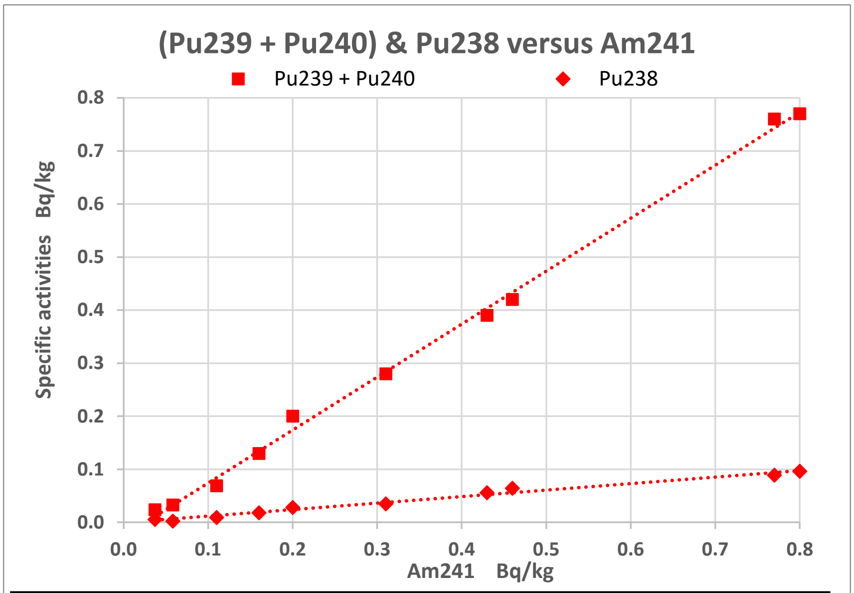


**Fig. 3.** Specific activities of Pu(239+241) and Pu(238) plotted against the Am241 measured in the same sample for the 10 core samples with highest plutonium concentration.

## 3) Evidence in Table 4 for Plutonium Microparticles in the core samples

Ref.2 presents plutonium alpha emission data from the report NRPB-M173 which shows two large peaks in the plutonium contribution in the liquid waste discharged from Hinkley Point A (HPA) in 1969 and 1982. All the plutonium discharged from HPA recorded in NRPB-M173 must have been in particulate form of less than 5 micron diameter, as this is the aperture size of the filters at the exit of the HPA cooling ponds. This is where the accidents that compromised the spent fuel cladding occurred in 1969.

Ref. 5, describes the definitive study of PMPs in estuaries around Sellafield resulting from historic plutonium emissions from Sellafield. Radiochemical separation of the sediment samples resulted in a range of grain sizes. The plutonium content of the grains was measured by alpha spectroscopy. Table I presents their results for the largest and the two smallest grain sizes.

The 5.5 µm PMP size had the highest specific activity of the grain sizes studied. All the plutonium that leaked through the HPA filters into the sediment will still, in 2021, have these extremely high specific activities given Pu(239) has a 24,000 year half-life.

| Table 1 | Pu(239+240) | Pu(238)/P(239+240) |
|---|---|---|
| Grain size | Bq/kg | Ratio |
| < 63 µm | 306 +/- 14 | 0.25 +/- 0.02 |
| < 5.5 µm | 15,642 +/- 59 | 0.27 +/- 0.01 |
| < 2.0 µm | 13,279 +/- 240 | 0.26 +/- 0.26 |

Table 1 shows the specific activity falls dramatically if grain size increases to 63 µm, suggesting that the PMPs giving the alpha signals in Table 4 are near the surface. There must be a comparable concentration of PMPs throughout the sample each emitting many thousands of alpha particles a second that never reach the surface and detectors. Specific activities in CEFAS Table 4 are a gross underestimate of the PMPs throughout each sample.

In Fig.4 the Pu(238) data points have been scaled up by the inverse of the average branching ratio given by the trend lines in Fig.3. The scaled Pu(238) points add 10 more determinations of the plutonium concentration at the surface. The regular increase in the Pu(239+240) level in Fig.4 is possibly due to different PMP grain sizes. A random increase is expected from variations in sediment absorption. This hypothesis is supported by noting in the 10 core samples there are two pairs of cores with similar values. This regularity is consistent with the plutonium signal in the sediment being entirely particulate.

The Pu(238)/Pu(239+Pu240) branching ratio from the trendlines in Fig. 3 is 0.123. This is around half the values in the table observed in the PMPs from Sellafield in the table. This is consistent with the Hinkley Point PMPs resulting from the low burn-up, weapon's grade Plutonium discharge in liquid waste from HPA due to the 1969 cooling pond accident.

