Assessment of the Performance of the ^{210}Pb-Based Dating Models with a Challenging Sediment History in Maryport Harbour (UK)

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SUPPLEMENTARY MATERIAL

Supplementary S1. A summary of the radiometric dating models.

This annex presents a brief summary of the model equations for the CRS, CFCS, and TERESA models which are used in this work. Their basic assumptions have been presented in the Introduction and Methods sections. All these models share the assumptions of no post-depositional redistribution, continuity of the sedimentary sequence, and ideal deposition of fluxes and matter at the SWI. Additional assumptions are highlighted in this summary. The models are formulated in terms of the mass depth scale, m (Eq. 3).

CRS model

The CRS model assumes a constant $^{210}Pb_{exc}$ flux onto the SWI and a steady state for the total inventory. It works with the $^{210}Pb_{exc}$ inventory below the sediment horizon at mass depth m, Σ_m:

$$\Sigma_m = \int_m^\infty A(m')dm'. \quad \text{(S1)}$$

The total inventory below SWI, Σ_0, is given by Eq. S1 for m = 0. Assuming a steady state Σ_0, the age of the sediment horizon at the mass depth m is:

$$T(m) = \frac{1}{\lambda}\ln\left(\frac{\Sigma_0}{\Sigma_m}\right) \quad \text{(S2)}$$

The assumption of steady state Σ_0 is mandatory for the model construction and relates the radioactive decay of Σ_0 with the constant flux at the SWI: $F = \lambda\Sigma_0$.

CFCS model

The CFCS model assumes a constant $^{210}Pb_{exc}$ flux on the SWI, F, and a constant SAR, w. Then the age of the sediment horizon at mass depth m is $t_m = m/w$. The above assumptions imply a constant initial activity concentration $A_0 = F/w$. Downcore this initial activity concentration only decreases with radioactive decay, and thus for the sediment horizon at mass depth m:

$$A(m) = A_0 e^{-\frac{\lambda}{w}m}. \quad \text{(S3)}$$

The exponential fit of the $^{210}Pb_{exc}$ vs. mass depth profile to Eq. S3 allows estimating the sedimentation rate and then solving the chronology from the mass depth scale.

TERESA model

The fundamentals of the TERESA model and its validation against synthetic cores and real data from varved sediments can be found in the work of Abril (2016) [20], and a multimodal version has been presented in Abril (2020) [18].

The model assumes that $^{210}Pb_{exc}$ fluxes are governed by 'horizontal inputs', so there is a statistical correlation between fluxes and SARs [5].

For a sediment core that has been sectioned into N slices of mass thickness Δm_i, (i =1, 2,...N), each has an associated age interval, ΔT_i, a mean SAR value wi, and an associated initial activity concentration $A_{0,i}$ (the one encountered for the sediment slice in SWI). The TERESA model operates with SARs and initial activities for which slice i adopts the values (w_i, $A_{0,i}$), both varying along the core, but closely following normal distributions around their respective arithmetic

mean values, $\bar{w}$ and $\bar{A}_0$, with standard deviations σ_w and σ_A, respectively, being s_w and s_A their typified values (standard deviations divided by mean values). Providing a first estimation of $\bar{w}$, $\bar{A}_0$, s_w and s_A, the model generates independent random distributions for w_i and $A_{0,i}$ by using two randomly sorted sets of N normal-typified $z_{1,i}$ and $z_{2,i}$ values:

$$A_{0,i} = \bar{A}_o(1 + z_{1,i}s_A) \; ; w_i = \bar{w}(1 + z_{2,i}s_w) \qquad \text{(S4)}$$

An algorithm solves their best arrangement down-core to fit the experimental $^{210}Pb_{exc}$ versus mass depth profile, generating then solutions for the chronological line and for the histories of SAR and fluxes [20]. The result depends on the first estimation of $\bar{w}$, $\bar{A}_0$, s_w, and s_A. The model applies a mapping technique by iterating the whole process for each parameter varying over a wide range. The error function, Q^2, measures the overall quality of the fit for each individual run of the model.

$$Q^2 = \sum_{i=1}^{N} \frac{(A_{th,i} - A_i)^2}{\sigma_i^2} \; ; \quad \chi^2 = Q^2/f; \qquad \text{(S5)}$$

where A_i and σ_i are, respectively, the measured value and the analytical error of the activity concentration of the slice with index i; $A_{th,i}$ is the corresponding value estimated by the model and f is the number of degrees of freedom. Parametric maps of the χ-function serve to find the best solution and to support error estimates [20,35]. Alternatively, the model's output can be better constrained with time markers, when available.

It should be noted that TERESA does not need the total inventory of $^{210}Pb_{exc}$, but activity measurements must be continuous over the sediment slice sequence.

The codes written in Quick-Basic and all the associated files required for TERESA and the newer version Multimodal-TERESA are available in the multimedia component of the paper Abril (2020) [18]: https://doi.org/10.1016/j.quageo.2019.101032

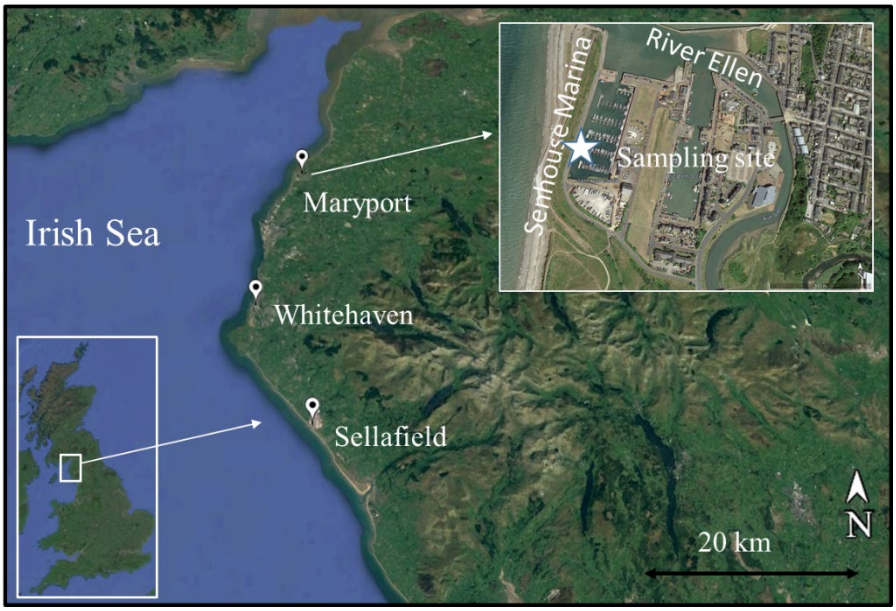

Figure S1. Map showing the location of the core sampled in February 1988 (the former Senhouse dock was transformed to a marina in 1990). Radioactive liquid wastes are released to the Irish Sea by the Sellafield Nuclear Reprocessing Plant since 1952. In Whitehaven, the phosphate industry has released its wastes to the sea since 1954. They contained enhanced levels of natural-occurring radionuclides. Both sites are also sources of gaseous emissions.