**This is further evidence, additional to that in Section 1) that CEFAS' extraction of a plutonium concentration from the AM(241) gamma data by a model based on the isotopic composition and spent fuel extraction dates of Sellafield plutonium is incorrect.**

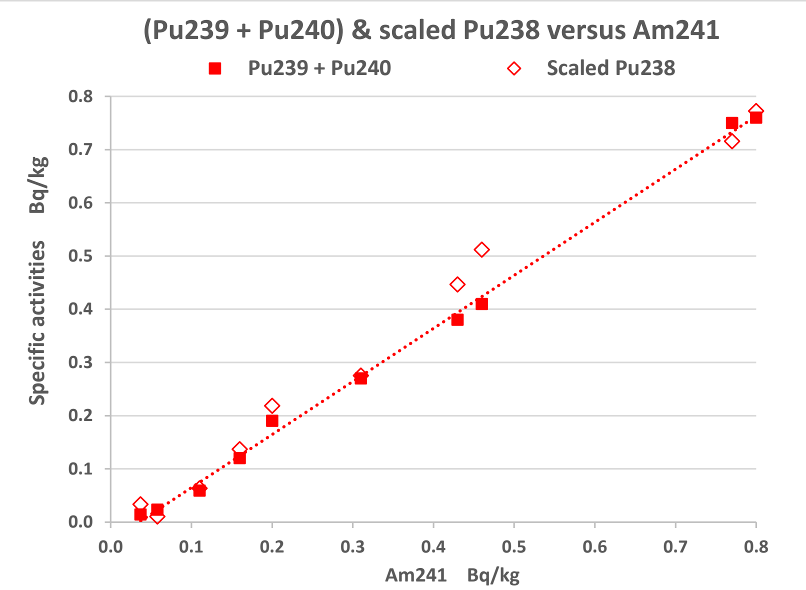


**Fig.4.** Open diamonds represent the Pu(238) data points in Fig. 3 scaled by the inverse of the average branching fraction of the trend lines in Fig.3.

## 4) Summary of errors in the CEFAS interpretation of alpha measurements

i. Averaging data sets that vary by factors of more than 1000.
ii. Not noticing the alpha and gamma results for the Pu/Am ratio in Table 2 differed significantly
iii. Comparing alpha and gamma results without sediment penetration corrections.
iv. Using the isotopic ratios appropriate to Sellafield high burn up plutonium rather than weapon's grade plutonium from HPA in modelling the extraction of a plutonium signal from gamma measurements.
v. Not using 1968 as the date the bulk of the weapons grade plutonium in the sediments was removed from the HPA core in the modelling of a plutonium signal from the gamma measurements.
vi. Not being aware that the plutonium in the Sellafield discharges is in the form of PMPs which need to be extracted from the sediment by well-known techniques **before** the alpha spectrometry measurements graphic analysis is performed.
vii. Not sending the samples for testing by particulate sensitive techniques such as the radiochemical grain separation techniques of Ref. 5 as soon as an alpha spectroscopy signal was detected at the surface of the core samples.

## 5) Conclusions and Recommendations

The plutonium in the Hinkley Point sediment consists of microparticles less than 5 microns in diameter. Such PMPs represent a very serious health risk as they can be inhaled. They have been observed around Sellafield, blown back in sea spray. If inhaled into the lungs the short range of their high energy alpha radiation can severely damage internal organs. PMPs less than 1 micron in size can readily pass through the lung to reach all the organs in the body including the brain and a fetus.

Our analysis shows that CEFAS estimates of plutonium concentration are likely to be a gross underestimate of the plutonium content of the sediments dumped into Cardiff Bay in 2018.

We request that National Resources Wales forward this report, and re-submit Ref. 2, to the Environment Agency, for their urgent attention. We recommend the following:

i. EDF to halt all dredge and dump preparations and all civil engineering work on HPC involving excavations and tunnelling, while the workers concerned receive lung-burden tests for alpha emitters, as is routine for some Sellafield workers.
ii. NRW to request the Environment Agency order the Marine Management Organisation to reconsider their approval of EDF's application.
iii. NRW to request the EDF pass the samples to an independent body to perform radiochemical grain separations and alpha spectroscopy as in Ref. 5.
iv. NRW to request the Westminster Government urgently institute an independent, public enquiry of how the historic evidence for plutonium leaks from Hinkley Point A was covered up and why the sediment dumped in Cardiff Bay was not analysed for

PMPs given the known plutonium leaks resulting from the cooling pond accidents in 1969.

## References


1) EDF, TR534, “Radiological Assessment of Dredging application for Hinkley Point C. Part 2”, (2020).
2) Keith Barnham, “Misleading information from the Environment Agency concerning evidence for Plutonium Microparticles in the Hinkley Point sediment
3) K.W.J.Barnham et al., *Nature*, **407**, 833, (2000).
4) K.W.J.Barnham et al., *Nature*, **317**, 213, (1885).
5) S.R.Aston. D.J.Assinder & M.Kelly, *Estuarine, Coastal & Shelf Science* (1985), **20**,761