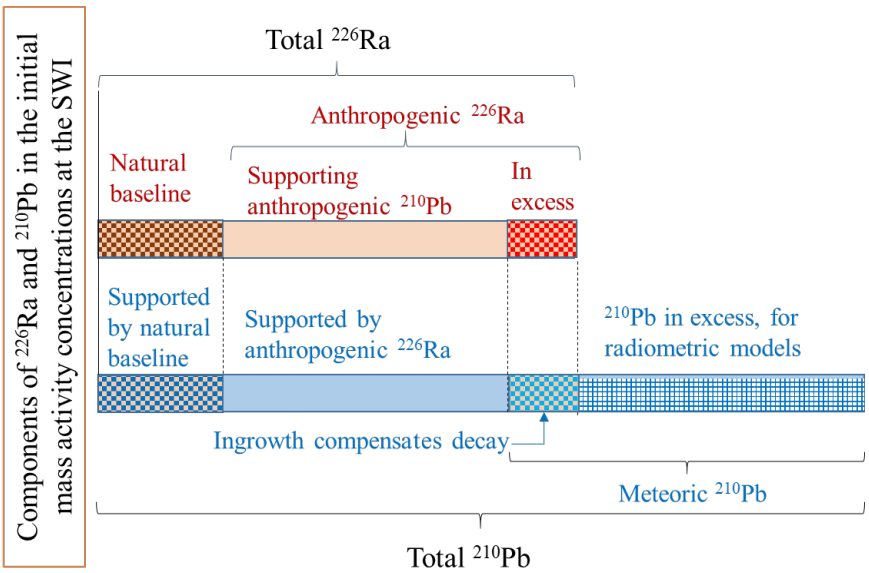

Figure S2. Sketch illustrating the different components of ^{226}Ra and ^{210}Pb in the initial mass activity concentrations at SWI for the core studied, impacted by ^{226}Ra releases from the phosphate plant at Whitehaven, in radioactive disequilibrium with its progeny radionuclide ^{210}Pb.

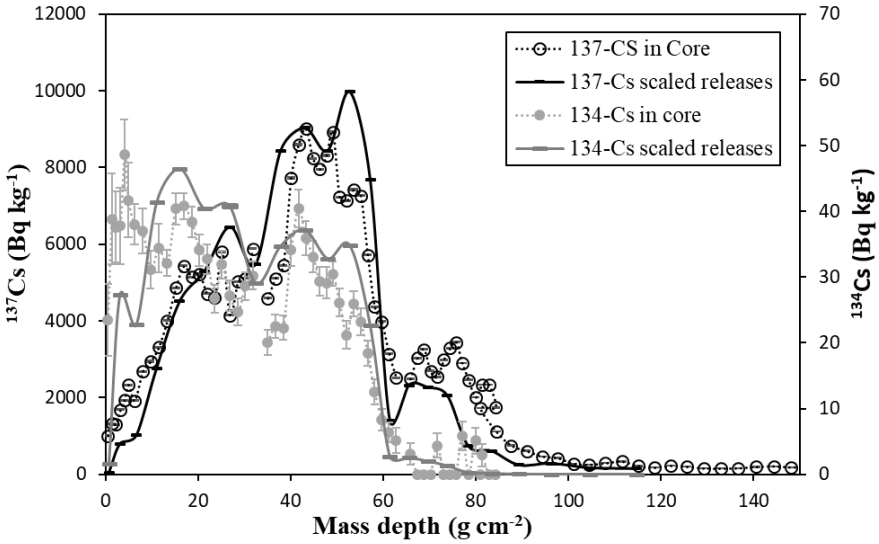

Figure S3. ^{137}Cs and ^{134}Cs versus mass depth in the sediment core from Maryport, compared with the records of their decay-corrected releases in Sellafield (data from Gray et al., 1995 [23]) using the age model of Fig. 4, a lag time of 1.5 yr. and a global scaling factor of $2.6 \cdot 10^{-12}$ yr. kg^{-1}. Data in core and chronology adapted from Kershaw et al. (1990) [22].

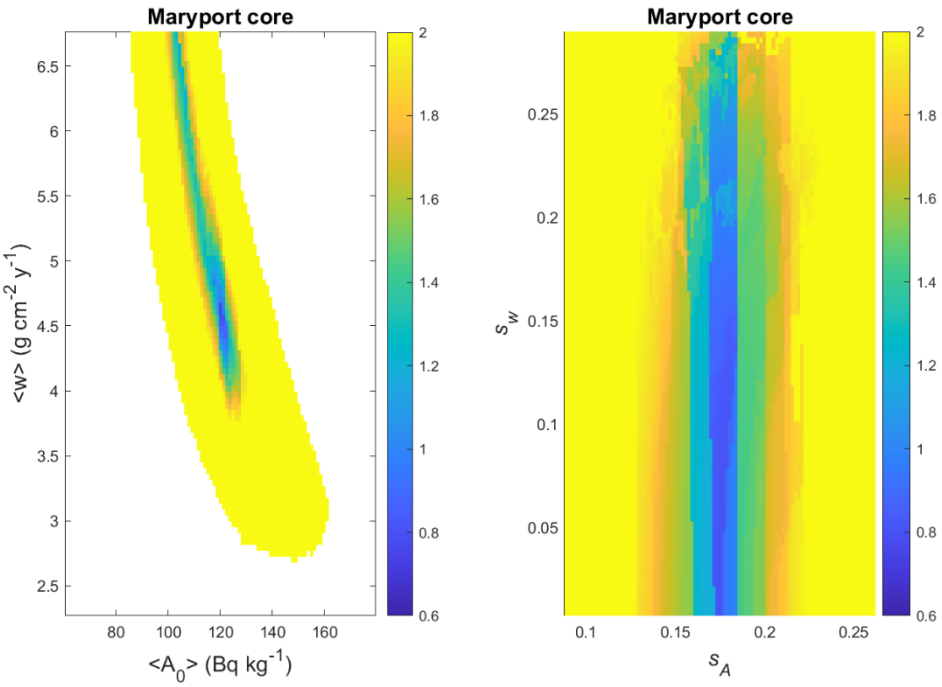

Figure S4. Mapping of the χ function (Eq. A5) for the sediment core of Maryport (100x100 pixels per plot). The left panel shows its projection onto the ($\bar{A}_0$, $\bar{w}$) plane (arithmetic means are denoted here as < >), while its projection onto the (s_A, s_w) plane appears in the right panel. The darkest areas correspond to the lowest χ values. They define the best values for the fitting parameters shown in Table 2.

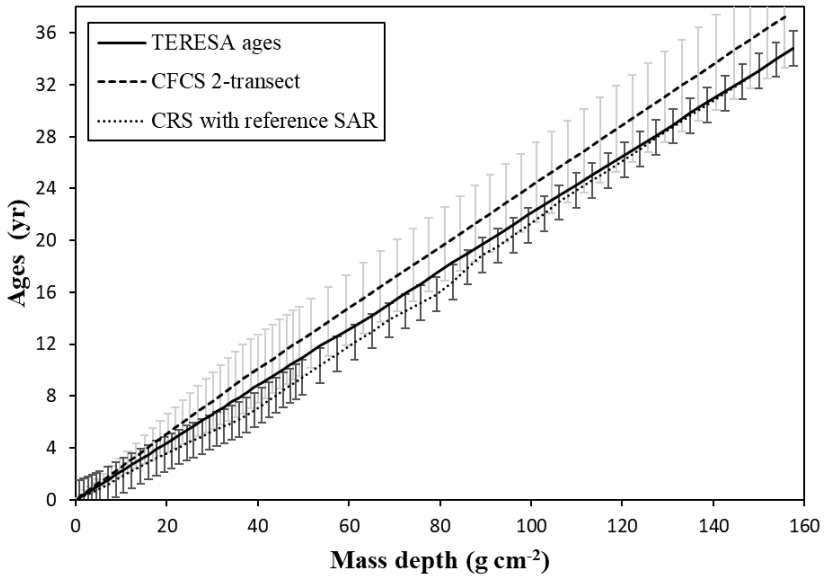

Figure S5. Chronologies for the Maryport sediment core produced with the models TERESA, piecewise CFCS with 2-transects (Fig. 5), and CRS with the reference SAR method, this last estimated from the second transect of the previous model. Note that the age of 38 years is the upper limit for the base of the retrieved core, as it did not reach the level of the dredging in the early 1950s.